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У збірнику викладено результати досліджень з розробки та застосування високоглиноземних, глиноземно-хромоксидно-цирконійоксидно-кремнеземних, глиноземно-цирконійоксидно-кремнеземних, периклазовуглецевих, шамотних вогнетривів та бетонів, а також технічної кераміки. Наведено дані з розробки нормативної документації на вогнетриви.

Збірник призначений для науковців, аспірантів, студентів, спеціалістів із виготовлення та використання вогнетривів і технічної кераміки, що застосовуються у різних галузях промисловості.

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Evaluation of some methods for calculation of the enthalpies of formation of some inorganic compounds in the system $CaO—BaO—Al_2O_3—Fe_2O_3—SiO_2$

Introduction

It is known that calcium aluminate cement concretes for their heat resistance and chemical stability exceed portland cement concretes considerably. But calcium aluminate cement, as a portland cement, is not a radiation-resistant material. Partial replacement of calcium oxide with barium oxide increases the binder's resistance to simultaneous action of ionizing radiation and high temperatures. That is why the systems corresponding to ordinary portland or alumina types of cement in which part of calcium oxide is substituted for barium oxide are paid great attention [1—3]. Such substitution will give ordinary cements several useful properties: increased fire resistance, high resistance to attack by sulfate corrosion and seawater, protection properties, as well as for x-rays and gamma-radiation, etc. Taking into consideration all mentioned above the quaternary system $CaO—BaO—Al_2O_3—Fe_2O_3—SiO_2$, containing both high-melting hydraulic active compounds and phases, which are characterized by high-protection properties for x-rays and gamma-radiation, is of potential interest in the field of special binders.

To create the above-mentioned materials first of all it is necessary to predict the preliminary interaction of components in the system and, as a consequence, the properties of products obtained. From this point of view, the thermodynamic method is preferable since it allows by calculation approach, without carrying out numerous experiments, to determine optimum synthesis conditions and composition of products. However, the absence of experimental data for standard enthalpies of formation (ΔH^0_{298}) of some inorganic binary and ternary compounds does not allow us to obtain thermodynamic characteristics of all processes being studied [4, 5].

Nowadays several methods are known for the calculation of the standard enthalpies of formation of inorganic compounds. It should be noted that, as a rule, most of these methods are used for the (ΔH^0_{298}) calculation of the binary compounds and demand the knowledge of additional data (for example, melting point, structure features, etc.). Considering all mentioned above, the assessment of the accuracy of known methods for calculating the standard enthalpies of formation of some little-known inorganic compounds in comparison with the method proposed by the authors of this article is actual and urgent.

Theoretical part

To evaluate the accuracy of some known methods the standard enthalpies of formation of all binary and ternary compounds (the silicates, aluminates and ferrites of calcium and barium) in the systems $CaO—BaO—SiO_2$, $CaO—BaO—Al_2O_3$ and $CaO—BaO—Fe_2O_3$ have been calculated by following techniques: Shchukarev's isoatom method, Lagzdinya's method, Kleptsova's method, Kasenov's method and Sladkov—Morachevsky's method.

Shchukarev's isoatom method [6] is based on the consideration (in general form) relation between the composition of compounds (n) in the binary system and corresponding values of enthalpies of formation (ΔH^0_{298}). That is why a plot of $(\Delta H^0_{298})/n$ as a function of the composition of compounds (in mole fractions) yields a smooth curve — isoatom. The smooth shape of the isoatom shows the smooth variation of bond fraction between atoms of different elements and bond fraction between the same ones. Thus, the unknown values can be found by interpolation (and sometimes even by extrapolation).

The idea of the interrelation of compound compositions and their thermodynamic characteristics was used by Lagzdinya and coworkers [7]. They have revealed that in the binary oxide systems, the formation enthalpies of inorganic compounds and their melting points are connected by a relationship. Proceeding from the assumption that the higher the melting point of a compound the higher its gram-atom heat of formation, Lagzdinja *et al* proposed to consider the formation enthalpies of the rest compounds in the system being studied, which have melting points, as additive quantities, and to calculate them from proportion. However, authors note that the method is applicable only for oxygen-containing compounds whose melting points (in the range of specific classes of compounds,

e.g. phosphates, borates, germanates) differ no more than by 30 %. Besides, the mentioned calculation method like Shchukarev's method requires graphic representation.

The method based on the correlation of unknown thermochemical data for some compounds, whose compositions are expressed by stoichiometric amounts of oxides, has been developed by Kleptsova [8]. The basis of correlation is the following assumptions: 1) the equality of energy contributions of oxides, which form the compound; 2) additive growth of thermochemical values with the increase of oxide amount. It should be noted that the complicated body of mathematics is used by a given method. Thus, the awkwardness and duration of calculations make difficult the application of the method.

Kasenov [9] proposed the method for calculating the (ΔH^0_{298}) of inorganic compounds, which takes into account not only the melting point of a compound but also sometimes unknown energy characteristics — such as the average ionization potential of metals of given compounds. The accuracy of the method is equal to ± 5 %, as the author reports.

Sladkov and Morachevsky [10] supposed that the compound can be presented as complex oxide (aluminate, chromite, sulfate, ferrite). The stoichiometric composition of the compound is considered as combination of basic and acid oxide. Taking into account the thermal effect under standard conditions, standard formation enthalpies of compound and parameters of basic and acid ions the thermal effect of the reaction is calculated.

It should be noted that all of the above-mentioned techniques are used for calculation of formation enthalpies of binary compounds only.

Taking into consideration all mentioned above in our investigation a simple and highly exact method for the (ΔH^0_{298}) calculation has been proposed [11]. The given method takes into account *the mean gram-atom heat of the formation* of compounds, as well as *the amount of atoms* in the compounds of the binary or ternary systems being studied. It is revealed that the relation of the sum of gram-atom enthalpies of formation for compounds of a specific class $\left(\sum \Delta H^{\circ}_{\text{f }298}\right)$ to the sum of atoms in these compounds $\left(\sum N_{\text{comp.}}\right)$ is constant for some compounds of the same type:

$$\bar{n}_{\text{gr.-at.}} = \frac{\sum \Delta H^{\circ}_{298\text{comp.}}}{\sum N_{\text{comp.}}} = \text{const.} \qquad (1)$$

The mean gram-atom heat of the formation of compounds of the being considered type is calculated by using known experimental data only. To determine the unknown formation enthalpy of a compound (providing the compound composition is known) it is necessary to solve the reverse problem i.e. to multiply the mean gram-atom heat of the formation of compounds of the specific class (e.g. barium silicates) on the amount of atoms in the being considered compound (e.g. Ba_4SiO_6):

$$\Delta H^{\circ}_{298\text{comp.}} = \overline{n}_{\text{gr.-at.}} \cdot N_{\text{comp.}} \qquad (2)$$

It should be emphasized that the appreciable difference between the proposed method and methods, considered in the given investigation, is the possibility of calculating values of formation enthalpies for ternary compounds. In this case, the mean gram-atom heat of formation is determined as the relation of the sum for formation enthalpies of all binary compounds in the ternary system ((ΔH^0_{298}) of which is determined experimentally) to the sum of atoms in these compounds.

Due to the absence of experimental data on the standard formation enthalpies of some inorganic compounds included in the binary and ternary systems of the multi-component system CaO—BaO—Al_2O_3—Fe_2O_3—SiO_2, the values of the formation enthalpies for calcium and barium silicates, aluminates and ferrites have been calculated by the techniques described.

Experimental part

To evaluate the accuracy of some known methods the standard formation enthalpies of all binary and ternary compounds (the silicates, aluminates and ferrites of calcium and barium) in the systems CaO—BaO—SiO_2, CaO—BaO—Al_2O_3 and CaO—BaO—Fe_2O_3 have been fulfilled by following techniques: Shchukarev's isoatom method [6], Lagzdinya's method [7], Kleptsova's method [8], Kasenov's method [9], Sladkov—Morachevsky's method [10], and proposed technique [11].

The (ΔH^0_{298}) calculations for calcium and barium silicates are fulfilled according to the techniques described (the necessary thermodynamic data are taken from [4, 12—14]) and the results obtained are given in Table 1.

The necessary thermodynamic data for calculation of the standard enthalpies of formation for the calcium and barium aluminates in the system CaO—BaO—Al_2O_3 were taken from [4, 15, 16] and the obtained results are presented in Table 2.

Table 1

Formation enthalpies of calcium and barium silicates

Compound	Enthalpies of formation, ΔH^0_{298}, kJ/mole					
	Calculated by:					Experimental data
	Shchukarev's method [6]	Lagzdinya's method [7]	Kleptsova's method [8]	Kasenov's method [9]	Proposed method [11]	
Ca_3SiO_5	2930.98	2981.18	29.68	3419.42	2953.99	2930.60 [12]
Ca_2SiO_4	2308.65	2308.65	2312.58	2691.02	2297.56	2306.91 [12]
$Ca_3Si_2O_7$	3956.47	3921.96	3969.30	2189.19	3938.64	3956.54 [13]
$CaSiO_3$	1635.69	1608.50	1655.40	1291.56	1641.13	1635.73 [12]
Ba_4SiO_6	3484.64	3509.75	3721.78	—	3560.12	—
Ba_3SiO_5	2900.98	2913.53	3025.40	2597.51	2910.77	—
Ba_2SiO_4	2297.23	2297.23	2329.03	2530.69	2263.92	2297.14 [14]
$BaSiO_3$	1628.20	1620.67	1632.65	1346.12	1617.07	1628.33 [14]
$Ba_2Si_3O_8$	4194.67	4177.93	4201.58	2245.47	4204.42	4194.33 [14]
$Ba_5Si_8O_{21}$	10960.41	10911.45	10971.24	3192.39	10996.22	—
$Ba_3Si_5O_{13}$	6755.28	6728.50	6770.50	2691.57	6791.76	—
$BaSi_2O_5$	2552.87	2550.98	2568.93	1635.27	2587.34	2552.99 [14]
$BaCa_2Si_3O_9$	—	—	—	—	4887.12	—
$Ba_5Ca_3Si_4O_{16}$	—	—	—	—	9122.63	9298.83[4]
Relative error of method, Δ, %	0.006	0.652	0.716	26.110	0.720	—

Table 2

Formation enthalpies of calcium and barium aluminates

Compound	Enthalpies of formation, ΔH^0_{298}, kJ/mole					
	Calculated by:					Experimental data
	Shchukarev's method [6]	Lagzdinya's method [7]	Kleptsova's method [8]	Sladkov—Morachevsky's method [10]	Proposed method [11]	
$CaAl_{12}O_{19}$	10642.84	10747.13	10742.82	11117.52	10608.96	10742.84 [15]
$CaAl_2O_4$	2327.18	2326.18	2319.03	2382.29	2320.71	2326.18 [15]
$CaAl_4O_7$	4089.61	4010.84	4003.78	4130.49	3978.36	4029.61 [15]
$Ca_3Al_2O_6$	3527.57	3608.88	3587.57	3604.64	3646.83	3587.57 [15]
$Ca_{12}Al_{14}O_{33}$	19628.81	19490.00	19404.56	19392.16	19560.27	19428.82 [15]
$BaAl_{12}O_{19}$	10740.33	11056.30	10931.28	11472.47	10584.00	10730.00 [16]
$BaAl_2O_4$	2311.24	2388.65	2311.24	2377.19	2315.25	2338.00 [16]
$Ba_3Al_2O_6$	3485.69	3640.03	3485.7	3397.37	3638.69	3515.00 [16]

The end of table 2

Compound	Enthalpies of formation, ΔH^0_{298}, kJ/mole					
	Calculated by:					Experimental data
	Shchukarev's method [6]	Lagzdinya's method [7]	Kleptsova's method [8]	Sladkov—Morachevsky's method [10]	Proposed method [11]	
$Ba_4Al_2O_7$	4054.42	4254.42	4072.93	3942.03	4299.75	—
$Ba_5Al_2O_8$	4620.81	4747.59	4660.16	4494.99	4961.25	—
$Ba_7Al_2O_{10}$	5742.59	6017.07	5834.62	5602.68	6284.25	—
$Ba_8Al_2O_{11}$	6300.48	6683.73	6421.85	6158.66	6945.75	—
$Ba_{10}A1_2O_{13}$	7411.01	7895.01	7596.31	7271.35	8268.75	—
$Ba_3CaAl_2O_7$	—	—	—	4011.65	4304.82	4110.10 [4]
$BaCaAl_4O_8$	—	—	—	4650.55	4635.96	—
$BaCa_2Al_8O_{15}$	—	—	—	8610.34	8603.64	8680.48 [4]
Relative error of method, Δ, %	1.28	1.46	0.36	2.17	1.36	—

In Table 3 the results of the evaluation of the (ΔH^0_{298}) for the compounds in the system CaO—BaO—Fe_2O_3 are given. Standard enthalpies of formation for calcium and barium ferrites have been calculated following some known techniques. Thermodynamic data for calculation were taken from [12, 17, 18].

Table 3

Formation enthalpies of calcium and barium ferrites

Compound	Enthalpies of formation, ΔH^0_{298}, kJ/mole				
	Calculated by:				Experimental data
	Lagzdinya's method [7]	Kleptsova's method [8]	Sladkov—Morachevsky's method [10]	Proposed method [11]	
$CaFe_4O_7$	2302.58	2468.58	2666.84	2741.40	—
$CaFe_2O_4$	1476.03	1530.93	1650.88	1599.15	1530.93 [17]
$Ca_2Fe_2O_5$	2124.26	2124.22	2188.65	2056.05	2124.22 [12]
$BaFe_{12}O_{19}$	—	4720.74	6166.80	6462.72	5534.18 [18]
$Ba_2Fe_2O_5$	—	1878.03	2174.89	1817.64	1763.97 [12]
$Ba_3Fe_2O_6$	—	2473.25	2652.36	2221.56	2640.73 [18]
$Ba_5Fe_2O_8$	—	3663.69	3706.94	3029.40	—
$Ba_2Fe_6O_{11}$	—	3253.20	4124.71	3837.24	—
$BaFe_2O_4$	—	1282.81	1854.00	1413.72	1282.81 [12]
$Ba_7Fe_4O_{13}$	—	5541.72	5687.27	4847.40	5541.71 [12]
$CaBaFe_4O_8$	—	—	3179.67	3012.87	—
Relative error of method, Δ, %	—	0.040	0.100	0.105	—

Results and discussion

It is revealed that Sladkov—Morachevsky's method [10] cannot be used for the calculation of standard enthalpies of formation for calcium and barium silicates because the given authors do not present the necessary data on silicon dioxide.

It is ascertained that Shchukarev's isoatom method [6] is not applicable for determining the ΔH^0_{298} for calcium and barium ferrites owing to a deficit of experimental data. This fact does not allow us to plot a smooth curve isoatom.

Besides technique proposed by Lagzdinya [7] cannot be applied for the calculation of formation enthalpies of barium ferrites due to the location of two extreme points on a plot of (ΔH^0_{298}) for compounds of a given class as a function of mole relation of compound component. Thus, data obtained in this way is impossible to use for calculation.

Moreover, the possibility of using Sladkov—Morachevsky's technique for ΔH^0_{298} calculation of the ternary compounds, which contain two alkaline-earth oxides and one acidic one is shown (in this case the basic oxides forming the compound is considered as complex cation that is the cation parameter is defined as the arithmetic mean of two cations of alkaline-earth elements).

As a result of the evaluation of the accuracy of the described methods and proposed technique following values of relative errors are obtained (%): — for silicates of calcium and barium — 0.006 for Shchukarev's method, 0.652 for Lagzdinya's method, 0.716 for Kleptsova's method, 26.11 for Kasenov's method, and 0.72 for our technique; — for aluminates of calcium and barium — 1.28 for Shchukarev's method, 1.46 for Lagzdinya's method, 0.36 for Kleptsova's method, 2.17 for Sladkov—Morachevsky's method, and 1.36 for proposed technique; — for ferrites of calcium and barium — 0.04 for Kleptsova's method, 0.1 for Sladkov—Morachevsky's method, and 0.105 proposed method, respectively. The obtained results show that the convergence in the experimental data and calculated values of formation enthalpies (except Kasenov's method, whose error is inadmissible for thermodynamic calculations) is considerable. Thus, the value of relative error for the proposed method is in the range of 0.105—1.36.

Thus, the proposed method takes precedence over known techniques:

— does not require graphic representation (unlike Shchukarev's and Lagzdinya's methods);

— does not demand knowledge of numerous data, but only the mean gram-atom enthalpy of formation and number of atoms in the compound (unlike Sladkov—Morachevsky's and Kasenov's methods);

— does not require a complicated body of mathematics (unlike Kleptsova's method);

— has a high convergence in the experimental data and calculated values (relative error of method does not exceed 1.5 %);

— can be applied for the (ΔH^0_{298}) calculation of the oxygen-containing inorganic binary compounds (for example, silicates, borates, aluminates, etc.) whose melting points differ more than by 30 % (unlike Lagzdinya's and Kasenov's method);

— can be used to determine the formation enthalpies of ternary compounds.

The data obtained are used for thermodynamic estimation of solid-state reactions of phase-formation processes as well as for investigation of sub-solidus composition in the ternary systems $CaO—BaO—SiO_2$ [19], $CaO—BaO—Al_2O_3$ [20], $CaO—BaO—Fe_2O_3$ [21] and quaternary system $CaO—BaO—Al_2O_3—Fe_2O_3$ [22].

Conclusions

Thus, by using the various methods the unknown values of standard enthalpies of formation for binary compounds Ba_4SiO_6, Ba_3SiO_5, $Ba_5Si_8O_{21}$, $Ba_3Si_5O_{13}$, $Ba_4Al_2O_7$, $Ba_5Al_2O_8$, $Ba_7Al_2O_{10}$, $Ba_8Al_2O_{11}$, $Ba_{10}A1_2O_{13}$, $CaFe_4O_7$, $Ba_5Fe_2O_8$, $Ba_2Fe_6O_{11}$ as well as ternary compounds $BaCa_2Si_3O_9$, $BaCaAl_4O_8$, $CaBaFe_4O_8$ have been calculated. An assessment of the accuracy of calculations fulfilled by the various methods is given. The possibility of using the modernized Sladkov—Morachevsky's method for calculating the formation enthalpies of ternary compounds is shown. Also, the advantages of the technique proposed by authors for the calculation of heat formation of complex oxygen-containing inorganic compounds are presented. The given method allows to calculation of the standard heat formation for ternary compounds using the mean gram-atom heat of the formation of compounds for the specific class $\bar{n}_{\text{gr.-at.}}$, which is defined as the ratio of the sum of the formation enthalpies of all binary compounds of the ternary system (ΔH^0_{298}), whose have been determined experimentally, to the number of atoms which these compounds consist of.

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