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SOME ASPECTS OF THE THERMODYNAMICS OF THE EXTRACTION
 OF URANIUM, THORIUM, AND RARE EARTHS FROM MOLTEN
 LiF-BeF₂ INTO LIQUID Li-Bi SOLUTIONS

L. M. Ferris

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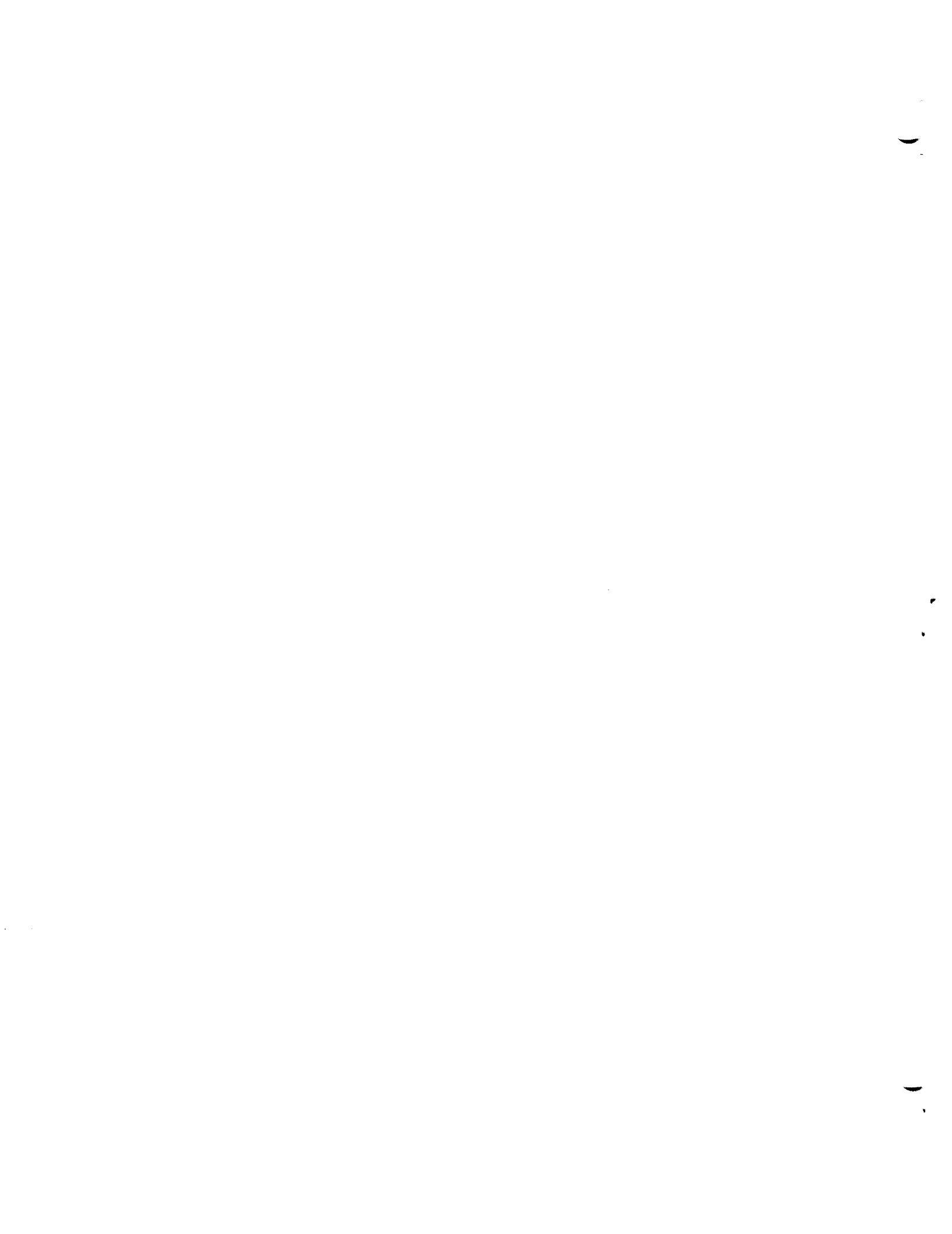
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CONTENTS

	<u>Page</u>
Abstract	1
1. Introduction	1
2. General Thermodynamic Treatment	2
3. The Thermodynamics of Uranium Extraction	5
4. The Thermodynamics of Thorium Extraction	7
5. The Thermodynamics of Lanthanum Extraction	9
6. The Thermodynamics of Sodium Extraction.	10
7. The Thermodynamics of Europium Extraction	10
8. Discussion	11
9. References	12

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ABSTRACT

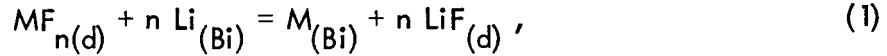
Expressions for the equilibrium distribution of uranium, thorium, lanthanum, and other solutes between LiF-BeF₂ solutions and lithium-bismuth solutions at 600 to 700°C were calculated, using thermodynamic data from the literature. The results obtained experimentally for uranium were in reasonably good agreement with the calculated values. However, the results for thorium and lanthanum reflect the high degree of uncertainty that exists in the available thermodynamic data for these solutes. It is concluded, therefore, that an accurate measure of the relative extractability of the various solutes can be obtained only by experimental means.

1. INTRODUCTION

One method that has been considered for separating uranium and rare-earth fission products in the processing of the fuel carrier salt, LiF-BeF₂ (66-34 mole %), from a two-fluid molten-salt breeder reactor is reductive extraction of the respective elements into liquid bismuth.¹⁻³ During the course of process development, we measured the equilibrium distribution of uranium, thorium, sodium, and certain rare earths between LiF-BeF₂ solutions and Li-Bi solutions at 600 to 700°C to determine the relative ease of extraction of the various elements.³ It was possible to predict the extraction behavior of several of the solutes by using the system of thermodynamics developed by Baes^{4,5} for LiF-BeF₂ systems, and the activity coefficients reported for the various metals in liquid bismuth. In this report, these calculated results are compared with those obtained experimentally with two salts: LiF-BeF₂ (66-34 mole %) and LiF-BeF₂ (56.9-43.1 mole %). Activity coefficients for ThF₄ and LaF₃ in the latter salt at 600°C were also computed from the experimental data.

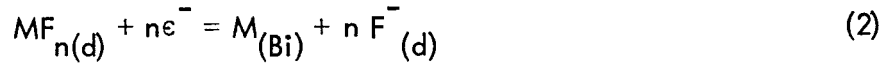
2. GENERAL THERMODYNAMIC TREATMENT

The extraction of a solute MF_n , which is present in low concentration in molten $LiF-BeF_2$, into liquid bismuth containing lithium can be expressed in terms of the general reaction



in which the subscripts (d) and (Bi) denote the salt and bismuth phases, respectively.

This reaction is actually the sum of the two half-reactions



Discontinuing the use of the subscripts, we can write the equilibrium constant for Eq. (1) as

$$K = \frac{a_M a_{LiF}^n}{a_{MF_n} a_{Li}^n} = e^{\frac{nF\Delta E_o}{RT}}, \quad (4)$$

in which a is the activity, F is the Faraday constant, R is the gas constant, T is the absolute temperature, and $\Delta E_o = E_{o,M} - E_{o,Li}$. From Eq. (4), we obtain

$$\frac{nF\Delta E_o}{RT} = \ln \frac{a_M a_{LiF}^n}{a_{MF_n} a_{Li}^n} = n \ln \frac{a_{LiF}}{a_{Li}} + \ln \frac{a_M}{a_{MF_n}} . \quad (5)$$

Let $a = X\gamma$, where X = mole fraction and γ is the activity coefficient; then

$$\Delta E_o = \frac{RT}{F} \ln \frac{X_{LiF}}{X_{Li}} + \frac{RT}{F} \ln \frac{\gamma_{LiF}}{\gamma_{Li}} + \frac{RT}{nF} \ln \frac{X_M}{X_{MF_n}} + \frac{RT}{nF} \ln \frac{\gamma_M}{\gamma_{MF_n}} . \quad (6)$$

If we define the distribution coefficient for component M as

$$D_M = \frac{X_M}{X_{MF_n}} \quad (7)$$

Eq. (6) can be written as

$$\Delta E_o = -\frac{RT}{F} \ln D_{Li} + \frac{RT}{nF} \ln D_M - \frac{RT}{F} \ln \frac{Y_{Li}}{Y_{LiF}} + \frac{RT}{nF} \ln \frac{Y_M}{Y_{MF_n}} \quad (8)$$

Moulton⁶ has defined the quantity E'_o for component M as

$$E'_{o,M} = E_{o,M} - \frac{RT}{nF} \ln \frac{Y_M}{Y_{MF_n}} \quad (9)$$

Rearranging Eq. (8), we get

$$E_{o,M} - \frac{RT}{nF} \ln \frac{Y_M}{Y_{MF_n}} - \left(E_{o,Li} - \frac{RT}{F} \ln \frac{Y_{Li}}{Y_{LiF}} \right) = \frac{RT}{nF} \ln D_M - \frac{RT}{F} \ln D_{Li} \quad (10)$$

If we define $\Delta E'_{o,M} = E'_{o,M} - E'_{o,Li}$, Eq. (10) becomes

$$\Delta E'_{o,M} = \frac{RT}{nF} \ln D_M - \frac{RT}{F} \ln D_{Li} \quad (11)$$

or

$$\Delta E'_{o,M} = E_{o,M} - E_{o,Li} - \frac{RT}{nF} \ln \frac{Y_M}{Y_{MF_n}} + \frac{RT}{F} \ln \frac{Y_{Li}}{Y_{LiF}} \quad (12)$$

The experimental determination of distribution coefficients allows values of $\Delta E'_{o,M}$ to be calculated from Eq. (11). The use of reported activity coefficients for metals in bismuth, and the activity coefficients and standard reduction potentials for the metal fluorides as given by Baes^{4,5} permits an independent calculation of $\Delta E'_{o,M}$, using Eq. (12). In Baes' treatment, LiF-BeF₂ (66-34 mole %) was used as the reference salt, and partial molal free energies of formation in this salt were calculated

for various solutes from the available thermochemical and equilibrium data. Standard reduction potentials were then computed from the free energy data. The activity coefficient for each solute (at low concentration) was defined as unity in this salt; however, the activity coefficients for LiF and BeF₂ were defined as 1.5 and 3, respectively. The changes in the values of these activity coefficients as the LiF/BeF₂ ratio in the salt varies were also estimated by Baes.

The standard states for the bismuth solutions are the pure metals; the activity coefficients, which are actually Henry's law constants, are practically constant when the solute is present in bismuth in low concentrations. The activity coefficient at infinite dilution is the one used throughout this report when referring to the metal phase.

Activity coefficients for solutes can be calculated for an LiF-BeF₂ composition other than 66-34 mole % if distribution coefficient data for the solute in both the new salt and the reference salt are obtained. Since activity coefficients for the metals in bismuth change only slightly with concentration, we get from Eq. (12) for LiF-BeF₂ compositions 1 and 2:

$$\Delta E'_{o,M,1} = E_{o,M} - E_{o,Li} - \frac{RT}{nF} \ln \frac{Y_M}{Y_{MF_n,1}} + \frac{RT}{F} \ln \frac{Y_{Li}}{Y_{LiF,1}} \quad (13)$$

$$\Delta E'_{o,M,2} = E_{o,M} - E_{o,Li} - \frac{RT}{nF} \ln \frac{Y_M}{Y_{MF_n,2}} + \frac{RT}{F} \ln \frac{Y_{Li}}{Y_{LiF,2}} \quad (14)$$

Subtracting Eq. (14) from Eq. (13), we get

$$\Delta(\Delta E'_o) = \Delta E'_{o,M,1} - \Delta E'_{o,M,2} = \frac{RT}{F} \ln \frac{Y_{LiF,2}}{Y_{LiF,1}} - \frac{RT}{nF} \ln \frac{Y_{MF_n,2}}{Y_{MF_n,1}} \quad (15)$$

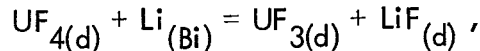
Let composition 1 be LiF-BeF₂ (66-34 mole %), where $Y_{MF_n} = 1$; then, rearrangement of Eq. (15) yields

$$\log \gamma_{MF_{n,2}} = n \log \frac{\gamma_{LiF,2}}{\gamma_{LiF,1}} - \frac{nF \Delta(\Delta E'_o)}{2.303 RT} \quad (16)$$

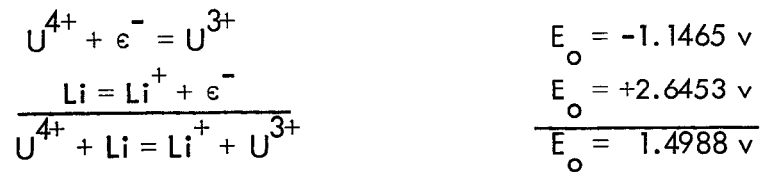
3. THE THERMODYNAMICS OF URANIUM EXTRACTION

Early in the development of the reductive extraction process, uranium was believed to exist primarily as a tetravalent species in the salt.^{3,6,7} However, the results of recent experiments^{8,9} indicate that the uranium is actually, for the most part, in the trivalent state in the salt, especially when the distribution coefficient is greater than about 0.1. These experimental results can be compared with those calculated by using the standard potentials given by Baes^{4,5} and reported values for the activity coefficient for lithium in bismuth.

Consider the reaction



in which the subscripts (d) and (Bi) refer to LiF - BeF₂ (66-34 mole %) and bismuth solvents, respectively. From Baes' treatment of the system we get, at 600°C:



The equilibrium constant

$$K = \frac{a_{Li^+}^X a_{U^{3+}}^Y}{a_{Li}^X a_{U^{4+}}^Y}$$

can be written as

$$K = \left[\frac{X_{U^{3+}}}{X_{U^{4+}}} \right] \cdot \left[\frac{1}{D_{Li}} \right] \cdot \left[\frac{Y_{Li^+}}{Y_{Li}} \right]$$

if we define $D_{Li} = \frac{X_{Li}}{X_{Li^+}}$ and use $\gamma_{UF_4} = \gamma_{UF_3} = 1$.

Rearrangement gives

$$\left[\frac{X_{U^{3+}}}{X_{U^{4+}}} \right] = D_{Li} K \left(\frac{\gamma_{Li}}{\gamma_{Li^+}} \right).$$

From the standard potential of the reaction (given above), we find that $K = 4.51 \times 10^8$. According to Baes' convention, $\gamma_{Li^+} = 1.5$; and from data reported by Argonne National Laboratory,¹⁰ we get $\gamma_{Li} = 5.9 \times 10^{-5}$. Thus,

$$\left[\frac{X_{U^{3+}}}{X_{U^{4+}}} \right] = (4.51 \times 10^8) \left(\frac{5.9 \times 10^{-5}}{1.5} \right) D_{Li}$$

$$\left[\frac{X_{U^{3+}}}{X_{U^{4+}}} \right] = (1.774 \times 10^4) D_{Li}.$$

The average uranium valence in the salt is

$$\bar{n} = \frac{3 \left(\frac{X_{U^{3+}}}{X_{U^{4+}}} \right) + 4}{\left(\frac{X_{U^{3+}}}{X_{U^{4+}}} \right) + 1}.$$

Values of \bar{n} at different values of D_{Li} are shown in the following table.

Li Conc. in Bi (at. %)	Li Conc. in Bi (ppm)	D_{Li}	$\left[\frac{X_{U^{3+}}}{X_{U^{4+}}} \right]$	\bar{n}
0.0037	1.23	5.6×10^{-5}	1.0	3.50
0.004	1.33	6.06×10^{-5}	1.08	3.48
0.01	3.32	1.515×10^{-4}	2.69	3.27
0.02	6.64	3.03×10^{-4}	5.38	3.16
0.1	33.2	1.515×10^{-3}	26.9	3.04

Using $\gamma_{\text{Li}} = 9.8 \times 10^{-5}$ at 600°C, as determined at Oak Ridge National Laboratory,⁶ the calculated $\text{U}^{3+}/\text{U}^{4+}$ values are slightly lower than those shown in the table. However, over the range where we can experimentally determine distribution coefficients for uranium and lithium (lithium concentration in the bismuth of greater than 1 ppm), we would expect the uranium in the salt to be primarily trivalent.

The agreement between distribution coefficients that were obtained experimentally and those that were calculated from the available thermochemical and equilibrium data is easily seen by comparison of the respective $\Delta E'_0$ values. In calculating $\Delta E'_0$ for U^{3+} , we used Baes' values^{4,5} for γ_{LiF} and $E_{\text{o,Li}}$, the activity coefficient for uranium in bismuth as calculated from the expression¹¹

$$\log \gamma_{\text{U}} = 0.7107 - (3995/T),$$

and either ANL or ORNL values for γ_{Li} (the values from the ANL work¹⁰ at various temperatures, and the value at 600°C reported by ORNL⁶). In the following table the calculated values are compared with those determined experimentally:⁹

Temp. (°C)	Calculated $\Delta E'_0$ (volt)		Experimental $\Delta E'_0$ (volt)
	From ORNL γ_{Li}	From ANL γ_{Li}	
600	0.66	0.62	0.66
675	-	0.60	0.66

The agreement is surprisingly good, considering the inherent inaccuracies involved and the variety of sources from which the data were obtained.

4. THE THERMODYNAMICS OF THORIUM EXTRACTION

Distribution coefficients for thorium have been measured⁹ at 600°C, using both LiF-BeF_2 (66-34 mole %), where $\gamma_{\text{ThF}_4} = 1$, and LiF-BeF_2 (56.9-43.1 mole %); these coefficients gave $\Delta E'_0$ values of 0.43 and 0.475 volt, respectively. Baes^{4,5} gives no value for the activity coefficient for ThF_4 in the latter salt; however, it can be readily calculated from the measured $\Delta E'_0$ values, using Eq. (16):

$$\log \gamma_{\text{ThF}_4} = 4 \log (0.866/1.5) - (-0.045/0.0433) = 0.0849$$

$$\gamma_{\text{ThF}_4} = 1.22 .$$

It is interesting to note that the corresponding activity coefficient for UF_4 (the only tetravalent species treated by Baes^{4,5}) is about 1.7.

Values of $\Delta E'_o$ were also calculated for thorium for both salts given above. Data from Brookhaven National Laboratory^{12,13} yielded the following expression:

$$\log \gamma_{\text{Th}} = 1.289 - 7565/T ,$$

from which γ_{Th} is calculated to be about 4.3×10^{-8} at 600°C . The use of this activity coefficient, the value of -1.77 volts for $E_{o,\text{Th}}$, as originally given by Baes,^{4,5} and the ANL and the ORNL values for γ_{Li} resulted in the following values for $\Delta E'_o$ at 600°C :

Salt Composition (mole %)		Calculated $\Delta E'_o$ (volt)		Experimental $\Delta E'_o$ (volt)
		From ANL γ_{Li}	From ORNL γ_{Li}	
LiF	BeF ₂			
66	34	0.432	0.470	0.43
56.9	43.1	0.476	0.514	0.475

The agreement among the calculated and experimental values is quite good. However, Baes now reports¹⁴ $E_{o,\text{Th}}$ to be -1.89 volts at 600°C . The use of this new value, along with the activity coefficients given above, results in calculated $\Delta E'_o$ values that are about 0.12 volt lower than those determined experimentally. If the new value for $E_{o,\text{Th}}$ is really more accurate than the value reported originally, and if we assume that all the other quantities used are reasonably valid, then γ_{Th} at 600°C would have to be in the range of 7×10^{-11} to 5×10^{-10} (instead of the value of 4.3×10^{-8} , which was obtained from the BNL work).

5. THE THERMODYNAMICS OF LANTHANUM EXTRACTION

Distribution coefficients for lanthanum were determined at 600°C with both LiF-BeF₂ (66-34 mole %), where $\gamma_{\text{LaF}_3} = 1$, and LiF-BeF₂ (56.9-43.1 mole %); these coefficients gave $\Delta E'_o$ values of 0.428 and 0.475 volt, respectively.⁹ Since Baes^{4,5} does not give a value for γ_{LaF_3} in the latter salt, it was calculated from the $\Delta E'_o$ values by using Eq. (16):

$$\log \gamma_{\text{LaF}_3} = 3 \log (0.866/1.5) - (-0.047/0.05773) = 0.098$$

$$\gamma_{\text{LaF}_3} = 1.25 .$$

This value is slightly higher than the value that was reported^{4,5} for CeF₃ in the same salt.

Values of $\Delta E'_{o,\text{La}}$ at 600°C were calculated for both salts, using the original $E_{o,\text{La}}$ of -2.314 volts as given by Baes^{4,5} and the activity coefficient for lanthanum in bismuth as calculated from the expression¹⁵

$$\log \gamma_{\text{La}} = 0.844 - (11070/T) ,$$

which gives $\gamma_{\text{La}} = 1.46 \times 10^{-12}$ at 600°C. The calculated $\Delta E'_o$ values were lower than the experimentally determined values by about 0.2 volt. This rather poor agreement is probably the result of the large uncertainty in the value for $E_{o,\text{La}}$. Assuming this to be the case, a new value of $E_{o,\text{La}} = -2.157$ volts at 600°C was calculated by using the measured $\Delta E'_o$ for LiF-BeF₂ (66-34 mole %), the values of γ_{LiF} and $E_{o,\text{Li}}$ as given by Baes,^{4,5} the value for γ_{La} as given by Kober et al.,¹⁵ and an average of the ORNL and ANL values for γ_{Li} . If it is assumed that Baes' temperature coefficient for $E_{o,\text{La}}$ (0.82 mv/°C) is valid, a value of -2.0955 volts is calculated for $E_{o,\text{La}}$ at 675°C. The revised values for $E_{o,\text{La}}$ lead to the following comparison of calculated and experimentally determined $\Delta E'_o$ values:

Temp. (°C)	Salt Composition (mole %)		Calculated $\Delta E'_o$ (volt)		Experimental $\Delta E'_o$ (volt)
	LiF	BeF ₂	From ANL γ_{Li}	From ORNL γ_{Li}	
600	56.9	43.1	0.46	0.49	0.475
675	66	34	0.41	-	0.43

As seen, the use of the revised standard potentials for lanthanum, along with the activity coefficient data cited above, gives much better agreement between the calculated and the experimentally determined quantities than was obtained with the original standard potential.

6. THE THERMODYNAMICS OF SODIUM EXTRACTION

A $\Delta E'_o$ value of 0.2 volt was determined experimentally³ at 600°C for sodium with LiF-BeF₂ (66-34 mole %). Using this value, along with Baes' values^{4,5} for $E_{o,Li}$ and γ_{LiF} , the activity coefficient for sodium in bismuth¹⁶⁻¹⁸ as determined from the expression

$$\log \gamma_{Na} = 0.4892 - (3512/T) ,$$

and the activity coefficient for lithium in bismuth, yields a value of about -2.3 volts for $E_{o,Na}$ at 600°C.

7. THE THERMODYNAMICS OF EUROPIUM EXTRACTION

A value of 0.33 volt has been determined⁹ at 600°C using LiF-BeF₂ (66-34 mole %). No comparison with calculated values can be made because of the lack of activity coefficient and standard potential data.

8. DISCUSSION

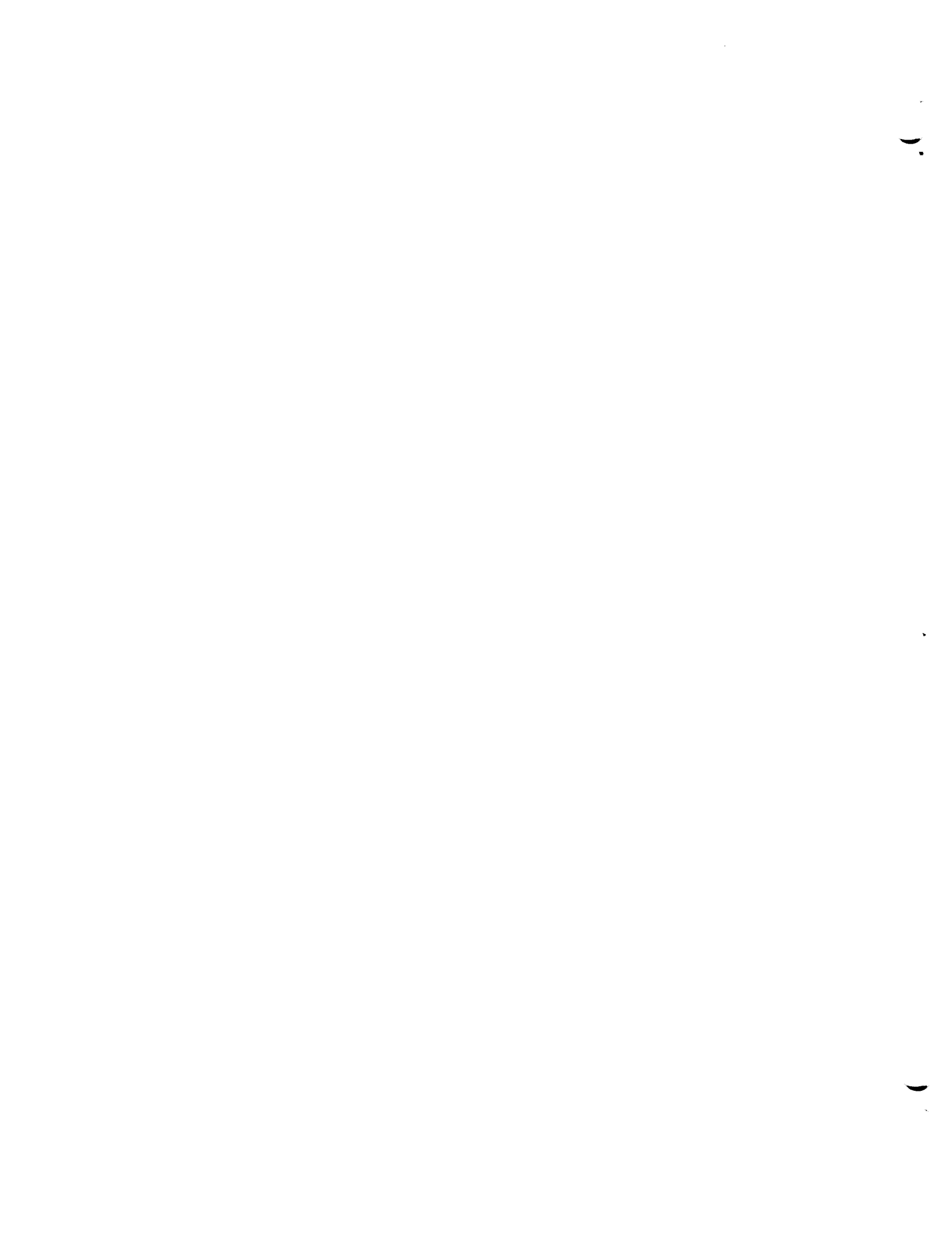
The information presented in this report indicates that uranium in the salt exists primarily as UF_3 during its extraction from $LiF-BeF_2$ solutions with $Li-Bi$ solutions. This is in accordance with the available thermodynamic data. Actually, the calculated distribution coefficients are in reasonable agreement with those that have been determined experimentally. The results of other experiments with $LiF-BeF_2-ThF_4$ solutions⁹ at 600 to 700°C strongly indicate that the potentials for the $U^{4+} \rightarrow U^{3+}$ and $U^{3+} \rightarrow U^0$ half-cell reactions are about the same as those in $LiF-BeF_2$ solutions.

A comparison of calculated and measured $\Delta E'_0$ values for solutes such as ThF_4 and LaF_3 illustrates the high degree of uncertainty that exists in the standard potentials and/or the activity coefficients for these components in bismuth. As a result, the predicted relative extractability of these two elements is greatly influenced by the particular sets of data used in the calculations. In general, the use of the available thermodynamic data gives only a rough indication of the relative extractability of the solutes of interest in the processing of molten-salt breeder reactor fuels, particularly in the case of $LiF-BeF_2-ThF_4$ systems of high ThF_4 concentration. Data for $LiF-BeF_2$ systems are scarce and inaccurate; in addition, no reliable method for extrapolating these data to $LiF-BeF_2-ThF_4$ systems has been devised. It is concluded, therefore, that an accurate measure of the relative extractability of the various solutes can be obtained only by direct experimentation. At present, a program to determine distribution coefficients for uranium, protactinium, zirconium, rare earths, and other fission products in a variety of molten fluoride salts and several liquid metal systems is under way at ORNL. The results will be of great value, not only in defining the process chemistry but also in supplying additional data from which the thermodynamics of these systems can be refined and extended.

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