

(21) Application No **7904888**

(22) Date of filing **12 Feb 1979**

(23) Claims filed **12 Feb 1979**

(30) Priority data

(31) **878849**

(32) **17 Feb 1978**

(33) **United States of America  
(US)**

(43) Application published

**10 Oct 1979**

(51) **INT CL<sup>2</sup>**

**B01D 59/00**

(52) Domestic classification

**G6P 6 8**

**B1L 214 501 502 505 507**

**510 GE**

(56) Documents cited

**None**

(58) Field of search

**G6P**

(71) Applicants

**Westinghouse Electric**

**Corporation,**

**Westinghouse Building,**

**Gateway Center,**

**Pittsburgh,**

**Pennsylvania,**

**United States of America**

(72) Inventor

**Peter Myer Castle**

(74) Agents

**Ronald Van Berlyn**

(54) **Isotopic separation**

(57) In a method for separating isotopes a desired isotope is separated mechanically from an atomic or molecular beam formed from an isotope mixture utilising the isotropic recoil momenta resulting from selective excitation of the desired isotope species by radiation followed by ionization or dissociation thereof by means of radiation or electron attachment. In an alternative method, matrix formation of UF<sub>6</sub> in HBr so as to collapse the  $\nu_3$  vibrational mode of the UF<sub>6</sub> molecule is used in conjunction with selective isotopic excitation to promote reduction of UF<sub>6</sub> molecules containing U<sup>235</sup> and facilitate separation.

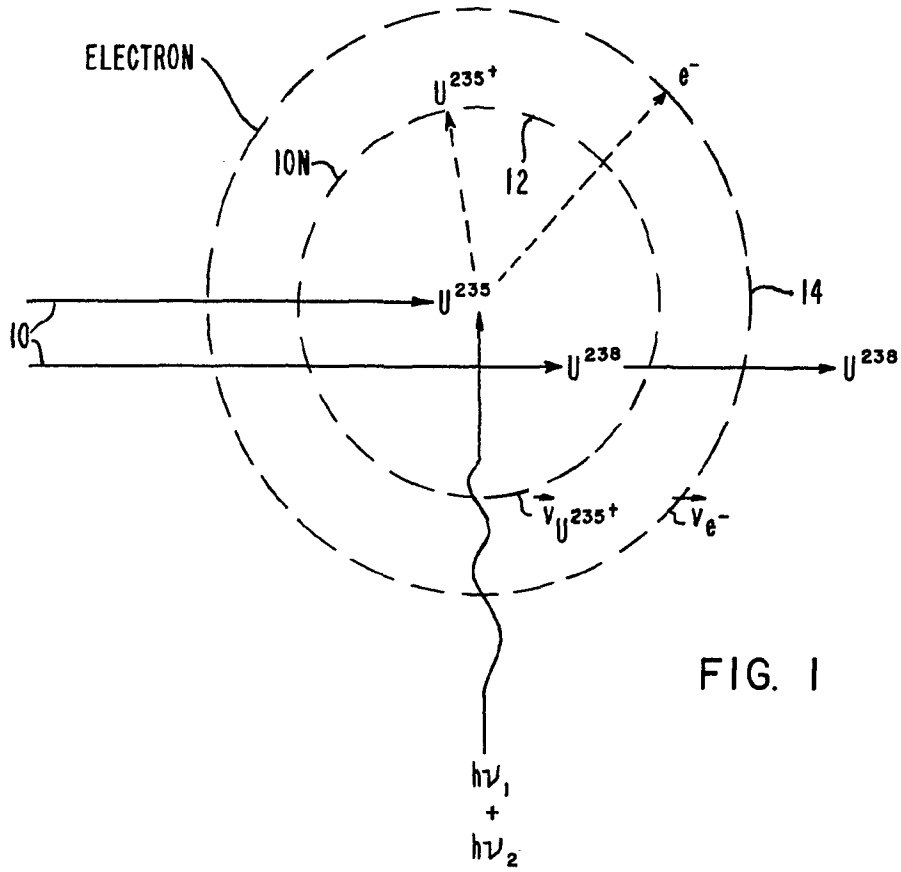


FIG. 1

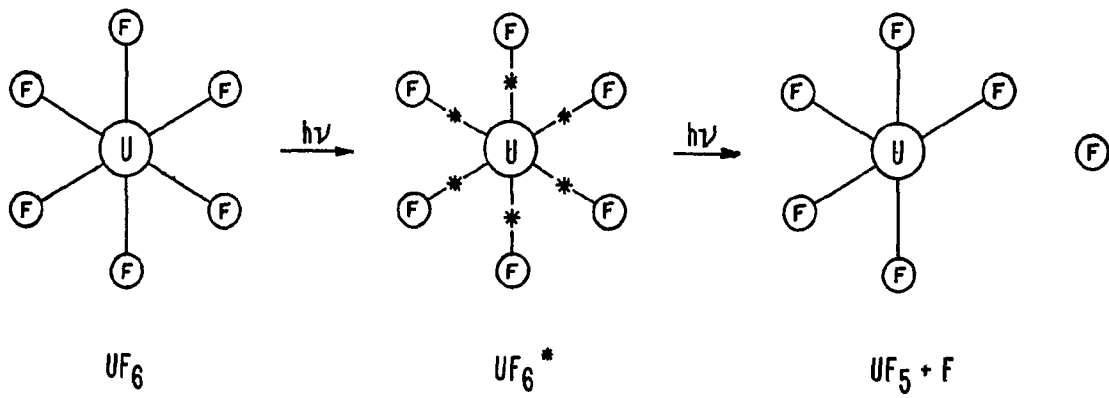


FIG. 3

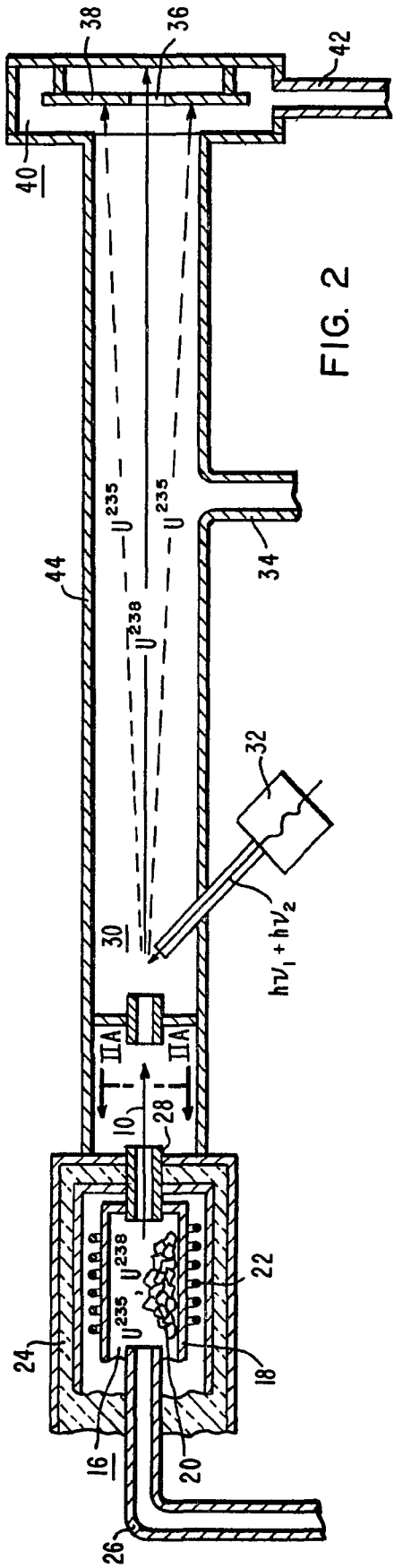


FIG. 2

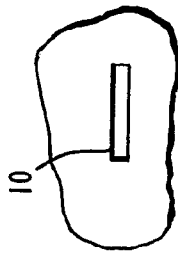


FIG. 2A

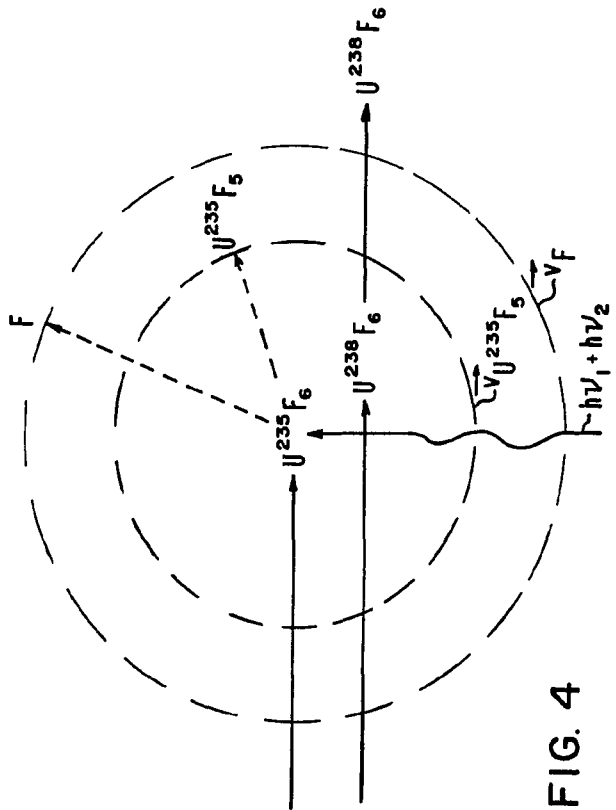


FIG. 4

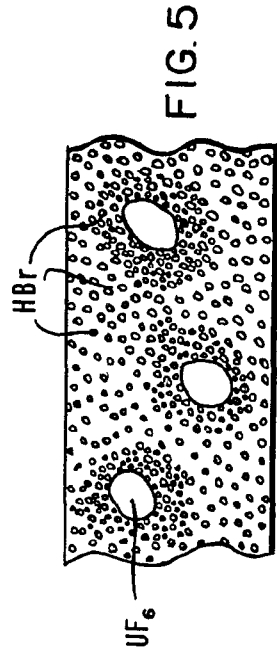


FIG. 5

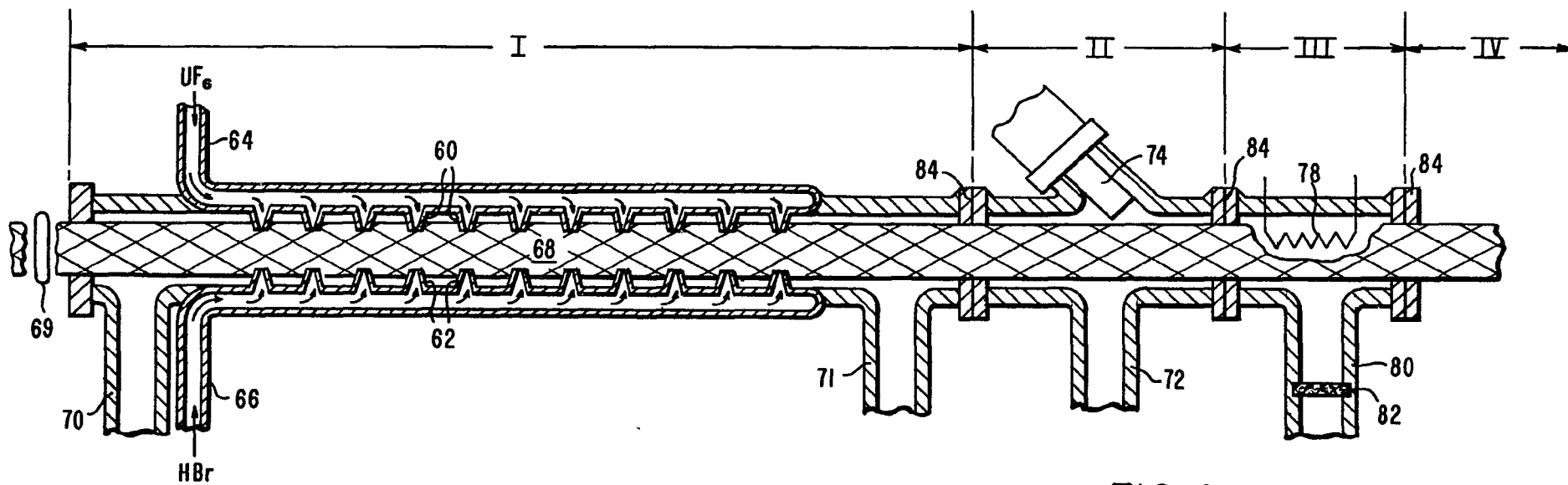


FIG. 6

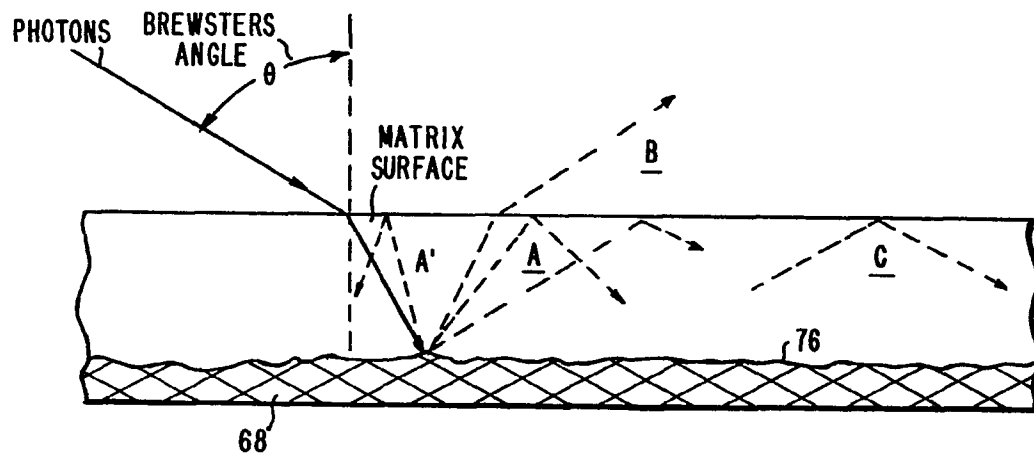


FIG. 7

3/3

2018012

## SPECIFICATION

**Isotopic separation**

5 This invention relates to molecular and atomic isotope separation, particularly applicable to separation of Uranium-235 from other uranium isotopes including Uranium-238. 5

Specific isotopes of a given element are used for many purposes including medical treatment, tracer studies of chemical and biological processes, and preparation of target materials and fuels for nuclear reactors. One of the most common and desirable processes is the separation or enrichment of Uranium-235 from other uranium isotopes, particularly Uranium-238. The basic process presently in use for such separation is gaseous diffusion, requiring a complex cascading network and large energy inputs. 10

Alternative methods being considered include centrifugal separation and, more recently, processes taking advantage of the isotope shifts in atomic or molecular mixtures so as to form ions of a desired isotope, and then separating the ions. Examples of the latter processes include those described in U.S. Patent 3,772,519 and also U.S. Patent No. 3,443,087. Another U.S. Patent, No. 3,558,877 teaches the use of a light beam to deflect selected isotopes to promote separation. Still another U.S. Patent, No. 3,944,825 utilizes selective ionization and an expanding plasma to achieve separation. The references, while representing desirable improvements in the art, are not without deficiencies. Among the deficiencies is the necessity for separation of ions from a neutral background requiring electric or magnetic fields formed from complex energy consuming separating devices. Another deficiency is the power required in some proposed processes to ionize to a sufficiently high energy state. Further, the occurrence of charge exchange reactions can complicate the proposed methods. 15 20

It is the principal object of the present invention to alleviate the complexities and requirements associated with such separating procedures and to provide further alternatives in the field of isotope separation. With this object in view, the present invention resides in a method of separating isotopes of an element in an atomic mixture containing first and second isotopes, wherein a gaseous atomic beam of said mixture is produced and said first isotopes are selectively exciting by radiation of a predetermined wavelength in preference to said second isotopes in said beam, characterized in that said excited first isotope is selectively ionized in preference to said second isotope so as to impart isotropically distributed recoil momenta to said ions of said first isotope, thereby changing the direction of motion of said ions from the direction of motion of said beam and said ions of said first isotope are then separated from said beam by a mechanical separation structure. 25 30

The invention eliminates reliance upon electric and magnetic field separation means and is generally unaffected by charge exchange possibilities. A basis relied upon for several of the preferred embodiments is that recoil momenta are imparted to atoms or molecules undergoing selective irradiation resulting in ionization, dissociation or dissociative electron attachment reactions. Accordingly, a molecular or atomic isotopic gaseous beam having velocity in a given direction can be selectively irradiated such that a desired isotopic species, because of isotropically distributed recoil momenta, acquires a velocity in a direction different than the direction of the beam. The desired products of these reactions can therefore be collected downstream of the irradiation area by simple mechanical means such as a slit plate upon which the products condense or from which they are continuously removed. The balance of the beam, containing a higher abundance of undesired species, passes through the slit. 35 40

In one embodiment a gaseous atomic isotopic beam containing, for example, a low abundance of a first isotope and a high abundance of a second isotope is irradiated so as to selectively excite the first isotope. The excited first isotope is further irradiated at a wavelength which takes advantage of the isotope shift and selectively ionizes the isotope and imparts isotropically distributed recoil momenta to the ions. The ions therefore move in a direction different than the direction of the beam and can be collected downstream with comparatively simple mechanical separation means. Once the ions acquire momenta and a new direction from the recoil reaction, the occurrence of neutralization or charge exchange reactions is of little consequence since it is the direction of the flow, and not the charge, which provides the basis for separation. 45 50

In another preferred embodiment the recoil momenta from a dissociative electron attachment reaction provides the basis for separation of  $U^{235}F_6$  from an isotopic mixture of  $UF_6$  including  $U^{238}F_6$ . The  $UF_6$  molecules are formed as a gaseous beam and the  $U^{235}F_6$  is selectively excited by narrow band radiation of a predetermined wavelength. The molecular beam is then exposed to a beam of electrons promoting dissociative electron attachment and production of  $U^{235}F_5^-$  and other products including a free F (fluorine) fragment and an energy release. A substantial portion of the energy is distributed as kinetic recoil momenta, thereby changing the direction of flow of the  $U^{235}F_5^-$ . Mechanical separation of molecules containing  $U^{235}$ , as ions or neutrals, is then performed downstream of the electron attachment reaction region. 55

In another similar embodiment, an isotopic mixture of  $UF_6$  is again formed into a gaseous beam and exposed to multi-photon, preferably 2-photon, irradiation. The first photon selectively excites  $U^{235}F_6$  molecules and the second photon promotes dissociation to products including  $U^{235}F_6$ . The energy released upon breaking the bond freeing the fluorine is distributed as vibrational motion and recoil momenta, changing the direction of the molecules containing  $U^{235}$  and thereby facilitating mechanical separation. 60

In yet another preferred embodiment, an abundance of  $U^{235}$  is recovered from an isotopic mixture of  $UF_6$  by an isotopically selective irradiation initiated reaction of HBr (hydrogen bromide) and  $U^{235}F_6$ .  $UF_6$  and HBr 65

molecules are deposited onto a non-reactive surface in a concentration providing a substantially greater abundance of HBr than  $UF_6$ . The  $UF_6$  and HBr are formed into a solid matrix so that the  $UF_6$  molecules are distributed throughout the HBr and so that the rotational structure of the  $\nu_3$  mode of vibration of the  $UF_6$  is collapsed. In the condensed state, the  $U^{235}F_6$  molecules in the matrix are then selectively excited. By proper  
5 adjustment of the molecular temperature by irradiation of the  $\nu_3$  mode, a reduction of  $U^{235}F_6$  molecules to products such as  $U^{235}F_5$  and  $U^{235}F_4$  is enhanced and, because of the differences in volatility, the other reaction products can be driven off as gases while the  $U^{235}F_5$  and  $U^{235}F_4$  remain as solids on the deposition surface.

It will be apparent to those skilled in the art that the various steps summarized above each have inherent  
10 inefficiencies and present technological limitations. It therefore is to be understood that the term "separation" and the like refer to increasing the concentration of a desired molecule or isotope, such as  $U^{235}$ , as compared to the feed concentration. It will also be understood that each of the disclosed embodiments is compatible with multiple repetitive stages, as desired for a chosen end-point concentration. And, it is also to be understood that while the following description refers to the separation of a desired species, such as  
15 Uranium-235, in the presence of an undesired species, such as Uranium-238, the actions and reactions directed toward the desired species can similarly be directed toward the undesired species, also accomplishing separation.

The invention will become more readily apparent from the following description of a preferred embodiment thereof shown, by way of example only, in the accompanying drawings, in which:  
20 *Figure 1* is a schematic representation of the isotropic velocity vector distribution of an ion and electron formed from an atomic beam;

*Figure 2* is a side view, in cross-section, of apparatus in accordance with the invention;

*Figure 2A* is a view taken at IIA-IIA of *Figure 2*;

*Figure 3* is a schematic representation of a photo-dissociation reaction;

25 *Figure 4* is a schematic representation of the isotropic velocity vector distribution of the reaction of *Figure 3*;

*Figure 5* is a schematic representation of  $UF_6$  molecules distributed in an HBr matrix;

*Figure 6* is a top view, in cross-section, of apparatus in accordance with another embodiment of the invention; and  
30 *Figure 7* is a schematic representation of photon interaction with a matrix surface on a substrate.

Isotopic separation of an atomic or molecular isotopic mixture can be achieved by selectively imparting added momentum to the desired atomic or molecular species. One method of obtaining added momentum, taking advantage of the isotopic shift, is through recoil, typically isotropic, upon fragmentation of the desired atom or molecule resulting from selective irradiation. Fragmentation, as used herein, refers to loss of an  
35 electron from an atom or molecule, or breaking apart of a molecule into plural parts. The fragmentation can be of various forms, forming such products as ions, neutrals and electrons. The recoil phenomenon is common to those reactions typically termed photo-ionization, dissociative electron attachment, and photo-dissociation.

In any of these processes, conservation of momentum and energy requires, upon fragmentation, that any  
40 excess energy be distributed among the resulting products, in forms such as vibrational, rotational, and translational motion. The translational motion imparted is referred to as recoil.

#### *Photo-Ionization*

In the photo-ionization process, a selected isotope in an atomic mixture is ionized through multi-photon  
45 absorption. Two-photon absorption is typically utilized in, for example, the selective ionization of Uranium-235 in an atomic mixture including other uranium isotopes, such as Uranium-238. Uranium is used herein as an exemplary element, although the procedure is applicable to other elements.

In accordance with this invention uranium vapor or gas is formed into an atomic beam 10 illustrated in *Figure 1*. The beam is preferably collimated and ribbon-shaped. The Uranium-235 in the beam is then  
50 selectively irradiated, in accordance with the isotope shift, by radiation of a predetermined wavelength which raises the electronic state but is insufficient to ionize the Uranium-235. The initial photon can have an energy of, for example, 3.5 eV. This initial selective irradiation of Uranium-235 in preference to Uranium-238, represented as a photon  $h\nu_1$  in *Figure 1*, results in a beam mixture in which the population of excited atoms is enriched in Uranium-235. The second photon, represented as  $h\nu_2$ , selectively ionizes the excited  
55 Uranium-235. This second photon can have any energy sufficient to ionize the excited atoms, up to about 6 eV, the ionization limit for both Uranium-235 and Uranium-238.

When the sum of the two photon energies is in excess of the ionization energy of Uranium-235, an electro,  
represented as  $e^-$  in *Figure 1*, leaves the Uranium-235 atom with a kinetic energy about equal to the difference between the total absorbed energy and the ionization potential. The excess momentum vector has  
60 a random direction, as depicted by the circular broken lines of *Figure 1*. Line 12 represents the distribution of the velocity vectors of the ionized  $U^{235+}$ , and line 14 represents a similar distribution of the freed electrons. The electron line 14 is of larger diameter than the ion line 12 to illustrate that the electron velocity ( $\vec{v}_e$ ) is substantially higher than the Uranium-235 ion velocity ( $\vec{v}_{235}$ ) in accordance with Equations (1) and (2) wherein  $m$  refers to mass and I.P. is the ionization potential.

$$hv_1 + hv_2 - \text{I.P.} = 1/2 m_U |\vec{v}_{U235}|^2 + 1/2 m_e |\vec{v}_e|^2 \quad (1)$$

As a result of conservation of momentum:

$$m_{U235} v_{U235} = m_e v_e \quad (2) \quad 5$$

It is apparent that the mass of the uranium fragment, or any other atomic or ionization fragment, is substantially greater than the mass of the electron. Accordingly, a substantial portion of the excess energy is given to the electron. If it is assumed, for example, that the excess energy is 1 eV, the electron velocity is:

$$1 \text{ eV} = 1.602 \times 10^{-12} \text{ ergs} = 1/2 m_e |\vec{v}_e|^2 \quad 10$$

$$|\vec{v}_e| = 5.931 \times 10^7 \text{ cm/sec}$$

15 And, from the conservation Equations (1) and (2): 15

$$|\vec{v}_{U235}| = \frac{m_e}{m_{U235}} |\vec{v}_e| = 137.5 \text{ cm/sec-eV}$$

If it is further assumed that the atomic beam is formed at about 2000K and a pressure of approximately 20 0.01 Torr, the isotopes in the beam have an average velocity given by: 20

$$\vec{v} = (\frac{8RT}{\pi M})^{1/2} = 4.195 \times 10^4 \text{ cm/sec}$$

where R is the universal gas constant; T is the absolute temperature; and M is the atomic mass.

25 The change in momentum, the recoil momentum, imparted to the isotopes by the ionization process is isotropic; hence the momentum vector has a random direction, as illustrated in Figure 1. 25

As the recoil  $U^{235+}$  ions acquire a differential velocity in random directions of about 137.5 cm/sec-eV, most of the ionized  $U^{235}$  will move out of the beam volume, in a direction of motion different than that of the beam. Because the added momentum is small compared to the isotope's initial momentum in the beam, the 30 flow path of the  $U^{235+}$  ions will fan out, maintaining a substantial forward directional component in the direction of the beam, as depicted in Figure 2. With a drift region downstream of the ionization of, for example, 300 cm, the  $U^{235+}$  recoil ions will diverge to maximum of 0.987 cm per eV of excess photon energy. 30

One form of apparatus consistent with carrying out the inventive method is shown in Figure 2. It includes a means for forming the atomic isotopic gaseous beam 10 such as an oven 16. The oven shown includes a 35 crucible 18 for holding a supply of the feedstock 20, such as natural uranium. It further includes means for vaporizing the feedstock 20, such as induction heating coils 22 and heat shield 24. To facilitate vaporization the oven is maintained at vacuum conditions through conduit 26, connected to vacuum apparatus not shown. For uranium feedstock the vacuum is preferably maintained at about  $10^{-8}$  Torr and the feedstock heated to approximately 2000K. The atom flux obtained can be in the region of  $10^{22}$  atoms  $\text{cm}^{-2}\text{sec}^{-1}$ . 35

40 In order to form the high temperature uranium gas into a beam of desired configuration, collimating means such as the elongated collimator 28 are used. Although the beam 10 can be of various geometric cross-sections, including circular, the preferred geometry is ribbon-shaped, as shown in Figure 2A. The beam 10 then enters an elongated photo-ionization region 30, where it is irradiated with photons  $hv_1$  and  $hv_2$  of preselected wavelength from one or more lasers 32. The irradiating photons preferably are oriented to 45 intersect the atomic beam 10 at an angle to the beam direction. The efficiency of the irradiation process can be increased by use of mirrors which reflect the photons back and forth through the atomic beam 10. The efficiency and throughput of the system can be similarly be increased by utilizing mirrors and a wide ribbon beam 10 or a plurality of beams arranged side by side. 45

The photo-ionization region 30 is also maintained at vacuum conditions, about  $10^{-8}$  Torr, through conduit 50 34 connected to vacuum maintaining means not shown. Upon obtaining recoil momentum, the selectively ionized Uranium-235 diverges from the beam 10 in a high vacuum field free area. The mainstream of the beam 10 therefore continues through a slit 36 in mechanical separating means such as the condensing 5 collecting surface 38. The region 40 about the collecting surface is also maintained at vacuum conditions through conduit 42. To increase collection efficiency, the beam 10 mainstream can continue through another 55 axially aligned photo-ionization and collection region. It will also be noted that since the basis for collection is the differing direction of the Uranium-235 ions, neutralization of the ions prior to impact upon the collecting surface does not affect the separation process. The collecting surface can be easily removed when a desired buildup of uranium, increased in the Uranium-235 concentration, is achieved. The uranium can then be removed from the collecting surface by chemical, scraping or other well known processes. 55

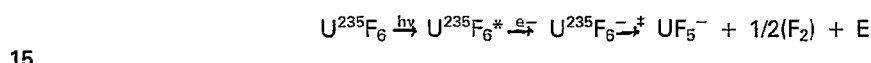
60 Dependent upon the geometry of the apparatus, a buildup of uranium may also occur on the walls 44 of the photo-ionization region 30, particularly downstream of the point of ionization. The walls 44 are therefore preferably segmented to facilitate removal and subsequent processing to recover the uranium product. 60

#### Dissociative Electron Attachment

65 In addition to photo-ionization, the recoil momentum upon fragmentation can also be utilized in 65

dissociative electron attachment reactions. The process and apparatus utilized can be similar to that described above, additionally requiring means for exposing the molecular beam to an electron beam.

In a dissociative electron attachment reaction an isotopic mixture of molecules of, for example,  $UF_6$  is formed into a gaseous collimated beam flowing in a preset direction. The beam is then exposed to irradiation at a predetermined wavelength so as to selectively excite the vibrational  $\nu_3$  or other suitable combination mode of the  $U^{235}F_6$  molecules in preference to other molecules such as  $U^{238}F_6$ . The energy of the irradiation should be less than the ionization potential of the molecule and the excitation can be performed through single or multiple photon absorbing steps. The  $UF_6$  beam is then exposed to a beam of electrons of sufficient energy to selectively promote dissociative electron attachment of the excited  $UF_6$  molecules in preference to other  $UF_6$  molecules. With an asterick denoting an excited state, alternatively stated as a state of population inversion, and a double plus denoting excess energy, the reaction can be written as:



While the reaction identifies  $U^{235}F_6^-$ , free fluorine, and energy (E) as the products, it will be understood that other products, such as  $U^{235}F_4^-$  are also possible. The dissociative electron attachment reaction offers some advantages with respect to the above-described photo-ionization process. First, it will be recognized that since the fragments include free fluorine atoms as opposed to electrons, the percentage distribution of recoil momentum to the  $UF_5$  ion is greater than the percentage distribution to a Uranium-235 atom. Accordingly, the  $UF_5^-$  will move out of the molecular  $UF_6$  beam with a greater separation. Also, the temperatures required for working with  $UF_6$  are much lower and would generally be below 1000C for other compounds.

#### Photo-Dissociation

In addition to photo-ionization and dissociative electron attachment reactions, recoil momentum can also be advantageously utilized, similar to the processes discussed above, in a photo-dissociative procedure in conjunction with a molecular isotopic beam. Here, molecules of, for example,  $SF_6$  or  $UF_6$  are formed into a gaseous collimated molecular beam. The molecular beam is selectively excited with a first photon irradiation of the vibrational  $\nu_3$  or suitable combination mode so as to increase the excited population in  $U^{235}F_6$  concentration. Neither the first or second photon irradiation, again, should be of sufficient energy to, alone, cause dissociation of the  $U^{238}F_6$  or  $U^{235}F_6$  molecules. The molecular beam is then further irradiated at a predetermined wavelength at an energy sufficient to dissociate the excited  $U^{235}F_6$  molecules. It has been established by Rockwood, S.D. and Rabideau, S.W., in the IEEE J. Quantum Electronics, QE-10, 789 (1974), that two-photon irradiation of  $SF_6$  is achievable, and can be written as:



Since the molecular makeup of  $SF_6$  is similar to that of  $UF_6$ , it can be presumed that two-photon dissociation is also achievable with  $UF_6$ . The process is illustrated in Figure 4. The bond energies of  $UF_6$  and  $SF_6$  are 35.9 kcal/mole and 45.6 kcal/mole, respectively.

Upon breaking of the bond, a substantial portion of the energy released goes into recoil motion of the fragments and, as in the above discussions, facilitates separation of the recoil products from the mainstream of the molecular beam by relatively simple mechanical separation structure. Since the recoil products are neutrals, electric or magnetic fields need not be used for separation.

#### Matrix Isolation

Figure 6 illustrates apparatus useful in separation of a molecular mixture of uranium isotopes by matrix isolation also utilizing selective excitation principles. The basis for the separation is the isolation of  $UF_6$  molecules in a precisely defined environment characteristically reactive above a preselected threshold temperature. The matrix can be formed by condensing a molecular species, such as  $UF_6$ , onto a cold surface at the same time that a matrix molecule or atom, inert or reactive, such as HBr, is condensed. While varying ratios of the matrix constituents can be used, the procedure as applied in infrared spectroscopy usually involves the matrix molecules or atoms at a number density in excess of 50 to 100 times the primary molecule or atom.

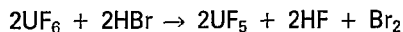
In accordance with one embodiment of the invention,  $UF_6$  and HBr, preferably in a gaseous state, are codeposited on a cold unreactive surface, thereby creating a thin solid layer of individual  $UF_6$  molecules trapped in an HBr matrix. As illustrated in Figure 5, the matrix formation makes each  $UF_6$  molecule generally unable to interact with other  $UF_6$  molecules. Additionally, the freezing of the  $UF_6$  into the matrix substantially lessens the molecular rotational transitions, and specifically collapses the rotational structure of the  $\nu_3$  fundamental vibration of  $623.5 \text{ cm}^{-1}$  into a single sharp line at a slightly different frequency (see BAR-Ziv, E., Frieberg, M., and Weiss, S., Spectrochimica Acta, 1972, 28A, 2025-2028).

The substantial elimination of rotational contributions greatly increases the efficient utilization of the vibrational frequency difference of about  $0.05 \text{ cm}^{-1}$  between the  $\nu_3$  vibrational modes of  $U^{235}$  and  $U^{238}$ . An efficient selective excitation of the  $U^{235}F_6 \nu_3$  fundamental frequency can therefore be realized.

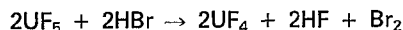
Although other isolating species may be used, HBr is known, through work developed at the Union



Carbide Corporation, Oak Ridge Gaseous Diffusion Plant reported by Wolf, A.S., Hobby, W.E., and Rapp, K.E., in *Inorganic Chemistry* 1965, 4, #6, 755-757, to undergo the following reactions:



5



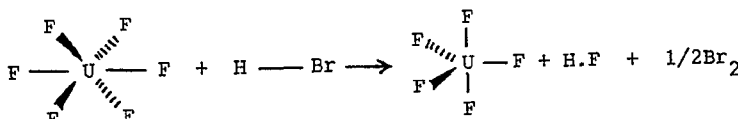
5

- It was found that while liquid  $\text{UF}_6$  which was allowed to stand overnight in contact with liquid  $\text{HBr}$  at room temperature produced only a small amount of reaction products, the reactions proceed vigorously at 65C. The reactions are therefore strongly temperature dependent. Although the mechanism of the reaction has not been elucidated, in all probability it proceeds along the same axis as the  $\nu_3$  fundamental mode:

10

10

15



15

Accordingly, with  $\text{U}^{235}\text{F}_6$  isolated and surrounded by  $\text{HBr}$  molecules, selectively exciting the  $\text{U}^{235}\text{F}_6$   $\nu_3$  mode with sufficient energy to produce an effective vibrational temperature of 65C will result in the molecules' reduction to  $\text{UF}_5$ ,  $\text{UF}_4$ , and so forth. Once the  $\text{U}^{235}\text{F}_6$  molecules have been selectively reduced in the presence of  $\text{U}^{238}\text{F}_6$  molecules, separation of the isotopic species is accomplished with relative ease. For example, because of differences in volatility, a mild heating of the matrix under reduced pressure to about 300 Torr will volatilize the  $\text{U}^{238}\text{F}_6$ ,  $\text{HBr}$ ,  $\text{HF}$  and  $\text{Br}_2$ , while the solid  $\text{U}^{235}\text{F}_5$  and some  $\text{U}^{235}\text{F}_4$  remain on the deposition surface. Chemical reactions carried out under matrix isolation conditions are also discussed in "Direct Synthesis And Characterization of Debenzenochromium(0) In An Argon Matrix At 14K", John W. Boyd, John M. Lavoie and Dieter M. Gruen, *Journal of Chemical Physics*, Vol. 60, No. 10, May, 1974; "Matrix Isolation Infrared Study Of The Reaction Between Germanium Vapor And Molecular Oxygen. The Characterization And Mechanism Of Formation Of Molecular Germanium Dioxide And Ozone", A. Bos, J.S. Ogden, and L. Orgee, *Journal of Physical Chemistry*, Vol. 78, No. 17, 1974; and "Infrared Spectra Of Matrix-Isolated Uranium Oxide Species. I. The Stretching Region", by S.D. Gabelnick, G.T. Reedy, and M.G. Chasanov, *Journal of Chemical Physics*, Vol. 58, No. 10, 1973.

20

20

25

25

30

30

It will be apparent that the disclosed invention offers substantial advantages as compared to other laser excitation methods. The exciting radiation is in the infrared region, which, under present technology, is believed to be the most economical and efficient region for photon production. Further enhancing the efficiency of the procedure is that the reactions are carried out in a solid state, thereby enabling a high density of reacting sites. Also, since no ionization processes are involved, a relatively high density of excited molecules can be achieved without concern for space charge effects. Further, selective isotopic occurrences are less likely to be scrambled by intermolecular collisions as a result of the dilution factor and general immobility in the solid matrix. And, since the final separation can merely be the process of removing volatiles from a solid product, complex field separation devices are unnecessary.

35

35

40

40

The apparatus shown in Figure 6 can be used to carry out the inventive method, and includes four basic regions denoted I through IV. Region I is the matrix formation area and includes a series of nozzles 60 and 62 which respectively deposit  $\text{UF}_6$  and  $\text{HBr}$  from the inlets 64 and 66 at a molecular number ratio of  $\text{UF}_6$  and  $\text{HBr}$  in the range of 1:50 to 1:100. The  $\text{UF}_6$  and  $\text{HBr}$  are deposited (as liquids or possibly gases) onto a moving substrate 68 which is cooled to a temperature less than 100K. The spraying region is preferably maintained at a pressure of about  $10^{-4}$  Torr or less, through vacuum maintaining means (not shown) connected to conduits 70 and 71. Cooling means such as a refrigeration coil 69 in contact with or near the moving substrate can be utilized to cool the substrate.

45

45

The substrate is non-reactive with the matrix materials and can include materials such as gold or platinum deposited on a copper or other band. Copper is preferred because of its good thermal heat transfer characteristics. The thickness of the matrix mixture can be adjusted by varying the amount deposited and/or the speed of the substrate 68. The actual mass throughput of the entire system is also controlled by the number of spray nozzles, the mass flow through the nozzles, the irradiation absorption efficiency, and the laser power. The typical advantageous density differential between this embodiment and the molecular beam teachings will be apparent from a comparison of the number of density in the beams at 1 Torr and the number density in the diluted matrix. For example, for a uranium feed having approximately 0.7%  $\text{U}^{235}$ , at 20C and a dilution of 100:1, the number density of  $\text{U}^{235}$  as  $\text{UF}_6$  in the solid matrix is approximately  $9.3 \times 10^{16}/\text{cc}$ . A molecular beam maintained at 1 Torr, which represents the maximum pressure reasonably applicable to molecular beams, will have a  $\text{U}^{235}$  number density of about  $2.6 \times 10^{14}/\text{cc}$ . The differential constitutes approximately a 350 times greater number density in a matrix than in a molecular beam, which advantageously allows higher throughput and greater efficiency.

55

55

60

60

Selective excitation and chemical reaction of the deposited solid matrix occurs in region II. The matrix in region II is preferably maintained at temperature and pressure conditions similar to region I to alleviate the potential for additional side reactions. A conduit 72 connected to appropriate apparatus can be used for this purpose. Selective irradiation of the matrix is performed by irradiation means such as a laser head 74. In this

65

65

region, the system efficiency is effected by the irradiation absorption efficiency and the power density of the laser 74. The substrate speed can be compatibly adjusted with the laser power available, being correspondingly faster for a high power and lower for a low power.

Since the output of many lasers is polarized, the laser head is preferably aligned, with respect to the surface of the substrate, at Brewster's Angle. As illustrated in Figure 7, this arrangement lessens reflective losses in the laser beam transmission. At Brewster's Angle substantially full transmission of the radiation is into the matrix. With a platinized or gold coated copper substrate, the beam of photons will be reflected back into the matrix as at "A" with only small losses as at "B". Deviations from the Brewster's Angle compliment at the matrix-substrate interface 76 will cause additional reflections back into the matrix as shown at "A'". Transmission of photons at Brewster's Angle thus creates a high efficiency configuration for photons subsequent to entering the matrix, as well as assuring essentially 100% penetration of the photon beam into the matrix. Further assisting the photon reaction efficiency, illustrated at "C", is the result that photons re-emitted by an unreacting relaxation are subjected to the same internal reflections, enhancing the reactive absorption probability for  $U^{235}F_6$  selective excitation.

Upon excitation causing a  $U^{235}F_6$  molecular temperature in excess of 65C, the reduction process dicussed above rapidly occurs, producing  $U^{235}F_5$  and  $U^{235}F_4$ .

The matrix and substrate then move to region III where separation of the reduction products occurs. Heating means such as a heating coil 78 warm the substrate and matrix to a temperature in the range of 60C. Because of the differing volatilities of the constituents, this mild heating drives off the unreacted  $U^{238}F_6$ , excess HBr, HF, and  $Br_2$ , leaving behind the  $U^{235}F_5$  and some  $U^{235}F_4$  as a solid residue. The volatilized compounds are pumped from the warm substrate through apparatus connected to conduit 80.

The substrate then continues to region IV, where the products having an increased  $U^{235}$  concentration are removed and the substrate is further cleaned and dried prior to returning to region I. The removal of desired products can merely involve washing, dissolving, or scraping of the products from the substrate.

The  $U^{235}F_6$  molecules left on the substrate in region III should be coalesce into stranded particles during vaporization of the other materials. However, it can also be carried away as small particulates with the  $U^{238}F_6$  and HBr gases. In this event, a particulate filtration system, including a filter 82, can be utilized to trap the solid  $U^{235}F_5$  and  $U^{235}F_4$ .

The apparatus also preferably includes baffles 84 between the respective regions which assist in maintaining the desired pressures and separation of the four regions. The baffles 82 can be maintained at low temperatures through cooling by liquid nitrogen or liquid carbon dioxide.

#### Example

An exemplary throughput utilizing matrix isolation can be shown analytically assuming, for example, that the  $UF_6$  deposition nozzles 60 have an effective orifice area of one square centimeter with a  $UF_6$  pressure of 1 Torr at 273K. The nozzles can be distributed over approximately 20 cm. Further assuming a substrate velocity of 0.5 cm/sec., a deposition of approximately 2.09 grams per hour of trapped  $U^{235}F_6$  is available for photo-excitation, corresponding to approximately 1.41 grams per hour of pure  $U^{235}$ , as shown by the following:

To determine the molecular density (n) of  $U^{235}F_6$  where  $n = \text{molecules/cm}^3$ , at, for example, 273K and 1 Torr (1/760 atmosphere), it is known that

$$Pv = \frac{n}{6.023 \times 10^{23}} RT$$

Where  $v = 1 \text{ cm}^3$ ,  $T = 273K$ ,  $P = 1 \text{ Torr}$  and  $R = 82.057 \text{ cm}^3 \text{ atm mole}^{-1}K^{-1}$ , and accordingly

$$n = \left( \frac{1/760 \times 1}{82.057 \times 273} \right) 6.023 \times 10^{23} = 3.65 \times 10^{16} \text{ molecules/cm}^3,$$

corresponding to  $n = 2.59 \times 10^{14} U^{235}F_6/\text{cm}^3$ .

The average velocity ( $\bar{v}$ ) of the  $U^{235}F_6$  can be defined as  $\bar{v} = (8RT/\pi M)^{1/2}$  where  $R = 8.317 \times 10^7$ ,  $T = 273$  and  $M = 349$ , corresponding to  $\bar{v} = 1.55 \times 10^4 \text{ cm/sec}$ .

The flux (F) at these exemplary conditions is defined by  $F = \eta_4 \bar{v}$ , or  $1.55 \times 10^4 \times 2.59/4 \times 10^{14} = 1.006 \times 10^{18} \text{ molecules/cm}^2 \text{ sec}$ . Accordingly, to determine the number of moles and grams deposited during one hour, assuming one  $\text{cm}^2$  of orifice at 1 Torr and 273K, the deposition rate is

$$\frac{1.006 \times 10^{18}}{6.023 \times 10^{23}} \times 3600 = 6 \times 10^{-3} \text{ moles } U^{235}F_6/\text{hr}.$$

This corresponds to  $6 \times 10^{-3} \times 349 = 2.09 \text{ g U}^{235}\text{F}_6/\text{hr}$ . In terms of pure  $\text{U}^{235}$  the deposition rate is  $6 \times 10^{-3} \times 235 = 1.41 \text{ g U}^{235} \text{ hr}$ .

The exemplary process can be carried out under reasonable energy requirements. To exemplify the power output required for the laser to perform the selective excitation in a single photon excitation process assuming  $1 \text{ cm}^2$  of orifice and the other exemplary conditions above, it was shown that the deposition rate of  $\text{U}^{235}\text{F}_6$  is  $1.006 \times 10^{18}$  molecules/sec. Accordingly, the minimum number of selective photons ( $h\nu_3$ ) would be  $1.006 \times 10^{18}/\text{sec}$ . The energy (Joule/ $h\nu_3$ ) of each photon is  $1.986 \times 10^{-23} \times 625 = 1.24 \times 10^{-20} \text{ J}/h\nu_3$ . Accordingly, the minimum desirable energy on a time basis is  $1.006 \times 10^{18} \times 1.24 \times 10^{-20} = 1.25 \times 10^{-2} \text{ J}/\text{sec}$ , or a laser power of only  $1.25 \times 10^{-2}$  watts, or 12.5 milliwatts. Assuming a one percent quantum yield and one percent wall plug efficiency for the laser, the power requires is 125 watts, or 319 kw/g of separated  $\text{U}^{235}$ . The electrical power expended by the laser for separation of 1g  $\text{U}^{235}\text{F}_6$  is approximately 216 kw and, for a yield of 1 kilogram of three percent enriched  $\text{UF}_6$ , the power required is 3.32 Mw/kg.

There have therefore been described a number of systems and methods useful in isotopic separation of atomic and molecular mixtures. It will be apparent that many modifications and additions are possible in view of the above teachings. It therefore is to be understood that within the scope of the appended claims, the invention may be practiced other than as specifically described.

#### CLAIMS

1. A method of separating isotopes of an element in an atomic mixture containing first and second isotopes, wherein a gaseous atomic beam of said mixture is produced and said first isotopes are selectively exciting by radiation of a predetermined wavelength in preference to said second isotopes in said beam, characterized in that said excited first isotope is selectively ionized in preference to said second isotope so as to impart isotropically distributed recoil momenta to said ions of said first isotope, thereby changing the direction of motion of said ions from the direction of motion of said beam and said ions of said first isotope are then separated from said beam by a mechanical separation structure.
2. A method as claimed in claim 1, characterized in that said ions are neutralized prior to said separation.
3. A method as claimed in claim 1 or 2, characterized in that for separation, said beam is passed through a slit of a collimator and said ions are collected on the surfaces forming said slit.
4. A method as claimed in claim 1 wherein one of said isotopes is Uranium-235 and the other is Uranium-238, the  $\text{U}^{235}$  being present as  $\text{U}^{235}\text{F}_6$  which is to be separated from a mixture of  $\text{UF}_6$  molecules including  $\text{U}^{235}$ , characterized in that said gaseous atomic beam is an isotopic gaseous beam of  $\text{UF}_6$  molecules, said  $\text{U}^{235}\text{F}_6$  molecules in said beam are selectively excited by irradiation at a predetermined wavelength in preference to said  $\text{UF}_6$  molecules, and said isotopic beam is exposed to a beam of electrons of sufficient energy so as to promote dissociative electron attachment of said excited  $\text{U}^{235}\text{F}_6$  molecules in preference to other  $\text{UF}_6$  molecules thereby producing  $\text{U}^{235}\text{F}_5^-$  and other products having isotropically distributed recoil momentum in a direction different than the direction of said beam, whereupon said products are separated from said beam by mechanical separation structure.
5. A method as claimed in claim 4, characterized in that a matrix of said  $\text{UF}_6$  molecules distributed in a substantially greater number of HBr molecules is formed so as to collapse the  $\nu_3$  vibrational mode of said  $\text{UF}_6$  molecules, said  $\text{U}^{235}\text{F}_6$  molecules in said matrix are selectively excited in preference to the other  $\text{UF}_6$  molecules so as to adjust the molecular temperature of said  $\text{U}^{235}\text{F}_6$  molecules and reduce said  $\text{U}^{235}\text{F}_6$  to products including  $\text{U}^{235}\text{F}_5$ , before separation of said products including  $\text{U}^{235}\text{F}_5$  from the balance of constituents of said matrix.
6. A method as claimed in claim 5, characterized in that the ratio of HBr to  $\text{UF}_6$  in said matrix is greater than 50:1.
7. A method as claimed in claim 5 or 6, characterized in that said matrix is formed by depositing said  $\text{UF}_6$  and HBr molecules onto a surface sufficiently cold to form a solid matrix.
8. A method as claimed in claim 7, characterized in that said  $\text{UF}_6$  and HBr are deposited onto said surface in a gaseous state, the temperature of said surface being below 100K.
9. A method as claimed in any of claims 5 to 8, characterized in that said selective excitation is performed by photon irradiation at a frequency in the range of  $623.5 \text{ cm}^{-1}$  adjusting the molecular temperature of said  $\text{U}^{235}\text{F}_6$  molecules to approximately 65C.
10. A method as claimed in any of claims 5 to 9, characterized in that said separation is performed by heating said matrix to a temperature sufficient to selectively volatilize said balance of constituents in preference to said products including  $\text{U}^{235}\text{F}_5$ .
11. A method as claimed in claim 10, characterized in that said products including  $\text{U}^{235}\text{F}_5$  are filtered from said volatilized constituents.