LASER ISOTOPE SEPARATION IN NUCLEAR WASTE BY-PRODUCT UTILIZATION*

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ABSTRACT

Various by-products in spent nuclear fuels including strategic metals are uniquely useful and of high intrinsic value. Isotope separation is necessary to achieve the full benefits of fission product partitioning, increasing the specific activity of radioactive modifications or reducing the intrinsic radiation associated with various elements. The atomic vapor laser isotope separation process, under large scale development for uranium enrichment, applies to most of the spent fuel nuclides and offers attractive benefit to costs.

INTRODUCTION

Spent nuclear fuels contain on the order of 30 different fission product elements and several actinides in addition to uranium and plutonium. Although the fission products constitute only a minor portion of the discharged fuel mass, their elementally separated value can be an important fraction of the uranium fissile fuel value. These by-products represent a potentially significant national resource for a variety of processing industries (e.g., agriculture, chemical and petroleum) and, for example, may find large scale applications in the waste sanitization and medical fields. Furthermore, removal of selected fission products (e.g., Sr and Cs) from the waste stream can significantly offset certain waste management costs.

For several cases of interest, isotope separation is necessary to achieve the full benefits of fission product partitioning. Although many of the elements and specific isotopes in the waste stream are uniquely useful and of high intrinsic value, conventional isotope separation technologies are either not applicable or, in general, prohibitively expensive. Advanced isotope separation processes, including laser isotope separation (LIS), offer major cost reductions for the isotopic enrichment/purification of the myriad of by-product nuclides. The atomic vapor laser isotope separation (AVLIS) process, under large scale development for uranium enrichment at the Lawrence Livermore National Laboratory, is directly applicable to most of the spent fuel elements. We outline here the generic benefits of isotope separation, the AVLIS technology status and some of the associated economic factors.

BENEFITS SUMMARY

We illustrate in Table I several fission product isotope separation applications. Figure 1 provides the approximate isotopic assays referenced to ∿1 year after discharge. The most notable application is the isotopic cleanup of the radioactive components in the strategic platinum group metals (PGM). In the absence of isotopic cleanup, severe usage restrictions are expected or equivalently the worth of the reactor materials may be substantially below the world market prices. The composite value of PGM in a typical commercial spent fuel is potentially ∿30% of the value of uranium in the spent fuel, based on current market prices.

TABLE 1. FISSION PRODUCT ISOTOPE SEPARATION APPLICATIONS

Element	Isotope	Key applications	Isotope separation benefits
Palladium Rhodium Ruthenium	Stable	• Catalysts • Electrical contacts	Decrease intrinsic radiation
Krypton	Kr-85	• NDT • Self-powered lighting • RTG	Increase specific activity
Strontium	Sr-90	• RTG	Increase specific activity
Cesium	Cs-137	• Irradiator – Sewage/sludge – Food	Increase specific activity
Promethium	Pm-147	• RTG • Self-powered lighting	Decrease intrinsic radiation

For Sr-90, the benefits of isotope separation are a more effective utilization of the available Sr-90 resources and lower radio-thermoelectric-generator (RTG) system costs. Isotope separation would enable the use of substantial quantities of low-grade Sr-90 (assay degraded by decay and/or chemical contamination during processing), and enrichment to high assays would allow extended RTG operation or extensive recycle. Kr-85 is in comparatively minor isotopic abundance and certain self-powered lighting applications could benefit from higher specific activity material. Radio-thermoelectric-generators represent a potentially large scale application for Kr-85, but highly enriched Kr-85 will be required to achieve adequate power densities. In connection with RTG applications, Pu-238 and Sr-90 are in roughly comparable

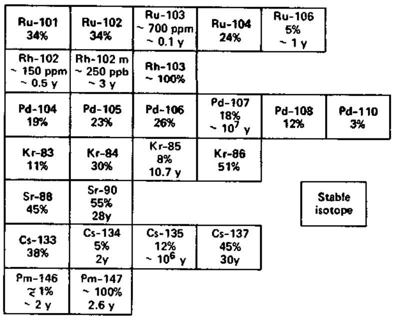

Fig. 1. Isotope abundances and half-lifes.

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quantities in commercial spent fuels, but the former is in minor isotopic content (∿1%). A suitable isotope separation process could enrich Pu-238 and also remove the undesirable Pu-236 component.

ATOMIC VAPOR LASER ISOTOPE SEPARATION

The physical principle behind LIS is the selective absorption of laser radiation by atoms/molecules containing the isotope of interest. In the specific enrichment process under development for the DOE at LLNL, we use precisely tuned (laser pumped) dye laser radiation to selectively photoionize the U-235 component in the atomic vapor stream. Three visible laser beams are produced with sufficiently precise wavelengths to distinguish between the U-238 and U-235 electronic energy levels which are slightly shifted with respect to one another. This and alternate one- and two-step processes are shown in Fig. 2. Following ionization, the U-235 atoms are extracted from the neutral vapor by pulsed electromagnetic fields and collected as enriched products. Figure 3 shows the basic elements of the process.

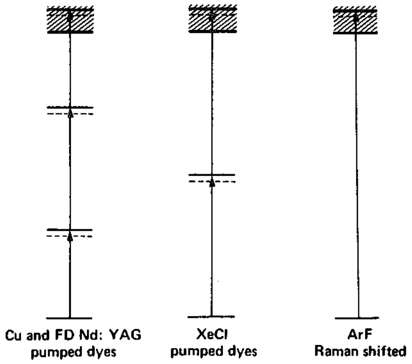


Fig. 2. Laser photoionization options utilize copper vapor laser, frequency doubled neodymium YAG (FD Nd:YAG) and XeCl pumped dye lasers, as well as Raman shifted ArF lasers.

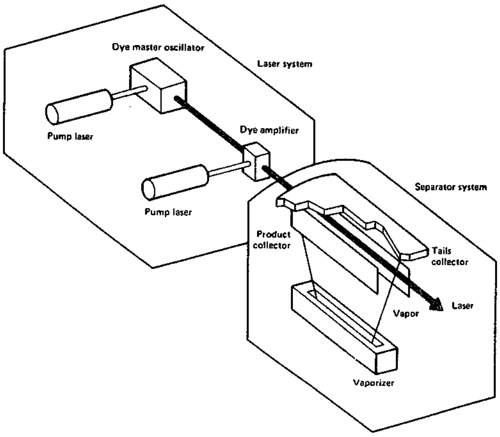


Fig. 3. Atomic vapor laser isotope separation -- major systems.

Since 1974, we have concentrated on developing the laser, vaporizer and extractor systems that can be scaled to large sizes. We have identified a set of baseline laser transitions and accurately measured all parameters in the photoionization process. Most effort has been directed toward the three-step photoionization process (Fig. 2), enabling cumulative selectivities in excess of 10^7. We have also successfully completed macroscopic enrichment experiments confirming that the baseline process agrees with our detailed model predictions. Finally an extensive development program, carried out at both LLNL and Union Carbide Corporation-Nuclear Division (Oak Ridge), has demonstrated that atomic uranium vapor of the required densities can be vaporized and collected reliably.

The ability of the AVLIS process to obtain a large separation factor and mass throughput in a single stage gives it its projected cost advantage over conventional processes (Table II). The specific capital investment for AVLIS is an order-of- magnitude lower, and the AVLIS power consumption is minor compared to that for the gaseous diffusion process.

	CAPITAL COSTS	ENERGY REQUIRE-MENTS	OPERAT-ING COSTS
Gaseous diffusion	high (~$600/SWU/yr)	high (~2400 kWh/SWU)	low
Gas centrifuge	high (~$800/SWU/yr)	low (~100 kWh/SWU)	moderate
Laser isotope separation	low (~$60/SWU/yr)	low (~100 kWh/SWU)	low

TABLE 2. COMPARISON OF URANIUM ENRICHMENT COSTS.

The design approach for our copper vapor-pumped-dye laser system that satisfies plant availability and reliability requirements is shown in Fig. 4. The laser system consists of two major modules, the waveform generator (oscillator and preamplifiers) and the power amplifier. The waveform generator provides the temporal, spatial, and spectral format required to drive the photoionization process. The waveform generator output is amplified by the power-amplifier module, which in turn provides sufficient power to fully illuminate the uranium vapor in the separator modules.

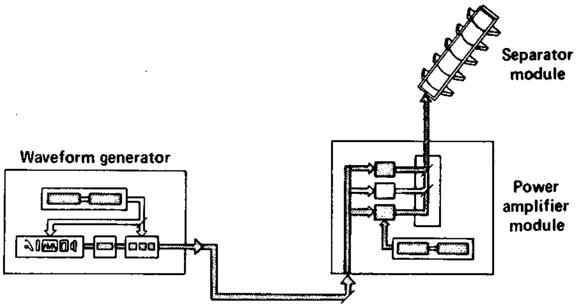


Fig. 4. Conceptual relationship between major AVLIS modules.

Development of adequate copper vapor lasers has been an important goal of the AVLIS program. A progression of lasers that have been developed in recent years is shown in Fig. 5, while Fig. 6 shows one of the ∿100 W lasers in operation.

Frequency conversion and amplification is accomplished using dye lasers which also have been extensively developed at LLNL. A dye laser amplifier is shown in Fig. 7. To date, the highest output power we have obtained from this type of dye laser is 55 W, operating at 6 kHz with 45% conversion effi-

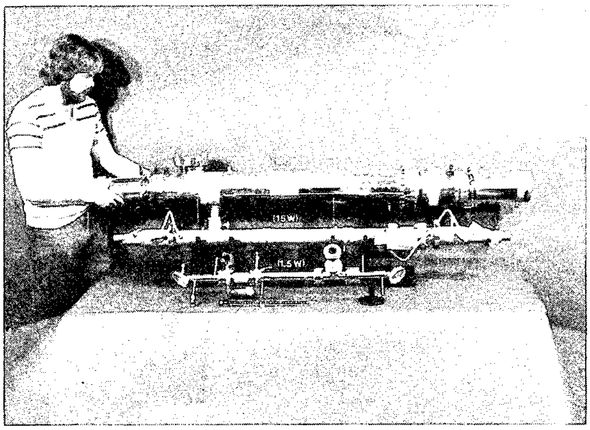


Fig. 5. Copper vapor laser development.

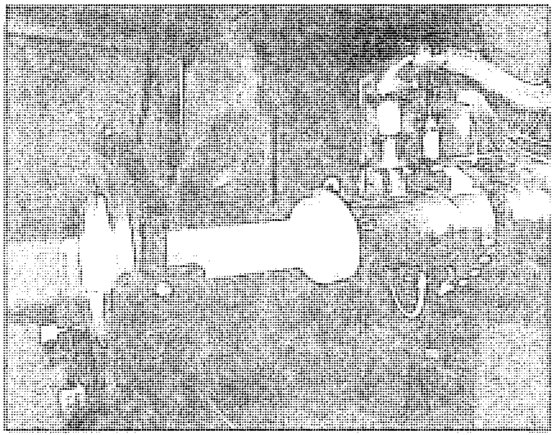


Fig. 6. Large bore copper vapor laser oscillator operating at approximately 90 W output power.

ciency. For the fission product applications, this power (reduced for laser system and process losses) equivalently corresponds to a product rate of ∿10-1000 kg/yr, depending on the specific application.

To enable the development of the laser electro-optic systems, we have constructed an integrated system of enrichment facilities which are operated on a routine basis. These are represented in Fig. 8. As in our AVLIS plant design, the laser system consists of the two major modules: the waveform generator (SPP-II) and a power amplifier (Venus). The composite system demonstrates all of the functions of a plant laser system. The system includes 40 copper vapor lasers, 5 dye master oscillators, 7 dye preamplifiers, and a full complement of beam transport optics, support structures and diagnostics/controls.

Our physics experiments are conducted in the low throughput separator (Regulis), while the high throughput separator (Mars) is used to test process characteristics at large scale and is also used to test production process technologies related to materials handling and reliability. Both separator systems use electron beam evaporation, optimized for the AVLIS process. Figure 9 shows the large scale separator.

The AVLIS technology base developed over the past 10 years is very broad in scope and applicable to many elements of interest other than the actinides. We have extensively explored a variety of scalable laser systems (e.g., Fig. 2), vaporizers, extractors, and collectors, as well as various process excitation/discrimination schemes. For the fission product applications, detailed process physics data bases must be acquired to set precise bounds on the economics. From a technical standpoint, fission product AVLIS is easier in some respects but more difficult in other respects to uranium-AVLIS. To illustrate, for strontium the intrinsic materials handling (vaporization and collection) is easier due to its comparatively low melting point and high volatility. On the other hand, the achievement of adequate isotopic resolvability for strontium and the PGM is more challenging due to their comparatively small isotope shifts. In this connection, we have identified several scientifically viable approaches for obviating this restriction.

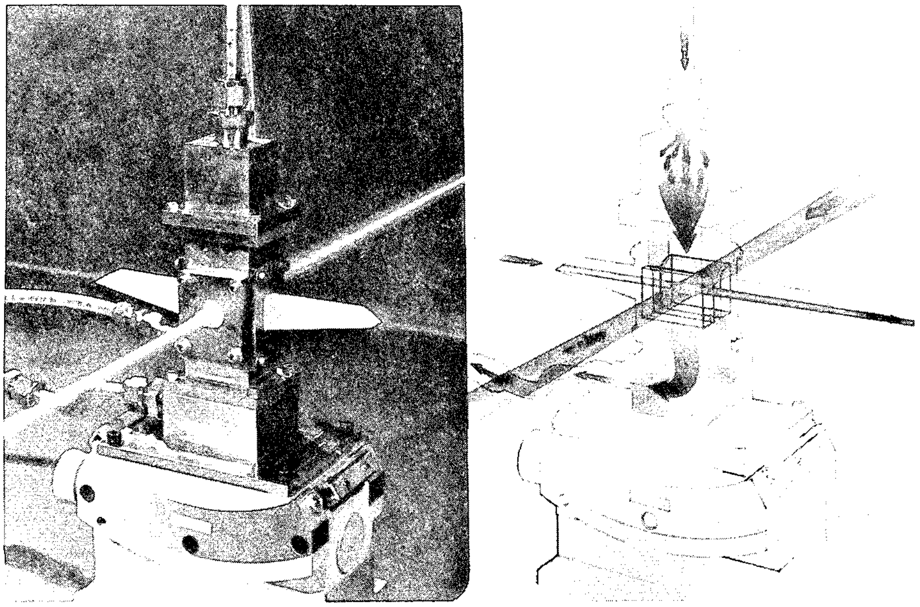


Fig. 7. Dye laser amplifier shown with mutually orthogonal directions for excitation, flow, and amplification.

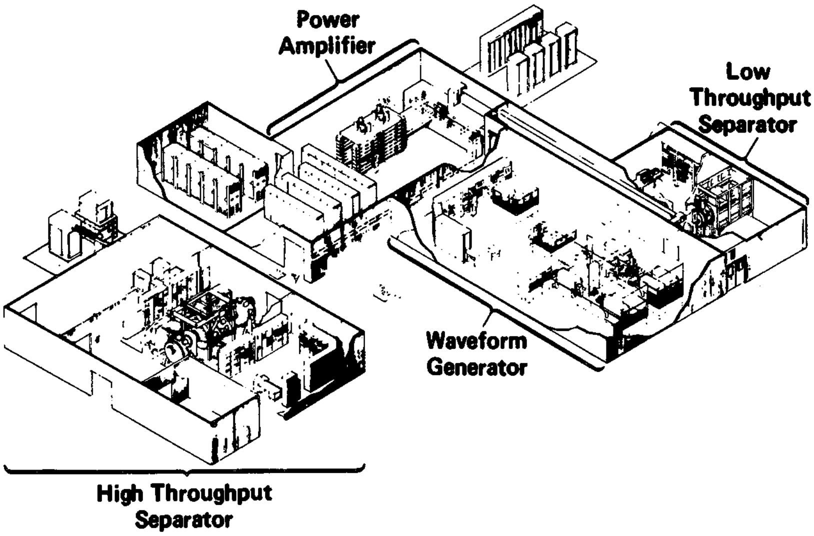


Fig. 8. Major on-line facilities of the LLNL AVLIS program.

ECONOMIC ASSESSMENT

The added value or benefit of isotope separation depends on the detailed analyses of the various applications. Much of the data required here for such analyses are not precisely known. Nonetheless, for our present objectives, order-of-magnitude estimates of the plausible benefits suffice: \$1-\$10/gram (PGM), \$10-\$100/gram (Sr-90) and \$1800/gram (Pu-238: DOE price guide, June 1981). We note that at current market prices the economic incentive for even recovering the PGM from the nuclear waste stream will depend on achieving lower extraction costs (non-aqueous processing). In any event, here we concern ourselves only with the plausible add-on costs for LIS.

The unit product cost of isotope separation is governed by the specific enrichment requirements (product and tails assay) and throughput set by product demand. We have found a simple expression to be quite useful for the general evaluation of laser material processing:

$$\left[\frac{\$}{\text{unit}}\right]_{\text{product}} = \left\{\left[\frac{\$}{\text{MJ}}\right] \cdot \left[\frac{\text{MJ}}{\text{unit}}\right]\right\}_{\text{laser}} + \left[\frac{\$}{\text{unit}}\right]_{\text{handling}}$$

This expression relates the unit product cost to the life-cycle cost of the laser energy, to the laser energy required per unit of product, and to the cost of handling the material throughout the process in a form compatible with laser processing.

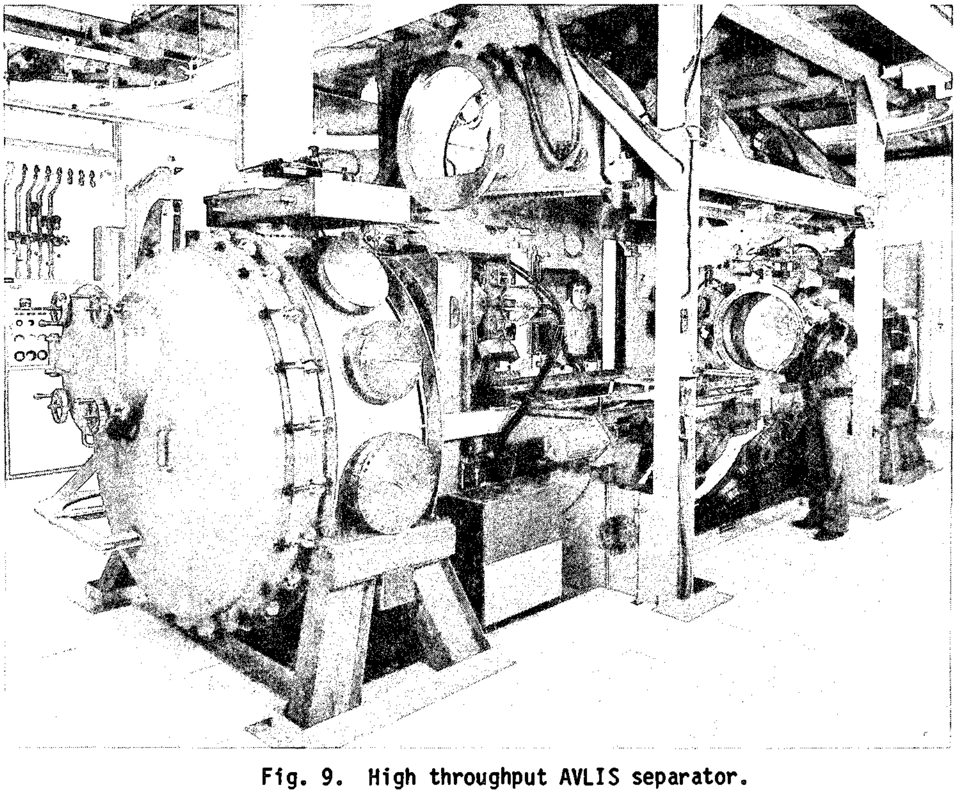
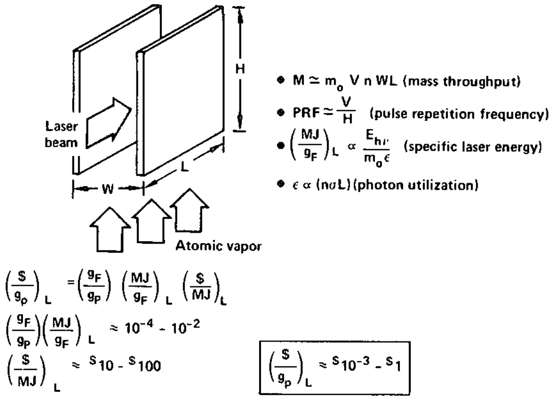


Fig. 9. High throughput AVLIS separator.

The laser energy required per unit of product depends on the stoichiometric amount of material that must be converted and the selectivity of the irradiation step. With the aid of Fig. 10, we can relate the specific laser energy requirements for the AVLIS process to the energy ($E_{h\nu}$) to photo-ionize the vapor, the atomic weight of the element (m_o), the photon utilization efficiency (ε) and the product yield (g_P/g_F). For purificiation applications, the laser energy requirements are generally much smaller than for applications involving the enrichment of an isotope in minor feed abundance. This leads to a broad possible range of laser energy requirements. The characteristic AVLIS laser system cost for delivering a megajoule of highly coherent tunable radiation to the separator module lies in the \$10-\$100 range. This is a total life-cycle system cost (electrical, refurbishment, and amortized capital investment costs) and takes into account system losses due to frequency conversion, beam combination and beam transport. For the range of applications, we then find a laser system related cost contribution of $\lesssim$\$1/gram product.

The materials handling contribution includes all separator related costs associated with feed, product and tails processing operations. This cost center is a strong function of: (1) the stoichiometric and staging requirements, the latter which in turn depends on the single stage laser conversion yield, and (2) the specific radiation characteristics of the feed, which determine the added (non-direct) containment, shielding and maintenance costs. With few exceptions, the nuclear by-products are significantly radioactive, and the incremental handling costs can be quite significant, if not dominant for Sr-90 and Cs-137. A rough analysis leads to an overall materials handling cost in the range of \$0.05-\$0.5/gram product.

Our rudimentary assessment seemingly indicates attractive benefit-to-costs for reactor by-product isotope separation. However, the analyses implicitly assumed economies of scale characteristic of uranium enrichment, $\sim 10^6$kg/yr product per enrichment plant (low enriched uranium is also a high value product, the gaseous diffusion enrichment cost nearing \$1/gram). It is instructive then to speculate on the possible throughputs for these by-product applications. For the PGM, feed availability (nuclear economy size) would set the maximum throughput at least for the foreseeable future. In particular, a 100 GW$_e$ installed nuclear capability with complimentary reprocessing could provide $\sim 10^4$ kg/yr feed ($\sim$product), roughly 5-10 fold lower than domestic imports (PGM). Conversely, the throughput for other applications will be limited by demand, expected to fall in the range of several to perhaps several hundred kg/yr. Clearly, the "total economic incentive" for uranium enrichment is orders-of-magnitude greater.



- $M \simeq m_o V n WL$ (mass throughput)
- $PRF \simeq \frac{V}{H}$ (pulse repetition frequency)
- $\left(\frac{MJ}{g_F}\right)_L \propto \frac{E_{h\nu}}{m_o \epsilon}$ (specific laser energy)
- $\epsilon \propto (n\sigma L)$ (photon utilization)

$$\left(\frac{\$}{g_p}\right)_L = \left(\frac{g_F}{g_p}\right)\left(\frac{MJ}{g_F}\right)_L\left(\frac{\$}{MJ}\right)_L$$

$$\left(\frac{g_F}{g_p}\right)\left(\frac{MJ}{g_F}\right)_L \approx 10^{-4} - 10^{-2}$$

$$\left(\frac{\$}{MJ}\right)_L \approx \$10 - \$100$$

$$\boxed{\left(\frac{\$}{g_p}\right)_L \approx \$10^{-3} - \$1}$$

Fig. 10. Laser system related product costs.

Simple cost scaling considerations indicate that on a stand-alone basis, these by-product isotope separation applications (limited throughputs) in general are not economically attractive. However, in analogy to many industrial chemicals and other industrial products, acceptable costs can be achieved by integration with an existing large scale laser process. This naturally raises the question on the nature and time frame for the large scale use

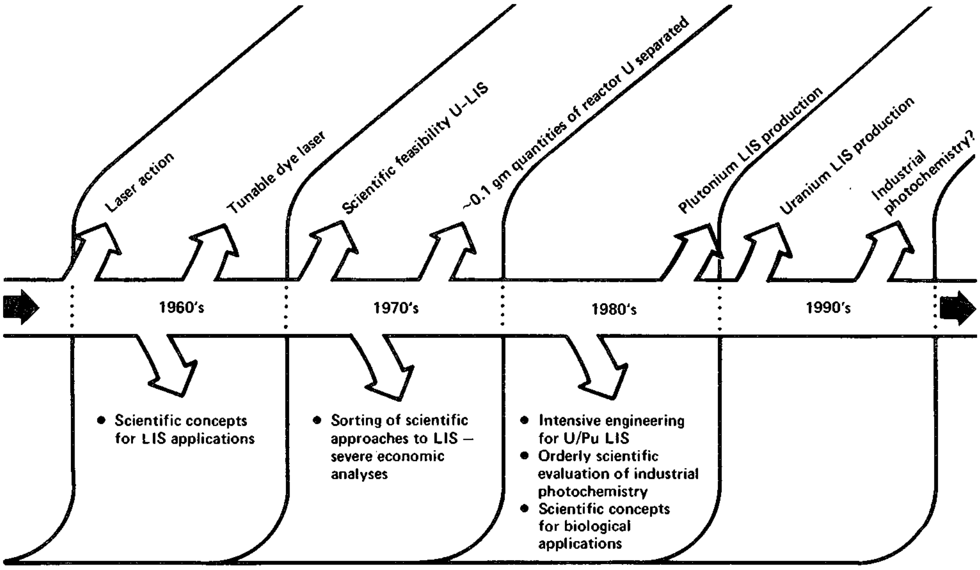


Fig. 11. History and anticipated progress in the science, technology, and application of industrial laser systems.

of lasers in material processing. Figure 11 summarizes our perspective on how long it takes to get a process into production and shows what we think might happen in the future. Uranium-AVLIS is expected to be the first use of lasers on a large scale basis and occuring in the early 1990s. This is also the plausible period for commercial reprocessing, essential for a substantial and sustained supply of nuclear by-products. The development and implementation of a significant nuclear by-product LIS capability with a firm economic basis is consistent with these schedules.

DISCLAIMER