OAK RIDGE NATIONAL LABORATORY
operated by
UNION CARBIDE CORPORATION
for the
U. S. ATOMIC ENERGY COMMISSION

UNION CARBIDE

ORNL- TM- 1129

COPY NO. - 70

DATE - May 7, 1965

OXIDE CHEMISTRY AND THERMODYNAMICS OF MOLTEN LITHIUM FLUORIDE-BERYLLIUM FLUORIDE BY EQUILIBRATION WITH GASEOUS WATER-HYDROGEN FLUORIDE MIXTURES

A. L. Mathews*
C. F. Baes, Jr.

*Present address: Western Carolina College, Cullowhee, North Carolina.

A dissertation submitted to the Faculty of The University of Mississippi in partial fulfillment of the requirements for the Degree of Doctor of Philosophy in the Department of Chemistry.

ACKNOWLEDGMENTS

This report is based upon a dissertation submitted to the University of Mississippi in partial fulfillment of the requirements for the doctoral degree. The report describes research carried out in the Reactor Chemistry Division of the Oak Ridge National Laboratory, which is operated by the Union Carbide Corporation for the Atomic Energy Commission. The research was supported by the Oak Ridge Graduate Fellowship Program of the Oak Ridge Institute of Nuclear Studies and was directed by a committee appointed by Dean Lewis Nobles of the University of Mississippi Graduate School which was composed of Dr. George Vaughan and Dr. Allen Cahill of the Department of Chemistry at the University of Mississippi and Dr. C. F. Baes, Jr., and Dr. C. H. Secoy of the Reactor Chemistry Division of the Oak Ridge National Laboratory.

CONTENTS

Page

LIST OF FIGURES

LIST OF TABLES

OXIDE CHEMISTRY AND THERMODYNAMICS OF MOLTEN LITHIUM FLUORIDE-BERYLLIUM FLUORIDE BY EQUILIBRATION WITH GASEOUS WATER-HYDROGEN FLUORIDE MIXTURES

A. L. Mathews C. F. Baes, Jr.

ABSTRACT

The transpiration method was used to equilibrate dilute gaseous mixtures of HF and H_2O in hydrogen carrier gas with molten LiF-BeF_2 mixtures varying in composition from 0.25 to 0.80 BeF_2, both saturated and unsaturated with crystalline BeO, in the temperature range 500 to 700°C. The partial pressure data were used to evaluate the equilibrium quotient for the reaction of HF and H_2O with solid BeO and dissolved BeF_2. Equilibrium quotients were also obtained for the formation of oxide and hydroxide ions in the liquid phase.

These equilibrium quotients were employed to determine: (1) thermodynamic activities of LiF and BeF_2 in the mixtures; (2) thermodynamics of liquid BeF_2; (3) stability of hydroxide in the melt; and (4) solubility of BeO in the LiF-BeF_2 system as a function of temperature.

I. INTRODUCTION

Molten mixtures of LiF and BeF_2 have been the subject of numerous investigations in recent years primarily because of their suitability as a carrier solvent for UF_4 in fluid fueled nuclear reactors. In addition, these solutions are especially worthy of study because the components - highly ionic LiF and highly associated, more covalent BeF_2 - represent extreme types of fluoride salts. Although the molten LiF-BeF_2 system has received considerable attention from both a practical and a theoretical point of view, the study of its chemistry is still far from complete. According to Everest,[1] many of the investigations of beryllium fluoride systems did not take into account the role of moisture and the resulting hydrolysis products. If this information were available, future investigators could make appropriate experimental adjustments and corrections.

The purpose of the present investigation was to study heterogeneous reactions of the type

$H_2O(g)$ + fluoride species(soln) $\rightleftharpoons$ $HF(g)$ + oxygen species(soln)

in the molten $LiF-BeF_2$ system. From such a study information could in principle be obtained about: (1) the thermodynamic activities of LiF and BeF_2, (2) the solubilities and stabilities of oxides, and (3) the interaction of oxide with the proton and perhaps other cations in this molten fluoride system. Information about the chemical reactivity of the components in the molten $LiF-BeF_2$ system could be obtained from the thermodynamic activities of LiF and BeF_2. The oxide chemistry of this system is of interest because oxide is a principal impurity to be dealt with in preparative work and because metal oxides are known to be only sparingly soluble in $LiF-BeF_2$ mixtures.

The presence of oxide species in the molten $LiF-BeF_2$ system, which is currently being used as the solvent in the Molten-Salt Reactor Experiment at ORNL, constitutes an undesirable impurity since inadvertent precipitation of sparingly soluble uranium (and other) oxides might result in unstable reactor operation. One of the steps in the purification of melts for reactor operation is sparging with a mixture of HF and H_2 to remove oxide. The present study of the equilibria involved would yield additional guidelines for this treatment.

The reactivity of oxide with beryllium and other cations has been investigated as a possible means of removal of reactor products, or of uranium to be reprocessed for later use. In order for proper evaluation of these methods to be carried out, a thorough understanding of the interactions occurring in melts would be desirable. For example, the use of

H_2O as the source of reactive oxygen for oxide precipitating schemes and the use of HF for removal of oxide require that the stability of the intermediate hydroxide be evaluated in the melts.

In the present study the transpiration method was used to equilibrate dilute gaseous mixtures of HF and H_2O in hydrogen carrier gas with molten LiF-BeF_2 mixtures varying in composition from 0.25 to 0.80 BeF_2, both saturated and unsaturated with crystalline BeO, in the temperature range 500 to 700°C. The primary data from the measurements were used to evaluate the equilibrium quotient for the reaction of HF and H_2O with solid BeO and dissolved BeF_2. Equilibrium quotients were also obtained for the formation of oxide and hydroxide ions in the liquid phase. These quantities in turn could be used to obtain the thermodynamic activities of LiF and BeF_2 as well as the solubility of BeO and the stability of hydroxide. Use of the HF-H_2O equilibria as a much needed analytical tool for the determination of oxide in such melts was also indicated.

Physical Properties of the LiF-BeF_2 System

Beryllium fluoride is frequently found in the form of a glass rather than a crystalline solid. The beryllium fluoride glass consists of a random network structure in which the beryllium atoms are surrounded tetrahedrally by four fluorine atoms and each fluorine atom by two beryllium atoms.[1] Liquid beryllium fluoride retains the polymeric character of the glass as indicated by its high viscosity. In contrast to the covalent nature of beryllium fluoride, lithium fluoride is a highly ionic salt. The addition of LiF to liquid BeF_2 causes a breaking down of the polymeric structure, but apparently the tetrahedral BeF_4^{2-} groups are retained.

Although it isn't proof of structure in the liquid phase, the fact that a compound Li_2BeF_4 can be precipitated from the melt may be some indication of the short range order in the liquid phase.

Phase behavior of the LiF-BeF_2 system has been studied extensively by Thoma, et al.[2] A copy of their published diagram (Figure 1) is included here to illustrate the major characteristics of the system.

The melting point of BeF_2 is 548°C; the melting point of LiF is 848°C. The liquidus temperatures for the BeF_2-rich region have been difficult to obtain because of the high viscosity of these solutions.[3] A brief summary of the phase studies of various BeF_2 systems is included in reference 1. Many of these systems parallel those of the much higher melting silicate glasses.

Studies of the LiF-BeF_2 system in the temperature range of the present work (500 to 700°C) are restricted to the region between the high liquidus temperatures at low BeF_2 concentrations and high viscosity at high BeF_2 concentrations.

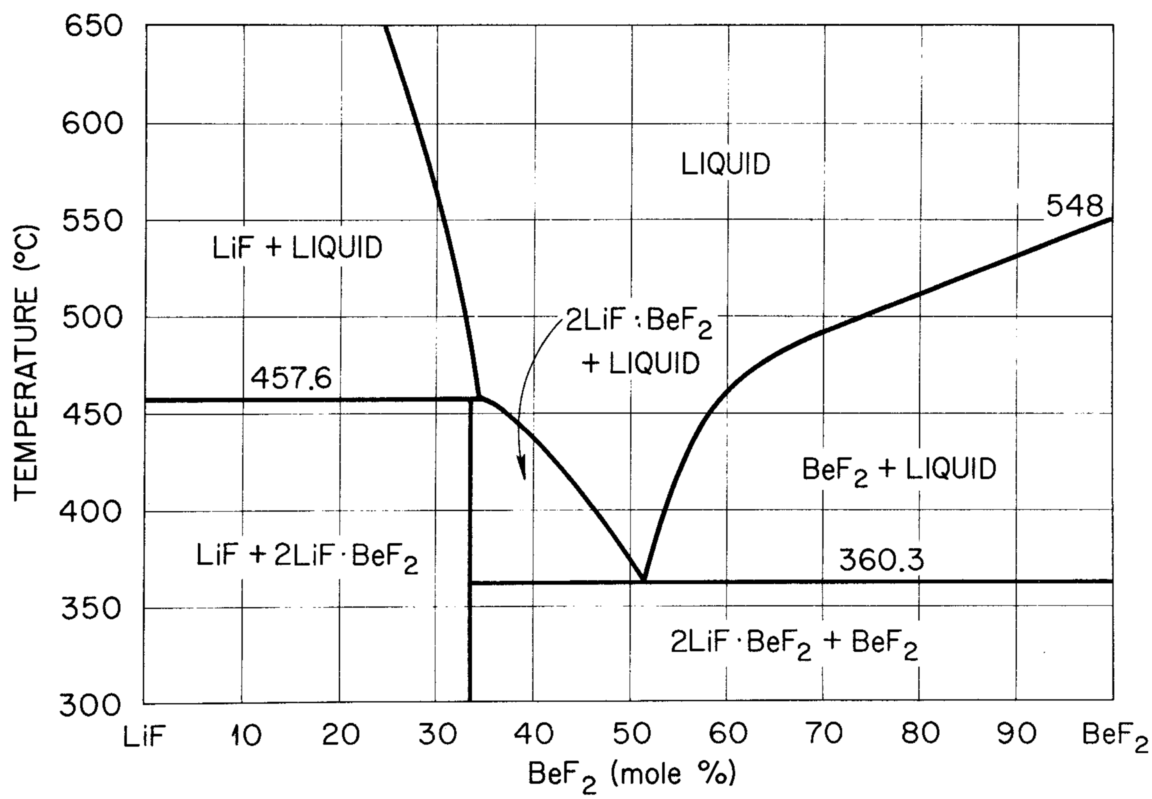

Fig. 1. Phase Diagram of the $LiF-BeF_2$ System (From Thoma, ref. 2).

Thermodynamic Properties of the Various Possible Species

From previous studies of LiF, BeF_2, and the other possible species present, some information can be drawn about the expected behavior of the system. The thermochemical data for BeF_2 are summarized in the JANAF Tables.[4] There is considerable uncertainty in ΔH_f° of $BeF_2(\underline{l})$, which was derived from the $\Delta H_f^\circ{}_{298.15}$ for the crystal and the appropriate heat capacity functions for both the solid and liquid phases. Two sources of error were cited in JANAF. The heat capacity studies were made on samples of BeF_2 which contained BeO and H_2O. Also, the heat of fusion is uncertain. The value of 2 kcal/mole was used in the tabulation (because of the similarity of BeF_2 glass to B_2O_3 and SiO_2) even though a value of 12.9 kcal/mole was determined from the vapor pressures over solid and liquid BeF_2. Determination of ΔH_f° of $BeF_2(\underline{l})$ would help resolve some of these difficulties.

The thermochemical properties of $BeF_2(g)$ have been based on properties of the liquid and the heat of vaporization of $BeF_2(\underline{l})$ except for the work of Greenbaum, <u>et al</u>.[5] who determined equilibrium constants for the reaction

$BeO(s) + 2HF(g) \rightleftharpoons BeF_2(g) + H_2O(g)$

over the temperature range 670 to 970°C. They report that a plot of log K vs 1/T yields a least squares slope corresponding to 20.5 ± 1.7 kcal/mole for ΔH_r over the temperature range studied, and that a plot of ΔF_r vs T gives a value of 6.0 ± 0.3 cal deg^{-1} $mole^{-1}$ for ΔS_r by a least squares analysis. If the calculated line for log K vs 1/T using these parameters were drawn and the reported equilibrium quotients plotted, the line would fall on the same side of all points. A least squares analysis using all published data, with equal weighting for all points, gave values of ΔH_r = 20.28 ± 0.84 kcal/mole and ΔS_r = 6.67 ± 0.77 cal deg^{-1} $mole^{-1}$. From their values Greenbaum, <u>et al.</u>[5] reported the $\Delta H_f^o{}_{298}$ of $BeF_2(g)$ as -191.3 ± 2.0 kcal/mole and $S^o{}_{298}$ of $BeF_2(g)$ as 52.4 ± 0.3 cal deg^{-1} $mole^{-1}$. Based on the recalculation, the values would be -191.5 ± 1.1 and 53.1 ± 0.8, respectively.

The vapor pressure of $BeF_2(\underline{l})$ has been studied extensively.[6-10] Sense, <u>et al.</u>[6,7] studied the vapor pressure from 745 to 1021°C. Two Russian groups[8,9] have reported vapor pressure studies. All of these are in general agreement but have slight differences. The most extensive, and probably the best, study (550 to 950°C) was that of Greenbaum, <u>et al.</u>[10] Since the enthalpy and entropy of vaporization are reasonably well known from these measurements, a combination of this information with independently determined entropy and enthalpy of formation of $BeF_2(\underline{l})$ should provide a new means for evaluating the thermodynamic properties of $BeF_2(g)$.

The thermodynamic values of $H_2O(g)$, $HF(g)$, and $BeO(s)$ are well characterized throughout the temperature range of the present measurements (see reference 4). Thus, if the ΔH_r can be determined for the reaction

$$H_2O(g) + BeF_2(\underline{l}) \rightleftharpoons 2HF(g) + BeO(s) ,$$

then ΔH_f^o of $BeF_2(\underline{l})$ can be calculated. The greatest uncertainty in ΔH_f^o for $HF(g)$ is the correction which should be applied for the imperfection at room temperature. Several recent publications have dealt with the subject.[11-15] Franck and Spalthoff[11] reported that the enthalpy of vaporization rises from 89.5 cal/g at 19.4°C to a max of 146 cal/g at 130°C and decreases at higher temperature. Armitage, <u>et al.</u>[12] show that the various thermodynamic properties can best be explained by assuming that HF exists in the gas phase principally as monomers and hexamers, but no actual indication of the average molecular weight as a function of temperature or pressure is given. Yabroff, *et al.*[13] have summarized most past work in their report. They conclude that HF molecules are strongly associated into polymeric forms and that dissociation is accompanied by large changes in enthalpy. Armstrong[14] and Feder, *et al.*[15] have considered the effect of this association on the heat of formation of HF(g). The average molecular weight of HF(g) at 1 atm and 25°C is 54, at 0.4 atm and 25°C is 22, and at 1 atm and 80°C is 20.[16] For pressures as low as a few hundredths of an atmosphere both HF and H_2O are reasonably ideal at 25°C and undoubtedly are ideal at melt temperatures.

From the thermodynamic quantities tabulated in JANAF, equilibrium constants were calculated for several conceivable reactions involving HF, H_2O, BeF_2, BeO, and LiF. These are presented in Table 1 along with the

Table 1. Equilibrium Constants Predicted from Thermodynamic Data

Reaction		K at 800°K	K at 1000°K	ΔH_r kcal
BeO(s) + 2LiF(s)	⇌ Li_2O(s) + BeF_2(l)	3×10^{-14}	2×10^{-11}	52.1
BeO(s) + H_2O(g)	⇌ $Be(OH)_2$(s)	5×10^{-4}	1×10^{-4}	-12.5
BeF_2(l) + $2H_2O$(g)	⇌ $Be(OH)_2$(s) + 2HF(g)	1×10^{-5}	6×10^{-5}	12.8
2BeO(s) + 2HF(g)	⇌ BeF_2(l) + $Be(OH)_2$(s)	2×10^{-2}	2×10^{-4}	-37.9
$Be(OH)_2$(s) + 2LiF(s)	⇌ 2LiOH(l) + BeF_2(l)	1×10^{-8}	3×10^{-6}	46.2
BeF_2(l) + H_2O(g)	⇌ BeO(s) + 2HF(g)	2×10^{-2}	6×10^{-1}	25.4
BeF_2(g) + H_2O(g)	⇌ BeO(s) + 2HF(g)	5×10^{5}	1×10^{3}	-21.7
BeO(s) + H_2O(g)	⇌ $Be(OH)_2$(g)	2×10^{-10}	2×10^{-8}	41.6
BeF_2(l)	⇌ BeF_2(g)	1×10^{-6}	2×10^{-4}	47.1

calculated heats of reaction. Although the equilibrium constants refer to reactants and products which are pure solids or liquids, they could be applied to reactions in solution if appropriate activities were used. From these data the following predictions are made:

(1) Oxygen containing compounds of Li should react to form Be compounds. If Li_2O were added to an LiF-BeF_2 melt, the Li_2O should react almost quantitatively to form BeO and LiF. If LiOH were added to an LiF-BeF_2 melt, the LiOH should react almost quantitatively to form $Be(OH)_2$ and LiF.

(2) The formation of $Be(OH)_2$ as a separate solid phase at temperatures as high as 800°K is very unlikely.

(3) All of the stable compounds have low volatilities at the temperatures of interest.

A more precise determination of the ΔH_f° of $BeF_2(\underline{l})$ and of the activities of BeF_2 and LiF in the melt would allow more quantitative predictions of reactions in the LiF-BeF_2 system.

Thermodynamic Studies of Molten Salt Mixtures

The determination of thermodynamic activities of melt components in molten salts has received considerable attention. Blander has summarized much of the work through 1962.[17]

The relationships between activities and activity coefficients depend on the choice of concentration units. The most frequent unit for expressing concentration of mixed solvents is the mole fraction. In molten salts the ion fraction is frequently used. For systems in which all salts contain the same anion, mole fraction and ion fraction are equal. The term "mole fraction" will be used in the text.

According to the Temkin model,[18] in which salts are considered to be completely ionic, the ideal activity of a component is equal to the product of ion fractions of its constituents ($a_{ij} = X_i X_j$). For a solution which contains only one anion, j, the ion fraction, X_j, equals one. The ion fraction of each cation is equal to the mole fraction of that component. The activities of components are usually referred to the pure liquid (supercooled if necessary) as the standard state. Occasionally, the activities are referred to the pure crystalline solid for experimental convenience.

The activities of components in solution have been measured by the following methods: vapor pressures, freezing point depression, electrode potentials, and heterogeneous equilibria. The vapor pressure method is complicated by the formation of complex species in the vapor phase. Determination of activities from freezing point depressions requires that the heat of fusion and the ΔC_p for the pure solid and liquid solvent be known. Electrode potential measurements of activities are often made in cells with liquid junction. Such measurements are limited to dilute solutions, which are expected to give small liquid junction potentials.

The use of heterogeneous equilibria has thus far been limited. The activity of $MgCl_2$ has been determined[19,20] in mixtures with KCl and NaCl by use of the equilibrium

$$MgCl_2(soln) + \frac{1}{2}O_2(g) \rightleftharpoons MgO(s) + Cl_2(g) \ .$$

Blood, _et al._[21] determined the activities of various metal fluorides in LiF-BeF_2 mixtures using the equilibrium

$$M(s) + xHF(g) \rightleftharpoons MF_x(soln) + (x/2)H_2(g) \ .$$

In studies of this type it is important that the solid present be well characterized and relatively insoluble and, of course, that all phases are in equilibrium.

If the activity of one of the components of a binary mixture is known as a function of composition, the other one can be determined by integration of the Gibbs-Duhem equation. From the activities, such properties as molar heats of mixing, excess chemical potential, vapor pressure, and phase behavior can be derived.

At present, a general theory of the behavior of melts has not been developed to the point that activities can be predicted for a system such as LiF-BeF_2.

The activities of LiF and BeF_2 have been reported for a limited number of cases. Berkowitz and Chupka in 1960 reported the activities in an equimolar mixture from relative ion intensities during mass-spectral analysis.[22] Recently, Büchler has reported determinations by a more careful mass-spectral analysis and emf measurements. The emf measurements were conducted at two temperatures in a concentration cell containing pure BeF_2 in one compartment and an LiF-BeF_2 mixture in the other.[23] Büchler used a twin crucible assembly in his determination of activities with the mass spectrometer to facilitate comparison of pure compound and mixture.[24] The results of these experiments are compiled in Table 2.

Table 2. Activities of LiF and BeF_2 from Literature

Conc X_{BeF_2}	Temp (°C)	a_{BeF_2}*	γ_{BeF_2}	a_{LiF}*	γ_{LiF}
0.50[a]	627	0.443	0.89	0.0246	0.049
0.25[b]	633	0.027	0.11		
0.25[b]	692	0.039	0.16		
0.26[c]	604	0.016	0.06	1	
0.67[c]	604	0.86	1.3	0.076	0.23

*Activities referred to $BeF_2(\underline{l})$ and LiF(s), respectively.

[a]From Berkowitz and Chupka, mass spectrometry.

[b]From Büchler, emf.

[c]From Büchler, mass spectrometry.

Solubilities of Gases in Melts

In addition to the studies of activities in melts, the solubilities of gases have also been of interest. Watson, et al.[25] have studied the solubility of the inert gases in molten fluorides, including the LiF-BeF_2 system. All solubilities obey Henry's law and increase with increasing temperature and with decreasing atomic weight of gas.

Burkhard and Corbett[26] reported the solubility of water in molten LiCl-KCl mixtures. Apparently Henry's law was obeyed for low pressures, with the deviations above a few millimeters attributed to hydrolysis. However, no analyses were reported on the gases to determine the amount of H_2O and HCl in the gas phase in equilibrium with the melt.

Shaffer, et al.[27,28] have studied the solubility of HF in molten fluoride mixtures. In the LiF-BeF_2 system[28] solubility of HF increases with decreasing temperature and with decreasing BeF_2 concentration. The Henry's law constants indicate that the solubility of HF in the $LiF-BeF_2$ system should be less than 150 ppm per atm HF above the melt. Thus, for the present studies with partial pressures less than 0.02 atm HF, the solubility should be less than 3 ppm.

Determination of Oxides in Melts

Since the presence of oxide in molten halides affects many physical and chemical properties, quantitative procedures for determination of the amount of oxide present have been widely sought. Goldberg, et al.[29] proposed high-temperature fluorination with $KBrF_4$ with liberated O_2 measured tensimetrically. This method works reasonably well, but the greatest uncertainties were found with fluoride melts and with samples containing less than 300 ppm.

Porter and Brown[30] used an inert-gas fusion technique (described by Banks, et al.[31]) in determining oxide concentrations in molten fluorides. Samples of the melt were withdrawn through graphite filters and the entire sample was used in the analysis. The reported analyses indicated the relative precision was ± 10% for oxide concentrations of about 0.5%. No indication was made of applicability to lower concentrations.

Delarue[32,33] has determined relative oxide solubilities in LiCl-KCl melts electrochemically. The concentration of oxide was determined in the melt with Pt-C electrodes vs a $Pt-PtCl_2$ reference electrode. These results permitted identification of a variety of reactions involving oxide.

None of the methods cited are adequate for determining oxide in the $LiF-BeF_2$ system. From studies of oxide solubility by Baes, et al.[34] in which weighed samples of BeO were added to fluoride melts containing uranium and zirconium, it was estimated that the solubility of BeO was less than 1000 ppm. Determination of oxide content in filtered samples by the Goldberg method,[29] however, gave very inconclusive results.

Suitable Experimental Approach

The molten salt production facility at the Oak Ridge Y-12 Plant has made use of HF-sparging to remove oxide from solution in the form of water.[35] Since the production procedure parallels the transpiration method for determining vapor pressures of liquids, the feasibility of transpiration techniques for an equilibrium study of the LiF-BeF_2 system was considered. Much of the feasibility study was conducted by M. K. Kemp who determined partial pressures over the ZrO_2-saturated LiF-BeF_2-ZrF_4 system and was able to estimate the activity of ZrF_4 in solution.[36]

Transpiration Method

The transpiration method for determining vapor pressures of liquids is well established.[37] In order for the transpiration method to work satisfactorily in studies of heterogeneous equilibria, the following conditions should be met:

(1) Reactions must be fast enough and flow rates must be slow enough to allow equilibration between gas and condensed phases.

(2) Adequate stirring of liquid phases must be maintained to provide uniform concentrations of reactants.

(3) The vapor pressures of condensed phases must be low enough to prevent significant loss by transport in the carrier gas.

(4) Reacting gases should be unassociated.

(5) An adequate means of removal and measurement of reactive gases from the flowing gas stream must be available.

The best evidence that these conditions have been met in the present system is described in Chapter III. The variation in flow rates and in the amount of reaction necessary to maintain equilibrium was used to test the validity of the first two conditions. The vapor pressure data for BeF_2[10] and LiF-BeF_2 mixtures[38] indicated that very little BeF_2 would be vaporized. The vapor pressure of BeO at the experimental temperatures is insignificant.[39] Although both H_2O and HF are at least partially associated at room temperature, the dilute gases at melt temperatures are expected to be ideal.

Determinations of the partial pressures of HF and H_2O over aqueous hydrofluoric acid solutions have been made by means of the transpiration method.[40] Measurements were made over solutions of 10, 20, 30, 50, and 70% hydrofluoric acid, at temperatures from 0 to 70°C. The HF in the carrier gas stream was absorbed by an aqueous KOH solution. The difference between weight lost by the saturator and weight of HF found in gas stream was assumed to be the amount of water removed from the saturator. Since this difference could not be used in the present studies, a suitable method for measuring water was needed. Kemp investigated several methods of analysis. Condensation of samples in a cold trap with subsequent weighing and analysis for per cent HF was not satisfactory primarily because of the small size of the samples. Adsorption of the water by a suitable dessicant in a weighed drying tube was not satisfactory because HF was also adsorbed.

Finally, it was observed that a mixture of methanol and pyridine was suitable for removal of water from a flowing stream even in the presence of HF. Titration of the water with Karl Fischer Reagent could be performed in the vessel and since Karl Fischer Reagent contains excess methanol and pyridine, several successive titrations could be performed. Apparently the HF reacted with excess pyridine to form a fairly soluble pyridinium hydrofluoride which did not interfere with the titration.

Karl Fischer Reagent consists of I_2 and SO_2 dissolved in pyridine and methanol (actually the "stabilized reagent" contains methylcellosolve instead of methanol). Although the titration is straightforward,[41] the reaction occurs in two steps:

$$H_2O + C_5H_5N + C_5H_5N \cdot SO_2 + C_5H_5N \cdot I_2 \rightarrow 2C_5H_5NH^+I^- + C_5H_5\overset{+}{N}\begin{matrix} SO_2 \\ | \\ O^- \end{matrix}$$

$$C_5H_5\overset{+}{N}\begin{matrix} SO_2 \\ | \\ O^- \end{matrix} + CH_3OH \rightarrow C_5H_5NHOSO_2OCH_3 \quad .$$

When titrating H_2O with Karl Fischer Reagent the endpoint corresponds to the first appearance of I_2. For macrotitrations the reagent serves as its own indicator; however, for microtitrations a more precise method is needed. Several authors have reported suitable circuits for the amperometric determination of the endpoint. The one actually used was quite similar to the one described by Nernitz.[42]

Equilibria

If the molten LiF-BeF_2 system comes into contact with water vapor, hydrolysis occurs releasing some HF into the gas space and leaving some oxygen-containing species dissolved in the melt. For a closed system at equilibrium, the amount of HF and H_2O in the gas phase will depend on (1) the amounts of HF and H_2O initially introduced, (2) the solubility of HF and H_2O in the melt, (3) the concentration of oxide in the melt, (4) the concentration of hydroxide, a reactive intermediate (in the sense that it can exist only in the presence of HF and H_2O), in the melt, and (5) the amount of HF and H_2O consumed in side reactions which do not influence the oxide reaction scheme.

If the above quantities are properly controlled or measured, useful equilibrium data can be obtained. The reaction of HF and H_2O at high temperatures with the structural metal used (nickel) can be suppressed by the presence of H_2. Side reactions are essentially eliminated if the purity of melts is sufficiently high. The solubility of HF in melts in the absence of oxygen species has been mentioned and is small compared with the best estimates of oxide solubility. It seems reasonable to assume that water solubility would be of the same order of magnitude as that of HF and the experimental results supported this. One method of controlling the oxide concentration would be saturation of the melt with a sparingly soluble oxide such as BeO. With the above conditions set, transpiration experiments can be used to control the amounts of HF and H_2O introduced and to measure the hydroxide concentration indirectly through the material difference between influent and effluent gas streams.

For a BeO-saturated, LiF-BeF_2 melt with excess solid BeO present the following equilibrium is valid:

$$H_2O(g) + BeF_2(soln) \rightleftharpoons 2HF(g) + BeO(s) .$$

If both HF and H_2O are assumed to be ideal gases and if the thermodynamic activities of the condensed species are represented by "a", the equilibrium constant for the reaction would be

$$K_a = (P_{HF})^2 a_{BeO}/P_{H_2O} a_{BeF_2} .$$

Since a_{BeO} is 1 by definition,

$K_a = (P_{HF})^2/P_{H_2O}a_{BeF_2} = Q/a_{BeF_2}$.

This equilibrium is independent of the presence or absence of hydroxide in the melt.

The quantity $(P_{HF})^2/P_{H_2O}$ for BeO saturated melts, defined above as Q, can be obtained from determination of partial pressures in the gas phase alone. Thus Q, which is proportional to the activity of BeF_2, would be equal to K_a for the equilibrium involving pure liquid BeF_2 at the same temperature. Determination of Q for various temperatures and compositions of BeF_2 provides the data necessary for thermodynamic calculations involving the melts.

The following additional equilibria involving oxide and hydroxide ions in the melt should also be considered. (The corresponding equilibrium quotients, which are shown, may be considered constant for a given LiF-BeF_2 composition and temperature since at the low concentrations of oxide and hydroxide involved, the activity of F^- and the activity coefficients $\gamma_{O^{2-}}$ and γ_{OH^-} can reasonably be assumed constant.) Since $[F^-]$ is reasonably constant for a given melt composition, $[F^-]$ is incorporated into the equilibrium quotients.

$H_2O(g) + 2F^-(soln) \rightleftharpoons 2HF(g) + O^{2-}(soln)$

$Q_O = (P_{HF})^2[O^{2-}]/P_{H_2O}$

For oxide saturated melts $[O^{2-}]$ is constant, hence

$Q_O/[O^{2-}]_{sat} = (P_{HF})^2/P_{H_2O} = Q.$

There are three reactions involving hydroxide in solution.

$H_2O(g) + F^-(soln) \rightleftharpoons HF(g) + OH^-(soln)$

$Q_A = (P_{HF})[OH^-]/P_{H_2O}$

$H_2O(g) + O^{2-}(soln) \rightleftharpoons 2OH^-(soln)$

$Q_b = [OH^-]^2/(P_{H_2O})[O^{2-}]$

and for a melt saturated with oxide where $[O^{2-}]$ is constant,

$Q_B = [OH^-]^2/P_{H_2O}$.

$HF(g) + O^{2-}(soln) \rightleftharpoons OH^-(soln) + F^-(soln)$

$Q_c = [OH^-]/(P_{HF})[O^{2-}]$

and for a melt saturated with oxide,

$Q_C = [OH^-]/P_{HF}$.

Note that not all of the equilibrium quotients are independent. Since Q_O is independent of $[OH^-]$ and Q_A is independent of $[O^{2-}]$, these two equilibrium quotients are the most conveniently employed. Both Q_b and Q_c can be expressed as functions of Q_O and Q_A.

$Q_b = (Q_A)^2/Q_O$ and $Q_c = Q_A/Q_O$

The relationships for BeO saturated melts are given by

$Q_B = (Q_A)^2/Q$ and $Q_C = Q_A/Q$.

Stoichiometric relationships show that the number of protons in a given volume of effluent gas will be less than the number in the influent gas by the amount that has appeared as OH^- added to the melt. If the proton level in the effluent gas stream exceeds the level in the influent stream, OH^- is being removed from the melt. The difference in H_2O content between the influent and effluent gas streams corresponds to the total amount of oxide plus hydroxide being added to or removed from the melt. Likewise, the difference in HF content between the influent and effluent gas streams corresponds to the amount of fluoride being added to or removed from the melt. For all experiments conducted, the change in fluoride was small enough so that the LiF-BeF_2 concentration in the melt was assumed to be constant throughout a given experiment.

For simplicity in handling equations the following notation will be used:

wt of melt = w(kg)

vol of gas (measured at T) through melt = V(liter)

$V/wRT = W(\text{mole kg}^{-1}\ \text{atm}^{-1})$

$P_{HF} = a + bW$ and $P_{H_2O} = c + dW$ (atm) for influent pressures

(NOTE: When the aqueous HF saturator was employed b and d were always 0. When gas mixing was employed b and d were small negative numbers because small opening in valve tended to gradually close.)

$P_{HF} = x$ and $P_{H_2O} = y$ (atm) for effluent partial pressures

$[O^{2-}] = r$

$[OH^{-}] = s$

The equilibrium quotients may now be expressed as

$Q = x^2/y$, $Q_O = x^2r/y$, and $Q_A = xs/y$.

Saturated Melts

For constant influent partial pressures of H_2O, HF, or both there will be an ultimate steady state condition in which the effluent partial pressures are not changing, hence r and s are constant. In fact r will be constant at all times in a given experiment unless rate of reaction to form hydroxide and water exceeds the dissolving rate of solid BeO. The speed with which x and y approach a steady state is controlled primarily by Q and Q_A. According to the equations described earlier, the difference in proton level of influent and effluent streams is equal to the change in hydroxide concentration. Thus,

$$ds/dW = (a + 2c) - (x + 2y) \quad .$$

Both ds and y may be eliminated by using the relationships $Q = x^2/y$ and $Q_A = xs/y$. Rearrangement of the former gives $y = x^2/Q$. Rearrangement and substitution into the latter gives $s = Q_A x/Q$, which may be differentiated to give $ds = (Q_A/Q)dx$. Thus

$dx/dW = (Q/Q_A)[(a + 2c) - (x + 2x^2/Q)]$.

Separation of variables gives

$$\frac{dx}{(a + 2c) - x - (2/Q)x^2} = (Q/Q_A)\ dW \quad .$$

Defining (a + 2c) as A, this equation may be integrated to give

$$\frac{1}{[1 + (8A/Q)]^{\frac{1}{2}}} \ln \frac{(4/Q)x + 1 + [1 + (8A/Q)]^{\frac{1}{2}}}{(4/Q)x + 1 - [1 + (8A/Q)]^{\frac{1}{2}}} = (Q/Q_A)W + \text{constant} \quad .$$

The correlation of experimental measurements with this equation is described in Chapter III. The computer program for this correlation was written by R. J. McNamee, Operations Analysis Division, ORGDP.

Unsaturated Melts

For unsaturated melts the change in r and s can be determined by material balance. The difference between influent and effluent partial pressures of H_2O corresponds to the change in total oxide concentration

$(dr + ds)/dW = (c - y)$.

The difference in proton levels corresponds to the change in hydroxide concentration just as in saturated melts

$ds/dW = (a - x) + 2(c - y)$.

By difference

$dr/dW = (x - a) + (y - c)$.

Combination of these equations with $Q_A = xs/y$ and $Q_O = x^2r/y$ could, in principle, yield integral equations expressing Q_A and Q_O as a function of the measured quantities. However, the resulting pair of simultaneous differential equations could not be integrated. The following simultaneous differential equations were obtained by elimination of x and y from the expressions for dr/dW and ds/dW:

$$ds/dW = (a + 2c) - (Q_O s/Q_A r)(1 + 2s/Q_A)$$

and

$$dr/dW = (Q_O s/Q_A r)(1 + s/Q_A) - (a + c) \quad .$$

These equations could be solved in the differential form to obtain values of r and s as a function of W for specified values of Q_O and Q_A. If r, s, Q_O and Q_A are specified at a given W, values for x and y may be calculated.

The method of solution of the equations and the correlation of experimental data by this method are described in Chapter III. The computer program for this correlation, "FIASCO", was written by M. T. Harkrider, ORNL Mathematics Division.

II. EXPERIMENTAL

Chemicals

Gases

Commercial H_2 was purified before use by passage through a deoxo unit, a magnesium perchlorate drying tube and, finally, a liquid N_2 trap. Commercial He was passed through an ascarite trap, a magnesium perchlorate drying tube and, finally, a liquid N_2 trap. Anhydrous HF was used directly from a commercial cylinder without further purification. B & A reagent grade 48% hydrofluoric acid was used as the source of HF(g) and H_2O(g).

Melt Components

Beryllium fluoride was from Brush Beryllium Company (material to be used in preparing the Molten-Salt Reactor Experiment fuel salt). Lithium fluoride was B & A reagent grade. Various mixtures were prepared by melting together weighed samples of the two components. Samples of the mixtures were withdrawn in filter sticks and analyzed for Be, Li, F and impurities (Fe, Cr, Ni, Cu and S). High purity BeO from Brush Beryllium Company was added to the melts which were used in studies of oxide saturated melts.

Standard Reagents

Reagent grade KOH was used in the preparation of 0.1 N base which was standardized with potassium acid phthalate. Karl Fischer Reagent from Fisher Chemical Company (So-K-3) was standardized by direct addition of weighed aliquots of water.

Apparatus

Experiments were carried out by means of a transpiration method. The stepwise sequence of processes was as follows:

Control of flow of H_2 carrier gas

↓

Addition of HF(g), H_2O(g), or both → Equilibration with melt

↘ ↙

Removal of HF or H_2O in standardized reagent

↓

Measurement of volume of H_2 carrier gas

A complete flow diagram of the apparatus is shown in Figure 2.

<u>Flow Control Panel</u>

The manifold gas pressures (H_2 and He) were not constant but varied, generally, between 5 and 10 lb gauge. A pressure relief valve (Moore Products Company differential type flow controller, Model 63BD, modified form) was used to reduce this pressure to a constant value of 3.0 lb gauge. The gas next passed through a Fisher-Porter Rotameter which was used only as a visual check of the flow rate. The pressure was measured with a 4-in. Ashcroft gauge graduated in 1/4-lb divisions from 0 to 15 lb above atmospheric pressure. The brass needle valve with micrometer handle was from Nuclear Products Company as was the 1/3-lb check valve which was used to

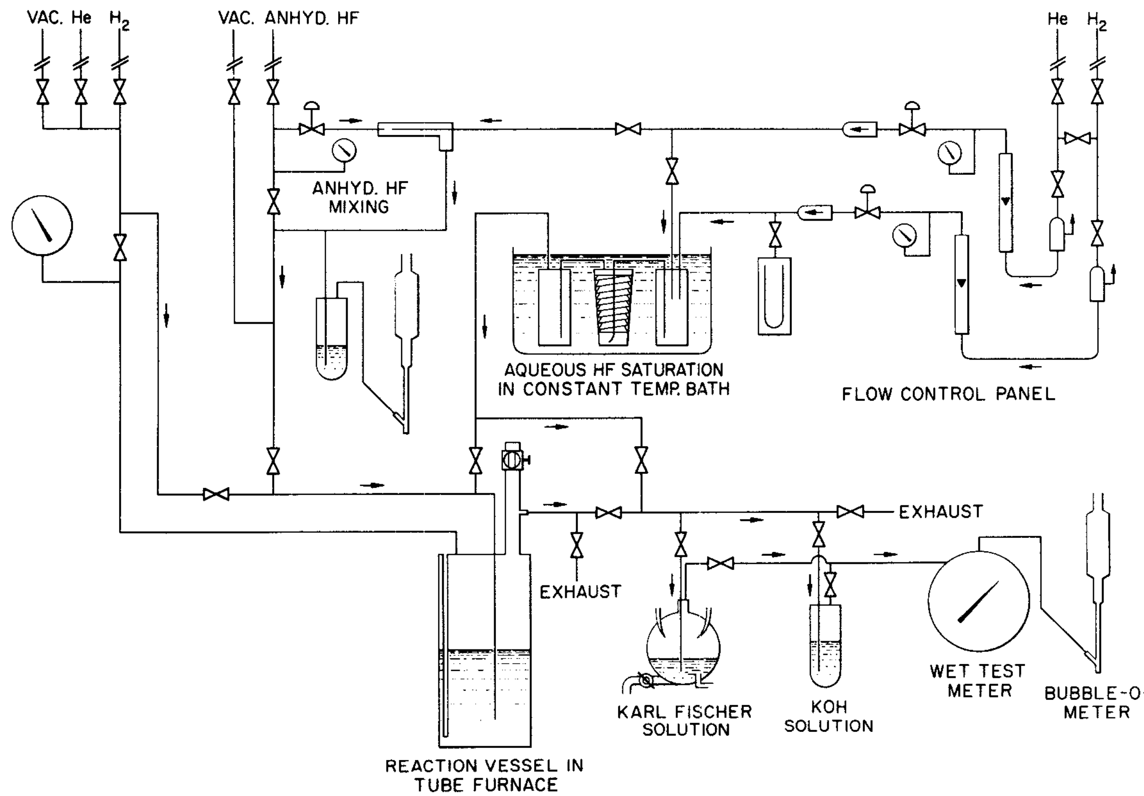

Fig. 2. Complete Flow-Diagram for Apparatus.

prevent back flow of gas. The H_2 line to the aqueous HF saturator was also connected to a water filled manometer from which the pressure immediately upstream of the saturator could be determined. The various pieces on the panel were connected by 1/4-in. Cu tubing using mechanical Swagelok fittings.

An alternate flow control system, identical with the above except for the H_2O manometer, was used to control the carrier gas for the anhydrous HF system.

Aqueous HF Saturator

When HF and H_2O were both desired in the influent gas, aqueous solutions of HF were employed. The H_2 from the flow control panel passed first through a dry bottle, next the filled saturator, and then another dry bottle which acted as a trap for liquid droplets. Both the bottles and the saturator were machined from Teflon bar stock. The two bottles were $2\frac{1}{2}$-in. diameter with a capacity of 300 ml. The saturator was a two-piece spiral gas-washing bottle (3-in. outside diameter top, 2-in. bottom) with approximately the same volume as the dry bottles. Lids for the three were screw-type with 45° shoulders which made pressure seals. The tube openings were threaded for compression-type nuts which sealed the tubes in place when tightened. The connecting line from the flow control panel and between the bottles was of 1/4-in. Teflon tubing.

The saturator and dry bottles were in a constant temperature bath, the temperature of which was used to control the partial pressures of HF and H_2O. The temperature could be controlled $\pm$ $0.1^{\circ}C$ from 15 to $25^{\circ}C$. The bath was continuously cooled by a water coil while heating was controlled by a mercury-mercury contact thermostat and an electronic relay.

Anhydrous HF Mixing System

For studies requiring anhydrous HF(g), controlled gas mixing was employed. The manifold HF pressure was controlled by regulating the temperature of the HF supply cylinder. The flow of HF was controlled by a small-orifice Monel diaphragm valve. The HF flowed from the valve through a 6-mil capillary to the mixing chamber, a 1/4-in. Cu tube about 3 in. long. The HF entered at one end of the mixing chamber; the H_2 entered at the other end through a 1/8-in. Cu tube which extended the length of the chamber to the point where the HF capillary ended. The mixed gases flowed out through a sidearm. This system was adapted from one designed by G. Long which was suitable for HF pressures from about 3×10^{-2} atm downward.[43]

Reaction Vessel

The melts were contained in cylindrical vessels (2-in. diam, 15-in. long; or 4-in. diam, 15-in. long) constructed of Grade A nickel. Each vessel was equipped with the following: a thermocouple well and a gas-inlet tube (1/4-in. tubing) each of which reached to within 1/2 in. of the bottom of the vessel; a cover gas line (1/4-in. tubing) with an attached 6-in. Ashcroft Duragauge graduated from 30-in. vac to 30-lb gauge; and a sampling port (a 3/8-in. pipe with the end closed by a 1/2-in. Worcester ball valve). A sidearm on the sampling port of 1/4-in. tubing served as the gas-outlet line.

The sampling port could be connected to either a sampling apparatus or an addition apparatus. These apparatus (developed and in common use in molten salt studies at ORNL[44]) allow removal of samples from the melt in Cu filter sticks with subsequent transfer to a dry box under He atmosphere, and addition of weighed salt samples to the melt under a dry He atmosphere.

The reaction vessel was located inside an upright tube furnace, the temperature of which was controlled by an L & N Speedomax H Temperature Controller with Model S Recorder and a Chromel-Alumel thermocouple. The temperature was checked periodically with a calibrated Pt vs Pt-10% Rh thermocouple and an L & N K-2 Potentiometer.

The gas lines from the saturator and the HF mixing chamber to the reaction vessel and from the vessel to the titration assembly were of 1/4-in. Cu tubing. Monel sealed-diaphragm valves were connected by means of silver-soldered joints. Both the lines and valves were heated to about 100°C with heating tape as precaution against condensation or surface adsorption of the HF or H_2O.

Titration Assembly

The Karl Fischer titration vessel was a 300-ml round-bottom flask with the following additions: two Pt wires fused through the bottom of the flask about 3/4 in. apart; a sidearm added to the bottom to allow drainage of the flask; two buret tips (for 5-ml Koch microburets with 150-ml reservoir) fused through the top of the vessel about 3 in. apart. The solution was stirred by a small bar magnet under the Pt wires. Teflon gas-inlet and gas-outlet lines were fitted through a rubber stopper.

The endpoint was determined by an amperometric method using the "dead-stop" technique.[45] The indicator circuit consisted of the following: 1.5-v dc source, a 10^4 ohm variable resistor, a 500 ohm variable resistor, the Pt electrodes, and a microammeter (all connected in series). With excess Karl Fischer Reagent present the variable resistors were adjusted so that a current of more than 100 μa was observed. With excess water present the current was less than 2 μa. The endpoint was taken as the point at which the current was 50 μa.

The KOH titration vessel was a 175-ml test tube. Teflon gas-inlet and gas-outlet lines, and the tip of a 10-ml Lab-Crest microburet were fitted through a rubber stopper. Phenolphthalein indicator was used.

Gas Volume Measurement

The carrier gas from the titration assembly was bubbled through a bottle containing KOH solution (to remove acidic vapors from the Karl Fischer Apparatus) and then through a bottle containing H_2O [to saturate with $H_2O(g)$ at room temperature] before its volume was measured in a Precision Wet Test Meter. The gas-exit line from the Wet Test Meter was connected to a Bubble-O-Meter which was used to measure flow rates.

Procedure

Measurements

The primary experimental quantities needed were the partial pressures of HF and H_2O over the melt.

Titrations.--Measurements for determination of partial pressures were made as follows:

(1) A measured volume of standardized reagent was added to a titration vessel.

(2) The time required for the flowing gas to neutralize the reagent was determined.

(3) The flow rate of gas was determined at room temperature by timing a soap bubble for 100 ml. Since diffusion of H_2 through the bubble

would lead to lower indicated flow rates than the true flow rates, at least one bubble before and one bubble after the one being timed were employed to help decrease diffusion of H_2.

(4) Usually, at least three successive titrations were carried out with a given reagent and then the gas flow was switched to the other titration vessel for a similar number of titrations.

(5) The initial volume for a series of titrations was recorded from the wet-test meter, which indicated continuously the total gas volume which had passed through the reaction vessel during an experiment. The meter could be read only to the nearest 10 ml, which was a precise enough reading for total volume but not for individual titrations. The gas volumes titrated were calculated from the flow rate and time of titration.

(6) The temperature of the wet-test meter, which was the temperature of the gas as it was being measured, was recorded.

Calculations.--The required calculations to evaluate the partial pressures were carried out as follows:

(1) The measured volume of gas was calculated by

(Time of titration)/(Time per 100 ml) x 100 = ml of gas.

(2) The number of millimoles of the component removed from the gas stream by the titration was calculated by

(ml reagent)(conc reagent) = millimoles component (HF or H_2O).

(3) The partial pressure of the component was calculated from ideal gas law by

$$P_{component} = (\text{millimoles component})(0.08206)(\text{temp wet-test meter})/(\text{ml gas}) .$$

Limitations.--The apparatus as employed did pose some limitations on the procedure. The aqueous HF saturator as designed could not accommodate flow rates greater than 200 ml/min without bumping. For consistency, comparable flow rates were used with the anhydrous HF system.

Under operating conditions both the Karl Fischer titration vessel and the KOH titration vessel were checked to verify that they were able to remove quantitatively the H_2O and HF, respectively, from the flowing gas stream. In both instances two vessels were set up in series and the leak-through from the first determined the second. The amounts of HF and H_2O titrated in the downstream vessels were less than 0.1% of the amounts measured in the upstream vessels.

The Karl Fischer titration vessel was also checked against in-leakage of moisture from the atmosphere. The amount of Karl Fischer Reagent needed to maintain the endpoint with no gas flowing through the vessel was about 0.07 ml/hr. This correction was applied to pressures below 1×10^{-3} atm when it altered the observed results by as much as 1%.

Since the titration vessels were operated at room temperature, in order to avoid condensation, the max pressures of HF and H_2O were limited to the vapor pressures above aqueous HF solutions at the same temperature.[40]

Even for pressures below these limits some surface adsorption was encountered. When the flowing gas stream was switched through the Karl Fischer apparatus, the P_{H_2O} determined for the first few tenths of a liter would be lower than the following determination. Therefore, the first titration of about 0.3 ml Karl Fischer Reagent was not recorded after the switch was made. Dehydration of the Teflon was observed after the flowing gas was stopped. Additional Karl Fischer Reagent was necessary to maintain the endpoint. (This was much in excess of the amount due to in-leakage discussed above and due to the untitrated gaseous water in the dip-line.) The same problem was not observed with P_{HF} measurements, but usually a small aliquot of KOH was added to vessel during switching to allow stabilization of flow before titrations were begun.

Systematic Errors

The preceding method of calculating partial pressures included the assumption that the pressure in the apparatus from the saturator to the wet-test meter was constant and that the amount of material was the same at every stage. The assumption was also made that the measured volume was equal to the volume of carrier gas plus reactive components, HF and H_2O. These assumptions were not completely valid, but it will be shown in the following sections that the resulting systematic errors were small.

The following notation will be used in the discussion of systematic errors:

T_m = temperature of melt, $^{\circ}K$

T_w = temperature of wet-test meter, $^{\circ}K$

P_w = total pressure at wet-test meter

p_w = vapor pressure of water at T_w

x and y = partial pressures of HF and H_2O, respectively, at titrator

n_{HF} and n_{H_2O} = moles of HF and H_2O as measured at T_w, P_w

V_w = measured volume on wet-test meter

V_a = volume of dry carrier gas going through system measured at T_w, P_w

V_b = volume of gas entering titration assembly at T_w, P_w.

Measured Volume.--The measured gas volume, V_w, was of the carrier gas which had been saturated with water at T_w, immediately prior to entering the wet-test meter. The volume which should have been used in the pressure calculations was V_b. The volume of carrier gas corresponding to the measured volume is given by

$$V_a = V_w(P_w - P_w)/P_w \quad .$$

The volume of HF and H_2O entering the titration assembly may be calculated from the gas laws to be

$$(n_{H_2O}RT_w)/P_w + (n_{HF}RT_w)/P_w .$$

(The effect of any non-ideality of HF or H_2O at the low partial pressures involved here is expected to be negligible.)

From Dalton's law of partial pressures

$$\frac{y}{(n_{H_2O}RT_w)/P_w} = \frac{x}{(n_{HF}RT_w)/P_w} = \frac{P_w - (x + y)}{V_a} \quad .$$

Substituting the value of V_a from the above equation,

$$y = \frac{[P_w - (x + y)]\ P_w n_{H_2O}RT_w}{V_w(P_w - p_w)P_w} \quad ,$$

note that P_w cancels, and by a parallel method

$$x = \frac{[P_w - (x + y)]\ n_{HF}RT_w}{V_w(P_w - p_w)} \quad .$$

According to these calculations, the x and y values calculated in the preceding section should be multiplied by the factor f, where

$$f = \frac{P_w - (x + y)}{P_w - p_w} \quad .$$

The max and min values for f can be estimated. P_w = atmospheric pressure; $(x + y)$ varies from 0 to 0.033 atm, but generally is between 0.013 and

0.020 atm; p_w is between 0.029 and 0.035 depending on T_w. The maximum value for f would be 1.00/0.965 = 1.037. The minimum value for f would be 0.967/0.971 = 0.996. For the majority of measurements,

$$0.980/0.970 \leq f \leq 0.987/0.970 \; ; \quad 1.013 \leq f \leq 1.0196 \; .$$

Thus the calculated partial pressures are too low by 1.0 to 2.0% due to the removal of HF and H_2O and subsequent saturation with H_2O vapor before the volume of carrier was measured.

Another effect, which exerts a minor influence on pressure measurements, is the pressure drop, between the gas space above the melt and atmospheric pressure, required to maintain bubbling through the titrator and wet-test meter. This drop was less than 0.01 atm, and would cause the measured values to be low by as much as 1.0%.

Influent Pressure.--The influent partial pressures were measured with the gas bypassing the reaction vessel. The influent partial pressures are controlled by the saturator temperature and are independent of the total pressure. The total pressure in the saturator was less during the measurements of the influent gas composition than when the gas was passing through the melt by the pressure drop required to maintain bubbling, which was about 0.015 atm. This pressure drop results in an expansion of the gas as it enters the reaction vessel, hence the actual influent partial pressures of HF and H_2O are less than the measured influent partial pressures by 1.5%.

Hydrogen Diffusion.--According to published diffusion coefficients[46] the diffusion of H_2 out of the Ni vessel could be a few milliliters per minute at the elevated temperature of the vessel. Only the reaction vessel was sufficiently hot to allow diffusion of hydrogen.

The rate of diffusion was measured experimentally by pressurizing an empty vessel to 5 lb gauge and following the decrease in pressure as a function of time. The pressures were measured down to 10 in. vacuum. A control experiment with He was used to check against leakage. The following rates of diffusion (expressed as the volume of gas per second which would be measured at the wet-test meter; i.e. at T_w and P_w) were determined: 700°C, 0.035 ml/sec; 650°C, 0.025 ml/sec; 600°C, 0.015 ml/sec. The rate of diffusion of H_2 out of a vessel containing salt should be no greater and probably not much less than for an empty vessel.

The influent partial pressures from the aqueous saturator were determined without loss of H_2 by diffusion since the gas bypassed the vessel. If the influent gas were passed through a vessel with no salt present, the measured partial pressures would be greater due to this effect, by an amount dependent on the temperature and flow rate.

Most experiments were conducted with flow rates between 2.0 and 2.5 ml/sec. This means the apparent influent pressures would be from 1.4 to 1.75% higher at 700°C; 1.0 to 1.25% higher at 650°C; and 0.60 to 0.75% higher at 600°C than the measured influent pressures. The difference would be less at lower temperatures.

Hydrogen diffusion did not play the same role when the anhydrous HF supply system was used. The flow of HF through the control valve was independent of the flow rate of H_2, hence the amount of HF was determined as a function of time and the influent P_{HF} was calculated from this value and the effluent flow rate.

The effect of diffusion of H_2 out of the reaction vessel need not have been as great as indicated above. The mixing of the effluent gas, primarily through turbulence, could have permitted re-equilibration of the gas with the melt. This would reduce the error caused by loss of hydrogen from the system by diffusion.

Dead-volume.--The preceding errors reflect the variations in pressure and mass of flowing gas. There is one additional error in the pressure determination at a given volume. This is the lag in measured pressures because of the volume of the gas space above the melt. The following additional notation will be used:

T_m = temperature of gas above the melt, $^{\circ}K$

V_m = volume of gas space above the melt

P_1 = partial pressure of HF or H_2O leaving the melt

P_2 = measured partial pressure

dV = increment of gas flowing through system as measured at T_w, P_w .

From the mixing process,

moles of substance entering $V_m = (P_1 dV)/(RT_w)$

moles of substance leaving $V_m = (P_2 dV)/(RT_w)$

change of moles in $V_m = (V_m dP_2)/(RT_m)$.

Thus,

$$(V_m dP_2)/(RT_m) = (P_1 dV)/(RT_w) - (P_2 dV)/(RT_w)$$

and rearranging,

$$P_1 = V_m(T_w/T_m)(dP_2/dV) + P_2 \quad .$$

For most experiments, $V_m = 0.400$ liters and $T_w = 300^{\circ}K$. When $T_m = 773^{\circ}K$, $(T_w/T_m) = (300/773) = 0.388$ and when $T_m = 973^{\circ}K$, $(T_w/T_m) = (300/973) = 0.308$.

Therefore, at $500^{\circ}C$

$$P_1 = 0.1552(dP_2/dV) + P_2$$

and at 700°C

$P_1 = 0.1232(dP_2/dV) + P_2$.

The most dramatic change in P_2 occurred in the experiments when water was introduced into melts which contained very little oxide. The maximum rate of pressure change noted was 0.01 atm/liter. Therefore,

$P_1 = 1.55 \times 10^{-3} + P_2$.

This large change occurred only during the first liter and only in the HF measurements. During this time P_2 increased to about 1×10^{-2} atm so that the error in these unusual circumstances was only about 10%. At all other times the change was less than 1×10^{-4} atm/liter and often less than 1×10^{-5} atm/liter. For all of these conditions the error would be much smaller than 1.0%.

This treatment assumes no further equilibration after the bubble leaves the melt. If any did occur then application of the above equation would be an overcorrection.

Summary.--The combined effect of these systematic errors on the overall accuracy of the results will now be considered.

Due to the measured-volume error all observed partial pressures were 1.0 to 2.0% low. The other errors do not affect the observed partial pressures but do affect the material balance relationships used in treating the data.

The calculated values of Q (i.e., x^2/y) are 1.0 to 2.0% low for all cases. This range is within the experimental scatter of the data. No bias is introduced in calculation of activities because of the extrapolation to pure BeF_2. The extrapolated thermodynamic values for $BeF_2(l)$ are well within the precision of literature data. No comparison of influent and effluent pressures is required and diffusion of H_2 should not influence results.

The expected effects on Q_A, $(x/y)[OH^-]$, are within the scatter of the data. The quantity, (x/y), will not be affected by any of the errors. The hydroxide concentration is based on proton material balance between influent and effluent gas. Thus, due to the error in measured influent pressures, the hydroxide concentration should be decreased by 1.5%. Due to the error caused by diffusion of hydrogen, the hydroxide concentration should be increased by 0.5 to 1.75% depending on temperature and flow rate. Since the hydroxide concentration is proportional to Q_A, the calculated values for Q_A could range from 1.0% high to 0.25% low. This range is well within the indicated scatter of data, with Q_A generally known only to ± 10%.

For unsaturated melts, Q_O is based on the difference between oxygen material balance and proton material balance. Q_O is subject to the same corrections as Q_A, hence it is from 1.0% high to 0.25% low. The quantities Q_B and Q_C are derived from Q_A and Q, hence they are in error in the same manner.

Random Errors

In order to give a satisfactory estimate of the expected precision of the experimental measurements, the various random errors and their probable magnitudes are considered.

Melt Composition.--The melts were prepared by adding weighed samples of BeF_2 and LiF to the reaction vessel. The precision of weighing and transferring samples to the vessel was about ± 0.2%. The weight per cent of Li, Be and F was determined chemically on filtered samples. The variation in analyses of successive samples from the same melt was generally ± 4% for each constituent, which was clearly greater than the uncertainty based on weights of components. Since both LiF and BeF_2 are hygroscopic, small amounts of water were included in the weighed samples. Determination of moisture content of typical samples by the thermogravimetric method indicated 0.1 and 0.4% moisture, respectively. Thus, on the basis of the above uncertainties in weight and moisture content, the melt compositions, expressed as mole fraction BeF_2, were probably known within ± 0.5%.

The amounts of the impurities Cr, Cu, Fe, Ni and S were determined in the starting materials, in filtered samples of the melt after it had been sparged with H_2 for more than 48 hr, and again immediately after sparging for 6 hr with a dilute mixture of HF and H_2O in H_2. All analyses were within the following range: Cr always less than 20 ppm, Cu always less than 100 ppm, Fe usually between 100 and 200 ppm, Ni usually less than 20 ppm, and S always less than 5 ppm. Total measured impurities only amount to about 350 ppm which is not significant in terms of melt composition.

The impurities could, however, cause some uncertainty in the partial pressure measurements if the impurities reacted to liberate or consume HF or H_2O during an experiment. Consumption of HF would have occurred if sulfide had been present in concentrations greater than 5 ppm by the reaction of S^{2-} with HF in a manner analogous to the reaction of oxide with HF.[47] Since the impurity levels were not appreciably decreased during the H_2 sparging, the impurities apparently were not reduced to the metallic state. The results of Blood's study on the stability of the difluorides of nickel, iron and chromium[21] indicate they should eventually be reduced. However, most of the reactions he studied required several days for equilibration. His results indicate that: the Ni reaction vessel would not react with HF to produce NiF_2 concentrations greater than 0.1 ppm with the highest $(P_{HF})^2/P_{H_2}$ ratios employed here; the Cr would have been in the CrF_2 state for all $(P_{HF})^2/P_{H_2}$ ratios above 1×10^{-8} (true at all times except for extended stripping with H_2); the FeF_2 concentration could have varied between the time when H_2 was used and when $(P_{HF})^2/P_{H_2} = 4 \times 10^{-4}$ atm.

The Fe probably did not react rapidly enough to interfere (or the analyses were in error) as indicated by the following experiment. Anhydrous HF-H_2 mixtures (approximately 0.01 atm HF) were employed to remove oxide and hydroxide from melt. After the melt was essentially free of oxide, the effluent P_{HF} equaled the influent P_{HF}. The HF was then stopped and H_2 continued. The total amount of HF stripped out was about 5 ppm and was probably due to HF solubility. If the Fe had been appreciably reduced, more HF would have been detectable in the stripping gas.

Melt Temperature.--The temperature of the melt varied about $\pm$ 0.5° during the furnace heating and cooling cycle. The observed temperature with the thermocouple at different depths in the melt did not vary more than $\pm$ 1.0°. Overall, the temperature of a melt was known and constant to $\pm$ 1.5°. One exception to the low temperature variation with dept occurred when large amounts of solid LiF were in the melt. In this case the temperature read about 15 to 20° lower at complete insertion of the thermocouple than when the thermocouple was withdrawn about 1 in. from the bottom. Even in this case the temperature varied no more than $\pm$ 2° when the thermocouple was withdrawn further. Apparently the solid LiF settled to the bottom and was not stirred by the bubbling, and as a result, loss of heat out of the end of the vessel was faster than the transfer of heat from the liquid to unstirred solid.

Titer Precision.--A 25-liter reservoir of KOH was standardized with potassium acid phthalate before initial use and again approximately 3 and 6 months later. All values agreed with the average value ± 0.1%.

The 150-ml reservoir of the Karl Fischer apparatus was filled as needed from quart bottles of Karl Fischer Reagent. The reagent was standardized each time the reservoir was filled and frequently thereafter. Water (0.050 to 0.080 g) was added directly to the titration vessel from a weighing buret and the Karl Fischer titer determined. Successive titrations agreed within ± 0.3%. Usually the concentration change from one standardization to the next would not exceed 1%. The precision in measuring the concentration of Karl Fischer Reagent was ± 0.5%.

The volumes of both KOH and Karl Fischer Reagent used in standardizations were large enough so that the uncertainties in reading the burets could be ignored. However, the much smaller volumes used during experimental titrations were known only to ± 1.0%.

Wet-test Meter Temperature.--The wet-test meter temperature, which was the temperature of the gas as its volume was being measured, varied no more than ± 0.5° during a given experiment. Due to the variation in temperature, the variation in measured volume is about ± 0.2%, including the effect due to the change in vapor pressure of water.

Endpoint Precision.--The precision in the timing of endpoints was constant at ± 0.5% of the measured value.

Flow-rate Precision.--The precision in the timing of flow rates was 0.1 second per 100 ml. For a typical value of 50 seconds per 100 ml, this would amount to 0.2%.

Statistical Error Analysis

The probable error in the calculated quantities can be determined from the magnitude of random errors in the various measurements. If the contribution of each observable, q_i, to the calculated function, Q, is known and expressed as

$$Q = f(q_1^a, q_2^b, \ldots\ldots, q_n^m) \quad ,$$

and $(\Delta Q/Q) \times 100$ is % probable error in Q and $(\Delta q_i/q_i) \times 100$ is % probable error in q_i, the contribution of errors can be calculated. If the errors follow a normal distribution, it can be shown[48] that the following equation allows for partial cancellation of errors of opposite sign:

$$(\Delta Q/Q)^2 = a^2(\Delta q_1/q_1)^2 + b^2(\Delta q_2/q_2)^2 + \ldots\ldots + m^2(\Delta q_n/q_n)^2 \quad .$$

Evaluating the above equation numerically for each observable and taking the square root of both sides will give the probable error in the calculated quantity.

Volume of Gas Titrated.--

ml of gas = f(time of titration, time of flow measurement)

$(\Delta \text{ml gas}/\text{ml gas})^2 = (0.005)^2 + (0.002)^2 = 29 \times 10^{-6}$

% probable error in vol of gas = 0.54%.

Millimoles H_2O Titrated by Aliquot.--

millimoles H_2O = f(conc Karl Fischer Reagent, vol Karl Fischer Reagent)

$(\Delta \text{millimoles } H_2O/\text{millimoles } H_2O)^2 = (0.005)^2 + (0.010)^2 = 125 \times 10^{-6}$

% probable error in millimoles H_2O = 1.11%.

Millimoles HF Titrated by Aliquot.--

millimoles HF = f(conc KOH, vol KOH)

$(\Delta \text{millimoles HF}/\text{millimoles HF})^2 = (0.001)^2 + (0.010)^2 = 101 \times 10^{-6}$

% probable error in millimoles HF = 1.00%.

Pressure H_2O.---

P_{H_2O} = f(vol gas, millimoles H_2O, wet-test meter temp)

$(\Delta P_{H_2O}/P_{H_2O})^2 = (0.0054)^2 + (0.0111)^2 + (0.002)^2 = 154 \times 10^{-6}$

% probable error in pressure H_2O = 1.24%.

Pressure HF.---

P_{HF} = f(vol gas, millimoles HF, wet-test meter temp)

$(\Delta P_{HF}/P_{HF})^2 = (0.0054)^2 + (0.010)^2 + (0.002)^2 = 133 \times 10^{-6}$

% probable error in P_{HF} = 1.15%.

Equilibrium Quotients.---The exact relationship of Q, Q_A and Q_O to all of the variables cannot be expressed in the proper form to calculate the expected uncertainty. However, use of only the dependence on the pressure measurements would lead to 2.5, 4.0 and 4.7%, respectively, as the most precise possible determinations. Both Q_A and Q_O depend on integral relationships between influent and effluent gas which increase the uncertainty still further.

III. RESULTS

Tabulation

Saturated Melts

Appendix A contains the data from each experiment performed on a saturated melt. The experiments are arranged according to a three digit "Run No." which indicates chronological order. Here, and throughout the text, numbers preceding "BeF_2" denote mole fraction. In the 100 series the first experiments (101-123) were run in a 2-in.-diam vessel on 305 g of 0.316 BeF_2 and 6 g BeO, whereas, for the later experiments (125-129), enough BeF_2 was added to give 0.333 BeF_2.

In the 200 series a 4-in.-diam vessel was charged with 1522 g of 0.80 BeF_2 and 10 g BeO for the first experiments (201-213). After this composition was studied, enough LiF was added to give 0.70 (215-223), 0.60 (225-233), 0.50 (235-241), 0.40 (245-255) and 0.333 BeF_2 (257-273).

In the 400 series the first experiments (401-409) were run on 0.30 BeF_2 obtained by adding 44 g LiF and 3.0 g BeO to the melt used in the 300 series. For the later experiments (413-423) 87 g additional LiF was added to give 0.25 BeF_2.

The parameters listed in Appendix A are:

w = wt of melt, kg

T = temperature of wet-test meter, ^oK

a = influent P_{HF}, atm x 10^3

c = influent P_{H_2O}, atm x 10^3 .

The effluent partial pressures, P_{HF} = x and P_{H_2O} = y, are tabulated (atm x 10^3) with the corresponding initial and final values of W. Note that the use of W allows consistent comparison of experiments independent of the weight of melt or temperature of gas-measurement.

Unsaturated Melts

Appendix B contains the data from each experiment performed on an unsaturated melt. These experiments are also arranged according to a three digit "Run No." In the 300 series a 2-in.-diam vessel was charged with 500 g of 0.333 BeF_2 and anhydrous HF was used to remove oxide from the melt. Measurements then were made while an aqueous-HF gas stream was being passed through the melt.

In the 500 series the first experiments (501-527) were run in a 2-in.-diam vessel containing 500 g of 0.333 BeF_2. For the later experiments (529-539) enough LiF was added to give 0.273 BeF_2. Measurements were made both while anhydrous HF was being used to remove oxide from solution and while an aqueous-HF gas stream was being used to add oxide to solution.

In the 600 series a 2-in.-diam vessel was charged with 424 g of 0.60 BeF_2 for the first experiments (601-615). For the later experiments (617-627) LiF was added to give 567 g of 0.40 BeF_2. Measurements were made during both the removal and addition of oxide.

The format of Appendix B is identical to that of Appendix A except that influent pressures are reported as $P_{HF} = a + bW$ and $P_{H_2O} = c + dW$ when gas-mixing method was employed.

Determination of Equilibrium Quotients

The experimental data were correlated according to the model presented in Chapter I. Computer methods were developed for these correlations.

Saturated Melts

The experiments fall into the following three groups:

Case I. Measurements were made while both P_{HF} and P_{H_2O} were still changing but were stopped before steady state was reached.

Case II. Partial pressure measurements were made only after P_{HF} and P_{H_2O} had reached steady state.

Case III. Measurements were made in both the changing- and steady-state regions.

The program was designed to handle all three cases with only Case I requiring additional input information. In all cases, since x and y were determined alternately, direct calculation of Q would require interpolation, which was not very satisfactory where x and y were volume dependent.

The procedure of solution is outlined in the following steps:

1. For Case I, Q was specified as part of input information. For Cases II and III, Q was not specified; the computer started with the largest value of W and compiled average x and y as it moved to successively lower values of W until the partial pressures became lower than the average to that point by a specified amount (usually 4%). Safeguards were build in to prevent the computer prematurely terminating this process because of unusual scatter.
2. Using all of the partial pressures selected above, the average value of x and the average value of y were calculated and their standard deviations were expressed at the 99% confidence level. The best Q was calculated from these averaged partial pressures and its standard deviation was calculated from the deviation of its component parts.

3. Using the Q calculated (or Q provided if Case I), each observed y was converted to a corresponding x using the relationship $x = (Qy)^{\frac{1}{2}}$, for the initial region not used in the determination of Q.

4. Next, for each x (observed or calculated from observed y) the quantity

$$\log \frac{(4/Q)x + 1 + [1 + (8A/Q)]^{\frac{1}{2}}}{(4/Q)x + 1 - [1 + (8A/Q)]^{\frac{1}{2}}} \equiv f(W)$$

(this relationship was derived in Chapter I) was determined and tabulated along with the midpoint, W_a, for each observation.

5. These f(W) were now correlated according to the equation

$$f(W) = \frac{[1 + (8A/Q)]^{\frac{1}{2}} Q}{2.303} \ (1/Q_A)W + \text{constant}$$

to obtain the best value of Q_A. The standard deviation in Q_A was calculated by normal method for slope in a straight line correlation.

6. At this point a consistency check was made - the deviations from the best fit were determined to see if the observed values of x were consistently on one side of the line and values of x calculated from observed values of y on the other side. If a difference had occurred, Q could be readjusted or Q_A determined from x and y separately.

Steps 1 and 2 complete the calculation for Case III. Steps 1 through 6 complete the calculation for each experiment.

Several examples of the various cases are given in Figures 3, 4, and 5. An example of Case I is given in Figure 3a, with the observed values of x and y indicated by short bars. The line is generated by using the values for Q and Q_A determined by the correlation. Figure 3b is a plot of f(W) vs W, indicating the linearity of the relationship. Figure 4a gives an example of Case III. The flat portion was used to determine Q. Q_A was determined from the initial region by holding Q constant. A plot of f(W) vs W is given in Figure 4b for the applicable region of the above run. Another example of Case I is given in Figure 5a showing the decrease in x and y when influent HF and H_2O were stopped and hydrogen sparging continued. Figure 5b gives a plot of f(W) vs W showing that the function also holds when influent pressures are zero.

The values obtained for Q and Q_A from all of the saturated-melt experiments are presented in Table 3. The values are arranged according to composition and temperature of melt. Run numbers are provided for cross reference with original data in Appendix A.

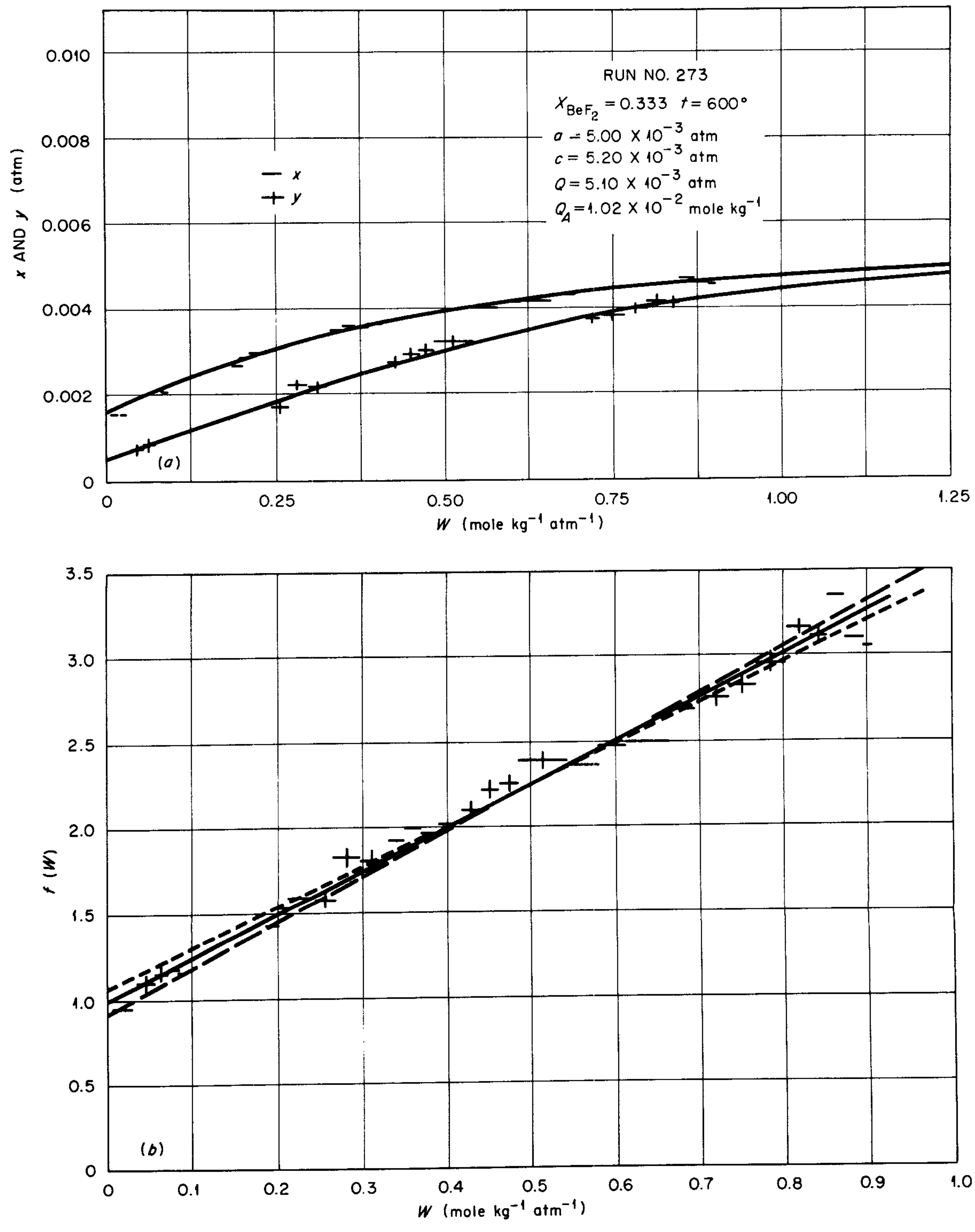

Fig. 3. BeO-saturated 0.333 BeF_2 Showing (a) Calculated and Observed Partial Pressures, and (b) Linear Correlation of Pressures.

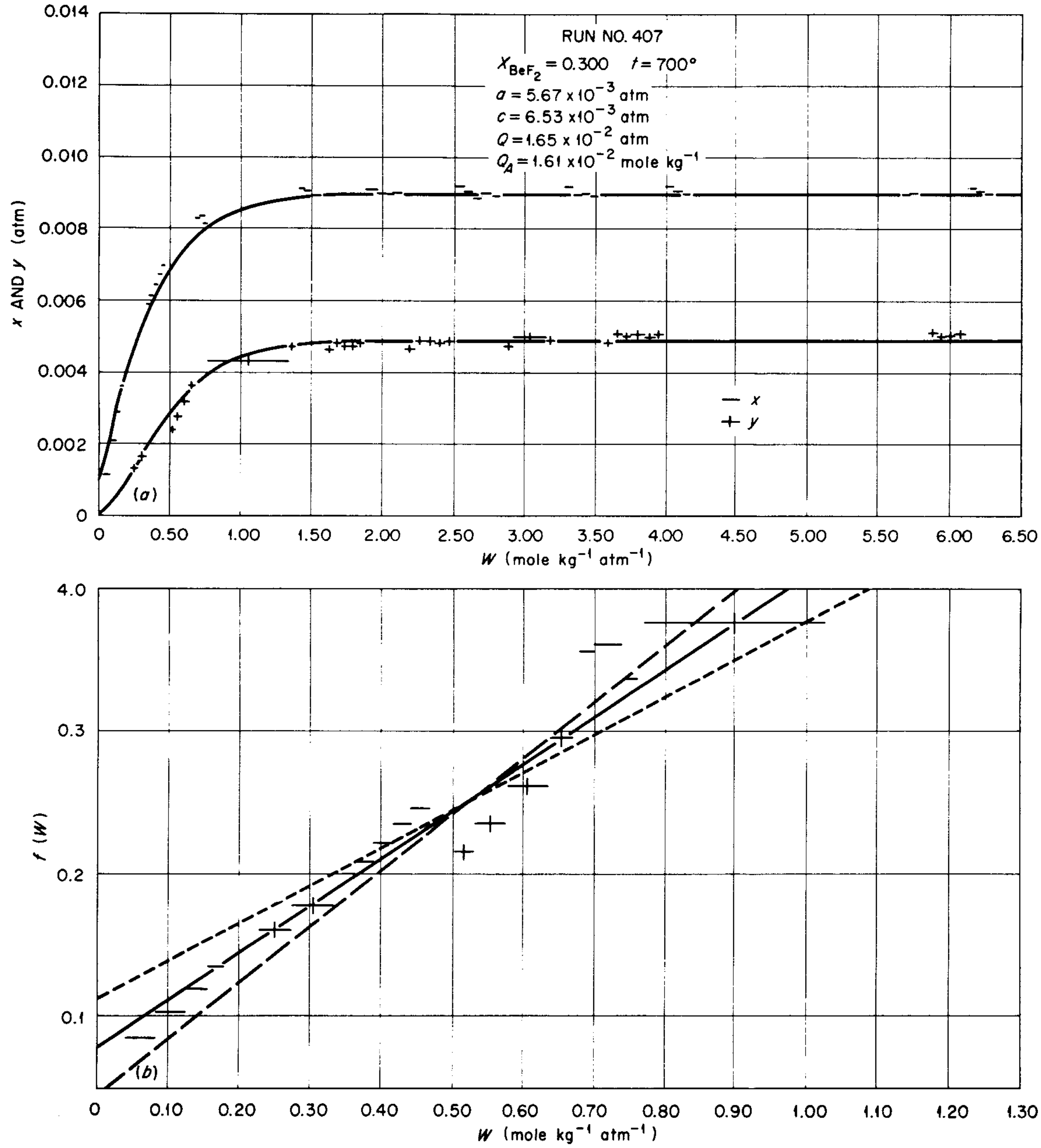

Fig. 4. BeO-saturated 0.300 BeF_2 Showing (a) Calculated and Observed Partial Pressures, and (b) Linear Correlation of Pressures in Applicable Region.

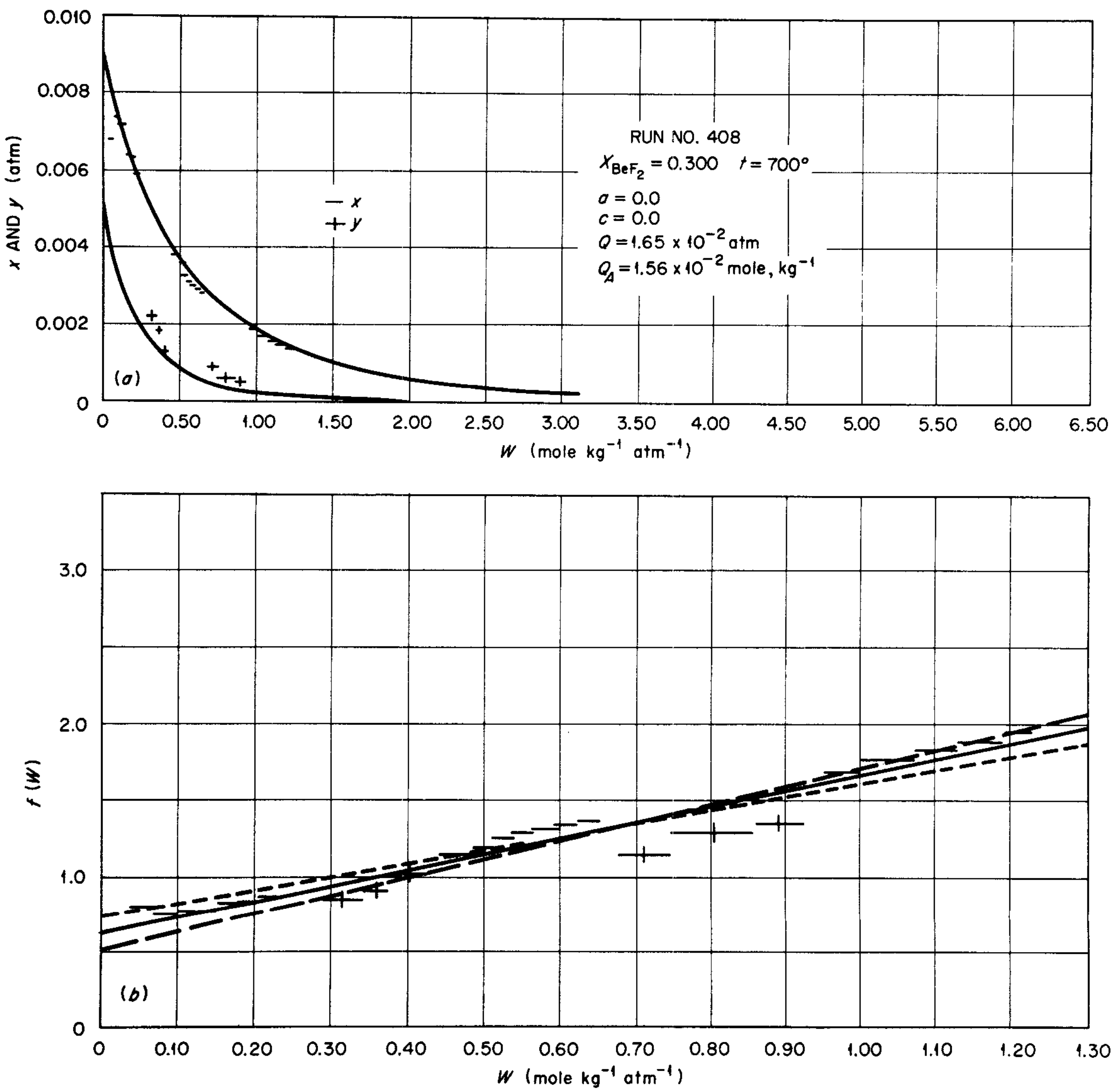

Fig. 5. BeO-saturated 0.300 BeF_2 during H_2 Sparging, Showing (a) Calculated and Observed Partial Pressures, and (b) Linear Correlation of Pressures.

Table 3. Equilibrium Quotients, Q and Q_A, Calculated from Data on Oxide-Saturated Melts

Comp BeF_2	Temp (°C)	Run No.	Q ± σ (atm)	Q_A ± σ (mole/kg)
0.250	520	419[a]	(7.53 ± 0.26) x 10^{-4}	(2.77 ± 0.30) x 10^{-3}
0.250	543	423[a]	(1.16 0.02) x 10^{-3}	(3.94 0.76) x 10^{-3}
0.250	607	413[a]	(2.33 0.05) x 10^{-3}	(6.08 1.10) x 10^{-3}
0.250	655	421	(4.52 0.09) x 10^{-3}	(9.37 3.00) x 10^{-3}
0.250	702	415	(9.29 0.22) x 10^{-3}	(1.14 0.28) x 10^{-2}
0.300	500	409	(4.19 ± 0.07) x 10^{-4}	(3.82 ± 0.29) x 10^{-3}
0.300	540	403	(8.61 0.27) x 10^{-4}	(4.86 0.57) x 10^{-3}
0.300	601	405	(3.15 0.07) x 10^{-3}	(8.34 2.60) x 10^{-3}
0.300	601	406	(3.15[b]) x 10^{-3}	(7.04 0.78) x 10^{-3}
0.300	652	401	(7.98 0.17) x 10^{-3}	(1.35 0.16) x 10^{-2}
0.300	652	402	(7.98[b]) x 10^{-3}	(1.23 0.15) x 10^{-2}
0.300	700	407	(1.65 0.03) x 10^{-2}	(1.61 0.32) x 10^{-2}
0.300	700	408	(1.65[b]) x 10^{-2}	(1.59 0.21) x 10^{-2}
0.316	500	113	(6.44 ± 0.43) x 10^{-4}	
0.316	500	117	(6.03 0.26) x 10^{-4}	
0.316	550	115	(1.66 0.08) x 10^{-3}	
0.316	600	105	(4.61 0.27) x 10^{-3}	
0.316	600	107	(4.07 0.20) x 10^{-3}	
0.316	600	109	(4.09 0.22) x 10^{-3}	
0.316	600	111	(4.69 0.16) x 10^{-3}	
0.316	650	119	(9.94 0.43) x 10^{-3}	
0.316	650	121	(9.29 0.55) x 10^{-3}	
0.316	700	101	(2.18 0.14) x 10^{-2}	
0.316	700	103	(2.47 0.17) x 10^{-2}	
0.316	700	123	(2.13 0.04) x 10^{-2}	
0.333	500	129	(8.16 ± 0.39) x 10^{-4}	
0.333	500	261	(7.30 0.42) x 10^{-4}	
0.333	550	127	(1.96 0.09) x 10^{-3}	
0.333	550	263	(2.39 0.06) x 10^{-3}	
0.333	550	264	(2.30[b]) x 10^{-3}	(6.53 ± 0.75) x 10^{-3}
0.333	600	125	(5.05 0.16) x 10^{-3}	
0.333	600	259	(5.24 0.12) x 10^{-3}	
0.333	600	265	(5.50[b]) x 10^{-3}	(9.40 0.84) x 10^{-3}
0.333	600	269	(5.36 0.20) x 10^{-3}	(1.27 0.17) x 10^{-2}
0.333	600	270	(5.36[b]) x 10^{-3}	(7.85 0.92) x 10^{-3}
0.333	600	273	(5.10[b]) x 10^{-3}	(1.02 0.06) x 10^{-2}
0.333	650	267	(1.33 0.04) x 10^{-2}	
0.333	650	268	(1.30[b]) x 10^{-2}	(1.34 0.08) x 10^{-2}
0.333	700	257	(2.52 0.06) x 10^{-2}	
0.333	700	271	(2.53 0.12) x 10^{-2}	(3.03 0.38) x 10^{-2}
0.333	700	272	(2.53[b]) x 10^{-2}	(1.62 0.17) x 10^{-2}

(continued)

Table 3. (continued)

Comp BeF_2	Temp (°C)	Run No.	$Q \pm \sigma$ (atm)	$Q_A \pm \sigma$(mole/kg)
0.400	500	251	$(2.07 \pm 0.08) \times 10^{-3}$	
0.400	550	252	$(5.76\ \ 0.46) \times 10^{-3}$	
0.400	600	249	$(1.47\ \ 0.04) \times 10^{-3}$	
0.400	650	253	$(3.27\ \ 0.12) \times 10^{-2}$	
0.400	700	245	$(6.15^{b}\ \ ) \times 10^{-2}$	$(3.71 \pm 0.57) \times 10^{-2}$
0.400	700	247	$(5.00\ \ ) \times 10^{-2}$	$(2.37\ \ 0.22) \times 10^{-2}$
0.400	700	255	$(6.15\ \ 0.21) \times 10^{-2}$	
0.400	700	256	$(6.15^{b}\ \ ) \times 10^{-2}$	$(2.43\ \ 0.59) \times 10^{-2}$
0.500	500	239	$(4.80 \pm 0.20) \times 10^{-3}$	$(2.85 \pm 0.21) \times 10^{-2}$
0.500	550	241	$(1.49\ \ 0.05) \times 10^{-2}$	
0.500	600	237	$(3.27\ \ 0.11) \times 10^{-2}$	
0.500	650	244	$(7.00^{b}\ \ ) \times 10^{-2}$	$(2.96\ \ 0.82) \times 10^{-2}$
0.500	700	235	$(1.40\ \ 0.15) \times 10^{-1}$	$(8.50\ \ 1.00) \times 10^{-2}$
0.600	500	229	$(8.07 \pm 0.69) \times 10^{-3}$	$(3.99 \pm 0.40) \times 10^{-2}$
0.600	550	231	$(2.22\ \ 0.12) \times 10^{-2}$	$(8.05\ \ 2.00) \times 10^{-2}$
0.600	550	232	$(2.22^{b}\ \ ) \times 10^{-2}$	$(2.71\ \ 1.20) \times 10^{-2}$
0.600	600	227	$(5.10\ \ 0.43) \times 10^{-2}$	$(7.77\ \ 0.50) \times 10^{-2}$
0.600	650	233	$(1.05\ \ 0.10) \times 10^{-1}$	$(1.14\ \ 0.13) \times 10^{-1}$
0.600	650	234	$(1.05^{b}\ \ ) \times 10^{-1}$	$(8.03\ \ 3.70) \times 10^{-2}$
0.600	700	225	$(2.11\ \ 0.09) \times 10^{-1}$	
0.600	700	226	$(2.05^{b}\ \ ) \times 10^{-1}$	$(1.43\ \ 0.10) \times 10^{-1}$
0.700	550	221	$(2.78 \pm 0.09) \times 10^{-2}$	
0.700	550	222	$(2.78^{b}\ \ ) \times 10^{-2}$	$(3.47 \pm 2.30) \times 10^{-2}$
0.700	600	217	$(5.81\ \ 0.19) \times 10^{-2}$	$(3.30\ \ 1.10) \times 10^{-1}$
0.700	600	218	$(5.81^{b}\ \ ) \times 10^{-2}$	$(6.78\ \ 3.90) \times 10^{-2}$
0.700	650	223	$(1.16\ \ 0.16) \times 10^{-1}$	
0.700	700	215	$(2.15\ \ 0.10) \times 10^{-1}$	$(2.22\ \ 0.21) \times 10^{-1}$
0.800	550	211	$(2.98 \pm 0.11) \times 10^{-2}$	$(1.65 \pm 0.93) \times 10^{-1}$
0.800	600	201	$(6.68\ \ 0.36) \times 10^{-2}$	$(2.56\ \ 1.90) \times 10^{-1}$
0.800	600	209	$(5.91\ \ 0.23) \times 10^{-2}$	$(3.30\ \ 1.50) \times 10^{-1}$
0.800	650	203	$(1.40\ \ 0.18) \times 10^{-1}$	$(5.79\ \ 1.90) \times 10^{-1}$
0.800	650	204	$(1.40^{b}\ \ ) \times 10^{-1}$	$(1.85\ \ 1.30) \times 10^{-1}$
0.800	650	213	$(1.25\ \ 0.05) \times 10^{-1}$	
0.800	700	207	$(2.20\ \ 0.15) \times 10^{-1}$	$(6.15\ \ 2.10) \times 10^{-1}$
0.800	700	208	$(2.35^{b}\ \ ) \times 10^{-1}$	$(2.08\ \ 0.68) \times 10^{-1}$

[a]Solid LiF present, mole fraction BeF_2 actually higher than indicated.

[b]Case I, Q value specified for Q_A determination.

Unsaturated Melts

Determinations of equilibrium quotients involving oxide and hydroxide were based on material balance between influent and effluent gas. The differential equations needed for this correlation were developed in Chapter I. These equations were:

$$dr/dW = (Q_O s/Q_A r)(1 + sQ_A) - [a + c + (b + d)W]$$

and

$$ds/dW = a + 2c + (b + 2d)W - (Q_O s/Q_A r)(1 + 2s/Q_A) \quad .$$

A computer program was developed for the simultaneous solution of these differential equations using the Runga-Cutta method of solution.[49] This calculation required that the adjustable parameters, Q_O, Q_A, r_o, and s_o, and the fixed parameters, a, b, c, and d, be specified. With all of these quantities chosen, the change in r and s with respect to W was calculated and new values estimated for r and s after the increment ΔW. The cycle was then repeated stepwise and the process continued until a value of W larger than a specified limit, W_{max}, was reached. In order to determine r and s precisely without significant accumulative errors the step size, ΔW, had to be restricted so that neither r nor s could change by more than 10% for any step. Values for r and s were tabulated at the various values of W corresponding to the end of each step.

With both r and s known, both x and y could be calculated using the equilibrium relationships. If the relationships

$$Q_A = xs/y \qquad \text{and} \qquad Q_O = x^2 r/y$$

are solved simultaneously,

$$x = Q_O s/Q_A r \qquad \text{and} \qquad y = Q_O s^2/Q_A{}^2 r \quad .$$

These calculated values of x and y form smooth continuous curves although exact equations for the curves could not be expressed. The deviation of the observed x and y values from the calculated values was determined. A general least squares method was used to improve the fit. The partial derivatives of the x and y curves were determined numerically by making slight changes in each adjustable parameter (Q_O, Q_A, r_o, s_o) successively. From the deviations and partial derivatives the adjustments in the parameters needed to minimize the squares of the deviations were determined.

With new values for Q_O, Q_A, r_o, and s_o the new values for x and y were calculated. The least-square subroutine was repeated until the values for Q_O, Q_A, r_o, and s_o were essentially constant as indicated by their being changed less than a preset limit (usually 1%) in successive steps.

The method would not work when the initial guesses were not reasonably close to the correct values. Upper and lower limits were preset for the value of each parameter. If these limits were reached the calculation was terminated and the data resubmitted with different first guesses.

The "best" values for the parameters Q_O, Q_A, r_o, and s_o are reported in Table 4. The indirect method for determining these values did not permit calculation of the standard deviations of these parameters. Since a large number of runs are reported for 0.333 BeF_2, the spread in the Q_O and Q_A values may be used as an indication of the uncertainties with which these are known. The values for r_o and s_o represent concentrations of oxide and hydroxide at the start of an experiment, hence their variations are mainly the result of differing histories of the melts. The runs are arranged according to composition and temperature with run numbers given for cross reference with the original data in Appendix B.

A measure of the goodness of fit is given in Table 5. The mean of the calculated x curve is given with the standard deviation of the observed values of x from the calculated values along with the number of observed x points. The corresponding information is also given for y. This tabular information does not give a complete picture of the correlation, hence the following figures are included to further show the relationships. Each figure consists of two plots: one showing the calculated values of x and y as a function of W with the pressure data indicated by short bars, the other showing the values of oxide and hydroxide (r and s) at the same values of W needed to obtain the calculated values of x and y for the parameters specified on each.

Figures 6 through 12 are all for 0.333 BeF_2. Figures 6 and 7 (Runs 303 and 305) show similar conditions (600°C) except for the initial oxide concentration in the melt. Figure 8 (Run 306) shows the decrease in x and y when the influent HF and H_2O were stopped at the end of Run 305 and H_2 sparging continued. Figures 9 and 10 (Runs 307 and 313) are for 700 and 500°C, respectively. Figures 11 and 12 (Runs 501 and 511) show the behavior during higher-temperature (650°C) and lower-temperature (550°C) removal of oxide from the melt.

Figures 13 through 15 are for 0.273 BeF_2. Addition of oxide is shown in Figure 13 (Run 533) and removal of oxide is shown in Figures 14 and 15 (Runs 535 and 539). Figure 16 (Run 607) shows removal of oxide from 0.600 BeF_2 which is near the upper limit of this technique. Figure 17 (Run 621) shows the addition of oxide to 0.400 BeF_2.

Table 4. Parameters for Unsaturated Melts from Least Squares Program

X_{BeF_2}	Temp (°C)	Run No.	Q_O (atm moles kg^{-1})	Q_A (moles/kg)	r_O (moles/kg)	s_O (moles/kg)
0.273	600	529	3.23×10^{-5}	4.11×10^{-3}	2.23×10^{-2}	1.00×10^{-4}
0.273	600	539	2.57×10^{-5}	6.08×10^{-3}	1.56×10^{-2}	1.00×10^{-4}
0.273	650	535	8.38×10^{-5}	9.36×10^{-3}	1.59×10^{-2}	3.66×10^{-4}
0.273	650	537	8.54×10^{-5}	9.22×10^{-3}	4.53×10^{-3}	1.11×10^{-3}
0.273	700	533	2.72×10^{-4}	1.08×10^{-2}	2.66×10^{-3}	1.92×10^{-4}
0.333	500	313	5.11×10^{-6}	5.23×10^{-3}	7.76×10^{-4}	1.20×10^{-3}
0.333	500	314	1.94×10^{-6}	7.78×10^{-3}	3.31×10^{-3}	3.89×10^{-2}
0.333	500	503	4.39×10^{-6}	6.34×10^{-3}	8.46×10^{-3}	2.87×10^{-3}
0.333	544	509	1.12×10^{-5}	8.69×10^{-3}	8.25×10^{-4}	1.78×10^{-3}
0.333	550	315	1.58×10^{-5}	1.02×10^{-2}	7.23×10^{-4}	7.99×10^{-4}
0.333	550	316	1.11×10^{-5}	8.53×10^{-3}	7.11×10^{-3}	2.79×10^{-2}
0.333	550	511	1.84×10^{-5}	6.77×10^{-3}	1.08×10^{-2}	7.52×10^{-4}
0.333	550	525	2.55×10^{-5}	6.89×10^{-3}	2.69×10^{-2}	1.07×10^{-5}
0.333	600	301	5.15×10^{-5}	1.34×10^{-2}	6.85×10^{-4}	1.23×10^{-4}
0.333	600	302	3.90×10^{-5}	1.34×10^{-2}	6.27×10^{-3}	1.57×10^{-2}
0.333	600	303	6.39×10^{-5}	1.50×10^{-2}	1.31×10^{-2}	4.82×10^{-3}
0.333	600	305	6.05×10^{-5}	1.06×10^{-2}	7.08×10^{-4}	8.73×10^{-5}
0.333	600	306	3.93×10^{-5}	1.15×10^{-2}	8.10×10^{-3}	1.55×10^{-2}
0.333	600	309	6.33×10^{-5}	1.16×10^{-2}	1.04×10^{-3}	2.41×10^{-4}
0.333	600	310	4.73×10^{-5}	1.10×10^{-2}	1.29×10^{-2}	1.34×10^{-2}
0.333	650	319	2.63×10^{-4}	2.76×10^{-2}	1.01×10^{-3}	1.00×10^{-4}
0.333	650	501	2.05×10^{-4}	1.27×10^{-2}	2.01×10^{-2}	6.61×10^{-4}
0.333	650	513	2.55×10^{-4}	9.78×10^{-3}	2.35×10^{-3}	1.56×10^{-4}
0.333	650	514	1.00×10^{-4}	1.93×10^{-2}	8.00×10^{-3}	1.36×10^{-2}
0.333	651	527	2.53×10^{-4}	1.62×10^{-2}	5.42×10^{-3}	1.11×10^{-3}
0.333	700	307	6.82×10^{-4}	2.92×10^{-2}	1.29×10^{-3}	5.37×10^{-6}
0.333	700	311	9.87×10^{-4}	2.90×10^{-2}	2.10×10^{-3}	2.24×10^{-4}
0.333	700	523	7.24×10^{-4}	1.57×10^{-2}	3.07×10^{-3}	8.28×10^{-5}
0.333	700	524	6.00×10^{-4}	2.15×10^{-2}	1.80×10^{-2}	8.00×10^{-3}
0.400	550	619	1.48×10^{-4}	2.27×10^{-2}	1.36×10^{-2}	1.61×10^{-2}
0.400	550	625	9.10×10^{-5}	1.55×10^{-2}	9.20×10^{-4}	3.37×10^{-4}
0.400	604	621	4.94×10^{-4}	1.58×10^{-2}	3.84×10^{-3}	4.49×10^{-4}
0.400	702	627	6.00×10^{-3}	3.01×10^{-2}	1.50×10^{-2}	1.65×10^{-4}
0.600	500	607	1.96×10^{-4}	2.39×10^{-2}	2.09×10^{-2}	1.83×10^{-2}
0.600	600	611	1.14×10^{-3}	6.97×10^{-2}	2.66×10^{-3}	5.79×10^{-4}

Table 5. Comparison of Calculated and Observed Partial Pressures for Unsaturated Melts

Run No.	(Mean of x $\pm\sigma$) x 10^3 (atm)		No. of Points	(Mean of y $\pm\sigma$) x 10^3 (atm)		No. of Points
301	10.252	± 0.309	35	5.979	± 0.128	26
302	4.442	0.634	10	3.906	1.189	7
303	6.325	0.247	30	7.571	0.208	25
305	9.726	0.285	42	6.843	0.230	38
306	3.030	0.155	13	4.109	0.383	10
307	15.177	0.355	47	4.478	0.454	32
309	6.974	0.260	39	4.559	0.141	33
310	2.908	0.294	10	3.157	0.381	12
311	11.307	0.302	60	2.866	0.258	39
313	6.296	0.267	33	11.434	0.497	32
314	1.207	0.170	8	6.469	0.929	12
315	9.490	0.462	44	12.455	0.679	36
316	2.728	0.273	18	16.710		1
319	16.884	0.915	40	7.747	1.404	33
501	5.076	± 0.126	36	2.181	± 0.192	26
503	2.149	0.095	22	1.596	0.117	20
509	7.031	0.387	47	5.474	0.422	38
511	5.921	0.298	39	2.554	0.298	39
513	11.164	0.349	41	3.939	0.264	30
514	4.988	0.462	15	2.568	0.333	10
523	12.871	0.362	49	2.895	0.138	34
524	5.921	1.143	15	1.848	0.956	8
525	3.302	0.198	51	3.124	0.268	47
527	10.085	0.247	40	4.089	0.305	33
529	7.953	± 0.557	40	5.582	± 0.993	40
533	11.392	0.207	39	3.416	0.182	31
535	7.471	0.330	23	3.599	0.282	25
537	7.911	0.159	40	5.034	0.287	40
539	7.975	0.286	46	3.372	0.335	44
607	9.400	± 0.300	21	5.760	± 0.359	23
611	12.910	0.709	46	1.600	0.326	31
619	10.520	± 0.236	17	6.902	± 0.249	12
621	11.745	0.700	32	2.320	0.178	22
625	10.366	0.523	41	2.907	0.159	33
627	14.812	0.651	33	1.131	0.210	18

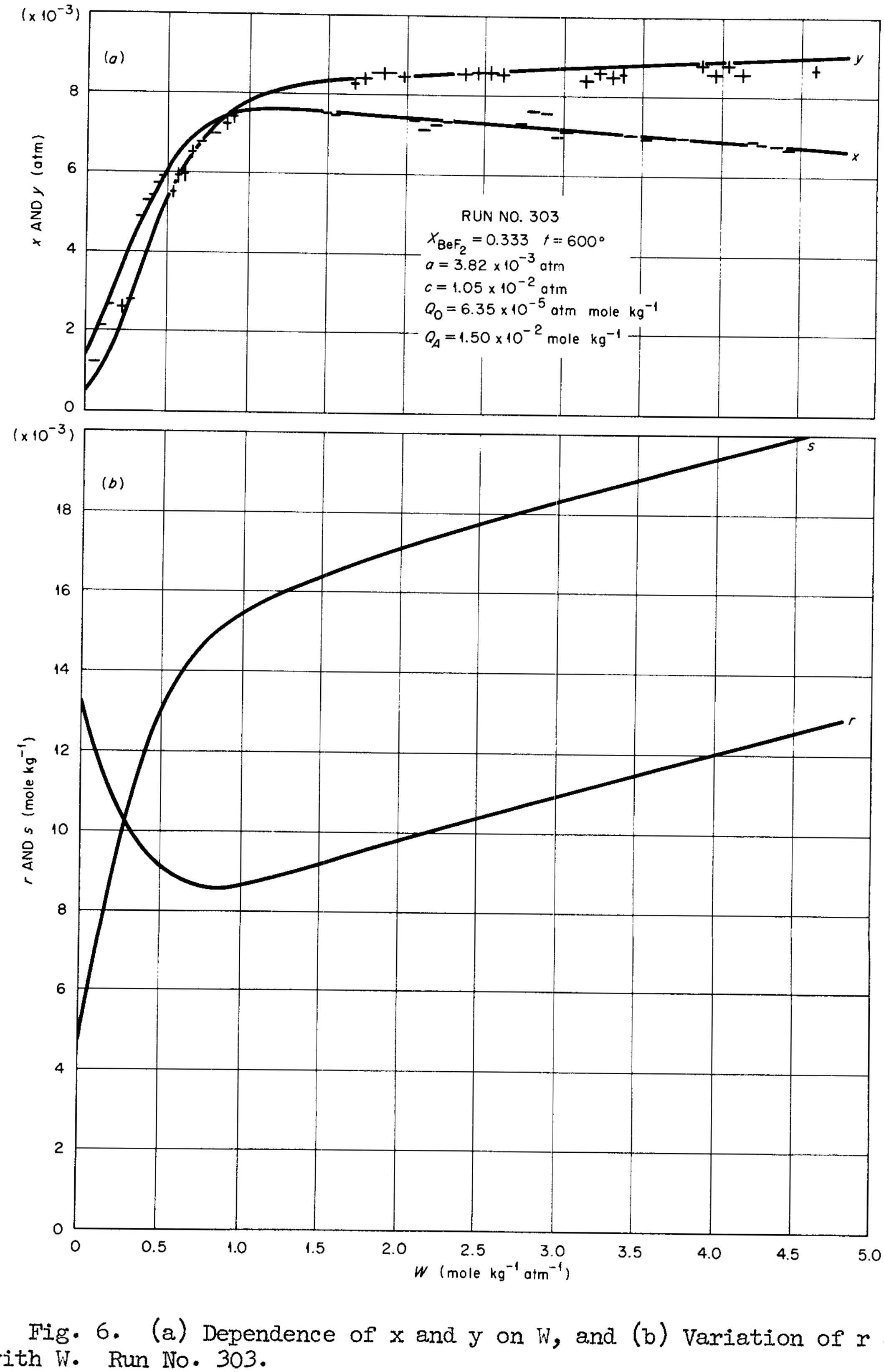

Fig. 6. (a) Dependence of x and y on W, and (b) Variation of r and s with W. Run No. 303.

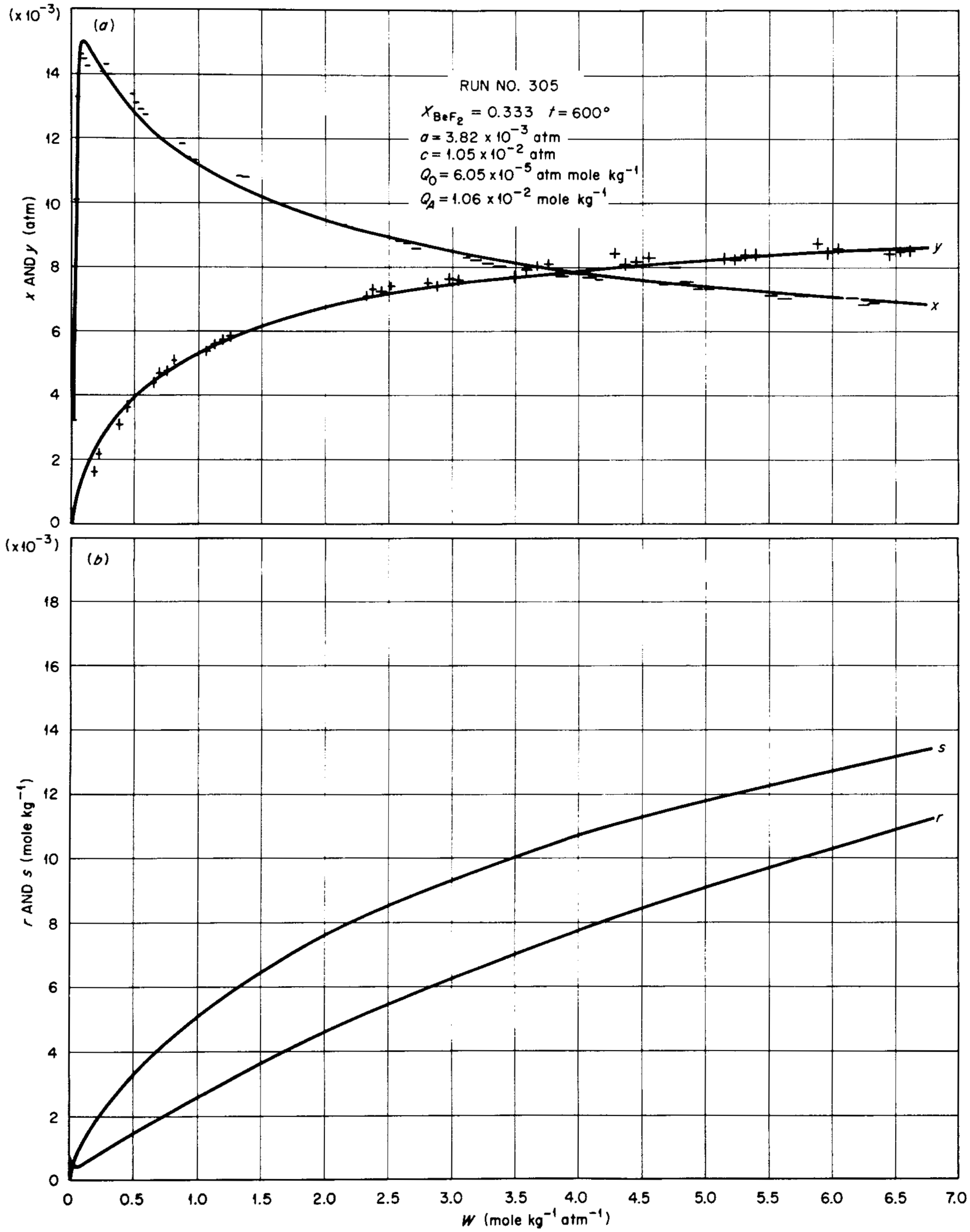

Fig. 7. (a) Dependence of x and y on W, and (b) Variation of r and s with W. Run No. 305.

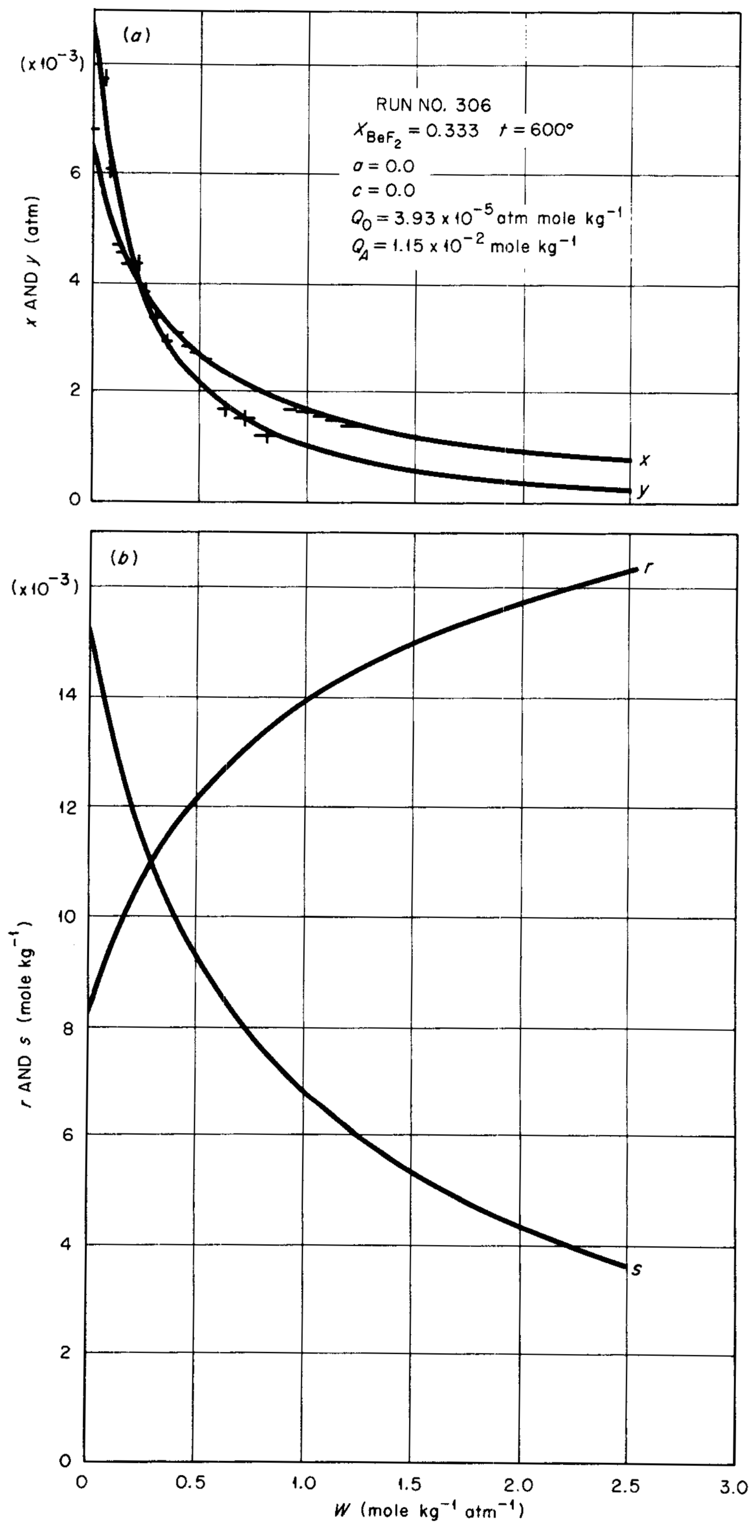

Fig. 8. (a) Dependence of x and y on W, and (b) Variation of r and s with W. Run No. 306.

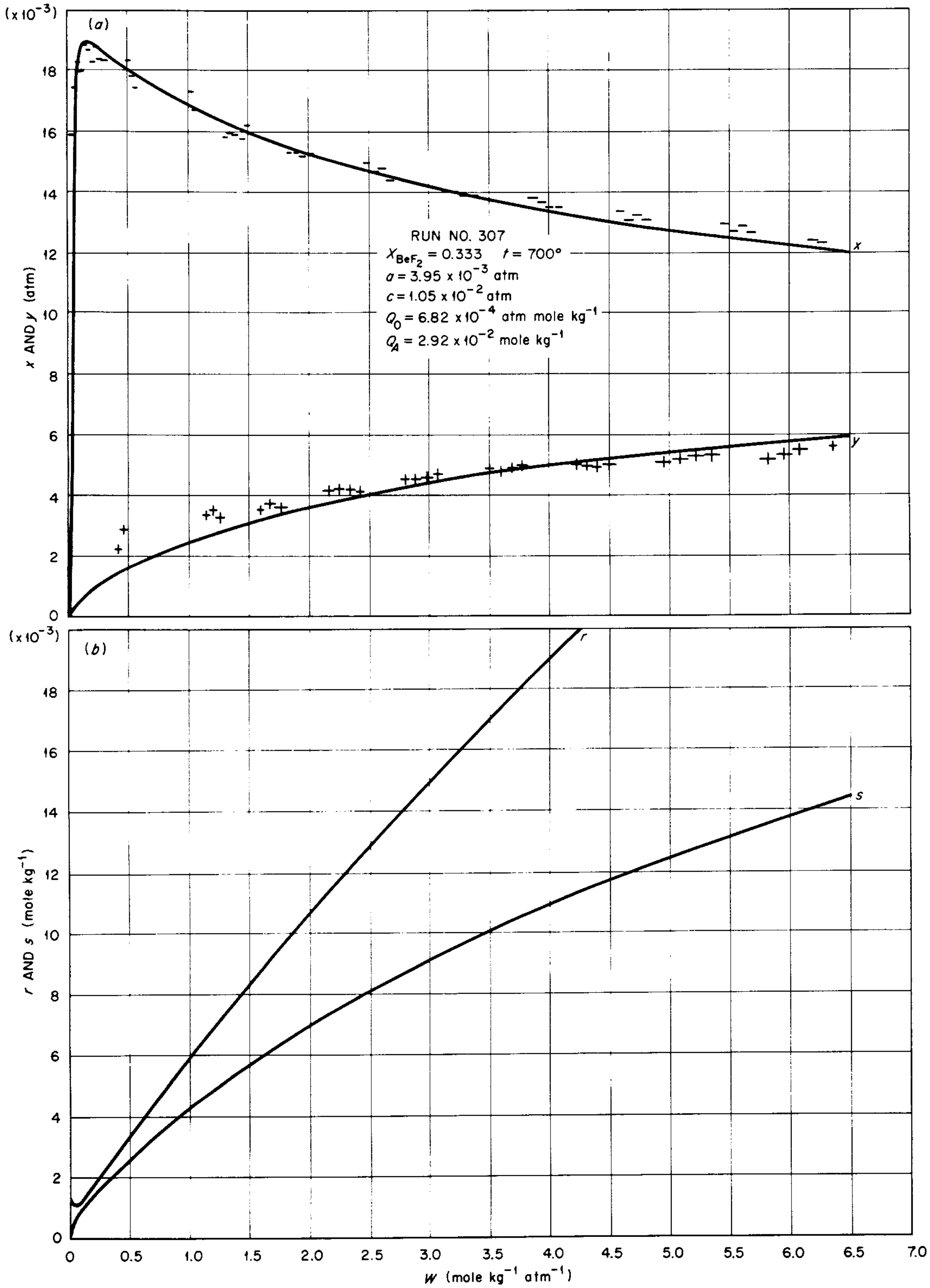

Fig. 9. (a) Dependence of x and y on W, and (b) Variation of r and s with W. Run No. 307.

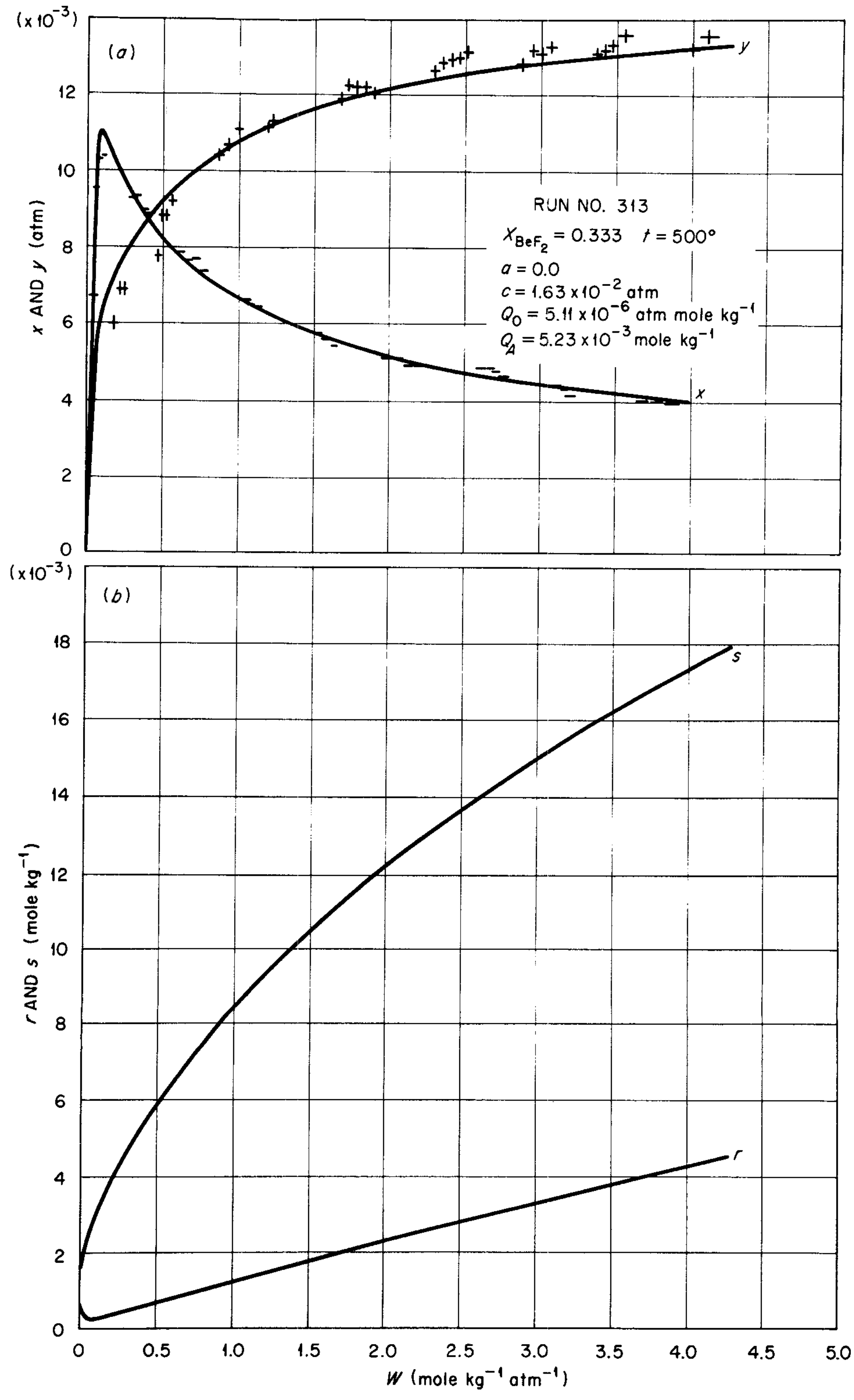

Fig. 10. (a) Dependence of x and y on W, and (b) Variation of r and s with W. Run No. 313

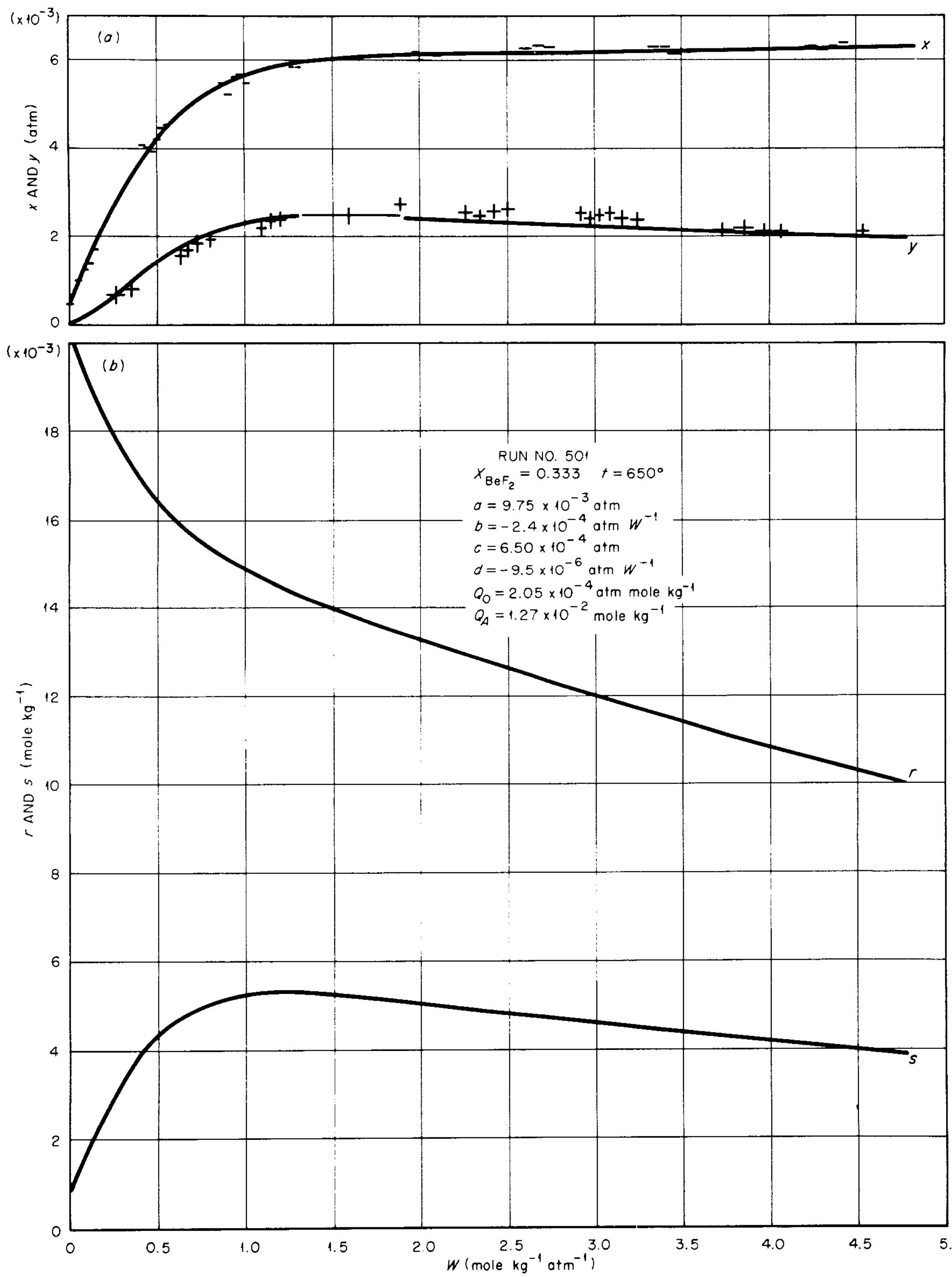

Fig. 11. (a) Dependence of x and y on W, and (b) Variation of r and s with W. Run No. 501.

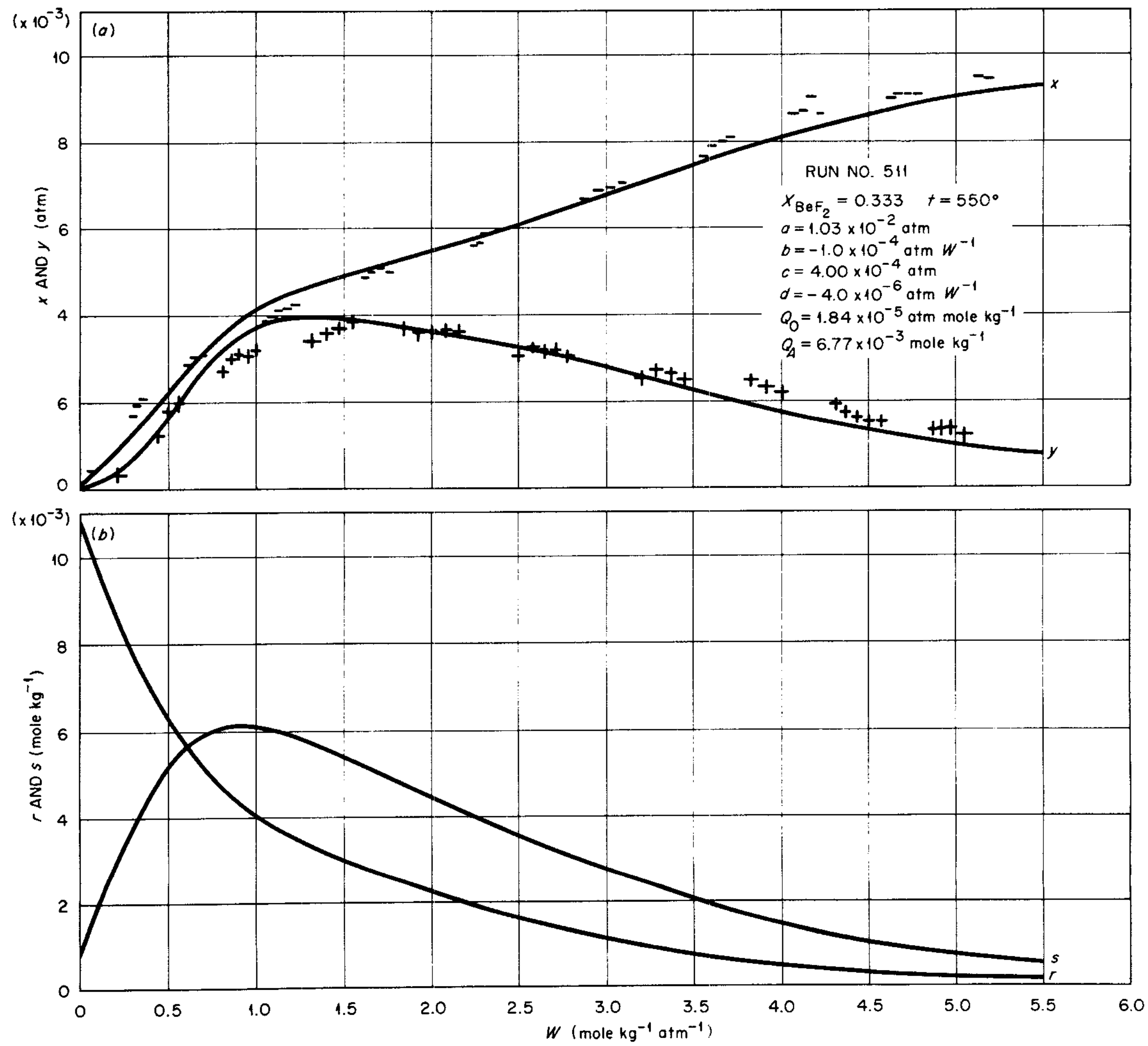

Fig. 12. (a) Dependence of x and y on W, and (b) Variation of r and s with W. Run No. 511.

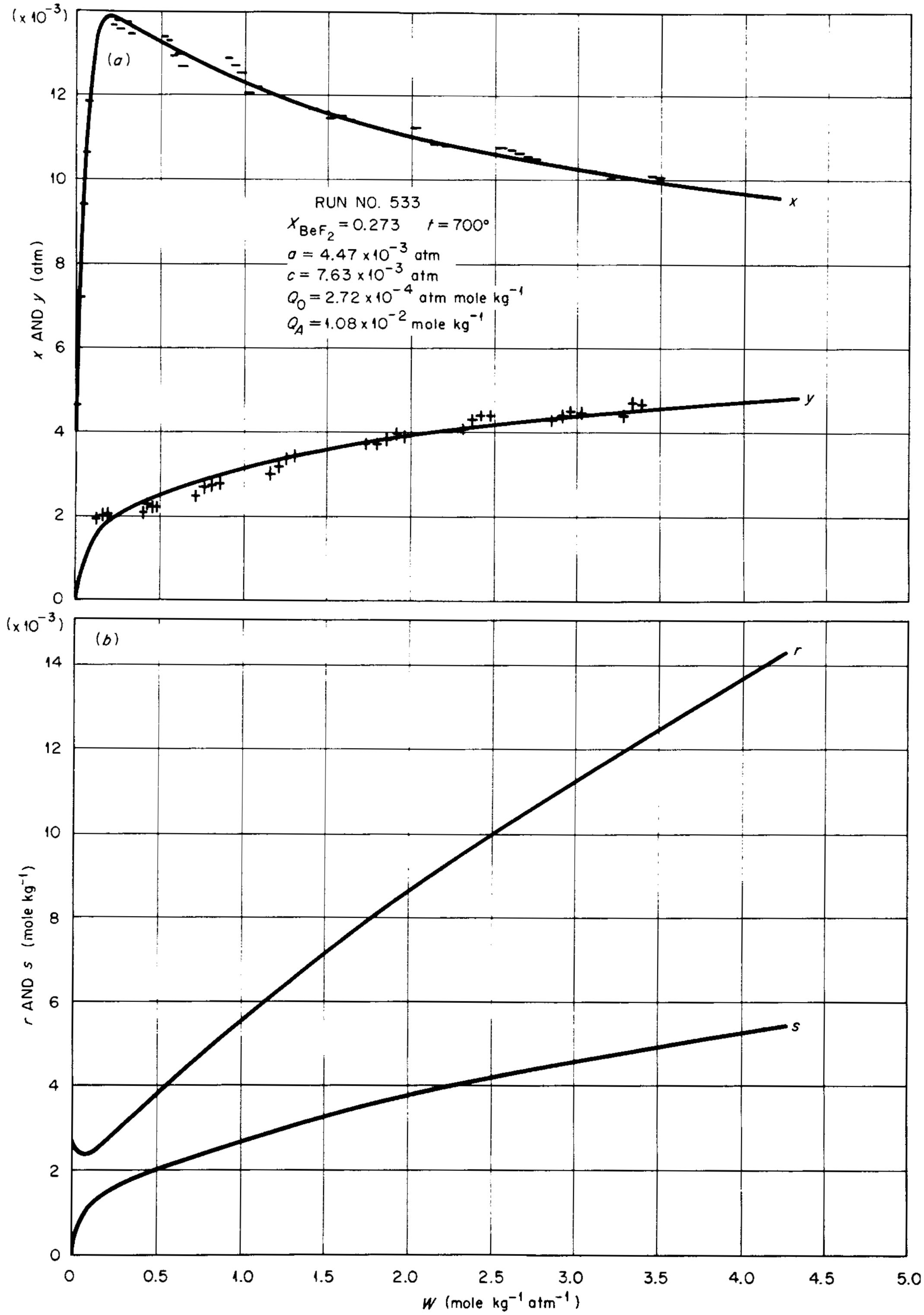

Fig. 13. (a) Dependence of x and y on W, and (b) Variation of r and s with W. Run No. 533.

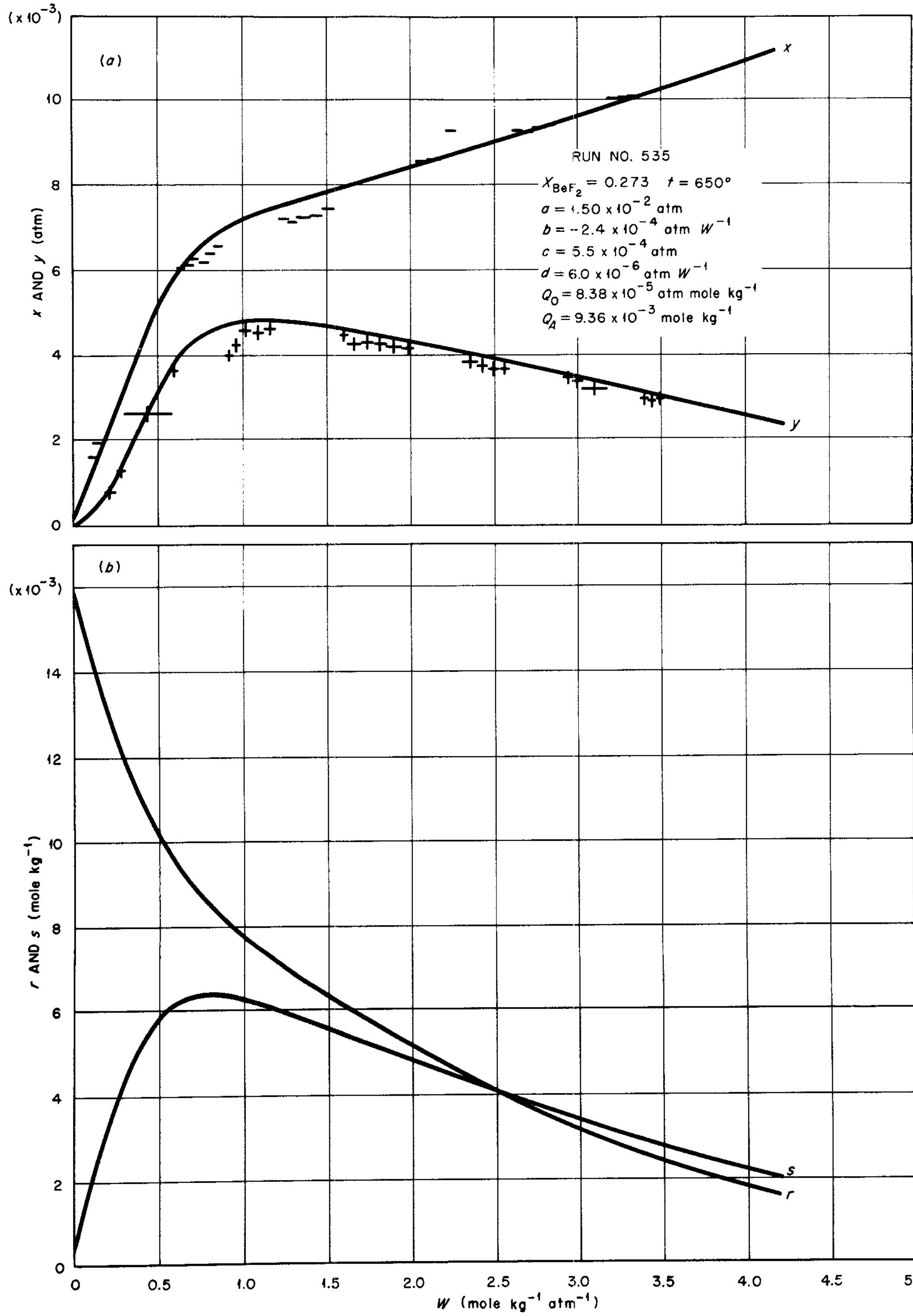

Fig. 14. (a) Dependence of x and y on W, and (b) Variation of r and s with W. Run No. 535.

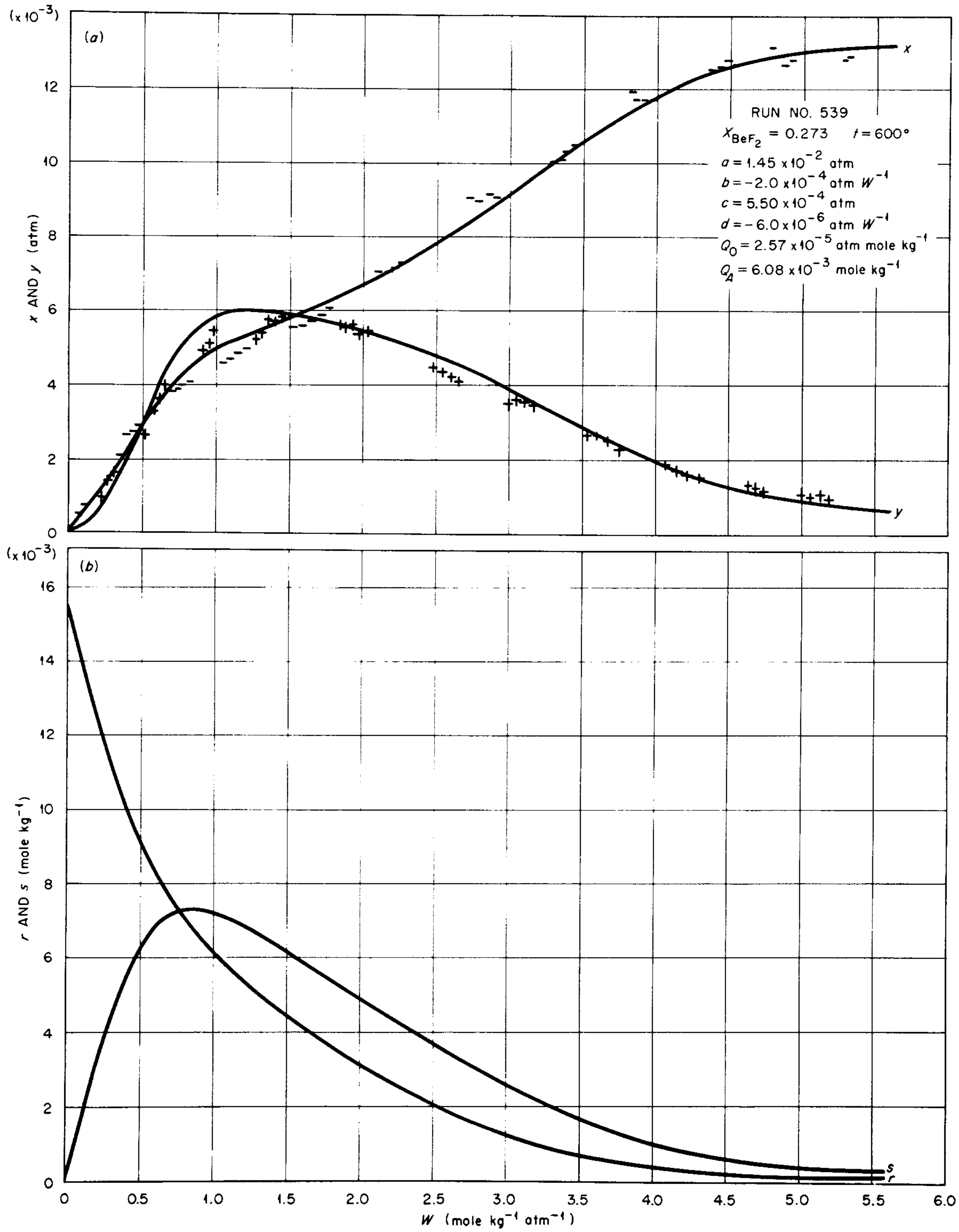

Fig. 15. (a) Dependence of x and y on W, and (b) Variation of r and s with W. Run No. 539.

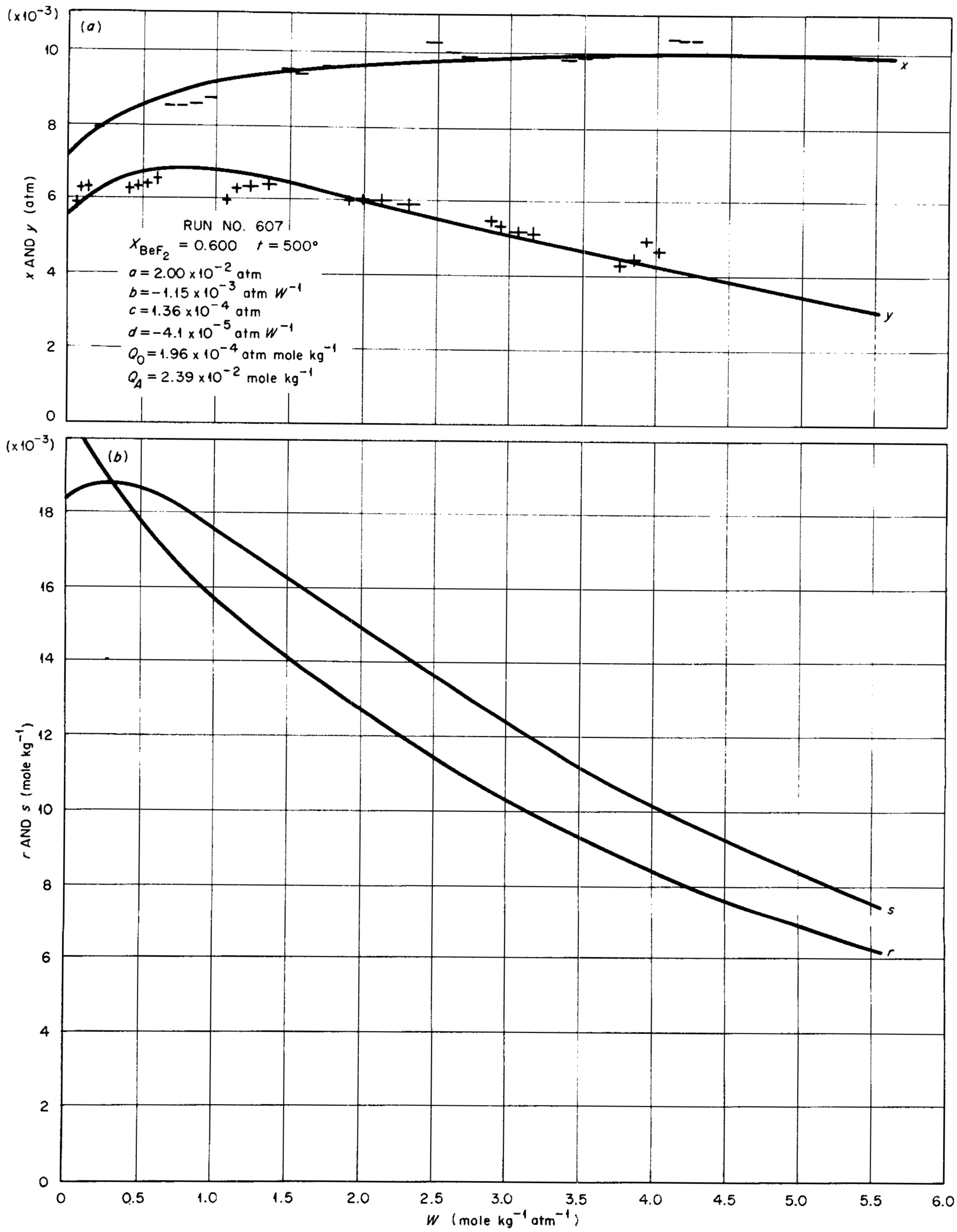

Fig. 16. (a) Dependence of x and y on W, and (b) Variation of r and s with W. Run No. 607.

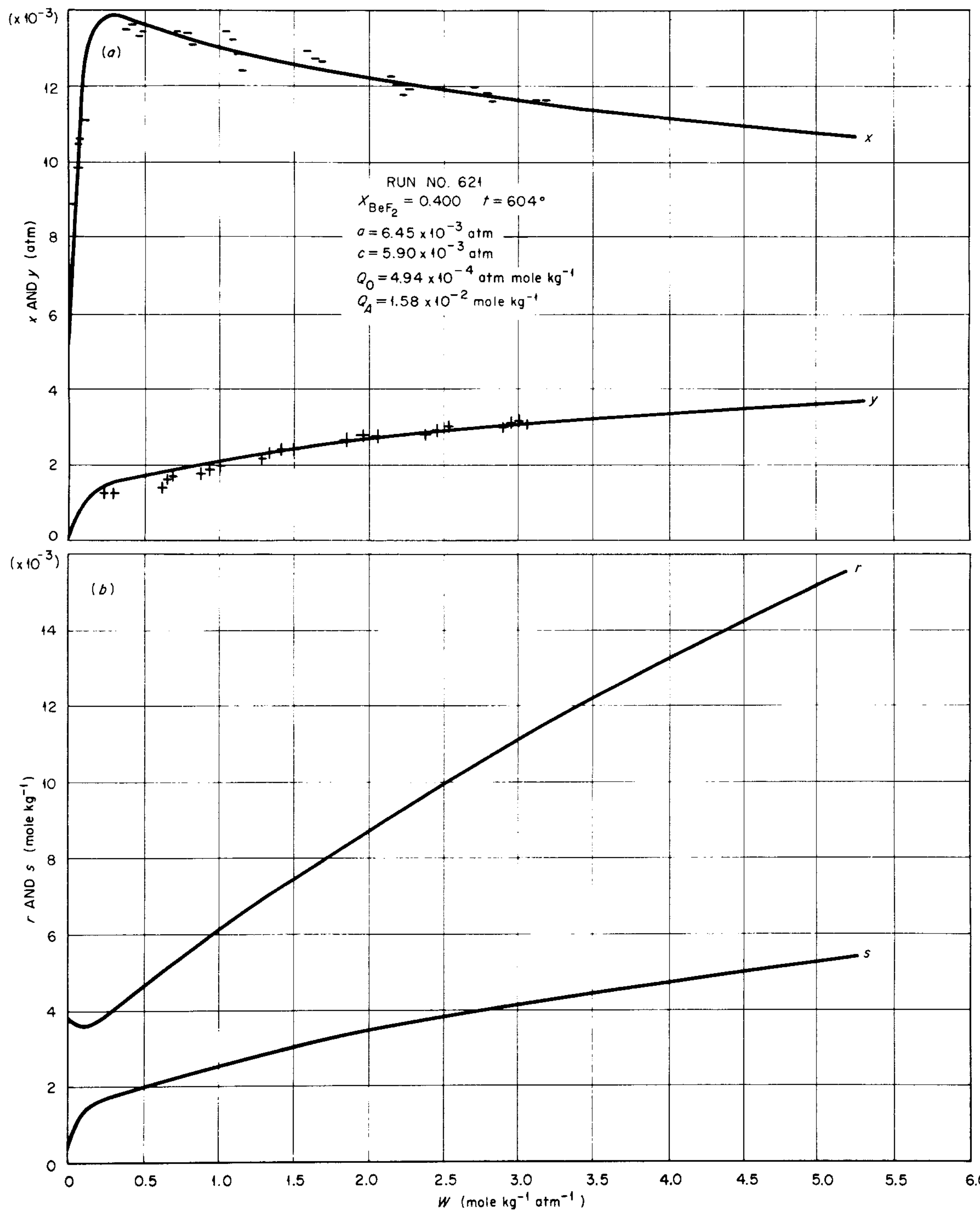

Fig. 17. (a) Dependence of x and y on W, and (b) Variation of r and s with W. Run No. 621.

Validity of Results

The general agreement between the calculated and observed effluent partial pressures of HF and H_2O, in view of the variety of experimental conditions, supports the method of measurement and indicates that the equilibria assumed were correct and sufficient. Some specific examples which support these equilibria are given below.

Saturated Melts

The first clearcut indication of the formation of hydroxide in measurable amounts came from the variation of effluent pressures with respect to volume of carrier gas. When a mixture of HF and H_2O was introduced into a BeO-saturated melt containing little or no hydroxide, the effluent partial pressures of HF and H_2O (x and y) increased with the increase in volume of gas passed in such a manner that x^2/y remained constant, while x/y decreased. Since $Q_O = x^2r/y$ and $Q_A = xs/y$, the quantity x^2/y would be constant when r is constant (fixed by BeO saturation) and x/y would decrease as s increases.

The influent partial pressures of HF and H_2O were varied and the resulting Q's compared. Runs 101, 103, and 123 for 0.316 BeF_2 at 700°C (with reported Q values of 2.18×10^{-2}, 2.47×10^{-2}, and 2.13×10^{-2}, respectively) had influent pressures such that influent $(P_{HF})^2/(P_{H_2O})$ were 4.36×10^{-2}, 4.46×10^{-2}, and 4.25×10^{-3}, respectively. Runs 105, 107, 109, and 111 for 0.316 BeF_2 at 600°C (with reported Q values of 4.61×10^{-3}, 4.07×10^{-3}, 4.09×10^{-3}, and 4.69×10^{-3}, respectively) also indicated that the observed Q's were independent of the influent pressures, with influent $(P_{HF})^2/(P_{H_2O})$ values being 6.52×10^{-2}, 2.28×10^{-2}, 5.54×10^{-3}, and 4.98×10^{-3}, respectively.

These runs include cases where the influent and effluent pressures were nearly identical and also where quite a bit of HF was required to react with oxide to form water and the reverse (H_2O reacting with fluoride to form HF). All three gave Q's in agreement. In the early runs, the flow rate of carrier gas was varied from 50 to 150 ml/min and no variation in the effluent pressures was observed.

The Q_A values determined using saturated melts were similarly found to be independent of the ratios of HF and H_2O in the influent stream. Also, Q_A was shown to be independent of whether hydroxide was being added or removed by the gas stream. For example, see the odd-even pairs 401-402, 405-406, and 407-408 in Table 3 for 0.300 BeF_2 at 652, 601 and 700°C, respectively.

The proton balance between influent and effluent gas streams, after the initial region where hydroxide was being formed, also attests to the validity of the measurement.

After completion of the 200 series the melt was transferred from the vessel through a sintered nickel filter and the material on the filter examined by x-ray diffraction. The characteristic peaks for LiF-BeF_2 and BeO were observed. There was no indication of compound formation between BeO and the fluoride melt or of other constituents [such as Li_2O or $Be(OH)_2$].

Unsaturated Melts

In addition to the tests performed with saturated melts, the validity of Q_O was checked by conducting experiments in which oxide was being added and also when oxide was being removed. The former, more precise results, and the latter were in reasonable agreement.

The total pressure of the reactive gases was varied by changing the temperature of the saturator with no noticeable effect on the Q_O and Q_A values.

IV. DISCUSSION

The measured equilibrium quotients, Q, Q_A, and Q_O, were correlated with temperature and melt composition in terms of simple algebraic expressions. In the case of Q, wherein $K_a = Q/a_{BeF_2}$, the resulting correlation can be used to derive a considerable amount of thermodynamic information about the LiF-BeF_2 solutions and, by extrapolation, about pure BeF_2 liquid. Combination of Q and Q_O can be used to obtain the concentration of oxide at BeO saturation. The variation of Q_A with melt composition and temperature reflects the stability of hydroxide in LiF-BeF_2 melts.

Correlation of Q

For each composition, the Q values were correlated according to the equation log Q = slope(1/T) + constant. The Q's were weighted by an amount inversely proportional to their variance since all measurements did not follow the same parent distribution. The values of the parameters are presented in Table 6.

Since one of the primary objectives was determination of thermodynamic properties of $BeF_2(\underline{l})$, extrapolation of these data as a function of X_{BeF_2} to the pure liquid BeF_2 was desirable. For a given temperature,

$\log (Q/X_{BeF_2})$ vs $(X_{LiF})^2$

forms a parabola for the composition range 0.30 to 0.80 BeF_2, thus the Q values at various compositions were correlated by the function

$$\log (Q/X_{BeF_2}) = k + l(X_{LiF})^2 + m(X_{LiF})^4 \quad .$$

From definition in Chapter I, log K_a = k, therefore, correlation of k as a function of 1/T will yield the thermodynamics of the desired reaction. Both l and m also show linear dependence upon 1/T. For the coefficients expressed according to $k = k^{o} + k'(1/T)$, etc.

$k^{o} = 3.900 \pm 0.019$ $\quad$ $k' = -4418 \pm 17$

$l^{o} = 7.819 \pm 0.225$ $\quad$ $l' = -5440 \pm 218$

$m^{o} = -12.66 \pm 0.60$ $\quad$ $m' = 5262 \pm 513$.

From the above parameters, equations were generated for each experimental composition as a function of (1/T). The smoothed parameters obtained are tabulated in Table 7. The lines generated by the smoothed parameters for the various experimental compositions are shown in Figure 18 along with log K_a vs (1/T) for pure $BeF_2(\underline{l})$ based on the extrapolation. Figure 19 provides a measure of this correlation. The quantity Q_{obs}/Q_{calc} is shown for each experimental run. The standard deviation for each point is represented by a vertical bar.

<u>Activity of BeF_2 and LiF</u>

Since Q is equal to $K_a(a_{BeF_2})$,

$$\log (Q/X_{BeF_2}) = \log K_a(a_{BeF_2})/(X_{BeF_2}) = \log K_a + \log (a_{BeF_2}/X_{BeF_2}) \quad .$$

By definition, a_{BeF_2} is unity for pure liquid BeF_2 ($X_{BeF_2} = 1$), therefore, $\log (a_{BeF_2}/1) = 0$. Since $X_{LiF} = 0$ at this point,

$$\log K_a = k^{o} + k'(1/T) \quad .$$

In order to solve for $\log (a_{BeF_2}/X_{BeF_2})$, which is $\log \gamma_{BeF_2}$, at any composition the value of $\log K_a$ must be subtracted from $\log (Q/X_{BeF_2})$ leaving

$$\log \gamma_{BeF_2} = [l^{o} + l'(1/T)]X^2_{LiF} + [m^{o} + m'(1/T)]X^4_{LiF} \quad .$$

At a specified temperature, the variation in γ_{BeF_2} with composition is given by

$$\log \gamma_{BeF_2} = \alpha X^2_{LiF} + \beta X^4_{LiF} \quad .$$

Values for the parameters at three temperatures are presented below:

Temp (°C)	α	$\pm\sigma$	β	$\pm\sigma$
500	0.783	± 0.380	-5.85	± 0.89
600	1.589	0.356	-6.66	0.85
700	2.229	0.338	-7.25	0.80

The quoted uncertainties in α and β lead to an average uncertainty of ± 0.10 units in log γ_{BeF_2}. From these uncertainties the values 100° apart would overlap within the quoted uncertainty, but the values for 500 and 700°C definitely do not overlap.

According to the Gibbs-Duhem Equation, if the activity (or activity coefficient) of one component is known as a function of composition, the other can be determined by integration of the equation

$$d \ln (\gamma_2) = -X_1/X_2 \, d \ln (\gamma_1) \quad .$$

From the equations given for log γ_{BeF_2} as a function of composition at specified temperatures, integration was carried out as follows:

$$\log \gamma_{BeF_2} = \alpha X^2_{LiF} + \beta X^4_{LiF}$$

$$d \log \gamma_{BeF_2} = (2\alpha X_{LiF} + 4\beta X^3_{LiF}) \, dX_{LiF}$$

$$d \ln \gamma_{LiF} = -\left[\frac{(1 - X_{LiF})}{X_{LiF}}\right](2.303)(2\alpha X_{LiF} + 4\beta X^3_{LiF}) \, dX_{LiF}$$

$$\ln \gamma_{LiF} = (2.303) \int (-2\alpha + 2\alpha X_{LiF} - 4\beta X^2_{LiF} + 4\beta X^3_{LiF}) \, dX_{LiF}$$

$$\log \gamma_{LiF} = -2\alpha X_{LiF} + \alpha X^2_{LiF} - (4/3)\beta X^3_{LiF} + \beta X^4_{LiF} + C \quad .$$

Two methods for evaluating C for various temperatures were considered. For a given temperature, if γ_{LiF} is known for one composition, C can be determined and, hence, γ_{LiF} determined for all compositions. First,

γ_{LiF} was calculated from the reported composition at LiF liquidus at various temperatures where $a_{LiF} \equiv 1$ and $\gamma_{LiF} \equiv 1/X_{LiF}$. The values for γ_{LiF} at the various temperatures were determined from the published phase diagram[2] which was presented in Chapter I. The second method involved the use of the Q values for BeO-saturated melts in contact with solid LiF to determine the X_{LiF} at saturation. (The measured Q's are tabulated in Table 3 and the parameters for smoothed fit are given in Table 6.) The equations for log a_{BeF_2} as a function of composition and log a_{BeF_2} at LiF saturation were solved simultaneously to obtain X_{LiF}. With the concentration of LiF at $a_{LiF} = 1$ known, γ_{LiF} could be evaluated. The following values for C were obtained by the two methods.

	From Thoma's Phase Diagram			From Solution of Equations		
Temp (°C)	X_{BeF_2}	γ_{LiF}	C	X_{BeF_2}	γ_{LiF}	C
500	0.329	1.490	-0.299	0.321	1.473	-0.328
600	0.280	1.389	+0.083	0.281	1.391	+0.086
700	0.208	1.263	+0.283	0.256	1.344	+0.451

Agreement of values of C determined by both methods is very good at 500 and 600 but not at 700°C. Although LiF saturation at 700°C occurs at 0.208 BeF_2, the value of C corresponding to a calculated saturation value of 0.256 BeF_2 was considered more acceptable because the parabolic function does not fit the data well below 0.30 BeF_2. In fact, at 0.25 BeF_2 (data at 650 and 700°C) the fit is very poor.

Other empirical expressions were tried in an attempt to improve the fit in this region. No improvement was obtained using expressions containing additional odd-powers of X_{LiF}, but a very good fit was obtained from 0.25 to 0.50 BeF_2 by using an equation containing X^2, X^4, and X^6

terms. Correlation of the data at 700°C gave the following equation:

$$\log (Q/X_{BeF_2}) = -0.723 + 4.094X^2_{LiF} - 16.64X^4_{LiF} + 12.51X^6_{LiF} \quad .$$

This equation should be compared with the correlation using only X^2 and X^4 terms:

$$\log (Q/X_{BeF_2}) = -0.640 + 2.229X^2_{LiF} - 7.25X^4_{LiF} \quad .$$

Both fit equally well between 0.33 and 0.50 BeF_2, but the former fits better for concentrations below 0.33 BeF_2 and the latter fits better for concentrations above 0.50 BeF_2. In order to obtain values for γ_{BeF_2} from the former equation, the constant term was adjusted to yield the same value of γ_{BeF_2} (at 0.40 BeF_2) as was given by the simpler expression used to express activities above 0.30 BeF_2. The expression obtained for log γ_{BeF_2} was given by:

$$\log \gamma_{BeF_2} = -0.0382 + 4.094X^2_{LiF} - 16.64X^4_{LiF} + 12.51X^6_{LiF} \quad .$$

The Gibbs-Duhem integration was carried out on this equation in order to obtain the expression for log γ_{LiF}:

$$\log \gamma_{LiF} = -8.188X_{LiF} + 4.094X^2_{LiF} + 22.20X^3_{LiF} - 16.64X^4_{LiF} - 15.01X^5_{LiF} + 12.51X^6_{LiF} + C \quad .$$

If this equation is solved simultaneously with the equation for log a_{BeF_2} at LiF saturation, the concentration equals 0.208 BeF_2 (the same as reported by Thoma) and the value of C equals 1.124.

Figure 20 shows a plot of log γ_{BeF_2} and log γ_{LiF} as a function of composition at 500, 600, and 700°C. The dashed lines indicate the values at 700°C (below 0.30 BeF_2) when the equation with the X^6_{LiF} term was used. (The overall fit was not improved by using the extra term, hence the simpler form was used for the more general correlation.) The dotted line indicates the values of log γ_{LiF} at which the solid LiF phase would appear and the values of log γ_{BeF_2} at which the pure liquid BeF_2 phase would appear.

The (-·--·-) line shows the predicted behavior of log γ_{BeF_2} on the basis of a simple model in which, for the region 0.33 - 1.0 BeF_2, the components are redefined as BeF_2 and Li_2BeF_4 and the activity of BeF_2 is assumed equal to the mole fraction X'_{BeF_2}. The value of X'_{BeF_2} is related to X_{BeF_2}, the mole fraction in LiF-BeF_2 mixtures, by

$$X'_{BeF_2} = (3X_{BeF_2} - 1)/2X_{BeF_2} \quad .$$

Since $a_{BeF_2} = X'_{BeF_2}$ as defined above, the activity coefficient of BeF_2 in LiF-BeF_2 mixtures is given by

$$\gamma_{BeF_2} = (3X_{BeF_2} - 1)/2X^2_{BeF_2} \quad .$$

This procedure of redefining components is similar to the method of Flood and Urnes;[50] however, the assumed proportionality of a_{BeF_2} to its mole fraction is purely arbitrary and the resulting correlation, which is rather good, is, therefore, entirely empirical. It might be more reasonable in this model to expect a_{BeF_2} to be proportional to the product of the cation fraction of Be^{2+} and the anion fraction of (F^-) squared [i.e., a_{BeF_2} proportional to $(X'_{BeF_2})^3$], the melt being considered to be a simple mixture of the four ions Li^+, Be^{2+}, F^-, and BeF_4^{2-}. In order to rationalize the proportionality of a_{BeF_2} to X'_{BeF_2} it seems necessary to adopt one of the following unattractive models for the components

Li_2BeF_4 (undissociated) + BeF_2 (undissociated)

or

$Li_2^{2+}\ BeF_4^{2-} + Be^{2+}\ BeF_4^{2-}$.

A more realistic approach to an interpretation of the behavior of a_{BeF_2} and a_{LiF} can perhaps be made in terms of models such as have been proposed by Førland.[51] According to this model $BeF_2(\underline{l})$ is assumed to be a three-dimensional polymer of BeF_4^{2-} tetrahedra with common corners, and the addition of LiF is assumed to break the Be-F-Be bonds. However, no such correlation will be attempted here.

Two features of the observed variation of a_{BeF_2} are especially noteworthy:

(1) The rapid drop in γ_{BeF_2} as X_{BeF_2} approaches 0.33, at all temperatures, suggests the formation of BeF_4^{2-} ion.

(2) The temperature coefficient of γ_{BeF_2} indicates increasing positive deviations from Raoult's law with increasing temperature at high X_{BeF_2} values.

The activities of BeF_2 and LiF were calculated from the activity coefficients as a function of composition and temperature. The activities at 500, 600, and 700°C are shown in Figure 21. The plot of activities, which should be proportional to the partial pressures of BeF_2 and LiF, corresponds to the predicted behavior of a system showing incomplete miscibility.[52] Since the positive deviations increase with temperature, these data seem to indicate that the system has a lower consolute temperature near 700°C and 0.80 BeF_2. Although the number of determinations of Q in this area is not extensive enough to definitely show the formation of two liquid phases, the indication is strong enough to warrant further investigation of this possibility. To date there is no clearcut evidence of partial immiscibility in the LiF-BeF_2 system although various investigators have considered the possibility of such occurrence.[53-55] The MgO-SiO_2 system, which has frequently been compared with the LiF-BeF_2 system,[1] indeed does show a miscibility gap. At least one case has been reported for a similar fluoride melt. At low LiF concentrations in the system, $LiF-BeF_2-ZrF_4$, evidence was obtained from quenched samples that two immiscible liquids were formed above the ZrF_4 primary phase field.[56]

The LiF activity exhibits large positive deviations at low BeF_2 concentrations; however, it should be kept in mind that these activities are referred to the solid rather than the supercooled liquid (if the latter were used as the reference state, all of the activities would be lower). The activities reported in the present work are in general agreement with previous measurements (summarized in Table 2). The largest discrepancy is in the value for LiF at 0.50 BeF_2 reported by Berkowitz and Chupka.[22] The value for LiF by Büchler[24] is in reasonable agreement. Also, Büchler obtained greater activities for BeF_2 at higher temperatures.[23] Special attention should be given to the value reported for 0.67 BeF_2 (displaying very large positive deviations) which corresponds very closely to present values. This agreement is particularly gratifying since three completely different techniques have been used to obtain the values. Also, the previous studies were performed with melts which did not have BeO present. The presence of BeO would not be expected to affect the activities since BeO is only slightly soluble. Beryllium oxide could not have affected the Gibbs-Duhem integration since BeO was always present as a saturating phase, thereby restricting its activity to unity.

The expression for the relative partial molal heat content is given by:

$$\frac{\partial \ln \gamma_1}{\partial T} = - \frac{(\overline{H}_1 - H_1^{\circ})}{RT^2} \quad .$$

From the expression for log γ_{BeF_2}, the appropriate differentiation may be carried out to obtain

$$\frac{\partial \log \gamma_{BeF_2}}{\partial T} = -(l'X^2_{LiF} + m'X^4_{LiF})(1/T)^2 \quad .$$

Therefore,

$$(l'X^2_{LiF} + m'X^4_{LiF}) = (\bar{H}_{BeF_2} - H^o_{BeF_2})/2.303R$$

and

$$(\bar{H}_{BeF_2} - H^o_{BeF_2}) = 2.303R\ [(-5440 \pm 218)X^2_{LiF} + (5262 \pm 513)X^4_{LiF}] \quad .$$

Values for the relative partial molal heat contents at various compositions are tabulated below:

X_{BeF_2}	$(\bar{H}_{BeF_2} - H^o_{BeF_2})$ cal/mole	X_{BeF_2}	$(\bar{H}_{BeF_2} - H^o_{BeF_2})$ cal/mole
0.80	- 961 ± 40	0.40	-5839 ± 471
0.70	-2045 ± 92	0.333	-6309 ± 640
0.60	-3363 ± 224	0.30	-6420 ± 745
0.50	-4718 ± 228		

Since the activity coefficient of BeF_2 is given by an analytical expression, the excess chemical potential of BeF_2 can be directly calculated from

$$\mu^E_{BeF_2} = RT \ln \gamma_{BeF_2} \quad .$$

Since $\ln \gamma_{BeF_2}$ is a function of temperature, this equation does not lend itself to easy tabular or graphic presention (unless in a plot similar to Figure 20); however, the analytical expression should be useful for further calculations involving the excess chemical potential.

Table 6. Parameters from Correlation of Q as a Function of Temperature at Specified Compositions

X_{BeF_2}	(slope $\pm\sigma$) x 10^{-3}		constant $\pm$ σ	
(LiF saturation)	-3.662	0.184	1.534	0.549
0.25	-6.025	0.365	4.147	0.384
0.30	-6.173	0.136	4.565	0.147
0.316	-5.832	0.075	4.322	0.083
0.333	-5.674	0.116	4.233	0.133
0.40	-5.565	0.101	4.528	0.114
0.50	-5.529	0.257	4.857	0.309
0.60	-5.274	0.046	4.745	0.052
0.70	-4.740	0.050	4.199	0.057
0.80	-4.667	0.215	4.140	0.249

Table 7. Smoothed Parameters from Correlation of Q as a Function of Composition and Temperature at the Specified Compositions

X_{BeF_2}	slope x 10^{-3}	constant
0.25	-5.813	3.690
0.30	-5.821	4.168
0.316	-5.812	4.288
0.333	-5.797	4.396
0.40	-5.694	4.676
0.50	-5.449	4.763
0.60	-5.153	4.605
0.70	-4.865	4.346
0.80	-4.628	4.096

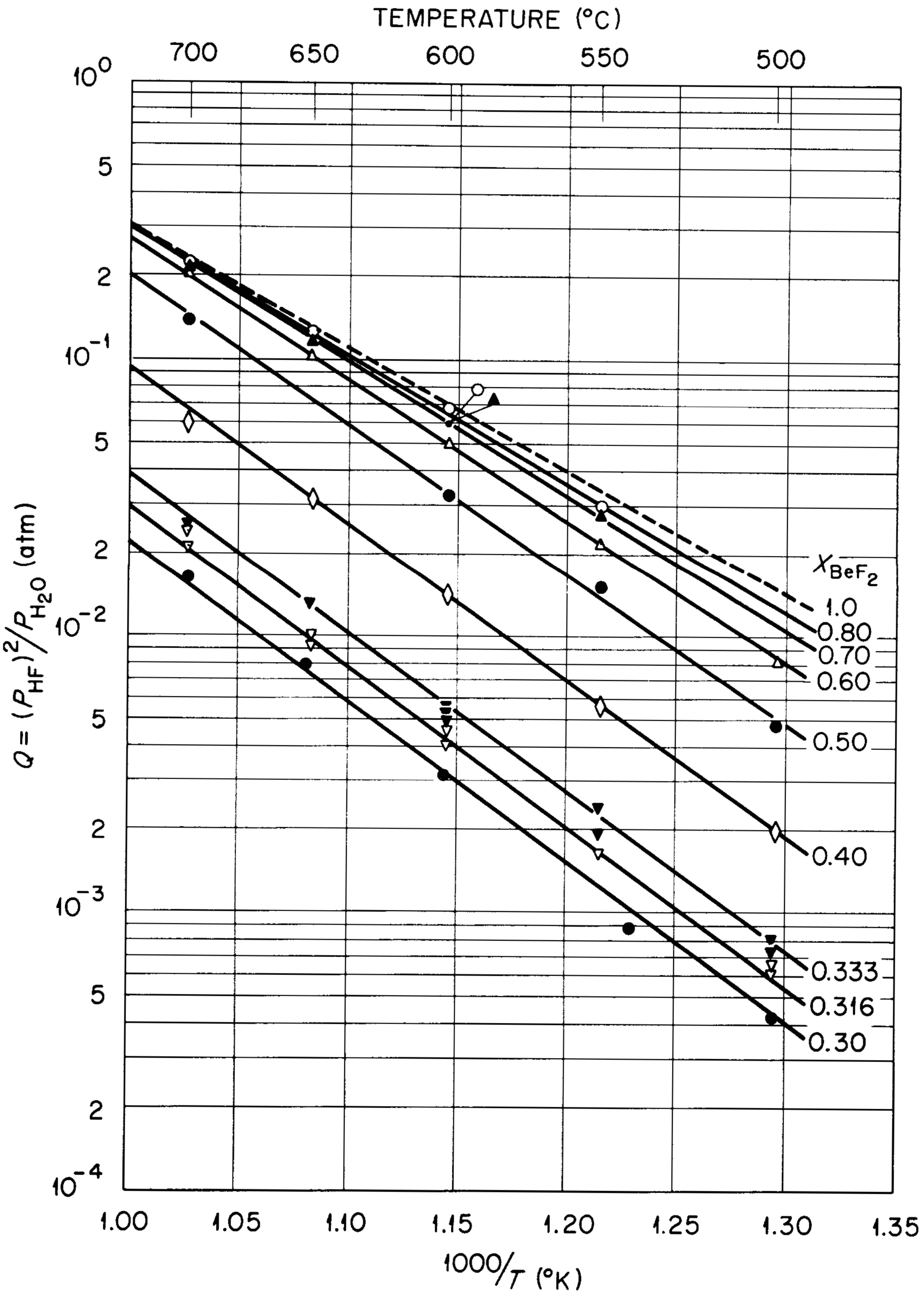

Fig. 18. Correlation of log Q as a Function of Melt Composition and Temperature.

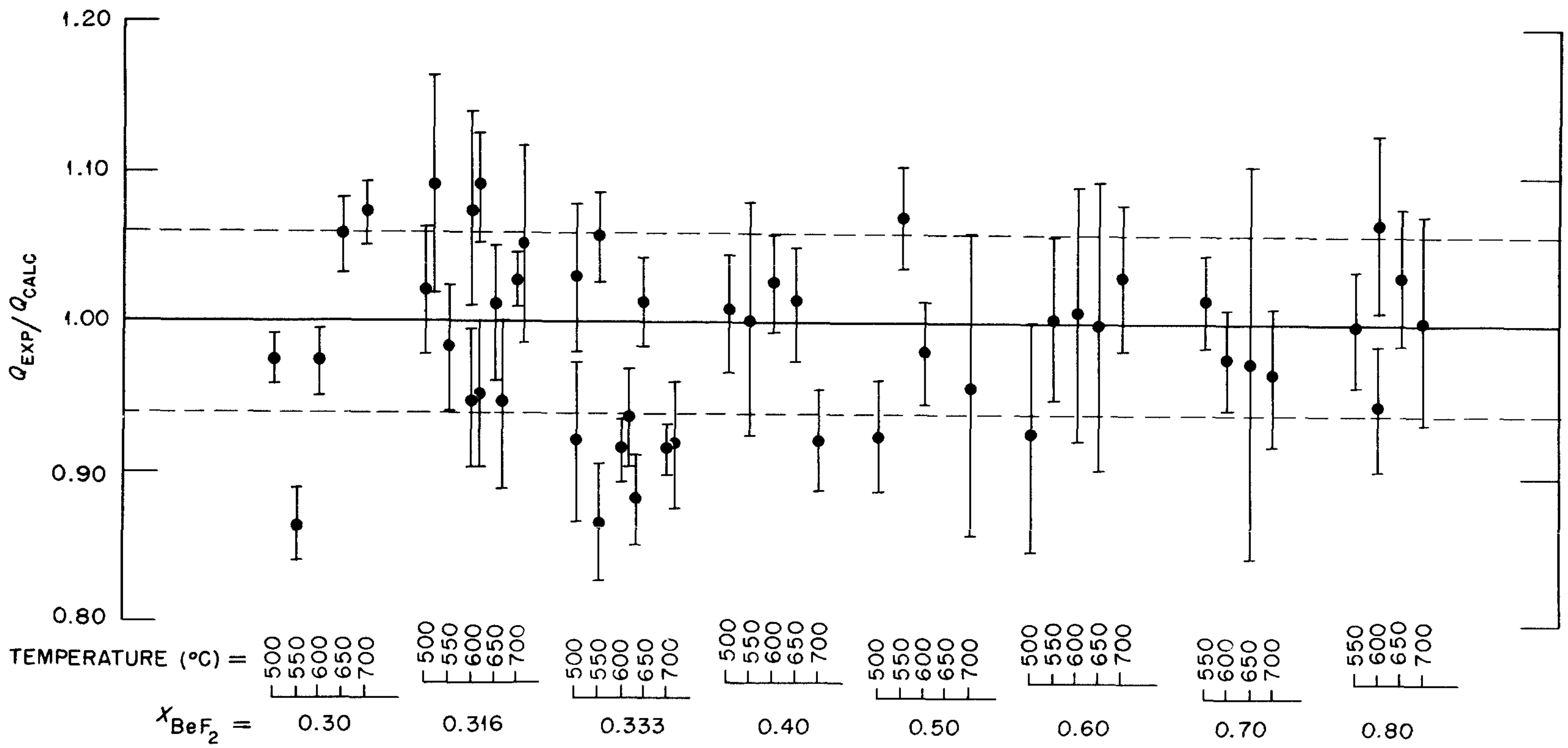

Fig. 19. Agreement between Observed Q and Value of Q from Correlation.

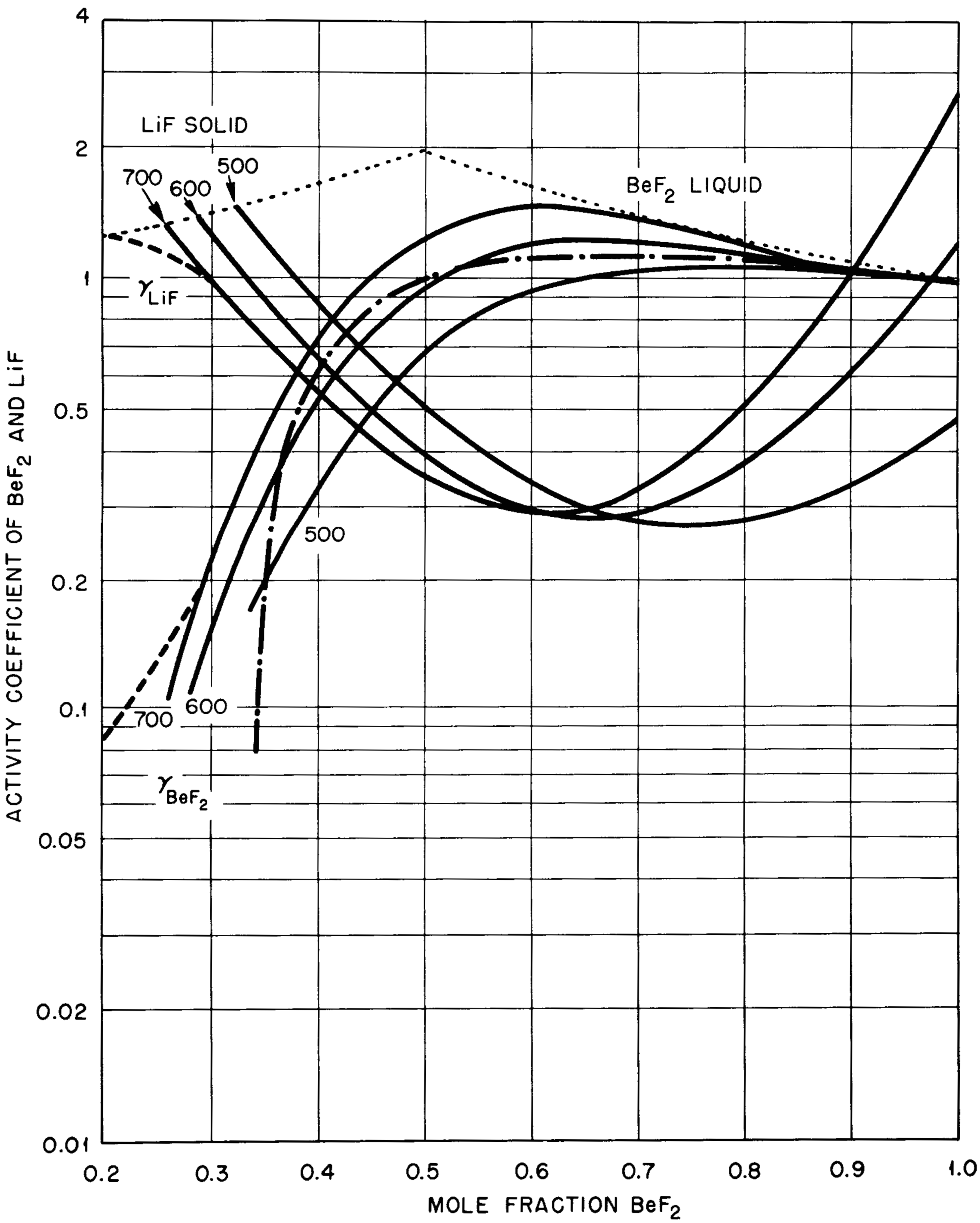

Fig. 20. Activity Coefficients of LiF and BeF_2 in Mixtures.

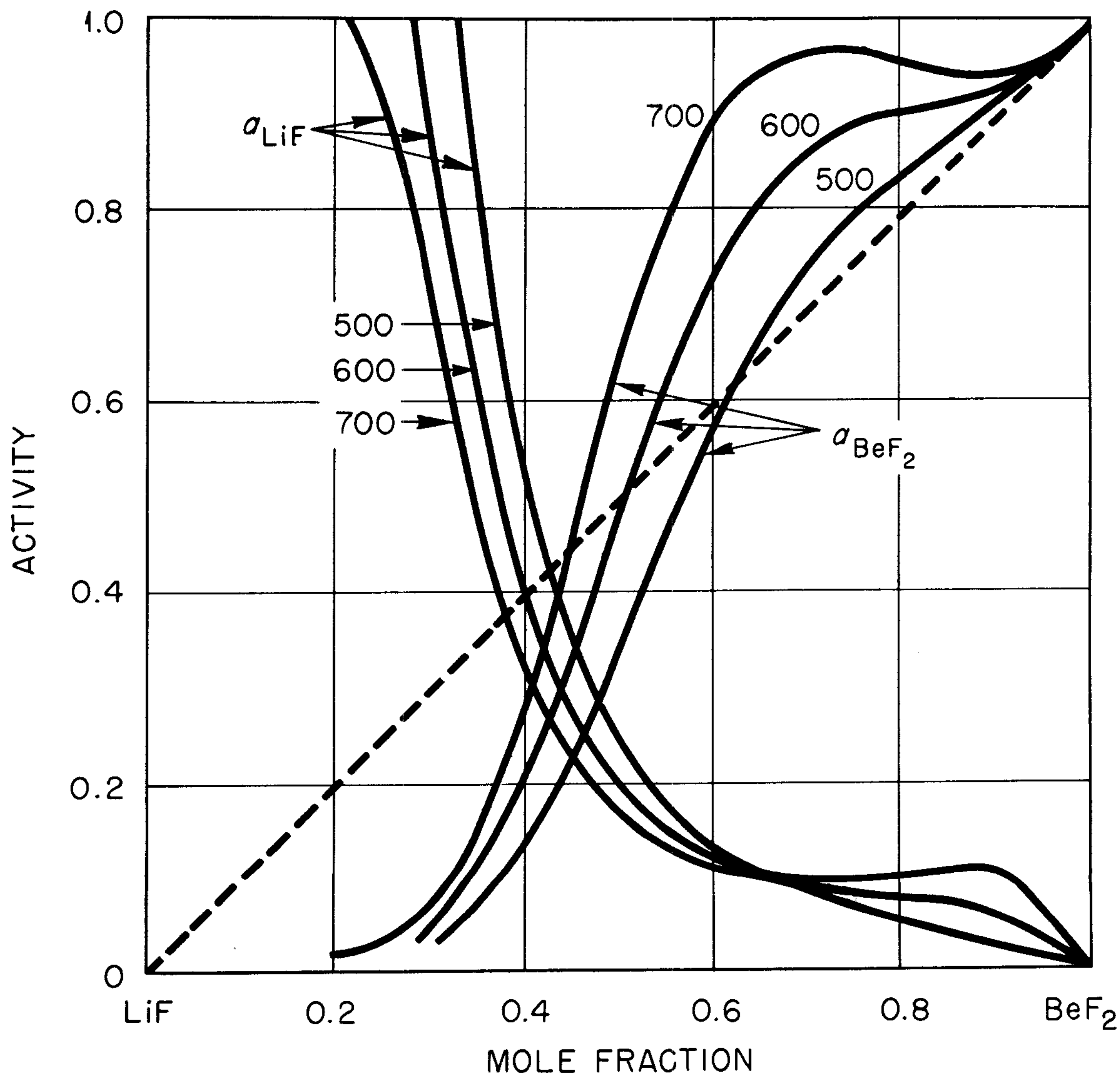

Fig. 21. Thermodynamic Activities of LiF and BeF_2 in Mixtures.

Thermodynamics of $BeF_2(\underline{l})$

From the correlation of Q as a function of temperature and composition for the reaction

$BeF_2(\underline{l}) + H_2O(g) \rightleftharpoons BeO(s) + 2HF(g)$

involving pure liquid BeF_2, the equation expressing equilibrium constant values is

$\log K_a = (3.900 \pm 0.019) - (4418 \pm 17)(1/T)$

or

$\Delta F = -(17.85 \pm 0.09)T + (20{,}217 \pm 78)$.

From this equation, the $\Delta H_{reaction}$, and $\Delta S_{reaction}$ are 20.22 kcal/mole BeF_2 and 17.85 eu/mole BeF_2, respectively. As indicated in Chapter I, the thermodynamic values of $H_2O(g)$, HF(g), and BeO(s) are fairly well established. Thus, by using these values and the relationships from the above reaction, new values for ΔF^o_f, ΔH^o_f, ΔS^o_f, and S^o of $BeF_2(\underline{l})$ can be calculated. These are tabulated below along with the reported values from JANAF.[4]

$BeF_2(\underline{l})$ at	800°K JANAF	800°K Present Work	1000°K JANAF	1000°K Present Work
ΔF^o_f	-213.57	-213.60	-207.08	-208.46
ΔH^o_f	-240.11	-234.49	-238.92	-234.26
ΔS^o_f	-33.18	-26.11	-31.84	-25.80
S^o	30.66	37.74	35.36	41.35

While the present values are not in close agreement with the published values, they provide the most direct experimental method of evaluating the thermodynamic properties of $BeF_2(\underline{l})$. If these values for $BeF_2(\underline{l})$ are combined with the data of Greenbaum, et al.[10] for the vapor pressures of $BeF_2(\underline{l})$, new values for the thermodynamics of $BeF_2(g)$ can be obtained. These are tabulated below along with the values reported in JANAF.

$BeF_2(g)$ at	800°K JANAF	800°K Present Work	1000°K JANAF	1000°K Present Work
ΔF^o_f	-192.02	-191.31	-192.92	-193.91
ΔH^o_f	-192.29	-181.24	-192.63	-181.01
ΔS^o_f	-0.34	1.25	-0.71	1.29
S^o	63.50	76.4	66.49	80.1

The values of ΔH^o_f and S^o of $BeF_2(g)$ reflect significant difference between these results and those of Greenbaum, et al.[5] for the reaction

$$BeO(s) + 2HF(g) \rightleftharpoons BeF_2(g) + H_2O(g) \quad .$$

Since the values of ΔF^o_f from the two methods are in reasonable agreement, the greatest source of difference must be in the temperature dependence of the equilibrium quotients.

In addition to the thermodynamic properties of $BeF_2(l)$ and $BeF_2(g)$, the present study provides a means of determining thermodynamic properties of $BeF_2(s)$ through determination of the heat of fusion, ΔH_{fusion}. If pure solid BeF_2 separates upon cooling the LiF-BeF_2 system, the relation between a_{BeF_2} and the freezing temperature T at a given concentration X_{BeF_2} in the liquid is given by

$$d(\ln a_{BeF_2}) = d \ln (X_{BeF_2}\gamma_{BeF_2}) = (\Delta H_{fusion}/RT^2)\, dT \quad .$$

If ΔH_{fusion} is independent of T, then

$$\ln (X_{BeF_2}\gamma_{BeF_2}) = -(\Delta H_{fusion}/R)(1/T - 1/T^o)$$

with T_o as the melting point of pure BeF_2. The values of T for various compositions X_{BeF_2} were tabulated from the published phase diagram.[2] The values of γ_{BeF_2} were calculated for the specified temperatures and compositions from the derived expression given earlier. A plot of

$$\log (X_{BeF_2}\gamma_{BeF_2}) \text{ vs } 1/T$$

is given in Figure 22. The slope of this correlation leads to a value of 11 ± 3 kcal/mole for ΔH_{fusion}. The other lines presented in the figure include: the line obtained if ΔH_{fusion} = 1.6 kcal/mole; the line obtained if $\gamma_{BeF_2} = 1$ (i.e., log X_{BeF_2} vs 1/T); the line obtained if ΔH_{fusion} = 7.5 kcal/mole.

The value of 1.6 kcal/mole for ΔH_{fusion} was included for comparison because this value was recently reported[57] from comparison of ΔH_{subl} with ΔH_{vap}. Both measurements were from the variation of vapor pressures with temperature over solid and liquid, respectively. One discrepancy in these data, which is not mentioned in the article, is that the vapor pressures reported for the solid near the melting point are greater than the extrapolated vapor pressures of the liquid by a factor of three. The fact that the reported vapor pressures for the solid and liquid do not intersect even near the melting point is indicative of a systematic error in one or the other sets of measurements since both sets seem internally consistent.

The value of 7.5 kcal/mole for ΔH_{fusion} was included for comparison because this is the value obtained if ΔH_{fusion} is determined to be the difference between ΔH^o_f of BeF_2(l) calculated here and ΔH^o_f of BeF_2(s) reported in JANAF.[4] The ΔH^o_f of BeF_2(s) is based on the heat of solution measurements on BeO and BeF_2 in aqueous HF by Kolesov, et al.[58] Since there is some question regarding the accuracy of the reported results, the value of ΔH_{fusion} is about 7.5 ± 6 kcal/mole. Although the present results do not definitely establish the value of ΔH_{fusion} of BeF_2(s), they do tend to support the value of 12 kcal/mole reported in JANAF rather than that of 2 kcal/mole used in the JANAF tabulation or the more recently reported value of 1.6 kcal/mole.

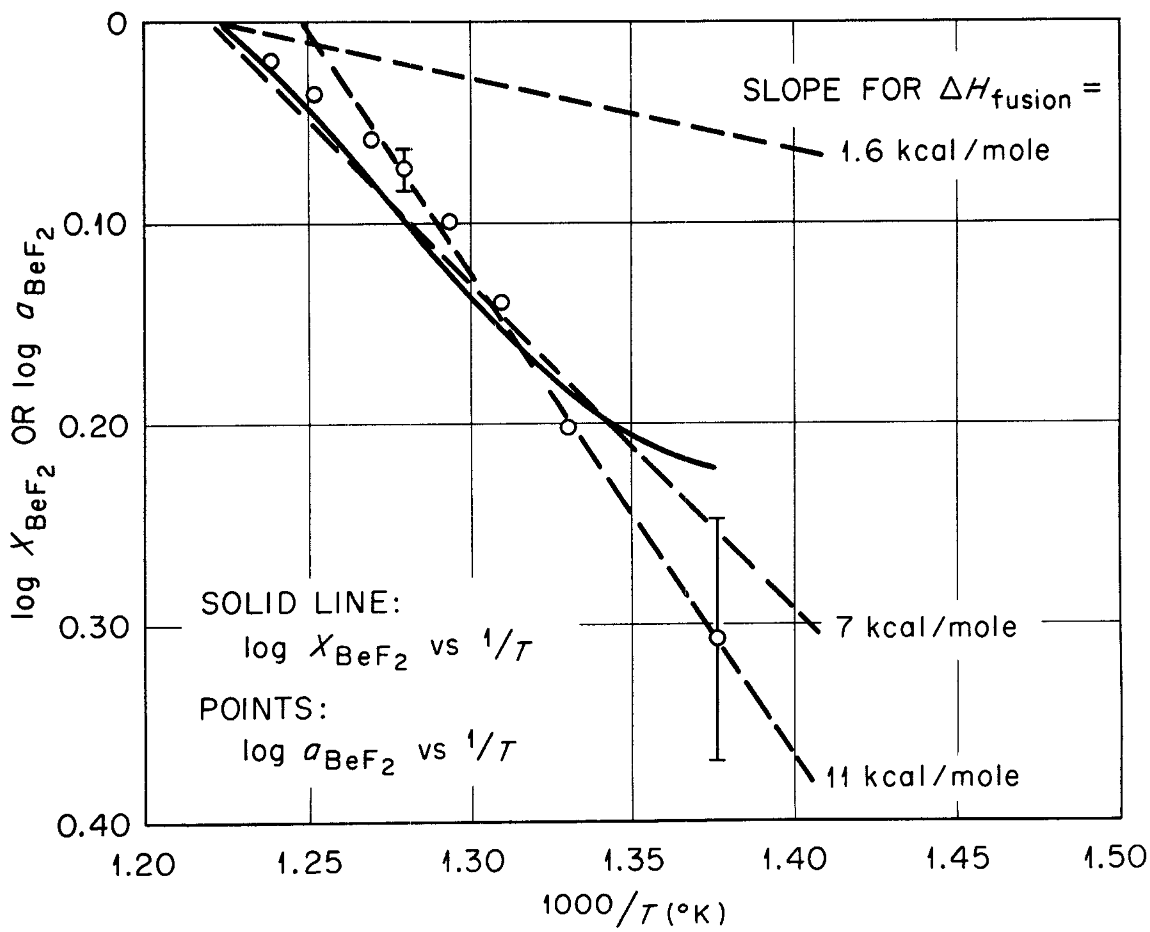

Fig. 22. Heat of Fusion of BeF_2 from Activities at Freezing Temperatures.

Correlation of Q_A

The values of Q_A reported in Tables 3 and 4 were correlated with respect to temperature and composition. Visual inspection indicated that the values exhibited linear dependence on both temperature and composition.

The general equation for the correlation was obtained in the following manner:

(1) The values at each temperature were grouped to determine the slope of the line

$$\log Q_A = (\text{slope})X_{BeF_2} + \text{constant} \quad .$$

The least-square slopes at the five experimental temperatures were as follows:

Temp (°C)	Slope	No. of Values
500	2.695 ± 0.473	6
550	2.480 ± 0.357	11
600	3.123 ± 0.174	22
650	2.847 ± 0.207	16
700	2.775 ± 0.175	20

(2) Using weights equal to the number of data points for each line, the average slope was determined to be 2.843. All of the values agree with this slope within the quoted standard deviations except the value at 600° which is slightly larger.

(3) With the slope of all lines set at 2.843, the value of $\log Q_A$ at the composition intercept ($X_{BeF_2} = 0$) was determined at each temperature to give best fit to data as shown below:

Temp (^{o}C)	log Q_A	Temp (^{o}C)	log Q_A
500	-3.138	650	-2.757
550	-3.056	700	-2.638
600	-2.893		

(4) These end values were used to calculate the function of log Q_A vs 1/T, which is log Q_A = -2085(1/T) - 0.503. Thus, the best smoothed values for Q_A over the experimental range are given by

$$\log Q_A = 2.843X_{BeF_2} - 2085(1/T) - 0.503 \quad .$$

The observed values of Q_A and the calculated values based on the above function are shown in Figure 23. Since the data at the various temperatures overlap, the coordinates are specified for each line independently. The standard deviations of the points are not shown in the figure, but most points agree within the quoted standard deviations in Table 3. The values determined in BeO-saturated melts are shown by closed symbols and those in unsaturated melts by open symbols.

The concentration of hydroxide present at equilibrium for known partial pressures of HF and H_2O above a melt of known composition could be calculated using the relationship for Q_A,

$$Q_A = (P_{HF}/P_{H_2O})[OH^-] = \text{mole/kg} \quad ,$$

derived from the reaction

$$H_2O(g) + F^-(soln) \rightleftharpoons HF(g) + OH^-(soln) \quad .$$

Other equilibria involving hydroxide were also introduced in Chapter I. One reaction is

$$H_2O(g) + O^{2-}(soln) \rightleftharpoons 2OH^-(soln)$$

from which

$$Q_B = [OH^-]^2/P_{H_2O} = (Q_A)^2/Q$$

for a BeO-saturated melt. Since expressions for both Q and Q_A are known, the values of Q_B can be derived. If the equation given earlier for $\log (Q/X_{BeF_2})$ is rearranged in terms of X_{BeF_2} rather than X_{LiF}, the following equation is obtained

$$\log (Q/X_{BeF_2}) = -0.940 - 4596(1/T) + [35.00 - 10168(1/T)]X_{BeF_2} + [-68.14 + 26132(1/T)]X^2_{BeF_2} + [50.64 - 21048(1/T)]X^3_{BeF_2} + [-12.66 + 5262(1/T)]X^4_{BeF_2} .$$

If this equation is combined with the expression for Q_A, the following holds:

$$\log Q_B = 2 \log Q_A - \log Q = -0.066 + 426(1/T) + [-29.31 + 10168(1/T)]X_{BeF_2} + [68.14 - 26132(1/T)]X^2_{BeF_2} + [-50.64 + 21048(1/T)]X^3_{BeF_2} + [12.66 - 5262(1/T)]X^4_{BeF_2} - \log X_{BeF_2} .$$

The parameters of the equation

$$\log Q_B = \text{slope}(1/T) + \text{constant}$$

are tabulated below for various experimental compositions:

X_{BeF_2}	Slope	Constant	X_{BeF_2}	Slope	Constant
0.30	1649	-3.367	0.60	983	-2.197
0.333	1629	-3.513	0.70	695	-1.370
0.40	1524	-3.415	0.80	456	-0.571
0.50	1279	-2.925			

Another reaction involving hydroxide is

$$HF(g) + O^{2-}(soln) \rightleftharpoons OH^-(soln) + F^-(soln)$$

from which

$$Q_C = [OH^-]/P_{HF} = Q_A/Q \text{ for a BeO-saturated melt.}$$

Therefore,

$$\log Q_C = \log Q_A - \log Q = 0.437 + 2511(1/T) + [-32.16 + 10168(1/T)]X_{BeF_2}$$
$$+ [68.14 - 26132(1/T)]X^2_{BeF_2} + [-50.64 + 21048(1/T)]X^3_{BeF_2}$$
$$+ [12.66 - 5262(1/T)]X^4_{BeF_2} - \log X_{BeF_2} \quad .$$

The parameters of the equation

$$\log Q_C = \text{slope}(1/T) + \text{constant}$$

are tabulated below for various experimental compositions:

X_{BeF_2}	Slope	Constant	X_{BeF_2}	Slope	Constant
0.30	3734	-3.819	0.60	3068	-3.404
0.333	3714	-3.960	0.70	2780	-2.862
0.40	3609	-4.052	0.80	2541	-2.348
0.50	3364	-3.847			

If the above functions are used to evaluate equilibrium quotients for 0.333 BeF_2, the values obtained and ΔH_r based on integration of the van't Hoff equation are given below:

Equilibrium Quotients	Q at 800°K	Q at 1000°K	ΔH_r (kcal/mole)
Q_A	6.90×10^{-3}	2.29×10^{-2}	9.5
Q_B	3.33×10^{-2}	1.30×10^{-2}	-7.5
Q_C	4.80	5.70×10^{-1}	-17.0

Each of these equilibrium quotients is in terms of moles of hydroxide per kilogram of melt due to experimental expediency, but they could be converted to the mole fraction scale by dividing each by moles of solvent per kilogram of melt. This conversion would not affect the ΔH_r values. The above values of equilibrium quotients involving hydroxide may not provide enough information to establish the complete nature of hydroxide in the LiF-BeF_2 system, but they do allow quantitative prediction of the extent of conversion of oxide to hydroxide, which is the principal controlling step in the removal of oxide by HF-sparging.

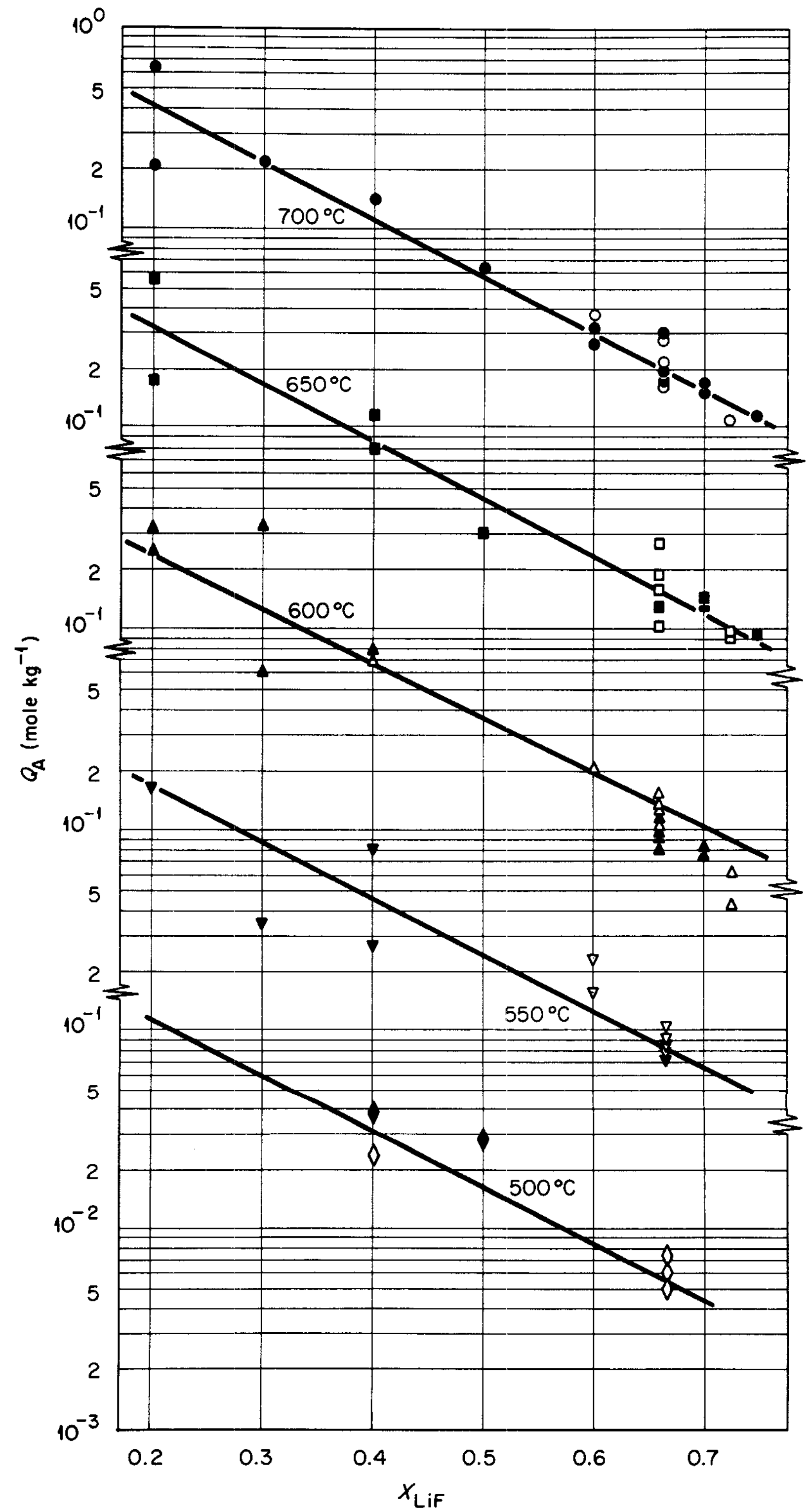

Fig. 23. Correlation of log Q_A as a Function of Composition and Temperature.

Correlation of Q_O

The values of Q_O reported in Table 4 were correlated as a function of temperature at each composition. The parameters of the equation

$\log Q_O = \text{slope}(1/T) + \text{constant}$

are given below:

X_{BeF_2}	slope $\pm\ \sigma$	constant $\pm\ \sigma$	No. of Points
0.273	-8158 ± 500	4.791 ± 0.549	5
0.333	-8637 ± 349	5.673 ± 0.401	24
0.400	-9032 ± 706	7.026 ± 0.812	4

The values of Q_O and the corresponding correlations as a function of temperature are shown in Figure 24. Since the two values for 0.60 BeF_2 would not yield a line with slope or intercept similar to the others, a line was drawn parallel to the line at 0.40 BeF_2 so as to be equidistant between the two values at 0.60 BeF_2.

Measurement of Q_O at high BeF_2 compositions is limited because the rate of change in P_{HF} and P_{H_2O} with respect to W approaches the precision of measurements of the low P_{H_2O} and high P_{HF} encountered. Although additional experiments were performed on 0.60 BeF_2, convergence in the solution of equations to obtain Q_O, Q_A, r_o, and s_o was not reached due to the experimental limitation mentioned above.

The data on unsaturated melts were neither extensive nor precise enough to permit a meaningful correlation with respect to melt composition. However, the solubility of BeO could be determined by comparison of these measurements with those of BeO-saturated melts.

As described in Chapter I,

$Q_0 = (P_{HF})^2[O^{2-}]/P_{H_2O}$

and since for a BeO-saturated melt

$(P_{HF})^2/P_{H_2O} = Q$,

$[O^{2-}]_{sat} = Q_0/Q$.

The value of Q at BeO saturation was established in the earlier work. Division of Q_0 by Q for the same melt composition and temperature would provide the concentration of oxide at saturation.

The values of Q needed to determine BeO solubility are given by the following parameters for the equation

log Q = slope(1/T) + constant:

X_{BeF_2}	Slope	Constant
0.273	-5900	4.150
0.333	-5674	4.233
0.400	-5565	4.528
0.600	-5274	4.745

The solubilities in moles per kilogram of oxide in the various experiments, as determined from Q_0/Q using observed Q_0 values and smoothed Q values from the above relation, are given in Table 8. Since

$[O^{2-}] = Q_0/Q$,

$\log [O^{2-}] = \log Q_0 - \log Q$,

and as both log Q_0 and log Q show linear dependence with (1/T), log $[O^{2-}]$ should also show linear dependence. Combination of the parameters for Q_0 and Q leads to

$\log [O^{2-}]$ = slope(1/T) + constant:

X_{BeF_2}	Slope	Constant
0.273	-2258	0.641
0.333	-2963	1.440
0.400	-3467	2.498

Each of the observed solubilities is shown in Figure 25 along with the calculated expressions for the three compositions tabulated above.

Since many more experiments were performed on 0.333 BeF_2, the range of measured solubilities at each temperature provides a rough measure of just how well these are known. A clear trend of solubility with temperature is shown:

500°C	(2.5 - 6.0) $\times 10^{-3}$ moles/kg
600	(7.2 - 11.8) $\times 10^{-3}$
700	(2.3 - 3.8) $\times 10^{-2}$

The other values indicate that BeO solubility may be increasing slightly with BeF_2, but the increase is not very great.

Table 8. Solubility of BeO in Molten LiF-BeF_2 System

X_{BeF_2}	Temp (°C)	Run No.	$[O^{2-}]_{sat}$ (mole/kg)	X_{BeF_2}	Temp (°C)	Run No.	$[O^{2-}]_{sat}$ (mole/kg)
0.273	600	529	1.30×10^{-2}	0.333	600	309	1.16×10^{-2}
0.273	600	539	1.04×10^{-2}	0.333	600	310	8.70×10^{-3}
0.273	650	535	1.46×10^{-2}	0.333	650	319	2.14×10^{-2}
0.273	650	537	1.48×10^{-2}	0.333	650	501	1.68×10^{-2}
0.273	700	533	2.13×10^{-2}	0.333	650	513	2.08×10^{-2}
0.333	500	313	6.67×10^{-3}	0.333	650	514	8.16×10^{-3}
0.333	500	314	2.53×10^{-3}	0.333	651	527	1.98×10^{-2}
0.333	500	503	5.72×10^{-3}	0.333	700	307	2.61×10^{-2}
0.333	544	509	5.85×10^{-3}	0.333	700	311	3.78×10^{-2}
0.333	550	315	7.25×10^{-3}	0.333	700	523	2.77×10^{-2}
0.333	550	316	5.09×10^{-3}	0.333	700	524	2.30×10^{-2}
0.333	550	511	8.45×10^{-3}	0.400	550	619	2.53×10^{-2}
0.333	550	525	1.17×10^{-2}	0.400	550	625	1.56×10^{-2}
0.333	600	301	9.47×10^{-3}	0.400	604	621	3.23×10^{-2}
0.333	600	302	7.18×10^{-3}	0.400	702	627	9.02×10^{-2}
0.333	600	303	1.18×10^{-2}	0.600	500	607	2.39×10^{-2}
0.333	600	305	1.11×10^{-2}	0.600	600	611	1.78×10^{-2}
0.333	600	306	7.23×10^{-3}				

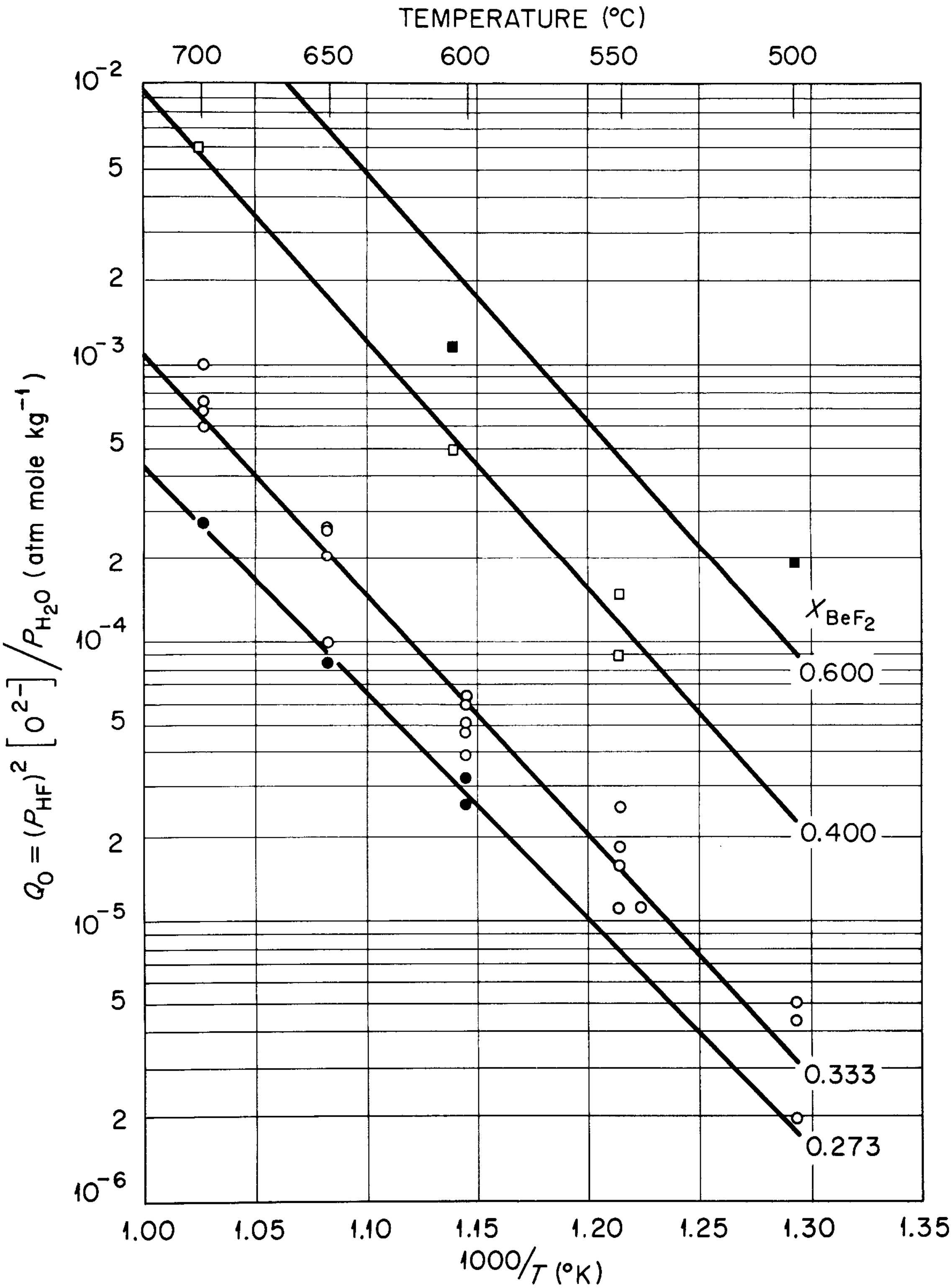

Fig. 24. Correlation of log Q_0 as a Function of Temperature for Various Melt Compositions.

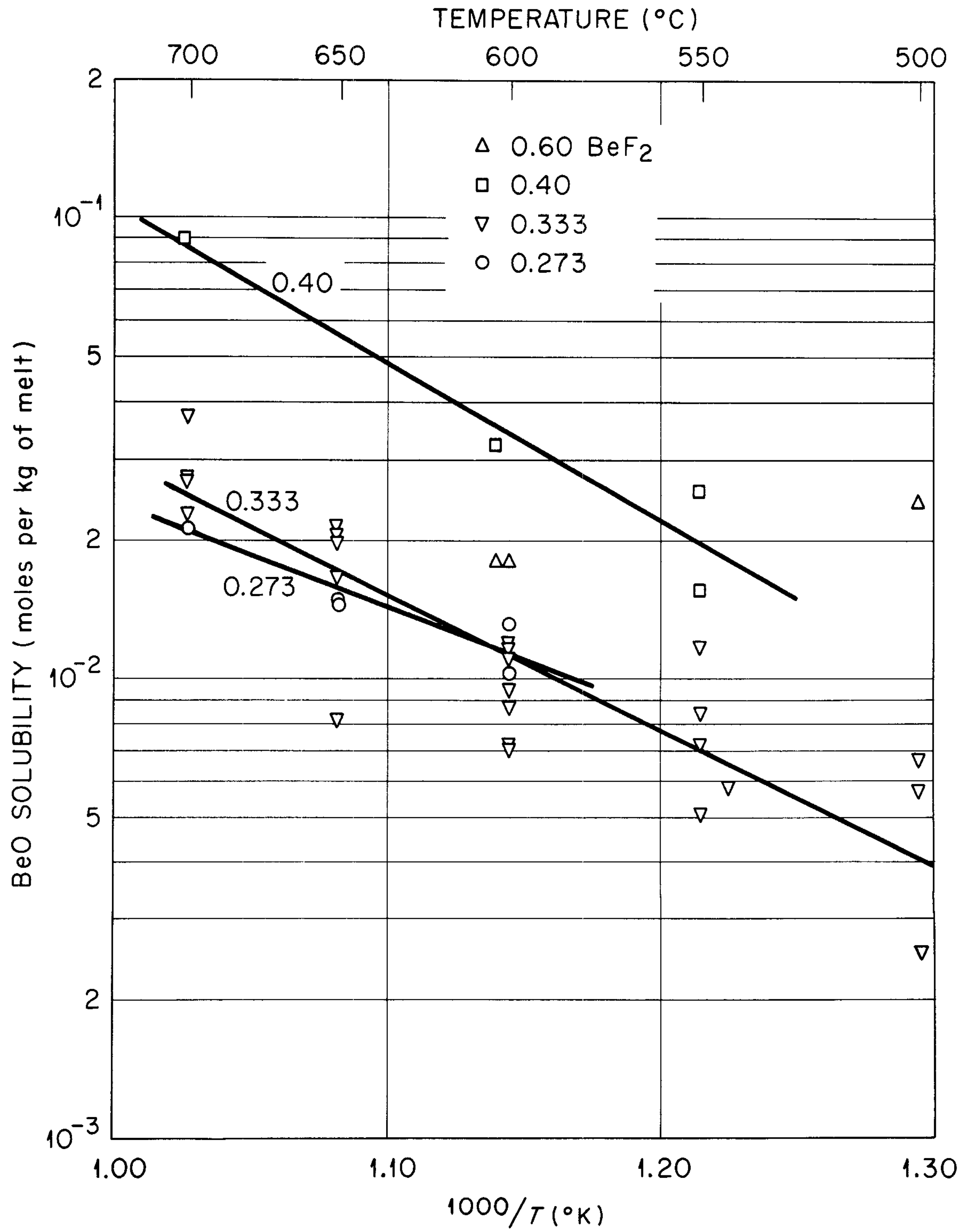

Fig. 25. Solubility of BeO as a Function of Temperature for Various Melt Compositions.

Summary

These studies of equilibria between HF, H_2O, and molten LiF-BeF_2 had a twofold purpose: (1) to determine as much as possible about the thermodynamics of the LiF-BeF_2 system, and (2) to determine as much as possible about the oxide chemistry of the system.

The thermodynamic activities of LiF and BeF_2 were determined as a function of temperature and composition. The major characteristics of the system include:

(1) Positive deviations from ideality at high BeF_2 concentrations.

(2) Increasing positive deviations of the activity with temperature indicating the probability of a miscibility gap near 700°C.

(3) Combination of activities with previously published phase diagram indicate a higher value for ΔH_{fusion} of BeF_2 than currently used in thermodynamic compilations.

(4) Activities determined with BeO present are in agreement with limited previous data.

Extrapolation of data to pure liquid BeF_2, which is inaccessible experimentally because of high viscosity, provides a means of determining thermodynamic properties of BeF_2(l).

The equilibria between H_2O, HF, and dissolved hydroxide were studied by estimating, from material balance calculations on the gas phase, the amount of OH^- formed in or removed from the melt upon reaction. The results from measurements on both BeO-saturated and unsaturated melts were consistent. The results indicate that significant amounts of hydroxide were formed in the presence of 0.01 atm HF, but OH^- could not exist in the absence of HF and H_2O.

The equilibria between H_2O, HF, and dissolved oxide were also evaluated with melts not saturated with BeO. The solubility of BeO was estimated by combination of these results with those for BeO-saturated melts.

These results suggest methods to attack at least two allied problems:

(1) Determination of thermodynamic activities of other fluorides dissolved in the LiF-BeF_2 system; and

(2) Determination of the amount of oxide in fluoride melts.

The procedure used here to determine activities of BeO-saturated melts could be applied to melts saturated with other sparingly soluble oxides. Variation in Q with amount of corresponding fluoride dissolved in melt would provide information necessary to calculate change in activities.

The experiments performed here on unsaturated melts required determination of the concentration of oxide present at the start of an experiment in addition to the equilibrium quotients. If oxide analyses were required on similar melts, the equilibrium quotients could be evaluated and treated as fixed parameters, thus the variation in effluent P_{HF} and P_{H_2O} to a small value of W (about 1.0) could be used to calculate the amount of oxide in the sample prior to treatment. In principle, this should work no matter whether oxide were being added or removed. The presence of saturating solid oxide would complicate the analysis, but the total amount of oxide present could be determined by treatment with anhydrous HF and measurement of the evolved H_2O.

BIBLIOGRAPHY

1. See: D. A. Everest, The Chemistry of Beryllium, Elsevier Publishing Company, New York, 1964, p. 38, for a survey of the chemistry and phase studies of beryllium fluoride.

2. R. E. Thoma et al., Reactor Chem. Div. Ann. Progr. Rept. Jan. 31, 1965, ORNL-3789, p. 3.

3. W. T. Ward and S. Cantor, Reactor Chem. Div. Ann. Progr. Rept. Jan. 31, 1964, ORNL-3591, p. 52.

4. JANAF Thermochemical Tables, The Dow Chemical Co., Midland, Michigan, 1963.

5. M. A. Greenbaum, M. L. Arin, and M. Farber, J. Phys. Chem. 67, 1191 (1963).

6. K. A. Sense, M. J. Snyder, and J. W. Clegg, J. Phys. Chem. 58, 223 (1954).

7. K. A. Sense and R. W. Stone, J. Phys. Chem. 62, 453 (1958).

8. A. V. Novoselova, F. Sh. Muratov, L. P. Reshetnikova, and I. V. Gordeev, Vestnik Moskov. Univ., Ser. Mat., Mekh., Astron., Fiz., Khim. 13, No. 6, 181 (1958).

9. N. E. Khandamirova, A. M. Evseev, G. V. Pozharskaya, E. A. Borisov, A. N. Nesmeyanov, and Ya. Il Gerasimov, Zhur. Neorg. Khim. 4, 2192 (1959).

10. M. A. Greenbaum, J. N. Foster, M. L. Arin, and M. Farber, J. Phys. Chem. 67, 36 (1963).

11. E. U. Franck and W. Spalthoff, Zeit. Elektrochem. 61, 348 (1957).

12. J. W. Armitage, P. Gray, and P. G. Wright, J. Chem. Soc. 1963, 1796.

13. R. M. Yabroff, J. C. Smith, and E. H. Lightcap, J. Chem. Eng. Data 9, 178 (1964).

14. G. T. Armstrong, "Fluorine Flame Calorimetry", p. 144 in Experimental Thermochemistry, vol. II, ed. by H. A. Skinner, Interscience, New York, 1962.

15. H. M. Feder, W. N. Hubbard, S. S. Wise, and J. L. Margrave, J. Phys. Chem. 67, 1148 (1963).

16. Hydrofluoric Acid, Anhydrous and Aqueous, p. 8, Stauffer Chemical Company, New York, 1957.

17. M. Blander, "Thermodynamic Properties of Molten Salt Solutions", p. 127 in Molten Salt Chemistry, ed. by M. Blander, Wiley, New York, 1964.

18. M. Temkin, Acta Physicochim. URSS 20, 411 (1945).

19. W. D. Treadwell and A. Cohen, Helv. Chim. Acta 22, 433, 1341 (1939).

20. I. L. Reznikov, Zh. Prikl Khim. 23, 897 (1950).

21. C. M. Blood, F. F. Blankenship, W. R. Grimes, and G. M. Watson, Abstr. 7th Intern. Congr. Coordination Chem., 1962.

22. J. Berkowitz and W. A. Chupka, Ann. N. Y. Acad. Sci. 79, 1073 (1960).

23. A. Büchler, Study of High Temperature Thermodynamics of Light Metal Compounds, Army Research Office (Durham), Prog. Report No. 7 (Contract No. DA-19-020-ORD-5584)(Sept. 30, 1963).

24. A. Büchler, Study of High Temperature Thermodynamics of Light Metal Compounds, Army Research Office (Durham), Prog. Report No. 9 (Contract No. DA-19-020-ORD-5584)(March 31, 1964).

25. G. M. Watson, R. B. Evans, III, W. R. Grimes, and N. V. Smith, J. Chem. Eng. Data 7, 285 (1962).

26. W. J. Burkhard and J. D. Corbett, J. Am. Chem. Soc. 79, 6361 (1957).

27. J. H. Shaffer, W. R. Grimes, and G. M. Watson, J. Phys. Chem. 63, 1999 (1959).

28. J. H. Shaffer and G. M. Watson, Reactor Chem. Div. Ann. Progr. Rept. Jan. 31, 1960, ORNL-2931, p. 31.

29. G. Goldberg, A. S. Meyer, Jr., and J. C. White, Anal. Chem. 32, 314 (1960).

30. C. V. Banks, J. W. O'Laughlin, and G. J. Kamin, Anal. Chem. 32, 1613 (1960).

31. B. Porter and E. A. Brown, Determination of Oxide Solubility in Molten Fluorides, U.S. Bur. Mines, Report of Invest. No. 5878 (1961).

32. G. Delarue, Silicates Ind. 27, 69 (1961).

33. G. Delarue, Chim. Anal. (Paris) 44, 91 (1962).

34. (a) C. F. Baes, Jr., Molten-Salt Reactor Program Semiann. Progr. Rept. Jan. 31, 1963, ORNL-3419, p. 110.
(b) C. F. Baes, Jr., J. H. Shaffer, and H. F. McDuffie, Trans. Am. Nuclear Soc. 6, 393 (1963).

35. (a) J. E. Eorgan, F. A. Doss, G. J. Nessle, J. Truitt, and C. R. Croft, Reactor Chem. Div. Ann. Progr. Rept. Jan. 31, 1960, ORNL-2931, p. 64.
(b) W. R. Grimes, "Materials Problems in Molten Salt Reactors," pp. 96-129 in Materials and Fuels for High-Temperature Nuclear Energy Applications, ed. by L. R. Zumwalt, M.I.T. Press, Cambridge, Mass., 1964.

36. A. L. Mathews, C. F. Baes, Jr., and M. K. Kemp, Reactor Chem. Div. Ann. Progr. Rept. Jan. 31, 1964, ORNL-3591, p. 46.

37. G. W. Thomson, "Determination of Vapor Pressure," pp. 446-451 in Physical Methods of Organic Chemistry, vol. I-part I, 3d ed., ed. by A. Weissberger, Interscience, New York, 1959.

38. D. R. Cuneo and S. Cantor, Reactor Chem. Div. Ann. Progr. Rept. Jan. 31, 1963, ORNL-3417, p. 46.

39. L. P. Firsova and A. N. Nesmeyanov, Zhur. Fiz. Khim. 34, 2615 (1960).

40. P. A. Munter, O. T. Aepli, and R. A. Kossatz, Ind. Eng. Chem. 41, 1504 (1949).

41. H. A. Laitinen, Chemical Analysis, p. 421, McGraw-Hill, New York, 1960.

42. G. Nernitz, Chemiker-Ztg. 82, 222 (1958).

43. G. Long, Reactor Chem. Div. Ann. Progr. Rept. Jan. 31, 1965, ORNL-3789, p. 65.

44. W. T. Ward, R. A. Strehlow, W. R. Grimes, and G. M. Watson, Solubility Relations Among Some Fission Product Fluorides in NaF-ZrF_4-UF_4 (50-46-4), ORNL-2421 (Jan. 15, 1958).

45. J. J. Lingane, Electroanalytical Chemistry, 2nd ed., p. 286, Interscience, New York, 1958.

46. S. Dushman, Scientific Foundations of Vacuum Technique, pp. 607-618, Wiley, New York, 1949.

47. H. H. Stone and C. F. Baes, Jr., Reactor Chem. Div. Ann. Progr. Rept. Jan. 31, 1965, ORNL-3789, p. 72.

48. H. D. Young, Statistical Treatment of Experimental Data, p. 96, McGraw-Hill, New York, 1962.

49. H. Margenau and G. M. Murphy, The Mathematics of Physics and Chemistry, p. 486, D. Van Nostrand, Princeton, N. J., 1956.

50. H. Flood and S. Urnes, Z. Elektrochem. 59, 834 (1955).

51. T. Førland, "Thermodynamic Properties of Fused-Salt Systems," p. 156 in Fused Salts, ed. by B. R. Sundheim, McGraw-Hill, New York, 1964.

52. S. Glasstone, Textbook of Physical Chemistry, 2nd ed., p. 721, D. Van Nostrand, Princeton, N. J., 1946.

53. E. Thilo and H. A. Lehmann, Z. Anorg. Allgem. Chem. 258, 332 (1949).

54. D. M. Roy, R. Roy, and E. F. Osborn, J. Am. Cer. Soc. 37, 300 (1954).

55. J. E. Ricci, "Phase Diagrams of Fused Salts," p. 269 in Molten Salt Chemistry, ed. by M. Blander, Interscience, New York, 1964.

56. Molten-Salt Reactor Program Semiann. Progr. Rept. July 31, 1963, ORNL-3529, p. 106.

57. J. A. Blauer, M. A. Greenbaum, and M. Farber, J. Phys. Chem. 69, 1069 (1965).

58. V. P. Kolesov, M. M. Popov, and S. M. Skuratov, Zhur. Neorg. Khim. 4, 1233 (1959).

APPENDIX A

Partial Pressures from Studies on BeO-Saturated Melts

The experiments are arranged according to a three digit "Run No." which indicates chronological order. There are three series (100, 200, 400) included. A brief history of each series is given in Chapter III.

The information provided for each experiment includes:

Run No.

Composition of melt (expressed as mole fraction BeF_2)

Temperature of melt, $^{\circ}C$

w = weight of melt, kg

T = temperature of wet-test meter, $^{\circ}K$

a = influent P_{HF}, atm x 10^3

c = influent P_{H_2O}, atm x 10^3

The effluent partial pressures, P_{HF} = x and P_{H_2O} = y, are tabulated (atm x 10^3) with the corresponding initial and final values of W (V/wRT). Note that the use of W allows consistent comparison of experiments independent of the weight of melt or temperature of gas-measurement. The equilibrium quotients evaluated from each experiment are presented in Table 3.

Run No. 101

0.316 BeF_2 at 700°C, w = 0.305, T = 300°K, a = 15.45, c = 5.48

W_i	W_f	x	y	W_i	W_f	x	y
0.000	0.075	8.22		0.935	1.030		6.51
0.075	0.103	11.32		1.030	1.119		7.03
0.103	0.181	11.90		1.119	1.203		7.42
0.181	0.253	12.76		1.203	1.286		7.49
0.253	0.325	12.88		1.286	1.374		7.09
0.325	0.394	13.38		1.374	1.457		7.54
0.394	0.462	13.78		1.457	1.538		7.67
0.462	0.526	14.34		1.545	1.641	12.88	
0.526	0.589	14.64		1.641	1.712	13.00	
0.589	0.655	14.20		1.712	1.787	12.42	
0.733	0.836		6.00	1.787	1.859	12.88	
0.836	0.935		6.28				

Run No. 103

0.316 BeF_2 at 700°C, w = 0.305, T = 300°K, a = 15.15, c = 5.14

W_i	W_f	x	y	W_i	W_f	x	y
0.000	0.203	4.57		1.838	2.046		6.59
0.203	0.343	6.63		2.046	2.234		6.60
0.343	0.437	9.87		2.234	2.414		6.93
0.437	0.531	9.87		2.414	2.596		6.86
0.531	0.600	10.68		2.596	2.772		7.09
0.600	0.677	12.00		2.772	2.916		6.93
0.677	0.758	11.50		2.916	3.061		6.87
0.758	0.862	11.90		3.130	3.177	13.25	
0.862	0.938	12.09		3.177	3.248	13.00	
0.938	1.016	12.00		3.248	3.342	13.16	
1.016	1.092	12.20		3.342	3.415	12.76	
1.092	1.170	12.20		3.415	3.485	13.25	

Run No. 105

0.316 BeF_2 at 600°C, w = 0.305, T = 301°K, a = 17.63, c = 4.77

W_i	W_f	x	y	W_i	W_f	x	y
0.929	1.002		8.50	1.709	1.805	6.45	
1.002	1.075		8.53	1.805	1.902	6.37	
1.075	1.148		8.82	1.902	1.995	6.63	
1.148	1.219		8.91	2.190	2.390		9.32
1.219	1.287		9.10	2.390	2.458		9.20
1.327	1.517	6.53		2.458	2.526		9.10
1.517	1.613	6.40		2.526	2.650		10.04
1.613	1.709	6.45					

Run No. 107

0.316 BeF_2 at 600°C, w = 0.305, T = 300°K, a = 11.58, c = 5.87

W_i	W_f	x	y	W_i	W_f	x	y
0.866	0.943		8.12	2.162	2.379	5.70	
0.943	1.016		8.50	2.379	2.543	5.63	
1.016	1.169		8.12	2.543	2.706	5.68	
1.169	1.323		8.13	2.706	2.872	5.60	
1.323	1.398		8.22	2.872	3.037	5.59	
1.398	1.546		8.45	3.170	3.284		7.68
1.546	1.695		8.34	3.284	3.442		7.88
1.732	1.891	5.80		3.442	3.599		7.91
1.891	2.052	5.75		3.599	3.759		7.79
2.052	2.162	5.64					

Run No. 109

0.316 BeF_2 at 600°C, w = 0.305, T = 300°K, a = 6.55, c = 7.75

W_i	W_f	x	y	W_i	W_f	x	y
0.866	1.012	6.36		2.531	2.677		9.26
1.012	1.149	6.72		2.677	2.794		9.71
1.149	1.286	6.75		2.794	2.918		10.04
1.399	1.547		8.41	2.918	3.040		10.18
1.547	1.688		8.80	3.064	3.163	6.20	
1.688	1.855		9.00	3.163	3.262	6.24	
1.898	2.044	6.37		3.262	3.362	6.20	
2.044	2.142	6.26		3.596	3.729		9.42
2.142	2.241	6.26		3.729	3.857		9.72
2.241	2.342	6.14		3.857	3.986		9.64

Run No. 111

0.316 BeF_2 at 600°C, w = 0.305, T = 300°K, a = 6.78, c = 9.23

W_i	W_f	x	y	W_i	W_f	x	y
0.799	0.943	6.45		2.205	2.302	6.41	
0.943	1.037	6.54		2.302	2.399	6.33	
1.037	1.135	6.34		2.464	2.600		9.14
1.135	1.230	6.46		2.600	2.737		9.10
1.265	1.403		9.07	2.737	2.875		9.08
1.403	1.541		9.03	2.875	3.010		9.22
1.541	1.675		9.28	3.010	3.146		9.13
1.675	1.810		9.26	3.146	3.282		9.21
1.810	1.926	6.53		3.282	3.418		9.13
1.926	2.019	6.63		3.463	3.555	6.75	
2.019	2.111	6.72		3.555	3.648	6.59	
2.111	2.205	6.57		3.648	3.742	6.59	

Run No. 113

0.316 BeF_2 at 500°C, w = 0.305, T = 300°K, a = 6.54, c = 9.33

W_i	W_f	x	y	W_i	W_f	x	y
0.799	0.862		9.95	2.168	2.284		10.83
0.862	0.996		9.26	2.331	2.449	2.60	
0.996	1.129		9.38	2.449	2.567	2.63	
1.129	1.258		9.66	2.567	2.683	2.67	
1.258	1.383		9.96	2.683	2.798	2.67	
1.432	1.674	2.55		2.824	2.943		10.43
1.674	1.794	2.57		2.943	3.060		10.70
1.812	1.931		10.46	3.060	3.175		10.83
1.931	2.050		10.45	3.175	3.233		10.68
2.050	2.168		10.60				

Run No. 115

0.316 BeF_2 at 550°C, w = 0.305, T = 300°K, a = 6.50, c = 9.58

W_i	W_f	x	y	W_i	W_f	x	y
0.932	1.090	3.92		2.398	2.546	4.17	
1.090	1.246	3.96		2.546	2.695	4.13	
1.246	1.401	3.99		2.695	2.847	4.07	
1.401	1.551	4.12		2.847	3.014	4.08	
1.551	1.705	4.00		3.014	3.151	4.04	
1.732	1.862		9.57	3.197	3.325		9.70
1.862	1.990		9.72	3.325	3.451		9.95
1.990	2.116		9.91	3.451	3.573		10.21
2.116	2.242		9.88	3.573	3.701		9.79
2.242	2.368		9.90	3.701	3.825		10.05

Run No. 117

0.316 BeF_2 at 500°C, w = 0.305, T = 300°K, a = 6.25, c = 9.22

W_i	W_f	x	y	W_i	W_f	x	y
1.332	1.452	2.57		2.322	2.437		10.91
1.452	1.572	2.57		2.437	2.551		10.93
1.572	1.815	2.54		2.551	2.677		10.85
1.815	1.935	2.57		2.731	2.848	2.63	
1.935	2.058	2.50		2.848	2.969	2.54	
2.091	2.205		10.92	2.969	3.090	2.55	
2.205	2.322		10.64				

Run No. 119

0.316 BeF_2 at 650°C, w = 0.305, T = 300°K, a = 6.20, c = 8.98

W_i	W_f	x	y	W_i	W_f	x	y
1.798	1.869	8.75		2.890	2.996	8.82	
1.869	1.939	8.83		2.996	3.099	8.92	
1.939	2.009	8.82		3.130	3.294		7.59
2.009	2.080	8.68		3.294	3.373		7.92
2.398	2.551		8.13	3.373	3.453		7.76
2.551	2.709		7.90	3.453	3.615		7.67
2.709	2.865		7.95				

Run No. 121

0.316 BeF_2 at 650°C, w = 0.305, T = 300°K, a = 6.04, c = 9.24

W_i	W_f	x	y	W_i	W_f	x	y
0.799	0.968		7.38	2.065	2.219		8.08
0.968	1.180		7.66	2.219	2.391		8.03
1.180	1.338		7.86	2.391	2.545		8.08
1.338	1.495		7.95	2.571	2.641	8.82	
1.495	1.651		7.99	2.641	2.747	8.70	
1.665	1.735	8.83		2.747	2.857	8.49	
1.735	1.809	8.36		2.857	2.965	8.54	
1.809	2.036	8.17		2.965	3.109	8.60	

Run No. 123

0.316 BeF_2 at 700°C, w = 0.305, T = 301°K, a = 6.23, c = 9.14

W_i	W_f	x	y	W_i	W_f	x	y
1.328	1.424		6.41	2.838	2.917	11.72	
1.424	1.519		6.50	2.917	2.996	11.66	
1.519	1.710		6.47	3.053	3.245		6.45
1.710	1.805		6.53	3.245	3.438		6.41
2.522	2.601	11.82		3.448	3.530	11.72	
2.601	2.680	11.66		3.530	3.609	11.83	
2.680	2.759	11.71		3.609	3.688	11.63	
2.759	2.838	11.79		3.688	3.768	11.63	

Run No. 125

0.333 BeF_2 at 600°C, w = 0.316, T = 300°K, a = 5.93, c = 9.14

W_i	W_f	x	y	W_i	W_f	x	y
0.900	1.035		8.68	1.671	1.760	6.68	
1.035	1.166		8.88	1.760	1.895	6.63	
1.166	1.301		8.70	1.895	1.984	6.63	
1.301	1.433		8.86	1.993	2.131		8.62
1.446	1.582	6.60		2.131	2.265		8.78
1.582	1.671	6.70					

Run No. 127

0.333 BeF_2 at 550°C, w = 0.316, T = 300°K, a = 6.00, c = 9.10

W_i	W_f	x	y	W_i	W_f	x	y
0.900	0.970	4.22		1.832	1.975	4.17	
0.970	1.113	4.18		1.975	2.113	4.32	
1.113	1.253	4.25		2.113	2.184	4.18	
1.286	1.416		9.12	2.250	2.382		9.04
1.416	1.549		9.00	2.382	2.512		9.13
1.549	1.680		9.09	2.512	2.643		9.12
1.680	1.810		9.20				

Run No. 129

0.333 BeF_2 at 500°C, w = 0.316, T = 300°K, a = 6.04, c = 8.97

W_i	W_f	x	y	W_i	W_f	x	y
0.064	0.174	2.71		2.186	2.305		9.72
0.174	0.283	2.72		2.305	2.423		9.87
0.283	0.393	2.71		2.423	2.537		10.10
0.450	0.585		8.57	2.537	2.655		9.84
0.585	0.719		8.70	2.655	2.772		9.97
1.157	1.283		9.18	2.828	2.933	2.84	
1.283	1.408		9.33	2.933	3.035	2.91	
1.543	1.649	2.80		3.035	3.139	2.87	
1.649	1.753	2.88		3.214	3.329		10.07
1.753	1.967	2.78		3.329	3.446		9.96
1.967	2.071	2.86		3.446	3.562		9.99

Run No. 201

0.800 BeF_2 at 600°C, w = 1.522, T = 301°K, a = 18.20, c = 4.62

W_i	W_f	x	y	W_i	W_f	x	y
0.040	0.057	11.04		0.755	0.788		3.62
0.057	0.072	12.25		0.788	0.822		3.55
0.077	0.092	13.07		0.822	0.859		3.32
0.367	0.388	14.55		0.859	0.895		3.37
0.388	0.412	14.28		0.895	0.931		3.30
0.412	0.431	14.41		0.931	0.964		3.60
0.431	0.453	14.50		0.964	0.999		3.46
0.468	0.507		3.10	1.005	1.026	15.22	
0.507	0.544		3.22	1.026	1.046	14.95	
0.544	0.579		3.46	1.046	1.067	14.97	
0.612	0.635	13.33		1.067	1.088	14.84	
0.635	0.655	15.79		1.088	1.109	15.03	
0.655	0.674	15.72		1.112	1.151		3.10
0.674	0.695	15.07		1.151	1.187		3.29
0.718	0.755		3.28				

Run No. 203

0.800 BeF_2 at 600°C, w = 1.522, T = 300°K, a = 17.10, c = 5.00

W_i	W_f	x	y	W_i	W_f	x	y
0.077	0.144		1.81	0.630	0.648	16.74	
0.155	0.174	16.03		0.648	0.667	16.83	
0.174	0.193	16.55		0.974	1.032		2.06
0.193	0.212	16.67		1.032	1.085		2.27
0.400	0.419	16.71		1.085	1.140		2.21
0.419	0.438	16.59		1.140	1.196		2.13
0.443	0.480		1.89	1.214	1.232	17.95	
0.480	0.539		2.04	1.232	1.249	17.95	
0.539	0.598		2.02	1.249	1.266	17.82	

Run No. 204

0.800 BeF_2 at 600°C, w = 1.522, T = 300°K, a = 0.000, c = 0.00

W_i	W_f	x	y	W_i	W_f	x	y
0.000	0.000	17.89		0.121	0.164	2.95	
0.013	0.027	9.09		0.164	0.209	2.75	
0.027	0.049	5.76		0.209	0.255	2.70	
0.049	0.081	3.79		0.388	0.413	2.50	
0.081	0.101	3.20		0.413	0.463	2.48	
0.101	0.121	3.01		0.463	0.514	2.42	

Run No. 207

0.800 BeF_2 at 700°C, w = 1.522, T = 300°K, a = 15.80, c = 5.26

W_i	W_f	x	y	W_i	W_f	x	y
0.003	0.020	7.24		0.814	0.831	18.46	
0.020	0.030	12.71		0.831	0.847	18.67	
0.122	0.206		1.42	0.847	0.864	18.76	
0.214	0.231	17.51		0.864	0.880	18.72	
0.231	0.248	17.72		0.880	0.897	18.67	
0.494	0.511	18.32		0.934	1.009		1.60
0.511	0.528	18.03		1.009	1.082		1.66
0.528	0.545	18.05		1.082	1.158		1.59
0.545	0.562	18.32		1.158	1.234		1.59
0.568	0.647		1.53	1.241	1.257	19.79	
0.647	0.729		1.46	1.257	1.273	19.49	
0.729	0.810		1.52				

Run No. 208

0.800 BeF_2 at 700°C, w = 1.522, T = 300°K, a = 0.0, c = 0.0

W_i	W_f	x	y	W_i	W_f	x	y
0.000	0.000	19.61	1.61	0.200	0.241	3.00	
0.001	0.008	18.18		0.241	0.284	2.92	
0.008	0.018	12.39		0.284	0.327	2.84	
0.018	0.033	8.06		0.327	0.347	3.12	
0.033	0.058	5.07		0.347	0.396	2.53	
0.058	0.089	4.03		0.396	0.448	2.38	
0.089	0.124	3.49		0.448	0.503	2.24	
0.124	0.160	3.28		0.503	0.617	2.18	
0.160	0.200	3.16					

Run No. 209

0.800 BeF_2 at 600°C, w = 1.522, T = 300°K, a = 15.40, c = 5.26

W_i	W_f	x	y	W_i	W_f	x	y
0.053	0.063	12.74		0.448	0.488		3.08
0.063	0.073	13.03		0.488	0.527		3.09
0.073	0.096	13.11		0.527	0.566		3.09
0.096	0.120	12.83		0.566	0.604		3.16
0.347	0.370	13.26		0.604	0.642		3.18
0.370	0.393	13.42		0.642	0.680		3.17
0.393	0.416	13.38		0.686	0.708	13.72	
0.416	0.440	13.45		0.708	0.731	13.86	

(continued)

Run No. 209 (continued)

W_i	W_f	x	y	W_i	W_f	x	y
0.731	0.753	13.71		0.881	0.903	13.86	
0.753	0.776	13.71		0.903	0.930	13.80	
0.801	0.839		3.12	0.930	0.952	13.99	
0.839	0.877		3.17				

Run No. 211

0.800 BeF_2 at 550°C, w = 1.522, T = 300°K, a = 15.00, c = 5.46

W_i	W_f	x	y	W_i	W_f	x	y
0.029	0.072		2.83	0.558	0.585	11.41	
0.075	0.081	10.04		0.585	0.612	11.66	
0.081	0.093	10.33		0.745	0.772		4.38
0.093	0.111	10.32		0.785	0.812	11.51	
0.111	0.135	10.43		0.812	0.838	11.57	
0.139	0.170		3.93	0.841	0.868		4.42
0.170	0.197		4.45	0.868	0.894		4.62
0.197	0.224		4.45	0.894	0.921		4.50
0.419	0.446		4.46	1.028	1.054		4.50
0.446	0.472		4.66	1.094	1.21	11.71	
0.472	0.498		4.55	1.121	1.147	11.78	
0.504	0.531	11.67		1.147	1.174	11.57	
0.531	0.558	11.30					

Run No. 213

0.800 BeF_2 at 650°C, w = 1.522, T = 300°K, a = 14.40, c = 5.53

W_i	W_f	x	y	W_i	W_f	x	y
0.128	0.185		2.14	0.750	0.769	16.67	
0.187	0.207	16.08		0.782	0.836		2.18
0.207	0.225	15.97		0.836	0.893		2.18
0.534	0.591		2.12	0.893	0.952		2.08
0.591	0.647		2.16	0.961	0.980	16.59	
0.694	0.713	16.59		0.980	0.998	16.43	
0.713	0.732	16.43		0.998	1.018	16.68	
0.732	0.750	16.45					

Run No. 215

0.700 BeF_2 at 700°C, w = 1.650, T = 300°K, a = 14.00, c = 5.46

W_i	W_f	x	y	W_i	W_f	x	y
0.094	0.117	11.97		0.643	0.660	16.61	
0.117	0.141	12.13		0.660	0.677	16.80	
0.313	0.333	14.33		0.677	0.694	17.20	
0.333	0.352	14.58		0.702	0.813		1.39
0.352	0.372	14.49		0.813	0.890		1.44
0.379	0.483		1.07	0.890	0.965		1.48
0.483	0.584		1.10	0.965	1.040		1.49
0.591	0.608	16.26		1.040	1.056	18.43	
0.608	0.626	16.70		1.056	1.071	18.26	
0.626	0.643	16.66		1.071	1.087	18.20	

Run No. 217

0.700 BeF_2 at 600°C, w = 1.650, T = 300°K, a = 13.80, c = 5.80

W_i	W_f	x	y	W_i	W_f	x	y
0.081	0.107	10.97		1.225	1.246	13.37	
0.107	0.134	10.88		1.246	1.268	13.41	
0.166	0.205		2.76	1.280	1.317		3.01
0.566	0.591	11.39		1.317	1.353		3.07
0.739	0.762	12.33		1.353	1.389		3.09
0.762	0.784	12.55		1.389	1.425		3.04
0.788	0.829		2.68	1.435	1.456	13.66	
0.829	0.870		2.72	1.456	1.477	13.58	
0.870	0.909		2.82	1.477	1.498	13.42	
0.911	0.933	13.00		1.732	1.759		3.10
0.933	0.954	13.34		1.759	1.795		3.09
1.182	1.203	13.17		1.802	1.823	13.37	
1.203	1.225	13.17		1.823	1.845	13.42	

Run No. 218

0.700 BeF_2 at 600°C, w = 1.650, T = 300°K, a = 0.000, c = 0.00

W_i	W_f	x	y	W_i	W_f	x	y
0.000	0.000	13.42	3.09	0.077	0.102	4.54	
0.000	0.010	11.66		0.102	0.128	4.47	
0.010	0.024	8.10		0.128	0.153	4.45	
0.024	0.043	6.08		0.153	0.179	4.38	
0.043	0.054	5.16		0.179	0.206	4.32	
0.054	0.065	4.93		0.206	0.232	4.26	
0.065	0.077	4.84					

Run No. 221

0.700 BeF_2 at 550°C, w = 1.650, T = 300°K, a = 13.20, c = 6.00

W_i	W_f	x	y	W_i	W_f	x	y
0.368	0.396	10.18		0.652	0.680		4.12
0.396	0.424	10.26		0.680	0.706		4.26
0.429	0.455		4.28	0.706	0.732		4.18
0.455	0.481		4.40	0.732	0.758		4.28
0.481	0.507		4.16	0.758	0.784		4.30
0.507	0.534		4.12	0.785	0.810	10.03	
0.540	0.566	10.80		0.810	0.837	10.79	
0.566	0.592	10.82		0.837	0.863	10.83	
0.592	0.619	10.74		0.863	0.890	10.83	
0.619	0.645	10.67					

Run No. 222

0.700 BeF_2 at 550°C, w = 1.650, T = 300°K, a = 0.000, c = 0.00

W_i	W_f	x	y	W_i	W_f	x	y
0.000	0.000	10.82	4.24	0.090	0.116	2.20	
0.000	0.015	7.71		0.116	0.143	2.11	
0.015	0.027	4.74		0.143	0.171	2.05	
0.027	0.044	3.40		0.171	0.229	1.99	
0.044	0.065	2.63		0.229	0.262	1.96	
0.065	0.090	2.29					

Run No. 223

0.700 BeF_2 at 650°C, w = 1.650, T = 300°K, a = 12.70, c = 6.00

W_i	W_f	x	y	W_i	W_f	x	y
0.515	0.577		1.81	0.938	0.958	14.79	
0.613	0.633	14.61		0.958	0.976	15.05	
0.633	0.653	14.34		0.993	1.028		1.90
0.653	0.672	14.33		1.028	1.060		2.10
0.686	0.857		1.95	1.067	1.085	15.75	
0.857	0.913		2.00	1.085	1.103	15.68	
0.920	0.938	15.38		1.103	1.121	15.62	

Run No. 225

0.600 BeF_2 at 700°C, w = 1.827, T = 300°K, a = 12.20, c = 6.05

W_i	W_f	x	y
0.004	0.014	22.36	
0.014	0.022	23.37	
0.022	0.032	22.90	
0.032	0.040	23.07	
0.111	0.120	22.46	
0.120	0.129	22.67	
0.194	0.235		2.45
0.236	0.245	22.29	
0.245	0.254	22.41	
0.254	0.266	21.67	
0.266	0.278	22.00	
0.309	0.335		2.07
0.335	0.382		2.15
0.382	0.429		2.16

W_i	W_f	x	y
0.489	0.535		2.18
0.535	0.582		2.16
0.582	0.628		2.19
0.634	0.646	21.43	
0.646	0.658	21.12	
0.658	0.670	21.12	
0.670	0.682	21.00	
0.682	0.737		2.10
0.737	0.783		2.21
0.783	0.829		2.17
0.829	0.875		2.18
0.878	0.898	21.40	
0.898	0.902	21.08	

Run No. 226

0.600 BeF_2 at 700°C, w = 1.820, T = 300°K, a = 0.000, c = 0.00

W_i	W_f	x	y
0.000	0.000	21.18	2.16
0.000	0.005	20.25	
0.005	0.011	17.43	
0.011	0.017	17.12	
0.017	0.024	15.83	
0.024	0.030	14.91	
0.030	0.037	14.76	
0.037	0.044	14.49	
0.044	0.052	14.36	
0.052	0.059	14.09	
0.059	0.067	13.49	
0.067	0.074	13.80	
0.074	0.082	13.25	
0.082	0.095	12.14	

W_i	W_f	x	y
0.095	0.103	12.33	
0.103	0.111	13.03	
0.111	0.120	12.43	
0.120	0.128	12.54	
0.128	0.136	12.20	
0.136	0.145	12.08	
0.145	0.154	11.83	
0.154	0.171	11.58	
0.171	0.195	10.83	
0.195	0.220	10.55	
1.607	1.647	1.31	
1.647	1.685	1.33	
1.685	1.725	1.30	

Run No. 227

0.600 BeF_2 at 600°C, w = 1.820, T = 300°K, a = 12.10, c = 6.05

W_i	W_f	x	y	W_i	W_f	x	y
0.000	0.000	1.30		0.986	1.054		2.97
0.011	0.085	3.51		1.056	1.078	11.86	
0.085	0.097	4.14		1.078	1.099	11.83	
0.288	0.308	5.20		1.099	1.121	12.05	
0.308	0.316	6.25		1.121	1.142	12.05	
0.316	0.332	6.59		1.183	1.214		3.24
0.332	0.348	6.54		1.214	1.245		3.25
0.348	0.362	7.18		1.245	1.276		3.24
0.397	0.458		1.67	1.283	1.303	13.00	
0.458	0.491		1.85	1.303	1.324	12.62	
0.496	0.514	8.41		1.324	1.344	12.63	
0.514	0.532	8.59		1.344	1.364	12.83	
0.532	0.550	8.54		1.403	1.434		3.36
0.550	0.568	8.82		1.434	1.463		3.38
0.781	0.796	10.67		1.463	1.493		3.45
0.796	0.810	10.68		1.495	1.515	13.36	
0.810	0.825	10.61		1.515	1.534	13.46	
0.830	0.914		2.49	1.534	1.554	13.18	
0.914	0.986		2.82				

Run No. 229

0.600 BeF_2 at 500°C, w = 1.820, T = 300°K, a = 12.00, c = 6.18

W_i	W_f	x	y	W_i	W_f	x	y
0.042	0.071	3.63		0.862	0.883		4.59
0.071	0.098	3.83		0.883	0.905		4.63
0.098	0.125	3.88		0.915	0.941	6.13	
0.134	0.203		2.92	0.941	0.966	6.09	
0.203	0.236		3.03	0.966	0.991	6.17	
0.246	0.270	4.21		0.991	1.016	6.25	
0.270	0.306	4.36		1.022	1.043		4.86
0.580	0.612	5.04		1.043	1.063		5.01
0.616	0.641		4.10	1.063	1.083		5.03
0.641	0.665		4.21	1.083	1.103		5.09
0.665	0.689		4.09	1.107	1.131	6.55	
0.692	0.720	5.60		1.131	1.154	6.57	
0.720	0.748	5.58		1.154	1.178	6.58	
0.748	0.785	5.59		1.183	1.202		5.24
0.785	0.812	5.63		1.202	1.222		5.14
0.817	0.840		4.46	1.228	1.251	6.57	
0.840	0.862		4.62	1.251	1.275	6.62	

Run No. 231

0.600 BeF_2 at 550°C, w = 1.820 T = 300°K, a = 11.80, c = 6.18

W_i	W_f	x	y	W_i	W_f	x	y
0.837	0.861		4.14	1.451	1.471	10.12	
0.861	0.886		4.13	1.471	1.492	9.99	
0.886	0.911		4.03	1.492	1.512	10.26	
0.915	0.944	8.91		1.512	1.532	10.22	
0.944	0.973	9.01		1.536	1.557		4.78
0.973	0.996	9.09		1.557	1.582		4.75
1.004	1.071		4.53	1.582	1.603		4.88
1.250	1.273		4.41	1.603	1.623		4.99
1.273	1.294		4.67	1.625	1.644	10.59	
1.299	1.320	9.97		1.644	1.664	10.49	
1.320	1.341	9.95		1.664	1.684	10.41	
1.341	1.361	9.95		1.687	1.709		4.75
1.363	1.384		4.80	1.709	1.729		5.05
1.384	1.405		4.76	1.732	1.751	10.63	
1.405	1.425		4.91	1.751	1.771	10.57	
1.425	1.447		4.68				

Run No. 233

0.600 BeF_2 at 650°C, w = 1.820, T = 299°K, a = 11.50, c = 6.45

W_i	W_f	x	y	W_i	W_f	x	y
0.007	0.029	7.09		0.821	0.868		2.14
0.029	0.048	8.33		0.868	0.913		2.21
0.470	0.489	13.74		0.918	0.938	15.93	
0.489	0.508	13.58		0.938	0.954	15.96	
0.508	0.527	13.74		0.954	0.970	15.80	
0.595	0.648		1.90	0.970	0.988	15.92	
0.648	0.678		1.97	0.997	1.031		2.34
0.683	0.701	14.99		1.013	1.065		2.40
0.701	0.721	14.92		1.065	1.098		2.38
0.721	0.742	14.84		1.114	1.129	16.34	
0.742	0.759	14.93		1.129	1.145	16.25	
0.762	0.821		2.03	1.145	1.161	16.21	

Run No. 235

0.500 BeF_2 at 700°C, w = 2.075, T = 298°K, a = 10.86, c = 6.71

W_i	W_f	x	y	W_i	W_f	x	y
0.030	0.051	2.11		0.707	0.721	16.28	
0.051	0.072	3.30		0.721	0.735	15.87	
0.072	0.089	4.13		0.735	0.750	15.86	
0.147	0.234		0.61	0.758	0.789		1.75
0.237	0.242	9.17		0.789	0.818		1.84
0.242	0.251	9.75		0.818	0.845		1.92
0.425	0.439	13.00		0.845	0.900		1.95
0.439	0.456	13.30		0.901	0.914	17.04	
0.456	0.473	13.50		0.914	0.927	16.75	
0.473	0.493	13.62		0.917	0.941	16.87	
0.494	0.551		1.24	0.948	0.988		1.83
0.551	0.618		1.58	0.988	1.014		2.04
0.618	0.673		1.61	1.015	1.028	17.22	
0.678	0.693	14.91		1.028	1.042	17.07	
0.693	0.707	16.25					

Run No. 237

0.500 BeF_2 at 600°C, w = 2.075, T = 299°K, a = 9.74, c = 5.92

W_i	W_f	x	y	W_i	W_f	x	y
1.304	1.344		4.41	1.709	1.727	12.57	
1.344	1.383		4.54	1.727	1.745	12.13	
1.387	1.405	12.58		1.745	1.764	12.13	
1.405	1.424	11.97		1.768	1.787		4.64
1.424	1.442	12.12		1.787	1.806		4.57
1.442	1.461	12.17		1.806	1.825		4.66
1.461	1.480	12.18		1.825	1.843		4.67
1.493	1.512		4.53	1.848	1.866	12.36	
1.512	1.531		4.58	1.866	1.885	12.08	
1.531	1.550		4.58	1.885	1.904	12.26	
1.550	1.570		4.55				

Run No. 239

0.500 BeF_2 at 500°C, w = 2.075, T = 298°K, a = 10.26, c = 6.97

W_i	W_f	x	y	W_i	W_f	x	y
0.047	0.066	2.43		0.107	0.130		2.25
0.066	0.084	2.57		0.130	0.153		2.34
0.084	0.102	2.47		0.153	0.176		2.35

(continued)

Run No. 239 (continued)

W_i	W_f	x	y	W_i	W_f	x	y
0.182	0.199	2.71		0.814	0.854	4.60	
0.199	0.215	2.80		0.854	0.893	4.63	
0.215	0.231	2.86		0.893	0.931	4.80	
0.405	0.432	3.43		0.958	0.975		4.99
0.432	0.463	3.62		0.975	1.013		5.10
0.463	0.488	3.62		1.013	1.058		5.14
0.494	0.520		3.37	1.058	1.075		5.22
0.520	0.544		3.60	1.076	1.094	5.12	
0.544	0.569		3.66	1.094	1.130	5.04	
0.569	0.593		3.70	1.130	1.166	5.09	
0.598	0.621	3.99		1.166	1.184	5.07	
0.621	0.644	4.04		1.207	1.226		5.53
0.644	0.671	4.17		1.226	1.239		5.37
0.671	0.694	4.09		1.239	1.255		5.55
0.705	0.731		4.12	1.255	1.271		5.62
0.731	0.752		4.29	1.274	1.291	5.29	
0.752	0.772		4.24	1.291	1.309	5.22	
0.772	0.793		4.32	1.309	1.326	5.18	
0.794	0.814	4.57					

Run No. 241

0.500 BeF_2 at 550°C, w = 2.075, T = 299°K, a = 9.35, c = 6.35

W_i	W_f	x	y	W_i	W_f	x	y
1.439	1.454		5.67	2.391	2.405	9.50	
1.454	1.463		5.75	2.465	2.477		5.93
1.463	1.472		6.09	2.477	2.488		6.30
1.472	1.481		6.09	2.488	2.499		6.18
1.483	1.492	9.93		2.499	2.511		6.33
1.492	1.501	10.05		2.514	2.528	9.50	
1.501	1.510	9.92		2.528	2.553	9.32	
1.510	1.519	9.88		2.553	2.567	9.32	
1.528	1.537		6.10	2.567	2.582	9.54	
1.537	1.545		6.45	2.614	2.626		5.93
1.545	1.553		6.68	2.626	2.638		6.17
1.553	1.566		6.64	2.638	2.649		6.41
1.567	1.577	9.63		2.649	2.660		6.43
1.577	1.586	9.92		2.662	2.672	9.68	
1.586	1.595	9.47		2.672	2.681	9.79	
1.595	1.605	9.57		2.681	2.690	9.74	
2.354	2.363	9.85		2.700	2.712		6.38
2.363	2.377	9.92		2.712	2.723		6.33
2.377	2.391	9.50					

Run No. 245

0.400 BeF_2 at 700°C, w = 2.432, T = 298°K, a = 8.49, c = 7.63

W_i	W_f	x	y	W_i	W_f	x	y
0.013	0.025	3.22		0.370	0.384		2.17
0.025	0.037	3.42		0.384	0.397		2.30
0.037	0.047	3.68		0.397	0.410		2.37
0.067	0.102		0.87	0.410	0.438		2.64
0.102	0.109	5.72		0.439	0.459		2.97
0.109	0.122	6.08		0.460	0.466	12.13	
0.122	0.134	6.41		0.466	0.472	12.13	
0.134	0.146	6.68		0.472	0.485	12.08	
0.329	0.337	10.41		0.494	0.520		2.96
0.337	0.344	10.75		0.520	0.539		3.10
0.344	0.352	10.46		0.590	0.599	13.30	

Run No. 247

0.400 BeF_2 at 700°C, w = 2.432, T = 298°K, a = 8.29, c = 7.63

W_i	W_f	x	y	W_i	W_f	x	y
0.180	0.188	9.26		0.499	0.511	13.38	
0.188	0.197	9.21		0.511	0.528	13.30	
0.197	0.205	9.13		0.528	0.540	13.51	
0.218	0.245		2.32	0.576	0.596		3.97
0.245	0.269		2.51	0.596	0.615		3.96
0.269	0.298		2.58	0.615	0.638		3.88
0.298	0.320		2.83	0.638	0.657		4.03
0.323	0.339	11.62		0.659	0.673	14.11	
0.339	0.353	11.68		0.673	0.686	14.30	
0.353	0.366	11.75		0.686	0.700	14.25	
0.366	0.379	11.87		0.706	0.725		4.07
0.379	0.382	11.67		0.725	0.743		4.05
0.392	0.420		3.25	0.743	0.762		4.09
0.420	0.441		3.51	0.762	0.773		4.08
0.441	0.463		3.54	0.777	0.787	15.08	
0.463	0.484		3.64	0.787	0.800	14.95	
0.487	0.499	13.21		0.800	0.810	15.08	

Run No. 249

0.400 BeF_2 at 600°C, w = 2.432, T = 299°K, a = 8.35, c = 7.63

W_i	W_f	x	y	W_i	W_f	x	y
0.415	0.427		6.32	0.444	0.452	9.71	
0.427	0.439		6.30	0.452	0.460	9.82	

(continued)

Run No. 249 (continued)

W_i	W_f	x	y	W_i	W_f	x	y
0.460	0.476	9.55		0.841	0.852		6.64
0.476	0.492	9.74		0.852	0.864		6.57
0.501	0.513		6.37	0.864	0.875		6.66
0.513	0.525		6.36	0.880	0.896	9.88	
0.525	0.537		6.41	0.896	0.911	9.93	
0.540	0.559	9.79		0.913	0.921	9.79	
0.559	0.580	9.62		0.921	0.929	9.92	
0.587	0.599		6.32	0.977	0.985	9.84	
0.599	0.610		6.43	0.985	0.993	9.92	
0.610	0.622		6.46	1.094	1.110	10.04	
0.623	0.640	9.62		1.110	1.126	9.76	
0.640	0.656	9.66		1.144	1.155		6.63
0.656	0.672	9.57		1.155	1.166		6.76
0.672	0.688	9.57		1.170	1.186	9.86	
0.830	0.841		6.75	1.186	1.201	9.86	

Run No. 251

0.400 BeF_2 at 500°C, w = 2.432, T = 299°K, a = 8.40, c = 7.70

W_i	W_f	x	y	W_i	W_f	x	y
0.000	0.022	4.32		0.294	0.302		9.41
0.022	0.041	4.28		0.369	0.386	4.43	
0.047	0.057		9.05	0.386	0.404	4.45	
0.057	0.074		8.87	0.404	0.421	4.49	
0.074	0.091		9.03	0.424	0.441		8.92
0.091	0.107		9.07	0.441	0.457		9.49
0.201	0.219	4.43		0.457	0.473		9.67
0.219	0.236	4.46		0.473	0.488		9.60
0.236	0.253	4.42		0.488	0.504		9.64
0.253	0.262	4.38		0.506	0.523	4.54	
0.277	0.294		9.16	0.523	0.540	4.55	

Run No. 252

0.400 BeF_2 at 550°C, w = 2.432, T = 299°K, a = 8.40, c = 7.70

W_i	W_f	x	y	W_i	W_f	x	y
0.671	0.689		9.79	0.733	0.747	8.25	
0.689	0.704		10.55	0.747	0.761	8.21	
0.704	0.719	7.67		0.788	0.803		9.97
0.719	0.733	8.24		0.803	0.817		10.72

(continued)

Run No. 252 (continued)

W_i	W_f	x	y	W_i	W_f	x	y
0.817	0.831		10.89	0.959	0.979		9.87
0.833	0.847	8.07		0.979	0.994		9.86
0.847	0.862	8.09		0.994	1.010		9.70
0.862	0.876	7.95		1.012	1.027	7.71	
0.876	0.891	7.90		1.027	1.043	7.54	
0.905	0.929		9.49	1.043	1.058	7.47	
0.929	0.944		10.12	1.058	1.074	7.37	
0.944	0.959		10.05				

Run No. 253

0.400 BeF_2 at 650°C, w = 2.432, T = 299°K, a = 8.40, c = 7.70

W_i	W_f	x	y	W_i	W_f	x	y
0.000	0.012	13.14		0.207	0.222	13.12	
0.012	0.026	13.66		0.222	0.237	13.07	
0.026	0.040	13.51		0.237	0.251	13.10	
0.040	0.071	13.50		0.268	0.296		5.36
0.092	0.105		5.42	0.296	0.331		5.32
0.105	0.133		5.43	0.331	0.359		5.38
0.133	0.161		5.45	0.360	0.375	13.18	
0.161	0.189		5.43	0.375	0.388	13.07	
0.193	0.207	13.53					

Run No. 255

0.400 BeF_2 at 700°C, w = 2.432, T = 299°K, a = 8.40, c = 7.70

W_i	W_f	x	y	W_i	W_f	x	y
0.000	0.023	16.87		0.299	0.332		4.67
0.025	0.067		4.33	0.335	0.346	17.33	
0.067	0.098		4.88	0.346	0.358	17.01	
0.098	0.132		4.92	0.358	0.369	16.99	
0.132	0.163		4.84	0.369	0.383	16.95	
0.168	0.178	18.00		0.383	0.394	16.95	
0.178	0.190	17.20		0.419	0.452		4.63
0.190	0.201	16.84		0.452	0.485		4.60
0.201	0.213	16.55		0.486	0.497	16.92	
0.235	0.267		4.67	0.497	0.509	16.32	
0.267	0.299		4.66				

Run No. 257

0.333 BeF_2 at 700°C, w = 2.751, T = 299°K, a = 8.40, c = 7.70

W_i	W_f	x	y	W_i	W_f	x	y
0.000	0.022	1.54		1.126	1.137	12.07	
0.022	0.039	2.01		1.150	1.162		5.67
0.039	0.053	2.46		1.162	1.185		5.83
0.053	0.076	3.04		1.185	1.208		5.87
0.164	0.176	5.71		1.307	1.318	12.28	
0.176	0.188	6.01		1.318	1.330	12.13	
0.188	0.209	6.40		1.330	1.341	12.26	
0.948	0.960	12.22		1.341	1.352	12.13	
0.960	0.971	12.14		1.380	1.391		5.92
0.961	0.982	12.14		1.391	1.414		5.93
1.012	1.024		5.70	1.414	1.436		5.93
1.024	1.035		5.86	1.436	1.459		5.92
1.035	1.058		5.80	1.472	1.483	11.88	
1.058	1.082		5.76	1.483	1.498	11.86	
1.086	1.097	12.25		1.498	1.509	12.34	
1.097	1.115	12.08		1.509	1.520	12.07	
1.115	1.126	12.16		1.520	1.531	12.05	

Run No. 259

0.333 BeF_2 at 600°C, w = 2.751, T = 299°K, a = 8.40, c = 7.70

W_i	W_f	x	y	W_i	W_f	x	y
1.162	1.177		8.64	1.526	1.536		8.63
1.177	1.193		8.80	1.536	1.552		8.47
1.193	1.208		8.72	1.552	1.567		8.55
1.214	1.244	6.70		1.567	1.583		8.59
1.244	1.260	6.64		1.584	1.599	6.82	
1.260	1.275	6.78		1.599	1.615	6.70	
1.289	1.297		8.96	1.615	1.630	6.67	
1.297	1.305		8.62	1.630	1.646	6.64	
1.305	1.320		8.71	1.657	1.672		8.80
1.320	1.336		8.59	1.672	1.688		8.45
1.336	1.343		8.62	1.688	1.704		8.40
1.346	1.356	6.72		1.704	1.719		8.55
1.356	1.366	6.79		1.722	1.738	6.55	
1.472	1.486	6.93		1.738	1.753	6.63	
1.486	1.502	6.78		1.753	1.769	6.74	
1.502	1.517	6.72		1.769	1.784	6.62	

Run No. 261

0.333 BeF_2 at 500°C, w = 2.751, T = 299°K, a = 8.40, c = 7.70

W_i	W_f	x	y	W_i	W_f	x	y
0.905	0.918		10.41	1.312	1.342	2.78	
0.918	0.931		10.60	1.342	1.367	2.82	
0.931	0.943		10.78	1.380	1.392		11.22
0.943	0.955		10.68	1.392	1.404		11.26
0.996	1.015	2.66		1.404	1.415		11.33
1.015	1.040	2.71		1.415	1.427		11.28
1.040	1.066	2.66		1.430	1.454	2.88	
1.066	1.092	2.64		1.454	1.478	2.88	
1.097	1.109		10.67	1.478	1.501	2.91	
1.109	1.121		10.88	1.507	1.513		10.92
1.121	1.133		10.99	1.513	1.537		11.29
1.133	1.151		11.17	1.537	1.549		11.28
1.151	1.163		11.03	1.550	1.574	2.92	
1.166	1.176	2.76		1.574	1.591	2.93	
1.176	1.201	2.76					

Run No. 263

0.333 BeF_2 at 550°C, w = 2.751, T = 299°K, a = 8.40, c = 7.70

W_i	W_f	x	y	W_i	W_f	x	y
0.931	0.952	4.78		1.292	1.320	4.86	
0.952	0.973	4.82		1.320	1.334	4.71	
0.973	0.995	4.82		1.356	1.370		9.22
1.041	1.048		9.84	1.370	1.391		9.68
1.064	1.078	4.90		1.391	1.405		9.72
1.078	1.093	4.80		1.405	1.425		9.80
1.093	1.107	4.82		1.430	1.444	4.82	
1.107	1.121	4.80		1.444	1.472	4.87	
1.134	1.148		9.46	1.472	1.501	4.82	
1.148	1.162		9.49	1.504	1.511		9.84
1.249	1.263	4.93		1.511	1.518		9.93
1.263	1.277	4.91		1.518	1.534		9.72
1.277	1.292	4.74		1.534	1.548		9.82

Run No. 264

0.333 BeF_2 at 550°C, w = 2.751, T = 299°K, a = 0.00, c = 0.00

W_i	W_f	x	y	W_i	W_f	x	y
0.000	0.000	4.87	9.80	0.004	0.008		9.59
0.000	0.004		9.51	0.008	0.013		9.36

(continued)

Run No. 264 (continued)

W_i	W_f	x	y	W_i	W_f	x	y
0.013	0.017		9.36	0.186	0.199	2.68	
0.017	0.021		8.90	0.199	0.212	2.64	
0.021	0.026		8.34	0.212	0.225	2.64	
0.026	0.032		7.67	0.225	0.251	2.62	
0.032	0.037		7.28	0.251	0.265	2.53	
0.037	0.043		7.10	0.276	0.288		3.38
0.043	0.049		6.53	0.288	0.316		3.29
0.050	0.059	4.07		0.318	0.661	2.50	
0.059	0.068	3.82		0.661	0.682	1.66	
0.068	0.078	3.47		0.682	0.704	1.55	
0.078	0.088	3.36		0.704	0.726	1.57	
0.091	0.098		5.86	0.738	0.766		1.44
0.098	0.106		5.47	0.766	0.796		1.32
0.106	0.122		4.99	0.796	0.828		1.25
0.122	0.138		4.79	0.828	0.894		1.21
0.138	0.147		4.66	0.894	0.921		1.29
0.147	0.183		4.43	0.921	0.949		1.24

Run No. 265

0.333 BeF_2 at 600°C, w = 2.751, T = 300°K, a = 8.55, c = 7.76

W_i	W_f	x	y	W_i	W_f	x	y
0.022	0.039	2.08		0.422	0.444		6.00
0.039	0.054	2.26		0.444	0.466		6.20
0.061	0.079		1.45	0.466	0.488		6.21
0.190	0.207	4.07		0.498	0.519	6.07	
0.207	0.222	4.49		0.519	0.536	6.04	
0.222	0.237	4.55		0.536	0.564	6.00	
0.237	0.251	4.74		0.564	0.587	6.07	
0.254	0.269		4.25	0.587	0.609	6.14	
0.269	0.311		4.75	0.609	0.631	6.22	
0.311	0.330		5.16	0.633	0.653		6.78
0.334	0.353	5.28		0.653	0.672		6.97
0.353	0.366	5.42		0.672	0.691		7.10
0.366	0.378	5.40		0.691	0.700		7.07
0.378	0.391	5.58		0.704	0.715	6.45	
0.394	0.411		5.62	0.715	0.726	6.38	
0.411	0.422		5.99				

Run No. 268

0.333 BeF_2 at 650°C, w = 2.751, T = 300°K, a = 0.00, c = 0.00

W_i	W_f	x	y	W_i	W_f	x	y
0.000	0.000	9.93	7.30	0.129	0.152		3.62
0.000	0.007		6.68	0.152	0.164		3.08
0.007	0.013		6.64	0.164	0.180		2.54
0.013	0.019		7.07	0.180	0.194		2.93
0.019	0.025		6.55	0.196	0.202	5.75	
0.025	0.032		5.86	0.202	0.208	5.45	
0.032	0.040		5.38	0.208	0.220	5.71	
0.042	0.045	8.99		0.220	0.233	5.37	
0.045	0.049	9.03		0.233	0.245	4.99	
0.049	0.053	8.20		0.623	0.635	2.80	
0.053	0.058	7.75		0.635	0.647	2.71	
0.058	0.062	7.50		0.647	0.686	2.67	
0.062	0.067	7.38		0.686	0.713	2.53	
0.067	0.072	7.34		0.713	0.741	2.46	
0.072	0.076	7.17		0.756	0.864		0.61
0.076	0.081	7.03		0.864	0.919		0.49
0.081	0.086	6.86		0.919	0.978		0.45
0.090	0.099		4.64	0.982	1.047	1.70	
0.099	0.109		4.13	1.047	1.107	1.59	
0.109	0.119		3.86	1.153	1.213	1.35	
0.119	0.129		3.79				

Run No. 269

0.333 BeF_2 at 600°C, w = 2.751, T = 300°K, a = 9.75, c = 10.75

W_i	W_f	x	y	W_i	W_f	x	y
0.028	0.041	2.60		0.401	0.423	6.12	
0.041	0.053	2.79		0.428	0.519		8.05
0.053	0.065	2.99		0.519	0.542		8.70
0.077	0.088		2.51	0.542	0.558		8.70
0.088	0.098		2.47	0.558	0.572		8.96
0.214	0.221	4.80		0.574	0.589	6.84	
0.221	0.235	4.79		0.589	0.604	6.92	
0.235	0.249	5.00		0.854	0.863	7.26	
0.268	0.299		5.66	0.863	0.873	7.49	
0.299	0.320		6.08	0.887	0.901		9.13
0.320	0.341		6.34	0.901	0.908		9.80
0.342	0.354	5.74		0.910	0.920	7.54	
0.354	0.366	5.86		1.371	1.377		11.37
0.366	0.378	5.96		1.383	1.392	7.91	
0.378	0.401	6.03		1.392	1.408	8.40	

(continued)

Run No. 269 (continued)

W_i	W_f	x	y	W_i	W_f	x	y
1.408	1.425	8.46		1.617	1.625	7.83	
1.425	1.441	8.18		1.748	1.759		11.46
1.441	1.458	8.05		1.759	1.771		11.64
1.497	1.509		11.67	1.771	1.782		11.86
1.509	1.521		11.66	1.783	1.801	7.92	
1.521	1.532		11.79	1.801	1.819	7.67	
1.532	1.544		11.93	1.819	1.827	8.14	
1.548	1.564	8.43		1.827	1.836	7.96	
1.564	1.581	7.95		1.836	1.853	7.79	
1.581	1.599	7.87		1.853	1.867	7.75	
1.599	1.617	7.80					

Run No. 270

0.333 BeF_2 at 600°C, w = 2.751, T = 300°K, a = 0.00, c = 0.00

W_i	W_f	x	y	W_i	W_f	x	y
0.000	0.000	7.90	11.71	0.125	0.139	5.08	
0.004	0.009	7.36		0.139	0.152	5.07	
0.009	0.014	7.55		0.152	0.160	4.79	
0.019	0.025	6.59		0.162	0.172		6.13
0.025	0.030	6.46		0.172	0.198		5.25
0.030	0.035	6.46		0.198	0.226		4.63
0.039	0.043		8.59	0.226	0.256		4.38
0.043	0.047		10.16	0.256	0.274		3.88
0.047	0.051		10.63	0.276	0.284	4.18	
0.051	0.055		10.76	0.284	0.301	4.03	
0.055	0.061		10.51	0.301	0.320	3.66	
0.061	0.068		9.88	0.320	0.338	3.70	
0.068	0.083		8.33	0.338	0.368	3.50	
0.083	0.093		7.05	0.375	0.398		2.87
0.093	0.103	5.45		0.398	0.425		2.49
0.103	0.112	5.50		0.427	0.437	3.29	
0.112	0.125	5.28		0.437	0.448	3.13	

Run No. 271

0.333 BeF_2 at 700°C, w = 2.751, T = 300°K, a = 10.90, c = 11.50

W_i	W_f	x	y	W_i	W_f	x	y
0.001	0.010	3.88		0.050	0.066		2.47
0.010	0.018	4.37		0.067	0.072	6.41	
0.018	0.025	4.78		0.072	0.077	6.53	

(continued)

Run No. 271 (continued)

W_i	W_f	x	y	W_i	W_f	x	y
0.077	0.087	7.00		0.711	0.726	14.05	
0.090	0.111		2.55	0.726	0.740	14.05	
0.111	0.129		2.93	0.740	0.760	13.87	
0.129	0.147		3.00	0.839	0.865		7.59
0.150	0.157	9.04		0.865	0.883		7.59
0.157	0.165	9.13		0.886	0.896	14.29	
0.291	0.296	11.54		0.896	0.918	13.86	
0.296	0.306	11.33		1.186	1.197	15.12	
0.306	0.318	11.61		1.197	1.208	15.71	
0.328	0.342		4.79	1.208	1.217	15.67	
0.342	0.352		5.63	1.217	1.229	15.12	
0.353	0.386		5.99	1.229	1.237	15.57	
0.386	0.418		6.29	1.237	1.246	15.72	
0.418	0.438		6.60	1.260	1.279		9.08
0.440	0.451	13.16		1.279	1.293		9.47
0.451	0.461	12.92		1.293	1.307		9.21
0.461	0.475	13.08		1.307	1.325		9.29
0.475	0.491	12.83		1.325	1.339		9.41
0.578	0.596		7.24	1.339	1.367		9.40
0.596	0.618		7.40	1.367	1.382		9.49
0.618	0.639		7.51	1.386	1.397	15.61	
0.639	0.662		7.58	1.397	1.413	15.07	
0.662	0.679		7.79	1.413	1.425	15.12	
0.679	0.695		7.99	1.425	1.436	15.05	
0.697	0.711	14.45					

Run No. 272

0.333 BeF_2 at 700°C, w = 2.751, T = 300°K, a = 0.00, c = 0.00

W_i	W_f	x	y	W_i	W_f	x	y
0.000	0.000	15.13	9.34	0.064	0.070	10.93	
0.009	0.013		9.03	0.070	0.076	10.57	
0.013	0.018		8.83	0.076	0.083	10.30	
0.018	0.022		9.34	0.083	0.090	9.95	
0.022	0.026		9.24	0.090	0.097	9.80	
0.026	0.030		9.60	0.101	0.107		6.68
0.030	0.035		9.30	0.107	0.122		5.28
0.035	0.039		9.64	0.122	0.141		4.21
0.039	0.043		9.54	0.141	0.162		3.68
0.043	0.047		9.60	0.162	0.186		3.40
0.047	0.052		8.82	0.187	0.196	7.95	
0.052	0.056		8.67	0.196	0.204	7.95	
0.058	0.064	12.08		0.204	0.217	7.83	

(continued)

Run No. 272 (continued)

W_i	W_f	x	y	W_i	W_f	x	y
0.217	0.231	7.63		0.332	0.376		1.53
0.231	0.255	7.24		0.376	0.427		1.29
0.255	0.280	6.68		0.427	0.449	4.78	
0.282	0.298		2.51	0.471	0.494	4.55	

Run No. 273

0.333 BeF_2 at 600°C, w = 2.751, T = 300°K, a = 5.000, c = 5.20

W_i	W_f	x	y	W_i	W_f	x	y
0.006	0.017	1.53		0.417	0.441		2.74
0.017	0.028	1.54		0.441	0.463		2.93
0.037	0.055		0.73	0.463	0.485		3.00
0.055	0.071		0.83	0.485	0.543		3.21
0.074	0.082	2.10		0.546	0.580	4.01	
0.082	0.091	2.07		0.580	0.613	4.13	
0.187	0.200	2.66		0.613	0.663	4.14	
0.200	0.212	2.86		0.663	0.695	4.30	
0.212	0.235	2.96		0.709	0.734		3.72
0.245	0.268		1.70	0.734	0.768		3.80
0.268	0.298		2.21	0.768	0.802		3.95
0.298	0.328		2.17	0.802	0.834		4.14
0.331	0.350	3.50		0.834	0.850		4.10
0.350	0.370	3.58		0.851	0.873	4.68	
0.370	0.389	3.55		0.873	0.895	4.57	
0.389	0.408	3.63		0.895	0.903	4.53	

Run No. 401

0.300 BeF_2 at 652°C, w = 0.544, T = 300°K, a = 5.77, c = 6.40

W_i	W_f	x	y	W_i	W_f	x	y
0.030	0.079	1.06		0.568	0.659		3.50
0.079	0.128	1.76		0.659	0.694		3.60
0.128	0.164	2.40		0.694	1.180		4.74
0.198	0.246		1.32	1.180	1.216		5.22
0.246	0.333		1.46	1.232	1.283	6.84	
0.344	0.395	4.34		1.283	1.334	6.80	
0.395	0.421	4.60		1.334	1.385	6.78	
0.421	0.458	4.74		1.385	1.437	6.68	
0.458	0.492	5.05		1.456	1.503		5.45
0.523	0.568		2.80	1.503	1.550		5.45

(continued)

Run No. 401 (continued)

W_i	W_f	x	y
1.550	1.595		5.59
1.605	1.680	6.99	
1.680	1.782	6.82	
1.782	1.832	6.86	
1.832	1.884	6.67	
1.904	1.960		5.68
1.960	2.016		5.70
2.016	2.071		5.78
2.071	2.126		5.83
2.147	2.197	6.90	
2.197	2.247	6.86	
2.247	2.298	6.82	
2.298	2.361	6.86	
2.361	2.412	6.78	
2.412	2.463	6.88	
2.494	2.604		5.79
2.604	2.714		5.79
2.714	2.769		5.87
2.769	2.823		5.87
2.837	2.887	7.03	
2.887	2.962	6.90	
2.962	3.039	6.75	
3.039	3.090	6.78	
3.106	3.161		5.82
3.161	3.238		5.79
3.238	3.293		5.79
3.293	3.347		5.97
3.368	3.419	6.78	
3.419	3.470	6.75	
3.470	3.521	6.78	
4.809	4.859	6.96	
4.859	4.909	6.88	
4.909	4.959	6.86	
5.010	5.064		6.00
5.064	5.116		6.07
5.116	5.169		6.03
5.169	5.223		5.86
5.223	5.276		6.09
5.302	5.352	6.93	
5.352	5.401	7.03	

Run No. 402

0.300 BeF_2 at 652°C, w = 0.544, T = 300°K, a = 0.00, c = 0.00

W_i	W_f	x	y
0.000	0.000	6.91	5.99
0.000	0.013	6.45	
0.013	0.029	5.58	
0.029	0.044	5.71	
0.044	0.071	5.21	
0.071	0.088	5.04	
0.088	0.109	4.99	
0.123	0.150		4.80
0.150	0.184		3.79
0.184	0.228		2.86
0.297	0.318	4.04	
0.318	0.338	4.50	
0.338	0.358	4.34	
0.358	0.379	4.13	
0.379	0.403	3.53	
0.426	0.498		1.76
0.498	0.548		1.28
0.548	0.660		1.13
0.756	0.793	2.33	
0.876	0.909	2.63	
0.909	0.942	2.60	
0.942	0.980	2.29	
0.980	1.024	1.96	
1.024	1.078	1.59	
1.078	1.135	1.53	
1.135	1.219	1.45	
1.219	1.280	1.42	

Run No. 403

0.300 BeF_2 at 540°C, w = 0.544, T = 300°K, a = 5.59, c = 6.33

W_i	W_f	x	y	W_i	W_f	x	y
0.037	0.132	0.55		1.979	2.061	2.53	
0.149	0.264		1.11	2.061	2.135	2.58	
0.264	0.308		1.46	2.135	2.194	2.60	
0.308	0.345		1.72	2.194	2.265	2.46	
0.362	0.411	1.41		2.265	2.333	2.55	
0.411	0.464	1.66		2.360	2.425		7.82
0.464	0.513	1.75		2.425	2.465		7.93
0.530	0.550		3.26	2.465	2.531		7.79
0.550	0.585		3.67	2.531	2.651		7.92
0.585	1.182		5.55	2.651	2.733		7.82
1.182	1.217		6.96	2.733	2.812		8.04
1.217	1.237		6.96	2.837	2.904	2.59	
1.295	1.331	2.43		2.904	2.971	2.58	
1.331	1.401	2.47		2.971	3.037	2.64	
1.401	1.472	2.43		3.037	3.104	2.58	
1.472	1.614	2.43		3.121	3.204		7.72
1.628	1.671		7.32	3.204	3.285		7.91
1.671	1.706		7.46	3.285	3.366		7.84
1.706	1.748		7.60	3.366	3.446		7.92
1.748	1.817		7.37	3.480	3.546	2.62	
1.817	1.884		7.60	3.546	3.611	2.64	
1.909	1.979	2.49		3.611	3.677	2.63	

Run No. 405

0.300 BeF_2 at 601°C, w = 0.544, T = 300°K, a = 5.59, c = 6.33

W_i	W_f	x	y	W_i	W_f	x	y
0.000	0.045	0.78		1.211	1.250	4.47	
0.045	0.105	0.87		1.250	1.288	4.53	
0.105	0.145	1.30		1.288	1.365	4.51	
0.145	0.182	1.86		1.381	1.434		6.00
0.224	0.303		1.60	1.434	1.486		6.16
0.303	0.334		2.08	1.486	1.537		6.22
0.334	0.361		2.34	1.549	1.587	4.55	
0.373	0.394	3.30		1.587	1.625	4.58	
0.394	0.425	3.38		1.625	1.662	4.66	
0.425	0.465	3.47		1.680	1.761		6.30
0.465	0.492	3.87		1.761	1.840		6.41
0.515	0.589		3.45	1.840	1.939		6.49
0.589	1.101		4.96	1.939	2.017		6.54
1.101	1.146		5.75	2.031	2.106	4.64	
1.172	1.211	4.49		2.106	2.180	4.67	

(continued)

Run No. 405 (continued)

W_i	W_f	x	y	W_i	W_f	x	y
2.180	2.275	4.55		3.778	3.835		6.83
2.275	2.349	4.64		3.835	3.891		6.71
2.367	2.446		6.42	3.891	3.947		6.80
2.446	2.523		6.59	3.958	4.012	4.79	
2.523	2.601		6.57	4.012	4.076	4.60	
2.601	2.677		6.70	5.134	5.189	4.70	
2.677	2.753		6.71	5.189	5.245	4.59	
2.770	2.844	4.67		5.245	5.282	4.74	
2.844	2.919	4.67		5.374	5.430		6.83
2.919	2.994	4.59		5.430	5.485		6.90
2.994	3.068	4.64		5.485	5.541		6.87
3.068	3.145	4.53		5.541	5.596		6.90
3.192	3.266		6.86	5.615	5.671	4.67	
3.266	3.350		6.80	5.671	5.726	4.72	
3.350	3.441		7.00	5.726	5.782	4.59	
3.441	3.515		6.90	5.782	5.838	4.64	
3.528	3.603	4.63		5.928	5.983		6.86
3.603	3.675	4.78		5.983	6.038		6.95
3.675	3.750	4.63		6.038	6.095		6.74

Run No. 406

0.300 BeF_2 at 601°C, w = 0.544, T = 300°K, a = 0.00, c = 0.00

W_i	W_f	x	y	W_i	W_f	x	y
0.000	0.000	4.67	6.84	0.433	0.497		1.99
0.022	0.040		6.68	0.497	0.573		1.67
0.040	0.059		6.74	0.573	0.660		1.46
0.059	0.082		5.53	0.720	0.763	1.62	
0.082	0.106		5.28	0.763	0.806	1.62	
0.106	0.135		4.43	0.806	0.853	1.45	
0.135	0.167		4.01	0.853	0.901	1.46	
0.183	0.204	3.34		0.933	1.036		0.93
0.204	0.227	2.93		1.036	1.121		0.75
0.227	0.252	2.75		1.157	1.209	1.36	
0.252	0.279	2.63		1.209	1.275	1.05	
0.279	0.305	2.66		1.275	1.340	1.06	
0.305	0.332	2.54		1.340	1.408	1.02	
0.332	0.360	2.45					

Run No. 407

0.300 BeF_2 at 700°C, w = 0.544, T = 300°K, a = 5.67, c = 6.53

W_i	W_f	x	y	W_i	W_f	x	y
0.037	0.083	1.13		2.223	2.302		4.86
0.083	0.125	2.05		2.302	2.368		4.83
0.125	0.156	2.87		2.368	2.434		4.80
0.156	0.180	3.59		2.434	2.500		4.83
0.227	0.276		1.30	2.516	2.573	9.18	
0.276	0.335		1.63	2.573	2.630	9.04	
0.347	0.362	5.88		2.630	2.689	8.84	
0.362	0.390	6.10		2.689	2.766	9.00	
0.390	0.417	6.42		2.766	2.842	8.91	
0.417	0.443	6.70		2.852	2.920		4.70
0.443	0.468	6.95		2.920	3.152		4.95
0.504	0.531		2.38	3.152	3.217		4.90
0.531	0.577		2.75	3.288	3.345	9.16	
0.577	0.637		3.16	3.345	3.403	8.95	
0.637	0.673		3.60	3.403	3.461	8.97	
0.680	0.700	8.28		3.461	3.520	8.91	
0.700	0.742	8.32		3.558	3.625		4.79
0.742	0.763	8.13		3.625	3.687		5.05
0.773	1.337		4.29	3.687	3.751		4.99
1.337	1.377		4.70	3.751	3.853		5.03
1.411	1.449	9.10		3.853	3.917		4.95
1.449	1.487	9.05		3.917	3.980		5.04
1.487	1.526	8.93		3.995	4.051	9.18	
1.526	1.565	8.93		4.051	4.109	9.05	
1.598	1.653		4.62	4.109	4.167	8.96	
1.653	1.706		4.78	5.656	5.714	8.95	
1.706	1.761		4.70	5.714	5.772	9.00	
1.761	1.815		4.70	5.847	5.909		5.07
1.815	1.868		4.79	5.909	5.974		4.99
1.882	1.958	9.09		5.974	6.037		5.00
1.958	2.016	8.99		6.037	6.113		5.04
2.016	2.074	8.95		6.130	6.187	9.14	
2.074	2.131	9.00		6.187	6.245	9.04	
2.154	2.223		4.62	6.245	6.303	8.95	

Run No. 408

0.300 BeF_2 at 700°C, w = 0.544, T = 300°K, a = 0.00, c = 0.00

W_i	W_f	x	y	W_i	W_f	x	y
0.000	0.000	9.01	5.00	0.099	0.128	7.16	
0.040	0.070	6.78		0.128	0.152	7.16	
0.070	0.099	7.36		0.152	0.180	6.36	

(continued)

Run No. 408 (continued)

W_i	W_f	x	y	W_i	W_f	x	y
0.180	0.206	6.32		0.598	0.628	2.91	
0.206	0.236	5.87		0.628	0.658	2.82	
0.288	0.346		2.21	0.680	0.752		0.88
0.346	0.380		1.83	0.752	0.862		0.58
0.380	0.430		1.28	0.862	0.927		0.49
0.444	0.490	3.80		0.952	0.999	1.86	
0.490	0.514	3.58		0.999	1.072	1.67	
0.514	0.541	3.24		1.072	1.128	1.54	
0.541	0.569	3.08		1.128	1.187	1.45	
0.569	0.598	3.00		1.187	1.252	1.34	

Run No. 409

0.300 BeF_2 at 500°C, w = 0.544, T = 300°K, a = 5.67, c = 6.53

W_i	W_f	x	y	W_i	W_f	x	y
0.083	0.177		0.68	1.974	2.033	1.76	
0.213	0.260	0.73		2.080	2.142		8.09
0.260	0.305	0.78		2.142	2.219		8.30
0.305	0.345	0.87		2.219	2.279		8.34
0.373	0.407		1.88	2.279	2.354		8.49
0.407	0.436		2.21	2.464	2.517	1.97	
0.436	0.482		2.72	2.517	2.605	1.97	
0.482	0.526		2.90	2.605	2.698	1.87	
0.556	0.587	1.13		2.698	2.772	1.87	
0.587	0.649	1.26		2.793	2.853		8.38
0.649	0.701	1.33		2.853	2.928		8.49
0.709	0.754		4.28	2.928	3.003		8.49
0.754	0.795		4.64	3.003	3.077		8.51
0.795	0.820		5.00	3.077	3.152		8.43
0.820	0.845		4.90	3.196	3.288	1.87	
0.866	0.912	1.51		3.288	3.362	1.88	
0.912	0.958	1.50		3.362	3.436	1.88	
0.958	1.490	1.63		3.436	3.510	1.86	
1.490	1.528	1.83		3.536	3.596		8.38
1.576	1.610		7.29	3.596	3.704		8.29
1.610	1.644		7.63	3.704	3.764		8.40
1.644	1.677		7.60	3.778	3.834	1.88	
1.677	1.710		7.75	3.834	3.888	1.88	
1.710	1.775		7.72	3.909	3.955		8.22
1.792	1.855	1.67		3.955	4.000		8.51
1.855	1.914	1.74		4.000	4.046		8.25
1.914	1.974	1.74					

Run No. 413

0.250 BeF_2 at 607°C, w = 0.633, T = 299°K, a = 5.59, c = 6.33

W_i	W_f	x	y	W_i	W_f	x	y
0.043	0.090	0.64		1.881	1.944		7.04
0.090	0.143	1.12		1.944	2.008		7.04
0.180	0.222		1.32	2.089	2.159	4.18	
0.222	0.255		1.70	2.159	2.233	4.09	
0.255	0.284		1.91	2.233	2.306	4.04	
0.303	0.325	2.63		2.306	2.344	3.95	
0.325	0.347	2.78		2.366	2.428		7.13
0.347	0.378	2.79		2.428	2.492		6.97
0.378	0.408	3.00		2.492	2.586		7.13
0.408	0.437	3.12		2.586	2.649		7.12
0.484	0.509		3.43	2.672	2.708	4.12	
0.509	0.536		4.05	2.708	2.784	3.91	
0.536	0.562		4.40	2.784	2.868	4.22	
0.579	0.605	3.50		2.868	2.912	4.09	
0.605	0.630	3.63		2.978	3.017		7.18
0.630	0.655	3.53		3.017	3.081		7.00
0.655	0.680	3.55		3.081	3.121		7.03
0.717	1.075		5.92	3.144	3.180	4.13	
1.075	1.092		6.46	3.180	3.216	4.12	
1.110	1.134	3.83		3.216	3.252	4.18	
1.134	1.164	3.95		4.130	4.166	4.05	
1.164	1.194	3.95		4.166	4.203	4.07	
1.194	1.224	3.99		4.284	4.323		7.08
1.224	1.253	4.12		4.323	4.378		7.17
1.302	1.346		6.38	4.378	4.416		7.20
1.346	1.388		6.57	4.416	4.455		7.13
1.388	1.429		6.74	4.477	4.514	4.08	
1.429	1.485		7.05	4.514	4.588	4.01	
1.497	1.526	4.07		4.588	4.661	4.05	
1.526	1.572	3.88		4.709	4.748		7.21
1.572	1.608	4.14		4.748	4.786		7.28
1.608	1.645	4.07		4.786	4.825		7.10
1.645	1.681	4.03		4.835	4.872	3.96	
1.738	1.778		7.00	4.872	4.909	4.07	
1.778	1.817		7.08	4.909	4.946	4.00	
1.817	1.881		7.03				

Run No. 415

0.250 BeF_2 at 702°C, w = 0.633, T = 299°K, a = 5.40, c = 6.51

W_i	W_f	x	y	W_i	W_f	x	y
0.029	0.057	1.05		0.080	0.112	1.82	
0.057	0.080	1.33		0.145	0.200		1.01

(continued)

Run No. 415 (continued)

W_i	W_f	x	y	W_i	W_f	x	y
0.200	0.254		1.04	1.828	1.869	7.29	
0.254	0.302		1.32	1.869	1.910	7.22	
0.309	0.321	4.95		1.910	1.951	7.26	
0.321	0.338	5.12		1.951	1.994	7.03	
0.338	0.357	4.92		1.994	2.035	7.17	
0.357	0.374	5.13		2.113	2.163		5.57
0.399	0.430		1.82	2.163	2.213		5.60
0.430	0.474		2.50	2.213	2.283		5.58
0.474	0.514		2.84	2.283	2.332		5.63
0.539	0.558	6.10		2.332	2.382		5.64
0.558	0.577	6.26		2.401	2.442	7.34	
0.577	0.596	6.36		2.442	2.491	7.20	
0.634	0.668		3.25	2.491	2.542	7.01	
0.668	0.698		3.80	2.542	2.583	7.30	
0.698	0.741		4.00	2.583	2.623	7.36	
0.772	0.794	6.78		2.678	2.728		5.53
0.794	0.816	6.88		2.728	2.788		5.66
0.830	1.336		4.96	2.788	2.836		5.74
1.336	1.388		5.40	2.836	2.885		5.72
1.400	1.420	7.34		2.885	2.934		5.66
1.420	1.441	7.17		2.948	2.989	7.29	
1.441	1.482	7.24		2.989	3.031	7.17	
1.482	1.524	7.14		3.031	3.073	7.10	
1.555	1.605		5.50	3.073	3.115	7.08	
1.605	1.657		5.38	3.154	3.204		5.57
1.657	1.718		5.46	3.204	3.254		5.63
1.718	1.768		5.67	3.254	3.353		5.66
1.768	1.819		5.42				

Run No. 419

0.250 BeF_2 at 520°C, w = 0.633, T = 300°K, a = 5.40, c = 6.35

W_i	W_f	x	y	W_i	W_f	x	y
0.032	0.080	0.55		0.422	0.452	1.95	
0.080	0.119	0.75		0.452	0.484	1.89	
0.119	0.150	0.96		0.507	0.530		4.72
0.150	0.178	1.05		0.530	0.551		5.41
0.209	0.258		2.21	0.551	0.569		5.76
0.258	0.293		3.20	0.569	0.588		5.95
0.293	0.325		3.45	0.588	0.614		6.43
0.325	0.352		4.03	0.635	0.665	2.00	
0.372	0.390	1.65		0.665	0.692	2.17	
0.390	0.422	1.90		0.692	0.746	2.22	

(continued)

Run No. 419 (continued)

W_i	W_f	x	y	W_i	W_f	x	y
0.746	0.773	2.23		1.960	2.016		7.87
0.789	0.814		6.62	2.016	2.086		7.82
0.814	0.838		7.08	2.086	2.156		7.87
0.838	0.869		7.08	2.156	2.212		7.84
0.869	0.899		7.21	2.240	2.301	2.42	
0.900	0.937		7.43	2.301	2.388	2.39	
0.937	0.966		7.42	2.388	2.450	2.42	
0.990	1.017	2.23		2.450	2.510	2.48	
1.017	1.447	2.35		2.510	2.572	2.41	
1.566	1.601		7.74	2.637	2.707		7.93
1.601	1.636		7.92	2.707	2.777		7.82
1.636	1.664		7.75	2.777	2.847		7.86
1.685	1.714	2.50		2.847	2.916		7.95
1.714	1.777	2.36		2.952	3.011	2.51	
1.777	1.839	2.42		3.011	3.072	2.46	
1.839	1.912	2.43		3.072	3.133	2.44	
1.932	1.960		7.75				

Run No. 421

0.250 BeF_2 at 655°C, w = 0.633, T = 300°K, a = 5.60, c = 6.55

W_i	W_f	x	y	W_i	W_f	x	y
0.032	0.085	0.56		1.187	1.226		5.99
0.085	0.125	1.12		1.226	1.273		5.92
0.125	0.143	1.62		1.309	1.336	5.50	
0.143	0.159	1.91		1.336	1.363	5.58	
0.159	0.178	2.24		1.363	1.400	5.58	
0.238	0.281		1.27	1.400	1.427	5.53	
0.281	0.332		1.74	1.457	1.514		5.74
0.337	0.356	3.92		1.514	1.561		5.90
0.356	0.375	3.96		1.561	1.606		6.09
0.375	0.392	4.25		1.606	1.667		6.30
0.392	0.410	4.10		1.667	1.710		6.34
0.410	0.428	4.21		1.733	1.760	5.47	
0.475	0.498		2.41	1.760	1.787	5.55	
0.498	0.841		4.16	1.787	1.851	5.58	
0.841	0.863		5.00	1.851	1.905	5.53	
0.882	0.930	4.68		1.905	1.970	5.45	
0.930	0.959	5.21		2.028	2.069		6.63
0.959	0.986	5.42		2.069	2.126		6.76
0.986	1.013	5.46		2.126	2.167		6.64
1.013	1.041	5.28		2.167	2.208		6.82
1.081	1.133		5.37	2.208	2.248		6.68
1.133	1.187		5.74				

(continued)

Run No. 421 (continued)

W_i	W_f	x	y	W_i	W_f	x	y
2.249	2.289		6.80	2.638	2.720		6.76
2.317	2.424	5.57		2.720	2.761		6.71
2.424	2.478	5.53		2.801	2.841	5.60	
2.478	2.532	5.46		2.841	2.868	5.46	
2.599	2.639		6.83				

Run No. 423

0.250 BeF_2 at 543°C, w = 0.633, T = 298°K, a = 5.40, c = 6.35

W_i	W_f	x	y	W_i	W_f	x	y
0.021	0.061	0.73		1.716	1.760		7.38
0.061	0.093	0.94		1.760	1.804		7.58
0.093	0.116	1.28		1.804	1.892		7.53
0.116	0.136	1.48		1.922	1.993	3.12	
0.171	0.225		2.04	1.993	2.044	2.96	
0.225	0.249		2.37	2.044	2.104	2.93	
0.249	0.267		2.95	2.104	2.155	2.96	
0.267	0.284		3.41	2.155	2.204	3.03	
0.307	0.327	2.20		2.245	2.299		7.22
0.327	0.356	2.10		2.299	2.364		7.62
0.356	0.381	2.34		2.364	2.408		7.54
0.381	0.406	2.36		2.408	2.451		7.76
0.406	0.430	2.47		2.451	2.501		7.74
0.456	0.485		3.91	2.501	2.545		7.51
0.485	0.905		6.17	2.565	2.615	2.99	
0.905	0.921		6.93	2.615	2.665	2.99	
0.921	0.943		7.21	2.665	2.714	3.01	
0.963	0.984	2.76		2.714	2.764	2.97	
0.984	1.016	2.80		2.827	2.870		7.59
1.016	1.046	2.92		2.870	2.912		7.87
1.046	1.076	2.95		2.912	2.955		7.59
1.076	1.106	2.97		2.955	2.998		7.75
1.228	1.264		7.47	2.998	3.041		7.67
1.264	1.302		7.40	3.041	3.085		7.59
1.302	1.347		7.38	3.108	3.157	3.04	
1.370	1.395	2.95		3.157	3.207	2.97	
1.395	1.421	2.90		3.207	3.282	2.97	
1.421	1.471	2.93		3.282	3.332	2.99	
1.471	1.548	2.93		3.332	3.381	3.01	
1.548	1.598	2.97		3.431	3.468		7.34
1.641	1.678		7.45	3.468	3.505		7.58
1.678	1.716		7.29	3.505	3.540		7.72

APPENDIX B

Partial Pressures from Studies on Unsaturated (with respect to BeO) Melts

The experiments are arranged according to a three digit "Run No." which indicates chronological order. There are three series (300, 500, 600) included. A brief history of each series is given in Chapter III.

The information provided for each experiment includes:

Run No.

Composition of melt (expressed as mole fraction BeF_2)

Temperature of melt, ^{o}C

w = weight of melt, kg

T = temperature of wet-test meter, ^{o}K

$a + bW$ = influent P_{HF}, atm x 10^3

$c + dW$ = influent P_{H_2O}, atm x 10^3

The effluent partial pressures, $P_{HF} = x$ and $P_{H_2O} = y$, are tabulated (atm x 10^3) with the corresponding initial and final values of W. Note that the use of W allows consistent comparison of experiments independent of the weight of melt or temperature of gas-measurement. The equilibrium quotients evaluated from each experiment are presented in Table 4.

Run No. 301

0.333 BeF_2 at 600°C, w = 0.500, T = 299°K, a = 3.68, c = 11.00

W_i	W_f	x	y	W_i	W_f	x	y
0.000	0.000	0.107		1.700	1.753		6.51
0.000	0.030	3.72		1.753	1.807		6.46
0.030	0.053	11.55		1.814	1.852	9.71	
0.053	0.080	13.92		1.852	1.890	9.93	
0.080	0.106	14.39		1.890	1.930	9.53	
0.106	0.133	14.36		1.930	2.011	9.34	
0.133	0.159	14.26		2.038	2.090		6.72
0.249	0.300		2.70	2.090	2.141		6.71
0.307	0.334	14.05		2.141	2.272		6.92
0.334	0.361	13.88		2.272	2.321		7.03
0.361	0.389	13.67		2.327	2.369	9.08	
0.403	0.445		3.30	2.369	2.431	9.09	
0.445	0.484		3.59	2.431	2.494	9.00	
0.484	0.519		3.95	2.494	2.538	8.57	
0.519	0.571		4.01	2.568	2.616		7.28
0.582	0.612	12.70		2.616	2.722		7.22
0.612	0.642	12.49		2.722	2.825		7.40
0.642	0.673	12.26		2.845	2.888	8.64	
0.673	0.704	12.26		2.888	2.930	9.12	
0.725	0.769		4.75	2.930	2.995	8.71	
0.769	0.812		4.92	2.995	3.062	8.46	
0.812	0.856		4.76	3.102	3.192		7.66
0.856	0.897		5.10	3.192	3.284		7.62
0.897	0.937		5.21	3.284	3.373		7.82
0.946	0.978	11.55		3.373	3.464		7.66
0.978	1.011	11.43		3.464	3.508		7.84
1.492	1.528	10.41		3.509	3.579	8.14	
1.528	1.565	10.28		3.579	3.648	8.09	
1.565	1.601	10.20		3.648	3.719	8.03	
1.610	1.645		5.99	3.719	3.814	7.88	
1.645	1.700		6.32				

Run No. 302

0.333 BeF_2 at 600°C, w = 0.500, T = 299°K, a = 0.000, c = 0.00

W_i	W_f	x	y	W_i	W_f	x	y
0.000	0.000	7.90	7.90	0.244	0.297	3.58	
0.000	0.020	7.72		0.297	0.350	3.58	
0.020	0.056	5.17		0.360	0.419		2.37
0.056	0.091	5.45		0.419	0.506		1.62
0.091	0.127	5.13		0.506	0.601		1.46
0.137	0.159		6.32	0.635	0.722	2.17	
0.159	0.190		4.51	0.722	0.819	1.93	
0.190	0.234		3.17	0.819	0.924	1.79	

Run No. 303

0.333 BeF_2 at 600°C, w = 0.500, T = 299°K, a = 3.82, c = 10.53

W_i	W_f	x	y	W_i	W_f	x	y
0.008	0.068	1.26		2.165	2.217	7.26	
0.068	0.120	2.16		2.217	2.309	7.37	
0.120	0.162	2.70		2.335	2.417		8.54
0.193	0.246		2.64	2.417	2.498		8.57
0.246	0.295		2.82	2.498	2.579		8.55
0.306	0.344	4.90		2.579	2.661		8.53
0.344	0.380	5.30		2.706	2.783	7.33	
0.380	0.414	5.43		2.783	2.857	7.63	
0.414	0.447	5.74		2.857	2.932	7.57	
0.447	0.479	5.91		2.932	3.013	6.99	
0.522	0.547		5.51	3.013	3.066	7.08	
0.547	0.582		5.93	3.097	3.189		8.40
0.582	0.629		5.97	3.189	3.270		8.59
0.629	0.671		6.53	3.270	3.360		8.49
0.685	0.712	6.78		3.360	3.401		8.55
0.712	0.740	6.78		3.407	3.488	7.00	
0.740	0.794	7.00		3.488	3.570	6.93	
0.794	0.821	7.00		3.712	3.793	6.97	
0.844	0.892		7.24	3.839	3.919		8.78
0.892	0.939		7.42	3.919	4.008		8.54
1.492	1.542	7.54		4.008	4.088		8.78
1.542	1.592	7.50		4.088	4.185		8.54
1.654	1.696		8.28	4.198	4.253	6.87	
1.696	1.779		8.42	4.253	4.336	6.79	
1.779	1.942		8.54	4.336	4.420	6.76	
1.942	2.024		8.46	4.420	4.476	6.68	
2.035	2.086	7.37		4.582	4.623		8.67
2.086	2.165	7.17					

Run No. 305

0.333 BeF_2 at 600°C, w = 0.500, T = 299°K, a = 3.82, c = 10.53

W_i	W_f	x	y	W_i	W_f	x	y
0.000	0.023	3.22		0.263	0.290	14.32	
0.023	0.042	10.09		0.290	0.316	14.00	
0.042	0.065	13.30		0.337	0.404		3.08
0.065	0.090	14.63		0.404	0.462		3.63
0.090	0.116	14.49		0.469	0.497	13.39	
0.116	0.143	14.26		0.497	0.526	13.13	
0.156	0.200		1.60	0.526	0.569	12.91	
0.200	0.232		2.16	0.569	0.599	12.76	
0.236	0.263	14.09		0.621	0.669		4.38

(continued)

Run No. 305 (continued)

W_i	W_f	x	y	W_i	W_f	x	y
0.669	0.713		4.70	3.626	3.713		8.01
0.713	0.786		4.74	3.713	3.799		8.08
0.786	0.828		5.07	3.815	3.913	7.71	
0.842	0.890	11.83		3.913	4.022	7.79	
0.890	0.939	11.41		4.031	4.129	7.67	
0.939	0.989	11.34		4.129	4.179	7.62	
1.030	1.095		5.37	4.239	4.321		8.43
1.095	1.158		5.58	4.321	4.407		8.08
1.158	1.218		5.72	4.407	4.493		8.16
1.218	1.278		5.84	4.493	4.602		8.29
1.292	1.344	10.83		4.630	4.706	7.47	
1.344	1.396	10.80		4.706	4.800	7.99	
2.119	2.160	9.34		4.800	4.900	7.55	
2.160	2.221	9.25		4.900	5.003	7.32	
2.295	2.344		7.08	5.003	5.054	7.32	
2.344	2.391		7.30	5.103	5.187		8.26
2.391	2.488		7.21	5.187	5.272		8.22
2.488	2.535		7.40	5.272	5.355		8.37
2.543	2.607	8.82		5.355	5.438		8.38
2.607	2.672	8.74		5.453	5.560	7.10	
2.672	2.760	8.55		5.560	5.693	7.04	
2.771	2.836		7.51	5.693	5.800	7.09	
2.836	2.930		7.40	5.844	5.924		8.74
2.930	3.022		7.62	5.924	6.006		8.45
3.022	3.068		7.59	6.006	6.088		8.58
3.069	3.137	8.28		6.100	6.207	7.04	
3.137	3.229	8.20		6.207	6.289	6.83	
3.229	3.322	8.09		6.289	6.372	6.86	
3.322	3.416	8.01		6.407	6.490		8.43
3.448	3.538		7.71	6.490	6.572		8.47
3.538	3.626		7.91	6.572	6.654		8.47

Run No. 306

0.333 BeF_2 at 600°C, w = 0.500, T = 299°K, a = 0.000, c = 0.00

W_i	W_f	x	y	W_i	W_f	x	y
0.000	0.000	6.84	8.42	0.256	0.319		3.36
0.040	0.058		7.76	0.319	0.366		2.91
0.058	0.081		6.09	0.383	0.414	3.08	
0.089	0.110	4.70		0.414	0.447	2.84	
0.110	0.130	4.57		0.447	0.503	2.70	
0.130	0.174	4.34		0.503	0.546	2.60	
0.188	0.220		4.36	0.571	0.655		1.67
0.220	0.256		3.84	0.655	0.748		1.50

(continued)

Run No. 306 (continued)

W_i	W_f	x	y	W_i	W_f	x	y
0.748	0.865		1.18	1.022	1.082	1.55	
0.885	0.941	1.68		1.082	1.146	1.47	
0.941	1.022	1.64		1.146	1.284	1.37	

Run No. 307

0.333 BeF_2 at 700°C, w = 0.500, T = 297°K, a = 3.95, c = 10.50

W_i	W_f	x	y	W_i	W_f	x	y
0.000	0.031	6.08		2.288	2.388		4.21
0.031	0.055	15.95		2.388	2.455		4.14
0.055	0.076	17.46		2.458	2.508	14.96	
0.076	0.107	18.32		2.508	2.585	14.71	
0.107	0.138	17.99		2.585	2.649	14.78	
0.138	0.168	18.87		2.649	2.714	14.39	
0.168	0.199	18.71		2.761	2.838		4.54
0.199	0.230	18.32		2.838	2.931		4.53
0.230	0.260	18.82		2.931	3.023		4.59
0.260	0.290	18.41		3.023	3.112		4.70
0.290	0.331	18.37		3.114	3.195	13.99	
0.378	0.431		2.26	3.195	3.262	14.00	
0.431	0.478		2.92	3.262	3.330	13.88	
0.492	0.523	18.37		3.330	3.425	13.88	
0.523	0.555	17.84		3.455	3.541		4.87
0.555	0.587	17.47		3.541	3.644		4.76
1.018	1.039	17.33		3.644	3.730		4.90
1.039	1.073	16.71		3.730	3.814		4.97
1.108	1.170		3.40	3.827	3.909	13.80	
1.170	1.229		3.55	3.909	3.978	13.66	
1.229	1.292		3.30	3.978	4.048	13.50	
1.301	1.336	15.83		4.048	4.118	13.49	
1.336	1.372	15.97		4.185	4.269		5.01
1.372	1.431	15.92		4.269	4.354		4.95
1.431	1.479	15.78		4.354	4.439		4.93
1.479	1.525	16.22		4.439	4.551		5.01
1.563	1.623		3.53	4.563	4.633	13.33	
1.623	1.716		3.75	4.633	4.706	13.04	
1.716	1.813		3.63	4.706	4.777	13.20	
1.822	1.871	15.33		4.777	4.849	13.05	
1.871	1.920	15.33		4.883	5.020		5.09
1.920	1.982	15.20		5.020	5.155		5.18
1.982	2.056	15.29		5.155	5.288		5.29
2.121	2.205		4.17	5.288	5.419		5.32
2.205	2.288		4.22	5.433	5.505	12.93	

(continued)

Run No. 307 (continued)

W_i	W_f	x	y	W_i	W_f	x	y
5.505	5.580	12.67		6.020	6.147		5.50
5.580	5.653	12.87		6.163	6.239	12.37	
5.653	5.727	12.64		6.239	6.316	12.30	
5.753	5.888		5.17	6.331	6.394		5.57
5.888	6.020		5.32				

Run No. 309

0.333 BeF_2 at 600°C, w = 0.500, T = 297°K, a = 2.30, c = 7.45

W_i	W_f	x	y	W_i	W_f	x	y
0.000	0.037	2.55		5.107	5.188		5.20
0.037	0.051	6.49		5.188	5.256		5.21
0.051	0.075	7.92		5.256	5.337		5.21
0.075	0.118	8.92		5.337	5.403		5.32
0.118	0.157	9.49		5.424	5.489	5.82	
0.199	0.245		1.53	5.489	5.588	5.79	
0.253	0.291	9.93		5.588	5.688	5.66	
0.291	0.328	10.18		5.740	5.819		5.40
0.328	0.366	9.96		5.819	5.925		5.32
0.366	0.403	9.99		5.925	6.000		5.64
0.435	0.472		1.87	6.000	6.076		5.53
0.472	0.505		2.14	6.100	6.164	5.91	
0.505	0.534		2.38	6.164	6.265	5.63	
0.546	0.585	9.62		6.265	6.368	5.51	
0.585	0.664	9.53		6.368	6.464	5.95	
0.664	0.747	9.03		6.502	6.577		5.60
0.780	0.835		2.54	6.577	6.652		5.62
0.835	0.884		2.86	6.652	6.728		5.63
0.884	0.952		3.05	6.728	6.806		5.41
0.960	1.023	8.96		6.811	6.881	5.45	
1.023	1.089	8.60		7.574	7.645	5.36	
1.089	1.177	8.58		7.645	7.755	5.18	
1.211	1.276		3.26	7.804	7.878		5.75
1.276	2.017		3.78	7.878	7.951		5.74
2.035	2.110	7.59		7.951	8.026		5.67
2.110	2.186	7.37		8.026	8.101		5.58
2.186	2.265	7.22		8.124	8.192	5.57	
2.310	2.377		4.17	8.192	8.261	5.54	
2.377	2.428		4.09	8.261	8.334	5.18	
2.428	2.478		4.22	8.336	8.408	5.09	
2.478	2.559		4.33	8.440	8.515		5.63
2.564	2.670	7.14		8.515	8.591		5.54
2.670	2.750	7.04		8.596	8.671	5.08	
4.879	4.973	6.05		8.671	8.745	5.08	
4.973	5.036	6.01		8.764	8.827		5.66
5.036	5.099	6.00		8.827	8.890		5.57

Run No. 310

0.333 BeF_2 at 600°C, w = 0.500, T = 297°K, a = 0.000, c = 0.00

W_i	W_f	x	y	W_i	W_f	x	y
0.000	0.000	5.08	5.59	0.345	0.411		2.13
0.000	0.028		4.92	0.420	0.456	2.66	
0.028	0.054		5.43	0.456	0.494	2.49	
0.054	0.082		4.99	0.494	0.578	2.26	
0.082	0.115		4.32	0.616	0.712		1.46
0.127	0.148	3.68		0.712	0.835		1.14
0.148	0.180	3.59		0.835	0.903		1.03
0.180	0.209	3.22		0.903	0.978		0.94
0.209	0.240	3.10		1.016	1.078	1.53	
0.250	0.294		3.22	1.078	1.208	1.46	
0.294	0.345		2.72				

Run No. 311

0.333 BeF_2 at 700°C, w = 0.500, T = 300°K, a = 2.35, c = 7.35

W_i	W_f	x	y	W_i	W_f	x	y
0.016	0.029	7.29		1.637	1.707		1.99
0.029	0.054	11.37		1.773	1.818	12.70	
0.054	0.083	12.88		1.818	1.878	12.45	
0.083	0.111	13.49		1.878	1.939	12.46	
0.111	0.139	13.49		1.939	1.969	12.57	
0.255	0.311		1.35	2.017	2.124		1.96
0.317	0.344	14.14		2.124	2.225		2.05
0.344	0.370	14.09		2.236	2.281	12.49	
0.370	0.398	13.76		2.281	2.344	12.04	
0.398	0.425	13.68		2.344	2.406	12.12	
0.425	0.481	13.58		2.406	2.468	12.08	
0.481	0.508	13.80		2.504	2.605		2.07
0.556	0.667		1.25	2.605	2.700		2.18
0.667	0.793		1.66	2.700	2.796		2.17
0.812	0.885	12.95		2.811	2.874	11.86	
0.885	0.914	12.99		2.874	2.938	11.82	
0.914	0.943	13.03		2.938	3.003	11.70	
0.943	0.972	12.82		3.003	3.066	11.84	
1.032	1.121		1.55	5.386	5.453		3.10
1.121	1.204		1.68	5.453	5.520		3.10
1.210	1.239	13.13		5.616	5.666	11.32	
1.239	1.283	12.82		5.666	5.719	10.63	
1.283	1.328	12.67		5.719	5.771	10.86	
1.328	1.373	12.58		5.771	5.824	10.68	
1.444	1.526		1.71	5.862	5.972		3.16
1.526	1.637		1.87	5.972	6.127		3.12

(continued)

Run No. 311 (continued)

W_i	W_f	x	y	W_i	W_f	x	y
6.127	6.258		3.18	9.359	9.497		3.53
6.264	6.335	10.63		9.497	9.614		3.54
6.335	6.388	10.55		9.614	9.771		3.54
6.388	6.460	10.45		9.786	9.863	9.74	
6.460	6.515	10.37		9.863	9.942	9.49	
6.597	6.703		3.25	9.942	10.06	9.25	
6.703	6.813		3.18	10.06	10.14	9.43	
6.813	6.921		3.20	10.18	10.30		3.54
6.921	7.028		3.26	10.30	10.42		3.60
7.089	7.144	10.22		10.42	10.53		3.66
7.144	7.218	10.20		10.60	11.01	9.59	
7.250	7.353		3.36	11.02	11.13		3.57
7.381	7.998		3.37	11.13	11.25		3.59
7.998	8.099		3.43	11.26	11.32	9.40	
8.185	8.259	10.18		11.32	11.38	9.32	
8.259	8.335	9.92		11.38	11.44	9.16	
8.335	8.410	10.00		12.05	12.13	9.38	
8.410	8.487	9.84		12.13	12.21	9.10	
8.530	8.655		3.34	12.21	12.29	9.22	
8.655	8.775		3.46	12.33	12.45		3.59
8.775	8.898		3.40	12.45	12.56		3.72
8.898	8.958		3.49	12.56	12.68		3.72
8.965	9.041	9.90		12.68	12.75		3.88
9.041	9.119	9.72		12.75	12.84	9.28	
9.144	9.221	9.70		12.84	12.92	9.09	
9.221	9.301	9.50		12.92	13.01	9.08	

Run No. 313

0.333 BeF_2 at 500°C, w = 0.500, T = 298°K, a = 0.00, c = 16.30

W_i	W_f	x	y	W_i	W_f	x	y
0.023	0.051	6.70		0.501	0.542		8.79
0.051	0.071	9.51		0.542	0.580		9.17
0.071	0.089	10.20		0.593	0.641	7.83	
0.089	0.126	10.37		0.641	0.690	7.62	
0.154	0.202		5.97	0.690	0.740	7.64	
0.202	0.232		6.87	0.740	0.804	7.34	
0.232	0.263		6.88	0.830	0.898		10.39
0.270	0.310	9.26		0.898	0.965		10.66
0.310	0.351	9.30		0.965	1.029		11.07
0.351	0.393	8.95		1.039	1.067	6.60	
0.393	0.436	8.84		1.067	1.096	6.49	
0.450	0.477		7.75	1.096	1.155	6.42	
0.477	0.501		8.80	1.174	1.205		11.13

(continued)

Run No. 313 (continued)

W_i	W_f	x	y	W_i	W_f	x	y
1.205	1.237		11.28	2.685	2.725	4.75	
1.513	1.546	5.75		2.725	2.806	4.60	
1.546	1.613	5.58		2.834	2.917		12.82
1.613	1.648	5.43		2.917	2.971		13.16
1.660	1.690		11.86	2.971	3.036		13.07
1.690	1.748		12.22	3.036	3.089		13.25
1.748	1.806		12.18	3.100	3.143	4.40	
1.806	1.864		12.18	3.143	3.187	4.28	
1.864	1.923		12.03	3.187	3.233	4.10	
1.947	2.021	5.07		3.233	3.321	4.26	
2.021	2.095	5.08		3.340	3.394		13.08
2.095	2.172	4.88		3.394	3.448		13.16
2.172	2.211	4.88		3.448	3.501		13.32
2.265	2.322		12.62	3.501	3.606		13.57
2.322	2.377		12.83	3.635	3.730	3.99	
2.377	2.432		12.93	3.730	3.825	3.96	
2.432	2.486		12.97	3.825	3.921	3.93	
2.486	2.540		13.13	3.942	4.049		13.21
2.568	2.646	4.84		4.049	4.154		13.55
2.646	2.685	4.83					

Run No. 314

0.333 BeF_2 at 500°C, w = 0.500, T = 298°K, a = 0.00, c = 0.00

W_i	W_f	x	y	W_i	W_f	x	y
0.000	0.000	2.79	14.47	0.580	0.657		3.64
0.004	0.049		12.59	0.675	0.768	1.01	
0.049	0.078		12.39	0.768	0.867	0.95	
0.078	0.124		7.63	0.867	0.969	0.92	
0.124	0.175		6.91	1.002	1.142		2.53
0.204	0.272	1.39		1.142	1.244		2.08
0.272	0.358	1.32		1.244	1.373		1.93
0.388	0.445		4.97	1.399	1.511	0.67	
0.445	0.509		4.47	1.511	1.668	0.60	
0.509	0.580		4.00				

Run No. 315

0.333 BeF_2 at 550°C, w = 0.500, T = 300°K, a = 0.00, c = 20.00

W_i	W_f	x	y	W_i	W_f	x	y
0.020	0.039	10.08		1.584	1.729	7.78	
0.039	0.058	14.92		1.729	1.775	8.34	
0.058	0.075	16.25		1.775	1.821	8.07	
0.075	0.099	15.72		1.821	1.882	9.25	
0.099	0.121	17.30		1.933	1.984		13.47
0.121	0.132	16.80		1.984	2.034		13.87
0.158	0.189		4.51	2.055	2.105	7.62	
0.189	0.215		5.36	2.105	2.154	7.66	
0.219	0.244	15.11		2.154	2.204	7.49	
0.244	0.270	14.96		2.252	2.317		13.89
0.270	0.295	14.54		2.317	2.365		14.47
0.321	0.354		6.37	2.365	2.403		14.70
0.354	0.380		7.93	3.672	3.733	6.18	
0.380	0.406		7.84	3.733	3.795	6.09	
0.406	0.432		8.21	3.795	3.859	5.90	
0.434	0.464	13.07		3.942	3.988		14.96
0.464	0.492	13.07		3.988	4.033		15.39
0.492	0.522	12.92		4.033	4.077		15.79
0.522	0.567	12.32		4.077	4.121		15.79
0.585	0.609		8.71	4.143	4.206	6.04	
0.609	0.631		9.36	4.206	4.266	6.21	
0.631	0.652		10.01	4.266	4.328	6.07	
0.652	0.686		10.29	4.328	4.390	6.08	
0.686	0.718		10.54	4.440	4.506		15.76
0.731	0.764	11.51		4.506	4.570		16.26
0.764	0.798	11.12		4.570	4.633		16.42
0.798	0.833	10.63		4.633	4.697		16.34
0.833	0.887	10.55		4.716	4.780	5.91	
0.975	1.007		10.84	4.780	4.845	5.78	
1.007	1.040		10.62	4.845	4.909	5.88	
1.040	1.070		11.30	4.909	4.993	5.60	
1.070	1.101		11.21	5.017	5.082		16.00
1.113	1.151	9.80		5.082	5.145		16.38
1.151	1.212	9.37		5.145	5.221		16.39
1.212	1.285	9.05		5.221	5.283		17.04
1.285	1.348	8.93		5.313	5.358	5.12	
1.411	1.453		13.07	5.358	5.479	5.43	
1.453	1.491		12.80	5.479	5.525	6.16	
1.491	1.528		13.29	5.525	5.594	5.46	
1.528	1.580		13.18	5.594	5.646	5.42	

Run No. 316

0.333 BeF_2 at 550°C, w = 0.500, T = 300°K, a = 0.00, c = 0.00

W_i	W_f	x	y	W_i	W_f	x	y
0.000	0.000	4.74	16.71	0.268	0.308	2.36	
0.000	0.063	4.80		0.308	0.348	2.36	
0.063	0.079	4.63		0.348	0.390	2.24	
0.079	0.103	3.96		0.390	0.477	2.17	
0.103	0.131	3.29		0.477	0.574	1.95	
0.131	0.162	3.01		0.574	0.676	1.82	
0.162	0.195	2.87		0.676	0.796	1.58	
0.195	0.232	2.55		0.796	1.674	1.31	
0.232	0.268	2.62		1.674	1.808	0.84	

Run No. 319

0.333 BeF_2 at 650°C, w = 0.500, T = 299°K, a = 0.00, c = 19.90

W_i	W_f	x	y	W_i	W_f	x	y
0.024	0.038	13.92		1.007	1.028		6.45
0.038	0.067	19.53		1.028	1.049		6.82
0.067	0.092	22.14		1.049	1.078		7.00
0.092	0.118	22.51		1.084	1.106	17.32	
0.118	0.142	23.18		1.106	1.128	16.68	
0.142	0.166	23.66		1.128	1.163	16.20	
0.166	0.190	23.26		1.174	1.288		6.68
0.204	0.255		2.70	1.288	1.675		8.07
0.255	0.298		3.20	1.675	1.715		8.68
0.298	0.334		3.88	1.728	1.755	14.00	
0.342	0.359	22.21		1.755	1.782	14.05	
0.359	0.385	22.37		1.782	1.809	14.09	
0.385	0.411	21.66		1.809	1.834	14.63	
0.411	0.438	21.08		1.834	1.861	14.05	
0.438	0.464	21.20		1.891	1.948		8.60
0.465	0.512		4.42	1.948	1.984		9.50
0.512	0.541		4.83	1.984	2.023		8.91
0.541	0.567		5.29	2.023	2.060		9.29
0.567	0.593		5.30	2.066	2.095	13.36	
0.595	0.623	20.16		2.095	2.122	13.51	
0.623	0.652	19.26		2.122	2.149	14.45	
0.652	0.682	18.95		2.149	2.176	13.80	
0.682	0.712	18.68		2.176	2.204	13.51	
0.769	0.806		5.54	2.277	2.315		8.95
0.806	0.829		6.17	2.315	2.353		9.17
0.870	0.902	17.78		2.353	2.392		8.96
0.902	0.932	18.53		2.392	2.429		9.33
0.932	0.993	18.55					

Run No. 501

0.333 BeF_2 at 650°C, w = 0.500, T = 298°K, a = 9.75,
b = -0.24, c = 0.65, d = -0.0095

W_i	W_f	x	y	W_i	W_f	x	y
0.000	0.000	0.46		2.025	2.086	6.17	
0.038	0.075	1.01		2.086	2.117	6.13	
0.075	0.105	1.26		2.117	2.178	6.16	
0.105	0.132	1.40		2.225	2.305		2.55
0.132	0.154	1.71		2.305	2.388		2.49
0.215	0.313		0.70	2.388	2.468		2.58
0.313	0.398		0.82	2.468	2.546		2.64
0.409	0.427	4.08		2.564	2.595	6.16	
0.427	0.456	4.03		2.595	2.654	6.29	
0.456	0.484	3.93		2.654	2.714	6.34	
0.484	0.511	4.22		2.714	2.774	6.32	
0.511	0.536	4.46		2.897	2.952		2.53
0.536	0.569	4.53		2.952	3.009		2.42
0.613	0.657		1.59	3.009	3.064		2.49
0.657	0.697		1.71	3.064	3.118		2.54
0.697	0.771		1.86	3.118	3.203		2.42
0.771	0.841		1.95	3.203	3.289		2.41
0.859	0.886	5.49		3.312	3.372	6.33	
0.886	0.922	5.22		3.372	3.432	6.32	
0.922	0.956	5.62		3.432	3.493	6.17	
0.956	0.989	5.67		3.493	3.553	6.20	
0.989	1.030	5.49		3.553	3.614	6.25	
1.059	1.122		2.20	3.672	3.799		2.17
1.122	1.180		2.37	3.799	3.925		2.20
1.180	1.237		2.40	3.925	4.021		2.14
1.255	1.288	5.86		4.021	4.117		2.13
1.288	1.320	5.84		4.200	4.274	6.32	
1.336	1.855		2.51	4.274	4.335	6.25	
1.855	1.930		2.76	4.335	4.394	6.34	
1.957	1.987	6.20		4.394	4.453	6.42	
1.987	2.025	6.03		4.502	4.567		2.13

Run No. 503

0.333 BeF_2 at 500°C, w = 0.500, T = 298°K, a = 5.15
b = -0.10, c = 0.26, d = -0.0050

W_i	W_f	x	y	W_i	W_f	x	y
0.049	0.138	0.21		0.720	0.795		0.92
0.229	0.437		0.33	0.795	0.865		0.98
0.466	0.523	0.66		0.865	0.934		1.00
0.523	0.576	0.71		0.973	1.005	1.18	

(continued)

Run No. 503 (continued)

W_i	W_f	x	y	W_i	W_f	x	y
1.005	1.070	1.17		4.098	4.142	2.54	
1.070	1.157	1.29		4.142	4.183	2.75	
1.157	1.219	1.22		4.183	4.237	2.78	
1.237	1.305		1.52	4.237	4.292	2.78	
1.305	1.373		1.52	4.322	4.398		1.82
1.373	3.147		1.98	4.398	4.475		1.78
3.147	3.211		2.14	4.475	4.559		1.80
3.278	3.310	2.38		4.559	4.637		1.77
3.310	3.341	2.37		4.666	4.731	2.91	
3.341	3.405	2.36		4.731	4.796	2.88	
3.434	3.497	2.40		4.796	4.859	2.97	
3.680	3.724	2.61		4.879	4.958		1.73
3.724	3.766	2.66		4.958	5.046		1.56
3.795	3.860		2.10	5.046	5.142		1.43
3.860	3.928		2.03	5.142	5.232		1.53
3.928	3.997		1.99	5.261	5.319	3.20	
3.997	4.067		1.98	5.319	5.377	3.25	

Run No. 509

0.333 BeF_2 at 544°C, w = 0.470, T = 298°K, a = 4.35, c = 7.70

W_i	W_f	x	y	W_i	W_f	x	y
0.000	0.018	2.26		0.611	0.647	8.18	
0.018	0.034	4.92		0.674	0.717		4.22
0.034	0.058	6.75		0.717	0.748		4.51
0.058	0.078	7.84		0.748	0.780		4.54
0.078	0.104	7.83		0.780	0.810		4.62
0.104	0.128	8.22		0.823	0.857	8.36	
0.128	0.152	8.29		0.857	0.891	8.08	
0.161	0.193		2.24	0.891	0.927	7.91	
0.193	0.220		2.67	0.927	0.963	7.80	
0.220	0.244		2.88	0.963	0.999	7.80	
0.252	0.276	8.53		1.072	1.131		4.83
0.276	0.299	8.63		1.131	1.185		5.29
0.299	0.322	8.53		1.185	1.225		5.34
0.322	0.345	8.76		1.240	1.278	7.32	
0.345	0.368	8.72		1.278	1.316	7.38	
0.387	0.412		2.90	1.316	1.354	7.38	
0.412	0.434		3.20	1.354	1.393	7.25	
0.434	0.474		3.60	1.393	1.432	7.28	
0.474	0.511		3.88	1.493	1.543		5.71
0.518	0.541	8.63		1.543	1.593		5.71
0.541	0.574	8.97		1.593	1.642		5.83
0.574	0.611	8.30		1.642	1.692		5.76

(continued)

Run No. 509 (continued)

W_i	W_f	x	y	W_i	W_f	x	y
1.714	1.809	6.78		3.061	3.116		6.43
1.809	1.911	6.67		3.137	3.184	6.37	
1.951	1.997		6.16	3.184	3.231	6.33	
1.997	2.044		6.05	4.433	4.485	5.76	
2.044	2.093		5.91	4.485	4.537	5.76	
2.093	2.139		6.17	4.537	4.589	5.84	
2.139	2.186		6.03	4.651	4.703		6.90
2.206	2.246	6.92		4.703	4.756		6.82
2.246	2.288	6.67		4.756	4.807		6.92
2.288	2.330	6.67		4.807	4.912		6.83
2.330	2.373	6.55		4.938	4.989	5.86	
2.417	2.462		6.32	4.989	5.042	5.71	
2.462	2.508		6.24	5.042	5.094	5.70	
2.508	2.553		6.30	5.094	5.148	5.62	
2.553	2.644		6.29	5.214	5.265		6.99
2.662	2.723	6.60		5.265	5.368		6.99
2.723	2.796	6.63		5.368	5.468		7.13
2.796	2.858	6.42		5.468	5.567		7.22
2.858	2.921	6.41		5.590	5.668	5.66	
2.947	3.005		6.17	5.668	5.740	5.57	
3.005	3.061		6.41				

Run No. 511

0.333 BeF_2 at 550°C, w = 0.470, T = 298°K, a = 10.30, b = -0.10, c = 0.40, d = -0.004

W_i	W_f	x	y	W_i	W_f	x	y
0.004	0.095	0.44		0.974	1.019		3.21
0.161	0.263		0.35	1.035	1.066	3.90	
0.274	0.309	1.70		1.066	1.097	3.96	
0.309	0.340	1.95		1.097	1.145	4.10	
0.340	0.379	2.09		1.145	1.194	4.17	
0.413	0.471		1.23	1.194	1.241	4.25	
0.471	0.531		1.78	1.262	1.365		3.42
0.531	0.584		2.01	1.365	1.424		3.58
0.605	0.640	2.87		1.424	1.520		3.70
0.640	0.679	3.05		1.520	1.576		3.83
0.679	0.718	3.10		1.592	1.633	4.88	
0.718	0.754	3.32		1.633	1.673	5.00	
0.783	0.836		2.70	1.673	1.736	5.09	
0.836	0.883		3.00	1.736	1.776	5.01	
0.883	0.928		3.10	1.805	1.881		3.68
0.928	0.974		3.08	1.881	1.961		3.57

(continued)

Run No. 511 (continued)

W_i	W_f	x	y	W_i	W_f	x	y
1.961	2.039		3.62	3.692	3.742	8.05	
2.039	2.117		3.64	3.780	3.867		2.47
2.117	2.195		3.62	3.867	3.958		2.32
2.219	2.254	5.62		3.958	4.055		2.20
2.254	2.290	5.67		4.061	4.107	8.60	
2.290	2.358	5.88		4.107	4.154	8.66	
2.358	2.425	5.95		4.154	4.209	8.99	
2.450	2.543		3.05	4.209	4.244	8.59	
2.543	2.608		3.26	4.285	4.341		1.90
2.608	2.676		3.14	4.341	4.403		1.72
2.676	2.742		3.20	4.403	4.469		1.61
2.742	2.812		3.04	4.469	4.538		1.52
2.836	2.915	6.66		4.538	4.608		1.52
2.915	2.985	6.87		4.622	4.666	8.97	
2.985	3.054	6.93		4.666	4.711	9.08	
3.054	3.111	7.04		4.711	4.755	9.05	
3.158	3.243		2.51	4.755	4.799	9.05	
3.243	3.321		2.72	4.838	4.892		1.30
3.321	3.402		2.62	4.892	4.945		1.33
3.402	3.488		2.47	4.945	4.997		1.37
3.524	3.576	7.62		4.997	5.115		1.20
3.576	3.643	7.87		5.129	5.171	9.46	
3.643	3.692	8.00		5.171	5.214	9.42	

Run No. 513

0.333 BeF_2 at 650°C, w = 0.470, T = 298°K, a = 4.60, c = 7.90

W_i	W_f	x	y	W_i	W_f	x	y
0.000	0.016	2.55		0.504	0.536		2.26
0.016	0.037	7.53		0.552	0.582	13.43	
0.037	0.072	11.34		0.582	0.612	13.71	
0.072	0.104	12.68		0.612	0.656	13.39	
0.104	0.137	13.20		0.656	0.702	13.18	
0.137	0.167	13.39		0.781	0.832		2.79
0.187	0.220		2.18	0.832	0.878		3.10
0.220	0.253		2.13	0.878	0.922		3.20
0.270	0.298	14.22		0.941	0.989	12.66	
0.298	0.326	14.00		0.989	1.050	12.43	
0.326	0.356	13.79		1.050	1.102	12.30	
0.356	0.385	13.71		1.136	1.181		3.18
0.405	0.440		1.99	1.181	1.222		3.41
0.440	0.473		2.17	1.222	1.262		3.54
0.473	0.504		2.25	1.262	1.303		3.47

(continued)

Run No. 513 (continued)

W_i	W_f	x	y	W_i	W_f	x	y
1.320	1.370	12.04		2.686	2.724	10.68	
1.370	1.421	11.91		4.185	4.227	9.63	
1.421	1.472	11.79		4.227	4.268	9.63	
1.558	1.595		3.82	4.350	4.389		5.43
1.595	1.629		4.21	4.389	4.428		5.41
1.629	1.681		4.07	4.428	4.467		5.45
1.697	1.749	11.38		4.467	4.507		5.38
1.749	1.802	11.38		4.529	4.591	9.71	
1.802	1.874	11.14		4.591	4.654	9.46	
1.874	1.929	11.05		4.654	4.718	9.40	
1.997	2.048		4.17	4.718	4.783	9.33	
2.048	2.134		4.13	4.846	4.905		5.41
2.134	2.230		4.42	4.905	4.953		5.88
2.253	2.307	11.25		4.953	5.005		5.49
2.307	2.363	10.82		5.005	5.055		5.63
2.363	2.419	10.66		5.073	5.137	9.40	
2.458	2.505		4.51	5.137	5.201	9.37	
2.505	2.552		4.53	5.201	5.266	9.17	
2.552	2.599		4.54	5.266	5.309	9.33	
2.615	2.650	11.29		5.309	5.353	9.25	
2.650	2.686	11.13					

Run No. 514

0.333 BeF_2 at 650°C, w = 0.470, t = 298°K, a = 0.00, c = 0.00

W_i	W_f	x	y	W_i	W_f	x	y
0.000	0.000	9.34	5.59	0.287	0.343	4.26	
0.000	0.010	8.38		0.343	0.374	3.87	
0.010	0.025	7.86		0.374	0.406	3.76	
0.025	0.041	7.28		0.406	0.439	3.64	
0.041	0.059	6.84		0.478	0.529		1.41
0.059	0.077	6.62		0.529	0.590		1.15
0.115	0.151		3.96	0.590	0.660		1.01
0.151	0.171		3.47	0.660	0.742		0.86
0.171	0.194		3.09	0.761	0.814	2.30	
0.194	0.221		2.63	0.814	0.869	2.16	
0.221	0.249		2.50	0.869	0.931	1.96	
0.261	0.287	4.62		0.931	0.972	1.92	

Run No. 523

0.333 BeF_2 at 700°C, w = 0.470, T = 298°K, a = 4.30, c = 7.65

W_i	W_f	x	y	W_i	W_f	x	y
0.000	0.016	2.43		2.279	2.317	13.00	
0.016	0.036	8.18		2.317	2.359	12.93	
0.036	0.061	11.96		2.359	2.395	12.89	
0.061	0.092	13.20		2.395	2.434	12.86	
0.092	0.120	13.92		2.590	2.637		3.09
0.120	0.148	14.21		2.637	2.685		3.04
0.148	0.189	14.80		2.685	2.731		3.14
0.322	0.348	15.57		2.731	2.777		3.12
0.348	0.386	15.78		2.793	2.840	12.70	
0.386	0.425	15.36		2.840	2.914	12.32	
0.425	0.465	15.12		2.914	2.946	12.49	
0.496	0.549		1.37	2.958	3.006	12.59	
0.549	0.596		1.55	3.006	3.054	12.51	
0.596	0.644		1.51	3.054	3.103	12.29	
0.644	0.691		1.52	3.141	3.185		3.29
0.705	0.738	15.00		3.185	3.228		3.36
0.738	0.772	14.72		3.228	3.292		3.41
0.772	0.806	14.72		3.292	3.355		3.43
0.806	0.841	14.58		3.355	3.418		3.47
0.841	0.875	14.39		3.432	3.482	12.21	
0.886	0.976		1.64	3.482	3.531	12.12	
0.976	1.054		1.85	3.531	3.581	12.04	
1.054	1.127		2.00	3.581	3.631	12.01	
1.170	1.205	14.50		3.631	3.682	11.91	
1.205	1.240	14.25		3.741	3.802		3.62
1.240	1.275	14.14		3.802	3.862		3.60
1.275	1.311	13.88		3.862	3.941		3.67
1.311	1.347	13.95		3.941	4.020		3.70
1.432	1.496		2.28	4.020	4.097		3.74
1.496	1.556		2.41	4.107	4.158	11.82	
1.556	1.617		2.41	4.158	4.209	11.79	
1.617	1.675		2.49	4.209	4.260	11.71	
1.701	1.737	13.74		4.260	4.312	11.63	
1.737	1.775	13.45		4.312	4.363	11.62	
1.775	1.836	13.12		4.420	4.493		3.99
1.836	1.881	13.18		4.493	4.566		3.96
1.965	2.019		2.68	4.566	4.640		3.95
2.019	2.072		2.70	4.640	4.712		3.99
2.072	2.124		2.80	4.845	4.898	11.33	
2.124	2.176		2.82	4.898	4.952	11.26	
2.176	2.226		2.86	4.952	5.005	11.33	
2.240	2.279	13.14					

Run No. 525

0.333 BeF_2 at 550°C, w = 0.470, T = 298°K, a = 10.20,
b = -0.19, c = 0.40, d = -0.007

W_i	W_f	x	y	W_i	W_f	x	y
0.039	0.181	0.28		3.171	3.246		3.86
0.213	0.301		0.41	3.276	3.340	3.10	
0.301	0.395		0.39	3.340	3.407	3.00	
0.435	0.479	0.90		3.437	3.500	3.14	
0.479	0.519	1.00		3.500	3.565	3.13	
0.519	0.553	1.18		3.585	3.662		3.75
0.553	0.586	1.24		3.662	3.738		3.80
0.653	0.715		1.16	3.738	3.813		3.88
0.715	0.762		1.53	3.813	3.886		3.96
0.762	0.808		1.58	3.920	4.048	3.42	
0.808	0.870		1.76	4.048	4.107	3.42	
0.899	0.934	1.71		4.107	4.164	3.51	
0.934	0.980	1.75		4.185	4.240		3.99
0.980	1.037	1.75		4.240	4.313		3.96
1.037	1.092	1.82		4.313	4.387		3.90
1.114	1.177		2.32	4.387	4.461		3.93
1.177	1.232		2.63	4.498	4.553	3.64	
1.232	1.284		2.75	4.553	4.634	3.72	
1.284	1.351		2.71	4.634	4.713	3.80	
1.396	1.446	2.45		4.713	4.766	3.75	
1.446	1.519	2.17		4.785	4.864		3.68
1.519	1.594	2.16		4.864	4.943		3.66
1.594	1.665	2.26		4.943	5.024		3.59
1.679	1.747		3.20	5.024	5.105		3.60
1.747	1.809		3.50	5.105	5.187		3.55
1.809	1.871		3.51	5.207	5.280	4.14	
1.871	1.932		3.62	5.280	5.355	4.01	
1.958	2.029	2.51		5.355	5.428	4.12	
2.029	2.094	2.49		5.428	5.500	4.18	
2.094	2.157	2.54		5.500	5.571	4.20	
2.157	2.249	2.62		5.595	5.684		3.24
2.275	2.333		3.78	5.684	5.771		3.33
2.333	2.390		3.82	5.771	5.858		3.33
2.390	2.465		3.87	5.882	5.948	4.54	
2.465	2.560		3.82	5.948	6.014	4.57	
2.584	2.655	2.83		6.014	6.057	4.66	
2.655	2.726	2.83		6.084	6.147	4.80	
2.726	2.799	2.75		6.166	6.241		2.91
2.799	2.869	2.84		6.241	6.312		3.08
2.869	2.969	2.80		6.312	6.383		3.05
2.984	3.040		3.90	6.383	6.454		3.05
3.040	3.096		3.92	6.454	6.527		3.00
3.096	3.171		3.86	6.554	6.614	5.00	

(continued)

Run No. 525 (continued)

W_i	W_f	x	y
6.614	6.674	4.84	
6.676	6.737	5.00	
6.737	6.797	4.96	
6.797	6.856	5.12	
6.856	6.916	5.03	
6.952	7.035		2.60
7.035	7.115		2.72
7.115	7.197		2.67
7.197	7.278		2.68
7.309	7.344	5.62	
7.344	7.399	5.51	
7.399	7.453	5.54	

Run No. 527

0.333 BeF_2 at 651°C, w = 0.470, T = 298°K, a = 4.30, c = 7.63

W_i	W_f	x	y
0.017	0.037	4.08	
0.037	0.058	5.88	
0.058	0.085	7.20	
0.085	0.135	8.04	
0.135	0.169	9.38	
0.169	0.203	9.64	
0.203	0.234	10.21	
0.234	0.264	10.78	
0.264	0.294	10.78	
0.294	0.322	11.17	
0.402	0.438		1.99
0.438	0.470		2.29
0.470	0.499		2.53
0.509	0.526	11.82	
0.526	0.552	12.12	
0.552	0.593	11.87	
0.593	0.627	11.87	
0.627	0.661	11.84	
0.718	0.769		2.87
0.769	0.816		3.07
0.816	0.863		3.10
0.912	0.947	11.51	
0.947	1.000	11.51	
1.000	1.052	11.46	
1.052	1.087	11.34	
1.120	1.164		3.30
1.164	1.206		3.49
1.206	1.268		3.54
1.268	1.326		3.78
1.326	1.383		3.79
1.401	1.428	11.18	
1.428	1.481	11.20	
1.481	1.536	10.96	
1.536	1.592	10.82	
1.592	1.655	10.79	
1.718	1.773		4.01
1.773	1.827		4.05
1.827	1.879		4.20
1.879	1.930		4.28
1.930	1.981		4.30
2.019	2.075	10.59	
2.075	2.133	10.42	
2.133	2.192	10.21	
2.192	2.251	10.22	
2.251	2.318	10.12	
2.401	2.452		4.36
2.452	2.502		4.38
2.502	2.568		4.41
2.568	2.640		4.57
2.640	2.703		4.63
2.741	2.802	9.80	
2.802	2.864	9.71	
2.864	2.947	9.70	
2.947	3.030	9.58	
3.076	3.140		4.55
3.140	3.203		4.64
3.203	3.264		4.78
3.264	3.325		4.75
3.325	3.386		4.80
3.386	3.447		4.83
3.459	3.541	9.67	
3.541	3.625	9.63	
3.625	3.709	9.51	
3.709	3.793	9.47	

(continued)

Run No. 527 (continued)

W_i	W_f	x	y	W_i	W_f	x	y
3.863	3.931		4.80	4.167	4.225		5.07
3.931	3.991		4.92	4.246	4.311	9.17	
3.991	4.050		4.96	4.311	4.377	9.10	
4.050	4.109		4.91	4.377	4.444	9.01	
4.109	4.167		5.00				

Run No. 529

0.273 BeF_2 at 600°C, w = 0.550, T = 298°K, a = 19.00, b = -0.20, c = 0.70, d = -0.007

W_i	W_f	x	y	W_i	W_f	x	y
0.056	0.088	1.05		1.647	1.690	5.84	
0.088	0.115	1.24		1.690	1.734	5.88	
0.161	0.203		0.76	1.734	1.776	6.10	
0.203	0.231		1.09	1.832	1.947		8.14
0.253	0.272	2.62		1.947	1.986		8.01
0.272	0.299	2.54		1.986	2.025		8.01
0.299	0.354	2.82		2.025	2.064		7.97
0.383	0.404		2.92	2.089	2.137	7.16	
0.404	0.431		3.50	2.137	2.185	7.22	
0.431	0.454		4.00	2.185	2.232	7.24	
0.454	0.674		5.40	2.232	2.279	7.33	
0.674	0.701		6.84	2.279	2.325	7.41	
0.714	0.734	4.22		2.342	2.389		6.70
0.734	0.774	4.26		2.389	2.436		6.63
0.774	0.814	4.30		2.436	2.485		6.37
0.814	0.853	4.43		2.485	2.536		6.04
0.853	0.891	4.51		2.554	2.593	8.79	
0.943	0.985		7.42	2.593	2.632	8.71	
0.985	1.024		7.96	2.632	2.671	8.76	
1.024	1.063		7.96	2.671	2.710	8.84	
1.063	1.102		8.07	2.751	2.799		5.21
1.102	1.140		8.25	2.799	2.853		4.59
1.145	1.179	5.09		2.853	2.911		4.32
1.179	1.245	5.14		2.911	2.956		4.12
1.262	1.294	5.42		2.963	2.995	10.68	
1.294	1.326	5.38		2.995	3.042	10.92	
1.364	1.401		8.46	3.042	3.088	11.14	
1.401	1.439		8.36	3.088	3.133	11.37	
1.439	1.476		8.38	3.156	3.190		3.75
1.476	1.512		8.64	3.190	3.221		3.91
1.512	1.548		8.55	3.221	3.252		4.07
1.558	1.602	5.79		3.252	3.300		3.90
1.602	1.647	5.76		3.300	3.332		3.93

(continued)

Run No. 529 (continued)

W_i	W_f	x	y	W_i	W_f	x	y
3.346	3.384	13.39		3.622	3.656		3.60
3.384	3.422	13.68		3.669	3.702	15.63	
3.434	3.457	14.76		3.702	3.734	15.88	
3.457	3.494	13.96		3.734	3.765	16.57	
3.517	3.553		3.42	3.765	3.797	16.22	
3.553	3.586		3.78	3.876	3.904		3.36
3.586	3.622		3.51	3.904	3.932		3.37

Run No. 533

0.273 BeF_2 at 700°C, w = 0.520, T = 298°K, a = 4.47, c = 7.63

W_i	W_f	x	y	W_i	W_f	x	y
0.016	0.024	4.66		1.152	1.196		2.96
0.024	0.039	7.20		1.196	1.248		3.16
0.039	0.058	9.37		1.248	1.296		3.36
0.058	0.075	10.62		1.296	1.344		3.41
0.075	0.106	11.84		1.400	1.462	11.67	
0.118	0.152		1.90	1.462	1.509	11.59	
0.152	0.185		1.99	1.509	1.556	11.43	
0.185	0.217		2.03	1.556	1.619	11.47	
0.228	0.241	13.63		1.619	1.683	11.39	
0.241	0.268	13.75		1.730	1.783		3.71
0.268	0.294	13.55		1.783	1.836		3.70
0.294	0.321	13.71		1.836	1.904		3.82
0.321	0.348	13.43		1.904	1.954		3.96
0.393	0.425		2.07	1.954	2.004		3.90
0.425	0.453		2.28	2.005	2.070	11.21	
0.453	0.483		2.21	2.070	2.136	10.92	
0.483	0.512		2.20	2.136	2.203	10.83	
0.523	0.550	13.36		2.203	2.270	10.79	
0.550	0.577	13.28		2.304	2.352		4.08
0.577	0.606	12.91		2.352	2.413		4.30
0.606	0.634	12.93		2.413	2.472		4.41
0.634	0.662	12.68		2.472	2.531		4.40
0.700	0.753		2.46	2.540	2.590	10.76	
0.753	0.802		2.67	2.590	2.641	10.71	
0.802	0.850		2.71	2.641	2.692	10.64	
0.850	0.897		2.78	2.692	2.744	10.55	
0.904	0.946	12.89		2.744	2.795	10.50	
0.946	0.989	12.68		2.831	2.892		4.28
0.989	1.033	12.50		2.892	2.951		4.41
1.033	1.070	12.05		2.951	3.009		4.51
1.070	1.115	12.16		3.009	3.067		4.46

(continued)

Run No. 533 (continued)

W_i	W_f	x	y	W_i	W_f	x	y
3.075	3.128	10.20		3.323	3.378		4.70
3.128	3.182	10.16		3.378	3.434		4.67
3.182	3.236	10.04		3.441	3.498	10.12	
3.264	3.323		4.42	3.498	3.552	10.08	

Run No. 535

0.273 BeF_2 at 650°C, w = 0.520, T = 298°K, a = 15.00, b = -0.24, c = 0.55, d = -0.006

W_i	W_f	x	y	W_i	W_f	x	y
0.098	0.121	1.63		1.712	1.788		4.30
0.121	0.158	1.95		1.788	1.864		4.29
0.177	0.260		0.79	1.864	1.957		4.21
0.260	0.310		1.29	1.957	2.035		4.18
0.310	0.581		2.64	2.045	2.108	8.59	
0.581	0.616		3.67	2.108	2.192	8.60	
0.629	0.665	6.09		2.226	2.284	9.30	
0.665	0.709	6.16		2.312	2.414		3.83
0.709	0.752	6.28		2.414	2.466		3.75
0.752	0.796	6.21		2.466	2.538		3.67
0.796	0.838	6.43		2.538	2.609		3.67
0.838	0.879	6.59		2.632	2.681	9.32	
0.912	0.945		4.01	2.681	2.740	9.25	
0.945	0.991		4.25	2.740	2.817	9.38	
0.991	1.062		4.58	2.817	2.875	9.43	
1.062	1.134		4.54	2.922	2.978		3.47
1.134	1.204		4.63	2.978	3.036		3.37
1.231	1.281	7.22		3.036	3.178		3.21
1.281	1.332	7.14		3.193	3.247	10.08	
1.332	1.406	7.26		3.247	3.301	10.08	
1.406	1.481	7.28		3.301	3.355	10.11	
1.481	1.554	7.45		3.382	3.426		2.96
1.592	1.636		4.49	3.426	3.470		2.92
1.636	1.712		4.28	3.470	3.514		2.96

Run No. 537

0.273 BeF_2 at 650°C, w = 0.520, T = 298°K, a = 4.08, c = 7.63

W_i	W_f	x	y	W_i	W_f	x	y
0.016	0.028	3.07		0.071	0.090	5.64	
0.028	0.048	3.62		0.110	0.139		2.20
0.048	0.071	4.62		0.139	0.169		2.07

(continued)

Run No. 537 (continued)

W_i	W_f	x	y	W_i	W_f	x	y
0.169	0.198		2.17	1.958	2.010		5.51
0.216	0.259	8.42		2.010	2.075		5.34
0.259	0.299	9.05		2.075	2.131		5.63
0.299	0.339	9.03		2.159	2.226	8.10	
0.339	0.379	9.20		2.226	2.294	7.99	
0.379	0.419	9.05		2.294	2.362	8.01	
0.476	0.495		3.29	2.362	2.430	7.96	
0.495	0.546		3.68	2.430	2.499	7.86	
0.546	0.594		3.97	2.548	2.595		5.43
0.594	0.641		4.09	2.595	2.642		5.38
0.649	0.687	9.47		2.642	2.698		5.60
0.687	0.725	9.55		2.698	2.765		5.70
0.725	0.764	9.34		2.765	2.822		5.51
0.764	0.802	9.34		2.839	2.902	7.79	
0.802	0.842	9.10		2.902	2.979	7.70	
0.869	0.912		4.37	2.979	3.050	7.67	
0.912	0.954		4.50	3.050	3.121	7.63	
0.954	0.995		4.64	3.157	3.243		5.93
0.995	1.036		4.66	3.243	3.309		5.76
1.036	1.075		4.83	3.309	3.374		5.79
1.093	1.133	9.14		3.374	3.440		5.74
1.133	1.173	9.03		3.440	3.506		5.83
1.173	1.214	8.84		3.593	3.692	7.36	
1.214	1.255	8.84		3.692	3.766	7.36	
1.255	1.297	8.64		3.766	3.840	7.32	
1.317	1.356		4.90	3.840	3.914	7.32	
1.356	1.408		4.84	3.948	4.012		5.95
1.408	1.459		4.99	4.012	4.076		5.90
1.459	1.510		5.01	4.076	4.140		5.95
1.510	1.558		5.18	4.140	4.203		6.00
1.581	1.623	8.47		4.203	4.265		6.12
1.623	1.675	8.40		4.286	4.360	7.32	
1.675	1.719	8.33		4.360	4.436	7.21	
1.719	1.763	8.24		4.436	4.512	7.12	
1.763	1.828	8.30		4.545	4.598		6.03
1.864	1.911		5.33	4.598	4.650		6.05
1.911	1.958		5.38	4.650	4.702		6.09

Run No. 539

0.273 BeF_2 at 600°C, w = 0.520, T = 298°K, a = 14.50, b = -0.20, c = 0.55, d = -0.006

W_i	W_f	x	y	W_i	W_f	x	y
0.055	0.104	0.56		2.522	2.579		4.41
0.104	0.151	0.76		2.579	2.639		4.24
0.224	0.256		0.98	2.639	2.700		4.13
0.256	0.300		1.44	2.721	2.761	9.10	
0.300	0.338		1.66	2.761	2.809	9.01	
0.350	0.384	2.12		2.854	2.893	9.17	
0.384	0.425	2.67		2.893	2.953	9.10	
0.425	0.465	2.74		2.980	3.034		3.53
0.465	0.502	2.90		3.034	3.087		3.62
0.523	0.547		2.66	3.087	3.140		3.55
0.547	0.576		3.26	3.140	3.213		3.46
0.576	0.614		3.29	3.224	3.278	10.04	
0.614	0.649		3.63	3.278	3.332	10.09	
0.649	0.681		3.99	3.332	3.386	10.12	
0.696	0.729	3.84		3.386	3.438	10.34	
0.729	0.766	3.88		3.438	3.490	10.51	
0.766	0.812	3.99		3.507	3.578		2.67
0.812	0.856	4.08		3.578	3.649		2.68
0.909	0.947		4.96	3.649	3.725		2.51
0.947	0.984		5.12	3.725	3.807		2.29
0.984	1.019		5.45	3.822	3.867	11.95	
1.038	1.077	4.62		3.867	3.914	11.75	
1.077	1.135	4.70		3.914	3.960	11.75	
1.135	1.191	4.86		3.960	4.021	11.76	
1.191	1.246	4.96		4.050	4.116		1.91
1.266	1.302		5.22	4.116	4.190		1.72
1.302	1.349		5.40	4.190	4.269		1.60
1.349	1.393		5.76	4.269	4.351		1.54
1.393	1.437		5.74	4.365	4.422	12.55	
1.437	1.481		5.82	4.422	4.480	12.61	
1.502	1.567	5.57		4.480	4.536	12.78	
1.567	1.632	5.60		4.536	4.592	12.68	
1.632	1.695	5.72		4.624	4.670		1.37
1.695	1.756	5.90		4.670	4.720		1.29
1.756	1.816	6.10		4.720	4.773		1.18
1.848	1.870		5.64	4.785	4.827	13.13	
1.870	1.916		5.59	4.827	4.869	12.89	
1.916	1.961		5.64	4.869	4.912	12.68	
1.961	2.008		5.37	4.912	4.954	12.78	
2.008	2.077		5.45	4.982	5.041		1.07
2.096	2.147	7.08		5.041	5.103		1.01
2.147	2.198	7.10		5.103	5.163		1.06
2.198	2.248	7.16		5.163	5.230		0.94
2.248	2.298	7.33		5.272	5.314	12.87	
2.465	2.522		4.50	5.314	5.342	12.93	

Run No. 601

0.600 BeF_2 at 600°C, w = 0.424, T = 297°K, a = 13.7, c = 3.88

W_i	W_f	x	y	W_i	W_f	x	y
0.000	0.024	5.50		0.714	0.751		2.16
0.024	0.055	7.24		0.760	0.789	11.36	
0.055	0.091	8.54		0.789	0.819	11.09	
0.091	0.128	9.10		0.819	0.849	11.21	
0.128	0.164	9.20		0.849	0.879	11.13	
0.164	0.200	9.34		0.879	0.908	11.13	
0.266	0.312		1.73	1.027	1.094		2.41
0.312	0.357		1.80	1.094	1.158		2.49
0.373	0.406	9.91		1.158	1.256		2.46
0.406	0.439	10.14		1.256	1.320		2.49
0.439	0.472	10.21		1.331	1.358	12.37	
0.472	0.504	10.30		1.358	1.385	12.20	
0.504	0.537	10.14		1.385	1.439	12.17	
0.571	0.614		1.86	1.439	1.494	12.17	
0.614	0.675		1.96	1.494	1.566	12.38	
0.675	0.714		2.09	1.597	1.663		2.41

Run No. 603

0.600 BeF_2 at 701°C, w = 424, T = 297°K, a = 13.7, c = 3.88

W_i	W_f	x	y	W_i	W_f	x	y
0.000	0.035	2.54		1.140	1.191	17.59	
0.035	0.056	4.22		1.239	1.353		1.35
0.056	0.087	5.74		1.353	1.471		1.31
0.087	0.120	6.64		1.471	1.581		1.40
0.120	0.149	7.75		1.581	1.693		1.38
0.149	0.181	8.45		1.723	1.769	19.05	
0.237	0.301		1.20	1.769	1.818	18.95	
0.301	0.362		1.26	1.818	1.866	18.74	
0.362	0.433		1.31	1.866	1.913	18.89	
0.440	0.456	14.07		1.913	1.960	18.97	
0.456	0.487	14.38		2.153	2.265		1.37
0.487	0.534	14.30		2.265	2.322		1.36
0.534	0.579	14.66		2.335	2.358	19.21	
0.579	0.638	15.08		2.358	2.406	18.74	
0.694	0.752		1.32	2.406	2.453	18.70	
0.752	0.808		1.38	2.453	2.477	18.74	
0.808	0.865		1.36	2.564	2.623		1.32
0.865	0.949		1.40	2.623	2.680		1.36
0.987	1.038	17.39		2.685	2.709	19.13	
1.038	1.089	17.46		2.709	2.732	19.13	
1.089	1.140	17.29					

Run No. 605

0.600 BeF_2 at 500°C, w = 0.424, T = 297°K, a = 13.7, c = 3.88

W_i	W_f	x	y	W_i	W_f	x	y
0.010	0.046	2.45		2.143	2.201		3.99
0.046	0.070	3.76		2.201	2.253		4.50
0.070	0.104	3.82		2.253	2.306		4.34
0.164	0.235		1.09	2.306	2.355		4.70
0.235	0.304		1.12	2.400	2.483	5.34	
0.304	0.372		1.15	2.483	2.563	5.58	
0.372	0.434		1.24	2.563	2.657	5.66	
0.434	0.494		1.27	2.657	2.734	5.74	
0.494	0.548		1.44	2.734	2.810	5.84	
0.549	1.019		1.96	2.896	2.946		4.97
1.113	1.174	3.66		2.946	2.988		5.54
1.174	1.233	3.72		2.988	3.033		5.22
1.233	1.293	3.75		3.033	3.076		5.38
1.293	1.350	3.86		3.097	3.166	6.40	
1.350	1.408	3.86		3.166	3.235	6.43	
1.408	1.464	3.96		3.235	3.304	6.43	
1.481	1.508		2.80	3.387	3.427		5.82
1.508	1.557		3.17	3.427	3.465		6.12
1.557	1.629		3.20	3.465	3.531		5.79
1.660	1.771	4.40		3.531	3.607		6.12
1.771	1.867	4.60		3.668	3.726	7.58	
1.867	1.963	4.62		3.726	3.813	7.64	
1.963	2.056	4.79		3.813	3.902	7.53	

Run No. 607

0.600 BeF_2 at 500°C, w = 0.424, T = 297°K, a = 20.0
b = -1.15, c = 0.136, d = -0.041

W_i	W_f	x	y	W_i	W_f	x	y
0.039	0.065		5.91	0.908	1.009	8.76	
0.065	0.114		6.30	1.045	1.110		5.97
0.114	0.163		6.33	1.110	1.171		6.28
0.174	0.230	7.95		1.171	1.293		6.33
0.230	0.286	7.99		1.293	1.413		6.41
0.286	0.341	7.99		1.432	1.525	9.54	
0.377	0.439		6.29	1.525	1.620	9.42	
0.439	0.500		6.34	1.620	1.713	9.55	
0.500	0.572		6.38	1.713	1.805	9.62	
0.572	0.631		6.54	1.858	1.948		5.97
0.648	0.726	8.54		1.948	2.038		6.03
0.726	0.804	8.54		2.038	2.218		6.00
0.804	0.908	8.58		2.218	2.401		5.91

(continued)

Run No. 607 (continued)

W_i	W_f	x	y	W_i	W_f	x	y
2.419	2.527	10.29		3.452	3.564	9.90	
2.551	2.662	10.03		3.564	3.677	9.91	
2.662	2.774	9.93		3.706	3.796		4.29
2.835	2.906		5.47	3.796	3.900		4.47
2.906	2.978		5.34	3.900	3.978		4.95
2.978	3.127		5.18	3.978	4.060		4.66
3.127	3.202		5.13	4.084	4.191	10.39	
3.227	3.339	9.91		4.191	4.255	10.37	
3.339	3.452	9.84		4.255	4.319	10.37	

Run No. 611

0.600 BeF_2 at 600°C, w = 0.424, T = 297°K, a = 6.45, c = 5.90

W_i	W_f	x	y	W_i	W_f	x	y
0.000	0.019	2.29		1.669	1.701	13.91	
0.019	0.038	7.13		1.701	1.733	13.87	
0.038	0.065	10.01		1.733	1.765	13.67	
0.065	0.088	11.34		1.853	1.912		1.31
0.088	0.123	11.47		1.912	1.973		1.26
0.123	0.160	12.05		1.973	2.034		1.27
0.160	0.196	12.25		2.034	2.140		1.45
0.227	0.314		1.34	2.148	2.180	14.17	
0.314	0.371		1.34	2.180	2.212	13.75	
0.371	0.430		1.31	2.212	2.245	13.67	
0.443	0.475	13.79		2.245	2.277	13.63	
0.475	0.508	13.63		2.277	2.310	13.46	
0.508	0.541	13.54		2.395	2.556		1.44
0.541	0.573	13.66		2.556	2.653		1.59
0.573	0.606	13.83		2.661	2.693	13.79	
0.677	0.750		1.06	2.693	2.726	13.67	
0.750	0.827		1.00	2.726	2.759	13.46	
0.827	0.974		1.05	2.797	2.950		1.51
0.992	1.023	14.43		2.950	3.090		1.66
1.023	1.054	14.21		3.090	3.219		1.79
1.054	1.086	13.79		3.219	3.389		1.82
1.086	1.118	13.91		3.389	3.551		1.90
1.118	1.150	13.75		3.576	3.609	13.50	
1.244	1.315		1.08	3.609	3.643	13.01	
1.315	1.382		1.16	3.643	3.676	13.42	
1.382	1.515		1.16	3.676	3.710	13.12	
1.515	1.578		1.21	3.710	3.744	13.12	
1.606	1.637	14.43		3.818	3.941		1.68
1.637	1.669	14.13		3.941	4.057		1.99

(continued)

Run No. 611 (continued)

W_i	W_f	x	y	W_i	W_f	x	y
4.057	4.171		2.03	5.022	5.073	13.25	
4.171	4.282		2.09	5.073	5.123	13.25	
4.297	4.348	13.07		5.123	5.174	13.07	
4.348	4.399	12.89		5.174	5.225	12.97	
4.399	4.451	12.86		5.250	5.354		2.21
4.451	4.504	12.71		5.354	5.456		2.28
4.572	4.681		2.13	5.468	5.518	13.09	
4.681	4.788		2.16	5.518	5.570	12.87	
4.788	4.897		2.13	5.570	5.622	12.95	
4.897	5.003		2.18				

Run No. 619

0.400 BeF_2 at 550°C, w = 0.567, T = 299°K, a = 21.0, c = 0.68

W_i	W_f	x	y	W_i	W_f	x	y
0.000	0.000	7.63	5.00	0.884	0.929	11.13	
0.000	0.050	8.34		0.929	0.975	10.87	
0.050	0.148	8.50		0.975	1.035	11.04	
0.148	0.195	8.83		1.035	1.095	11.11	
0.294	0.337		6.66	1.169	1.210		6.99
0.337	0.379		6.90	1.210	1.266		7.18
0.379	0.419		7.17	1.266	1.314		7.25
0.431	0.481	9.96		1.366	1.477	11.93	
0.481	0.531	9.96		1.477	1.545	12.29	
0.531	0.582	9.92		1.545	1.613	12.17	
0.582	0.632	9.99		1.694	1.746		6.57
0.742	0.782		7.28	1.746	1.799		6.64
0.782	0.819		7.64	1.815	1.868	12.63	
0.819	0.865		7.55	1.868	1.921	12.54	

Run No. 621

0.400 BeF_2 at 604°C, w = 0.567, T = 299°K, a = 6.45, c = 5.90

W_i	W_f	x	y	W_i	W_f	x	y
0.004	0.017	2.57		0.216	0.261		1.27
0.017	0.026	7.22		0.261	0.329		1.24
0.026	0.041	8.84		0.385	0.409	13.47	
0.041	0.058	9.79		0.409	0.458	13.59	
0.058	0.083	10.47		0.458	0.496	13.29	
0.083	0.107	10.57		0.496	0.520	13.39	
0.107	0.129	11.08		0.597	0.637		1.42

(continued)

Run No. 621 (continued)

W_i	W_f	x	y	W_i	W_f	x	y
0.637	0.672		1.63	1.797	1.907		2.63
0.672	0.707		1.67	1.907	2.014		2.75
0.722	0.747	13.43		2.014	2.099		2.72
0.747	0.785	13.32		2.131	2.172	12.24	
0.785	0.822	13.36		2.172	2.214	12.00	
0.822	0.847	13.09		2.214	2.256	11.75	
0.863	0.896		1.75	2.256	2.298	11.87	
0.896	0.975		1.88	2.333	2.416		2.76
0.975	1.034		1.97	2.416	2.496		2.88
1.042	1.079	13.43		2.496	2.574		2.95
1.079	1.117	13.21		2.685	2.726	11.97	
1.117	1.156	12.82		2.726	2.769	11.75	
1.156	1.183	12.39		2.769	2.811	11.79	
1.247	1.300		2.17	2.811	2.847	11.59	
1.300	1.375		2.32	2.857	2.916		2.95
1.375	1.447		2.41	2.916	2.972		3.07
1.447	1.544		2.38	2.972	3.027		3.13
1.574	1.626	12.91		3.027	3.084		3.08
1.626	1.678	12.72		3.098	3.141	11.63	
1.678	1.730	12.64		3.141	3.184	11.63	

Run No. 625

0.400 BeF_2 at 550°C, w = 0.567, T = 299°K, a = 6.45, c = 5.90

W_i	W_f	x	y	W_i	W_f	x	y
0.004	0.020	2.08		0.649	0.677	11.95	
0.020	0.034	6.93		0.677	0.706	11.42	
0.034	0.052	9.47		0.706	0.734	11.83	
0.052	0.076	10.38		0.734	0.779	11.26	
0.076	0.099	10.80		0.779	0.807	11.64	
0.099	0.121	11.04		0.834	0.873		2.16
0.154	0.200		1.23	0.873	0.947		2.28
0.200	0.242		1.33	0.947	0.981		2.47
0.242	0.284		1.36	0.981	1.014		2.51
0.297	0.317	12.38		1.035	1.065	11.22	
0.317	0.337	12.47		1.065	1.094	11.28	
0.337	0.365	12.04		1.094	1.125	10.91	
0.365	0.392	11.92		1.125	1.155	10.80	
0.392	0.421	11.59		1.186	1.231		2.51
0.464	0.501		1.52	1.231	1.271		2.75
0.501	0.533		1.72	1.271	1.324		2.64
0.533	0.563		1.88	1.324	1.361		3.09
0.563	0.609		1.83	1.361	1.396		3.13
0.622	0.649	12.01		1.402	1.431	11.20	

(continued)

Run No. 625 (continued)

W_i	W_f	x	y	W_i	W_f	x	y
1.431	1.462	10.78		2.256	2.291	9.60	
1.462	1.494	10.53		2.325	2.374		3.50
1.494	1.590	10.38		2.374	2.418		3.78
1.625	1.662		3.01	2.418	2.463		3.75
1.662	1.699		3.03	2.480	2.514	9.66	
1.699	1.733		3.25	2.514	2.549	9.57	
1.733	1.767		3.29	2.549	2.584	9.57	
1.790	1.821	10.51		2.584	2.619	9.49	
1.821	1.854	10.37		2.670	2.712		4.07
1.854	1.886	10.34		2.712	2.754		3.96
1.886	1.918	10.21		2.754	2.797		3.93
1.955	1.990		3.25	2.818	2.853	9.53	
1.990	2.037		3.53	2.853	2.897	9.45	
2.037	2.069		3.50	2.897	2.932	9.36	
2.069	2.118		3.50	2.932	2.968	9.25	
2.138	2.172	10.05		2.997	3.040		3.96
2.172	2.205	9.96		3.040	3.081		4.08
2.205	2.526	9.75		3.081	3.122		4.14

Run No. 627

0.400 BeF_2 at 702°C, w = 0.567, T = 299°K, a = 6.70, c = 6.05

W_i	W_f	x	y	W_i	W_f	x	y
0.000	0.011	3.00		0.744	0.808		0.88
0.011	0.022	5.82		0.808	0.856		0.91
0.022	0.033	9.18		0.856	0.914		0.97
0.033	0.048	11.12		0.942	0.961	17.12	
0.048	0.068	12.43		0.961	0.992	16.32	
0.068	0.087	13.08		0.992	1.022	16.28	
0.087	0.106	13.49		1.022	1.043	16.17	
0.106	0.123	14.88		1.053	1.142		0.95
0.123	0.143	14.64		1.142	1.197		1.02
0.288	0.308	16.28		1.197	1.255		1.07
0.308	0.328	16.16		1.283	1.312	17.29	
0.328	0.349	16.51		1.312	1.341	16.87	
0.349	0.380	16.13		1.341	1.381	16.82	
0.413	0.472		0.96	1.381	1.421	16.49	
0.472	0.542		0.81	1.421	1.462	16.13	
0.542	0.602		0.93	1.495	1.576		1.03
0.611	0.631	16.87		1.576	1.648		1.17
0.631	0.651	16.51		1.648	1.717		1.22
0.651	0.671	16.28		1.736	1.766	16.59	
0.671	0.692	16.34		1.766	1.796	16.51	
0.692	0.712	16.05		1.796	1.837	16.20	

(continued)

Run No. 627 (continued)

W_i	W_f	x	y	W_i	W_f	x	y
1.837	1.878	16.30		2.265	2.296	16.01	
1.905	1.975		1.20	2.318	2.382		1.32
1.975	2.041		1.28	2.382	2.442		1.40
2.041	2.102		1.37	2.442	2.502		1.41
2.120	2.151	16.36		2.502	2.561		1.43
2.151	2.181	16.36		2.581	2.611	16.43	
2.181	2.223	15.96		2.611	2.642	15.87	
2.223	2.265	16.05		2.642	2.674	15.80	

APPENDIX C

Glossary

a (or a + bW) - influent partial pressure of HF

A - (a + 2c)

a_{BeF_2} - thermodynamic activity of BeF_2

α,β - parameters expressing the variation in γ_{BeF_2} with composition at a specified temperature according to the equation

$$\log \gamma_{BeF_2} = \alpha X^2_{LiF} + \beta X^4_{LiF}$$

c (or c + dW) - influent partial pressure of H_2O

C - integration constants

ΔC_p - heat capacity change at constant pressure

f - correction factor $= \dfrac{P_w - (x + y)}{P_w - p_w}$

ΔF_r - free energy of reaction

γ_{BeF_2} - activity coefficient of $BeF_2 = a_{BeF_2}/X_{BeF_2}$

ΔH^o_f - standard heat of formation

ΔH_{fusion} - heat of fusion

ΔH_r - heat of reaction

ΔH_{subl} - heat of sublimation

ΔH_{vap} - heat of vaporization

$\bar{H}_{BeF_2}$ - partial molal heat content of BeF_2 in solution

$H^o_{BeF_2}$ - partial molal heat content of pure liquid BeF_2

K - thermodynamic equilibrium constant

K_a - $X^2/ya_{BeF_2} = Q/a_{BeF_2}$

k, l, m - parameters used in correlation of Q as a function of melt composition according to the equation

$$\log (Q/X_{BeF_2}) = k + l(X_{LiF})^2 + m(X_{LiF})^4$$

k^o, l^o, m^o - temperature independent portions of k, l, and m, respectively

k', l', m' - temperature dependent portions of k, l, and m, respectively

n_{HF} and n_{H_2O} - moles of HF and H_2O as measured at T_w, P_w

P_{HF} and P_{H_2O} - partial pressures of HF and H_2O, respectively.

p_w - vapor pressure of water at T_w

P_w - total pressure at wet-test meter

P_1 - partial pressure of HF or H_2O leaving melt

P_2 - measured partial pressure of HF or H_2O

Q - x^2/y for BeO saturated melt

Q_A - xs/y

Q_b - $s^2/yr = (Q_A)^2/Q_O$

Q_B - $s^2/y = (Q_A)^2/Q$

Q_c - $s/xr = Q_A/Q_O$

Q_C - $s/x = Q_A/Q$

Q_O - x^2r/y

r - $[O^{2-}]$ - oxide concentration in moles per kilogram of melt

s - $[OH^-]$ - hydroxide concentration in moles per kilogram of melt

r_o and s_o - values of r and s at W = 0 in unsaturated experiments

S^o - standard entropy at specified temperature

ΔS_r - entropy of reaction

σ - standard deviation

t - degrees Centigrade (generally used to indicate melt temperatures)

T - degrees Kelvin (used to indicate wet-test meter temperatures)

T_w - temperature of wet-test meter, ^oK

$\mu^E_{BeF_2}$ - excess chemical potential of BeF_2

V_a - volume of dry carrier gas going through system measured at T_w, P_w

V_b - volume of gas entering titration assembly at T_w, P_w

V_m - volume of gas space above the melt

V_w - volume of gas through melt measured at T_w, P_w (liters)

dV - increment of gas flowing through system as measured at T_w, P_w

w - weight of melt (kg)

W - V/wRT_w (mole kg^{-1} atm^{-1})

X_{BeF_2} - mole fraction BeF_2

x - effluent partial pressure of HF

y - effluent partial pressure of H_2O

INTERNAL DISTRIBUTION

1. R. F. Apple
2-6. C. F. Baes, Jr.
7. J. T. Bell
8. C. M. Blood
9. F. F. Blankenship
10. G. E. Boyd
11. M. A. Bredig
12. R. B. Briggs
13. D. W. Cardwell
14. G. I. Cathers
15. E. L. Compere
16. J. L. Crowley
17. F. L. Culler
18. J. M. Dale
19. R. B. Evans III
20. D. E. Ferguson
21. R. A. Gilbert
22. W. R. Grimes
23. M. T. Harkrider
24. B. F. Hitch
25. S. S. Kirslis
26. K. A. Kraus
27. R. B. Lindauer
28. D. L. Manning
29-31. H. F. McDuffie
32. R. J. McNamee
33. R. L. Moore
34. K. A. Romberger
35. M. W. Rosenthal
36. H. C. Savage
37. D. Scott
38. D. R. Sears
39. J. H. Shaffer
40. S. H. Smiley
41. P. G. Smith
42. H. H. Stone
43. A. Taboada
44. R. E. Thoma
45. G. M. Watson
46. A. M. Weinberg
47. M. E. Whatley
48. J. C. White
49. B. J. Young
50. L. Brewer (consultant)
51. J. W. Cobble (consultant)
52. D. G. Hill (consultant)
53. H. Insley (consultant)
54. G. Mamantov (consultant)
55. T. N. McVay (consultant)
56. R. F. Newton (consultant)
57. J. E. Ricci (consultant)
58. G. Scatchard (consultant)
59. H. Steinfink (consultant)
60. T. F. Young (consultant)
61-62. Central Research Library
63. Document Reference Section
64-66. Laboratory Records Department
67. Laboratory Records, ORNL R.C.
68. ORNL Patent Office

EXTERNAL DISTRIBUTION

69. Research and Development Division, ORO
70-84. Division of Technical Information
85. F. A. Anderson, Chemical Engineering Dept., University of Miss., University, Mississippi
86. M. J. Blander, North American Aviation Science Center, 8437 Fallbrook Avenue, Canoga Park, California
87. A. Büchler, Arthur D. Little, Inc., 30 Memorial Drive, Cambridge 42, Massachusetts
88. S. F. Clark, University of Mississippi, University, Mississippi
89. P.A.D. deMaine, University of California, Santa Barbara, Calif.
90. G. Dirian, Commissariat a l'Energie Atomique (CEN Saclay) 69, Rue de Varenne, Paris, France

91. C. F. Dodson, Western Carolina College, Cullowhee, North Carolina.
92. F. R. Duke, Texas A and M College, College Station, Texas
93. J. E. Eorgan, Kawecki Chemical Co., Boyertown, Pennsylvania
94. H. Flood, Institute of Silicate Science, Norwegian Institute of Technology, Trondheim, Norway
95. Tormod Førland, The Institute of Theoretical Chemistry, Trondheim, Norway
96. R. M. Fuoss, Yale University, New Haven, Connecticut
97. M. A. Greenbaum, Rocket Power Inc., Research Laboratories, Pasadena, California
98. Marwin Kemp, University of Arkansas, Fayetteville, Arkansas
99. O. J. Kleppa, Chemistry Department, University of Chicago, Chicago, Illinois
100. S. Langer, General Atomics, P.O. Box 608, San Diego, California
101. G. Long, Chemistry Dept., UKAEA, Harwell, Didcot, Berks., England
102. C. R. Masson, National Research Council of Canada, Atlantic Regional Laboratory, Halifax, Nova Scotia
103-112. A. L. Mathews, Western Carolina College, Cullowhee, North Carolina
113. D. A. Mathewes, Western Carolina College, Cullowhee, North Carolina
114. J. L. Margrave, Rice University, Houston, Texas
115. N. J. Meyer, Department of Chemistry, Bowling Green State University, Bowling Green, Ohio
116. G. Nessle, Kawecki Chemical Co., Boyertown, Pennsylvania
117. S. Pizzini, Euratom, CCR, Ispra, Italy
118. P. A. Reid, Western Carolina College, Cullowhee, North Carolina
119. Rustum Roy, Materials Research Laboratory, The Pennsylvania State University, University Park, Pennsylvania
120. S. D. Squibb, Asheville-Biltmore College, Asheville, North Carolina
121. B. R. Sundheim, New York Univeristy, New York, New York
122. E. L. Topol, 19806 Gilmore Street, Woodland Hills, California
123. W. J. Wallace, Muskegon College, Muskegon, Michigan
124. J. Zarzycki, Compagnie de Saint-Gobain, 52 Bd De La Villette, Paris 19e, France