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Hydrothermal Synthesis and Thermodynamic Property of the Zeolite-Like Galloborate of $K_2[Ga(B_5O_{10})]\cdot 4H_2O$

Sa-Ying Li and Zhi-Hong Liu*

Key Laboratory for Macromolecular Science of Shaanxi Province, School of Chemistry and Materials Science, Shaanxi Normal University, Xi'an 710062, P.R. China

ABSTRACT: The pure zeolite-like galloborate of the $K_2[Ga(B_5O_{10})]\cdot 4H_2O$ sample has been synthesized by a mild hydrothermal method and characterized by means of chemical analysis, XRD, FT-IR, and DTA-TG techniques. The molar enthalpies of solution of $KGa(SO_4)_2\cdot 12H_2O(s)$ in the mixture of 1 $mol\cdot kg^{-1}$ HCl and calculated amount of $K_2B_4O_7\cdot 4H_2O$, of $K_2SO_4(s)$ in the mixture of 1 $mol\cdot kg^{-1}$ HCl and calculated amount of H_3BO_3, of $K_2[Ga(B_5O_{10})]\cdot 4H_2O(s)$ in the mixture of 1 $mol\cdot kg^{-1}$ HCl and calculated amount of K_2SO_4 and H_3BO_3 at 298.15 K were measured, respectively. With the incorporation of the previously determined enthalpies of solution of $H_3BO_3(s)$ and $K_2B_4O_7\cdot 4H_2O(s)$ in 1 $mol\cdot kg^{-1}$ HCl, together with the use of the standard molar enthalpies of formation for $K_2B_4O_7\cdot 4H_2O(s)$, $KGa(SO_4)_2\cdot 12H_2O(s)$, $K_2SO_4(s)$, $H_3BO_3(s)$, and $H_2O(l)$, the standard molar enthalpy of formation of $-(5768.5 \pm 9.1)$ $kJ\cdot mol^{-1}$ for $K_2[Ga(B_5O_{10})]\cdot 4H_2O(s)$ was obtained on the basis of the appropriate thermochemical cycle. In addition, the standard molar enthalpy of formation of $-(5757.6 \pm 0.8)$ $kJ\cdot mol^{-1}$ for $KGa(SO_4)_2\cdot 12H_2O(s)$ was also obtained according to its molar enthalpy of solution of (45.15 ± 0.11) $kJ\cdot mol^{-1}$ in H_2O at 298.15 K.

INTRODUCTION

Microporous materials have attracted considerable attention due to their widespread applications in catalysis, ion exchange, and adsorption. The discovery of aluminophosphate analogues of zeolites[1] has greatly stimulated the interest in designing other novel inorganic microporous materials with different topological structures. Recently, we have reported the mild solvothermal synthesis and crystal structure of a zeolite-like galloborate of $K_2[Ga(B_5O_{10})]\cdot 4H_2O$.[2]

Thermodynamic properties play very important roles in scientific research and industrial applications. Thermochemical data can provide information on stabilities and reactivities of molecules that are used and also are a key factor in the safe and successful scale-up of chemical processes in the chemical industry. Navrotsky's group has done much work on the thermochemistry of microporous compounds such as zeolites, pure silica, gallosilicate, and aluminophosphates by using high-temperature calorimetry.[3,4] As for the thermochemistry of main group borates, the standard molar enthalpies of formation of many alkaline and alkaline-earth metal borates have been reported, such as several potassium borates.[5−7] However, studies of the thermochemistry of the zeolite-like galloborates are not reported in the literature. This paper reports a new synthesis method and the determination of standard molar enthalpy of formation of $K_2[Ga(B_5O_{10})]\cdot 4H_2O$, by using a heat conduction microcalorimeter.

EXPERIMENTAL SECTION

Hydrothermal Synthesis and Characterization of the $K_2[Ga(B_5O_{10})]\cdot 4H_2O$ Sample. All reagents used in the synthesis were of analytical grade. A mixture of 0.314 g of Ga_2O_3, 2.012 g of B_2O_3, 1.522 g of KOH, 0.03 g of NH_4F, and 4.0 cm^3 of H_2O was sealed in a Teflon-lined stainless steel vessel and heated at 433 K for 7 days, then cooled to room temperature. The resulting crystals were filtered, washed with absolute alcohol and absolute ether, and finally dried at room temperature to constant mass. The synthetic sample was characterized by X-ray powder diffraction (Rigaku D/MAX-IIIC X-ray diffractometer with Cu target at $8°\cdot min^{-1}$), FT-IR spectroscopy (Nicolet NEXUS 670 FT-IR spectrometer by using KBr pellets at room temperature), and TG-DTA (TA-SDT Q600 simultaneous thermal analyzer under N_2 atmosphere with a heating rate of 10 $K\cdot min^{-1}$). The chemical compositions of the sample were determined by NaOH standard solution in the presence of mannitol for B_2O_3 and by the mass loss in the TG curve for H_2O.

Thermochemical Cycles. $K_2[Ga(B_5O_{10})]\cdot 4H_2O$ can be regarded as the product of the reaction (7) in the designed thermochemical cycle (Figure 1).

The approximately 1 $mol\cdot kg^{-1}$ HCl(aq) solvent can rapidly dissolve all components of the reaction (7). It was prepared from analytical grade hydrochloric acid and deionized water, and its concentration, 1.0182 $mol\cdot kg^{-1}$, was determined by titration with standard sodium carbonate. Its concentration can also be expressed as the form of $HCl\cdot 54.561H_2O$. In all these determinations, strict control of the stoichiometries in each step of the calorimetric cycle must be observed, with the objective that the dissolution of the reactants gives the same composition as those of the products in the reaction (7).

Applying Hess's law, the enthalpy of reaction (7) ($\Delta_r H_m°(7)$) can be calculated according to the following expression

$$\Delta_r H_m°(7) = \Delta_r H_m°(1) + \Delta_r H_m°(2) - \Delta_r H_m°(3) - \Delta_r H_m°(4) - \Delta_r H_m°(5) - \Delta_r H_m°(6)$$

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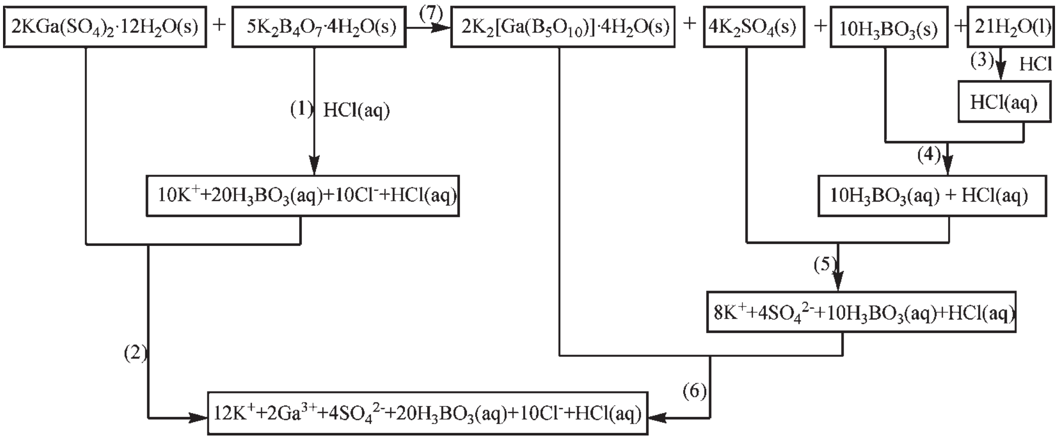

Figure 1. Schematic drawing of the thermodynamic circle.

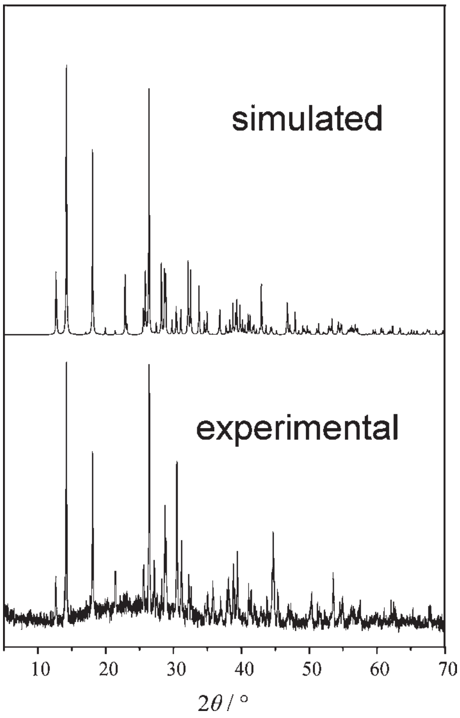

Figure 2. X-ray powder diffraction pattern of the synthetic sample.

The standard molar enthalpy of formation of $K_2[Ga(B_5O_{10})] \cdot 4H_2O$ can be obtained from the value of $\Delta_r H_m^\circ$ (7) in combination with the molar enthalpies of formation of $KGa(SO_4)_2 \cdot 12H_2O(s)$, $K_2B_4O_7 \cdot 4H_2O(s)$, $K_2SO_4(s)$, $H_3BO_3(s)$, and $H_2O(l)$.

Calorimeter. All the enthalpies of solution were measured with an RD496-III heat conduction microcalorimeter (Southwest Institute of Electron Engineering, China), which has been described in detail previously.[8,9] The total time required for the complete dissolution reaction was about 0.5 h. There were no solid residues observed after the reactions in each calorimetric experiment.

To check the performance of the calorimeter, the enthalpy of solution of KCl (mass fraction ≥ 0.9999) in deionized water was determined to be (17.54 ± 0.10) $kJ \cdot mol^{-1}$, which was in agreement with that of (17.524 ± 0.028) $kJ \cdot mol^{-1}$ reported in the literature.[10] This shows that the device used for measuring the enthalpy of solution in this work is reliable.

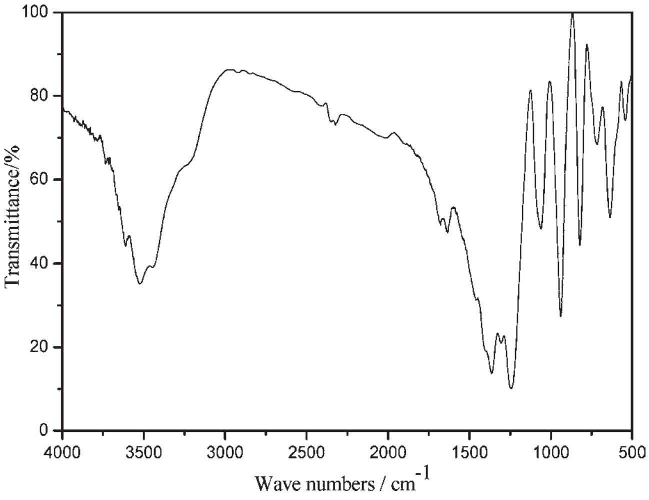

Figure 3. FT-IR spectrum of the synthetic sample.

RESULTS AND DISCUSSION

Characterization of the Synthetic Sample. Figure 2 shows the powder XRD pattern of the as-synthesized compound and the simulated pattern on the basis of the single-crystal structure of $K_2[Ga(B_5O_{10})] \cdot 4H_2O(s)$. The diffraction peaks on patterns corresponded well in position, indicating the phase purity of the as-synthesized sample.

The FT-IR spectrum (Figure 3) of the sample exhibited the following absorption bands, and they were assigned referring to the literature.[11] The band at 3525 cm^{-1} was the stretching mode of O−H. The band at 1633 cm^{-1} was the H−O−H bending mode, which shows that the compound contains crystal water. The bands at 1364 cm^{-1} and 939 cm^{-1} might be the asymmetric and symmetric stretching mode of B(3)−O, respectively. The band at 1244 cm^{-1} might be attributed to Ga−O vibration in the GaO_4 group. The bands at 1061 cm^{-1} and 822 cm^{-1} were the asymmetric and symmetric bending modes of B(4)−O. The band at 637 cm^{-1} is the out-of-plane bending mode of B(3)−O.

The simultaneous TG-DTA curves of the synthetic sample (Figure 4) indicate that the mass fraction of total mass loss is

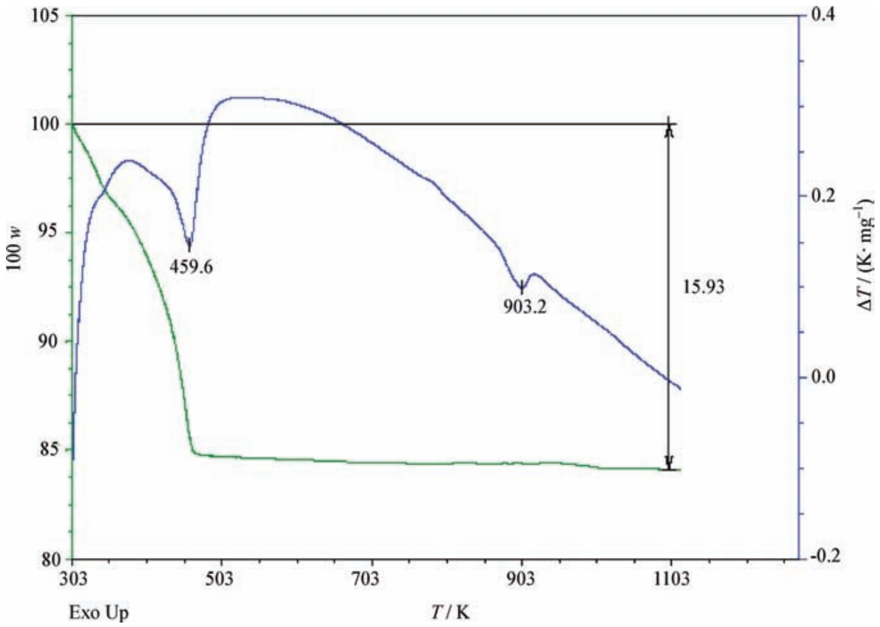

Figure 4. Simultaneous TG-DTA curves of the synthetic sample.

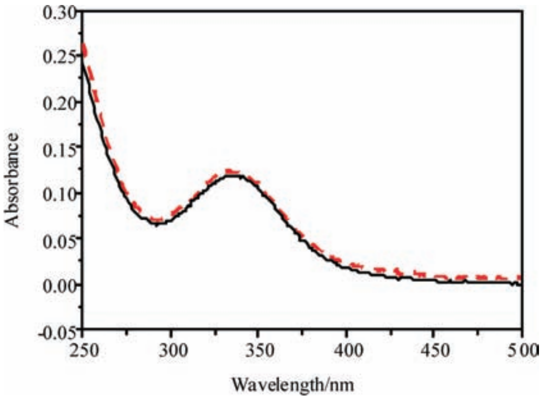

Figure 5. UV−vis spectra of solutions obtained from the dissolution of the reactants and products in reaction (7) at 298 K: —, the solution from reactants; - - -, the solution from products.

Table 1. Molar Enthalpies of Solution of KGa$(SO_4)_2 \cdot 12H_2O$(s) in H_2O at 298.15 K[a]

no.	m/mg	$\Delta_r H$/mJ	$\Delta_{sol}H_m$/(kJ·mol^{-1})
1	10.10	881.238	45.12
2	10.09	878.925	45.05
3	10.09	880.984	45.15
4	10.11	881.347	45.08
5	10.14	889.614	45.37
mean			45.15 ± 0.11[b]

[a] In each experiment, 8.00 cm^3 of H_2O was used. [b] Uncertainty is estimated as twice the standard deviation of the mean.

0.1593 from (303 to 473) K, which corresponds to the loss of four water molecules and agrees with the calculated mass fraction of 0.1659. In the DTA curve, the endothermic peaks appearing at (460 and 903) K are related to the dehydration and the melt of $K_2[Ga(B_5O_{10})]$, respectively.

Table 2. Molar Enthalpies of Solution of $KGa(SO_4)_2 \cdot 12H_2O$(s) in the Mixed Solvent of 1 mol·kg^{-1} HCl(aq) and $K_2B_4O_7 \cdot 4H_2O$ (aq) at 298.15 K[a]

no.	m/mg	$\Delta_r H$/mJ	$\Delta_{sol}H_m$/(kJ·mol^{-1})
1	7.12	960.250	69.74
2	7.18	958.282	69.02
3	7.17	961.230	69.33
4	7.20	961.975	69.09
5	7.22	963.739	69.03
mean			69.24 ± 0.27[b]

[a] In each experiment, 2.00 cm^3 of HCl (aq) was used. [b] Uncertainty is estimated as twice the standard deviation of the mean.

Table 3. Molar Enthalpies of Solution of K_2SO_4(s) in the Mixed Solvent of 1 mol·kg^{-1} HCl(aq) and H_3BO_3(aq) at 298.15 K[a]

no.	m/mg	$\Delta_r H$/mJ	$\Delta_{sol}H_m$/(kJ·mol^{-1})
1	5.02	1157.382	40.18
2	4.91	1143.266	40.18
3	4.91	1138.547	40.18
4	4.83	1120.478	40.18
5	4.98	1152.042	40.18
mean			40.38 ± 0.13[b]

[a] In each experiment, 2.00 cm^3 of HCl(aq) was used. [b] Uncertainty is estimated as twice the standard deviation of the mean.

The chemical analytical data of the synthetic sample are (calcd/found, mass fraction) B_2O_3 (0.4010/0.4000) and H_2O (0.1659/0.1593), which are consistent with the theoretical values.

All of the above results indicate that the synthetic sample is pure and suitable for the calorimetric experiments.

Confirmation of the Rationality of the Designed Thermochemical Cycle. The UV−vis spectra of the solutions obtained from reactions (2) and (6) in Figure 1 are identical (Figure 5).

This result indicates that the chemical components and physical-chemical property of solution obtained from the dissolution of the reactants were consistent with those of solution obtained from the dissolution of the products in the reaction (7), which shows that the designed thermochemical cycle (Figure 1) is reasonable and reliable and can be used to calculate the standard molar enthalpy of formation of $K_2[Ga(B_5O_{10})]\cdot 4H_2O(s)$.

Standard Molar Enthalpy of Formation of $KGa(SO_4)_2\cdot 12H_2O(s)$. The molar enthalpy of solution ($\Delta_{sol}H_m$) of $KGa(SO_4)_2\cdot 12H_2O(s)$ of (45.15 ± 0.11) kJ·mol^{-1} in H_2O at 298.15 K is listed in Table 1. The molar ratio of used $H_2O(l)$ and $KGa(SO_4)_2\cdot 12H_2O(s)$ was $2.3\cdot 10^4$, which can be thought of as an unlimitedly dilute solution. So, the standard molar enthalpy of formation of $-(5757.6 \pm 0.8)$ kJ·mol^{-1} for $KGa(SO_4)_2\cdot 12H_2O(s)$ can be obtained according to the following equations

$$KGa(SO_4)_2\cdot 12H_2O(s) = K^+(aq) + Ga^{3+}(aq) + 2SO_4{}^{2-}(aq) + 12H_2O(l)$$

$$\Delta_f H_m{}^\circ(KGa(SO_4)_2\cdot 12H_2O, s) = \Delta_f H_m{}^\circ(K^+, aq) + \Delta_f H_m{}^\circ(Ga^{3+}, aq) + 2\Delta_f H_m{}^\circ(SO_4{}^{2-}, aq) + 12\Delta_f H_m{}^\circ(H_2O, l) - \Delta_{sol}H_m$$

where $\Delta_f H_m{}^\circ(K^+, aq) = -(252.14 \pm 0.08)$ kJ·mol^{-1}, $\Delta_f H_m{}^\circ(Ga^{3+}, aq) = -(211.7 \pm 0.1)$ kJ·mol^{-1}, $\Delta_f H_m{}^\circ(SO_4{}^{2-}, aq) = -(909.34 \pm 0.40)$ kJ·mol^{-1}, and $\Delta_f H_m{}^\circ(H_2O, l) = -(285.830 \pm 0.040)$ kJ·mol^{-1}, which were taken from the CODATA Key Values.[12]

Table 4. Molar Enthalpies of Solution of $K_2[Ga(B_5O_{10})]\cdot 4H_2O(s)$ in the Mixed Solvent of 1 mol·kg^{-1} HCl(aq), K_2SO_4(aq), and H_3BO_3(aq) at 298.15 K[a]

no.	m/mg	$\Delta_r H$/mJ	$\Delta_{sol}H_m$/(kJ·mol^{-1})
1	6.18	−677.114	−47.56
2	6.25	−684.525	−47.54
3	6.04	−655.476	−47.10
4	6.20	−676.602	−47.37
5	6.03	−658.387	−47.39
mean			−47.39 ± 0.16[b]

[a] In each experiment, 2.00 cm^3 of HCl(aq) was used. [b] Uncertainty is estimated as twice the standard deviation of the mean.

Standard Molar Enthalpy of Formation of $K_2[Ga(B_5O_{10})]\cdot 4H_2O(s)$. The molar enthalpy of solution of $KGa(SO_4)_2\cdot 12H_2O(s)$ in the mixture of 2.00 cm^3 of 1 mol·kg^{-1} HCl(aq) and calculated amount of $K_2B_4O_7\cdot 4H_2O$ at 298.15 K is listed in Table 2. The molar enthalpy of solution of $K_2SO_4(s)$ in the mixture of 2.00 cm^3 of 1 mol·kg^{-1} HCl and calculated amount of H_3BO_3 at 298.15 K is listed in Table 3. The molar enthalpy of solution of $K_2[Ga(B_5O_{10})]\cdot 4H_2O(s)$ in the mixture of 2.00 cm^3 of 1 mol·kg^{-1} HCl and calculated amount of K_2SO_4 and H_3BO_3 at 298.15 K is listed in Table 4. In the tables, the m is the mass of samples; $\Delta_{sol}H_m$ is the molar enthalpy of solution of solute; and the uncertainty is estimated as twice the standard deviation of the mean with $n = 5$.

Table 5 gives the thermochemical cycle used for the derivation of the standard molar enthalpy of formation of $K_2[Ga(B_5O_{10})]\cdot 4H_2O(s)$. The mean molar enthalpy of solution of $H_3BO_3(s)$ of (21.84 ± 0.05) in 2.00 cm^3 of 1 mol·kg^{-1} HCl(aq) was taken from our previous work.[9] The mean enthalpy of solution of $K_2B_4O_7\cdot 4H_2O(s)$ of (53.36 ± 0.22) kJ·mol^{-1} was taken from the literature.[5] The enthalpy of dilution of HCl(aq) was calculated from NBS tables.[13] The enthalpy change of (120.56 ± 1.46) kJ·mol^{-1} for the formation of $K_2[Ga(B_5O_{10})]\cdot 4H_2O$ from the reagents in the solid phase (reaction (7)) was calculated on the basis of the thermochemical cycle. The standard molar enthalpies of formation of $H_3BO_3(s)$ and $H_2O(l)$ were taken from the CODATA Key Values,[12] namely, $-(1094.8 \pm 0.8)$ kJ·mol^{-1} and $-(285.830 \pm 0.04)$ kJ·mol^{-1}, respectively. The standard molar enthalpy of formation of $K_2SO_4(s)$ was taken from NBS tables,[13] namely, $-(1437.79 \pm 0.03)$ kJ·mol^{-1}. The standard molar enthalpy of formation of $K_2B_4O_7\cdot 4H_2O(s)$ of $-(4568.77 \pm 3.24)$ kJ·mol^{-1} was taken from the literature.[5]

From these data, the standard molar enthalpy of formation of $K_2[Ga(B_5O_{10})]\cdot 4H_2O(s)$ was calculated to be $-(5768.5 \pm 9.1)$ kJ·mol^{-1}.

CONCLUSIONS

Through an appropriate thermochemical cycle, the standard molar enthalpy of formation of $K_2[Ga(B_5O_{10})]\cdot 4H_2O(s)$ has been obtained from measured enthalpies of solution, together with the

Table 5. Thermochemical Cycle and Results for the Derivation of $\Delta_f H_m{}^\circ$ ($K_2[Ga(B_5O_{10})]\cdot 4H_2O(s)$, 298.15 K)[a]

no.	reaction	$\Delta_{sol}H_m$/(kJ·mol^{-1})
(1)	$5K_2B_4O_7\cdot 4H_2O(s) + 289.384(HCl\cdot 54.561H_2O) =$ $10KCl(aq) + 20H_3BO_3(aq) + 279.384(HCl\cdot 56.496H_2O)$	266.88 ± 1.10
(2)	$2KGa(SO_4)_2\cdot 12H_2O(s) + 10KCl(aq) + 20H_3BO_3(aq) + 279.384(HCl\cdot 56.496H_2O) =$ $4K_2SO_4(aq) + 4KCl(aq) + 20H_3BO_3(aq) + 2GaCl_3(aq) + 279.384(HCl\cdot 56.582H_2O)$	138.48 ± 0.54
(3)	$21H_2O(l) + 289.384(HCl\cdot 54.561H_2O) = 289.384(HCl\cdot 54.634H_2O)$	−(0.42 ± 0.04)
(4)	$10H_3BO_3(s) + 289.384(HCl\cdot 54.634H_2O) =$ $10H_3BO_3(aq) + 289.384(HCl\cdot 54.634H_2O)$	218.40 ± 0.50
(5)	$4K_2SO_4(s) + 10H_3BO_3(aq) + 289.384(HCl\cdot 54.634H_2O) =$ $4K_2SO_4(aq) + 10H_3BO_3(aq) + 289.384(HCl\cdot 54.634H_2O)$	161.52 ± 0.52
(6)	$2K_2[Ga(B_5O_{10})]\cdot 4H_2O(s) + 4K_2SO_4(aq) + 10H_3BO_3(aq) + 289.384(HCl\cdot 54.634H_2O) =$ $4K_2SO_4(aq) + 4KCl(aq) + 20H_3BO_3(aq) + 2GaCl_3(aq) + 279.384(HCl\cdot 56.582H_2O)$	−(94.78 ± 0.32)
(7)	$2KGa(SO_4)_2\cdot 12H_2O(s) + 5K_2B_4O_7\cdot 4H_2O(s) =$ $2K_2[Ga(B_5O_{10})]\cdot 4H_2O(s) + 4K_2SO_4(s) + 10H_3BO_3(s) + 21H_2O(l)$	(120.56 ± 1.46)[b]

[a] $\Delta_f H_m{}^\circ(K_2[Ga(B_5O_{10})]\cdot 4H_2O, s) = 1/2[\Delta_r H_m{}^\circ(7) + 2\Delta_f H_m{}^\circ(KGa(SO_4)_2\cdot 12H_2O, s) + 5\Delta_f H_m{}^\circ(K_2B_4O_7\cdot 4H_2O, s) - 4\Delta_f H_m{}^\circ(K_2SO_4, s) - 10\Delta_f H_m{}^\circ(H_3BO_3, s) - 21\Delta_f H_m{}^\circ(H_2O, l)]$. [b] Uncertainty of the combined reaction is estimated as the square root of the sum of the squares of uncertainty of each individual reaction.

standard molar enthalpies of formation of $H_3BO_3(s)$, $KGa(SO_4)_2 \cdot 12H_2O(s)$, $K_2SO_4(s)$, $K_2B_4O_7 \cdot 4H_2O(s)$, and $H_2O(l)$.

AUTHOR INFORMATION

Corresponding Author

*Tel.: +86-29-8530-7765. Fax: +86-29-8530-7774. E-mail: liuzh@snnu.edu.cn.

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