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LASER ISOTOPE SEPARATION*

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ABSTRACT

A brief background on the history and motivation of laser isotope separation is presented. Methods of molecular laser isotope separation are reviewed, and the Los Alamos process for separation of uranium isotopes as well as the general problems with this approach are covered. A short summary on critical uv cross-section-enhancement results is given and the implications of infrared cross-section dependence on laser fluence is discussed. The laser requirements for the LASL process and the projected economics for this process are presented. The future prospects for molecular laser isotope separation as well as some speculation on general advances in photochemistry are presented.

INTRODUCTION

In 1971 the University of California Los Alamos Scientific Laboratory (LASL) initiated a comprehensive research program aimed at demonstrating that lasers could be used to efficiently separate the isotopes of uranium. Very early in this program the decision was made to concentrate on molecular species and in particular to center the process around the use of UF_6.(1-3) This allows the

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*Work performed under the auspices of the U.S. Department of Energy.

use of the existing UF_6 capital plant and exploits the physical, chemical and spectroscopic simplicity of UF_6 as compared to other compounds. Several excellent technical reviews and discussions(4-10) on laser isotope separation have appeared, and it will be difficult in this paper to add substantially to what has already been published. More general reviews of the subject of laser isotope enrichment and applied photochemistry are also available.(11-13)

To date, the use of lasers has resulted in the enrichment of the isotopes of over 13 elements ranging from hydrogen to uranium. A summary table of the isotopes enriched at LASL is presented as Table I. In two cases presented in Table I, an ir laser was used to generate a physical difference between the absorbing and nonabsorbing isotopic molecules. Following the selective laser step the molecules are subjected to a successive step, which results in the cleavage of a chemical bond within the molecule. As can be seen from the table, a variety of techniques can be used to achieve chemical-bond rupture ranging from a second laser to the use of uv flash lamps. Techniques that utilize single uv and single ir lasers have been demonstrated during the course of the investigations at Los Alamos. The discovery(14-19) that the dissociation of a complex molecule such as SF_6 could be achieved in a selective fashion by the absorption of 35 photons of about 0.1-eV energy has since led to the application of this phenomena to the separation of other elements. The mechanism of collisionless multiple-photon dissociation is still under extensive investigation by the scientific community.(20-23)

The discovery of the multiphoton dissociation process was especially helpful in providing a photochemical method for dissociating SF_6 because the compound has no absorption in the accessible regions of the visible or ultraviolet spectrum. The same is true for SiF_4 and the freons that have also been selectively dissociated by this process. In these cases it has been found that secondary reactions between daughter molecules and initial reactants do not lead to loss of initial selectivity. However, it is often the

TABLE I

LASER ISOTOPE SEPARATION: ELEMENTS ENRICHED AT LASL IN AP DIVISION

COMPOUND IRRADIATED	ISOTOPES ENRICHED	LASERS
H_2CO (IN LIQUID Xe)	H, D	319 nm FREQUENCY-DOUBLE DYE; 325 nm HeCd
BCl_3	^{10}B, ^{11}B	10 μm CO_2 + uv FLASHLAMP
	^{10}B, ^{11}B, ^{35}Cl, ^{37}Cl	10 μm CO_2
CF_2Cl_2	^{12}C, ^{13}C	10 μm CO_2
CS_2	^{12}C, ^{13}C, ^{32}S, ^{34}S	193 nm ArF
CS_2 (IN LIQUID N_2, Ar OR Kr)	^{12}C, ^{13}C	206 nm IODINE RESONANCE LAMP
O_2	^{16}O, ^{17}O, ^{18}O	193 nm ArF
UO_2F_2 (IN LIQUID METHANOL)	^{16}O, ^{18}O	448 nm DYE; 455 nm DYE
SiF_4	^{28}Si, ^{29}Si, ^{30}Si	9 μm CO_2
SF_6, SF_5Cl, S_2F_{10}	^{32}S, ^{33}S, ^{34}S	10 μm CO_2
MoF_6	^{92}Mo, ^{94}Mo, ^{95}Mo, ^{96}Mo ^{97}Mo, ^{98}Mo, ^{100}Mo	9 μm CO_2
UF_6	^{235}U, ^{238}U	16 μm ir + uv

case that processes based on acids do suffer isotopic scrambling because of the lability of acid protons. Tiee and Wittig(24) have employed one- and two-frequency processes to dissociate UF_6.

It is interesting to observe that, even with the laboratory successes such as those presented in Table I, the most significant efforts for commercialization of laser isotope separation for any element other than uranium are efforts on sulfur and carbon being pursued in the USSR.

In this paper we cover only molecular laser isotope separation phenomena. Information on atomic laser isotope separation and on laser-assisted chemistry can be found in the references previously cited.

DISCUSSION OF PROCESSES AND MECHANISMS

Laser processing of molecules can be broadly grouped into the application-oriented categories shown in Fig. 1. The first step in laser isotope separation is selective excitation of one of the isotopic species, and if the exciting photon energy is sufficient,

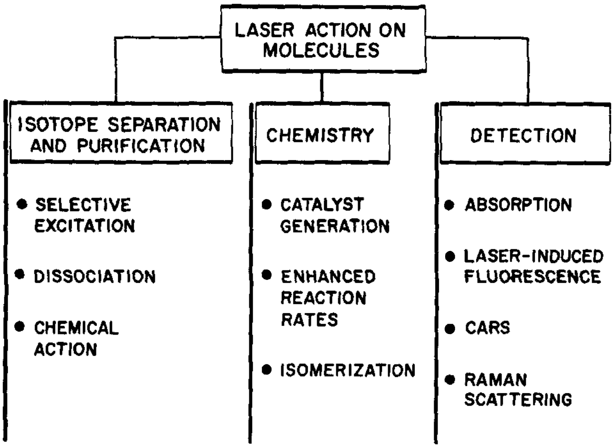


FIGURE 1 General classification of laser action on molecules

this step can lead to dissociation or ionization. The selective excitation is made possible by the difference in vibrational and rotational spectral features of the different isotopic molecules brought about by the different masses. Cold gaseous compounds have the best understood vibrational spectra, and, generally, the best understood kinetics. Volatile compounds of heavy elements are generally polyatomic and, therefore, have low-lying vibrational states that produce an anharmonicity in the progression of levels of all other vibrational modes. These levels also are often highly degenerate so that their population is large and the population of the ground state is much less than half of the total.

Transitions beginning on these many low-lying levels are called hot bands, and they account for considerable broadening of spectral features. These features become a wide virtual continuum for compounds such as UF_6. An experimental technique for achieving dramatic spectral simplification is to mix the material with a high gamma (ratio of specific heats) gas-like helium and then to expand the mixture in a nozzle.(25) D. N. Travis, <u>et al</u>. have recently described results of such experiments for UF_6.(26)

In the attempt to obtain more volatile gases so that they may be cooled at equilibrium by static techniques, many highly complex compounds have been proposed. It should be noted that these compounds have even lower-lying states and more complex spectra than the fully-fluorinated compounds. This added complexity will have to be compensated by dramatically higher vapor pressures at a given temperature to provide a competitive process. If the initial selective excitation step is carried out with low-energy photons, then additional steps are necessary to achieve separation. An additional step that has been successful is molecular dissociation through the addition of a uv photon or through the addition of sufficient ir photons. A second alternative for a second step in laser isotope separation is to take advantage of the increased chemical activity normally associated with the more excited isotopic molecule. To date the most successful methods have been the photodissociative methods with some variance in the degree of initial excitation as shown in Figs. 2 and 3.

In addition to laser interaction selectivity, there are many other problems that must be faced if a practical laser separation scheme is to be developed to commercial status. A set of criteria for a successful separation process is presented below.

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 feedstock density as possible.

Overall Process Efficiency: The net energy expended in separating the desired product molecule from the mixture should be minimized. If we consider a photodissociative process, then the minimum energy required to effect a separa-

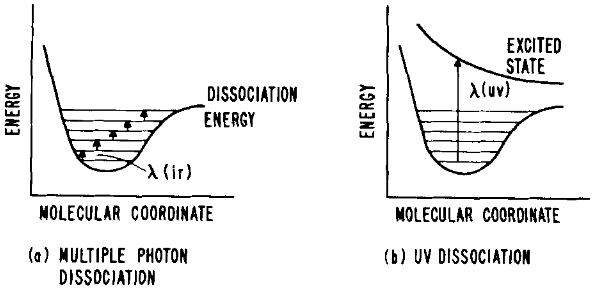
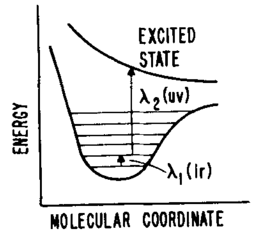
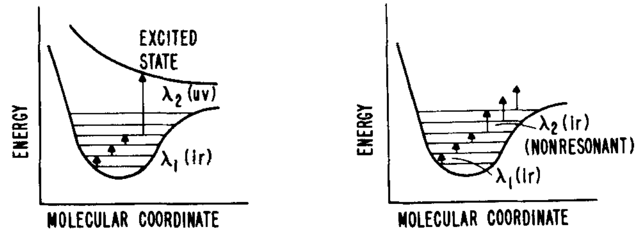


FIGURE 2 Single frequency processes



(c) TWO PHOTON DISSOCIATION



(d) MULTIPLE IR SINGLE UV DISSOCIATION

(e) TWO IR FREQUENCY DISSOCIATION

FIGURE 3 Two frequency processes

tion for UF_6 is the dissociation energy of 3.2 eV per molecule or 309 kJ/mole of $^{235}UF_6$ separated. For gaseous diffusion the energy consumption is about 3 x 10^8 kJ/mole of $^{235}UF_6$ separated, which is more than six-orders-of-magnitude greater than the minimum energy. This leaves a large margin for improvement and is one of the reasons that photolytic methods, which offer the possibility of greatly reduced energy consumption, are so attractive.

A measure of the effectiveness of an isotope separation process is the separation factor α which is defined by the relationship

$$\alpha = \frac{(N_a/N_b)\ \text{Product}}{(N_a/N_b)\ \text{Waste}} = \left(\frac{X_p}{1 - X_p}\right)\left(\frac{1 - X_w}{X_w}\right)$$

where N stands for number density and X stands for mole fraction. A high value of α assures a minimum number of stages for a desired enrichment level.

The complexity and the number of other considerations for a laser isotope separation process are illustrated in Fig. 4. Not shown in Fig. 4 are the considerations of plant design and overall process economics for a laser process. It is apparent that the development of a laser-based isotope separation process is a considerable undertaking requiring extensive basic research, laboratory tests and engineering design, and testing.

THE LASL PROCESS FOR URANIUM ENRICHMENT

Scientists at Los Alamos Scientific Laboratory have been investigating a laser-based process for the separation of ^{235}U from natural material. The process uses natural UF_6 as the feed material, which is dissociated by using one or more ir photons coupled with a uv photon. Spectral simplification and high selectivity are achieved by expanding the UF_6 and a carrier gas through a supersonic nozzle as illustrated schematically in Fig. 5. The

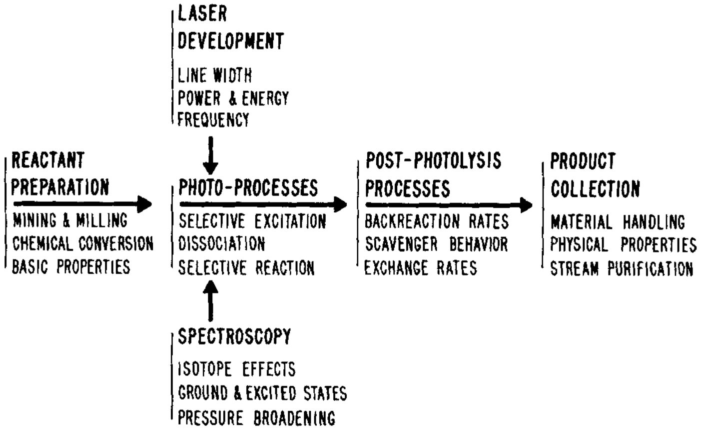


FIGURE 4 Illustration of considerations important to laser isotope separation

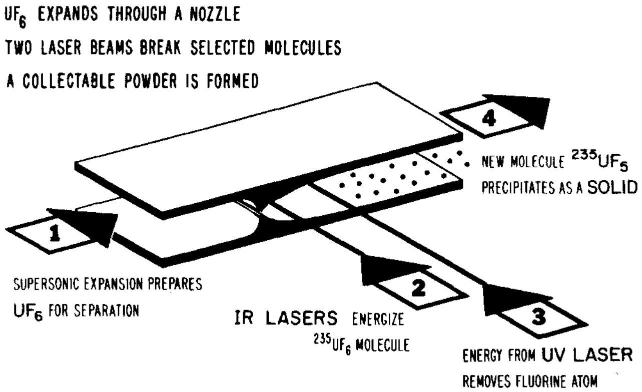


FIGURE 5 Laser isotope separation process

UF_5 formed in the process agglomerates into a fine solid powder, which can be removed from the stream by an appropriate filter.

In the summer of 1976 the LASL process was used to enrich uranium. The feed material, along with a carrier gas, was expanded through a deLaval-type nozzle and then operated on with a 16-μm ir laser and a uv laser. In these experiments milligram quantities of slightly enriched uranium were collected.

The features, which make the LASL process unique and commercially attractive are:

1. Utilization of a feed stock with a well-developed supply, distribution, and handling industry.
2. The enrichment physics are straightforward.
3. Economic studies predict a factor-of-three-to-five reduction in the cost of fuel-grade uranium using the laser process.
4. Predicted energy consumption for the process is about a factor-of-20 less than that for gaseous diffusion.
5. The process is one with a high throughput and low plant inventory.
6. The process is readily scalable to large [9×10^6 SWU (separative work units)/yr] plants and is projected to be cost effective over a wide range of plant sizes.

The decision to use UF_6 for the feed material has resulted in an extensive research effort directed at obtaining ir lasers appropriate for selective excitation of any of the ir-active vibrational modes of this molecule. Because of the high absorption cross section for the ν_3 band of UF_6, much of the laser research effort has concentrated on 16-μm devices. This effort has been very productive and has resulted in tunable as well as fixed-frequency ir lasers that have application in basic research. In addition, the advancement of rare-gas-halide lasers as the second laser for this process has helped to further this technology to the point where 1-joule devices with kilohertz repetition rates will soon be available. Laser development and research continues to be a significant part of the advanced isotope separation program.

PROCESS PHYSICS

There are two central physics issues in the separation of uranium isotopes through selective dissociation of UF_6. The first issue is whether highly-selective vibrational excitation can be achieved through ir irradiation, and second is whether a large increase in uv-photolysis cross section will result from the vibrational excitation.

That isotopically selective vibrational excitation is attainable is indicated by the high-resolution ir spectroscopy of UF_6.(8,26,27) These spectra were obtained by using high-resolution tunable laser diodes along with the technique of gas-dynamic cooling to achieve depopulation of excited states, and consequently, simpler spectra. Selective vibrational excitation turns out to be much more complex than originally conceived because of the existence of multiple-photon excitation.(19,28,29,30) A further complication of the multiple-photon absorption is the fact that the infrared absorption per molecule at the resonant frequency decreases with increasing laser fluence for most transitions.(31-34) The fluence dependence of the ir cross section is shown in Fig. 6 for SF_6.(32)

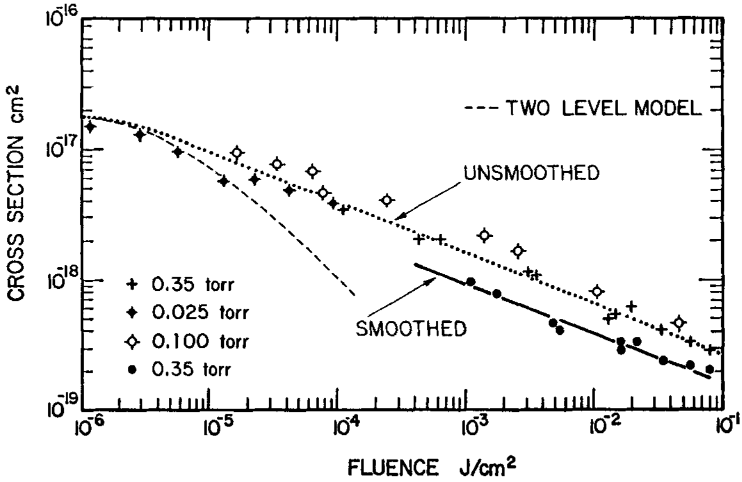


FIGURE 6 Absorption of the P(16) line of the CO_2 laser by SF_6

Also plotted on this figure is the prediction from a simple two-level model of the absorption. The smoothed data is for a single-mode laser, and the unsmoothed data is for a laser that is mode-hopping. As is seen in Ref. 32 and others, the transitions with the largest absorption cross sections show the largest decrease in cross section for a given increase in fluence. Consequently, weak transitions in unwanted molecules that lie under or near strong transitions of desired molecules, tend to become nearly equal to the high cross-section transitions as the laser fluence is increased.

To maintain high selectivity in the vibrational excitation step in the face of the decreasing absorption cross section, it is expedient to use additional infrared frequencies. This process exploits the fact that there is a dramatic enhancement of the ir cross section over a broad frequency range once the molecule has absorbed several ir photons.(24)

Knyazev et al.(35) have recently exploited this technique to demonstrate isotopic selectivity of over 60 for separation of carbon isotopes in CF_3I. Results of their work are presented as Fig. 7. Thus the ir absorption spectrum, even of large polyatomic molecules, is a sensitive, dynamic function of the irradiation field, but still affords the means of highly selective vibrational excitation by proper control of temperature and collision frequency coupled with proper sequencing of frequency- and intensity-controlled lasers.

The second crucial issue in laser isotope separation is the dependence of uv-photolysis cross section on vibrational excitation. The work of Knyazev demonstrates an increase in uv-photolysis cross section of CF_3I with increasing vibrational energy. A similarly convincing example is the observation of a twenty-fold increase in uv absorption cross section of OsO_4 as a result of strong irradiation at 10.6 μm.(36)

More to the point, it has been shown(37) that the uv absorption by UF_6 in the 420 to 550-nm range is increased fifty-fold

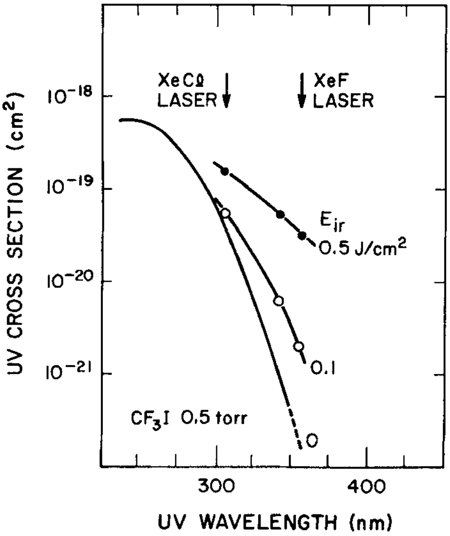


FIGURE 7 Ultraviolet absorption cross section for CF_3I pre-irradiated in the infrared

upon high-power irradiation in the $\nu_2 + \nu_3$ combination band at 8.6 µm. These results are elucidated and qualitatively predicted by the earlier ab initio theoretical treatment of R. T. Pack.(38) Therefore, it is evident that the two basic physics issues are verified by both theory and experiment to be favorable to laser isotope separation of UF_6.

LASERS FOR MOLECULAR LASER ISOTOPE SEPARATION

Due to the need for tunable ir lasers for basic studies in laser photochemistry, as well as for the molecular laser isotope separation process for uranium, several important new ir laser systems have been developed. Among these new laser systems are the HF-pumped optical parametric oscillator,(39) the CO_2-pumped CF_4 laser,(40,41) the CO_2-bending-mode laser(42) and other systems based on optical pumping and Raman scattering from para-hydrogen. Lasers, due to their complexity and the number required, dominate economic considerations for molecular laser isotope separation. To minimize costs and at the same time have reliable and efficient lasers, the laser-based molecular uranium isotope separation program has concentrated on gas lasers for the ir as well as the uv radiation required for this process. Current candidate lasers for the selective ir step are CO_2-pumped devices similar to the CF_4 laser. In the uv, attention is focused on the rare-gas halogen lasers. The most efficient of the rare-gas lasers include XeCl and KrF. Output wavelengths for typical rare-gas-halide lasers are shown in Fig. 8 along with the characteristic energy output per pulse and peak power generated by a typical half-meter device described by Tallman.(43)

In addition to the fundamental-frequency lasers listed above, Raman scattering from H_2, D_2, and CH_4 using these lasers has generated additional uv frequencies that are useful for MLIS studies. Some of the wavelengths generated using this technique are shown in Fig. 9. Full use of Raman scattering from all of the

LASER TYPE	OUTPUT WAVELENGTH (nm)	ENERGY OUTPUT (mJ)	PEAK POWER (MW)
ArF	193	350	12
KrCl	222	45	4.5
KrF	248	720	18
XeBr	282	60	2
XeCl	308	180	5
XeF	354	500	12

FIGURE 8 Primary lasers in the ultraviolet

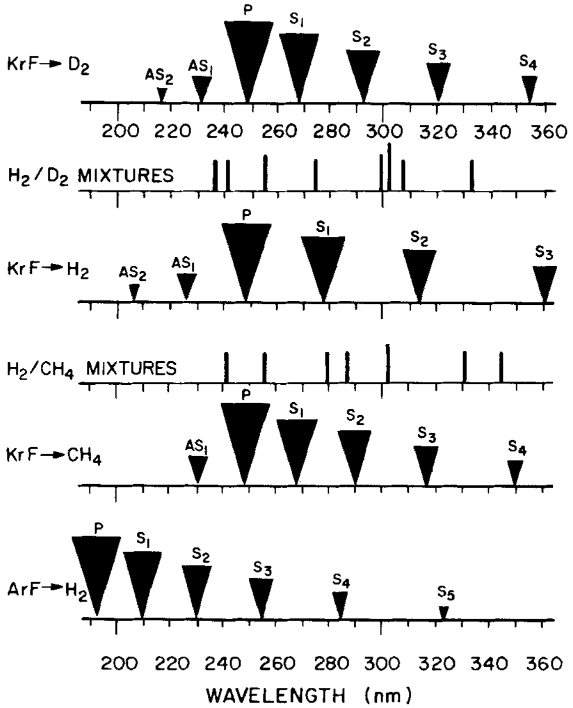


FIGURE 9 New uv wavelengths generated by Raman scattering

primary lasers shown in Fig. 8 provide coverage of virtually all of the useful uv region.

With the recent record of continuing progress in scaling and operation, the major issues in the development of high-average-power and reliable RGH lasers now lies in the optics and in the pulse-power circuit. The pulse power requirements are essentially identical in all the rare gas halide lasers so that research on one is applicable to all. The KrF laser has received the most attention. Several lines of R&D are being pursued and continued progress can be anticipated in the near term.

The characteristics of optical elements vary significantly among the rare-gas halide lasers. Optics will be much more of a limitation on a RGH laser such as ArF at 193 nm than for XeCl or XeF lasers. An unanswered question about RGH laser optics is related to damage thresholds at high average pulse energies (1 to 10 J/pulse) combined with high-pulse rates (0.1 - 5 kHz). At high-pulse rates, gas recirculation and control assumes greater significance. The net laser efficiency at high-pulse rates will be lowered by power requirements for gas recirculation, relative to the net efficiencies obtainable at low-pulse repetition rates.

Laser requirements for laser isotope separation were outlined in Ref. 8, and a more recent update on uv lasers was presented by Rockwood.(44) Typical lasers being developed are 1-J/pulse, 1-kHz, KrF-gas-discharge lasers and 1-J/pulse, 1-kHz, CO_2-pump lasers. The CO_2 laser has the flexibility of line tuning and single-mode-frequency stabilization about line center.

ECONOMICS

The first major application of lasers to chemical processing and separation promises to be in uranium enrichment in support of the nuclear fuel cycle. Most nuclear reactors require a higher concentration of the fissile ^{235}U isotope than is present in naturally occurring uranium ores. The isotopic ratios of ^{235}U and ^{238}U are currently modified in gaseous diffusion enrichment plants.

In these facilities UF_6 is allowed to repeatedly diffuse through barriers to achieve separation. The ideal separation factor for a single separation stage, derived from the ratio of the diffusivities for $^{235}UF_6$ and $^{238}UF_6$, is very small. In practice, many stages are needed to achieve concentrations of ^{235}U in the range of 3-4%, required for light-water moderated and cooled nuclear reactors (LWR's). This leads to large capital-intensive plants, with a high electric-power demand for circulating the gas through all the stages, and with a significant net cost for the enriched product.

Because the spectral features of the two isotopes of uranium as UF_6 are well separated when compared to laser bandwidths, relatively high separation factors are achievable in a laser-based process for enrichment of ^{235}U. The high separation factors will allow enrichment of either natural uranium or diffusion-plant tails to reactor-grade material in a few stages, while simultaneously effectively extracting the fissile ^{235}U from the feedstock.

Current research and development efforts are focused on scaling laser devices to the powers required for industrial scale applications. Gas lasers, such as those discussed in the previous section, have the advantage of being inherently scalable. Carbon-dioxide lasers are among the most highly developed of all lasers and are being used in a number of industrial applications at the present time. These lasers have also established a credible record for operability and reliability. A cost scaling curve for CO_2 lasers is presented at Fig. 10. From Fig. 10 it may be seen that for high-average-power lasers, the net cost of optical power can be substantially reduced relative to that obtainable with small-scale devices. Similar scaling relationships may be expected with other types of gas lasers.

An estimated cost breakdown for possible MLIS uranium plants is presented in Fig. 11. From this figure it may be seen that laser costs constitute a major portion of the capital costs of the facility and that total capital costs account for about one-third of the net cost of separation with the laser-based process.

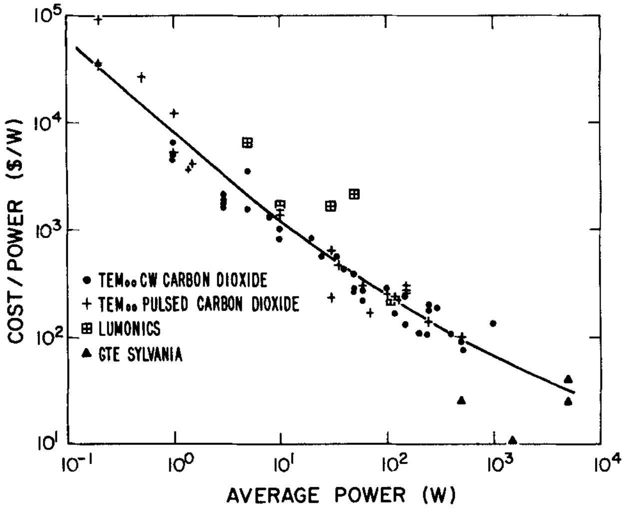


FIGURE 10 Carbon dioxide laser costs from 1978 Laser Focus Buyers Guide

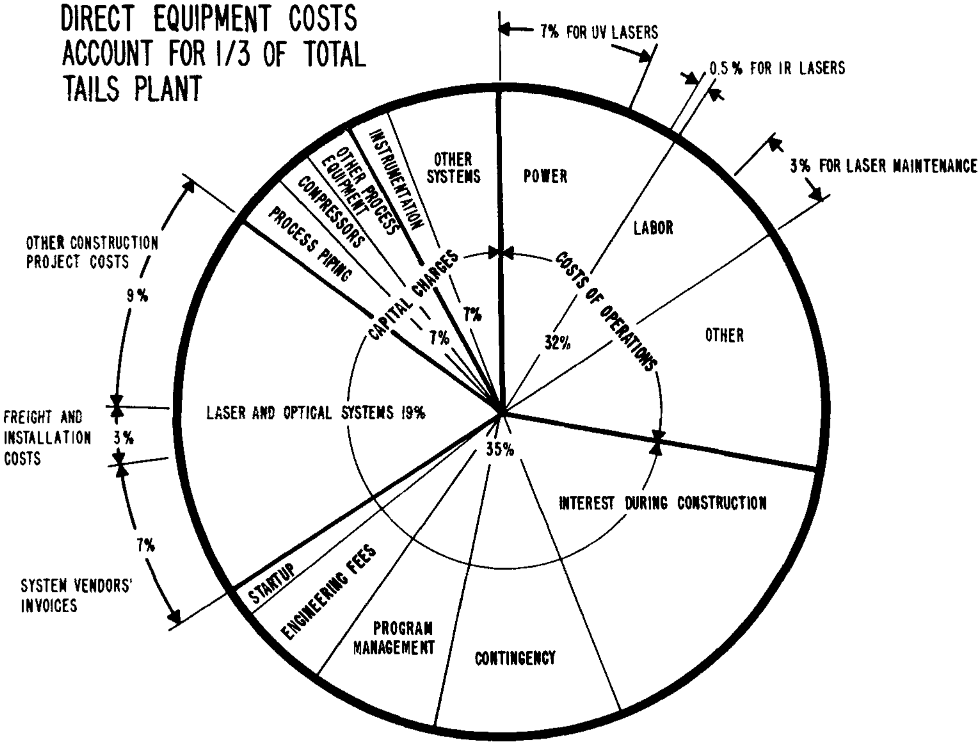


FIGURE 11 Direct equipment costs account for one-third of total tails plant

Current projections, based on several independent evaluations made by LASL, Rockwell International, Union Carbide-Nuclear Division, and Garrett AiResearch, indicate that the laser-based enrichment process should result in significantly lower net separation costs and electrical-power requirements. The projected values for molecular-based laser isotope separation are compared with the estimated costs and power requirements for new diffusion plants in Table II. The direct savings to utilities with MLIS technology working in a supplementary role of tails stripping could be over

TABLE II

COMPARISON OF MOLECULAR LASER ISOTOPE SEPARATION WITH GASEOUS DIFFUSION

	Gaseous Diffusion	Molecular MLIS
Feed Material	UF_6	UF_6
Selectivity (α-1)(a)	2×10^{-3}	1-15
Energy (kWh/SWU)(b)	2600	50-120
Net Separation Cost ($/SWU)	90-140(c)	15-30(d)
Power Costs ($/SWU)	68	∿2
Status	Production	R&D(e)

(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) The costs for LIS are calculated with 10-year amortization rather than the longer terms used for diffusion- and centrifuge-plant evaluations.

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

700 million dollars per year. This conclusion is based on the assumption of \$40/lb yellow cake and an MLIS capacity of 20 million SWU/yr to service the diffusion and centrifuge facilities expected to be in place after 1990. Replacement of the existing diffusion facilities, for which the power costs alone are several times the projected total costs of the laser-based systems, should yield savings of over 900 million dollars annually on an investment of about 2.5 billion dollars. In addition to the reduction in direct cost for electrical-power, MLIS will make possible indirect savings through reduction of mining and milling requirements by operation at a significantly lower tails mole fraction.

INDUSTRIALIZATION

The introduction of lasers into industrial chemical processes has been slow. This is due to the high cost of laser photons, the lack of an adequate data base, and in part to the high sophistication of laser-based technology. A comparison of the relative costs of laser photons is shown in Fig. 12. Based on the projected costs from Fig. 12, which assumes amoritization of the laser capital costs over a ten-year period, it is clear that ir photons cost a few cents per Einstein whereas uv photons range from 50 cents to \$1.50 per Einstein. Current production methods for chemicals result in costs that range from 2¢/mole to \$2.00/mole. Considering the fact that the laser costs would represent only a fraction of the cost of a chemical production plant it is easy to see why the development of laser-based processes has been concentrated on high-cost products such as isotopes. A more complete discussion of possibilities and limitations to the application of lasers in industrial processes is given in Ref. 45.

With the advent and rapid development of rare-gas-halide lasers, new areas of application have been opened. These new areas include purification,(46) detection,(47) and catalyst generation.(48) The silane-purification research described in Ref. 46

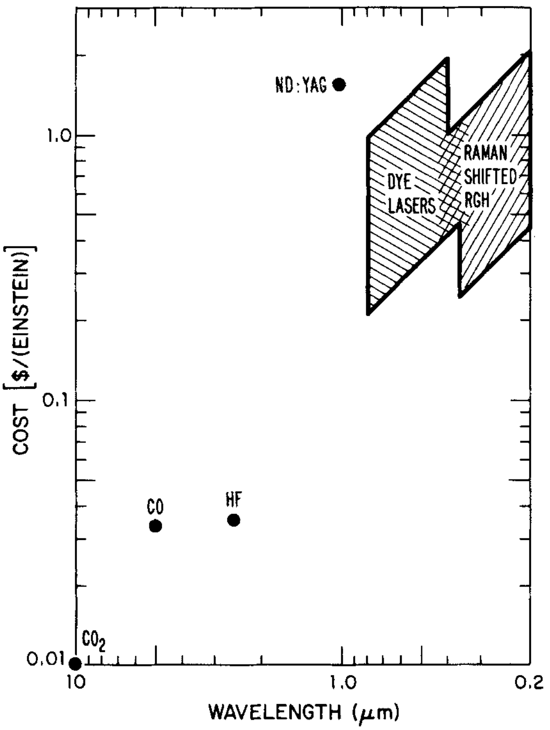


FIGURE 12 Laser photon costs in $/Einstein

has demonstrated that laser processes for the removal of impurity molecules that are present in small quantities in a process stream can be quite competitive. Cost projections for a laser add-on process for the purification of silane, which is the feedstock material for the production of silicon solar cells and silicon wafers for the electronics industry, are in the range of a few cents per kilogram of purified material. Because the DOE 1986 price goal for purified silane is $10/kg the laser add-on would be insignificant costwise, but would yield much higher resistivity material. The application of lasers to other purification problems holds high promise and is currently an area of high interest to the laser community.

Multiphoton-laser processes have thus far found limited practical application outside of the area of isotope separation. However, it is reasonable to predict that other important areas of application will be found for this process and its various derivatives.

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