Isotope Separation with Drift Tube, (I)

Basic Analysis and Experiment

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A new method of isotope detection and separation based on transport phenomena of ions in gases, which consist of a drift tube, is proposed and the analytical expression of resolution power of the method has been derived from a diffusion equation. The expression shows that the resolution power increases with the square root of the voltage supplied to the tube. A simple drift tube was constructed and natural Li was isotopically separated in Ne 100 Torr with the maximum resolution power, about 2. The variations of the resolution power with the applied voltage were measured by using the ion source of $^6$Li and $^7$Li separately and compared with calculations.

**KEYWORDS:** isotope separation, isotope detection, resolution power, drift tube, lithium, lithium 6, lithium 7

I. INTRODUCTION

Separation of isotopes tends to be indispensable not only in nuclear industry, but in other industrial and scientific fields and accordingly, a variety of elements are considered to be separated. In nuclear industry, a major concern has been concentrated on U. Nuclear fusion, however, requires a large amount of tritium which may be produced from $^6$Li. Many other nuclides such as $^{30}$Si, $^{50}$Ti and $^{90}$Zr are also valuable. Some are planned to be used in semiconductor and others are useful as tracers in compensation for lack of proper radionuclides. A number of techniques have been proposed and developed. However, a technique based on ions drifting in gases has been rarely published.

The present technique may be characterized by intermediate features between distillation and electromagnetic separation. That is, ions in gases collide so many times with gas molecules that their movement is affected by an interaction between ion and gas molecule. On the other hand, ions are forced to move along an applied electric field similar to electromagnetic separation. The intermediate features of the technique may be preferable in higher operational ionic density to electromagnetic separation and may have a higher separation factor than that of distillation.

In this paper we will report a basic analysis of the technique and preliminary experimental results of Li ions.

II. RESOLUTION POWER

Ions in gases drift along an applied electric field, while drift velocities of ions in gases depend on polarizabilities of gases and reduced masses of ion and gas. When we introduce ions consisting of a mixture of isotopes in a very short duration as compared with a flight time of ions into a region where an electric field is applied, they may arrive separately at the end of the region because of different drift velocities.

1. **Full Width at Half-Maximum (FWHM) of Arrival Time Profile of Ions**

The drift velocity of ions in gases $v_d$ is described by

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\[ v_d = K(E/N), \]  
where \( K \) is the mobility and \( E/N \) the electric field strength divided by gas number density. Using ordinary units, we may obtain a following expression:

\[ v_d = 268.68 \times K_0 \times (E/N), \quad \text{(cm/s)}, \]  
where \( K_0 \) is reduced mobility and \( (E/N) \) is in units of Td (Townsend),

\[ 1 \text{Td} = 1 \times 10^{-17} \text{V} \cdot \text{cm}^2. \]  

Assuming the configuration of electrodes in Fig. 1, a disk shaped ion swarm introduced into the drift region from the hole of A electrode moves down under the influence of an electric field and diffuses in a gas. An observable ion current \( I \) at time \( t \) and the position of 1 cm in the z-direction, is

\[ I = C t^{-4/3} \exp \left[ -\frac{(l-v_d t)^2}{4 D_L t} \right], \]  
where \( C \) is a constant and \( D_L \) the diffusion coefficients\(^{(2)(3)}\). The variation of the ion current with time gives us a spectrum, arrival time spectrum (ATS). Under usual conditions, where the flight time of ions surpasses appreciably FWHM, we can neglect the \( t^{-4/3} \) factor in the equation\(^{(3)}\) and with sufficient accuracy FWHM, i.e., \( t_{1/2} \) is

\[ t_{1/2} = \frac{4(D_L \ln 2 t_p)^{1/2}}{v_d}, \]  
where \( t_p \) is the arrival time at the maximum current of ATS.

\[ 2. \text{ Resolution Power} \]

The resolution power of the arrival time spectrum in drift tube, \( F_s \) may be defined by

\[ F_s = \frac{2(t_p - t_p')}{t_{1/2} + t_{1/2}'}, \]  
where primes indicate the lighter isotopes as shown in Fig. 2.

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where $\alpha$ is polarizability of buffer gas and $M_r$ reduced mass and assuming the ratio of the mass of ion to that of buffer gas is zero, we have found the limiting resolution power $F_s(\text{max})$

$$F_s(\text{max}) = \frac{1}{2} \left( \frac{eV}{kT \ln 2} \right)^{1/3} \left( \frac{1 - (m'/m)^{1/3}}{1 + (m'/m)^{1/3}} \right),$$

where $m$ is the mass of the ion.

### III. EXPERIMENTAL

1. **Apparatus**

   (1) **Applied Voltages to Drift Tube**

   Equation (9) implies that the resolution power of drift tube increases with increasing applied voltage to it with keeping a constant temperature of buffer gases. Applying a higher voltage, however, is restricted by starting an electrical gas discharge, which depends on various factors such as gas pressure, gas species, configuration of electrodes, and shape of insulators and electrodes.

   (2) **Gas Pressure**

   With increasing gas pressure the tube can be applied a higher voltage, while ions are clustered with buffer gas molecules at a higher gas pressure and the Langevin prediction (10) is no longer valid for these cluster ions. Usually, the mobility ratio of clustered ions is smaller than the square root of the mass ratio. The pressure that ion clustering reactions become significant, depends on various factors such as gas and ion species, flight time, gas temperature and $E/N$. Some of the rate coefficients for clustering reactions of Li ions have been published(3).

   (3) **Length of Drift Region**

   Equation (9) does not indicate a proper length of drift tube explicitly because the resolution power depends solely on the applied voltage. Taking account of electrical discharges, the tube having a longer length may be applied a higher voltage.

   (4) **Design of Drift Tube**

   Considering the factors described above we have constructed a drift tube shown in Fig. 3 in order to demonstrate that it can separate isotopes. All the components used in this tube were made of high vacuum material; stainless steel, ceramics, Cu metal gaskets, and hermetic sealed feedthroughs. At the bottom of the tube a feedthrough having 13 pins was mounted and electric power for ion source, and electrical potential to electrodes were supplied through these pins. A thermionic Li ion emitter consisted of a few turns of 0.1 mm Pt wire coated with Li$_2$O-Al$_2$O$_3$·SiO$_2$. The two stainless steel rings having a 20 mm orifice covered with 50 mesh gauze of 0.05 mm stainless steel wire set apart 0.7 mm. The pair of the rings worked as a electrical ion shutter.

   At the end of the tube a simple ion current collector was located and connected to a shielded wire for high vacuum that transferred signals with a minimum noise. A vacuum envelope was made of aluminum ceramics tube and the inner diameter of it was about 80 mm.

![Fig. 3 Schematic view of drift tube](image-url)
(6) Electronic Measuring System
A similar system used has been reported previously(2) and will be only briefly described here. A schematic diagram is shown in Fig. 4. The proper potentials for the electrodes were supplied from a voltage divider consisting of variable resistors. A emission regulator controlled the filament current to get stable Li ion current (emission current 1~0.1 μA). The upper pair of the electrodes in the figure, the first ion shutter, introduced an ion swarm into the drift region and the lower pair of the electrodes, the second shutter, allowed a part of ions arrived in a delayed time from the time of the opening of the first shutter to move to the collector (maximum collector current, about 1~0.1 nA). Both the shutters were opened in a short duration (about 1% of ion flight time) by triggering pulses from a digital delay scanner. Scanning the delay time between the first and the second shutters we obtained ion current profiles as a function of arrival time of ions ATS (arrival time spectra).

2. Experimental and Results
(1) In Case of Natural Li
With increasing gas pressure the higher voltage can be applied to the tube. A preliminary test shows that Li ions significantly cluster with Ar atom over 4 Torr. Even at the highest pressure the break down voltage still was not enough to separate distinctively Li isotopes. Then, we used Ne, which has a lower reaction rate coefficients than that of Ar, as a buffer gas. We obtained three ATSs in the pressure 5, 20 and 100 Torr at room temperature. The three ATSs are shown in Fig. 5(a)~(c).

![Fig. 4 Wiring diagram of drift tube](image)

![Fig. 5 ATS of Li ions in Ne with various resolution powers](image)
Fitting the right-hand side of a composite ATS with a calculated one, we obtained an ATS corresponding to \(^6\)Li with subtracting the calculated ATS from the composite one. The experimental resolution powers were derived from both the FWHM of the ATSs, the calculated and subtracted ATSs. The values of \(E/N\), the maximum resolution powers, and the peak current ratios are listed in Table 1, together with the applied voltages to the drift tube.

<table>
<thead>
<tr>
<th></th>
<th>Fig. 5(a)</th>
<th>Fig. 5(b)</th>
<th>Fig. 5(c)</th>
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</thead>
<tbody>
<tr>
<td>Exp. (F_s)</td>
<td>1.0</td>
<td>1.4</td>
<td>2.0</td>
</tr>
<tr>
<td>(F_s) (max)</td>
<td>1.53</td>
<td>2.73</td>
<td>3.72</td>
</tr>
<tr>
<td>Press. (Torr)</td>
<td>5.0</td>
<td>20</td>
<td>100</td>
</tr>
<tr>
<td>Voltage (V)</td>
<td>110</td>
<td>349</td>
<td>648</td>
</tr>
<tr>
<td>(E/N) (Td)</td>
<td>9.46</td>
<td>7.13</td>
<td>2.66</td>
</tr>
<tr>
<td>(K_6-6/K_6-7) (mobility ratio)</td>
<td>1.088</td>
<td>1.068</td>
<td>1.061</td>
</tr>
<tr>
<td>(I(\text{'Li})/I(\text{total})(%))</td>
<td>6.1</td>
<td>9.6</td>
<td>10.1</td>
</tr>
<tr>
<td>(I(\text{'Li})/I(\text{total})(%)) (intensity ratio)</td>
<td></td>
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As can be seen in Table 1, the experimental resolution powers are smaller than the calculated ones and the current ratio of \(^6\)Li to \(^{7}\)Li exceeds the natural abundance ratio of Li. We may ascribe the reason to the pulse width of the shutters and a drift of baseline.

(2) In Case of \(^6\)Li and \(^{7}\)Li

In order to obtain the further detail of dependence of the applied voltage and of mass of buffer gas on the resolution power we modified the ion source to be able to emit \(^6\)Li and \(^{7}\)Li separately without displacing the ion source to ensure the identical experimental conditions. The accurate concentrations of the enriched \(^6\)Li, \(^{7}\)Li used were not known. Each arrival time spectrum showed that the isotopical purity exceeded 95%. At several pressures the FWHM of each isotope were measured separately and the resolution powers were obtained as illustrated in Fig. 6(a)~(c).

At the low applied voltage region, the resolution power increases linearly with the square root of the voltage and then deviates from the straight line, which is expected with Eq. (9). In the high voltage region, rigorously in the high \(E/N\) region, the Einstein relation is no longer valid. The modified expression, which are valid in high \(E/N\) region, has been published\(^4\). We have derived the resolution power using the expression as follows:

![Fig. 6(a)~(c) Variations of resolution power of ATS as a function of applied voltage to drift tube](image)
where $V$ is an applied voltage, $K_0$ the reduced mobility, $N_0$ the gas number density at NPT (0°C, 760 Torr), $K_0(0)$ the zero-field reduced mobility, $m$ the ion mass, $M$ the mass of buffer gas and $T$ the gas temperature. The calculation shows that the $F_s$ varies with the square root of applying voltage as can be seen in Fig. 7. We did not superimpose the calculated results on experimental one because experimental initial condition such as initial pulse widths, and initial delay times were not known. According to our experimental results, the initial effective ion width and delay time depend on various factors such as ion mass, gas pressure, bias voltage of the shutter, pulse width of triggering pulse. However, it is noted that the calculated $F_s$ values varies quite similarly to the experimental ones.

$$F_s = \frac{1}{2} \left( \frac{V}{\ln 2} \right)^{1/2} \left( \frac{1 - K/K'}{1 + K/K'} \right) \frac{K_0 N_0}{2.31 \times 10^{14} K_0(0) T + 0.35 \times 10^{17} M(M+3.72m)K_0^2(E/N)^4} (M+1.91m)$$

IV. CONCLUSION

We have proposed a new technique for separation and detection of isotopes and demonstrated that natural Li was separated isotopically with the technique. The resolution power of pulse mode in the low ion energy region has been obtained. We have also derived the valid expression of the resolution power in the high ion energy region. The expression was not proved quantitatively but the experimental results are qualitatively in good agreement with our calculation. It is proved that the technique can be used in separation and detection of isotopes with proper equipments.

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(3) idem: ibid., 76, 5283 (1982).