

DISSOCIATION OF ANIONS  $\text{AlF}_6^{3-}$   
IN SYSTEMS CONTAINING  $\text{Li}_3\text{AlF}_6$ . I.

THE SYSTEMS  $\text{LiF-Li}_3\text{AlF}_6$  AND  $\text{Li}_3\text{AlF}_6\text{-AlF}_3$

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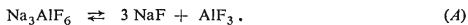
Grjotheim's method of calculating the degree of dissociation of the complex anions  $\text{AlF}_6^{3-}$  was applied to the systems,  $\text{LiF-Li}_3\text{AlF}_6$  and  $\text{Li}_3\text{AlF}_6\text{-AlF}_3$ . In the calculations the following fundamental data were used:

$$T_f^f(\text{Li}_3\text{AlF}_6) = 782^\circ\text{C}, \quad \Delta H_{(\text{Li}_3\text{AlF}_6)}^f = 20\,500 \text{ cal mol}^{-1}.$$

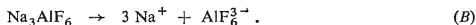
The experimentally established and calculated liquidus curves of  $\text{Li}_3\text{AlF}_6$  in the system  $\text{LiF-Li}_3\text{AlF}_6$  do not differ by more than  $3^\circ\text{C}$  within the concentration interval (100, 60 mol%)  $\text{Li}_3\text{AlF}_6$  if the degree of dissociation of  $\text{AlF}_6^{3-}$ ,  $\beta$ , is taken to be 0.35. However, in the system  $\text{Li}_3\text{AlF}_6\text{-AlF}_3$ , for the same value of  $\beta$ , the deviation of  $3^\circ\text{C}$  between the experimental and computed liquidus curves holds only within the concentration interval (100–90 mol%)  $\text{Li}_3\text{AlF}_6$ .

Since both  $\text{Na}_3\text{AlF}_6$  and  $\text{Li}_3\text{AlF}_6$  manifest themselves as a rounded maximum on the liquidus curves  $\text{NaF (LiF)-AlF}_3$  this is to be expected that the complex anions  $\text{AlF}_6^{3-}$  dissociate partially during the melting process. The literature is concerned mostly with the problem of dissociation of sodium cryolite, for two reasons: first, because of its industrial importance, and second, because only for this cryolite the necessary experimental data has been established.

Pearson and Waddington<sup>1</sup> suggested that  $\text{Na}_3\text{AlF}_6$  dissociated during melting according to Eq. (A)



The degree of dissociation of sodium cryolite was estimated to be 0.15–0.20. The remaining molecules of  $\text{Na}_3\text{AlF}_6$  dissociate as follows



The molecules of  $\text{NaF}$  formed by Eq. (A) yield  $\text{Na}^+$  and  $\text{F}^-$ , and the formed molecules of  $\text{AlF}_3$  react together



Abramov and coworkers<sup>2</sup> reported that using density data the degree of dissociation in Eq. (A) was found to equal 0.15 at 1000°C. Sodium cryolite was assumed to form ions by Eq. (B). The anions  $\text{AlF}_6^{3-}$  react further either by Eq. (D)



or by Eq. (E)



Belyaev and coworkers<sup>3</sup> believed that there were only ions present in the cryolite melts, namely  $\text{Na}^+$  and  $\text{AlF}_6^{3-}$ , the latter being partially dissociated into  $\text{Al}^{3+}$  and  $\text{F}^-$  according to Eq. (D).

A classical thermodynamic method of determination of the degree of dissociation of  $\text{AlF}_6^{3-}$  in molten sodium cryolite was applied by Grjotheim<sup>4</sup>. The most probable scheme of dissociation of  $\text{AlF}_6^{3-}$  ions was shown to be



The degree of dissociation was reported<sup>4</sup> to be 0.30, or, using a more exact value of the enthalpy of melting of sodium cryolite<sup>5</sup>,  $\beta = 0.25$ .

Similar conclusions were made by Frank and Foster<sup>6</sup> and Rolin<sup>7,8</sup>, and later by Vetyukov<sup>9</sup>. Another method for calculating the degree of dissociation of  $\text{AlF}_6^{3-}$  was suggested by Cochran<sup>10</sup>. However, the two latter methods are open to criticism.

The only direct measurement of the degree of dissociation of  $\text{AlF}_6^{3-}$  in molten sodium cryolite was reported by Solomons and coworkers<sup>11</sup>. They investigated the Raman spectrum of molten  $\text{Na}_3\text{AlF}_6$  at 1030°C using laser excitation. The results suggested the presence of both  $\text{AlF}_6^{3-}$  and  $\text{AlF}_4^-$  particles in the melt, the degree of dissociation of  $\text{AlF}_4^-$ , according to Eq. (F), being 70–75%. This appears to be a surprisingly high value.

Thus the presence of  $\text{AlF}_4^-$  anions in molten sodium cryolite is well-established. The equilibrium coexistence of different anions in the melt means that the experimentally determined melting point of pure sodium cryolite corresponds to the nonvariant equilibrium between the solid  $\text{Na}_3\text{AlF}_6$  (in which there is either no dissociation of  $\text{AlF}_6^{3-}$  ions at all, or if such a dissociation exists then only to a comparatively small extent, of order of 1%), and a mixture of ions in a fixed

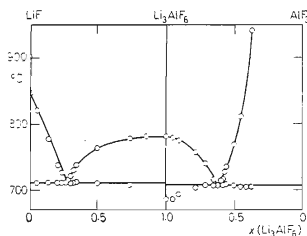


FIG. 1

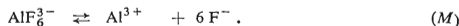
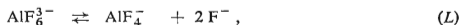
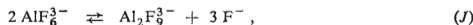
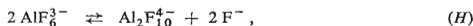
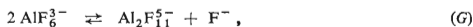
The Phase Diagrams of the Systems  
 $\text{LiF-Li}_3\text{AlF}_6$  and  $\text{Li}_3\text{AlF}_6\text{-AlF}_3$

proportion at a given temperature. The ideal melting point of an absolutely undissociated cryolite is therefore considerably higher than the experimentally determined melting point of the above-mentioned "ionic mixture".

The phase diagram of the system  $\text{LiF}-\text{AlF}_3$  has been re-established by Malinovský and coworkers<sup>12</sup> and it has been verified that the solid solutions are absent in this system (Fig. 1). The same author has determined the unknown value of the enthalpy of melting of lithium cryolite<sup>13</sup>. This enables Grjotheim's procedure to be applied to the lithium cryolite in the systems  $\text{LiF}-\text{Li}_3\text{AlF}_6$  and  $\text{Li}_3\text{AlF}_6-\text{AlF}_3$ , respectively.

Recently, two publications dealing with the dissociation of  $\text{AlF}_6^{3-}$  anions in lithium cryolite at its melting point appeared: a paper by Rolin and coworkers<sup>14</sup> and a thesis by Jenssen<sup>15</sup>. In the former the degree of dissociation of  $\text{AlF}_6^{3-}$  was found to be 0.30, in the latter 0.20.

Grjotheim and his coworkers<sup>5</sup> suggested a complete dissociation of sodium cryolite according to Eq. (B) and a partial dissociation of  $\text{AlF}_6^{3-}$  anions according to one of six different schemes:



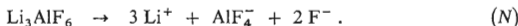
Assuming that the molten mixture under consideration can be regarded as an ideal ionic solution and choosing one of these dissociation schemes and a certain value of the dissociation degree,  $\beta$ , ( $0 < \beta < 1$ ), it is possible to calculate the theoretical course of the liquidus curve of sodium cryolite in the systems with either  $\text{NaF}$  or  $\text{AlF}_3$ . It was found that the most probable scheme is Eq. (L), this being supported by other published results<sup>16-18</sup>, in which the existence of the compound  $\text{NaAlF}_4$  was established. The degree of dissociation of  $\text{AlF}_6^{3-}$  anions was reported to be 0.23–0.25 (23–25%) and the corresponding dissociation constant 0.03–0.04.

## THEORETICAL

### *Dissociation of $\text{AlF}_6^{3-}$ Anions in Molten $\text{Li}_3\text{AlF}_6$*

We assume that each molecule of  $\text{Li}_3\text{AlF}_6$  yields three cations  $\text{Li}^+$  and a mixture of anions  $\text{AlF}_6^{3-}$ ,  $\text{AlF}_4^-$  and  $\text{F}^-$  according to the scheme (L). Two arguments support this suggestion. These are the fact that the compound  $\text{LiAlF}_4$  has been detected in the gaseous phase<sup>19-21</sup>, and also the results of cryoscopic measurements in the system  $\text{Li}_3\text{AlF}_6-\text{KCl}$  by Malinovský<sup>22</sup>. The latter measurements showed that within the concentration range 0.5–2.0 mol%  $\text{Li}_3\text{AlF}_6$  in  $\text{KCl}$ , each molecule of  $\text{Li}_3\text{AlF}_6$

brought six new particles into the molten KCl. The most probable explanation is the complete dissociation of lithium cryolite according to the scheme



If the concentration of lithium cryolite in the mixture with potassium chloride is higher than 2 mol%, then only a partial dissociation of  $\text{AlF}_6^{3-}$  anions takes place. Therefore the presence of  $\text{AlF}_4^-$  anions in the melt of pure  $\text{Li}_3\text{AlF}_6$  seems to be the obvious one.

Let us denote the degree of dissociation of  $\text{AlF}_6^{3-}$  anions according to the scheme (L) as  $\beta$ . We shall assume that the melt is regulated by the laws of ideal ionic solutions formulated by Temkin<sup>23</sup> and that within the considered temperature interval (less than 75°C) both the dissociation constant of  $\text{AlF}_6^{3-}$  and the enthalpy of melting of  $\text{Li}_3\text{AlF}_6$  are not temperature dependent.

If one mol of  $\text{Li}_3\text{AlF}_6$  melts, then a mixture of 3 mol  $\text{Li}^+$ ,  $(1 - \beta)$  mol  $\text{AlF}_6^{3-}$ ,  $\beta$  mol  $\text{AlF}_4^-$  and  $2\beta$  mol  $\text{F}^-$  are formed. The sum of the anionic mol is  $1 + 2\beta$ . Thus the activities of the individual ions are

$$\begin{aligned} a(\text{Li}^+) &= 1 & a(\text{AlF}_4^-) &= \beta/(1 + 2\beta) \\ a(\text{AlF}_6^{3-}) &= (1 - \beta)/(1 + 2\beta) & a(\text{F}^-) &= 2\beta/(1 + 2\beta) \end{aligned}$$

and the dissociation constant of  $\text{AlF}_6^{3-}$  is

$$K^{\text{dis}}(\text{AlF}_6^{3-}) = 4\beta^3/(1 - \beta)(1 + 2\beta)^2 . \quad (1)$$

For the liquidus curve of lithium cryolite the generalized form of the Le Chatelier-Schröder equation holds

$$\ln a(\text{Li}_3\text{AlF}_6) = \frac{\Delta H^f(\text{Li}_3\text{AlF}_6)}{R} \left[ \frac{1}{T_{\text{id}}^f} - \frac{1}{T_{\text{exp}}^f} \right], \quad (2)$$

where  $a(\text{Li}_3\text{AlF}_6)$  is the activity of  $\text{Li}_3\text{AlF}_6$  in the molten lithium cryolite (*i.e.*, in the mixture of  $\text{Li}^+$ ,  $\text{AlF}_6^{3-}$ ,  $\text{AlF}_4^-$  and  $\text{F}^-$  ions);  $\Delta H^f(\text{Li}_3\text{AlF}_6)$  is the enthalpy of melting of one mol of lithium cryolite;  $T_{\text{id}}^f$  is the ideal, hypothetical melting point of the pure  $\text{Li}_3\text{AlF}_6$ , if no dissociation of  $\text{AlF}_6^{3-}$  occurs; and  $T_{\text{exp}}^f$  is the experimentally established melting point of the pure  $\text{Li}_3\text{AlF}_6$ . Since  $a(\text{Li}_3\text{AlF}_6) = a^3(\text{Li}^+) a(\text{AlF}_6^{3-})$  we obtain after some rearrangement

$$\frac{1}{T_{\text{id}}^f} = \frac{1}{T_{\text{exp}}^f} + \frac{R}{\Delta H^f(\text{Li}_3\text{AlF}_6)} \ln \frac{1 - \beta}{1 + 2\beta} . \quad (3)$$

*Dissociation of  $\text{Li}_3\text{AlF}_6$  in the System  $\text{LiF}-\text{Li}_3\text{AlF}_6$* 

We assume that each molecule of  $\text{Li}_3\text{AlF}_6$  yields three cations  $\text{Li}^+$  and that  $\text{AlF}_6^{3-}$  ions dissociate partially according to the scheme (L), the degree of dissociation being changed from  $\beta$  to  $\delta$ . If the mole fraction of  $\text{Li}_3\text{AlF}_6$  in the system with  $\text{LiF}$  is equal to  $x$  then in the liquid mixture there are  $(1 + 2x)$  mol  $\text{Li}^+$ ,  $x(1 - \delta)$  mol  $\text{AlF}_6^{3-}$ ,  $x\delta$  mol  $\text{AlF}_4^-$  and, finally,  $[2x\delta + (1 - x)]$  mol  $\text{F}^-$  in equilibrium.

Therefore

$$K^{\text{dis}}(\text{AlF}_6^{3-}/\text{LiF}) = \frac{(1 + 2x\delta - x)^2 \delta}{(1 - \delta)(1 + 2x\delta)^2}. \quad (4)$$

The liquidus temperature,  $T$ , of  $\text{Li}_3\text{AlF}_6$  is expressed by

$$\frac{1}{T} = \frac{1}{T_{\text{id}}^f} - \frac{R}{\Delta H^f(\text{Li}_3\text{AlF}_6)} \ln \frac{x(1 - \delta)}{1 + 2x\delta}. \quad (5)$$

*Dissociation of  $\text{Li}_3\text{AlF}_6$  in the System  $\text{Li}_3\text{AlF}_6-\text{AlF}_3$* 

We assume that the added  $\text{AlF}_3$  will interact with molten lithium cryolite which consists of  $\text{Li}^+$ ,  $\text{AlF}_6^{3-}$ ,  $\text{AlF}_4^-$  and  $\text{F}^-$  according to the scheme



We assume such an addition of  $\text{AlF}_3$  that a certain excess of free  $\text{F}^-$  ions will remain in the molten mixture of  $\text{Li}_3\text{AlF}_6 + \text{AlF}_3$ . If the mol fraction of  $\text{Li}_3\text{AlF}_6$  is  $x$  in this mixture then in the melt at equilibrium there are present  $3x$  mol  $\text{Li}^+$ ,  $x(1 - \delta)$  mol  $\text{AlF}_6^{3-}$ ,  $[x\delta + (1 - x)]$  mol  $\text{AlF}_4^-$  and  $[2x\delta - (1 - x)]$  mol  $\text{F}^-$ . Thus

$$K^{\text{dis}}(\text{AlF}_6^{3-}/\text{AlF}_3) = \frac{(1 - x + x\delta)(2x\delta + x - 1)^2}{x^3(1 - \delta)(1 + 2\delta)^2}. \quad (6)$$

The equation for the liquidus temperature,  $T$ , of  $\text{Li}_3\text{AlF}_6$  is

$$\frac{1}{T} = \frac{1}{T_{\text{id}}^f} - \frac{R}{\Delta H^f(\text{Li}_3\text{AlF}_6)} \ln \frac{1 - \delta}{1 + 2\delta}. \quad (7)$$

There is no term representing the activity of cations in Eq. (7) because the concentration of free  $\text{Al}^{3+}$  cations in the melt is thought to be zero and hence the activity of  $\text{Li}^+$  equals one.

The procedure for calculating is the following one: a) we choose a value of the degree of dissociation,  $\beta$ , and inserting it into Eq. (1) and (2), we compute

$K^{\text{dia}}(\text{Li}_3\text{AlF}_6)$  and  $T_{\text{id}}^f$ ; b) since for ideal ionic mixture the following equations are valid

$$K^{\text{dia}}(\text{AlF}_6^{3-}) = K^{\text{dia}}(\text{AlF}_6^{3-}/\text{LiF}) = K^{\text{dia}}(\text{AlF}_6^{3-}/\text{AlF}_3), \quad (8)$$

it is possible to calculate the degree of dissociation  $\delta$  from Eqs (4) and (6) for a fixed value of  $x$ ; c) inserting the  $x$  values and the corresponding  $\delta$  values into Eqs (5) and (7) we obtain the theoretical liquidus temperature of lithium cryolite in the mixture in which  $x$  represents the mole fraction of  $\text{Li}_3\text{AlF}_6$ . The agreement between the calculated and experimental data within the limit of experimental errors is the criterion for the right choice of the degree of dissociation,  $\beta$ , of the pure molten  $\text{Li}_3\text{AlF}_6$ .

### RESULTS AND DISCUSSION

The comparison of calculated using the digital computer Minsk 22 and measured temperature of lithium cryolite for some selected values of  $\beta$  is given in Tables I and II. The data concerning the degree of dissociation of  $\text{AlF}_6^{3-}$ ,  $\delta$ , in dependence on composition in the melt are also shown there.

In the system  $\text{LiF}-\text{Li}_3\text{AlF}_6$  the best agreement between the calculated and measured liquidus temperature within the concentration range down to 40 mol.%  $\text{Li}_3\text{AlF}_6$  occurs if  $\beta$  is chosen to be 0.35 (and hence the corresponding dissociation constant equals 0.06. In this case the absolute value of the maximum deviation, i.e.,  $|T_{\text{calc}} - T_{\text{exp}}|$  does not exceed 3.1°C. However, if we consider that the accuracy of establishing the liquidus curve of  $\text{Li}_3\text{AlF}_6$  in the system  $\text{LiF}-\text{Li}_3\text{AlF}_6$  is equal to  $\pm 1.5^\circ\text{C}$ , then the agreement within the limit  $|1.5^\circ\text{C}|$  takes place only down to 50 mol.%  $\text{Li}_3\text{AlF}_6$ .

TABLE I  
Data on the System  $\text{LiF}-\text{Li}_3\text{AlF}_6$  ( $T$ , °C)

Mol fraction $x(\text{Li}_3\text{AlF}_6)$	$T_{\text{exp}}$	Grjotheim's procedure			LeChatelier Schröder $T$	
		$\beta = 0.05$ $T$	$\beta = 0.35$ $T$ $\delta$	$\beta = 0.95$ $T$		
1.0	782	782	782	0.35	782	782
0.9	781.5	779.4	781.5	0.33	781.7	770.7
0.8	780.5	771.0	780.3	0.30	781.3	758.4
0.7	778	758.5	777.8	0.27	780.4	744.8
0.6	773	743.3	773.1	0.24	778.8	729.6
0.5	764	725.6	765.1	0.21	775.9	712.1
0.4	749	704.6	752.1	0.18	770.9	691.6
0.3	721	678.7	731.6	0.15	761.7	666.3

TABLE II  
Data on the System  $\text{Li}_3\text{AlF}_6$ - $\text{AlF}_3$  ( $T$ , °C)

Mol fraction $x(\text{Li}_3\text{AlF}_6)$	$T_{\text{exp}}$	Grjotheim's procedure					$\beta = 0.95$	LeChatelier Schröder $T$	Temkin model $T$
		$\beta = 0.05$	$\beta = 0.10$	$\beta = 0.15$	$\beta = 0.35$	$\delta$			
		$T$	$T$	$T$	$T$	$\delta$	$T$	$T$	$T$
1.0	782	782	782	782	782	0.35	782	782	782
0.9	776.1	772.1	775.1	776.7	779.25	0.36	780.8	770.7	740.9
0.8	762.4	754.1	759.5	763.25	770.75	0.39	776.9	758.4	702.3
0.7	738.6	730.9	737.7	742.7	754.9	0.44	767.9	744.8	664.4
0.65	721.5	717.1	724.1	729.6	743.6	0.48	760.3	737.5	645.3
0.6	710.0	701.3	708.4	714.1	729.3	0.53	749.4	729.6	625.7

In the system  $\text{Li}_3\text{AlF}_6$ - $\text{AlF}_3$  the accuracy of determination of the liquidus temperature of  $\text{Li}_3\text{AlF}_6$  is somewhat less,  $\pm 3^\circ\text{C}$ . Then the best agreement between the calculated and measured curve within the difference  $|3^\circ\text{C}|$  occurs if  $\beta = 0.10$ . In this case both curves agree within the range 100–65 mol.%  $\text{Li}_3\text{AlF}_6$ . This means there is an inconsistency between the most suitable values of  $\beta$  in both systems. We are of the opinion that the results based on the data from the system  $\text{LiF}$ - $\text{Li}_3\text{AlF}_6$  are more reliable than those from the system  $\text{Li}_3\text{AlF}_6$ - $\text{AlF}_3$  because the behaviour of the former is closer to the ideal behaviour than in the case of the latter, as it has been shown by Paučířová and coworkers<sup>24</sup>. We therefore prefer the value  $\beta = 0.35$ . In this case an agreement within the difference  $|3^\circ\text{C}|$  between the calculated and measured liquidus curves of  $\text{Li}_3\text{AlF}_6$  in the system  $\text{Li}_3\text{AlF}_6$ - $\text{AlF}_3$  is restricted to the mixtures with 90 and more mol.%  $\text{Li}_3\text{AlF}_6$ .

Quite recently the degree of dissociation  $\beta$  was computed by Paučířová and coworkers<sup>25</sup> on the basis of density data and the magnitude 0.30 was found which is closed to the reported value 0.35.

The ideal melting point of an absolutely undissociated  $\text{Li}_3\text{AlF}_6$  calculated from Eq. (3) using  $\beta = 0.35$  has been found to be  $897.1^\circ\text{C}$ .

It should be stressed that all simplifying assumption which have been made in the calculation are fulfilled the better the larger is the concentration of lithium cryolite in the mixture. But just in such regions the liquidus temperature is only slightly dependent on the degree of dissociation  $\beta$ , especially in the system  $\text{LiF}$ - $\text{Li}_3\text{AlF}_6$ . If data from the concentration interval (100–90 mol.%)  $\text{Li}_3\text{AlF}_6$  should be taken as a basis for calculating the quantity of  $\beta$ , then the liquidus temperature of  $\text{Li}_3\text{AlF}_6$  has to be established with an accuracy better than  $|0.1^\circ\text{C}|$ .

From data presented in Tables I and II it follows that among different possibilities of calculating the path of liquidus curves of  $\text{Li}_3\text{AlF}_6$  in the system under considera-

tion only Grjotheim's method based on the concept of a partial dissociation of  $\text{AlF}_6^{3-}$  anions yields results which are in general in good agreement with experiments.

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