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HISTORY OF THE OAK RIDGE NATIONAL LABORATORY
1943 – 1963

FIRST ROUGH DRAFT

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Compiled by
W. E. Thompson

August 23, 1963

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HISTORY OF THE OAK RIDGE NATIONAL LABORATORY 1943 - 1963

PART I THE PLUTONIUM PILOT PLANT

INTRODUCTION

The Oak Ridge National Laboratory, originally known as the Clinton Laboratories, was established in 1943 as a direct result of several events which had occurred in the preceding two years. In 1939 scientists discovered that when uranium atoms were bombarded with neutrons, they would split into approximately equal parts with the release of enormous quantities of energy. This phenomenon of nuclear fission was investigated at many laboratories, and it was soon discovered that the uranium 235 isotope was the particular atom that was fissionable. The uranium 238 isotope, which makes up 99.3% of natural uranium, was found not to be fissionable with slow neutrons. On the basis of work carried out in the interests of pure science in 1939 and 1940, the possibility of obtaining vast quantities of power from uranium fission was visualized by the scientists. Cyclotron experiments showed that when uranium is bombarded with neutrons, a new element, plutonium, is produced. It was discovered that plutonium is produced from nonfissionable uranium 238 and that the plutonium 239 isotope thus produced is fissionable. A group of leading scientists recommended to President Roosevelt that the government undertake a program of research and development aimed at providing a method for the exploitation of the tremendous potential power of nuclear fission to contribute to the war effort. As a consequence, it was decided that the Office of Scientific Research and Development (OSRD) should sponsor a program of research and development in this field.

Theoretical considerations had indicated that plutonium 239 could be produced in quantity by neutron bombardment of the nonfissionable

uranium 238 isotope, which is 140 times as plentiful in natural uranium as the fissionable uranium 235 isotope. If a nuclear chain reaction could be made to occur in natural uranium, some of the neutrons generated by the chain reaction would be absorbed in uranium 238, changing it by nuclear reactions to plutonium 239. Under the Office of Scientific Research and Development, a "Metallurgical Laboratory" project was established early in 1942 at the University of Chicago to develop a system in which natural uranium could be made to sustain a nuclear chain reaction. Also important among the objectives of the Metallurgical Laboratory were the development of methods for chemically separating and purifying the plutonium produced in a natural uranium chain reaction and for using purified fissionable material to effect a chain reaction of explosive characteristics which would make a super bomb.

In December, 1942, at the University of Chicago, work performed under the project established by the Office of Scientific Research and Development led to successful operation of the first nuclear reactor, which, although it was a very low-power laboratory research device, nevertheless clearly demonstrated that a self-sustaining nuclear chain reaction could be established under certain conditions with natural uranium. The chain reaction was found, as expected, to supply tremendous numbers of neutrons which, by their nuclear reaction with uranium 238, produced plutonium 239.

Thus by the end of 1942, it was clearly recognized that nuclear fission offered exciting possibilities as a new source of almost unbelievable power. It was also known, at least theoretically, that two fissionable isotopes, uranium 235 and plutonium 239, could be produced in sufficiently large quantities to be used in a new type of weapon that

would unleash the tremendous power available from nuclear fission in a devastating explosion. While natural uranium, containing only seven-tenths of one percent of fissionable material, requires several tons of uranium to establish a nuclear chain reaction, scientists reasoned that if fissionable material could be produced in pure form, the quantity required for a nuclear chain reaction might be quite small. Production of purified fissionable isotopes thus became an important objective.

Fissionable uranium 235 can be separated from the uranium 238 which comprises over 99% of natural uranium only by processes which exploit the difference in mass of the isotopes - 235 versus 238 - because their chemical behavior is identical. Experiments showed several physical processes which offered promise for separating the U-235 isotope from natural uranium. The experiments also showed that production of large quantities of U-235 would be a huge industrial operation requiring the enormous expenditures.

Fissionable plutonium appeared equally as promising for making a super bomb and had the advantage of being, not an isotope of uranium, but a different element which could be separated by chemical means from the uranium in which it was produced. Since plutonium was a new element which had not existed in nature, its chemical properties were unknown; but theoretical considerations made it nearly certain that the chemical separation could be accomplished. In this case also, the production of large quantities of plutonium would require huge industrial facilities involving large expenditures of money and scientific effort.

Although it was not known whether the technological problems of large-scale production of uranium 235 or plutonium 239 could be solved, experimental results available by the end of 1942 indicated that they

could. Four methods of producing fissionable material appeared promising: (1) electromagnetic separation of uranium isotopes, (2) separation of uranium isotopes by gaseous diffusion through a porous barrier, (3) separation of uranium isotopes by centrifugation, and (4) production of plutonium 239 by the interaction of neutrons with uranium 238 in a nuclear chain reacting system. At the end of 1942 it was not possible to state definitely that any one method appeared superior to the others. It was considered unsafe, in view of the recognized gaps in information available at that time, to concentrate on any one method of achieving the production of fissionable material.

With the increasingly promising results that were being obtained during 1942, it was decided to proceed with plans for a major national effort to produce fission bombs. Theoretical and experimental studies indicated strongly that it would be possible to produce a super bomb which would have a decisive effect on the outcome of the war. In the latter part of 1942 the Army organized a new Manhattan District, under the Corps of Engineers to manage the activities directed toward producing an atomic bomb.

Land was acquired in 1942 in the area between Clinton, Kingston, and Oliver Springs, Tennessee, under the guise (for security reasons) of establishing the Kingston Demolition Range. The Corps of Engineers started construction of a town and administrative office buildings in 1942 under the name Clinton Engineer Works. This was the area planned for the large-scale production activities required to produce an atomic bomb. It was originally planned that all atomic bomb project activities would be carried out at this site.

With the successful operation of the "Chicago Pile" in December, 1942, construction of a pilot plant for the production and chemical separation of plutonium was considered justified. The Metallurgical Laboratory at the University of Chicago, having responsibility for this phase of the atomic bomb project, was also responsible for the operation of the pilot plant. Because of the urgency of the work, the ordinary procedure of completing pilot plant tests prior to the undertaking of full-scale production activities was not to be followed. A possibility was recognized that something might cause a large nuclear chain reaction to release fission products which are highly radioactive, and therefore it was decided that large-scale plutonium production facilities should not be located at the Tennessee site, but at some remote location where fewer people would be endangered by an accident. Plans were made for the construction of a full-scale production plant in Hanford, Washington, a site chosen because of its isolation and the abundance of cold, pure water in the Columbia River and the availability of large quantities of electric power. In order that the pilot plant might be enough ahead of the production plant to permit experience gained in pilot plant operations to be applied in the design, construction and operation of the production plant, steps toward construction of the pilot plant were accelerated as soon as successful operation of the "Chicago Pile" was achieved. The production plant at Hanford, Washington was to be a large-scale enterprise, and a large industrial organization was needed to manage the design, construction and operation of such a huge plant. The duPont Company was considered by the Manhattan District to be the best qualified organization for the job. DuPont agreed to accept the responsibility for the plutonium production plant, and because of this, also accepted responsibility for

the design and construction of the pilot plant at Clinton Engineering Works in Tennessee. Although the pilot plant was to be actually operated by the Metallurgical Laboratory, it was expected that the duPont Company would train personnel at the pilot plant for later assignment to the production facilities. Construction of the pilot plant started on February 1, 1943, the first plant construction in the Clinton Engineer Works area. Because of the location in the Clinton Engineer Works, the name Clinton Laboratories was chosen for the pilot plant being constructed by duPont for operation by the Metallurgical Laboratory of the University of Chicago.

It had been demonstrated that nuclear chain reactions could be achieved in natural uranium and that plutonium 239 was produced from uranium 238 in the chain reacting system. However, all that was known about plutonium production was based on theoretical studies, or at best on very small-scale laboratory experiments, and it was quite apparent that a great deal of research and development would be required before large-scale production activities could be started. The Clinton Laboratories pilot plant was established to carry out the research and development in preparation for large-scale production of plutonium. The Metallurgical Laboratory at Chicago performed basic research on plutonium chemistry and worked out the basic processes to be tested in the pilot plant for separating plutonium from uranium and from fission products. It is interesting to note that all research performed until the successful operation of the pilot plant yielded larger quantities of plutonium, was performed with cyclotron-produced plutonium, of which never more than one millogram was available for all research at the Metallurgical Laboratory.

Early in 1943 scientists were confident that a nuclear chain reaction utilizing highly purified fissionable material could be made to cause an explosion of unprecedented magnitude. Top priority projects were initiated under the Manhattan District to produce purified uranium 235 and plutonium 239 by several methods. Plants for separation of uranium isotopes by gaseous diffusion and electromagnetic processes were started early in 1943 at the Clinton Engineer Works in Tennessee.

The Metallurgical Laboratory of the University of Chicago, in these early days, was responsible for all phases of research and development on fission chain reactions, for plutonium production and chemical processing methods, and for the development of methods for making a super bomb using either fissionable uranium 235 or plutonium 239. The plutonium production and chemical processing phases of the atomic bomb project were to be tested in a pilot plant known as Clinton Laboratories, constructed especially for this purpose.

Simultaneously another new laboratory was set up at a very remote location in Los Alamos, New Mexico to pursue development of the bomb itself. The work which had been started under the Metallurgical Laboratory of the University of Chicago had actually been carried out by a group working in laboratories at the University of California. When plans for the new laboratory at Los Alamos were made, the University of California accepted responsibility for this phase of the work, leaving the Metallurgical Laboratory free to concentrate on plutonium production and chemical processing.

The Oak Ridge National Laboratory had its beginning in the pilot plant, Clinton Laboratories, set up to test the plutonium production methods planned by the Metallurgical Laboratory of the University of

Chicago. The need for purified fissionable material was urgent, less for the production of bombs than for research and development on how to produce a bomb. Clinton Laboratories would supply the first gram quantities of purified fissionable material as well as test the production processes.

THE ESTABLISHMENT OF CLINTON LABORATORIES

In January, 1943, the decision was made to construct an intermediate energy reactor at the Clinton Laboratories location to serve as a pilot plant for the large-scale production operations at Hanford, Washington. During the early discussions, a reactor operating at 30 to 100 KW was considered; however, the decision to enclose the uranium metal in protective "cans" to reduce the oxidation of uranium and to minimize the release of fission products allowed the reactor to operate at higher temperatures and thus made possible a power level of several hundred kilowatts. The design power level of 1,000 KW was agreed on during February, and considerable work was done toward designing the reactor and chemical processing facilities for Clinton Laboratories. At the time, early in 1943, it was planned that the large plutonium production reactors would be helium cooled. The Clinton Laboratories reactor was to operate at a much lower power level, and air cooling was considered acceptable and more expedient. (Later it was decided that the large production reactors at Hanford should be water cooled.) The chemistry and process engineering work at the Metallurgical Laboratory had provided the essential background for a chemical process that would separate the plutonium from uranium and from the highly radioactive fission products. Equipment design for the Process Building was well under way in the early months of 1943.

The research studies and experimental investigations performed under the supervision of the Office of Scientific Research and Development had been conducted on the campus of the University of Chicago by scientists who were connected with the University. When the Manhattan District took over the atomic bomb project, they considered it essential that the

assistance of the existing Metallurgical Laboratory organization, which was one of few in the world experienced in the new field of nuclear energy research, be maintained in order to assure optimum progress in continuing the program into pilot plant and production phases. Therefore, the University of Chicago was chosen as the contractor to operate the Clinton Laboratories pilot plant, under the Manhattan District. For security reasons, it was not desirable to have the University of Chicago's name associated with the work which was to be performed in Tennessee; therefore, an organization known as the "Clinton Laboratories" was formed to operate the pilot plant.

The scope of work for which the Metallurgical Laboratory of the University of Chicago was responsible under the Manhattan District contract consisted of the following functions:

(1) Procure and train additional personnel as may be required to perform research and development on plutonium production and processing.

(2) Conduct the required research and engineering development work necessary to accomplish the development of processes and design of facilities for producing plutonium in a nuclear chain reaction and to separate and purify the plutonium.

(3) Train personnel and operate a pilot plant for the purpose of producing a small amount of plutonium.

(4) Make the necessary studies and develop a workable and dependable method of chemical separation and isolation of plutonium from uranium metal and from fission products.

(5) Organize and operate a training school for the technical personnel who would serve as a nucleus for operating the large production plant at the Hanford Engineer Works.

(6) Develop a process for the recovery of partially depleted uranium metal which had been irradiated and used in the development work at the pilot plant.

(7) Develop methods and produce certain radioisotopes for use at other locations on the project.

(8) Procure the necessary personnel to conduct studies and investigations with respect to the medical and biological problems associated with work utilizing radioactive materials.

CONSTRUCTION OF FACILITIES

While plans and arrangements were being made for the work to be performed at Clinton Laboratories, the Engineering Division of the E. I. du Pont de Nemours and Company, Incorporated was assigned the responsibility for constructing the Clinton Laboratories facilities. Construction was actually started on February 1, 1943, and proceeded on a rush schedule, which enabled the reactor to be completed and placed in operation on November 4, 1943. The reactor, of course, was the major facility around which the research and development activities at Clinton Laboratories were to be centered. Even though it was only the second reactor ever to be constructed, indeed the first to operate at a power level high enough to provide an appreciable potential for producing plutonium, the first to employ forced air cooling, and the first to incorporate complete provisions for shielding, controls, and other operating requirements, the Clinton Laboratories reactor operated from the start with better than expected performance.

A second major facility constructed at Clinton Laboratories was the chemical pilot plant where the process for separating and purifying plutonium was to be tested. Because chemical operations involving radioactive materials had never before been attempted on anything even approaching the scale planned in the pilot plant, the design and construction of these facilities was an engineering achievement of the first magnitude.

All equipment for the operations was enclosed in "hot cells" surrounded by five feet of concrete. Remote control was required for even the simplest operations, which were accomplished by workers who

could not even see the operation they were performing. At one stage of the process, a television set was used to allow observation of a critical operation. For the most part, the performance of equipment was determined by the readings on a bank of instruments which covered the walls of the "operating gallery".

Other facilities constructed at the same time included laboratories for chemistry, physics, and medical (health physics) research; machine shops, instrument shops; and several administration buildings and warehouses. One hundred and fifty buildings, large and small, were constructed at a cost of about $13,000,000. More than 3,000 construction workers were on the job during the peak construction period, which lasted from February 1, 1943, through all of that year and well into the summer of 1944.

ORGANIZATION OF CLINTON LABORATORIES

The organization set up by the University of Chicago provided for research and development activities to be carried out in the fields of medicine, health physics, biology, chemistry, engineering, and physics. The organization actually set up for carrying out these responsibilities provided a medical division with health physics and biology groups under this division, a chemistry division, a separations development division, an analytical division, an engineering development division, and a physics division. The work carried out by these divisions may be outlined as follows:

Medical Division: The medical staff had the responsibility for pre-employment, interim, and termination examinations, and for general plant medical work associated with the special hazards existing in the plant. A large fraction of this latter work involved careful clinical studies that were made in an attempt to establish the proper relationship between detectable clinical conditions of employees and the physical conditions under which they work.

Health Physics Section: The health physics group, working in close cooperation with the medical group and under the supervision of the Medical Director, was concerned with the general problem of controlling radiation and contamination hazards. Their major activities included radiation surveys of all working space, clothing, water, and of hands (especially); radiation monitoring, including maintenance, calibration and reading of pocket ionization chambers and film meters; and research on methods of decontaminating

individuals, clothing and laboratories. They also developed and calibrated physical instruments used in their work.

Biology Section: The work of the Biology Section was also closely related to that of the Medical Section. The work consisted of a study of the metabolism of short-lived fission products and the preparation of material to be used in metabolism studies on longer-lived fission products. This section maintained biological monitors in the hazardous areas of the Laboratory and also studied the biological effects of fast and slow neutrons and gamma and beta rays on various species of animals with the idea of extrapolating this information to man as effectively as possible.

Chemistry Division: The chemistry research and process development work at Clinton Laboratories was performed in three divisions: The Chemistry Division, the Separations Development Division and the Analytical Division. The Separations Development Division was expected to complete most of its work on the pilot plant demonstration of the Hanford process by October 1, 1944, and then to be transferred almost in a body to Hanford. After the transfer of men to Hanford, it was planned that operation of the plant would be put on a research basis. It was recognized that the facilities available at Clinton Laboratories would be of primary importance in laying out a program of research in the new fields of nuclear science. It was expected that the research program could be broadened considerably by having the available facilities used in part for the preparation of selected tracer elements for use in chemical, biological, and medical work elsewhere.

Engineering Development Section: The Engineering Development Section (later the Technical Division) was primarily concerned with Hanford problems and improvement of Clinton operations. It was planned that the technical personnel of this division would also contribute to the design of future reactors, particularly light water reactors and possibly others using heavy water or enriched uranium. A reactor designed for production of usable power was recognized in the very beginning as being one that should certainly be given serious consideration as soon as personnel could be diverted from the more urgent production research activities.

Physics Division: The Physics Division at Clinton Laboratories was concerned with research in pure physics and reactor physics. Initially, the division was expected to devote its entire effort to the start-up and operation of the reactor. Later, it was planned that the reactor physics work would probably emphasize the development of light water and enriched fuel reactors to the point where the selection of the most interesting possibilities could be made. Detailed design studies could be carried out by the physicists and engineers to the point where construction of designs could be undertaken.

ACCOMPLISHMENT OF THE ORIGINAL OBJECTIVES

Since the University of Chicago did not have on its staff personnel trained in plant operation, and due to the great importance of getting the work under way as quickly as possible, a sub-contract was negotiated with the duPont Company whereby the latter agreed to make available to the University the services of as many of its experienced employees as could be reasonably spared from other war plants until such time as new personnel could be recruited and trained. The duPont Company provided a total of 337 employees, all of whom were experienced in some phase of plant operations. As originally planned, many of these employees, after gaining additional training in particular phases of this type of plant operation, were returned to the duPont Company to serve as a nucleus for the operating personnel at the Hanford Engineer Works.

Reactor Operations

One of the main objectives covered in the scope of the work at Clinton Laboratories was to produce, separate and purify a few grams of plutonium. It was extremely important that this relatively small amount of material be made available as soon as possible in order that fundamental experiments, having to do with its physical, chemical, metallurgical, and nuclear properties could be performed. With this objective in view, the reactor structure, having a theoretical capacity of about 1,000 KW, was designed and built with the expectation that after it was put into operation and tested, improvements could be made to permit higher operating levels.

Because the reactor designed for construction at Clinton Laboratories was so much larger and more complex than the small, experimental model

operated at the University of Chicago, there were many features of the larger reactor which had never been demonstrated. While theoretical and experimental studies had been performed to provide as much information as possible for design of the reactor, it was not absolutely certain that the final design would be such that the desired performance characteristics could be achieved. Therefore, a horizontal section, two feet square, running exactly through the center of the reactor, was constructed in such a way as to be removable, so that if the reactor, as originally constructed, did not perform as well as desired, it would be possible to remove this section and replace it with another of improved design. It was known that the center region of the reactor was the portion that was most critical in determining the reactor's performance, and therefore, it was believed that if necessary, this section could be altered in such a way as to make the reactor work. As it turned out, it was never necessary to remove this center section, and the original installation is in place today.

Another illustration of some of the problems and uncertainties that were encountered in the design of a reactor so much larger than any other ever tried was the system devised for the control of the chain reaction under all conceivable conditions. For control during normal start-up operation and shutdown of the reactor, electrically driven control rods were installed, running horizontally through the center region of the reactor. In order that the rods could still be driven in when an electrical power failure occurred, two large tanks filled with sand were installed in a hydraulic press arrangement so that the tanks were supported by hydraulic pressure supplied from electrically driven compressors. A power failure would, of course, stop the compressors and

permit the tanks to fall. In falling, they would generate hydraulic pressure that could be used to drive the control rods into the reactor, thus stopping the chain reaction.

There was some concern that the horizontal control rods might warp as a result of the reactor temperature and radiation to which they would be exposed. In the event that the rods warped sufficiently to jam so that they could not be inserted to stop the chain reaction, an alternate set of control rods were installed vertically in the top of the reactor. Under normal conditions, these rods would be held up, out of the reactor, by electromagnets. In the event of an electrical power failure or upon the decision of the operating personnel, the electromagnets could be released, dropping the rods into the reactor to stop the chain reaction. Here again it was recognized that warpage or misalignment might cause the rods to jam before they dropped completely into the reactor. Therefore, it was decided to install as a "last ditch" safety device a large slanted tube filled with boron shot, located near the top of the reactor core inside the shield. This tube was to be supported by a simple mechanical trigger which could be tripped manually to dump the shot out of the tube through the active lattice of the reactor, thus effectively poisoning it and stopping the chain reaction. It was expected that in the extremely remote possibility that all other control devices failed simultaneously, the boron shot could be dumped from the tube under any circumstance and could be counted on to stop the chain reaction.

It was planned that the reactor would be operated for plutonium production and also for the purpose of carrying out, simultaneously, certain fundamental physical measurements to provide nuclear data that was urgently needed for the development and progress of other phases of

the Manhattan Project. The design and construction of the reactor incorporated certain facilities, such as special experimental holes for inserting materials into the reactor, beam holes for bringing radiation beams out of the reactor, and special radiation measuring instruments, which provided sufficient flexibility to permit the carrying out of various types of investigations.

Basically, the reactor, as originally designed and constructed, consists of a twenty-foot cube of graphite, pierced with horizontal holes arranged eight-inch centers to contain the aluminum-jacketed uranium fuel pieces. The fuel "slugs" were cylindrical, approximately one inch in diameter and four inches long. The fuel channels were square, slightly larger than the slugs, and arranged with the corner downward so that the slug rested in a groove with open space on the sides so that cooling air could pass around it. The twenty-four foot cube of graphite that made up the reactor core was surrounded on all sides by high density concrete, seven feet thick, to provide shielding for radiation originating in the reactor.

A canal was constructed adjacent to the reactor at a level below its base so that the fuel slugs could be pushed manually from the channels in the graphite into an open space between the graphite cube and shield. They would fall through the open space into a funnel arrangement that guided them into a chute and carried the slugs through the reactor shield to a point approximately twenty feet below the surface of the water in the canal. After the highly radioactive slugs reached a point outside the reactor shield, they would be shielded by water in the canal. The canal was arranged so that fuel from the reactor could be transported in buckets under water from the reactor to the first shielded cell of the

adjacent chemical processing pilot plant building. There a mechanical device was provided to pick up the bucket by remote control and dump the fuel slugs from it into the chemical dissolver tank so that chemical processing operations could be started.

The reactor structure was loaded with approximately 35 tons of uranium metal and placed in operation on November 4, 1963, this being only a few weeks later than had been anticipated when the construction work was first begun. After a brief period of operation at low power for checking the performance characteristics of the reactor, full power operation was started in order to produce sufficient quantities of plutonium as rapidly as possible. Beginning in December, 1943, irradiated fuel was discharged from the reactor for use in the chemical process, development and testing operations in the pilot plant. The first significant quantities of plutonium were separated in purified form in the months that followed.

Even though the operating level of the reactor was as great as had been anticipated, the results of engineering studies made during the early part of 1944 clearly indicated that the rate of production could be greatly increased by making certain changes, such as the use of uranium slugs with argon arc-welded jackets. A change in the distribution of the uranium in the reactor resulted in a more favorable operating condition in that it gave a better temperature distribution. It was also evident that the operating level could be greatly increased by passing through the reactor a greater quantity of air. Two large fans, having much greater capacities than the first ones, were obtained for this purpose. They were installed and put into operation

in July, 1944. The combined effect of these changes made it possible to operate at an average power level of slightly greater than 4,000 KW.

At no time were difficulties encountered in connection with the operation of the reactor proper. However, interruptions were introduced as a result of certain mechanical failures in the cooling air system, particularly with the fan bearings. In spite of these interruptions, the production schedule was maintained and at no time was behind that originally estimated.

Until about December 1, 1944, the reactor was kept in operation for the purpose of producing plutonium for experimental purposes. By that time, sufficient material, about 20% more than originally estimated, had been produced to meet the initial requirements for experimental purposes. Since that time, the reactor has been operated for the purpose of producing other radioactive materials which were of great importance in carrying on the research programs here and at other locations.

Chemical Process Development and Pilot Plant Demonstration

Before the large production plant at the Hanford Engineer Works could accomplish its mission, it was necessary that a workable and reliable chemical separations process be developed and tested under plant conditions. It was known that after the reactor fuel (pure uranium) was irradiated, it would contain plutonium, and in addition, over one hundred other radioactive fission products which would have to be removed by a chemical separation in order to obtain the final product in the desired form. Processes based on volatility, absorption, solvent extraction and precipitation were investigated for the separation and purification of plutonium. In pioneering research with radioactive materials, scientists often used co-precipitation processes in which a small quantity of a radioactive element is precipitated with a "carrier" of some other element. A process using bismuth phosphate as the "carrier" was developed for plutonium separations. Precipitation processing had the advantages of being well-known, of being easily broken down into repeated steps for processing in "batches", and of requiring only comparatively simple equipment.

By June, 1943, plutonium separation processes had been sufficiently investigated by the Metallurgical Laboratory to warrant a decision to use the bismuth phosphate process in the large scale production work to be undertaken at the Hanford Engineer Works. Additionally, corrosion tests had demonstrated that the bismuth phosphate process would be much less severe than some other processes investigated in regard to corrosion of equipment which had to be operated and maintained in good repair by remote control methods. Other experimental work indicated that satisfactory separation and high yield could be attained.

Even while construction of the Clinton Laboratories pilot plant was in progress, equipment changes in the pilot plant were frequently made. Since the plant was intended for the purpose of demonstrating and improving the reliability of a process for the Hanford Engineer Works, all refinements which could be developed to improve the process were included, if possible, for testing in the pilot plant so that the best possible process would be used for production operations at Hanford.

Actual processing operations in the pilot plant started in December, 1943, using uranium irradiated at the Washington University (St. Louis) cyclotron, because it contained smaller amounts of the highly radioactive fission products and, therefore, would leave the equipment less seriously contaminated with radioactivity in case operational difficulties were encountered and repairs or modifications became necessary. From these first runs, a few milligrams of purified plutonium were prepared and shipped to the Metallurgical Project in Chicago for research uses. The first shipment was made on December 30, 1943.

Following this, uranium irradiated at low power in the Clinton Laboratories reactor was used, and shortly thereafter, processing of uranium irradiated at full power in the Clinton reactor was started and plutonium shipments became correspondingly larger. By the end of January 1944, the chemical pilot plant was processing one-third ton per day of irradiated uranium from the reactor. By the first of March 1944, several grams (still less than one ounce) of plutonium had been delivered.

During the succeeding year of production, a number of process and equipment modifications were made to improve yield and decontamination, but no basic change in the process was required. The plant served as a

proving ground for refinements indicated by the laboratory and pilot plant research which was continued as part of the essential work at Clinton Laboratories. Valuable information was gained in operation technique, and a training program was carried on for key personnel charged with the responsibility of starting and operating the Hanford Engineer Works Plant.

The final tests performed on full plant scale for the purpose of developing and testing the process for use at the Hanford Engineer Works were completed in August, 1944. Based on the results of these tests, recommendations for the detailed process to be used at Hanford were submitted in a formal report in September, 1944. Further tests designed to improve the over-all efficiency of plutonium recovery were completed in November, 1944. Recommendations based on these results were submitted to Hanford in a supplementary report.

At the conclusion of the operating period in January, 1945, experienced personnel carried out a thorough program to remove radioactive substances from all equipment before the separations plant was placed in standby condition.

Personnel Training

The unique nature of the various process for production, separation and isolation of plutonium, and the fact that so few people had received training in work with radioactive materials, made it necessary to provide adequate facilities for the training of personnel. In order to accomplish this mission, Clinton Laboratories organized and operated a training school for a group of du Pont employees who eventually were transferred to the Hanford Engineer Works to form the nucleus of the operating

personnel. In addition to the regular classroom work, covering the basic principles, all of the trainees were required to devote part of their time to the actual operation of the facilities in the pilot plant.

A total of 183 du Pont employees received training in the particular part of the work in which they would be engaged at the Hanford Engineer Works. The training program was completed, and the transfers effected well in advance of Hanford startup.

In addition to the trainees, there were also 83 du Pont employees who supervised the operations of certain phases of the work at Clinton Laboratories in order to gain experience before eventual transfer to the Hanford Engineer Works.

Physics Research and Development

The major functions of the Physics Division at Clinton Laboratories were three:

(a) Determine the operating characteristics of the Clinton Pile and devise means for improving its operations.

(b) Develop information essential to the safe and efficient operation of the Hanford water cooling arrangement.

(c) Supply certain technical information as requested by Los Alamos.

During the first few months after the startup of the pile, a number of physical measurements were carried out at low power levels to determine temperature stability, control rod behavior, and neutron distribution throughout the pile, as well as to calibrate the various power measuring instruments in order that routine operations could proceed without interruption.

A large section of the proposed shield for the Hanford Unit was inserted in a special opening in the top of the concrete shield for the purpose of determining the adequacy of the proposed Hanford shield for stopping the radiation that would be present at the higher operating levels. The measurements indicated the proposed shield to be adequate for reducing the intensity of all dangerous radiation well below the tolerance level.

After the completion of the test on the adequacy of the Hanford shield, a large block of uranium slugs was installed in the hole at the top of the pile and a series of measurements made at the urgent request of Los Alamos. The object of this work was to gain more information concerning the behavior of neutrons and the physical effects produced when traversing relatively large distances in uranium. The measurements

were carried out by a group of physicists over a period of several weeks and were completed to the satisfaction of Los Alamos about April 1, 1944. A report, incorporating the results of the tests, was compiled and transmitted to Los Alamos shortly after the experimental work was completed.

Among the by-products formed from uranium in the fission process were certain substances which absorbed neutrons and thus produced a poisoning or inhibiting effect upon the operation of the pile. It was realized that an accumulation of these might create a condition such as to make it impossible for the pile to operate. Little was known about these poisoning effects at the time the pilot plant was started. The two elements, samarium and gadolinium were known to be produced and to have a very high poisoning effect, but the extent to which they were formed as fission products in a reactor was not known, nor was the exact value of their poisoning effect. In view of this, Clinton Laboratories, in cooperation with a group of physicists at the University of Chicago, undertook a study of the problem by bombarding samples of the rare earth elements in the reactor and analyzing them in the mass spectrograph. From this work, it was concluded that these two elements would not give rise to any serious difficulties in connection with the Hanford operations.

Shortly after the Hanford startup an entirely unexpected isotope of xenon formed in the fission process was discovered to be thousands of times more detrimental as a poisoning agent than anything previously encountered. The effect was discovered when, soon after startup, the large Hanford reactor shut down of its own accord. The chain reaction simply stopped. At that time it was not known that xenon was the cause. This calamitous problem was given immediate and very intensive study

by the physicists and chemists at Hanford, Clinton Laboratories, and the Metallurgical Laboratory. Measuring the poisoning effect in the reactor over a period of time, they determined a short half-life for the poison and from tables of half-lives of the fission products guessed the identity as xenon. Physicists calculated that the Clinton Laboratories reactor had not encountered the xenon poisoning effect because it operated at a much lower power level which produced less xenon in the fuel and because the Clinton Laboratories reactor had "extra" fuel which could create enough "extra" neutrons to overcome the poisoning effect of xenon. The problem of the Hanford reactor was solved by adding more uranium fuel. The highly conservative reactor design developed by the du Pont Company had provided extra fuel channels all around the edges of the reactor so that adding more fuel created no problems. A method of operation was worked out which eliminated the xenon poisoning difficulty in a satisfactory manner.

A large number of miscellaneous investigations of direct interest to Hanford were carried out at Clinton Laboratories by the physicists during the first part of 1944 and prior to the time of the startup of the Hanford units. These included: a study of the radioactivity produced in a variety of commercial materials of construction by exposure to radiation in the Clinton Laboratories reactor; measurement of the heat output from irradiated uranium slugs after removal from the reactor; development of methods for detecting slug jacket failures (leakage of fission products) during operations; and development of neutron thermocouples for use in power measurements.

Chemistry Research and Development

The major part of the Chemistry Division's effort from its inception in the fall of 1943 to the fall of 1944 was devoted to the study of the bismuth phosphate separations process. The program was carried out in conjunction with the Separations Development Division which shared responsibility for its success. The goals were:

(a) The elaboration and improvement of the basic processing procedure developed by the Metallurgical Laboratory, by the study of the process variables.

(b) The establishment of the reproducibility of the most favorable flowsheets to permit process predictions concerning Hanford operations.

(c) The testing of alternate flowsheets and reagents for substitution in the event of difficulty at Hanford.

(d) The increase of security against partial failure, by the basic study of the process from the standpoint of the chemical mechanisms involved.

Considerable improvement in the efficiency of the process was achieved. During the course of the program, the decontamination factor (by which the amount of the undesirable radioactive fission products accompanying the plutonium was decreased) rose many thousand-fold, and at the same time, production yields were increased from less than 80% to better than 90%.

Since plutonium is an element which prior to Clinton startup existed in milligram amounts only, its chemical characteristics were little known. Important research on the chemistry of plutonium was started as soon as production operations yielded even minute amounts of plutonium for research. The development of the separation process

depended upon the data obtained from the laboratory. Further, the general chemical properties demonstrated by this work suggested other and better methods for isolating plutonium. Finally, effective exploitation of the peculiar nuclear properties of plutonium demanded a full understanding of its chemistry. A number of immediate problems attacked included the study of the suitability of various plutonium compounds for shipment, storage, ease of dissolution, bulk, purity, and ease of preparation.

The preparation of radioactive elements was assigned the Chemistry Division originally for the purpose of supplying tracers for plutonium separation process studies. Soon afterward, radioisotopes production developed into a major service. The demand for special radioisotope preparations for research very rapidly exceeded the capabilities of ordinary laboratory equipment. The first new building to be constructed at Clinton Laboratories, in addition to those originally planned for the pilot plant, was a "Hot Laboratory" providing shielded cells in which highly radioactive materials could be processed. The Hot Laboratory building was completed and put into operation in March 1944. Almost immediately an addition was constructed to provide larger processing areas with thicker shielding so that still greater quantities of radioisotopes could be processed. The addition to the Hot Laboratory building was constructed especially to provide facilities for producing curie quantities of a fission product radioisotopes, barium 140, which was needed for research and development at Los Alamos. Methods and facilities were then available for filling requests for almost any of the long lived fission products in amounts approaching the curie level, free of carriers and other solids.

A number of radioactive species other than fission products were made available as a result of study of the reaction of neutrons with pure isotopes and of the chemistry required to separate the products so formed from the material bombarded. These included radio-phosphorous and radio-iron, of especial interest for biological tracer work, and tritium, the extra heavy isotope of ordinary hydrogen, in cubic centimeter amounts (the first preparation in appreciable quantities of a radioactive light element).

Fundamental research in the Chemistry Division began in September 1943, at a low level of activity because of the urgency of process development problems and its manpower demands. With the discharge of responsibilities in this field, basic chemical research increased until it occupied a significant fraction of the Division's total effort. The earliest fields for study were research on the radioactive fission products to identify them, determine their yields from phenomena associated with fission and establish the characteristics of their radioactivity. The effects of radiation upon chemical reactions and compounds constituted another area of research.

Study of the process of fission led to a more accurate idea of the proportion of the total energy of fission in the radioactive products and of the rate of release of this energy. The products of fission, of which about 150 were then known, were further characterized with respect to amounts produced, chemical properties, nature of radiation, and rate of decay. Information of this type, collected in conjunction with the Metallurgical Laboratory chemists, was used in improving the plutonium separation procedure, and in planning at Hanford for personnel protection and uranium waste disposal.

Attention was given the question of the interactions of neutrons with various chemical elements with a view of exploring the possibilities of using the reactor for the preparation of new isotopes. The production of tritium and radio-phosphorous by this method was started in 1944. Methods were developed for the purification of radio-antimony from fission products to provide a source of intense gamma radiation. Previous work in this field placed the chemist in a favorable position for identifying the xenon isotope produced in fission as the chief substance responsible for decreasing the reactivity of the Hanford reactor. The amounts of xenon produced by fission, its rate of growth and decay, and its capacity for absorbing pile neutrons were determined. The interpretation of these data in cooperation with the Physics Division was important in establishing an operating procedure at Hanford to minimize the effects of xenon poisoning of the reactor.

Methods were developed for the analytical determination of nearly all of the long lived fission activities in solutions of bombarded uranium or in the complex solutions resulting from the plutonium separation process. The utilization of these analytical methods for the determination of specific radioactive contaminants in separation process fractions contributed materially to the elimination of these undesirable elements.

Engineering Development

The Technical Division, staffed largely with chemical engineers, was formed for the purpose of providing technical engineering assistance to pilot plant operations and to gather information necessary for the successful design and operation of the Hanford production plant.

In collaboration with the Physics Division, investigations were made to determine the adequacy of the shielding material which was to be used in the construction of the Hanford reactors. For this purpose, provisions had been made in the construction of the Clinton reactor for a large opening in the top through which could be placed various arrangements of the shielding material. The Hanford shield was thoroughly tested and found to be quite adequate.

Various other materials to be used in and about the Hanford reactors were subjected to the conditions produced in the pilot plant. The necessary equipment was designed and installed in the reactor for studying the effects of intense radiation on the rate of corrosion on Hanford water tubes and on film formation within the tubes. In connection with this, determinations were made of the activity which could be expected in the cooling water under the Hanford operating level based on the results at Clinton. Further work included the determination of the extent of activity in the cooling water resulting from bare uranium slugs and from slugs with broken jackets, and the development of a method for detecting slugs which failed while in the reactor.

Consideration was given to the hazards which would arise in case of failure of the aluminum cans on the uranium slugs used for charging the Clinton reactor. The first slugs used in the pilot plant were jacketed with light, 15-to 20-mil, aluminum and sealed with a stitch weld. The maximum temperature set for this type of slug in the reactor structure was 150°C. Not only did many of these jackets fail when subjected to the heat test given them before insertion into the reactor, but the maximum allowable temperature was inadequate for higher operating levels. After considerable research was carried out, in cooperation

with the Metallurgical Laboratory, a new jacket was developed of heavier aluminum on which the argon-arc weld, a more positive seal, was used.

The testing of the entire quantity of argon arc welded slugs received was completed in February 1945. Of approximately 104,000 slugs tested, less than 4% were rejected because of jackets swollen or broken by oxide formation, excessive gain in weight, and pitted, scarred or concave jackets.

Since water was used as the coolant for the Hanford production reactors, the susceptibility of the aluminum cans to corrosion under operating conditions was investigated. Through a special experimental channel constructed to simulate a Hanford tube, water of composition similar to that in the Columbia River was passed through the tube and the corrosion rate of the slug jackets noted. Although the radiation did affect corrosion slightly, no unusual difficulty with slug failures was anticipated as a result.

The Technical Division, in close cooperation with the Physics Division, devoted a great deal of time and effort to the problem of increasing the operating power level of the Clinton reactor. This was quite important as one of the main objectives was to produce the maximum amount of plutonium in the Clinton reactor before the Hanford plant went into production. Improved aluminum cans for the fuel slugs permitted higher operating temperatures, which allowed operation at higher power. Other improvements included the installation of two large, 50,000-cfm fans for increasing the quantity of cooling air through the reactor, sealing off unused fuel channels to give better air flow through the tubes containing the uranium fuel, and rearranging the fuel loading to equalize the generation of heat. The combined

results of these various changes permitted reactor operation at a power level of approximately 4000 KW, four times the original design power level.

Provisions were made for storing the irradiated uranium after the plutonium had been recovered in the pilot plant. The uranium was stored in large, underground tanks in order that it might be recovered and made available for reuse at some later date. The Technical Division was charged with the responsibility of performing fundamental research and engineering development work required to develop a process whereby the uranium could be recovered.

Radiation Protection and Hazards Evaluation

It was early recognized that there would be serious radiation hazards associated with the processing of large quantities of radioactive materials, and also that the number of personnel experienced in handling radioactive materials was quite limited. Therefore, it was necessary to procure qualified personnel and conduct an intensive training program to prepare them to cope with the many problems which were anticipated. This area of responsibility was assigned to the Medical Division.

The first important function of the Medical Division was to lend assistance in the design of most of the facilities at the pilot plant. For each installation it was necessary to investigate the special hazards involved and to incorporate in the design sufficient protection for personnel in the plant area. It was usually necessary, at this time, to include a large factor of safety since certain radiation effects were not completely understood.

The reactor was surrounded by concrete walls seven feet thick, and the separation plant, most of which was underground, was controlled in its intricate chemical processes from behind five feet of concrete. The large quantities of highly radioactive waste material evolved were stored in underground tanks. Special research laboratories were constructed so that chemical research could be carried on by remote control, and the design and operation of this remote control research equipment marked a definite technical advance in coping with health hazards.

To provide ample protection to personnel, it was necessary to develop and construct special types of instruments for measuring the different forms of radiation. As soon as operations started, a service was initiated for the purpose of checking the radiation level at regular intervals throughout the plant at all locations which were frequently inhabited. Each individual who entered the hazardous area was required to carry on his person a small instrument of special design to record the amount of radiation to which he had been exposed while in the area. These instruments were read daily and the results evaluated by personnel trained in this particular field. All personnel subjected to the hazards were given a rigid physical examination at frequent intervals with special reference to the symptoms which one might expect from an over-exposure to radiation. Waste gases formed in the production processes and exhausted into the air by means of high stacks, were continually monitored to determine the quantities of hazardous gases which were released into the atmosphere. Likewise, the waste water from the plant was continually analyzed for hazardous elements. Automatic recording instruments were also placed in the various buildings

and at strategic points in the vicinity of the plant to assure that safe working conditions prevailed.

All these precautions required a great deal of research and development work. The special types of instruments required were, almost without exception, unavailable before the start of the project. The work required the formation of an instrument development section whose work included the design and development of instruments for use here and elsewhere on the project.

It was recognized when the pilot plant was first planned that very little information concerning the biological effects of radiation was available. This information was urgently needed before the design of the pilot plant was undertaken, however, a great deal of it was of such a nature that it could not be obtained until after the plant had been put in operation. It was necessary to work as fast as possible after the startup, and therefore it was decided to obtain most of the data by means of experiments on animals. The necessary facilities for accommodating some 10,000 animals were provided. Rather high radiation intensity exposures were used to obtain results as quickly as possible.

Radiation hazards studies were assigned to two groups to facilitate and speed research. The Biology Division at Clinton Laboratories concentrated its efforts on radiations produced externally which penetrates the body and produce a biological change. There were several different types of radiation to be considered in this class, each with its own characteristics and hazard. A group at the Metallurgical Laboratory was assigned to study radioactive elements which enter the body by respiration, ingestion, or through contamination of wounds and which

h. A satisfactory health program was instigated and adequate measures adopted and enforced to provide safe working conditions for the personnel.

PART II POSTWAR TRANSITIONS

REORIENTATION OF RESEARCH AND DEVELOPMENT ACTIVITIES AT CLINTON LABORATORIES

By June, 1945, all the initial objectives for which Clinton Laboratories was established had been successfully accomplished, and the University of Chicago, having fulfilled its responsibilities at Clinton Laboratories, withdrew from its operating responsibilities. The Monsanto Chemical Company was selected as the new operating contractor. The contract negotiated by the Manhattan Engineering District with the Monsanto Chemical Company for operation of the Clinton Laboratories became effective July 1, 1945.

Because of the unique facilities for pilot plant and research work at Clinton Laboratories and also because the unusual talents of the scientific research team already working at Clinton Laboratories could not be duplicated anywhere else at that time, the accomplishment of the original objectives did not mark the end of operations at Clinton Laboratories as originally expected. Instead, the research groups embarked on new projects of fundamental research and engineering development concerned, primarily, with reactor design.

The transition from war-time plutonium production, processing, development, and testing had begun in December, 1944, when both the reactor and the chemical pilot plants were changed over from plutonium production operations to experimental use. During the first half of 1945 under the University of Chicago, scientists at Clinton Laboratories gave serious consideration to determining what were the most important and valid research and development activities that should be pursued in order to make the greatest contributions to the new fields of nuclear

can remain for long periods continuing to emit harmful radiations. It was understood that the Biology Division would supply the Metallurgical Laboratory group with most of the special materials needed to carry out their research program. The preparation of this material was one of the most important functions of the Biology Division, and this work was expanded to include the preparation of special materials for other groups in Physics and Chemistry.

Radiations which caused some concern were those which were suspected of being present in minute quantities but could not be detected with the instruments available. To investigate this possibility, animals were subjected to the unknown radiation 24 hours a day, whereas the personnel were exposed for a maximum of 8 hours per day. Under constant observation, and periodic sacrifice and examination, it was not possible to detect any abnormalities in the animals. It was concluded that if any unknown radiations were present, they were certainly not in sufficient quantities to be harmful.

In addition, a program was undertaken in which animals were allowed to breathe gases which contained some of the radiation producing elements. Results showed that these gases are much less of a hazard than had been surmised.

While animal exposure methods were being developed and information accumulated it became apparent that the possibility of having personnel exposed to very large doses of radiation was remote. Experience taught the value of precautionary measures and showed that any operation could be carried out with adequate safety if the rules were observed. It was also determined that it is possible to devise methods for doing practically all hazardous work by remote control, when necessary, so

that in almost all cases it was possible to keep the exposure of personnel well below the danger level.

The results of animal experiments and the development of operating techniques and precautionary measures gave an insight into the way the various radiations cause damage, and thus allowed a more accurate evaluation of the hazards to be expected and the steps to be taken for the protection of personnel both at the pilot plant and the Hanford Engineer Works.

Brief Summary of Results Obtained Through June 1945

a. A pilot plant was designed, constructed, and successfully operated.

b. A sufficient quantity of plutonium was produced and concentrated for the purpose of investigating its fundamental chemical, physical, and metallurgical properties.

c. A dependable separations process for use in the pilot plant and at Hanford was developed and thoroughly tested.

d. Sufficient personnel were trained in various phases of plant operation to operate the pilot plant successfully and to form the operating nucleus at the Hanford Engineer Works.

e. The operating characteristics of the graphite reactor were determined and methods devised for improving its production of plutonium.

f. A method of recovering partially depleted uranium metal was investigated.

g. A process was developed and facilities were designed and constructed for the preparation of numerous radioactive sources.

science and technology. The need for many fundamental research activities could be clearly recognized on the basis of experience in coping with the problems encountered in developing production processes. Other needs, arising from the requirements of the weapons development groups and from the generally recognized desirability of obtaining basic information for reactor design, were considered important and were reflected in the plans for research and development activities to be undertaken at Clinton Laboratories.

Reactor Development

In particular, major effort was devoted to research and development work leading to the design of a high flux experimental reactor and to larger scale preparation of radioisotopes for special uses within the Manhattan Project, as well as to basic research in physics, chemistry, and biology.

Under Monsanto, these programs were materially increased in scope. The reactor development activities had been initiated late in 1944 by the chemists who proposed the construction of a 50-kw homogeneous reactor, utilizing an aqueous fuel solution containing 5 kgs. of uranium enriched to 12.5% uranium 235, or containing 500 grams of plutonium. They visualized this reactor as a research tool for the preparation of large quantities of radioactive tracers and radiation sources, for studies of chemical radiation effects at high power levels, and for the accumulation of data on the operating characteristics, chemical stability, and general feasibility of homogeneous reactors. It was pointed out that the aqueous fuel solution could be utilized very effectively for chemical processing studies and that the high neutron

flux of the reactor would be useful for irradiating thorium in connection with studies of the preparation and extraction of uranium 233.

The physicists were also interested in the homogeneous reactor, particularly as a research facility which would provide a high neutron flux for various experimental uses. The desirability of studying, or demonstrating, if possible, the process of breeding had been made especially attractive by the recent data indicating that uranium 233 emitted more neutrons for each one absorbed than either uranium 235 or plutonium 239; and the physicists were quick to point out the possibility of establishing a uranium 233-thorium breeding cycle which would create more uranium 233 from the thorium than was consumed in the reactor. These potentialities were very convincingly presented in November, 1944, in a report entitled, "The Case for an Enriched Pile", (CF-44-11-236).

The power output of such a breeder with a three-year doubling time is about 10,000 kw, and this was established as a new goal for the homogeneous reactor. The reactor, then, was conceived to be a prototype homogeneous reactor and thermal breeder; in addition, it was conceived as an all-purpose experimental tool with a neutron flux higher than any other reactor.

Work on the 10,000 kw homogeneous reactor was pursued vigorously through 1945, and at the end of that year, several major problems had not been solved. Perhaps the most serious of these problems was the formation of bubbles in the homogeneous solution. These bubbles appear as a result of the evolution of gas formed by fission fragments and other energetic particles. Because the bubbles cause fluctuations in the density of the fuel solution, they make it difficult to control the operating level of the reactor. Nuclear physics calculations made

at the time indicated that under certain conditions, it might be possible to set up a power oscillation which, instead of being damped, would get larger each cycle until the reactor was completely out of control. Minimizing the bubble problem by operating at elevated temperature and pressure was not considered seriously for two reasons. First, beryllium, aluminum, and lead were the only possible tank materials with sufficiently low neutron absorption characteristics to be useful in a breeder reactor. Of these metals, only lead was acceptable because of corrosion, and lead is not strong enough to sustain elevated temperatures and high pressures. Second, there had been essentially no previous experience in handling highly radioactive materials under pressure, and consequently, the idea of constructing a completely new type of reactor to operate under high pressure was not attractive.

Other major unsolved problems at the end of 1945 were those of corrosion, solution stability, and large external holdup of fissionable material. Because it appeared that the solution of these problems would require extensive research and development without assurance of success, it was decided to return to the earlier idea of a heterogeneous reactor proposed by physicists at the Metallurgical Laboratory. Therefore, effort was shifted to a high flux heterogeneous reactor for experimental purposes, one of which was to be obtaining the additional information needed to continue homogeneous reactor development.

In the latter part of 1945 and in 1946, at the same time work on the high flux experimental reactor was being vigorously pursued, a smaller research effort on the Daniels gas-cooled power reactor was undertaken. Also, the radioisotopes production program was expanding, and by July 1946, production capacity was sufficiently large to justify

making radioisotopes available to users outside the Manhattan Project installations. On August 2, 1946, the first radioisotope shipment was made under the new radioisotope distribution program. The shipment was one millicurie of carbon 14, which was sent to the Barnard Free Skin and Cancer Hospital in St. Louis, Missouri. At the same time, the reactor development and radioisotope production activities were expanding, fundamental research was being pursued on an increasing scale in nuclear physics, chemistry of the heavy elements, radiation chemistry, and biology; and applied metallurgical research was initiated on materials problems in reactor technology.

On January 1, 1947, the newly formed Atomic Energy Commission, created by the Atomic Energy Act of 1946, took over the responsibility for the atomic energy program, including the Clinton Laboratories. It is interesting to note that during the eighteen months from the time Monsanto Chemical Company assumed the responsibility for operation of the Clinton Laboratories under the Manhattan District until the Atomic Energy Commission was established, 90% of the research and development effort at Clinton Laboratories was centered in the Chemistry, Technical and Physics Divisions, with Chemistry making up nearly 45% of the total effort. The remaining 10% was divided among the power pile (5%), biology (3%), and training school (2%). On a program basis, 38% of the total research and development effort was expended on the high flux reactor, 27% on fundamental research, 15% on service functions for other Manhattan Project installations, 13% on radioisotope development and production, 5% on the power reactor, and 2% on the training school. The accomplishments under the major research projects initiated since the original objectives were accomplished are summarized below.

Heavy Element Chemistry

The spectrum, stability, and extractability of plutonium in perchlorate, nitrate, and chloride solutions were determined. Work was done on the identification of rare earths by fluorescence spectrum of aqueous solutions of their salts. A method of preparing a crystalline uranium trioxide hydrate was determined. The reactions of hexavalent uranium compounds in alkaline solutions were studied and reported. The basic chemistry of plutonium was studied and results giving the stability of the valence states as a function of pH, solution concentration, and other ions present were determined. Quantitative methods for the recovery of plutonium from laboratory wastes were determined and put in operation. Methods for the recovery of plutonium and uranium from aqueous reactor fuels were investigated. Studies were made of the complexes formed by plutonium under various solution conditions.

Separations Processes

The work on separations processes was divided between the Technical and Chemistry Divisions. Three main classes of separation have been studied:

1. Separation, decontamination, and recovery of uranium 235 from fission products.

2. Separation of uranium 233 from irradiated thorium.

3. Recovery of uranium and plutonium from large volumes of stored acid waste solution of Clinton pile slugs.

Class 3 has been a very minor part of the activity.

The theory of solvent extraction was extensively studied and extended. The equilibrium constants, decontamination factors, and other fundamental data were determined for several organic solvents. The theory of ion-exchange absorption processes, using resins, was also studied, and many constants were determined. The effect of radiation on organic solvents and resins was studied.

Solvent extraction columns were designed, constructed, and tested. Emphasis was placed on developing continuous processing methods, but batch processes have also been developed.

The separation processes were recognized as being of fundamental importance to the operation of proposed experimental and power piles because of the value of the large quantities of enriched fissionable material that must be processed. The primary objectives were to achieve high recovery and good decontamination. The development and design work on the separations processes for the experimental reactor was estimated to be 61% complete in January, 1947.

Analytical methods were developed for the heavy elements encountered in the chemical research and process development. Typical of these methods were those developed for the detection of (1) uranium and plutonium in urine, (2) plutonium in K_2CO_3 and Na_2CO_3 solutions, and (3) 100-milligram concentrations of thorium.

Physical Chemistry

Spectrophotometric determinations were made of the coloration due to electron and X-ray irradiation of various translucent crystals. The possibility of using atomic beams for the determination of magnetic properties of radioactive species was investigated. Apparatus for the

quantitative measurement of the molecular beams of alkali compounds was developed. A number of types of proton sources were built and tested. Colorimetric measurements of (n,γ) reactions were made as a method of determining the binding energies of neutrons to various nuclei. The effect of a number of variables on the steady state pressure of gaseous products over pure water exposed to radiation was studied.

Reactor Chemistry

This work was done primarily on the chemical problems of homogeneous reactor solutions. Investigations were made to determine what compounds of uranium and plutonium were suitable for aqueous fuel solutions. The mechanism of gas formation and recombination was studied, as well as other physico-chemical problems of the dissociation of compounds under irradiation. The results of this work indicated that many serious difficulties would be encountered in the operation of a high flux homogeneous reactor.

Reactor Physics

Reactor physics included the development of the nuclear data needed by the reactor design groups. Two major items claimed attention in this field: control studies and critical experiments. Critical size calculations were checked against actual measurements in critical assemblies, using both heavy and light water as the moderator. The effects of variations of many factors were measured. Reactor control calculations were made for various assumptions, and an electronic pile simulator was built for ready solution of control problems. Minor investigations of neutron temperature and fissionability of plutonium 239 were carried

out. A report describing the methods available for calculating critical sizes of slow and resonance chain reacting systems was published. A method of calculating multiple reflectors was developed and a report summarizing the existing knowledge of an "average fission" was published.

Theoretical and Experimental Nuclear Physics

A sensitive method of measuring neutron absorption cross-sections by causing oscillations of the reactor flux was developed, and a program of cross section measurements on elements and separated isotopes was started. The production of photoneutrons by gamma rays from Na^{24}, Ga^{72}, La^{140}, and others acting on D_2O was measured. The range of thermal neutrons in water from a Ra - - Be source was measured, and from this, the activity of the source was determined.

A crystal spectrometer, using single or double crystals, was developed, calibrated, and used for neutron diffraction and cross-section measurements. The characteristics of various nuclides produced in fission and by neutron absorption were investigated, and a search was conducted for hard gamma rays from short lived fission products, resulting in several being found and measured. Various isotopes were examined for the existence of short-lived isomeric states. An experimental program was undertaken to verify the radioactive decay of the neutron predicted by theory. The criticality of mixtures of fissionable materials in various vessels was investigated, and safe dimensions and concentrations were determined.

Compilations and interpretations were made of available data on cross-sections, fissions, characteristics, and practical phases of reactor design. Measurements were made of neutron absorption in a model layer of proposed thorium blanket for a reactor. Also, measurements

were made of the average energies, and the most probable energies of delayed neutrons from uranium 235. A search for three-particle fission was made. No definite evidence for this phenomenon was found.

High Flux Reactor

The engineering design and development of a high flux experimental reactor was a major effort of Clinton Laboratories. Some consideration was given to a homogeneous reactor, but this type was abandoned. A heterogeneous reactor, using heavy water moderator and light water cooling, was next studied and developed to the point of preliminary design. This design was modified to use light water as moderator and coolant and beryllium as a reflector. This reactor, in the preliminary design stage in 1946, appeared to be the final choice. Two comprehensive preliminary design reports were issued; the design and development work was estimated to be 51% complete by January, 1947.

Power Reactor

The Power Pile Division was formed in July, 1946, and was made up almost wholly of engineers on loan from industry (graduates of the Clinton Laboratories Training School). Its purpose was to design, construct, and operate a pile to produce useful power, using the ideas and the results of preliminary investigations made at Argonne National Laboratory. The education of the personnel of the Division in this new field took much of the time in the first months. The first preliminary design study was issued in 1946, and a second was started. No decisions as to the final form of the reactor were made, however.

Radioisotopes and Radiochemistry

The radiochemistry work involved the determination of the chemical consequences of isomeric transition, selective precipitation of

radioactive compounds, ion exchange in radioactive solutions, and daughter decay product ion forms in solution. Extensive work was done on the identification of various fission product activities and the determination of their decay chain. Element "43" was produced in weighable quantities for the first time, and was isolated and studied. The element "61", unknown in nature, was discovered at Clinton Laboratories among the radioactive fission products, was identified as the heretofore "missing" element, and was separated in weighable quantities. It was given the name of promethium, symbol Pm.

Physical Metallurgy

Metallurgical development work was undertaken in connection with the preparation of fuel assemblies for the experimental high flux pile. Uranium-aluminum alloy fuel assemblies were investigated. Some work was also done on the use of thorium and its alloys in pile control rods. Data on the results of this developmental work was included in the experimental high flux pile reports. Aluminum alloys were tested for water corrosion resistance under conditions simulating reactor operating conditions. A separate Metallurgy Division was formed in 1946.

Biology Research

The Biology Division was established in 1946 and carried out extensive experimentation with animals in a search for better methods of radiation injury detection, and used animals as radiation monitors in various parts of the area. This work contributed substantially to the meager information on the effects of β-ray, γ-ray, and neutron irradiation on living organisms. A separate Health Physics Division was established for personnel monitoring and the detection and control of radiation hazards.

CLINTON LABORATORIES RESEARCH AND DEVELOPMENT UNDER THE ATOMIC ENERGY COMMISSION

At the end of the transfer of atomic energy activities from the Manhattan District to the Atomic Energy Commission on January 1, 1947, serious plans were being made for permanent research facilities to be constructed at the Clinton Laboratories site in place of the temporary war-time buildings which had been intended to last only until the original objectives were accomplished. The name of Clinton Laboratories was changed to Clinton National Laboratory to reflect the new status of the organization under the Atomic Energy Commission. At the same time, consideration was being given to the long range research and development activities that should receive major emphasis at Clinton National Laboratory and to the size and composition of the scientific staff required to carry out these programs.

It was recommended by the laboratory staff that the high flux experimental reactor development program and the power reactor continue to be the Laboratory's major activity with strong emphasis being given to the radioisotopes program and to fundamental research, particularly in physics and chemistry. During 1947, the research and development activities did, in fact, continue more or less along the lines recommended.

Important developments in the program aimed toward the construction of a high flux experimental reactor occurred during 1946 and 1947 as the development and planning activities were intensified.

First High Flux Reactor Design

In 1946, the heterogeneous reactor first proposed was a heavy water moderated, heavy water cooled enriched reactor designed for operation at 20,000 KW. However, investigations of this proposal led to the conclusions that the corrosion of the stainless steel and aluminum parts of the system might introduce enough iron, chromium, nickel, and aluminum to cause deposition of these materials on the heat transfer surfaces, and that about 15 tons of heavy water would be required for the pile. The large heavy water requirement was a particularly serious drawback, since the national supply at that time was only slightly greater than 15 tons.

There was also some question as to the safety of the pile at the 20,000 KW power level, although it was unquestionably safe at lower power levels such as those planned for the present Argonne National Laboratory CP-5 research reactor, which was essentially a copy of this first proposed high flux pile.

The decision was made to change to cooling with ordinary water in order to avoid the first two drawbacks mentioned above.

Second High Flux Reactor Design

In May, 1946, a feasibility report (Mon N-108) was published to describe the heavy water moderated, light water cooled, enriched heterogeneous reactor to operate at a power level of 30,000 KW. The fuel was aluminum clad U-Al alloy made into thin, flat plates and rolled in a spiral inside a two-inch aluminum tube, through which a high velocity stream of ordinary water was circulated for cooling purposes. About 30 aluminum tubes, containing the fuel, were arranged vertically

in a triangular lattice immersed in the heavy water moderator. Around the lattice were other aluminum tubes, containing thorium for breeding.

This reactor, called the high flux experimental reactor, was intended to be built with its attendant chemical and metallurgical plants, at Clinton National Laboratory. The proposed construction was to start on July 1, 1946 and to reach completion of all facilities on October 1, 1947, with the reactor scheduled to start operation on July 1, 1947. The reactor was designed "to have the highest thermal neutron flux obtainable with present knowledge". It was intended primarily as a research tool and secondarily as a facility for the production of radioisotopes. A third objective was to serve as a sort of pilot plant for breeder and converter reactors.

Re-examination and evaluation of this proposed reactor indicated that the amount of heavy water required for the moderator, even with light water cooling, was rather large and resulted in a large reactor volume. Since the fast neutron flux is proportional to power per unit volume, it was decided that, in order to get a better fast neutron flux, the reactor volume should be reduced.

Third High Flux Reactor Design

In order to increase the fast neutron flux and at the same time maintain the highest possible thermal neutron flux, it was decided to eliminate the heavy water, replacing it with ordinary water; to fabricate the fuel elements in the form of flat, aluminum clad U-Al alloy plates and place the plates closer together; and to make the reflector of beryllium. Using ordinary water, which is a better moderator than heavy water, would decrease the volume of moderator required and would allow the fuel plates to be placed closer together, giving a smaller reactor

volume and a higher fast neutron flux. Beryllium was chosen as a reflector material because it again gives a smaller volume than heavy water, its corrosion resistance is good, it is not so susceptible as graphite to radiation damage, and, like heavy water, it supplies additional neutrons by virtue of the gamma-neutron reaction in which neutrons are emitted by beryllium under gamma irradiation.

Thus, in a rather natural series of changes, there evolved, by the summer of 1946, the essential design of the MTR — a design which was quite similar to the original Metallurgical Laboratory U^{233} converter of 1944. This reactor, still intended to be constructed at Clinton National Laboratory, was felt to be justified on the basis of the following objectives:

1. To produce a slow neutron flux equal to that required for large-scale breeding and for the production of mobile thermal power. This would allow testing of homogeneous reactor fluids at operating conditions.
2. To produce uranium 233 at a sufficient rate to permit evaluation of the advisability of basing a large segment of the atomic energy program on this isotope.
3. To provide Clinton National Laboratory and the nation with an experimental reactor "second to none". (The Chalk River reactor was nearly ready to operate at this time.)
4. To provide a fast neutron flux of magnitude comparable to that encountered in a small reactor in order to test radiation damage effects.

These were major motivations — the relative importance of the four shifted from time to time. In addition, there were certain secondary justifications:

5. To provide an alternate polonium source.

6. To provide an isotope production source.

These objectives were sufficient to justify the Manhattan District's assigning the Kellex Corporation to work on the reactor as design contractor; Monsanto Chemical Company, the contractor for operating Clinton National Laboratory, was to be the construction manager. Throughout 1947, design work continued at a substantial rate, although Kellex's effectiveness was somewhat impaired by the fact that it had many other project responsibilities in addition to reactor design. Nevertheless, it was believed in the fall of 1947 that design was sufficiently advanced that by the end of the year, the first actual construction drawings would be finished. In anticipation of the new reactor, a water reservoir, an electric substation, and a new steam plant were built at Clinton National Laboratory.

Long Range Program Plans

During 1947, the Atomic Energy Commission, as well as the Clinton National Laboratory staff, gave serious consideration to the permanent long-range programs that should be pursued at various installations, to the facilities already available, to new facilities that would have to be constructed, and to the most logical division of responsibility for research and development activities among the National Laboratories, so that duplication of effort would be avoided and each laboratory would pursue those lines of effort in which it was best qualified. In December, 1947, the Commission announced its plan to consolidate reactor development activities at the Argonne National Laboratory near Chicago and

to maintain the Clinton National Laboratory as a strong center for basic research, applied chemical research, and isotope production and research. As a part of the Commission's over-all plan, it was intended that the Clinton National Laboratory's efforts on the high flux experimental reactor and on the power reactor would be transferred to Argonne, and that most of the technical people who had been carrying out these programs at Clinton National Laboratory would also be transferred to the Argonne National Laboratory. It was expected that most of the people would be transferred from the Technical and Power Pile Divisions, together with others from the Chemistry, Physics, and Metallurgy Divisions.

It was recognized that long-range plans for the Clinton National Laboratory research programs in chemistry, physics, biology, health physics, metallurgy, and other fields had been developed on the basis of the assumption that the high flux experimental reactor would be constructed at the Clinton National Laboratory and would be available for experimental use in the comparatively near future. The change in plans for the high flux experimental reactor necessitated a complete revision of plans for future research and development activities at Clinton National Laboratory. The Atomic Energy Commission requested that new plans be made on the basis of major efforts devoted to basic research in the fields of physics, chemistry, biology, health physics, metallurgy, and others of direct interest to the Atomic Energy Commission program. The Commission recommended that these programs be planned on the basis of "continuing indefinitely and growing in strength at a healthy rate". A second aspect of the program recommended by the Commission for the Clinton National Laboratory was a vigorous effort in applied chemical engineering directed toward the solution of current problems that were vital to the atomic

energy operations. It was the Commission's intention that the Clinton National Laboratory, starting from its chemical engineering work already under way, should develop into a center of chemical technology for atomic energy activities.

A third aspect of the work at the Clinton National Laboratory was that of radioisotope production and development. It was planned that this work should continue along the lines already established and should be expanded as rapidly as possible by the development of new methods of production, the discovery of new and more useful isotopes, and the progress in finding new methods for their application.

In anticipation of this fundamental change in the program carried out at the Clinton National Laboratory, the Kellex Corporation was requested by the Atomic Energy Commission in November, 1947, to cease all design work on the high flux experimental reactor. When the Commission's decision to move the high flux experimental reactor to the Argonne National Laboratory was announced, those who had been working on the project at the Clinton National Laboratory pointed out that for safety reasons, they would not consider it advisable to locate a reactor of 30,000 KW close to Chicago.

The first meeting with the Reactor Safeguard Committee was in January, 1948. Prior to that, the Laboratory had made some safety investigations of its own, and had decided that the reactor was safe for the Oak Ridge site. The opinion of the Safeguard Committee was that, as a matter of principle, such high-powered reactors should be kept away from Chicago; but the Committee never stated its views on the suitability of Oak Ridge as a site. As an outgrowth of this view on safety, the

Atomic Energy Commission proceeded with its plans for establishing a new reactor test station at some remote location.

The Materials Testing Reactor

Work on the high flux reactor was continued by the Laboratory, although the responsibility for the construction and operation of the reactor no longer rested with the Laboratory. Actually, since the main features of design were already finished, the major achievements of 1948 were the experimental verification of design choices and construction of a full-scale mock-up of the reactor for further testing.

There ensued, during 1948, a series of conferences between the Laboratory and the Atomic Energy Commission, culminating in a proposal that the Commission reconsider the site for the high flux reactor. The Laboratory proposed a site about 20 miles from Oak Ridge, provided the Safeguard Committee considered Oak Ridge itself unsuitable as a site for the reactor. These conferences ended with a statement by the Commission that the high flux reactor was to be constructed at the Arco, Idaho reactor testing station.

The reactor to be constructed at Arco was given the name Materials Testing Reactor (MTR) and was essentially identical with the high flux experimental reactor designed at the Laboratory. An MTR Steering Committee was organized in November, 1948, composed of representatives of Argonne and Clinton National Laboratory, and was given the responsibility for general direction of the project. The Laboratory was given specific responsibility for completing the final reactor design, while Argonne was responsible for the design of the reactor building and associated facilities.

REORIENTATION OF THE RESEARCH AND DEVELOPMENT PROGRAM IN 1948

Since most of the research and development work at Clinton National Laboratory had been concerned directly with reactor development or strongly influenced by plans for the construction of the high flux experimental reactor, the Atomic Energy Commission's decision to move the reactor development to Argonne National Laboratory and to construct the high flux experimental reactor at another site required that the entire research and development program planned for Clinton National Laboratory be reconsidered. Another factor involved in the reorientation of the research and development program was the change of operating contractors on March 1, 1948, at which time Carbide and Carbon Chemicals Company assumed the operating responsibilities. At the same time, the name of the Laboratory was changed to the Oak Ridge National Laboratory. Carbide was already the operating contractor for the gaseous diffusion plant and the electromagnetic plant in Oak Ridge, and also had the advantage of its strong corporation background and interests in chemical engineering research and development, which were to be major activities of the Oak Ridge National Laboratory.

During most of the year of 1948, reorganization of the scientific groups, transferring of personnel to Argonne National Laboratory, and other activities associated with the changes that were being made at the Laboratory contributed to a rather large turnover in the scientific staff. Toward the end of the year, most of the major changes had been accomplished, and the new research and development program was not as much different from the one previously in progress as might have been expected. The research and development of the power reactor was moved

completely to Argonne National Laboratory, but the Oak Ridge National Laboratory continued its responsibility for the detailed design and testing of the high flux experimental reactor which came to be known under the Atomic Energy Commission as the Materials Testing Reactor. The full scale mock-up that was constructed at the Oak Ridge National Laboratory was used for tests of hydraulic and mechanical characteristics of the reactor core and controls systems. Particular attention was devoted to detailed study of the cooling water flow through the reactor core, and some modifications were made to improve the flow characteristics.

As an outgrowth of its own original interests and problems and in line with the Atomic Energy Commission's broad objectives, the Oak Ridge National Laboratory gradually developed a new program of research and development. The progress that has been made, the advancements and contributions coming out of the Laboratory's activities since 1948 will be covered in some detail.

OAK RIDGE NATIONAL LABORATORY

MAJOR FACILITIES COMPLETED OR TRANSFERRED BY CALENDAR YEAR

Year Completed	Bldg. No.	Title	Original Cost
1943			
	2011	Original Steam Plant	$ 186,000
	2013	Medical and Biological Building (now storage)	157,000
	2068	Administration Building (now ORSORT)	222,000
	2069	Cafeteria (now ORSORT)	104,000
	2500	Patrol and Fire Headquarters	51,000
	2506	Instrument Shops	135,000
	2516	Central Stores	122,000
	2517	Safety Department Offices	45,000
	3001	Pile Building (including Graphite Reactor)	3,485,000
	3019	Separations Building	681,000
	3022	Training School (now torn down)	192,000
	3026 C	By-Product Process Building and Chemistry Separations Laboratory	423,000
	3550	Chemistry Laboratory	804,000
1944			
	2005	Physics Laboratory (now torn down)	246,000
1945			
	3026 D	Dismantling Cell for Power Reactor Development Experiments	473,000
1946			
	1000	Administration Building (now P & E Division Offices)	370,000
	2008	Health Physics Low Level Analysis Laboratory	33,000
1947			
	3012	Rolling Mill	180,000
	3024	Research Shop	103,000
	9207, 9210 and surrounding Buildings	Biology Research Facilities Taken over from Y-12	6,759,000

Year Completed	Bldg. No.	Title	Original Cost
1948			
	0902	Reservoir	$ 304,000
	0907	Interim Low Level Facility	130,000
	2000	Metallurgy Laboratories	765,000
	2001	Health Physics Laboratories	397,000
	2519	New Steam Plant	912,000
	3503	High Radiation Level Chemical Laboratory	566,000
1950 (Transferred from Y-12)			
	9213	Criticality Laboratory	448,000
	9201-3	Reactor Design and Engineer Development	3,558,000
	9204-1	Reactor Experimental Engineering	3,857,000
	9704-1	Reactor Division Offices	93,000
1951			
	2007	Health Physics Test Building	79,000
	2010	New Cafeteria	284,000
	2518	Change House	137,000
	2521	Sewage Treatment Plant	109,000
	3010	Bulk Shielding Building	308,000
	3019	Addition to Separations Building	706,000
	3025	Solid States Laboratory	878,000
	3074	North Field Service Shop	42,000
	3500	Instrument Laboratory	435,000
	3504	Health Physics Waste Research Laboratory	204,000
	3505	Reactor Fuels Processing Plant	487,000
	3508	Chemical Technology Alpha Laboratory	313,000
	4500N	Central Research Building	4,966,000
	4501	High Level Radiochemical Laboratory	3,070,000
	7001	General Stores*	179,000
	7002	Garage and Utility Shop*	129,000
	7500	Homogeneous Reactor Experiment Building	320,000
	3029-3038	Radioisotope Area	2,843,000

00 Area Buildings built by J. A. Jones for construction headquarters and transferred to ORNL after completion of construction.

Year Completed	Bldg. No.	Title	Original Cost
1951 (Transferred from Y-12)			
	9201-2	Thermonuclear	$ 2,542,000
	9204-3	Electronuclear	3,479,000
	9711-4	Technical Library - Ecology Laboratory	278,000
	9731	Stable Isotope Separations	968,000
	9734	Spectroscopy Research Laboratory	137,000
	9735	Mass Spectrometer Laboratory	160,000
	9766	Ceramic Laboratory - Photographic Laboratories	237,000
1952			
	3005	LITR (including Reactor)	913,000
	3017	Reactor School Laboratory	146,000
	3592	Unit Operations Volatility Laboratory	34,000
	5500	High Voltage Laboratory	1,044,000
	7503	Reactor Experiments Building (ARE)	375,000
1953			
	7012	Central Machine Shop	344,000
1954			
	7702	Tower Shielding Facility (including TSR-1)	1,672,000
1955			
	2523	Decontamination Laundry	70,000
	3027	Source and Special Materials Vault	17,000
	3044	Special Materials Machine Shop	68,000
1956			
	2024	Metallurgy Laboratory Annex	95,000
	3025	Addition to Solid States Laboratory	622,000
	3019	High Radiation Level Analytical Facility	285,000
1957			
	2525	Research Shops	465,000
	3518	Process Waste Treatment Plant	287,000

Year Completed	Bldg. No.	Title	Original Cost
1958			
	3042	ORR (including Reactor)	$ 4,607,000
	3517	Fission Product Development Laboratory	1,916,000
	4507	High Radiation Level Chemical Development Laboratory	281,000
1959			
	2528	Low Level Waste Pilot Plant	84,000
	7018	Salvage Yard Facility	65,000
1960			
	3103 Cooling Tower	Increase in ORR power to 30 Mw	892,000
	3010	BSF II (reactor)	104,000
	7702	TSR II (reactor	577,000
	4500N	4500N - Wing 5 - Administration	1,577,000
	3500	Addition to Instrument Laboratory	989,000
	9207	Biology Additions	
		Mammalian Radiation Injury and Protection Facility	523,000
1961			
	2621	Tool Stores	67,000
	3104	Reactor Services Field Shop	94,000
	9204-3	Expansion of Stable Isotope Production Facilities	846,000
	9207	Biology Additions	
		Chemical Protection and Immunogenetics Laboratory	170,000
1962			
	4500S	Central Research Building Additions (including Compressor House and Cooling Towers)	7,495,000
	4508	Metals and Ceramics Building	6,500,000
	9201-2	Project Sherwood Relocation	612,000
		Biology Additions	
	9207	Biochemistry Laboratory	511,000
	9207	Low Level Radiation Experimental Facility	443,000
	9207	Pathology and Physiology Laboratory	690,000

Year Completed	Bldg. No.	Title	Original Cost
1963			
	3047	Radioisotope Development Laboratory	1,489,000
	3525	High Radiation Level Examination Laboratory	4,202,000
	Addition to Bldg. 5500	10 Mev Tandem Van de Graaff Accelerator (including accelerator)	2,400,000
	6000	Oak Ridge Relativistic Isochronous Cyclotron (including cyclotron)	3,718,000
	7709, 7710	Health Physics Research Reactor	1,409,000
		Biology Additions	
	9207	Cell Physiology Laboratory	500,000
	9210	Mammalian Genetics Laboratory	757,000

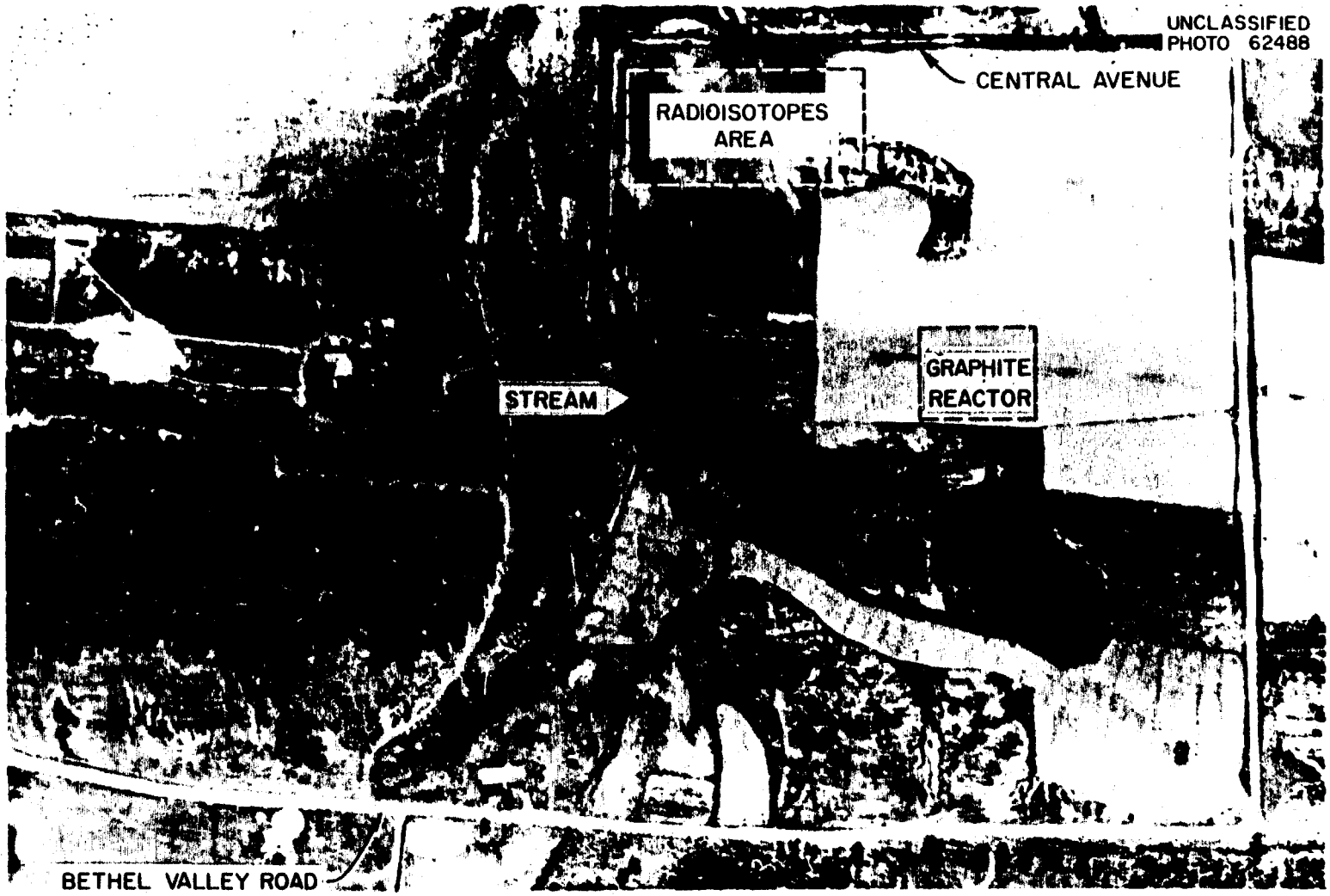

Fig. 1 X-10 SITE AS IT WAS IN 1942

The site selected for the Plutonium Pilot Plant was a remote farm area which is shown in this photograph prior to the start of construction. Bethel Valley Road in the lower part of the picture is located in approximately the same place today. The grove on top of the hill in front of Building 4500 can be seen at left in the photograph much as it appears twenty years later. The farm road in the upper right portion of the photograph became the central avenue of the Laboratory and the Graphite Reactor was located in the area appearing as a freshly plowed field upper right.

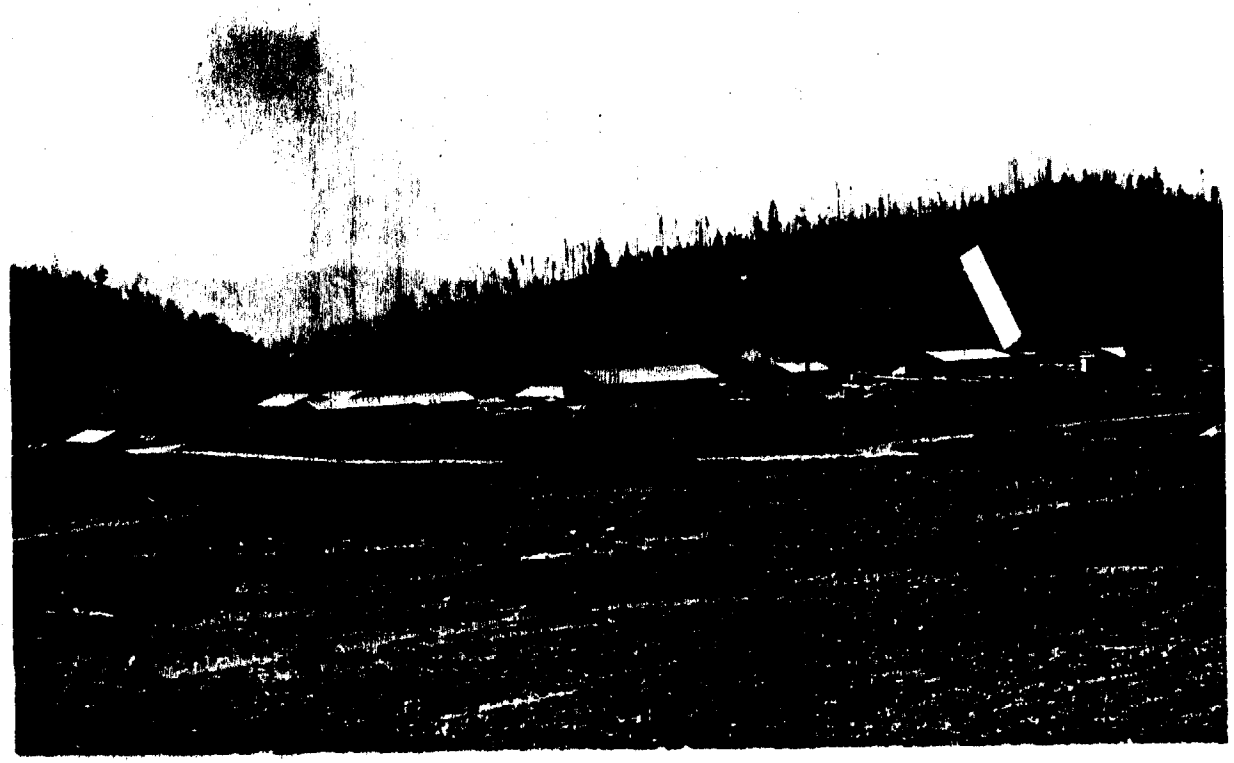

Fig. 2 START OF CONSTRUCTION – MARCH 1, 1943

Construction warehouses, shops and offices were erected quickly to provide materials and facilities for a rapidly expanding construction work force. Note that only a rough access road has been graded through the open field, which often became a sea of mud in the spring rains.

Fig. 3 SITE OF CLINTON LABORATORIES ON APRIL 15, 1943

This is ten weeks after the start of construction. The road through the center will almost divide the plant site in half, with production and research facilities on the right, administrative and service buildings on the left.

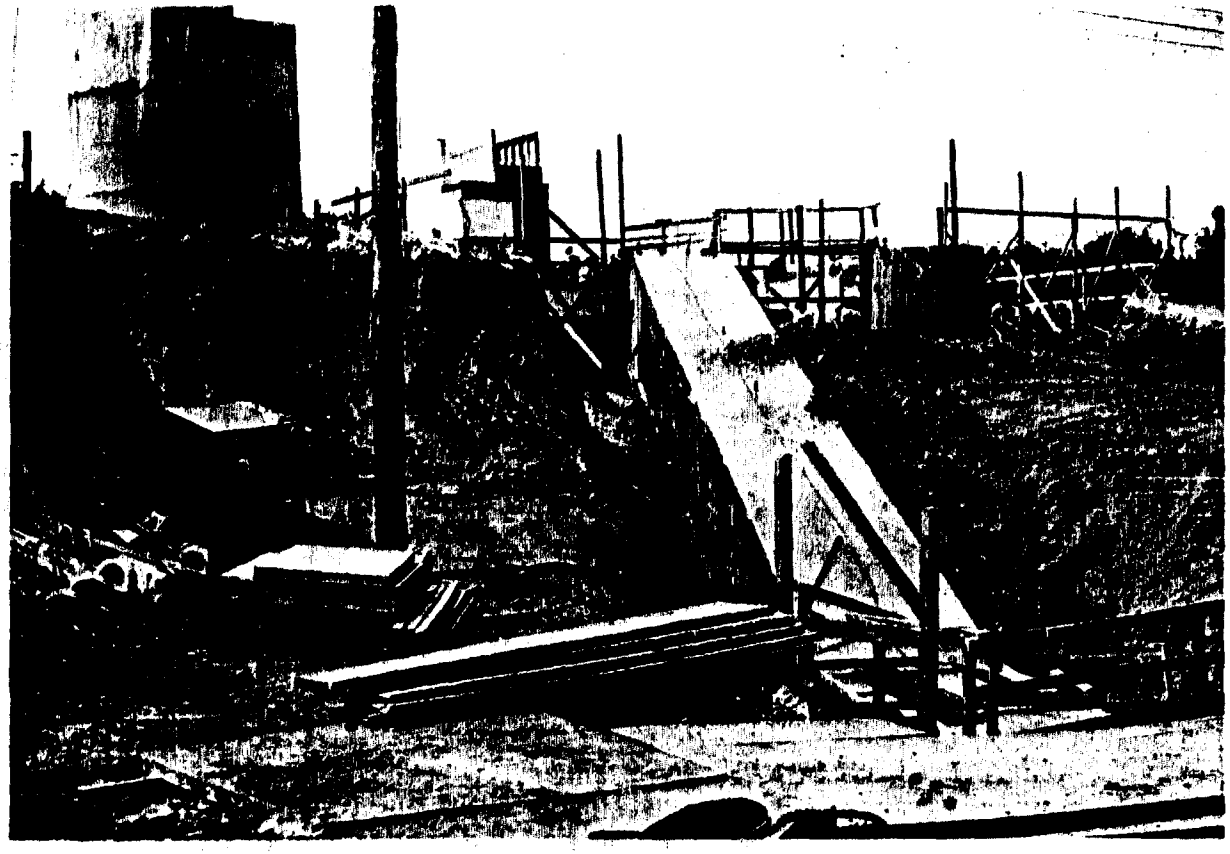

Fig. 4 EXCAVATION FOR THE GRAPHITE REACTOR

This photograph shows the duct for reactor cooling air to be exhausted through the fan house and out the stack which can be seen at upper left.

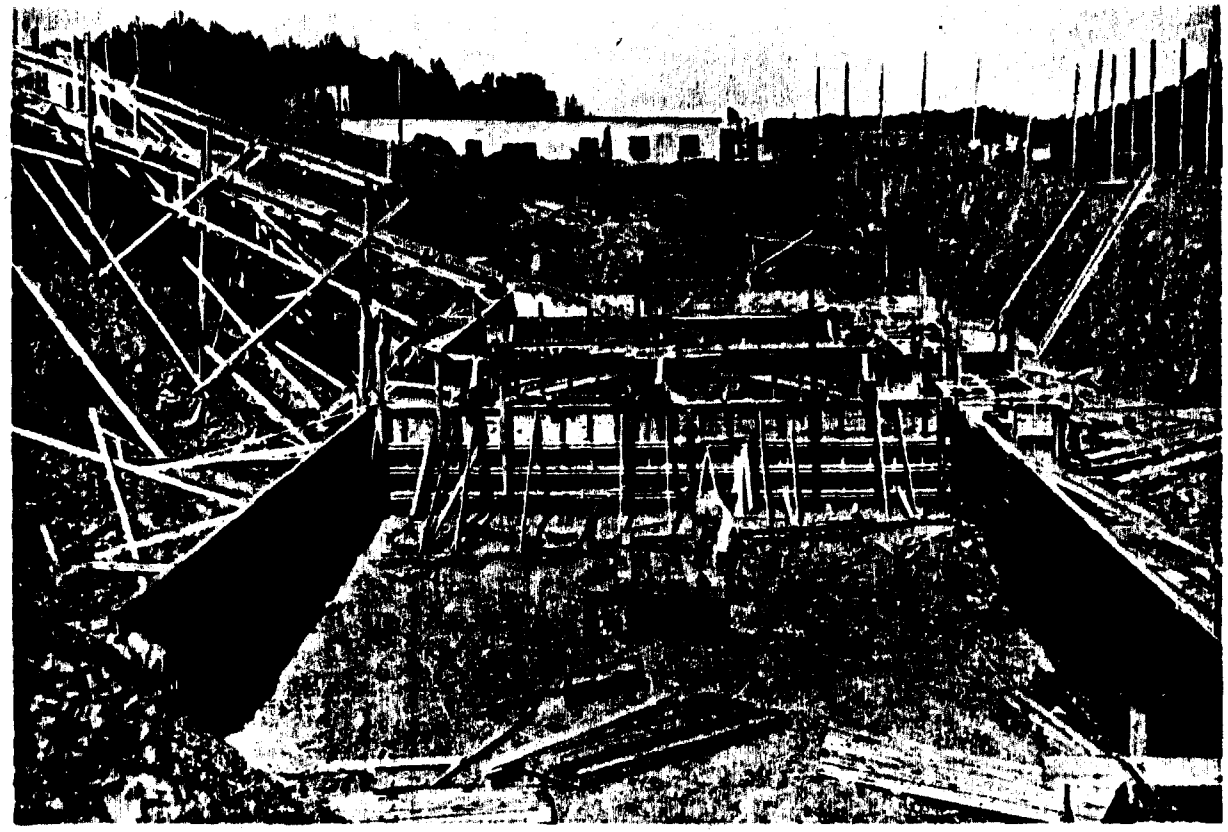

Fig. 5 FOUNDATIONS FOR THE CHEMICAL PILOT PLANT

The forms for heavy concrete foundations and shielding are being erected in May 1943. Note that the excavation extends to the Graphite Reactor site. An underground canal provided water shielding for highly radioactive fuel to be moved from the reactor to the dissolver cell where chemical processing began.

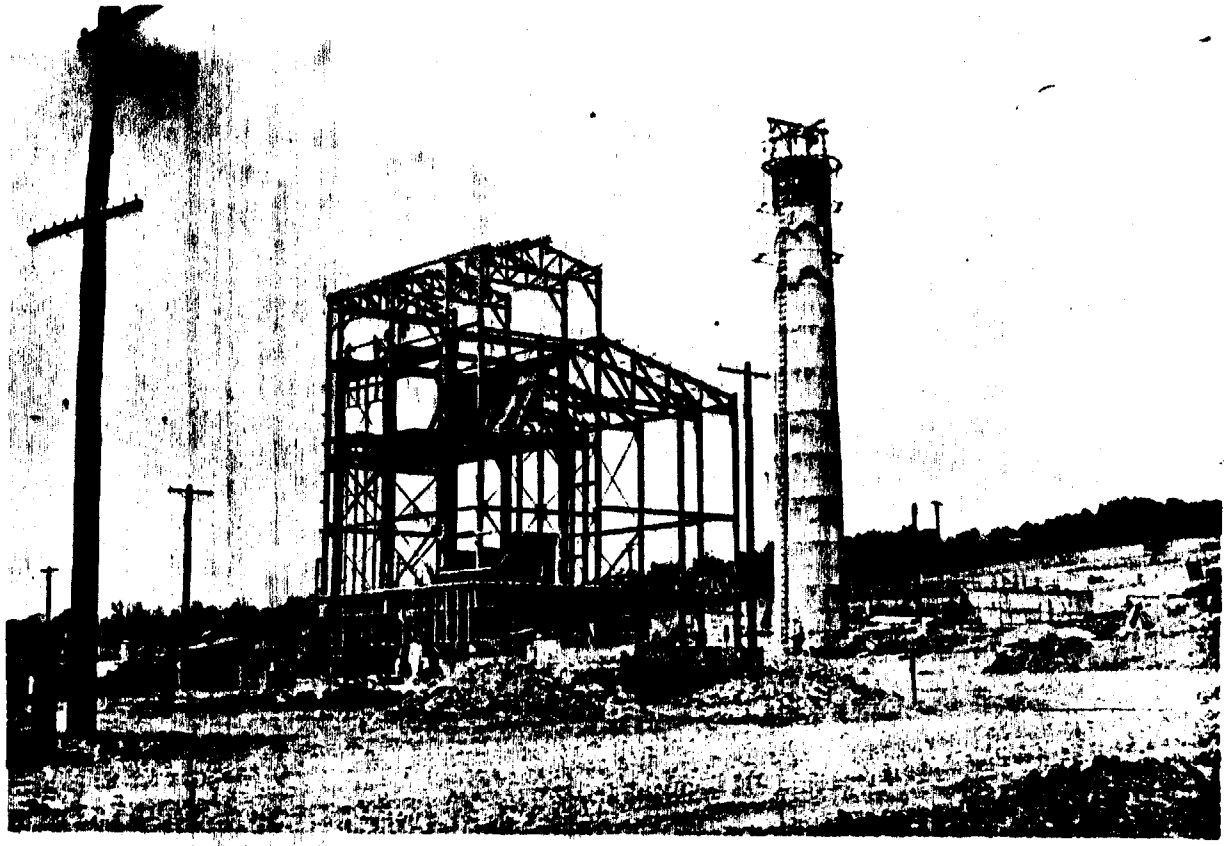

Fig. 6 THE STEAM PLANT

This building, the Graphite Reactor Building and the Chemical Pilot Plant were the only ones not of wood construction. The Clinton Laboratories pilot plant operation was originally expected to be completed and closed after Hanford production operations were progressing smoothly.

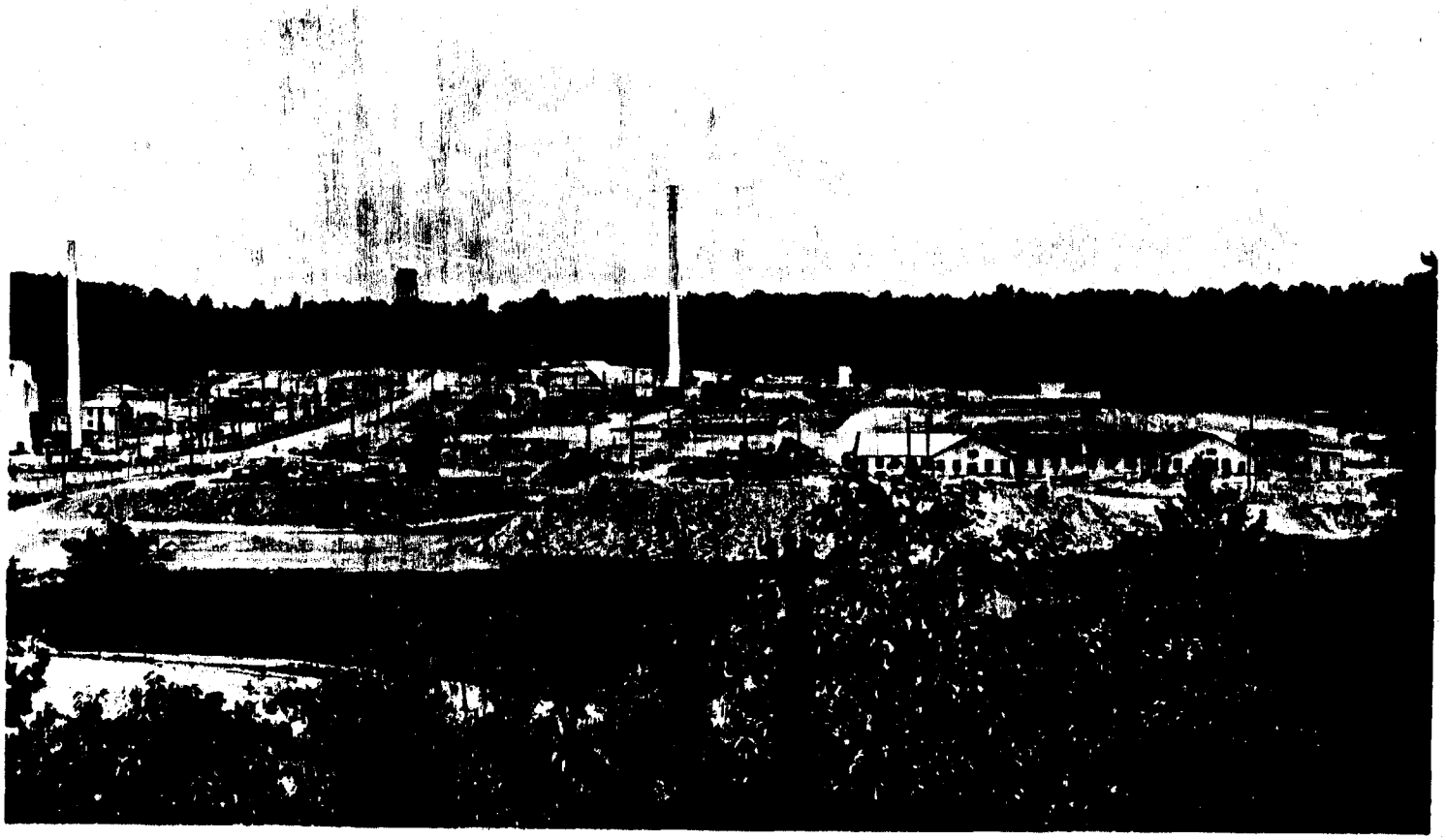

Fig. 7 STATUS OF CLINTON LABORATORIES CONSTRUCTION ON JUNE 27, 1943

The large building with the smoke stack is the steam plant. Note the tents used for storage and for construction shops until additional buildings could be completed. The Health Division Building and some of the shop buildings are complete and can be seen in the background to the left of the steam plant.

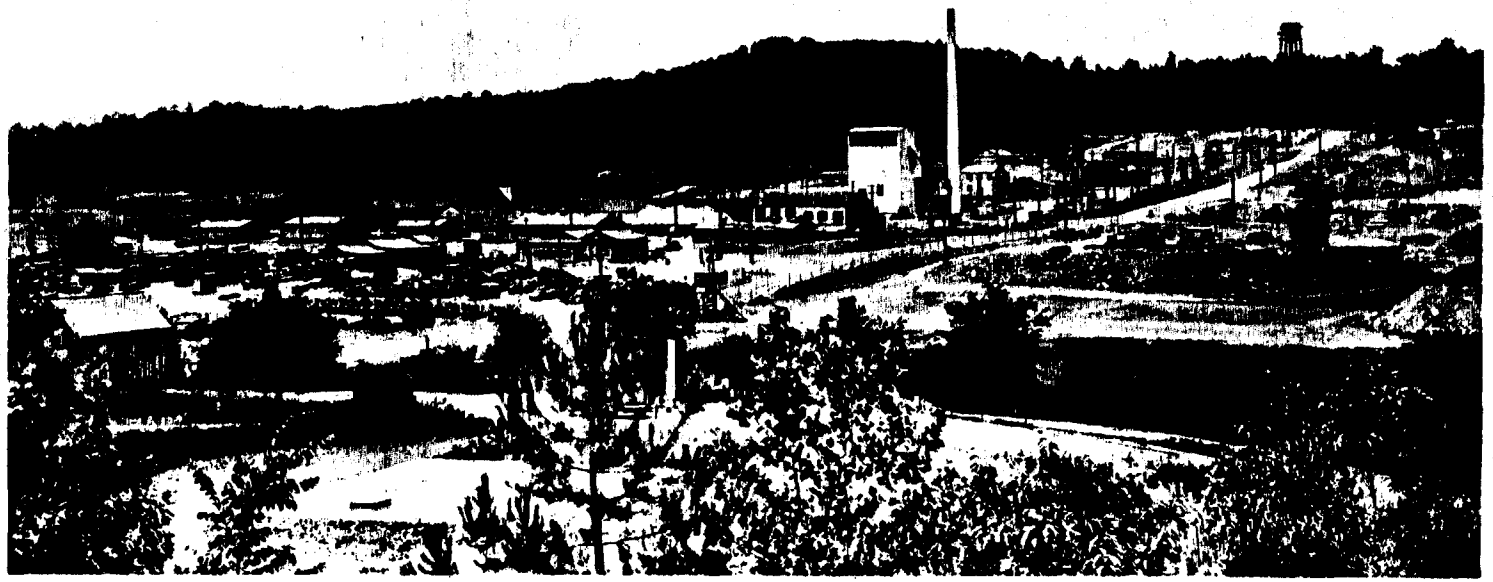

Fig. 8 STATUS OF CLINTON LABORATORIES CONSTRUCTION ON JUNE 27, 1943

The large building with the smoke stack is the steam plant. Note the tents used for storage and for construction shops until additional buildings could be completed. The Health Division Building and some of the shop buildings are complete and can be seen in the background to the left of the steam plant.

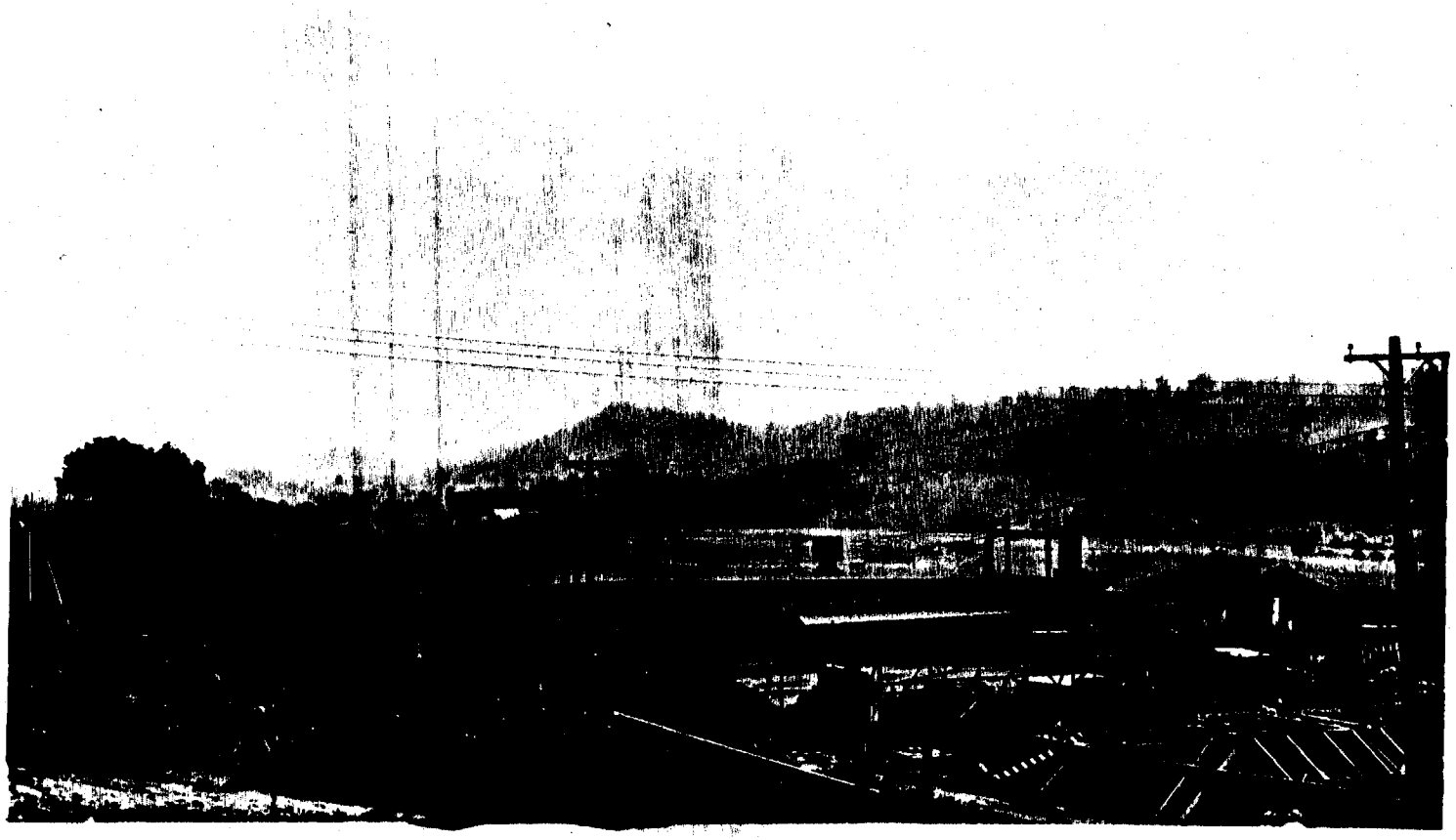

Fig. 9 THE MACHINE SHOP BUILDING

The building was complete at the time of this photograph, June 27, 1943. The 4-inch square graphite bars were machined here to make the 24-foot cube that served as moderator and reflector for the reactor. Uranium fuel slugs were loaded in horizontal holes through the graphite cube. The forms for pouring concrete foundations of the reactor building are in the right foreground.

ORNL 594-84

Fig. 10 UNDERGROUND TANKS FOR THE STORAGE OF RADIOACTIVE WASTES

The tanks were nearing completion in July, 1943. Six of these 150,000-gallon tanks were constructed in a tank farm just west of the Chemistry Building, and southward down the hill from the Chemical Pilot Plant. The forms for pouring the concrete shield for the reactor can be seen in the left background.

ORNL 593-84

Fig. 11 AERIAL VIEW OF THE RESEARCH AND PRODUCTION AREA, AUGUST 31, 1943

The Chemistry Building is in the center; to the left are the radioactive waste storage tanks; and at the upper left corner is the reactor building. At lower right can be seen two retention ponds for holding radioactive waste that might leak from the underground tanks.

ORNL-592-84

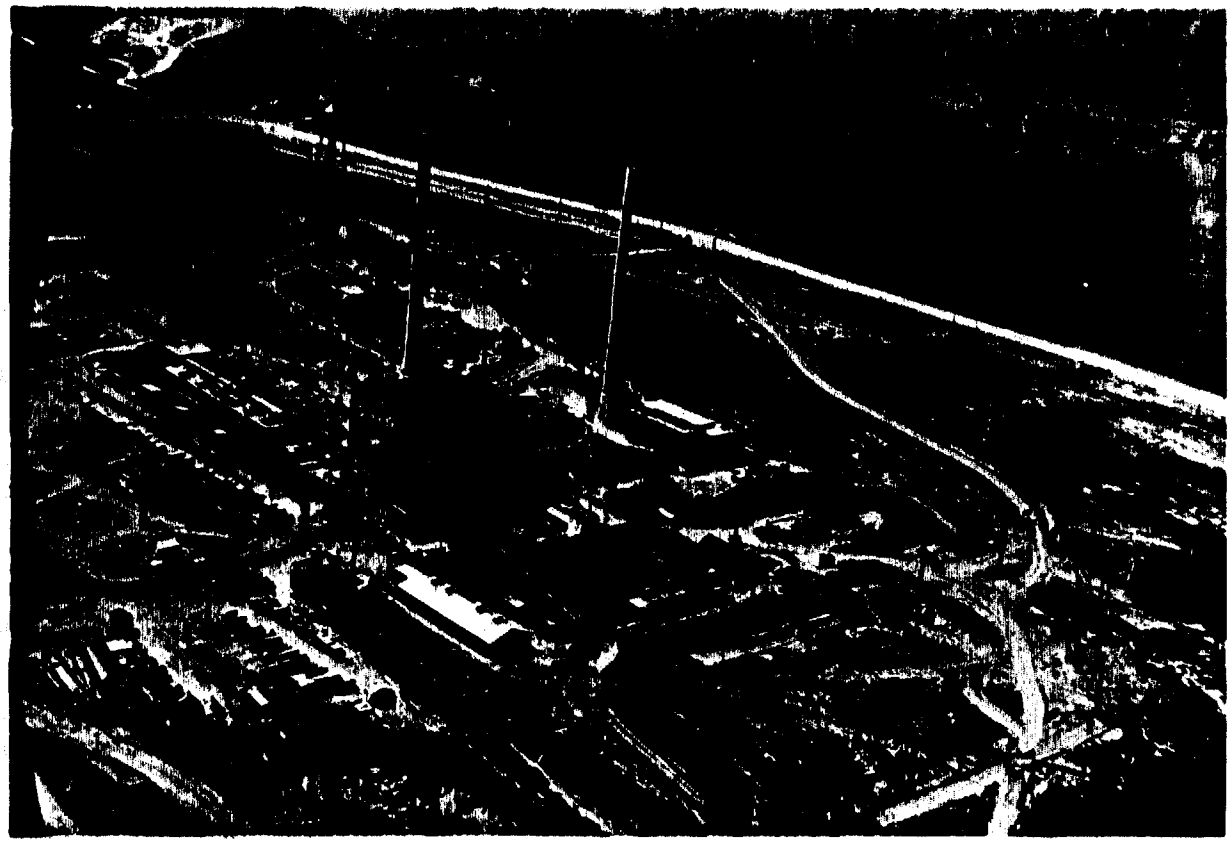

Fig. 12 VIEW OF REACTOR BUILDING AND CHEMICAL PILOT PLANT ON AUGUST 31, 1943

The concrete shielding for the radioactive materials processing cells of the pilot plant can be seen to the left of the reactor building. The openings in the top indicate the locations and sizes of individual cells. The fan house for drawing cooling air through the reactor is at the base of the stack on the right. The graphite machine shop is in the foreground. The Physics Building is to the left of the water tank.

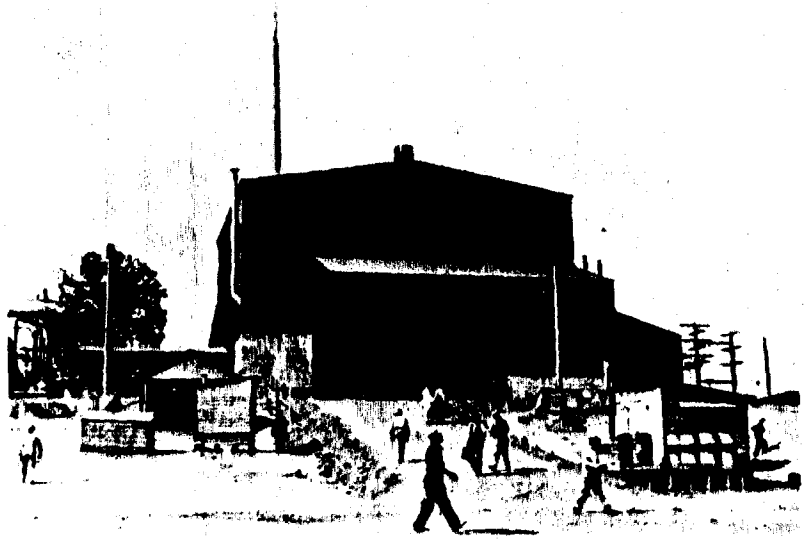

Fig. 13 THE REACTOR BUILDING ON OCTOBER 11, 1943

Construction work had proceeded on a 24-hours-a-day basis in order to get the reactor in operation as soon as possible. Operation started about three weeks after this photograph was made.

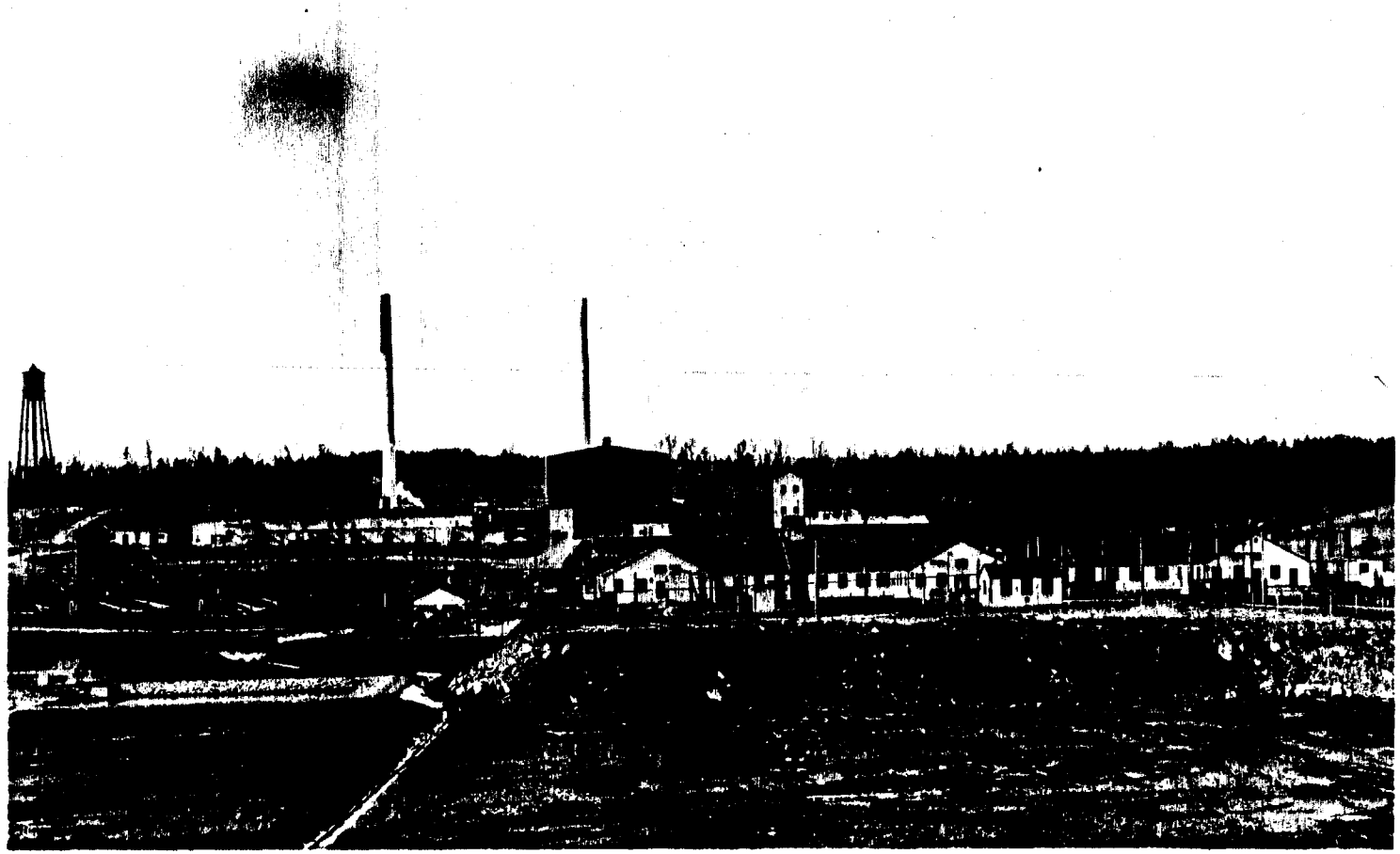

Fig. 14 CONSTRUCTION STATUS ON DECEMBER 20, 1943

The reactor and chemical pilot plant in the background have been operating for about six weeks. The Chemistry Building in the foreground has been occupied even longer.

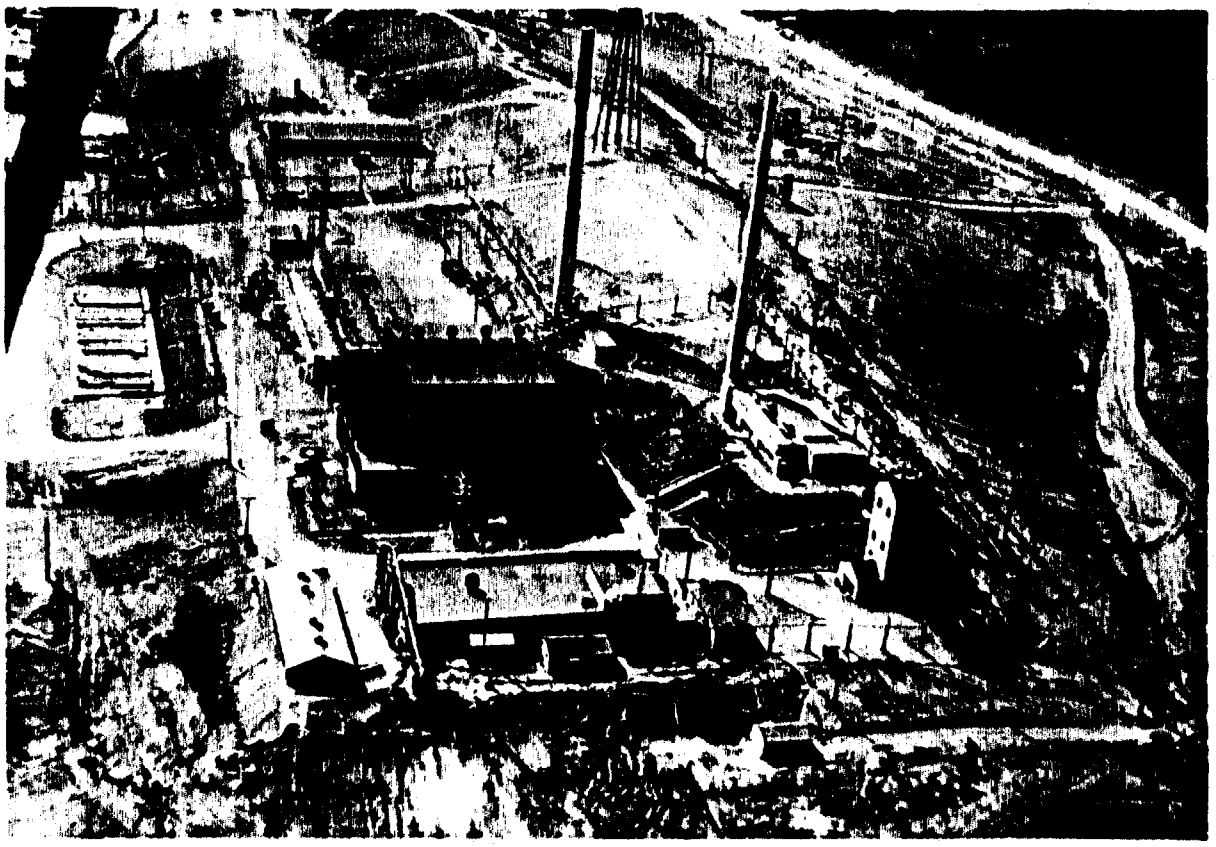

Fig. 15 - REACTOR BUILDING AND CHEMICAL PILOT PLANT ON MARCH 10, 1944

The machine shop in the foreground had, by this time, been converted to a "hot shop" for doing work on uranium fuel slugs and other hazardous materials. To the right of the shop is the tall water demineralizer building. The Physics Building is in the far background.

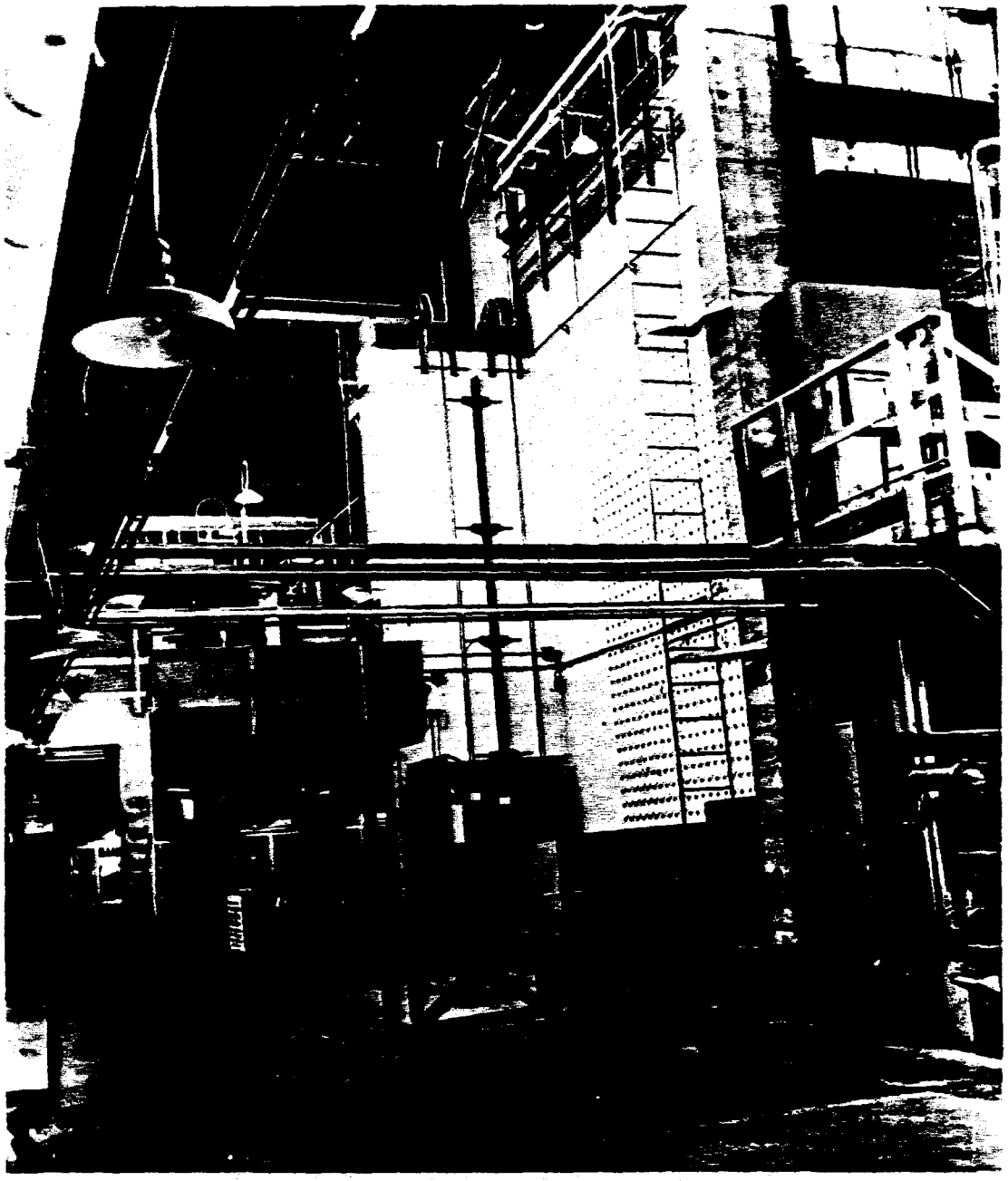

Fig. 16 FUEL-LOADING FACE OF THE GRAPHITE REACTOR

This photograph shows the reactor as it was at the time of startup in November, 1943. The elevator, shown near floor level, permits new fuel slugs to be inserted manually into the holes on the front of the reactor and pushed into the fuel channels in the graphite cube. The reactor control room is located on the balcony.

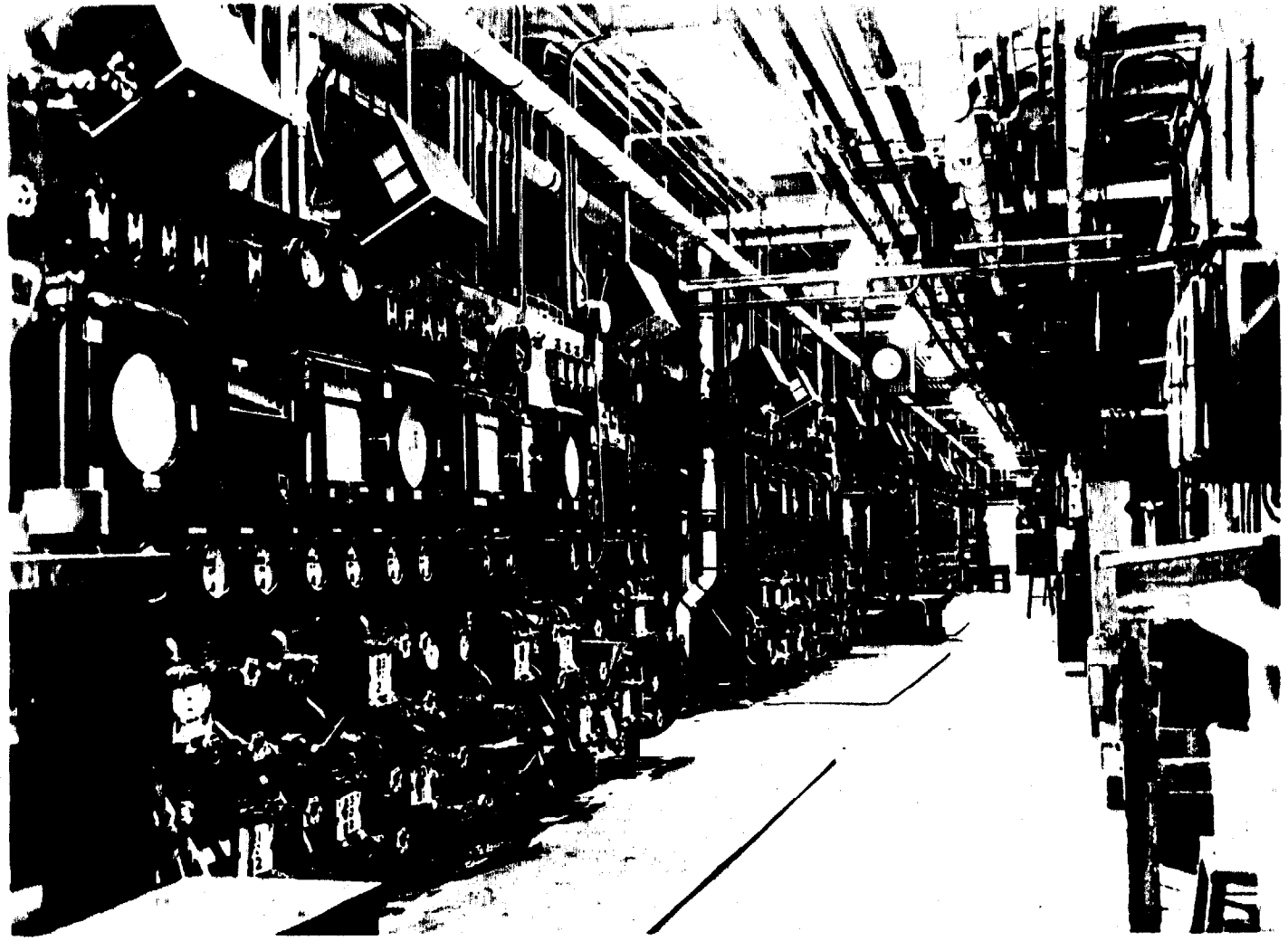

Fig. 17 OPERATING GALLERY OF THE PILOT PLANT

Here were located all the instruments and controls for remote operation of the chemical processing equipment which was contained in cells completely surrounded by 5-foot-thick concrete walls.

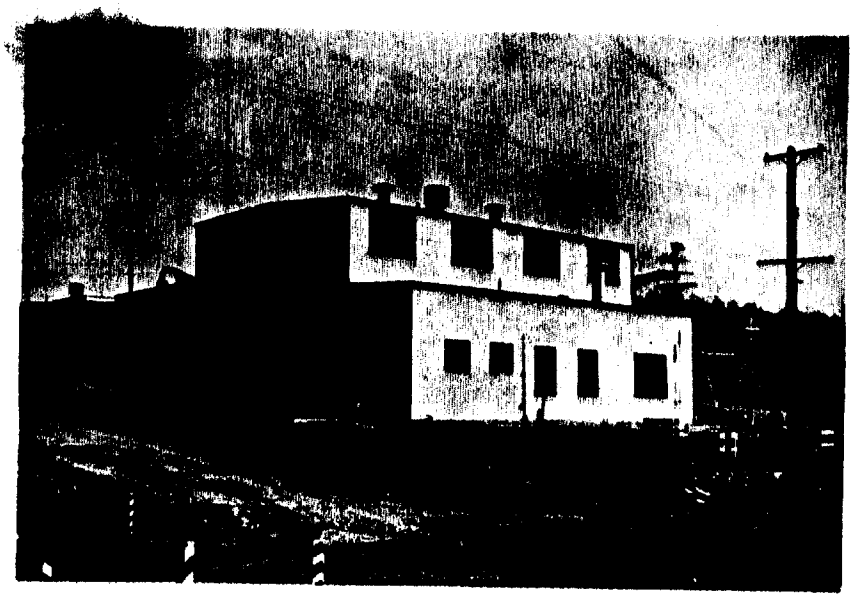

Fig. 18 THE HOT LABORATORY BUILDING

This special facility was constructed early in 1944 to provide shielded work areas which would permit the chemists to utilize large quantities of radioactive materials in their research. The building, located just across the road north of the Chemistry Building, had just been completed when this picture was taken on March 13, 1944.

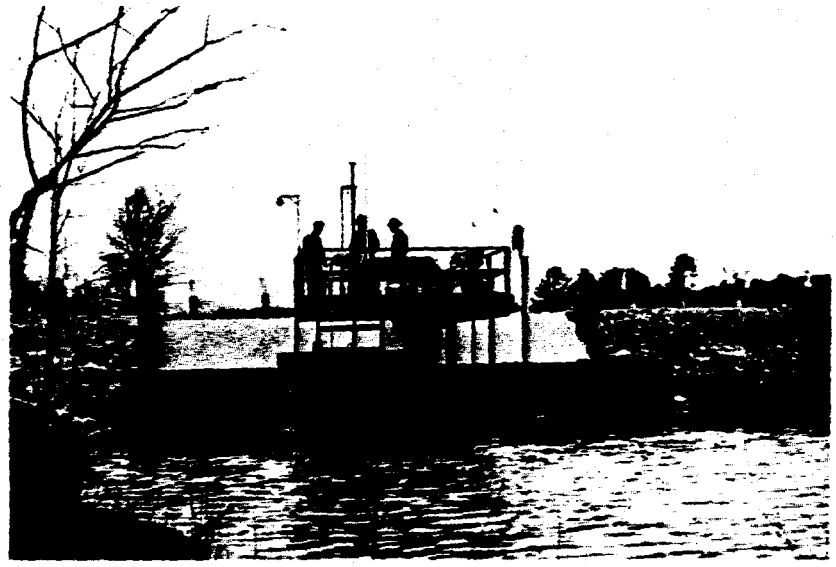

Fig. 19 WHITE OAK CREEK DAM

The White Oak Creek carries drainage from the Clinton Laboratories site. The dam was constructed in 1943 about a mile downstream to provide a means of holding up radioactive materials that might escape in the event of an accident at the plant. Water passing through the dam was routinely monitored to detect any radioactivity that might be escaping.

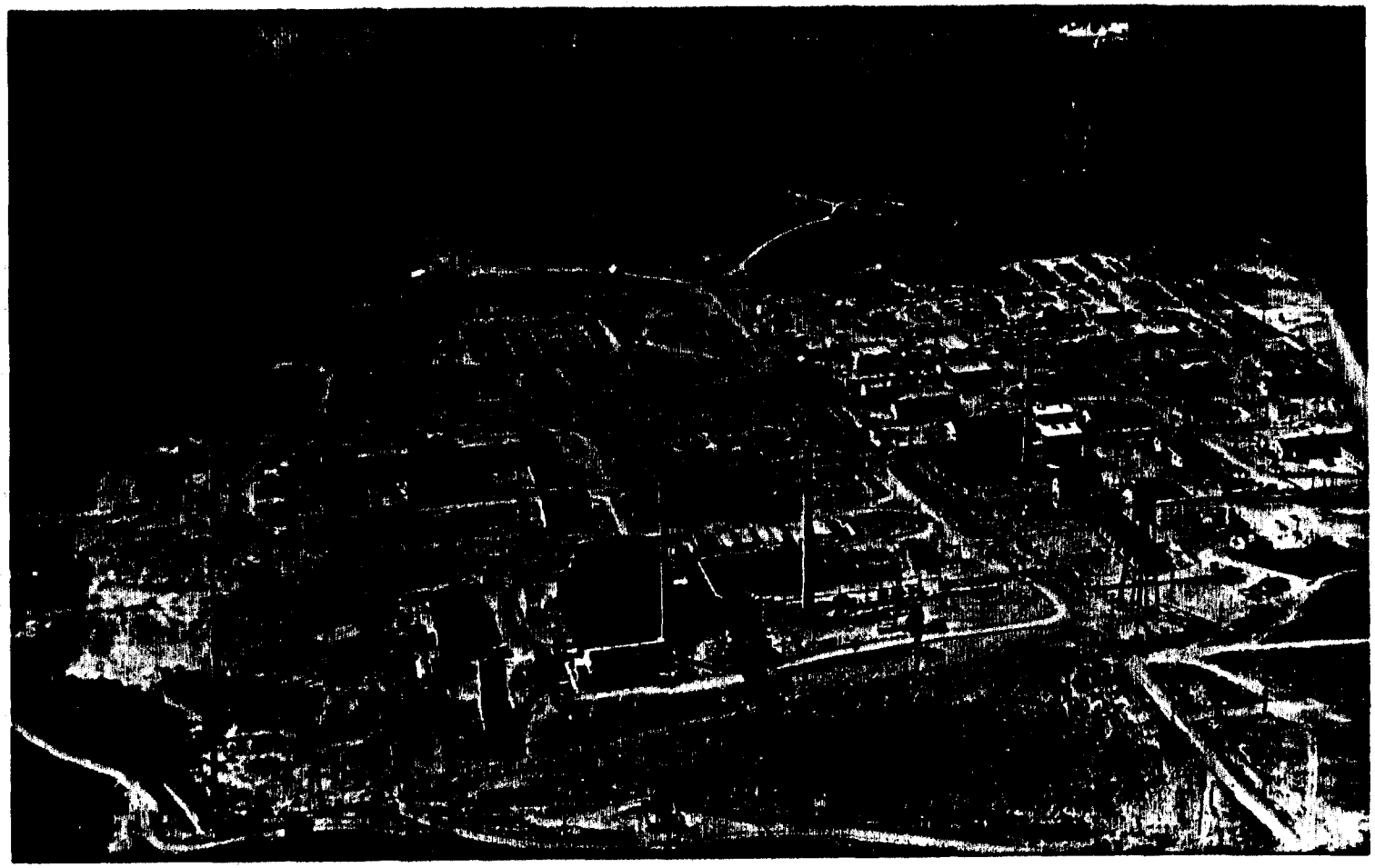

Fig. 20 CLINTON LABORATORIES IN 1944

In this photograph taken in the summer of 1944 all of the originally planned facilities for the Clinton Laboratories Pilot Plant have been completed, and for the first time very little construction is in progress. Soon after this photograph was taken additional construction was started. Note that the Physics Building behind the water tank has only one wing and that the Hot Laboratory Building located between the Graphite Reactor and the Chemistry Building has been completed, but the Fission Products Separation Building which was added to the east end of the Hot Laboratory has not yet been started.

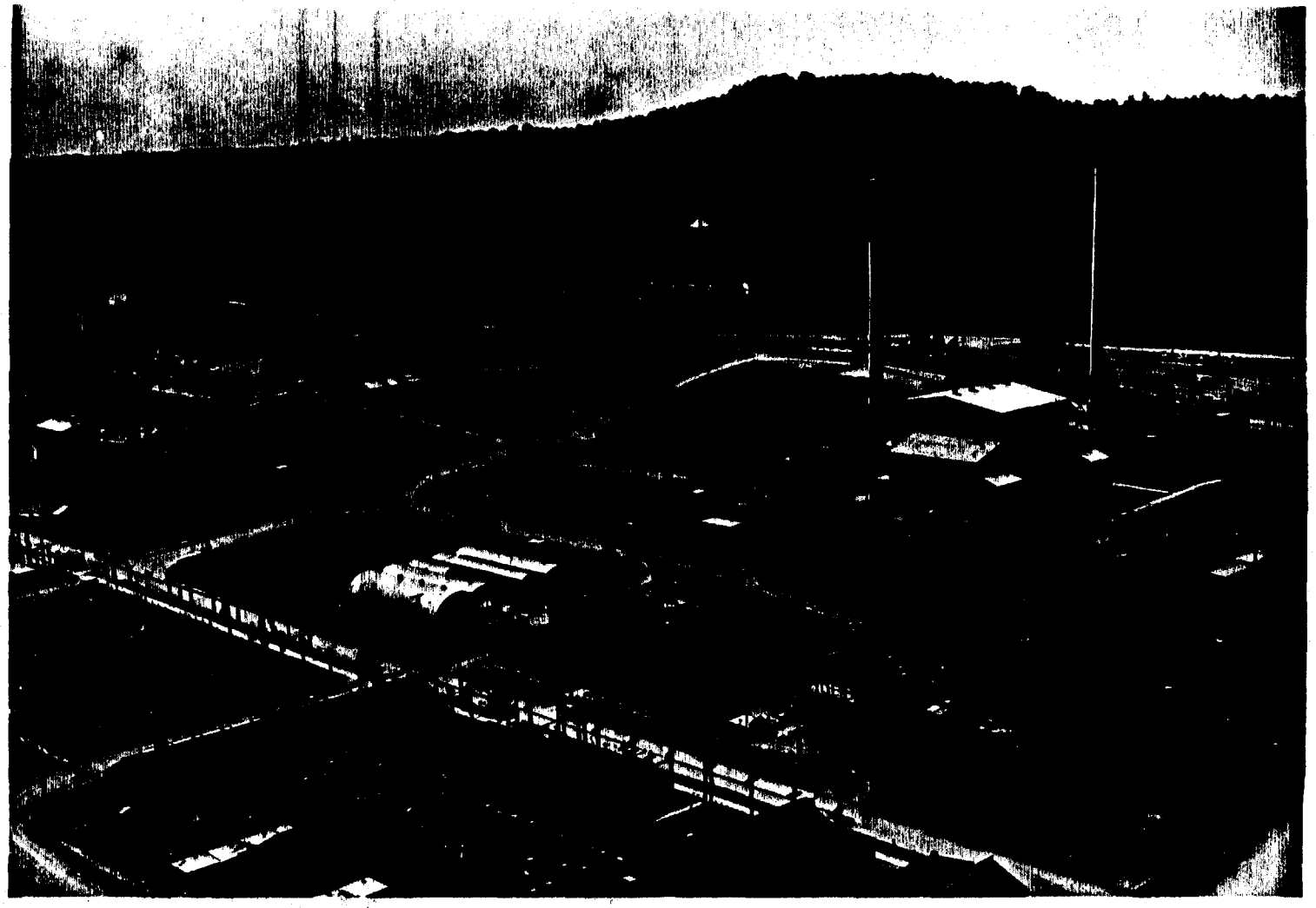

Fig. 21 LABORATORY SITE IN 1947

The Clinton Laboratories Training School Building was erected in 1946 on the chemicals storage concrete pad just across the road (downhill) from the Pilot Plant. The quonset huts which became the central machine shop were under construction in 1947, and the Fission Products Separations Building had been completed as an addition to the East end of the Hot Laboratory. The wartime black of the Graphite Reactor Building was repainted. Note that the Physics Building now has three wings and that the construction materials storage area behind the water tower indicates that construction activities are proceeding at a vigorous pace.

Fig. 22 CLINTON LABORATORIES IN THE SUMMER OF 1947

This photograph shows the Metallurgical Rolling Mill in the left foreground under construction. Note other signs of construction activity in the right foreground area.

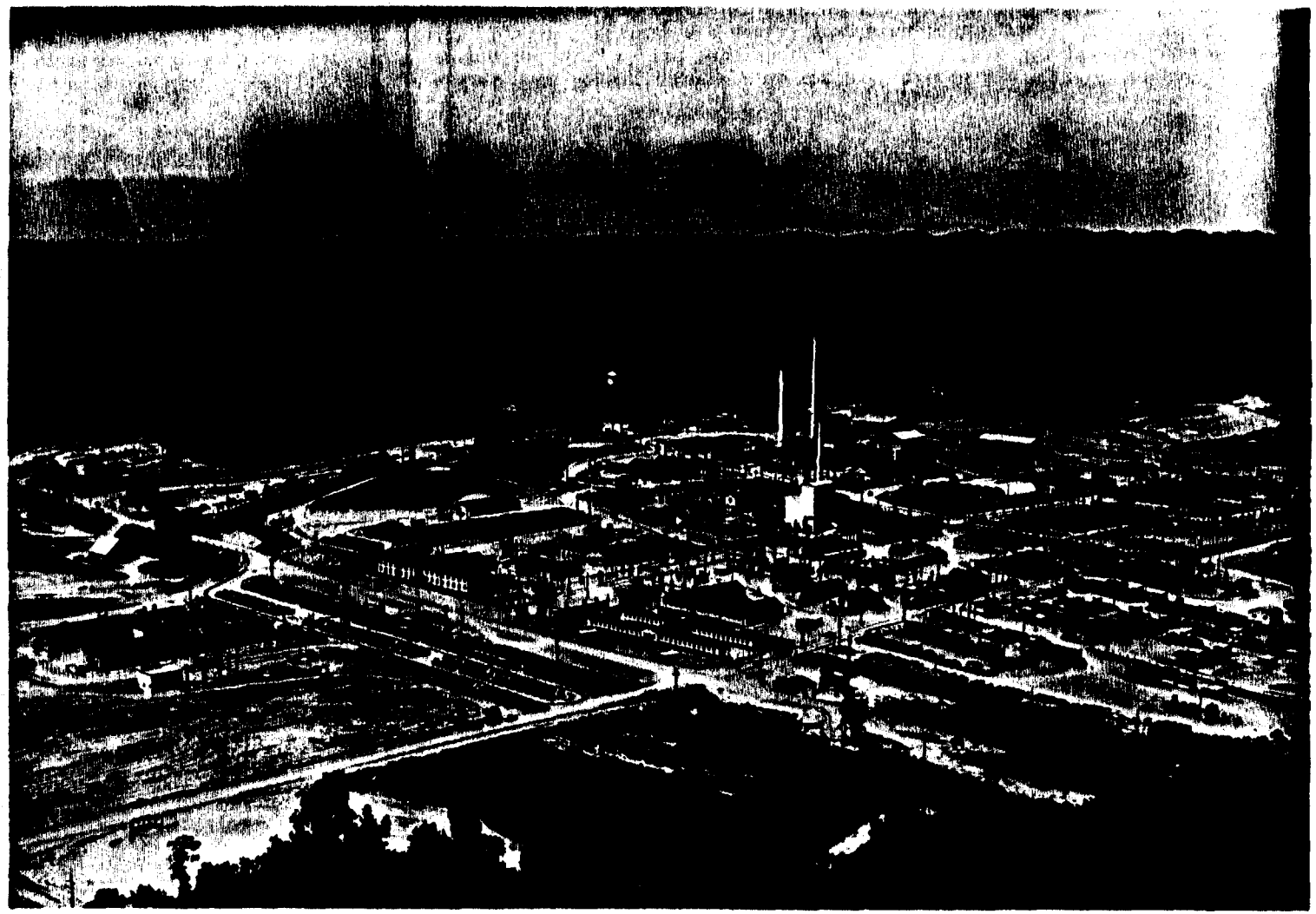

Fig. 23 CLINTON LABORATORIES SITE IN 1947

An "H" shaped frame building had been dismantled at K-25 and re-erected at the west end of the Clinton Laboratories site to provide additional office and administrative areas. In this photograph clearing and grading of the grounds around the building are still in progress.

Fig. 24 LABORATORY SITE IN 1948

In anticipation of construction of a High Flux Research Reactor at the X-10 site a new steam plant had been constructed — the first permanent masonry building at the Laboratory site. Note that the quonset huts which later housed the Metallurgy and Health Physics Divisions are nearly completed in the right foreground. The road in the left corner of the picture leads to a new reservoir also constructed in anticipation of the increased capacity required for a High Flux Research Reactor. Note the temporary steam plant which had been erected near Building 1000 when the original steam plant could not meet the load imposed by heating requirements. It is also interesting to note that even in 1948 there were no paved roads in the Laboratory site.

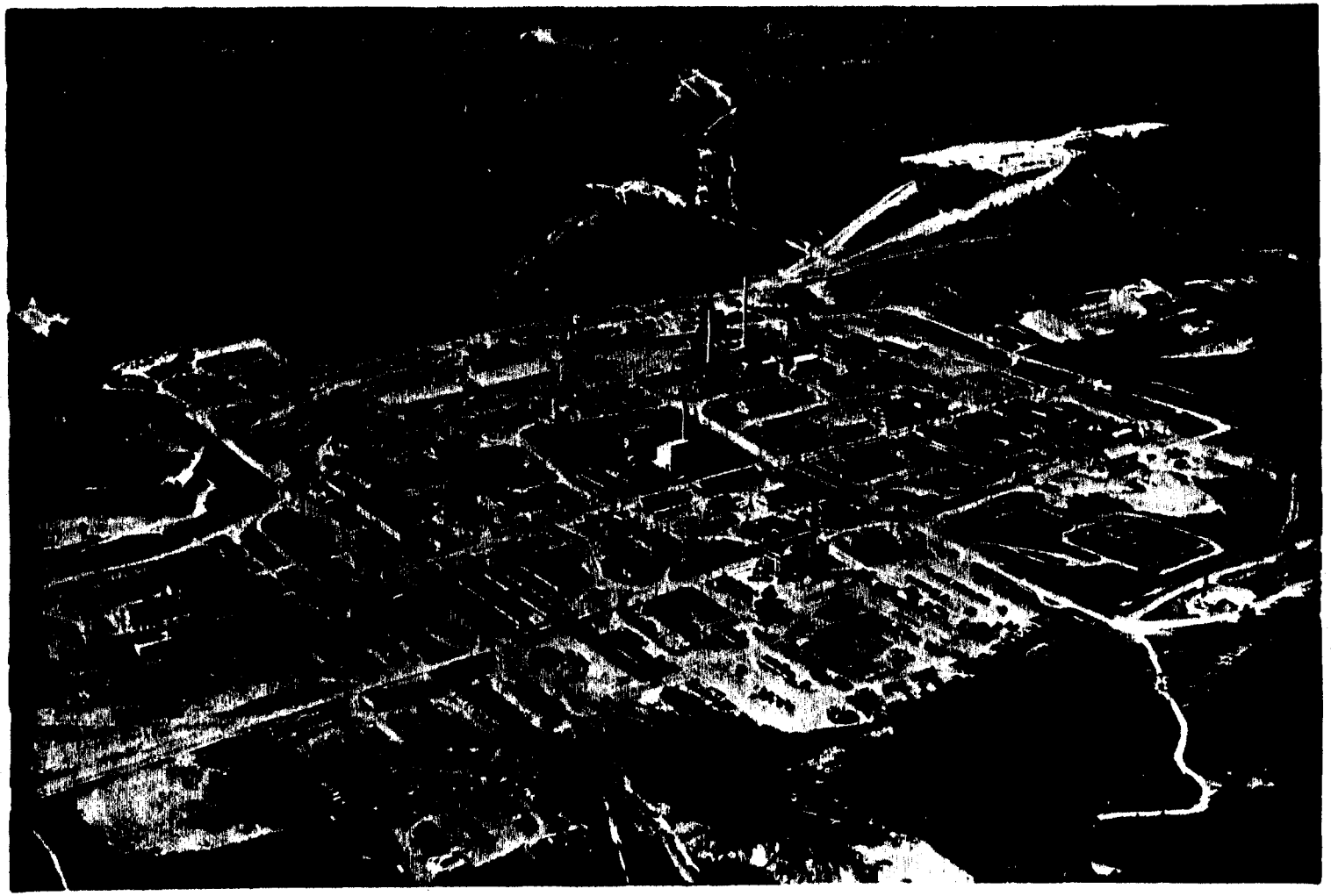

Fig. 25 LABORATORY SITE IN 1948

The new steam plant in the center is not quite completed. Note the temporary steam plants at the East and West ends of the Laboratory area. The new reservoir can be seen at upper right. A new chemical processing development building can be seen behind the Settling Basin, south of the old Chemistry Building.

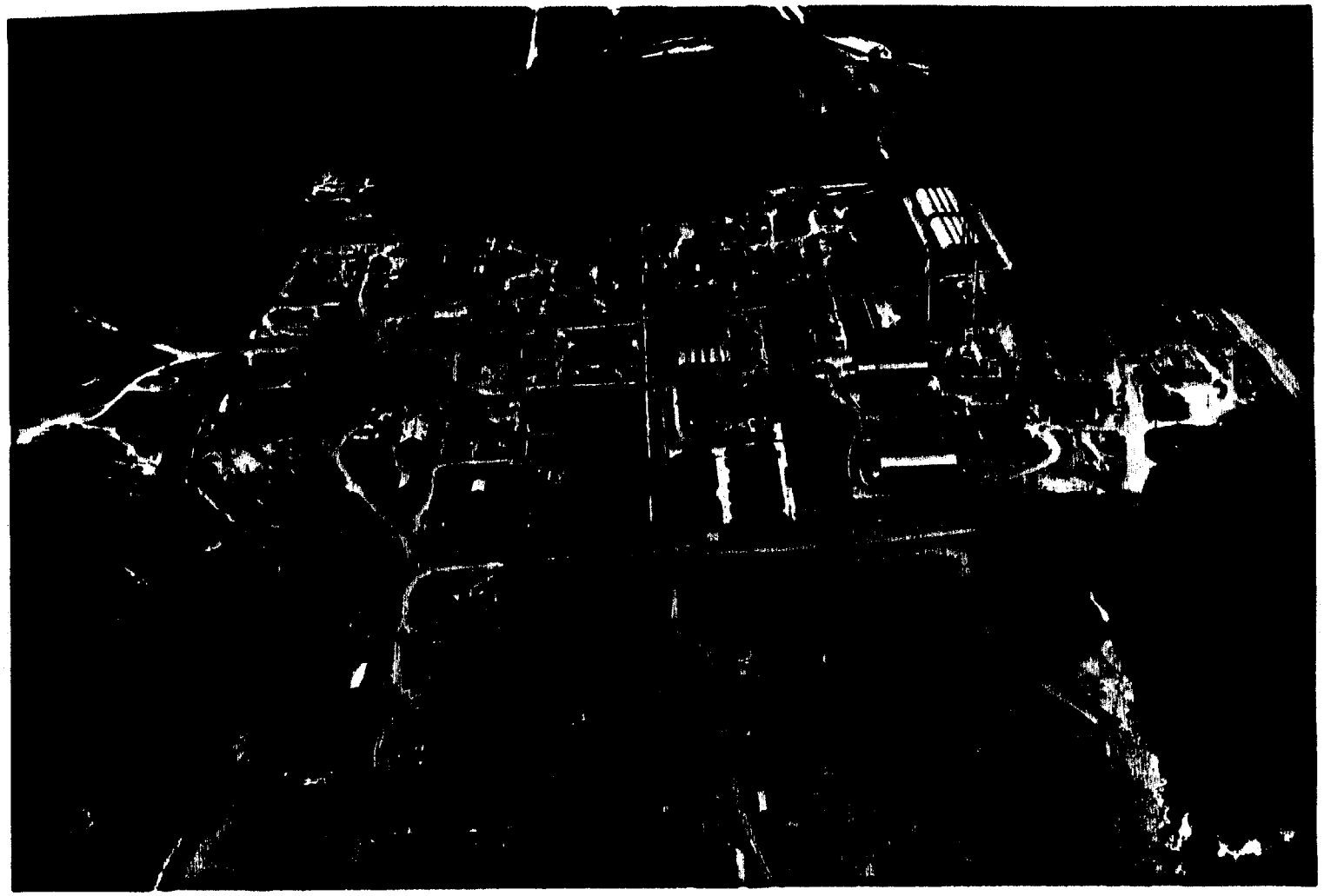

Fig. 26 CONSTRUCTION IN 1950

By the summer of 1952 the Central Research Laboratory Building 4500 and the Radiochemical Laboratory Building 4501 were nearing completion. The new Instrument Laboratory Building 3500 had been completed, as had the buildings of the Radioisotopes Area. The new cafeteria was finished and a new, permanent Change House had been constructed near the West end of Central Street. The LITR and Bulk Shielding Reactor buildings have been completed. The Metal Recovery Plant is under construction in the Tank Farm area.

Fig. 27 LABORATORY SITE IN 1950

Note how "permanentization" has changed the Pilot Plant Building 3019. The Solid State Laboratory Building 3025 has been completed, south of the Graphite Reactor. The new Health Physics Calibration Laboratory Building 2007 has been completed, south of the Health Physics quonset-type building.

Fig. 28 NEW RESEARCH FACILITIES, 1950

Details of construction activities in the summer of 1952 can be seen in this photograph. Note the foundations for the High Voltage Laboratory Building 5500 in the right foreground.

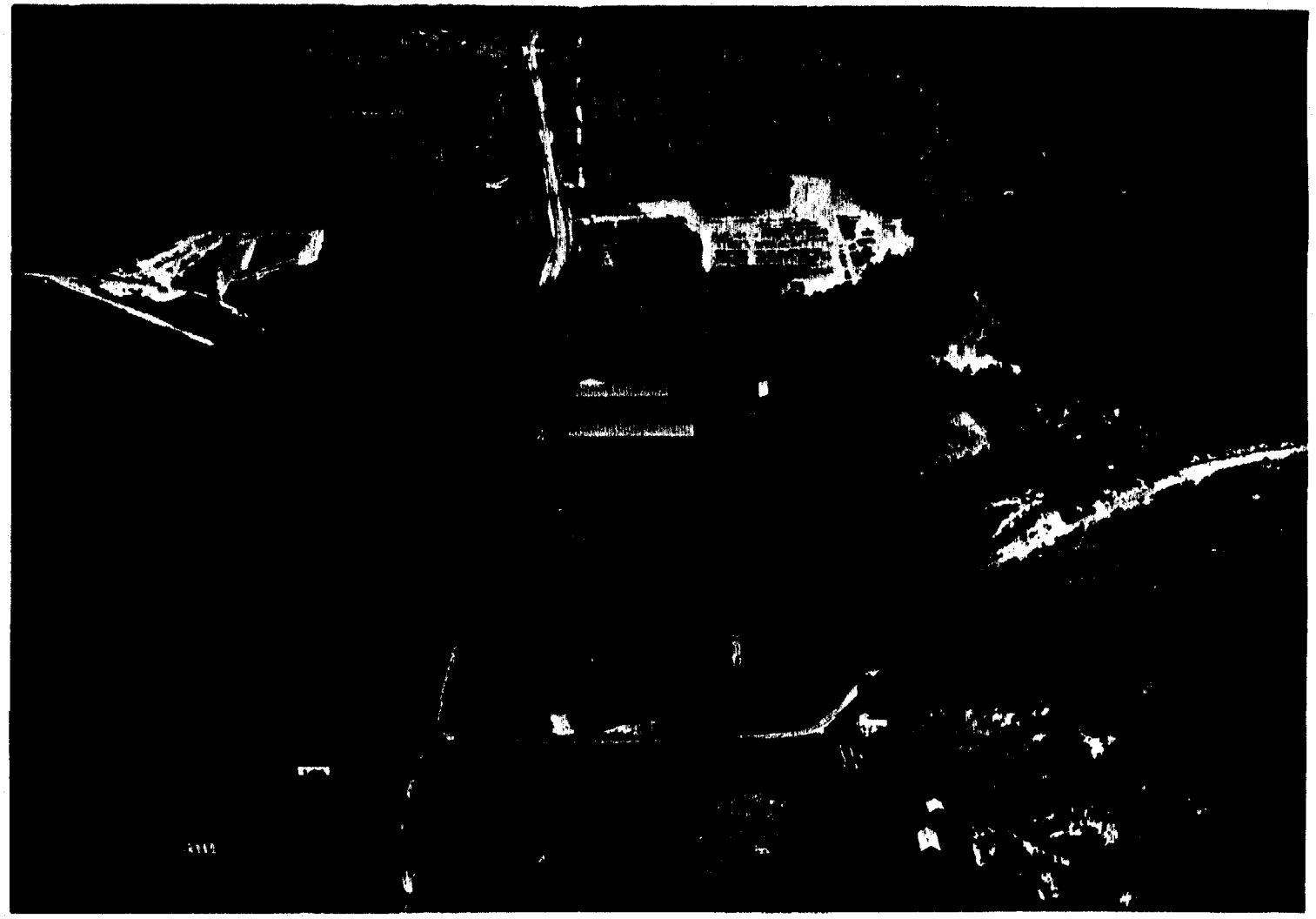

Fig. 29 CONSTRUCTION HEADQUARTERS, 7000 Area

The J. A. Jones Construction Company, contractor for the 4500 and 4501 Buildings established a shops and headquarters area east of the Laboratory site. After completion of construction activities, these buildings were turned over to the Laboratory in 1951. Note the old farm buildings still in existence.

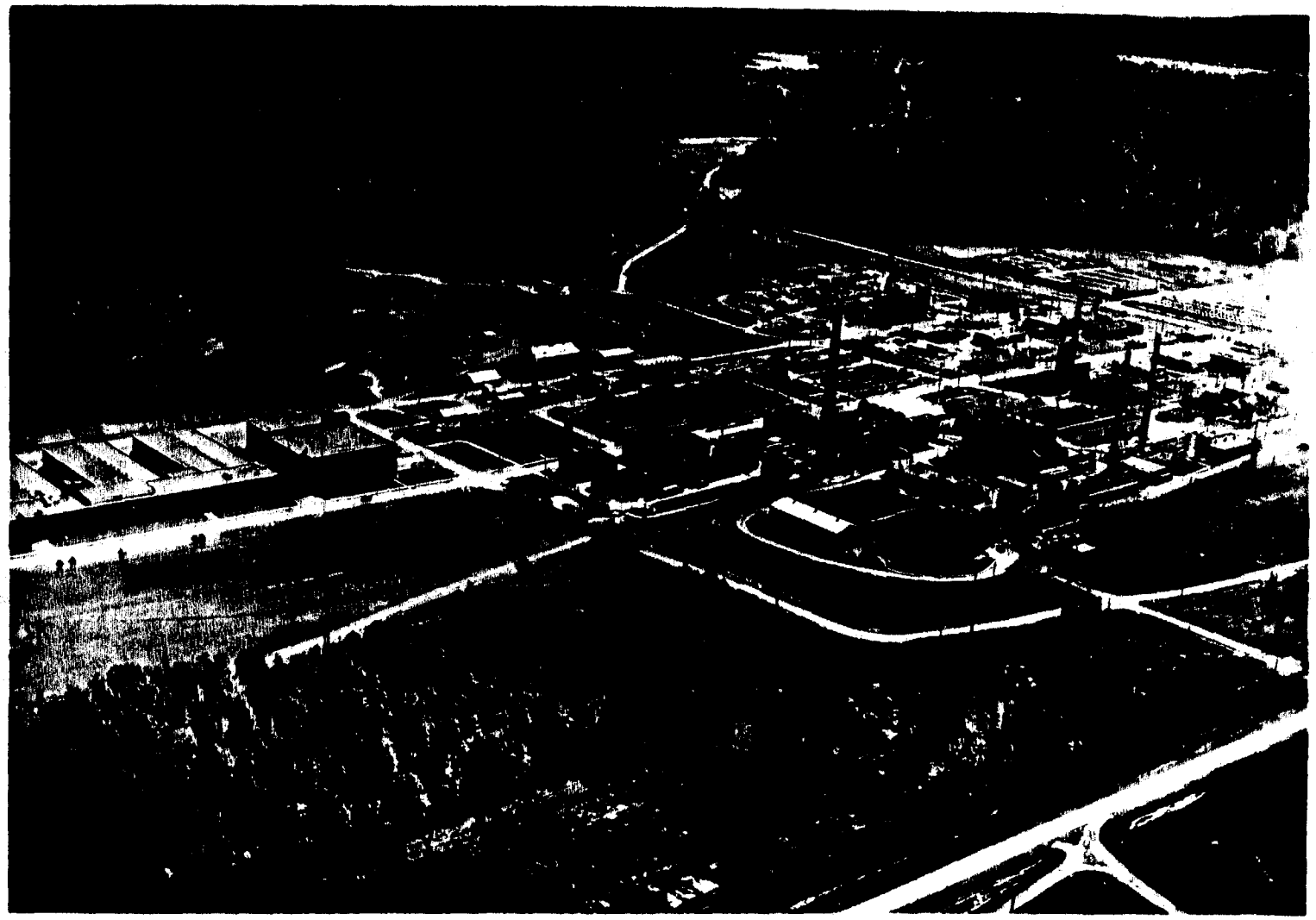

Fig. 30 LABORATORY SITE IN 1954

The new buildings added since 1952 are the Reactor School Laboratory Building 3017, to the right of the two stacks for the Graphite Reactor and the Pilot Plant, the Chemical Technology Alpha Laboratory Building 3508, and the Health Physics Waste Research Laboratory Building 3504 near the Settling Basin. Note the improvement to the grounds and the paving on some main streets.

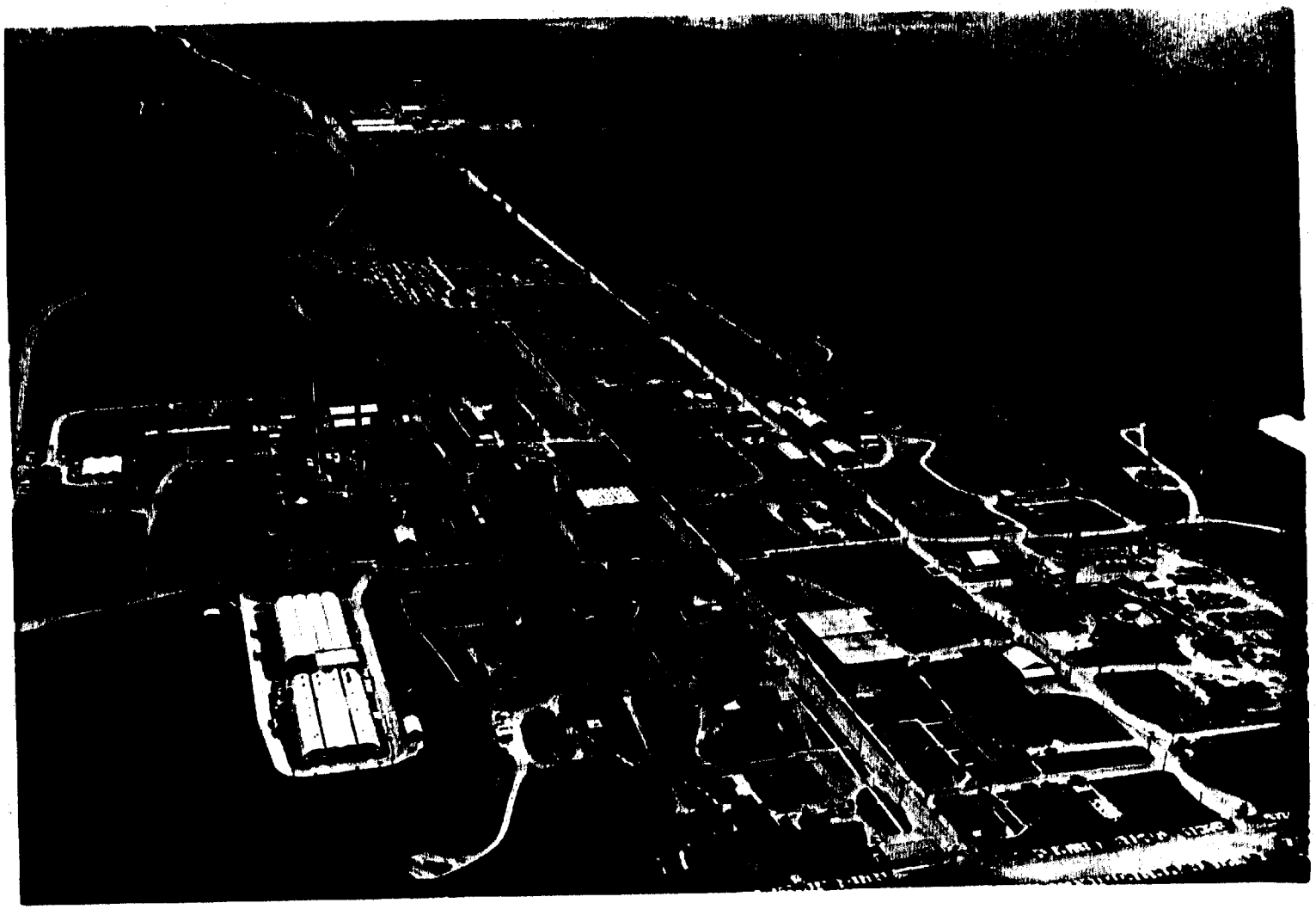

Fig. 31 LABORATORY SITE IN 1959

In 1959 the ORR had been completed, as had the addition to the Solid States Laboratory Building, the Fission Products Development Laboratory, the addition to the Instrument Laboratory, the Central Research Shops, the Process Waste Water Treatment Plant, and the High-Radiation-Level Analytical hot cells addition at the west end of Building 3019. Note the removal of a number of wartime wooden buildings plus the further improvement of grounds and streets. Part of the old Chemistry Building has been removed and the site cleared for the High-Radiation-Level Analytical Laboratory.

PART III THE OAK RIDGE NATIONAL LABORATORY 1948 to 1958

A PERMANENT BASIS

In March of 1948, the newly formed Atomic Energy Commission, then just over a year old, gave to the Carbide and Carbon Chemicals Company, a Division of Union Carbide and Carbon Corporation, the responsibility for operating the Oak Ridge National Laboratory. Since that time, the Laboratory has been operated by Carbide for the Atomic Energy Commission.

In the first year after its establishment, the Atomic Energy Commission took positive action "toward the repair of those areas of administrative and physical erosion which resulted from the period of national uncertainty following the cessation of hostilities of World War II." The Commission took a number of specific steps "to establish the (atomic energy) program on a sound, permanent basis of operation in both physical plant and organization." Under the Commission's new plan, the Oak Ridge National Laboratory was conceived to be a center for chemical and chemical engineering research, both basic and applied, with emphasis upon industrial applications.

In addition, the Laboratory was responsible for broad programs in physics, biology, and metallurgy and for the production and processing of stable and radioactive isotopes under the Commission's Isotopes Distribution Plan. Another major function of the Laboratory strongly supported by the AEC was the training program carried out in cooperation with the Oak Ridge Institute of Nuclear Studies.

RESEARCH PROGRAM IN 1948

The major fields of research at ORNL in 1948, were well established in accordance with the plans of the Atomic Energy Commission and with the special qualifications of the Laboratory from the standpoint of buildings, equipment, and experienced scientific personnel. Major emphasis during World War II had been placed upon the development of a chemical process for use at the Hanford plutonium production plant and upon demonstrating its successful operation on a pilot-plant scale. For this purpose, the graphite reactor had been built to produce small amounts of plutonium and serve other research needs, a chemical pilot plant had been built and operated for the development and testing of chemical processes, and programs of fundamental research in physics and chemistry had been established to aid in the solution of difficult problems that arose. During the war years, the need for fundamental information in other fields became apparent as the basic problems inherent in atomic energy operations were learned from experience. For example: failures of uranium fuel pieces emphasized the need for metallurgical research on reactor fuel elements and other reactor components; as the high intensities of radiation associated with reactors and radioactive materials were encountered with increasing frequency, the need for information relating to radiation exposure and its effects became urgent; expansion of the national atomic energy effort required the training of scientists in this field so new and so clouded with secrecy that it was not taught in the universities. To meet these needs, ORNL had established research programs in metallurgy and biology and a training program.

Thus, by 1948 ORNL research programs in chemistry, chemical engineering, physics, metallurgy, and biology had been established as well as a training program and an isotope-production program. Because of its major wartime efforts in chemical and chemical engineering research and development, the Laboratory was designated by the AEC as center for these activities and for isotope production, which had been entered into to meet research needs of atomic energy installations during the war.

REACTOR DEVELOPMENT

In 1948, the Oak Ridge National Laboratory's reactor development efforts were concentrated entirely upon the design of the Materials Testing Reactor (MTR) which was to be constructed at the new National Reactor Testing Station in Idaho. At this time in 1948, ORNL had no reactor of its own, nor plans for any, other than the original graphite reactor. As a part of the MTR development and design program, a full-scale mock-up of the reactor tank and major core components was constructed at ORNL for the performance of hydraulic tests to assure that the design provided adequate cooling for the reactor core. Since the MTR was designed to be the highest performance reactor to date, it was particularly important that the cooling system be designed to avoid "hot spots" in which inadequate cooling might allow the high power density to melt the fuel. When hydraulic experiments had been completed and had demonstrated the adequacy of the design, ORNL had a full-scale mock-up of the MTR with a cooling system and all the basic features necessary for an operating reactor.

The Laboratory obtained AEC authorization to perform critical experiments in the MTR mock-up to check out the nuclear characteristics of the new reactor design. A small amount of beryllium was installed to simulate the beryllium reflector of the MTR, and fuel assemblies and a simplified control system were provided for the critical experiments. The critical experiments demonstrated that the reactor would indeed perform as planned.

Having all the other essential ingredients of a reactor, the Laboratory requested AEC authorization to provide shielding around the MTR mock-up and make other modifications necessary to operate it routinely as a research reactor. AEC authorization was granted for operation at power levels to 1000 kw, and the mock-up was provided with a shield containing provisions for experiments. The reactor was called the Low Intensity Testing Reactor (LITR) and was placed in operation in 1949. Because it had a comparatively small core of highly enriched uranium, it offered the highest neutron flux available and therefore was a particularly valuable addition to the ORNL research and radioisotope production facilities. In this reactor the beautiful blue glow of Cerenkov radiation surrounding the fuel of a reactor operating in water was seen and photographed for the first time. The "blue glow" photograph of the LITR was widely acclaimed as the first photograph of a reactor core during normal operation. Modifications and improvements have been made to permit the LITR to operate at higher power levels, and in 1963 it is still in routine operation.

A serious problem in the early post-war years was the fact that there were no universities with the necessary facilities and teaching staff to offer training in nuclear energy fields. The problem was complicated still further by the fact that much of the information developed under the Manhattan District during the war was still classified secret under the security regulations. The problem of providing university training in nuclear energy fields was considered acute, especially from the standpoint of assuring the availability of trained personnel for expanding AEC operations throughout the country. ORNL undertook a minor study to adapt the basic MTR design to a reactor

which would be inexpensive, unusually safe and stable in operation, and as flexible as possible for teaching and research use in universities. With a minimum of effort, the basic features of the MTR were incorporated into such a reactor which was designed to operate submerged in a large pool of water which provided shielding, cooling and moderation for the neutrons. Because of the pool in which it operates, the reactor was popularly known as the "swimming pool" reactor from its earliest days. A swimming pool reactor was constructed at ORNL in 1950, primarily for Laboratory research activities, but, almost as important, also for the purpose of demonstrating a low cost, versatile research tool which universities could use in their nuclear education programs. This was the third reactor to be constructed at ORNL. The same blue glow around the fuel, first seen in the LITR, can also be seen during operation of the swimming pool reactor. The new swimming pool research reactor was immediately used to study, in bulk, various materials for use in improved radiation shields. The reactor therefore became known officially as the Bulk Shielding Reactor.

MTR Type Reactors

The basic design developed at ORNL for the high performance Materials Testing Reactor has proved, in actual operation of the MTR, the LITR, and the Bulk Shielding Reactor to be very flexible and to possess numerous advantages. ORNL has continued efforts to improve various aspects of the design of MTR-type reactors and out of these efforts have come fairly standardized

designs for research reactors of the "swimming pool" variety. It is gratifying to the Laboratory to note that many reactors of this type have been constructed at universities and other research establishments.

A continuing program at ORNL has been pursued not only to improve this reactor type, both in utility and safety of operation, but also to facilitate the design of similar reactors. An example of the improvements is the development of better control rods and mechanisms for their manipulation. As a result, it was possible to increase the approved operating power level for the ORNL swimming pool reactor from the original 10 kw to 1000 kw. The higher power made necessary a special scheme to disperse the radioactive cooling water rising from the reactor, and water jets were designed and installed for this purpose. The reactor improvement program also included development of stainless-steel-clad UO_2 fuel elements, which have a greatly lengthened lifetime against corrosion. The original aluminum elements lasted only about two years.

In addition, an effort was made to reduce the number of costly experiments required for the design of each new swimming pool type reactor. As part of this program, a method was developed for predicting theoretically the critical masses of such reactors. A number of calculations were performed for comparison with three experimental loadings of the Bulk Shielding Reactor which were arranged in as simple a geometry as was possible. The predicted critical masses agreed to within 1% with the experimental masses.

In addition, a Pool Critical Assembly was designed and constructed in a corner of the Bulk Shielding Reactor Pool so that critical

experiments or others requiring very little power can be conducted without tying up the full facilities of the larger reactor. In this way the critical assembly can be operated while experiments are being installed or other work is being done on the Bulk Shielding Reactor, although when the Bulk Shielding Reactor is in operation, the PCA must be shut down.

TSF Reactor

The Tower Shielding Facility was developed for shielding research at ORNL. It incorporates a reactor of modified MTR design, developed for this specific application at ORNL. Constructed in 1953, the reactor is completely enclosed in a spherical container and is suspended on cables between 300-foot towers so that it can be raised from ground level to nearly the full height of the tower. It is a tribute to the designers of the reactor that it has operated at full design levels very satisfactorily in every respect even though the reactor is suspended in midair and is controlled from a control room a considerable distance away. Since the reactor was constructed for use in shielding studies related to the development of improved aircraft reactor shields, there is no built-in shielding around the reactor proper. Therefore, when the reactor is in operation, operators and observers must stay inside a shielded control room some distance away, from which point they can view the reactor by means of a television system installed for that particular purpose. The Tower Shielding Facility has contributed significantly to the outstanding achievements in shielding research that have made possible the design of improved reactor shields that are far more compact

and lighter than previously thought possible. This achievement in shielding research is one of the technological advances now finding application in the nation's space program.

Army Package Power Reactor

Still another modification of the MTR design is embodied in the Army Package Power Reactor developed at ORNL. In the fall of 1952, a suggestion was made that another way to exploit the compactness of the nuclear power unit would be the development of a small nuclear power plant that could be installed at remote or relatively inaccessible locations where the nuclear power costs would be competitive with conventional power costs for the area and where the fuel, because of its compactness and potentially long life, would have logistic advantages over conventional fuels.

Early in 1953 a small group at the Oak Ridge National Laboratory started work on selecting a reactor type which would permit the design of a reliable, inexpensive system which could operate long periods of time without refueling. The reactor chosen was a heterogeneous, pressurized water, stainless steel system. The conceptual design on the system was completed by July 1954. An artist's conception of the plant is shown in Figure 1. The net electrical power and steam heat were selected to be 1000 kw and 3500 kw respectively. The reactor core contained highly enriched uranium in flat, plate-type elements which were clad with stainless steel. Standard components were used wherever possible, and the special components were designed for reliability and long life.

At the time the ORNL study was being made, the U. S. Army Corps of Engineers, because of the military interest in remote bases,

established a group for investigating nuclear power for application in remote military stations. This group became interested in the package reactor work at ORNL and decided to have a full-scale prototype generating station based on the ORNL conceptual design built at Fort Belvoir, Virginia. The ORNL package reactor conceptual design was used, after certain modifications of the specification, as the basis for obtaining bids for construction of the Army Package Power Reactor (APPR-1). The contractor selected was Alco Products, Inc., and a fixed price contract was awarded to them on December 10, 1954. Ground breaking at the Fort Belvoir site occurred on October 5, 1955. A year and one half later on April 8, 1957, the APPR-1 went critical (see Figure 2). During the design, development, and construction stages, the Package Reactor Group at ORNL advised on the reactor program, reviewing the design and performing experimental, development, and testing work including the critical experiment, simulator studies, the development of the fuel element and control rod, control rod drive development, and the materials irradiation testing program. Fuel elements and control rods for the first core loading were fabricated by ORNL.

The APPR-1 operation performance exceeded expectation. When the reactor was taken up to full power (10 MWR), the load response characteristics were found to be excellent.

ORNL Research Reactor

Additional improvements in the basic MTR design have been developed by the Laboratory and incorporated into the very flexible high performance research reactor (ORR) constructed at ORNL. This reactor incorporates features that make it particularly useful for

engineering research as well as irradiations and neutron beam studies of various types. Although the ORR will not reach the high power levels and neutron fluxes of the MTR, it has far more flexibility for experimental use and provides greater access to the reactor core for irradiations and experiments.

The ORR went critical on March 21, 1958. The complete reactor installation, including buildings and auxiliary equipment, cost approximately five million dollars. It is a high-flux, light-water-moderated and cooled reactor with the core enclosed in a tank so that it can be cooled by a high velocity flow of water. The entire reactor tank is also immersed in a pool of water. The reactor can therefore operate at high power, similar to the MTR, and still have the ready access of a swimming pool type reactor.

The ORR is designed to operate at a power level of 30 megawatts with a maximum neutron flux of approximately 4×10^{14} n/cm^2/sec. The power level was limited at first to 20 megawatts by the external water cooling system, but the cooling capacity was subsequently increased to permit operation at 30 mw.

The ORR has been given thorough performance tests and has been operated routinely at 20 to 30 megawatts since July 1, 1958. The reactor provides unusual flexibility for performing engineering scale tests of reactor systems or components under conditions of reactor operation. The MTR-type reactor core is contained in a cylindrical tank which is flattened on one side. The reactor core tank itself is surrounded by water. One side is used for beam hole tubes which extend through the concrete shield; the flat side is clear of obstructions and faces the main pool of water. This permits experiments or

materials to be irradiated in a zone of high neutron flux simply by lowering them through the water to the level of the reactor core. Irradiations can be started or terminated in this location without requiring that the reactor be shut down. Two very large holes, 18" x 24" in cross section, provide access for experiments to be placed along other sides of the reactor core for high flux irradiations.

In most high flux research reactors, there is a delay of from 10 to 15 hours in starting up after a shutdown because of the time required for the xenon poison in the fuel to decay. Xenon continues to build up in concentration after the reactor is shut down because other fission products form xenon in their decay chains. However, after a few hours the xenon concentration reaches a peak and thereafter decreases rapidly. Thus, after a shutdown, a reactor could be started up again in a short time - before the xenon concentration builds up - or it must wait 10 to 15 hours for the xenon to pass its maximum concentration and start to decay faster than it is formed. In the ORR a unique and very valuable feature is that the rods for reactor control are operated by mechanisms located below the reactor core level, leaving the top of the reactor unobstructed and readily accessible for easy replacement of fuel. This feature permits fuel to be changed completely within two or three hours so that by having a spare fuel loading on hand, the buildup of xenon poison when the reactor is shut down is not a problem. The poisoned fuel elements can simply be replaced and held in storage in the pool around the reactor tank until the xenon decays.

Gas-Cooled Power Reactors

In 1958 ORNL initiated development work on gas-cooled power reactors. Initially a design study was made to determine whether reactors of the general type built by the British could be advanced in design and technology to compete economically with reactors of the other types being developed in the United States. The principal conclusions from this study were:

1. Gas-cooled, graphite-moderated power reactors have good future prospects for application in the United States.
2. Enriched gas-cooled reactors will produce power more cheaply than natural uranium reactors.
3. Gas-cooled reactors are technologically and economically competitive with pressurized-water reactors for power production.
4. Helium-cooled, graphite-moderated reactors utilizing 2% enriched UO_2 fuel elements clad with stainless steel and having a maximum gas temperature of 1000°F represent a good starting configuration comparable with current technology.

Following this study, ORNL accepted responsibility for developing the fuel element to be used in the first gas-cooled power reactor planned for construction in the United States and for continuing advanced studies aimed toward developing a reactor of improved performance.

Homogeneous Reactors

It is probably no exaggeration to say that the MTR idea (parallel plates with water moderator and coolant) has had as profound an influence on the whole development of power reactors as any concept advanced. But when the MTR design and development program was nearing completion in 1949, ORNL had no other major reactor development projects planned for the future.

New information had been developed and technological advances had occurred in materials and components fields since the early homogeneous reactor development efforts had been dropped in 1945 because the problems appeared insurmountable. It was decided, therefore, to take a new look at the feasibility of homogeneous reactors to see whether the problems which had stymied further development in 1945 could be overcome in 1949.

Perhaps the most serious of the problems which caused the abandonment of homogeneous reactors in 1945 was the formation of bubbles in the fuel solution as a result of the decomposition of water in the strong radiation field. Minimizing the bubble problem by operating at elevated temperatures and pressure was not considered seriously in 1945 because the only materials then known to have sufficiently low neutron absorption cross sections to be useful in a breeder reactor were beryllium, aluminum and lead. These materials were not considered suitable for a reactor system operating at high temperature and pressure. By 1949 the very low neutron absorption cross section of pure zirconium (free of hafnium) had been discovered and zirconium was considered a very promising structural material for homogeneous reactors. Its strength and corrosion resistance, as well as its low neutron absorption

cross section, appeared very favorable for homogeneous reactor applications. Also, considerable experience had been gained in handling radioactive liquids at high temperature and pressure and many components for high pressure, high temperature systems had been tested in actual use. Further, considerable research had been performed on uranyl sulfate-water solutions and had indicated that these solutions showed favorable characteristics for use as fuel solutions in aqueous homogeneous reactors. Conceptual designs of possible systems for homogeneous reactors indicated their feasibility and appeared very promising for further development.

By the end of 1949, the Atomic Energy Commission had given approval for a program of research and development leading to the construction of a homogeneous reactor. About the middle of 1950, the status of the entire homogeneous reactor development program was reviewed and a proposal was made to construct a small homogeneous reactor as an experiment. It was proposed to construct the experimental reactor as quickly and cheaply as possible to demonstrate the feasibility and operating characteristics of homogeneous reactors. It was expected that the homogeneous reactor experiment could accomplish the following objectives:

1. To demonstrate the operational feasibility of a circulating fuel chain reaction at fairly high power.
2. To study radiation decomposition and corrosion in a homogeneous chemical reactor at power densities of the same order as those which might be encountered in a power reactor.
3. To demonstrate electrical power production from a circulating fuel system.

4. To obtain direct and complete operating experiences in the handling of a homogeneous reactor at high temperature and high pressure.

Approval for the construction of the HRE was received from the Atomic Energy Commission in September 1950. The HRE building was completed in March 1951, and construction of the reactor was completed in January 1952.

During the later stages of construction, a thorough and rigorous program of testing and inspection was carried out to assure the satisfactory operation of the entire system. Since leaks were considered to be a major problem in the high temperature, high pressure system which was designed to contain large quantities of radioactive materials, major emphasis was placed on inspections and tests to assure that the system would be leak-free under full operating pressure.

Simulated operation with natural uranium fuel solution at 250°C and 1000 psi pressure was carried out to continue testing the system. The testing program lasted through January, February and March of 1952, and at the end of this period it was believed that the system had been demonstrated to be leak-tight and to be in good operating condition. Critical experiments were carried out through April, May and June with low power, high temperature operations and required several months for cleaning out the system, making repairs and retesting the entire system.

In October, operation was resumed, going almost immediately to the kilowatt power level to create sufficient activity in the fuel to facilitate the detection of leaks. Minor leaks were discovered and repaired without difficulty, and operation continued at increasing power levels. The HRE was brought to its full design power of one

megawatt on February 24, 1953. At this power level, high pressure steam was generated by the reactor to operate a turbine generator and produce 150 kilowatts of electricity. This was only two months after the first demonstration of electric power production from nuclear energy in the Argonne National Laboratory's Experimental Breeder Reactor. The HRE was operated through a complete program of tests to determine its characteristics and behavior. Operations continued most successfully through all of 1953 and the early part of 1954. In 1954 the HRE, having accomplished all of its objectives with very promising results, was dismantled so that a larger homogeneous reactor could be built in its place to continue the development of large-scale power reactors.

HRE

Among the important results obtained from operation of HRE-1 were demonstrations of (1) a remarkable degree of inherent nuclear stability, (2) the lack of need for mechanical control rods, (3) the direct dependence of reactor power upon turbine demand, (4) flexibility and simplicity of fuel handling, (5) the ability to attain and maintain leaktightness in a small high-pressure reactor system, (6) the safe handling of the hydrogen and oxygen produced by the radiation decomposition of the water and (7) the use of copper sulfate as a homogeneous catalyst for recombining these gases as they formed in the fuel. These results, plus other encouraging results from the concurrent development programs, resulted in the expansion of the program, with the development of a large thorium breeder power reactor as the ultimate goal.

HRT

As a step toward this goal, the Homogeneous Reactor Test (HRT) or Homogeneous Reactor Experiment No. 2 (HRE-2) was built. Homogeneous Reactor Experiment No. 2 was a two-region, forced-circulation aqueous homogeneous reactor. Located at the site formerly occupied by HRE-1 at Oak Ridge National Laboratory, it was an advancement over its predecessor in power, physical size, and quality of construction. Operating at the design level of 300°C and 2000 psia, the reactor produced 5 megawatts of heat. Uranyl sulfate solution containing 10 grams of uranium (93% U^{235}) per kilogram of heavy water circulated through the reactor core at a rate of 400 gallons per minute. The fluid entered the core at 256°C and leaves at 300°C. Copper sulfate added to the fuel solution served as a dissolved catalyst for the internal homogeneous recombination of the radiolytic gas. The first experiment with HRE-2 utilized heavy water as the blanket.

The objectives of HRE-2 were (1) to demonstrate that a homogeneous reactor of moderate size can be operated with the continuity required of a power plant, (2) to establish the reliability of fuels, engineering materials, and components with features which can be adapted to full-scale power plants, (3) to evaluate equipment modifications which will lead to simplifications and economy, (4) to test maintenance procedures and, in particular, maintenance under water, and (5) to develop and test methods for the continuous removal of fission and corrosion contaminants, for which HRE-2 was supplied with an integrated fuel processing plant.

Design of HRE-2 was started in January 1954 and construction was begun in July of the same year. Construction of the reactor and the

high-pressure system of the associated processing plant was completed in the summer and early fall of 1956. Both plants went through periods of cleaning, preliminary testing, and non-nuclear operation before the first critical experiment was performed. The reactor reached cricicality on December 27, 1957, and was first operated at full power in February of 1958.

Construction of HRE-2 would not have been possible without the development of new or improved equipment for handling the uranyl sulfate solution fuel at high pressure and temperature. These developments were made by Laboratory personnel or by Laboratory personnel in cooperation with equipment manufacturers. Notable examples were the 5-ft-diameter 2000-psi reactor vessel which contained the first reactor core tank made of Zircaloy, the canned-motor circulating pumps which were modified especially for HRT use, the bellows-seated high-pressure valves and liquid-level indicators, the feed pumps used to inject fluids into the reactor against a pressure of 2000 psi, the hydroclones used to remove fission product and corrosion product solids at high temperature and pressure from the reactor system, and many instruments and other items of special reactor equipment. All of the reactor equipment was designed and fabricated to achieve reliability and leak-tightness much superior to those found in normal industrial operations. Non-radio-active testing facilities in which many of the reactor operating conditions could be simulated were constructed and operated for thousands of hours in developing and testing equipment.

Maintenance of a homogeneous reactor system presented many new and unique problems because the equipment was large and highly radioactive. Special tools and techniques were developed; and HRE-2 was

designed so that the equipment can be submerged and maintained while using water for shielding against radiation. These direct maintenance methods were an ORNL innovation which promised to solve many of the most difficult problems of making repairs on highly radioactive systems.

At the time that HRE-1 was built the severity of corrosion at 250°C was believed to limit the operating temperature and the feasibility of homogeneous reactors. It was later established that the corrosion is less severe as the temperature is increased above 250°C to 300°C, and HRE-2 was designed to operate in that temperature range. The higher operating temperature greatly increased the attractiveness of homogeneous reactors for producing power.

HRT Operations

The power experiments were started during the last week in March and were continued through April 4, 1958. The power level was raised to 3 Mw and then up to 5 Mw. The reactor had operated for only a short time at the 5-Mw level when a leak developed that permitted fluid to be transferred from the core to the blanket. Operation was continued for a short time with fuel transferring between core and blanket while it was being determined that a leak existed, and then the reactor was shut down to permit an examination of the damage.

The reactor was inspected to determine the location and the cause for the leak. Periscopes were used to view both the inside and outside of the core tank, but it was not possible to identify positively a hole or crack large enough to account for the leak, even though the location in the lower diffuser section of the core vessel appeared to have been established by means of liquid level measurements in the system. After many unsuccessful attempts, the work on viewing was

discontinued, the reactor system was sealed again and experiments were performed to determine how best to resume nuclear operation. These tests involved circulating heavy water and uranyl sulfate solutions in both the core and blanket systems while investigating the problems of maintaining liquid level and chemical stability in the systems. It was shown that the reactor could operate satisfactorily with fuel in both the core and blanket regions and that fuel concentrations could be controlled to produce 60% of the power in the core.

On the basis of these tests, it was concluded that no modifications were necessary for reactor operation to continue. On June 4, 1958 the reactor was started up again and the reactor behavior was explored at core average temperatures of 240, 260, and 275°C and at total power outputs ranging from 250 to 3500 kw. Following this, the reactor was operated routinely for 30 days at 280°C and 1750 psi. During the run, 1600 Mw/hr of thermal power was generated, bringing the total thermal power generation by the HRT to approximately 2000 Mw/hr.

The period from July 7 through July 18 was spent in attempting to make satisfactory examination of the reactor core tank and the interior of the pressure vessel. Although detailed observations were made of parts of the interior and exterior of the core tank, it was not possible to identify the region of failure. Accumulations of solids were found in the core at the junction between the diffuser screens and the wall. There was nothing unusual in the appearance of the metal surfaces or the welds that could be observed.

The HRT operated intermittently from August 1958 through 1959 at power levels as high as 5 Mw. The reactor performance was free of major mechanical difficulties. Circulating pumps, feed pumps, valves,

and the many other items of equipment within the reactor cell performed exceptionally well. Experience with reactor maintenance was surprisingly good. The simple tools for remote maintenance performed well when replacing valves, removing samples from the circulating pump lines, opening and closing the piping systems, and inspecting the reactor vessel, core tank and piping.

However, operational experience with the HRE-2 during 1959 served to call attention to important questions that were not fully resolved. These questions were related to the phase stability of uranyl sulfate solution fuels under reactor operating conditions, the use of austenitic stainless steel for containing uranyl sulfate solutions, and the design, operation and metallurgy of the Zircaloy core tank. Experience in the operation of HRE-2 and in the research and development program both in- and out-of-pile indicated that the uranium was separating from solution and concentrating at spots along the core tank wall. Fission of the uranium that was concentrated at spots on the tank walls evidently generated more heat than the normal flow of fuel through the core could remove and overheating of the tank walls occurred at these spots. Evidence accumulated that the mixing of fuel and blanket observed after the first high-power runs had occurred because at high power levels uranium on the tank wall had generated enough heat at one point to melt a small hole through the wall.

During 1959, experimental operation of HRE-2 achieved a unique record for reactors of all types by operating continuously for 100 days. This achievement demonstrated clearly a unique advantage of the fluid fuel system: new fuel could be added and fission product poisons removed while the reactor continued normal operation. The continuous

run of the HRE-2 actually lasted for 105 days, after which it was terminated because of clear indications that a second hole had developed in the core tank. The hole actually developed on January 21, 1960 after several weeks of operation at high power levels (about 5 Mw), which suggested at the outset that again uranium deposits on the tank walls had overheated, burning a hole through the core tank.

The most probable explanation for the burning of the hole was based on the relationship between the hydrodynamics of the reactor core and the chemistry of the uranyl sulfate solution fuel. The upper two-thirds of the reactor core tank was a spherical shell and it was joined to a truncated cone. Fuel entered the core tank at the bottom, was diffused by perforated plates, flowed upward through the cone so there would be a uniform temperature rise as it flowed through the sphere and out the top. Flow velocities were so low that corrosion- and fission-product solids on which uranium was sorbed could be trapped in the core. The flow distribution caused the solids to accumulate along the wall. During nuclear operation, the conditions of low flow, flow separation and recirculation where diffuser screens were attached to the wall, and accumulation of uranium-bearing solids along the wall are believed to have caused local wall temperatures to exceed the maximum design temperature. As the power level was raised, local boiling occurred and uranium deposited as a solid or, if the pressure was high enough, as a heavy liquid phase. A characteristic of uranyl sulfate solutions is that they separate into a heavy phase rich in uranium and a light phase depleted in uranium at high temperatures which depend on the concentrations of salts in the solution and on the ratio of sulfate to excess sulfuric acid. Power excursions

resulted from rapid movement of some of the material into or through regions of higher nuclear importance. When local accumulations became large enough, the heat production rates exceeded those which could be removed by boiling heat transfer, and the tank wall melted at the point of overheating.

The reactor core tank was inspected by the use of a periscope which showed a new hole just below the equator of the core tank. With a radiation field of about 100,000 rads in the reactor core region, workers were nevertheless able to perform a detailed examination of the core tank using remote control devices. The flow-directing screens in the bottom part of the core tank were removed, all surfaces of the tank walls were systematically photographed for corrosion examinations, wall thicknesses were measured, and the two holes in the core tank wall were plugged, all by remote maintenance techniques.

The direction of fuel flow (originally from bottom to top) through the core was reversed so that solids which entered or were formed in the core would be flushed out. Introducing the fuel into the top of the core as a jet was expected to induce higher circulation rates over the core tank wall surfaces and to promote better cooling. The acidity of the fuel was increased to raise the phase separation temperature.

Work on modifications to the HRE-2 lasted through most of 1960, and the reactor was ready to resume operations early in November 1960, with the flow reversed (now from top to bottom) in the core. During the period of the shutdown and repair of the HRE-2 Congressional hearings on the AEC budget were in progress. Congressional reaction to the overall homogeneous reactor situation was unfavorable because

it appeared that the large expenditures for homogeneous reactor development over a period of ten years had culminated in an experimental reactor which had encountered some fundamental problems. Under sharp attack from Congress, the AEC instructed ORNL on December 28, 1960 to terminate the homogeneous reactor development and testing program by July 1, 1961 and to close out research activities as quickly as could be done in an orderly manner. The AEC also requested that the HRE-2 be operated at a near full power for two or three months prior to shutdown.

ORNL vigorously protested these decisions, pointing out that the problems encountered in HRE-2 had been greatly clarified by the recent studies and examinations and that solutions for these problems seemed straightforward and easily incorporated into plans for future homogeneous reactors. The Laboratory argued that the long range goal of developing a thorium breeder reactor to produce economical electric power should still be pursued as vigorously as before.

In the end, the AEC agreed to continuing support for thorium breeder technology, but insisted that the aqueous homogeneous reactor research and development be dropped. The HRE-2 operated at full power in January, February, March and April until operations were terminated on April 28, 1961, when it appeared that the plug in the hole of the core tank wall had disintegrated. HRE-2 was then dismantled for detailed inspection of its components to determine the full extent of corrosion, wear, radiation damage and other changes throughout the system. Final reports were prepared on all important phases of the work to close out the homogeneous reactor program in 1961.

Research and development on thoria slurries and other aspects of thorium breeder reactor development became the objective of the new

thorium utilization program which was started in 1961 to pursue promising systems indicated by the work of the previous ten years. The thorium utilization program was greatly reduced in level of effort from the homogeneous reactor program.

AIRCRAFT NUCLEAR PROPULSION PROJECT

In 1946, the U. S. Air Force awarded to the Fairchild Engine and Airplane Corporation a contract which established Fairchild as the responsible directing agency of the Nuclear Energy for Propulsion of Aircraft (NEPA) Project. The purpose of the project was twofold: (1) to perform feasibility investigations and research leading toward the adaptation of nuclear energy to the propulsion of aircraft, and (2) to educate the aircraft engine industry in the field of nuclear science and its adaptation to aeronautical propulsion.

From midyear in 1946 until the early part of 1948, the NEPA Project in Oak Ridge, Tennessee, was the only agency actively investigating and developing nuclear propelled aircraft. In 1948 the Lexington Project, a group assembled by the Massachusetts Institute of Technology at the request of the Atomic Energy Commission to evaluate the possibilities of nuclear-powered flight, spent the summer in reviewing the existing knowledge of the subject and in appraising the problems to be solved and the prospects for success. The members of the Lexington Project, in their report, "Nuclear-Powered Flight" (Lex P-1), reached the conclusions that although success cannot be guaranteed, there is a strong possibility that some version of nuclear-powered flight can be achieved if adequate resources and competent manpower are put into the development; that intensive effort will be needed if a nuclear-powered aircraft is to fly within fifteen years; that integration of work on reactors, power plants, materials, and other components is essential for efficient progress toward the goal; and that a vigorous and realistic

aircraft reactor development program during the next few years should contribute to and benefit from other aspects of the Reactor Development Program of the Atomic Energy Commission."

The Lexington Report also made the recommendation that a strong development program on nuclear-powered flight be undertaken, if it is decided that as a national policy the high cost in technical manpower, fissionable material, and money can be justified.

On April 27, 1949, a conference was held at Oak Ridge, attended by representatives from the AEC Division of Reactor Development, the Office of Oak Ridge Operations, the Carbide and Carbon Chemicals Division, the Air Force, and the NEPA Project. The purpose of the meeting was to consider the part which the Oak Ridge National Laboratory could play in the Aircraft Nuclear Propulsion program.

As a result of the meeting, it was decided that the Oak Ridge National Laboratory would submit recommendations to the AEC regarding the participation of ORNL in the Aircraft Nuclear Propulsion program.

The ANP Program at ORNL

Numerous conferences between representatives of ORNL and the AEC took place in the following months in an effort to establish a suitable ANP program at the Laboratory. During this period the Laboratory increased its areas of cooperative effort with the NEPA Project. There had been cooperative programs of research and development in shielding and radiation damage whereby both ORNL and NEPA personnel worked together, making use of ORNL facilities. Several other projects at ORNL were so closely related to ANP that their major emphasis could be applied to this program with very little change in plans. The number of NEPA employees working cooperatively with groups at ORNL increased appreciably.

On September 1, 1949, the Oak Ridge National Laboratory received from the AEC instructions for the establishment of an Aircraft Nuclear Propulsion program at ORNL. On September 20, 1949, the Oak Ridge National Laboratory notified the AEC in writing of its willingness to accept the proposed ANP responsibilities and to carry out the program to the best of its ability with a priority second only to that of the MTR project.

Having accepted rather heavy responsibilities for the nuclear aspects of the Aircraft Nuclear Propulsion program, the Oak Ridge National Laboratory established an ANP Project and made plans for building up suitable groups of research and development personnel. One of the major initial activities planned for the early stages of the ANP program was the establishment of a Technical Advisory Board of outstanding scientists who would meet at ORNL during the summer of 1950 for an intensive session to evaluate the various aircraft reactor designs under consideration and attempt to establish certain basic design points from which an aircraft reactor could be developed. The Technical Advisory Board consisted of scientists whose competence in their respective fields was nationally recognized. Chosen for their ability to meet and cope with new problems in their fields as well as their known contributions to recent scientific advancement, these men were perhaps the most highly qualified group of specialists assembled for concerted effort on a special scientific project since the end of the war.

At the conclusion of their intensive review of the status of all factors influencing the feasibility of nuclear powered aircraft, the Technical Advisory Board issued a report reflecting optimism for achievement of supersonic flight. They recommended that an experimental

aircraft reactor be constructed in Oak Ridge as soon as possible. The work performed in the bulk shielding (swimming pool) facility had permitted significant refinements and improvements in shielding for an aircraft reactor and had given encouragement that further improvements could be realized. Because the shield for an aircraft reactor was recognized as being by far the heaviest component of a nuclear-powered plane, the progress toward reducing shield weight was a particularly important advance which greatly improved the outlook for feasibility of nuclear-powered flight.

With the initiation of a major effort on aircraft nuclear propulsion under the AEC at ORNL, the Air Force program at the NEPA Project in the K-25 area was closed out. In 1950 NEPA groups were brought into cooperative efforts with ORNL ANP groups and in 1951 the NEPA facilities at K-25 were closed, with most technical groups transferring to ORNL.

The ORNL idea of building reactor experiments was considered especially well suited for an aircraft reactor. This philosophy represented a return to the more normal industrial practice of building a pilot plant for thorough testing and further development before a major new type of installation is constructed. The Laboratory staff, therefore, formulated plans for constructing an experimental aircraft reactor to operate at a power level of 1000 KW, but otherwise to duplicate as nearly as possible the essential features of a full-scale aircraft reactor. This method of approach was based upon the belief that by constructing and operating a small-scale aircraft reactor, it will be possible to speed up the development program and thereby assist in establishing a feasible design which will lead to the construction of a full-scale test stand aircraft reactor.

Since no reactor operating at such high temperatures as those required for an aircraft reactor had ever been constructed and since liquid metal cooling in the reactor had never been utilized before, it was expected that the ultimate performance of a full-scale aircraft reactor could be pursued with more confidence if a pilot model

embodying these and other unusual features had been operated and thoroughly evaluated. The Laboratory gained much confidence in the ultimate operation of the MTR as a result of experiments with the full-scale MTR mock-up.

With the optimistic TAB report, the Laboratory obtained AEC support and initiated construction of an aircraft reactor experiment. A new building to house the aircraft reactor experiment was constructed close to the homogeneous reactor experiment building which at that time was also under construction. Installation of the components of the experimental aircraft reactor was started in 1953 and completed about the middle of 1954.

The ARE

Operation of the ARE took place in October 1954. The ARE operated at full design power for approximately 100 hours, as planned. Operation was completely satisfactory in every respect and provided a most convincing demonstration of the feasibility of a very high temperature fluid fuel reactor. Perhaps the simplest way to illustrate the magnitude of the engineering achievement reflected in successful operation of the ARE is to point out that while the reactor is in operation, every part of the fuel system is literally red hot, including the reactor core, piping, pumps, valves, heat exchangers, and all other components.

The fuel consisted of a molten mixture of UF_4 with NaF and ZrF_4. The fuel flowed in serpentine Inconel tubes through the reactor, surrounded by BeO blocks which acted as a moderator. The liquid fuel was pumped by means of a high temperature centrifugal pump and, after

leaving the reactor, went through a heat exchanger. The heat ultimately was removed by water, however, between the fuel system heat exchanger and the water heat exchanger was a stream of recirculating high-velocity helium which served as an intermediate heat exchanger fluid so that water and fuel could not mix in the event of a heat exchanger leak. The heat generated in the BeO reflector-moderator was removed by circulated sodium. This heat also was removed by water after passing through an intermediate helium heat exchanger. Due to the inherent self-regulating characteristics of the reactor, the only controls needed were a stainless steel regulating rod and three boron carbide safety rods. The active region of the reactor was a right circular cylinder, 33.3 inches in diameter and 35.8 inches high. The reactor was designed to operate at 1500 kw but actually ran at a peak output of 2500 kw. The exit temperature of the fuel from the reactor to the heat exchanger was about 1500°F; the inlet fuel temperature was about 1200°F. A total of about 90 megawatt hours of high-power, high-temperature operation was accomplished before the scheduled shutdown and dismantling of the ARE.

Research and Development for the ARE

Successful operation of the ARE represented major achievements in numerous fields of research and development activities. For example, the development of a molten salt fuel mixture that was chemically stable, non-corrosive, fluid over the proper range of temperature, and satisfactory in its heat transfer properties, was an outstanding accomplishment. The molten salt fuel had an attractive advantage over the aqueous fuel of the homogeneous reactor in that it

could be used in a reactor system operating with little increase over normal atmospheric pressure. The development of materials that would resist corrosion by red hot fluoride salt mixtures, and the development of methods for fabricating these materials into a reactor system that would operate reliably at 1500°F were metallurgical achievements of the greatest importance. The design of a reactor of this comparatively new type, to operate under these heretofore unheard of conditions, and the development of components to go into the reactor system were engineering problems of unparalleled difficulty which were solved with very encouraging success.

In operation, the ARE demonstrated again the advantageous features of fluid fuel reactors, including excellent nuclear stability, strong coupling between power demand and power level, and ease of operation and controllability. It is not exaggerating to say that the ARE was the most advanced reactor type the Laboratory had developed and, consequently, its successful operation represented perhaps the greatest achievement of the combined research and development staffs of the Laboratory.

The ART

Following the successful operation of the ARE, research and development efforts on a molten salt reactor for aircraft propulsion were intensified with the objective of building a 60 megawatt prototype reactor at the earliest possible date. The ARE building was enlarged and modified for the installation of the second experimental reactor. As the work progressed, an improved aircraft reactor design was developed incorporating the benefits of experience with the operation

of the ARE, plus other advances, particularly in materials and components development, achieved since the operation of the first reactor. Within two years, design of the ART was essentially complete, and orders for the fabrication of major components had been placed. An Engineering Test Unit, designed to be essentially a non-nuclear replica of the ART, was being constructed on a schedule slightly ahead of the ART so that the benefits of experience in the construction and operation of this test unit could be incorporated into the ART.

Early in FY 1958, the national program of aircraft nuclear propulsion development came under Congressional fire because of the high cost of the program and because changing military requirements made the achievement of nuclear powered aircraft less important as a national goal. The President of the United States ordered an extensive review of the current status of the program, re-evaluation of the military need for nuclear powered aircraft, and consideration of the additional time, effort and cost involved in developing and constructing a nuclear powered aircraft. Upon completion of this program review, the President cancelled major portions of the national ANP program and the Commission curtailed the aircraft reactor development program, eliminating the molten salt reactor phase entirely. With this decision the Oak Ridge National Laboratory ANP effort was changed in purpose and scope to provide direct support for the main lines of attack being pursued by Pratt Whitney and General Electric. Design of the ART was completed and "put on the shelf." The enlarged and modified aircraft reactor experiment building in which the ART was to be installed was put in standby for possible use to house a future reactor experiment, and the components and other materials on order for the ART were cancelled

and closed out in as orderly a manner as possible. The national ANP program was terminated on June 30, 1961. ORNL's research and development efforts in this field were shifted to the Space Power Program and to high temperature materials research.

Because it had already been shown that many of the most difficult technological problems associated with the molten salt aircraft reactor arose from the ultra high performance requirements and the stringent limitations on size and weight of the entire reactor system, it was apparent that the adaptation of this type of reactor to serve as a central station electric power plant would involve significantly less difficult technological problems.

MOLTEN SALT REACTOR

During the ORNL development of ANP technology for molten salt reactor systems, it became apparent that this type of reactor offered inherent advantages that made it attractive for electric power production. ANP work was pursued on a high priority basis with an urgency of scheduling that left no opportunity to divert effort into civilian power reactor adaptations. This opportunity became available, however, in 1958 when the decision was made to drop the development of a molten salt aircraft reactor and to concentrate ORNL ANP effort on support for the Pratt-Whitney and General Electric ANP efforts.

The ANP molten salt reactor groups were able to provide personnel for a much smaller scale effort to adapt ANP technology to achieve the goals of a civilian power reactor. The decision to continue development of molten-salt reactors for civilian power production came as a result of the fact that they, almost uniquely, combine the advantages of very high temperature, wide solubility limits, and low pressure in a liquid system. Because of their low pressure, the mechanical parts of the system were relatively uncomplicated. There was no need for complicated core structures, for control mechanisms, or an explosion containment vessel, and this basic simplicity, it was hoped, would offset the cost of the required heating and remote maintenance equipment. The molten-salt reactor capital costs thus might be nearly equal to those of other power reactors when compared on a heat generation basis. The higher thermal efficiency ($\sim$40%) of the salt system would then give an appreciable advantage in capital charges over aqueous fuel systems. The use of a fluid fuel had always been expected to achieve

lower fuel cycle costs through the elimination of fuel element refabrication costs. The very high thermal efficiency also tends to reduce fuel costs. Thus there was substantial reason to believe that the molten salt system could lead to over-all power costs which would be less than those of other, lower efficiency, nuclear power systems. It was these basic considerations which justified the original interest of ORNL in molten salt systems for civilian power, and which justified continued pursuit of the development effort.

Advances in Technology Under the ANP Program

ORNL developed a nickel-molybdenum alloy, called INOR-8, for containment of molten salts at high temperature. Out-of-pile INOR-8 thermal convection corrosion loops had been operated for thousands of hours at 1250°F peak temperature with no corrosion attack whatsoever. The INOR-8 alloy was considered to be a most promising structural material for molten salt reactors. Commercial production of the alloy had been achieved, with several firms willing to supply tubing, sheet and bar stock on a purchase order basis. ORNL had established INOR-8 that made it resistant to oxidation as well as corrosion, gave it good welding properties, and provided high-temperature strength. Wide interest had been shown in the alloy for other important high-temperature uses.

Phase studies of salt mixtures of LiF, NaF, BeF_2, UF_4, and ThF_4 had shown ranges of composition with suitable melting points for both blanket and fuel uses. Gas solubility studies indicated a basis for the continuous removal of fission product gases from molten-salt reactors. PuF_3 solubility studies established that plutonium can be burned in a molten-salt reactor.

Although reactor cost studies showed that it was economically feasible to use only the fluoride volatility process for chemical reprocessing and to discard the uranium-free salt with its contained fission products at each cycle, it was obvious that recovery of the salt with its Li^7 is desirable. Two salt reprocessing schemes affording recovery of the Li^7 had been demonstrated in small-scale laboratory work.

Remote maintenance experiments revealed two types of flange worthy of further development. Hand welding by remote manipulators was demonstrated in such operations as assembling and disassembling a molten-salt pump by using a remote manipulator.

A conceptual design study of a power reactor showed a feasible arrangement of a reactor cell equipped for remote maintenance, suitable methods of handling the off-gas, and arrangements for draining and otherwise handling the fuel. A cost study indicated power costs somewhat lower than those calculated for gas-cooled or water-cooled reactors.

GENERAL REACTOR TECHNOLOGY

The MTR, being the first truly high performance reactor constructed for purposes other than the production of plutonium, incorporated many features which were the result of extensive research and development performed at ORNL from the end of the war until the early 1950's. As research and development groups completed their responsibilities on the MTR project, many of them continued their fruitful work on a more general basis to benefit all reactors rather than a specific reactor. For example, the reactor control system developed for the MTR was the most advanced of its day and after its performance in the routine operation of the MTR had been assured, the reactor control development group undertook the improvement of controls for existing reactors at ORNL and the design of a modified MTR control system "package" which could be used with little change for other reactors of a similar type. Most swimming pool and other research reactors constructed since 1950 have made use of the generalized reactor control system developed by ORNL as an outgrowth of its work on the MTR controls. Similarly, studies of corrosion, heat transfer, shielding, metallurgy and radiation damage all began or were redirected as a result of the particular information needed for the development and design of the MTR. In all of these areas of research, work continued after the completion of the MTR project.

The work in general reactor technology was responsive not only to problems encountered in existing reactors but also to needs for information which would result in advances in technology. While many reactors had been built and operated during the war, much fundamental

information of great importance to reactor technology had never been developed because the urgency of wartime projects allowed no time for the research to be performed. In the early postwar years in particular a great deal of research effort was needed simply to provide fundamental data which had never previously been developed.

From the fundamental studies of this sort performed at ORNL, numerous continuing research and development programs were started to pursue areas of research and development where particularly difficult problems were encountered or where opportunities for advancement of technology appeared especially promising.

Removal Cross Sections

The phenomenological concept of an experimentally derived fast-neutron removal cross section, first introduced at ORNL, has constituted the practical basis of most fast-neutron shielding since its inception. In addition, the concept has given a physical picture of the attenuation process which in large measure has served as a guide in developing more recondite theories. The experimental data from which the removal cross sections have been derived were obtained at ORNL in the Lid Tank Shielding Facility.

Fission Gamma-Ray Spectral Measurements

The complex emission of gamma rays from fission represents a major source of these radiations in reactors and atomic weapons and gives rise to important problems in reactor shielding, reactor heating after shutdown, and civil defense shelter design. Before the ORNL measurements, only the spectra of the long-lived portion of these gamma rays, which represents less than 10% of the total energy release, had been studied. Now data are available for almost all time regions from a preliminary analysis of the ORNL data. The final analysis is still in preparation but the results of the preliminary analysis have already been applied very widely.

Proof of Feasibility of Breeding in Nuclear Reactors

The possibility of making more fuel than is consumed in nuclear reactors or "breeding" is dependent fundamentally on the ratio of neutrons produced to neutrons absorbed in the fuel material itself. This ratio called "eta" was in serious question in 1958 for thermal neutrons on the most promising reactor fuel, U^{233}, as a result of measurements made in the United Kingdom which gave much lower values than had previously been measured in the United States. The Oak Ridge National Laboratory undertook a broad program to settle the matter with finality and accuracy. In one series of experiments a sample of uranium was made to fission by thermal neutrons and the resultant neutrons were counted by the activation they produced in a very large surrounding bath of a manganese salt.

In another series of experiments, large containers of solutions of U^{233} or U^{235} salts were adjusted in concentration so that they just sustained a chain reaction. From this concentration and from estimates of the neutron leakage (which was kept low because of the difficulty of this estimation) it was possible to derive the value of eta for the two uranium isotopes. An even more accurate result was the comparison of eta for the two isotopes, which gave eta for U^{233} in terms of the better known eta of U^{235}.

In the third series of measurements, the effect of inserting small samples of U^{233}, U^{235}, and Pu^{239} on the reactivity of a small chain-reacting system gave an additional indication of the comparison of the three fuel isotopes in regard to their values of eta.

These experiments, plus considerable cross-calibration of samples with the British, served to reveal the reason for the original

discrepancy and to establish firmly that, at least on this fundamental score, breeding reactors with the thorium-uranium-233 cycle were feasible.

Neutron Absorption Cross Sections

The problem of xenon poisoning in the Hanford reactors very dramatically demonstrated the need for fundamental information on neutron absorption cross sections of all materials which might be used or formed in a nuclear reactor. At the end of the war, very little was known about the neutron cross sections except for the commonest structural materials. The most essential cross sections had been measured during the war at least for some neutron energies, but such work really had only been started. The Laboratory initiated a program of neutron cross section measurements in the graphite reactor using a "pile oscillator" to measure absorption cross sections to 5% accuracy for all elements which could be obtained in pure form. As this program progressed through the chemical elements which were available in pure form, it was extended to include measurements on the separated isotopes which had been prepared by electromagnetic processing at the Y-12 Plant. For research purposes, calutron isotope separators had been used to prepare significant quantities of separated stable isotopes of most elements that were amenable to electromagnetic separation. In the course of cross section measurements on the separated stable isotopes, it was found that zirconium exhibited a remarkably low neutron absorption cross section — much lower than the cross section which had been measured for chemically pure zirconium in the previous experiments. Further investigations showed that the hafnium which

invariably accompanies zirconium as an impurity which can only be removed with great difficulty has such a high neutron absorption cross section that even though it is present in zirconium only in small amounts, it made the zirconium cross section appear much higher than it actually was. The new cross section observed for hafnium-free zirconium was so low that it made zirconium appear most attractive for use as a reactor structural material. A process was developed at ORNL for removing the hafnium impurity from zirconium, and commercial production of hafnium-free zirconium was soon undertaken by industrial organizations so that low cross section zirconium became available in quantity for use in reactor construction.

Determination of the Sources of Radiation

In order to design shields for reactors accurately it is essential to know the amount and nature of the radiations produced. Many loose ends which were left in the wake of the wartime development program are now being cleared up. Among these are the radiations from very short half-life fission products, which are of great importance to the new circulating-fuel reactors (e.g., aqueous homogeneous, or fused salt reactors). Two experiments, one with a rotating uranium-laden belt and another with a fast pneumatic tube, have been carried out to measure these short-lived radiations.

Another poorly known quality is the number and energy per photon of the prompt gamma rays produced by a fissioning nucleus. A three-crystal spectrometer is currently being used to measure this component.

The energies of neutrons produced in fission, while quite well known for U^{235}, have been somewhat uncertain for U^{233}, which will be

produced from thorium in breeder reactors. Some research has improved this situation and more is anticipated in the near future.

Development of the Li^6I (Eu) Spectrometer

A thorough study of a fission reactor includes the cataloging by energy of the neutrons given off by the reactor. Further, the change in energy of those neutrons as they penetrate various materials must be known. The lack of a suitable spectrometer to measure neutrons in the range of from 1 to 14 Mev in the presence of the high gamma-ray fields encountered in and near reactors has in the past precluded such an investigation; therefore, a program to develop a satisfactory instrument was initiated. Two types of spectrometers were studied: (1) proton-recoil spectrometers, which, though useful, are limited by very low detection efficiency, and (2) scintillation spectrometers in which the energy released in a neutron-induced nuclear reaction is measured. Of the few reactions available for the second type of spectrometer, the $Li^6(n,\alpha)H^3$ reaction appeared the most promising, having both a large cross section and large energy release. A number of experiments were performed with single crystals of europium-activated Li^6I crystals and various monoenergetic sources. It was found that the response of these crystals was most satisfactory for crystal temperatures below about -140°C; therefore, a system was devised in which the crystal was cooled by liquid nitrogen. The experiments indicated that a Li^6I(Eu) crystal will be suitable for measuring fission-like neutron spectra above an energy of approximately 1.5 Mev.

Theoretical Reactor Research

A series of reactor codes for use with high-speed computers has been developed. The most elaborate of these, which is a consistent P_1 approximation to the Boltzmann equation, will enable an improved representation of the slowing down of neutrons in water. This code will be used to analyze the results of clean critical experiments at ORNL and should improve our understanding of the behavior of critical assemblies.

A greatly improved theory of resonance absorption has been obtained by including the effects of the statistical fluctuations of level widths and of Doppler broadening. Further progress has recently been made by including interference between potential and resonance scattering and by extending the theory to wide resonances.

Radiation Damage

A question which caused serious concern in connection with the operation of the Hanford reactors was whether radiation would cause changes in the materials inside the reactor in such a way that the performance of the reactor would be impaired. It had been recognized from the earliest studies with radioactive materials that radiation would cause changes in the physical properties of many materials and under some circumstances in their chemical behavior. Although the Hanford reactors were built and operated at high power without the benefit of information on the damaging effects of radiation, no serious operational difficulties were encountered. However, it was not known how long the reactor could be operated until the cumulative effects of radiation would create a dangerous situation, or whether radiation

effects would even be dangerous. Consequently, this was considered a very important field for research.

One of the earliest postwar research activities was the study of radiation damage in materials used in reactors. ORNL research groups were the first to undertake extensive studies of radiation damage making use of the graphite reactor facilities for irradiating samples of various materials. It was quickly recognized that post-irradiation examination of materials which had become highly radioactive in the graphite reactor required specialized facilities with heavier shielding and more extensive provisions for remote control operations. Lack of such facilities would require that radiation damage studies be limited to the low levels of radiation and the small samples which could be handled behind comparatively lightweight shields with the simple remote control tools that were then available. These limitations meant that new facilities would be required for the type of research that was needed to answer questions on radiation damage. Accordingly, a "hot laboratory" for physical and chemical measurements on irradiated materials was included as an adjunct to a Physics of Solids research laboratory in the construction program to provide the first permanent facilities at ORNL.

The radiation damage hot cells were completed and placed in operation during 1952. Thereafter the work proceeded on a much broader scale. It had been known that such physical properties as length, electric current resistance and elastic modulus would change quite significantly with radiation in many materials. The radiation damage research program yielded data not only on the way various materials reacted to radiation damage but also on the mechanisms by which

radiation damage occurs in various types of materials. As knowledge was gained in this new field, it became possible to select materials which would show least damage with exposure to radiation and to devise new materials with special characteristics to minimize radiation damage.

Aluminum Fuel Element Technology

Successful operation of the MTR in 1952 brought to fruition the advances in reactor design and materials development, and also demonstrated the technical feasibility of the High Flux Reactor concept. One important product of the materials effort was the development of an aluminum-base fuel element that embodied the following unique features: (1) exploitation of the compactness of nuclear energy by employing highly enriched fuel; (2) a major deviation from the classical natural uranium slug canned in aluminum toward an extended surface element to facilitate heat removal; and (3) dilution of the fuel in a suitable diluent and use of composite plate-type construction with metal-to-metal bonding to further maximize the heat transfer capability of the product. Practically all of the domestic and foreign pool- and tank-type reactors built for the purposes of education, radioisotope production, engineering testing, and research employ aluminum-base fuel units in their operation. In addition, the criteria established served as the underlying basis for the development of stainless steel and zirconium core components for the pressurized-water reactor programs of the Army and Navy. The fabrication facility at the Oak Ridge National Laboratory was the first integrated plant designed specifically for the manufacture of nuclear fuel and control components and served as the principal source of supply in the United States during the period 1948-1955, supplying the initial loading to some thirty-five different reactors.

An important adjunct of the materials effort for the MTR was the development of beryllium metal of suitable quality for initial reactor application in the reflector. Prior to that time, beryllium metal was

produced via the melting-casting-extrusion route, which resulted in a product with poor ductility when compared to beryllium produced via the powder route. Despite the reservations of many, the inherent advantages of the powder product were expounded and were finally used with the Brush Beryllium Company to produce massive pieces and with the Y-12 Mechanical Operations Division to machine the toxic metal into a variety of shapes to fit around beam holes and other perturbances in the reflector area. A key accomplishment was the development of a reliable deep-hole drilling technique at ORNL to provide the 3/16-in.-dia, 40-in.-long cooling channels in beryllium metal. This latter achievement permitted the use of massive beryllium pieces and thereby saved an estimated cost of $ 1.5 x 10^6 from the over-all cost of the beryllium reflector.

Thorium and Its Alloys

In 1950 an extensive review of the project literature covering the metallurgy of thorium was completed. This was used as a basis for starting a comprehensive program on the physical metallurgy of thorium and its alloys. A four-prong attack as follows was to be made:

1. An elucidation of the effect of impurities such as oxygen, nitrogen, carbon, and hydrogen on the mechanical and physical properties of thorium.
2. An extensive investigation of the mechanical properties of thorium and its alloys.
3. An alloy development program.
4. A fabrication and cladding program.

This program was quite successful and showed that thorium had a yield point similar to that of iron, which was due to the impurities in thorium. The first determinations of yield strength, tensile strength, modulus of elasticity, Poisson's ratio, and creep strength came out of this work. The extensive investigation of the alloys and fabrication procedures of thorium was extremely useful in the Materials Testing Accelerator Program; however, at the demise of this program, and because of the lack of interest in the U^{233} cycle, all work on thorium was stopped for several years until interest revived in breeder reactors utilizing U^{233} fuel and a thorium blanket.

Liquid Metal Technology

It was recognized very early in the study of reactor technology that liquid metals offered the most attractive heat-transfer media for high-performance reactors. This focused attention on the fact that virtually no information existed on the high-temperature compatibility of structural materials and various alkali metals. Techniques were developed by which the many facets of the problem could be studied and pioneering work was carried out on lithium, sodium, and NaK, and later the first work on rubidium and potassium was undertaken.

One of the most significant results from this work is the observed phenomenon of mass transport of metal and the effect of numerous variables on the rate of mass transport. These variables include solubility, maximum temperature, temperature gradient through the system, dissimilar metals, and impurities. Based on these results, it has been possible to select appropriate materials to contain various liquid metals and to predict the limiting operating conditions for the heat-transfer system.

The techniques and knowledge developed have continued to serve as the basis upon which subsequent research groups at other sites have formulated their programs.

Advances in Metallography

The utilization of color in metallography to a practical advantage in the nuclear field was pioneered at the Laboratory in the early 1950's. Techniques for the anodization of some metals and alloys were established to permit preferred orientation studies which otherwise would not have been possible by conventional methods. Metallography in color has also proved to be a tremendous asset in grain-size determinations and in the identification of microconstituents.

Vibratory polishing is another noteworthy achievement in which the Laboratory played a dominant role. This recent development has automated the arduous task of specimen preparation and is destined to contribute significantly in the field of remote metallography. Two supply houses are now marketing vibratory polishers which are patterned after the Oak Ridge National Laboratory model, and both give recognition to the Laboratory in their sales literature.

Stainless Steel Dispersion Fuel Elements

Another significant advance in the field of reactor materials was the development and manufacture of an experimental core loading for powering the SM-1, formerly designated as APPR.

The core consisted of enriched UO_2-stainless steel fuel elements with a high burnup capability and control components of B_4C dispersed in iron for neutron absorption. The loading, containing a burnable poison dispersed in the fuel, has performed exceptionally well by exceeding the design lifetime of 15 Mwy and achieving a burnup greater than 100,000 Mwd/ton.

More recently, two innovations have been developed for incorporation into the second core loading. These are europium-bearing absorber rods and control-fuel section with an integral flux suppressor to avoid flux perturbations at the absorber-fuel interface due to the "Wilkins Effect." The chief advantages in the use of the costly europium oxide are (1) radiation stability, by virtue of the fact that europium does not undergo and (n, α) reaction, and (2) extension of burnout lifetime because europium inherently generates a chain of four high neutron-absorbing isotopes in its decay chain.

Mechanical Metallurgy

There were three areas relating to mechanical metallurgy in which virtually no information existed prior to 1955. One was the effect of gaseous and liquid environments on creep, a second was an understanding of thermal fatigue, and third was the effect of multiaxial stresses on the creep, fracture, and ductility of metals. In order to study these variables, it was first necessary to develop new and unique testing devices. The popular analytical models were examined against data obtained from the new test conditions with the result that new and important information regarding the behavior of metals under general as well as nuclear-service conditions was revealed for the first time. Therefore, new analytical tools were provided the engineer and stress analyst to assist them in solving their design problems.

Nondestructive Test Development

One of the most persistent problems which beset the engineer is that of obtaining high-quality materials for constructional purposes.

The problem becomes more acute in nuclear technology because of the sensitivity of the public regarding reactor failures and because of the unusual nature of many of the materials which are used. The Non-destructive Test Development Group in the Metallurgy Division was formed to develop methods to ensure that all materials for use in the Oak Ridge National Laboratory would meet the necessary standards of quality. This need was recognized when industry was unable to inspect tubing to the high level of quality required by the Aircraft Nuclear Propulsion Program.

Research and development for both ultrasonic and eddy-current types of inspection have resulted in significant improvements in the sensitivity and interpretation of such tests. Thus, it has become possible to reliably inspect very thin-wall tubing and clad materials. In addition, techniques were developed for inspecting beryllium and graphite; two materials which present unusual inspection difficulties. Many of the techniques and equipment developments of this group have been adopted by commercial vendors. Remote inspection of the HRT core, accurate gaging of the HFIR fuel plates, and low-voltage radiography of beryllium are among the recent specific accomplishments of this group.

Reactor Materials Development

The primary purpose of reactor materials research and development at ORNL has been the selection and development of suitable materials of construction for reactor components. The requirements of high temperature reactors and converter or breeder reactors concerning mechanical strength and corrosion resistance, in addition to neutron

economy, are frequently beyond the capabilities of the usual engineering materials. Consequently, in addition to the conventional metallurgical assistance in the selection of materials for particular applications, the program comprises an intensive study of the metallurgical properties of elements of special interest (uranium, thorium, zirconium, titanium) and the development of fabrication methods for the unusual shapes and combinations of materials required in nuclear reactors. As much of the work as possible is done under controlled and simple conditions leading to an understanding of the phenomena concerned.

Homogeneous Reactor Materials

In homogeneous reactors reliability of structural materials under rather severe operating conditions is of particular importance. Metallurgical development and testing of materials, accordingly, has received much attention, emphasizing the development of better characteristics of corrosion resistance, strength, ease of fabrication, and weldability. The scope of the metallurgical effort extends from the development of new and improved alloys through the establishment and control of suitable metallurgical processing methods (fabrication, heat treatment, and welding) to the inspection and testing of the final components of the reactor system.

Materials explored in this connection have been stainless steel, zirconium, and its alloys, and titanium. Of outstanding importance has been the development of welding techniques that do not require use of dry boxes, thus permitting construction of large vessels, and the discovery of the usually good corrosion resistance alloys of the Zr-Nb

system in uranyl sulphate under irradiation. It has also been found that the usual correlations in mechanical properties determined by conventional means do not apply to the hexagonal metals.

Molten Salt Reactor Metallurgy

The successful construction and operation of a high-temperature reactor for aircraft propulsion depends upon the solution of associated materials problems. The purpose of the metallurgy group is to determine the suitability, properties, and methods of fabrication and handling of structural, moderator, fuel carrier, control rod, shielding, and coolant materials for use in such reactor systems.

The very high heat fluxes involved require thin-walled metal in all heat exchange situations. The use of such thin-walled metal is opposed by the combination of corrosion and mass transfer by the high temperature liquids (fused fluorides and liquid metals) employed, by the metallurgical defects occurring in commercial materials, and by the mechanical stresses imposed. As a consequence the metallurgical work has proceeded chiefly along the following lines:

(1) The mechanism of corrosion and mass transfer in fused fluorides and liquid metals has been investigated and the effect of additives to the liquid and alloy additions to the container materials determined. Among the commercially available materials Inconel and Hastelloy B have been found to be best suited but have certain deficiencies which have been overcome by the development of a new alloy INOR-8 (Ni-Mo-Cr-Fe).

(2) Continued progress has been made in the development of new methods and materials for welding and brazing heat exchangers and radiators for high temperature service. An outstanding new development has been in methods for brazing cermets to metals for valves and bearings.

(3) Entirely new and sensitive methods for non-destructive testing of small, thin-walled tubing have been devised and applied in the inspection of all material used in test assemblies. These are of the ultrasonic- and eddy current-types.

(4) An enormous amount of data has been obtained on the high temperature creep and stress rupture properties of Inconel and other materials in various environments, particularly under multiaxial and cycling stressing. Application of these data should permit better design of component parts.

In addition, two special high-temperature shielding materials have been developed for this reactor: B_4C-Cu for the neutron shield and WC-Hastelloy C for the γ shield.

Heterogeneous Reactor Materials

The feasibility of exploiting the compactness and low fuel transportation requirements of nuclear energy to power plants to provide economical power in remote locations, such as the arctic, has been demonstrated by the construction and successful operation of the 10 mw, pressurized plant at Fort Belvoir, Virginia. The ORNL-designed plant is powered with highly enriched UO_2 incorporated into stainless steel components of compact-core-design. Control of the high

uranium-investment core is accomplished by the employment of a bank of seven control rods, and the utilization of a burnout poison uniformly dispersed in the fuel.

Metallurgical assistance required in support of the program included material evaluation and selection, development of specifications and procedures for powder-metallurgy processing, cladding and the joining of the composite fuel plates into an integral assembly by brazing, preoperational corrosion, hydraulic and mechanical testing, as well as a final demonstration of performance under irradiation. A similar program was carried out in the development of the enriched boron- 10- bearing stainless steel rod for the absorber section of the control rod assembly.

The initial core loading was fabricated in accordance with the specifications developed and adopted, and delivered to the site for startup of the reactor.

Plate-Type Fuel Element Development

In support of the Atoms for Peace Program there was an urgent need to develop a highly reliable aluminum plate-type fuel element which utilized 20% enriched U-235 materials for service in foreign reactors. Three types of materials have been investigated in an attempt to arrive at an optimum serviceable and economical fuel element. These combinations include fuel plates containing Al-U-Si alloy, U_3O_8-Al cermet, and UC_2-Al cermet. Procedures have been established for melting and casting highly concentrated U-Al alloys and for fabricating these into fuel elements. The UAl_4 compound in such alloys is completely suppressed by a 3% Si addition resulting in improved

workability. Highly concentrated U_3O_8-Al and UC_2-Al cermets have been found to be more compatible than UO_2-Al and UC-Al cermets.

For the Special Power Excursion Reactor Test requiring stronger and more rigid fuel elements of the MTR-type, it has been demonstrated that age-hardening aluminum alloys 6961 and 6061 could be substituted for 1100 aluminum with the desired results.

There also have been developed several other new composite plates which appear promising for fuel or control components:

uranium clad with zirconium

thorium clad with zirconium

thorium clad with aluminum

dispersion of B_4C in copper clad with stainless steel

dispersion of boron in iron clad with stainless steel

dispersion of Eu_2O_3 in iron clad with stainless steel

REACTOR FUEL PROCESSING

Wartime Program

The strong tradition in chemical engineering begun during the war years when the entire Laboratory was operated as a pilot plant for the Hanford, Washington, plutonium-production plant, influenced the AEC in establishing ORNL as the center of chemical and chemical engineering research in 1948. The bismuth phosphate process developed at the University of Chicago Metallurgical Laboratory for separating and purifying plutonium from the reactor fuel in which it is produced was carried through successful pilot-plant stages at ORNL. The first gram of pure plutonium was produced, separated, and purified here. In addition, the Laboratory had wartime responsibility for producing thousand-curie amounts of the radioisotopes barium and lanthanum, for use at Los Alamos as a very strong radiation source. Process-improvement research and development on these processes continued through the war years, even after successful performance had been demonstrated.

Postwar Development — The Redox Process

The successful operation of the Hanford plutonium-production facility attests the contribution of wartime research at ORNL to the atomic energy program. Soon after the war, ORNL was requested to use its pilot-plant facilities to test and improve the Redox process developed at Argonne National Laboratory for separating and purifying uranium and plutonium by solvent-extraction methods. The Argonne process showed great promise of being much more simple and economical than the bismuth phosphate precipitation process, which served to separate

and purify only plutonium, but it yielded uranium of insufficient purity. ORNL modified the process and demonstrated on pilot-plant scale how a sufficiently pure uranium product could be produced. A new chemical plant to use this process was constructed at Hanford in 1952 at a cost of $60,000,000.

"25" Process

The Materials Testing Reactor at Arco, Idaho, required a new chemical process to recover the highly enriched uranium from its used uranium-aluminum alloy fuel elements. ORNL chemists and chemical engineers developed a process, called the "25" process, and demonstrated it in the pilot plant. The Laboratory was responsible for the design of the Idaho chemical plant and for supervising its construction and initial operation. The philosophy of direct maintenance was used in this plant for the first time, all previous chemical plants having used remote-control maintenance. The $28,000,000 plant was completed in 1952. The plant and process were sufficiently versatile to permit fuel elements from the EBR, STR, or other reactors to be processed routinely, as well as MTR fuel elements.

TBP Process (Metal Recovery)

During the war and afterward, large amounts of radioactive wastes containing many tons of uranium were stored in tank farms at ORNL and at Hanford. In 1949, ORNL undertook the development of a process for the recovery and purification of uranium from its metal wastes in the tank farm. The process that was developed utilized tributyl phosphate (TBP) as a solvent for extracting uranium, and was later adapted to processing Hanford wastes. Still later, the process was further modified

to permit the recovery of plutonium as well as uranium from ORNL wastes. The TBP metal-recovery process was adopted at Hanford and installed at a cost of $35,000,000 in an unused facility originally built for the bismuth phosphate process, which was replaced by the Redox process. A new waste-processing facility, the Metal Recovery Facility, was constructed at ORNL at a cost of $650,000 to use the TBP process. The new facility was first used in the Commission's high-priority program for separating uranium and plutonium from Chalk River reactor fuel elements purchased from the Canadians and was later used for recovery of the uranium stored in waste tanks at ORNL. The Metal Recovery Facility was also used for the recovery of uranium and plutonium from the Brookhaven National Laboratory reactor and Argonne National Laboratory's CP-2 and CP-3 reactors.

Purex Process

On the basis of the work carried out in developing the TBP process for recovering uranium from waste solutions, it was decided to investigate the possibilities of using this type of process for recovering, separately, plutonium and uranium from Hanford plutonium-production metal. It was shown in the summer of 1949 that such a process was feasible, and that it probably would be more economical than the other processes which had been or were being developed at that time. In addition to uranium and plutonium, the Purex process also isolated the fission products, thus greatly simplifying radioactive waste storage problems. In 1950, the process was selected as the one to be used by du Pont at the Savannah River Project.

The major chemical development work for Savannah River was done at ORNL. Some supplementary studies were carried out at Knolls Atomic Power Laboratory and at Argonne National Laboratory. KAPL developed the pump mixer-settler to be used at the Savannah River Project as the contacting device.

In addition to the main-line solvent-extraction process, called the Purex process, ORNL was responsible for the development of an ion-exchange Plutonium-isolation scheme, a process for recovering plutonium from miscellaneous metallurgical wastes, a method for recovering nitric-acid from the solvent-extraction wastes, and a process (fumeless dissolving) for removing radioactive fission gases from dissolver fumes, permitting the recovery of the oxides of nitrogen as nitric acid which could then be reused.

The Purex process was used in the two separations plants at Savannah River in 1954 costing about $75,000,000 each. Also, a Purex plant was constructed at Hanford at a cost of about $78,000,000 in 1956, using a slightly modified process.

Thorex Process

The Thorex process separates protactinium, uranium-233, and thorium from fission products and from each other. The process is of interest for the isolation of fissionable uranium-233 for weapons development and for power-breeder reactors which are expected to employ thorium blankets. It will make it possible to return irradiated thorium to production channels, to provide quantities of protactinium-233, and to provide a source of isotopically pure uranium-233.

The process was demonstrated at ORNL on a pilot plant and semi-production scale in 1954.

Interim-23 Process

The Interim-23 process isolates uranium-233 from irradiated thorium and associated fission products.

The INT-23 process was developed for the production of kilogram quantities of uranium-233 required for weapons testing. An existing development facility was modified to provide adequate shielding, and supplementary shielded waste tanks were constructed to receive the radioactive thorium wastes. Slugs irradiated at Hanford were shipped in special shielded carriers to ORNL and processed at a rate of 70 kilograms of thorium per day.

A total of about 2.5 kilograms of uranium-233 was isolated in 1952.

Slurrex Process

The Atomic Energy Commission's uranium raw-materials purification program was expanded in 1951, and a search was made for an alternate to the ether extraction process being used by Mallinckrodt Chemical Works. In 1950, ORNL was asked to make a preliminary evaluation of various solvents which could be used for the recovery and purification of uranium from ore concentrates. Ethyl ether had been used for this purpose, but a less hazardous and less volatile solvent was highly desirable. TBP proved to be the most attractive alternate.

The Slurrex process using TBP as solvent was developed in conjunction with Mallinckrodt Chemical Works and Catalytic Construction Co.,

and demonstrated at ORNL. In addition to the use of a less volatile solvent, the process permitted the use of ore-digest slurries as feed material in pulse-column contactors. The process was adopted by The National Lead Company for use in the Feed Materials Production Center at Fernald, Ohio. A plant costing approximately $20,000,000 was constructed.

Excer Process

The uranyl nitrate product of extraction processes, such as Purex and Redox, must be converted to uranium tetrafluoride as the first step of its preparation for recycle through the gaseous diffusion plant at K-25. This was accomplished by costly reduction with hydrogen and hydrogen fluoride. The Excer process converts uranyl nitrate to uranium tetrafluoride at a much lower cost.

Fuel and Blanket Processing

Since 1953, studies of reactor fuel processing have included the continuing development of the Thorex process for uranium-233 recovery and the initiation of the Fluoride Volatility process for uranium-235 recovery.

Thorex studies were made to evaluate the effects of irradiation level, up to 4000 Mwd/t, and decay times as short as 30 days. Various solvent extraction procedures were studied to achieve maximum separation with a minimum number of process operations. During the development work approximately 80 kg of uranium-233 was separated along with 45 metric tons of thorium. The separation of protactinium-233 as a specific product was an early objective of this program that was not achieved. However, in the runs on short-decayed material, isotopically pure uranium-233 resulting from the decay of the protactinium was recovered by reprocessing the waste.

The Fluoride Volatility process used the high volatility of uranium hexafluoride to separate the uranium from less volatile fluoride salts. At ORNL this process was developed for the recovery of uranium-235 from molten salt reactor fuels and from other reactor fuels soluble in molten salts. In the initial phase of the pilot plant test 56.6 kg of uranium-235 was recovered from the ANP molten salt fuel. This represented a recovery of greater than 99.5% with a decontamination factor of greater than 10^5.

The 25-TBP process was developed for the recovery and decontamination of enriched uranium-235 for use at Savannah River. This was carried through experimental chemical and engineering development along with a detailed design study.

Processes for the continuous removal of fission and corrosion products from the Homogeneous Reactor fuel system were tested on a pilot plant scale. Some of the fission and corrosion products precipitate in the fuel solution. These precipitates were continuously removed by centrifugation in a hydraulic cyclone separator. Iodine was removed by chemical absorption on silver while the rare gas fission products, xenon and krypton, were adsorbed on a charcoal bed. Homogeneous Reactor studies also included the definition of the properties required to produce a satisfactory thorium oxide slurry to be used as a blanket to produce uranium-233. The studies involved chemical and engineering development to produce thorium oxide slurries with specified properties, to develop techniques to evaluate the behavior of these slurries under conditions of high temperature, pressure, and radiation, and finally to develop large-scale procedures for production of satisfactory thorium oxide. Results of large scale pumped loop tests and in-pile bomb experiments indicated that this work produced a satisfactory thorium slurry. The Thorex process can be used for the recovery of the uranium-233; however, alternate procedures were studied to achieve a more direct recycle of both uranium and thorium.

The development of reprocessing methods for nuclear power reactor fuels has been a major effort. The principal work covered dissolution procedures for the stainless steel- and zirconium-clad fuels. The resulting solutions were converted to forms compatible with the established solvent extraction purification technology. The Darex process has been developed for stainless steel-clad fuels, and it is being considered for use at Hanford and Idaho.

An alternate process for stainless steel jacketed fuels, called Sulfex, dissolved the stainless steel jacket with sulfuric acid. The fuel material was then dissolved directly in nitric acid. This reduced the quantity of material going through solvent extraction and may simplify the waste disposal problem. A similar process for zirconium jacketed fuel, Zirflex, used $NH_4F \cdot NH_4NO_3$ solution. The coating removal step was again followed with the nitric acid dissolution of the core. It appeared that both Sulfex and Zirflex could be carried out in the same equipment. These procedures were particularly applicable to fuels with uranium and thorium oxide cores such as Commonwealth Edison, Yankee Atomic, and Consolidated Edison.

Additional studies with the Darex Process showed that complete dissolution of the stainless steel fuel with a mixture of HCl and HNO_3 was possible, and that chloride ion could be removed in either a batch or continuous system. The Zircex Process, in which HCl gas is used to remove zirconium from clad or alloy fuels, was operated successfully on unirradiated zirconium-bearing fuel elements to give a zirconium-free solution of uranium.

The Thorex process development program at Oak Ridge National Laboratory has been completed; the final phase of this study proved the feasibility of the tributyl phosphate solvent extraction process in a 200 kg thorium/day pilot plant. Other studies during this period were concerned with processes for separation of the transuranic elements. Processes were developed for the transuranic elements. Processes were developed for the separation of neptunium-237 from irradiated enriched uranium-235 fuel elements, from irradiated natural uranium by modification of the Purex process, and from the fluorination ash of recycled

Hanford and Savannah River uranium. In pilot plant tests of the processes about 400 grams of neptunium-237 were separated. The separation of another transuranic element, americium-241, has been studied. Pilot plant tests on its separation from plutonium metallurgical waste has produced about 40 grams of americium.

RADIOACTIVE WASTE DISPOSAL

Control of Waste Off-Gases

The handling and disposal of high-level radioactive liquid wastes presented major problems of air contamination. Airborne radioactivity must be removed from the air or off-gases, trapped in some way, and contained sufficiently to prevent its being dispersed to the atmosphere. Information and techniques from earlier basic investigations of aerosols were utilized in the development of methods for the removal and containment of off-gas contaminants.

A pilot scale aerosol entrainment well was developed and operated to investigate the evolution of radioactive aerosols from hot and boiling radioactive solutions, and to study the effectiveness of sand and other filter media for wet aerosols. The decontamination factor by evaporation (condensate vs liquid in well) ranged from about 10^4 to 10^6. Experimental evaluation of sand and glass fibers showed that these filter media when dry provide decontamination factors greater than 10^5 per foot of filter; but when the filter media became saturated with condensable vapors, the decontamination factor was reduced to 10^3 per foot or less.

For decontamination of high temperature sintering off-gases, a simple multibed filter system was developed. The gases flowing at a velocity of 0.1 centimeters per second were passed through a deep sand bed and then through soda-lime and activated carbon. More than 99.99% of a particulate matter was removed in the dry portion of the sand bed. Nitrogen oxides, iodine, and other reactive gases were absorbed and

retained in the soda-lime or activated carbon. A laboratory model of a recirculating caustic-absorber system for off-gas contaminants from sintering processes was developed and evaluated. In this system aerosols and reactive gases were retained in a caustic solution and nonreactive gases held in a separate container.

Process Waste Water Treatment Plant

The process waste system at ORNL is made up of a network of underground pipes, tanks, and surface collection basins which collects the large volume (750,000 gallons per day) of waste water throughout the Laboratory area. This large volume, including water from laboratory floor drains, operating areas, and process cooling, is normally low in radioactivity but is potentially subject to greater contamination from accidental spills or faulty equipment. In the original system the waste passed through an excavated settling basin and, after sampling, was discharged to the Clinch River through White Oak Creek.

Built during World War II with only minimum monitoring and control facilities, this system has contributed the bulk of the contamination discharged into the Clinch River by the Laboratory. In earlier years the discharge of radioactive materials to White Oak Creek averaged 2 to 5 curies per day which, diluted by normal river flow, did not exceed permissible concentrations of radioactivity for drinking water. With improved monitoring and control, and more complete segregation of intermediate-level effluents for soil disposal, the quantity of radioactivity released to the river was reduced to approximately one-half curie per day. Even this ten-fold reduction in radioactivity released to the Clinch River was not considered adequate on a long range basis

because the system lacked the necessary hold-up capacity to control accidental or emergency discharges. It was considered essential to guarantee that even in the event of a major accidental release of large quantities of radioactivity in the Laboratory, the radioactivity reaching the Clinch River could be kept at very low levels. For these reasons treatment of the waste before release to the river was considered essential.

A new process waste water treatment plant with a capacity of 500,000 gallons per day was added to the waste system in 1957. An 800,000-gallon equalization and hold-up basin was provided for the incoming wastes that require treatment. A radiation monitoring station was installed to actuate an automatic valve which would divert the more radioactive flows to the equalization basin and bypass the uncontaminated wastes directly to White Oak Creek without treatment.

This plant utilized a modified lime-soda softening process, which proved effective in the removal of strontium by coprecipitation. Operating experience showed the plant to be capable of removing 90% of the radioactive strontium and rare earths; and in case of abnormally heavy contamination the effluent can be recycled for further treatment to obtain higher efficiencies if necessary. This waste treatment plant is unique in that it is the first to be designed specifically for the removal of radioactive strontium from large volumes of low-level wastes. After treatment the wastes are monitored, sampled, and discharged to White Oak Creek.

Soil Disposal of Intermediate-Level Wastes

The continually increasing quantity of intermediate-level wastes at ORNL required either an expansion of existing tank storage facilities

or a change in disposal methods. Extensive laboratory research and field investigations were carriedout to determine the feasibility of soil disposal of these wastes. Consequently, an experimental soil disposal system consisting of three one-million-gallon surface pits was constructed in the local shale formation.

The radioactive chemical-process wastes with other highly radioactive liquids were at one time collected in the underground storage tanks, reduced in volume by evaporation, and hauled to the disposal pits by truck. A significant change in the system was the construction of a 1 1/2-mile pipeline and pumping station by which the wastes are pumped directly to the disposal pits. Pumping wastes to the pits eliminated the radiation hazards of hauling the wastes in trucks.

Through December 1957 a total of 8.5 million gallons of waste containing 68,000 curies of Cs^{137}, 19,000 curies of Ru^{106}, and lesser amounts of Sr^{89}, Sr^{90}, Co^{60}, Sb^{125}, and the rare earths were discharged to the pits. The predominant stable ions in the waste were sodium and nitrate. The dispersion of wastes seeping from the pits was studied extensively. The quality and extent of waste movement through the shale formation, primarily through the weathered zone and along the strike, was correlated with separate and total pit inventories, surface runoff, and basal ground-water flow. The purpose was to determine the safety factors and probable period of usefulness of the present pits, and to develop more satisfactory methods of using the capacity of soils for disposal of radioactive wastes.

Fixation of High-Level Liquid Wastes by Sintering

Potentially one of the best means of ultimate disposal of radioactive wastes is to fix the hazardous fission products in a relatively insoluble solid form and store them under conditions that will assure minimal dispersion for several hundred years. Ceramic materials and techniques have been used in the development of methods for incorporating fission products in a mixture of waste chemicals, shale, limestone, and soda ash which, when dried and heated to a high temperature, will produce a durable nonleachable sinter. The radioactive sinter may be stored near the earth's surface above the ground-water table or deep underground, as in dry caves or cavities in natural salt formations.

The self-fixation process, which has been studied in the laboratory and in field experiments, would utilize the heat of radioactive decay to evaporate the water from the waste mixture, decompose the unstable compounds, and raise the temperature of the ceramic sinter to about 800 to 900°C. The studies already completed, using simulated acid aluminum-nitrate wastes and artificial heating, have determined the thermal requirements of sintering. They have also provided data on the engineering problems involved in large-scale field experiments which would utilize the energy in a high-level waste to provide the heat for sintering.

A large number of ceramic materials and various mixtures have been investigated for their effectiveness in fixing the fission products present in waste solutions. From these studies it appears that illitic clay materials are most promising for the retention of cesium which is one of the most leachable long-lived radioelements.

Disposal of Wastes in Natural Salt Formations

Ultimate disposal of highly radioactive wastes by storage underground in natural salt formations is considered a promising method because of the impermeability, tensile strength, and thermal conductivity of salt, and its widespread distribution and abundance in areas of a low degree of seismicity in the United States. A major concern in salt disposal is the structural stability of the cavity or opening in the salt formation.

The physical properties of salt are influenced by pressure, temperature, radiation, and chemical interactions. The degree to which the desired structural properties and stability of waste storage facilities would be affected by liquid waste chemicals, pressure, heat generation, and radiation will in large part determine the role that salt is to play in the disposal of radioactive wastes. Preliminary investigations have shown that the solubility of sodium chloride in the already highly salted waste solutions varies from 50 to 90% of its solubility in distilled water. The plasticity of salt is increased by higher temperatures and pressures. The heat generated in the wastes and heat losses by natural conduction in salt as a function of fission product concentrations have been calculated and are to be checked experimentally. The effects of prolonged exposures to intense radiation are being studied.

Disposal of Liquid Wastes in Deep Wells

The disposal of liquid radioactive wastes at depths of several thousand feet through injection wells has great potential if the technical problems involving waste pretreatment, plugging of the

injection well, and reactions of specific wastes with the connate waters or constituents of the aquifer matrix can be solved. Large volumes of oil-field brines have been desposed of cheaply and safely by this method.

Extensive studies must be completed in order to develop deep well disposal practically and safely. The natural movement of ground water in most deep porous formations can be shown to be a few feet per year, far too slow to be hazardous. The wastes and typical disposal formations must be studied to determine the danger of plugging from suspended matter or precipitates, the possibility of excessive temperatures due to adsorption of fission products near the point of injection, the pressure increases due to injection of wastes into the formation, and the preliminary treatment of dilution of the wastes that may be essential.

Studies have been carried out to characterize the aged, neutralized, and filtered wastes that may be amenable to deep well disposal and the types of geological formations that will be suitable.

PHYSICAL RESEARCH

Physical research activities at ORNL include major efforts in physics, chemistry, metallurgy, and solid state physics. In each of these major fields, ORNL's responsibilities have increased steadily as the Laboratory has pursued promising fields of research which were particularly suited to investigation with the Laboratory's facilities and scientific staff capabilities. The flexibility that ORNL has always sought to maintain in its research organization and research facilities made it possible for the Laboratory also to undertake research on new problems of particular urgency as they arose in various Commission programs. While the scope of research efforts in each field is broad, nevertheless there are areas in which ORNL has specialized because of its own interests or program needs, because of the fruitfulness of the work, or the particular suitability of facilities available at ORNL.

In the field of chemistry, ORNL's major efforts have been on high temperature inorganic systems of interest to reactor technology and on the fundamental chemistry of separations process for nuclear energy materials. In physics, ORNL's work is mainly neutron physics and low energy accelerator physics. In the field of metallurgy, the development of plate-type solid fuel elements and the development of new structural material alloys have occupied major attention, along with the study of welding techniques and other factors involved in the fabrication of materials of particular interest.

Major efforts in the solid state physics have been devoted to the study of radiation effects in pure metals and special materials such as the semi-conductors in which radiation effects can be studied

especially fruitfully. ORNL solid state research has placed emphasis on temperature effects such as the annealing of radiation damage and the low temperature "freezing in" of radiation effects. Specific activities in these fields will be discussed in the following sections.

PHYSICS

The wartime physics program dealt with research and development related to reactor design and operation. Since then, the program has broadened to include basic studies which exploit reactors as neutron sources, as well as applied studies of importance in reactor development. In the first category are neutron diffraction and radiation damage to semiconductors; in the latter category are reactor criticality experimentation, shielding research, and measurement of such important cross sections as that of radioactive xenon-135.

Xenon Experiment

At the startup of the first Hanford reactor, it was found that the pile "went to sleep" after a few hours of operation, and "woke up" again a few hours later. This peculiar performance was found to be due to the buildup of a radioactive fission-product poison, identified as xenon-135, and its subsequent decay. The existence of this poison has affected the design of all high-powered reactors that have subsequently been contemplated, and it was clearly of vital interest to learn with accuracy the neutron absorption cross section of xenon and how the cross section varies with neutron energy.

At ORNL, the xenon experiment was performed in 1950 over as much of the energy range as could be covered by the use of a neutron crystal spectormeter. The source of the xenon-135 was solid fuel slugs which were processed in a hot cell to obtain fission product xenon for study. Later work was conducted to repeat and extend the measurements by making use of the mechanical velocity selector. The second experiment

utilized thousand-curie quantities of radioactive xenon drawn off from the aqueous fuel of the homogeneous reactor experiment. The xenon was purified in chemical hot cells and compressed into a small cell about one cubic centimeter in volume. The tiny capsule of xenon was placed in a lead shield weighing twelve tons, and moved to the LITR, where the sample was taken from the shield and placed in the neutron beam ahead of the velocity selector. This machine chops the neutron beam into intermittent bursts. The neutrons travel a distance of 40 feet, and in doing so they sort themselves out according to their velocities, so that some arrive at the detector earlier than others. The absorption of the neutrons in the xenon was determined as a function of energy and accurate data were developed for use in reactor physics. All of this work had to be done within the time permitted by the nine-hour half-life of xenon-135.

Reactor Physics

It was recognized that, to facilitate studies like these, a research reactor providing a higher neutron flux than any in existence was needed, so the Laboratory in 1946 embarked on a program of design and development for a "High-Flux Pile." Out of this work came the basic information used in the design of the Materials Testing Reactor (MTR), for which ORNL was given responsibility under the AEC's reactor-development program. The MTR, designed in cooperation with Argonne National Laboratory and constructed at Arco, Idaho, under ORNL supervision, incorporated many new features that have been adopted for other reactors. The basic idea of the MTR has been used in the Low Intensity Test Reactor, originally a mock-up of the MTR, the Bulk Shielding Reactor,

the Oak Ridge Research Reactor, the Tower Shielding Reactor, the Submarine Thermal Reactor, and low-cost reactors for university research.

To assure continued advancement in reactor technology, the Laboratory established programs of research and development in instrumentation and controls, critical-mass studies, reactor shielding, and radiation-damage physics. In shielding and critical-mass studies the Laboratory performs major work for all AEC installations. These applied-physics programs supplement the studies in basic physics, exploiting the information already available and making important new contributions in the specialized fields. The control system developed at ORNL for the MTR received wide acceptance and was used almost without change in many other research reactors.

Neutron Physics

Basic physics research since the war has been primarily concerned with studies of neutrons and their interactions with matter. Out of this research have come many significant contributions to nuclear science.

From theoretical considerations, it was predicted that neutrons should be radioactive and decay by the emission of an electron to form a proton. At ORNL, evidence of neutron decay was found in a complex experiment in which a beam of neutrons from the graphite reactor was analyzed to detect the electrons and protons resulting from meutron decay. It was definitely established that neutrons decay in the manner predicted, and the half-life associated with the neutron-decay process was determined to be about 15 minutes.

A program of measuring the neutron absorption cross sections of all elements and isotopes was carried out first in the rather simple pile oscillator in the Graphite Reactor and later in more sophisticated ways using neutron velocity selectors, accelerators and improved instrumentation. The discovery of the low cross section of pure zirconium was the starting point of an intensive program of development that led to the use of zirconium-clad fuel elements in the submarine reactor and its application in other reactors.

Neutron diffraction studies of crystal structure have become an exceedingly powerful tool for determining how atoms are arranged in crystals. The neutron diffraction technique overcomes some limitations of X-ray and electron-diffraction methods and permits the structural analysis of materials that cannot be studied satisfactorily by any other method. One of the earliest contributions of the neutron diffraction studies was to establish conclusively the crystal structure of ordinary ice — a matter of some controversy which could not be settled by previously known experimental techniques.

A program of low-temperature physics research was initiated to show the effects of polarization upon the interactions between neutrons and atomic nuclei. By strong magnetic fields at temperatures within a tenth of a degree of absolute zero, atomic nuclei can be polarized, and their reactions with a beam of polarized neutrons can be studied. Differences between "polarized reactions" and those in which the neutrons and target atoms are not polarized were demonstrated. In particular, it was observed that the absorption of polarized neutrons in a polarized target is somewhat less when the polarization of the beam and the

target is antiparallel than in the same situation without polarization, and somewhat greater when the polarizations are parallel.

Charged Particle Physics

Studies of nuclear reactions were extended to include those involving bombarding particles other than neutrons. The Laboratory designed and constructed three cyclotrons of 44-inch, 63-inch, and 86-inch sizes, respectively, for use in its nuclear physics research program. These nuclear particle accelerators, coupled with two Van de Graaff accelerators which develop potentials of 2.5 and 6 million volts respectively, permit the study of nuclear forces and nuclear reactions as well as the production of certain radioisotopes obtainable in no other way.

The 86-inch cyclotron produced the world's most intense proton beam, four times higher than that of any other cyclotron, and was used in studying the basic physics of proton-induced nuclear reactions, in supplying a source of fast neutrons for biological research, and in producing certain isotopes that cannot be produced in nuclear reactors. This cyclotron proved to be the most economical machine for making cyclotron-produced isotopes.

The 63-inch cyclotron was used for basic physics research on the reactions between accelerated heavy particles and light element targets. It was this cyclotron in which the fusion of nitrogen atoms was demonstrated.

The 44-inch cyclotron was used primarily for technological development work, testing new ion sources, new beam-focusing methods, and methods of increasing the intensity of the accelerated beam.

These ORNL cyclotrons were probably the most economical ones ever built because the magnetic-field facilities from the Y-12 electromagnetic process equipment were used with very little change. Other unique features were the vertical mounting (instead of horizontal) of the "dee" units that are the heart of the cyclotron, and the high beam currents that were produced. These cyclotrons were used to develop the high beam current technology which was the basis for the high performance ORIC constructed at ORNL in 1962.

The Van de Graaff accelerators have been primarily used in fundamental studies of the reactions between charged particles and target nuclei. Both the Van de Graaff accelerators and the cyclotrons have been used in radiation-damage research to show the effects of radiation under various conditions.

These research tools supply a large proportion of the basic data for increased understanding of the field of nuclear physics.

New Research Tools

The study of nuclear reactions involves the measurement of radiation of all types, and consequently, physics research at ORNL includes strong emphasis on instrument development and modification. Of the many new instruments developed at ORNL, perhaps the outstanding examples are the scintillation spectrometer and the multichannel analyzers. The scintillation spectrometer was developed at ORNL after it had been observed in Germany that certain types of crystals emitted a flash of light when struck by a beam of radiation and that the intensity of the light flash is proportional to the energy of the radiation. The scintillation spectrometer measures the number and intensity of light

flashes emitted by a crystal exposed to radiation, and by electronic analysis of the measured flashes gives a precise determination of the intensity of the radiation and its energy distribution. Multichannel analyzers are a part of the electronic equipment used in analyzing the data from scintillation spectormeters and other radiation counters. The multi-channel analyzers count simultaneously and separately the number of particles or gamma rays of each of many energies detected by the crystal, and thus enable a complete energy analysis of the radiation beam to be made in a short time.

Further information about atomic structure and nuclear forces is obtained through a strong program of research in spectroscopy. This research has been particularly successful in demonstrating the effects of isotopic mass differences upon the spectrum of light emitted by an element under the excitation of an electric arc. The "isotope shift" observed in hyperfine spectroscopy is proving a valuable tool for isotopic analysis.

The analysis and interpretation of large quantities of nuclear data are aided by well-qualified groups of theoretical physicists and mathematicians who have access not only to the most recent data but also to the Laboratory's high-speed computing machines. The Laboratory obtained an electronic computer designed and constructed by the NEPA project and turned over to ORNL at the termination of its work.

An advanced type of digital computer, the ORACLE, was constructed for ORNL in 1954 by the Argonne National Laboratory. At that time it was perhaps the fastest machine in existence and for several years it

was surpassed by only a relatively few. After first being installed, it was improved in several ways to increase its versatility: storage was doubled, from 1024 words of 40 bits each to 2048 words; a magnetic tape auxiliary memory of extremely rapid access was added; an on-line cathode ray tube curve plotter with a photographic recording, an improved console with monitoring typewriter, a rapid magnetic tape output, and facilities for handling alphabetic inforamtion were added.

Such a machine can, of course, carry out only the laborious, repetitive steps of a computation, and effective utilization was possible only after careful and detailed mathematical analysis. In particular, the evaluation of certain standard functions, the solution of systems of equations, the determination of eigenvalues and eigenvectors, were types of computation that recur in many physical settings, and the design of efficient routines for them was a major activity of the mathematicians. Many such routines were developed, and, to mention a single example, that for computing the eigenvalues of a symmetric matrix was widely used in computing centers, here and abroad.

Reversibility in Nuclear Reactions

The failure of some time-honored symmetry theorems (such as parity conservation) re-emphasized the need for careful experimental evaluation of the validity of all such theorems. The theorem of "time reversal invariance" has been of particular interest, not only for "weak interactions" (such as radioactive beta decay and certain reactions with mesons) but also for "strong interactions" (for example, neutron capture by a nucleus followed by charged particle emission). One method of determining the validity of "time reversal invariance" for the case of

strong interaction is the measurement of a nuclear reaction and its inverse. First, let particle A bombard nucleus B, producing particle C and nucleus D. The (measured) behavior of this reaction, if time reversal is indeed invariant, can be used to compute the behavior of the "inverse" reaction where particle C bombards nucleus D, producing particle A and nucleus B.

At the Oak Ridge National Laboratory, the development of a flat-response neutron flux integrator has made possible the investigation of total neutron yields from charged particle reactions. The results on the H^3 (p,n)He^3, Li^7(p,n)Be^7, $Li^7(\alpha,n)B^{10}$, $H^3(\alpha,n)Li^6$ and $Si^{29}(\alpha,n)S^{32}$ reactions as compared with the reactions going in the reverse direction are, within experimental errors, entirely consistent with the assumption of time reversal invariance.

Orientation of Nuclei

Nuclear orientation has been produced through the use of enormous crystalline electric and magnetic fields occurring in suitable solids and by the direct application of the field of a large magnet. Using these techniques, studies of the spin dependence of neutron capture by Mn^{55}, Sm^{149}, and In^{115} have been made. Also, these techniques have been applied to U^{233}, U^{234}, Am^{241} and Np^{237} alpha particle emission and to U^{233} fission. These experiments help to establish a connection between the "cigar-like" shape of these nuclei and the detailed nature of the alpha particle emission and fission processes.

Germanium Fission Counters

The study of spin dependence in nuclear reactions is facilitated by forcing the spin axes of the nuclei in a sample to orient in a direction

common to all. These nuclear orientation techniques have been generally developed and, in particular, extended to fission studies in work at the Oak Ridge National Laboratory. These investigations must be performed at temperatures near the absolute zero where the fission fragment detectors that are useful at room temperatures are found to become unsatisfactory or inoperative. For this reason, a very compact and efficient germanium solid state counter has been developed, which detects alpha particles and fission fragments over the temperature range 1 - 77° K. This device has an exceptionally fast response time and may be used to measure fragment energies up to one hundred million volts or more.

Fission

The fission cross sections of all the isotopes of uranium have been measured in the Mev range with relatively good energy resolution and with good statistics. These careful measurements have shown up the maximum and minimum, and the inflections in the cross sections of the even isotopes which heretofore had been overlooked. In seeking an explanation of this phenomena, Bohr and Wheeler have re-investigated theoretically the fission process. Their explanations are based on the ideas that inelastic neutron scattering is competing with various modes of fission. As a further check on their ideas, angular distributions of fission fragments are being measured as a function of energy for several of the uranium isotopes.

Fission Fragment Spectrograph

A fission-fragment magnetic spectrograph utilizing the ORNL graphite reactor was constructed and operated. Measurements were made of the

energy distribution and nuclear charge distribution of fragments of a given mass--this information is of great importance in fission theory--of the scintillation respose, range-energy curves, and equilibrium charge of fission fragments in gases, and of the energy distribution and frequency of long-range alpha particles from fission. A new principle for fission fragment mass separation was evolved, and its limitations were studied.

Sensitive Neutrino Recoil Experiment

A sensitive recoil experiment has been carried out using the electron-capturing radioisotope A^{37}, a monatomic gas, that was allowed to flow through a vessel under essentially high vacuum conditions. The energy of the recoils was found to agree within about one per cent with that expected from the emission of a neutrino having a mass of less than one per cent of the mass of an electron (the next lightest particle).

Neutron Cross Sections

It has been found that neutrons and protons bound in certain nuclei are usually stable (O^{16} is such an example), so that for interpreting many phenomena, one can consider this stable core of nuclear matter as behaving as an average attractive force to a neutron or a proton. Thus, by studying the scattering of neutrons from O^{16} and by use of knowledge of the bound states of O^{17}, one is able to obtain clues as to the size and shape of the neutron and effective strength of the interaction of the O^{16} core on a neutron.

For elements above the very lightest ones, the cross sectional area of a nucleus, as seen by neutrons used as probes, shows resonance peaks

at various neutron energies. From the measured properties of these resonances, one can deduce other properties of the nucleus such as, for example, its probability of capture of a neutron with emission of gamma radiation. For neutrons at around room temperature, with velocities of about 2200 meters/sec, capture cross sections deduced from analyses of neutron resonances agree reasonably well with that found by experiment. Recent results with neutrons at higher energy, having velocities around 2.2 million meters/sec, show much higher probability of capture than expected, which indicates that information on resonances in this higher energy region is needed.

Neutron-Capture Cross-Section Measurements

The significant work in this field, especially that of importance in reactor design, is centered on the rare isotopes and those obtainable only with difficulty, both radioactive and stable. For these measurements of relatively high precision, the pile oscillator, time-of-flight spectrometer, and crystal spectrometer are utilized. Among the important contributions are, (1) the xenon-135 cross section (σ) vs. energy data, which are extremely important for the kinetics of high-flux reactors; (2) the complete survey of separated stable isotopes, with, for example, the discovery that elemental zirconium, potentially an important structure material, has a low cross section; and (3) the data on radioactive isotopes ionium (thorium-230), actinium-227, uranium-234, and uranium-236.

Milli-microsecond Time-of-Flight

At ORNL, a new instrument has been developed for measuring cross sections for neutrons with velocities around 2 million meters/sec. This

involves accelerating pulses of charged protons 5 to 10 milli-microseconds in duration (.000,000,005-0.00000001 sec) with a Van de Graaff machine, and measuring the time it requires for neutrons, produced by the protons in the $Li^7(p,n)Be^7$ reaction, to traverse a distance of a few meters. In obtaining the data on the neutron cross sections, time intervals of a fraction of a millionth of a second in duration are measured to an accuracy of a few per cent.

Neutron Diffraction

Investigations by neutron diffraction have furnished a unique approach to a study of magnetic phenomena at the atomic level. This is the only method for obtaining the detailed magnetic structures of magnetic materials and for obtaining the spatial distributions of the electrons which contribute to the atomic magnetic moments. Over sixty different magnetic materials have been investigated, including elements and compounds of nearly all the types of atoms in which the electronic configuration produces an atomic magnetic moment. Results of the neutron diffraction experiments are primarily responsible for recent theories of the mechanisms involved in the indirect coupling of the magnetic electrons via the nonmagnetic anions which surround them.

Nuclear-Alignment Studies

The spin dependence of nuclear forces is being studied in a direct manner by measurements of the cross sections of polarized nuclei, bombarded with neutrons polarized by passing them through highly magnetized iron. By using a method which utilizes the enormous magnetic field that can be created at the nucleus by the atomic electrons and a sample temperature of 0.2 degree above absolute zero, a 4% change in

cross section of manganese has been observed. Results in this field of experimentation may give an increased understanding of nuclear forces and nuclear structure.

Instrumentation

The basic physics research at ORNL contains a program of instrument development and modification. In addition to the development of equipment for the physicists' use, a program of instrumentation for medical diagnostic use of radioisotopes has been carried out. This program includes a one-chassis scintillation spectrometer, focusing collimators for high resolution, in vivo localization of radioisotopes and a surgical scintillation counter probe for use in surgical procedures.

Coulomb Excitation

Excitation of nuclei by a long-range electric interaction which does not involve a direct collision with the nucleus is a process known as coulomb excitation. This process has recently been developed into a powerful tool for the systematic study of nuclei. Because this excitation depends on the well-understood electric interaction, the measured probabilities for producing excited states are more easily and reliably interpreted. Hence one is able to extract from the experiments information on some of the finer details of nuclear structure.

Effect of Radioactive Decay Upon Atomic and Molecular Structures

New evidence has recently been obtained relative to the structural changes that take place in the electron shells of atoms following a radioactive change in the nucleus. Two kinds of effects are observed.

The first is encountered with pure beta emitters, or beta-gamma emitters in which there is no internal conversion, and in these areas there is usually no loss of extra-nuclear electrons. The second is encountered when one or more of the inner atomic electrons is removed, and this effect therefore accompanies radioactive decay by electron capture and decay processes that involve internal conversion. In these events the damage to the atomic electron structure can be severe. When more molecules have been surveyed, embodying different kinds of radioactivities, the results promise to be of great interest to basic hot-atom chemistry.

Neutron Time-of-Flight Chopper

In completing our understanding of nuclear structure, as well as in predicting the behavior of high flux reactors, it is necessary to accumulate information on the nuclear properties of radioactive nuclei. Significant research with such nuclei has been performed by use of the neutron chopper at ORNL. Neutron total cross sections of the radioactive fission products, Zr^{93}, I^{129}, Pm^{147} and Sm^{151}, have been measured for neutron shaving velocities in range from 2,200 meters/sec to about 12,000 meters/sec (thermal energies to 30 ev).

For carrying out this type of measurements on unstable as well as on stable nuclei, a greatly improved mechanical chopper allowing considerably better resolution has been installed in the LITR.

Molecular Studies Using Tritium

Careful measurements carried out on the spectra of the isotopic species of hydrogen have been of considerable value in probing the secrets of molecular structure and binding forces. The relatively large

scale availability of tritium, the radioactive hydrogen isotope of mass three, has now made possible a further valuable extension of this research technique.

The current program in molecular spectroscopy is concerned largely with the study of simple molecules, such as ammonia and some of its derivatives into which both tritium and deuterium have been selectively incorporated in various combinations. Through mathematical analysis, using the Oracle, additional insight has been provided into the properties of these fundamentally important compounds.

Cyclotron Development

Over a period of years Oak Ridge National Laboratory has established a position of leadership in the design of cyclotrons of high performance. The 44-inch cyclotron constructed at ORNL holds the world's record for beam strength at one Mev. During the course of performance tests of the 44-inch cyclotron, a maximum beam of 200 milliamperes was attained. Work on a new approach to cyclotron design culminated in the design of the Oak Ridge Relativistic Isochronous Cyclotron (ORIC) whose performance, first tested experimentally in electron analogs, and later demonstrated in actual operation of the ORIC itself, far outstrips that of any existing cyclotron in regard to strength of beam and flexibility in accelerating a variety of particles over a wide range of energy. This cyclotron design represents a technological breakthrough of the first magnitude and promises to open areas of nuclear research which previously could only be explored on a limited basis. The electron analog tests demonstrated that the energy limit encountered with fixed-frequency cyclotrons can be overcome by the introduction of azimuthal variations

in the magnetic field. In the conventional fixed-frequency cyclotron the relativistic increase in the mass of the ions with energy makes it increasingly difficult to keep the ions both in focus and in resonance. As a result, the practical energy limit for protons is about 25 Mev. In the frequency-modulated cyclotron the frequency of the accelerating voltage is programmed to maintain resonance for the orbiting ions. It is then possible to go to much higher energies (over 700 Mev), but at the expense of a considerable reduction in beam current. In the new development, the use of an azimuthally varying field introduces strong focusing forces which make it possible to design the magnetic field to keep the ions in resonance to high energies, up to 900 Mev.

The AVF cyclotron retains the great advantage of both fixed frequency and fixed magnetic field. The azimuthal variations provide sufficiently strong focusing to overcome the defocusing effect of the radially increasing average magnetic field which is necessary to compensate for the relativistic increase in mass. Large currents of ions can thus be accelerated to very high energies.

The AVF cyclotron has been under investigation at the Oak Ridge National Laboratory since 1950. Theoretical studies and calculations with the ORACLE showed the feasibility of such high energy fixed-frequency cyclotrons. A Cyclotron Analog, operated to test the theoretical ideas of new designs, accelerated electrons to an energy of 190 kev (comparable to 350 Mev for protons). That the Analog performed exactly as predicted was strong substantiation of the basic theoretical work. A larger magnet, with an eight-fold azimuthal configuration, was built to be used in an analog for a proposed 850-Mev

proton cyclotron. A 10-Mev electron synchrotron analog was moved to Oak Ridge from Brookhaven National Laboratory for use in studying problems associated with injecting protons from such a high energy cyclotron into a proton synchrotron.

High Energy Accelerator Studies

Theoretical studies of the new type of accelerator system indicated its promise of very high performance. The system was composed of a spiral-sector fixed-frequency cyclotron, acting as the injector for a rapidly pulsed alternating gradient synchrotron. Each component was complex by comparison with previous accelerators, and careful studies were performed, both analytically and numerically, of the orbits in the cyclotron, beam extraction from the cyclotron and injection into the synchrotron. The studies indicated that it would be possible to perform the extraction and injection much more efficiently than has previously been possible, and detailed plans were made for experimental tests with an electron analog.

AVF Proton Cyclotron-Cyclotron Analog

The Laboratory pursued the design and development of an unusual accelerator system which consisted of an 850-Mev-high-intensity, azimuthally-varying-field cyclotron injecting its beam into a pulsed synchrotron ring. The system would yield 12-Gev protons at an intensity considerably exceeding that available from conventional machines. To gain design experience and to study some of the problems of orbit dynamics for the 850-Mev injector, a cyclotron analog was built and operated. The analog accelerated electrons to an energy of 200 kev

utilizing an air-cored magnetic field designed entirely by analytical techniques with the aid of the ORACLE. The low threshold voltage and the fact that operation is almost exactly as predicted proves the soundness of both the theory and the design techniques. A new analog with 8 spiral poles was designed to model the 850-Mev AVF cyclotron almost exactly. The outstanding performance of the more advanced analog provided the basis for ORNL's proposal to construct a full-scale accelerator of this sort. The large accelerator proposed for construction to incorporate these advances in cyclotron technology has been named the Mc^2 Cyclotron in the Laboratory's proposal for authorization to construct the machine.

86-Inch Cyclotron Beam Deflection

A beam deflection system was incorporated into the 86-inch Cyclotron to provide an intermediate-intensity focused proton beam inside the shielded area and a less intense beam outside the shield. Both beams are used for nuclear physics experiments. The deflector does not interfere with the use of high-current internal targets for isotope production. The deflector system contains many novel features which increase its flexibility and reduced the cost.

25-Mev Nitrogen Accelerator

The 63-inch-pole-face-diameter, fixed-frequency cyclotron for accelerating triply charged nitrogen ions provides a new and unique means of studying nuclear structure. For light nuclei especially, the direct experimental observation of the geometrical properties (i.e., angular distribution, momentum, etc.) of nuclear-reaction products

permits important deductions to be drawn concerning the nature of coulomb interaction of nuclei and the detailed mechanism of nuclear reactions. Additionally, important studies of range-energy relationships and the charge-equilibrium mechanism of the interaction of heavy fast-moving ions in matter are possible.

Internal-Conversion Coefficients

One of the most important procedures for the determination of spins and parities of nuclear energy states is the comparison between measured and calculated values of the internal-conversion coefficient. The values of these coefficients, representing the relative yields of K-conversion electrons to gamma rays emitted in a transition from an excited nuclear state, were obtained by using the exact quantum mechanical theory of the electron. These results have been used extensively for the purpose of establishing decay schemes of radioisotopes, thereby providing the data for testing current theories of nuclear structure.

Internal-Conversion Electron Spectra

Radioactive nuclides produced by proton irradiation of rare earths (Gd through Lu) were chemically separated with ion-exchange resin columns and their internal-conversion spectra studied with a 180-deg permanent magnet spectrograph. More than a dozen transitions proceeding between postulate rotational levels in even-even nuclei were observed. For the region of neutron number 94 to 108, it appears that the value of the moment of inertia has a dependence on proton number, and is slightly smaller for higher Z elements with the same neutron number.

Light Particle Emission From Nitrogen-Induced Nuclear Reactions

Light reaction products, proton, deuterons, tritons, and alpha-particles, were detected from nitrogen reactions on Li^6, Li^7, Be^9, Al^{27}, and O^{16}. The energy distributions of these particles were measured to provide information on the level densities of the residual nuclei. Angular distributions, in the case of particles from Al^{27} targets, proved to be nearly isotropic.

Anomalous Inelastic Scattering in Heavy Elements

An unexpected peak in the energy distribution of inelastically scattered protons was found. There are striking regularities in its cross section and angular distribution in neighboring elements. The effect has been studied in great detail in a variety of experiments but as yet no completely satisfactory explanation has been evolved.

Neutron Transfer Reactions

Transfer reactions were investigated in which a neutron transfers from the incident nucleus to the target nucleus. Excitation functions were measured in the 63-in. cyclotron for ten such reactions and a systematic dependence of the cross section on the energy was found. Angular and energy distributions were measured in one case. The angular distribution was found to be peaked at 60 deg in the barycentric system.

Excessive Charged-Particle Emission by Medium and Heavy Nuclei

In a series of experiments, a greatly excessive emission of protons and alpha particles was observed from proton-induced nuclear reactions in medium weight and heavy nuclei. Various possible explanations for the effect were studied but almost all of these have proven incorrect; no good explanation has been found yet.

Fusion of Heavy Nuclei

A new type of nuclear reaction in which heavy nuclei fuse was observed and described for the first time. These fusion reactions are produced in the 63-inch cyclotron by bombarding various target nuclei with 28-Mev nitrogen ions. The two nuclei fuse and result in a final nucleus a few mass units lighter than the sum of the masses of the two incident nuclei. An example of such a reaction is $O^{16} + N^{14} \longrightarrow Al^{28} + 2p$. The total cross sections for many such reactions have been measured.

Range-Energy and Charge Distribution for Heavy Ions

The behavior of fast (28-Mev) nitrogen ions passing through matter was investigated. The range-energy relations in various materials were measured. The average charge, the charge distribution, and the electron capture and loss cross sections were also determined.

Diffraction Scattering of Protons by Nuclei

The diffraction nature of the scattering of protons by nuclei was observed for the first time in the 86-in. cyclotron; all of the qualitative features of the process were studied. The maximum was found to occur at the same energy, and with the same cross section and angular distribution in a large number of elements. The evidence indicates that it is probably due to some type of collective motion of the protons and neutrons in the nucleus. This has led to a large amount of similar work at other laboratories, and its theoretical analysis has provided most of the basic elements of the optical model of the nucleus.

Elastic Scattering of Nitrogen by Nitrogen

The elastic scattering of nitrogen by nitrogen was investigated. The differential cross section for this process, at several energies,

was measured and the results were explained by a simple semi-classical total absorption model.

CHEMICAL PROCESSING TECHNOLOGY

The chemical technology phase of the Laboratory's research and development program has been in support of the processing requirements of nuclear energy. Its purpose has been to develop processes which will prepare the unique materials required for nuclear reactor components, processes to separate and recover the valuable materials discharged from reactors, and to treat radioactively contaminated wastes discharged from the reactors and associated processes. These processing operations represent a new industrial activity which has become an important part of the chemical industry in the United States.

Raw Materials

A very valuable chemical technology discovery in the raw materials area was solvent extraction reagents for recovering uranium from sulfate solutions used to leach uranium from ore. Ore processing plants using these solvent extraction reagents, in the Dapex and Amex processes, are now in operation in the United States and Canada (see table). The solvent for the Dapex process is a dialkyl phosphoric acid and is used for both uranium and vanadium recovery. The solvent for the Amex process is a long-chain alkyl amine and has been used for uranium recovery. These solvent extraction processes have increased uranium recovery with decreased chemical cost.

A second development in this field was the application of the Higgins continuous ion exchange equipment to uranium recovery from ore leach liquors. This promised significant reduction in cost since the uranium can be extracted directly, without the necessity of clarifying

the liquor. Following successful pilot plant tests, the process was "put on the shelf" because uranium processing plants were operating well below capacity and further improvements were not of interest.

The Monex process for the recovery of thorium from Brazilian sludge was developed and carried through engineering tests with satisfactory results. This solvent extraction process employed tributyl phosphate to extract thorium from unclarified leach liquor. The program was terminated when the Commission's requirements for thorium were reduced.

Feed Materials

An ion exchange technique for the recovery of uranium from metal-reduction bomb liners and slag was developed through a pilot plant scale. A pilot plant for recovery of slightly enriched uranium from this waste is now in operation at Y-12. A Higgins continuous ion exchange unit is used with the unclarified sulfate leach liquor.

The Excer process study was completed. This involved aqueous-phase hydro-fluorination of uranium to uranium tetrafluoride. The process did not have sufficient economic advantage to justify replacement of the existing gas-phase process plants in the United States. However, a pilot plant employing this process is being constructed in Japan.

The chemical kinetic studies of uranium tetrafluoride (UF_4) reaction with oxygen to produce uranium hexafluoride (UF_6) and uranyl fluoride (UO_2F_2), the Fluorox process, have been completed, and engineering tests are in progress. This process promises economies by using oxygen instead of elemental fluorine to convert UF_4 to UF_6.

Continuous procedures for direct reduction of uranium hexafluoride and thorium tetrachloride to metal by sodium metal and sodium or lithium

have been studied. Results of chemical and engineering tests now in progress are very promising. The Metallex process for reduction of $ThCl_4$ with sodium amalgam appears to be competitive with other proposed reduction methods. The reduction of UF_6 by sodium, the Druhm process, or by lithium amalgam have demonstrated yields of 80% and 85%, respectively.

Chemical Plant Criticality Studies

In the design of chemical plants for processing fluids containing fissionable material, it is essential to ensure that the fluids not collect in either an individual container or several contiguous containers in such a way as to constitute a critical mass. A new type of tank has been experimentally verified which utilizes the annular space between two process pipes for solution storage, with water and cadmium as neutron absorbers in the interior pipe. This arrangement will afford a considerable saving in required container and building space.

Transuranic Elements

Chemical separations studies for recovery of neptunium and americium have been carried through pilot plant scale operation at ORNL. Gram quantities of both elements were successfully separated as a result of these studies. The neptunium was recovered from the fluorination ash of recycle uranium going from nuclear reactors back to the gaseous diffusion plant for enrichment. The americium was recovered from plutonium.

Uranium-233

A special separation of high purity uranium-233 was carried out at Oak Ridge National Laboratory to provide material to Argonne National

Laboratory for nuclear cross-section measurements and for other research. Waste streams from the Thorex process which contained uranium produced exclusively from protactinium decay was reprocessed in the Thorex pilot plant for uranium recovery. This product was further purified by electromagnetic isotope separation at Y-12 to give a U-233 product essentially free of U-232, U-234, U-235, and U-238.

CHEMICAL RESEARCH

The general field of chemistry and chemical technology research covers ORNL's largest basic research efforts. This stems from the fact that ORNL has been regarded from the earliest days of the AEC as the center for chemistry and chemical technology research and also from the fact that ORNL's major program interests in reactor development, radioisotope production, and other activities involve the application of methods or techniques that are largely in the field of chemistry or chemical technology.

New Research Techniques

Research on the chemistry of heavy elements has been especially fruitful in connection with the development and improvement of chemical processes for use in the fissionable and raw materials production plants. An example is the discovery that, by causing uranium to form a complex negative ion in solutions instead of its usual positive ion, one can effectively separate and purify uranium using the ion-exchange technique.

Research on the fission-product elements required the development of improved methods of separating them from each other and from the heavy elements. The ion-exchange process developed at ORNL was the first method to be used successfully to separate gram amounts of the rare-earth elements in pure form. These fifteen elements are so much alike chemically that their separation is exceedingly difficult. In the progress of this work, a new element, technetium, was prepared in weighable amounts for the first time by other methods developed at ORNL in the fission-product research. The ion-exchange technique of separation

and purification has been used with outstanding success in widely different applications, such as the separation of nucleic acids and other biological compounds and the final product purification step in the Purex process for separating and purifying uranium and plutonium.

Anion Exchange. A new technique involving the use of anion exchange resins and solutions of the metallic elements has been developed and intensively studied. This technique has important applications in analytical chemistry and the isolation and purification of uranium, including recovery of uranium from extremely dilute solutions, and shows great promise for general industrial application.

Some elements are very strongly held by these resins, permitting the processing of large volumes of solution with very small amounts of resin. For example, iron could be removed from tons of commercial hydrochloric acid with only a pound of resin and, similarly, tons of uranium-bearing solutions under the appropriate conditions could also be processed with only relatively small amounts of resin.

Many elements which are generally considered very similar show large differences in their ability to form negatively charged complexes. Anion exchange takes advantage of these differences, thus permitting many separations with extremely simple equipment in a very short time. For example the essentially complete separation of cobalt and nickel can be achieved with a very short column of resin by passing the mixture in the appropriate solution through the column. The nickel appears immediately, while the cobalt is held by the resin but then can be removed rapidly by the use of a second reagent (water). Many similar separations are possible. In some cases, for example zinc and uranium, separation from practically all elements is possible in less than an hour using only common chemicals.

Ion-Exchange Research

The basic concepts involved in ion-exchange phenomena, with both cationic and anionic resins, have been extended and used in important and diversified applications, such as (1) the separation of the rare-earch elements in up to multigram amounts for chemical and physical properties research; (2) the separation and isolation of nucleic acids and other biological compounds important in metabolism; (3) the separation, as positively or negatively charged complexes, of elements ordinarily separable only with extreme difficulty, and (4) the concentration and recovery of valuable trace elements from large volumes of solution. The technique of separation in all cases involves the sorption of the substance on the resin under one set of conditions (i.e., temperature, concentration, pH, complex behavior, etc.) and its elution under another set of conditions.

Pure Quadrupole Spectra

The new field of quadrupole spectroscopy in solids is proving to be a powerful tool for elucidating the nature of chemical binding, in addition to being a highly precise method for determining nuclear quadrupole-moment information. Under certain conditions, the exact radiofrequency for absorption of electromagnetic radiation is a function of the interaction between the nuclear-charge distribution and the electrical-charge distribution in the chemical bonds. Thus, the nucleus is used as a probe for sampling features of this interaction which, in turn, may be related to the nature of chemical bonds.

A cooperative program with Duke University was set up to measure by microwaves some nuclear properties of radioactive isotopes available

at ORNL. One of the properties studied was the way in which the electric charge in the nucleus is distributed (that is, like a football, or like a sphere, or like a saucer). The exact frequency of radiation which a molecule absorbs in the radio-frequency region under certain conditions is the result of an interaction between this nuclear charge distribution and the distribution of electrical charge in the chemical bonds which hold the atoms of the molecule together. Hence, a measurement of the frequency of this line in a series of molecules gives direct information about the differences between analogous bonds in the series, a result of the highest interest to chemists. It was already known that the frequencies of the lines were so related to chemical bonding in gaseous molecules, and it was known that similar lines could be ovserved in crystals, but it was discovered at the Laboratory that the same direct correlation between frequency and bond nature applied to the lines measured in crystals. Thus, a very wide field of interest to chemists and physicists has been opened up, for the range of compounds measurable as crystals is very large.

Production Processes

Research on basic chemistry led to the development of a method of separating hafnium and zirconium, which always occur together in nature and are very difficult to separate. The ORNL-developed process was selected out of a great many developed at various AEC installations as the one to be used in a production plant. This process is of great importance to reactor development, where zirconium is a very desirable structural material but is made unattractive by the presence of hafnium with its large capacity for absorbing neutrons. Production plants using

this ORNL process have been built to supply hafnium-free zirconium to meet the Atomic Energy Commission's needs for the reactor program.

During the war, purified radioisotopes were needed for research and were supplied mostly by ORNL. Radioisotope-production processes developed in the research laboratories formed the basis for the Commission's program of radioisotope sales and distribution centered at ORNL after the war. This research has been expanded to include the preparation of chemical compounds "labeled" with radioactive atoms, particularly organic compounds containing carbon-14. It should be noted that a fairly large private industry has been built up, depending upon the radioisotopes produced at ORNL.

Studies of chemical reactions and reaction equilibria led to the development of a chemical method for separating certain isotopes, based upon the slightly different distribution of the isotopes when reaction equilibrium is reached. An ORNL-developed process for the chemical separation of lithium isotopes was used in the production plant constructed in Y-12.

Chemistry of Technetium

The properties of fission-product Tc^{99} continue to be of importance to nuclear energy applications because of the long half-life, the high fission yield and the large thermal neutron cross-section of this isotope. A considerable advance in the understanding of the chemistry of technetium has been made at the Oak Ridge National Laboratory. Oxidation states of 7, 5, 4 and 3 have been shown to exist in aqueous solutions. Thermochemical and electrochemical measurements have led to a quantitative oxidation-reduction scheme inter-relating the stabilities

in aqueous solutions of several of these oxidation states to that of technetium metal at 25°. Low temperature specific heat measurements have yielded values for the entropy of several technetium compounds. Magnetic measurements have shown that certain complex compounds of the tetravalent state of technetium contain three "unpaired" electrons. Extensive solvent extraction measurements have revealed that heptavalent technetium may be removed efficiently from a wide variety of aqueous solutions by organic compounds containing a basic oxygen or nitrogen atom. An examination of the ultra-violet absorption spectrum of technetium in these organic solutions indicates that a sensitive optical method for analysis is possible.

Organic Chemistry

Techniques of multiple labeling with carbon-14 which have been developed at ORNL now permit a much more penetrating study of organic reactions than has ever before been possible. By the use of these techniques 1) the mechanism of the pinacol rearrangement has been clarified; 2) in combination with stereochemical experiments it has been shown that the energy barriers to free rotation about the carbon-carbon bond, contrary to general belief, are determining factors in many reactions and 3) it has been demonstrated that the conformations, — or the arrangements in space — assumed by certain cyclohexane derivatives — are the important qualities which differentiate their reactions from those of noncyclic compounds.

Inorganic Ion Exchange

The standard organic ion exchanger resins, though of extreme importance and usefulness, suffer from a number of serious handicaps,

particularly when viewed from possible atomic energy applications. For example, they have relatively low selectivity for similar ions, making some separations difficult; and they are unstable in intense radiation fields and in the presence of strong oxidizing agents. In an attempt to develop new ion exchangers a large group of relatively simple inorganic materials was found satisfactory. The classes of compounds which have received most attention in the past few years are the insoluble hydrous oxides and certain insoluble salts containing polyvalent anions such as phosphates, tungstates, molybdates and arsenates. Most of the oxides studied, which include those of Ti(VI), Zr(IV), Th(IV), Nb(V), Ta(V) and Sn(IV), appear to be useful anion exchangers in acidic and cation exchangers in basic solutions. The oxide of Bi(III) and mixed oxides containing excess Bi(III) are principally anion exchangers with a remarkable selectivity for halide ions. The polyvlent salts, as well as the oxides of V(V), and U(VI) are attractive cation exchangers.

These inorganic exchangers have unusual selectivities and hence permit some otherwise difficult separations, e.g., of the alkali metals from each other or of the alkaline earths with very small columns. The inorganic anion exchangers have special selectivities for polyvalent anions and thus should prove useful in the recovery of chromates, molybdates, tungstates, etc. In view of their anticipated radiation stability, the new inorganic ion exchangers should find special application for the processing of highly radioactive solutions.

Inorganic Polymers

Hydrolysis of metal ions is frequently accompanied by aggregation or colloid formation, and the course of such reactions is important for

an understanding of the chemistry of these solutions. During the last few years estimates of the degree of aggregation and of the change of the aggregates have been obtained from equilibrium ultracentrifugations, and more recently through light scattering, techniques not previously used in this field. The existence of a wide variety of hydrolytic reactions was confirmed. However, contrary to the opinion of some, many elements (e.c., Pb(II), Bi(III), Zr(IV), Hf(IV) exist as discrete low molecular weight polymers over wide ranges of acidity, metal ion concentration, and temperature, rather than as colloidal aggregates of large and indefinite size. Continuous distribution of small ploymers was observed in some cases, particularly Th(IV). Complexing of polymeric cations by simple anions (e.g., ClO_4^-) occurs under conditions where it is not expected for monomeric ions. The species formed by the amphoteric ions Sn(IV) and Pb(II) in basic solution are monomeric, rather than colloidal, as some have held. However, polymeric aggregates were observed for W(VI) and Mo(VI) on the alkaline side of the isoelectric point, in confirmation of usual notions on iso-poly acid formation.

An extremely soluble (ca. 3 kg Th(IV) per kg water) high molecular weight hydrolytic polymer of Th(IV) containing ca. 1000 Th(IV) ions per particle was discovered. It can be prepared by controlled drying of precipitates of hydrous thorium oxide. Though it is very stable at low temperatures, attempts to preserve the dispersions at temperatures above 200°C have so far been unsuccessful.

Anion Exchange Studies of Metal Complexes

The technique of separating metals by anion exchange through control of the medium and control of the degree of complexing, which was discovered

earlier at ORNL, has been broadly extended. Studies have been carried out in solutions containing most of themetallic ions of the periodic table. Particularly detailed is the study in chloride solutions which presently encompasses almost 90 elements. These studies have helped to establish anion exchange of metal complexes as an important part of analytical chemistry and of hydro-metallurgy. A considerable amount of new information on properties of the resins and on complexing of metals by various anions such as chloride, nitrate, fluoride, sulfate, citrate and EDTA (ethylene diamine tetraacetic acid) has been obtained as a by-product of these studies.

On the basis of information now available, it appears practical to devise a general ion exchange separations scheme for the metallic elements, and a preliminary scheme involving approximately 20 elements has been tested.

Neptunium-237 Production and Purification

Interest in the production and isolation of neptunium (element 93) of mass 237 has been limited in the past to an occasional gram-scale operation sufficient to serve the most urgent requirements. The fact that large quantities are actually formed along with the production of plutonium (U^{238} n2n $U^{237} \xrightarrow{\beta} Np^{237}$) presents an attractive possibility for continuing routine by-product production of this important element.

An initial analytical inspection of one isolated plant process at ORNL resulted in the discovery that neptunium in an essentailly pure state was present as a waste product. From this process the first 100 grams of neptunium (ten times as much as all previous production) was eventually isolated. Inspection of other production processes failed

to reveal a similarly fortunate situation; however, after a reexamination of the solvent extraction chemistry of neptunium, a highly significant feature, the catalytic oxidation of neptunium (V) to an extractable state, neptunium (VI), involving a trace concentration of nitrite ion was found. It was then finally possible to account for its often erratic distribution.

Continued investigation has led to the recovery of neptunium from what is likely to become a significant and permanent source of kilogram quantities of neptunium. This is the nonvolatile residue following fluorination of depleted uranium in the Gaseous Diffusion Process. The routine reprocessing of this residue from one plant has already produced the second 100 grams of neptunium for an apparently unending research demand.

The Preparation of Kilocurie Quantities of Xenon-135

In the initial direct measurement of the xenon resonance cross-section it was possible to cover only a narrow range (0.03 to 0.20 ev.) because of the limited amount of xenon available. In reactor technology it continues to be important to extend this energy range of xenon cross-section measurements; therefore, as soon as the Homogeneous Reactor Experiment (HRE) became operative, there was renewed interest in further direct measurement. This reactor provided a unique source of even the shorter-lived mixed fission gases and was actually capable of producing up to 40,000 curies of 9.2 hr Xe-135 per day at the operating level of 1 megawatt.

Using an underground holding-tank, the entire gas stream containing added oxygen and the fission gases produced by the reactor was safely

diverted for collection. Intensely radioactive, shorter-lived products were then allowed to decay a few hours during the period required for the necessary adsorption and chromatographic separation. The ultimately purified samples of 500 or more curies were sealed in 1 cc volumes in a special transmission capsule for neutron attenuation measurements. Precision assay of the absolute number of xenon atoms present in a sample was accomplished by recovering the radiogenic cesium-135 from the capsule following xenon decay and applying mass-spectrographic isotopic dilution analysis. The remeasurement of the xenon cross-section with this quantity of source-material fulfilled a significant need.

Boron Isotope Separation

A new and improved system for the enrichment of boron isotopes has been developed and successfully demonstrated. This countercurrent gas-liquid system utilizes chemical exchange between boron trifluoride gas and the anisole boron trifluoride complex to enrich B^{10} in the liquid phase. The bas and liquid streams form closed cycles and refluxing is accomplished by adding and removing heat.

The new system, named ANCO and ANisole-COmplex, has two principal advantages over previous methods of boron isotope enrichment: (1) the separation factor is much larger, 1.03 at 25°C, and (2) the new system can be operated at atmospheric or higher pressures. Since the new system represents improvement in both the enrichment and through-put capacities of precious processes, a considerable reduction in the unit cost of B^{10} is anticipated by use of the new method.

Radiation-Induced Formation of Nitric Acid in Moist Air

Most of the interest of radiation chemists since the beginning of the atomic energy project has been focused on the reactions of liquids. Only one gaseous system has been of real practical interest, that of air. Early work showed that radiation-fixation of nitrogen (to form nitric acid) in the air over a water-moderated reactor was responsible for the increasing acidity of the water. More recently, there have been suggestions that this reaction might eventually have practical use in the manufacture of nitric acid. However, very little was learned about the mechanism of the reaction because of the large number of intermediate products for which no satisfactory analytical methods were available.

Experiments at this Laboratory have demonstrated that a detailed analysis for almost all of the intermediate compounds of nitrogen and oxygen can be made by infrared spectroscopy. By a combination of this with conventional measurements of pressure change (both carried out on the irradiation cell itself), it has proved possible to unravel most of the details of this process. In particular, the role of water has been clarified, and it will now be possible to choose rationally the best conditions for nitric acid synthesis, and to evaluate with greater certainty the ultimate economic prospects of this particular radiation synthesis.

Short-Lived Fission Products Studies

In 1954 it became possible to begin detailed studies of short-lived fission products because of refinements in the technique of quantitative beta- and gamma-ray scintillation spectrometry, and the development of pulse height analyzers with large numbers of channels, capable of rapidly recording complete energy distributions.

A common characteristic of short-lived fission products is their high decay energy which, in turn, makes it possible to excite many nuclear levels in the daughter nuclides at relatively high energies. Some of the most energetic radiations ever seen in radioactive decay have been characterized in these studies. The complexity resulting from this situation is illustrated in an analysis of a gamma-ray spectrum of 86-second I^{136}; since the beta-decay energy has been measured in these studies as 7.0 Mev, it is quite reasonable to find the gamma-ray spectrum complicated by transitions between the numerous, energetic levels of the daughter, Xe^{136}.

The principal accomplishment has been a systematic study of the decay schemes of the short-lived isotopes of rubidium, 17.8-minute Rb^{88}, 14.9-minute Rb^{89}, and 2.6-minute Rb^{90}, and of the halogen isotopes 32-minute Br^{84}, 52-minute I^{134}, and the 86-second I^{136} already mentioned. It is important to certain circulating fuel reactor and continuous reactor fuel processing concepts that in all of the short-lived fission products studied, both beta and gamma rays of high energy and intensity were found.

Nuclear Chemistry

A program has been started to determine cross-sections of heavy nuclides of particular interest to reactors and to try to correlate some of the observed transmutations of heavy nuclides in reactors with reported cross section data. The following are examples of results obtained to date: An effective reactor cross-section of 80 ± 20 b has been found for Np-239. A value of 290 ± 30b for the thermal cross-section of Pu-240 has been both measured in the ORNL pile oscillator and calculated from

known resonance parameters; an infinitely dilute resonance integral of about 8500 b has also been calculated from the latter. It was found that observed transmutation rates of plutonium isotopes in graphite, heavy water and light water moderated reactors were consistent with these figures and not consistent with a single effective cross-section of about 500 b, as has been previously reported by several investigators. A value of 320 b was calculated for the (infinitely dilute) resonance integral of U-236. Using a thermal cross-section of 6 b, an effective capture cross-section of 33 b was calculated for MTR neutrons, as compared with an observed value of 34 $\pm$ 6 b. Since the effective value may then vary from 6 to about 35 b depending on the extent of thermalization of the neutrons involved, a possible explanation is hereby seen for the spread of 5 to 35 b previously reported by other investigators.

Fused-Fluoride-Salt Systems

In the realm of reactor fuels for mobile reactors, the inorganic fluorides have become increasingly prominent, in contrast to chloride or hydroxide systems. In the constant search to find fused-salt systems with better physical, nuclear, and chemical properties, the Oak Ridge National Laboratory has since mid-1950 undertaken the technologically difficult study of ternary and quaternary systems, as well as many binary salt mixtures. From the phase diagrams of systems utilizing mainly zirconium or beryllium fluorides as base materials, together with important physical data (e.g., viscosity, thermal conductivity, vapor pressure, radiation stability, etc.), it has been possible to propose salt mixtures suitable for the aircraft reactor.

High Temperature Reactor Chemistry

A continuing program of research has been devoted to the behavior of fluoride mixtures at temperatures up to 1000°C. This program, which has generated a considerable body of fundamental knowledge of the chemistry molten fluorides, has demonstrated the general suitability of uraniferous fluoride melts as fuels for high temperature nuclear reactors.

An experimental study has been devoted to the phase behavior of fluoride mixtures with especial emphasis on those containing UF_4 and/or ThF_4. Detailed phase diagrams have been developed, each based on data for more than 30 binary and ternary systems. Nearly 50 other systems have been given sufficient examinations to disclose their probable lack of value as reactor materials. The systems $NaF-ZrF_4 - UF_4$, which furnished the fuel for the Aircraft Reactor Experiment, and $LiF - BeF_2 - ThF_4 - UF_4$ seem most promising for reactor exploitation. A similar study of phase behavior of mixtures containing the fluorides of plutonium has been carried out.

A method in which the molten mixture is alternately treated with gaseous HF and H_2 has been developed for preparation of $NaF - ZrF_4 - UF_4$ mixtures of reactor purity. This method, which was used to produce material for the ARE and for the numerous engineering experiments with only minor modifications to many fluoride systems.

Equilibrium constants have been determined experimentally for chemical reactions of several metals with components of molten fluoride solutions. Activities of species in the molten fluorides have been evaluated from such data as well as from emf's of concentration cells using molten fluoride electrolytes and from vapor pressure measurements.

Activities of metals in solid alloys have been estimated from emf of cells with molten electrolytes and alloy electrodes. Measurement of rate of exchange between radiotracer ions of a metal in molten fluoride solution and normal atoms of the metal in a solid alloy has permitted calculation of diffusion coefficients for the test metal in the alloy. Data from all these sources have been incorporated into a theory of corrosion of metallic alloys by circulating molten fluorides which correlated the data obtained from many empirical corrosion tests.

To serve as the fuel for a long-lived nuclear reactor the molten fluoride must withstand buildup of fission products as elements or as their fluorides. The fission porduct fluorides have been found to be sufficiently soluble in the molten fuel to avoid precipitation during reasonable burnup of uranium. The noble gases are very sparingly soluble in the fuel; the important poisons Xe and Kr may, as a consequence, be removed readily and continuously. Several methods, including selective precipitation of oxides, chemisorption on solid oxide beds, and chemical exchange between the melt and solid fluorides show promise for partial decontamination of the fluoride fuel mixture.

Miscibility of Metals with Salts in the Liquid State

The increasing technological use of liquid metals and molten salts as heat transfer agents has created a strong interest in the properties of mixtures of these two chemically and physically rather different groups of substances. The miscibility, in the liquid state, of transition metals with some of their compounds, namely oxides or sulfides, and limited solubility of some alkali and alkaline earth metals in their molten halides has been known. Work at ORNL has

demonstrated that for the alkali metals, complete miscibility with their molten halides is a common occurrence. The minimum temperature of complete miscibility depends on the particular system, and in general decreases in these systems from lithium to cesium and from fluoride to iodine, i.e., parallel with the decrease in cohesive forces in the metal and the salt. The bromides represent an interesting and as yet not fully explained exception.

At moderate metal concentrations, large differences between the electrical conductivity of the alkali metal solutions and that of others, such as cadmium and bismuth, in their molten halides, indicate a rather different structure: alkali metals form polymerized metal atoms and molecules, such as K_2, which donate electrons for electrical or thermal conductances, whereas other metals form stable complex ions, such as Cd_2^{2+} which had been shown not to contribute conducting electrons. These findings are expected to have significant applications both to a theory of the dilute metallic state at thigh temperature and in a strongly polar medium (somewhat reminiscent of the dilute metallic state in liquid ammonia solutions at low temperatures), and to the technology of heat transfer media.

High Temperature Aqueous Solution Chemistry

A concerted effort has been made during the last few years to elucidate the properties of aqueous solutions at elevated temperatures. The cells Ag, AgCl, $HCl_{(m)}$, Hg_2Cl_2, Hg and Ag, $Ag_2SO_{4(m)}$, Hg_2SO_4, Hg have been studied up to 250°C and have shown to give the expected EMF values when the acidity was sufficiently high to prevent hydrolysis of the mercury salts. The cell $PbSO_4$, PbO_2, $H_2SO_{4(m)}$, Ag_2SO_4, Ag was

studied up to about 150°C. The solubility of Ag_2SO_4 has been studied in various electrolyte media up to 250°C; good agreement was found between the calculated and observed solubilities in H_2SO_4 media. An isopiestic unit for determining osmotic and activity coefficients of aqueous solutions up to 150°C or higher has been designed and constructed. Preliminary data show that the osmotic coefficients of alkali chlorides and bromides (except for the lithium salts) change very little with temperature; those of sodium fluoride and sulfate increase with temperature; while those of magnesium and uranyl sulfate decrease markedly with increasing temperature. A recording spectrophotometer has been adapted for high temperature work. It has been shown that the absorption bands for neptunium (at least the pentavalent state) remain sharp and change little in magnitude up to at least 250°C. An analytically useful absorption band has been found for hexavalent neptunium — in the near infrared region at 12,230 Å. The absorption by the solvent D_2O in this region is small, in contrast to H_2O, making the former a much more useful solvent for spectrophotometric studies; this factor also affords a sensitive method for analyzing for H_2O impurities in D_2O.

Spectrophotometry of Aqueous Solutions of Neptunium up to 250°C

The operation of aqueous homogeneous reactors at temperatures of 250°C and above made unusual demands upon current knowledge of chemistry. Before even considering such a reactor seriously, it was necessary to determine experimentally the limits of stability of the proposed fuel solution, uranyl sulfate in water. If such reactors are operated for reasonable times at significant power levels, they will accumulate sizable quantities of transmutation products of uranium. An important

one of these is neptunium, and it is therefore urgent to learn as much as possible about its chemical behavior in water at these high temperatures. Since neptunium may occur in several valence states, a prerequisite of such studies is an analytical method for each valence state separately at the temperature of interest. Spectrophotometry early suggested itself as such a method, but no instrument existed for making such studies safely. Dilute aqueous solutions at 250°C exert pressures of around 30 atmospheres, and neptunium-237, the long-lived isotope of the element, is a hazard in experimental studies because of its radioactivity.

A suitable light absorption cell and associated equipment has been designed and built for operation under these conditions, using a commercial spectrophotometer for the light measurement. With it, a solution of a neptunium salt has been studied from room temperature to 250°C. This is the first time that such studies have been possible, and the results demonstrate that spectrophotometry can indeed be used to follow the concentrations of the various valence states of neptunium.

Physical Chemistry of Ion Exchange

An important and relevant general objective of the program of basic investigations of the properties of synthetic organic ion exchangers is to reduce the large degree of empiricism now existing in the numerous AEC applications of these materials. At the Oak Ridge National Laboratory a basic thermodynamic equation for the prediction of cation and anion exchange equilibria has been established. This equation shows how the ionic selectivity of an exchanger at constant temperatures will depend on its polymeric structure, on its ionic composition (i.e., "loading")

and on the composition and concentration of the external electrolyte solution with which it is in equilibrium. The generalization afforded by this equation is most important because it is now possible to estimate desired ion-exchange equilibrium constants for a very wide range of conditions.

An understanding of the factors governing the rate of attainment of ion-exchange equilibrium is also of great importance to the practical applications of ion exchangers. The transport of ions in an anion or cation exchanger either in the presence or absence of an electric field gradient has been shown to be governed by a diffusional and not by a chemical exchange mechanism. This diffusion rate is dependent on the polymeric structure of the exchanger, on the temperature and on the charge carried by the ion to a marked degree. The theory of absolute reaction rates has been applied successfully to the interpretation of these ionic diffuison phenomena.

The ultimate aim of these basic researches is to connect the thermodynamic and rate theories of ion exchangers to theories of polyelectrolytes upon which statistical thermodynamic calculations of all the relevant properties of ion exchangers may be based.

High Temperature Ion Exchange

A systematic evaluation of ion exchangers in high temperature aqueous solutions (up to 200°C) has been performed. The temperature stability of these exchangers was found to be considerably better than anticipated. The standard cation exchangers of the Dowex-50 type in the sodium form showed little decomposition even at 200°C; in the hydorgen form they were less stable; and noticeable, though not severe, decomposition

occurred at 150°C. The anion exchanger Dowex-1 in a salt form (chloride) was readily usable up to at least 150°C. Since, in addition, the temperature coefficients of approximately a dozen simple ion exchange equilibria were found to be small, operation of ion exchangers in pressure systems and prediction of their performance should not offer any special difficulties.

Because of their stability, ion exchangers can be used at high temperatures for elucidation of the properties of the aqueous phase in much the same way as they are currently widely used at room temperature. Thus ion exchange becomes one more technique for the detailed study of solutions at high temperature.

Studies on the Action of Corrosion Inhibitors

It has long been known that the corrosion of mild steel in water containing air may be inhibited by the presence of a surface layer only a few atoms thick. This layer forms spontaneously when certain substances, called inhibitors, are present in quite small amounts in the water. Previously, little was known about how inhibitors act to cause such a great reduction in the rate of corrosion. At ORNL, studies of the chemistry of corrosion led to a hypothesis which suggested that, of all the chemical elements, the one having the most suitable combination of properties for an inhibitor should be the man-made element technetium. This element is not known to occur naturally in the earth, but is produced in nuclear fission. By use of technetium recovered at Oak Ridge from fission products, the prediction was fully confirmed. Specimens of mild steel have been kept for over five years without detectable corrosion, and the inhibitor is effective even at

250°C, at which temperature chromates, one of the best inhibitors, are destroyed by steel. The radioactivity and long life of technetium made it possible to conduct various studies not previously made, and these with related experiments, have contributed considerably to the understanding of corrosion inhibition. Among other results, it was demonstrated that the inactive state produced by these inhibitors is electrochemically the same as that on iron "passivated" by nitric acid or passage of an electric current.

Studies on the Electrochemistry of Stainless Steel

The unique property of the stainless steels is their ability to become "passive", or resistant to corrosion, under conditions where mild steels corrode rapidly. The corrosion resistance of stainless steels depends upon the relative rates of different possible electrochemical processes, some of which may be conducive to passivity and others to activity. A theoretical study was made in which the equations applicable to the rate of a simple electrochemical process have been amplified to cover complicated systems such as a composite metal (alloy) in a complex, corrosive, environment. In experimental studies, the conditions leading to passivation of the type of stainless steel used in the homogeneous reactor were determined. Very precise measurements of current-potential relationships were made for both stainless steel and Zircaloy-2 in sulfuric acid, and the application of the mathematical theory to these data yielded information of value regarding the mechanism of the electrochemical processes involved. In a few instance, measurements were made while the system was exposed to intense gamma radiation, in order to determine the effect of such a change in the environment.

Neutron-Diffraction Studies

A new technique in the field of structural chemistry is the use of intense neutron beams for the crystallographic examination of hydrogen-containing compounds, which cannot be studied effectively by any other method. The experiments utilize conventional diffraction techniques, but make use of the wavelike properties of the neutron. From the information on spatial distribution of hydrogen atoms in molecules, more may be learned about hydrogen bonding. Allied to these studies is that of the magnetic scattering of atoms in magnetic materials, from which data may be obtained about the detailed electronic structure of the atoms in a crystalline lattice.

Neutron Diffraction

Neutron diffraction studies were extended over an increasing range of inorganic and organic materials. Among the outstanding results were the clarification of the ice structure, the elucidation of the nature of the ferroelectric transition in KH_2PO_4, the analysis of the molecular structure of oxalic acid dihydrate, and the extension of neutron diffraction to the biologically interesting amino acid derivative, acetyl glycine. The determination of thermal vibration parameters allowed considerable insight into the thermal motions of the crystal.

The structure of ice is of great intrinsic interest not only because of its widespread occurrence, but also because of the intimate relationship of water to much of chemistry. Theoretical predictions of the distribution of hydrogen atoms along oxygen-oxygen bond lines had been challenged on several occasions. The neutron diffraction study established that on the average hydrogen atoms were indeed equally distributed in two

equivalent positions along O-O bond lines and that this was true at widely separated temperatures. These findings were thus in essential agreement with a picture of a dynamically disordered ice structure. A careful analysis of the thermal motions in ice gave clear indications of hydrogen-oxygen bond stretching and bending vibrations in excellent agreement with expectation.

Infrared observations of hydrogen bonded crystals indicated a functional relationship between O-H stretching frequency and the length of the O-H--O bond. Neutron diffraction bond length measurements made on similar crystals over the past several years showed that there is indeed also a relationship between O-H bond length and O-H--O distance. Thus the shorter the O-H--O bond, the longer the O-H bond and, apparently, the greater the tendency for the proton to be equidistant from the two oxygen atoms.

The Structure of Liquids by X-Ray and Neutron Diffraction

A study, by means of X-ray and neutron diffraction, of the structure of various liquid systems at both room and elevated temperatures was performed at the Oak Ridge National Laboratory. Because of the importance of high temperature fluids, the nature of the arrangement of the ions in fused salts, in particular the more simple ones, the molten alkali halides, was investigated. As generally expected, the absence of a long-range order was ascertained, and the packing of ions of opposite charge as nearest neighbors around a given ion was found to be less dense than in the crystalline solid at the melting point, yet with a smaller average interionic distance. These results were in good agreement with the concept of the importance of both disorder and

existence of holes as particular characteristics of the structure of a liquid.

Aqueous solutions containing tightly bound groups of atoms, such as the uranyl group, were also studied, with particular emphasis on the question of polymerization of such groups. In confirmation of chemical and thermodynamic evidence, X-ray diffraction showed uranyl fluoride solutions to contain polymers embracing at least two, and probably three, uranyl groups, but also proved that polymers were absent in uranyl nitrate or perchlorate, and in acid bismuth chloride solutions. The results for bismuth oxyperchlorate solutions were consistent with a proposed hexamer containing octahedrally arranged bismuth atoms.

Pure Quadrupole Spectroscopy

Studies in pure quadrupole spectroscopy have been pursued since shortly after the announcement of the first successful observation at ORNL. The method has been applied to physical and chemical problems. In the latter case the quadrupolar nucleus has been used as a probe to evaluate features of the electric charge distribution in chemical bonds. The various studies have included a precise determination of the nuclear quadrupole moment of I^{129} (compared to I^{127}) which was the first such measurement on a radioactive isotope. The earlier work on chlorine resonances in substituted methanes has been extended to other alkyl halides as well as to bromide and iodine resonances in simple molecules. The data have been correlated with features of the chemical bond in these substances.

Zeeman studies of the quadrupole spectrum for chlorine in sodium, potassium and barium chlorates have been carried out. The results for

the sodium salt which has very high symmetry were shown to be consistnet with existing theory and the known crystal structure. In the case of potassium and barium chlorates the measurements showed the electric charge distribution about the chlorate ion to be remarkably axially symmetric even though it is not required by the crystal structure, and it was demonstrated that the orientation of the chlorate ions in the crystal lattice could be accurately determined. Zeeman studies on the iodine resonance in iodic acid showed a number of unusual features for the first time. Among these were extra spectroscopic lines that originated from a very weak interaction of the proton with the iodine nucleus, and a detailed study has showed that the location of the proton in the crystal lattice can essentially be determined. At this time this is the only known case of a direct magnetic dipole-dipole splitting that has been studied in quadrupole spectroscopy.

Paramagnetic Resonance Studies of Free Radicals Produced by Radiation

The paramagnetic resonance method has been applied to radiation chemistry in an effort to learn something of the basic processes taking place immediately after the absorption of radiation but before the final chemical products have formed. Often one of the early radiation steps is the breaking down of molecules into smaller fragments which are chemical free radicals. The paramagnetic resonance method is a physical technique that in some cases will allow a direct observation of the free radicals to be made by virtue of their magnetic properties. In applying the method, the sample containing free radicals is placed in a magnetic field and the frequencies at which microwaves are absorbed are measured.

Radiation chemical studies have been made by cooling various samples of interest in liquid nitrogen (-196°C) and irradiating with gamma rays from cobalt-60. In this way free radicals are frequently formed and stably trapped in the rigid, cold solid. The sample is then examined while still cold in a paramagnetic resonance spectrometer.

Various concentrations of sulfuric, phosphoric and perchloric acids irradiated in this way have shown a number of characteristic microwave absorptions, or lines, but one pair of lines was common in the spectra of all three acids. This pair of lines has been clearly demonstrated to arise from the presence of free atoms of hydrogen stably trapped in the cold materials. Subsequently atomic hydrogen has been found in a variety of other substances irradiated at a low temperature.

A number of properties of atomic hydrogen prepared in this way have been studied. Upon prolonged irradiation the atomic hydrogen concentration increases to limiting steady state value. In the three acids this concentration is reached after about 150 hours of irradiation in a 1000 curie cobalt-60 source and amounts to roughly 0.1 per cent of the hydrogen converted to atoms. Upon warming, the atomic hydrogen is found to rapidly disappear as it enters chemical reactions to form final chemical products. In sulfuric acid, for example, the atoms disappear in a matter of minutes when warmed to -175°C. The rates of disappearance have been measured at various temperatures which give valuable clues as to the mechanism for the disappearance.

Gaseous hydrogen is known to be one of the products formed in the irradiation of these acids at room temperature. In order to ascertain whether or not the mechanism for the disappearance of atomic hydrogen was the reaction of two such atoms to form a molecule of hydrogen gas,

analyses have been made on the amounts of atomic hydrogen formed by a known amount of irradiation and also analyses made on the amount of hydrogen gas released upon warming the acids. In sulfuric acid there is a very striking correspondence between atomic hydrogen formed and hydrogen gas released upon warming which substantiates the proposed mechanism. This is probably the only radiation chemical study in which an intermediate has been identified and measured by a direct method and its relation to the final chemical product demonstrated.

Use of Molecular Beams in Studying the Mechanisms of Chemical Reactions

It has long been recognized that much could be learned about the details of gaseous chemical reactions if the reactants could be brought together as crossed molecular beams. Then each product molecule would result from a single collision, and the complications of chain reactions and of wall effects could be obviated. Such an experiment has been carried out at ORNL, between beams of potassium and of hydrogen bromide. The usual parameters of reaction kinetics, activation energy and steric factor, were measured with good precision under these ideal conditions. In addition, the angular distribution of the potassium bromide produced was measured, and some conclusions drawn therefrom about the relative effectiveness for reaction of various orientations of the colliding reactants. This is one of the kinds of information that can be obtained only by this technique.

METALLURGY

The program of metallurgical research was established at ORNL in 1946 to carry out applied research primarily related to the reactor program. The need for such work was stressed by experience with the Laboratory's reactor. During the previous years, operation of the ORNL graphite reactor had been complicated by ruptures in the aluminum jackets around the uranium fuel pieces. Metallurgical research resulted in the development of a new technique for canning the uranium that materially reduced the rupture problem.

Fuel-Element Development

The design of the MTR developed at ORNL called for a type of fuel element that had never been made before — plates of uranium-aluminum alloy clad with aluminum. Successful methods of fabricating these new fuel elements were developed, and today ORNL-developed fuel elements are used in the MTR, the LITR and in most swimming-pool reactors as well, since the latter also use MTR-type fuel elements. Improved fuel elements have also been developed at ORNL for the Argonne CP-3 and CP-5 reactors and the Convair Shield Test Reactor.

Beryllium Research

Another problem that arose in connection with the MTR was that of producing and fabricating beryllium for the reflector. A method of producing beryllium of suitable quality by powder-metallurgy techniques was developed at the Brush Beryllium Company, who then produced the MTR beryllium pieces under ORNL supervision. Successful methods of fabricating beryllium metal pieces to close tolerances were developed

at the Laboratory, and a beryllium-machining shop was set up at the Y-12 plant to do this work. The highly successful operation of the Materials Testing Reactor would have been impossible but for the accomplishments of research and development metallurgists working on fuel elements and beryllium production and fabrication.

Powder-Metallurgy Applications

Since powder-metallurgy techniques are readily adaptable to preparation of solid fuel elements and control rods for high-temperature reactors, the Metallurgy Division has developed a well-equipped facility for this work. Solid fuel elements, consisting of uranium oxide dispersed in a stainless steel matrix, have been fabricated. Control-rod elements have been prepared for the Aircraft Reactor Experiment, the Homogeneous Reactor Experiment, and the General Electric Aircraft Reactor. The control rods for the Aircraft Reactor Experiment consisted of boron carbide dispersed in iron and canned in stainless steel.

Reactor Materials Research

The homogeneous reactor project introduced new materials requirements which had to be met by materials developed through metallurgical research. Operation at 250°C under 1000 pounds per square inch pressure and in a corrosive medium imposed severe strains upon the reactor structural materials. Methods of treatment to reduce corrosion and successful procedures for fabrication, welding, and inspection were developed to assure that the reactor system would stand up under conditions of operation. Similar problems were encountered in connection with the aircraft reactor, where the structural material had to have strength and corrosion resistance while operating at red heat (1500°F). Here again, problems

of fabrication, welding and improving corrosion resistance were solved successfully.

Welding and Brazing

The requirement that complicated fuel systems and heat exchangers for liquid fuel reactors be leak-free and reliable in operation demonstrated the need for research and development in improved welding and brazing techniques. Techniques were developed for high-quality, reproducible welding of small-diameter tubing to header plates. This technique, known as "cone-arc welding" was extremely useful in fabricating the type of liquid sodium-to-air heat exchanger used for heat-transfer experiments in the Aircraft Reactor Program.

Research and development on high-temperature brazing alloys yielded several new brazing alloys having excellent corrosion resistance to various liquid coolants.

Physical Properties Research and Alloy Development

A great many possible structural materials were tested for strength at high temperatures, corrosion resistance, and other metallurgical properties. From these tests, materials were selected and improved for use in the two liquid-fuel reactors. Further work is being performed to develop still better materials and methods of fabricating them into reliable reactor systems.

Zirconium is a metal which has an attractively low neutron-absorption cross section and which has a sufficiently high melting point to give promise of its use in high-temperature reactor systems. Research was directed toward developing high-strength, heat-treatable zirconium alloys which are analogous to the high-strength, heat-tractable steels.

This program developed certain high-strength zirconium alloys which can be used in reactors where the high cross section of Inconel or stainless steel is undesirable and where the temperatures are too high to permit the use of magnesium or aluminum alloys.

A similar program of metallurgical research and development on titanium was undertaken because of its high-temperature characteristics and corrosion resistance.

Ceramic Materials

Ceramics offer a possible solution to some of the materials problems encountered in the construction of high-efficiency, compact reactors operating around 1000°C and above. The Ceramics Laboratory at ORNL is the only integrated group devoted to ceramics research among the various installations of the Atomic Energy Commission. The program of ceramics research and development included the investigation of new or unique ceramic materials for possible application in the nation's nuclear energy program, and adaptation of existing materials for this service.

An outstanding accomplishment of the Ceramics Laboratory was the development of a fabrication technique for the weapons program which greatly simplified the preparation of certain weapon components, at substantial savings in costs and manufacturing time, over other proposed methods.

Fundamental Metallurgy

Although almost all metallurgical research and development has been related to the reactor program, fundamental metallurgical research was undertaken on a small scale, particularly for important metals, like

thorium and uranium, whose metallurgy had not been completely explored.

One of the most important difficulties encountered in production reactors arises from dimensional instability of uranium. This dimensional instability was shown to vary markedly with the degree and nature of the orientation of the crystals which make up the uranium. A method was devised for determining preferred orientations in uranium both more rapidly and more accurately than was previously possible. Significant progress was made toward connecting various preferred orientations with the techniques used in fabricating uranium. This work contributed valuable advances with dimensional stability problems encountered in nuclear reactors.

In practical application, a rolling procedure has been developed for zirconium alloy plate which gives essentially no preferred orientation and therefore uniform properties in all directions, a condition desired in metals for some reactor vessels. On the other hand, it has been found possible by control of extrusion conditions to produce a texture which gives a desirable 70% increase in the longitudinal strength of aluminum and thorium rod.

Fundamental Physico-Metallurgical Research

The phase relationship in the alloys of the important reactor metals, zirconium and titanium, have been rationalized on the basis of the electron concentration and of the difference in size of the solute and solvent atoms. The correlations so obtained should permit prediction of alloy behavior in systems not yet investigated.

The response of metals to fabrication and service stresses is in large measure related to the preferred grain orientation induced by mechanical deformation and recrystallization. An intensive study of a

number of metals of interest in reactor technology, aluminum, thorium, and uranium indicated that a duplex texture with the attendant complex behavior may be a result of partial recrystallization rather than a complicated mechanism of deformation.

Some of the lanthanide and actinide elements, such as cerium and thorium, were found to show novel allotropic transformations at low temperatures. A rather thorough kinetic study of the reactions in cerium revealed very unusual behavior in the so-called diffusionless transition category, e.g., coexistence of three phases, two of which have the same crystal structure.

Thorium Research

Since thorium is of importance as a source material for breeding of U-233, an extensive investigation has been made of its metallurgical characteristics, such as physical and mechanical properties, alloying effects, and the effects of hot and cold working. The fabricability of thorium by extrusion, rolling, drawing, and swaging has been demonstrated.

High Temperature Reactions of Metals

Oxidation is a limiting factor in the reactor application of a number of potentially useful metals and is an important problem in high temperature reactors that employ aqueous solutions. It has been found that the non-protective oxidation of niobium and tantalum is related to microscopic cracks in the oxide films, presumably having their origin in the stresses induced by anion diffusion.

Other work, on the oxidation of sodium, potassium, and rubidium, has proved that the theoretical criterion for protective oxide formation on metals needs to be drastically revised.

Fused NaOH is a very useful stable fluid moderator, but its utility is limited by its reaction with the metals and alloys employed to contain it. A study of the corrosion mechanism has revealed the container materials to suffer a novel form of stress corrosion. NiMo alloys have been found to be most resistant to this attack.

The interactions and the atomic and electronic configurations of the species existing in fused salts have been studied by means of absorption spectrophotometry. Complex ions and other forms of cation-anion interaction have been observed.

Ceramic Research

The desire to operate reactors at high thermal efficiency makes ceramic materials of keen potential to this field, which is always in want of more suitable materials of construction. ORNL has produced UO_2 crystals which are compatible with aluminum and of high density (95%) UO_2 for fuel elements of the proposed gas-cooled and maritime power reactors.

Reprocessing of Fuel Elements

To assist chemical reprocessing of stainless steel-base fuel elements a metallurgical method of carburization was developed to destroy the chemical inertness of the stainless steel, making it amenable to selective dissolution and permitting high uranium recoveries by conventional chemical reprocessing techniques. The large fraction of stainless steel retained as chemically inert solid waste alleviated the long-term waste disposal problem.

Disposal of Radioactive Wastes

Ceramic techniques have been used to develop a method of radioactive waste disposal whereby fission products are incorporated in a non-leachable sintered mass of shale, limestone, and soda ash, which can be conveniently stored or buried.

SOLID STATE PHYSICS RESEARCH

Originally a facet of the metallurgical research program, solid state physics research at ORNL has grown in scope, as the severity of radiation damage problems was recognized with increasing clarity. Like the metallurgical research effort, solid state physics research is strongly influenced by the problems of specific reactor systems but with increasing attention to the fundamental physics of solids involved in understanding the effects of radiation.

Radiation Damage Investigations

Accumulated experience with existing reactors has demonstrated that prolonged exposure of a solid in a nuclear radiation field (i.e. within a reactor or other nuclear device) may markedly alter the desirable properties of the solid, often in a definitely deleterious fashion. These changes in physical properties result from the production of defects in the crystalline lattice of solids by collisions of the energetic particles (neutrons, fission fragments, photons, etc.) which make up the nuclear radiation field. Therefore, if existing materials are to be used to their fullest extent and new ones are to be found which will be even more radiation resistant, it is essential that the fundamental nature of radiation damage be well understood. A knowledge of

the nature of radiation-induced lattice imperfections and the mechanisms by which these influence the behavior of solids will suggest means whereby the deleterious effects of radiation on existing materials may be minimized and, in addition, will indicate the most profitable approach for future developmental research devoted to radiation resistant materials. A long range benefit which should not be overlooked also derives from these studies as a by-product in that radiation damage studies will yield and have already yielded much valuable information about the fundamental behavior of solids in the absence of radiation.

Irradiation Effects in Semiconductors

Because of the sensitivity of their electrical properties to lattice disorder produced by fast-neutron bombardment, the semiconductors silicon and germanium have been chosen as important tools in studies of irradiation effects in solids. By correlating the changes in electrical properties with lattice disorder that occur during irradiation in the nuclear reactor, information is gained that, (1) aids in the construction of a valid model of radiation damage, and (2) yields of fundamental information concerning the modification of properties, such as concentration and mobility of charge carriers, due to lattice defects.

Basic Irradiation Studies in Alloys

Neutron irradiation affects the properties of alloys in two ways. First, collisions between the impinging neutrons and the atoms of the solid produce lattice defects such as vacancies and interstitials, which reduce the crystallinity of the solid. This effect takes place in pure metals as well as alloys and is largely independent of the temperature of irradiation. The second effect of neutron irradiation

is to enhance certain diffusion-controlled solid state reactions. These reactions are of particular importance for alloys, since many of their special properties, such as hardness, electrical resistivity, or magnetic permeability, depend upon the extent to which such reactions are permitted to progress during the preparation of the material. Examples of such diffusion-controlled reactions are ordering and precipitation from solid solution.

The effect of neutron irradiation on a diffusion-controlled reaction of this type was studies in Cu-Al alloys. It was found that the electrical resistivity of Cu-Al alloys decreased sharply in the early stages of neutron irradiation at 40°C. This effect decreases with decreasing aluminum content and is absent in pure copper. Thus, the effect was established as one having to do with the alloy and not with the pure metal. Furthermore, the decrease in resistance was not observed when the irradiation was carried out at -120°C instead of 40°C. Thus, it was shown that the reaction was diffusion-controlled, because at the lower temperature atomic mobilities would otherwise have been severely reduced. It was observed that irradiation effect can remain latent in the material, for when the alloy was allowed to warm after having been irradiated at -120°C, a decrease in resistance was observed. The rate at which the decrease in resistance took place as a function of annealing temperature was studied and the activation energy for the process was found to be about one electron-volt. It was found that a similar decrease in resistance took place after rapidly cooling (quenching) the alloy from a high temperature. This process also had an activation energy of about one electron-volt. However, the decrease in resistance after quenching took place at temperatures about 65°C higher than the

temperatures for the decrease in resistance after cold irradiation. The inference was that essentially the same process was being observed in the quenching and irradiation experiments but the neutron irradiation endows the material with the ability to carry out the process at lower temperatures than are possible in the absence of radiation. The apparent explanation was that diffusion in solids occurred chiefly by a vacancy mechanism and was accelerated as a result of the additional vacancies produced by irradiation.

The significance of this work primarily concerns the fact that alloys are frequently prepared to have optimum properties by allowing diffusion-controlled reactions such as ordering or precipitation to occur to a critical point. Then the solid is returned to room temperature and diffusion is relied upon to be sufficiently slow at service temperatures that the properties of the solid remain stable. However, the experiments indicated that reaction temperatures may be significantly lowered by neutron irradiation and, as a consequence, the important properties of irradiated alloys may deteriorate at temperatures at which they are normally thought to be stable.

Decomposition of Metastable Alloys

Many structural alloys derive their useful properties from the fact that by appropriate pretreatment they can be maintained in a metastable condition. The so-called austenitic stainless steels are an excellent example of such metastability. By metastable it is meant that the alloy would prefer to exist in a different structural form but is prevented from doing so by minor alloying constituents or by the very small rate of diffusion, or motion, of constituent atoms permitted in the solid

state. It has been conclusively demonstrated that lattice imperfections, in particular vacant lattice sites, play a dominant role in solid state diffusion, since transport of atoms can occur by the easy exchange of positions of the vacant site and an adjacent atom. Bombardment with fast neutrons introduces lattice vacancies into the solid by simply knocking atoms out of their normal positions. This increased concentration of mobile defects markedly increases the rate of diffusion and hence decomposes the metastable state, often causing it to lose desirable characteristics by increasing the rate of reversion into the more thermodynamically stable alloy form.

Studies on the behavior of a number of alloy systems including copper-beryllium, nickel-beryllium, copper-gold, copper-zinc, and copper-aluminum have demonstrated that phase transformations occur and that fast neutron bombardment enhances to a considerable degree the rate of solid state reactions. These results indicate that considerable care must be exercised in choosing alloys for reactor construction in order to prevent unwanted metallurgical changes from occurring.

Mechanical Behavior of Metals

The tensile strength and plastic deformation of metals arise from a specific type of one-dimensional lattice imperfection called a dislocation, which is able to move under a critical applied stress, thus allowing the metal to yield. If the metal is subject to an alternating or cyclic stress field, the dislocation oscillates, thereby degrading the vibrational energy in the form of heat. This degradation is known as internal friction. It is well known that fast particle bombardment causes metals to harden, thus increasing the stress necessary to cause

the metal to flow. This hardening effect is particularly pronounced in the case of a pure metal such as copper. Experiments which were conducted at both low temperature ($20^{o}K$) and room temperature have thrown considerable light not only on the mechanism of radiation hardening but on the mode of dislocation as well. It was shown that radiation hardening by fast neutron bombardment is caused by the interaction of lattice imperfections with the dislocations. The imperfections act as barriers to the motion of dislocations. Studies of internal friction, using cyclic stress fields in the kilocycle range with very small amplitude, demonstrated that at room temperature imperfections migrate to the dislocations. Consequently, this property is extremely sensitive to radiation and perceptible changes in internal friction have been observed at exposures as low as 10^9 neutrons/ cm^2 ($\sim$0.001 second at the center of the ORNL graphite reactor).

This reduction of internal friction formed the basis of an instructive exhibit at the Geneva Conference. Two high-purity, single-crystal copper tuning forks were prepared, one of which was bombarded with fast neutrons in the graphite reactor. When struck with a wooden clapper the irradiated fork rang as though it were bronze; whereas the unirradiated crystal did not.

Experiments on Semiconductors

The great sensitivity of certain semiconductors to the disordering effects of nuclear radiation has made them excellent materials with which to study the nature of lattice imperfection produced by reactor radiation. The investigation of the influence of lattice defects on the electrical behavior of germanium has been extended to include many

other semiconductors such as silicon, indium antimonide, gallium antimonide, indium arsenide, etc., which show promise for use in electronic devices. A sensitive electrodynamic balance (sensitive to weight changes of 2×10^{-7} grams) has been used to measure the magnetic susceptibility of semi-conductors. These data have elucidated the electronic structure of semiconductors and, after bombardment, have revealed the magnetic nature of lattice defects introduced by fast neutron bombardment. Other experiments have shown that gamma rays from a cobalt-60 source are capable of introducing defects which alter the electrical properties of germanium in much the same way as, but at a considerably smaller rate than, fast neutrons. Information gained from these fundamental studies has been used to explain the drastic and deleterious effect of reactor radiation on semiconductor electronic components (diodes and transistors) and have indicated possible ways in which the radiation resistance of such devices might be improved.

Thermal Conductivity of Non-Metallic Crystals at Low Temperatures for the Study of Lattice Defects

The thermal conductivity, K, of single crystals is very sensitive to the presence of lattice defects. Point defects, such as interstitial atoms, vacancies, or impurity atoms result in a reduction in the value of thermal conductivity in an otherwise perfect crystal. Dislocations, introduced by bending a crystal, result in a decrease in K at low temperatures. However, there is a qualitative difference in the thermal resistivity introduced by point defects on the one hand and dislocations on the other. Furthermore, it is possible by measurements of K to ascertain whether the lattice defects are clustered together, thereby acting as a scattering region that is large compared to isolated point

defects.

The low temperature thermal conductivity has, consequently, been a useful property in studying the nature of lattice defects, whether present in the original crystal or purposely introduced by irradiation with Co^{60} gamma rays, fast neutrons, or thermal neutrons.

The difference in the type and distribution of defects produced by Co^{60} gamma ray bombardment and bombardment by fast neutrons is being studied by examining the resulting thermal conductivity changes in high purity single crystals of KCl, CaF_2, MgO and SiO_2. Using LiF single crystals, the effects of successive thermal neutron bombardment on the resulting K decrease at low temperatures are being studied to obtain information with regard to reported structural changes in LiF. The difference between thermal neutron damage of LiF and Co^{60} gamma ray damage of the same material will be ascertained.

The structure of quartz crystal and fused silica are altered by fast neutron bombardment. Both, upon bombardment, approach a limiting density intermediate between that of the two forms, suggesting an intermediate state of order. The low temperature thermal conductivity, which is sensitive to crystalline order was measured on fused silica before and after successive exposures to a maximum dosage of 7×10^{19} fast neutrons/cm^2. The resulting increase in K and density indicates that the short range order in silica improves with fast neutron bombardment.

Low Temperature Studies of Radiation Damage in Detail

The disordering effects of fast particle bombardment on the crystalline lattices of solids may be almost completely removed by appropriate heat treatment. In the case of copper, annealing above 350°C is sufficient to remove the radiation hardening. In fact, for a soft

material such as aluminum, radiation effects anneal almost completely at room temperature. A more careful investigation has shown that the lattice damage retained by most solids bombarded in the room temperature range is only a small portion of that expected to result from a given bombardment in the reactor. This indicates that a considerable portion of the damage tends to "heal" itself. In order to understand more completely the nature of the lattice disordering processes and to gain some idea as to the extent and configuration of the lattice damage remaining at room temperature, it is necessary to conduct bombardments in the reactor at temperatures sufficiently low to "freeze-in" all of the lattice defects resulting from a fast neutron collision within the specimen and, in addition, to study the kinetic processes involved in the annealing or "healing" of damage as the specimen is warmed to room temperature. Therefore, a refrigerated reactor chamber was constructed in which specimens could be irradiated at temperatures as low as $10^{\circ}K$. The reactor cryostat consisted of a vacuum-jacketed tube extending 20 ft into a vertical hole of the reactor. Cooling is supplied by a cold stream of helium gas which circulates in a heat exchanger surrounding the specimen chamber. The circulating helium is the refrigerant fluid in an expansion-engine type of refrigerator unit, and the cooling capacity is such that temperatures less than $10^{\circ}K$ under no heat load can be obtained.

This low temperature facility has been employed in the study of a variety of metals during exposure. The rate of damage as indicated by the change of electrical resistance has been shown to vary by as much as a factor of 10^3 in going from good metals such as copper to poor ones such as bismuth. Measurements of tensile properties and internal friction

during and subsequent to exposure at the low temperature and upon warming have been of great assistance in understanding the nature of radiation hardening of metals. Finally, in the case of copper and aluminum, the energy stored in the form of lattice disorder during irradiation below 20°K has been measured on warming the specimens at a controlled rate. A comparison of the surprisingly low amount of energy released with the recovery of the radiation-induced increase in resistivity reveals that current theories of the nature of fast neutron induced lattice disorder and of the recovery or healing process that occurs during warming are inadequate.

Radiation Damage in Refractory Non-Metals

Since the materials problems for high temperature reactor design must be solved before economical power reactors can be constructed, a knowledge of the behavior of refractory solids in intense radiation fields is essential to progress in this area. For this reason much effort has been spent in fundamental studies of certain nonmetals which may be considered prototypes of refractories. Both electronic properties and structural behavior have been examined in some detail. The former, in particular the optical and magnetic properties, are important since changes in these may be interpreted to give information about the nature of the bombardment-produced defects. Studies of changes in structure also yield such information; but, even more important from an applied standpoint, these yield information about the dimensional and phase stability of certain crystals and crystal structures which is essential in choosing materials for reactor construction. By means of density measurements and X-ray diffraction techniques, a wide variety

of refractory crystals has been studied. It has been found that nearly all crystals studied expand, i.e. decrease in density. One of them, namely quartz, expands by as much as 14% and at the same time loses its crystal structure completely, becoming a glass. Other materials of which zirconia and barium titanate are examples change from a structure of low symmetry to one of high symmetry which is normally thought to be stable only at high temperatures. Here, as in the case of studies on alloys, fundamental research has indicated that care must be exercised in choosing ceramic materials for reactor construction.

Fusion Research

In 1953 ORNL initiated a small theoretical study of some approaches to the achievement of controlled thermonuclear reactions. Utilizing mainly the services of consultants, ORNL pursued these studies to a point where it appeared that experimental research and development on a new approach to the achievement of controlled thermonuclear fusion was justified by the promise indicated from the theoretical studies. The broad background of experience with the electromagnetic separations and with the development and operation of high current cyclotrons led ORNL rather naturally into the use of energetic ion injection as its major approach to achievement of a hot plasma. Small scale research and development activities were started in 1955 and were significantly expanded in 1956 as results continued to show promise. By early 1957 a technological breakthrough had been achieved in the development of high current carbon arc that would efficiently disassociate D_2^+ ions so that they would be effectively trapped inside the magnetic field. With this scheme, molecular ions could be accelerated to an energy well in excess of that needed

in the high temperature plasma. An experimental thermonuclear device called the Direct Current Experiment (DCX) was constructed in the early part of 1957 to permit more detailed investigations of the phenomena encountered in a fusion device of this type. Molecular deuterium ions were injected in the DCX at energies up to 625 kev. Near the edge of the magnetic field, the arc breaks up the beam, yielding 300 kev deuterium ions which are trapped, plus energetic deuterium atoms which pass on through the field. The deuterium ions are trapped in the magnetic field initially in an organized ring, which gradually spreads out as the motion of the ions becomes random through collisions. Experimental operation of the DCX was achieved in July 1957, and successful trapping of the accelerated beam was accomplished on August 21, 1957.

Later in 1957 another technological breakthrough came as a result of development of the concept of "burnout" of neutral atoms as a necessary initial condition for the formation of a thermonuclear plasma in the DCX. This concept was elucidated until it was possible to predict the injected beam current and vacuum that would have to be achieved before burnout would occur. For example, with a spherical plasma volume of a radius equivalent to that of 300 kev deuterons in a magnetic field of 10,000 gauss and with a pressure of 10^{-6} millimeters of mercury in the plasma region, a deuterium ion injection rate of 80 milliamperes is required to achieve burnout. The better the vacuum, the lower the injection current can be.

The vacuum conditions in DCX, which have reached 3×10^{-8} mm Hg, will be improved further and the incoming beam current increased until the residual gas in the plasma region can be completely ionized by the incoming beam. When this condition (called "burnout") is reached, the

trapped ions will be lost much more slowly, and a plasma of hot ions and electrons is expected to form. The attainment of "burnout" condition will be a major event.

The plasma formed after burnout is reached should have an effective temperature which may be as great as 250 kev, and thermonuclear neutrons should be produced to such an extent that the neutron level, which before burnout will be principally produced in the walls and neutral gas of DCX, will rise by a factor of 10^4 and 10^5. This distinct rise in neutron level will indicate the production of a definite thermonuclear reaction, which will be another major event.

The magnetic fields in the DCX machine are too small to contain the reaction products of the thermonuclear reactions, and a self-sustaining reaction cannot be produced in it. Another facility must be constructed with a stronger magnetic field and a higher mirror ratio is a self-sustaining thermonuclear reaction, fed by cold gas, is to be demonstrated. This second machine, ORION, will have a central field value of about 26 kilogauss and a mirror field of about 91 kilogauss. These fields in DCX are 10 and 20 kilogauss, respectively. The coils will be designed to permit increasing the field rapidly. The purpose of increasing the field is to compress the plasma associated with the trapped atomic ions, and to contain reaction products. It has been shown that molecular-ion injection must stop when the field is increased and neutral-atom injection will then be used. It will be possible to use energetic neutrals if they are needed. These neutrals will be ionized by collisions in the compressed plasma.

When the plasma has reached its maximum density and impurities in the working volume have been "cleaned up", cold deuterium and tritium

gas will be fed into the plasma and the energetic neutral beam stopped. A self-sustaining plasma, heated by thermonuclear reactions, is expected to result from this procedure. The system is extremely flexible, and several procedures will be tried. It may be possible to compress the trapped ions and inject cold gas immediately, thus eliminating the energetic-neutral injection phase.

When a self-sustaining plasma has been created, various schemes of power extraction will be studied. In addition to the power represented by the production of energetic charged particles, which most desirably would be converted directly into electrical power, that large amount of power that is dissipated by neutrons and radiation must be used. Many of the neutrons can presumably be trapped in a surrounding blanket containing lithium, for the further generation of nuclear power (about 5 Mev per neutron captured) and regeneration of the tritium burned in the reaction. A heat cycle will be necessary in order to convert power from these sources to a useful form.

Geneva Conferences

At the two international conferences on peaceful uses of atomic energy held in Geneva, Switzerland, in 1955 and 1958, ORNL placed a major role in providing exhibits to show recent advances in nuclear science and technology in the United States. The first conference emphasized the peaceful applications of reactor technology, and for this conference ORNL designed and constructed an operating research reactor of the "swimming pool" type. The reactor was designed, constructed, and test-operated at ORNL. It was then shipped to Geneva where it was installed in a building constructed under ORNL supervision at the conference site.

The complete job of providing the reactor and the building, from initial design to final public demonstration during the conference, was carried out in the five months between March and September, 1955. The Geneva reactor proved to be a major attraction among the exhibits and was called "the most beautiful reactor ever built."

For the 1958 Geneva conference, which emphasized progress in fusion research, ORNL again assumed responsibility for overseeing construction of the exhibit building and for providing major exhibits of fusion devices. ORNL constructed two full scale operating models of the DCX device designed for visual display of the basic operating principles. Through viewing windows, it was possible to see the injected ion beam and the trapped ring during actual operation of the device. Again in 1958, the Geneva exhibits were outstanding and provided a high point of interest during the conference. As in 1955, the building and exhibits for the 1958 conference were designed, constructed, and opened to the public in a period of five months.

BIOLOGY AND MEDICINE

Biology — Effects of Radiation on Living Cells

In the field of cytogenetics, it has been found at ORNL that the presence of oxygen increases the likelihood of chromosome breaks in living cells, that such an effect can be counteracted by a variety of chemicals, and that most of these chemicals seem to protect against radiation damage by removing oxygen. It has also been found that bacteria can be made to recover from the damaging effects of radiation by post-exposure treatment consisting of incubation at lower-than-normal

temperatures in the presence of certain nutritional factors. These factors can be isolated from meat extract or yeast extract, and appear to be related to the spleen factor which enables irradiated mice to recover from X-ray damage.

The photographs show three of the kinds of radiation effects which the Cytogenetics Section is using to study the basic nature of biological radiation damage and the mechanisms by which it is brought about. The center of interest is damage to those parts of cells - the chromosomes and nucleus - which control cellular processes and inheritance. Much attention is currently being paid to the role of molecular oxygen in the production of this damage. These studies have given important information about the complex processes by which radiation damage is produced.

In mammalian genetics and development research, it has been established that mutation rates in mice are considerably higher than expected on the basis of Drosophila (fruit flies) experiments, and this discovery requires reconsideration of the present tolerance dosage for man. Studies of the effects of radiation on embryonic mice give much information about the mechanisms of embryonic development, and data obtained are of considerable importance in determining the possible radiation effects on women exposed to atomic explosions or other radiation.

Extensive studies on radiation-induced cataracts and leukemis in mice are in progress. It has been found that cataracts in mice can be induced by as little as 15 rep of fast neutrons.

Radioiodine in larger doses depresses or destroys the thyroid gland, and research at ORNL has shown that this causes a disturbance in the normal hormonal balance and the thyroid-regulating gland (the pituitary) undergoes a tumorous enlargement. These pituitary tumors are capable

of growing in hosts whose thyroid function is depressed, and, if uncontrolled, in time they become autonomous cancers. These tumors secrete thyroid-stimulating hormones and cause great enlargement of the thyroid gland. Studies are in progress to learn about the factors of initiation and control of these and other tumors which can be caused by total-body irradiation or by radioisotopes.

New Research Tools

The technique of ion-exchange chromatography, well known in the field of inorganic chemistry (especially for rare earth separations), has been applied to the detection of the breakdown constituents of the nucleic acids, which are polymers approximating the size of proteins and which, like proteins, are largely known through their breakdown products. It quickly developed that this excellent tool, not before applied to the problem, could detect many fragments never before considered to be constituents of nucleic acids. These new natural products have now been prepared for further study. They have led to the concept that one kind of nucleic acid (ribonucleic acid) has the same general structure as the other (desoxyribonucleic acid) and not an entirely different one as previously supposed. Although it is yet to be demonstrated that the former type (concerned with cell growth and protein synthesis) is converted to the latter (a constituent of chromosomes), or the reverse, it is still a matter of considerable significance that the two types have so similar a basic structure.

Biological Discoveries

These methods have not only revised the concepts of biosynthesis of nucleic acids but have made possible new approaches to biological

problems. An additional by-product has been the development of methods for the separation by ion exchange of the neutral sugars by means of their borate complexes and also of the biologically important sugar phosphates.

A new phenomenon, the luminescence of green plants during photosynthesis, has been discovered. Luminescence from extracts coming from luminescent bacteria, a phenomenon searched for by many of the most outstanding scientists during the last 50 years, has been found in this laboratory. Studies on microbial metabolism have elucidated some of the mechanisms by which bacteria synthesize certain important compounds.

The Biophysics Group developed new methods of preparing and measuring beta-radiation source plaques and obtained much new information on diffusion of potassium into and out of irradiated erythrocytes. A difference in the effectiveness of 250 kv X rays and cobalt gamma rays in causing radiation damage has been found. New methods have been developed to obtain nucleic acids in practically native condition from a variety of cells. This highly polymerized nucleic acid showed extreme sensitivity to X rays very similar to the sensitivity of chromosomes in vivo.

Radiation-Induced Genetic Damage in Mammals

Large-scale experiments on the genetic effects of radiation in mice have been carried on for the past 10 years at the Oak Ridge National Laboratory. The most recent findings come from two groups of relatively long term experiments which use two different approaches to the problem

of genetic radiation hazard.

The first approach is essentially built around this question: "What basic factors affect the frequency with which mutations are induced by radiation?" The second group of experiments centers around the question: "What do mutated genes do to the individual and the population?"

Using these two approaches, some of the earlier Oak Ridge findings have been confirmed and extended. Thus, by the first approach, it has been confirmed that the mutation rate in the mouse is higher than in fruit flies; that there is no decrease with time in the probability of transmitting a mutation; and that the dose-curve drops from linearity at high doses, possibly because, at these doses, the rate may be measured in only the more resistant, surviving cells.

Using the second approach, earlier results indicating deleterious effects in first generation offspring of irradiated animals have been extended to include shortening of total life span and other effects.

New work with chronic gamma radiation has brought two major surprises. The first comes from extensive data on the induction of mutations in male mice that indicate that the mutation per total dose is considerably lower than it was in the earlier Oak Ridge experiments with acute X-irradiation. This appears to contradict the results of genetic experiments in other organisms where it has been shown that mutation rate is determined only by total dose and is independent of whether that dose is given over a short or long interval. However, possible explanations of the results that are compatible with the orthodox view must be considered.

One possible interpretation is that chronic irradiation at the particular dose and intensity levels used destroys relatively more

sensitive cells in the testis than does acute irradiation and that the mutation rate observed is that characteristic of the more resistant surviving cells. This explanation is similar to that invoked to account for the drop in mutation rate at high doses in the acute X-irradiation experiments. Since this hypothesis is at least plausible, it would be incautious, at the present time, to reach the sweeping conclusion that chronic gamma irradiation of males will, at all doses and intensities, be less effective in the induction of mutation than acute X-irradiation.

Whatever the final explanation turns out to be, it is apparent that the measurement of radiation-induced mutation rates in the immature male sex cells of a mammal is affected by factors that were not anticipated from the results obtained with the mature sex cells of the fruit fly. Since it is the immature cell stage that is important in man, these findings are clearly of vital significance in the estimation of genetic hazards of radiation.

The second major surprise in the new body of results is that the mutation rate found in female mice exposed to chronic gamma radiation is much lower than that used heretofore as the yardstick for genetic hazards, namely, the mutation rate obtained from acute X-irradiation of male mice. The difference could be due to intensity or quality of radiation or to sex.

Radiation Protection and Recovery

At the Oak Ridge National Laboratory research has been carried out on radiation protection and recovery and consists both of basic studies at the cellular level and methods of protecting irradiated mammals. These studies may very well be considered to lead to practical medical

applications, which would be valuable in accidental irradiation from a reactor or other sources.

Cellular Level

An important new finding in protection and recovery at cellular level is the enhancement of rejoining of chromosome breaks by supplying energy within a short time after exposure to radiation. This indicates the feasibility of reducing radiation injury by appropriate treatments shortly after irradiation. The protective effect of certain chemical agents varies widely among different species and strains of micro-organisms. In some cells, protective compounds actually increase the yield of radiation-induced mutations; the mechanism and significance of this effect remain to be determined.

Chemical Protection in Irradiated Mice

Among the many classes of chemicals that have been tested as protective against radiation damage, the most effect in mice seems to be the aminoalkylisothioureas and their corresponding mercaptoalkylguanidines. The radiation $LD_{50}/30$ days is proportional to the log of the oral dose of compound up to a maximum of 1500 r for (101 X C3H)F_1 mice. Unprotected mice show an $LD_{50}/30$-day value of approximately 700 r. The systems protected are apparently saturated at 1500 r, and the administration of additional compound does not further increase the $LD_{50}/30$ days. A maximum oral dose of AET remains effective for 6 hours against the acute lethal effects of 900 r.

Chemical modification of the AET structure established a relation between protective activity and molecular structure; the major requirement for maximum activity is that the nitrogen and sulfur atoms not be

separated by more than three methylene groups. Thirty compounds conforming to these general requirements varying widely in their chemical toxicity have given good 30-day survival when administered before X irradiation. Although no single mechanism can completely explain the functions of these compounds in protecting complex mammalian systems, it seems likely that their main function is the neutralization of the free radicals formed by ionizing radiation.

Bone Marrow Treatment in Irradiated Mice

Mice given whole-body irradiation near the level that causes 100% death in 30 days can usually be kept alive if they receive an intravenous injection of normal, living isologous bone marrow cells after the exposure. Isologous bone marrow transplantation means that the animal donating the marrow is genetically like an identical twin to the irradiated animal receiving the marrow. Homologous bone marrow transplantation will prevent acute radiation death but the radiation injury to the immune system that allows it to grow in the first place (since it is genetically different from the host) is gradually healed and a new disease complication develops called the foreign bone marrow reaction. Homologous fetal bone marrow was tried and proved to be greatly superior to homologous adult bone marrow. The radiated mice developed very mild foreign bone marrow reactions from which they usually recovered. It appears that fetal tissues will be an important material for study of homotransplantation problems.

Modification of Immune Status of Irradiated Mice

Studies of the antibody response of lethally irradiated mice pretreated with MEG or posttreated with isologous bone marrow performed

1,15, and 30 days after irradiation revealed that the recovery rate of the immune mechanism of mice injected with MEG and then exposed to 950 r of X rays was almost comparable to that of the mice given 475 r without other treatment. Lethally irradiated mice receiving foreign bone marrow usually undergo a delayed reaction 30-90 days after irradiation. Bone marrow chimeras that survive this reaction for more than 90 days appear normal outwardly but their response to antigenic stimulus is subnormal.

The radiosensitive mechanism of antibody-producing cells of normal mice can differentiate and respond to closely and distantly related antigens with equal effect, but radiation damage to the mechanism for recognizing and responding to the more closely related antigen is greater than to that for a distantly related antigen. From these studies a wealth of information about transplantation immunity and the passive transfer of antibody-forming systems has been derived. Determination of the chemical nature of the antigens responsible for heterograft (bone marrow) rejection should provide the immunologist with new ways of studying tissue transplantation.

Modification of Delayed Radiation Effects

Mice of the LAF_1 and $(101 \times C3H)F_1$ strains were exposed to whole-body 250-kvp X radiation at 10-13 weeks of age with and without concomitant administration of AET or isologous bone marrow or a combination of the two. The preliminary results of combined treatment with AET and bone marrow shows that the $LD_{50}/30$ days was nearly doubled by treatment with either agent along, and the use of both agents in combination further increased the $LD_{50}/30$ days to 2.5-3.0 times its value in untreated animals. Comparable protection was also afforded against the life-shortening action

of radiation, the mean longevity of 30-day survivors being more closely associated with the per cent (fraction) of their 30-day survival than with the dose of radiation they received. Induction of leukemia was also greatly inhibited by AET and by bone marrow. The results of these studies demonstrate that AET and bone marrow protect against delayed effects of radiation as well as against 30-day mortality.

Nucleic Acids, Genes, Viruses, and Bacteriophage

Cell nucleic acids appear to be directly related to both genetic stability and growth of organisms. At ORNL a number of years ago a new class of mononucleotide constituents of nucleic acids was discovered. The continued development of new analytical techniques allows a progressively greater insight into the structure of nucleic acids, their chemical properties and their susceptibility to specific enzymes.

It has thus become increasingly possible to attack the problem of the function of the nucleic acids in relatively simple biological systems, such as the bacteriophage virus in its host bacterium or in ascites tumor cells. The approach is designed to study biochemical relationships of the nucleic acids and protein during the synthesis and reproductive cycle of the virus, the composition of the latter being almost totally nucleic acid and protein.

It has been demonstrated that during the infection process, with concomitant virus synthesis, a small but highly active pool of ribonucleic acid (RNA) serves as a phosphorous precursor, presumably to viral deoxyribonucleic acid (DNA). If verified, this would be the first demonstration of such a biological precursor relationship. The major portion of the ribonucleic acid is either inert or turns over much more slowly.

Agents (for example, chloramphenicol) that prevent the synthesis of virus deoxyribonucleic acid, but not of cellular ribonucleic acid, have been used to separate the two events; upon removal of the inhibitor, synthesis of the former and turnover of the latter are initiated, which permits study of the interrelationships between these compounds.

The protein synthesis inhibitor, chloramphenicol, has apparently two effects on RNA metabolism on phage-infected bacteria. If chloramphenicol is added shortly after infection, a new kind of RNA similar to that found in control cultures is formed but is not degraded. If chloramphenicol is added before infection, the new kind of RNA is not formed. In both cases, when chloramphenicol is removed, RNA turnover and DNA synthesis occur. RNA turnover is not directly related to DNA synthesis since the addition of chloramphenicol at later times after infection allows DNA synthesis but still prevents RNA turnover.

A theory that fits the data is that RNA turnover is intimately connected to protein synthesis. Thus, as suggested by other investigators, a new protein must be formed shortly after infection before new phage DNA can be synthesized. We suggest that once this protein is formed, chloramphenicol would have no effect on DNA synthesis but would inhibit the RNA turnover associated with the synthesis of nonmetabolic phage protein.

Amino Acid Activation and Protein Synthesis

From a consideration of available information, a working hypothesis of the mechanism of protein synthesis is that is is a four-stage process: primary activation of amino acids; secondary activation (anhydride exchange); transport; and finally polymerization of the active constituents

to protein. Experiments at the Oak Ridge National Laboratory indicate that only about ten of the thirty-odd amino acids seem to participate in the primary activation step. Hence, the secondary activation, or anhydride exchange reaction, has been introduced as a possible mechanism for the activation of those amino acids which do not participate in the primary activation. Such anhydride exchanges are not uncommon in biological systems, for example, the activation of acetoacetic acid by interaction with succinyl-coenzyme A. The biological activation of amino acids has been measured by the amino acid-dependent exchange of pyrophosphate (labeled with radioactive phosphorous) with adenosine triphosphate and has been studied in rat liver, rabbit liver, mouse and rat bone marrow, and chicken erythrocytes. All tissues so far studied carry out this reaction. The catalysis of the pyrophosphate-ATP exchange by 18 individual amino acids has been studied in rabbit liver. Only nine amino acids, phenylalanine, histidine, tryptophan, valine, isoleucine, cysteine, methionine, tyrosine, and leucine, were active. This is the same group that has been found to be active with several microbial extracts, and this finding adds further evidence that these amino acids are the ones that undergo primary activation. The remaining amino acids may be activated in another manner, for instance as suggested earlier, by anhydride exchange. The rather general distribution of the amino acid-dependent exchange of pyrophosphate with ATP suggests that it may indeed by involved in protein synthesis. A detailed investigation is being made of several of the individual amino acid-activating enzymes.

Effect of Extremely High Gamma Irradiation on Green Plant Photosynthesis

The photosynthesis process in green plants is known to be relatively

insensitive to ionizing and ultra-violet irradiation. In addition, the formation of the photosynthetic process (greening) in etiolated plants upon exposure to light is also γ radiation insensitive. In two ORNL experiments, dosages of 150,000 r of γ radiation were necessary to produce a temporary inhibition of photosynthesis. After 24 hours green plants returned to a normal rate of photosynthesis and etiolated plants had greened normally. In this report are given some analyses on these irradiated plants for the subsequent period which they survived.

Etiolated plants, after exposure to an acute dose of 150,000 r of γ radiation, were kept in pots of soil in a constant environment plant growth chamber. For the first two to three weeks the plants appeared normal, but there was no new growth except for some general enlargement of the plant, probably from cell elongation. By the fourth week, the plants were chlorotic and fungal growth appeared on the dying leaves. At intervals, the respiration was measured, photosynthesis was examined by fixation of $C^{14}O_2$, and the products of the photosynthesis experiments were examined chromatographically. The respiration rate increased at the end of the second week, part of which may have been due to bacterial contamination on the dying leaves. The trend even after the second day was away from the normal pattern of sucrose synthesis to incorporation of C^{14} into glycine and serine. This same trend had previously been noted to a lesser degree in green plants immediately after radiation. During the last two weeks several substances, such as glutamic acid, were not labeled during one hour of exposure of $C^{14}O_2$ in the light. Though the wheat plants had received a lethal acute dose of radiation, there was neither inhibition of respiration nor photosynthesis, but growth was prevented. During the first two weeks after radiation, there

appeared on the epidermis of the leaves numerous droplets of a gummy concentrate, which dried as beads and was collected and analyzed. Had the plants been watered by rain or spray, this phenomenon would not have been detected, since the exudate would have been washed off. The excreta contained much glucose, glutamine and serine, as well as substantial amounts of fructose and many other amino acids. The exudation ceased between the second and third week when the photosynthetic rate dropped. During the first two weeks after irradiation, the leaves probably accumulated large amounts of the products of photosynthesis, since the process was not inhibited while growth, which would have utilized the products, ceased. This accumulation of photosynthesis products may also have been partially responsible for the drop in rate of photosynthesis and change in the products of photosynthesis.

Between the second and fourth weeks, the plants showed a slow decline. No definite period of death occurred, rather fungal growth slowly took over the leaves. This pattern of death in plants is in marked contrast to that found with animals exposed to massive dosages of acute radiation.

Role of Peroxide in Bacterial Metabolism

The mechanism of the oxygen effect on the radiation sensitivity of biological material is still unknown. It is therefore of interest to understand the detailed mechanism of peroxide metabolism in living organisms. To this end, microorganisms are among the most suitable material for study.

Electron transport enzymes from the strict anaerobe *Clostridium perfringens* are being used to study the role of peroxide in the metabolism

of anaerobic bacteria, with special reference to the mechanism of oxygen toxicity.

The reduced diphosphopyridine nucleotide (DPNH) oxidase of this organism has been further characterized with respect to: (1) variety of H acceptors utilized, (2) inhibitors, (3) ultra-violet inactivation, (4) kinetics of inactivation by substrate (CPNH), and (5) role of peroxide in the DPNH inhibition reaction. The results demonstrate that even extremely low peroxide concentrations may be toxic to certain anaerobes and that these peroxide concentrations can be the result of DPNH oxidation reactions. The behavior of DPNH oxidase indicates that some enzymes may show enhanced sensitivity to peroxide when they are combined with substrate, reduced, or 'turning over'.

The Relation Between Structure and Activity in Radiation Protection Sulfhydryl Compounds

The oral administration of mercaptoethylguanidine (MEG) and mercaptopropylguanidine (MPG) has been shown in this laboratory to reduce radiation damage in mice. Since both compounds must be prepared from their respective thiuronium salts - AET or APT - by neutralization, and since they are susceptible to oxidation to the less-active disulfides, it would be obviously advantageous to administer them as the more stable thiuronium salts either as a solid or in solution. The pH of the animal is such that it could be expected that both AET and APT would be rearranged to MEG and MPG upon absorption into the body fluids.

Indeed, both compounds were found to be just as effective when given in this manner, and in addition, it was found that they were effective over a longer period of time, since they were probably absorbed more slowly. Since there is a relatively wide margin of activity for both

AET and APT, a series of radiation LD_{50} experiments at different drug doses were performed. The elevation of the radiation LD_{50} is proportional to the logarithm of the dose of drug and APT is more effective at lower dose levels than AET. A compound - 3-aminopropyl-N' -methylisothiuronium'B·HBr (APMT)- that had been found to be very effective at 900 r in screening tests, and to have a very desirable therapeutic ratio at this level, was subjected to the same test. It was found that APMT very rapidly saturated whatever sites it was protecting and that even increasing the drug dose 16 times did not bring about any further increase in the radiation LD_{50}. These facts illustrate how dependent the protective activity is on the structure of the compound. The simplest one, AET, apparently is widely distributed, and small changes in structure are sufficient to alter the selectivity and distribution of the compounds markedly.

The period for which a single, maximum, oral dose of AET or APT remained effective was found by the determination of the radiation $LD_{50}/30$ days at several time intervals after administration of the drug. The protective activity remained at its maximum for an hour and fell off with time, until at 5 hours the $LD_{50}/30$ days was 900 r. An estimate of the rate of excretion of these compounds indicates that at 5 hours about 20% of the AET is still present, while the APT was more rapidly excreted with only about 10% remaining in the animal.

Biophysics Research

Biophysics research at the Oak Ridge National Laboratory is concerned with the detection and evaluation of radiation hazards and with protection of personnel from these hazards. The scope of work includes

all types of radiation hazards to employees in the nuclear energy establishments and to the general public, including hazards of normal plant operations, unusual accidents, or military uses of atomic weapons. About equal emphasis is placed on the three aspects of biophysics research: detection of radiation, evaluation of the hazard, and protection of personnel from the damaging effects of the radiation hazard.

The ORNL biophysics research program is strongly influenced by the needs of applied health physics groups concerned explicitly with the protection of ORNL employees and inhabitants of the general area around ORNL. Consequently, emphasis is placed on detection and control of radiation hazards of the type to which personnel in nuclear energy establishments are most likely to be exposed.

Radiation Protection for Employees

In health physics, research has covered a wide field, ranging from the development of new radiation-detection and monitoring instruments to studies of the distribution of radioactive waste materials in the environment of White Oak Creek, into which low-radioactivity waste solutions are drained. The protection of Laboratory employees from radiation hazards is a routine service carried out so well by the health physics groups that since the Laboratory started operation in 1943 there has been no known case of injury to a worker resulting from radiation exposure.

Radiation Detection Instruments

In carrying out and improving its program of radiation protection for employees, the Laboratory developed new instruments for the detection and measurement of radiation. Many of these instruments have been

adopted for use at other AEC installations and are being manufactured from ORNL designs by private industry.

A scintillation counter developed for radiation monitoring proved so sensitive that with modifications it has been adapted to prospecting for uranium from a low-flying airplane. Other very successful instruments have been a portable neutron survey instrument, underground radiation detectors for use in core drillings, and a "pocket screamer" that sounds an alarm when the wearer has received the maximum permissible radiation exposure.

Civilian Defense Contributions

A method of adapting army field water-purification equipment to the removal of radioactive contaminants has been developed as a part of the health physics program. The successful adaptation of standard city water-purification procedures to the removal of radioactive material from a water supply has also been demonstrated.

New items of protective equipment for use at the Laboratory and in civilian defense against atomic bomb attacks have been developed at ORNL and made available for widespread distribution to civilian defense centers. The constant improvements of radiation-detection instruments to give good service under rough use has provided several simple and reliable instruments that are now being manufactured by private industry under an agreement set up by the Atomic Energy Commission.

Waste Disposal Research

In seeking to improve the control of radioactive materials released into an area surrounding radiochemical processing plants, it was decided to concentrate liquid wastes by evaporation. The first radiochemical

waste evaporator was designed and operated at ORNL. This philosophy of concentrating radioactive wastes for storage by evaporation has since been adopted at other AEC installations.

Basic Research

Fundamental health physics research has been successful in providing new information relating to permissible radiation exposure and to the effects of radiation on body tissues. A "permissible internal dose" for radioisotopes ingested by breathing or eating has been proposed by ORNL health physicists and established as a standard national code for radiation protection. In addition, basic research has resulted in the development of satisfactory instruments for the measurements of dose rate of fast and thermal neutrons.

Interaction of Radiation with Matter

Electron Attachment Studies

Most measurements by the health physicist of absorbed radiation dose require gas-filled counters complying with the Bragg-Gray principle. A knowledge of the number of electrons which become captured while crossing the gas under the influence of an electric field makes possible an evaluation of the effects of electronegative gases in the operation of proportional counters, Geiger-Mueller Counters, and ionization chambers. An apparatus was developed at the Laboratory for the purpose of measuring attachment coefficients of electrons to various gases. Typical results from this apparatus give the required information in suitable form for practical application. Further interpretation of the results yields basic information regarding the probability for the exchange of vibrational energy by collisions between molecules.

Electron Transport and Slowing Down

Determination of the spectra and spatial distribution of electron flux in solids is important for establishing the potential damage to man from beta-emitting isotopes. Measurement of the energy spectrum of electrons in a solid containing a uniformly distributed P^{32} source has been made using a solenoidal beta-ray spectrometer. The depth dose distribution due to electrons incident on a solid has been measured and is of importance in determining the possibility of beta burns from external exposure and in interpreting beta dosimeter readings.

The Physics of Tissue Damage

The concept of Information Theory has been applied in this Laboratory to the mathematical study of radiation damage to living organisms. The role of noise in the genetic specificity message has been investigated and has resulted in a general treatment of survival from this point of view. An equation relating radiation injury, dose and recovery, originally suggested by H. A. Blair, has been derived by application of these ideas.

Characteristic Energy Losses by Charged Particles in Matter

Experimental and theoretical researches on the energy loss of charged particles in matter have been carried out. Discrete losses by fast electrons traversing metallic foils have been observed and have been attributed to the excitation of plasma oscillations of electrons in the conduction band of the metal. The observed values of the energy loss per unit path length in the foil agree well with theories of the plasma effect. A theory of the effect of the boundaries of the foil upon the plasma

losses has been developed.

Fast-Neutron Dose Calculations

The difficult problem of estimating the energy released by neutrons in their passage through living tissue has been solved rather accurately by the "Monte Carlo" method. This mathematical technique involves calculating, step by step, possible paths of neutrons through the tissue in such a way that the interactions, occurring along a large number of such paths (2000), will be a statistically reliable sample of the reactions occurring under actual irradiation. Such calculations are important for understanding and predicting biological effects of irradiation, which in turn are important for establishing standards of protection.

Instrumentation and Dosimetry Development

Radsan Fast Neutron Dosimeter

Around reactors and accelerators there is often an acute need for an instrument which will measure the fast neutron dose in the presence of gamma radiation. The "Radsan" instrument, developed at ORNL gives a direct measure of fast neutron dose and is essentially insensitive to gamma rays at rates less than 5 r/hr. It is suitable for radiation protection measurements and for research in radiobiology, neutron physics, and shielding.

Neutron Threshold Detector System

Another very important development in the measurement of neutron dose in the presence of high gamma fields is the threshold detector technique. It employs a series of foils which respond to neutrons above certain energy levels. The amount of induced activity in the detectors

is determined with scintillation counters. Differences in the fluxes indicated by the foils give a measure of the spectral distribution of the neutron flux. This method works well for high intensity neutron bursts and in high gamma fields and may be accomplished under conditions where other methods, e.g., the ionization chamber and proportional counter, would lead to questionable data.

Beta-Ray Dosimeters for Personnel Monitoring

An ORNL-developed personnel dosimeter system for beta radiation has many applications. These pocket chambers, in conjunction with the standard Victoreen chambers, permit accurate measurement of beta-ray dosage for personnel monitoring. The chambers are nearly energy independent and are calibrated in water solutions of several beta-emitting isotopes.

Another dosimeter of the general size and shape of a wrist watch has been developed to measure the radiation exposure to the hands. The device is rugged, easily and cheaply fabricated, and records the exposure from soft beta radiation which normally escapes detection, and from X-rays and gamma rays as well. General use of this dosimeter will enable a more careful assessment of beta-ray exposure to be made, which in turn will do much to reduce the hazard of beta burns on the hands of workers in atomic energy and related programs.

Applications

Instruments developed in the Radiation Dosimetry Section have been extensively applied in measurements in Applied Health Physics, and in radiobiological experiments.

Radiation Dosimetry for Human Exposures

A most important application of radiation dosimetry is accurate determination of the neutron bombings of Hiroshima and Nagasaki. A correlation of the doses received with their medical effects will lead to the important goal of establishing maximum permissible levels of exposure of man to radiation. A far-reaching program at ORNL involves continued liaison with the Atomic Bomb Commission in Japan and participation in weapons tests to determine structure shielding.

X-Ray Dosimetry

Radiation doses to the skin and gonads of patients during routine diagnostic X-ray procedures have been determined for various techniques and equipments. The doses vary widely with technique and are tabulated as functions of filtration, cone or diaphragm, size, distance, kilovoltage and milliamp seconds. The data obtained have been used to determine the gonadal dose from occupational medical exposure of employees of the Oak Ridge National Laboratory. A method has been initiated for determining this dose from all medical sources.

Human Tissue Studies for MPC Determination

One of the most important problems of Health Physics is the establishing of maximum permissible concentration (MPC) values of radioactive materials in the body and in air, water, and food. A knowledge of the distribution of stable trace elements in the human body is of great value in this connection, because the principal organ of deposition for a radionuclide is the same as for the stable isotope. During the past several years spectrographic analysis has been performed for 37 elements in 35 human tissues from about 300 autopsies from nine cities in the

USA and from fifteen foreign countries. In addition to their application in determining MPC levels, these data are proving to be of value to studies of cancer and heart disease.

Internal Dosimetry of Uranium in Man

The potential large-scale industrial use of uranium as a source of power has led to concern about the hazard to man from chronic or accidental exposure to this element. Valuable data on the excretion of uranium and its distribution in the body tissues has been obtained from the injection in terminal brain tumor patients at the Massachusetts General Hospital, where the potentialities of uranium-neutron capture therapy in the treatment of inoperable brain tumors is being studied.

Radioparticulate Inhalation Studies

In order to evaluate the hazard of airborne particles of uranium, preliminary measurements of the retention of inhaled uranium in dogs have been made. Data obtained in this way will be of great interest for the determination of MPC values for airborne uranium in man's environment.

Radioactive Wastes and Environmental Studies

Water Decontamination Studies

An extensive program of research and development on decontamination of radioactive water supplies for civilian and military use and work on related problems was begun in 1949. The U. S. Public Health Service, U. S. Army Corps of Engineers, Tennessee Valley Authority, and graduate students from several universities cooperated in these studies. The general purpose was: (1) to define the sources and significance of

radioactive contaminants in water supplies; (2) evaluate conventional, nonconventional, and modified water treatment processes for the removal of radioactive materials from water; and (3) to develop effective applications of these processes for the decontamination of water supplies and low-level radioactive wastes. Most of the experimental work was completed in 1954.

In experimental laboratory and pilot plant studies of conventional methods of water treatment the efficiencies of various processes singly or in combination ranged from zero to 99 per cent in the removal of individual radionuclides or mixtures of radioactive materials from water. The ion exchange process, where applicable, is one of the most efficient methods for removing radioactive contaminants from water supplies. By co-precipitation in the lime-soda softening process strontium was removed with efficiencies of 95 to 99 per cent or higher.

In studies of nonconventional water treatment processes, phosphate coagulation removed up to 98 per cent or more of radiostrontium and more than 90 per cent of most other radionuclides studied. Efficiency of decontamination in water treatment plants can be increased by modified pre-treatment procedures, as by the addition of clay for removal of cesium, or of metallic dusts for absorption of certain other radionuclides.

A thorough evaluation of military mobile water treatment equipment was completed in 1954. It was found that the several military field units, including coagulation and filtration, ion-exchange, and distillation, could be operated routinely with satisfactory efficiencies and without undue radiation exposure of operating personnel.

Aerosol Studies

Basic studies of aerosols have been carried out in cooperation with the U. S. Army Chemical Corps. This work has contributed greatly to an understanding of aerosol kinetics and has been helpful in meeting problems of personnel protection and off-gas decontamination. A diffusion battery was developed which is applicable for determining the size of homogeneous particles from molecular dimensions to radii of about 0.3 microns; however, battery construction difficulties essentially limit the method to particles under 0.1 micron radius. The Naval Research Laboratory photoelectric particle size meter (Owl) was automatized to provide continuous recording of the size of homogeneous particles in the range of 0.1 to 0.2 micron radius. This instrument was useful, particularly in studies of particle generator characteristics. A modified LaMer type aerosol generator improved upon the stability and homogeneity of particles produced in the submicron to several micron range. An improved model of the Wilson cloud chamber particle counter, based on Green's (Britain) apparatus, was developed which could enumerate particles as small as 10^{-3} micron radius. An original development was a lead shot-column particle sizer to speed up determinations of the size of aerosol particles. Another accomplishment was the establishment of an optimum particle size for maximum filter penetration which has aided in filter design problems. It was found that the maximum penetration in fiberglass and sand filters resulted from particles of about 0.2 to 0.5 micron radius.

Ecology Program

Nuclear power programs are expected to produce, in addition to high-level wastes, large volumes of low- and intermediate-level wastes.

It may be possible to release some of these to the natural environment. The purpose of the Ecology Program is to provide data which will aid in the determination of safe release concentrations. Data are being obtained on the distribution and movement of fission products in soil and food chains, on their ultimate concentration in plants and animals, and on the long-term ecological effects of these concentrations. Many investigations are being carried out in the drained White Oak Lake bed and in the environs of the radioactive waste pits with their significant concentrations of Sr^{90} and Cs^{137}.

Ecological Research

Ecological studies at ORNL are mainly in the contaminated White Oak Lake Bed that was formerly the radioactive waste impoundment area and now is one of the truly unique tracts of land in the world. Twelve years of low-level waste impoundment have resulted in terrain that contains more strontium-90 and cesium-137 than any comparable land now know. Average concentrations of strontium-90 are about 400 millicuries per acre and those of cesium-127 range up to 20 curies per acre. In addition, there are significant concentrations of cobalt-60, ruthenium-106, cerium-144, and the trivalent rare earths. The presence of these materials in the soil makes the tract suitable for studies of the distribution and uptake of these fission products by both natural and cultaivated vegetation. Radiation fields in excess of 100 times natural background make the area suitable for studies of the long-term effects of low-level radiation on plant and animal populations.

Major field ecology research effort has been applied to the further characterization of the complex physico-chemical soil system;

to the characterization of the natural flora and fauna developing on the lake bed; and the determination of concentrations of strontium-90 and cesium-137 in the soil, in the natural vegetation, and in corn grown in the bed.

The first results of the corn analyses indicate that the concentrations of cesium-137 may differ between the various plant organs. The ratios of cesium-137 to potassium indicate a considerable discrimination against cesium-137. Preliminary analyses of strontium-90 concentrations in these plants indicate less discrimination against this radionuclide than for cesium-137. The evidence also indicates that strontium is differentially deposited in the various plant organs, with the highest concentrations being found in the leaves.

Preliminary trapping studies have shown that several species of mammals have moved onto the lake bed. Since the radiation field over the ground surface averages above 0.5 roentgen per day plus additional exposure due to ingestion of contaminated food materials, it is believed that there is opportunity here to obtain information on long-term effects of low, continuous doses of radiation.

TRAINING AND EDUCATION

As one of the first research establishments to undertake a major nuclear energy program involving large quantities of radioactive materials, ORNL has, from its very beginning, devoted much attention to training and education in the fields of nuclear science and technology. Major activities have included the original training of personnel to operate the Hanford production paltn, the early postwar training of personnel from private industry in reactor technology, and the subsequent operation of formal training programs in the Oak Ridge School of Reactor Technology and in Radiological Physics.

A program of research participation for university faculty members is conducted; and, in cooperation with the Oak Ridge Institute of Nuclear Studies, a program of graduate training is offered. A Traveling Lecture Program jointly sponsored by ORINS and ORNL provides speakers from ORNL's scientific staff to visit southern universities and colleges for lectures to students and faculty on recent scientific advances. From time to time, specialized short courses are offered by ORNL to meet particular needs.

Also, ORNL accepts employees from private industry and other government agencies to receive on-the-job training by working with ORNL research and development groups. The highly successful operation of the Oak Ridge School of Reactor Technology was discontinued at the end of the 1957-58 school year, since it appeared by then that adequate educational opportunities were available through the universities and colleges. Training in reactor technology is continuing

on a limited scale to meet special needs for advanced training or for concentrated short courses in specialized areas such as reactor operations and reactor hazards evaluation.

Also as a part of its training and educational activities, ORNL participates in the preparation of scientific reference books and handbooks on various phases of nuclear science and technology, in consultation with foreign countries on nuclear energy programs, and in the evaluation of, or assistance with, reactor construction projects. Special conferences on nuclear energy are often held on a regional, national, or international basis. Many regional conferences are sponsored by ORNL, while for the national and international conferences, the Laboratory often plays a major role in providing reports, displays or exhibits.

Early Postwar Program

One of the early postwar programs at the Laboratory was a training school, which operated for one year in 1964 and 1947 as a nuclear-technology school for scientists interested in entering the new field of atomic energy. Approximately 40 scientists from universities and industry attended the training school, including several who are now among the best-known figures in the American atomic energy effort.

Oak Ridge School of Reactor Technology

The success of the training school gave impetus to the plans for establishing a full-fledged training program. In 1950, the Oak Ridge School of Reactor Technology started operation on a permanent basis. The first "interim" class while the school was getting started included

18 students from governmental agencies and industry. Subsequent class enrollments for the one-year course have been 46, 68, 80, 74, 84, 88, and 111 making a total of 569 engineers and scientists who successfully completed the full program. About 75 additional people attended portions of the program during this period. The original program was discontinued with the graduation of the 1956-1957 class. A two-part program composed of six months of fundamentals in a cooperating university and six months of advanced reactor technology at ORSORT was initiated in March 1957. The participating universities are Carnegie Institute of Technology, Case Institute of Technology, Northwestern University, Union College, University of California at Los Angeles, and the University of Florida. The first two cooperative classes numbered 69 and 50. Applications for the 1958-1959 class were much fewer in number, and it was decided that university and colleges were meeting the major educational needs in this field.

A small but important by-product of ORSORT is the preparation of University faculty members for teaching in the nuclear field. Each year, the ORSORT faculty is augmented by several loanees from various universities; after 6-12 months, these men return to the universities prepared to take active parts in the growing nuclear programs on the campuses.

In the summer of 1957, ORNL offered a special 8 weeks institute in reactor physics under the joint sponsorship of the AEC and the American Society for Engineering Education. This advanced course was attended by 14 university teachers and 7 representatives of industry.

Research Participation Program

Another program which contributes to the training of university faculty members as well as to the Laboratory's research is the research participation program, under which faculty members work on research projects with established ORNL groups during the summer when they are not teaching. The faculty members learn as they work, and quite frequently are able to make noteworthy contributions to the research from their own experience. The exchange of ideas is beneficial to all concerned. Under this program, research participants from the universities work at ORNL each summer.

Graduate Study Program

The cooperative program carried out with the Oak Ridge Institute of Nuclear Studies (ORINS) allows university graduate students to do their thesis research at the Laboratory, contributing to the research program as well as to their own education.

Traveling Lecture Program

The traveling lecture program is also carried out in cooperation with ORINS. Speakers on uncalssified scientific subjects are supplied from the Laboratory staff to speak to technical groups at universities according to arrangements made cooperatively with ORINS.

Education and Training Activities

In cooperation with Vanderbilt University during the period 1953-1957, ORNL has trained 108 people under the U. S. Atomic Energy Commission's Radiological Physics Fellowship Program. Approximately 80% of these fellows are now engaged in health physics activities.

Two International Courses in Health Physics have been conducted by ORNL personnel, one at Stockholm, Sweden, in 1955 and one at Mol, Belgium, in 1957. These were sponsored by the World Health Organization and the U. S. Atomic Energy Commission. There were 51 participants from 29 countries. A course in Health Physics was conducted by members of the Health Physics Division of the Oak Ridge National Laboratory at the Centre D'Etudes Nucleaire, Mol, Belgium, and was attended by 27 students from 19 countries. The course, sponsored by the World Health Organization, the Atomic Energy Commission, and the Oak Ridge National Laboratory, extended over a five-week period and covered much of the same material presented to the AEC Fellows in Radiological Physics plus elements of reactor theory as applied to the study of reactor safety. The Belgium course did much to widen the field of Health Physics in Europe where the people associated with the nuclear energy developments are anxious to obtain information on radiation protection and the training of health physicists.

A special short course (eight weeks) in Health Physics Fundamentals was offered at ORNL in 1958 to fill an immediate need for personnel who have some understanding of the nature, scope and magnitude of Health Physics problems. The course included formal classroom work concurrent with Applied Health Physics Training.

Numerous individuals and groups from industry, universities, the U. S. Armed Forces, and from ORNL, have been trained during this period. The Health Physics personnel from the Nautilus received much of their training here.

ISOTOPE PRODUCTION

Radioisotopes

The Atomic Energy Commission's isotope-distribution program was inaugurated in 1946 with the first radioisotope shipment to a non-project user. The Laboratory had been the main source of supply for radioisotopes to meet research needs during the war, and was established by the AEC as the center for radioisotope production and distribution. Radioisotopes have proved very valuable in a wide range of uses, from industrial applications in such devices as thickness gauges, to research programs using radioactive tracers, and therapeutic treatments where the radiation from radioactive atoms has been used in combating cancer. Radioisotopes have been called "the most valuable research tool since the invention of the microscope." Since 1946, the number of radioisotope shipments has increased greatly each year; the total number of shipments through April 1953 was 43,572.

Stable Isotopes

The calutron units used for separation of uranium isotopes on a production scale at the electromagnetic plant have been adapted to the separation of the isotopes of other elements, and a stock of those naturally occurring stable isotopes has been built up, and is available for research uses under the isotope-distribution plan. Stable isotopes can be traced by their mass differences, and are extremely useful in tracer studies where it is undesirable to use radioactive atoms. Also, stable isotopes provide excellent sources for preparing certain pure radioisotopes. When ordinary iron is irradiated with neutrons, two

radioisotopes, iron-55 and iron-59 are formed from the stable isotopes present. By using a separated stable isotope for the irradiation, iether pure iron-55 or pure iron-59 can be prepared without contamination by the other. Approximately 50 elements have been processed for isotope separation, and the total number of separated isotopes now available from these elements is 185. Through April 1953, 1,759 shipments of stable isotopes were made.

Radioisotopes Separations Development

The expanding use of radioisotopes has resulted in increased efforts to develop new processes and new products. The production and sale of separated and analyzed short-lived radioisotopes made by irradiating target materials in high-flux zones is a major program. Since 1953, 26 short-lived new radioisotopes have been made available as analyzed products. These products range in half-life from 12.44 hours to 19.5 days.

Processes for the recovery and purification of radioactive gases were developed. Typical products are 5.3-day xenon-133, 35-day argon-37 and 10.27-year krypton-85. Small quantities of xenon-131 are produced for research activities at ORNL. Iodine-129, with a half-life of 17,000,000 years, is recovered from the scrubbers for the off-gas from dissolution of irradiated uranium. The iodine-129 can be used as essentially a stable isotope or as a target material for the production of 12.5-hour iodine-130 by neutron irradiation. An improved process for the recovery of phosphorus-32 uses highly purified sulfur as the target material; the excess sulfur is vacuum distilled from the phosphorus-32 product in quartz equipment. The product has a high

purity and low total solids. Improved techniques have been developed for the preparation of tritium-zirconium targets for use with particle accelerators to provide monoenergetic neutrons for research purposes.

Radioisotopes with much higher specific activities than can be obtained by irradiating naturally occurring elements are being produced by using target materials obtained from the stable isotope separations program. A particular example is the production of chromium-51 with a specific activity of about 15,000 millicuries per gram of chromium. The target material is chromium in which the chromium-50 isotope has been enriched from the naturally occurring 4.31% to more than 75%. This development has been of particular interest in medical applications.

Development has continued on improvement of processes for the recovery of fission products from waste solutions. Processes for the recovery and purification of Cs-137, Tc-99, Np-237, and Pm-147 were developed.

Isotope Production

During the period 1954 to 1957, radioisotope sales increased 400% in terms of annual total number of millicuries shipped while unit production costs per millicurie sold were reduced 76%. In the calendar year 1957, the Laboratory made over 104,000 shipments containing a total of 375,000 curies of radioactivity. These reduced unit production costs in spite of increasing "costs of living" have permitted the Commission to pass the benefits of increased production to the customers in unit price reductions. Prices for cobalt-60 were essentially cut in half in 1957.

The conversion of a major portion of the stable isotopes distribution program from a loan to a sales basis has permitted recovery of a substantial portion of production costs.

Radioisotope source production increased almost 600% from 1954 to 1957 in the total number of curies shipped while the activity per average individual source increased almost 300%. This expansion took place during a time when permissible radiation exposure tolerances for the production personnel were reduced by 75%. In keeping with this trend more heavily shielded processing facilities with advanced remote-operating techniques were built. A manipulator cell with 3 feel of barytes concrete as shielding and equipped with master-slave manipulators and a lead glass viewing window was constructed to handle greater quantities and higher specific activities of cobalt-60. Up to 10,000 curies at one time can be handled safely in this cell.

A plant for the separation of short-lived fission products was built to operate at a capacity of 400 curies per month. This plant extracts fission products with half-lives of 12 to 60 days. Typical products are 40-day ruthenium-103, 65-35-day zirconium-95-niobium-95, 12-day barium-140, 53-day strontium-89, and the rare earths 32-day cerium-141, 11-day neodymium-147, 14-day praseodymium-143, and 58-day yttrium-91. Completion of the new fission product pilot plant will allow the production of up to 100,000-curie quantities of long-lived separated fission products. Typical uses are teletherapy sources of cesium-137, cerium-144, or ruthenium-106, small batteries powered by promethium-147, and corrosion protection by technetium-99, an element not found in nature. This pilot plant will also permit the testing of separation processes at plant levels of radioactivity to accumulate

information that could be used in designing a megacurie-level separation plant.

Remote source-fabrication techniques and equipment have been developed. The radioactive powder is compressed into dense pellets by a remotely operated hydraulic press. A remotely operated welder which seals stainless steel source capsules by a fission weld under blanketing inert gas was developed and is used routinely to seal sources containing cesium-137 chloride.

Another outgrowth of the fission product plant program is the development of large shielded carriers to transport fission product waste solutions from other locations to ORNL for test runs in the pilot palnt. A prototype unit of 250 gallons capacity was built and tested by transferring waste from the Idaho Chemical Processing Plant to ORNL. Based on the data obtained from this transfer, six tanks of 500 gallon capacity are being fabricated.

A cobalt-60 irradiation facility has been constructed to use radiographic and teletherapentic sources which are stored at ORNL prior to sale. The heat released by the 200,000 to 300,000 curies in storage is removed by air flowing around the storage tubes.

Special Isotopes Services

During the early part of the period from 1953-57, while essentially all enriched stable isotopes were distributed on a loan basis, it was noted that considerable waste of valuable isotopic material was resulting from losses when researchers unfamiliar with the techniques of quantitative chemical recoveries prepared their targets. In numerous cases more than half the sample was lost during conversions of target preparations.

It appeared that considerable savings to the overall program could be effected if ORNL were to provide certain special services in connection with some of the target fabrication and other related needs. Not only would there be a reduction in the amount of isotope used (and/or lost) per target, but there would be available a service for many users who perfer not to have to prepare their own targets.

Since the inception of this program, the number of special services performed on a cost-recovery basis has increased until it now comprises about 15% of all stable isotope shipments. There is a wide variety of techniques and services available and the items prepared have run the gamut from a 1 mg/cm^2 electroplated unsupported copper foil to 4-kilogram targets of elemental lithium-6 and 7. In general, isotopes from inventory may be converted to more desirable chemical compounds; large and small coupons of many elements can be made by powder metallurgy techniques; thin foils are vacuum-evaporated, rolled, or electroplated; wires have been drawn and thin metallic strips fabricated. And upon completion of each service, there is available a competent isotope chemist to assure efficient recovery of any unplated, unconverted, or unreduced material.

86-Inch Cyclotron Isotope Production

Marked reductions in the cost of cyclotron-produced isotopes have been achieved; for example, the cost of arsenic-74 has been reduced ten-fold in the past two years. This positron-emitting isotope is now being used in the detection and localization of brain tumors.

Formerly, arsenic-74 cost $200 per millicurie; it is now available commercially as a biological preparation at only $20 per millicurie.

The price includes the full cost of cyclotron operation as well as the processing costs at the pharmaceutical laboratory (Abbott).

Cost reductions are made possible by the high performance of the ORNL 86-Inch Cyclotron. This machine accelerates large currents of protons to about 25 Mev. The arsenic isotope is produced by bombarding germanium with protons, a current of 200 microamperes is used on a water-cooled target. The yield is now approximately 15 millicuries per hour. With the increased success in the use of the isotope as a diagnostic tool, the production of arsenic-74 has been increased to a rate of over 2000 millicuries per year.

Calutron Production Improvements

Improvements in calutron ion sources have made possible the electromagnetic separation of isotopes that could not previously be vaporized nd ionized sufficiently well for calutron processing. For example, rare earths were processed as soon as suitable charges became available but only after development of the M-14 source. Separations of the platinum-palladium group were the result of development of the bombardment-heated source. The production rate achieved in the separation of isotopes of these four elements was approximately five times that predicted through use of the more conventional cathode-sputtering source types. In general, production rates of elements previously separated can be increased by proper choice of ion source. A two-fold increase in separation rates for chromium and calcium resulted from development of the modified Alpha-2, M-56, and modified grid ion sources.

Another technique for increasing isotope separations is the simultaneous collection of certain "side-band" isotopes such as those

of nickel and copper which results in an overall increase in productivity per calutron tank hour. In this case, 106 grams of copper isotope were separated as a "by-product" of a nickel collection, simply by adding copper to the charge material and providing proper collectors.

Improvements in methods of cooling receiver pockets has increased the retention of material to such an extent that productivity shows marked improvement even for a fixed ion output. Silver and magnesium isotopes show a two-to five-fold increase in collection rates as a result of this development.

The separation of high-purity samples provides a real service for research, but such special separations would be expected to be at the expense of production rate. High-purity Li^6 and Li^7, however, are noteworthy exceptions to this general rule. Improvements in isotopic purity without loss in production rate are at times achieved by unusual operating techniques and equipment design. The increase in purity of selenium, for example, was achieved through use of alcohol for refrigeration of the calutron components; the increase in cadmium purity resulted from the use of inter-pocket shielding and improved operating techniques.

Extension of the use of graphite collector pockets to replace copper in all possible instances has reduced chemical processing time. Quidk assay can be obtained and used as a basis for pocket combinations prior to complete recovery and sample processing. The quick assays have also made possible intra-run control of the separations.

Separation of Plutonium Isotopes

The electromagnetic separation of the plutonium isotopes was successfully undertaken. Gram quantities of enriched Pu^{239}, Pu^{240},

and Pu^{241} were separated with maximum purities of 99.8%, 92.2%, and 62.0%, respectively. The feed plutonium used was obtained from the NRX reactor at Chalk River, Canada. To obtain isotopes of higher purity, plutonium is beinghighly irradiated in the MTR to achieve a feed of optimum isotopic distribution. Also, a high resolution isotope separator has been developed. The products of the isotope separation program are distributed on a Commission-wide basis for cross-section measurements and other basic research uses.

High Resolution Isotope Separator

A new isot pe separator developed for heavy-element-isotope work, and also for research with cyclotron or reactor-produced radionuclides, gives large separation factors and high efficiency with relatively small amounts of feed material. This unit, which has the same mass dispersion and uses the same megnetic gap and shims as the Beta calutron operates satisfactorily at a feed rate of approximately one milli-mole of element per hour with an efficiency of 5-10%. In one test, U^{235} (0.7% normally abundant) was enriched to 88% purity. The enrichment factor was 1000, a factor of ten greater than obtained with conventional high enrichment calutrons.