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URANIUM MATERIAL ACCOUNTING AT THE GASEOUS DIFFUSION PLANT

U.S. DEPARTMENT OF ENERGY DECLASSIFICATION REVIEW	
1st Review - Date: 4-23-97	Declassification (Circle Number(s))
Authority: <input type="checkbox"/> ADC <input checked="" type="checkbox"/> ADD	1. Classification Retained
Name: mshum	2. Classification Changed To: SECRET
2nd Review - Date: 4/23/97	3. Contains No DOE Classified
Authority: ADD	4. Coordinate With:
Name: George Downing	5. Classification Cancelled *
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M. Benedict
A. M. Squires

1. Introduction

* with Deletions

This memorandum reports the findings of a two-day visit with the USAEC and G & CCC personnel in Oak Ridge who are concerned with uranium material accounting at the gaseous diffusion plant. The purpose of the visit was to form an opinion concerning the reliability of present material accounting practices, and the degree of improvement in them which can reasonably be expected.

The study has been restricted to the cascade U-235 material balance, the most important contributor to uncertainty in the unexplained loss of U-235 from the K-25 area. Time did not permit study of the cascade uranium balance, or U-235 material accounting procedures in various other operations of the K-25 area (laboratory, decontamination, etc.).

An opinion is given concerning the present situation as regards unexplained losses of U-235 and the limit of error in estimating these losses. Means of reducing these quantities are suggested.

It is recommended that the following objectives be set for the unexplained losses of U-235 from the gaseous diffusion plant, which may ultimately be attained:

- (1) In no one year should the unexplained U-235 loss exceed 30 kgm.
- (2) Over a period of years, the cumulative unexplained loss, together with its upper limit of error evaluated for the full period of time, should not exceed a value judged by the Commission to represent a potentially intolerable hazard.

Attainment of the first objective cannot reasonably be expected for at least another year, and may be further delayed unless there is some sacrifice of production in favor of material accounting reliability.

2. Objectives of Uranium Material Accounting

The primary objective of uranium material accounting is to provide assurance that significant quantities of valuable, dangerous, highly enriched U-235 have not been diverted from the plant.

Unexplained losses of U-235 should be a source of real concern to the Commission because of the possibility that such losses may represent actual theft of fissionable material by individuals who intend to use it against the interests of the United States. The magnitude of unexplained losses should set the intensity of effort to determine their cause.

A judgment of the probable magnitude of the unexplained losses of U-235 may be obtained from observable data as follows:

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to special restrictions and chemical reactions.

<u>Quantity</u>	<u>Significance of Quantity</u>	<u>Jan.</u>	<u>Feb.</u>	<u>March</u>	<u>etc.</u>
12 x material acctg. deficiency for month.	Current observed annual rate of consumption plus thefts.	D_1	D_2	D_3	etc.
Material acctg. deficiency for preceding yr.	Current estimate of annual rate of consumption plus regular thefts, from past material accounting.	E_1'	E_2'	E_3'	etc.
Calculated annual rate of consumption for current operating conditions.	Estimate of annual rate of consumption, from consumption test data.	E_1''	E_2''	E_3''	etc.

Entries for total uranium losses, average U-235 assay of lost material, etc. could be added. Other tables for different time intervals could also be carried. Rules could be established for the extent to which discrepancies between values in rows 1, 2, and 3, and in cumulative averages, should lead to additional effort to locate the unexplained losses which they indicate. These rules should take into account the two types of error which could be made in interpreting unexplained losses, and which the Commission wishes to avoid: (1) the cumulative average value of the unexplained losses (or the losses for a particular year) may be judged higher than the tolerance limit set by the Commission when actually no illegitimate losses have occurred, and effort and expense may be devoted to determining the cause of non-existent losses; (2) the average unexplained losses may be judged not significantly different from zero when illegitimate losses have actually occurred. A balance between the expense of locating an unexplained loss and the expense of the hazard resulting from such a loss is necessary before it will be possible to formulate, by statistical methods, consistent rules for the proper action following the reporting of unexplained losses.

A systematic quality control program for uranium accounting should be worked out by a trained statistician, who should keep under continuous critical review the recommendations of this memorandum.

3. Conclusions

3.1 Adequacy for Carbide Organization for U-235 Accounting Control

The uranium material accounting methods now employed at the gaseous diffusion plant are well conceived and well executed, and reliable results are being secured for the monthly and annual U-235 material balance deficiency. On the other hand, far too little information is now available concerning the rate of consumption of U-235 by plant surfaces and other legitimate process losses, so that no reliable estimate can now be given of the unexplained U-235 losses, i.e., the portion of the material balance deficiency which cannot be attributed to consumption or other measurable types of loss.

Material accounting procedures result in a value for the annual material balance U-235 deficiency, D , with its limit of error $\pm d$. This deficiency is computed so as to represent an estimate of the sum of legitimate U-235 losses, such as consumption within the plant, plus improper diversion; the annual material balance should not include credits for U-235 returned to the cascade from decontamination operations which recover U-235 lost sight of by the material accounting procedure during previous years. From D must be subtracted an estimate of the legitimate, explainable annual U-235 losses (such as consumption) $E \pm e$, to obtain a value for the unexplained annual losses of U-235, $L \pm l$, where $l = \sqrt{d^2 + e^2}$.

Judgments concerning the magnitude of the annual legitimate U-235 losses, E , may come from one of two sources. First, so long as plant operating conditions are steady, values of the material balance deficiency D for previous operating periods provide an estimate of E for current operations. To obtain this estimate there must be considerable sacrifice in operating flexibility to achieve long periods of steady operating conditions. This estimate of E is subject to two uncertainties, to which statistical weights cannot be assigned: the possible existence of a regular bias in the value of E caused by a source of error unthought of by the material accounting staff, or possible regular thefts of U-235 by a clever person who is aware that irregularities in the value of D may call attention to his activities. A second judgment of consumption losses E , not subject to these uncertain biases, can be obtained from actual consumption test data on plant equipment; stages may be periodically removed from the plant and inspected for uranium content, or stages may be operated in isolation from the rest of the plant and the decline in visible uranium observed. *if possible get two samples*

In a given year, a value of the unexplained annual loss L greater than its limit of error l , should be regarded as improbable, and as a signal for effort to locate the reason. Over a period of years, the average unexplained losses of U-235, \bar{L} , should stabilize to a value considerably less than the upper limit of error l for a single year's operation. (The limit of error of the average is $\pm l/\sqrt{\text{number of years}}$.) The average unexplained annual losses, \bar{L} , may result from biases in observations entering the uranium balance, actual losses which are unknown to the material accounting staff and hence not included in its estimate of E , or diversion. If \bar{L} is larger than a value judged to represent a potentially intolerable hazard, effort should be expended to locate and determine the unknown biases or unknown legitimate sources of loss, if any, which contribute to these unexplained losses.

Monthly judgments of the need for additional effort to locate unknown biases or sources of loss can be formed if material accounting is put on a quality control basis. The following schematic procedure is offered only as an example of what might be done.

3.2 Importance of Steady Operating Conditions

Attainment of reliable estimates of U-235 loss in the past have also been hampered by adoption of frequent changes in operating conditions which have affected the U-235 inventory and consumption rate in ways which are almost impossible to estimate. A notable example of this is the introduction of a in April 1947, and change in location of in October 1947 and March 1948. should not be changed again. Such experimenting with the operation of the plant may increase production, but it is certain to confuse interpretations of material balance records.

The barrier replacement program planned for 1948 will have a similar adverse effect on reliability of knowledge of U-235 losses for 1948. Uncertainties will result from the initial take-up of UF₆ on the fresh metal surfaces and from the changes in plant pressures and cascade U-235 concentration gradient which will occur when the new barrier is put into operation. For these reasons, it must be expected that the material accounting record for 1948 will have a limit of error much larger than the ultimate target recommended in this report.

Changes in operation should be made only when it is established that an increase in production will result and that this increase will more than compensate for the loss in control of U-235 accountability.

3.3 Need for Criteria to Judge Relative Importance of Production and Accounting Control

The lack of information regarding consumption and the frequent changes in plant operation is partly a consequence of the emphasis placed in current K-25 operations on the maximum possible U-235 production, without the slightest sacrifice of production for the measurement of consumption or for improving the precision of the material balance. Carbide needs and wants criteria which will enable them to judge the relative importance of maintaining U-235 production on the one hand, and of reducing the unexplained loss of U-235 on the other. Among the recommendations of Section 4 of this report are listed specific operating changes for reducing unexplained losses at the sacrifice of small amounts of production, which could be developed by the Commission into definite criteria of the type Carbide needs.

3.4 U-235 Material Balance Deficiency

The U-235 material balance deficiency of the K-25 cascade for the year 1947 was 100 kgm., when the 88 kgm. deficiency reported by Carbide is adjusted for the use of the experimentally determined U-235 content of feed (0.711 wt.%), instead of the less reliable nominal value (0.705 wt.%). It is possible that a lower figure will be reported in future years as consumption is reduced, but in setting a target for the unexplained loss it is assumed that the 100 kgm. annual deficiency will continue.

3.5 Precision of U-235 Material Balance Deficiency

With operational and material accounting techniques presently in use at K-25, the limit of error in the annual U-235 material balance deficiency is about ± 30 kgm./yr. Statistical analysis of causes of variation of material balances for previous periods, tests on plant instruments, and relatively minor changes in operating techniques, which Carbide now have under way, should reduce this limit of error to ± 20 kgm. U-235/yr. at the 95% probability level.

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3.6 Estimate of Losses of U-235 by Consumption

No reliable data on consumption are now available. Measurement of the uranium content of ten convertors from the section of the K-25 cascade handling material near normal uranium in isotopic assay suggests that if uranium has been consumed at a uniform rate since the plant was put into operation, the rate of U-235 loss in the entire cascade would be of the order of 150 kgm./yr. at the level of product purity in effect since January 1, 1947. This estimate is extremely unreliable. Much more data on consumption are needed before an estimate with known limit of error can be given.

3.7 Present Estimate of U-235 Losses for 1947

Subjectively, it seems probable that the entire material balance deficiency of 100 kgm./yr. reported in 1947 is attributable to consumption, but this point will not have been established until a much larger number of convertors and other cell equipment have been cut out of the cascade and analyzed for uranium than has yet been done.

3.8 Plans for Measurement of Consumption

In the barrier replacement program planned by Carbide for 1948 almost all of the convertors in Plant III will be removed for retubing. It is estimated that about three-quarters of all the U-235 lost by consumption has been deposited in the stages of Plant III. By decontaminating a sufficient fraction of these convertors and other components of these stages, and sampling stages from other sections of the plant, it should be possible to obtain as reliable a measure of consumption from the time the plant went into operation to the time the stage was opened as one would wish. By making plausible assumptions about the distribution of these losses with time, it should be possible to estimate the consumption for the year 1947 with a limit of error of ± 25 kgm. U-235/yr.

3.9 Ultimate Estimate of Unexplained Loss for 1947

When this retubing and decontamination program has been completed (which should take place in 1949), it is expected that the unexplained loss of U-235 during 1947 will have been reduced to a small fraction of the 100 kgm. material balance deficiency for that year. The limit of error for the unexplained loss for this period will be ± 40 kgm., ~~± 30 kgm.~~ for the material balance deficiency itself and ± 25 kgm. for consumption losses.

3.10 Limit of Error for Unexplained Losses in Future Years

After this barrier replacement program has been completed it will be important to continue to remove a sufficient number of representative stages for decontamination and uranium analysis to permit an estimate of current consumption with whatever reliability the Commission specifies. A limit of error on consumption of ± 25 kgm. U-235 per year seems attainable without serious sacrifice of production, but at some cost for maintenance and materials. This, combined with the limit of error in the material balance deficiency ± 20 kgm. U-235 per year deemed ultimately attainable, leaves a limit of error of ± 30 kgm. U-235 per year in the unexplained losses as an ultimate goal.

4. Recommendations

4.1 Target for Unexplained U-235 Loss

This limit of error is the basis for recommending the following targets for unexplained U-235 loss:

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- (1) In no one year should the unexplained U-235 loss exceed 30 kgm.
- (2) Over a period of years, the cumulative unexplained loss, together with its upper limit of error evaluated for the full period of time, should not exceed a value judged by the Commission to represent a potentially intolerable hazard.

4.2 Quality Control of U-235 Accountability

Carbide should institute and maintain a systematic quality control program on the U-235 material balance deficiency and on measurements of consumption and other process losses.

4.3 Systematic Consumption Measurements

It appears that the only presently known way of measuring U-235 consumption with a limit of error of ± 25 kgm. per year is by removing parts of a representative sample of a number of stages on a regular schedule, and decontaminating them to determine the amount of uranium deposited in them. The greater the number of stages removed per year the lower will be the limit of error in consumption, but the greater will be the cost and the interference with production. After such a program has been instituted, it will be possible to state from the variability of results on individual stages what the limit of error in the estimate of consumption will be as a function of the cost and the loss in production. The barrier replacement program now under way will provide evidence of the variability of consumption among stages, and will give the Commission a concrete case for developing a criterion of the relative importance of U-235 accountability compared with cost and production rate. When the results of decontamination of converters removed in the present barrier replacement program become available, it is recommended that the Commission determine the number of converters to be removed annually, and request Carbide to institute a systematic program of measuring consumption by regular removal and decontamination of stage parts.

Another possible method of measuring the consumption rate is to valve off cells or buildings from the cascade without physically removing them for decontaminating and measure the rate of loss of uranium from the visible cell gas inventory. Up to the present time, measurements of this type have given extremely variable and inconsistent results. Notwithstanding these discouraging results, tests of this method should be continued in the hope of bringing it under control and of obtaining information supplementing that obtained by the program of equipment removal.

4.4 Separate Material Accounting for Plant III

One of the most important factors contributing to uncertainty of the interpretation of the material balance deficiency is the impossibility of localizing U-235 losses within the cascade as presently constituted. It is thus impossible to tell at what concentration U-235 losses have taken place, except for such indirect evidence as is afforded by the uranium balance. If a separate material balance could be made on Plant III, it would be possible to determine how much U-235 had been lost at concentrations above 15%, and the limit of error of the material balance and the unexplained loss could be materially reduced because of the smaller volume of material and equipment to be watched.

With the present type of connections between Plant II and Plant III, about 150,000 lbs./day of heads containing 15% U-235 enters Plant III, and the same amount of tails of almost identical composition flows back to Plant II. This makes a separate

material balance on Plant III impossible. However, if the junction between Plants II and III were altered to permit the overlap of one or more buildings, as done in connecting the K-25 and K-27 cascades, these flow rates could be reduced to 500 lbs./day, a manageable quantity, and an independent material balance on Plant III might become possible. It is estimated that reduction of the interplant flow to this extent would reduce the product purity around 1% without affecting the U-235 production rate, or reduce the production rate around 2% without affecting the product purity.

It is recommended that the Commission request Carbide to study the cost and feasibility of altering the connections between Plants II and III in this way, and to estimate the reduction in output and the increase in precision of material accounting which might be obtained from various numbers of buildings overlapped. Such a study would provide the Commission with another concrete case for developing a criterion of the relative importance of production vs. accountability.

4.5 Change in Nominal U-235 Content of Feed

Ample evidence has been obtained at the K-25 Works and Research Laboratories to establish that the U-235 content of present plant feed is 0.711 wt.% when the assay is made by the same methods used for the waste and product streams, instead of 0.705 wt.%, the value now specified by the Commission. Even if there is some doubt as to the "true" value of this assay, it is recommended that Carbide be allowed to use their measured value in U-235 material balances, to eliminate bias in the feed assay.

4.6 Daily Analysis of Feed

It is remotely possible that an occasional lot of UF₆ from Harshaw may have been contaminated with an isotopic content different from normal feed. To guard against this possibility, it is recommended that either

- (1) A composite sample of feed be taken each day at the K-25 plant and analyzed by comparison with standard (0.711%) uranium, or
- (2) A sample be taken from each lot of Harshaw's production and sent to K-25 for analysis as under (1).

4.7 Initial Consumption

One important source of uncertainty in future consumption estimates will be the abnormally high consumption rate which will occur when a converter not previously exposed to UF₆ is filled with this gas. For example, a high unexplained loss will be observed in 1948 because of installation of new barrier, unless the high initial consumption on this barrier is measured and allowed for in computing the unexplained loss. It should be possible to measure the initial consumption by filling a cell containing new barrier with UF₆ of the current isotopic content from a weighed container (instead of from adjacent cells of the cascade as is usually done) until normal operating conditions have been established. The difference between the weight of UF₆ charged and the cell inventory computed as done in the uranium material balance will give the effective initial consumption. It is recommended that Carbide be asked to develop a procedure of this type to measure initial consumption. If this is not done, the unexplained U-235 losses for 1948 will be very high, probably outside of the target set in 4.1

4.8

It is hard to determine whether the has been effective in preventing consumption or in removing uranium from plant surfaces. If it is effective,

Leadings 1
Source 14-A

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Its use will adversely affect uranium accounting unless it is kept steady. It is recommended that the present technique be continued without change.

5. Appraisal of U-235 Material Accounting Problem

U-235 material accounting at the K-25 plant is much more difficult than in conventional chemical plants because:

- (1) The cascade inventory is many times greater than the daily production rate.
- (2) The cascade inventory is contained in a piping network of quite unprecedented complexity and extent, at widely variable temperatures, pressures and UF_6 concentrations.
- (3) The cascade contains many dead-ended volumes whose isotopic assay may differ from the main streams.
- (4) Concentration of UF_6 by plant surfaces proceeds at an imperfectly known and variable rate.
- (5) Exchange of U-235 and U-238 gradually takes place between gas and condensed (solid or adsorbed) phases.
- (6) The UF_6 inventory in contact with metal surfaces is greater than predicted by the gas laws because of adsorption.
- (7) Isotopic analysis is less accurate than chemical analysis.
- (8) Accidental losses may occur at a great variety of points from so complex a plant.

For these reasons, attainment of satisfactorily low unexplained U-235 losses will be a difficult and costly undertaking. Nevertheless, given encouragement by the Commission and diligence on the part of Carbide, it is believed that the unexplained loss of U-235 can be brought and kept below 30 kgs. per year by 1949.

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