The Chemla Effect in the Internal Mobilities of $(Li\text{-}NH_4)NO_3$ Melts

Isao Okada
Department of Electronic Chemistry, Tokyo Institute of Technology, Nagatsuta,
Midori-ku, Yokohama, Japan

Tetsuo Ayano and Kazutaka Kawamura
Research Laboratory for Nuclear Reactors, Tokyo Institute of Technology, O-okayama,
Meguro-ku, Tokyo, Japan

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Internal mobility ratios in melts of the system $(Li\text{-}NH_4)NO_3$ were measured with the Klemm method. Densities and electric conductivities of this system were also measured. From these data the internal mobilities b_{Li} and b_{NH_4} were calculated. On account of the inevitable thermal decomposition of NH_4NO_3 the range of temperatures and concentrations was limited. In these the mobility of NH_4^+ is considerably greater than that of Li^+. In contrast to other binary nitrate systems, the isotherms of the internal mobility of the larger cation distinctly decrease with a decrease of its concentration, in which the fact that NH_4NO_3, whose cation is moderately light though large in size, melts at relatively low temperature is reflected. The isotherms of b_{Li} and b_{NH_4} and the temperature dependence are discussed on the basis of the autor's interpretation so far presented.

Introduction

In previous papers [1, 2] we have proposed a method for enriching ^6Li by countercurrent electromigration in which NH_4NO_3 is placed in a large cathode compartment while $LiNO_3$ is located in a separation tube connected to it. If the mobility of Li^+ were greater than that of NH_4^+, this method would not work because Li would enter the cathode compartment where isotopic mixing takes place. It was the main aim of the present study to ascertain that the mobility of NH_4^+ is greater than that of Li^+ in all accessible mixtures of the two salts.

It is known [3, 4] that in some binary molten salt mixtures with a common anion the larger cation migrates faster than the smaller one, particularly at high concentrations of the larger cation. At temperatures in the range of the melting point of $LiNO_3$ (~ 264 °C) NH_4NO_3 decomposes vigorously; for example, it has been reported that a 15 g NH_4NO_3 sample looses 0.12% in 6 hr at 170 °C and 15% in 40 min at 240 °C [5]. Thus this system is only relatively stable at low temperatures, i.e. low concentrations of $LiNO_3$ (see Figure 1). We therefore had expected that NH_4^+ would be more mobile than Li^+ under all accessible conditions.

The internal mobility ratios were measured with the countercurrent electromigration method introduced by Klemm. Since no data on the molar conductivities of this binary system were available, the densities and the conductivities were also measured in order to enable the determination of the internal cation mobilities.

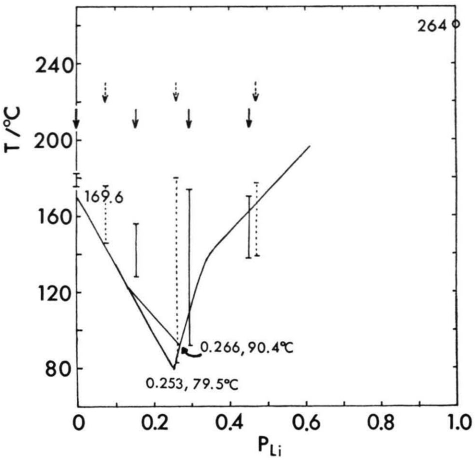

Fig. 1. Concentration and temperature ranges of the measurements of densities (-------) and conductivities (———), and liquidus lines [7]. The lower line corresponds to the metastable state.

Experimental

The chemicals $LiNO_3$ and NH_4NO_3 (p.a. grade) were dried at 120 °C for about 10 hr under vacuum

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Reprint requests to Dr. Isao Okada, Department of Electronic Chemistry, Tokyo Institute of Technology, Nagatsuta 4259, Midori-ku, Yokohama 227, Japan.

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before use. It was inferred from the generation of bubbles that NH_4NO_3 in mixtures with $LiNO_3$ starts to decompose slightly even below the melting point of NH_4NO_3 (~ 169.6 °C). One of the decomposition products is water [6]. It is therefore impossible to completely dehydrize these melts.

In the methods employed in this study, the accuracy of the measurements of the densities and conductivities is affected by the evolution of bubbles while that of the mobility ratios is not seriously affected. Therefore, the highest temperature was about 185 °C in the former measurements and about 210 °C in the latter ones. The lowest temperatures followed from the phase diagram [7] (see Figure 1). Since this system easily supercooles, some measurements were also done in this state.

The density was measured with the Archimedes method. A platinum bob (volume about 0.76 cm^3) was immersed in the melt and the buoyance was measured with a balance from which the bob was suspended with a thin platinum wire of 0.2 mm ∅. The bob was put into the melt at the beginning of each measurement. In the measurements above ca. 140 °C the readings of the balance slowly decreased on account of adhesion of bubbles to the bob and therefore these readings were extrapolated to zero time.

The conductivity was measured with a cell of the type proposed by Kochinashvili and Barzakovskii [8]. The cell constant, as measured with a standard KCl solution, was about 140 cm^{-1}. The temperature of the cell was kept within ±0.5 °C by keeping it in a melt bath. The cell was shaken before each measurement to remove bubbles sticking to the capillary wall (internal diameter was 2 mm). The resistance was measured at frequencies from 1 to 10 kHz; the frequency dependence was not clear.

The electromigration cell for the measurement of the internal mobility ratios from the concentration changes near the anode was similar to that previously used for the study of isotope effects [9]. In a large cathode compartment NH_4NO_3 was added to a mixture of $LiNO_3$, $NaNO_3$ and KNO_3. The presence of NH_4NO_3 led to the evolution of gas at the cathode and prevented electrodeposition of metals. The separation tube (internal diam. 4 mm, packed with quartz powder) and the small cathode compartment were filled with a mixture of $LiNO_3$ and NH_4NO_3 of known composition. The cell was made of Pyrex glass. At the relatively low temperatures of the experiments the Pyrex glass was not attacked by $LiNO_3$. Both anode and cathode were platinum wire of 1 mm ∅. The temperature was kept within ±3 °C in most runs.

After electromigration of 6—8 hr, the separation tube was taken out, allowed to cool, cleaned thoroughly on the outside and cut into several pieces for chemical analysis. Flame spectrophotometry and the Kjeldahl method were employed for the quantitative determination of Li^+ and NH_4^+, respectively. At least 3 trials were made for the NH_4^+ ions of each fraction.

In order to check that chemical fractionation did not occur during the filling process and that evaporation of NH_4NO_3 at the top of the separation tube would be negligibly small, blank tests were also made twice, in which the tube was filled with a melt of a certain composition and kept in the cell for about 8 hr at 200 °C without electric current: the distribution of the chemicals had remained uniform.

Results

The density data for every composition can be expressed by a linear function of temperature. The results are given in Table 1.

Since the range of temperatures was limited, the conductivities could be expressed by a linear func-

Table 1. Densities $d = a - (b \times 10^{-3})(T - 170)$; d in g cm^{-3}, T in °C.

p_{Li}	a (g cm^{-3})	b (g cm^{-1} $°C^{-1}$)	Temp. range (°C)	Maximum uncertainty[a] (%)
0.075 ± 0.003	1.466 ± 0.001	0.778 ± 0.056	146—176	0.1
0.260 ± 0.003	1.530 ± 0.001	0.675 ± 0.007	83—180	0.1
0.470 ± 0.002	1.587 ± 0.001	0.552 ± 0.029	139—177	0.1

[a] $\left| \frac{\text{Experimental value} - \text{Tabulated value}}{\text{Tabulated value}} \times 100 \right|$.

Table 2. Conductivities $\varkappa = a' \times 10^{-1} + (b' \times 10^{-3})(T-170) + (c' \times 10^{-6})(T-170)^2$ ($\varkappa$ in S cm^{-1}; T in °C).

p_{Li}	a' (S cm^{-1})	b' (S cm^{-1} °C^{-1})	c' (S cm^{-1} °C^{-2})	Temp. range (°C)	Maximum uncertainty (%)
0.154 ± 0.003	2.93 ± 0.03	2.50 ± 0.10	—	128–156	2.1
0.295 ± 0.008	2.86 ± 0.02	2.63 ± 0.05	—	86–174	4.5
	2.90 ± 0.02	3.01 ± 0.13	4.66 ± 1.06		2.0
0.451 ± 0.002	2.77 ± 0.02	2.84 ± 0.10	—	138–170	1.9

In pure NH$_4$NO$_3$, the $\varkappa$'s are 0.335 ± 0.001, 0.349 ± 0.001 and 0.356 ± 0.001 S cm^{-1} at 175°, 180° and 182 °C, respectively.

Table 3. The relative differences in internal mobilities.

Run No.	T (°C)	Mole fraction of LiNO$_3$	Electromign. duration (hr)	Transported charge (C)	$(b_{\mathrm{Li}} - b_{\mathrm{NH_4}})/\bar{b}$
1	183	0.075 ± 0.001	6.2	1128	− 0.317 ± 0.016
2	173	0.086 ± 0.001	6.3	1145	− 0.329 ± 0.011
3	155	0.088 ± 0.002	6.6	1181	− 0.246 ± 0.013
4	195	0.096 ± 0.002	6.0	1065	− 0.271 ± 0.016
5	192	0.107 ± 0.001	6.3	1249	− 0.278 ± 0.009
6	182	0.111 ± 0.002	6.3	1093	− 0.288 ± 0.017
7	164	0.128 ± 0.002	6.4	1230	− 0.289 ± 0.015
8	211	0.161 ± 0.003	6.5	1210	− 0.461 ± 0.029
9	181	0.167 ± 0.003	7.0	989	− 0.294 ± 0.021
10	182	0.167 ± 0.004	8.5	1144	− 0.255 ± 0.025
11	195	0.182 ± 0.003	6.6	1077	− 0.426 ± 0.023
12	193	0.186 ± 0.003	7.0	1152	− 0.378 ± 0.023
13	212	0.205 ± 0.003	6.0	1043	− 0.300 ± 0.021
14	182	0.239 ± 0.005	7.3	850	− 0.400 ± 0.019
15	197	0.242 ± 0.003	7.3	1094	− 0.427 ± 0.030
16	183	0.244 ± 0.003	6.0	1207	− 0.307 ± 0.008
17	197	0.244 ± 0.004	8.5	1027	− 0.379 ± 0.020
18	154	0.248 ± 0.002	6.4	1303	− 0.289 ± 0.009
19	182	0.246 ± 0.003	6.6	1344	− 0.320 ± 0.010
20	153	0.246 ± 0.002	6.9	1357	− 0.261 ± 0.005
21	165	0.257 ± 0.006	7.5	1428	− 0.295 ± 0.017
22	161	0.258 ± 0.003	6.5	1204	− 0.399 ± 0.013
23	197	0.259 ± 0.005	6.6	1390	− 0.319 ± 0.012
24	190	0.260 ± 0.003	8.3	1687	− 0.348 ± 0.013
25	138	0.262 ± 0.003	6.0	1097	− 0.371 ± 0.013
26	180	0.272 ± 0.003	9.7	1833	− 0.240 ± 0.009
27	200	0.272 ± 0.003	8.2	1242	− 0.308 ± 0.025
28	168	0.272 ± 0.003	6.0	1174	− 0.379 ± 0.008
29	142	0.276 ± 0.002	6.2	1216	− 0.303 ± 0.007
30	197	0.282 ± 0.003	6.0	1134	− 0.383 ± 0.014
31	185	0.283 ± 0.003	6.0	1132	− 0.369 ± 0.012
32	140	0.289 ± 0.001	8.0	1577	− 0.196 ± 0.005
33	194	0.294 ± 0.002	6.0	1204	− 0.355 ± 0.013
34	190	0.295 ± 0.003	8.0	1598	− 0.399 ± 0.010
35	173	0.300 ± 0.002	7.0	1295	− 0.274 ± 0.007
36	195	0.317 ± 0.003	9.0	2084	− 0.244 ± 0.006
37	154	0.329 ± 0.003	5.9	1123	− 0.308 ± 0.008
38	158	0.333 ± 0.003	6.0	1218	− 0.304 ± 0.009
39	191	0.356 ± 0.003	5.8	1086	− 0.337 ± 0.008
40	182	0.362 ± 0.002	6.0	1125	− 0.282 ± 0.005
41	182	0.466 ± 0.002	5.9	1029	− 0.358 ± 0.012
42	181	0.472 ± 0.003	5.8	1210	− 0.328 ± 0.009
43	194	0.491 ± 0.003	6.0	1264	− 0.292 ± 0.006
44	194	0.496 ± 0.003	5.9	1249	− 0.257 ± 0.007
45	175	0.517 ± 0.003	7.4	1019	− 0.289 ± 0.015
46	180	0.521 ± 0.003	7.6	1800	− 0.237 ± 0.007
47	170	0.560 ± 0.004	9.4	1482	− 0.280 ± 0.006
48	190	0.564 ± 0.004	8.4	1798	− 0.289 ± 0.005

tion of temperature too (Table 2). For $p_{\mathrm{Li}} = 0.295$ (p: mole fraction), a quadratic equation appeared to give a better fit.

The relative differences in internal mobilities of Li$^+$ and NH$_4^+$ are defined as

$$\varepsilon_{12} = (b_1 - b_2)/\bar{b}\,, \tag{1}$$

where b is the cation mobility relative to the anion, and $\bar{b}$ is the average mobility, that is, $\bar{b} = p_1 b_1 + p_2 b_2$; the subscripts 1 and 2 stand for Li$^+$ and NH$_4^+$, respectively. The measured values of ε_{12} are given along with the main experimental conditions in Table 3. The reproducibility is not as good as in other nitrate systems so far studied with the Klemm method, partly because of the thermal decomposition and partly because of the inevitably low current densities.

Discussion

The molar volumes calculated from the densities at 170 °C are shown in Fig. 2, where the density of pure LiNO$_3$ is estimated from that of the melt:

$$d(\mathrm{g\ cm^{-3}}) = 2.068 - 0.546 \times 10^{-3}\,T$$

(T: temperature in K) [10]. It is interesting to note that the linear extrapolations for pure LiNO$_3$ with respect to temperature and for 170 °C with respect to concentration yield the same molar volume of a fictive supercooled molten LiNO$_3$ at 170 °C.

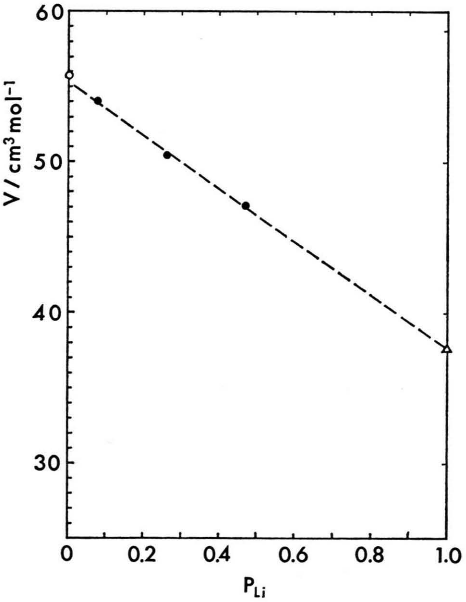

Fig. 2. Molar volume isotherm at 170 °C. The values for pure NH$_4$NO$_3$ (○) and pure LiNO$_3$ (△) [10], are taken from [11] and [10], respectively, by extrapolation.

Reliable measurements of the density of pure NH$_4$NO$_3$ could not be achieved owing to the evolution of bubbles. The molar volume of NH$_4$NO$_3$ at 170 °C extrapolated with respect to concentration is 55.10 cm^3 mol^{-1}; this is in good agreement with 55.17 cm^3 mol^{-1}, which is extrapolated with respect to concentration and temperature from the values obtained in the system NH$_4$NO$_3$—H$_2$O [11]. This value for pure NH$_4$NO$_3$ is comparable with the corresponding extrapolated values for pure RbNO$_3$ and TlNO$_3$, which are 56.32 and 53.60 cm^3 mol^{-1}, respectively. The order of these values is approximately in accord with that of the Pauling ionic radii of these cations (Rb$^+$: 148 pm, Tl$^+$: 140 pm, and NH$_4^+$: 148 pm).

The conductivity of pure NH$_4$NO$_3$ was measured at 3 temperatures (see Table 3), and is in satisfactory agreement with very old values [12]. To our knowledge there are no literature data on the conductivity of (Li-NH$_4$)NO$_3$ melts.

The molar conductivity of pure LiNO$_3$ at 170 °C, extrapolated with respect to temperature by use of the recommended values of conductivity and density [10], is

$$1.38 \times 10^{-3}\ \mathrm{S\ m^2\ mol^{-1}}$$

($b_{\mathrm{Li}} = 1.43 \times 10^{-8}\ \mathrm{m^2\ V^{-1}\ s^{-1}}$). The molar conductivity of a pure NH$_4$NO$_3$ at 170 °C is estimated from the present data to be $1.75 \times 10^{-3}\ \mathrm{S\ m^2\ mol^{-1}}$ ($b_{\mathrm{NH_4}} = 1.81 \times 10^{-8}\ \mathrm{m^2\ V^{-1}\ s^{-1}}$). Thus, the molar conductivity of pure NH$_4$NO$_3$ melt is greater than that of an imaginary supercooled LiNO$_3$ melt, whereas the ionic radius of an NH$_4^+$ ion is much greater than that of a Li$^+$ ion. Such an inverse relationship has been found, for example, for some alkali fluoride melts [13].

The molar conductivities of the present mixtures deviate negatively from additivity as in most of other binary melt systems.

As seen from Table 3, ε_{12} is negative at all accessible conditions, that is, the mobility of the NH$_4^+$ ion is greater than that of the Li$^+$ ion. The internal mobilities of these ions are calculated from

$$b_1 = (\Lambda/F)\,(1 + p_2\,\varepsilon_{12})\,, \tag{2}$$

$$b_2 = (\Lambda/F)\,(1 - p_1\,\varepsilon_{12})\,, \tag{3}$$

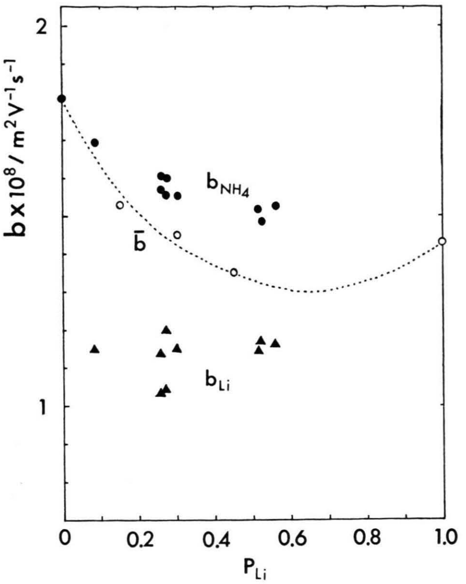

Fig. 3. The isotherms of b_{Li} (▲) and b_{NH_4} (●) and the average b (○) at 170 °C. The internal mobility of a fictive supercooled pure LiNO$_3$ is also shown.

where Λ is the molar conductivity, and F the Faraday constant.

The isotherms of the internal mobilities of the two cations at 170 °C are shown in Fig. 3 together with that of the average ones ($\bar{b}$). For the evaluation of the isotherms of the two cations, 9 data of ε_{12} in Table 3 ranging from 160 °C to 180 °C are adopted, since the temperature dependence of ε_{12} appears to be rather small.

In a previous paper [14], we defined the Chemla effect as follows: the difference of internal mobilities of two pure salts with a common ion changes sign on mixing in certain ranges of temperature and composition. This definition holds only if the two pure salts are molten at the temperature of the mixture. The following definition can be also applied in our case: in a charge symmetric binary system with a common ion, the mobility of the larger ion is greater than that of the smaller one in certain ranges of temperature and composition. According to the latter definition, the Chemla effect occurs at all coverable conditions under ambient pressure in the present system. Our general interpretation for this effect has already been given in previous papers [14, 15]; in brief, it takes into consideration the motion of ions based on the assumption that the internal mobility is related with the separating motion of neighbouring unlike ions.

In contrast to most other binary nitrate systems thus far studied, the internal mobility of the larger cation, i.e. the NH$_4^+$ ion in the present case, distinctly decreases with decreasing concentration. We have pointed out that two factors, acting in opposite directions, rule the internal mobility of the larger cation, namely (a) agitation of the shape of free space by the smaller and hence (in most cases) lighter cation and (b) decrease of free space with addition of the salt containing the smaller cation [14, 15]. It should be noted that free space is also needed, to some extent, for the agitation to be effective. In the present case, the mass of an NH$_4^+$ ion is moderately small and not vastly different from that of a Li$^+$ ion. Therefore, the addition of LiNO$_3$ to NH$_4$NO$_3$ would not result in an appreciable enhancement of the agitation effect. On the other hand, it will surely reduce the free space, which is small in melts containing Li$^+$ ions as main cations particularly at low temperature. This is also expected from the decreasing isotherm of the molar volume illustrated in Figure 2. These would be the reasons why b_{NH_4} decreases with decreasing concentration. In other words, the fact that NH$_4$NO$_3$, whose cationic species is moderately light though large in size, melts at rather low temperature is explicitly reflected in this isotherm of the internal mobility.

Some 10 years ago Moynihan [16] summarized the data on monovalent cation internal mobilities in mixtures with a common anion to produce two generalizations. One of them, mainly based on which the polarization model [17] is claimed [16, 18], is as follows: an increase in the mole fraction of the salt with the larger cation will result in a decrease in both cationic mobilities. The system (Li-K)$_2$SO$_4$ is an exception (b_K decreases with decreasing p_K [19, 20]), as pointed out by himself [16]. Also alkali fluoride systems such as (Li-Na)F, (Li-K)F and (Li-Cs)F have been assumed to be exceptional cases [13]. The present system gives rise to another clear exceptional example. The polarization model could not explain these "exceptional" cases.

As for the internal mobility of the Li$^+$ ion, the imaginary b_{Li} seems to decrease with decreasing concentration of Li$^+$ in the high concentration region, as in other binary nitrate systems. In the low concentration region, the isotherm appears to be nearly horizontal. Such a trend has been observed also in the system (Li-Na)NO$_3$ [15], which could not be accounted for in terms of the polarization model. This would be mainly due to the movement of NO_3^- ions, which is facilitated by an increase of free space with a decrease in Li^+ concentration, since the agitation effect does not decrease so much by the replacement of Li^+ ions by moderately light cations such as NH_4^+ or Na^+ ions.

It has been found [15] that b_{Li} in the system $(Li\text{-}M)NO_3$ ($M = K^+$, Rb^+ and Cs^+) is well expressed by

$$b_{Li} = \{A/(V - V_0)\} \exp(-E/RT), \quad (4)$$

where

$$A = 2.84 \times 10^{-11}\, m^5\, V^{-1}\, s^{-1} \quad \text{and}$$
$$E = 17.8\, kJ\, mol^{-1},$$
$$V_0 = 2.47 \times 10^{-5}\, m^3\, mol^{-1},$$

V is the molar volume, T the temperature and R the gas constant. If this is applied to the present system, the b_{Li}'s at 170 °C are, for example, 0.86 and $1.04 \times 10^{-8}\, m^2\, V^{-1}\, s^{-1}$ at $p_{Li} = 0.25$ and 0.50, respectively, whereas they are estimated to be ca. 1.10 and $1.15 \times 10^{-8}\, m^2\, V^{-1}\, s^{-1}$, respectively, as seen from Figure 3. Thus, b_{Li} in the present system is greater than expected from (4), particularly at higher concentration of NH_4NO_3. This is another consequence of the above stated fact that b_{Li} is nearly constant in this concentration range.

For pure $LiNO_3$ the value

$$b_{Li} = 1.74 \times 10^{-8}\, m^2\, V^{-1}\, s^{-1}$$

expected from (4) deviates positively from the fictive "experimental" value of supercooled $LiNO_3$ $b_{Li} = 1.43 \times 10^{-8}\, m^2\, V^{-1}\, s^{-1}$. This seems reasonable, because even at higher temperatures in pure $LiNO_3$ or at high concentrations of $LiNO_3$ in other systems b_{Li} deviates negatively from the values expected from (4), probably owing to the fairly small free-space [15].

For the purpose of learning the temperature dependence of ε_{12}, 21 data (Run No. 14—34 in Table 3) at the concentrations $0.239 \leqq p_{Li} \leqq 0.295$ are selected for plotting against temperature (Fig. 4), since a relatively wide range of temperatures can be spanned around this concentration range. It seems that the absolute values of ε_{12} tend to increase slightly with temperature. This trend can be expected, since the expansion of the free space due to an increase in temperature is more favourable to the movement of NH_4^+ than of Li^+ ions.

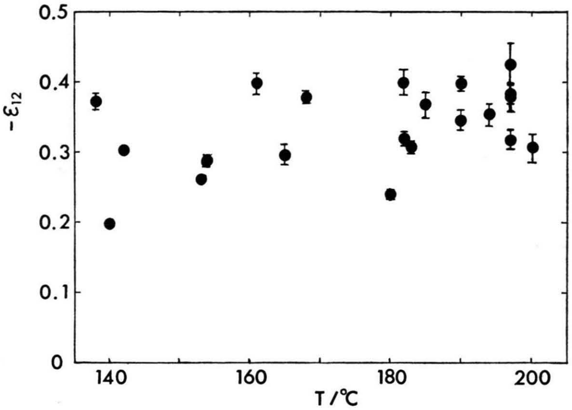

Fig. 4. Temperature dependence of ε_{12} in a concentration range $0.239 \leqq p_{Li} \leqq 0.295$.

Conclusion

The Chemla effect occurs under all accessible conditions in the molten system $(Li\text{-}NH_4)NO_3$. Therefore, as far as their mobilities are concerned, there will arise no problems in the previously proposed method for enrichment of ^6Li.

The isotherms of the internal mobilities of the two cations present rather strange features. (1) The internal mobility of the larger cation is greater than that of the smaller one even in their respective pure salts if the mobility of Li^+ is assumed to be that of an imaginary supercooled $LiNO_3$ melt. (2) The internal mobility of the larger cation decreases with decreasing concentration. This reflects the fact that NH_4NO_3 melts at relatively low temperature and that the NH_4^+ ion is moderately light though large in size.

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[1] I. Okada and N. Saito, J. Nucl. Sci. Technol. **11,** 314 (1974).
[2] I. Okada, Z. Naturforsch. **33a**, 498 (1978).
[3] J. Périé and M. Chemla, C. R. Acad. Sci. Paris **250,** 3986 (1960).
[4] F. Lantelme and M. Chemla, Bull. Soc. Chim. Fr. **1963,** 2200.
[5] J. D. Brandner, N. M. Junk, J. W. Lawrence, and J. Robins, J. Chem. Eng. Data **7,** 227 (1962).

[6] Gmelin Handbuch der anorganischen Chemie, 8. Auflage, System-Nummer 23 Ammonium, Verlag Chemie, Berlin 1936, p. 108.
[7] E. O. Holmes, Jr., E. O'Connell, and F. Hankard, J. Amer. Chem. Soc. **73**, 2885 (1951).
[8] V. A. Kochinashvili and V. P. Barzakovskii, Zhur. Priklad. Khim. **30**, 1755 (1957).
[9] N. Saito, K. Hirano, K. Okuyama, and I. Okada, Z. Naturforsch. **27a**, 288 (1972).
[10] G. J. Janz, F. W. Dampier, G. R. Lakshminarayanan, P. K. Lorenz, and R. P. T. Tomkins, Molten Salts: Vol. 1, Electrical Conductance, Density, and Viscosity Data, NSRDS-NBS **15**, Nat. Bur. Stand., Washington 1968.
[11] F. M. A. Hoeg, Z. Anal. Chem. **81**, 115 (1930).
[12] L. Poincaré, C. R. Acad. Sci. Paris **108**, 138 (1889).
[13] R. Takagi, Y. Uchiyama, I. Okada, and K. Kawamura, Denki Kagaku **45**, 666 (1977).
[14] I. Okada, R. Takagi, and K. Kawamura, Z. Naturforsch. **35a**, 493 (1980).
[15] C. Yang, R. Takagi, and I. Okada, Z. Naturforsch. **35a**, 1186 (1980).
[16] C. T. Moynihan, Ionic Interactions, ed. S. Petrucci, Vol. **1**, Academic Press, New York **1971**, Chapter 5, p. 261.
[17] C. T. Moynihan and R. W. Laity, J. Phys. Chem. **68**, 3312 (1964).
[18] S. I. Smedley, The Interpretation of Ionic Conductivity in Liquids, Plenum, New York and London 1980.
[19] A. Kvist, Z. Naturforsch. **21a**, 1221 (1966).
[20] V. Ljubimov and A. Lundén, Z. Naturforsch. **21a**, 1592 (1966).