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LASERS FOR ISOTOPE SEPARATION*

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Abstract

The Los Alamos Scientific Laboratory is conducting research on uranium enrichment. All processes being studied employ uranium molecules and use lasers to provide isotopic selectivity and enrichment. There are four well-defined infrared frequencies and two ultraviolet frequency bands of interest. The infrared frequencies are outside the range of the available lasers and an extensive research and development activity is currently underway. Lasers are available in the uv bands, however, much development work remains. The specification for the commercial uranium enrichment plant lasers will depend upon the results of the current enrichment experiments, the laser capital cost, reliability, and maintenance cost. For the processes under investigation there are specific photon requirements but latitude in how these requirements can be met. The final laser selections for the pilot plant need not be made until the mid-1980's. Between now and that time as extensive as possible a research and development effort will be maintained.

Introduction

The present energy crisis has focused attention on all of the energy sources available within the United States. Nuclear electrical power plants using enriched uranium as fuel for light water reactors represents a proven technology that can be expanded to reduce the use of natural gas. The enrichment of the natural uranium to 3% uranium 235 is presently done by the three government-owned gaseous diffusion plants. The gaseous diffusion enrichment step requires 19% of the energy output from the power plant.(1) The other steps in the fuel cycle: mining, milling, conversion, fuel fabrication, fuel and waste storage, amount to about 3% of the output energy. Thus, about one-fourth of the output from a nuclear power plant is used to generate fuel for the plant.

The uranium enrichment processes currently under investigation at the Los Alamos Scientific Laboratory (LASL) have the potential for greatly reducing the energy required for the enrichment step. From inhouse calculations, our enrichment processes will require only 1% of the power plant output energy, thus reducing the overall requirement to 4%. This lower energy usage would greatly enhance the already competitive position of nuclear plants, whem compared to other electrical energy generation schemes.

During the past four years, LASL has been engaged in research directed toward the enrichment of natural uranium to 3% uranium 235. The LASL approach employs uranium molecules and uses lasers to provide isotopic selectivity and enrichment. The details of the process are classified; however, during the past year articles have been published on certain unclassified aspects of the research.(2-5)

The near-term research on the LASL process is directed toward establishing the economic feasibility and preliminary engineering description of candidate processes by the end of fiscal 1978. The following phases of the work lead toward a mid-1980 preproduction plant. The laser requirement will progress from the small "bench-top" lasers to prototype production lasers during this period of time.

In this paper the laser needs for the research phase and for a commercial uranium enrichment plant are presented. The recent laser research and development are summarized and evaluated. The various options for the final laser requirements for a commercial plant are discussed.

Photon Requirements

For the LASL processes under investigation there is a need for coherent radiation at certain frequencies in order to perform enrichment experiments. One of the objectives of the near-term experiments will be to evaluate the effectiveness of the various frequencies for each process. The important point is to evaluate the photon characteristics and not necessarily a specific laser concept. The low energy and single-pulse-per-second

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requirements presented for the near-term needs are adequate to perform the physics experiments and at the same time minimize the laser development effort required. During the next two years LASL has a requirement for one-pulse-per-second lasers with the following characteristics:

Infrared Frequencies	Energy per Pulse
628.0 ± 1.0 cm^{-1}	0.1 - 1 mJ
823.0 ± 1.0 cm^{-1}	1 - 25 mJ
1160.0 ± 1.0 cm^{-1}	1 - 25 mJ
1294.0 ± 1.0 cm^{-1}	1 - 25 mJ ,

with a bandwidth of 0.03 to 0.07 cm^{-1} in the frequency ranges listed; and

Ultraviolet Bands	Energy Per Pulse
4000 - 3800 Å	150 - 500 mJ
3000 - 2350 Å	150 - 500 mJ ,

with a bandwidth less than 4 Å. For both the ir and uv lasers, there are certain specific frequencies within the ranges given that are more desirable. Various frequency shifting techniques, such as Doppler-shifting by rotating mirrors and Raman-shifting in hydrogen, for the ir and uv, are under investigation in order to extend the usefulness of a given laser concept.

For the 1978 research goals, at lease one ir and one uv laser, with increased pulse rate and energy per pulse, will be required. These lasers will be used in conjunction with scaled process equipment to establish the economic feasibility of the processes. This set of lasers will be the first set of at least two or three sets of large-scale lasers that will be evaluated and used in large-scale enrichment experiments during the 1978-81 time period. The exact frequencies of these lasers will depend on the small-scale experiments.

The pulse repetition rate required to do meaningful enrichment experiments must be greater than 100 pps and can be as high as 5000 pps. The latter is the pulse rate requirement for the final commercial uranium enrichment plant lasers. In describing these sets of lasers we have used a nominal number of 200 pps. This number was arrived at by considering the possible cost of the lasers and the relative ease of manufacture. Of course, consideration would be given to any lasers within this pulse repetition range; with the additional restriction that both the ir and uv lasers must have the same pulse rate.

Although there is a wide range for the pulse rate, the energy per pulse is much more limited. Beyond a certain level, additional energy is of minimal value. The energies desired are:

628 cm^{-1}	5 mJ
823, 1160, 1294 cm^{-1}	100 mJ
4000-3800, 3000-2350 Å	1-2 J ,

for the large-scale lasers. The energy required per pulse for commercial enrichment plant lasers is the same as listed above.

Current Laser Research

There are currently at LASL a total of fifteen experimental programs being pursued. Ten are involved with ir lasers and five with uv lasers. These programs involve a staff of twenty-one principal investigators. LASL and ERDA-funded contracts, in direct support of the program at industrial, university and government laboratories, make up another 29 programs, 19 of which are devoted to ir research, and the remaining ten to uv lasers. There are 26 principal investigators involved in this area.

Most of the current laser research has been reported in at least a preliminary manner in the open literature. The following discussion will be devoted to an enumeration of the laser research efforts. The principal investigator, his institution and the area of laser research are displayed in the following tables. Table 1 is devoted to the inhouse research and Table 2 to the industrial, university, and government contractors.

Table 1. LASL Laser Research

Topic	Principal Investigator and Work Report
1. Optical parametric oscillator, pumped by HF laser using cadmium selenide tunable between 14 and 18 microns	G. Arnold and R. Wenzel LASL/ERDA Conf. on LIS, Albuquerque 4/76 Applied Optics 15, 1322 (1976)
2. Optically-pumped ir lasers using HF excitation	M. Buchwald and C.R. Jones LASL/ERDA Conf. on LIS, Albuquerque 4/76 Applied Phys. Letts. to be pub. 9/1/76
3. Tunable ir diode lasers for spectroscopy	H. Flicker and N. Nereson LASL/ERDA Conf. on LIS, Albuquerque 4/76 J. Quantum Electron. 12, 326 (1976)
4. Raman shifting of 1.06 micron lasers in high pressure gases	T. Loree LASL/ERDA Conf. on LIS, Albuquerque 4/76 Optics Comm. 17, 160 (1976)
5. Infrared difference frequency mixer using CO and CO_2 laser, $CdGeAs_2$ mixer crystal	N. Barnes LASL/ERDA Conf. on LIS, Albuquerque 4/76 Opt. Comm. 15, 112 (1975); CLEOS, 1976
6. Advanced ir nonlinear optical materials, CdTe	M. Piltch LASL/ERDA Conf. on LIS, Albuquerque 4/76 J. Appl. Phys. to be pub. 8/76
7. Four-wave Raman mixing in gases	N. Kurnit and N. Levinos LASL/ERDA Conf. on LIS, Albuquerque 4/76
8. Electronic to vibrational energy transfer ir lasers	J. Telle LASL/ERDA Conf. on LIS, Albuquerque 4/76
9. Growth of nonlinear optical materials	R. Eckhardt LASL/ERDA Conf. on LIS, Albuquerque 4/76
10. Rare-gas halide and metal vapor uv lasers pumped by electric discharges	R. Sze, L. Blair, R. Begley and I. Bigio LASL/ERDA Conf. on LIS, Albuquerque 4/76 Appl. Phys. Letts. 28, 263 (1976)
11. Rare-gas halide lasers pumped by e-beams	P. Mace, J. Weinbrecht, W. Willis, G. Erickson LASL/ERDA Conf. on LIS, Albuquerque 4/76
12. Dye lasers for the uv	G. Balog, R. Butcher, and L. Sherman LASL/ERDA Conf. on LIS, Albuquerque 4/76

Table 2. Industrial and University Contractors

Installation	Topic	Principal Investigator and Where Reported
Industrial Contractors:		
Aerospace Corp.	Ultraviolet lasers, rare-gas halides and nitrogen lasers	S. Suchard; LASL/ERDA Conf. on LIS, Albuquerque 4/76 Appl. Phys. Letts. 28, 522 (1976)
GTE Sylvania	Infrared Raman scattering in crystals	E. Ammann; Appl. Phys. Letts. 27, 662 (1975)
Hughes Malibu	Infrared frequency conversion in gases	R. Abrams
Sandia Labs.	Rare-gas halide lasers pumped by e-beams	G. Tisone; LASL/ERDA Conf. on LIS, Albuquerque 4/76 Appl. Phys. Letts. 28, 538 (1976)

Table 2. (Contd.)

Installation	Topic	Principal Investigator and Where Reported
Mathematical Sciences NW	OCS and CS_2 lasers	S. Byron, LASL/ERDA Conf.on LIS, Albuq. 4/76; Appl.Phys.L.25, 517('74)
Laser Analytics	Tunable ir laser diodes for spectroscopy	K. Nill, LASL/ERDA Conf. on LIS, Albuquerque 4/76
University/Government Contractors:		
MIT Lincoln Laboratory	Infrared nonlinear optical materials	G. Isler
	Laser frequency measurements/ Laser frequency shifting	C. Freed
	Optically-pumped ir lasers	H. Kildal, T. Deutsch, R. Osgood, LASL/ERDA Conf. on LIS Albuquerque 4/76; Appl. Phys. Letts. 28, 342(1976)
Stanford	Infrared crystal growth laser	R. Feigelson
	Beam characterization	A. Siegman
	Wide range spectroscopic source using nonlinear optics	R. Byer; LASL/ERDA Conf. on LIS Albuquerque 4/76
USC	Electronic to vibrational (E-V) pumped ir lasers	C. Wittig, LASL/ERDA Conf. on LIS, Albuquerque 4/76; Appl. Phys. Letts. 27, 305 (1975)
MIT	High pressure ir lasers	A. Javan
	Tunable ir sources, uv CO laser	C. Dewey, LASL/ERDA Conf. on LIS, Albuquerque 4/76; Appl. Phys. Letts. 26, 442 (1975)
Univ. of Utah	Atomic nitrogen uv laser	G. Fowles, LASL/ERDA Conf. on LIS, Albuquerque 4/76
Univ. of Ill.	Raman shifting of ir lasers by liquids	P. Coleman, LASL/ERDA Conf. on LIS, Albuquerque 4/76
Univ. of NM	Scintillation materials as uv dye lasers	G. Daub, LASL/ERDA Conf. on LIS, Albuquerque 4/76
U. of Rochester	Ultraviolet laser coatings and damage studies	P. Baumeister, LASL/ERDA Conf. on LIS, Albuquerque 4/76
BYU	New uv laser dyes	J. Thorne, LASL/ERDA Conf. on LIS, Albuquerque 4/76
Texas Tech.	Infrared molecular lasers pumped by electric discharge	M. Gundersen, LASL/ERDA Conf. on LIS, Albuquerque 4/76; J. Quantum Electron. 12, 260 (1976)
Naval Res. Lab.	CS_2 laser - ir EDGDL 16 micron laser	F. O'Neill, T. Manuccia, LASL/ERDA Conf. on LIS, Albuquerque 4/76; Appl. Phys. Letts. 28, 539 (1976)
Stanford Research Institute	Infrared and uv frequency conversion	C. Rhodes, LASL/ERDA Conf. on LIS, Albuquerque 4/76

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Stanford	Infrared crystal growth laser	R. Feigelson
	Beam characterization	A. Siegman
	Wide range spectroscopic source using nonlinear optics	R. Byer; LASL/ERDA Conf. on LIS Albuquerque 4/76
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Naval Res. Lab.	CS_2 laser - ir EDGDL 16 micron laser	F. O'Neill, T. Manuccia, LASL/ERDA Conf. on LIS, Albuquerque 4/76; Appl. Phys. Letts. 28, 539 (1976)
Stanford Research Institute	Infrared and uv frequency conversion	C. Rhodes, LASL/ERDA Conf. on LIS, Albuquerque 4/76

In the tables we listed all the current laser research and development in support of the Program. The following will be an evaluation of the research on each of the six required wavelength regions presented in the previous section.

In the infrared at 628 cm^{-1} the required energy is in the 100 μJ range. It is currently being provided with the correct bandwidth by two devices operating at 1 pps. These are the CO - CO_2 nonlinear optical mixer and the HF-laser-pumped CdSe optical parametric oscillator. Although both devices are usable at the low pulse rates their reliance on solid nonlinear optical materials makes their scalability to high repetition rates doubtful. Lasers that use gaseous media with either optical or electric discharge pumping are clearly more favorable from a repetition rate scaling viewpoint. The devices under investigation are the HBr-laser-pumped CO_2 system at Lincoln Laboratory, the EDGDL at NRL, and the HF-laser-pumped CO_2 system at LASL. No engineering effort has been devoted to scale-up to this date, although there does not seem to be any major scientific barrier present. The primary future efforts must definitely be devoted to engineering.

The 823 cm^{-1} range around 12.2 microns wavelength has the following candidates for both near-term and intermediate-term systems. The high-pressure CS_2 e-beam laser seems the most probable candidate as it has already exhibited the required energy. The scalability of e-beam machines to high pulse-rates is considered uncertain, at best, because of foil heating problems and lifetimes of cold cathode emitters. The $^{14}CO_2$ laser can potentially be used and would make available all CO_2 technology. However, the large cost of the initial gas-fill and its radioactivity make this less than an ideal laser. A final candidate that makes use of the solid material InSb is the CO_2-laser-pumped Raman spin-flip laser.(6) This device can be operated at high pulse-rates by Q-switching the laser, and has the advantage of magnetic field tunability. It has not been developed as an engineering device and there is no effort underway to accomplish this at present.

The 1160 cm^{-1} region is covered by several interesting possibilities, and at least one demonstrated laser. The optically-pumped OCS lasing transition falls very close to the required wavelength. The disadvantage of this scheme is that it requires a high peak power frequency-doubled CO_2 laser. This makes use of a solid-state doubling crystal which severely limits achievable repetition rates. In the event that efficient CdTe plate doublers become feasible, this objection will no longer have to be considered. A system that has been demonstrated to produce usable 1160 cm^{-1} output is that using the process of noncollinear four-wave mixing of CO_2 lasers in germanium.(7,8) Although it is a solid material, Ge is characterized by low optical loss and relatively high damage threshold. This device, in addition, may be suitable for high repetition rate operation. Stimulated Raman scattering by a Nd:glass laser off high pressure hydrogen has been shown to be able to provide the correct wavelength and energy. It is limited, though, by the inability to operate glass systems at high repetition rates.

The situation at 1294 cm^{-1} is quite dismal in comparison to the previously mentioned wavelength regimes. High pressure CO lasers can be made to tune to this region. There has been virtually no research or development on this laser and it remains an area where very little in the way of usable devices is available.

In the uv regime, the region between 3800 and 4000 Å is best covered by flash lamp-pumped dye lasers. These have severe repetition rate limitations in addition to short dye lifetimes. The flash lamp lifetime is also problematic as they must be stressed severely to provide the necessary fast rise time excitation pulses. There is a definite need for gaseous lasers operating in this wavelength regime.

The 2350-3000 Å region is fairly well covered by the rare-gas halogen lasers, such as KrF and XeBr. The KrF transition at 2484 Å has been made to oscillate in an electric discharge-pumped system and is therefore capable of repetition rate scaling. Further wavelengths are attainable by using this laser to pump dyes. The ArF laser at 1950 Å(9) can also be used in this mode, thus making fairly dense coverage possible.

This review has tried to emphasize the many possibilities that exist for particular laser devices, while providing the caution that little or no engineering development has been accomplished on any of the systems.

Projections

With the above as an evaluation of the present status of laser and laser-component research and development, a projection is now made into the future. For this projection we assume that a LASL molecular uranium enrichment process has proven to be successful and that a process has been selected for one of the nation's uranium enrichment plants. The current physics experiments on the processes and those scheduled during the next two years will determine which of the following scenarios is correct:

single-stage plant
or
multi-stage plant.

From all our work to date, no physical phenomenon have been discovered that preclude the possibility of a single enrichment stage, capable of producing 3% enriched uranium 235 with 0.02% tails from a feed of natural 0.7% uranium 235. A single-stage plant, or a plant with two or three stages would be the most desirable; however, as a hedge against some unforeween deleterious phonomenon in the process, a multi-stage plant consisting of 25 stages will also be considered in this paper. As might be expected, the number of stages required in the production plant will have an impact on the extent of the laser development activity.

The typical approach to any commercial continuous feed, large quantity process plant is to design it for 24-hour, seven-day-a-week operation. The reasoning behind this usually is that the plants are capital intensive and must run continuously in order to recover the initial investment. A single-stage uranium enrichment plant using a LASL process will require much less capital than a gaseous diffusion plant; our estimate is $130 million, excluding the support facilities, as compared to $2 or $3 billion for a gaseous diffusion plant. Each of these plants would generate 9×10^6 SWU and produce the same quantity of enriched uranium. The impact of this capital cost differential is that a single stage plant using such processes need not operate continuously. Significant periods of time could be allowed for equipment maintenance. An operating schedule that would allow for eight hours of maintenance per day would impose less strenuous performance reliability requirements on equipment than one that required 24-hour operation. Thus, the results of the present enrichment physics experiments which determine the effectiveness of the separation process can have a significant impact on the process plant laser performance requirements.

The photon requirements enumerated in this paper for the process plant can be produced by several approaches. One approach is for a single ir and a single uv laser -- each operating at 5000 pps per stage. There are two apparent disadvantages to this approach. The first is in the increased development time and cost required to achieve the required performance, since in some subcomponent areas a significant advance in today's state-of-the-art is required. A second disadvantage is that associated with a failure or off-design performance of the laser. In either event, the effect might necessitate plant shut-down for repairs. A situation such as this would certainly have an impact on the economics of the plant operation, especially in the case of a plant that was required by cost and production considerations to operate 24 hours a day. An advantage of a single laser could be in reduced capital cost, beter "packaging" of the stage equipment, and fewer operating personnel.

A second approach is based upon a set of lasers that in total produce 5000 pps. As an example, this could be twenty multiplexed lasers operating at 250 pps each. (The energy per pulse must be the same for every laser pulse.) Two advantages are: 1) feasibility of manufacture using components and techniques more closely within the stage-of-the-art; and 2) reduced economic impact on plant operations if one laser fails for a period of time during plant operation. The disadvantages are: 1) the total capital cost of twenty lasers with spares could be greater than that of a single laser; 2) possible increased maintenance cost; 3) increased timing and jitter problems; and 4) the more difficult problem of "packaging" and integrating forty lasers around the stage equipment.

In these two situations a very wide range of requirements has been presented. The first plant was based upon one stage operating only 16 hours a day, using one ir and one uv laser at 5000 pps, producing the equivalent of 6×10^6 SWU/yr. The second was a plant operating 24 hours per day using 500 ir and 500 uv lasers and producing the equivalent of 9×10^6 SWU/yr. The physics of the process will determine the number of stages requires. The capital and operating cost of the various laser alternatives will establish whether or not single or multiple lasers are best for each stage. The general trend for the cost of lasers is a decrease in terms of dollars per watt as the average power increases. If this is also true of the plant lasers used in the uranium enrichment plants, then the cost of the twenty lasers per stage would be higher than that of a single laser. The initial capital cost for the lasers will also influence the desired lifetime of the lasers. The possibility exists that in a single stage plant using a single ir and uv laser, new and improved lasers could be installed as they became available. This technique would be less likely for the 500-laser situation.

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Conclusions

The present and near-term laser research and development is being driven by the physics experiment requirements. Once lasers have been provided to do the experiments and the relative impact of the various operational parameters have been determined the final configuration of the production plant will be established by optimizing all variables. This optimization includes the lasers. The developmental cost, purchase price, reliability, and lifetime of the various lasers at all the acceptable frequencies must be considered. The possibilities are numerous and in all likelihood an early clear winner will not be established. We anticipate a long and active period of laser research and development extending into the 1980's before any definite decisions are made on the lasers to be used in the first production plants. There is no reason to preclude the possibility that additional plants could use different laser systems; thus, there is a high probability of laser research and development extending into the 1990's.

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