The Enthalpies of Formation of BeO(c) and BeF₂(c)

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Two of the key compounds in the evaluation and synthesis of a consistent set of thermodynamic values for the Be compounds are BeO(c) and BeF₂(c). The available measurements on the enthalpies of formation of these two compounds are presented with a detailed outline of the approach used to select the "best" values, \( \Delta H_f^0 \) [BeO(c)] = \(-145.7 \pm 0.6 \) kcal \( \cdot \) mol\(^{-1}\) \(-609.6 \pm 2.5 \) kJ \( \cdot \) mol\(^{-1}\) and \( \Delta H_f^0 \) [BeF₂(c, quartz)] = \(-245.4 \pm 0.8 \) kcal \( \cdot \) mol\(^{-1}\) \(-1026.8 \pm 3.3 \) kJ \( \cdot \) mol\(^{-1}\).

Key words: BeF₂(c); BeO(c); beryllium fluoride; beryllium oxide; enthalpies of formation; \( \Delta H_f^0 \) [BeF₂(c)]; \( \Delta H_f^0 \) [BeO(c)]; thermochemistry.

1. Introduction

Two of the key compounds in the evaluation and synthesis of a consistent set of thermodynamic values for the Be compounds are BeO(c) and BeF₂(c).

One approach used in the preparation of compilations of thermochemical data is to start with a compound for which \( \Delta H_f^0 \) (or \( \Delta G_f^0 \)) is definitive and independent of \( \Delta H_f^0 \) of any other compound of that element and preferably involves a minimum of auxiliary \( \Delta H_f^0 \)'s, and to build from the selected value for this compound. An example of this is the direct oxidation of the metal to the oxide, e.g., BeO(c), or the halogenation of the metal to the halide, e.g., BeF₂(amorp), or a set of reactions that can be combined in such a way that only one \( \Delta H_f^0 \) is unknown, e.g., Be(c) + 2HF(aq) \( \rightarrow \) H₂(g) + BeF₂(aq) and BeO(c) + 2HF(aq) \( \rightarrow \) BeF₂(aq) + H₂O(liq) so that by difference we can write the possible reaction, Be(c) + H₂O(liq) \( \rightarrow \) BeO(c) + H₂(g); similarly Be₂N₂(c) + 3/2O₂(g) \( \rightarrow \) 3BeO(c) + N₂(g) and 3Be(c) + N₂(g) \( \rightarrow \) Be₃N₂(c) giving Be(c) + 1/2O₂(g) \( \rightarrow \) BeO(c). We may then relate the \( \Delta H_f^0 \)'s of other compounds of that element to the selected compound by enthalpies of reaction.

If however every subsequent \( \Delta H_f^0 \) calculated is dependent upon the value selected for one compound, although we have internal consistency, we have no crosscheck as to how good the original value is. We should then have a second compound whose \( \Delta H_f^0 \) can also be obtained independently and an enthalpy of reaction relating the two to corroborate the choices and to close the cycle.

Until the recent measurements of Kilday, Prosen, and Wagman [1] on the enthalpies of solution of BeO(c) in aqueous HF solutions and the measurements of Churney and Armstrong [2] on the direct determination of the \( \Delta H_f^0 \) [BeF₂(amorph)], the data available on the direct enthalpies of formation of BeO(c) and BeF₂(c) and the data linking these values were discordant. These new investigations are a significant aid in establishing the values for BeO(c) and BeF₂(c) with more certainty. Our main efforts then, after considering the direct determinations, center upon the use of the solution measurements of BeO(c) in HF(aq) together with the solution measurements of Be(c) in HF(aq) which previously could not be fully utilized to obtain indirectly a definitive value for the \( \Delta H_f^0 \) [BeO(c)] and to relate that value to the determinations on the \( \Delta H_f^0 \) [BeF₂(c)]. Figure 1 schematically presents the reactions and paths discussed in this paper.

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1 Figures in brackets indicate the literature references at end of this paper.
All auxiliary data and constants used in the calculations are given in Wagman et al. [3]. Unless otherwise specified the values quoted are at, or have been corrected to, 298.15 K. Our final selections are reported in both kJ·mol⁻¹ and kcal·mol⁻¹. However, since this evaluation is included in Parker, Wagman, and Evans [3], where values are expressed in kcal·mol⁻¹, we report the individual values and their corrections in the same units in order to preserve the consistency of the relationships.

2. Discussion of Data on BeO(c)

2.1. Bomb Combustion

The following values of $\Delta H$ (kcal·mol⁻¹) for the oxidation of Be(c) have been reported: Moore and Parr [4], −134.4; Roth, Börger, and Siemonsen [5], −147.3; Neumann, Kröger, and Kunz [6], −145.3; Mielenz and v. Wartenberg [7], −136.2; and Cosgrove and Snyder [8], −143.1. Neumann, Kroger, and Kunz [6] measured the enthalpy of combustion of Be₃N₂ (crystal form unspecified) to form BeO(c) and N₂(g) as −300.6 kcal·mol⁻¹ of Be₃N₂(c). Neumann, Kröger, and Haeblcr [9] directly determined $\Delta H_{f}^{o}[\text{Be3N2(c)}] = -134.1$. By difference we obtain $\Delta H_{f}^{o}[\text{BeO(c)}] = -144.9$. Of these determinations the Cosgrove and Snyder measurement appeared to be the best value and had been generally accepted; however, the measurements of Kolesov, Popov, and Skuratov [10] on the enthalpies of reaction of BeO(c) in aqueous HF and BeF₂(c) in aqueous HF indicate that the value for $\Delta H_{f}^{o}[\text{BeO(c)}]$ should be more negative. This is in line with the fact that Cosgrove and Snyder did not determine the completeness of the reaction; incomplete combustion would cause the value, based on the weight of metal taken, to be too positive. We turn therefore to the indirect determinations of $\Delta H_{f}^{o}[\text{BeO(c)}]$.

2.2. The Enthalpies of Solution of Be(c) and BeO(c) in Aqueous HF Solutions

Matignon and Marchal [11] measured the enthalpies of solution of Be(c) and BeO(c) in 30 percent HF solutions, as have Copaux and Philips [12]. By difference, we obtain for Be(c) + H₂O(lq) → BeO(c) + H₂(g), $\Delta H = -70.9$ and $-62.1$ kcal, or $\Delta H_{f}^{o}[\text{BeO(c)}] = -139.2$ and $-130.4$ kcal·mol⁻¹, respectively. The individual $\Delta H$’s for solution of Be(c) in 30 percent HF are −94.2 from Matignon and Marchal and −82.2 kcal·mol⁻¹ from Copaux and Philips. For solution of BeO(c) they are −23.3 and −20.1 kcal·mol⁻¹, respectively. More recently, Bear and Turnbull [13] measured the enthalpy of solution of Be(c) in 12, 22.6, 30, and 40 percent HF solutions. The values are $-101.5$, $-101.0$, $-100.5$, and $-100.5$ kcal. Armstrong and Coyle [14] reported −99.6 kcal for solution in 25 percent HF. For solution of BeO(c) in aqueous HF we also have the results of Kilday et al., e.g., −24.2 kcal in a 30 percent HF solution, the results of Kolesov et al., in a 23 percent HF solution, −24.1 kcal, and Fricke and Wüllhorst [15] −24.3 kcal in 12 percent HF. It appears that the measurements of Matignon and Marchal and Copaux and Philips are not reliable; they give little information as to the experimental details and purity of materials. We cannot rely upon the values for $\Delta H_{f}^{o}[\text{BeO(c)}]$ obtained from the data of either Matignon and Marchal [11] or Copaux and Philips [12], but a judicious combination of the other measurements can yield a more reliable value.

One of the problems associated with combining reactions of Be(c) and BeO(c) in aqueous HF is that in most cases the final solutions are not the same. Not only are there no quantitative data on what Be species are in the final solutions or the percent of each, there are also no direct data on the $\Delta H_{dil}^{o}$ or $\Delta H_{mix}$ of these species in HF solutions. The measurements of Kilday et al., however, provide some insight into the effect of having solutions that do differ in both the amount and concentration of the excess HF(aq).

They have also measured the differential enthalpy of dilution in two of their final solutions,

$$H_2O(lq) + \text{[BeF}_2 \text{ in excess HF} \cdot n\text{H}_2\text{O}] \rightarrow \text{[BeF}_2 + \text{H}_2\text{O in excess HF} \cdot n\text{H}_2\text{O}].$$

These measurements are important since $H_2O(in \text{BeF}_2 + \text{HF} \cdot n\text{H}_2\text{O})$ is formed in the reaction of BeO(c) with HF(aq), i.e.,

$$\text{BeO(c)} + 2\text{HF(aq)} \rightarrow \text{[BeF}_2 + \text{H}_2\text{O}] \text{ in excess HF}.$$  

We have compared the experimental data with those calculated from the slope $d\varphi_1/dn^{1/2}$ of $\varphi_1$ HF at $n_f$ H₂O from the values tabulated by Parker [16]. These values are: at $n_f = 2.68$, $L_1 (\text{cal} \cdot \text{mol}^{-1}) = -178$ (calc.), and $-160 \pm 10$ (experimental); at $n_f = 3.57$, $L_1 = -110$ (calc.) and $-85 \pm 5$ (experimental). Since the agreement is good, we decided to ignore the presence of the BeF₂ in the final solutions and treat the solutions as if they were HF solutions. Since $X$ (the ratio of HF to BeF₂ in the final solution) ≥ 50 this is not an unreasonable approach.

We can now set up the equation for the reaction³ in the form,

$$\text{BeO(c)} + (X+2)(\text{HF} + nh_2\text{O}) \rightarrow \text{[BeF}_2 + \text{H}_2\text{O} + X(\text{HF} + n_f \text{H}_2\text{O})].$$

and use the experimental $L_1$ values and the $\varphi_1$ values at the appropriate concentrations from [16] to calculate $\Delta(H_f^{o})$ which represents $\Delta H_{f}^{o}[\text{BeO(c)}] - \Delta H_{f}^{o}[\text{BeF}_2(aq)]$. Table 1 shows the results as a function of $n_f$ where $n_f$ is the final ratio of H₂O to HF. It includes the mole of H₂O formed. All of their experimental $\Delta H$’s

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³$\varphi_1$ is the relative apparent molal enthalpy. It is the negative of the integral enthalpy of dilution per mole of solute at a concentration $n$ to an infinitely dilute solution. $L_1$ is the partial or differential enthalpy of dilution, per mole of solvent, when it is added to a large volume of solution at the given concentration.

⁴$n_f = n(X + 2)/X$; it excludes the mole of H₂O formed. $n_i$ is the initial molar ratio of H₂O to HF. $X$ is the mole ratio of HF to BeF₂ in solution.
The number in the first column corresponds to the entries from Kilday et al.’s data, we obtain -24.158 kcal·mol⁻¹. Their reaction corresponds to:

\[ \text{BeO}(c) + 342 \text{(HF + 3.801 H}_2\text{O)} \rightarrow \]

\[ \text{[BeF}_2 + \text{H}_2\text{O} + 340 \text{(HF + 3.823 H}_2\text{O)} \] \]

are included, corrected to 298.15 K where necessary, using the temperature coefficient given in their work. The number in the first column corresponds to the number of the Kilday et al., experiment.

The values for \( \Delta(\Delta H_f) \) within each group are in excellent agreement with one another, within the precision of the experimental data although there are some differences in \( X \) and \( n_f \) in the final solutions. The values appear to be primarily dependent upon the concentration of HF within the range \( 50 \leq X \leq 250 \). Figure 2 shows a plot of \( \Delta(\Delta H_f) \) as a function of \( n_f \). For the \( \Delta(\Delta H_f) \) we obtain a smooth curve. The smoothed values are also given in table 1. The variation of \( \Delta H \) as a function of the concentration of HF without regard to the variation of \( X \) may be expressed as:

\[ \Delta H = -24.092 - 0.113(450 - n_f) \text{ kcal·mol}^{-1} \text{ for } 7.1 > n_f > 2.6. \]

Kolesov, Popov, and Skuratov [10] also measured the enthalpy of solution of BeO(c) in aqueous HF. Their reaction corresponds to:

\[ \text{BeO}(c) + 342 \text{(HF + 3.801 H}_2\text{O)} \rightarrow \]

\[ \text{[BeF}_2 + \text{H}_2\text{O} + 340 \text{(HF + 3.823 H}_2\text{O)} \] \]

The \( \Delta H = -24.158 \text{ kcal·mol}^{-1} \) of BeO(c) results in \( \Delta(\Delta H_f) = 107.085 \text{ kcal·mol}^{-1} \) (using the same treatment as before). From our straight line plot of \( \Delta H \) from Kilday et al.’s data, we obtain -24.168 kcal·mol⁻¹ where the final solution contains BeF₂ in 90(HF+3.826 H₂O). From our plot of \( \Delta(\Delta H_f) \) versus \( n_f \) we obtain \( \Delta(\Delta H_f) = 107.185 \text{ kcal·mol}^{-1} \). The agreement in \( \Delta H \) is excellent, fortuitously so; in \( \Delta(\Delta H_f) \) it is not, but still within the experimental uncertainties. In addition \( X \) differs by 250 moles HF in the two solutions.

Kilday and Churney, private communication (1971), also made some measurements on the enthalpy of solution of BeF₂(amorph) in HF concentrations of 3.63 H₂O and 5.06 H₂O where \( X \) varied from 400 to 2700. From these measurements we obtain the following:

\[ \Delta H_{\text{soln}} = -8.430 + 0.50 X \text{ cal·mol}^{-1} \text{ for } n = 3.63 \]

\[ \Delta H_{\text{soln}} = -8.680 + 0.60 X \text{ cal·mol}^{-1} \text{ for } n = 5.06. \]

Although the precision of these measurements is not high they do enable us to obtain an approximate correction to \( \Delta(\Delta H_f) \) for the variation in \( X \). As a reference solution we have chosen this \( X \) to be 100 and have added -0.55(100 - \( X \)) cal·mol⁻¹ (an
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<th>$n_f$</th>
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<th>Mean $\Delta (\Delta Hf)$ from smooth curve, fig. 3</th>
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* Corrected for BeSO₄ impurity.
TABLE 3. \( \Delta H^\circ \)\[BeO(c)] in kcal \cdot mol^{-1} as calculated by various methods from data in table 2

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<th>Method 1</th>
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average value) to the \( \Delta (\Delta Hf) \)'s (see table 1, columns 10, 11, and 12 and fig. 3). The new \( \Delta (\Delta Hf) \)'s are in slightly better agreement with one another. For the \( \Delta (\Delta Hf) \)'s from Kolesov et al., we obtain 107.217 kcal \cdot mol^{-1} as compared to 107.180 from figure 3, in much better agreement; this lends support to the use of this treatment on the Bear and Turnbull measurements [13] on Be(c) in excess HF solutions, where the final ratio of HF to BeF₂ is greater than 340.

Bear and Turnbull used solutions of 12, 22.6, 30, and 40 percent HF. Their results can be treated three ways:

1. Using their \( \Delta H_f \) with the \( \Delta H_f \) from their straight line plot of the Kilday et al., data on BeO in HF at the appropriate \( n_f \), with the experimental \( L_1 \), obtaining the reaction, Be(c) + \( H_2O(aq) \rightarrow BeO(c) + H_2(g); \)
2. calculating the \( \Delta H^\circ[BeF_2] \) in \( X(HF + n_fH_2O) \) and using the appropriate \( \Delta (\Delta Hf) \) [BeO - BeF₂(aq)]\ at \( n_f \) from figure 2;
3. correcting \( \Delta H_f[BeF_2(aq)] \) to \( X = 100 \) and using the comparable corrected \( \Delta (\Delta Hf) \) from figure 3.

The results are summarized in table 2 and 3. It is obvious that the values for \( \Delta H^\circ[BeO(c)] \) are in better agreement using the third way of calculating. Figure 3 shows a plot of \( \Delta H_f[BeF_2(aq)] \) in 100(HF + \( n_fH_2O) \) and also the uncorrected values derived from the Bear and Turnbull measurements.

For confirmation of the above values we may use the enthalpy of solution of the samples of BeF₂(amorph) cited earlier whose \( \Delta H^\circ \) is known (\( -244.3 \text{ kcal \cdot mol}^{-1} \), directly determined by Churney and Armstrong [2]) from which we obtain \( \Delta H_f[BeF_2(aq)] \) in 100(HF + \( 3.63H_2O) = -252.68 \text{ kcal \cdot mol}^{-1} \) and \( \Delta H_f[BeF_2(aq)] \) in 100(HF + 5.06 \( H_2O) = -252.92 \text{ kcal \cdot mol}^{-1} \) (cf. \( -252.75 \) and \( -253.38 \text{ kcal \cdot mol}^{-1} \) from the smoothed curve (fig. 3) of Bear and Turnbull's corrected \( \Delta H_f \)'s). Using \( \Delta (\Delta Hf) = 107.11 \) and \( \Delta (\Delta Hf) = 107.57 \text{ kcal \cdot mol}^{-1} \) from figure 3, we obtain \( \Delta H^\circ[BeO(c)] = -145.57 \) and \( -145.35 \text{ kcal \cdot mol}^{-1} \), respectively.

In all the above cases we have used the \( \Delta (\Delta Hf) \) [BeO(c) - BeF₂(aq)] derived from the Kilday et al. measurements. If we use \( \Delta (\Delta Hf) = 107.217 \text{ kcal \cdot mol}^{-1} \) from the Kolesov et al. measurements and \( \Delta H_f[BeF_2(aq)] = -252.93 \text{ kcal \cdot mol}^{-1} \) from figure 3 we obtain \( \Delta H^\circ[BeO(c)] = -145.71 \text{ kcal \cdot mol}^{-1} \).

There is another reaction involving the enthalpy of solution of Be(c) in HFaq), that by Armstrong and Coyle [14]. They reported that there was an unexplained deficiency of \( H_2 \) on the order of 0.5 percent. They ascribe an uncertainty of not more than 0.2 percent to the enthalpy of reaction for this contribution. The measurements are on the reaction: Be(c) + 6.470(HF + 3.309H₂O) \( \rightarrow \) [BeF₂ + 4.470(HF + 4.789H₂O)] + H₂(g) for which \( \Delta H = -99.64 \text{ kcal \cdot mol}^{-1} \) and \( \Delta H/[BeF_2(aq)] = -251.1 \text{ kcal \cdot mol}^{-1} \). Using the same three approaches as for Bear and Turnbull's data we obtain \( \Delta H^\circ[BeO(c)] = -143.9 \text{ kcal \cdot mol}^{-1} \); \( -143.6 \text{ kcal \cdot mol}^{-1} \); and \( -143.5 \text{ kcal \cdot mol}^{-1} \) (3), where \( X \) is corrected to 100. In all cases we assume that the species in the two solutions are the same; in the third case we also assume our correction is applicable to \( X = 4.47 \), which is probably not justified.

There are two other approaches we can use on their data:

4. Herefore we have neglected the effect of the BeF₂ on the HF dilution correction. If we assume that the BeF₂ present in the solution may be replaced by an equivalent number of moles of HF solution, then we may consider the HF to be in \( 3.913H_2O \) and using the same three approaches as for the initial solution, [HF + 3.309H₂O], to their final solution, using the \( \phi_i \) values of HF, and obtain:

\[
[BeF_2(aq) + 4.470(HF + 4.789H_2O)] \\
+ 95(HF + 3.309H_2O) \rightarrow [BeF_2(aq) + 99.470(HF + 3.375H_2O)] \]

\( \Delta H_{mix} = -0.264 \text{ kcal \cdot mol}^{-1} \). Combining this with their reported \( \Delta H \) leads to:

\[
\text{Be(c) + 101.470(HF + 3.309H_2O) \rightarrow [BeF_2(aq) + 99.470(HF + 3.375H_2O)] + H_2(g); } \\
\Delta H = -99.95 \text{ kcal \cdot mol}^{-1}.
\]

This value is now in closer agreement with Bear and
Turnbull’s measurements. For the same reaction with BeO(c) we obtain \( \Delta H = -24.22 \text{ kcal} \cdot \text{mol}^{-1} \), so that \( \Delta H^\circ \text{[BeO(c)]} = -144.1 \text{ kcal} \cdot \text{mol}^{-1} \). A variation of this would be to use a \( \Delta H_{\text{mix}} \) based on our fourth approach, i.e., to assume the HF is in 3.913 H2O, then \( \Delta H = -100.23 \) and \( \Delta H^\circ \text{[BeO(c)]} = -144.4 \text{ kcal} \cdot \text{mol}^{-1} \).

### 2.3. The Enthalpies of Solution of Be(c) and BeO(c) in Aqueous HCl Solutions

The problem of nonidentical final solutions is also present in the Be(c)–BeO(c)–HCl aqueous systems. Kilday et al. [1] have obtained \( \Delta H = -12.8 \) and -12.7 kcal \cdot mol^{-1} for the solution of BeO(c) in 18 and 22.6 percent HCl, respectively. The complete reactions are:

\[
\text{BeO(c)} + 40.9\text{(HCl + 9.21H}_2\text{O)} \rightarrow \text{[BeCl}_2\text{(aq)}] \\
\quad + \text{H}_2\text{O(aq)} + 38.9\text{(HCl + 9.69H}_2\text{O)}
\]

and

\[
\text{BeO(c)} + 47.8\text{(HCl + 6.93H}_2\text{O)} \rightarrow \text{[BeCl}_2\text{(aq)}] \\
\quad + \text{H}_2\text{O(aq)} + 45.8\text{(HCl + 7.23H}_2\text{O)}.
\]

(The experimental \( \tilde{L} \) value = -272 cal \cdot mol^{-1} of H2O (see ref. [1], sec. 5.4) checks well with \( \tilde{L} \) calculated from the \( \phi_t \) values of HCl [16] at \( n = 7.23\text{H}_2\text{O} \).

From these reactions we obtain for BeCl2(aq) in 38.3(HCl + 9.72H2O) \( \Delta H^\circ \text{[BeO(c) - BeCl}_2\text{(aq)]} = 18.6 \text{ kcal} \cdot \text{mol}^{-1} \) and 16.6 kcal \cdot mol^{-1} for BeCl2(aq) in 45.8(HCl + 7.25H2O).

Thompson, Sinke, and Stull [17] reported \( \Delta H = -89.61 \text{ kcal} \cdot \text{mol}^{-1} \) for Be(c) + 8.38(HCl + 8.11H2O) \rightarrow \text{[BeCl}_2\text{(aq)}] + 6.38\text{(HCl + 10.65H}_2\text{O)} + \text{H}_2\text{(g)} \), from which we obtain \( \Delta H^\circ \text{[BeCl}_2\text{(aq)]} = -163.79 \text{ kcal} \cdot \text{mol}^{-1} \). Blachnik, Gross, and Hayman [18] reported \( \Delta H_{388K} = -90.00 \text{ kcal/mol for:} \)

\[
\text{Be(c)} + 8.86\text{(HCl + 7.99H}_2\text{O)} \rightarrow \text{H}_2\text{(g)} + \text{[BeCl}_2\text{(aq)}] \\
\quad + 6.86\text{(HCl + 10.32H}_2\text{O)}.
\]

This value, corrected to 298.15 K is in good agreement with the Thompson et al. value.

Using the same approaches as for HF we have:

(1) With \( \Delta H \) of solution of BeO(c) = -12.8 kcal \cdot mol^{-1} when \( n_f = 10.65 \), \( \Delta H^\circ \text{[BeO(c)]} = -145.2 \text{ kcal} \cdot \text{mol}^{-1} \).

(2) From an extrapolation of \( \Delta \Delta H^\circ \text{[BeO(c) - BeCl}_2\text{(aq)]} \), we obtain \( \Delta \Delta H^\circ = 19.4 \text{ kcal} \cdot \text{mol}^{-1} \) where the BeCl2 is in 34(HCl + 10.65H2O) and \( \Delta H^\circ \text{[BeO(c) - BeCl}_2\text{(aq)]} = -144.4 \text{ kcal} \cdot \text{mol}^{-1} \).

(3) Not used since we have no correction for the variation in X.

As in our treatment of the Armstrong and Coyle [14] results we may try the following:

(4) If we assume that the BeCl2 present may be replaced by an equivalent number of moles of HCl solution we may consider the HCl to be in 9.21H2O. Then \( \Delta H^\circ \text{[BeCl}_2\text{(aq)]} = -165.0 \) and \( \Delta H^\circ \text{[BeO(c)]} = -145.6 \text{ kcal} \cdot \text{mol}^{-1} \).

(5) We can calculate a \( \Delta H_{\text{mix}} \) by adding more of the initial HCl solution to the final solution. We obtain:

\[
\begin{align*}
\text{[BeCl}_2\text{(aq)] + 6.38\text{(HCl + 10.65H}_2\text{O)} & + 35.54\text{(HCl + 8.11H}_2\text{O)} \\
& \rightarrow \text{[BeCl}_2\text{(aq)] + 41.92\text{(HCl + 8.499H}_2\text{O)}; } \Delta H = -0.64 \text{ kcal} \cdot \text{mol}^{-1} \\
& \text{and } \Delta H = -90.3 \text{ kcal} \cdot \text{mol}^{-1} \text{ for:}
\end{align*}
\]

\[
\begin{align*}
\text{Be(c)} + 43.92\text{(HCl + 8.11H}_2\text{O)} & \rightarrow \text{H}_2\text{(g)} \\
& + \text{[BeCl}_2\text{(aq)] + 41.92\text{(HCl + 8.499H}_2\text{O)}).
\end{align*}
\]

For the reaction with BeO(c) we can say \( \Delta H = -12.7 \text{ kcal} \cdot \text{mol}^{-1} \) for Be(c) + 43.92(HCl + 8.11H2O) \rightarrow \text{[BeCl}_2\text{(aq)]} + 41.92\text{(HCl + 8.499H}_2\text{O)} \), so that \( \Delta H = -77.56 \text{ kcal} \cdot \text{mol}^{-1} \) for Be(c) + H2O(aq) \rightarrow BeO(c) + H2(g) and \( \Delta H^\circ \text{[BeO(c)]} = -146.0 \text{ kcal} \cdot \text{mol}^{-1} \). A variation of this would be to calculate a \( \Delta H_{\text{mix}} = -1.81 \text{ kcal} \cdot \text{mol}^{-1} \) on the basis of (4). Then \( \Delta H^\circ \) for the reaction with Be(c) = -91.4 and \( \Delta H^\circ \text{[BeO(c)]} = -147.2 \text{ kcal} \cdot \text{mol}^{-1} \).

Averaging these five values we obtain -145.7 kcal \cdot mol^{-1} for \( \Delta H^\circ \text{[BeO(c)]} \).

### 2.4. Other data

From the cell measurements of Smirnov and Chukreev [19] in the temperature range 955 to 1313 K, we obtain a second law \( \Delta H^\circ = -94.6 \text{ kcal} \cdot \text{mol}^{-1} \) and a third law \( \Delta H^\circ = -93.7 \text{ kcal} \cdot \text{mol}^{-1} \) for Be(c) + 1/2CO2(g) \rightarrow BeO(c) + 1/2C(graphite), or \( \Delta H^\circ \text{[BeO(c)]} = -141.6 \) and \( -140.8 \text{ kcal} \cdot \text{mol}^{-1} \), respectively.

### 2.5. The Selection of the \( \Delta H^\circ \text{[BeO(c)]} \)

It is appropriate at this point to tabulate the values of \( \Delta H^\circ \text{[BeO(c)]} \), both the direct and indirect determinations.

<table>
<thead>
<tr>
<th>Investigator</th>
<th>( \Delta H^\circ \text{[BeO(c)]} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Moose and Parr [4]</td>
<td>-134.1</td>
</tr>
<tr>
<td>Roth et al. [5]</td>
<td>-147.3</td>
</tr>
<tr>
<td>Neumann et al. [6]</td>
<td>-145.3</td>
</tr>
<tr>
<td>Cosgrove and Snyder [8]</td>
<td>-143.1</td>
</tr>
<tr>
<td>Neumann et al. [6, 9]</td>
<td>-144.9</td>
</tr>
<tr>
<td>Smirnov and Chukreev [19]</td>
<td>-141.1</td>
</tr>
<tr>
<td>Kilday et al. [1] and Bear and Turnbull [13]</td>
<td>-145.6</td>
</tr>
<tr>
<td>Kilday et al. [2]</td>
<td>-146.20</td>
</tr>
<tr>
<td>Kilday et al. [1] and Bear and Turnbull [13]</td>
<td>-145.65</td>
</tr>
<tr>
<td>Kilday et al. [1] and Armstrong and Coyle [14]</td>
<td>-143.9</td>
</tr>
<tr>
<td>Kilday et al. [1] and Thompson et al. [17]</td>
<td>-145.7</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Investigator</th>
<th>( \Delta H^\circ \text{[BeO(c)]} )</th>
</tr>
</thead>
</table>
tions (table 4). As is evident, especially for the Smirnov and Chukreev value, all the indirect determinations support a more negative value than that of Cosgrove and Snyder. The “best” value now appears to be −145.7 kcal mol⁻¹ in good agreement with the Neumann et al. [6] direct determination. The uncertainties are discussed in section 4.

2.6. The Decomposition of Beryllium Hydroxide as Supporting Evidence

Bear and Turnbull [13] have also measured the enthalpy of solution of Be(OH)₂ (β, orthorhombic) and Be(OH)₂ (α, tetragonal) in 22.6 percent HF, [679(HF +3.80H₂O)]. From these measurements and their measurements on Be(c) we obtain ΔH = −79.83 kcal mol⁻¹ for Be(c) + 2H₂O(liq) → Be(OH)₂(β) + H₂O(g) and ΔH° = −216.5 kcal mol⁻¹; similarly for Be(OH)₂(α) we obtain ΔH = −79.10 kcal mol⁻¹ and ΔH° = −215.7 kcal mol⁻¹. Using these values for ΔH° and our tentative “best” value for Be(c) we obtain:

BeO(c) + H₂O(liq) → Be(OH)₂(c, β); ΔH° = −2.5 kcal mol⁻¹
BeO(c) + H₂O(liq) → Be(OH)₂(c, α); ΔH° = −1.7 kcal mol⁻¹.

Fricke and Wüllhorst [15] measured the enthalpies of solution of BeO(c), Be(OH)₂(c, β), and Be(OH)₂(c, α) in 11.59 percent HF. From these measurements we obtain ΔH° = −2.5 and −1.8 kcal mol⁻¹, respectively, in excellent agreement with our ΔH°. Matignon and Marchal [20, 21], from measurements in 30 percent HF, obtained ΔH°₅₀ = −3.2 kcal mol⁻¹, in fair agreement. Fricke and Severin [22] measured the equilibrium vapor pressure at 378 K to 100 mm H₂O(g) over Be(OH)₂(c, β). Using a Nernst equation they calculate ΔH = 15.5 kcal mol⁻¹ of H₂O(g), which results in ΔH° = −5.0 kcal mol⁻¹ for the hydration of BeO(c). However they reported that the BeO formed had a distorted lattice which should require a ΔH° more positive than −145.7 kcal mol⁻¹. Also since the ΔH calculated is based on only one point it can not be considered a definitive value.

It is evident that the related data are supportive of our value for ΔH°₅₀[Be(c)] = −145.7 kcal mol⁻¹.

3. The Enthalpy of Formation of BeF₂(c)

3.1. The Enthalpy of Transition of BeF₂(g) and BeF₂(amorph) to BeF₂ (α, quartz)

There are no published direct determinations of the ΔH°₅₀[BeF₂(c)] from combustion of Be(c) in F₂(g); the combustion of Be(c) in F₂(g) results in amorphous material. The indirect reactions, in which BeF₂ forms, produce a glassy state.

Taylor and Gardner [23] determined the enthalpy of solution of both the α, quartz form, and the glassy form in acetic acid-sodium acetate solutions to be −3.64 and −4.76 kcal mol⁻¹, respectively. This leads to a ΔH°₅₀[BeF₂(α, quartz)] = −257.0 kcal mol⁻¹.

3.2. The Reaction of Be(c) With F₂(g)

Churney and Armstrong [2] measured the enthalpy of reaction of Be(c) in F₂(g) to be −244.3 kcal mol⁻¹. They report BeF₂(amorph) to be their product. This results in ΔH°₅₀[BeF₂(c)] = −245.4 kcal mol⁻¹. In a preliminary report (1965) they cite the unpublished measurements of Simmons (1961) on the conversion of BeF₂ to partially glassy BeF₂, leading to ΔH°₅₀[BeF₂(c)] = −257.0 kcal mol⁻¹.

3.3. The Reaction of Be(c) With PbF₂(c)

Gross [24] reported ΔH° = −84.0 kcal for the reaction of Be(c) with PbF₂(c) to form BeF₂(c) and Pb(c). Although no crystallographic identification was made the direction of the results under varying conditions indicates that the value is for the formation of BeF₂(c). Since there is some uncertainty in our selection for PbF₂(c) we will avoid its use by relating the reaction to the reaction from Gross, Hayman, and Levi [25]:

3/2PbF₂(c) + Al(c) → AlF₃(c) + 3/2Pb(c); ΔH = −118.53 kcal from which we obtain:
3/2Be(c) + AlF₃(c) → Al(c) + 3/2BeF₂(c); ΔH = −7.47 kcal and using ΔH°₅₀[AlF₃(c)] = −359.5 kcal mol⁻¹ [3]. However, a more recent direct determination of ΔH°₅₀[AlF₃(c)] = −361.0 kcal mol⁻¹ by Rudzitis et al. [26] would lead to ΔH°₅₀[BeF₂(c)] = −246.5 kcal mol⁻¹.

3.4. Other Data

There is a path to Δ(H°₅₀)[BeO(c) − BeF₂(c)] and another to Δ(H°₅₀)[BeF₂(c)] both of which involve BeF₂(g) in high temperature gas phase equilibria. These can be referred to BeF₂(c) with the enthalpy of sublimation of BeF₂. Table 5 summarizes the second

<table>
<thead>
<tr>
<th>Investigator</th>
<th>Temp. Range of Measurements</th>
<th>Second Law ΔH°</th>
<th>Third Law ΔH°</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cantor [28]</td>
<td>1146-1372</td>
<td>kcal mol⁻¹</td>
<td>kcal mol⁻¹</td>
</tr>
<tr>
<td>Khandamirova et al. [29]</td>
<td>846-950</td>
<td>58.7</td>
<td>56.07</td>
</tr>
<tr>
<td>Sense and Stone [30]</td>
<td>1075-1293</td>
<td>55.37</td>
<td>55.65</td>
</tr>
<tr>
<td>Sense, Snyder, and Clegg [31]</td>
<td>1019-1076</td>
<td>60.8</td>
<td></td>
</tr>
<tr>
<td>Hildenbrand and Theard [32]</td>
<td>1076-1241</td>
<td>55.9</td>
<td></td>
</tr>
<tr>
<td>Blauer et al. [33]</td>
<td>821-942</td>
<td>55.98</td>
<td>55.40</td>
</tr>
<tr>
<td>Greenbaum et al. [34]</td>
<td>713-795</td>
<td>58.1</td>
<td>53.70</td>
</tr>
<tr>
<td>Novoselova et al. [35]</td>
<td>823-1053</td>
<td>56.66</td>
<td>54.86</td>
</tr>
<tr>
<td>Taylor and Gardner [23]</td>
<td>1040-1376</td>
<td>52.3</td>
<td>55.8</td>
</tr>
</tbody>
</table>
and third law $\Delta H^\circ_{\text{subl}}$ BeF$_2$ calculated from the vapor pressure measurements on the crystal and liquid. From these data we have chosen $\Delta H^\circ_{\text{subl}}$ = 55.7 kcal · mol$^{-1}$. Because the thermal functions of BeF$_2$(g) are based on an estimate for one of the three vibrational frequencies and the thermal functions of the condensed phases at high temperature are only approximate, we estimate an uncertainty of $\pm 2.0$ kcal · mol$^{-1}$. The gas phase equilibria calculations are summarized in table 6. Because of the many uncertainties inherent in these data, we have not assigned any weight to these measurements. The thermal functions used are given in [27].

Table 6. Gas phase equilibria involving BeF$_2$(g)

<table>
<thead>
<tr>
<th>Reference</th>
<th>Reaction</th>
<th>$\Delta(\Delta H^\circ)$[BeO(c) - BeF$_2$(c)]</th>
<th>$\Delta H^\circ$[BeF$_2$(c)]</th>
</tr>
</thead>
<tbody>
<tr>
<td>2d Law</td>
<td>BeO(c) + 2HF(g) $\rightarrow$ BeF$_2$(g) + H$_2$O(g)</td>
<td>kcal · mol$^{-1}$</td>
<td>105</td>
</tr>
<tr>
<td>3d Law</td>
<td>BeO(c) + 2HF(g) $\rightarrow$ BeF$_2$(g) + H$_2$O(g)</td>
<td>kcal · mol$^{-1}$</td>
<td>101</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>-242</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>-238</td>
</tr>
</tbody>
</table>

3.5. The Selection of $\Delta H^\circ$[BeF$_2$(c)] and $\Delta(\Delta H^\circ)$[BeO(c) - BeF$_2$(c)]

From the values for $\Delta H^\circ$[BeF$_2$(c)] in 3.2 and 3.3 our "best" value for BeF$_2$(c, quartz) appears to be $-245.4$ kcal · mol$^{-1}$. Tentatively, then our $\Delta(\Delta H^\circ)$[BeO(c) - BeF$_2$(c)] is 99.7 kcal · mol$^{-1}$.

As cited earlier the data from Kolesov et al. [9] yielded $\Delta(\Delta H^\circ)$[BeO(c) - BeF$_2$(aq)] = 107.085 kcal · mol$^{-1}$, where the BeF$_2$(aq) is in 340HF + 3.826H$_2$O. They also measured the enthalpy of solution of BeF$_2$(c, $\beta$-cristobalite) to the same final solution, $\Delta H = -8.07$ kcal · mol$^{-1}$. If we combine these results with our selected value for BeF$_2$(c), $\Delta H^\circ = -145.7$ kcal · mol$^{-1}$ we obtain for BeF$_2$(c, $\beta$-cristobalite) $-244.7$ kcal · mol$^{-1}$. This indicates an enthalpy of transition of 0.7 kcal · mol$^{-1}$ between the two forms. Reported values for similar transitions in SiO$_2$ [3], CaF$_2$ [37], and BeCl$_2$ [38] are 0.37, 1.14 and 1.32 kcal · mol$^{-1}$, respectively.

4. Assigned Uncertainties

We have tried to indicate some measure of the uncertainty in the reported values of $\Delta H$ and in the derived $\Delta H^\circ$'s by the number of significant figures given, following the convention that the overall uncertainty lies between 2 and 20 units of the last figure. The uncertainty in the $\Delta H^\circ$'s depends on the uncertainties of all the determinations in the total chain of reactions used to establish the value. But the values are also given so that the experimental data from which they are derived may be recovered with an accuracy equal to that of the original experimental quantities.

The overall uncertainties in the $\Delta H^\circ$'s are based on many factors—the experimental technique used, the details given, the number of measurements, the standard deviation of the reported results, the magnitude and reliability of the corrections to 298.15 K, and the reliability of previous work of the investigators. A strictly mathematical evaluation can therefore not be made. For this reason we shall consider only the discussion of the assignment of uncertainties to our "best" values for $\Delta H^\circ$[BeO(c)] and $\Delta H^\circ$[BeF$_2$(c)].

Kilday et al. reported the uncertainty in their measurements of $\Delta H$ of solution of BeO(c) in aqueous 24 percent HF to be $\pm 0.05$ kcal · mol$^{-1}$. Kolesov et al. state their uncertainty to be $\pm 0.12$ kcal · mol$^{-1}$. Bear and Turnbull state the uncertainties in their measurements to be $\pm 0.6$, $\pm 0.3$, $\pm 0.6$, and $\pm 0.9$ kcal · mol$^{-1}$ for the solution of Be(c) in 12, 22.6, 30, and 40 percent HF solutions, respectively. In calculating the $\Delta(\Delta H^\circ)$[BeO(c) - BeF$_2$(aq)] and $\Delta H^\circ$[BeF$_2$(aq)] we introduced errors due to the uncertainties in our values for $\Delta H^\circ$ F$^-$(aq) and $\varphi_1$ HF, but these errors essentially cancel in obtaining $\Delta H^\circ$[BeO(c)]. We introduce a $\pm 0.05$ kcal · mol$^{-1}$ uncertainty by our correction of $\Delta H^\circ$[BeF$_2$(aq)] to $X = 100$. This is negligible. The overall uncertainty obtained from the combined results of Bear and Turnbull and Kilday et al. is $\pm 0.6$ kcal · mol$^{-1}$; similarly from the results of Kolesov et al. and Bear and Turnbull. The $\Delta H^\circ$[BeO(c)] derived from the combination of the results of Kilday et al., and Churney and Armstrong, is dependent upon $\Delta H^\circ$[F$^-$(aq)]; hence the overall uncertainty must be $\pm 1.2$ kcal · mol$^{-1}$.

The uncertainty in the Armstrong and Coyle measurement is $\pm 0.2$ kcal · mol$^{-1}$. However, the final solution here is not dilute with respect to the BeF$_2$(aq). Even with the estimated mixing correction to the measured $\Delta H$, the uncertainty in the derived $\Delta H^\circ$[BeO(c)] must be $\pm 1.5$ kcal · mol$^{-1}$. Similarly, although the uncertainty in the measurements of Thompson et al., on Be(c) in HCl is $\pm 0.1$ kcal · mol$^{-1}$ (for BeO in HCl from Kilday et al. it is $\pm 0.5$ kcal · mol$^{-1}$) the overall uncertainty in the derived $\Delta H^\circ$[BeO(c)] is $\pm 1.5$ kcal · mol$^{-1}$. For our "best" value for $\Delta H^\circ$[BeO(c)] we assign an uncertainty of $\pm 0.6$ kcal · mol$^{-1}$, Churney and Armstrong assigned $\pm 0.8$ kcal · mol$^{-1}$ to their value for $\Delta H^\circ$[BeF$_2$(amorph)]. The indirect determination from the measurements of Gross et al. have an overall uncertainty of $\pm 0.8$ kcal · mol$^{-1}$. We assign an overall uncertainty of $\pm 0.8$ kcal · mol$^{-1}$ to our "best" value for $\Delta H^\circ$[BeF$_2$(c)].

5. A Key Assumption

The interpretation of the data and the values given are internally consistent with our value for $\Delta H^\circ$[HF(aq, std. state)] [3], and lend support to this value; eg., the values for $\Delta(\Delta H^\circ)$[BeO(c) - BeF$_2$(quartz)] from our 'selected' values, independent of HF(aq) are in excellent agreement with that derived from the Kolesov et al. difference, HF dependent, if one assumes a $\Delta H^\circ$[trans] of 0.7 kcal · mol$^{-1}$ for the $\beta$ cristobalite to the quartz form. Also the $\Delta H^\circ$[BeF$_2$(aq, in HF)] from the Bear and Turnbull measurements, dependent upon HF...
are in excellent agreement with that derived from the $\Delta H_{\text{soln}}$ (measured by Kilday and Churney) of a BeF$_2$ (amorph) sample whose $\Delta H_f^\circ$ was measured directly by Churney and Armstrong and is thus independent of $\Delta H_f^\circ$[HF(aq)]. However there is also evidence [39, 40, 41] that the ‘selected’ value for HF may be too positive by 0.3 to 0.4 kcal·mol$^{-1}$. If so, this would involve a reinterpretation of the data.

In summary:

$$\Delta H_f^\circ[\text{BeO(c)}] = -145.7 \pm 0.6 \text{ kcal} \cdot \text{mol}^{-1}$$

$$(-609.6 \pm 2.5 \text{ kJ} \cdot \text{mol}^{-1})$$

$$\Delta H_f^\circ[\text{BeF}_2(c, \text{quartz})] = -245.4 \pm 0.8 \text{ kcal} \cdot \text{mol}^{-1}$$

$$(-1026.8 \pm 3.3 \text{ kJ} \cdot \text{mol}^{-1}).$$

6. References


