(19) (CA) CANADIAN PATENT

(54) SEPARATION OF ISOTOPES

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ABSTRACT OF DISCLOSURE

A two or more stage enrichment process involving photolysis at two or more selected wavelengths of laser light irradiation. The overall separation factor is the product of the individual separation factors of each stage and is achieved with concurrent photolysis or with consecutive photolysis without physical separation between stages.
This invention relates to a method for the separation of isotopes.

The separation of isotopes using photochemical techniques especially laser irradiation is well known and fairly widely used. The following are representative patents in this field:

- 3,983,020 C.B. Moore Sept. 28, 1976 204-157.1
- 4,000,420 S.E. Harris Dec. 28, 1976 250-281
- 4,023,038 G.S.Janes et al May 10, 1977 250-423
- 4,038,549 G.S.Janes et al July 26, 1977 250-423
- 4,060,732 D. Rosenberger Nov. 29, 1977 250-432
- 4,120,767 S.N. Bittenson et al Oct. 17, 1978 204-158

Patent No. 4,038,549 is concerned with a multilaser system of irradiating gas systems having plural excited states and Patent No. 4,060,732 describes the use of two lasers to essentially promote hot band photoactivation of lighter isotopes in a gaseous isotope mixture.

It is an object of the present invention to provide an improved and more economic method of isotope separation.

This and other objects of the invention are achieved by a two or more stage enrichment process involving photolysis at two or more selected wavelengths of laser light irradiation. The overall separation factor is the product of the individual separation factors of each stage and is achieved with concurrent photolysis or with consecutive photolysis without physical separation between stages.

In drawings which illustrate an embodiment of the invention:

Figure 1 is a flow diagram of a multi-stage separation system.
The general scheme for this multistage process is as follows:

1st Excitation wavelength

Substrate (1) (+ Additives) $\xrightarrow{hv_1} \text{Substrate (2)} \alpha_1$

2nd Excitation wavelength

Substrate (2) (+ Additives) $\xrightarrow{hv_2} \text{Substrate (3)} \alpha_2$

and so on to n stages.

Overall separation factor $\alpha_{\text{overall}} = \prod_{x=1}^{n} \alpha_x$ .... (1)

Figure 1 is a flow diagram of a multistage process with a first laser 10 irradiating a first cell 11 into which has been introduced a primary substrate (feedstock) and a primary transfer agent e.g. halogen at a wavelength $\lambda_1$. The separation factor of this first stage is $\alpha_1$. The products from the first cell pass through a first absorber 12 to remove the transfer agent and then to a second cell 13 along with a suitable secondary transfer agent. This cell is irradiated by a second laser 14 at a wavelength $\lambda_2$ to give a separation factor of $\alpha_2$. The products from cell 13 pass through absorber 15 to remove the secondary transfer agent and then the final separator 16 to separate by mechanical means e.g. still, the enriched products and the depleted substrate 16. If more than two stages are required, additional lasers 17, cells 18, and absorbers 19 are added to the system. The overall separation factor of the multiple stages is given by equation (1).

There are several classes of systems which can be described in general terms.
Radical Transfer Class

Step 1

\[ R_1 - R_2 \xrightarrow{\text{nhv}_1} R_1 + R_2 \]
\[ R_1 + X_n \rightarrow R_1X + X_{n-1} \]

Remove transfer agent \( X_n \), add transfer agent \( Y_n \).

\[ R_1X \xrightarrow{\text{nhv}_2} R_1 + X \]
\[ R_1 + Y_n \rightarrow R_1Y + Y_{n-1} \]

Remove transfer agent \( Y_n \), add transfer agent.

Final Step

\[ R_1Z \xrightarrow{\text{nh}_1} R_1 + Z \]
\[ R_1 + R_1 \rightarrow R_1\rightarrow R_1 \]

\( R_1\rightarrow R_1 \) is the highly enriched final product.

Example:

Step 1

\[ \text{CF}_3\text{CO} \quad \text{CF}_3 \xrightarrow{\text{nhv}_1} 2\text{CF}_3 + \text{CO} \]
\[ \text{CF}_3 + \text{Br}_2 \rightarrow \text{CF}_3\text{Br} + \text{Br} \]

Remove \( \text{Br}_2 \) passage over copper foil.

Step 2

\[ \text{CF}_3\text{Br} \xrightarrow{\text{nhv}_2} \text{CF}_3 + \text{Br} \]
\[ \text{CF}_3 + \text{CF}_3 \rightarrow \text{C}_2\text{F}_6 \]
\[ \text{C}_2\text{F}_6 \text{ enriched in carbon-13} \]

Sub-Class Product Transfer

As above except stable product reacts with transfer agent.

Example:

Step 1

\[ 2\text{C}_3\text{F}_6 \xrightarrow{\text{nhv}_1} 3\text{C}_2\text{F}_4 \]
\[ \text{C}_2\text{F}_4 + \text{HCl} \rightarrow \text{C}_2\text{F}_4\text{HCl} \]
\[ \text{C}_2\text{F}_4\text{HCl} \xrightarrow{\text{nhv}_2} \text{C}_2\text{F}_4 + \text{HCl} \]
\[ \text{C}_2\text{F}_4 \text{ enriched in carbon-13} \]
Product Photolysis Class

\[ R_1 \rightarrow R_2 \xrightarrow{\text{nhv}_1} R_1 + R_2 \]

Step 1

\[ R_1 + R_1 \rightarrow R_1 \rightarrow R_1 \]

Step 2 \[ R_1 \rightarrow R_1 \xrightarrow{\text{nhv}_2} C + \text{by-product} \]

Final Step \[ C \xrightarrow{\text{nhv}_2} D + \text{by-product} \]

Example

\#1 IR:IR

Step 1 \[ C_2F_3Cl_2 \xrightarrow{\text{nhv}_1} C_2F_3H + Cl_2 \]
Step 2 \[ C_2F_3H \xrightarrow{\text{nhv}_2} C_2F_2 + HF \]

HF enriched in deuterium.

\#2 IR:UV

Step 1 \[ CF_3CO CF_3 \xrightarrow{\text{nhv}_1} C_2F_6 + CO \]
Step 2 \[ CO \xrightarrow{\text{hv}_2 + 3CO} C_3O_2 + CO_2 \]

\[ C_3O_2 \text{ enriched in carbon-13} \]

Fragment Photolysis Class

Step 1 \[ R_1 \rightarrow R_2 \xrightarrow{\text{nhv}_1} R_1 + R_2 \]
Step 2 \[ R_1 \xrightarrow{\text{nhv}_2} Ra + \text{by-product} \]

Final Step \[ Ra + Ra \rightarrow Ra \rightarrow Ra \]

The two steps must be closely synchronized so that the fragment is photolyzed before it undergoes reaction.

Example:

Step 1 \[ CF_3COCF_3 \xrightarrow{\text{nhv}_1} 2CF_3 + CO \]
Step 2 \[ CF_3 \xrightarrow{\text{nhv}_2} CF_2 + F \]
\[ CF_2 + CF_2 \rightarrow C_2F_4 \]
\[ C_2F_4 \text{ enriched in carbon-13} \]
Unstable Isomer Photolysis Class

Step 1  \[ A \xrightarrow{\text{nhv}_1} B \]  
(unstable isomer)

Step 2  \[ B \xrightarrow{\text{nhv}_2} C + D \]

Example:

Step 1  \[ \text{CF}_3 CO\text{CF}_2\text{H} \xrightarrow{\text{nhv}_1} \text{CF}_3 CO\text{HCF}_2 \]  
(ketone)  
(enant)

Step 2  \[ \text{CF}_2\text{H} + \text{CF}_3 \xrightarrow{\text{nhv}_2} \text{CF}_3 \text{H} + \text{CF}_3 + \text{CO} \]

\[ \text{CF}_2\text{H} + \text{CF}_3 \xrightarrow{\text{nhv}_2} \text{CF}_3 \text{H} + \text{CF}_3 + \text{CO} \]

\[ \text{C}_2\text{F}_5\text{H} \text{ enriched in either deuterium or carbon.} \]

There are a large number of possible systems. These may be merely of one class type or combinations of two or three classes. Various combinations of frequencies are also possible e.g. IR: IR, IR:UV, UV:UV etc. Also the technique covers most, if not all, isotopic systems and elemental enrichment schemes.
CLAIMS:

1. A method for the separation of isotopes comprising irradiating a gaseous mixture of the isotope with a laser beam at a first selected wavelength and irradiating the product formed with a laser beam at a second selected wavelength to produce a final product highly enriched in the desired isotope.

2. A method for the separation of isotopes comprising irradiating a gaseous mixture of the isotope primary substrate and a suitable primary transfer agent in a first cell with a laser at a first selected wavelength to obtain an enriched product, removing the primary transfer agent, irradiating the enriched product with a secondary transfer agent in a second cell with a laser at a second selected wavelength, separating the product obtained from the secondary transfer agent and the depleted substrate, said product being highly enriched in the desired product.
FIG. 1

LASER #1

LASER #2

LASER #n

CELL #1

ABSORBER #1

CELL #2

ABSORBER #2

CELL #n

ABSORBER #n

DEPLETED SUBSTRATE

FINAL SEPARATOR

ENRICHED PRODUCTS

PRIMARY SUBSTRATE

PRIMARY TRANSFER AGENT

SECONDARY TRANSFER AGENT

\( \text{REF. IG. 1} \)