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Recent Developments in Uranium Enrichment

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URANIUM MOLECULAR LASER ISOTOPE SEPARATION

The Molecular Laser Isotope Separation program is moving into the engineering phase, and it is possible to determine in some detail the plant cost terms involved in the process economics. A brief description of the MLIS process physics is given as a motivation to the engineering and economics discussion. Much of the plant cost arises from lasers and the overall optical system. In the paper, we discuss lasers as operating units and systems, along with temporal multiplexing and Raman shifting. Estimates of plant laser costs are given.

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During the past 20 years, there has been a revolution in optics, optic devices, and the application of optical techniques, which was triggered by the invention of the laser in the early sixties. This new technology has found novel and new applications in medicine, communications, chemical analysis, the metal and cloth fabrication industries, and the areas of military target acquisition, ranging, and weaponry as well as others. An area that has seen intensive research but has yet to demonstrate an industrial application, is the area of laser photochemistry. The fundamental research in laser photochemistry has led to the discovery of methods for highly efficient isotope separation, new methods for purification, the in situ generation of gas-phase catalysts, the driving of unimolecular reactions, and many other novel research discoveries. The frequency purity of the laser, coupled with the availability of a multitude of wavelengths ranging from 200 nm to greater than 100 μm , will ultimately result in applied industrial photochemical applications where the required photons and their cost compete favorably with conventional chemical technology. In the areas of laser separation of fissile isotopes, two processes, the Molecular Laser Isotope Separation (MLIS) process and the

Atomic Vapor Laser Isotope Separation (AVLIS) process, are rapidly advancing to the industrial demonstration phase. In this paper, we will touch briefly on the fundamentals of the MLIS process and present some of the laser and optical considerations that are attendant with industrialization of this process.

Excellent topical and review articles on applied photochemistry have been presented by V. S. Letokhov, (1,2) C. B. Moore, (3) J. P. Aldridge, et al, (4) and many others too numerous to mention here. Articles specific to molecular laser isotope separation include many articles by the team of scientists and engineers working on this process for uranium separation at the Los Alamos National Laboratory (5 to 10).

DISCUSSION

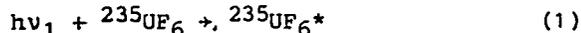
Molecular Laser Isotope Separation of Uranium

Because of the extensive industrial infrastructure and technology base associated with the use of UF_6 in the uranium fuel cycle, this molecule was chosen as the prime candidate around which a molecular laser isotope separation process should be built. The vapor pressure of UF_6 compared to other uranium bearing molecules (126 torr at room temperature), the isotope shift of about 0.6 cm^{-1} , and the reactivity of UF_6 permit

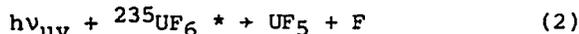
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ease of handling with conventional materials and separation with easily achievable laser bandwidths.

As discussed elsewhere, (9) the number of fundamental overtones and combination vibrational modes available to UF_6 make it necessary to simplify the vibrational spectra by aerodynamically cooling the molecule. This is accomplished within the bounds of conventional supersonic flow technology by mixing the UF_6 with a suitable carrier gas, such as Ar or N_2 , and expanding the mixture through a supersonic nozzle. Aerodynamic cooling results in a large fraction of the UF_6 occupying the ground state where it is amenable to selective photophysical processing. The basic process for the separation of uranium isotopes is one wherein the $^{235}UF_6$ is selectively excited with an ir laser according to the reaction,



The absorption of one or more infrared photons by the UF_6 results in a broadening and red shift of the vibrational absorption spectra. An increase in uv absorption cross section of the excited $^{235}UF_6$ results in preferential dissociation of the excited molecules.



The above sequence of events leading to photodissociation is depicted in Figure 1a.

An alternative method of selective dissociation of UF_6 wherein a single laser is used is shown in Figure 1b. This second method, which is known as multi-photon dissociation, has been discussed by Robinson (11) and by Judd (12) as well as others.

The criteria that must be satisfied by the interaction of one or more lasers with the UF_6 are:

Selectivity. The spectral difference between the isotopic molecules must be sufficient to permit a major excitation of one isotopic molecule over the others.

Quantum Efficiency. The absorbed laser energy must lead to the desired reaction or dissociation with reasonable efficiency.

Separation. After the selective step, separation of the product must be accomplished with high efficiency and with a minimum of scrambling reactions.

Throughput. The key to industrialization of selective laser-based separation processes is high throughput. In the gas-phase photochemical processes, this means working at as high a feed-stock density as possible.

Overall Process Efficiency. The net energy expended in separating the desired product molecule from the mixture should be minimized.

Table 1 presents a comparison of some of the parameters of molecular laser isotope separation with gaseous diffusion technology. The comparisons of Table 1 show that the deployment of MLIS technology would substantially reduce the staging requirements of gaseous diffusion, result in a factor of 20 to 40 reduction in power consumption, and result in end-product costs that are projected to be about one-fifth of gaseous diffusion.

A schematic of the main elements of MLIS process is presented as Figure 2. Basing the uranium MLIS process on UF_6 has resulted in the situation where all of the production plant parameters for the gas flow system can be specified with a high degree of confidence. This includes the UF_6 feed system, the nozzle, solid UF_5 collector, interstage equipment, and the gas cleanup and tails storage components.

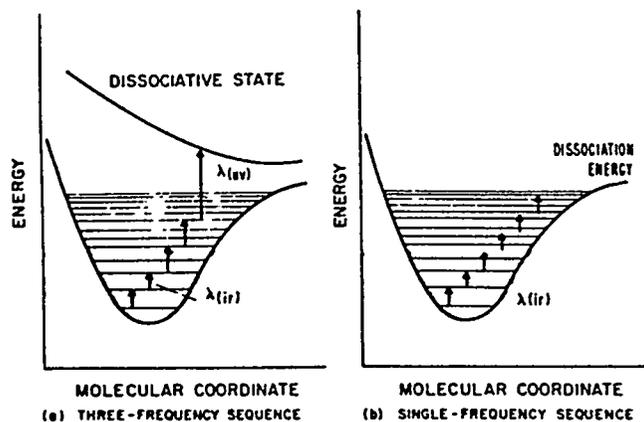


Figure 1. Methods of photophysical dissociation of UF_6
 a. Infrared plus uv sequence
 b. Single-frequency sequence

Table 1. Comparison of molecular laser isotope separation with gaseous diffusion.

Feed Material	Gaseous Diffusion	Molecular MLIS
	UF ₆	UF ₆
Selectivity (α-1) ^(a)	2 × 10 ⁻³	1-8
Energy (kWh/SWU) ^(b)	2600	50-120
Net Separation Cost (\$/SWU)	90-140 ^(c)	15-40
Power Costs (\$/SWU)	68	~2
Status	Production	R&D ^(d)

^a α is defined as the ratio of the isotopic abundance ratios of the product and tails streams of a single stage.

^b SWU refers to a kg-separative work unit as conventionally defined in uranium enrichment processes.

^c The range of costs varies for the existing US diffusion-plant complexes vs new US or European plants and by the method of financing.

^d Engineering-scale MLIS facilities are planned to be placed into the existing diffusion plants starting in the mid-eighties.

out on both ir and uv lasers. To minimize costs and at the same time realize the technology advancements of existing lasers, it would be highly desirable if the plant lasers for the MLIS uranium process could be based on CO₂ laser technology. The desirable result has been achieved through the use of highly efficient Raman cells to shift CO₂ laser wavelengths to those of the ν₃ vibrational band of UF₆ (15.9 μm). The invention of rare-gas-halide lasers has solved the laser problem in the uv with a free running gas laser very similar to CO₂ lasers.

The specific requirements for the ir and uv lasers are dictated by the process physics and the conditions that must prevail in the irradiation zone to optimize uranium monomer density. Figure 3 shows a schematic of the irradiation zone and indicates the important parameters that must be considered in designing the photolysis zone.

1. NOZZLE EXPANSION COOLS THE GAS TO REVEAL ISOTOPIC FEATURES:

2. LASER BEAMS BREAK SELECTED MOLECULES INTO A COLLECTABLE POWDER.

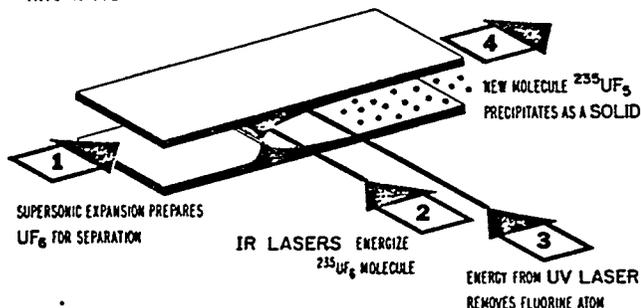


Figure 2. Molecular laser isotope separation process.

Optimization of the MLIS process for uranium requires further effort in laser development and in certain elements of the beam transport and multiplexing system.

LASER REQUIREMENTS FOR MLIS

The fundamental nature of the MLIS process for uranium, which depends on selective resonant and nonresonant vibrational excitation of UF₆ followed by enhanced photodissociation of the excited species, has dictated that laser development be carried

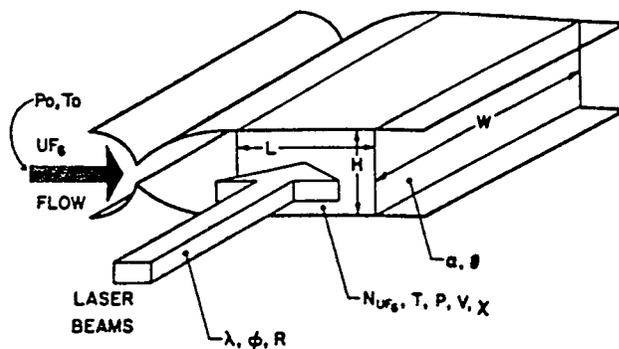


Figure 3. Nozzle and irradiation zone.

The wavelengths of the ir and uv lasers are determined by ir and uv spectroscopy measurements and are set at values that optimize enrichment (α) at a product cut (θ), which will result in an economical process. The cut for the process is determined by the ²³⁸UF₆ uv absorption cross section (σ) and the fluence (φ) of the uv laser. The number density for the process is optimized for the required α, which is temperature dependent, and for the temperature (T), which governs the length (L) over which the monomer number can be maintained without excessive condensation. The required irradiation zone temperature in turn fixes the flow Mach number since the nozzle inlet stagnation temperature is essentially fixed at ambient conditions or

slightly above ambient. The Mach number and the gas composition determine the required flow velocity (V), which in combination with the flow length (L), determines the laser repetition rate required to irradiate all of the flow passing through the irradiation zone. The depth (W) of the irradiation zone is governed by the necessity of maintaining a uniform enrichment over the depth; and since the ir absorption is a function of fluence, this requirement translates to one of maintaining a uniform fluence for the ir laser, which is most strongly absorbed. The height of the irradiation zone can be chosen to maintain reasonable beam sizes for the required fluences. The number of nozzles for a given stage in the plant is determined by the required throughput and the available flow area (H x W). The large number of variables involved in the process makes it necessary to perform several iterations to achieve an optimum irradiation zone and plant design.

For the MLIS uranium process, the objective is to design the flow system so that it conforms to the requirements of the lasers and the required uniformity of irradiation in the photolysis zone. To meet full production plant energy and repetition rate requirements, with moderate extrapolations of current technology, it is necessary to both spatially and temporally multiplex laser beams. The required values for laser parameters are indicated in Table 2.

Table 2. MLIS laser requirements.

Parameter	Symbol	Units	Parameter Value	
			ir CO ₂ Pump	uv Laser
Wavelength	λ	μm	15.9	0.308
Energy per pulse	E	J	3.4	3.6
Repetition rate	R	kHz	1.25	1.25
Efficiency	η	%	6.7	1.2-1/6

A typical configuration for the ir laser systems is shown in Figure 4. A MOPA arrangement is used to achieve the required beam energy. After conversion of the 10 μm CO₂ radiation to 16- μm radiation through the use of the efficient (about 50%) Raman cell, the beams from eight MOPA chains are multiplexed together to achieve a repetition rate of 10 kHz. The requirements indicated

in Table 2 are for a single amplifier stage of the MOPA chain shown in Figure 4.

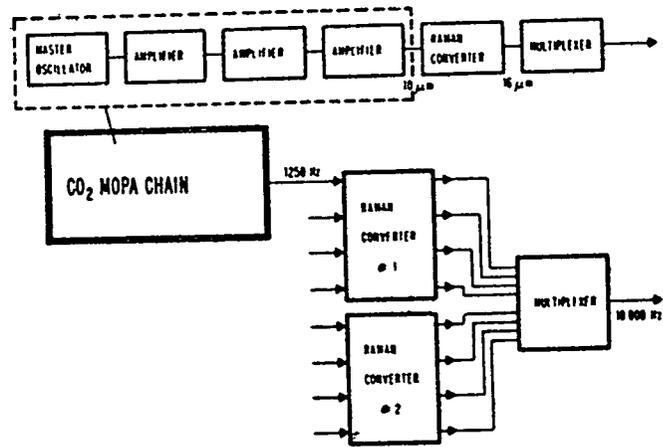


Figure 4. Infrared laser system.

A schematic of a typical uv laser system is shown in Figure 5 and consists of a rare gas halide master oscillator and a single power amplifier with the characteristics listed in Table 2.

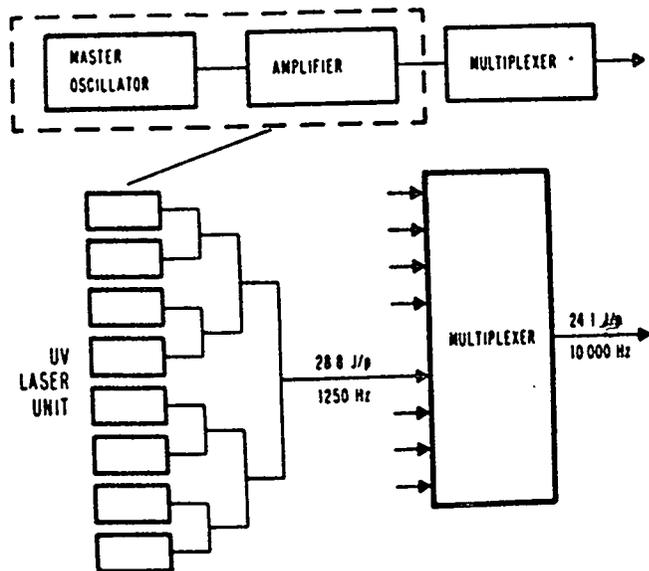


Figure 5. Ultraviolet laser system.

To achieve the correct energy in the uv beam, the beams from individual units are spatially multiplexed together, as indicated in Figure 6, using a mirror arrangement called a dihedral beam combiner.

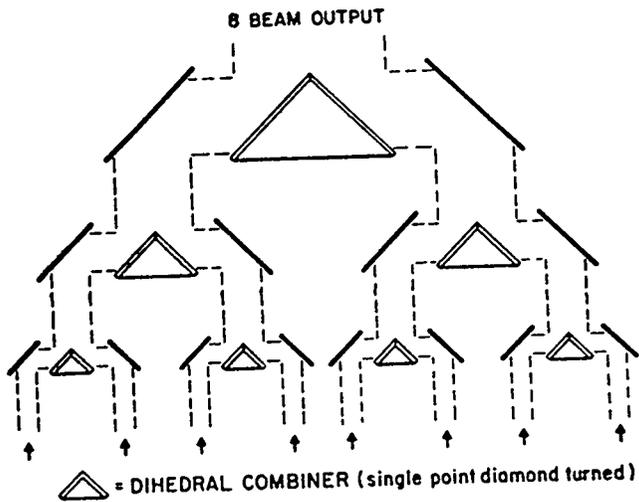


Figure 6. Spatial beam combiner.

A conceptual design of the ir laser systems is shown in Figures 7 and 8. As shown, the CO₂ pump laser consists of two discharge heads embedded in a common recirculating flow system. The conceptual design of the uv laser is very similar to the ir in appearance and is shown in Figure 9.

Temporal Multiplexing

As noted above, the industrialization of the MLIS uranium process requires both ir and uv lasers that deliver kilohertz repetition rates. To obtain the full repetition rate needed to irradiate all of the gas passing through the photolysis zone, the ir beams and uv beams must be temporally combined to form a single beam. A schematic diagram of a device that can accomplish this for all beams and serve as a master timer for laser triggering is shown in Figure 10. The device consists of a faceted wheel with each facet sized to accommodate the cross-sectional area of eight laser beams without overlap. The wheel is water-cooled and attached to a spindle that is driven with a constant speed electric motor through a timing belt. Figure 11 shows more detail of the water cooling and wheel design. The wheel facets will be machined by diamond turning and then coated for optimum reflection of the ir beams on one side and for the uv beams on the opposite side. This device, which consists of commercially available components except for the wheel, operates at a low rotational speed, is compact and simple in design, and is designed to maintain beam quality by limiting thermal distortion of the reflective surfaces.

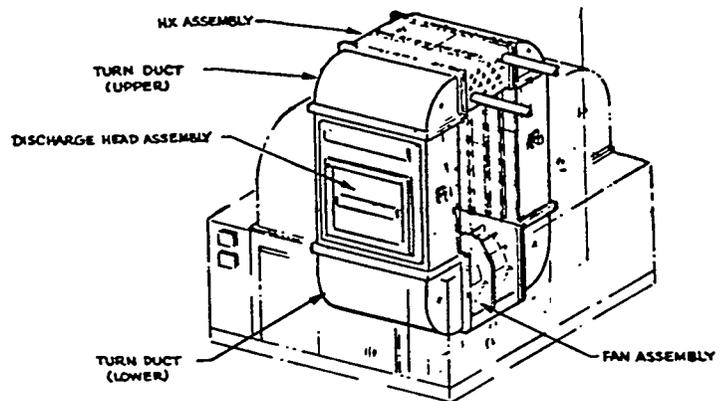


Figure 7. Flow system configuration for infrared laser system.

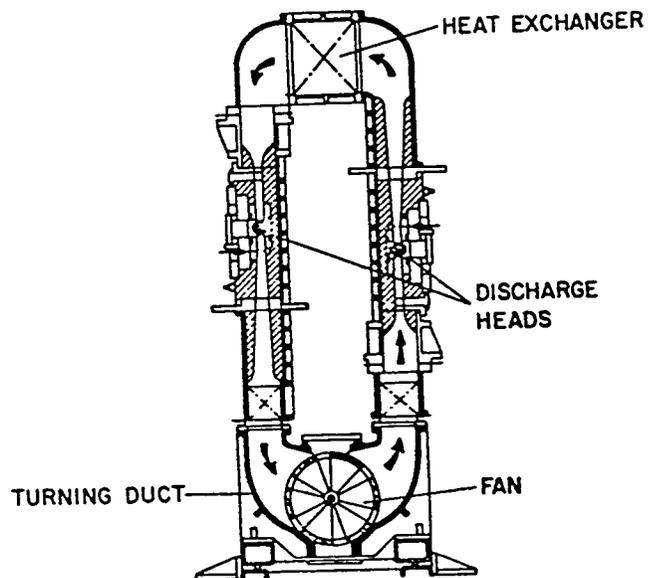


Figure 8. Schematic of dual discharge head ir laser system.

Raman Conversion

One of the key elements in the MLIS uranium process is the Raman wavelength conversion systems. Raman systems are highly efficient, with conversion efficiencies greater than 50% demonstrated. The simplest form of a Raman cell is a cylindrical tube filled with the scattering gas and fitted with end mirrors and beam insertion and extraction optics. A pump beam and a seed source are inserted into the cell and multipassed within the cell to achieve the desired output power. A schematic of a static two-stage Raman cell is shown in Figure 12.

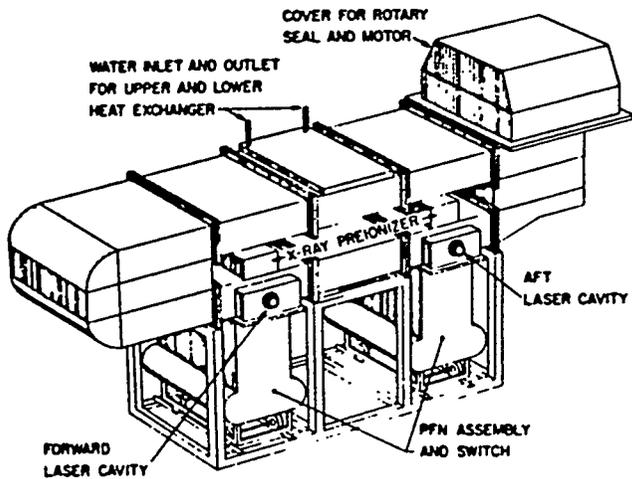


Figure 9. Ultraviolet laser concept.

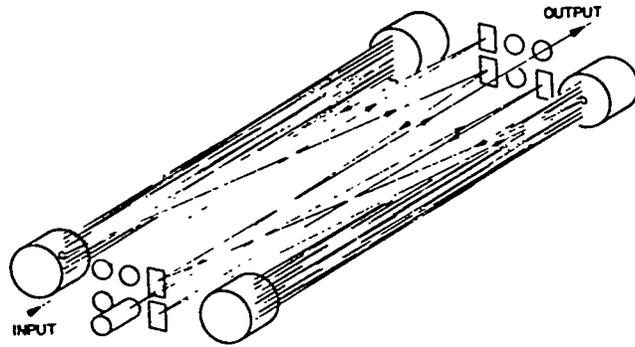


Figure 12. Two-stage multipass Raman converter.

Figure 13 shows the depletion of a CO₂ pump beam and the buildup of the 16- μ m output as a

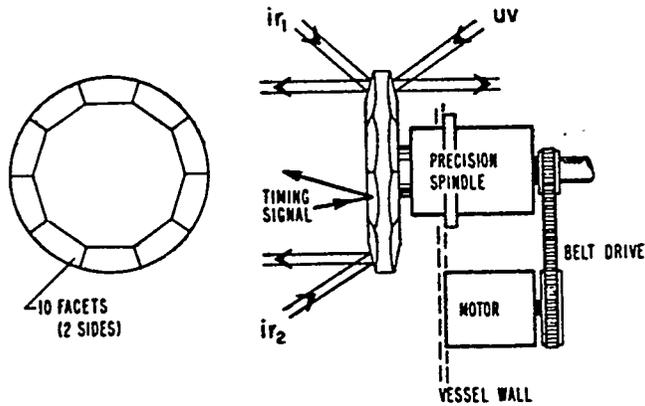


Figure 10. Multiplexer schematic.

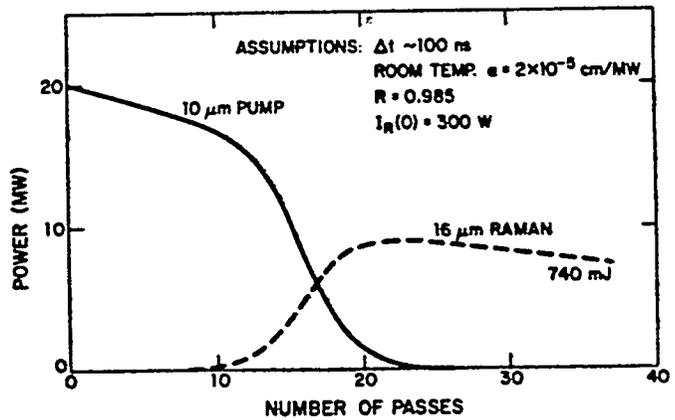


Figure 13. Calculated raman conversion.

function of number of passes through a hydrogen-filled Raman cell held at room temperature. As can be seen from the figure, about 20 passes are required to fully deplete the pump beam. The beam profile for the CO₂ pump, before and after depletion in the cell, and the 16- μ m output are shown in Figure 14.

To accommodate the beam powers needed in MLIS of uranium, the Raman gas must be circulated and cooled. A schematic of a large Raman converter that would provide four Raman cells that could operate simultaneously is shown in Figure 15, and an isometric of a similar two-cell system shown in Figure 16 will be tested at Los Alamos in early 1982.

Irradiation Chamber Considerations

The design of an MLIS process for uranium requires the design of a photolysis

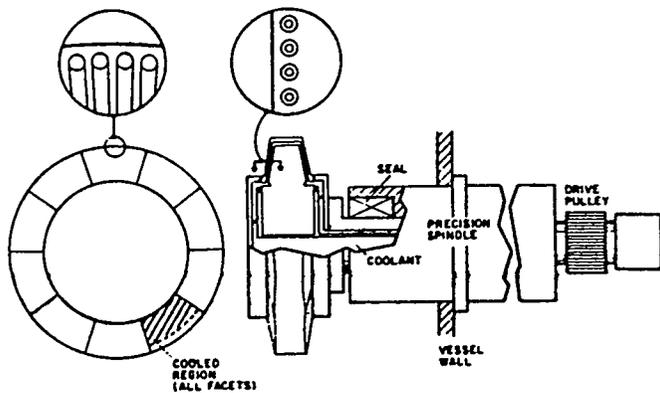


Figure 11. Multiplexer design approach

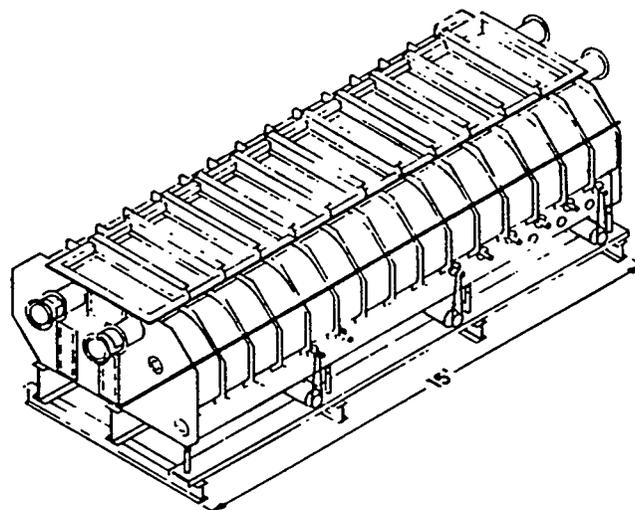
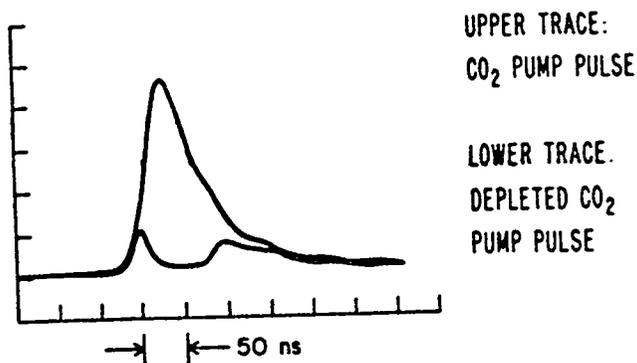


Figure 16. Two-cell room-temperature Raman converter.

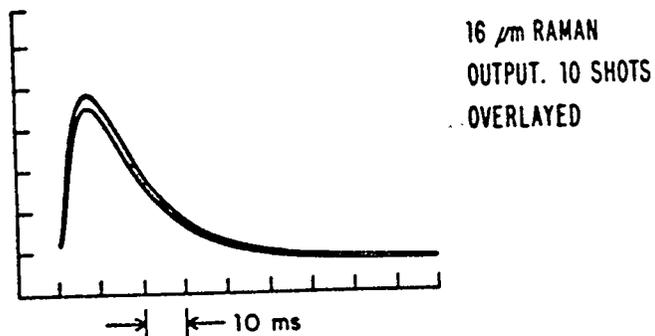


Figure 14. Raman conversion pump and output beam profiles.

chamber that integrates the process physics, engineering, and economics. The design must accommodate different laser wavelengths (with different UF₆ absorption cross sections for each wavelength) and variations in pulse-timing and in laser fluences. For a plant, the photolysis chambers must efficiently utilize the laser photons and at the same time provide a high enrichment over the full width of each nozzle of each stage. The correct fluences can be maintained by beam compression using two-stage telescopes between stages, and optical losses can be minimized by using reflective optics and by providing separate components for the ir and uv beams.

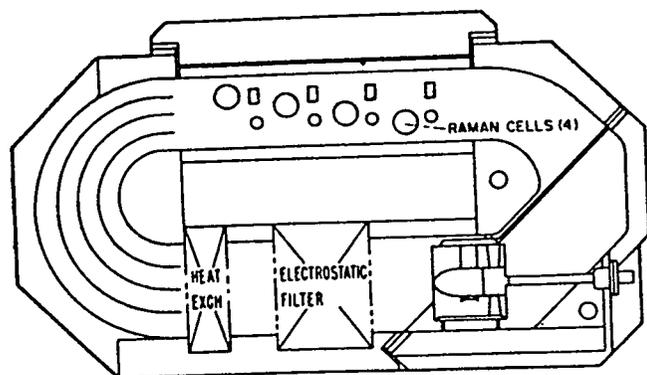


Figure 15. Four-cell room-temperature Raman converter.

The design must provide for good photon utilization and not introduce diffraction effects. A number of such designs are being developed at Los Alamos. Figure 17 shows the relative photochemical performance in a chamber as a function of clipping loss for a Gaussian profile. Figure 18 shows the propagation characteristics of clipped Gaussian beams. Figure 19 shows the intensity pattern for a side-by-side insertion scheme of two beams.

Figure 20 shows typical intensities across a centerline of the reaction chamber for Type I input where the side-by-side beams happen to line up in phase and Type II input where the side-by-side beams are out of phase. Figure 21 shows the performance obtainable with the use of super Gaussian

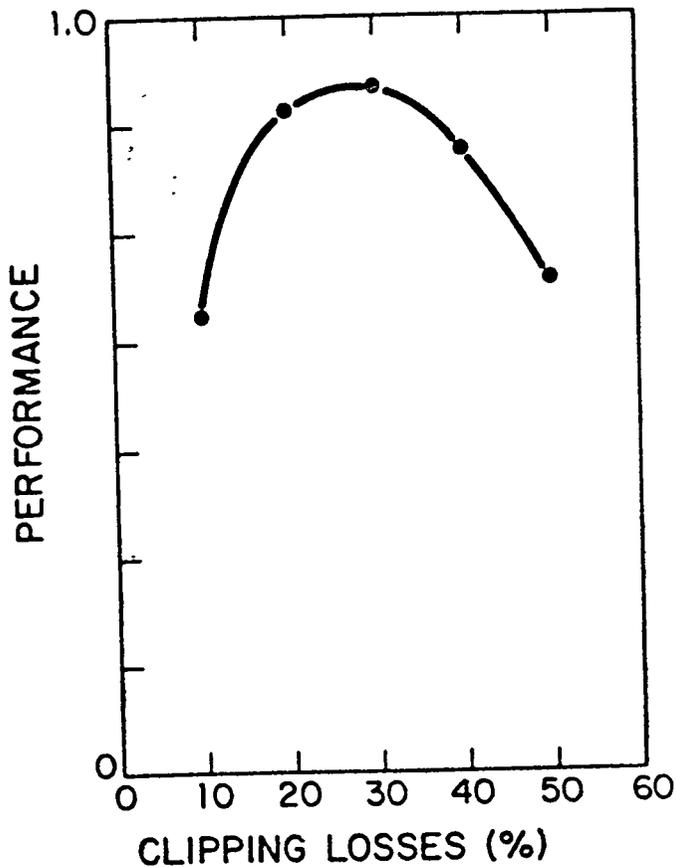


Figure 17. Average performance as a function of ir beam clipping.

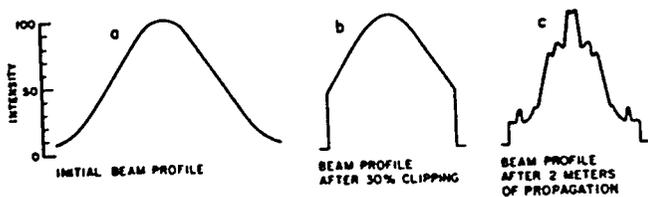
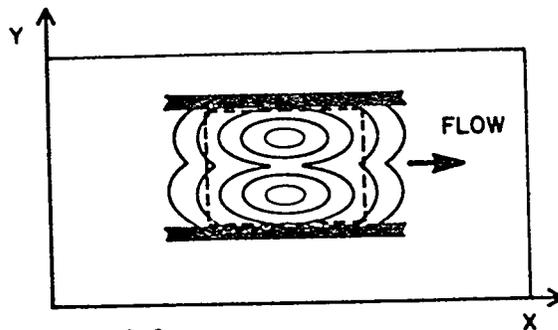


Figure 18. Infrared beam profiles in photolysis chamber.

beams. Obviously, these beams have a better filling factor than normal Gaussian beams, but they introduce the problem of rapid degradation of the beam as it propagates.

For the several meters of pathlength required for each photolysis chamber, theoretical calculations using a detailed multi-level model predict that self-focusing of the resonant ir_1 beam will not present a problem.



UV PHOTOLYSIS REGION

Figure 19. Profiles inside photolysis chamber.

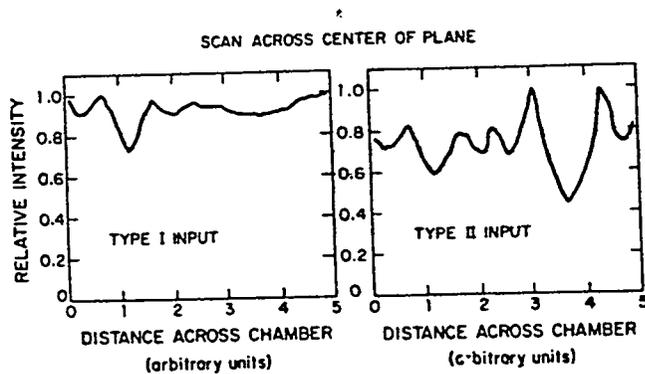


Figure 20. Intensity profile inside photolysis chamber.

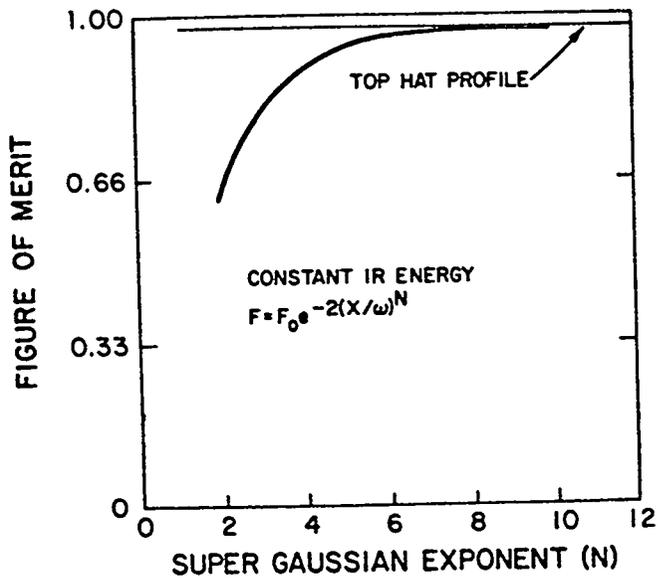


Figure 21. Separation vs. super Gaussian beam profile.

Production Plant Costs

The principal reason for extensive investigation of advanced isotope separation processes is the potential for significant reduction in both capital and operating costs paid for enriched uranium. Direct comparisons of advanced uranium isotope separation techniques (MLIS, AVLIS, and PSP) have consistently indicated that these new processes have the potential for producing reactor-grade uranium at costs per separative work unit of less than \$40.00. Figure 22 shows a comparison between the costs of conventional enrichment methods and advanced techniques. The basis for this comparison is a 9 M SWU/year production plant operating with natural feed. As can be seen from Figure 22, the projections for a new MLIS plant show separative work costs that are significantly less than the projected costs for diffusion-enriched material when only the

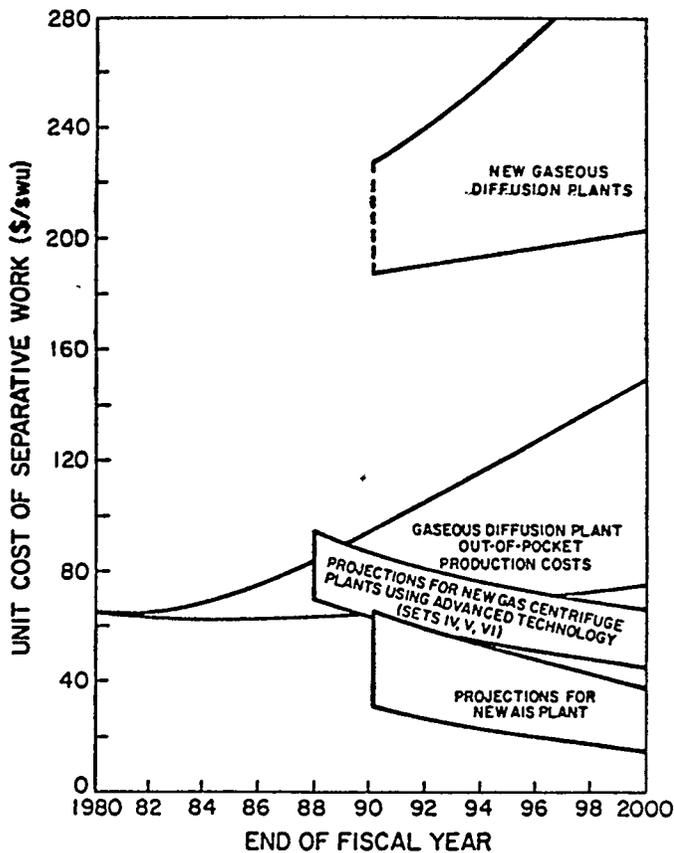


Figure 22. Projected costs for uranium enriched by standard and advanced technologies.

operating costs for diffusion are considered. This very favorable result is due to the low

operating costs projected for an MLIS plant when compared to gaseous diffusion. The cost of electrical power enters strongly into the comparison since a typical 9 M SWU/year gaseous diffusion plant requires about 2300 MW of electrical power per year, whereas an MLIS plant would only require about 100 MW. The comparison shown in Figure 22 also indicates that the MLIS technology will compete very favorably with advanced gas centrifuge production plants. The principal reason for the projected lower costs for an MLIS plant when compared to gas centrifuge is the significantly lower capital investment required to construct an MLIS facility. The costs for electrical power for the MLIS and gas centrifuge processes are nearly the same.

The capital cost breakdown for a typical MLIS production plant is presented in Table 3. From this cost estimate, it is clear that the process laser systems and gas flow systems each make up about one-third of the total capital cost for an MLIS plant.

Table 3. Typical MLIS production plant capital cost breakdown.

Total new buildings and building modifications	\$174.6M
Total support facilities	77.2
Site preparation	74.9
Plant startup expense	14.0
Special equipment	
Infrared lasers	\$165.4M
Ultraviolet lasers	161.3
Beam transport system	16.6
Photolysis chambers	14.9
Process Feed System	5.5
Product withdrawal system	21.9
Tails withdrawal system	9.4
Gas flow system	240.7
Gas purification	4.5
Process instrumentation	16.2
Data acquisition and control	51.1
Subtotal special equipment	707.5
Total direct capital costs	1048.2
Engineering (15%)	157.2
Contingency (45%)	<u>542.4</u>
Total Plant Capital Cost	\$1747.8M

The remaining one-third is devoted to new buildings, support facilities, and site preparation. The final total plant capital cost is significantly increased by the large 45% contingency that is added to account for the conceptual status of the design.

A breakdown of the estimated operating costs for an MLIS production plant is shown in Table 4.

Table 4. Typical MLIS production plant operating costs.

Plant staff	\$20.6M
Operating and maintenance materials	66.9
Electrical power	30.5
Utilities other than electrical	1.0
Uranium inventory	0.1
Interest on working capital	2.0
Total annual operating costs	\$121.1M

SUMMARY

From the material presented here, it can be seen that the design principles for deployment of the MLIS uranium process have been identified. The construction, testing, and optimization of larger scale lasers and flow systems are the next logical steps for the MLIS process.

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