Appl. Phys. B 46, 253–260 (1988)

Applied Physics B Photophysics and Laser Chemistry

Atomic Vapor Laser Isotope Separation*

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Received 19 April 1988/Accepted 25 April 1988

Abstract. Atomic Vapor Laser Isotope Separation (AVLIS) is a general and powerful technique applicable to many elements. A major present application to the enrichment of uranium for lightwater power reactor fuel has been under development at the Lawrence Livermore National Laboratory since 1973. In June 1985, the Department of Energy announced the selection of AVLIS as the technology to meet future U.S. needs for the internationally competitive production of uranium separative work. Major features of the AVLIS process will be discussed with consideration of the process figures of merit.

PACS: 32.80-T

Atomic Vapor Laser Isotope Separation (AVLIS) is a general process for converting a feed stream into a product stream in which a selected set of isotopes has been enriched or depleted [1]. The heart of the process is the selective multistep photoionization of an atomic vapor stream. The components of a generic AVLIS process are shown in Fig. 1. The process hardware is divided into a separator system and a laser system that are, to a great degree, mechanically independent. Atomic vapor is produced in the vaporizer and expands upwards in vacuum. Tunable laser frequencies are generated in a dye laser system that is in turn driven by a pump laser system. Copper lasers serve as the pump lasers in the major systems we have constructed. Both the pump lasers and tunable lasers are configured in master-oscillator/power-amplifier (MOPA) chains. The laser light illuminates the atomic vapor near the surface of an ion extractor. Photoions are drawn to and neutralized at these electrically biased surfaces. The remaining vapor streams through to the roof of the separator. In an enrichment mission the material from the extractor is enriched product; the material from the roof is depleted tails. In a stripping mission, product and tails are reversed. At Livermore, we have investigated the application of this laser isotope separation technology to a number of missions including those for defense programs. Of particular interest are applications with reasonable product demand, some of which are shown in Fig. 2. The dominant mission is clearly the enrichment of uranium for use in civilian light-water reactors. Laser isotope separation technology for uranium enrichment has been under development at Livermore since 1973 in a joint effort with

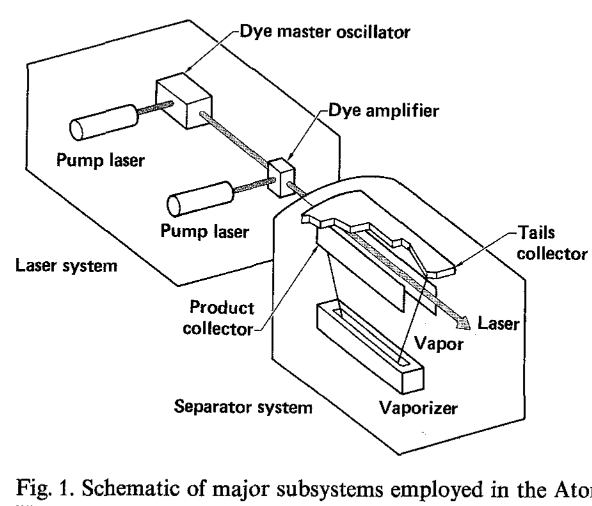

Fig. 1. Schematic of major subsystems employed in the Atomic Vapor Laser Isotope Separation (AVLIS) process at Livermore

* This paper is a synopsis of a presentation made at the Royal Swedish Academy of Engineering Sciences IVA Symposium "Laser Technology in Chemistry," Stockholm, Sweden, November 10, 1987

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

Fig. 2. Candidate elements for processing using atomic vapor laser isotope separation

Martin Marietta Energy Systems, Oak Ridge, Tennessee. In June 1985, following an intensive year-long review, the Department of Energy selected AVLIS as the technology to meet the nation's future need for competitive production of enriched uranium.

1. Process Figures of Merit

The figures of merit of an enrichment process can be understood by examining the SWU (Fig. 3). The SWU is a value function with units of mass that depends on the mass flows and assays. The expression comes from cascade theory. It is related to, but not proportional to, the entropy of mixing destroyed by the enrichment process. For natural uranium feed, light-water-reactor fuel product, and typical tails assay, 1 SWU converts 1.24 kg of feed into 0.21 kg of product. Consequently, a world demand of 7000 Mg of product/year corresponds to processing 40,000 Mg of feed/year and producing 3×10^7 SWU. Hence any uranium-enrichment process involves large-scale hardware.

A reasonable target cost for separative work is \$ 60/SWU. This corresponds to a processing cost of about \$ 50/kg of feed or approximately \$ 300/kg of product. Breaking the SWU cost into components as shown in Fig. 4, two components reflect the process engineering costs for the materials-handling and laser systems and two components reflect the process physics, i.e., the laser energy required to process one unit of feed and the separative work obtained from that feed.

To evaluate what is required of the laser system, it is necessary to estimate the MJ of laser energy required to process 1 kg of feed (Fig. 5). Outside of conversion factors, the laser energy is the product of the photon utilization (i.e., the fraction of the required photons that is actually absorbed – accounting for the saturation level necessary to drive the laser/atom kinetics and the light losses in optical elements), the absorbed photon energy needed to process one atom (about 6 eV for any typical photoionization or photodissociation process), and the fraction of the atoms that absorb the light. For a process such as uranium enrichment of natural material, where the isotope of interest is only a very small fraction of the vapor (0.0072 mole fraction), it is essential to have very high photoselectivity. Otherwise, absorption in unwanted isotopes will dominate and require a larger laser system. Putting in values characteristic of the AVLIS process, including the very high process photoselectivity, roughly 100 kJ of laser energy are needed to process 1 kg of uranium feed. The laser cost/kg feed is expected to be on the order of \$ 10.

$$\begin{aligned} \text{SWU (kg)} &= \text{Product (kg)} \cdot V(X_P) \\ &\quad + \text{Tails (kg)} \cdot V(X_T) \\ &\quad - \text{Feed (kg)} \cdot V(X_F) \\ V(X) &= (2X-1)\ln\left(\frac{X}{1-X}\right) \\ X &= \text{wt fraction } {}^{235}\text{U} \end{aligned}$$

At "standard" conditions

$X_F = 0.00711$
$X_P = 0.032$
$X_T = 0.002$
1 SWU converts 1.24 kg of feed into 0.21 kg of product

Fig. 3. Separative work units

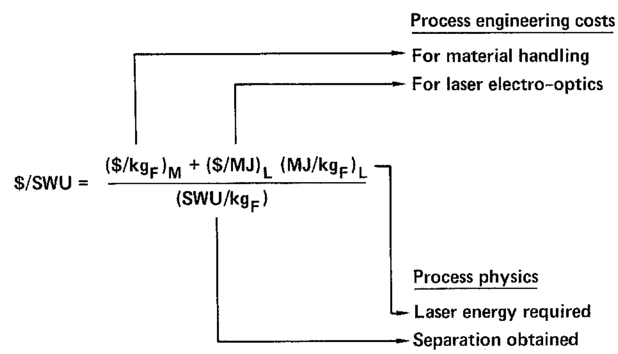

Fig. 4. Elements of separative work cost

$$\underbrace{(\text{MJ/kg}_F) = \underset{\substack{\text{Photon}\\ \text{utilization}\\ (0.2)}}{\frac{1}{\epsilon}} \times \underset{\substack{\text{Photon energy}\\ \text{(all steps)}\\ (6)}}{h\nu\left(\frac{\text{eV}}{\text{atom}}\right)} \times 1.6 \times 10^{-25}\left(\frac{\text{MJ}}{\text{eV}}\right) \times \underset{\substack{\text{Molecular}\\ \text{weight}\\ (238)}}{\frac{6 \times 10^{26}}{\text{MW}}}\left(\frac{\text{atoms}}{\text{kg}_F}\right) \times [\underset{\substack{\text{Mole fraction}\\ \text{of desired}\\ \text{isotope}\\ (0.007)}}{X_F} + \underset{\substack{\text{Photo-}\\ \text{selectivity}\\ (> 10^4)}}{\frac{1}{S}}(1 - X_F)]}_{(\text{MJ/kg}_F) \gtrsim 0.1 \text{ for uranium AVLIS}}$$

$(\$/\text{MJ})_L \lesssim \$100$ for an engineered electro-optics system

$\therefore (\$/\text{kg}_F)_L \lesssim \$10$ for uranium AVLIS

- High selectivity is essential to achieve low MJ/kg_F
  – Obtained in AVLIS

Fig. 5. Laser figures of merit

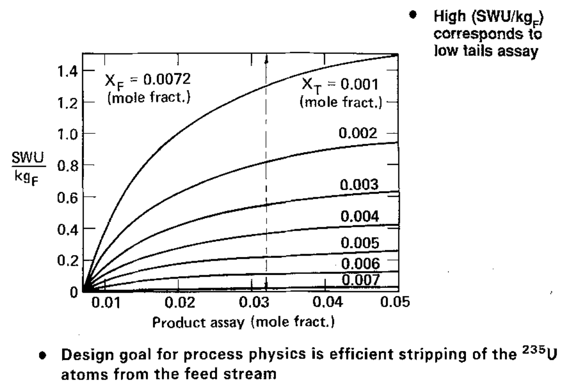

Fig. 6. Separative work performance

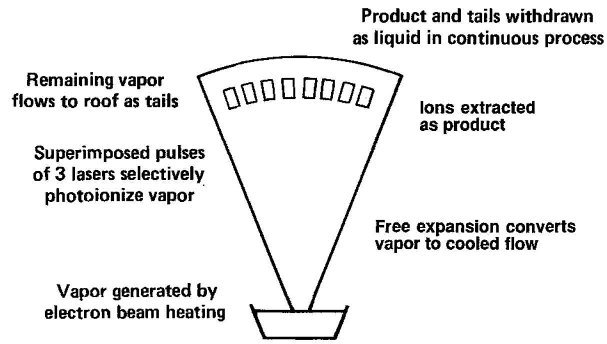

Fig. 7. AVLIS process steps

The other half of the physics performance is the separative work generated per kg of feed. Figure 6, which is a map of separative work performance vs product and tails assay, shows that high SWU production corresponds to low tails assays, not to high product assays. In other words, high selectivity is not essential to achieve high SWU/kg of feed. However, high selectivity is essential to achieve low MJ/kg feed and low \$/SWU. A process that has low feed assay X_F and low selectivity S will have a laser cost $1/(SX_F)$ 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 of feed. The AVLIS process is attractive because it can achieve extremely high process photoselectivity.

The goal for the physics of the AVLIS process is therefore clear; to strip a very high fraction of the ^{235}U atoms out of the feed, leaving few ^{235}U atoms in the tails. All modeling and experiments to date indicate that high stripping efficiency at high photoselectivity and consequently low separative work costs can be obtained using AVLIS.

2. AVLIS Process Steps

The physical steps an atom encounters as it passes through an AVLIS separator are illustrated in Fig. 7. Uranium vapor is generated by an electron beam striking uranium metal held in a cooled crucible. This type of vaporization is commonly used in the metal-plating industry. Nevertheless, uranium is highly refractory (the metal boils at 4100 K) and the technology demands high vaporization rates. The vapor then undergoes adiabatic free expansion into vacuum, in the course of which it transits all of the regimes of aerodynamics: continuum flow near the source, transition flow as the vapor expands, and eventually free molecular flow.

As the vapor passes near the extractor surfaces it is illuminated by three superimposed laser pulses of the wavelengths necessary to drive a three-step photoionization. The approach being used at Livermore 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, pulsed dye lasers pumped by copper lasers are used in the process. The temporal lengths of the laser pulses are shorter than the radiative lifetimes of the resonant levels in the transition sequence. Very high photoselectivity is attained on each step. Some of the methods of ionization that have been investigated to optimize performance and minimize process laser power requirements are shown in Fig. 8. The net photoselectivity in the process is extremely high as indicated in Fig. 9. The photoionization process can be described as an optical distillation column where work is done only on the isotope or species of interest.

The initial photoplasma contains, to a very good approximation, only ^{235}U ions. If the plasma were left unperturbed, resonant charge exchange between ^{235}U ions and ^{238}U neutrals would in time restore the ion population to natural abundance. This sets a time scale for efficient extraction. The ions go to negatively biased surfaces, and the electrons go to positively biased surfaces or to ground.

In order to obtain the economic advantages of continuous operation, the product stream on the extractors and the tails stream on the roof are collected by liquid flow. This obviously means that the collecting surfaces run at high temperature. In a sense precision laser physics is being performed inside a foundry.

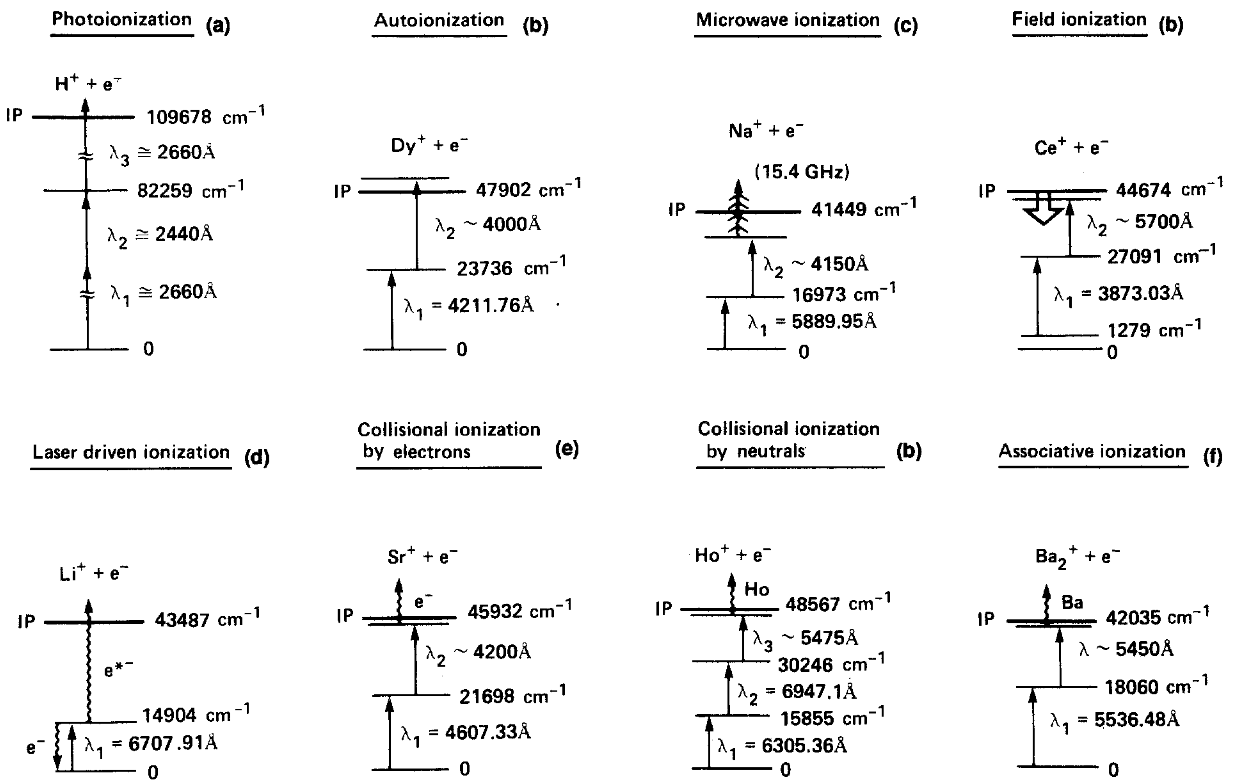

Fig. 8. Methods of atomic ionization after multistep excitation. (a) G.C. Bjorklund, C.P. Ausschnitt, R.R. Freeman, R.H. Storz: Appl. Phys. Lett. **33**, 54 (1978). (b) E.F. Worden, R.W. Solarz, J.A. Paisner, J.G. Conway: J. Opt. Soc. Am. **68**, 52 (1978). (c) P. Pillett, H.B. van Linden van den Heuvell, W.W. Smith, R. Kachru, N.H. Tran, T.F. Gallagher: Phys. Rev. A **30**, 280 (1984). (d) T.J. McIlrath, T.B. Lucatorto: Phys. Rev. Lett. **38**, 1390 (1977). (e) E.F. Worden, J.A. Paisner, J.G. Conway: Opt. Lett. **3**, 156 (1978). (f) R.W. Solarz, E.F. Worden, J.A. Paisner: Opt. Eng. **19**, 85 (1980)

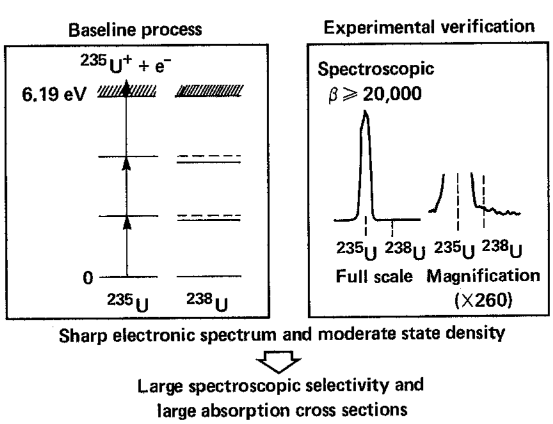

Fig. 9. Spectroscopic selectivity – AVLIS

3. Performance and Cost Modeling

The present generation of systems has been designed and tested after intensive study of the interplay of the physics and engineering that governs the AVLIS process. Each physics area has been modeled, in some cases from first principles, and benchmarked against results obtained in the laboratory or in large-scale enrichment systems described below.

Figure 10 shows an example of the detailed modeling performed in the area of photoionization and propagation physics. The theoretical approach includes a first-principles model based on integrating Schrodinger's equation. This computer model uses as input a complete set of experimentally measured spectroscopic and kinetic parameters corresponding to the relevant transitions of the atom of interest. The code treats the coherent multiple laser/atom interactions and accounts for the evolution of every magnetic sublevel and velocity class within each hyperfine level in the excitation sequence. The code also computes the atomic polarization driven by the light fields and thus facilitates analysis of resonant propagation effects.

These physics models have been incorporated, along with engineering models, in an integrated process model (Fig. 11). The engineering cost models include results from detailed bottom-up costing studies and data obtained from procurements for our demonstration facilities. Also included in the process model are operational parameters based on reliability,

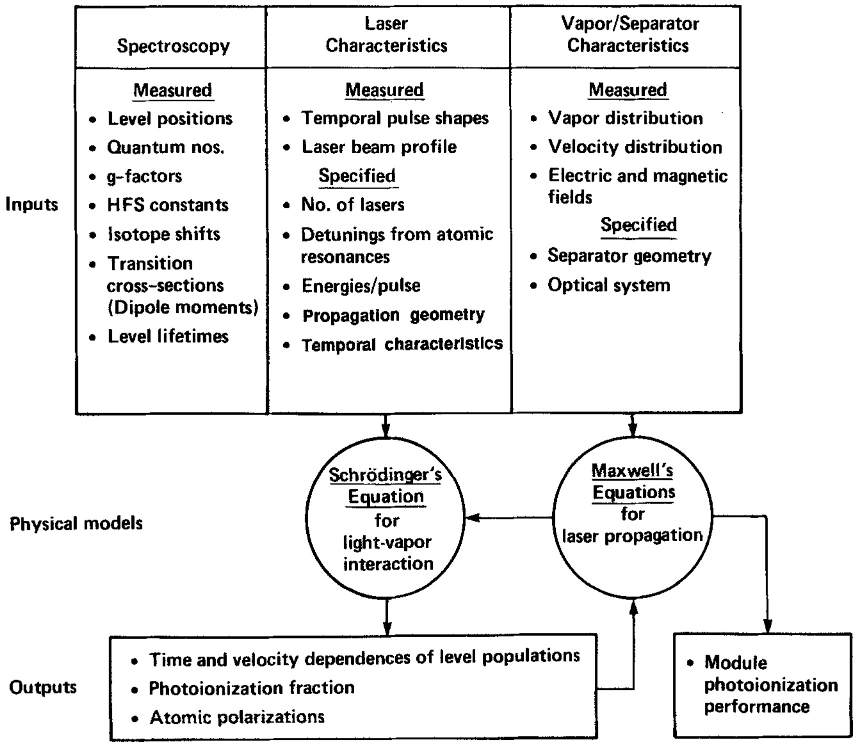

Fig. 10. Modeling of laser-atomic vapor interaction

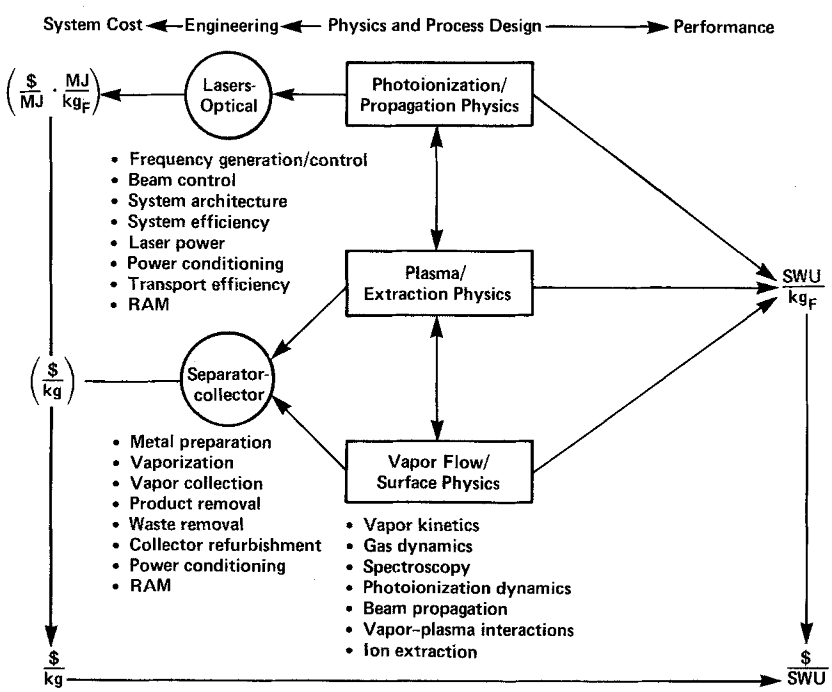

Fig. 11. AVLIS process morphology/structure of integrated process model (IPM)

availability, and maintainability of AVLIS subsystems. Essentially, the integrated process model contains all the fine detail of the AVLIS process and allows an examination of the sensitivity of cost and performance to variations in engineering, design, and physics parameters. There are literally hundreds of parameters that affect the separative work cost of the process. These range from the values of the optical-transition cross sections to the cost of labor. Each one of these parameters has an associated uncertainty and un-

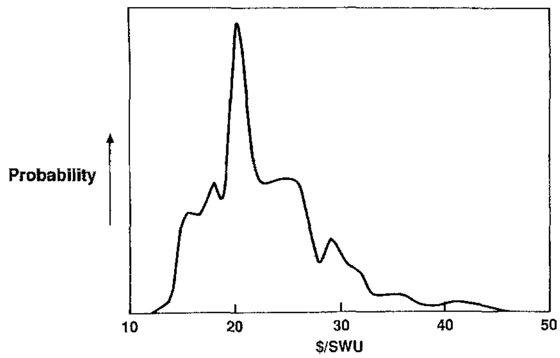

Fig. 12. AVLIS multivariable sensitivity study: distribution of projected separative work cost ~9 million SWU/year (1981 design, 1982 dollars)

certainty distribution. 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. Figure 12 illustrates a multivariable sensitivity study histogram for process separative work cost for the 1981 AVLIS engineering design.

4. Development Status

It has taken about 15 years for the AVLIS program to reach its current state of maturity. Process science studies were completed several years ago. In 1985 a full-scale demonstration facility was activated at Livermore. The building, shown in Fig. 13, houses a uranium separator module called the Separator Demonstration Facility. The building also contains a laser 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.

Fig. 13. AVLIS Full Scale Demonstration Facility consisting of a Laser Demonstration Facility (LDF) and a Separator Demonstration Facility (SDF) at Livermore. The building contains provisions for several copper laser and dye laser corridors and a prototype plant enrichment module.

Figure 14 shows the first completed corridor of copper laser MOPA chains installed in the facility in April 1985. There are 6 MOPA chains containing 30 laser heads (Fig. 15) with a total output capacity of several thousand watts. Figure 16 shows a section of the optical system in the dye laser corridor.

Figure 17 shows the separator module in the Separator Demonstration Facility. The tanks at the

Fig. 14. LDF corridor with copper laser master-oscillator/power amplifiers

Fig. 15. A copper laser operating on a test stand. It operates at approximately 5 kHz and is capable of generating several hundred watts in two colors, 5107 and 5782 Å

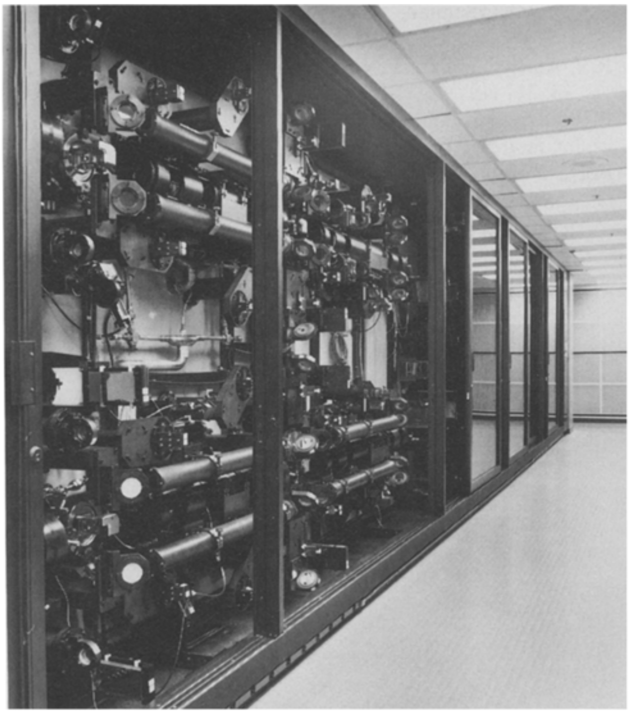

Fig. 16. Optical system in dye laser corridor in the Laser Demonstration Facility

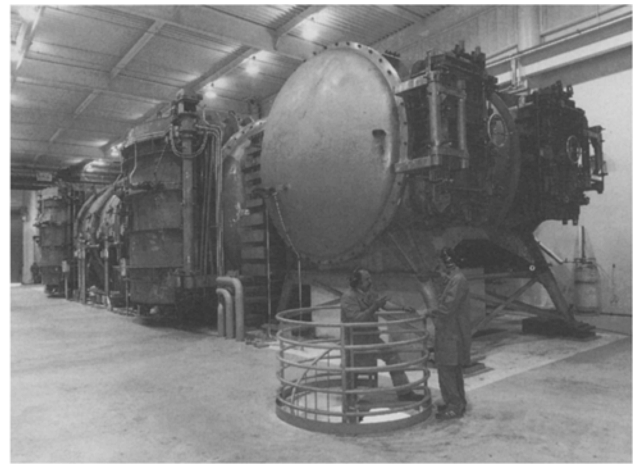

Fig. 17. Prototype AVLIS separator module in Separator Demonstration Facility

ends house the module for directing the laser beams through the uranium vapor. The module is essentially plant size and has a projected production rate of over 5×10^5 SWU/year, or 100 Mg of product.

5. Summary

As discussed in this paper, the program at Livermore has focused on laser isotope separation of atomic uranium because of the large demand and high product enrichment price for material used as fuel in commercial light-water nuclear power reactors. In support of that effort, a fundamental approach to the underlying physics was adopted and techniques to obtain the necessary data base were developed. Once developed and deployed for uranium, the AVLIS process can be applied directly to separating many elements economically on an industrial scale. Figure 18 shows the elements whose electronic transitions are accessible using the fundamental and harmonics of the dye system under development at Livermore.

AVLIS has reached industrial scale due to the dedicated efforts of several hundred scientists, engineers and supporting staff. Clearly, the broad range of technologies that comprise AVLIS offer exciting prospects for the future of laser driven processes.

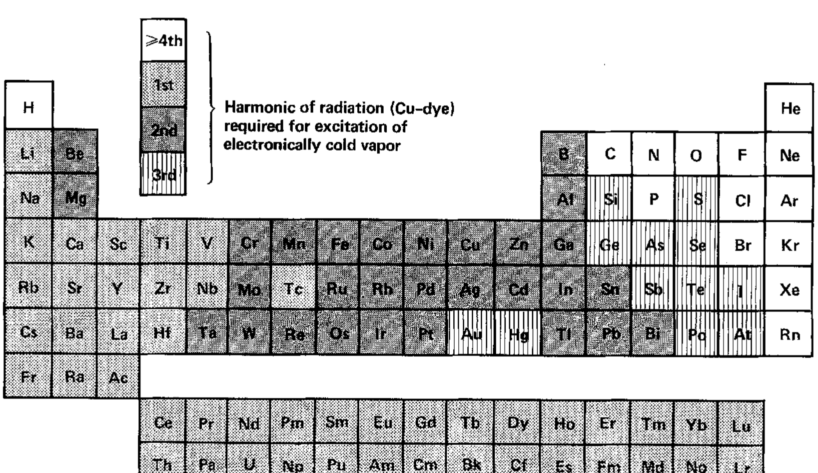

Fig. 18. Elements separable by AVLIS using copper-laser/dye-laser technology

Acknowledgement. Work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract W-7405-Eng-48.

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