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LASER ISOTOPE SEPARATION TECHNOLOGY

by

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ABSTRACT

The Molecular Laser Isotope Separation (MLIS) process currently under development at the Los Alamos National Laboratory is discussed as an operative example of the use of lasers for material processing. The MLIS process, which uses infrared and ultraviolet lasers to process uranium hexafluoride (UF_6) resulting in enriched uranium fuel to be used in electrical-power-producing nuclear reactor, is reviewed.

The economics of the MLIS enrichment process is compared with conventional enrichment techniques, and the projected availability of MLIS enrichment capability is related to estimated demands for U. S. enrichment service. The lasers required in the Los Alamos MLIS program are discussed in detail, and their performance and operational characteristics are summarized.

Finally, the timely development of low-cost, highly efficient ultraviolet and infrared lasers is shown to be the critical element controlling the ultimate deployment of MLIS uranium enrichment.

Enriched uranium for use as fuel in nuclear reactors is presently produced in the United States by the gaseous diffusion process. In addition to being very expensive, gaseous-diffusion enrichment of uranium is very energy intensive. The Department of Energy has initiated a study of advanced concepts that could potentially reduce the cost of enriching uranium fuel for electrical-power-generating reactors and lower the energy requirements to achieve the enrichment.

The national program for advanced uranium isotope separation comprises centrifuge and Advanced Isotope Separation (AIS) processes. A centrifuge facility is presently being added to the gaseous diffusion plant at Portsmouth, Ohio; and work is continuing on advanced centrifuge development. Three advanced isotope separation techniques are being studied: an atomic vapor process using lasers, a molecular process using lasers, and a laserless process based upon ion cyclotron resonance. It is planned that, in the next few months, one of the AIS programs will be chosen to advance to the development module (DM) stage where ton quantities of process material and plant equipment will be used in rather exhausting component and overall process tests. The DM program plan calls for operation of the DM in the 1987 time frame and for establishment of an operational production plant in the early 1990s. Although the demand for enrichment is uncertain at this time, an estimate is shown as the shaded band on Fig. 1. Also shown on Fig. 1 is the forecasted capacity of the gaseous diffusion plants and the gaseous centrifuge plants now under construction. In the international market, one must also consider the significant capacity of foreign facilities such as Urenco and Eurodif, which along with Japan and the USSR will have a capacity of over 20 million SWUs per year by 1995. Figure 2 summarizes the cost projections of the various enrichment options.

The Molecular Laser Isotope Separation (MLIS) program has been under development at the Los Alamos National Laboratory since the early seventies and has established a sound technology base for the uranium enrichment process. The program is now moving into the engineering phase and it is possible to determine in detail the cost distributions used in MLIS economic projections.

The basis of the MLIS process is a selective three-color dissociation of UF_6. The first infrared frequency is tuned to optimize isotopic selectivity. The second infrared frequency is chosen to address only those molecules strongly excited by the first laser, and the ultraviolet frequency of 308 nm photodissociates the vibrationally excited molecules. The ultraviolet frequency is chosen as an economic tradeoff of average cross section for the whole gas mix versus selectivity of photolysis for those molecules excited by the infrared.

Because the vibrational levels of UF_6 are highly degenerate and are low lying, they are heavily populated at room temperature and give rise to a large number of hot-band spectral features in addition to the fundamental band, but each participating molecule has its ν_3 quantum number changed by one integer. These hot bands result in a spectral widening that exceeds the magnitude of the spectral isotope shift between the ^{235}U and ^{238}U molecules. The obvious remedy for the room-temperature spectral overlap is to cool the gas to a low temperature. Gas-dynamic cooling is used to achieve spectral simplification. After the photodissociation by the uv laser, the isotopically selected ^{235}U is in the UF_5 molecule, which grows to particles that are readily removed from the UF_6 gas flow by standard techniques. The result is a physical separation of the ^{235}U from the ^{238}U uranium. A tabular comparison of gaseous diffusion and MLIS is given in Table I. The MLIS process is based on the same process gas, UF_6, as gaseous diffusion, thus the engineering development of MLIS can take full advantage of the many years of gaseous diffusion operating experience using UF_6.

Table I also indicates the process engineering considerations that arise from the laser processing of a nozzle-cooled gas.

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. The wavelengths of the ir and uv lasers are determined by ir and uv spectroscopic measurements and are set at values that optimize enrichment (ε) at a product cut (θ_{cut}), determined by the economics of the process. The cut fraction of UF_6 photolyzed for the process is determined by the $^{235}UF_6$ uv absorption cross section (σ) and the fluence (ϕ) of the uv laser. The number density for the process, which is temperature dependent, is optimized for the required α and for the temperature (T_0). The temperature determines the length (L_0) over which the monomer number density 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. The Mach number and the gas composition determine the required flow velocity (v_0), which, combined with the flow length (L_0), determines the laser repetition rate required to irradiate all of the gas flow passing through the irradiation zone. The depth (d_0) of the irradiation zone is governed by the necessity of maintaining a uniform enrichment over the depth, the ir absorption is a function of fluence, thus a unity fluence ratio for the ir laser which is most strongly absorbed, is required. The height (h_0) of the irradiation zone is chosen to establish reasonable beam sizes at the required fluences. The large number of variables involved in the MLIS process make it necessary to perform several iterations to achieve an optimum irradiation zone and plant design.

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

A typical configuration for the ir laser systems is shown in Fig. 3. A master oscillator-power amplifier (MOPA) arrangement is used to achieve the required beam energy. After conversion of the 10-μm CO_2 radiation to 16-μm radiation through the use of the efficient H_2O-CO_2 Raman converter, the beams from eight MOPA chains, each operating at 1.5 kHz, are multiplexed together to achieve a repetition rate of 10 kHz. The requirements indicated in Table II are for a single amplifier stage of MOPA chain shown in Fig. 4.

A typical uv laser system consists of a rare-gas-halide master oscillator and a single power amplifier. To achieve the correct energy in the uv beam, the outputs from individual amplifying units are spatially multiplexed together using a dihedral beam combiner.

A conceptual design of an ir laser amplifier unit is shown in Figs. 4(a) and (b). As shown, the CO_2 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.

A. Temporal Multiplexing

As noted above, the industrialization of the MLIS uranium process requires both ir and uv lasers that operate at kilohertz pulse repetition rates. To obtain the process repetition rate needed to irradiate all of the gas passing through the photolysis zone, the individual laser beams must be temporally combined to form a multiplexed single beam. A preliminary design of a temporal multiplexer, which will also serve as a master timer for laser triggering, has been accomplished. This device consists of a faceted wheel with each mirror 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 drive. The mirror facets are 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, built from commercially available components except for the wheel, operates at a low rotational speed, is compact and simple in design, and is designed to maintain laser beam quality by limiting thermal distortion of the reflective surfaces.

B. Raman Conversion

One of the key elements in the MLIS uranium process is the Raman system that converts the 10-μm output of the CO_2 lasers to the proper 16-μm wavelength. Raman systems are highly efficient, with conversion efficiencies greater than 50% demonstrated. The simplest form of a Raman converter is a cylindrical tube filled with the scattering gas and fitted with end mirrors and beam insertion and extraction optics. A 10-μm pump beam and a 16-μm seed beam are inserted into the cell and multipassed to achieve the desired 16-μm output power. Calculation of the depletion of a CO_2 pump beam and the buildup of the 16-μm output as a function of number of passes through a para-H_2-filled Raman cell at room temperature show that approximately 20 passes are required to fully deplete the pump beam. The beam profile for the CO_2 pump, both in and after depletion in the cell, and the 16-μm output are shown in Fig. 5.

To accommodate the high-average infrared power needed in the MLIS process, the Raman gas must be continuously recirculated and cooled. A large Raman converter that would provide four Raman cells operating simultaneously in a common gas flow has been designed. A similar two-cell system will be tested at Los Alamos in early 1982.

C. Irradiation Chamber Considerations

The MLIS photolysis chamber requires a design that integrates the process physics, engineering, and economics. The design must accommodate the different laser wavelengths (with different UF_6 absorption cross sections for each wavelength) and variations in pulse timing and laser fluences. For a plant, the photolysis chamber 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 from nozzle to nozzle by beam compression using cylindrical optics between stages; optical losses are minimized by using independent reflective optical components for each laser beam.

The design must provide good photon utilization and not introduce diffraction effects. A number of such designs are being studied at Los Alamos. Figure 6 shows the relative photochemical performance in a chamber as a function of clipping loss for a Gaussian profile. Clipping of the Gaussian beam is done to obtain a "quasi" top-hat beam profile.

Figure 6 shows the propagation characteristics of clipped Gaussian beams A side-by-side insertion scheme provides more uniform beam distributions over large areas

Figure 7 shows typical intensities across a center line 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 Finally improved performance is obtainable with the use of super-Gaussian beams of the form $f = f_c \exp[-2(x/w)^N]$. The super-Gaussian beams have a better filling factor than normal Gaussian beams for N as small as 5, 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 multilevel model predict that self-focusing in the process gas of the resonant $1r_1$ beam will not be a problem

Simple, straightforward gas-phase lasers are used in the MLIS process Operational requirements for these lasers, such as frequency stability, phase front distortion, and optical component requirements are minimal Likewise, minimal requirements are imposed on the plant environment by the MLIS lasers, temperature stability, humidity, and other environmental factors are easily obtainable for the MLIS infrared and ultraviolet lasers

There is a strong overlap between MLIS laser requirements and those for several DoD programs As an example, Table III compares MLIS CO_2 laser requirements with those of the DoD laser radar program Likewise, Table IV summarizes the excimer laser requirement overlap for MLIS and the Navy DARPA blue-green submarine communications program The commonality of MLIS and DoD laser requirements will allow the MLIS program to take full advantage of the technology development supported by other governmental agencies

As the MLIS program enters the engineering phase and the AIS selection date approaches, increasing attention is being directed to plant costs The ultimate economics of MLIS uranium enrichment is strongly dependent on the cost of the lasers used in the process Table V itemizes the estimated cost breakdown for an MLIS production plant Table VI summarizes MLIS annual costs and predicts a cost of $26 80 for each separative work unit Laser costs account for approximately 40% of the total direct capital cost of the plant Clearly, since laser costs constitute the single most important cost element of the MLIS plant, the projection of laser costs becomes a dominant factor in determining the economic feasibility of the MLIS process

Table VII presents an estimated cost breakdown for direct capital costs of the ultraviolet and infrared laser systems for an MLIS plant on a cost per watt of laser power basis The corresponding infrared laser costs on a 5 J/pulse basis is $1750 (1) assuming a Raman conversion efficiency of 40% The sensitivity of the cost of enriched material to laser capital costs is shown in Fig 8 Likewise, the sensitivity to laser maintenance costs is almost nonexistent for the first laser and slight for the second ir and uv lasers

The overall efficiency of the MLIS lasers is a strong determinant of the electrical power consumption of an MLIS plant The sensitivity of enrichment costs to laser efficiency is very slight for efficiencies greater than 0 5% An efficiency of 1 2% has been assumed for the XeCl ultraviolet laser system This efficiency compares favorably with the 1% demonstrated by commercially available lasers and the 0 5 to 1 0% demonstrated in low pulse repetition rate XeCl laser systems fabricated at Los Alamos The design goal has been exceeded by industry, where XeCl efficiencies greater than 2% have recently been demonstrated

The CO_2 laser efficiency assumed in MLIS plant designs is 6 7% Continuous-wave CO_2 lasers routinely operate at efficiencies exceeding 10% Low repetition rate pulsed CO_2 lasers have demonstrated approximately 9% efficiency Recent tests on a CO_2 laser system fabricated by United Technologies Research Center, which will be used in an MLIS prototype facility, yielded an intrinsic efficiency in the 9 to 10% range Thus, it is felt that the MLIS design efficiencies for the infrared and ultraviolet laser are achievable within the present laser technology

In summary, the basic physical principle of the MLIS process has been adequately demonstrated in integral enrichment experiments at Los Alamos. The technology required to extend the MLIS process to production plant scales exists today. The projected economics of an MLIS production plant compare very favorably with those of gaseous diffusion and gas centrifuge plants, and substantial electrical energy conservation may be realized by timely integration of the MLIS process into the existing uranium enrichment capacity in the United States.

The key to the ultimate success of MLIS uranium enrichment is the development of low cost, highly efficient ultraviolet and infrared lasers. The industrial laser community is challenged to become involved in an aggressive development program directed toward the production of the required laser systems.

TABLE I

COMPARISON OF MOLECULAR LASER ISOTOPE SEPARATION WITH GASEOUS DIFFUSION

	GASEOUS DIFFUSION	MOLECULAR MLIS
FEED MATERIAL	UF_6	UF_6
SELECTIVITY (α 1)	2×10^{-3}	1 15
ENERGY (kWh/swu)	2600	50 120
NET SEPARATION COST ($/swu)	90 140	15 30
POWER COSTS ($/swu)	88	~ 2
STATUS	PRODUCTION	R & D

TABLE II

MLIS LASER REQUIREMENTS

Parameter	Symbol	Units	Parameter Value: Infrared CO_2 Pump	Parameter Value: Ultraviolet Laser
Wavelength	λ	μm	15 9	0 308
Energy per pulse	E	J	2 - 5	2 - 5
Repetition rate	R	kHz	0 5 - 2	0 5 - 2
Efficiency	η	%	5 - 8	0 8 - 1 5

TABLE III

THERE IS A STRONG MILITARY NEED FOR A CO_2 INDUSTRIAL PROGRAM

MLIS CO_2 LASER	DOE LASER RADAR
• LINE TUNABLE	• [illegible] LINE
• [illegible] PULSE MAXIMUM	• [illegible]
• 1-5 KHz PRF	• 1 KHz PRF
• 10^6-10^{10} SHOT RELIABILITY	• 10^6-10^{10} SHOT RELIABILITY
• STABLE PLANT ENVIRONMENT	• [illegible]
• [illegible] MHz FREQUENCY STABILITY	• [illegible] MHz FREQUENCY STABILITY

TABLE IV

COMPARISON OF DOE/DOE AND MLIS ULTRAVIOLET LASER REQUIREMENTS

	MLIS REQUIREMENT	DARPA + NAVY REQUIREMENT
ENERGY/PULSE	4 J	4 J
ACTIVE MEDIUM	XeCl	XeCl
BEAM QUALITY	~5 × DIFFRACTION LIMITED	~5 × DIFFRACTION LIMITED
LIFETIME	> 10^6 SHOTS	10^9 SHOTS
PULSE RATE	1.25 kHz	100 Hz
EFFICIENCY	1-2%	~
LINEWIDTH	NONE	0.1 Å, 0.01 Å SHOT - SHOT
SPACE QUALIFIED	N	YES

TABLE V

MLIS PRODUCTION PLANT COST ESTIMATE

TOTAL PROCESS BUILDING AND AUX.		$188.3 M
TOTAL SUPPORT FACILITIES		84.5
SITE PREPARATION		20.8
PLANT STARTUP EXPENSE		11.9
SPECIAL EQUIPMENT		
INFRARED LASERS	$176.1 M	
ULTRAVIOLET LASERS	74.5	
NOZZLES	8.6	
COMPRESSORS, DRIVERS, COOLERS	46.5	
PRODUCT COLLECTORS	10.8	
HEAT EXCHANGERS	2.4	
OPTICAL BEAM CONTROL SYSTEM	18.0	
SUBTOTAL		335.9
TOTAL DIRECT CAPITAL COSTS		641.4
ENGINEERING (25%)		160.3
CONTINGENCY (55%)		440.9
INTEREST DURING CONSTRUCTION (18%)		223.7
TOTAL PLANT CAPITAL COSTS		$1466.3 M

TABLE VI

SUMMARY OF ANNUAL MLIS COSTS

• ANNUAL COSTS	
CAPITAL AT 0.11017/YR	$161.54 M
POWER 30.0 mills/kWH	19.56
OTHER OPERATING COST	71.46
TOTAL ANNUAL COSTS	$252.52 M
• UNIT COST OF SEPARATIVE WORK	28.86/swu
• UNIT COST OF PRODUCT	708.99/kgU

TABLE VII

DIRECT CAPITAL COST BREAKDOWN FOR MLIS ULTRAVIOLET AND INFRARED LASERS

LASER SUBSYSTEM	UV COST $/WATT	CO_2 COST $/WATT
POWER SUPPLY	9.24	20.83
MODULATOR	26.67	
PULSE FORMING LINE	2.67	
ELECTRICAL SWITCH	1.56	
GAS CLEAN-UP	2.00	1.50
FLOW SUBSYSTEM	86.56	17.42
PREIONIZER	11.11	
OPTICS	13.33	3.33
INSTRUMENTATION & CONTROLS	27.78	4.25
SYSTEM ASSEMBLY	8.89	14.17
QUALIFICATION TESTING	8.89	
DATA & DOCUMENTATION	1.11	8.25
PACKING & SHIPPING	22.22	8.33
INSTALLATION & ACCEPTANCE TESTING	22.22	3.00
TOTAL	213.25	79.16

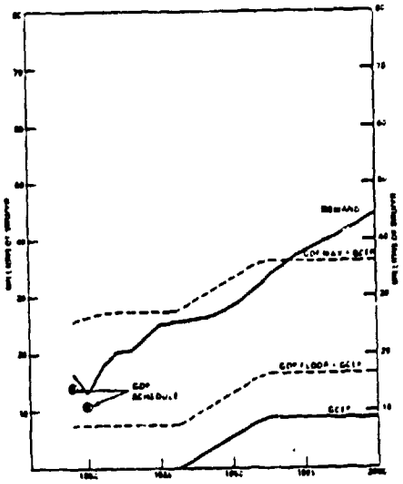

Figure 1. Projected demand for enriched uranium through the year 2000. The United States production capability for Gaseous Diffusion (GDP) and Gas Centrifuge Enrichment (GCEP) is also shown.

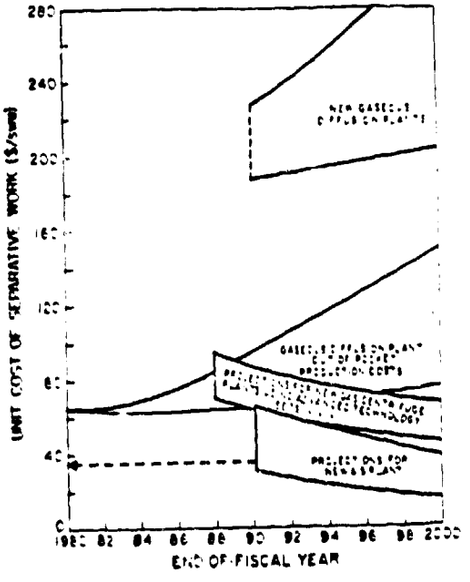

Figure 2. Cost projections for gaseous diffusion, gas centrifuge and advanced isotope separation uranium enrichment processes.

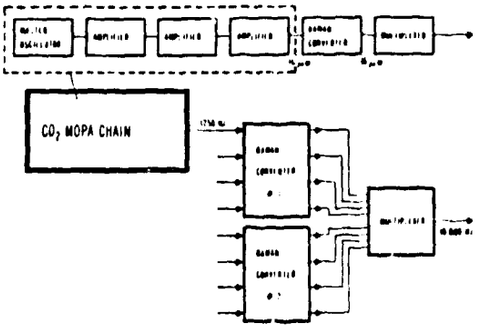

Figure 3. Schematic layout of the infrared laser system for an MLIS plant.

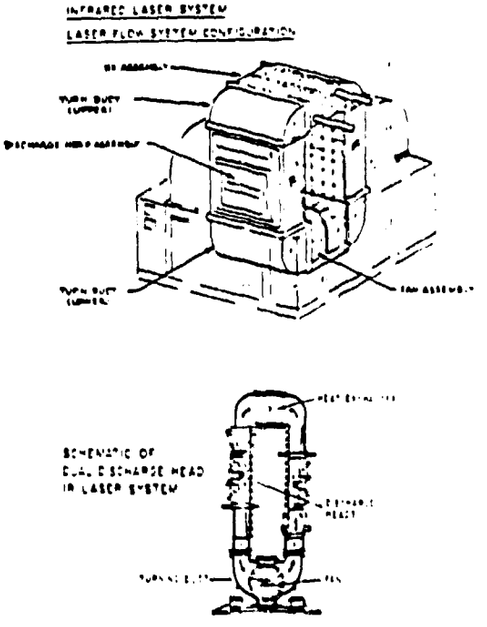

Figure 4. Conceptual design of CO_2 laser amplifier. (a) Isometric view showing the laser flow system configuration. (b) Cross-section view showing dual discharge heat configuration.

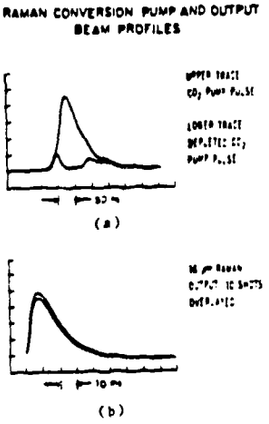

Figure 5. Oscilloscope traces depicting Raman conversion. (a) Input CO_2 pump pulse (upper) and depleted CO_2 pulse after Raman conversion (lower). (b) Outlay of ten 16-μm output pulses. Note: Temporal pulsewidth limited by the optical detector used.

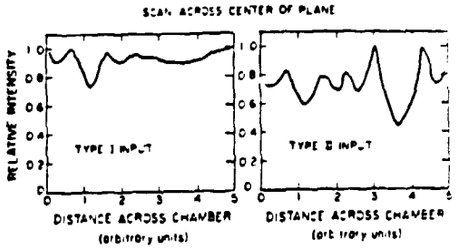

Figure 7. Typical intensity profiles across the photolysis chamber centerline.

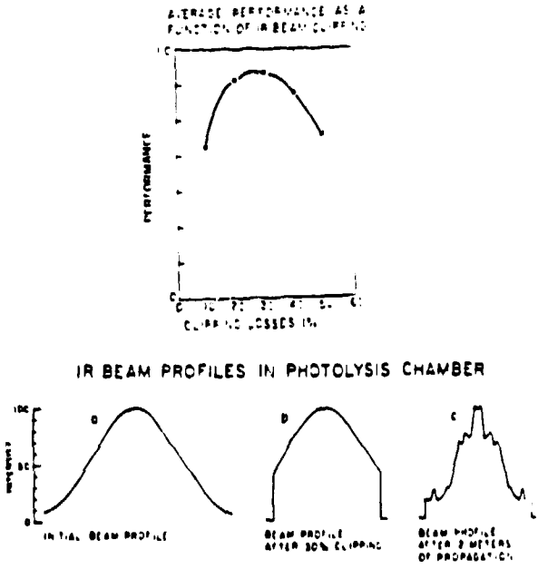

Figure 6. Infrared beam clipping. (a) Performance as a function of clipping losses. Approximately 25% clipping is optimum. (b) Infrared beam profiles at insertion and after 2 m of propagation.

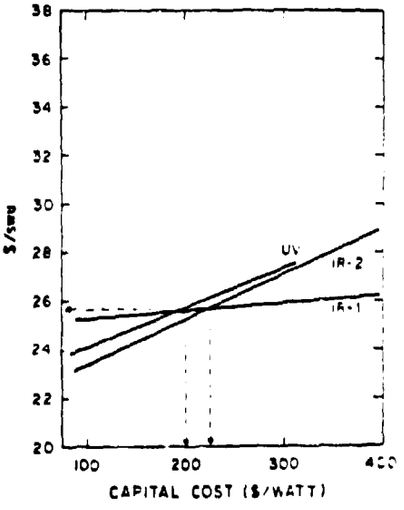

Figure 8. Enrichment cost sensitivity to laser capital costs.