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Science and Technology of isotope enrichment[†]

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Abstract : One of the toughest scientific challenges has been to effectively - and inexpensively - separate a desired isotope of an element from the remaining isotopes for uses ranging from medicine to energy to weapons applications. In this account, after briefly dealing with important basics of separation physics, the various separation methods broadly classified as physical and chemical methods are described. Emphasis has been given to the advanced separation processes using lasers and some results from our laboratory have been described in detail. It is quite clear that there is a continuous massive effort by scientists and engineers around the world for developing isotope enrichment technologies.

Keywords : Isotope enrichment, laser, AVLIS, MLIS, IRMPD.

Introduction

The discovery of stable "isotopes" began with J. J. Thomson's identification of neon-22 in 1912. More than 90 naturally occurring elements have been identified on the earth; they exist as nearly 270 stable isotopes – forms of the elements that do not decay or emit radiation and hundreds of radioactive isotopes. The various isotopes of a given element differ from one another only in the number of neutrons in their atomic nucleus (the number of protons in the nucleus differentiates the elements from one another), and even highly purified samples of an element are generally a mixture of several isotopes. Pure silver, for example, is composed of nearly equal amounts of silver-107 and silver-109. Iron is mostly iron-56 (92 percent), but it contains small amounts of three other isotopes as well, and tin is a mixture of 10 stable isotopes, the most abundant of which makes up only 33 percent of the total.

One of the toughest scientific challenges has been to effectively – and inexpensively – separate a desired isotope of an element from the remaining isotopes for uses ranging from medicine to energy to weapons applications. The main thrust is aimed at developing workable isotope separation method for nuclear industry : (i) uranium-235, the fissile isotope of uranium needed to fuel light water reactors and (ii) deuterium for heavy water moderated reactors. Enriched refers to material that consists largely or exclusively of a single isotope and is usually obtained by one of the enrichment techniques described in this article.

No doubt the common man today knows a lot more of the atom; all the same perhaps not enough to appreciate the role of atomic age affecting their daily activities. For many, bombs and power plants are their only associations with the term. The uses and benefits of isotopes, stable and/or radioactive, in medicine, agriculture, industry, and science are now widespread. In the last three decades the use of enriched isotopes has offered substantial advantages to scientists and clinicians involved in the rapid growth of research on human body composition, energy balance, protein turnover, and fuel utilization. Today, increased awareness of the role played by the elements in human health and as etiologic factors in diseases (osteoporosis, heart disease, cancer, diabetes, kidney disease) as well as diagnostic and therapeutic adjuncts (obesity, inborn errors of metabolism, heart disease) has created an explosion in the need for isotopes. Although stable isotopes occur naturally, their utility can

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[†] In honour of Professor Jai P. Mittal.

be greatly enhanced when they are isolated and enriched through processes such as electromagnetic separation, gaseous diffusion, centrifuge, chemical exchange, cryogenic distillation, thermal diffusion, or other advanced processes like laser isotope separation. Since in principle these processes can also be employed to enrich uranium to very high U-235 concentrations, required for the manufacture of nuclear weapons, the technical details of almost all enrichment processes are subjected to strict security regulations.

Basic facts of separation physics

Before going into more detail on the individual enrichment processes, it is necessary to explain some important basic facts of separation physics. A separation element (as shown in Fig. 1) divides the feed stream F with U-235 concentration N_F into two streams : the product stream P of enriched uranium with concentration N_P and a tails stream T of depleted uranium with concentration N_T. The concentration N is the U-235 content in the mixture of isotopes U-235 and U-238. The ability of the separation element to separate U-235 and U-238 is described by its separation factor. One distinguishes between the enrichment factor α, the depletion factor β and the total separation factor α.β, the definition of which is given in Fig. 1. The separation factor expresses the degree of concentration or depletion achieved in a single separation element. When a separation element, for example a centrifuge, is installed such that the enrichment factor α is equal to the depletion factor β and when the given total separation factor is 1.4, then natural uranium with a concentration of 0.71% U-235, fed in as feed, is divided into a product stream with 0.84% U-235 and a tails stream with 0.6% U-235.

However, the separation factor alone does not describe fully the efficiency of a separation element, because the material flows between the different concentrations must also be taken into consideration. To determine the "work" that must be applied for separation, the concentration of the feed material N_F and the depletion in the tails N_T must be given in addition to the product quantity P and the product enrichment N_P. In order to describe the effort required for the separation of isotopes, the term "separative work" was introduced. Since separative work has no quantifiable physical form, a formula had to be developed, using the starting data P, N_P, N_F and N_T, to calculate a value which is proportional to the technical and energy resources required for the separation of the isotopes.

The mass and isotopic balance of a separation element are the starting point for the mathematical description :

$$P + T - F = 0$$

$$PN_P + TN_T - FN_F = 0$$

These two equations give the amount of natural uranium F required from the amount of product and the concentration :

$$F = P\,(N_P - N_T)/(N_F - N_T)$$

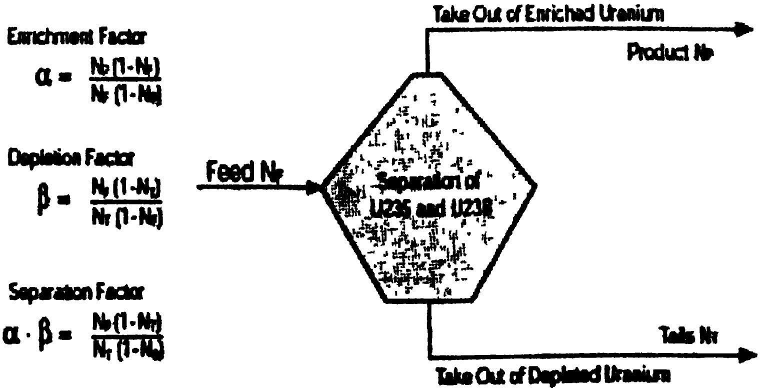

Fig. 1. Scheme of a separation element. The enrichment factor α, the depletion factor β and the total separation factor α.β is defined.

The feed and the two streams leaving the separating element are multiplied by a value function, which depends only on the concentration of each respective stream. Separative work is then defined by the balance :

$$\delta U = PV(N_P) + TV(N_T) - FV(N_F)$$

The value function V(N) is determined using mathematical methods so that the calculated separative work is independent of the U-235 concentrations in the separation element and depends only on the archieved change in concentration and throughput.

A second order differential equation is the mathematical solution of this problem. After double integration of the differential equation the value function depends on 2 freely chosen integration constants. The integration constants are chosen so that the expression of the value function takes the simplest form possible. Thus the expression for the value function in common practice is taken as :

$$V(N) = (2N - 1) \ln [N/(1 - N)]$$

Since the value function is dimensionless, the separative power has the dimension of mass over time and the separative work, calculated from the quantities of material, has the dimension of mass. Its units are kg separative work (kg SW) also known as "separative work unit (SWU)", whereby 1 SWU = 1 kg SW. The capacity of an enrichment plant is given in separative work units which can be produced each year, often using tSW (1000 kg SW) to give plant capacities of e.g. 1000 tSW/a or 1 MSWU/a.

The amount of separative work and the amount of natural uranium, required for a certain amount of enriched uranium, rise and fall considerably depending on the residual content of U-235 in the tails. The concentration of U-235 in the depleted uranium is called the tails assay. Fig. 2 illustrates this. With a reduction in the tails assay from 0.3% to 0.2% U-235 the separative work required for the production of 1 kg of uranium enriched to 3% increases by 25%, while approximately 17% less natural uranium is required. The optimum tails assay is that for which the total costs for enriched uranium are minimised depending on the costs for separative work and including the costs of natural uranium in the form of UF_6.

Different technologies for isotope enrichment

Almost all the methods used for separation in conventional industries can be used for separation of isotopes also with varying degree of success. The methods can be broadly classified into physical and chemical methods.

(i) Physical methods are those in which difference in physical properties e.g. inertia, density, electrical properties etc. is harnessed. Some of the methods in this cat-

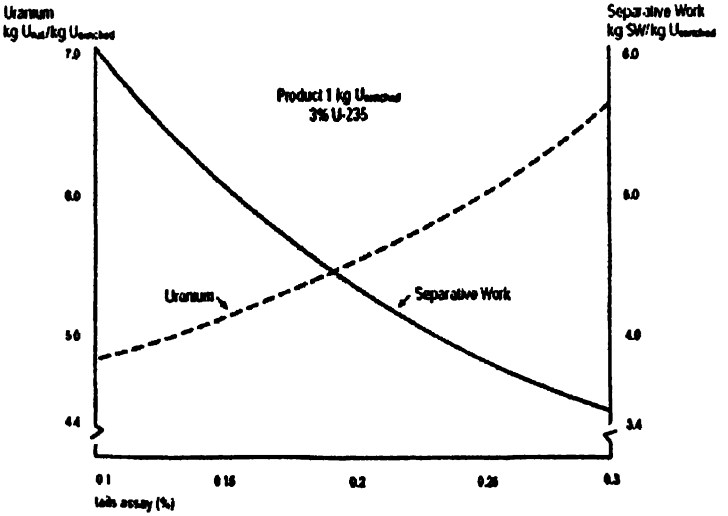

Fig. 2. Effect of tails-assay on separative work and uranium composition.

egory are :

- Gaseous diffusion
- Gas centrifuges
- Jet nozzle/aerodynamic separation
- Thermal diffusion
- Electrolysis
- Electromagnetic separation
- Laser isotope separation

(ii) Chemical methods are those in which difference in chemical properties e.g. chemical potential is harnessed. Some of the methods in this category are :

- Distillation
- Chemical exchange
- Exchange distillation
- Ion exchange
- Liquid-liquid extraction

Chemical methods are more suitable for light isotopes and are extensively used for separation of hydrogen isotopes e.g. production of heavy water largely by chemical exchange process. Physical methods are in general more suitable for separation of heavy isotopes e.g. uranium enrichment. The remaining part of this article will be dealing only with first three of physical process as applied to uranium enrichment and some of the LIS successes from our laboratory.

Gaseous diffusion

The gaseous diffusion process has been used to enrich nearly all of the low and highly enriched uranium that has been produced in the United States. It was first developed in the 1940s as part of the Manhattan Project and was used to enrich a portion of the uranium used in the bomb that was dropped on Hiroshima. All five acknowledged nuclear weapons states within the nuclear non-proliferation treaty (NPT) regime have operated gaseous diffusion plants at one time or another, but currently only the United States and France continue to operate such facilities. The diffusion process requires pumping uranium in a gaseous form through a large number of porous barriers and, as noted above, is very energy intensive.

In order to make the uranium into a gaseous form that can be used in the diffusion process, the natural uranium is first converted into uranium hexafluoride (UF_6). The uranium hexafluoride molecules containing U-235 atoms, being slightly lighter, will diffuse through each barrier with a slightly higher rate than those containing U-238 atoms. A simple analogy to help visualize this process is to imagine blowing sand through a series of sieves. The smaller grains of sand will preferentially pass through each sieve, and thus after each stage they would represent a slightly higher percentage of the total than they did before passing through the stage. The total separation factor $\alpha.\beta$ is given by :

$$\alpha.\beta = (M_2/M_1)^{1/2}$$

The difference in mass, and therefore velocity, between the UF_6 molecules containing either U-235 or U-238 is very small, and thus thousands of such stages are needed in order to enrich commercial or military amounts of uranium. In a gaseous diffusion plant, the stages are arranged into "cascades" that allow each stage to build on the enrichment achieved by the ones before it and also to more efficiently make use of the depleted uranium stream. The most challenging step in building a gas diffusion plant is to manufacture the permeable barriers required in the diffusers. The material for the barriers needs to be highly durable and able to maintain a consistent pore diameter for several years of operation. This is particularly challenging given the highly corrosive nature of the uranium hexafluoride gas used. Typical barriers are just 5 millimeters (less than 0.2 inches) thick and have openings that are only about 30 to 300 times the diameter of a single uranium atom.

Gas centrifuge

Gas centrifuges are the most commonly used technology today for enriching uranium. The technology was considered in the U.S. during the Manhattan Project, but gaseous diffusion and electromagnetic separation were pursued instead for full scale production. The centrifuge was later developed in Russia by a team lead by Austrian and German scientists captured during the Second World War. The head of the experimentation group in Russia was eventually released and took the centrifuge technology first to the United States and then to Europe where he sought to develop its use in enriching commercial nuclear fuel.

The centrifuge is a common technology used routinely in a variety of applications such as separating blood plasma from the heavier red blood cells. In the enrichment process, uranium hexafluoride gas is fed into rapidly spinning cylinders. In order to achieve as much enrichment in each stage as possible, modern centrifuges can rotate at speeds approaching the speed of sound. It is this feature that makes the centrifuge process difficult to master, since the high rate of revolution requires that the centrifuge be sturdy, nearly perfectly balanced, and capable of operating in such a state for many years without maintenance. Inside the rotating centrifuge, the heavier molecules containing U-238 atoms move preferentially towards the outside of the cylinder, while the lighter molecules containing U-235 remain closer to the central axis. The gas in this cylinder is then made to circulate bottom to top driving the depleted uranium near the outer wall towards the top while the gas enriched in U-235 near the center is driven towards the bottom. These two streams (one enriched and one depleted) can then be extracted from the centrifuge and fed to adjoining stages to form a cascade just as was done with the diffusers in the gas diffusion plants. The maximum separation factor of a counter current centrifuge is :

$$\alpha.\beta = \exp\,[\{\sqrt{2}(M_2 - M_1)V^2L\}/2RTd]$$

and the separative power :

$$\delta U = (n/2)\,\rho\,DL\,[\{(M_2 - M_1)V^2\}/2RT]^2$$

where M_1, M_2 : molecular weight of the molecules to be separated, R : gas constant, D : diffusion constant of the process gas, ρ : density of the process gas, T : temperature in degrees Kelvin, d : diameter of the rotor, L : length of the rotor, V : circumferential velocity.

Like the gas diffusion process, it requires thousands to tens of thousands of centrifuge stages to enrich commercially or militarily significant quantities of uranium. In addition, like the gas diffusion plants, centrifuge plants require the use of special materials to prevent corrosion by the uranium hexafluoride, which can react with moisture to form a gas of highly corrosive hydrofluoric acid. One of the most important advantages to the gas centrifuge over the gas diffusion process, however, is that it requires 40 to 50 times less energy to achieve the same level of enrichment.

Despite having a larger separative power in each stage compared to the gaseous diffusion process, the amount of uranium that can pass through each centrifuge stage in a given time is typically much smaller. Typical modern centrifuges can achieve approximately 2 to 4 SWU annually, and therefore in order to enrich enough HEU in one year to manufacture a nuclear weapon like that dropped on Hiroshima would require between three and seven thousand centrifuges. Such a facility would consume 580 to 816 thousand kWh of electricity, which could be supplied by less than a 100 kW power plant.

Electromagnetic isotope separation (EMIS)

The electromagnetic separation technique is a third type of uranium enrichment process that has been used in the past on a large scale. Developed during the Manhattan Project at Oak Ridge, Tennessee, the electromagnetic separation plant was used to both enrich natural uranium as well as to further enrich uranium that had been initially processed through the gaseous diffusion plant, which was also located at the Oak Ridge facility. The use of this type of facility was discontinued shortly after the war because it was found to be very expensive and inefficient to operate.

The electromagnetic separations process is based on the fact that a charged particle moving in a magnetic field will follow a curved path with the radius of that path dependent on the mass of the particle. The heavier particles will follow a wider circle than lighter ones assuming they have the same charge and are traveling at the same speed. In the enrichment process, uranium tetrachloride is ionized into a uranium plasma (i.e. the solid uranium tetrachloride) is heated to form a gas and then bombarded with electrons to produce free atoms of uranium that have lost an electron and are thus positively charged). The uranium ions are then accelerated and passed through a strong magnetic field. After traveling along half of a circle the beam is split into a region nearer the outside wall which is depleted and a region nearer the inside wall which is enriched in U-235. The large amounts of energy required in maintaining the strong magnetic fields as well as the low recovery rates of the uranium feed material and slower more inconvenient facility operation make this an unlikely choice for large scale enrichment plants, particularly in light of the highly developed gas centrifuge designs that are employed today.

Jet nozzle/aerodynamic separation

The final type of uranium enrichment process that has been used on a large scale is aerodynamic separation. This technology was developed first in Germany and employed by the apartheid South African government in a facility which was supposedly built to supply low enriched uranium to their commercial nuclear power plants as well as some quantity of highly enriched uranium for a research reactor.

The aerodynamic isotope separation (which includes the jet nozzle and helicon processes) achieves enrichment in a manner similar to that employed with gas centrifuges in the sense that gas is forced along a curved path which moves the heavier molecules containing U-238 towards the outer wall while the lighter molecules remain closer to the inside track. In the jet nozzle plants, uranium hexafluoride gas is pressurized with either helium or hydrogen gas in order to increase the velocity of the gas stream and the mixture is then sent through a large number of small circular pipes which separate the inner enriched stream from the outer depleted stream. This process is one of the least economical enrichment techniques of those that have been pursued, given the technical difficulties in manufacturing the separation nozzles and the large energy requirements to compress the UF_6 and carrier gas mixture. As with gaseous diffusion plants, there is a large amount of heat generated during operation of an aerodynamic separations plant which requires large amounts of coolants such as Freon.

Laser enrichment

There are a number of uranium enrichment technologies using lasers such as atomic vapor laser isotope separation (AVLIS), molecular laser isotope separation (MLIS), and chemical reaction by isotope selective laser activation (CRISLA) those have been developed as well, but they are mostly still in the experimental or demonstration stage and have not yet been used to enrich commercial or military quantities of uranium.

The AVLIS, CRISLA, and MLIS processes make use of the slight difference in atomic properties of U-235 and U-238 to allow powerful lasers to preferentially excite or ionize one isotope over the other. AVLIS makes use of uranium metal as a feed material and electric fields to separate the positively charged U-235 ions from the neutral U-238 atoms. MLIS and CRISLA on the other hand use uranium hexafluoride mixed with other process gases as a feed material and use two different lasers to excite and then chemically alter the uranium hexafluoride molecules containing U-235, which can then be separated from those molecules containing U-238 that remained unaffected by the lasers. AVLIS was pursued for commercial use by the U.S. Enrichment Corporation, but was abandoned in the late 1990s as being uneconomical while many other countries have also abandoned all known AVLIS and MLIS production programs as well.

Selective multistep photoionization and AVLIS :

Selective photoionization of atoms is the most universal photophysical method for selective separation of substances, particularly isotopes, at the atomic level. A common feature of all schemes for selective ionization is the sequence of the two processes : (a) isotopically selective excitation and (b) ionization of the excited atoms. The two-step photoionization scheme is the simplest. The three-step scheme can be used for atoms with a high ionization potential. Normally the small photoionization cross section which is about 10^{-4}–10^{-6} times the resonant excitation cross section, can be enhanced by tuning the frequency of last-step radiation to that of the transition to an autoionization [spontaneous or electric-field-induced] state (Fig. 3c). Finally, high-lying Rydberg states can be ionized by IR radiation or a pulsed electric field (Figs. 3d,e).

Besides uranium isotope separation, laboratory experiments are being carried out on selective ionization of other isotopes, potentially useful in much smaller quantities, such as K, Ca, Rb, rare-earth, and of course, transuranium elements. An effective ionization of excited atoms at an average, moderate intensity of the ionizing radiation presents a serious problem in these cases. The ionization schemes given in Figs. 3a-c cannot be practically applied here, so greater attention is paid to the ionization scheme of highly excited atoms shown in Figs. 3d-e. The above three methods enable one to ionize selectively excited isotopic atoms of any element in the periodic table. High separation factors like 10^2–10^5 can be achieved quite comfortably.

Development of AVLIS technology were mainly per-

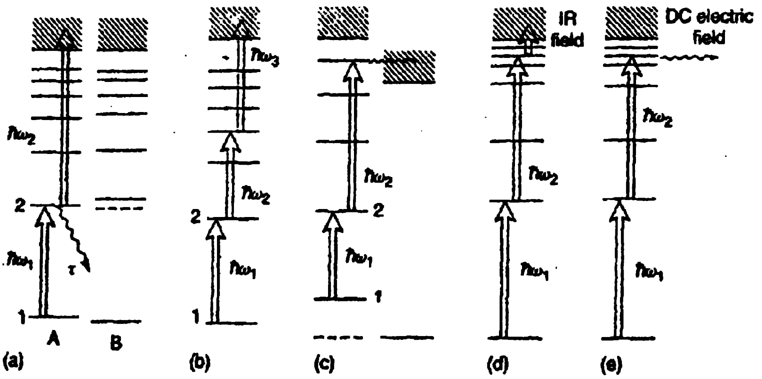

Fig. 3. Schemes of selective step-wise photoionization of atoms by laser radiation : (a) two-step photoionization; (b) three-step photoionization; (c) two-step photoionization through an autoionization state; (d) two-step selective excitation of a Rydberg state and its photoionization by IR laser radiation; (e) two-step selective excitation of a Rydberg state and its ionization by dc electric field.

formed in USA, France, Japan and few more countries. In the first half of the 1980s, an experimental separation plant with a potential output of around 10^3 kg of low-enrichment uranium per year was built at Lawrence Livermore National Laboratory (LLNL) in the United States. The production of a few kilograms of such uranium was reported. Metallic uranium is first vaporized in a separator unit contained in a vacuum chamber. Even this process makes demands on high technology, since the vapour pressure of uranium is very low even at high temperatures ((10^{-4} Torr at 2000 K). At the same time, molten and as-vaporized uranium are extremely powerful reactants. Therefore, the metal kept in a water-cooled crucible is evaporated with an electron beam from where it expands with a velocity of $\sim 5 \times 10^4$ cm/s. Another important parameter need to be considered is the share of uranium atoms in the ground state level, which is only 47% at 2800 K. A metastable state at 620 cm^{-1} is populated to the extent of 28% and the rest are distributed among higher levels due to thermal excitation. This means that to access most of the desired uranium isotopes, two laser frequencies must be provided, one from the ground and the other from 620 cm^{-1} level, just to reach the first excited state.

The vapor stream is then illuminated with laser light tuned precisely to a color at which 235-U absorbs energy. Three wavelengths of red-orange light of dye lasers were used as a radiation source. These were pumped by copper-vapor lasers that provided the necessary 20–30 ns pulse duration at a repetition frequency of around 10^4 Hz. Each color selectively adds enough energy to ionize or remove an electron from 235-U atoms (IP = 6.2 eV), leaving other isotopes unaffected. Because the ionized 235-U atoms are now "tagged" with a positive charge, they are easily collected on negatively charged surfaces inside the separator unit. The product material is condensed as liquid on these surfaces and then flows to a caster where it solidifies as metal nuggets. The unwanted isotopes, which are unaffected by the laser beam, pass through the product collector, condense on the tailings collector, and are removed.

Infrared multiphoton chemistry and MLIS :

Probably none of the phenomena in laser photochemistry discovered in the recent past is as exciting as infrared multi photon excitation and dissociation (IRMPE and IRMPD). Recognition of this method as a means by which selective photochemistry may be realised, has whetted the appetite of photochemists who are excited by its possible applications in dissociation, isomerisation, control of chemical equilibria, synthesis of complex molecules and finally in bond selective chemistry.

The nature and characteristics of the photophysical and photochemical processes induced by multiphoton excitation (MPE) of collisionless, isolated molecules triggered hectic experimental and theoretical activities. The following qualitative picture has emerged out of the numerous experimental and theoretical studies in this subject. The molecular energy levels are separated into three regions (Fig. 4). In the lowest energy range (region I), the density of molecular states is very low and laser field interacts with isolated discrete molecular states (isotope selec-

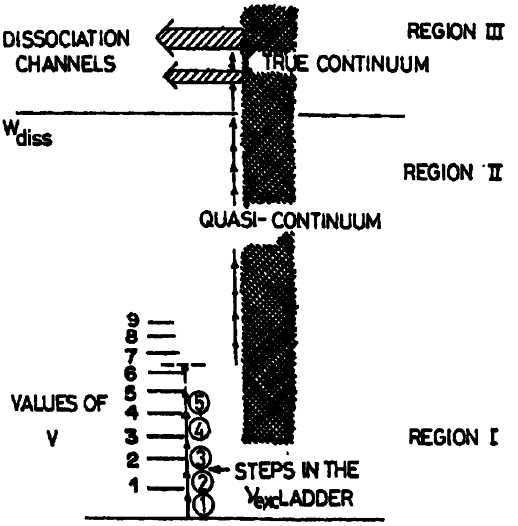

Fig. 4. Schematic for infrared multiple photon excitation (IRMPE) and dissociation (IRMPD), the working principle of MLIS.

tive excitation). Several possible mechanisms to overcome the anharmonic detuning at several successive transitions of the excited vibrational mode have been considered viz. (a) power broadening, (b) rotational anharmonicity compensation and (c) multi photon transitions. Once the molecule has absorbed a few quanta, the density of molecular states become very large and time evolution can no longer be described in terms of a few isolated molecular states. This region is denoted as quasi-continuum (region II) and its exact theoretical treatment is still a subject of intense investigation. Finally, when the molecule acquires enough amount of internal energy, it enters a true continuum (region III) where in addition to all the complications of region II, dissociation or predissociation or isomerisation channels open up.

Many experiments on isotopically selective MPD have thus been performed which covered many isotopes, from light ones (H, D, T) to heavy ones (Os, U) contained in very different molecules. Many of these became the basis of laser isotope separation methods developing in numerous laboratories of several countries. Table 1 presents some of the MLIS successes from our laboratory.

Conclusion

From this brief account, it is quite clear that there is a continuous massive effort by scientists and engineers around the world for developing isotope enrichment technologies. Judging from the development since the days of Manhattan Project, uranium enrichment is a dynamic field and even today new and interesting technology is being

Table 1. Isotope selective laser photochemistry at BARC

Isotopes	System pressure (Torr)	Typical results Laser freq.	 Fluence (J/cm^2)	 Selectivity
Sulphur	SF_6	10 P(20)	2.0	10
34/32	1.0	CO_2 laser		
Uranium	$UO_2[(CF_3CO)_2CH]_2.THF$	10 P(4)	0.08	1.2
235/238	Molecular beam	CO_2 laser		
Carbon	CF_2HCl	9 P(20)	4.0	60
13/12	100.0	CO_2 laser		
Stage I	CF_3Br/Cl_2 (1 : 4)	9 P(32)	4.5	65
	50.0	CO_2 laser		
Stage II	CF_3Cl/Br_2 (1 : 6)	9 P(8)	1.7	200
	20.0	CO_2 laser		
Deuterium	CHF_3/Ar (1 : 2)	10 R(10)	30.0	>40
D/H	75.0	CO_2 laser		
Tritium	CHF_3/Ar (1 : 2.5)	9 R(8)-9 R(14)	65.0	38
T/H	28.0 (0.2 ppm CTF_3)	CO_2 laser		
T/D	$CDCl_3/Ar$ (1 : 5)	828 cm^{-1}	21.0	5000.0
	12.0	NH_3 laser		

pursued throughout the world. Therefore, assessment of evolving technology must also be a continuing activity, not only to recognize the impact of the scientific achievements themselves, but also to keep the entire uranium enrichment endeavour in step with a changing world-wide economic and political environment.

Acknowledgement

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