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Science, Technology, and the Industrialization of Laser-Driven Processes

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Introduction

About 12 years ago, the laser program at Lawrence Livermore National Laboratory (LLNL) was given the charter to examine the potential use of lasers in industry for materials processing: in particular, to investigate those processes that could exploit the unique spectral, temporal, and spatial properties of laser light. One of the constraints implicit in this exercise was to focus on processes that were economically viable, i.e., ones that could generate support for the research and development of the laser and electro-optics technology. Our initial estimate was that it would take an investment of several hundred million dollars to accomplish this mission--to provide a reliable industrially hardened laser technology base.

Candidate Elements

Members of the laser program at LLNL reviewed potential applications of lasers in industry, some of which are listed in Fig. 1. Many of these areas are currently under active study in the community. We quickly focused on laser isotope separation of atomic uranium because of the large demand (>1000 tonnes/year) and high product enrichment price (>$600/kg of product) for material used as fuel in commercial light-water nuclear power reactors.

We also believed that once the technology was fully developed and deployed, it could be applied directly to separating many elements economically on an industrial scale. Figure 2 shows the elements whose electronic transitions are accessible using the fundamental and harmonics of the dye laser system under development at LLNL.

Some of the candidate elements are shown in Fig. 3. The Atomic Vapor Laser Isotope Separation (AVLIS) program at LLNL has an extensive uranium and plutonium program of >$100 M in FY85 and a minor research program for other elements.

Application	Economic Potential
• Isotope separation	• Demands today a high value product ≳$10/kg
• Cleanup of radioactive waste	• Extremely high payoff if, and only if, a fully integrated reprocessing cycle is realized, including military applications.
• Trace impurity removal	• Cannot alone support technology development but will be major spin off
• Selective chemical reactions	• Demands net gain in reaction (no. particles/no. photons) >> 1
• Photochemical activation or dissociation of gases	• Potential high leverage in many applications (e.g., reactive etching, coatings)
• Control of combustion particulates	• Will probably happen slowly as a spin off
• Crystal and powder chemistry	• Unique and interesting
• Laser induced biochemistry	• Inevitable but a decade at least before some clarification

Fig. 1 Some potential applications of lasers in industry.

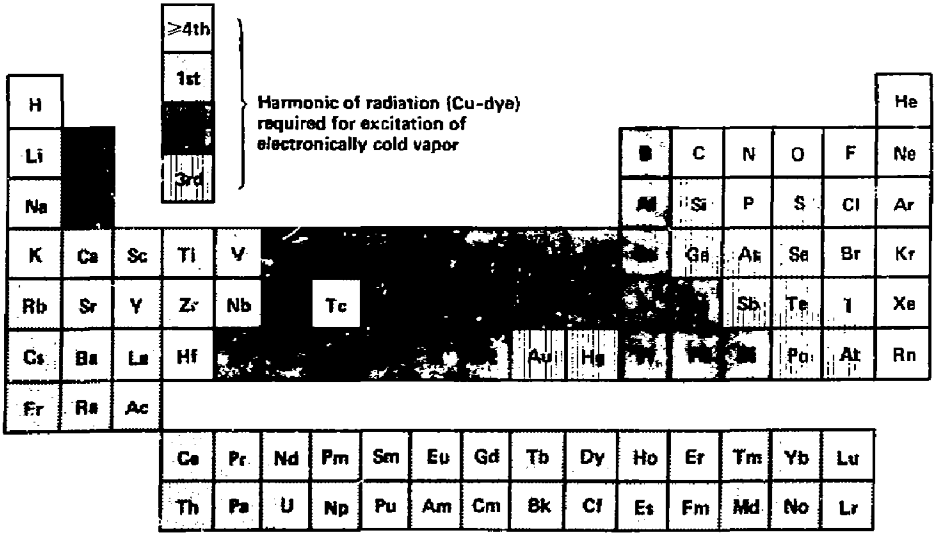


Fig. 2 Elements separable by AVLIS using copper-vapor (Cu)/dye laser technology.

Separative Work Units

Figures 4 and 5 focus on the figures of merit for laser-driven processes. Here S is the entropy of mixing ΔS expressed in terms of the constituent mole fractions X_a, X_b, . . . and normalized by the product of Boltzmann's constant k and the total number of particles N. The terminology used derives from the uranium-enrichment industry but is universally applicable to any chemical process. The productivity of a production module is given in terms of separative work units (SWUs). A thermodynamicist would recognize that separative work is just the difference between the Gibbs free energy of the product plus waste (tails) material, and the feed material. The relationships are fundamental and express the amount of work that must be employed to overcome the entropy of mixing or to alter the chemical potentials of the components of the system. Actually the SWU is normalized in mass units. Value is given to both the product and tails material. The importance of this will be seen later.

Figure 5 introduces a simple economics model for laser-driven processes. The numerator contains the materials-handling cost and laser-related costs per kg of feed. The costs are derived primarily from engineering considerations. Laser energy is expensive, three to four orders of magnitude more expensive than energy derived directly from oil or electricity. To establish the cost of the process, given in terms of $/SWU, the numerator must be divided by the intrinsic separative performance of the process, given in dimensionless units of SWU/kg feed. It is important to realize that separative performance is solely dependent on the process physics and plays an obvious role of process cost multiplier. Laser-related costs can be estimated as shown and depend sensitively on the photoselectivity of the process. Using the analysis for uranium AVLIS, the total laser-related processing costs can be less than $10/kg feed.

As summarized in Fig. 6, separative performance depends on the assays of both the product and tails material. Consequently, high SWU/kg feed can be achieved by depleting tails. Thus high selectivity is not essential for high separative work. However, as seen from Fig. 5, high photoselectivity is essential to achieve low MJ/kg feed and low $/SWU. All else being equal,

Element	Application	Demand (MT/y)
Uranium	Low-cost fuel for light water reactors	>1000
Plutonium	Low-cost flexible purification for defense applications	>1
Samarium Europium Gadolinium, etc.	Burnable poison for power reactors	>1
Mercury	More efficient fluorescent lamps	>1
Zirconium Titanium	Cladding for nuclear fuel elements	>1
Rhodium Palladium Platinum	Precious metal recovery from nuclear waste	>1

Fig. 3 Candidate elements for processing using atomic vapor laser isotope separation.

- **The entropy of mixing in an ideal mixture**

$$s = X_a \ln X_a + X_b \ln X_b + \cdots, \quad (s = \frac{-\Delta S}{kN})$$

- **The "value" of a mixture is given by the function, ψ:**

$$\psi = X_a \frac{\partial s}{\partial X_a} + X_b \frac{\partial s}{\partial X_b} + \cdots$$

- **The net "value" of a separation process is given by, Ψ:**

$$W_f \Psi = W_p \psi_p + W_t \psi_t - W_f \psi_f, \text{ (W in kilograms)}$$

- **This nomenclature has led to the following terminology:**

$$W_f \Psi \triangleq \text{SWUs, separative work units}$$

$$\Psi = \frac{\text{SWU}}{W_f}, \text{ dimensionless measure of process work}$$

Fig. 4 Definition of separative work in discrete processes.

Process engineering costs

Material handling subsystems

Laser electro-optic subsystems

Process performance

Laser vapor interaction

Separation physics (β_1, β_2)

$$\$/SWU = \frac{\overbrace{(\$/kg_F)_M}^{\text{Material handling subsystems}} + \overbrace{(\$/MJ)_L}^{\text{Laser electro-optic subsystems}}\,\underbrace{(MJ/kg_F)}_{\text{Laser vapor interaction}}}{\underbrace{SWU/kg_F}_{\text{Separation physics }(\beta_1,\beta_2)}}$$

- $(\$/MJ)_L \sim \$1 - 100$
- $(MJ/kg_F) \sim \underbrace{\frac{1}{\epsilon}}_{\text{Utilization}} \times \underbrace{\frac{h\nu(eV)}{particle}}_{\text{Photon energy}} \times 1.6 \times 10^{-16}\,\frac{MJ}{eV} \times 6 \times 10^{23}\,\frac{particles}{mole} \times \frac{\overbrace{X_F}^{\text{Mole fraction of excited material}}\ moles}{\underbrace{M(kg)}_{\text{Mass of 1 mole feed}}} \times \left[1 + \underbrace{\frac{1}{S}}_{\text{Photo-selectivity}}\,\frac{(1 - X_F)}{X_F}\right]$

$$(MJ/kg_F) \sim 10 \times X_F \times \left[1 + \frac{1}{S}\,\frac{(1 - X_F)}{X_F}\right] \sim 10^{-1} \text{ for uranium AVLIS}$$

Fig. 5 Elementary economics model.

- **High selectivity is not essential to achieve high SWU/kg_F**
- **High selectivity is essential to achieve low MJ/kg_F and low \$/SWU**
- **AVLIS process has very high process photo-selectivity**

Fig. 6 Photo-selectivity for a laser-driven separation process.

a process that has low feed assay X_F and low selectivity S will have a laser cost $1/(S \cdot X_F)$ times higher than a process with high photoselectivity. As an example, a uranium LIS process with a photoselectivity of 2 will require 70 times higher MJ/kg feed and have a concomitantly higher laser-related cost of >$100/kg feed. This is one of the reasons we chose the AVLIS process since it can achieve extremely high process photoselectivity, as shown in Fig. 7.

Spectroscopic Selectivity

The approach being used at LLNL is a three-step photoionization process that exploits the large isotope shifts in the electronic spectrum of atomic uranium. These isotope shifts derive primarily from nuclear volume effects. Since the ionization limit is about 6 eV, a three-step process involves lasers operating at about 2 eV or 6000 Å. Consequently, we use pulsed dye lasers pumped by copper-vapor lasers in the process. Very high photoselectivity is attained on each step. The net photoselectivity in the process is extremely high as indicated. The photoionization process can be described as an optical distillation column where work is done only on the isotope or specie of interest.

Major Systems

The major systems that comprise a module in the AVLIS process are shown in Fig. 8. Uranium is vaporized by a high-voltage electron beam from a crucible containing the metal. The uranium in the melt is heated to temperatures in excess of 3500 K. The vapor-pressure gradients established above the melt are sufficient to cause aerodynamic expansion of the vapor flow. The direct flow is then irradiated transversely by the photoionizing lasers. The pulse-repetition rate of the laser is sufficiently high to ensure that all the vapor is irradiated. The copper-vapor lasers are configured in sets of master oscillator/power amplifier (MOPA) chains, each operating at about 5 kHz. The dye lasers are also configured in MOPA chains. The ^{235}U photoions are extracted by a combination of

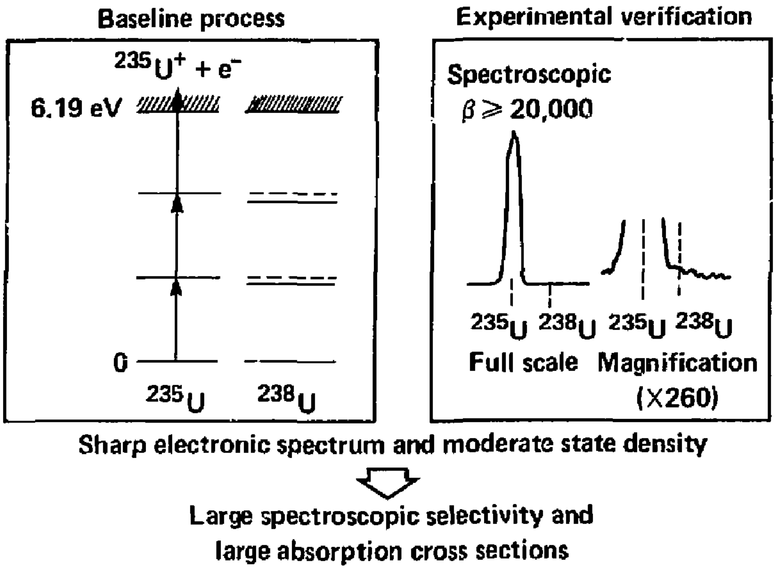


Fig. 7 Spectroscopic selectivity - AVLIS.

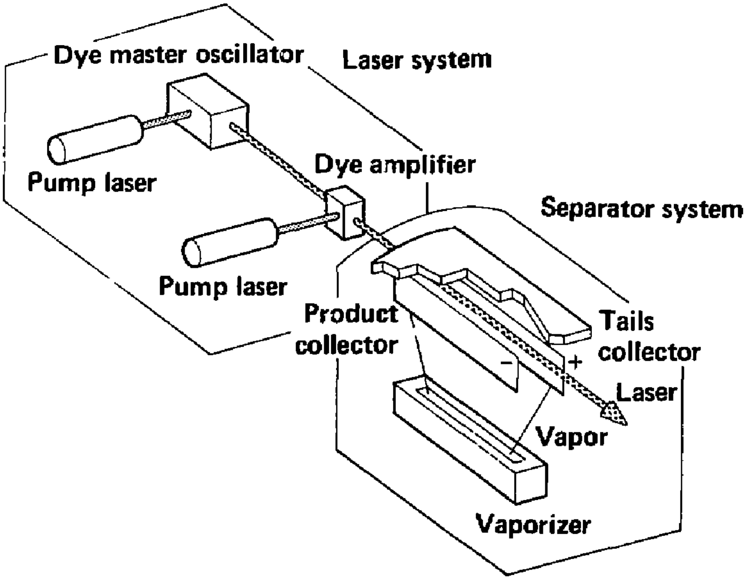


Fig. 8 Atomic vapor laser isotope separation - major systems.

electric and magnetic fields and collected on product plates. The remaining vapor depleted in ^{235}U flows to the tails collector. The assay of the tails material depends on the fraction of ^{235}U ionized and collected. The assay of the product depends not only on this quantity but also on the fraction of the feed material that deposits nonselectively on the product plates. A major feature of the AVLIS process is that both product and tails material are collected as liquids and are allowed to flow to collection pots. This aspect of the design has been developed by Martin Marietta Energy Systems in Oak Ridge, TN, who are an integral part of the AVLIS program.

It takes about 1 SWU/kg feed to achieve the enrichment needed to obtain material for light-water reactors (0.7% feed to 3.2% product) as shown in Fig. 9. The exact separative performance depends on the tails assay considered and is pretty much independent of product assay. The tails assay is a design specification depending on the relative costs for separative work ($/SWU) and feed costs. The gaseous diffusion plants operate at a tails assay to make enrichment costs ($600/kg product) about equal to feed costs ($600/kg product). The AVLIS process is capable of achieving high separative work (1 SWU/kg feed), the required product assay, and very low tails assay in very few stages. Consequently, the laser-related separative work costs can be less than $20/SWU, or less than $100/kg product and the feed costs can be significantly reduced.

Facilities

It has taken about 10 years for the AVLIS program to reach its current state of maturity. Figure 10 summarizes the uranium AVLIS program. We are presently in a production-scale systems-integration stage, having completed process science studies and developed and tested the laser and separator subsystems in stand-alone as well as in fully integrated enrichment operations.

Figure 11 illustrates the layout of the full-scale demonstration facility that has just been activated at LLNL. The building houses a uranium separator module called the separator demonstration facility. The

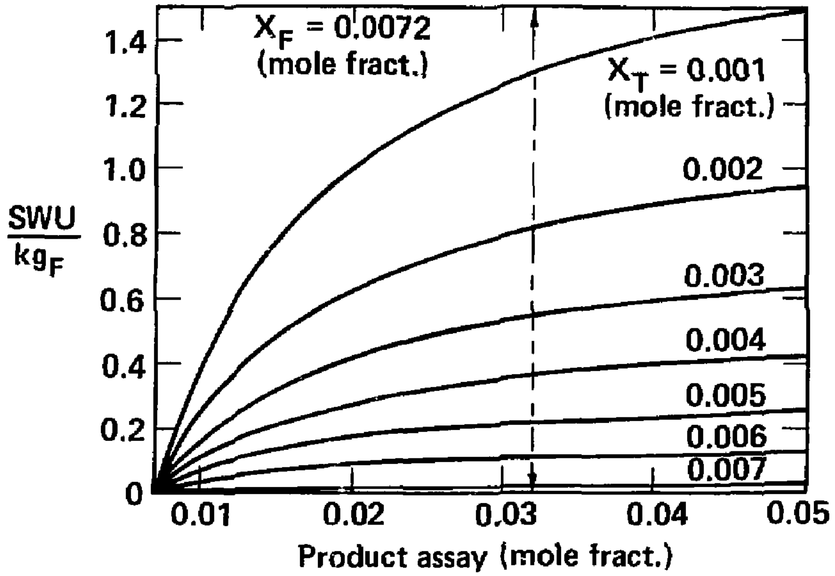


Fig. 9 Separative performance map for uranium enrichment.

- Process science (1974-1980)
  - Functional systems integration (1978-1985)
    - Production systems integration (1984-1990)
      - Economic production operation in 1990s

Fig. 10 Uranium AVLIS program - summary.

building also contains a laser system called the laser demonstration facility that will provide the laser power for the module. The balance of the building contains instrumentation and control systems and refurbishment facilities in support of the laser and separator systems.

Figure 12 is an aerial photograph of the facility taken a few months ago. The octagonal structures on the left are the office buildings that house the program scientists, engineers, and administration staff at LLNL.

Figure 13 is a photograph of the first completed corridor of copper-vapor MOPA chains installed in the facility this April. There are 6 MOPA chains containing 30 laser heads with a total output capability of several thousand watts.

Figure 14 shows the separator module in the full-scale demonstration facility. The tanks at the ends house the module optics for directing the laser beams through the uranium vapor. The module is essentially plant size and has a projected production rate of about 1×10^6 SWU/year, or 2×10^5 kg product/year.

For the past several years, we have transported the output of the laser subsystems over long distances (~1 km) to separator subsystems located in other buildings. Figure 15 shows an aerial view outlining the path of this optical-transport system. The distance between the laser demonstration facility and our special-materials testing facility is about 2 km, while that to our half-scale uranium separator is over 1 km.

Using the initial increment of light available from our new laser facility, we have recently conducted integrated enrichment demonstrations in our half-scale facility (Fig. 16). This facility has been operational for several years.

Integrated Process Model

Returning to some of the concepts introduced in the beginning of this report, we have proceeded to the present generation of systems after intensive study of the interplay of the physics and engineering that govern the AVLIS process. Figure 17 shows schematically some of these areas and how they influence process design and cost using the simple economics

Fig. 11 Production-scale AVLIS demonstration system.

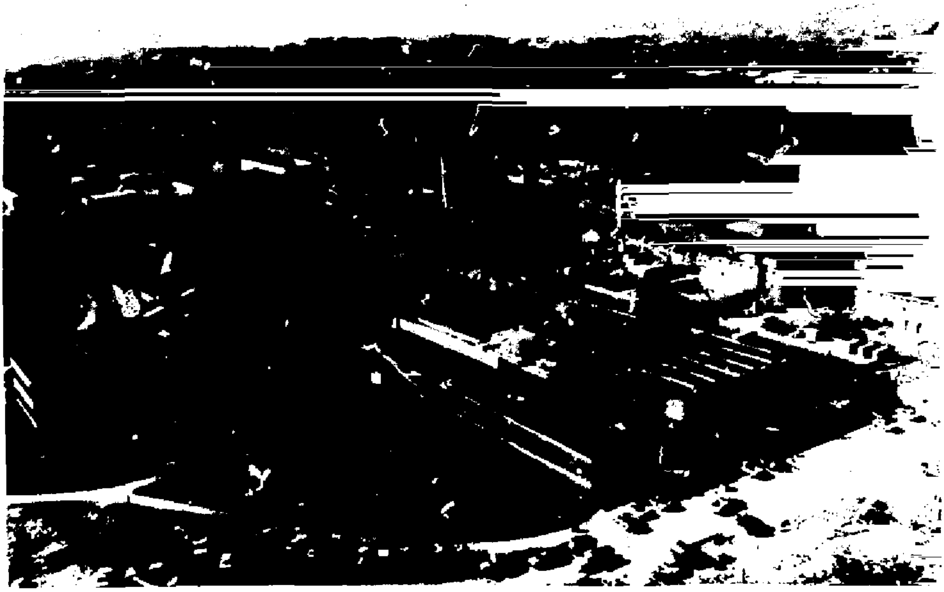


Fig. 12 Full-scale demonstration facility at LLNL (Feb. 1985).

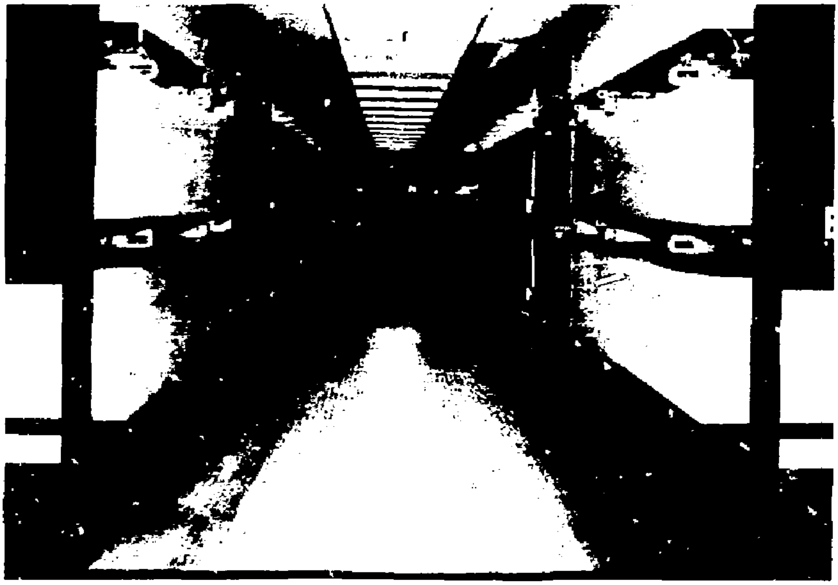


Fig. 13 Full-scale demonstration facility copper-vapor laser system (April 1985).

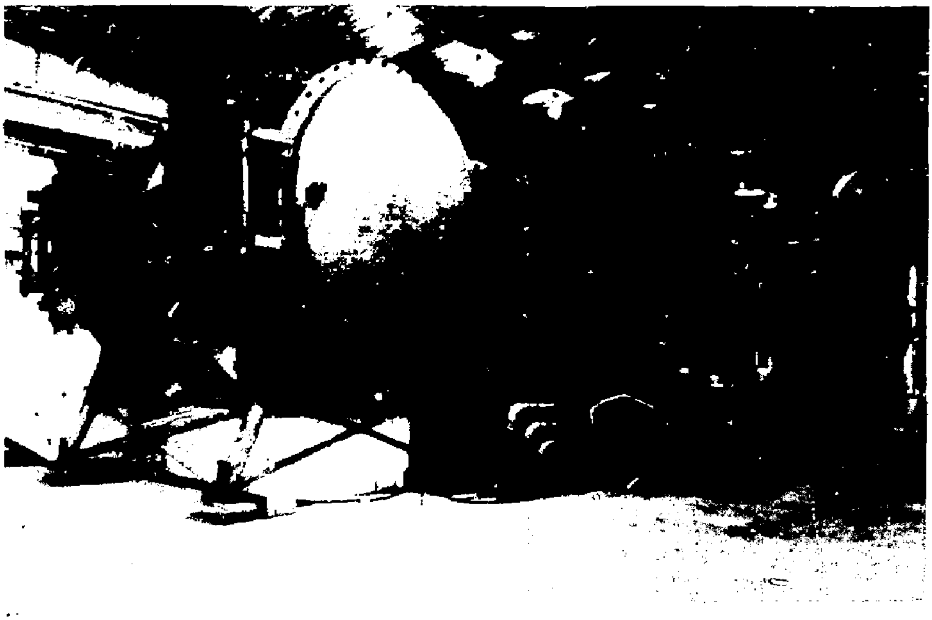


Fig. 14 Full-scale demonstration facility separator module (April 1985).

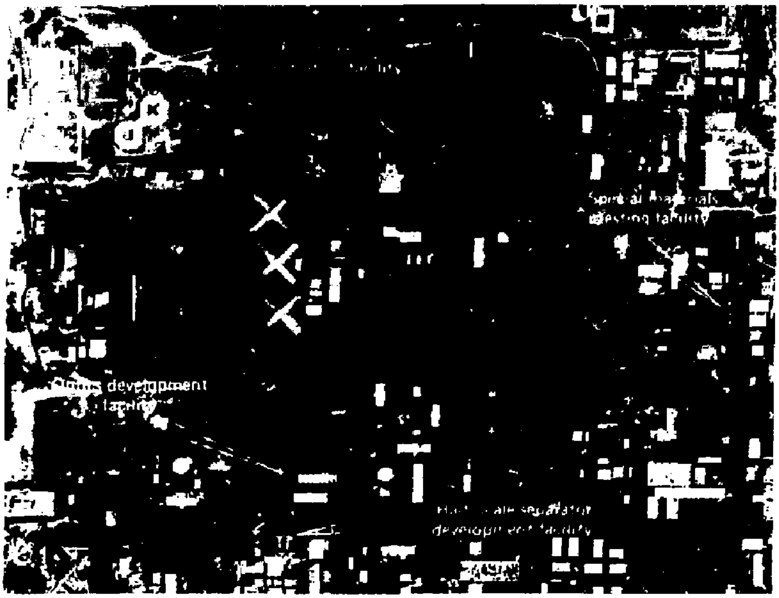


Fig. 15 Beam tube for optical transport between facilities at LLNL.

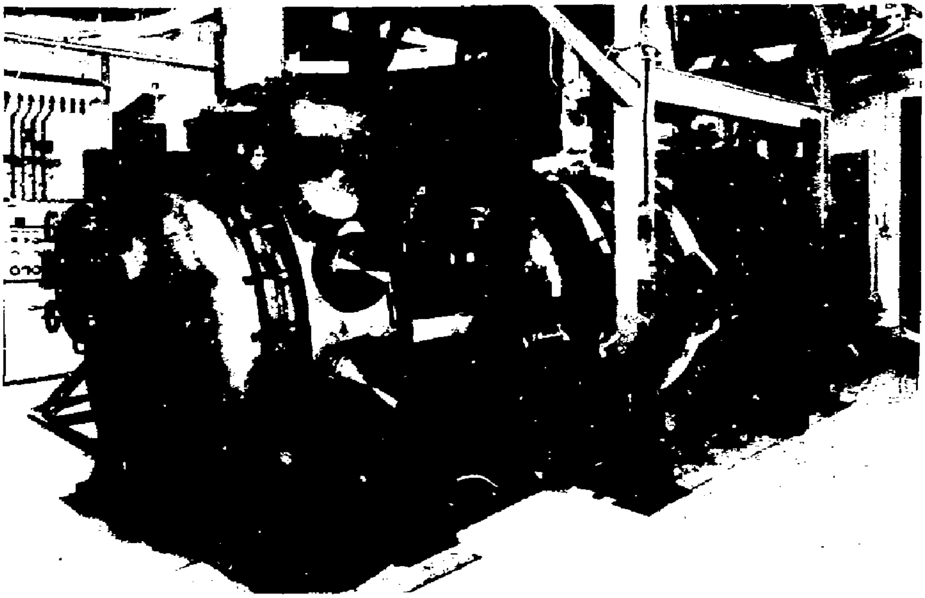


Fig. 16 Half-scale separator at LLNL.

construction shown earlier. Each physics area has been modeled, in some cases from first principles, and benchmarked against results obtained in the laboratory or in systems such as the half-scale separator.

Figure 18 shows an example of the detailed modeling that we do in the area of photoionization and propagation physics. One of the novel features of the process and of the laser system is that we can specify and control the spectral content of the tunable dye lasers to optimize overall photoionization performance.

We have incorporated these physics models along with engineering models in an integrated process model (Fig. 19). The engineering cost models include results from detailed bottom-up costing studies and data obtained from procurements for the full-scale demonstration facility. We also include in the process model operational parameters based on reliability, availability, and maintainability of AVLIS subsystems. Essentially, the integrated process model contains all the fine detail of the AVLIS process and allows us to examine the sensitivity of cost and performance to variation in engineering, design, and physics parameters. We use the code to guide process design. The code is also used to quantify the impact of uncertainties in process parameters, which range from the values for the optical-transition cross sections to the cost of labor. The code was originally constructed by the staff at Martin Marietta Energy Systems to perform multivariable sensitivity studies.

Multivariable Sensitivity Studies

There are literally hundreds of parameters that affect the cost of the process. Each one of these parameters has an associated uncertainty and uncertainty distribution (Fig. 20). The code is capable of using these distributions in a Monte Carlo calculation of performance and of comparing it to the performance using our base-case design values. In the example shown in Fig. 20, the parameter of interest may be the charge-exchange cross section. As our data base improves, the uncertainty associated with each of these parameters becomes smaller.

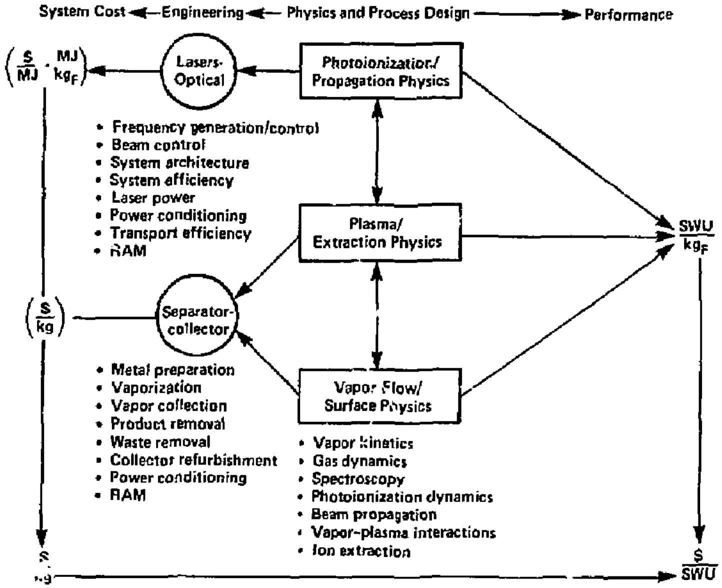


Fig. 17 AVLIS process morphology/structure of integrated process model (IPM).

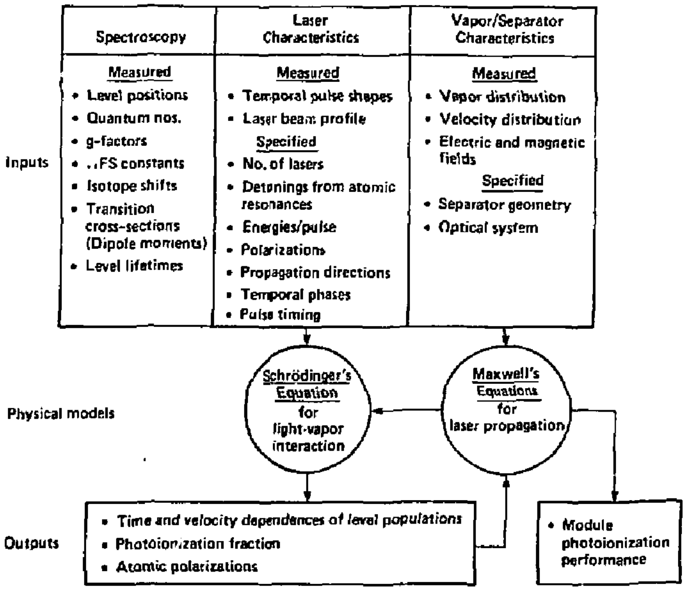


Fig. 18 Modeling of laser - atomic vapor interaction.

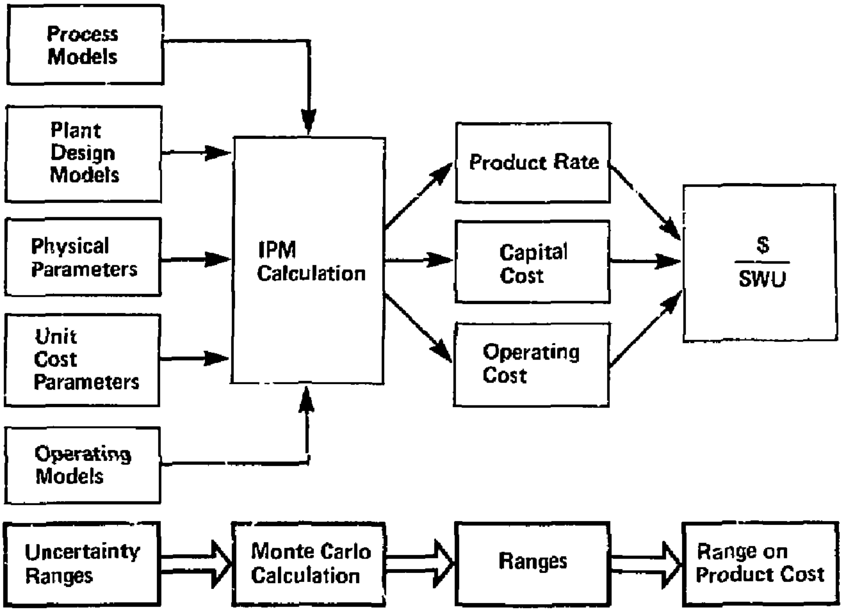


Fig. 19 Integrated process model and multivariable sensitivity studies.

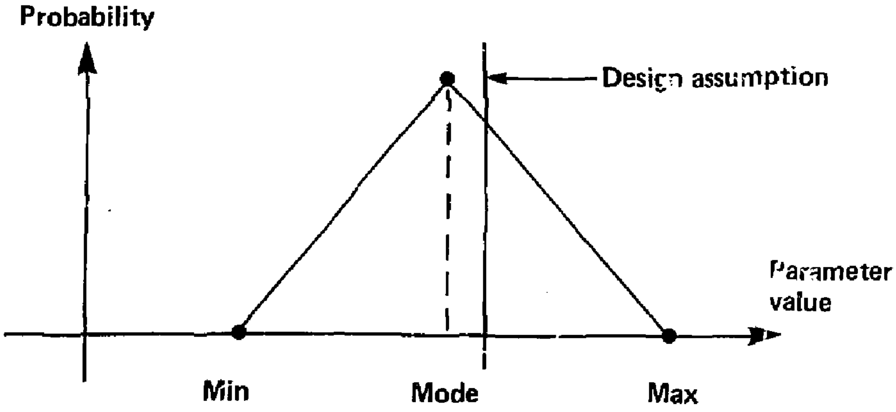


Fig. 20 Generic MVSS parameter description.

Figure 21 is an MVSS histogram for process separative work cost for the 1981 AVLIS engineering design.

World Market

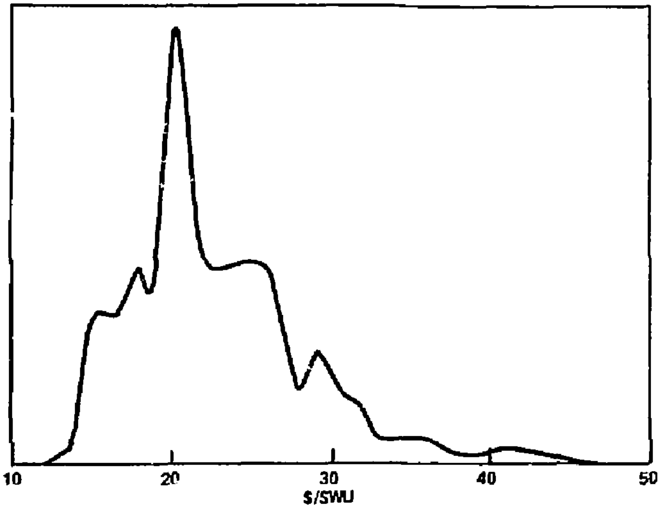
Up to this point we have emphasized process economics. To industrialize any process one must also be cognizant of market economics. Figure 22 summarizes the present market conditions for enriched uranium: prices are expected to drop. This results in part from world enrichment capability presently exceeding demand and the presence of a world inventory of several years' demand. A new technology for the U.S. enrichment enterprise is only attractive if its total cost is below the operating costs of existing capacity.

U.S. Enrichment Enterprise

Presently, there are two advanced technologies in the U.S. being considered for potential production: the Advanced Gas Centrifuge (AGC) and AVLIS (Fig. 23). Both of these programs are funded out of net revenues from sales of enriched material from the gaseous diffusion plant (GDPs). Although the profits from the GDPs are in the hundreds of millions of dollars, in order to ensure that the U.S. remains competitive in the world market in the future and achieves its target prices, the Department of Energy accelerated the selection date for an advanced technology to 1985. During the past year, the AVLIS program has been under intense technical scrutiny by our peers in the AGC program and by a process evaluation board. The board reports directly to the Secretary of Energy whose decision is imminent.

Summary

As a final note, dozens of scientists, engineers, and supporting staff have committed over a decade of their lives in trying to bring this technology to industrial scale. If AVLIS is chosen, we hope the rest of the laser and electro-optics community will share in the excitement and challenge this will offer for the future of laser-driven processes.



SEPARATIVE WORK COST ($/SWU)	
*MINIMUM	16
MEDIAN	22
*MAXIMUM	33
MODE	20

*MINIMUM AND MAXIMUM REFER TO THE MIDDLE 90 PERCENT INFER

Fig. 21 AVLIS MVSS results: distribution of projected separative work cost ~9 million SWU/y (1981 design, 1982 dollars).

- Prices expected to drop
  - World separative work capacity presently exceeds demand
  - World inventory of several years demand exists
- DOE has set target prices that will keep the enterprise competitive
  - Replacement capacity with new enrichment technology is attractive if its total cost is below the operating costs of existing capacity

Fig. 22 World market for uranium enrichment.

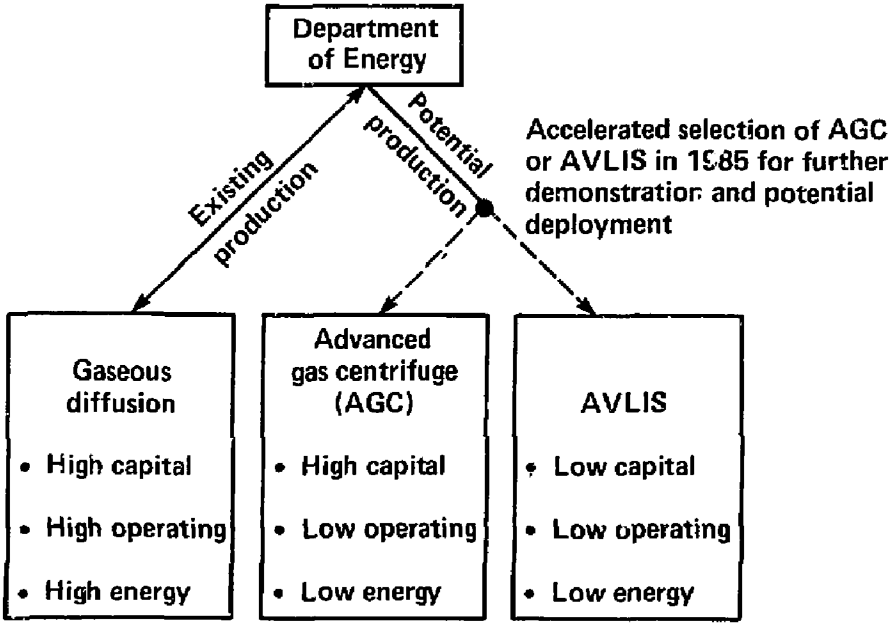


Fig. 23 Structure of U.S. enrichment enterprise.

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