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Laser Isotope Enrichment for Medical and Industrial Applications

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ABSTRACT

The principal isotope enrichment business in the world is the enrichment of uranium for commercial power reactor fuels. However, there are a number of other needs for separated isotopes. Some examples are: 1) Pure isotopic targets for irradiation to produce medical radioisotopes. 2) Pure isotopes for semiconductors. 3) Low neutron capture isotopes for various uses in nuclear reactors. 4) Isotopes for industrial tracer/identification applications.

Examples of interest to medicine are non-radioactive targets such as S-33, Mo-98, Mo-100, W-186, Sn-112 to produce radio-isotopes; while for MRI diagnostics, the natural Xe-129 isotope is wanted. For super-semiconductor applications desired industrial isotopes are Si-28, Ga-69, Ge-74, Se-80, Te-128, etc. An example of a low cross section isotope for use in reactors is zinc depleted of the Zn-64 isotope. Depleted zinc is used as a corrosion inhibitor in nuclear reactor primary coolant systems. Neutron activation of Ar isotopes is of interest in industrial tracer and diagnostic applications (e.g. oil-logging).

In the past few years there has been a sufficient supply of isotopes in common demand, because of huge Russian stockpiles produced with old electromagnetic and centrifuge separators previously used for uranium enrichment. Production of specialized isotopes in the USA has been largely accomplished using old “calutrons” (electromagnetic separators) at Oak Ridge National Laboratory. These isotope separation methods are rather energy inefficient.

Use of lasers for isotope separation has been considered for many decades. None of the proposed methods have attained sufficient proof of principal status to be economically attractive to pursue commercially. Some of the authors have succeeded in separating sulfur isotopes using a rather new and different method, know as condensation repression. In this scheme, a gas of the selected isotopes for enrichment, is irradiated with a laser at a particular wavelength that would excite only one of the isotopes. The entire gas is subjected to low temperatures sufficient to cause condensation on a cold surface or coagulation in the gas. Those molecules in the gas that the laser excited are not as likely to condense or dimerize (coagulate into a double molecule, called a dimer) as unexcited molecules. Hence in cold-wall condensation, gas drawn out of the system is enriched in the isotope that was laser-excited.

We have evaluated the relative energy required in this process if applied on a commercial scale. We estimate the energy required for laser isotope enrichment is about 30% of that required in centrifuge separations, and 2% of that required by use of "calutrons".

INTRODUCTION.

Nuclear medicine and industrial isotope users are relying on a growing number of enriched natural isotopes. Some nuclei of these isotopes can be transmuted by spallation reactions induced by particle accelerators or by neutron absorptions produced in research reactors, to yield valuable radioactive species for medical and industrial applications. Disruption in the supply of enriched isotope sources, which are currently provided by surplussed Russian and European ultra-centrifuges (UCF) or electromagnetic calutrons (EMC) at Oak Ridge, could seriously jeopardize such nuclear applications. It is therefore important that new laser isotope separation (LIS) techniques be developed to insure a secure supply of source isotopes for these important medical and industrial disciplines. Prices of isotopes produced by converted UCF or EMC plants, used earlier for Uranium enrichment operations, are rather high because of high energy consumptions. For example $^{33}SF_6$ enriched to 99% in a UCF presently sells for about $16,000 per gram of S-33. LIS schemes forecast ten-fold lower costs. Table 1 lists some isotopes used in modern medicine that require enrichment.

One of the main reasons for a need of pure isotopes in medical applications is to minimize the natural contamination in radioisotopes used in nuclear medicine. For instance, the most common nuclear medicine isotope is Tc-99, derived from radioactive Mo-99 shipped to hospitals weekly in a “cow.” The Mo-99 (half life 2.75 days) decays to Tc-99 (half life 6 hours), which is eluted chemically from the “cow” for injection into the patient. The elution is never perfectly free of contamination from some of the parent material (Mo), and hence the need to keep the quantity of Mo to a minimum. Current techniques for producing the Mo are by separating it from the fuel and fission products irradiated in a nuclear reactor. The chemical process cannot distinguish between the seven or more different Mo isotopes which come from the fission process, and hence all of these isotopes naturally come along into the product. This process results in a lot of high-level radioactive chemical waste. An alternative process would produce Mo-99 either from neutron irradiation of Mo-98 (24%) or neutron spallation on Mo-100 (9.6%), with essentially no high level waste. If indeed these processes became the ones preferred because they produce no high level waste, then pure targets of either Mo-98 or Mo-100 would be needed in order to minimize the “contamination” of the five other naturally occurring isotopes of Mo. In fact the one reason that medical production of Mo-99 uses the fission product process is that it has far less Mo contamination than the neutron

irradiation process on pure Mo-98. The latter, even in a high flux reactor, produces typically only 1 activated Mo-99 atom in every 50,000 Mo-98 atoms. However, the many other medical isotope applications that depend on neutron irradiation in reactors (and have no fission product production options) need to start with pure targets of the isotope that is exposed to neutron capture.

Medical isotope needs are rather small, in the gram range, with patient injections being in the milligram range. However, as has been pointed out above, there are needs for pure isotopes in industrial applications, and these come in the kg range need. When dealing with the need for very large quantities of an isotope, existing techniques used for uranium separation might be more feasible, at least until laser isotope separation techniques have been demonstrated to be sufficiently effective for commercial development. For that reason our focus in this paper is on laser techniques for isotope separations of potential use in nuclear medicine.

BACKGROUND.

After sufficiently powerful lasers became available in the 1970's for isotope-selective excitations, it appeared that quantum-action LIS techniques might enrich some isotopes at much lower cost than the old mass-action UCF and EMC schemes. A world-wide effort to develop LIS techniques ensued. The main advantage of LIS methods over mass-action UCF and EMC processes, is that in the latter case, all isotopes of a desired element must be energized, while in LIS one only energizes one isotope of interest. For example to separate S-33 whose natural abundance is 0.75%, a UCF or EMC device must energize 133 times more molecules or atoms than what is required in a LIS process. In addition, single-stage separation factors (β) are generally higher, that is $\beta > 2$ compared to $\beta \leq 1.3$ for UCF. (Note, in the traditional gaseous diffusion process used for uranium enrichment, the single stage $\beta = 1.0043$.) Therefore, LIS separator equipment offers smaller footprints, which allows small-quantity radioisotope separators to be mounted inside standard hot-cells. Thus LIS offers the possibility of providing new radioisotopes which hitherto were too difficult to extract from a "hot" product mixture.

Two different LIS approaches have evolved in the last decades. One labeled AVLIS (Atomic Vapor LIS) employs atomic vapors and utilizes isotope shifts of electronic excitation frequencies. The other called MLIS (Molecular LIS), uses gaseous molecules and takes advantage of isotope-shifts of vibrational absorption bands. In AVLIS, laser photons in the ultraviolet or visible spectrum are used, while MLIS requires laser excitations in the infrared. Using high-temperature furnaces and electron-beam evaporators of elemental Uranium, AVLIS has been developed at Livermore for Uranium enrichment. Compared to MLIS and UCF schemes however, AVLIS is uneconomic and too expensive to develop for most medical isotope separation applications.
Suitable infrared lasers for vibrational excitations in MLIS were developed in the 1970 to 1990 period, and a number of favorable laser/isotope spectral matches were found. While the first step of providing selective molecular laser excitation was straightforward, the second MLIS step of separating or "harvesting" excited isotopic species from unexcited ones, proved more difficult. Early MLIS harvesting involved molecular obliterations (MOLIS) and enhanced chemical reactions (CHEMLIS) with a mixed-in co-reactant. In these schemes, aside from selective excitation, laser photons induced dissociations and/or chemical reactions of selected isotopic molecules with mixed-in co-reactants, yielding enriched or depleted products that were chemically different and separable from feed molecules. Although a few MOLIS/CHEMLIS schemes using multi- photon absorption

were successful, they still required a large number of photons per separated isotopic molecule, partly negating the basic LIS promise of low energy consumption. Experiments showed further that many heavy molecules of interest form process-complicating dimers when they are cooled to improve spectral separation of adjacent isotopic absorption bands.

Rather than combating dimerization (coagulation into double molecules), subsequent research took advantage of it, leading to the more recent condensation repression (CR) harvesting techniques. A big advantage of CR-MLIS is that feed and product gas streams are chemically the same, so staging is simple. Furthermore one laser beam can irradiate three or more enriching chambers in series. Finally, quantum energies needed to affect CR are compatible with single photon energies of high-power infrared CO_2 and CO lasers. CR-MLIS can be activated by single infrared photons of only ~0.1 eV per monomer or dimer, which compares with 6.2 eV per atom for Uranium ionization in AVLIS, and 2 to 5 eV per UF_6 molecule for a laser-induced chemical change in MOLIS/CHEMLIS.

The throughput of a single supersonic free-jet CR-MLIS device (Fig. 1) is ~0.1 moles/hr, which is comparable to a single gas ultracentrifuge unit that processes 0.1 to 1 moles/hr. This flow rate is adequate in most medical or tracer isotope applications. A single cold-wall CR-MLIS unit (Fig. 2) on the other hand can process only $\sim 10^{-5}$ moles/hr and the surface physics is only favorable for a few isotopic molecules (see below). Nevertheless the latter technique can still be useful for separating small quantities of selected (radio)isotopes. Table 2 summarizes typical performance parameters and estimated product costs of UCF, EMC, and LIS separators. In what follows we outline the basic principles of free-jet and cold-wall CR-MLIS methods researched at the University of Missouri and Idaho State University.

CONDENSATION REPRESSION HARVESTING.

The two CR schemes that have been investigated use either a supersonic free jet and low-temperature dimer formation, or take advantage of cold-wall condensation of a subsonic gas stream. Both employ mixtures of a vapor with isotopic molecules such as $^iQF_6$, $^iQF_4$, or ^iQXYZ (e.g. MoF_6, SF_6, TeF_6, $XeOF_4$) with ^iQ a desired isotope, diluted in excess carrier gas G (G could be H_2, He, N_2, Ar, Xe, SF_6, etc). Both CR schemes are operated at low temperatures and pressures. In the free-jet method, a self-cooling QF_6/G gas stream expands adiabatically through a nozzle into a low-pressure chamber shown in Figure 1. After traversing the laser irradiation chamber, most of the jet core is captured by a skimmer, while rim gases that diffuse radially out of the core are evacuated separately. A tuned laser beam irradiates the jet coaxially or transversely and excites selected $^iQF_6$ isotopic molecules. Unexcited $^jQF_6$ molecules dimerize in the jet as it cools and tend to stay in the jet core longer because of their heavier mass. Excited $^iQF_6^*$ migrate out of the jet core more rapidly, following a sub-microsecond existence as a $^iQF_6^*$:G dimer that experiences (pre)dissociation, yielding epithermal $^iQF_6$ and G molecules that recoil off each other. As a result the rim gases are enriched by, and the skimmer gas stream is depleted of $^iQF_6$. The heavier the atomic mass M_G of carrier gas G, the higher the separation factor β_i is [1]. However to insure adequate jet cooling, the gas specific heat ratio of G must be $1.2 < (c_p/c_v)_G < 1.4$.

In the cold-wall approach, a coaxial laser beam selectively excites $^iQF_6$ in a QF_6/G gas stream that flows subsonically through a wall-cooled tube, shown in Figure 2. The temperature of the wall must be such that the corresponding equilibrium vapor pressure is below the partial pressure of the

incoming QF_6 vapor, allowing some QF_6 to condense out. The precise value of the wall temperature T is further selected to optimize isotope separation. The laser beam radius in the tube should be as close as possible to the inner radius of the cylindrical tube, but not touch it. Then, if sufficient numbers of excited $^iQF_6^*$ reach the cold wall (i.e. at low total pressures), and provided the vibrational excitation quantum ε_a of $^iQF_6^*$ exceeds the depth D_α of the attractive surface potential, $^iQF_6$ will desorb from the surface at a higher rate than unexcited $^jQF_6$. This is due to vibration-to-translation (VT) energy conversion and recoil of surface-captured $^iQF_6^*$ molecules, from (pre)dissociation effects. The exit gas stream is thereby isotope-enriched and the wall condensate isotope-depleted. However the condition $\varepsilon_a > D_\alpha$ and additional molecular surface orientation restrictions, eliminate some isotopic molecules from a possible CR-MLIS process.

Free-jet and cold-wall CR-MLIS are only effective at total gas mix pressures below 0.1 torr. Because of the supersonic speed, throughputs in the free-jet scheme are still reasonable. In the subsonic cold-wall scheme however, process gases move a thousand times more slowly. Free-jets are therefore preferred for most CR-MLIS separations. For milligram separations of (radioactive) medical isotopes however, the simpler cold-wall CR method may still be useful.

In both the subsonic cold-wall and supersonic free-jet case, earlier theories of cold-wall condensation, dimerization, and vibrational relaxation, were found inadequate and incapable of predicting experimental observations. Calculated optimum gas pressures were far too high. This greatly hindered validation of CR-MLIS concepts, which appeared fundamentally viable. In-depth studies were therefore undertaken to reexamine condensation, dimerization, and vibrational relaxation physics of QF_6 vapor molecules. The results are published in [1–4].

Cold-wall laser isotope separation of $^iBCl_3$ was first announced in 1975 by K.S. Gochelasvili e.a. [5]. However attempts at Los Alamos by G.K. Anderson and J.T. Lee to duplicate the Russian results failed [6], and in general the concept has received mixed reviews. Our new cold-wall condensation theory has thrown new light on the method and can explain previous ambiguities. The new condensation theory uses a new principle in accounting for the various possible outcomes or "events" when an excited or unexcited QF_6 molecule strikes a cold surface covered with QF_6 condensate. At gas pressures below about ten atmospheres, one can show that such interaction events occur primarily between one striking QF_6 molecule and one surface-captured QF_6. By listing all of the most probable one-on-one events such as surface capture, expulsion by high energy surface-striking gas molecules, ejection by condensate phonons, etc, and by recognizing that the sum of these probabilities must equal unity for an average QF_6 molecule, important parameters such as critical temperatures and vapor pressure curves can be deduced which agree well with experiment. For molecules like SF_6 with $\varepsilon_a > D_\alpha$, the theory predicts possible isotope separations at low pressures within small temperature windows with enrichment factors $\beta \sim 2$ as shown in Figure 3 [3,4]. CR-MLIS experiments with CO_2 laser irradiations at Idaho State University (ISU) and the University of Missouri (MU) have confirmed that cold-wall separations of $^iSF_6$ indeed take place [7]. Measured values of $\beta = 1.5$ to $\beta = 2$ agree with calculated values. Unfortunately because of low throughputs, long collection times (hours) are needed to enrich even micrograms; that is cold-wall isotope separation is less favorable than the free-jet scheme.

The possibility of laser-induced isotope separation by gas-phase dimerization repression in super-cooled supersonic free jets was first proposed by Y.T. Lee in 1977 [9], and experimentally verified

for SF_6 by H. VandenBergh in 1985 [10]. Lee's original proposal considered laser excitation of already formed dimers, which would thereafter pre-dissociate, whereas our work and VandenBergh's indicate that excitation of $^iQF_6$ monomers followed by dimerization (e.g. $^iQF_6^*$:Xe) and subsequent pre-dissociation of the dimer is more profitable. This is because photon absorption peaks of monomers have much higher cross-sections than those of dimers [2].

As mentioned, originally the major problem in diagnosing and predicting dimerization in free jets was the lack of a reliable theory. It was earlier believed that dimers only form in three-body collisions because of simultaneous energy and momentum conservation, which requires that one of the three interacting bodies must carry off any excess energy so that the other two can bond. If so, the high rates of dimer formation observed in cold supersonic free jets, would be theoretically impossible. If the three-bodies theory were correct, laser-pumped vibrational states of QF_6 molecules at low temperatures should last a relatively long time. According to the three-bodies-only dimer formation theory and well-established VT relaxation relations, for heavy molecules QF_6 and heavy carrier gases G, laser-excited QF_6^* molecules should last through some 10^5 collisions before loosing their vibrational energy by collisional VT transfers, when T<200K. Early CR-MLIS experiments relying on this theory therefore used process gas pressures that were much higher than what was found later to be effective.

A new examination of dimerization physics revealed that the three-bodies-only theory for creation of dimer populations is incorrect. This orbital mechanics theory assumes motions of point particles. Actually it was found that for finite-sized molecules, dimers are more frequently formed in two-body contact collisions for those with kinetic energies in the low-energy part of the Boltzmann distribution [1]. Excess energy is shed by exchange with vibrational quanta of the VanderWaals dimer potential and by induction of dimer rotation. Dimer formation rates are essentially the same for laser-excited and unexcited QF_6 molecules, since both migrate with the same thermal molecular speeds. However after a few rotations and dimer vibrations of a freshly formed QF_6^*:G dimer, stored vibrational energy ε_a is converted into kinetic energy by the pre-dissociation process [2]. This VT conversion forces the dimer partners to recoil off each other and is utilized in CR-MLIS free-jet isotope separation. The new dimer formation theory [1] shows that the probability for dimer formation increases exponentially with decreasing temperature. Thus vibrational relaxation rates of excited $^iQF_6^*$ also increase as the temperature drops since they are catalyzed by dimerization. This is opposite to earlier (dimer-less) collisonal VT theory where VT relaxation rates decrease exponentially with decreasing T. The new theory explains why earlier CHEMLIS schemes failed: the (pre)dissociation time for freshly formed QF_6^*:R dimers (R = coreactant) is shorter than the time it takes for a chemical rearrangement of the atoms in the reaction complex. With the new dimer theory, migrations of dimers, thermal, and epithermal monomers in and out of a supersonic free jet can be calculated with and without laser irradiation [2]. Optimum pressures and temperatures for isotope enrichment can be predicted, as shown for example in Figure 4, which plots the enrichment factor β versus temperature T for dimethyl-zinc, $^iZn(CH_3)_2$, excitable by a CO laser. Zn-67 and Zn-68 are used as targets in accelerators to produce Cu-67 and Ga-68, which are important for tumor diagnostics and treatment. Zn-64-depleted zinc is desirable as a low neutron absorbing corrosion inhibitor in BWR cooling systems.

CONCLUSIONS.

We believe that free-jet and cold-wall condensation repression harvesting in laser isotope separations are promising processes for enriching medical and industrial isotopes, and that the requisite physics has been developed with which to model the performance of these processes. Much research with different isotopic molecules is still needed to increase our data-base of CR-MLIS methods, and to further validate the new analytic theories we have developed. Because in CR-MLIS, feed and product gases are physically the same (except for isotopic composition), simple enriching stages can be used in series and be irradiated by a single laser beam to obtain desired overall enrichments. When compared with UCF's, the main advantage of CR-MLIS is the total energy consumed per separated isotope and the smaller chamber size. Ultimately CR-MLIS techniques might be used for the separation of a particular radioactive isotope from a "hot" mix of products generated by accelerators or reactors. This could be done in hot-cells with infrared transmitting windows through which a laser beam is passed from an outside high-power laser.

There is considerable laboratory experimental work to be done to demonstrate the conditions under which condensation repression will operate for a particular molecular isotope. Comparison with theory will help to ratify the adequacy of the theory. Such experiments would be appropriate to be done on several isotopes of medical interest. Such work is generally of the type that can be conducted in a university laboratory setting. The Idaho National Laboratory, with its Center for Advanced Energy Studies that is closely associated with the regional universities, would be an ideal venue for carrying out research of this type.

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TABLE I. MEDICAL RADIO-ISOTOPES.

DESIRED RADIO-ISOTOPE[*)]	NATURAL TARGET[**)]	APPLICATION
(A) SPALLATION - ACCELERATOR PRODUCED		
I-123 (13.2h)	Xe-124 (0.1%), {p,2p}	PET Scan[***)]
I-123 (13.2h)	Te-122(2.6%), {d,n}	PET Scan[***)]
Mo-99(66h)/Tc-99m (6h)	Mo-100 (9.6%), {p,pn}	Diagnostics; 150,000 US procedures weekly
Cu-67 (2.58d)	Zn-67 (4.1%), {p,n}	Radiotherapy with Monoclonal Antibodies
Ge-68(270d)/Ga-68 (68m)	Zn-68 (18.7%), {p,n}	Generator for PET[***)]
Co-57 (272d)	Fe-57 (2.1%), {p,n}	Radiotherapy
Pd-103 (17d)	Rh-103 (100%), {p,n}	Prostate "Thera-Seed"
Tl-201 (3.04d)	Tl-203 (29.5%),{p,t}/decay	Heart Scan
Co-58 (9.1h/71d)	Ni-58 (68%), {p,n}	Anemia Tracer
(B) NEUTRON ACTIVATION - PRODUCED IN RESEARCH REACTOR		
P-33 (25d)	S-33 (0.74%), {n,p}	Diagnotics/Therapy
Os-191(15d)/Ir-191 (4.9s)	Os-190 (26%), {n,γ}	Pediatric Cardiology
Os-194(6a)/Ir-194 (19.4h)	Os-192 (41%), {2n,γ}	Proposed Radiotherapy
Rh-105 (35.4h)	Ru-104(19%) indirct{n,γ}β	Radio-Immune Therapy
Mo-99(66h)/Tc-99m (6h)	Mo-98 (24%), {n,γ}); U-235 (0.7%),{n,fission}	Diagnostics; 150,000 US procedures weekly.
W-188(69d)/Re-188 (17h)	W-186 (28%), {2n,γ}	Radiotherapy Trials
Sn-113(115d)/In-113m (1.7h)	Sn-112 (1%), {n,γ}	Diagnostics
Dy-166(3.4d)/Ho-166 (117d)	Dy-164 (28%), {2n,γ}	Bone Marrow Ablation
Pm-149 (48h)	Nd(148) (17.2%), {n,γ} β	
Pd-103 (17d)	Pd-102 (1%), {n,γ}	Prostate "Thera-Seed"
Pt-195 (4.02d)	Pt-194 (33%), {n,γ}	Brain Scan
Sm-153 (1.93d)	Sm-152 (27%), {n,γ}	Bone Cancer Therapy
Re-186 (3.77d)	Re-185 (37%), {n,γ}	
Lu-177 (6.68d)	Lu-176 (2.6%), {n,γ}	Radio-Immune Therapy

*) Combinations indicate parent/daughter pairs. Half-lifes are in parentheses.
**) Parentheses ( ) are natural abundances of target isotopes; { } indicate nuclear reactions.
***) PET= Positron Emission Tomography

TABLE II. COMPARISON OF ENRICHMENTS OF ^{33}S FROM 0.74% F → 99%Y/0.2%W[1)]

	MASS ACTION[1)]		LASER[1)]	
	UCF (SF_6)	EMC (S)	MOLIS (SF_5Cl)	Free-Jet CRISLA[2)] (SF_6)
β_{stage}	1.09	10	4	2.2
Total No Stages[3)]	125 (110 + 15)	7 (5 + 2)	8 (7 + 1)	14 (12 + 2)
No of Unit Separators	3,157[4)]	140	23	14
Feed per Unit[5)] F_u ,moles/hr	0.250[4)]	0.011	0.040	0.064
Plant Total Feed, F, moles/hr	201.3[4)]	3.660	3.660	3.660
Plant Output Y, moles/hr	0.110[4)]	0.020	0.020	0.020
Footprint for Plant, m^2	1,250[4)]	16,000	25 for chambers + 975 for lasers= 1000	5 for chambers + 105 for lasers = 110
kWhr/mole	40,000	150,000	9,400[6)]	3,600
Consumed eV per S-33	1,492,400	5,596,500	350,714	134,316
Atomic Excit'n eV per S-33	----	1,392 (ionization)	3.0 (dissociation)	0.1 (dimer predissoc'n)
Operating Cost	$ 250/g	$ 6,000/g	$ 165/g	$ 95/g
Write-off Plant (10y)	$ 1,850/g	$ 6,000/g	$ 177/g	$ 125/g
Total Product Cost	$ 2,100/g	$ 12,000/g	$ 342/g	$ 220/g

[1)] F = Feed; Y = Product; and W = Tails Stream Flow Rates in moles/hr. xx% = S-33 abundance. All values are coarse estimates based on open literature publications. Note that W = F – Y.

[2)] CRISLA = Condensation Repression by Isotope Selective Laser Activation = CR-MLIS.

[3)] Ideal cascades are assumed with number of stages in enriching and stripping sections in parentheses.

[4)] For a UCF the minimum-sized plant requires 3,157 units, with Y_{UCF} = 0.11 moles/hr; F_{UCF} = 201 moles/hr; and a footprint of 1,250 m^2.

[5)] Based on allowed flow rate through a single separator unit, and $Y_u/F_u = \theta = (1+\beta_{stage})^{-1}$ for each unit.

[6)] Includes power for inter-stage chemical re-conversions in MOLIS.

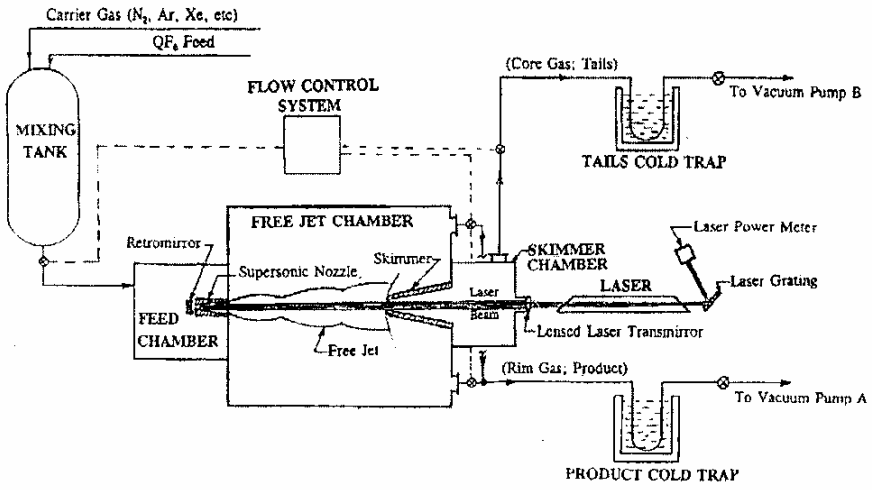

FIG. 1 SCHEMATIC OF FREE-JET LASER ISOTOPE SEPARATION EQUIPMENT.

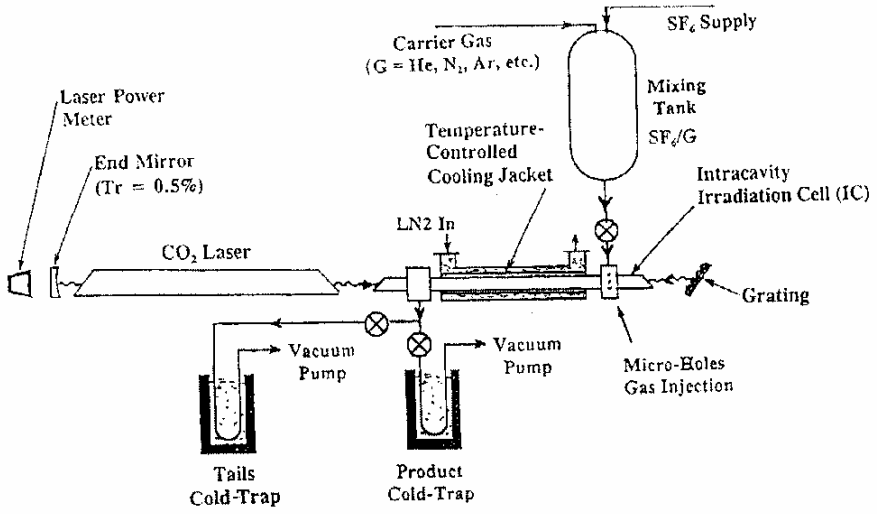

FIG. 2 SCHEMATIC OF COLD-WALL LASER ISOTOPE SEPARATION EQUIPMENT.

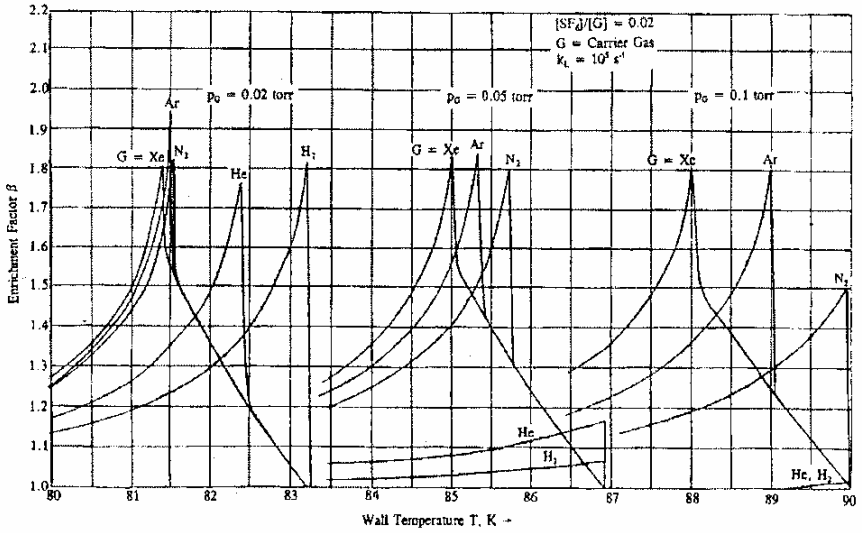

FIG. 3 CALCULATED ENRICHMENT FACTORS FOR $^{i}SF_6$ IN COLD-WALL CR-MLIS.

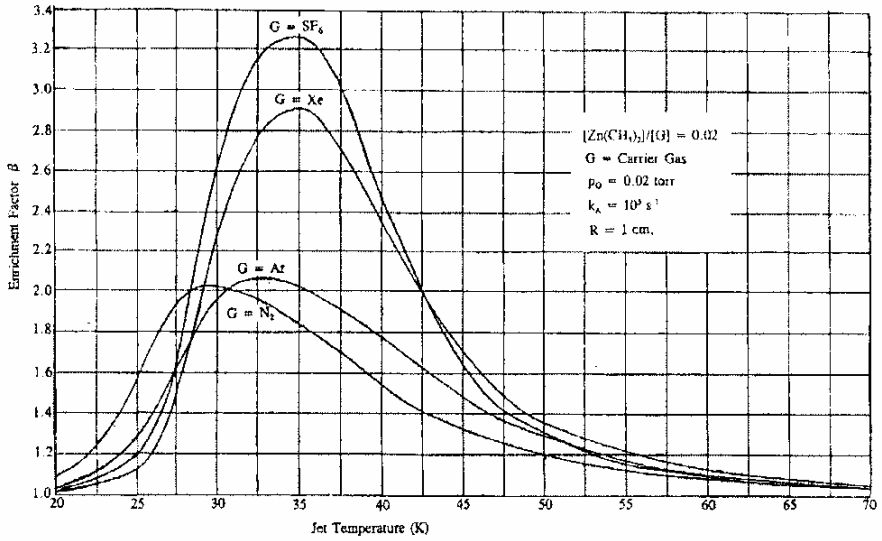

FIG. 4 CALCULATED ENRICHMENT FACTORS FOR $^{i}Zn(CH_3)_2$ IN FREE-JET CR-MLIS.