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A STATUS OF PROGRESS FOR THE LASER ISOTOPE SEPARATION (LIS) PROCESS

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16. ABSTRACT

Since the earlier days of the patent by the Israeli scientists (Nebenzahl and Leven, 1973), a virtual explosion of information on Laser Isotope Separation (LIS) has occurred. Research is apparently going on in several European countries and particularly in Russia. References vary from German patents to the Soviet Journal of Quantum Electronics, the American Science Journal, and then finally to the Science Fiction magazine, Analog.

An overview of the various categories of the LIS methodology is given together with illustrations showing a simplified version of the LIS technique, an example of the two-photon photoionization category, and a diagram depicting how the energy levels of various isotope influence the LIS process.

Applications have been proposed for the LIS system which, in addition to the use to enrich uranium, could in themselves develop into programs of tremendous scope and breadth. Such applications as treatment of radioactive wastes from light-water nuclear reactors, enriching the deuterium isotope to make heavy-water, and enriching the light isotopes of such elements as titanium for aerospace weight-reducing programs.

Economic comparisons of the LIS methodology with the current method of gaseous diffusion indicate an overwhelming advantage; the laser process promises to be 1000 times more efficient. The technique could also be utilized in chemical reactions with the tuned laser serving as a universal catalyst to determine the speed and direction of a chemical reaction.

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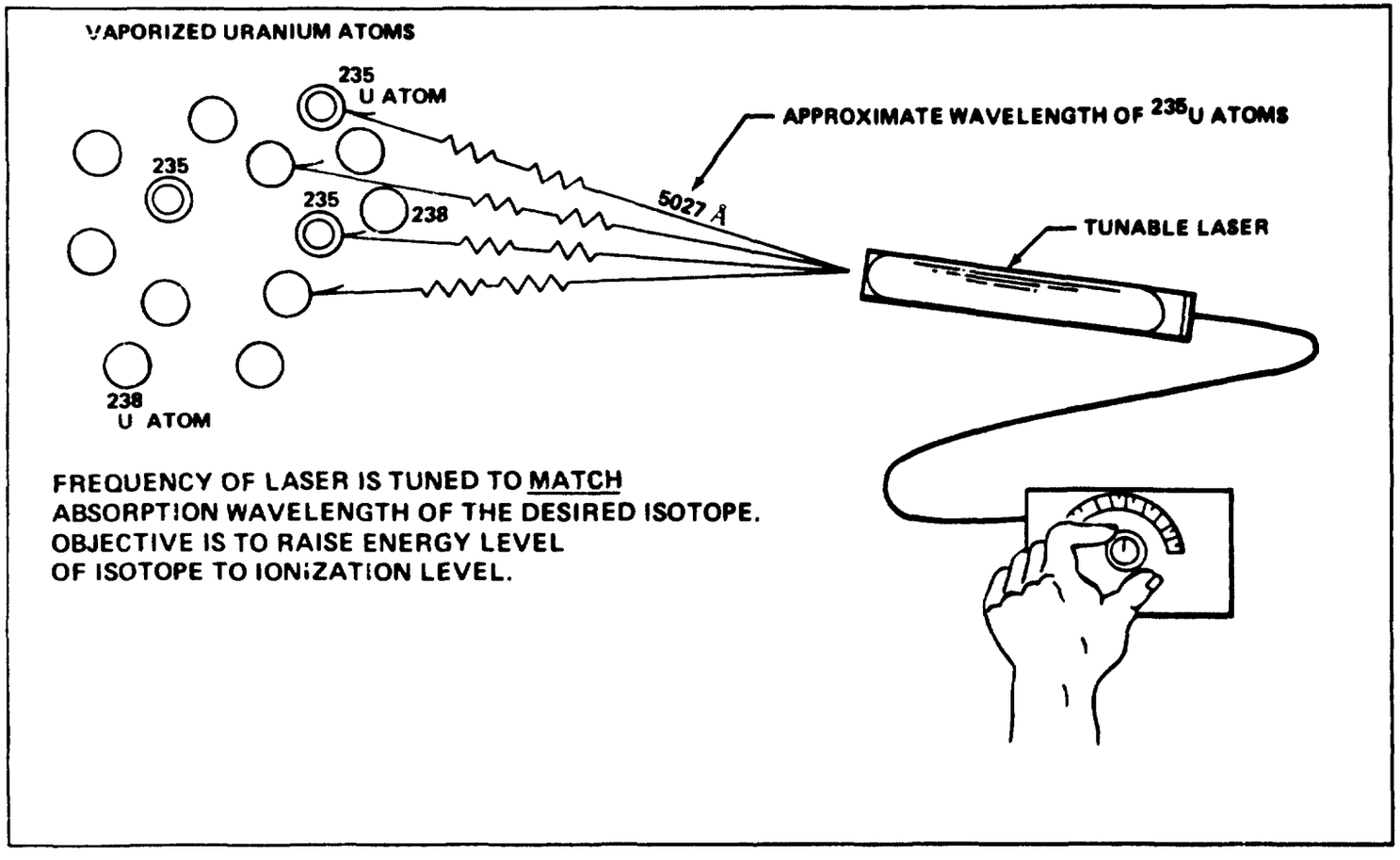
PREFACE

Since the publication of the first Technical Memorandum (TM X-64947) on the Laser Isotope Separation (LIS) Process in May 1975 [1], there has been a virtual explosion of available information on this process. Articles have been published in a widely varying degree of sophistication, ranging from the recent article in the popular science fiction magazine Analog to the prestigious Science Journal, all of which contain information on the LIS process. As a matter of fact, the LIS process is not a single methodology — but a series of techniques, each of which has its own unique set of critical parameters. Discussion will be primarily from an engineering point of view, although it is recognized that the field lies heavily in the discipline specialty of optical physics. An attempt will be made to simplify the technical descriptions and to present the information from the viewpoint of people with a diverse background of specialties.

The trade journal, Laser Focus, is to be commended for its role in serving as the source for all late technical information on the evergrowing list of laser applications.



SIMPLIFIED SCHEMATIC DIAGRAM OF THE LIS PROCESS

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INTRODUCTION

Apparently due to a change in the classification policy [2], there has been a noticeable change in the details concerning Laser Isotope Separation (LIS), technical and economic. Surprisingly, the principle of "detente" was never more accurately applied than in the Soviet Journal of Quantum Electronics [3]. The cost values of isotope enrichment by the gaseous diffusion process are released for public utilization. At one time this value ($ 5 per gram of U-235) was classified top secret, as were cost values pertaining to the consumption of electrical energy, capital costs of gaseous diffusion plants, and maintenance costs for the diffusion plant cascade.

Also, the classification or categorization of the several methodologies for LIS has shown a noticeable progress during the past 18 months [4]. The various subcategories of the LIS process are outlined approximately as follows:

Laser Isotope Separation:

a) Single-Photon Photoionization

b) Two-Photon Photoionization

c) Three-Photon Photoionization

d) Two-Step Photodissociation

e) Two-Step Photochemistry

f) Raman-Scattering Process

g) Autoionization

h) Others.

Of these several techniques, only those represented by b), c), and f) have indicated significant progress in the information which has been released up to the present time. Of the several processes outlined here, some are based on atomic unit processing while others are based on the separation of molecular units, e.g., UF_6 or P_UF_6. The efforts at the Los Alamos Scientific Laboratory have been concentrated on the molecular system of isotope separation, whereas the activities at Lawrence Livermore Laboratory (also Exxon, Avco, etc.) mainly utilize the atomic methodology in their laser separation schemes. The main point here is the question concerning the accumulation of technology related to the chemical processing of uranium to yield the desired molecular form of UF_6 versus the relative difficulty of processing uranium in the metallic state (atomic). Vast facilities already exist at Oak Ridge to process the U_3O_8 into the vaporizable UF_6 [5] and also to reduce the enriched $^{235}UF_6$ back to the parent metal; therefore, this part of the nuclear fuel cycle is considered well in hand.

Also, still to be reckoned with in the isotope separation sweepstakes is the category of processes for enrichment of isotopes by the method of photochemistry. Several groups have participated in this specialty [6,7,8]. Much promise is held for this approach which naturally falls into the molecular category, and applications to other areas of chemistry (other than nuclear) are virtually unlimited.

DISCUSSION OF LIS METHODOLOGY

Overall progress in application of the laser to separate isotopes has been excellent in the past 18 months. Much of this progress has been reported by the teams at Los Alamos and at Lawrence Livermore [7]. Laboratories in foreign countries have also made significant contributions [3,9,10].

The technique whereby a particular atom, e.g., ^{235}U, is selectively energized by the application of a laser tuned to the specific characteristic bandwidth of the isotope in question has become a principle subject of discussion. The application of the laser tuned to a discrete bandwidth at a predetermined power level and at a suitable pulse rate has become the standard approach.

Many variations in this approach have been reported and others are being added. Inherent in this LIS approach is the choice of a feed material. Basically. this is the choice between UF_6 or the use of metallic or atomic uranium. Many prefer the UF_6 form because this is the only compound of uranium that can be rendered in the gaseous form at a slightly elevated temperature (60°C). As was mentioned previously, facilities already exist to process the uranium into the UF_6 form and to reduce it to the metallic form if desired. The vast gaseous diffusion process is based on the UF_6 form of uranium feed. If a process in LIS based on a UF_6 feed material could be accomplished then that part of the nuclear fuel cycle would be simplified [5,11].

The approach by Lawrence Livermore Lab, Exxon, and Avco-Everette has by contrast been based upon using the atomic uranium as the feed material with the uranium in a metallic state being heated in an oven to the molten state (Fig. 1) [12]. A similar approach was used by Israel as described in a patent disclosure [9]. According to this patent disclosure, 7 gm of ^{235}U (at a purity level of 60 percent) were separated in a 24 h period. Due to the very corrosive characteristic of molten uranium (2000 K), it is necessary to have the atmosphere for the oven in a rare gas atmosphere, e.g. argon. In spite of the problem with the corrosive nature of molten uranium, there is at least one significant advantage to the use of atomic uranium as the feed material — the characteristic laser wavelengths are much better defined or distinct for the atomic uranium. Each isotope of uranium, or any other element, has a distinct wavelength or bandwidth for each of its isotopes. The energy level of a chosen isotope may be increased by matching the frequency or bandwidth of a suitable laser to the characteristic wavelength of the isotope. The energy level of a particular isotope may be increased by application of a suitable laser until a point is reached where the continuum is realized — at this point, the isotope in question emits an electron and becomes an ion. As illustrated in Figure 1, the ions are collected by the use of a negatively charged plate. Also, as shown in Figure 2, the atoms of uranium may be brought up to the continuum level in step-wise fashion by either two, three, or more lasers tuned to the appropriate bandwidth. As pointed out in the "Laser Focus" [2], the LIS could be accomplished with "a single highpower pulsed laser, tuned to one isotopic transition" [13]. The mode of actual recovery of the product will depend upon whether the isotope is being separated in atomic or molecular form.

In the photodissociation process, the significant action is the physics of selectively breaking chemical bonds in a two-step process. The first reported application of two-step photodissociation to isotope enrichment was the separation of nitrogen isotopes ^{14}N and ^{15}N [4]. Other molecular examples have also been reported [2,4,6,11].

LASER ISOTOPE SEPARATION (LIS)

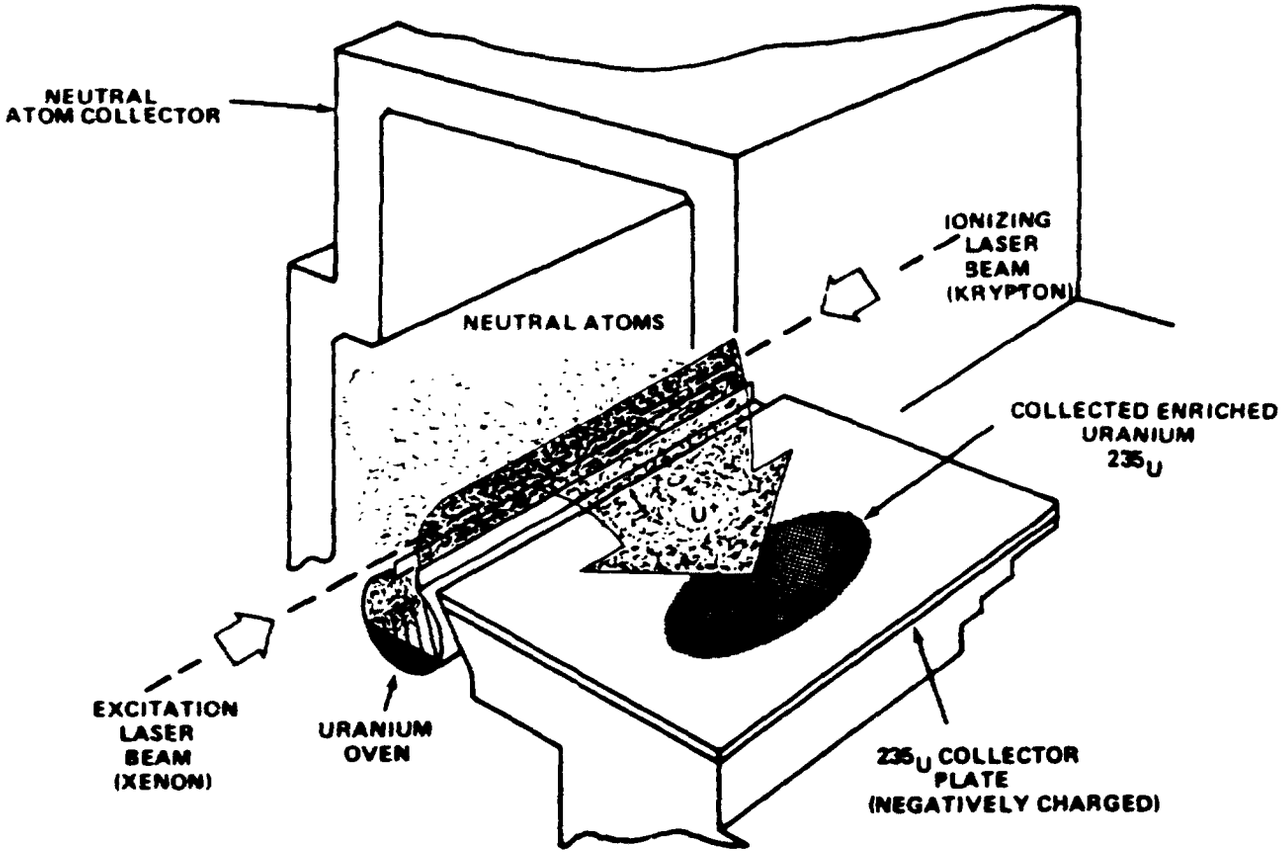

Figure 1. Two-photon photoionization.

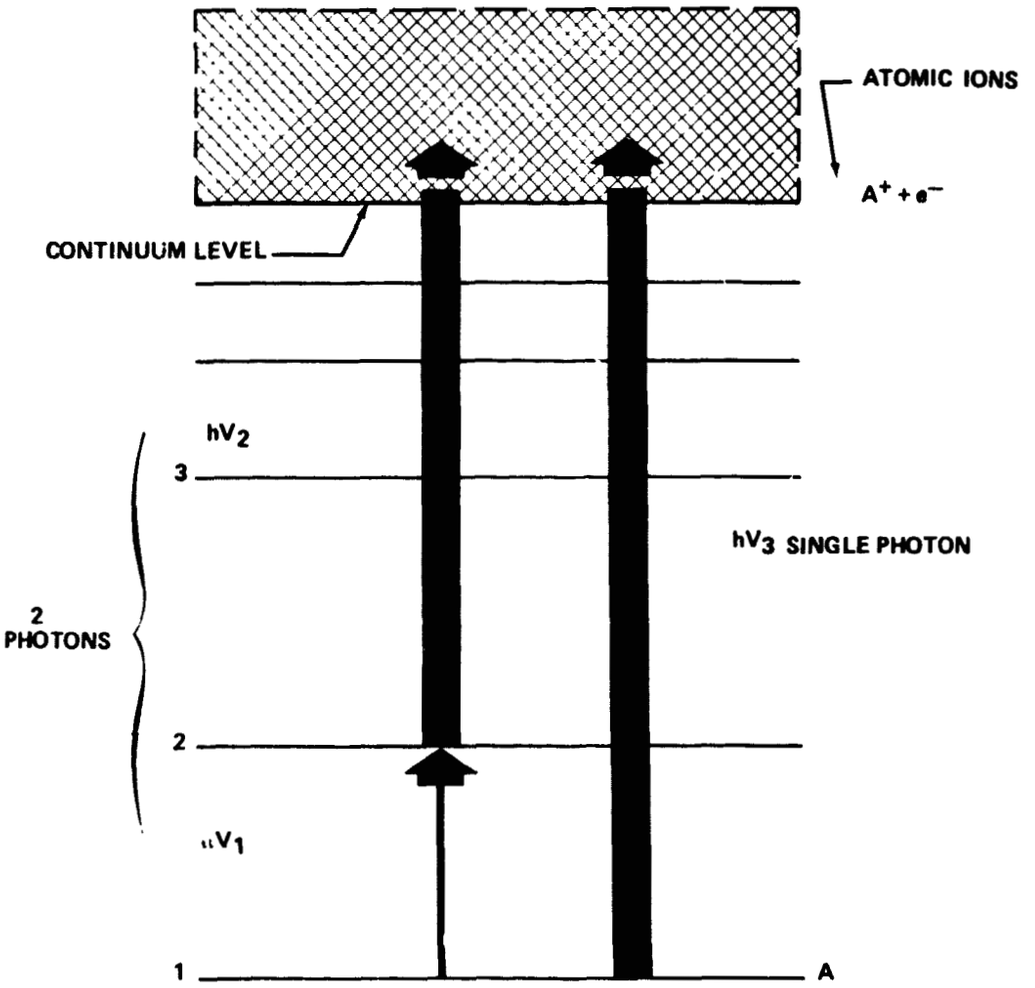

Figure 2. Energy level diagram.

In the selective photochemical process, the critical activity [8] is the influence of the laser on the chemical reactivity of a particular element during a chemical reaction. Several isotopes have been reported enriched during chemical reactions which involved enhancement by lasers (^{15}N, ^{35}Cl, ^{37}Cl).

In the autoionization scheme, the process relies upon the greater cross section for autoionization (by the isotopes) than for conventional photoionization. Consequently, a considerably weaker laser can be used for the final ionization step. This method is considered a variant of the photoionization technique [9].

In the Raman-scattering mode of laser isotope separation, the process depends upon an amplification process in which the simultaneous irradiation by a dye laser is tuned to the difference between the pump frequency and the frequency of the Raman-scattering component of the selected isotope.

In the final analysis, the "isotope separation sweepstakes" break down into two different camps. The advantages and disadvantages of each approach (as previously outlined) indicate that considerable development and some additional research will be needed before the process of LIS can be reduced to the pilot plant stage by specific methodology [11]. For the atomic approach, there are the problems of the corrosiveness of high temperature uranium vapor and the tendency of ionized ^{235}U atoms to recombine. In the case of the molecular approach, there are several problems: 1) the energy states of the molecular form UF_6 are close together, tending to overlap, so that it is almost impossible to find a wavelength at which $^{235}UF_6$ will absorb light (from a laser) but $^{238}UF_6$ does not; 2) few of the UF_6 molecules will be found in the "ground state" (a necessary condition for laser excitation); and 3) perhaps the most important drawback to the molecular method is the lack of commercially available lasers with sufficient power and at the desired wavelength to do the job.

The Los Alamos group has reported successful separation of isotopes of SF_6 by the LIS system, but for UF_6 proof of the methodology must await development of a more powerful laser with the desired bandwidth [11].

Recent reports have placed the desired wavelength for enrichment of uranium as approximately 16.1 μm (for UF_6) [14]. It has been reported that more precise specification of the wavelength beyond three digits is classified (e.g., 16.13483).

OTHER TECHNIQUES FOR SEPARATION OF ISOTOPES

Although considerable disclosures have been made, certain classified facts related to the isotope separation still remain. Naturally, only the information based on published reports can be discussed.

The mainstay of the American nuclear industry has been and continues to be the gaseous diffusion (also for the French). There have been several challenges to this process which are described in general in Reference 15

and in detail in Reference 5. The main advantage is that it is a proven technology and has been for over 25 years. The disadvantages are: a) capital investment cost, b) power consumption cost, c) land usage, and d) maintenance costs. A recent contender for the uranium enrichment business has been the centrifuge which has been used by a European consortium and by the Japanese. The centrifuge relies upon certain physical properties of the isotope to effect the separation. The process, though still under development in the United States (and is largely classified), does show considerable promise with enrichment factors in the range of 1.5 to 2.0, as compared with 1.004 for gaseous diffusion [11]. The power consumption for an equivalent amount of product would also be reduced considerably.

There is also another process, about which little is known, in Germany. This is based upon the passage of a gas (UF_6) over an airfoil by a nozzle. The reported disadvantage of this process is the excessive consumption of power. South Africa and Brazil are reportedly utilizing the technology of this isotope separation methodology.

PARTIAL LISTING OF APPLICATIONS OF THE LIS PROCESS

The principal candidate for application of the LIS has been to separate ^{235}U from natural uranium. This methodology promises to be 1000 times more efficient than the current technique of gaseous diffusion [15]. The enrichment of the ^{235}U isotope from its naturally occurring percentage of 0.711 percent to the 2 to 4 percent level is currently important because of the ever increasing demand as fuel elements for the light-water reactor.

Also potentially important is the application to enrich deuterium. The naturally occurring concentration of deuterium in water is only 0.015 percent, too low for a technique based on mass difference to work efficiently. Applications for heavy-water are well known, but there is, for example, the Canadian derived CANDU reactor (a heavy-water type) which uses heavy-water and also the well known application in the fusion process.

Treatment of nuclear waste materials was used to separate the isotopes of plutonium PU-238 and PU-239. Also, the laser could be very useful in the separation of certain actinides from the waste materials from reactor systems

to make the storage of those waste materials and subsequent handling easier. Since many of the actinides are so close together in the periodic table, separation by chemical means becomes very difficult. An alternative for this application is not known at the present time.

Other less important uses are as follows:

a) Separation of ^{10}B to use in light-water reactor control rods.

b) Separation of ^{6}Li for use in the fusion reactor system.

c) Savings in weight by separation of the light isotopes of, e.g., Titanium for aerospace application. Light isotopes could also be useful for applications where weight of a moving part limits its performance, e.g., turbine blades and rotors.

Since the gaseous diffusion method leaves a significant portion of the ^{235}U isotope ($\approx$ 40 percent) unseparated, a final consideration for application is that it is possible to remove practically all of the selected isotope with the laser methods. This fact alone will result in approximately a 67 percent increase of our effective uranium reserves [15]. The tails from the gaseous diffusion plants could all be worked to remove the remaining radioactive portion.

ECONOMIC COMPARISONS

One might ask "with a proven technology that has been operating for over 25 years," why take the chance with one more way to enrich uranium? This would be especially true when you consider that for power plants, the operating costs are already cheaper than for conventional fossil fuels. The reasons are rather obvious when you consider that the overall investment costs are roughly 30 times higher for the present method, which is gaseous diffusion. It would only be fair to say that these statistics are based on the currently reported information and are in the nature of projections because there is no LIS plant to compare directly with the gaseous diffusion plants located in Oak Ridge (Tennessee), Paducah (Kentucky), and Portsmouth (Ohio). The Table is, however, based on the best information available at the present time and is assumed on the basis of 1976 dollars [15].

TABLE. COST COMPARISON OF GASEOUS DIFFUSION VERSUS LIS ENRICHMENT PLANTS

Assumed Capacity = 8.75×10^6 Separative Work Units/Year, $ = Billions of 1976 Dollars

Capital Investment	Gaseous Diffusion	LIS
Enrichment Plant	$3.1	$0.130
Power Plant	1.4	0.013
Total Capital	$4.5	$0.143
Operating Costs		
Electric Power	$0.426	$0.004
Other	0.036	0.016
Total Operating Costs	$0.462	$0.020
Land Use	≈ 90 acres	≈ 1 acre

Note: The gaseous diffusion plant must also be located close to a river or some other source of cooling water. An LIS plant could presumably be located at any geographic location.

So far as enrichment of uranium is concerned, the LIS is the choice by a wide margin on most points of comparison — capital investment, operating costs, land use, or even choice of the plant site to be close to a river. The cost comparisons with the centrifuge process would be less impressive — but since the data are still classified, none can be made.

Cost comparisons concerning the application to treatment of nuclear wastes cannot be made because there is no known competitive system. The cost of deuterium enrichment is still unavailable, so no comparison can be made.

Based on current projections and assuming currently planned use of nuclear power plants, the demand for enriched uranium will begin to exceed supply sometime between 1980-1983. A decision must be made soon as to the type of enrichment plants that must be built and whether it will be financed by the United States Government or private enterprise.

CONCLUSIONS AND RECOMMENDATIONS

As has been outlined, the LIS process is really a group of related techniques rather than a single method. The inherent simplicity of the approach is the thing that attracts much attention. Also, the economic question is perhaps the aspect of the methodology that attracts the most interest. Millions and perhaps even billions of dollars might be saved by application of the LIS process to the uranium reactor fuel system alone.

The question of whether the molecular or the atomic form of separation will prove to be most practical must be answered. There are several disadvantages as well as advantages inherent to each of the forms of feed material. Also, there remains the problem of scaling the laboratory-sized operations up to the pilot plant size and then industrial plant size.

Other applications of the LIS process to such approaches as treatment of nuclear wastes to effect a separation of actinides, etc., and to enrich deuterium for heavy-water applications show exceptional promise. The treatment of the tails from the Oak Ridge gaseous diffusion appears to be a simple way to extend the volume of our uranium reserves.

In the area of recommendations, the laser part of the system appears to be the way to progress in most all of the categories which have been previously detailed. It would therefore appear frugal to concentrate development efforts on the laser itself. In such efforts there are two principal laser hardware parameters which should receive attention:

a) Tunability of the laser

b) Power levels.

The savings in uranium enrichment can be very important by application of the LIS process; however, laser-influenced chemistry holds forth a much broader application range because laser-induced chemistry can be applied to all the elements of nature and to complex molecules as well. The laser would in effect become a "super-catalyst," to influence the speed of chemical reactions, etc.

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APPROVAL

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The information in this report has been reviewed for security classification. Review of any information concerning Department of Defense or Atomic Energy Commission programs has been made by the MSFC Security Classification Officer. This report, in its entirety, has been determined to be unclassified.

This document has also been reviewed and approved for technical accuracy.

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