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MASTER

PHYSICAL PROPERTIES OF MOLTEN-SALT REACTOR FUEL,
COOLANT, AND FLUSH SALTS

Edited by S. Cantor

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CONTENTS

ABSTRACT

For seven molten salt mixtures:

four fuel mixtures, each containing LiF, BeF_2, ThF_4, UF_4
one flush salt, $LiF-BeF_2$ (66-34 mole %)
two coolant salts, $NaBF_4-NaF$ (92-8 mole %) and single-
    component $NaBF_4$

estimates and/or experimental values are given for the follow-
ing properties:

> viscosity,
> thermal conductivity,
> electrical conductivity,
> phase transition behavior,
> heat capacity,
> heat of fusion,
> density,
> expansivity,
> compressibility,
> vapor pressure,
> surface tension,
> solubility of the gases, He,Kr,Xe,BF_3 .

From the foregoing properties, the following have also been
calculated and appended:

> isochoric heat capacity (C_v)
> sonic velocity
> thermal diffusivity
> kinematic viscosity
> Prandtl number.

Composition of Salt Mixtures

	Symbol	Mole %				Liquidus Temp. (^{O}C)
		LiF	BeF$_2$	ThF$_4$	UF$_4$	
	F$_1$	73	16	10.7	0.3	500° ± 5°
Fuel-Breeder Mixtures	F$_2$	72	21	6.7	0.3	500° ± 5°
	F$_3$	68	20	11.7	0.3	480° ± 5°
	F$_4$	63	25	11.7	0.3	500° ± 5°
Flush Salt (present MSRE coolant)	L$_2$B	66	34	--	--	458° ± 1^O (peritectic)
		NaBF$_4$	NaF			
Coolants	C$_1$	92	8			385° ± 1^O (eutectic)
	C$_2$	100	-			407° ± 1^O (melting point)

INTRODUCTION

In this document we have compiled physical property information, either measured or estimated, on seven salt mixtures that are presently of importance in the design of advanced molten salt reactors. The primary user of this compilation will, no doubt, be the nuclear reactor engineer who requires these data for the design and development of molten salt reactors. Specialists in the chemistry of molten salts may be another audience interested in this report. We earnestly hope that all who critically examine or otherwise use these data will give us the benefit of their advice so that future versions of this document can be greatly improved.

Basis for Selecting the Salts

The choice of salt mixtures has been primarily governed by recent changes in the Molten Salt Reactor Program: (a) the combining of fissile and fertile material within the same circuit (the "single-region" concept), and (b) the testing of coolant salts which are mainly $NaBF_4$.

Four mixtures have been selected for possible use as single-region fuel melts. These are:

Salt Mixture	Composition (mole %)			
	LiF	BeF_2	ThF_4	UF_4
F_1	73	16	10.7	0.3
F_2	72	21	6.7	0.3
F_3	68	20	11.7	0.3
F_4	63	25	11.7	0.3

Salts F_1 and F_3 are fuel mixtures appropriate to a prismatic configuration of the graphite moderator; the lesser concentrations of BeF_2 and ThF_4 in F_1 may be more favorable with respect to rare-earth fission product removal by reductive extraction.

Salt F_2, containing a relatively low concentration of thorium, might be used in a reactor (e.g., with random-packed graphite spheres) where good breeding performance is not a prime consideration. Mixture F_4, on the other hand, could contribute to improved breeder performance mainly because the higher the beryllium concentration, the greater the opportunity to increase neutrons by the (n, 2n) reaction.

It is worthwhile noting that for the purposes of estimating physical properties of salts F_1-F_4, the effects of the small concentration of UF_4 was almost always assumed to be the same as for the corresponding increase in the ThF_4 concentration.

Although no firm decision has been reached as to the exact composition of the fuel salt for the next molten salt reactor, it is highly probable that the concentrations of LiF, BeF_2 and ThF_4 will be within the limits given for these components by the above four mixtures.

Physical property information is also provided for:

LiF-BeF_2 (66-34 mole %) symbolized as L_2B.
This mixture has been used in the MSRE as the coolant and as the flush salt for the fuel circuit. The inclusion of L_2B

in this report is justified by the good possibility that it will be a flush salt (and perhaps a coolant) in future molten salt reactors.

As intermediate coolant (in this case the fluid which transports heat from the fuel salt to the steam generators) the salts which presently appear attractive contain mostly $NaBF_4$. Two such salts are considered:

	Composition (mole %)	
Coolant	$NaBF_4$	NaF
C_1	92	8
C_2	100	

The salt symbolized as C_1 is a eutectic composition which melts at $385°C$ ($725°F$). Although a lower melting fluoroborate mixture would be desirable, it is not presently clear how much and which additive will substantially depress the melting temperature. Moreover, it seems likely that lower melting fluoroborate mixtures will not be very different from C_1; hence mixture C_1 seems, at present, the leading candidate for the next coolant to be tried in a molten salt reactor.

Another salt for which estimates are tabulated in this report is "pure" $NaBF_4$, symbolized as C_2. Since stoichiometric $NaBF_4$ does not exist in the molten state without a very high partial pressure of BF_3 gas, C_2 cannot be considered a practical coolant. However, estimations of the physical properties of hypothetically pure molten $NaBF_4$ are useful for evaluating the contributions of $NaBF_4$ as a component in

a salt mixture. In solution, $[BF_4]^-$ ion may be imagined to behave like a halide ion, slightly larger and more polarizable than iodide ion. By applying this analogy, several properties of C_2 were estimated from the measured properties of molten NaI.

For convenience, a list of salt compositions and their corresponding liquidus temperatures are given after the abstract (page 2) and at the end of this report (page 46).

Uncertainties Listed with the Physical Property Values

Each contributor has stated what he believes is the error associated with the experimental result or with the estimated quantity. For most cases, the uncertainty represents considerably more than either "goodness of fit" of an interpolation or internal consistency available from thermodynamics. Instead, the uncertainty may be considered as the largest probable combination of systematic and random errors associated with the value given for the property. Where the listing is a property-temperature equation, the uncertainty is for the property calculated at the temperature substituted in the equation. In properties where the number of significant figures are not justified by the specified uncertainties, the extra significant figures are given to aid the reader in judging whether a particular salt is "less than" or "greater than" another salt for the property in question.

Although the magnitudes of the uncertainties are highly intuitive and often disappointingly large, they should be

taken seriously. Each contributor, while not necessarily qualifying as "expert" in the physical property, either possesses long experience in measuring the property or has carefully (and usually critically) reviewed the literature for that property. In other words, for each property the person whose name is given is at least a very interested observer and may also be an active participant.

For Further Information ---

It is best to contact the person (or persons) listed under the property heading. The editor hopes to provide addenda to this report as newer, more reliable, data become available.

VISCOSITY

S. Cantor

Viscosity-Temperature Equation

Salt	η in Centipoise, T in ^{0}K	Uncertainty
F_1	$\eta = 0.084 \exp (4340/T)$	25%
F_2	$\eta = 0.072 \exp (4370/T)$	25%
F_3	$\eta = 0.077 \exp (4430/T)$	25%
F_4	$\eta = 0.0444 \exp (5030/T)$	25%
$L_2 B$	$\eta = 0.116 \exp (3755/T)$	15%
C_1 C_2	$\eta = 0.04 \exp (3000/T)$	50%

Sources of Data and Methods of Estimation

Salts F_1-F_4: Estimated empirically from viscosities in the
system LiF-BeF_2-UF_4 (ref. 1) and also from measurements of
LiF-BeF_2-ThF_4 (71-16-13 mole %).[2] It was assumed that the
effect of ThF_4 concentration on viscosity was the same as
that observed for UF_4.

$L_2 B$: Measured[3]

C_1 and C_2: The equation was derived from (a) preliminary
measurements of $NaBF_4$,[4] and (b) assuming that the temperature
variation of viscosity for $NaBF_4$ is equal to that of NaI.[5]
Given the rather large uncertainty, the contribution of NaF
(in C_1) to the viscosity may be considered negligible.

. Discussion

Viscosities of Reactor Fuel Mixtures

From the reported viscosity measurements[1] of the system

LiF-BeF$_2$-UF$_4$, two trends can be observed:

(a) for LiF concentrations of 60 mole % or greater, substitution of UF$_4$ for BeF$_2$ (at const. temp.) causes an increase in viscosity,

(b) increasing LiF from 60 to 70 mole %, at const. temp. and at const. UF$_4$ concentration, decreases the viscosity by, at most, a factor of 1/2; for most compositions the factor is closer to 3/4.

The data and trends observed for the system LiF-BeF$_2$-UF$_4$ can serve to predict reliably (i.e., to within 25%) the viscosities in the slightly different system, LiF-BeF$_2$-MF$_4$ (M is Th and/or U). Assuming that all single-region fuel mixtures will be restricted to the following ranges of component composition:

$$62 - 73 \text{ mole \% LiF}$$

$$15 - 30 \text{ mole \% BeF}_2$$

$$6 - 16 \text{ mole \% MF}_4 ,$$

then one may conclude that the predicted viscosities have a rather narrow range of values, e.g.,

at 600°C,   9 - 16 Centipoise

at 700°C,   5 - 9 Centipoise

References

1. B. C. Blanke et al., "Density and Viscosity of Fused Mixtures of Lithium, Beryllium, and Uranium Fluorides," MLM-1086, Dec. 1956.

2. Molten Salt Reactor Program Quar. Progr. Rept. Oct. 31, 1959, ORNL-2890, p. 21.

3. S. Cantor and W. T. Ward, Oak Ridge National Laboratory, unpublished measurements.

4. L. J. Wittenberg, Mound Laboratory, Miamisburg, Ohio. Oscillating-cup viscometry.

5. G. J. Janz et al., "Molten Salt Data. Electrical Conductance, Density and Viscosity," Technical Bulletin Series Rensselaer Polytechnic Institute, Troy, N. Y., July 1964, p. 79.

THERMAL CONDUCTIVITY

J. W. Cooke

Salt	Thermal Conductivity[a] in watt/(cm-$^{\circ}$C)	Uncertainty
F_1	0.01_0[b]	$\geq \pm 25\%$
F_2	0.01_1[b]	$\geq \pm 25\%$
F_3	0.008_3[b]	$\geq \pm 25\%$
F_4	0.007_0[b]	$\geq \pm 25\%$
L_2B	0.010	$\pm 10\%$
C_1	0.005_2	$\pm 50\%$
C_2	0.005_1	$\pm 50\%$

[a] As a first approximation, the temperature dependence of thermal conductivity may be neglected. Although the thermal conductivity of molten salts does vary somewhat with temperature, uncertainties in measurements at a given temperature are usually greater than the temperature dependence over the whole range of temperature (usually an interval of 200°C).

[b] Before assuming anything about the relative values of the four fuel melts, please read the caveat in the Discussion.

Sources of Data and Methods of Estimation

Salts F_1 - F_4: Estimated by means of a theoretical expression derived by Rao[1] and adapted to molten salts by Turnbull.[2] The expression is

$$k \ (\text{in w cm}^{-1} \ ^{\circ}\text{C}^{-1}) = 11.9 \times 10^{-3} \ \frac{T_m^{1/2} \ \rho_m^{2/3}}{(M/n)^{7/6}}$$

where T_m = melting point ($^{\circ}$K), ρ_m = liquid density in g cm^{-3} at T_m, M = average molar weight and n = average number of discrete

ions per molecule. Part of the expression,

$$11.9 \times 10^{-3} \; T_m^{1/2} \; \rho^{1/3} / (M/n)^{5/6} \quad ,$$

is a good approximation to the average maximum Debye lattice frequency for single ionic salts.[2] It was found for eleven molten mixtures (nitrates or chlorides) that the above expression agreed with experimental results, on the average, to within 15%. For two fluoride melts, one L_2B,[3] the other, $LiF-BeF_2-ThF_4-UF_4$ (71.2-23-5-.8 mole %),[3] the theoretical expression yielded values approximately 25% less than experimental. Note that the latter is very similar in composition to F_2.

In applying the theoretical expression the liquidus temperature was substituted for T_m; in computing n, the following ions were assumed: Li^+, F^-, $(BeF_4)^{2-}$, $(ThF_5)^{-1}$, $(UF_5)^{-1}$. Assumption of the more plausible ions, $(ThF_7)^{-3}$ and $(UF_7)^{-3}$ leads to a lower and less reliable estimated thermal conductivity. Also, 15% was added to the estimated value because of the previously noted discrepancy for the cases of the two similar fluoride mixtures.

L_2B: Measured[3]

C_1, C_2: Very preliminary measurement[3] on C_2 agrees with the theoretical expression.

Discussion

The relative conductivities of the four fuel mixtures, F_1-F_4, are <u>not</u> more reliable than the absolute values. The tabulated conductivities were obtained from a theoretical

equation that was greatly extended to apply to these mixtures. The dearth of accurate experimental data prevents adequate testing of the extended theoretical expression either absolutely or relatively.

References

1. M. Rama Rao, Indian Journal of Physics 16, 30 (1942).

2. A. G. Turnbull, Australian Journal of Applied Science 12, 324 (1961).

3. J. W. Cooke, Oak Ridge National Laboratory, unpublished experimental results. The method of measurement is given on p. 15 in Proceedings of the Sixth Conference on Thermal Conductivity, Dayton, Ohio, Oct. 19-21, 1966.

ELECTRICAL CONDUCTIVITY

G. D. Robbins

Salt	Specific Conductivity - Temperature Equation	Uncertainty
	κ in (ohm-cm)$^{-1}$, t in $^{\circ}$C	
F_1	κ = 1.72 + 8.0 x 10^{-3} (t-500)	± 20%
F_2	= 1.63 + 7.3 x 10^{-3} (t-500)	± 20%
F_3	= 1.66 + 6.4 x 10^{-3} (t-500)	± 20%
F_4	= 1.94 + 7.1 x 10^{-3} (t-500)	± 20%
$L_2 B$	= 1.54 + 6.0 x 10^{-3} (t-500)	± 10%
C_1	= 2.7 + 13 x 10^{-3} (t-500)	± 50%
C_2	= 1.92 + 2.6 x 10^{-3} (t-500)	± 20%

Sources of Data and Method of Estimation

For 6 salts κ was estimated empirically from data on related or analogous salt melts. Often the assumptions employed were not those which seemed physically most reasonable, but those which resulted in the most self-consistent correlation of the data. Therefore, estimated κ's are believed to have relatively large uncertainties. The number of significant figures in the equations for κ vs. t are not meant to contradict the listed uncertainties, but rather are intended to show differences between salt mixtures whose conductivities are predicted to be very similar.

Salts F_1 - F_4: The following equations were employed in these estimates:

$$\Lambda_{\Theta} = \kappa_{\Theta} \cdot \frac{M_e}{\rho_{\Theta}}$$

$$\Theta = \frac{T_{\Theta} \; (^{\circ}K)}{T_{liquidus} \; (^{\circ}K)}$$

$$M_e = X_{LiF}M_{LiF} + \tfrac{1}{4}X_{ThF_4}M_{ThF_4} + \tfrac{1}{2}X_{BeF_2}M_{BeF_2}$$

Λ_Θ = equivalent conductivity at a corresponding temperature Θ

κ_Θ = specific conductivity at Θ

ρ_Θ = density at Θ

M_e = equivalent weight of a mixture

M = formula weight of a component

X = mole fraction

X' = equivalent fraction

At several values of Θ smoothed curves of Λ_Θ <u>vs</u> X'_{ThF_4} were obtained from conductivities of the system LiF-ThF_4 measured by Brown and Porter.[1] Liquidus temperatures reported in references 2 and 3 were used in calculating Θ. Similar curves for LiF-BeF_2 were derived by plotting the experimental results for a single composition (66 mole % LiF)[4] and assuming that the variation of Λ_Θ with X' in the LiF-BeF_2 system was equal to that in LiF-ThF_4. (For these estimates UF_4 was treated as indistinguishable from ThF_4.) The equations of κ <u>vs.</u> t given above were then derived by assuming that Λ_Θ is additive in X'_{ThF_4} and X'_{BeF_2} for a given concentration of LiF.

$\underline{L_2B}$: Preliminary measurements.[4]

$\underline{C_2}$: The ratio $\Lambda_{\Theta NaI}/\Lambda_{\Theta KI}$ appeared relatively constant in the range Θ = 1.05 - 1.20 (data for NaI and KI from ref. 5). Assuming that $\Lambda_{\Theta NaBF_4}/\Lambda_{\Theta KBF_4} = \Lambda_{\Theta NaI}/\Lambda_{\Theta KI}$, specific conductance data of Winterhager and Werner[6] for KBF_4 were combined with density estimates for KBF_4 and $NaBF_4$[7] to obtain values of $\Lambda_{\Theta NaBF_4}$ <u>vs.</u> Θ (liquidus temperatures, from reference 8).

$\underline{C_1}$: Specific conductivity data in the range 47 to 77 mole % $NaBF_4$ in the $NaF-NaBF_4$ system[9] were combined with those calculated for pure $NaBF_4$ (see C_2) to interpolate κ for the composition $NaBF_4-NaF$ (92-8 mole %). The large uncertainty listed reflects a lack of confidence in the data reported in reference 9.

Discussion

Specific conductivity is determined from resistance measurements according to the relation

$$\kappa = \frac{1}{R_\infty} (\ell/a)$$

where (ℓ/a) is the cell constant. For a given apparatus and set of experimental conditions, the measured value of resistance can vary with the frequency of the applied potential wave form.[10] The values of κ listed above are valid for resistance extrapolated to infinite frequency (denoted as R_∞). Thus predicting the resistance of the melt which will be measured in a particular experimental arrangement not only requires a value for conductivity κ, but also presupposes a knowledge of the frequency dispersion characteristics of the measuring device.

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1. Brown, E.A. and B. Porter, U.S. Bureau of Mines Report of Investigations 6500 (1964).

2. Thoma, R.E., H. Insley, B. S. Landau, H. A. Friedman, and W. R. Grimes, J. Phys. Chem. 63, 1266 (1959).

3. Thoma, R. E., et al, ibid., 64, 865 (1960).

4. Robbins, G. D. and J. Braunstein, Molten Salt Reactor Program Semiannual Progress Report for Period Ending February 29, 1968, ORNL-4254.

5. Yaffe, I. S. and E. R. Van Artsdalen, J. Phys. Chem., 60, 1125 (1956).

6. Winterhager, H. and L. Werner, "Forschungsber. des Witschafts u. Verkehrsministeriums Nordrhein - Westfalen, No. 438, 1956.

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PHASE TRANSITION BEHAVIOR

R. E. Thoma

Salt	Type of Transition	Temp. ($^\circ$C)	Crystallization Sequence at Equilibrium
F_1	Liquidus	500±5	Liq $\rightleftharpoons$ LiF + L_3T[a] + Liq Btwn 500-444: LiF+L_3T+Liq
	Solidus	444±5	LiF+L_3T+Liq $\rightleftharpoons$ LiF+L_3T+Li_2BeF_4
F_2	Liquidus	500±5	Liq $\rightleftharpoons$ LiF + Liq Btwn 500-495: LiF + Liq Btwn 495-444: LiF+L_3T+Liq
	Solidus	444±5	Same as for F_1
F_3	Liquidus	480±5	Liq $\rightleftharpoons$ L_3T + LT[b] + Liq Btwn 480-448: L_3T+LT+Liq Btwn 448-440: L_3T + Liq
	Solidus	440[c]	L_3T + Liq $\rightleftharpoons$ L_3T + L_2B
F_4	Liquidus	500±5	Liq $\rightleftharpoons$ LT_2[d] + Liq Btwn 500-495: LT_2 + Liq Btwn 495-490: LT_2+LT+Liq 490: LT_2+LT+Liq $\rightleftharpoons$ L_3T+Liq Btwn 490-448: L_3T + Liq
	Solidus	448±5	Liq + L_3T $\rightleftharpoons$ Li_2BeF_4 + L_3T
L_2B	Peritectic	458±1	Liq $\rightleftharpoons$ Li_2BeF_4 + Liq Btwn 458-360: Li_2BeF_4+Liq
	Solidus	360±3	Li_2BeF_4+Liq $\rightleftharpoons$ Li_2BeF_4+BeF_2
C_1	Eutectic	385±1	Liq $\rightleftharpoons$ $NaBF_4$(cubic) + NaF
	Solid-Solid	245±1	$NaBF_4$(cubic)+NaF $\rightleftharpoons$ $NaBF_4$(orthorhombic) + NaF
C_2	Melting Point	407±1	Liq $\rightleftharpoons$ $NaBF_4$(cubic)
	Solid-Solid	245±1	$NaBF_4$(cubic) $\rightleftharpoons$ $NaBF_4$(orthorhombic)

a. L_3T is an abbreviation for the solid solution, $Li_3(Th,Be)F_7$, shown as the peppered triangle in the accompanying phase diagram of LiF-BeF_2-ThF_4 system.

b. LT is the abbreviation for $LiThF_5$.

c. No precision has been assigned because this temperature has not been experimentally established.

d. LT_2 is the abbreviation for $LiTh_2F_9$.

Sources of Data

Phase equilibria in the system, $LiF-BeF_2-ThF_4$ - see next page.

Phase equilibria in the system, $LiF-BeF_2$ - R. E. Thoma, H. Insley, H. A. Friedman, and G. M. Hebert, Journal of Nuclear Materials <u>27</u>, in press 1968.

Phase equilibria in the system, $NaBF_4-NaF$ - C. J. Barton, L. O. Gilpatrick, <u>et al.</u>, MSRP Semiann. Progr. Rept. Feb. 29, 1968, USAEC Report ORNL-4254. The phase diagram is given on page 21.

The LiF–BeF₂–ThF₄ System.

REF: J. PHYS. CHEM., 64, 865 (1960)
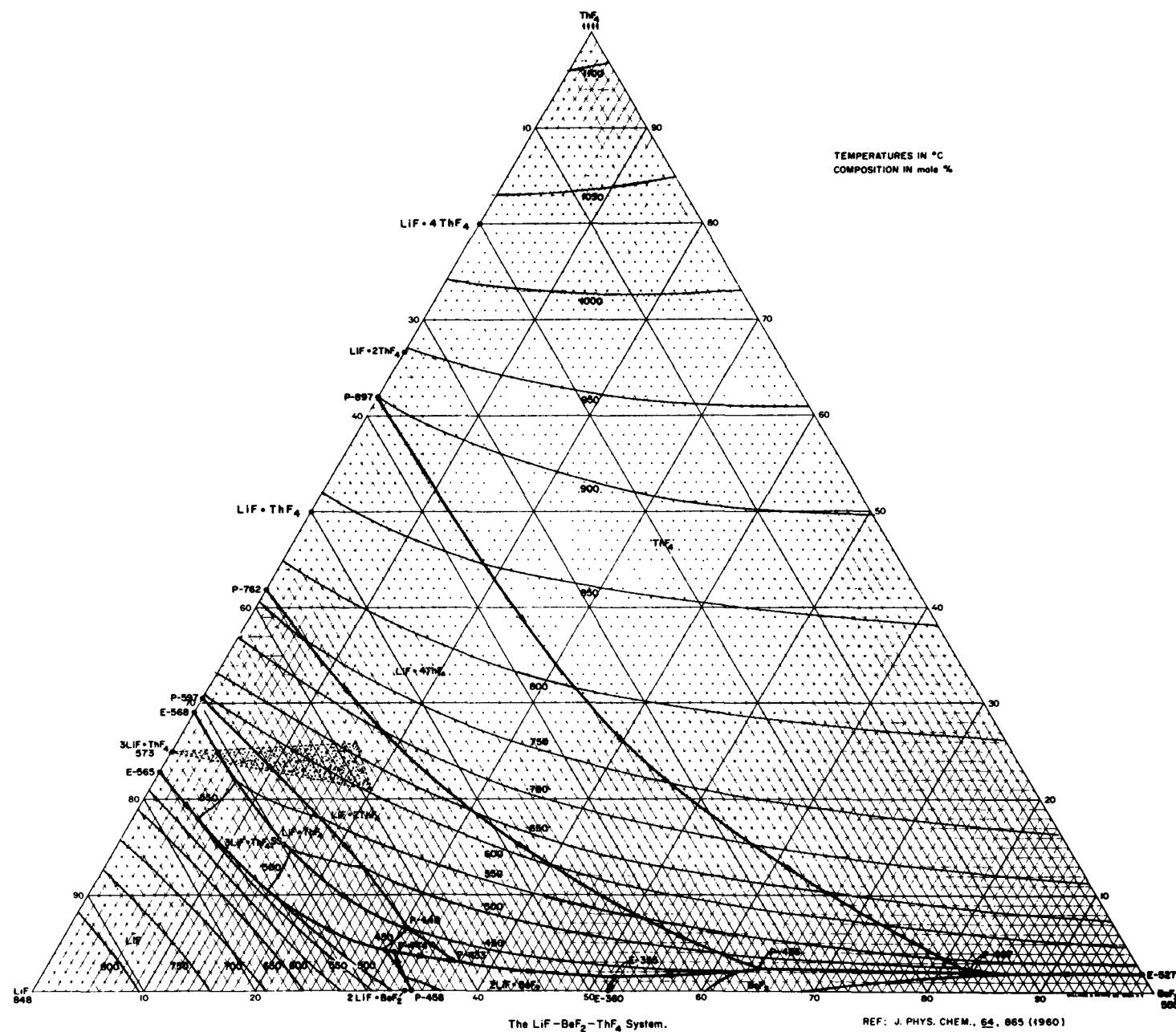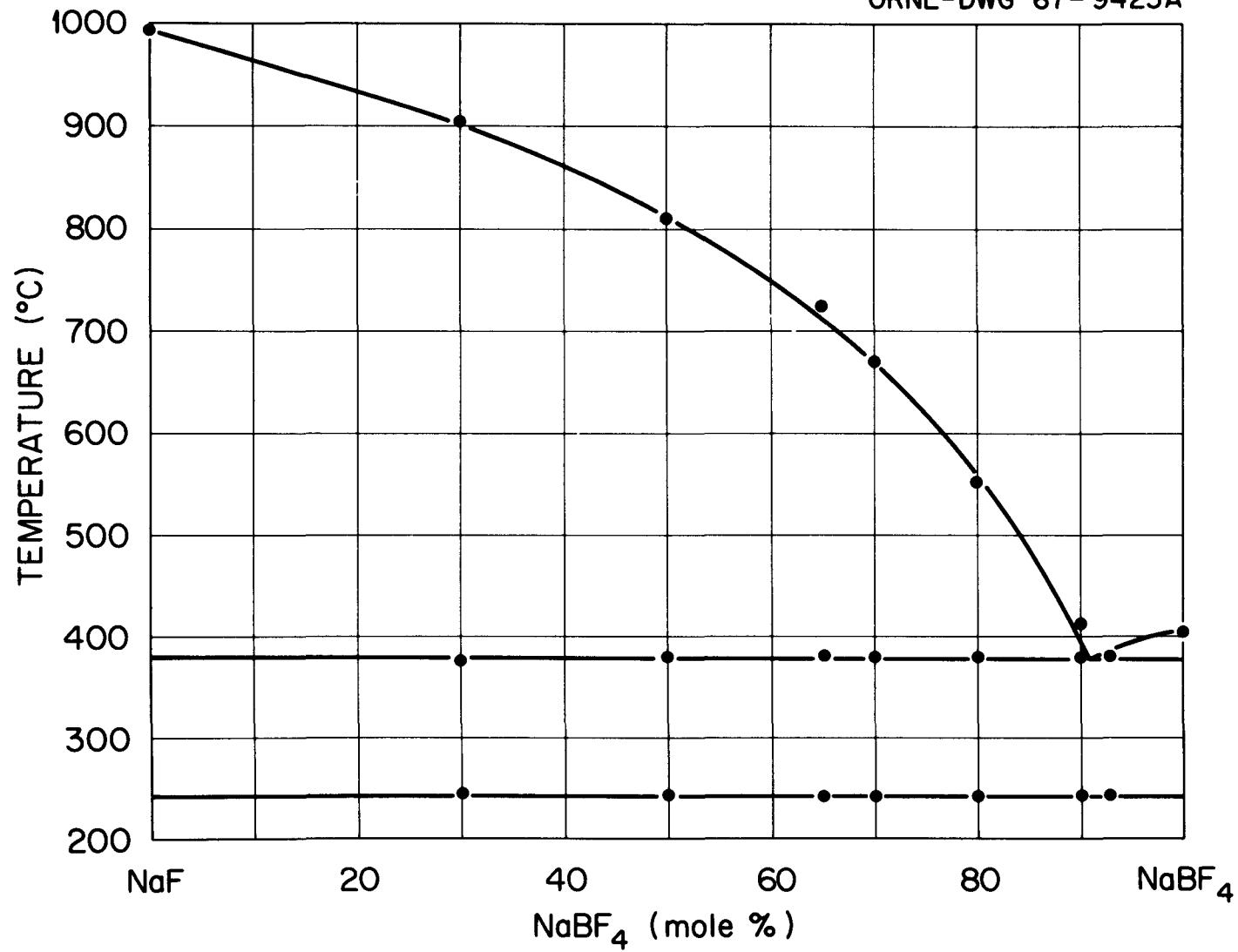
The System NaF – NaBF₄.

HEAT CAPACITY (at constant pressure)

A. S. Dworkin

Salt	C_p in cal. g^{-1} $^{O}C^{-1}$; t in ^{O}C	Uncertainty
F_1 liquid	0.34	± 4%
solid	$0.22 + 12.7 \times 10^{-5}$ t	± 10
F_2 liquid	0.39	± 4
solid	$0.27 + 12.7 \times 10^{-5}$ t	± 10
F_3 liquid	0.33	± 4
solid	$0.21 + 12.7 \times 10^{-5}$ t	± 10
F_4 liquid	0.33	± 4
solid	$0.21 + 12.7 \times 10^{-5}$ t	± 10
L_2B liquid	0.57	± 3
solid	$0.317 + 3.61 \times 10^{-4}$ t	± 3
C_1 liquid	0.360	± 2
solid (243-381°C)	0.34	± 3
solid (25-243°C)	$0.23 + 5.8 \times 10^{-4}$ t	± 6
C_2 liquid	0.36	± 2
solid (243-406°C)	0.33	± 3
solid (25-243°C)	$0.23 + 6.0 \times 10^{-4}$ t	± 6

Sources of Data and Methods of Estimation

Salts F_1 - F_4: Liquid heat capacities were estimated by assuming mole-fraction additivity and assigning 16, 24, and 44 cal mole^{-1} $^{O}C^{-1}$ for the respective contributions of LiF, BeF_2, and ThF_4. The heat capacities for the solids were estimated by assuming that (a) temperature coefficient and (b) difference in C_p between liquid and solid are the same as that measured for LiF-BeF_2-ThF_4 (72-16-12 mole %).[1]

L_2B: Liquid C_p is the average of two independent sets of measurements. Hoffman[2] obtained 0.577 cal. g^{-1} $^{O}C^{-1}$; Douglas and Payne[3] obtained 0.56 cal g^{-1} $^{O}C^{-1}$. The solid heat capacity

is that of Douglas and Payne.

$\underline{C_1}$: Measured[1]

$\underline{C_2}$: Measured[1] Agrees within experimental error with that derived from C_1 by subtracting enthalpy contribution of NaF[4] assuming negligible heat of mixing between $NaBF_4$ and NaF.

Discussion

The values of 16 and 24 cal mole^{-1} $^{o}C^{-1}$ were chosen for the respective C_p contributions of LiF and BeF_2 because 8 cal (g-atom)$^{-1}$ $^{o}C^{-1}$ is the average observed for alkali and alkaline earth halides.[5] The C_p of 44 cal mole^{-1} $^{o}C^{-1}$ for the contribution of ThF_4 was assumed from the average value of 8.8 cal (g-atom)$^{-1}$ $^{o}C^{-1}$ for lanthanide halides.[6]

The validity of using the indicated additive contributions for estimating liquid heat capacities was checked by comparing with measured values of three related salts:

Salt Mixture	Estimated C_p	Measured C_p	References
L_2B	0.57 cal g^{-1} $^{o}C^{-1}$	0.57	2,3
$LiF-BeF_2-ThF_4$ 72 - 16 - 12 m %	0.32_6	0.324	1
$LiF-ThF_4$ 75 - 25 m %	0.24	0.25	7

References

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HEAT OF FUSION

A. S. Dworkin

Salt	ΔH_{fusion} (cal g^{-1})	Uncertainty
F_1	62	± 10%
F_2	67	± 15
F_3	58	± 15
F_4	63	± 15
L_2B	107	± 3
C_1	31	± 2
C_2	29	± 2

	ΔH of solid transition (cal g^{-1})	
C_1	14.5 (at 243°C)	± 2%
C_2	14.7 (at 243°C)	± 2

Sources of Data and Methods of Estimation

Salts F_1 - F_4: Although there is no isothermal heat of fusion, estimations were made as if all the melting (or freezing) occurred at 500°C. The salts were treated as additive mixtures of the components, Li_2BeF_4, Li_3ThF_7, and LiF or ThF_4. Li_2BeF_4 was considered to be "formed" first from the BeF_2 present and the appropriate quantity of LiF. The remainder of the mixture was then considered to consist of Li_3ThF_7 and either LiF or ThF_4, whichever was "in excess." For example, for 1 mole of salt F_1, .16 moles of BeF_2 and .32 moles of LiF form .16 moles of Li_2BeF_4 while .11 moles of ThF_4 and the remaining .41 moles of LiF give .11 moles of Li_3ThF_7 and .08 moles of LiF. The estimation is then made on the basis of .16 moles Li_2BeF_4, .11 moles ThF_4 and .08 moles LiF.

The following heats of fusion were used in making the estimations:

Li_2BeF_4	10,600 cal mole^{-1}	(ref. 1)
Li_3ThF_7	13,960 cal mole^{-1}	(ref. 2)
LiF	6,470 cal mole^{-1}	(ref. 3)
ThF_4	11,000 cal mole^{-1}	estimated by
	assuming the entropy of fusion	
	is the same as that of UF_4	(ref. 4)

$\underline{L_2B}$: Measured.[1]

$\underline{C_1 \text{ and } C_2}$: Measured.[5] C_2 agrees within experimental error with that calculated by subtracting the contribution of the heat of fusion of NaF[6] from C_1.

Discussion

Although the assumptions used in estimating ΔH_{fusion} for salts F_1 - F_4 are highly intuitive, it is encouraging to note that the estimated and $\underline{\text{measured}}$[7] ΔH_{fusion} are respectively 57.5 and 59 cal g^{-1} for the salt mixture $LiF-BeF_2-ThF_4$ (72-16-12 mole %).

For salts F_1 - F_4, to obtain the heat necessary to convert the solid at the solidus temperature to the melt at the liquidus temperature, an additional 10 to 15 cal g^{-1} should be added to the above listed heats of fusion. For convenience in calculating the quantity of heat necessary to raise the salt from room temperature to any desired temperature, the following heat content equations (based on measurements) are included:

LiF-BeF$_2$-ThF$_4$ (72-16-12 mole %) - ref. 5

Solid: H_t-H_{25} (cal g^{-1}) = -5.28 + .207t + 6.33 x 10^{-5}t^2;

(25 - 440°C)

Liquid: H_t-H_{25} (cal g^{-1}) = 11.34 + .324t (500 - 750°C)

LiF-BeF$_2$ (66-34 mole %)

Solid: H_t-$H_{0^{\circ}C}$ (cal g^{-1}) = 0.3179t - 1.806 x 10^{-4}t^2;

(0 - 472°C) - ref. 1

Liquid: H_t-$H_{0^{\circ}C}$ (cal g^{-1}) = 32.632 + 0.561t; (472 - 600°C) -

ref. 1

H_t-H_{30} (cal g^{-1}) = 33.62 + 0.577 (t-30); ref. 7

NaBF$_4$-NaF (92-8 mole %) - ref. 5

Solid: H_t-H_{25} (cal g^{-1}) = -5.90 + .230t + 2.90 x 10^{-4}t^2;

(25 - 243°C)

H_t-H_{25} (cal g^{-1}) = 0.40 + .337t; (243 - 381°C)

Liquid: H_t-H_{25} (cal g^{-1}) = 22.1 + .360t; (381 - 600°C)

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DENSITY OF LIQUID

(S. Cantor)

Density-Temperature Equation

Salt	ρ (in g/cm^3) t (in $^\circ$C)	Uncertainty
F_1	$\rho = 3.628 - 6.6 \times 10^{-4}\ t$	3%
F_2	$= 3.153 - 5.8 \times 10^{-4}\ t$	3
F_3	$= 3.687 - 6.5 \times 10^{-4}\ t$	3
F_4	$= 3.644 - 6.3 \times 10^{-4}\ t$	3
L_2B	$= 2.214 - 4.2 \times 10^{-4}\ t$	2
C_1	$= 2.27 - 7.4 \times 10^{-4}\ t$	5
C_2	$= 2.26 - 7.4 \times 10^{-4}\ t$	5

Sources of Data and Methods of Estimation

Salts F_1 - F_4 - Estimated by additivity of molar volumes (see Ref. 1). The following molar volumes were used:

	600°C	800°C	Ref.
LiF	13.411 cm^3	14.142 cm^3	2
BeF$_2$	23.6	24.4	1,3
ThF$_4$ and UF$_4$	46.43	47.59	2

Salt L_2B - Three experimental determinations have been reported; refs. 5 and 6 were over a wide temperature range with the densities of ref. 6 averaging 3% higher than ref. 5. Reference 4 reports densities at 649°C which vary from 1.87 to 2.02 g cm^{-3}. The density-temperature equation given above

was derived from additive molar volumes; this equation yields densities that are approximately the average of the densities of refs. 5 and 6.

Salt C_1 - Preliminary pyknometric measurements.[7]

Salt C_2 - The relatively small concentration of NaF in C_1 would be expected to increase the density slightly over that for "pure" $NaBF_4$. The density-temperature equation was calculated by subtracting the contribution of NaF (ref. 1) from the molar volume of C_1.

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EXPANSIVITY (VOLUME COEFFICIENT OF THERMAL EXPANSION)

S. Cantor

Salt	Estimated Value at 600°C[a]	Uncertainty
F_1	$2.0_4 \times 10^{-4}/°C$	25%
F_2	2.0_7	25
F_3	1.9_7	25
F_4	1.9_3	25
L_2B	2.1_4	20
C_1	$4._0$	40
C_2	$4._0$	40

[a] For estimating the expansivity at other temperatures, please substitute in the appropriate density-temperature equation (see discussion below).

Sources of Data and Methods of Estimation

The expansivity is defined as

$$\alpha \equiv \frac{1}{V}\left(\frac{\partial V}{\partial T}\right)_P$$

where V, T and P are volume, temperature and pressure. Since density is inversely proportional to volume, the expansivity is usually derived from density-temperature data:

$$\alpha = -\frac{1}{\rho}\frac{d\rho}{dT} \quad \text{(P ordinarily one atm.)}$$

Most density data for liquids are linear with and decrease with temperature, i.e.,

$$\rho = \rho_0 - a\, t \tag{1}$$

ρ_0 and a are constants; t is usually in degrees Celsius. Thus,

expansivity is very simply

$$\alpha \;=\; \frac{a}{\rho} \hspace{5cm} (2)$$

The tabulated expansivities are consistent with the corresponding density-temperature equations in the "Density of Liquid" section of this report. To calculate the expansivity for any temperature, substitute in equations (1) and (2). As a rough approximation, the expansivity is one half to one third of the temperature coefficient of density as given by the constant $\underline{a}$ in eqn. (1).

References

Same as for the "Density of Liquid" section, page 29.

COMPRESSIBILITY (ISOTHERMAL)[a]

S. Cantor

Salt	Compressibility-Temperature Equation β_T in $cm^2 dyne^{-1}$, T in $^\circ K$
F_1 F_2 F_3 F_4 L_2B	$\beta_T = 2.3 \times 10^{-12} \exp (1.0 \times 10^{-3} \ T)$
C_1 C_2	$\beta_T = 9.0 \times 10^{-12} \exp (1.6 \times 10^{-3} \ T)$

The compressibilities pertain to the liquid and are all estimated; the uncertainty is a factor of 3.

[a]Isothermal compressibility is a function of pressure as well as temperature. The tabulated equations are less reliable at higher pressures (>50 atm).

Methods of Estimation

Salts F_1-F_4, L_2B: Estimated empirically from the compressibility-temp. equations of LiF and Li_2SO_4 (see ref. 1).

C_1 and C_2: Assumed to be slightly more compressible than NaI (see ref. 1).

Reasonable values derived for C_p/C_v and for sonic velocities (see Appendix A of this report) lend support to these estimated compressibilities.

References

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VAPOR PRESSURE

S. Cantor

Salt[a]	Pressure-Temperature Equation (P in torr, T in $^{\circ}$K)	Uncertainty in Pressure
F_1 F_2 F_3 F_4	$\log P = 8.0 - \dfrac{10,000}{T}$	A factor of fifty from 500-700°C
L_2B	$\log P = 9.04 - \dfrac{10,500}{T}$	A factor of ten from 500-700°C
C_1	$\log P \text{ (of } BF_3 \text{ vapor)}^{b} =$ $9.024 - \dfrac{5,920}{T}$	$\pm$ 10% from 400-700°C
C_2	Pressure of BF_3 depends on amount of salt and on vapor volume (see Discussion below)	

[a] In no case is the composition of the vapor congruent with the composition of the melt.

[b] The pressures given by the equation are those in equilibrium with a melt whose composition is fixed at $NaBF_4$-NaF (92-8 mole %).

Sources of Data and Methods of Estimation

Salts F_1-F_4: Estimated empirically from vapor pressure data of the LiF-BeF_2 system[1] and of LiF-UF_4 (73-27 mole %).[2] Although the uncertainty is relatively large, please note that the vapor pressures for the 500 - 700°C temperature range are quite low (between 10^{-2} and 10^{-5} torr).

L_2B: Estimated from data in the LiF-BeF_2 system.[2]

C_1 : Experimentally determined.[3]

Discussion - The Dissociation Pressure of $NaBF_4$

When $NaBF_4$ is thermally equilibrated at a temperature above its melting point the following dissociation occurs:

$$NaBF_4(\ell) = NaF(\ell) + BF_3(g) \qquad (1)$$
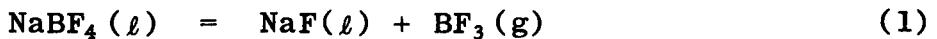
The dissociation product, NaF, dissolves in the $NaBF_4$. The system described by the above equation is bivariant; thus, a constant partial pressure of BF_3 above the melt requires that the temperature and the melt composition both be constant. For reaction (1) the BF_3 pressure is related to the composition of the melt by the equation:

$$P_{BF_3} = K \frac{a_{NaBF_4}}{a_{NaF}} \qquad (2)$$

where K is the equilibrium constant and a_i is activity. The temperature dependence of K has been derived from experimental data[3] and is given by

$$\ln K \text{ (in atm)} = \frac{-29,800}{RT \text{ (in } ^OK)} + \frac{26.41}{R} \qquad (3)$$

[29,800 cal and 26.41 cal $(^OK)^{-1}$ are the enthalpy and entropy of the reaction; R, the gas constant, is 1.98717 cal $(^OK)^{-1}$ $(g\text{-mole})^{-1}$].

A consequence of the bivariance of the $NaBF_4$-NaF system is that the equilibrium BF_3 vapor pressure is difficult to predict for melts in which the concentration of $NaBF_4$ is very large (>98 mole %). For these concentrations, a_{NaBF_4} is virtually unity, but a_{NaF} is very small (<0.1); hence, by

equation (2), P_{BF_3} tends to be quite high. Thus for any experiment in which crystalline $NaBF_4$ is encapsulated, the temperature of the sample should be kept as low as necessary or else sufficient vapor space should be included so as to permit the dissociation reaction (1) to occur.

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SURFACE TENSION

J. W. Cooke, S. Cantor

Salt	Surface Tension-Temperature Equation γ in dynes/cm, t in $^\circ$C	Uncertainty
F_1 F_2 F_3 F_4 L_2B	$\gamma = 260 - 0.12\ t$	$+30, -10\%$
C_1	$\gamma = 130 - 0.075\ t$	$\pm\ 30\%$
C_2	$\gamma = 120 - 0.075\ t$	$\pm\ 25\%$

Sources of Data and Methods of Estimation

Salts F_1-F_4, L_2B: Estimated primarily from maximum bubble pressure measurements on NaF-BeF$_2$,[1] LiF-BeF$_2$-ThF$_4$-UF$_4$,[2] LiF,[3] and ThF$_4$[3] melts. Measurements at one temp. (480°C) of LiF-BeF$_2$ (63-37 mole %)[4] by the ring method tends to support bubble pressure values. Sessile drop measurements[5] on L_2B, on LiF-BeF$_2$-ZrF$_4$-ThF$_4$-UF$_4$ (70-23-5-1-1), and on other fluoride melts would have led to higher predicted values. The higher uncertainty in the positive direction expresses the possibility that the sessile drop investigations might have yielded more accurate surface tensions.

Salt C_1 and C_2: Assumed that NaBF$_4$ (C_2) and NaI[6] exhibit (a) equal surface tensions at their melting points, (b) equal temperature coefficients of surface tension. Then

it was assumed that NaF in C_1 increased the surface tension

over that of C_2 by 10%.

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SOLUBILITY OF HELIUM, KRYPTON, AND XENON

G. M. Watson

Unit of solubility - $\underline{10^{-8}}$ moles of inert gas per (cm^3 melt-atm).

Salt	Temperature (°C)	He	Kr	Xe
F_1 F_2 F_3 F_4 L_2B	500	6.6	0.13	0.03
	600	10.6	0.55	0.17
	700	15.1	1.7	0.67
	800	20.1	4.4	2.0
C_1	500	44	20	12
	600	52	40	28
	700	60	69	54
	800	66	106	91
C_2	500	52	32	21
	600	61	61	46
	700	69	100	84
	800	75	148	136

All solubilities are estimated; the uncertainty is a factor of ten or greater.

Sources of Data and Methods of Estimation

Solubilities of noble gases were estimated by a method originally proposed by Blander et al.[1] The expression used in estimating the values given above is:

$$K_p = \frac{1}{RT} \text{ (polarization correction) } \exp\left(\frac{-18.08 \ r^2 \gamma}{RT}\right)$$

where

K_p = moles of gas/(cm^3 melt-atm)

r is the radius of the noble gas in Angstroms

γ is the surface tension of the liquid in dyne cm^{-1}

R in the pre-exponential term = 82.0561 cm^3-atm ($^{\circ}$K)$^{-1}$ (g-mole)$^{-1}$; in the exponential term R = 1.98717 cal ($^{\circ}$K)$^{-1}$ (g-mole)$^{-1}$

T is the absolute temperature in $^{\circ}$K .

The numerical values for the radii and for the "polarization corrections" are:

	He	Kr	Xe
Radius (Angstroms)	1.22	2.0	2.18
Polarization correction	0.14	1.0	1.34

The polarization corrections were determined empirically by comparison of experimental and calculated noble gas solubilities in NaF-ZrF$_4$ (53-47 mole %),[2] NaF-KF-LiF eutectic,[1,3] and LiF-BeF$_2$ (64-36 mole %).[3] The surface tensions used appear in this report on page 36.

The rather large uncertainty in the gas solubilities can be rationalized from the following considerations:

a. Experimental[1-3] and calculated (using the equation
    given in the previous paragraph) solubilities agreed
    to within a factor of three,

b. Calculated solubilities depend exponentially on the
    assumed value of the surface tension; for the salts
    of this report the surface tension, in each case
    estimated, has a large uncertainty.

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SOLUBILITY OF BF_3 GAS

S. Cantor, G. M. Watson

Unit of Solubility - $\underline{10^{-4}}$ moles BF_3 per $(cm^3$ melt-atm)

	Temperature ($^{\circ}$C)			
Salt	500	600	700	800
F_1	3.4	1.1	0.44	0.19
F_2	3.4	1.1	0.44	0.19
F_3	2.8	0.95	0.39	0.20
F_4	2.4	0.83	0.35	0.18
L_2B	3.2	1.0	0.38	0.18
C_1	See section on Vapor Pressures, page 33.			
C_2				

All solubilities are estimated; the uncertainty is a factor of ten or greater.

Sources of Data and Methods of Estimation

Solubilities of BF_3 were assumed to be analogous to solubilities of HF. For $LiF-BeF_2-ZrF_4-ThF_4-UF_4$ (65-28-5-1-1 mole %) the measured BF_3[1] and HF[2] solubilities both exhibited negative temperature dependence (inert-gas solubilities in fluoride melts are much smaller and show positive temperature dependence). The ratio of BF_3 to HF solubility in the range 500-800°C for this melt was the multiple used to estimate the

BF_3 solubility in L_2B from the measured values of HF solubility.[3]

Solubility of HF in F_1-F_4 was estimated by assuming the same "free fluoride" concentration dependence as had been observed for LiF-BeF_2 mixtures.[4] (For F_1-F_4, free fluoride is defined as X_{LiF} minus ($2X_{BeF_2}$ + $3X_{MF_4}$), where X is mole percent; for LiF-BeF_2 mixtures, free fluoride equals X_{LiF} minus $2X_{BeF_2}$). The BF_3 solubilities were then calculated by multiplying the estimated HF solubilities by the same ratios that were derived from the melt where both gas solubilities had been measured.

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APPENDIX A

ISOCHORIC HEAT CAPACITY (C_V), C_p/C_V, AND SONIC VELOCITY

Salt	Temp. °C	$\dfrac{cal}{g°K}$ C_V [a]	$\dfrac{cal}{(g\text{-mole})°K}$	$\dfrac{cal}{(g\text{-atom})°K}$	C_p/C_V	μ [b] $(m\ sec^{-1})$
F_1	500	0.29_5	$17._8$	7.1_5	1.1_5	2650
	600	0.29_2	$17._6$	7.0_8	1.1_6	2560
	700	0.28_8	$17._3$	6.9_7	1.1_8	2480
F_2	500	0.33_7	$16._9$	6.9_8	1.1_6	2850
	600	0.33_2	$16._6$	6.8_8	1.1_7	2760
	700	0.32_8	$16._4$	6.7_9	1.1_9	2670
F_3	500	0.28_9	$18._5$	7.2_2	1.1_4	2610
	600	0.28_5	$18._3$	7.1_3	1.1_6	2520
	700	0.28_2	$18._1$	7.0_6	1.1_7	2440
F_4	500	0.29_0	$18._9$	7.2_3	1.1_4	2620
	600	0.28_7	$18._7$	7.1_5	1.1_5	2530
	700	0.28_4	$18._5$	7.0_8	1.1_6	2440
L_2B	500	0.48_9	$16._2$	6.9_1	1.1_7	3420
	600	0.48_2	$15._9$	6.8_1	1.1_8	3310
	700	0.47_5	$15._7$	6.7_2	1.2_0	3200
C_1	500	0.31_2	$32._6$	5.7_4	1.1_5	1400
	600	0.30_8	$32._2$	5.6_7	1.1_7	1330
	700	0.30_5	$31._8$	5.6_0	1.1_8	1260

Theoretical C_V [c]
cal mole^{-1}(°K)$^{-1}$

Salt	Temp. °C	$\dfrac{cal}{g°K}$	$\dfrac{cal}{(g\text{-mole})°K}$		C_p/C_V	μ $(m\ sec^{-1})$
C_2	500	0.31_2	$34._2$	30.47	1.1_6	1400
	600	0.30_8	$33._8$	30.92	1.1_7	1330
	700	0.30_4	$33._3$	31.46	1.1_9	1260

a. Calculated from the equation,

$$C_V = C_p - \frac{\alpha^2 T}{\rho \beta_T}$$ where α is expansivity; ρ, density; β_T isothermal compressibility.

b. Calculated from the equation,

$$\mu = \left(\frac{C_p}{C_V \beta_T \rho}\right)^{1/2}$$ where μ is sonic velocity.

c. Calculated by assuming

$C_V = 6 R$ (harmonic oscillation of 2 ions) + 1.5R (free rotation of BF_4^- ion) + Vibrational*heat capacity of BF_4^-.

* Vibrational frequencies obtained from K. Nakamoto, Infrared Spectra of Inorganic and Coordination Compounds, John Wiley and Sons, N. Y., 1963, p. 106.

APPENDIX B

THERMAL DIFFUSIVITY,[a] KINEMATIC VISCOSITY,[b] AND PRANDTL NUMBER[c]

Salt	Temp. ($^{\circ}$C)	Therm. Diffy. ($cm^2\ sec^{-1}$)	Kin. Visc. ($cm^2\ sec^{-1}$)	Prandtl Number
F_1	500	$2.1_3 \times 10^{-3}$	$6.9_8 \times 10^{-2}$	$32._8$
	600	2.1_7	3.7_5	$17._2$
	700	2.2_2	2.2_9	$10._3$
F_2	500	$2.3_5 \times 10^{-3}$	$7.1_7 \times 10^{-2}$	$30._4$
	600	2.4_0	3.8_3	$15._9$
	700	2.4_5	2.3_4	$9._5$
F_3	500	$1.7_9 \times 10^{-3}$	$7.0_5 \times 10^{-2}$	$39._4$
	600	1.8_2	3.7_3	$20._5$
	700	1.8_6	2.2_6	$12._1$
F_4	500	$1.5_2 \times 10^{-3}$	$8.9_2 \times 10^{-2}$	$58._6$
	600	1.5_5	4.3_2	$27._8$
	700	1.5_8	2.4_4	$15._4$
L_2B	500	$2.0_9 \times 10^{-3}$	$7.4_4 \times 10^{-2}$	$35._6$
	600	2.1_4	4.3_6	$20._4$
	700	2.1_8	2.8_6	$13._1$
C_1	500	$1.8_2 \times 10^{-3}$	$1.0_2 \times 10^{-2}$	$5._6$
	600	1.8_9	0.6_8	$3._6$
	700	1.9_7	0.5_0	$2._5$
C_2	500	$1.7_9 \times 10^{-3}$	$1.0_2 \times 10^{-2}$	$5._7$
	600	1.8_6	0.6_8	$3._7$
	700	1.9_3	0.5_0	$2._6$

Calculated from the equations:

a. $X = \dfrac{k}{\rho C_p}$ where k is thermal conductivity; ρ, density; C_p, specific heat.

b. $\nu = \dfrac{\eta}{\rho}$ where η is viscosity in poise (g $cm^{-1}\ sec^{-1}$).

c. $Pr = \dfrac{\nu}{X} = \dfrac{\eta C_p}{k}$

APPENDIX C

CONVERSION FACTORS

	Multiply	By	To Obtain
Viscosity	centipoise	2.419	$lb_m/hr \cdot ft$
Thermal Conductivity	watts/$^\circ$C$\cdot$cm	57.8	Btu/hr$\cdot$ft$\cdot^\circ$F
Heat Capacity	cal/gm$\cdot^\circ$C	1.0	Btu/$lb_m \cdot^\circ$F
Heat of Fusion	cal/gm	1.8	Btu/lb_m
Density	gm/cm^3	62.43	lb_m/ft^3
Compressibility	cm^2/dyne	6.894×10^4	in $^2/lb_f$
Pressure	torr	0.019337	lb_f/in^2 (psia)
Surface Tension	dyne/cm	6.85×10^{-5}	lb_f/ft
	dyne/cm	2.203×10^{-3}	lb_m/sec^2

Composition of Salt Mixtures

| | Symbol | Mole % | | | | Liquidus Temp. (°C) |
		LiF	BeF$_2$	ThF$_4$	UF$_4$	
Fuel-Breeder Mixtures	F$_1$	73	16	10.7	0.3	$500° \pm 5°$
	F$_2$	72	21	6.7	0.3	$500° \pm 5°$
	F$_3$	68	20	11.7	0.3	$480° \pm 5°$
	F$_4$	63	25	11.7	0.3	$500° \pm 5°$
Flush Salt (present MSRE coolant)	L$_2$B	66	34	--	--	$458° \pm 1°$ (peritectic)
		NaBF$_4$	NaF			
Coolants	C$_1$	92	8			$385° \pm 1°$ (eutectic)
	C$_2$	100	-			$407° \pm 1°$ (melting point)

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