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Isotopes Separation Method using Physical Vapor Deposition Technique

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ABSTRACT

An isotope separation technique using effusive emission of vapors from the heated molybdenum boat is presented. The technique is applied for the separation of the lithium isotopes. Lithium fluoride with natural isotopic abundance was chosen for evaporation and it was achieved by resistive heating of the molybdenum boat with an exit orifice in the center that provides a point source emission. Glass substrates were placed in a semi-circle around the source of evaporation at different positions of peripheral region to deposit the evaporated material. A non-commercial laboratory developed linear Time of Flight (TOF) mass spectrometer was used for isotopic abundance measurements of lithium in the deposited thin films. The dependence of the size of exit orifice on the separation is also studied for the three exit orifices with diameters of 0.3, 0.6 and 1.0 mm. The separation factors of the isotopes as a function of different peripheral locations are calculated and presented. The abundance of the ^{6}Li isotope has been increased up to 16% on the peripheral positions.

1. Introduction

Different methods for isotope separation [1–8] have been studied in the last decades. However, each method has some limitations; for example in the gas centrifugation and diffusion methods, the limitation is the gas-phase problem that limits the production of many isotopic elements [1,2]; in the electromagnetic method, the limitations are a low output and high electricity consumption [3]. Laser isotope separation methods [4–7] are still in research phase and has small industrial production; chemical methods [8] of isotope separation are costly and have health hazards; the ion cyclotron resonance method [9] seems to be an universal isotope separation technique but is still under research and development phase. Due to the limitations in existing isotope separation techniques, the search for new, efficient and low cost methods is always demanded.

The present work elaborates a new, simple and very interesting process of isotope separation. The basic idea originated from the earlier work of Becker et al. [10,11] and in subsequent experiments on the same subject by Waterman and Stern [12]. The theoretical explanation is very interesting and it states that if a container has two different masses m_1 and m_2 in gaseous form such that $m_2 > m_1$ and both are effusing from an exit orifice, then the heavier masses will have maximum concentration in the forward direction whereas the lighter ones prefer to distribute themselves in the peripheral region. The reason is that if a gas mixture of different masses emerges from the exit orifice, the total velocity of any molecule is the vector sum of its stream and thermal velocities. The Maxwellian distributions in thermal velocity superposed on the stream velocity and it causes the beam of molecules to spread out or diverge from the stream direction. The average thermal velocities of the lighter constituent are larger than the heavier ones, so the flux will diverge more rapidly for the lighter ones than for the heavy molecules, producing an enrichment of the lighter constituent along the peripheral locations around the exit orifice.

Becker et al. [10,11] studied the free expanding jet of gas mixtures from circular or slit-shaped nozzles and applied it for a UF_6 jet with helium as the auxiliary gas deflected by a curved wall. They found that the degree of separation of the different masses depends upon many factors like beam exit geometry, mass difference of components and pressure difference between the inside and outside of the heating boat. Two important results were reported; first, the greater the mass difference between the gas constituents, the higher the separation factor, and secondly for good degree of separation, the flow of constituents must be effusive, i.e. collision free molecular flow.

Waterman and Stern [12] applied this technique for the separation of gas mixture from the hydro-dynamical kinetics of supersonic flow. They concluded that the separation is due to the manner in which stream and thermal velocities behave, rather than a hydrodynamic diffusion (supersonic expansion) flow. Mikami and Takashima [13] presented a mathematical treatment on separation of isotope mixtures in a free molecular jet and Anderson [14] described experimentally the spatial distributions of a flux in axisymmetric free jets of helium–argon mixtures. Later on Anderson

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[15,16] registered two patents on the process for separating gas mixtures, especially isotopes including uranium isotopes, i.e. $^{235}UF_6$ and $^{238}UF_6$. Severin and Lydtin [17] also reported the uranium isotope separation through chemical vapor deposition (CVD) method.

In this contribution, we, for the first time applied the idea described above for studying the isotope separation of the lithium through simple effusion from circular orifice. We also studied the dependence of the separation of the lithium isotopes on the size of the exit orifice. The separation factors for the lithium isotopes at different positions in the peripheral region around the point source are also determined from the experimental data.

2. Experimental setup

Lithium fluoride with natural isotopic abundance was chosen for evaporation because of its very low melting point and it was heated in a molybdenum boat with a cover having an exit orifice in the center. Fig. 1 shows the experimental setup for evaporation and deposition of samples on the glass substrates. The material was melted through resistive heating and deposited on glass substrates placed in a semi-circle dome of width 60 cm and height 50 cm around the source of evaporation at different angles to deposit the evaporated material. The exit orifice was circular to provide a point source emission. The molybdenum boat was heated at 773 K to produce the effusive flow for lithium atoms for thin film deposition. The flow of atoms from the exit orifice will be effusive [18] for $\lambda \ll a$ and $t \leqslant a$; here λ is the mean free path, a is the exit orifice diameter and t is the wall thickness of the exit orifice. In our case, for lithium, the mean free path at the boat temperature of 773 K was $\lambda \sim 4.8$ cm; $t \cong 0.1$ mm and three different diameters of 0.3, 0.6 and 1.0 mm of exit orifice of boat were used. For all the three exit orifice diameters of boat at the temperature of 773 K, the effusive flow of lithium atoms was therefore ensured. During the deposition process, the vacuum was of the order of 1×10^{-6} mbar. In the said way, we prepared three batches of thin film samples with exit orifices of diameters 0.3, 0.6 and 1.0 mm in the boat. Each batch held 19 glass substrates. The deposited thin films of lithium fluoride on the glass plates at different peripheral locations were analyzed in a non-commercial locally developed linear Time of Flight (TOF) mass spectrometer. It is a two-field acceleration linear mass spectrometer shown in Fig. 2. The thin film sample was placed between the ionization/extraction region of the TOF mass spectrometer. The grid G_2 was grounded to produce a pure field free region. A Nd:YAG laser (Quantel, Brilliant) at its fundamental wavelength (1064 nm) was used to ablate/ionize the thin films of lithium fluoride with 10 Hz repetition rate and laser pulse duration of 5 ns. The laser energy per pulse can be controlled up to the desired level by the flash lamp Q-switch delay pulse. The vacuum in the TOF analyzer was maintained at 1×10^{-7} mbar during the analysis of the isotopic composition. The photoions produced by the pulsed laser source were extracted by a continuous electric field E_{ext} and then accelerated by another continuous electric filed E_{acc} before entering TOF field free region. The lithium ions at the end of the field free region were detected by a channeltron (Burle USA, 4821G). A photodiode BPX65 was used for triggering the oscilloscope as well as for the Time of Flight (TOF) measurement of the isotopic signal peaks. Experimental data were captured on the digital interfaced oscilloscope TDS2024 (Tektronix) and transferred to the computer for further analysis.

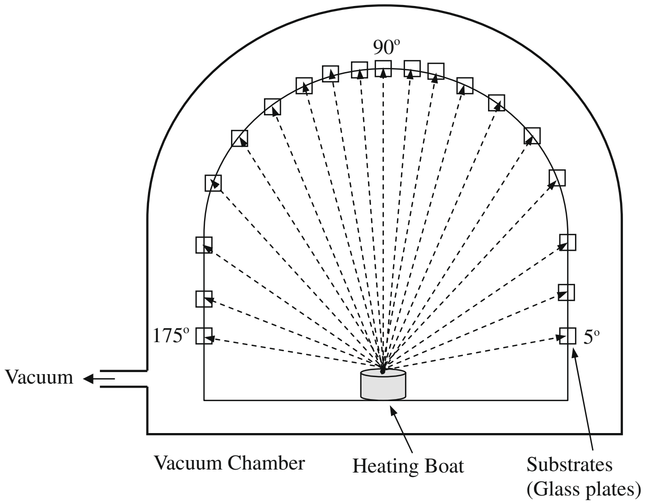

Fig. 1. Experimental setup for deposition of the lithium fluoride vapor on glass substrates in the peripheral locations around the exit orifice of the heated molybdenum boat.

3. Results and discussion

The isotope separation of the lithium isotopes is presented as a result of the effusive flow from the exit orifice of different diameters of the heated molybdenum boat. When both the lithium isotopes having masses 6 and 7 a.m.u. emerge effusively from the exit orifice with the same stream velocity parallel to the orifice axis and a randomly oriented Maxwellian thermal velocity distribution, then the total velocity of any isotope will be the vector sum of its stream and thermal velocities, as depicted in Fig. 3. Here u is the stream velocity and v is the thermal velocity of isotopes. Both the isotopes have same stream velocity and have Maxwellian distribution in thermal velocity, thereby, changing their paths around the peripheral locations. The Maxwell–Boltzmann distribution [19] of thermal velocities for both the lithium isotopes at the boat temperature of 773 K is plotted in the Fig. 4. The lighter lithium isotope ^{6}Li, on the average, has larger Maxwellian thermal velocities than that of ^{7}Li. The large average thermal velocities of ^{6}Li isotope causes it to diverge more from the stream axis than ^{7}Li, producing an enrichment of the lighter isotope on the peripheral locations. The results for the separation of the isotopes at peripheral positions are described in the following paragraphs.

Using the experimental setup, shown in Fig. 1, the thin films of lithium fluoride were deposited on the glass substrates through evaporation technique. We have produced three batches of deposited films of lithium fluoride effusing from the exit orifices with diameters of 0.3, 0.6 and 1.0 mm, each having 19 glass substrates. The thickness of lithium fluoride films was monitored by a quartz crystal and in all the three batches it was maintained at 50 nm with an error of ±10%. The deposited thin films of the three batches have been mass analyzed for both the lithium isotopes ^{6}Li and ^{7}Li with a locally developed TOF mass spectrometer. The detector applied voltages of the TOF mass spectrometer was optimized so that it distributes the isotopic signals on the time axis according to their isotopic abundance. The calibration of the TOF mass spectrometer for the lithium isotopes was performed by using a sample of known abundance. The abundance of the lithium isotopes of that sample was also confirmed from a magnetic sector mass spectrometer. Fig. 5 shows the TOF distribution of the lithium isotopes from the deposited thin film substrates only for three locations in the peripheral region for a batch of an exit orifice with the diameter of 0.6 mm. In Fig. 5, the middle trace is from the substrate at the axis of stream velocity (above the exit orifice), the upper and the lower traces are from the substrates at the extreme right and left peripheral positions, i.e. at (b) 5° and at (c) 175°. The reference laser pulse is used to designate the lithium isotopic masses. The isotopic concentrations of the ^{6}Li and ^{7}Li isotopes at each peripheral

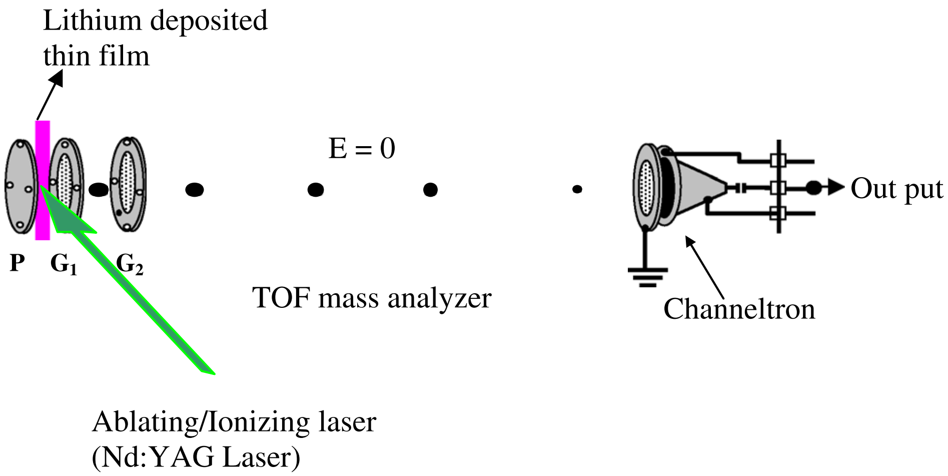

Fig. 2. Schematic layout of the laboratory developed linear Time of Flight (TOF) mass spectrometer with thin film sample holder.

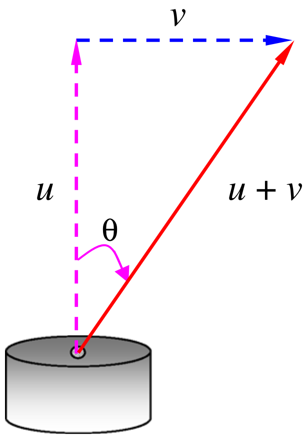

Fig. 3. A velocity vector diagram, showing effusively emerging of the isotope from the exit orifice of the heated molybdenum boat with the stream velocity u and thermal velocity v has a total velocity vector $u + v$ making an angle θ with the orifice axis.

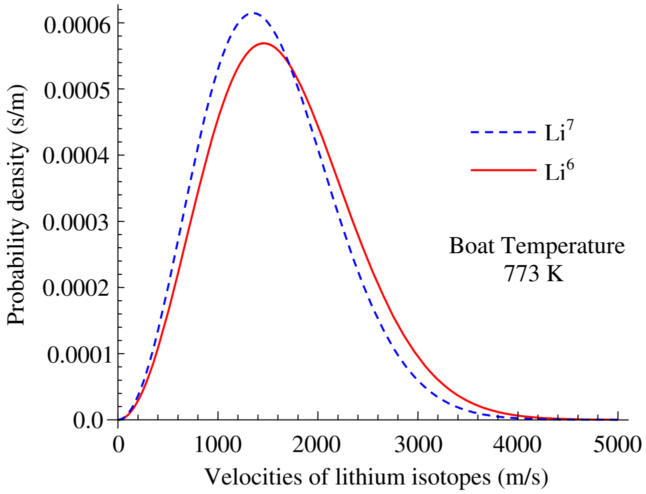

Fig. 4. The Maxwell–Boltzmann distribution of the velocities of lithium isotopes ^{6}Li and ^{7}Li in molybdenum boat at temperature of 773 K.

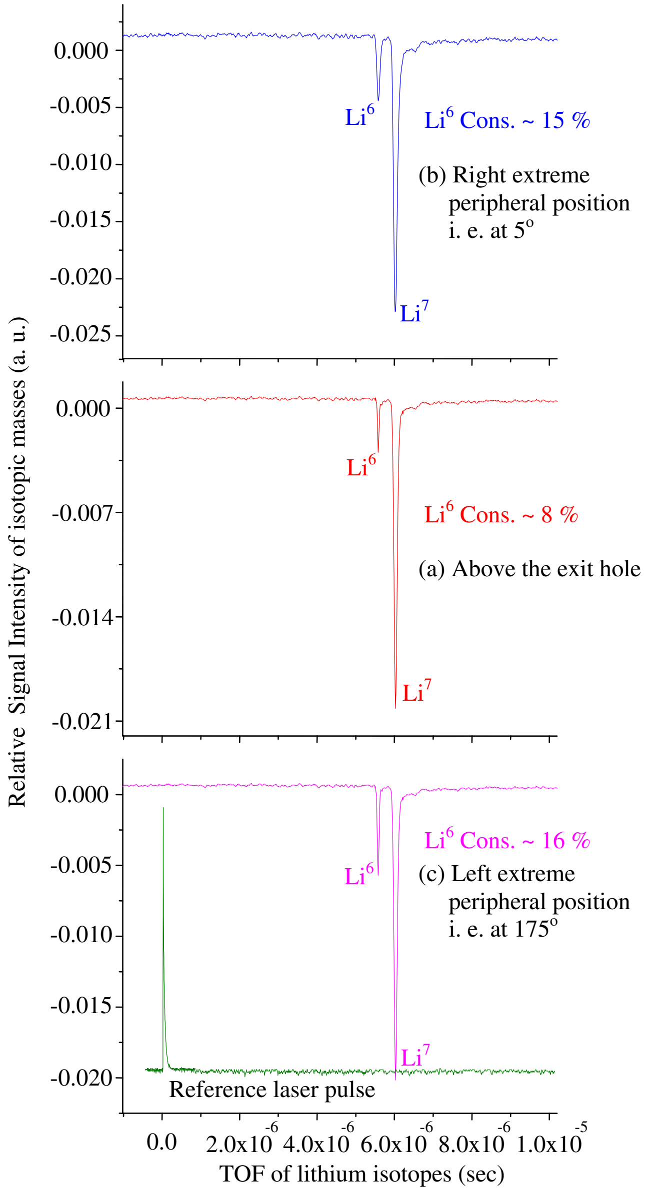

Fig. 5. TOF mass spectrum of the lithium isotopes ^{6}Li and ^{7}Li at three positions: (a) above the exit orifice, (b) at the left extreme and (c) at the right extreme.

Table 1
Separation factor for the lithium isotopes from different deposited thin films as a function of the angle from the oven exit hole of diameter 0.3, 0.6 and 1.0 mm.

Position of glass samples	Oven exit size		
(°)	0.3 mm	0.6 mm	1.0 mm
5	1.78 ± 0.27	2.19 ± 0.33	2.07 ± 0.31
15	1.64 ± 0.25	2.03 ± 0.30	2.07 ± 0.31
25	1.64 ± 0.25	1.89 ± 0.28	1.93 ± 0.29
35	1.51 ± 0.23	1.87 ± 0.28	1.78 ± 0.27
45	1.51 ± 0.23	1.72 ± 0.26	1.78 ± 0.27
55	1.39 ± 0.21	1.57 ± 0.24	1.64 ± 0.25
65	1.25 ± 0.19	1.57 ± 0.24	1.64 ± 0.25
75	1.12 ± 0.17	1.28 ± 0.19	1.51 ± 0.23
85	1.00 ± 0.15	1.14 ± 0.17	1.12 ± 0.17
90	1.00 ± 0.15	1.00 ± 0.15	1.00 ± 0.15
95	1.11 ± 0.17	1.25 ± 0.19	1.25 ± 0.19
105	1.25 ± 0.19	1.42 ± 0.21	1.25 ± .0.19
115	1.39 ± 0.21	1.57 ± 0.24	1.51 ± 0.23
125	1.51 ± 0.23	1.72 ± 0.26	1.51 ± 0.23
135	1.51 ± 0.23	1.72 ± 0.26	1.64 ± 0.25
145	1.64 ± 0.25	1.87 ± 0.28	1.64 ± 0.25
155	1.64 ± 0.25	1.87 ± 0.28	1.78 ± 0.27
165	1.78 ± 0.27	2.03 ± 0.30	1.93 ± 0.29
175	1.78 ± 0.27	2.19 ± 0.33	2.07 ± 0.31

location for the three batches were determined by integrating their signal peaks. For each spectrum the ratio of the isotopic concentration was determined by these integrating isotopic intensities. For the batch with exit orifice diameter of 0.6 mm, the concentration of ^{6}Li is enhanced up to ~15% at the right peripheral position, i.e. at 5° and 16% at the left peripheral position, i.e. at 175°. The accuracy of the measurement of isotopic signals lies within ±15%. Similar results with in experimental error were obtained from the other two batches having exit orifices of diameters 0.3 mm and 1.0 mm.

Using the integrating intensities of both the lithium isotopes ^{6}Li and ^{7}Li, we have calculated the separation factor α for the three batches using the relation [12]:

$$\alpha = \frac{N_H^C N_L^P}{N_L^C N_H^P}$$

where N_H and N_L represent the fraction of the heavy and light components of the mixture, and the superscripts C and P refer to the central and peripheral streams, respectively. The separation factors for the lithium isotopes from the deposited thin films in the three batches were calculated, as shown in Table 1. Table 1 predicts that nearly the same separation factors were found that may be due to the effusion flow [18] from the exit orifices with diameters 0.3, 0.6, 1.0 mm in the three batches. However, the best results were obtained with an exit orifice of 0.6 mm and are plotted in Fig. 6. In this figure, the separation factors of lithium isotopes from the 19 deposited films are displayed as a function of different peripheral locations around the exit point source. Fig. 6 clearly shows the separation trend of both the isotopes, i.e. the concentration of the ^{6}Li is smoothly increasing and that of ^{7}Li is decreasing at different peripheral positions. The error bar (±15%) results from the TOF mass spectrometric measurements. Evidently, there is a clear enhancement of the lighter isotope ^{6}Li at the peripheral positions and the enhancement in separation shows a smooth trend from center to peripheral locations. However, the concentration of both isotopes above the exit orifice remains as natural with in the experimental error. The results presented, have therefore a strong correlation with the theoretical basis.

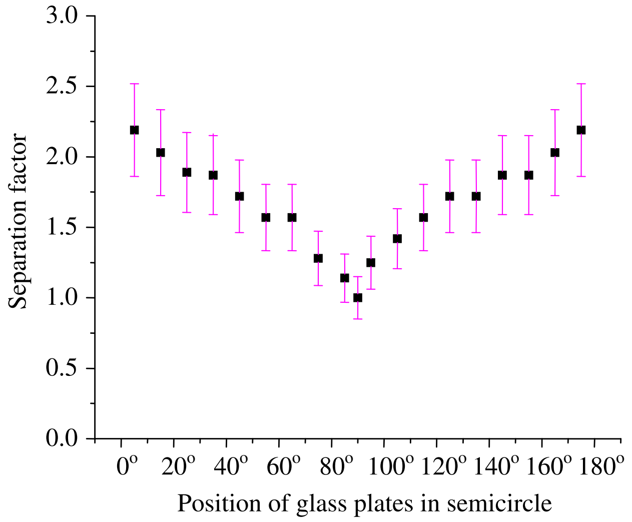

Fig. 6. Separation factor for the lithium isotopes ^{6}Li and ^{7}Li as a function of the peripheral locations around boat exit orifice of diameter 0.6 mm.

4. Conclusion

The experimental results presented above complement a similar investigation by Becker and co-workers but for metallic lithium as they have reported their results for gas mixtures. In this case the lithium metal has to be heated to convert it into gas phase. The other difference is that these results have been reported for lithium isotopes where the mass difference is only 1.0 amu. The results reflect the essence of the basic mechanism of the separation that the distribution of the thermal velocity of the lighter component is broader than the heavier. We consider this technique very simple and cheap to enhance the concentration of the isotopes of interest up to a certain extent. It is hoped that this paper may stimulate further work both in theoretical and experimental directions and in the end, to a better understanding of the separation phenomena.

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