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Overview of Selective Photo-reaction

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Abstract

Selective reaction process especially isotope separation is a key technology for the development of the technologies related to the nuclear energy. However only a few species are separated on a production scale using the conventional processes such as thermal diffusion, chemical exchange reaction and distillation for lighter isotopes ,and gas centrifuge and gaseous diffusion for uranium. As these methods are based on statistical thermodynamics and have low enrichment factors, they need repetitive operations of separation with many separating units combined together. Electro-magnetic separation method known as the one with high separation factor can be applied to most of the elements ,but extremely low production rate is realized, which is uneconomical.

From the above point of view, much attention has been paid to the laser process by which high selectivity is expected. This method can be applied to either gas ,liquid or solid phase, and high separation factors are basically realized only in gaseous phase. Since the beginning of the studies on isotope separation in early 1970s, many ideas have been proposed for the selective photo-reaction process such as photoionization, multiphoton dissociation and state selective chemical reaction. As a result of experimental and theoretical efforts, large scale production of some isotopes have been intended. Production of deuterium by infrared multi-photodissociation method was studied aiming at the replacement of the conventional dual temperature exchange process, and lots of experiments have been achieved intensively for the uranium enrichment. A stepwise selective photoionization method has also been studied for the isotopic separation of many elements, especially uranium enrichment.To implement the laser processes on a large scale production system, advanced performances are required not only for the tunable laser systems but also for many related technologies such as atomic/molecular source,photo-reactor and extractor of products.

Keywords:Laser Isotope Separation,Selective Reaction,Photo-ionization,Photo-dissociation,Selectivity

1.Introduction

Selective photoreaction is made based on the spectroscopic difference caused by the difference elements, isotopes,isomers and isobars. In the field of nuclear engineering, isotope separation technique special attention is paid to the wide area ranging from the uranium enrichment and isotope tailoring materials to the reprocessing of spent fuels. Among many conventional isotope separation methods the laser method is most promising as a technology of next generation in terms of both energy efficiency and selectivity. Laser Isotope Separation experiments have long been achieved since early 70's. In convenience the laser isotope separation methods are divided according to the wavelength range used. One of these is the selective photoreaction by tunable infrared laser and the other is the one made by tunable visible or ultra-violet laser.

Table1.Selective photoreaction by tunable IR lasers

Source Laser	Wavelength Conversion	Type of Reaction	Working Materials	
TEA CO_2 Laser	----------	Infrared Multi-photon Dissociation	Halogenated hydrocarbon	CF_3H,$CFCl_2H$,CF_3CClH, Freon123,CF_3COH
				CF_2Cl_2,CF_3I,CCl_3,CF_2ClH
			Hexahalide	Si_2F_6,SF_6,SeF_6,WF_6
			Tetrahalide	SiF_4,$TiCl_4$
			Trichloride	BCl_3
			Metal Alkoxide	$Zr(OC(CH_3)_3)_4$, $Ge(OC(CH_3)_4)$
			Others	OsO_4
		Infrared Excitation +UV Dissociation	Ammonia	NH_3
	Optical Pump NH_3 $CF4$	Single-colored IRMPD	Halogenated hydrocarbon	CCl_3H
			Hexafluoride	UF_6
	Raman $p\text{-}H_2$	Multi-colored IRMPD	Hexafluoride	UF_6
CW CO_2 Laser	----------	Laser Induced Thermal Diffusion	Hexafluoride	SF_6
	----------	Laser Assisted Nozzle Method	Halogenated hydrocarbon	CCl_2FH
			Hexafluoride	SF_6
CO Laser	----------	Laser Induced Chemical Reaction (CRISLA)	Hexafluoride	UF_6

1.1 Selective Reaction with Tunable Infrared Lasers

(1) TEA CO_2 laser based reaction

Infrared Multi-photon Dissociation method (IRMPD) is most widely used by taking advantage of

discrete tunability of CO_2 laser irradiating special molecules which have resonant spectroscopic absorption peaks. Especially halogenated hydrocarbon, hexahalide, tetrahalide ,trichloride and even metal alkoxide are used. Dissociation of molecule like ammonia by infrared excitation with ultraviolet excitation was also applied. In order to convert wavelength of CO_2 TEA laser, optically pumped NH_3 or CF_4 lasers ,especially Raman p-H_2 lasers are used for the dissociation of uranium hexafluoride based on the multi-colored IRMPD.

(2) Reaction by the other types of lasers

Continuous wave CO_2 laser is used for laser induced thermal diffusion and laser assisted nozzle method. In the very special case CO Laser is used for the laser induced chemical reaction (CRISLA).

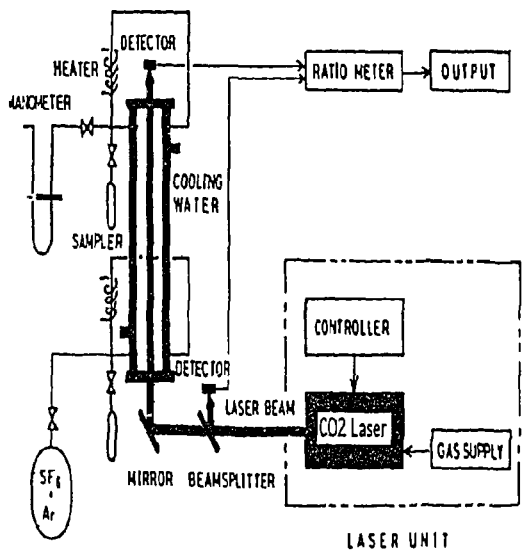

Fig.1 Laser Induced Thermal Diffusion[8]

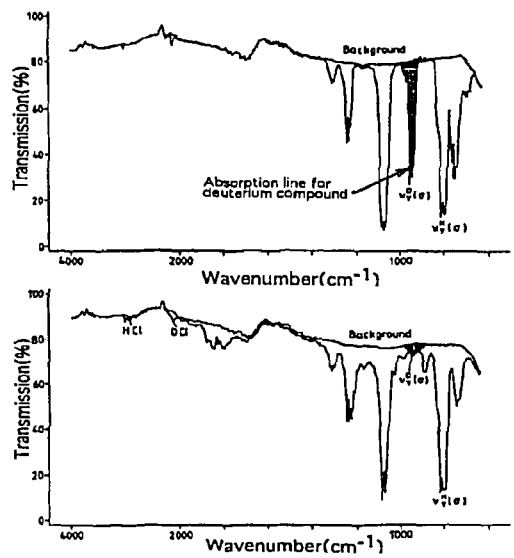

Fig.2 IR Multi-photon dissociation and hydrogen isotope exchange process[8]

Basic Reaction

$^{235}UF_6' + 3h\nu_3 \rightarrow {}^{235}UF_6^* \rightarrow UF_5Cl + HF'$

5.33 μm by CO laser $^{235}\sigma = 6x^{238}\sigma$

· V-T relaxation

$^{235}UF_6^* + HCl \rightarrow {}^{235}UF_6'' + HCl + K.E.$

· V-V scrambling

$^{235}UF_6^* + {}^{238}UF_6 \rightarrow {}^{235}UF_6 + {}^{238}UF_6^*$

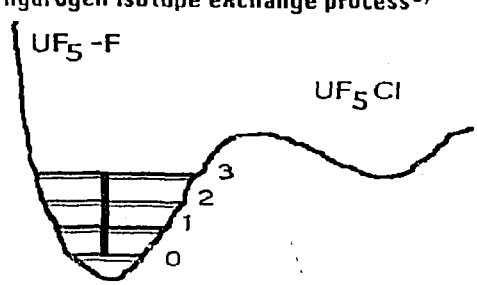

Fig.3 CRISLA method(IR Laser Induced Photochemical Reaction)

1.2. Selective Reaction with Tunable Visible or Ultraviolet lasers

Photochemical reaction by tunable visible lasers is made by a dye laser,which can emit tunable photons with many colors of dyes. CW laser pump dye lasers are used for laser induced chemical reaction photochemical reaction in gaseous/liquid phase and atomic beam deflection. Diode CW lasers are also used for radiation pressure. Pulsed laser are used for photo-dissociation, especially single photon dissociation and for laser induced chemical reaction. But most popular and widely applicable selective reaction is a multistep photoionization.

(1) Single photon dissociation

Single photon dissociation is seen in visible and ultra-violet wavelength region. Typical reaction systems are given for the dissociation of symmetric tetrazine and formaldehyde.

a.Dissociation by Visible Photon

s-Tetrazine($H_2C_2N_4$) + h $\nu \rightarrow$ HCN+N_2

b.Dissociation by Ultra-Violet Photon

H_2CO+ h $\nu \rightarrow H_2$+CO

Combination of ultraviolet dissociation and cascade is proposed by LLNL.[1] Physical and

operational parameters are given as follows.

Table 2. Selective photoreaction using tunable visible/ultra-violet lasers

Source Laser	Wavelength Conversion	Type of Reaction	Working Materials
CW Laser (Pump)	*Dye Laser*	Laser Induced Chemical Reaction	ICl
		Photo-dissociation	H_2CO,$C_2H_2N_4$
		Photochemical Reaction (Liq.)	UO_2F_2+HF+CH_3OH
		Atomic Beam Deflection	Ba
CWLD	----------	Radiation Pressure	Rb
Pulse Laser (Pump)	*Dye Laser*	Photo-dissociation	H_2CO,Br_2
		Laser Induced Chemical Reaction	Li+CCl_2FH,Yb+HCl,$CSCl_2$,$U(BH_4)_4$, $UO_2(HCOO)_2H_2O$
		Multistep Photoionization	Li,Mg,K,Ca,Ti,Fe,Cu,Ru,Zr,Pd,Ba,Hg,Tl, Nd,Sm,Eu,Gd,Dy,Er,Tu,Yb,U,Pu
Pulse Laser		Element Selective Photochemistry	$Eu^{3+}H_2O$

And the following enrichment is expected.

^{13}C 1.1%→~70%(single stage),95%(2 stages)

^{18}O 0.208%→~40%(single stage),95%(2 stages)

^{17}O 0.038%→~7%(single stage),95%(3 stages)

Table 3. Cascaded single photon dissociation process

Physical parameters		Operational Parameters	
Wavelength	290-355nm	Laser power	129W
Photoabsorption cross section	$5x10^{-17}cm^2$	Laser PRF	4350Hz
Spectroscopic selectivity	200(13C,17O),300(18O)	Laser pulse width	40ns
Quantum Yield	0.95	Temperature	300K
Geometry		Photolysis cell length Number of passes	10m 10

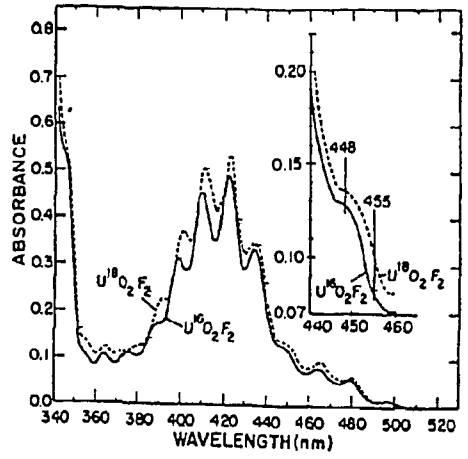

Fig.4 Oxygen isotope separation in liquid phase

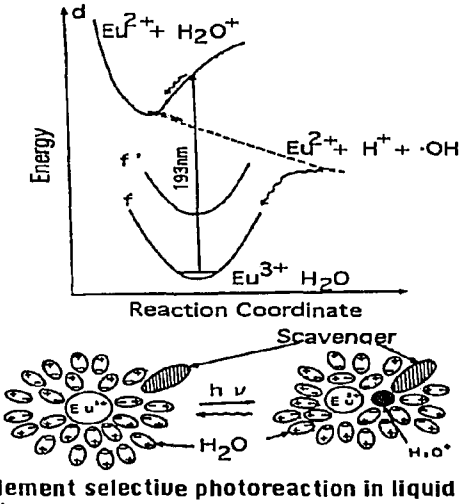

Fig.5 Element selective photoreaction in liquid phase

(2)Laser Induced Selective Chemical Reaction

Photo-decomposition was made with Uranium Boro-hydride (-deuteride)by 2-step Scheme by

E.R.Bernstein(COO-4118-1),in which 2-5% $U(BH_4)_4/Hf(Zr)(BH_4)_4$ was irradiated by tunable laser at low temperature to decompose it as follows;$U(BH_4)_4+h\nu \rightarrow U(BH_4)_3$.Photochemical reaction was also observed in Uranyl Formate in the following way[2],

$UO_2(HCOO)_2H_2O+h\nu \rightarrow U^{+4}, UO_2^{+2}$

(3)Other reaction methods

Laser induced sputtering of SF6 from the cold Kr matrix are made using TEA CO_2 laser ,and Isotopic Selectivity Å3 was obtained when laser illuminated non-target molecules.[3]

Isotope separation in liquid phase was carried out for hydrogen isotope separation based on the photolysis of formaldehyde in liquid xenon [4]

Oxygen isotope separation was made with UO_2^{2+} in methanol solution as follows.[5]

$UO_2F_2+2HF+CH_3OH \rightarrow UF_4+$Organic Products

Photochemical reaction selective to elements but not to isotope was achieved in liquid phase for europium.[6]

$Eu^{3+}H_2O+h\nu \rightarrow [Eu^{2+}H_2O]^* Eu^{2+}+H^++\cdot OH$

$Eu^{2+}+SO_4^{2-} \rightarrow EuSO_4$

2. Atomic Vapor Laser Isotope Separation

As this method is based on the resonant multiple photoionization process, atoms can easily be ionized with low photon flux,which is related to the high utilization efficiency of photons. and the atomic spectrum is enough narrow so that the photons with tunable wavelengths are easily absorbed by the ^{235}U without being absorbed by ^{238}U atoms.Therefore ,this method

- could achieve separation at high separation factor by one single operation.
- could be economical.
- could enrich uranium from the reprocessed uranium ,which leads to the effective utilization of natural resources.

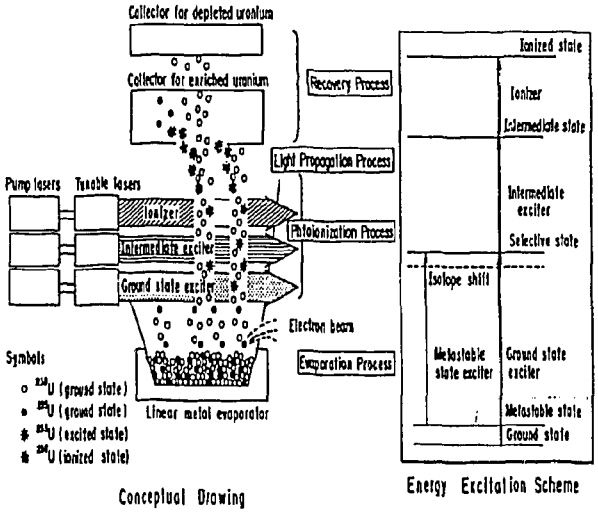

Fig.6 Concept of AVLIS process

Table4.MPI schemes for RIMS

Element	Ionization Potential(cm^{-1})	Types of RIS Schemes covered in the data sheet for this element		
Al	48278	$\omega_1+\omega_1$ $\omega_1+\omega_2+\omega_3$	$\omega_1+\omega_2$ $\omega_1+\omega_2+\omega_3^{AI}$	$\omega_1+\omega_3^{R}$
As	78950	$2\omega_1+\omega_1$	$2\omega_1+\omega_2^{AI}$	
Au	74409	$\omega_1+\omega_1$ $\omega_1+\omega_2+\omega_3$	$\omega_1+\omega_2$ $\omega_1+\omega_2+\omega_3^{AI}$	$2\omega_1+\omega_1$
B	66928	$\omega_1+\omega_2+\omega_3$		
C	90820	$2\omega_1+\omega_1$		
Ca	49306	$\omega_1+\omega_2+\omega_3$ $\omega_1+\omega_2+\omega_3^{AI}$	$\omega_1+\omega_2+\omega_2$ $2\omega_1+\omega_1$	$\omega_1+\omega_2+\omega_3$ $2\omega_1+\omega_2^{AI}$
Cd	72540	$\omega_1+\omega_1$ $\omega_1+\omega_2+\omega_3$ $2\omega_1+\omega_2$	$\omega_1+\omega_2$ $\omega_1+\omega_2+\omega_2$	$\omega_1+\omega_2+\omega_3$ $2\omega_1+\omega_1$
Co	63564	$\omega_1+\omega_1$	$\omega_1+\omega_2+\omega_3$	
Cr	54576	$\omega_1+\omega_1$	$\omega_1+\omega_2$	$\omega_1+\omega_2^{AI}$
Cs	31406	$\omega_1+\omega_1$	$\omega_1+\omega_2+\omega_3$	$2\omega_1+\omega_1$
Cu	62317	$\omega_1+\omega_1$ $2\omega_1+\omega_1$	$\omega_1+\omega_2$	$\omega_1+\omega_2^{AI}$
Fe	63737	$\omega_1+\omega_1$	$\omega_1+\omega_2$	$2\omega_1+\omega_1$
Ge	63713	$\omega_1+\omega_1$ $\omega_1+\omega_2+\omega_3$	$\omega_1+\omega_2+\omega_1$	$\omega_1+\omega_2+\omega_2$
Hg	84184	$2\omega_1+\omega_1$ $\omega_1+\omega_2+\omega_3$	$\omega_1+\omega_2^{AI}$ $\omega_1+\omega_2+\omega_2^{AI}$	$\omega_1+\omega_2+\omega_3$
Kr	12914	$\omega_1+\omega_2$	$\omega_1+\omega_2+\omega_3$	$2\omega_1+\omega_1$
Mg	61671	$\omega_1+\omega_1$ $2\omega_1+\omega_1$	$\omega_1+\omega_2^{AI}$	$\omega_1+\omega_1+\omega_1$
Ni	61619	$\omega_1+\omega_1$	$\omega_1+\omega_2+\omega_3$	
Pb	59820	$\omega_1+\omega_1$	$2\omega_1+\omega_1$	$\omega_1+\omega_2+\omega_3$
Si	65748	$\omega_1+\omega_1$	$\omega_1+\omega_2+\omega_3$	
Zn	75769	$\omega_1+\omega_1$ $\omega_1+\omega_2+\omega_3$	$\omega_1+\omega_2+\omega_1$ $2\omega_1+\omega_1$	$\omega_1+\omega_2+\omega_3$ $2\omega_1+\omega_2$

Whole separation process is divided into four major subprocesses for convenience, ie. evaporation process for generating atomic beam, photoreaction process for ionizing isotopes selectively by resonant multistep photoionization method,ion extracting process for recovering isotopic ions from laser induced plasma, and light propagation process for propagating laser

beams in the optically deep atomic medium with the interaction between photons and specific elements. Some of the typical results from our experiments under the AVLIS (Atomic Vapor Laser Isotope Separation) process are presented[8].

2.1. Versatile applicability

Isotopes of almost all the elements except noble gases can be separated by AVLIS process. Normally visible laser process is more convenient than the ultraviolet laser process. Because in the shorter wavelength region special optical components must be sometimes developed. It is easy to get tunable visible sources using a variety of laser dyes,but tunable ultraviolet sources are obtained by non-linear optical crystals which converts wavelength into shorter one. These crystals suffers from thermal stress or degradation, when the laser power is increased.

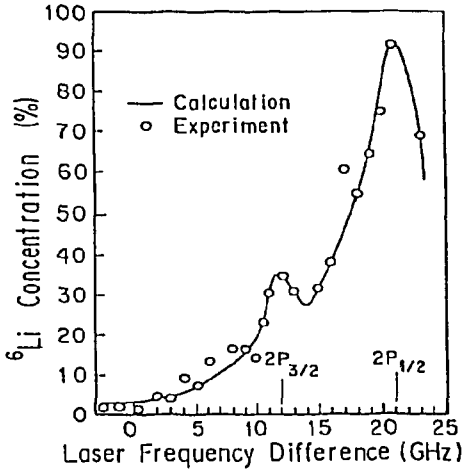

Fig.7 Lithium isotope separation[8]

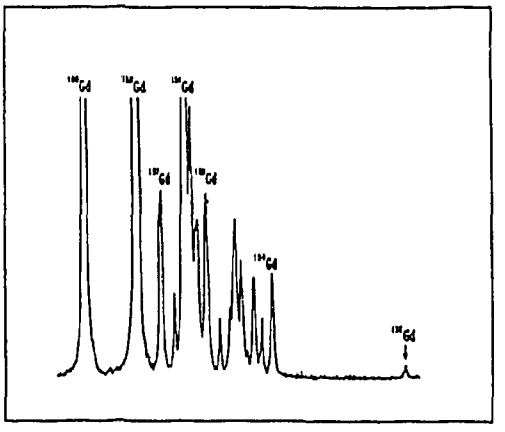

Fig8. Hyperfine structures of Gd[8]

Fig.6 shows the conceptual drawing of AVLIS process based on 3-step photo-ionization method ,in which four subprocesses are indicated. Evaporation process includes the electron beam gun heating and production of atomic vapor.

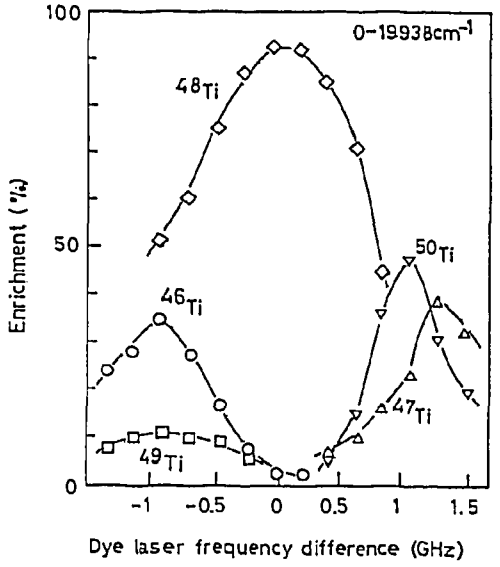

Fig.9 Ti isotope separation[8]

Levels	Allowed transitions	Average population distribution
Photoionization (J = 0)		0%
Second excited (J = 1)		13%
First excited (J = 2)		34%
Ground (J = 2)		53%
m_j (electronic)	−2 −1 0 1 2	m_j

Selection rules for even Gd isotopes with no magnetic field and parallel linear polarization.

Levels	Allowed transitions	Population flow relative to ground state
Photoionization (J = 0)		70%
Second excited (J = 1)		74%
First excited (J = 2)		67%
Ground (J = 2)		100%
m_F (nuclear plus electronic)		m_F
m_j (electronic)	−2 −1 0 1 2	m_j

Selection rules for odd Gd isotopes with no magnetic field and parallel linear polarization.

Fig.10 Polarization selection for Gd[8]

2.2 Selective MPI process

Well-tuned tunable laser beams with different three colors illuminate the isotopic medium at the same time after combining each color. The specified isotopic atom at the ground state is excited to the selective excitation level discriminating the isotope shift. Excited atom is then further excited to the ionized state through the two-step transition,i.e. intermediate excitation and ionization.

In the case that many atoms are thermally populated in their lower metastable states due to the high temperature evaporation,some of those states could be processed by adding another tunable laser beams excite them to the common selective excitation level from the ground state. Multiphoton ionization schemes of several atoms are listed in the NIST table,but precise spectroscopic studies are required for searching the economical isotope separation.[7]

2.3 Development of AVLIS process in large scale

(1) Process development

The photoionization process includes all the above processes. Recovery process consists of the collection of depleted metal and the extraction of product ions.Light propagation process deals the interaction of long traveling laser beams and near resonant medium to increase the production rate. Fig.11 shows the process evaluation scheme to optimize the sub-processes to construct the whole integrated process system.

Under the development program of evaporation process,data are taken on velocity of atoms in the vapor,internal energy of atoms,spatial distribution of vapor ,vapor density and evaporation efficiency. For the photoionization process, spectroscopic data are taken on the energy levels, photo-absorption cross section, excited state lifetime, quantum number,isotope shift and hyperfine structure which are used for optimizing the combination of wavelength,photon flux,detuning.

For ion extraction process,data are taken on the characteristics of laser induced plasma,charge exchange cross section ,scattering rate, sputtering rate to obtain short extraction period of time and low enrichment loss. For the light propagation process ,high density linear evaporator ,4-color tunable laser system are constructed to collect product ions onto the electrode and to evaluate the photon utilization. From these data energy efficiency and scale-up law for separative work are determined.

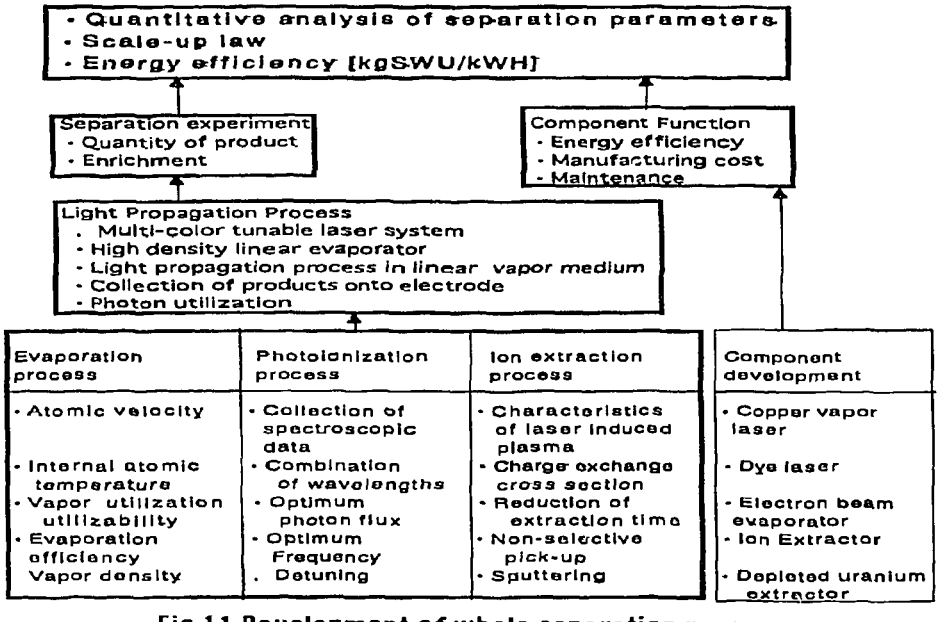

Fig.11 Development of whole separation process

(2) Evaporation process

Several methods are considered for providing atoms into a photo-reactor. Those may be a gas cell,long linear Knudsen cell,two-dimensional nozzle or electron beam gun evaporator with linear crucible. Among above devices an electron beam method is convenient because of its applicability to many kinds of elements with high melting point. In generating atomic vapor, following points are also considered;narrow Doppler width ,generation of high density atomic beam ,high directivity of atomic beam ,well cooled internal energy states ,high energy efficiency ,non-selective ionization of atoms by collision with electrons ,behavior of electrons on the surface of liquid metal etc.

The internal temperature decreases with the increase of the evaporation rate, and atoms can be cooled down more than 1500K. When feed metal is evaporated in the vacuum chamber by electron beam bombardment at very high temperature,which thermally excites atoms from the ground state to several lower metastable states. Highly populated lower metastable states are not appropriate for the efficient photoexcitation, because the laser beams illuminate atoms mainly in the ground state. Atomic vapor thus generated is cooled down by around 1000 K due to the adiabatic expansion,which increases the population of the ground state in the photoreaction zone. In the case that some lower metastable states are considerably populated even after cooling, some of the metastable states are used as target levels.Each population equilibrated at 2500K is around 70% (ground state) and 25% (lowest metastable state) in the case of uranium atom. With the 4-wavelength-3-step photo-ionization scheme more than 90% of feed atoms is ionized from both states. Heating the metal with high power electron beam may cause also thermal ionization and collisional ionization which dilutes the enriched photoions at the photo-reaction zone.

(3) Photoionization process

For AVLIS process, tunable visible lasers are used rather than UV because of their high energy conversion efficiency and easy to control the beam quality. As the visible photon has the energy of 2eV, normally 3-step photoionization scheme is employed to ionize the target isotopic atoms by taking into considerations the ionization potential around 6eV. This multistep process makes use of the real energy levels and is characterized by high ionization efficiency and high selectivity.To implement multi-step photoionization,data related to the energy levels and transition probability between those levels. The former data include the intermediate excitation and ionization levels,parity and total angular momentum represented by J-value. Photoionization includes the direct transition from the intermediate excitation level to the continuous energy spectrum and the autoionization to the high lying levels above ionization limit through the resonant interaction with the continuous spectrum of free electrons. The rate for the former process is faster than the latter,which makes it important to search for the autoionization levels with large photoionization transition probability. Data concerning transition include photoabsorption cross section caused by the electric dipole moment and the total integrated photoionization cross section calculated for the 3-step photoionization. As the former value especially control the total ionization rate, the most important issue is to find out the ionization scheme with large cross section. Spectroscopic data obtained so far are limited to the lower energy region and they were taken using hollow cathode lamps under poor resolution.

Measurements ranges over the energy levels,transition probabilities between levels,isotope shifts and hyperfine structures. Based on the fact that the oscillator strength is independent of isotopes, measurement is made at first stage for the isotopes without hyperfine structure and at the second stage isotope with hyperfine is measured after the characterization of the capability for ionization.

From the view point of the restriction on the wavelength of the tunable lasers pumped by copper vapor lasers,4-wavelength-3step method is adopted, which ionize both the ground sate and the lowest metastable state through excitation levels located at 14000~18000cm^{-1},intermediate levels at 32000~36000cm^{-1},and ionization over the ionization limit 49958cm^{-1}. Data to be taken concerning energy levels,total angular momentum(J-value),parity,autoionization width,hyperfine structure, isotope shift ,photoabsorption cross section ,lifetime, branching ratio ,etc. are not theoretically obtained for heavy elements even with the most advanced computer code. From this point of view JAERI has been collecting spectroscopic data on heavy metallic elements. Measurement method employed is mainly based on the resonant ionization using pulsed laser by which several kinds of optical properties can be obtained by scanning wavelength, power, pulse timing, polarization of tunable lasers. For the measurement which requires high resolution such as hyperfine structure stemming from the interaction between atomic nucleus and orbital electrons, Zeeman-effect, resonant fluorescent tunable lasers with narrow linewidth are employed.

Spectroscopic studies was first done for ^{238}U with zero nuclear spin to obtain data on level,cross section,J-value,lifetime and branching ration, and then optimization is made to give each wavelengths combination the integrated equivalent cross section. Succeedingly data are taken for ^{235}U with non-zero spin to obtain hyperfine structure and isotope shift for the further evaluation. Finally photoionization dynamics was studied by simulating the photoreaction process based on the quantum mechanical model to obtain both the optimum photon flux and detuning.

Photoionization dynamics is generally described by quantum mechanical equations. However when ion extraction rate is fast or when the laser linewidth and absorption spectrum width are wide,coherent interaction is not much dominant. In such cases multistep equation system is roughly converted to the rate equation model without noticeable error.There are two typical methods. One is to use single mode laser with fixed frequency so that the highest ionization rate can be obtained by scanning the laser frequency. Second method is to modify the laser frequency during one single laser pulse so that every part of the atomic spectrum feels the laser frequency for the various laser frequency scanning rate and direction. As a result of calculation, frequency modification method can generally increase the ionization rate efficiently depending the actual

hyperfine structure. Especially ionization of atom with widely spread hyperfine is considerably improved.

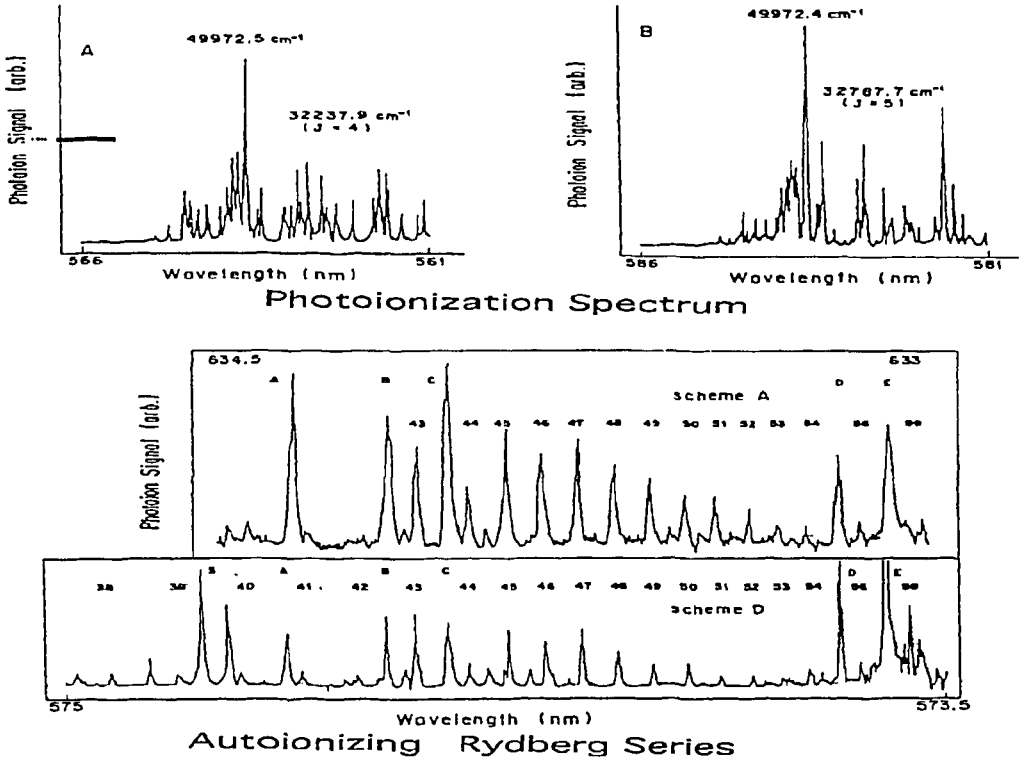

Fig.12 Examples of photoionization spectrum[8)]

(4) Ion extraction process

Generally speaking density of ions extracted with the static electric field from the relatively active plasma with the electron temperature of around 10eV should be lower than 10^{12}ions/cm^3,if effective extraction is expected. Space charge limited current is written by the Langmuir-Child's formula . On the other hand ion current is limited by the electron temperature of the plasma.Based on these experimental data we take the following measures to extract ions effectively from laser induced plasma at low electron temperature. Highest allowable extraction speed is realized by applying voltage as high as possible to collect as much as ions from the moving plasma at the similar speed with the neutral atom.

Higher speed is realized by increasing electron temperature. Avoidance of sputtering from the once collected ions on the electrode due to high momentum of ions, interaction period as short as possible to reduce the chance for charge exchange and ionization collision are also important. From these points of view plasma heating can be considered to enhance the electron temperature, but care must be taken not to heat up electron so high as the ionization limit.

As the AVLIS process is usually applied to the separation of trace amount of isotopic components, the degree of ionization is low, even if 100% ionization of specified isotope is achieved, that is, the ions coexist with huge amount of neutral atoms.For this reason during the extraction of isotopically pure ions from the laser induced plasma,charge exchange easily occur by the collision between ions. The charge exchange cross section is obtained as a function of collisional energy for the reaction system of $^{238}U + {}^{235}U^{+} \rightarrow {}^{238}U^{+} + {}^{235}U$, which is found to be similar to the semiempirical formula introduced by S.Sinha. Scattering of neutral atoms in the atomic beam is also important. We measure the adhesion of the neutral atoms as a function of atomic density. As a result of the experiment the adhesion rate of neutral atoms from the atomic beam onto the electrode is turned out to be mainly caused by two body scattering process. Special care should be taken in order to avoid charge exchange and scattering from the neutral atoms.

(5) Light propagation process

In the application of AVLIS process to the enrichment plant tunable laser beams interact with the specified atomic isotope in the long path of medium aiming at the enhancement of the reaction volume up to the order of m^3 which is the product of cross sectional area and path length of the laser beam. This long propagation also enhances the photon utilization

efficiency. During the propagation in the reaction medium the wavefronts of the laser beams are degraded by nonlinear effect.Taking into considerations the atomic beam velocity and the laser beam width, thermal blooming phenomenon does not occur because of the short pulse width, but the self (de-)focusing is observed due to the local change of the diffraction index.This phenomena are caused not only by the resonant photoreaction of the specified isotope but also by the near resonant interaction with nonspecified isotopes.After the self focusing and self trapping phenomena, laser beam might make a filamentation or decrease the upper state population by superradiance. These phenomena induce big changes in spatial distribution of laser beam intensity and frequency which brings out inefficiency in the separation process. We measured the change of a laser beam radius in a small detuning region of the tunable laser frequency for the two level system in the medium with the long propagation distance.

Measurement was preliminary made concerning the effect of light propagation on the temporal ,frequency and spatial region using heat pipes. After the experiment we started to measure the light propagation effect through the long atomic vapor of uranium atoms as a function of detuning.Simulation of spatial distribution of laser energy propagating through the near resonant isotopic medium was analyzed based on the density matrix equation of motion combined with Maxwell's propagation equation. Distinguished deformation is observed.

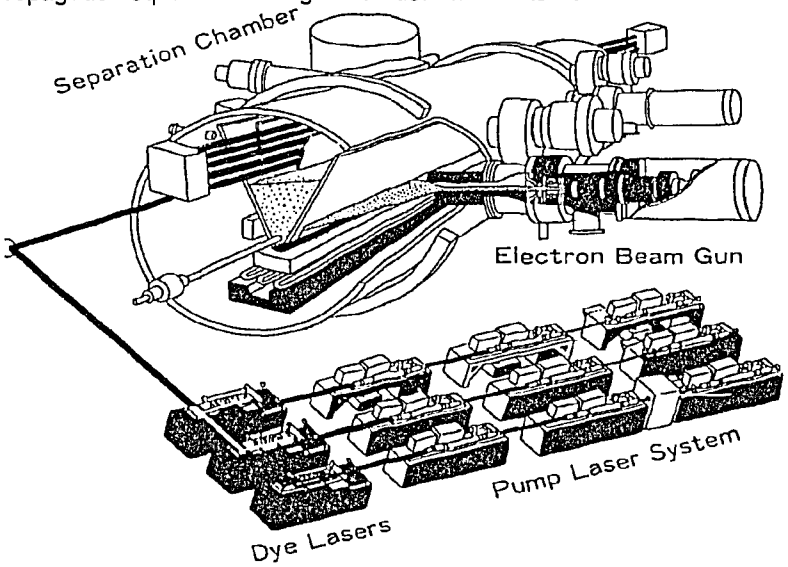

Fig.13 Experimental set-up for evaluating separative power

(6) Evaluation of separative power

Separator is a target cell for tunable laser beams in which long linear atomic beam is generated so that the laser beam could interact as much as atoms to increase the reaction volume. Fig13 shows the conceptual drawing of the electron beam bombardment type evaporator for generating metallic vapor. For most of atoms with high melting point, electron beam guns with either transverse type cathode or Pierce type cathode can be used. Linear cathode type has no scanning mechanism,whereas Pierce type has scanning mechanism to get uniformly distributed atomic density along the linear photoreactor. We constructed one meter long linear evaporator with scanning electron beam gun which is operated by two Pierce type electron beam guns. Cares must be taken so that the laser beam colors could cover all the atoms spread in the frequency field by inducing AC Stark effect to broaden spectrum, by chirping laser frequency or by operating lasers with multiple longitudinal modes.

After optimization in each subprocess mentioned above, many parameters regarding operational conditions are fixed based on the simulation for the construction of the most appropriate separation process. Then the enrichment experiment is achieved by extracting small amount of product, from which total separative power is obtained. When we look at the distribution of the product along the long linear electrode,we find that around the central part of the electrode the production rate is highest due to the high adhesion rate of scattered atoms. The distribution of the recovered products along the linear electrode was obtained. In the collection of ions from the moving plasma with the linear electrode, neutral atoms tend to scatter with each other, which makes neutral atoms to attach to the electrodes and dilute enriched product.

Separative power is calculated based on the experimental values.We also propose two unique parameters which can describe the process performance based on the above

separative power. One of them is the energy efficiency,which is defined by the separative power per unit energy consumption ,ie. gSWU/kWh. And the other one is the specific separative power,which is defined by the separative power per unit volume per pulse.Energy efficiency is naturally related to the energy consumption and specific separation power is related to the scaleup performance.

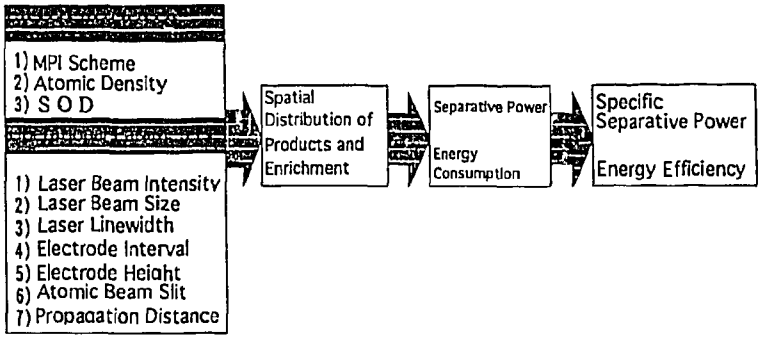

Fig.14 Procedures for evaluating separation process

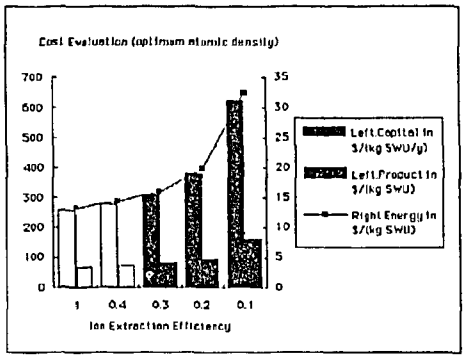

Fig.15 Example of cost estimation

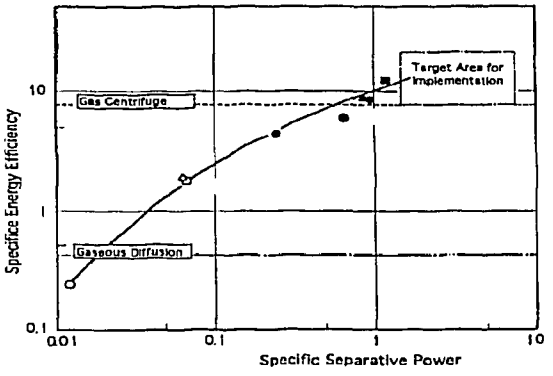

Fig.16 Status of whole process evaluation

3. Summary

Fig.15 shows the typical example of cost estimation where cost depends on how efficiently ions are extacted from the plasma towards electrode under the high atomic density. Several typical results are plot in fig.16 in the plane of these two parameters to show the progress status of AVLIS technology. AVLIS process may lower the uranium enrichment cost.In order to implement the laser isotope separation process economically for various kinds of elements, innovations are expected especially in the development of compact and efficient light sources,the photoreaction process and the low loss recovery process.

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