Lasers in Materials Processing

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Abstract—**We analyze the general requirements for an economically viable laser materials-processing application. Laser light is not only expensive relative to other forms of energy but at ~$10/kg of product for laser processing costs (corresponding to one 2 eV photon per product molecule) it is expensive relative to most bulk chemicals. We identify four criteria for a successful application that allows efficient utilization of this costly source of energy. In reviewing the status of uranium laser isotope separation (LIS) at the Lawrence Livermore National Laboratories (LLNL), we show how this program satisfies our criteria.**

Introduction

HOW can the unique properties of laser photons be best utilized in the production of materials and components despite the high cost of laser energy? To answer this question we need first to identify the characteristics of potential applications that are necessary for success. We will also point out those factors that have up to now frustrated attempts to find commercially viable laser-induced chemical and physical processes for the production of new or existing materials. Having identified the general criteria to be satisfied by an economically successful laser process and shown how these, in turn, imply the laser system requirements, we will present a status report on the uranium laser isotope separation (LIS) program at the Lawrence Livermore National Laboratory (LLNL). The LIS of actinides is the only large-scale application for bulk laser photochemical processing that thus far satisfies these criteria.

Process Economic Analysis

The crucial issue with which any potential application must contend is the cost of laser energy. At approximately $1–$100/MJ it is orders of magnitude more expensive than electricity or fossil fuels. The classical concept of laser photochemistry envisions the production of bulk chemical compounds through selective excitation or breaking of specific chemical bonds, leading to a preferred pathway for the ensuing chemical reactions. The laser may drive the reaction through this selective application of energy or merely catalyze it. Either way, one hopes to gain an advantage by achieving a higher process yield of the desired product by bypassing many steps in the reaction scheme and avoiding nonproductive chemical pathways. However, one must deal with both process- and laser-related issues in realizing this hope. Standard chemical processes utilizing temperature and pressure control of the reactions are often quite efficient. This is reflected in the selling price of under $1/lb (or $2/kg) for most bulk chemicals sold in large volume. The specific laser costs are typically quite high relative to this. Consider a chemical process requiring one 2 eV (visible) photon for each molecule of product, enough to break a typical chemical bond. At a typical molecular weight of 100, this translates into on the order of $10/kg for laser energy.

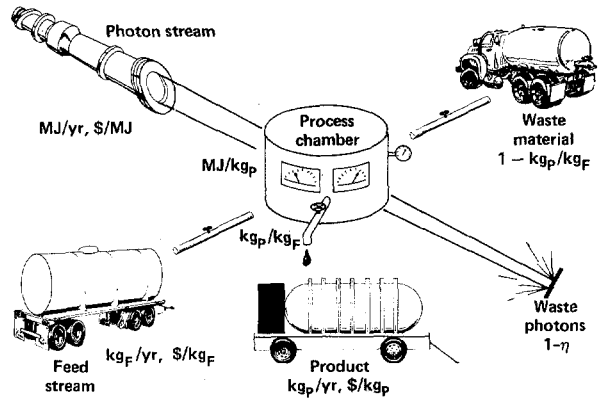

Fig. 1. Elements of a laser photoprocess expressed in terms of kilograms of product per kilogram of feed (kg$_p$/kg$_F$) and photon utilization efficiency (η).

In terms of the major process parameters listed in Fig. 1, which shows the elements of a generic laser photoprocess, how are currently successful applications of lasers in production able to overcome this high laser cost? The specific laser energy requirement in MJ of laser energy per kilogram of product (MJ/kg$_p$) is favorable in such applications as the cutting of fabrics, the welding of metals, and the heat treating of various materials because only a fraction, e.g., a surface layer or cross section, of the material is treated. In these applications the process yield in kilograms of product per kilogram of feed (kg$_p$/kg$_F$) is typically near one, while maintaining low specific laser energy per unit product. The value of the product is clearly also high relative to the laser costs for these fabricated products, as is the market size (kg$_p$/yr) allowing realization of economies of scale.

As opposed to fabricated products, the production of homogeneous substances such as bulk or high-value chemicals, pharmaceuticals, or reactor-grade enriched fuel, requires selective interaction within the volume of the feed stock. The selectivity can be intramolecular, picking out one bond to be broken, thus initiating a desired chemical reaction, or interspecies, picking out one isotope or distinct chemical compound to be activated. In either case, to realize the same advantage of low specific laser energy requirements (MJ/kg$_p$) enjoyed by the above fabrication processes, we again seek applications in which the number of product molecules per photon absorbed is large. This can occur through initiating chain reactions such

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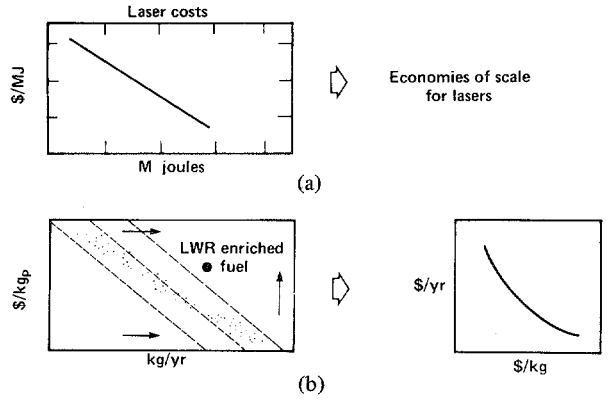


Fig. 2. (a) Yearly laser costs per megajoule are found to decrease with laser amplifier power. (b) High value chemicals typically have low demand not only in kg/yr but also in $/yr, making it difficult to find applications allowing economies of scale.

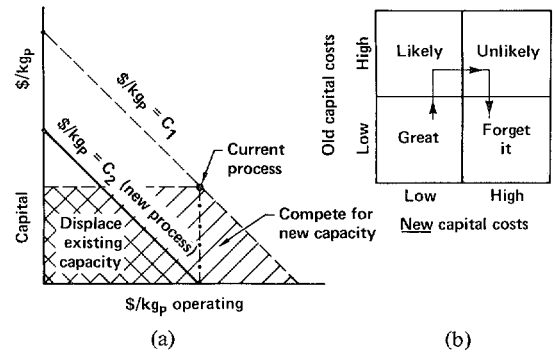


Fig. 3. (a) To be commercially competitive, the costs for a new laser photoprocess must fall in the (singly hatched) region with both lower cost/kg$_p$ and lower capital costs. To displace existing production capacity the new process must also have total costs lower than the operating costs of the existing process. (b) Low capital costs are always preferable for a new process, although an existing process with high (sunk) capital costs is more firmly entrenched because of the resulting lower forward (operating) costs for a given cost/kg$_p$.

as in polymerization, in creating catalysts that then promote many cycles of the ensuing reactions, or by selectively interacting with a minority species such as 1) H_2S in the laser purification of synthesis gas ($CO + H_2$) [1] or 2) ^{235}U in atomic vapor laser isotope separation as pursued at LLNL. The cost of the feed stream is minimized by seeking applications with high yield per kg of feed. Because of the high laser costs we again want applications in which both the product value ($/kg$_p$) is high and demand levels are high enough to allow some economies of scale. Unfortunately, these are often conflicting requirements, as seen in Fig. 2. Achieving economies of scale for lasers [Fig. 2(a)] implies processing many kg of product per year. Fig. 2(b) shows that materials with a large demand in kg/yr tend not only to have lower cost per kg (magnifying the specific laser processing cost problem) [2], but also tend to have lower dollar volume per year (making it a much less rewarding market to penetrate). The requirements of 1) sufficient selling price to offset the laser energy costs and 2) sufficient demand to obtain economies of scale are seen to exclude most chemicals produced.

Clearly, the unique selective characteristics of laser energy must be well utilized in any laser process in order to achieve the process and economic leverage necessary to justify its high cost. Unfortunately, rapid intramolecular vibrational relaxation rates can quickly (on a picosecond time scale) spread the energy selectively absorbed by one bond, leading to thermalization of the laser energy. The broad electronic (~vibronic) absorption bands of molecules also frustrate the selective intramolecular application of laser energy. In addition, few viable high-yield-per-photon applications have been identified.

For a new process to be adopted, the risk/reward ratio must also be favorable. The reward is measured by the reduction in unit cost of the product, achieved from a laser photoprocess, along with the size of the potential market. Requirements on these two factors have been covered above. The level of risk is measured by the probability of failure and by the up-front at-risk expenditures consisting of RD&D plus capital costs. In the capital-intensive chemical industry, the cost, availability, and possibility of loss of the capital investment must be carefully weighed not only against the potential benefits but also against the sunk costs in the existing process to be displaced. Hence, a new laser process may have a significantly lower unit cost, but, as shown in Fig. 3, unless it also boasts 1) lower capital costs and 2) lower cost/kg$_p$ than the forward (operating) costs of the existing process, it will have a low chance of acceptance. The diagonal lines on the graph in Fig. 3 are lines of constant cost/kg of product, which consists of amortized capital cost/kg plus the operating cost/kg. The two shaded regions on the graph point up the conclusion that it is much more difficult to compete against the forward costs of a capital-intensive existing process than to compete for new capacity against such a conventional process.

We can summarize the conclusions from our analysis in the form of four criteria for an economically successful laser photoprocessing application. These are: 1) produce "high value" or new products, 2) have markets allowing high volume/yr along with high value/unit, 3) achieve maximum leverage from laser photons by utilizing the unique laser properties with maximum yield per photon, and 4) require lower capital expenditures than existing processes, as well as lower $/kg$_p$. In scoping out a new process, these general criteria must be supported by a more quantitative, although approximate, estimate of the laser system requirements.

Atomic Vapor Laser Isotope Separation (AVLIS)

In the isotopic enrichment of uranium for light-water-reactor (LWR) fuel rods, we see that each of our four criteria can be satisfied. Furthermore, the added cost of enrichment, presently about $600/kg for LWR fuel, exceeds our $10/kg estimate of typical laser-processing costs by a favorably large margin. In fact, although AVLIS will reduce the enrichment cost to about $100/kg, we still have a high-value product.

The high cost of the fuel for light water nuclear reactors as well as the current and projected demand readily satisfy our first and second criteria for an economically viable laser materials-processing application. We also see that the unique, precisely definable properties of laser radiation can be applied to realize highly selective ionization of ^{235}U without also ionizing ^{238}U atoms [criterion 3], which are much more

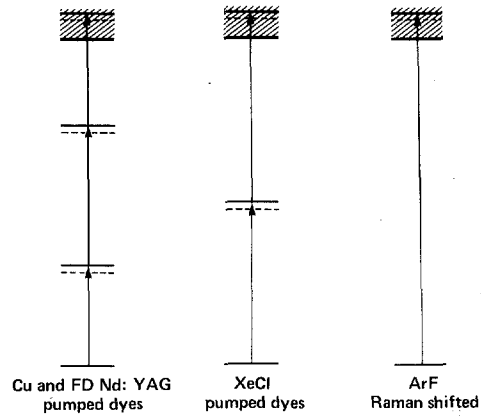


Fig. 4. Laser photoionization options utilize copper vapor laser, frequency doubled neodymium YAG (FD Nd:YAG) and XeCl pumped dye lasers, as well as Raman shifted ArF lasers.

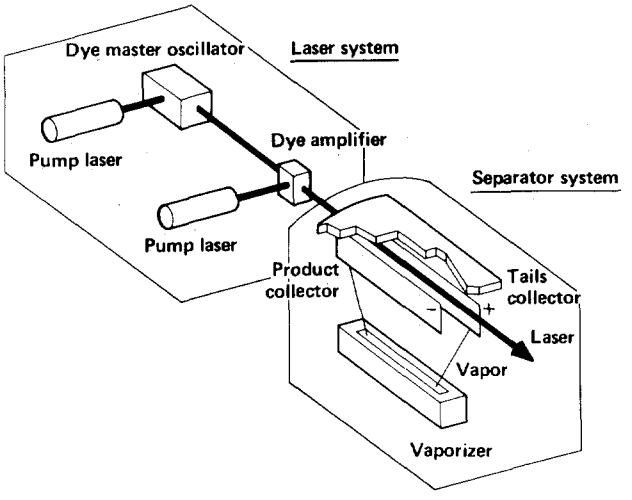


Fig. 5. Atomic vapor laser isotope separation–major systems.

abundant in both the feed and product. Finally, the fourth criterion is satisfied because LIS may profitably displace existing gaseous diffusion plants at only a fraction of their capital costs.

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 ^{235}U component in atomic vapor. Three visible beams are produced with sufficiently precise wavelengths to distinguish between the ^{238}U and ^{235}U electronic energy levels, which are slightly shifted relative to each other. This and alternate one- and two-step processes are shown in Fig. 4. Following ionization, the ^{235}U atoms are extracted from the neutral vapor by pulsed electromagnetic fields and collected as enriched product. Fig. 5 shows the basic elements of this process. The major elements of photon stream, feed stream, process chamber, and product stream in Fig. 1 are readily identified with their counterparts in this figure.

As contrasted with the financial perspective which we have just considered, there is the process-oriented perspective in which we may view the economics of the process. The materials-handling and laser system contributions to the cost/kg_p are calculated as

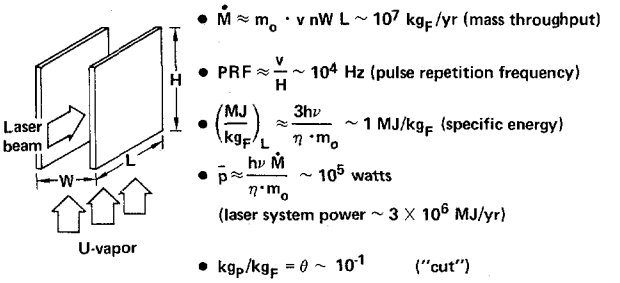


Fig. 6. AVLIS process and laser requirements.

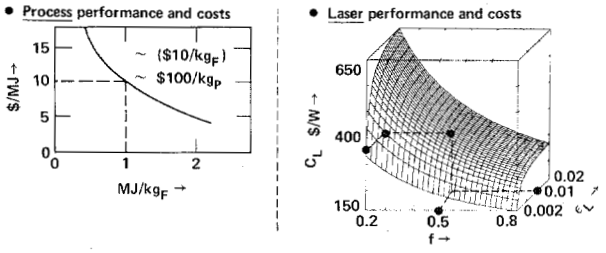
$$\$/kg_p = \frac{(\$/kg_F)_M + (MJ/kg_F)_L\,(\$/MJ)_L}{kg_p/kg_F}$$

where the process performance parameters are kg_p/kg_F (process yield) and $(MJ/kg_F)_L$ (specific laser energy requirements), and the process engineering costs are those for materials-handling systems and laser EO subsystems. Solving this equation for $(\$/MJ)_L$, we will determine acceptable laser system photon costs (\$/MJ) once the process parameters have been estimated. This along with laser power, frequency, and beam parameters will define the laser technology development goal and show the sensitivity of acceptable laser costs to some of the process physics parameters. In this way we obtain the projected range for \$/MJ and may select candidate lasers having a reasonable chance of development to the required engineering and cost specifications.

Fig. 6 presents an estimate of the order of magnitude of the important process and laser parameters. The total mass throughput is calculated in terms of the process-chamber geometry, atomic number density (n), vapor velocity (v), and average uranium molecular weight ($m_0 = 238$) to be on the order of 10^7 kg/yr. The requirements for the laser pulse repetition frequency (PRF) are determined by the inverse clearing time for the laser interaction region. Specific laser energy requirements are determined to be about 1 MJ/kg_F by combining the uranium ionization energy of about 6 eV, the molecular weight, and the system photon utilization efficiency η. The latter consists of the product of laser chain and frequency conversion efficiency, beam transport efficiency, and the fraction of photons absorbed by ^{235}U. Total system average power $\bar{p}$ is the product of mass throughput and the specific laser energy. Converted to appropriate units, it is on the order of 10^5 W. Finally, we can estimate the process yield from the desired feed, product, and tails assays to be about 10 percent.

With these estimates in hand, we can now use our simple economics model to determine acceptable photon costs. Fig. 7(a) shows the tradeoff of specific laser energy requirements with acceptable laser system costs, assuming the process yield in Fig. 6, ~\$10/kg_F for materials handling and ~\$100/kg_p, which is the approximate goal for enrichment costs for our process. While substantially less than the present \$600/kg_p quoted above, the latter figure is still many times our \$10/kg_p typical laser-processing estimate. Choosing our point estimate of 1 MJ/kg_F on this sensitivity curve corresponds to an acceptable photon energy cost of ~\$10/MJ.

The allocation of this life-cycle cost per MJ between capital and operating must be consistent both with attainable laser



(a) (b)

Fig. 7. (a) The specific laser energy requirement (MJ/kg_F) for the atomic vapor laser isotope separation (AVLIS) process defines acceptable laser energy cost goals. (b) A laser system cost of approximately \$10/MJ defines a surface in the parameter space of yearly operating cost (f) as a fraction of capital cost (C_L), and electrical efficiency (ϵ_L). Cost and performance tradeoffs are carried out to identify a viable laser system on this surface.

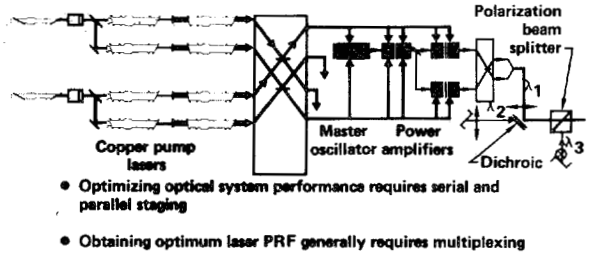


Fig. 8. Laser system architecture + process requirements = laser head requirements.

system lifetimes and efficiencies and with the particular market-acceptance requirements for capital expenditures we discussed above and in Fig. 3. Fig. 7(b) depicts the parameter space for total system capital cost per watt C_L, laser electrical efficiency ϵ_L, and yearly operating cost f, expressed as a fraction of capital cost. Laser cost/MJ may be expressed approximately as

$$(\$/\mathrm{MJ})_L \approx 0.011/\epsilon_L + (f + 0.163)\ .037\ C_L$$

where the factor 0.163 is for capital amortization over 10 years at 10 percent interest. Each point on the surface corresponds to our acceptable laser energy cost of \$10/MJ. The particular (not unreasonable) point indicated on the curve corresponds to \$6.66/MJ for operating cost and \$3.34/MJ for capital costs, certainly a favorable allocation (cf. Fig. 3), especially as compared with highly capital-intensive conventional enrichment methods.

Having scoped out the total laser system cost and power requirements and determined tradeoffs available between capital and operating expenses, we now indicate how to distribute these cost and performance parameters among laser-head, beam-transport, and beam-combination optics. Fig. 8 shows a representative AVLIS laser system design in which tradeoffs can be made to 1) optimize optical extraction efficiency versus laser-head economies of scale (Fig. 2), interstage losses, and beam-combination losses using series and parallel laser-chain staging, and 2) match laser-head PRF to process-chamber requirements by multiplexing. In an iterative design process, these tradeoffs are performed in order to minimize overall laser system costs and arrive at copper vapor laser (CVL) (our baseline pump laser) and dye laser technology development

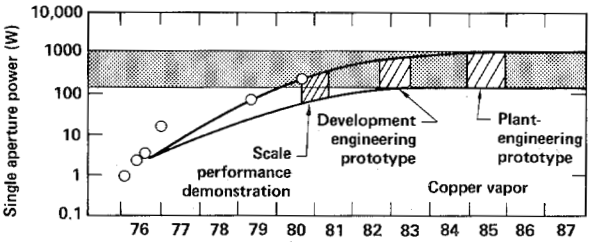


Fig. 9. Large bore CVL's have been demonstrated at plant scale represented by the horizontal dotted area.

goals. Fig. 8 shows the dye lasers pumped from both sides, since this optimizes beam quality. Following combination with the second- and third-step wavelengths (λ_2 and λ_3), the photons are transported to the process chamber with the correct energy fluence and PRF.

As mentioned above, our baseline AVLIS process consists of a three-step excitation to ionization. We are developing CVL's to pump dye lasers with frequencies tuned to each of these steps. As a result of the analysis procedure, we have established the horizontal dark band on the development schedule shown in Fig. 9 as the required single-aperture power for a commercial plant. The open circles denoting performances achieved at Livermore show that we have already demonstrated CVL's at plant scale. Future work will emphasize perfecting integrated operation and reliability, and reducing costs.

Three generations of CVL's have been developed at progressively larger scale (1-2 W, 10-20 W, and 100-200 W, respectively) at Livermore. The "Venus" laser facility consists of 32 CVL heads rated at 15 W that operate at 6 kHz PRF.[1] These lasers, based on 1977 technology, typically occur in six-head master-oscillator-power-amplifier (MOPA) units. The modular design CVL's are contained in a rigid space frame that allows each CVL head to slide out for servicing. The quick-disconnect features make it possible to repair or replace one CVL without interrupting the entire system operation. The electronics are also packaged in replaceable modules. By demonstrating integrated laser system operation we have achieved significant progress towards scaling up to the 100 kW level appropriate to a full-scale plant.

We made significant progress in scaling up the individual CVL heads when during the last year we achieved a major advance in laser technology with the first successful operation of a large-bore (7.5 cm diameter) longitudinal-discharge CVL. Fig. 10 shows the stainless-steel water-cooled vacuum housing surrounding the alumina plasma tube that comprises the CVL. We have obtained power levels appropriate to a full-scale plant with well over 100 W of extractable power at 5 kHz PRF. The achievement of high average powers at kHz PRF's is made possible by the discovery that volumetric deactivation of the lower metastable level between pulses is possible using high-pressure buffer gas, as reported by workers in the U.S.S.R. [3], [4] and in Israel [5]. Prior to this it was thought that deactivation of the metastables could only be obtained through diffusion to the walls. This had directed development to annular designs that had limited volume (and power) for a given

[1]We appreciate the help of General Electric Valley Forge Space Sciences Laboratory in the development of these copper lasers.

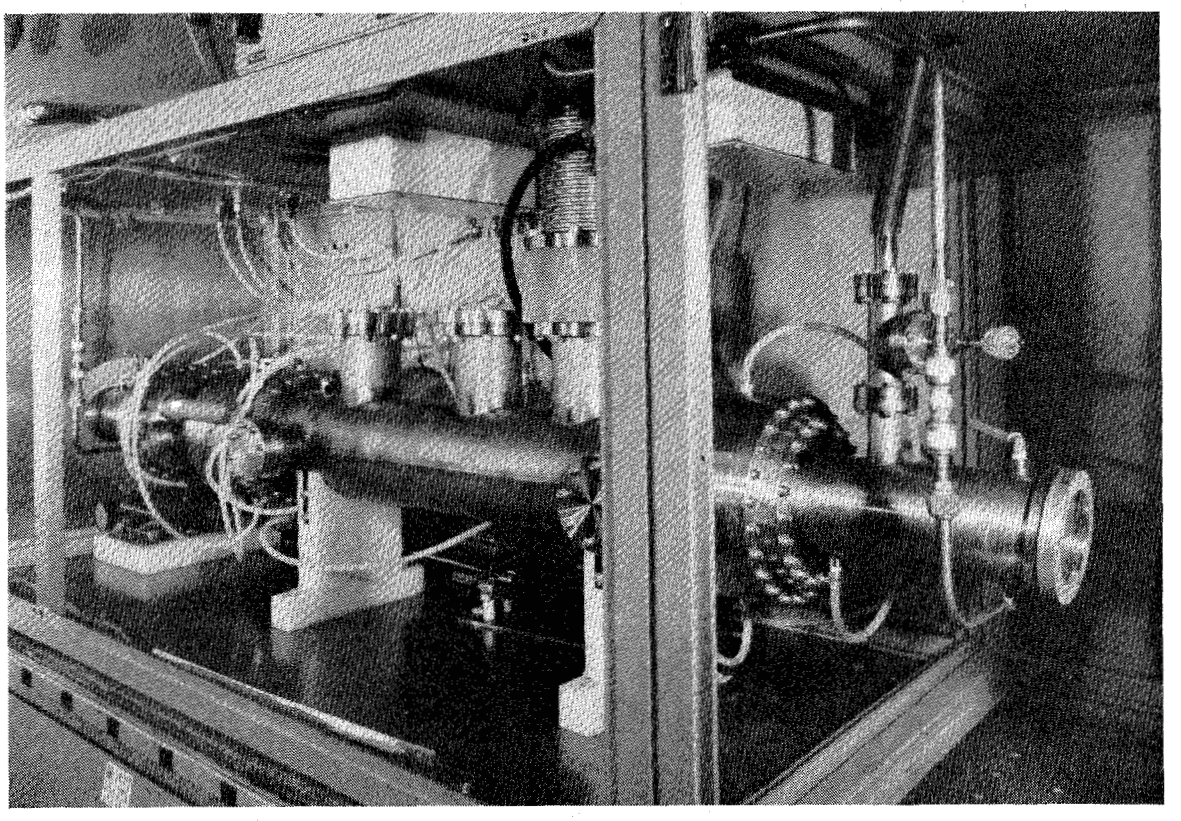


Fig. 10. The large bore CVL has achieved between 100–200 W of extractable power.

PRF. Along with obtaining high average power with large-bore CVL's, we are also able to avoid the optical difficulties inherent in transporting annular laser beams.

In both single-aperture power and in integrated operation, the feasibility of scaling CVL's to full-scale plant power requirements is essentially demonstrated. Copper laser life and reliability can be achieved with a systematic engineering and test program.

Given the flexibility in defining the process wavelengths afforded us by the many available transitions in atomic uranium, we have also been developing an advanced pump laser option. A two-step photoionization process (cf. Fig. 4) using XeCl laser-pumped dyes has been pursued with the development of the rare-gas halide (RGH) closed-cycle test bed (CCTB) laser shown in Fig. 11. This system comprises a flow loop that continuously circulates a rare-gas halogen mixture through the (rectangular cross section) discharge-excitation region. An electron gun is used for preionization, followed by the thyratron-triggered discharge. Operating the CCTB as a XeCl laser, we have achieved 70 W of average power at 850 Hz PRF. The high-power prototype (HPP), a scaled-up RGH laser system presently under construction, will be potentially capable of operating at power levels comparable to the "Venus" CVL system.

The dye laser system converts the fixed-frequency green-yellow CVL light (or the appropriate wavelength RGH light) into tunable radiation. A master oscillator feeds the desired frequency into a chain of highly saturated transversely pumped dye amplifiers, which we combine for transmission into the isotope-separation chamber. The dye fluid, flowing near vertically through the amplifier, passes through an active volume pumped by the green light from a CVL. Fig. 12 shows the layout being developed for this dye waveform generator (DWG). The CVL beam can be seen entering the DWG and pumping the dye oscillator and amplifiers.

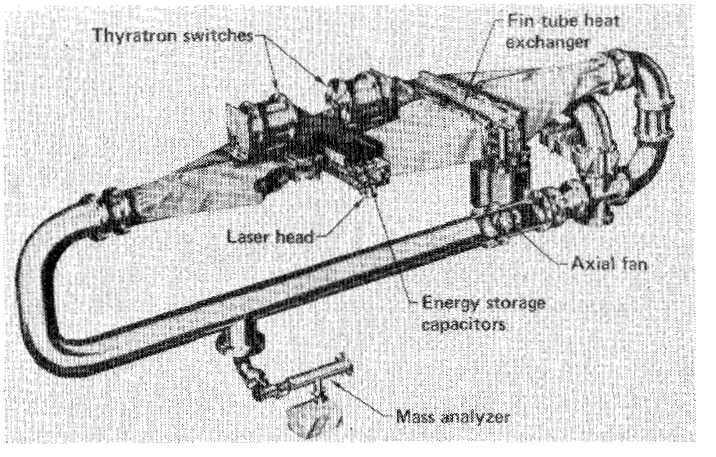


Fig. 11. Closed cycle test bed–RGH lasers.

Over the past year we developed an outstanding dye master oscillator that has significantly improved the dye laser system. The dye master oscillator defines the wavelengths precisely matching the appropriate ^{235}U transitions. Accomplishments in the dye laser area also include development of new dye compounds with improved stability and photon conversion efficiency, and demonstration of a significant portion of our efficiency and beam quality goals required for a plant. A recently developed dye amplifier has achieved improved beam

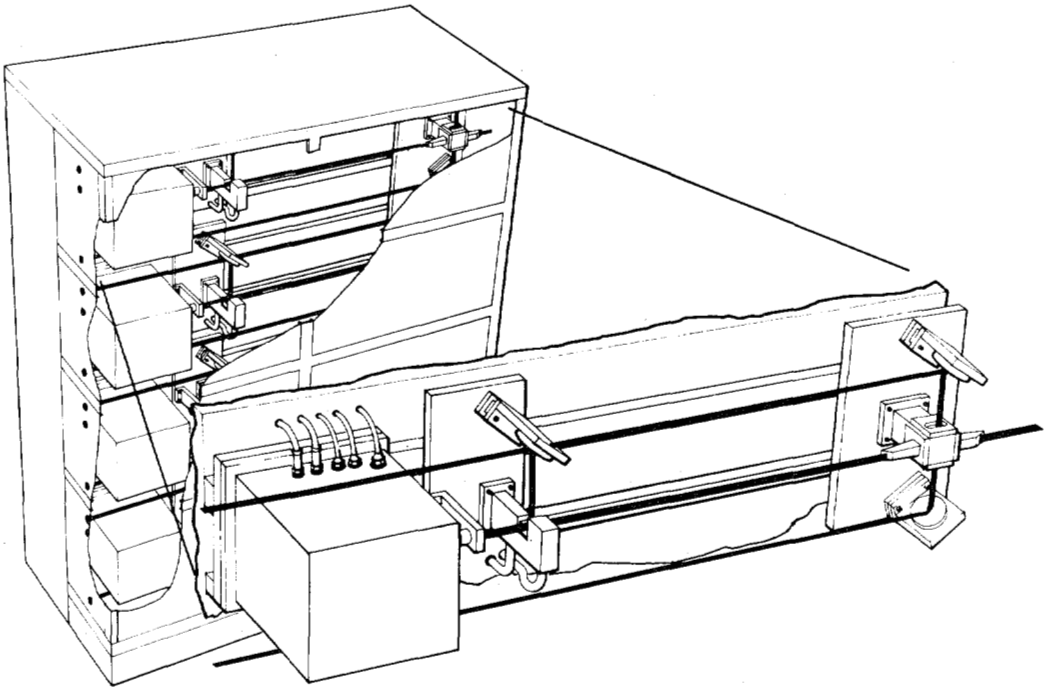


Fig. 12. Dye waveform module.

characteristics through engineering refinements such as suppression of internal reflections.

For the development of these lasers we have constructed an integrated system of enrichment facilities. A schematic of the systems in each facility is presented in Fig. 13. We have enriched macroscopic quantities of ^{235}U at progressively larger scales using the SPP-II lasers and the "Regulis" vaporizer. The "Venus" laser system enables us to illuminate a greater volume of the uranium vapor. The recently constructed "Mars" vaporizer and process chamber system, a significant step towards full-scale technology, is currently being brought on-line through integrated testing and evaluation. A factor of ten increase in vapor throughput will give us a great deal of developmental information at increased scale over the next several years.

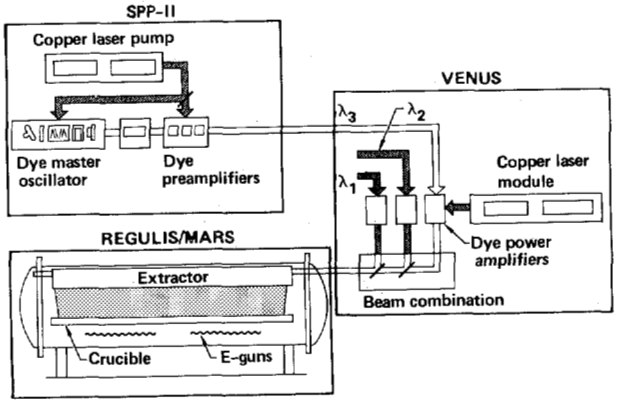


Fig. 13. Enrichment facilities schematic.

We have completed an intensive science and technology evaluation phase in which the process technology has been shown to be scalable to full production plant scale ($\sim 10^7$ kg/yr throughput and $\sim 10^5$ W laser power as in Fig. 6). While technology evaluation will continue on alternative options such as the RGH lasers, we are now in the technology demonstration phase in which AVLIS may be the advanced isotope separation process chosen by the DOE to be demonstrated at engineering scale in the late 1980's. Following a successful engineering demonstration, a full-scale isotope-separation plant could be in production in the 1990's.

The DOE is planning to select an advanced isotope-separation process in 1982 for engineering demonstration. If a laser process is chosen, we will have the first real opportunity to achieve large-scale application of lasers in a materials-processing application. Current plans include an advanced isotope separation facility (AISF) that will house a large-scale LIS system capable of demonstrating empirically the process performance we have projected. If this effort is fully funded in 1982, we would be able to verify full-scale process performance as early as 1985–1986. With realization of the projected savings in enrichment costs for LWR fuel, and the hundreds of millions of dollars per year of laser/EO business that could easily result, the acceptance and support of laser technology for a broader range of commercial applications should be considerably enhanced.

The future prospects for laser materials-processing applications may be expected to be improved by factors such as steadily improving laser performance, resulting in lower photon costs, and the identification of new high-photon-leverage processes as the result of continuing research in corporations, universities, and in the national laboratories. Finally, we should not entirely write off potential high-value material applications merely because of too small a market. With some reduction in laser costs we may yet find products with favorable price elasticities so that a reduction in price from an improved-efficiency laser process could result in significantly increased revenues.

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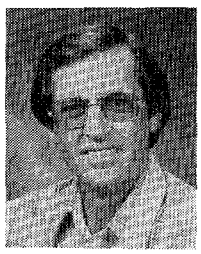

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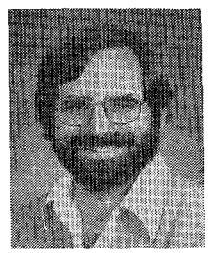

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The Stimulated Cerenkov Interaction and Its Applications

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Abstract—**Reviewed are the results of two experiments in which momentum modulation of a relativistic electron beam by laser fields using the stimulated Cerenkov interaction was measured, and coherent Cerenkov radiation from the optically bunched electron beam was observed. In the first experiment, light at 1.06 μm from a 30 MW Nd:YAG laser intersected 102 MeV electrons at an angle of 18 mrad in hydrogen gas, which was used as the phase-matching medium. The change in the electron-beam energy spectrum in the presence of the laser was measured, together with its functional dependence on the index of refraction of the phase-matching medium. In the second experiment, the same laser intersected 55.9 MeV electrons at an angle of 17 mrad, again in hydrogen gas. Coherent Cerenkov radiation at the second harmonic of the laser frequency (0.532 μm) was measured and indicated that the electrons were bunched on the order of the laser wavelength as a result of being velocity modulated by the laser. Applications of these results, such as development of optical klystrons and laser-driven particle accelerators, are considered. The characteristics of these devices and possible design configurations are discussed.**

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INTRODUCTION

THE success of the free-electron laser (FEL) developed by Madey and others [1] has generated a great deal of interest concerning the methods and problems of achieving momentum exchange between free electrons and laser light. Present efforts utilize a static magnetic field produced by a wiggler or undulator magnet system [2] to obtain wave-vector matching between the particles and photons [3]. We have demonstrated

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