PATENT SPECIFICATION

(11) **1 595 216**

(54) ISOTOPE SEPARATION

(71) We, JOHN HOWARD COLEMAN, of Overlook Road, Locust Valley, New York 11560, United States of America, and TOBIN JAY MARKS, of 1410 Chicago Avenue, Evanston, Illinois 60201, United States of America, both citizens of the United States of America, do hereby declare the invention, for which we pray that a patent may be
5 granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:-

This invention relates to isotope separation. More particularly, the invention is directed to methods irradiating the isotope compounds selectively to change their chemical (and/or physical) properties, thereby to facilitate separation.

10 Isotope separation is an area of profound importance not only to chemists and physicists but to the entire populace as well. The use of infra-red lasers to separate isotopic species is suggested in the prior art. Described techniques involve selectively exciting gaseous molecules of a given isotope with intense, highly monochromatic laser radiation. It is required that the frequency of the radiation be coincident with a fundamental,
15 combination, or overtone in the vibrational spectrum of the molecule. The vibrational excitement of the molecules may lead to spontaneous decomposition, or it may be necessary to introduce a second agent, which reacts selectively with the excited species. In some cases, simultaneous laser and ultraviolet or visible radiation have been employed. In all cases, the objective is selectively to transform the molecules of a single isotope into an
20 enriched material which can be separated (on the basis of solubility, volatility, electrical charge, etc) from the molecules containing the other isotope(s). To prevent energy transfer beween isotopically different molecules, it may be desirable to operate at low pressures and/or to add an inert diluent.

The above concepts have been proposed for the photo-excitation separation of the
25 uranium isotopes U^{235} and U^{238}. U.S. Patent No. 3,443,087 describes the selective ionization of UF_6 by a combination of infra-red (IR) and ultra violet (UV) laser irradition to facilitate electrostatic separation of U^{235} in the vapour phase, and Belgian Patent No. 821,823 describes selective dissociation of UF_6 by a combination of IR and UV, facilitating chemical separation of U^{235}.

30 However, the fundamental vibrational resonance frequencies of UF_6 are 624 cm^{-1}, and 186 cm^{-1}, values which are not presently available from electrically pumped transitions in gas lasers. Semiconductor lasers are available which can be tuned to this frequency, but their output power is limited to fractions of a watt. This low power results in an intensity that is insufficient for multiphoton absorption to cause dissociation. Any reactions induced
35 would occur at low rates which are impractical for production processes. Other known separation processes such as those according to UK Patent No. 1,284,620 describe a method of isotope separation by selectively stimulating chemical reactions effected by irradiating an isotope mixture with a laser frequency corresponding to the vibrational absorption frequency. Specifically, it is there disclosed to use a CO_2 laser in combination with the
40 molecules UF_6, UF_nCl_{6-m}, UCl_6, or $U(BH_4)_4$. However, none of these molecules is known to have fundamental vibration frequencies in the CO_2 laser output range. Since the most stable and reliable, and the most economical source of infra-red laser radiation is a carbon dioxide gas laser, the most effective range of which is 9.2-11.0μ, 1086-909 cm^{-1}, the above molecules would not absorb sufficient radiation for usable excitation. None is reported to
45 have fundamental or overtone metal-ligand vibrations in the appropriate region of the

spectrum.

The present invention provides a process for separating uranium isotopes, which comprises: preparing a volatile compound U-T in which U is a mixture or uranium isotopes and T is a chemical moiety containing at least one organic or deuterated borohydride group, and which exhibits, for at least one isotopic species thereof a fundamental, overtone or combination vibrational absorption excitation energy level at a frequency between 900 and 1100 cm^{-1}; irradiating the said compound in the vapour phase with energy emitted by a radiation source at a frequency between 900 and 1100 cm^{-1} and thereby differentially modifying properties of at least one isotopic species of the said compound to establish differences in properties of the irradiated isotopic species facilitating a separation of isotopic species by physical or chemical means applied to the irradiated compounds and to products derived therefrom; and utilizing the established differences in properties to selectively recover a specific uranium isotope from the irradiated compound.

The compound is preferably one in which the U-T bond has a fundamental, overtone or combination vibrational absorption frequency between 900 and 1100 cm^{-1}.

The invention can be performed in apparatus comprising: a reaction chamber for retaining a chemical composition including a plurality of isotopic species of uranium, a valve means for controlling introduction of the chemical composition into the reaction chamber, means for controlling the atmospheric composition within the reaction chamber, temperature control means for regulating the temperature in the reaction chamber, a generator including a power supply for producing a beam of radiated energy, and means including electrode means to direct the said beam to impinge upon the said chemical composition contained in the reaction chamber to interact with and to modify the properties of the chemical composition so as to facilitate the separation of isotopic components thereof.

It is an important feature of the present invention that it teaches the use, in laser induced separations of uranium isotopes, of compounds having vibrational sensitivities accessible to a CO_2 laser.

In achieving the aims of the present invention, the compounds must be sufficiently volatile to be irradiated in the gas phase. The molecules must have fundamental or overtone (preferably the former and preferably metal-ligand) vibrations in the region accessible to a CO_2 laser, i.e. 900-1100 cm^{-1}. These criteria encompass a surprisingly wide range of compounds.

Coordination number and coordinative saturation appear to play a major role in determining the volatility of neutrally charged uranium compounds. In low oxidation states, e.g. +4, a uranium complex with non-bulky ligands will attempt to expand its coordination number beyond 4 by coordinating additional ligands, or, when these are unavailable, by forming ligand bridging bonds to neighbouring uranium ions. This leads to polymeric structure, high lattice energies, and low volatility. Two approaches can be used to circumvent this problem. In higher oxidation states, the uranium will have a smaller ionic radius, hence a smaller desired coordination number, and generally more ligands per metal ion. This usually lead to higher volatility. This is illustrated by UF_4 and UF_6 although these particular compounds are not suitable for use in this invention.

UF$_4$

UF$_6$
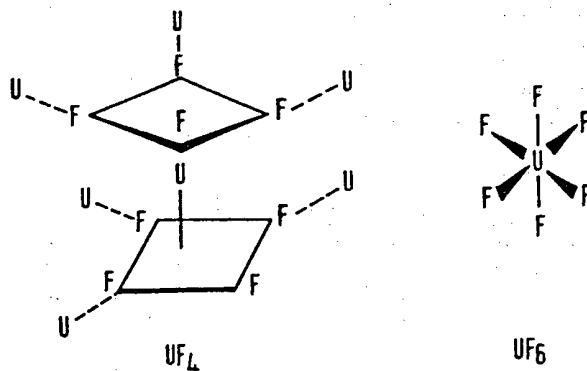
(distorted square antiprism)   (octahedral)

Another way to reduce intermolecular interactions is by employing sterically bulky ligands. For example, in the alkoxide series, U $(OCH_3)_4$ is essentially non-volatile, insoluble, and is probably extensively associated in the solid state. In contrast, $U[i\text{-}OC_3H_7]_4$ is more soluble and can be sublimed. Further confirmation of these concepts is found in $U(OC_2H_5)_5$ which

is dimeric and rather volatile (boiling point 123°/0.001 mm pressure) and in $U(OC_2H_5)_6$ which is monomeric and even more volatile (boiling point 72-74°/0.001 mm pressure). Thus, maximizing the oxidation state and the coordinative saturation are feasible approaches to increased volatility. Also important, but apparently less critical, is keeping the molecular

5   weight as low as possible.

The foregoing discussion is believed to be relevant to reaction design for uranium isotope separation. One attractive strategy is to employ those laser-induced reactions which dissassociate a ligand (or ligands) from the uranium, or which promote the substitution of a less bulky ligand. Starting with, for example, a U(VI) compound, laser promoted homolytic

10   cleavages would reduce the uranium to the accessible +5 then +4 oxidation states.

$$UL_6 \longrightarrow UL_5 + L$$
$$UL_5 \longrightarrow UL_4 + L.$$
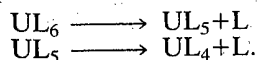
15   The UL_5 and UL_4 species are entities markedly less volatile than UL_6 and are separable. Alternatively, reactions such as

$$UL_4 \longrightarrow UL_4^* \xrightarrow{\;L'\;} UL_3L' \quad +L$$
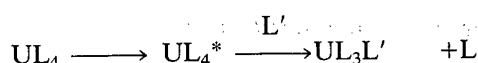
20   produce a U(IV) complex with different properties (e.g. volatility, solubility) than UL_4, since the L' is less bulky and may also form intermolecular bridges.

The compounds selected for use in the first embodiment must be stable to the thermal conditions necessary to volatilize them. In the Pearson terminology, uranium is a "hard"

25   acid. It will have the greatest affinity for negatively charged, non-polarizable ligands. Such qualitative rules are relevant in developing syntheses and also in promoting, via laser irradiation, ligand substitution reactions. Fortunately, many of the properties which impart volatility afford, in addition, some resistance to thermal degradation. Studies of the mechanism of thermal decomposition of several classes of uranium organometallics indicate

30   that coordinative saturation of the uranium ion can greatly retard thermal decomposition. This presumably arises because immobilization of the coordination sphere blocks the lower energy pathways for thermolysis. Thus, the approaches which should foster high volatility are, in addition, reasonable initial approaches to enhance thermal stability. Such concepts clearly illustrate that decomposition of the desired molecules by energetic photons

35   facilitates separation from undecomposed molecules by the resulting differences in chemical and physical properties.
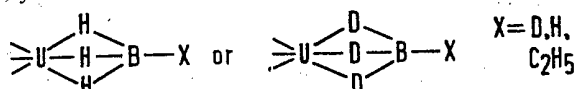
The following is a partial enumeration of useful classes of compounds, for the purpose of the present invention. The frequency of the metal-ligand stretching (and to a lesser extent, deformation) fundamental is important in the practice of the invention. The magnitude of

40   the frequency difference in $\nu \, 238_{U-X}$ versus $\nu \, 235_{U-X}$ will depend on the particular molecular system.

For uranyl complexes ($O=\overset{+2}{U}=O$) the difference should be ca. 0.7 cm^{-1} for the antisymmetric $O=U=O$ stretch. Application of the Teller-Redlich product rule yields a value for

45   the T_{1u} U-O stretching mode of ca. 1.5 cm^{-1} in $U(OCH_3)_6$, treating the methoxy ligands as point masses (a crude qualitative approximation). The most accurate analysis of metal ligand fundamentals is through high resolution metal isotopic substitution studies, an area which has recently seen rapid development.
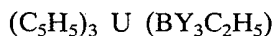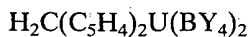
A number of known volatile uranium complexes, not suggested for such use in the

50   literature, are, in accordance with the invention, employed for the laser-induced separation of uranium isotopes. These compounds have metal-ligand fundamental or overtone vibrations in the spectral region accessible to a CO_2 laser, and, as herein disclosed, are susceptible to isotope-selective excitation and interception. Arranged by class, compounds useful in the practice of the invention include the following

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A. *Borohydride-Organo Complexes*

These complexes of the BH_4^- ligand have been found to have a tridentate metal-ligand geometry. Under C_{3v} local symmetry, the A_1 and E bridging modes, which are believed to have considerable ν M-H character, occur in the region ca. 1100-1230 cm^{-1}. All have

60   sufficient volatility to make them attractive.

65   Examples are given below.

$U(BY_4)_4$

$H_2C(C_5H_4)_2U(BY_4)_2$

$(C_5H_5)_3 \; U \; (BY_4)$ (Y=H,D)

$(C_5H_5)_3 \; U \; (BY_3C_2H_5)$

$(C_4H_4N)_3U \; (BY_4)$

Most of these complexes can be prepared by displacing halide with BH_4^- or BD_4^- in an ethereal solvent. Indeed, we have found borohydride derivatives to be some of the most accessible for a number of organoactinide systems studied.

$$\geqslant\text{U-Cl} + NaBY_4 \longrightarrow \;\; \geqslant UBY_4 + NaCl$$
$$(\text{or } LiBY_4)$$
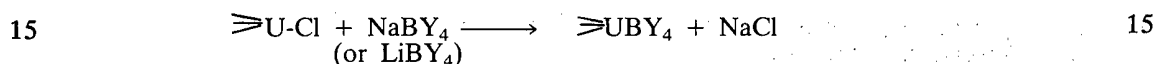
Additional volatile derivatives are prepared by sequences such as

$$U(OR)_n \xrightarrow{\;HCL\;} U(OR)_{n-1}Cl \xrightarrow{\;NaBH_4\;} U(OR)_{n-1}BH_4$$
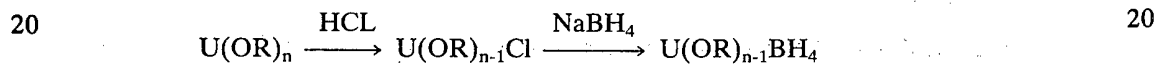
In several of the compounds, for example $U(BH_4)_4$, the desired normal mode is above the frequency accessible to the CO_2 laser (for $U(BH_4)_4 \; v = 1230 \; cm^{-1}$). Deuteration shifts this absorption to ca. 926 cm^{-1} and the deuterated compounds can thus be employed in this invention. Deuterated compounds are accessible via commercially available $NaBD_4$ or $LiBD_4$.
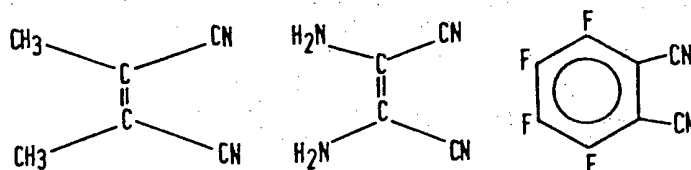
B. *Uranyl Complexes*

These are complexes which incorporate the O=U=O functionality, usually in a pantagonal (A) or hexagonal (B) bipyramidal arrangement (a few octahedral examples are also known). The antisymmetrically coupled v O=U=O stretching vibration usually occurs in the region 910-940 cm^{-1}.
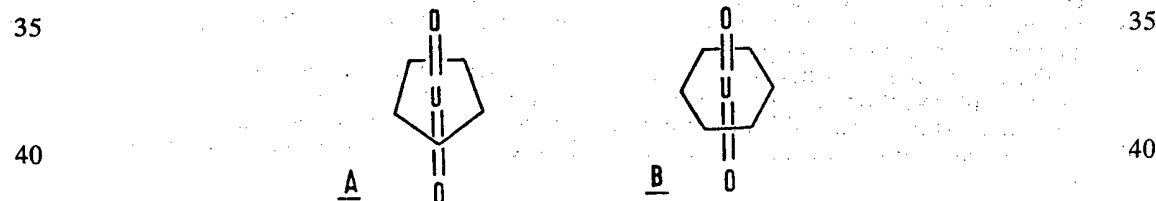

Another compound for laser irradiation is uranyl superphthalocyanine, prepared in a template reaction as shown below.

$M = UO_2^{+2}$
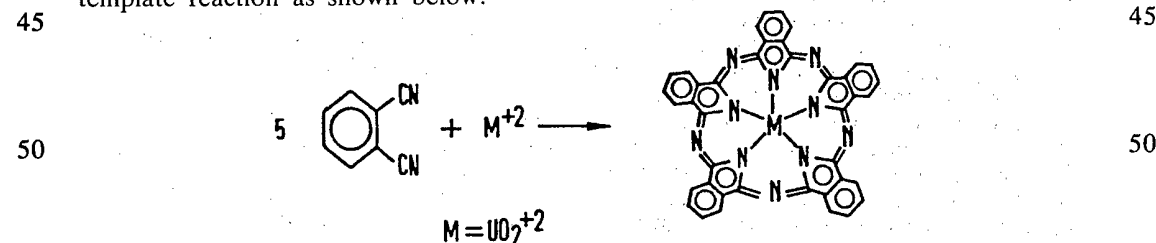
This compound has very high chemical and thermal stability and sublimes *in vacuo* at 400°. Still other organic "monomers" providing macrocyclic uranyl complexes of lower molecular weight, and greater volatility include:

Uranyl alkoxides, $UO_2(OR)_2$, also reported in the literature, are generally of low volatility, and are particularly suited for the second embodiment. Complexes with bulky fluorinated alcohols are also useful in the practice of the invention. While known uranyl β-diketonates are not appreciably volatile, very bulky ligands such as fod, shown below, complexed to uranium are of interest.
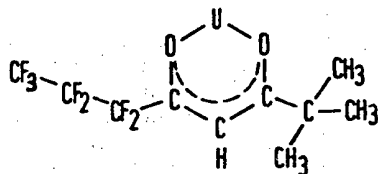
Also, aluminium alkoxide ligands impart volatility. A possible route to such complexes for U(VI) is

$$UO_2Cl_2 + 2NaAl(OR)_4 \longrightarrow$$

$$+ 2NaCl$$
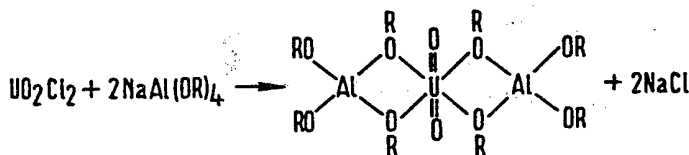
C. *Uranium Alkoxides and Siloxides*

Complexes with OR ligands may be prepared for uranium on a large scale and in a variety of oxidation states. Many are quite volatile; some are distillable liquids. The known analogous silicon compounds are also appreciably volatile. Representative species are shown below, and it is apparent that they offer a wide flexibility in terms of possible variation of oxidation state, symmetry, and volatility.

$U(OR)_4$ $U(OSiR_3)_4$

$U(OR)_5$ $U(OSiR_3)_5$

$U(OR)_5$ $U(OSiR_3)_6$

R = variety of organic groups

A U-O fundamental (except possibly for uranium in the +6 oxidation state) will not absorb in the desired frequency region. For example, the Ti-O stretches in $Ti(OCH_3)_4$ have been assigned at 588 and 553 cm^{-1}, those in $Ti(OC_2H_5)_4$ at 625 and 500 cm^{-1}, and those in $Nb(OEt)_5$ at 571 cm^{-1}. Though the fundamental U-O stretches in the aforementioned alkoxides may not be in the CO_2 laser range, overtones are. The fundamental U-O stretch occurs in the region 460-550 cm^{-1}. Hence, the first overtone is in the region 920-1100 cm^{-1}. By appropriate selection of ligands to "tune" the symmetry and various force constants, it is possible to bring a U-O overtone or combination band into Fermi resonance with another fundamental (e.g. ν C-O $\approx$ 1000 cm^{-1}), thereby increasing the excitation cross section.

The uranium alkoxides also appear to be excellent precursors to other derivatives. Typical reaction schemes to introduce a variety of new ligands are shown below.
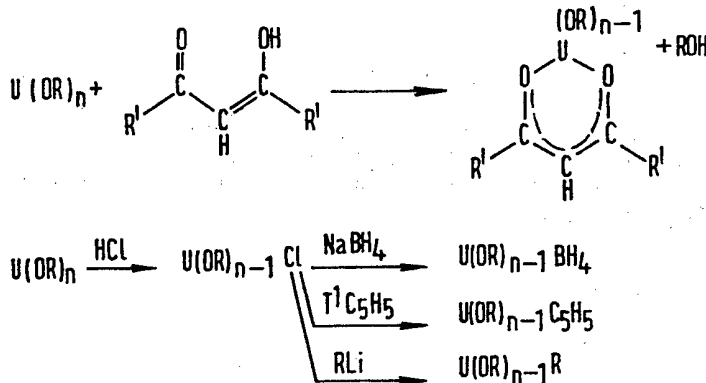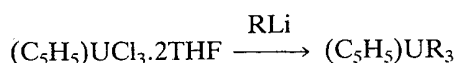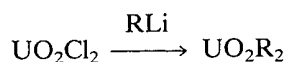
R = alkyl group

5   Mixed complexes in which one or more of the OR or $OSiR_3$ groups is replaced by a β-diketonate, a borodeuteride, or an alkyl group may also be used.   5

D. *Uranium Alkyl Compounds*

10   These compounds include complexes of the formula L_n UR_{n-4} where $L=C_5H_5$, an alkoxide or siloxide, or borodeuteride, and R is an alkyl (e.g. methyl, ethyl, isopropyl, t-butyl, benzyl, neopentyl) or aryl (e.g. phenyl) group. The U-C stretching - vibration   10 occurs in the region of 500 cm^{-1}. Thus, the first overtone is in the region of 1000 cm^{-1}. Uranium alkyl compounds are thermally stable only if sufficient coordinative saturation is present to hinder various intra-molecular decomposition processes. Thus $(C_5H_5)_3$ UR compounds (with π -bonded C_5H_5 ligands) possess rather high thermal stability. For R= 15   methyl or vinyl, the complexes are sublimable.   15
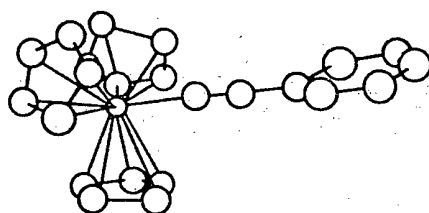

X-ray structure of $(\eta^5\text{-}C_5H_5)_3U(\text{-}C\equiv CC_6H_5)$ Systems such as $(C_5H_5)_3UR$ offer the attractive feature that products of a homolytic laser-induced cleavage would be $(C_5H_5)_3U$, which is nonvolatile, and R-radicals, which could be scavenged. Other attractive uranium 30   alkyl systems are presented below.   30

$$CH_2(C_5H_4)_2 \ UCl_2 \xrightarrow{\ RLi\ } CH_2(C_5H_4)_2UR_2$$

35          $$UO_2Cl_2 \xrightarrow{\ RLi\ } UO_2R_2$$ 35

$$(C_5H_5)UCl_3.2THF \xrightarrow{\ RLi\ } (C_5H_5)UR_3$$

40   THF = tetrahydrofuran   40

Particularly interesting, in terms of imparting thermal stability, are bulky R groups (e.g. t-butyl), those without β-hydrogen atoms (e.g. tri-methylsilylmethyl), and chelating alkyls 45   (e.g. those derived from 1,4-dilithiobutane).   45
The stability of $W(CH_3)_6$ suggest the utilization of $U(CH_3)_6$, which is expected to be quite volatile.

50        $$UCl_6 \xrightarrow{\ CH_3Li\ } U(CH_3)_6$$ 50
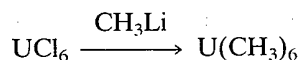
E. *Uranium Amides*

The diethylamide or uranium $U[N(C_2H_5)_2]_4$ is volatile enough to be distilled or sublimed. Until recently, it was the only known uranium dialkylamide. Since the U-N stretch falls in 55   the 500 cm^{-1} region, the first overtone is in the region of 1000 cm^{-1}. The amido complexes of   55 uranium contain the U-NR_2 functionality: examples include $U(NR_2)_4$, L_n $U(NR_2)_{n-4}$ where L = B diketonate, alkoxide, or siloxide, alkyl group, or borodeuteride. Some approaches to a variety of compounds are proposed below.

$$U(NR_2)_4 \xrightarrow{\ C_5H_6\ } (C_5H_5)_2 \ U(NR_2)_2$$
$$\searrow^{\ nROH} \ \ U(NR_2)_{4-n}(OR)_n$$
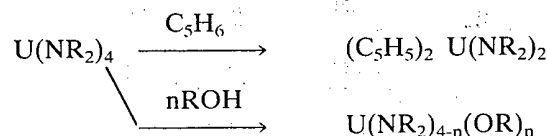
The existence of $W[N(CH_3)_2]_6$ is impetus to investigate uranium oxidation states higher than +4.

In the above cases, the complexes may be irradiated in the gas phase at the fundamental, combination, or overtone metal (either U^{238} or U^{235}) ligand frequency, with a high power CO_2 gas laser. This is carried out with or without simultaneous ultraviolet or visible irradiation. Reactive agents such as methanol, styrene and thiols, can also be added to trap the excited molecules. The reactions are preferably conducted at low pressures, and it is possible to add an inert diluent such as methane, neon or argon. In all cases, the selectively excited molecules react to form an isotopically enriched (in U^{238} or U^{235} depending on the irradiation frequency) product which is chemically different from the starting material. The product is then isolated from the starting materials on the basis of volatility, solubility, and/or electrical charge.

The invention will be further described, by way of example only, with reference to the accompanying drawings, in which:

Figure 1 illustrates apparatus for carrying out one embodiment of the invention in which a pulsed CO_2 laser, controlled in frequency and intensity, is used to irradiate a gaseous U-organic isotope mixture; and

Figure 2 illustrates apparatus for carrying out a second embodiment in which the radiation from a CO_2 laser is combined with a UV source to irradiate a gaseous U-organic isotope mixture.

Referring now more particularly to Figure 1 of the drawings, a gaseous mixture of U^{235} and U^{238} incorporated in a U-organic compound, for example, a uranium alkoxide $U(OEt)_6$, is irradiated in a reactor 10 by a laser beam 12 from a pulsed CO_2 laser source 16. The reactor includes a one meter long 'Monel' (Trade Mark) reaction tube 18 having an entry window 20 for entry of the laser beam 12 and an exit window 24 for exit of the beam 12. The beam entry and exit windows 20, 24 may be of NaCl or other suitable materials such as ZnSe that transmit the 9-11 micron CO_2 laser radiation. The windows 20, 24 are positioned at the Brewster angle to the beam 12 to minimize loss of beam energy. A photo-detector 26 monitors the unabsorbed portion of the beam 12, and a monitor window 28 of quartz, connected by a side tube 32 to the reaction tube 18, may be used to monitor the visible and UV fluorescence from the reaction. The U-organic composition is stored in a reservoir 34, fitted with a heat exchange coil 34a, and a reactive or buffer gas may be stored in a second reservoir 36.

The U-organic composition (or the U-silicon composition) is introduced into the reaction tube 18 through a metering valve 38 which may incorporate a converging-diverging nozzle 40 for expansion cooling, designed as is well known in the art to provide a velocity of approximately Mach 1 or larger. A reaction or buffer gas in the reservoir 36 is introduced (when desired) into the reaction tube 18 through a second metering valve 42. A heating coil 44 adjusts the temperature in the reactor 18 for the desired vapour pressure. A vacuum pump 46 exhausts the gases in the reaction tube 18 through an aperture 48 to a second chamber or zone 54 the temperature of which is controlled by a heat exchanger 56. A third chamber or zone 54a may be used to condense unreacted components as well as volatile by-products. The pump 46 exhausts into a storage tank 58. The tetra-alkoxide, $U(OEt)_4$, is a principal reaction product, together with organic fragments. The less volatile tetra-alkoxide is then separated from the more volatile constituents of the mixture to achieve the desired isotope enrichment. A focusing lens 60 focuses the laser beam 12 through the entry window 20 to a spot on the optical axis of the reaction tube 18 to position the reaction above the nozzle 40 of the valve 38.

The laser source 16 uses a pulsed electrical discharge between a pair of bronze electrodes 62, 64 to attain output power beween the limits of 10^4 and 10^{10} watts per cm^2. The upper value of beam intensity is limited in some cases by breakdown of the gases and formation of a plasma. The electrodes 62, 64, in a preferred embodiment of the invention, are 50 cm long x 2 cm wide and are separated by 1 cm. The lasing gas, which may be a 1,1,8 molar mixture of CO_2 N_2 and He stored in a pressurized tank 68, is introduced through a reducing valve 70 into a circulating and cooling system 72 of the laser source 16. The electrodes 62, 64 are symmetrically positioned above and below the axis of an acrylic laser tube 76. The tube ends are sealed by Brewster angle, NaCl end windows 78, 80. An optical resonator is formed between an adjustable grating 84 and a 95% reflecting germanium (Ge) mirror 88 which are centered about the optical axis (which is slightly off-set from the centre axis of the electrodes 62, 64 when the beam passes through the end window 78, 80). Depending on whether a stable or unstable resonator is desired, the Ge mirror 88 may be convex, flat, or concave, as presented to the grating 84. The grating 84 acts as a plane, frequency-selective mirror which is adjusted for the particular U-organic used. The grating 84 and the Ge mirror 88 may be positioned inside the laser tube 76 eliminating the end windows 78, 80. However, wide mirror spacing is desired to improve frequency characteristics.

Electrical power to the electrodes 62, 64 is supplied by a power supply 92, 94 through suitable leads 96, 98. The electrical pulse from supply 92, 94 is a fast rise-time pulse of a typical magnitude 47 KV per cm of spacing between the electrodes 62, 64 and per atmosphere of lasing gases. The power supplies 92, 94 may be a Blumlein or equivalent design with low-inductance circuit elements. Alternatively a commercial, triggered spark-gap may be used for the power supply 92, in which case a lead 96 is connected directly to the ground, eliminating supply 94.

To facilitate a uniform glow discharge at high lasing gas pressures in the laser tube 76, a thin (.007") tungsten wire 102 is positioned at mid-plane but outside the edges of the electrodes 62, 64 and connected through a small capacitor 104 (typically 50 pF or less) to a lead 98. Thus, the rising voltage from the power supply 92, which increases the potential difference between the electrodes 62, 64, also increases the potential difference between a wire 102 and the electrode 62. However, the Townsend breakdown voltage between the wire 102 and the electrode 62 is adjusted to be lower than between the electrodes 62, 64 thereby causing limited breakdown and preionization of the lasing gas prior to breakdown between the electrodes 62, 64. Other pre-ionization circuits, may be used such as naked spark-gaps positioned outside the electrodes 62, 64 along the mid-plane to UV-irradiate the gap between the electrodes 62, 64. Also, a low ionization-potential liquid such as tri-butylamine may be applied to the electrodes 62, 64 to reduce sparking.

Hot lasing gases from the gap between the electrodes 62, 64 are recirculated and cooled through a cooling tube 106 of a refrigerator 72 by ducts 110 112 by means of a pump (not shown). A cooling coil 116 extracts the heat from the lasing gases, added by power absorbed from the power supply 92 during the electrical discharge. The ducts 110, 112 are ideally located to circulate lasing gases directly through the space between the electrodes and may use a diffuser if turbulence is required to stabilize the discharge.

The most convenient operating pressure of lasing gases in the laser tube 76 is atmospheric. However, the value can be above or below one atmosphere, depending on desired output characteristics. In some cases it may be necessary to raise the pressure to attain tunability above the natural transitions as broadening of the CO_2 lines increases 3GHz per atmosphere up to 10 atmospheres where CO_2 laser lines overlap.

Thus, in operation, the laser source 16 is switched on aligned with the optical axis of the reaction tube 18 of the reactor 10. To obtain the proper output frequency selection of the laser source 16, the reaction tube 18 is evacuated and backfilled with U-organic compound enriched in U^{235}. Absorption is noted while laser lines are selected with the grating 84. The U-organic feedstock in the reservoir 34 is then admitted through the nozzle 40 and irradiated. Non-volatile reaction products are condensed in a chamber 54 and non-volatile products as well as reacted U-organic are condensed in a second cold zone 54a.

Of course, other U-organics may be used with the laser source 16 tuned by the grating 84 to the desired frequency and with the intensity selected to the proper value. In each case the reaction proceeds from the focal spot back upstream towards the laser source 16 until the intensity falls below the minimum value.

The apparatus shown in Figure 2 differs from that shown in Figure 1 primarily in the arrangement of the "laser source", two sources being used in the Figure 2 embodiment. For the most part, the reactor of Figure 2 is the same as for Figure 1 and, therefore, the corresponding mechanical components of Figure 2 are designated by prime numerals ('.)

Referring now to Figure 2, a tunable CO_2 infra-red laser source 120 and a tunable argon ion laser source 130 direct laser beams 134 and 136 respectively through Brewster angle input windows 140 and 142 respectively to a common focal spot. The second input window 142 is made from a silica or 'Vycor' (Trade Mark) plate, transparent to UV and visible radiation while the remaining parts, designated with subscripts "a" and "b" of the two laser apparatuses are similar to those described with reference to Figure 1. However, since fewer IR photons are required, sufficient intensity may be obtained from the laser source 120 without the use of a focusing lens, which is omitted, or by means of a CW CO_2 laser with sufficient output intensity (power per unit area).

In operation, on proper frequency tuning, the laser source 120 excites the vibrational states in the U-organic compound and the laser source 130 raises the electronic states sufficiently to dissociate the desired chemical bond attached directly to the U atom or to stimulate a chemical reaction. The laser 130 may be replaced with a filtered Hg lamp or other UV source of sufficient intensity to dissociate the desired bond in the cases when sufficient isotope selectivity is obtained through vibrational excitation by irradiating with the laser 120. A frequency doubler (not shown) may be used with the tunable laser 130, depending upon the particular U-organic.

WHAT WE CLAIM IS:-

1. A process for separating uranium isotopes, which comprises: preparing a volatile compound U-T, in which U is a mixture of uranium isotopes and T is a chemical moiety

containing at least one organic or deuterated borohydride group, and which exhibits for at least one isotopic species thereof a fundamental, overtone or combination vibrational absorption excitation energy level at a frequency between 900 and 1100 cm^{-1}; irradiating the said compound in the vapour phase with energy emitted by a radiation source at a frequency between 900 and 1100 cm^{-1} and thereby differentially modifying properties of at least one isotopic species of the said compound to establish differences in properties of the irradiated isotopic species facilitating a separation of isotopic species by physical or chemical means applied to the irradiated compounds and to products derived therefrom; and utilizing the established differences in properties to selectively recover a specific uranium isotope from the irradiated compound.

2.   A process as claimed in claim 1 wherein the U-T bond has a fundamental, overtone or combination vibrational absorption frequency between 900 and 1100 cm^{-1}.

3.   A process as claimed in claim 2 wherein the volatile compound is a borohydride-organo uranium complex or a uranyl-organo complex.

4.   A process as claimed in claim 2 wherein the chemical moiety T is selected from fluorinated organic groups, alkoxy groups, organo siloxy groups and amino or amido groups.

5.   A process as claimed in claim 2 wherein the volatile compound is $U(BD_4)_4$.

6.   A process as claimed in any preceding claim wherein the radiation source is a CO_2 laser.

7.   A process as claimed in any of claims 2 to 5, wherein the radiation source comprises an infra-red source in combination with a visible light or ultra-violet source.

8.   A process as claimed in claim 7 wherein the radiation source comprises in combination a CO_2 infra-red laser and an argon ion laser.

9.   A process as claimed in any preceding claim wherein irradiation of the said compound is conducted in a first reaction zone and the said compound in a vapour phase is pumped through an expansion nozzle to effect cooling thereof in the said first reaction zone.

10.   A process as claimed in claim 9 wherein the aperture diameter of the said nozzle and the velocity of pumping therethrough are regulated to control the pressure in the said first reaction zone.

11.   A process as claimed in claim 9 or 10 which further comprises introducing the irradiated compounds from the first reaction zone and reactive gas species into a second reaction zone, and reacting the irradiated compounds with the reactive gas species in the second zone.

12.   A process as claimed in any preceding claim which further comprises condensing reacted irradiated modified compound in a first temperature-controlled zone, and condensing unreacted irradiated compound in a second temperature-controlled zone.

13.   A process as claimed in any preceding claim wherein the temperature of the said compound is controlled during irradiation thereof.

14.   A process as claimed in claim 13 wherein the said differences in properties of the isotopic species derive from the said controlling of the temperature of the compound during irradiation thereof, and separation of the irradiated isotopic species is based upon controlling the temperature of one compound during irradiation thereof.

15.   A process as claimed in any preceding claim an isotopic species of the said compound is activated and the activated isotopic species caused to react.

16.   A process as claimed in any of claims 1, 2, 4 or 6 to 15 wherein the said compound is a uranyl fluorinated β-diketonate.

17.   A process as claimed in any of claims 1 to 4 or 6 to 15 wherein the said compound is a uranyl aluminium alkoxide complex.

18.   A process for separating uranium isotopes substantially as herein described with reference to Figure 1 or Figure 2 of the accompanying drawings.

19.   Uranium isotopes separated by a process as claimed in any preceding claim.
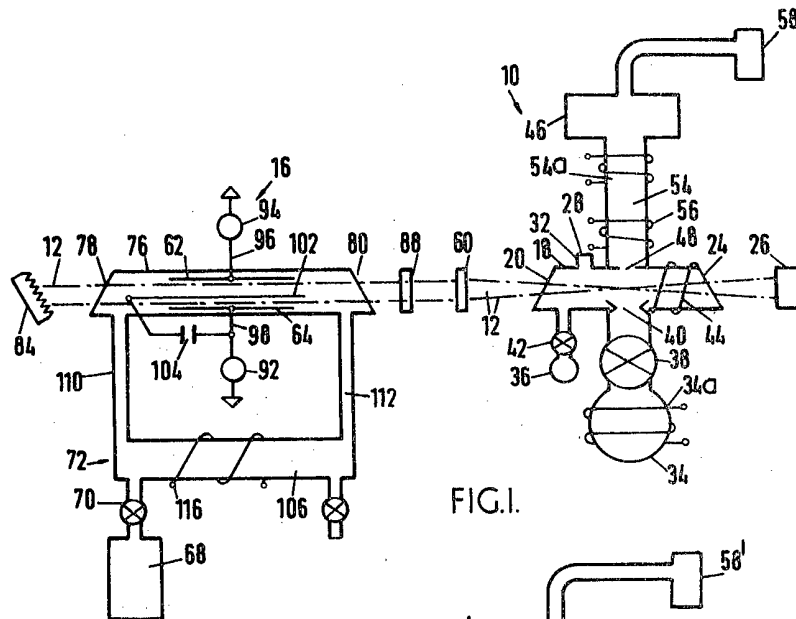
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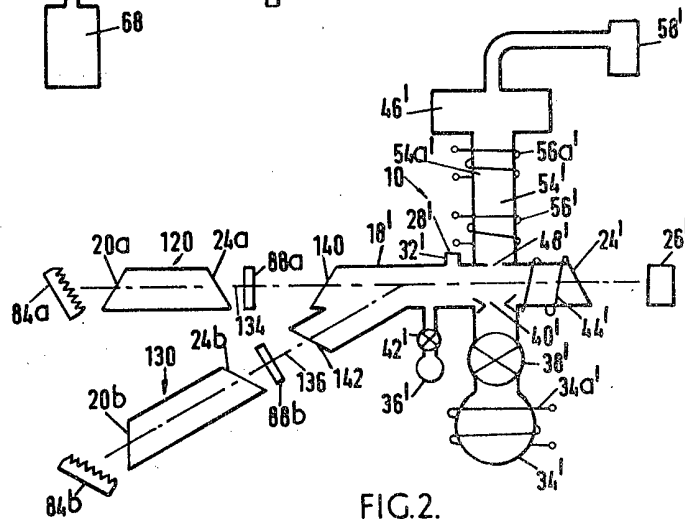Agents for the Applicants.

FIG.1.

FIG.2.