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Comprehensive Study of the Process of Formation of the Ceramic Material with the Composition $1.5MgO \cdot 0.5RO \cdot 2Al_2O_3 \cdot 5SiO_2$ [R = Mn(II), Fe(II), Cu(II), or Zn]

O. I. Salychits and S. E. Orekhova

Belarussian State Technological University, ul. Sverdlova 13-A, Minsk, 220050 Belarus
e-mail: oliSa_@list.ru

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Abstract—The results of the study of ceramic materials obtained by partial substitution of MgO in the $2MgO \times 2Al_2O_3 \times 5SiO_2$ (cordierite) with transition metal oxides FeO, MnO, CuO and ZnO, are presented The modification of the magnesium-aluminosilicate system led to intensification of the phase formation and improved the ceramics properties. In the systems modified with the oxides basic by their chemical nature (MnO, FeO) solid solutions formed $Mg_{2-y}R_yAl_4Si_5O_{18}$ ($0.5 < y < 1.5$), which, according to X-ray analysis, are close to high-and low-temperature modifications of cordierite, where R is Mn(II) or Fe(II). The qualitative phase composition of the materials modified with oxides of amphoteric nature (ZnO, CuO) is characterized by the presence of silicate and aluminate solid solutions $Mg_{1-x}R_xAl_2O_4$ ($0.25 < x < 0.75$), where R is Cu(II) or Zn. The activation energies of the studied processes and standard heats of formation of products were determined.

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Ceramic materials are widely used in technology and industry, particularly in engineering. These materials should obey a number of requirements, including mandatory the low thermal linear expansion coefficient (TLE) and high specific volume electric resistivity (ρ_V). Introduction of modifying agents into the initial composition at the synthesis of ceramic materials in some cases leads to improvement of the characteristics of the produced material. However, as a rule, information concerning the influence of modifying additives on the material properties are not systemic in nature and relate only to establishing the fact that the properties are changed.

The aim of our work was to establish the regularities of phase formation processes occurring during the calcinations of a system corresponding to the composition of cordierite $2MgO \cdot 2Al_2O_3 \cdot 5SiO_2$ (ceramic material used in mechanical engineering) with a partial substitution of MgO by transition metal oxides, determination of kinetic characteristics and thermal effects of the phase formation processes and the effect of the nature of modifying oxides FeO, MnO, CuO and ZnO on these characteristics.

The modifying oxides were chosen accounting for the the diagram of the state of magnesium-aluminosilicate system [1] and the size of ion in the composition of the modifying oxide, and hence by the possibility of partial isomorphous substitution of the Mg^{2+} cation by the cation of the metal of the modifying oxide.

The modifying oxides used are of different nature. According to the current classification of ceramics [2], the FeO and MnO are basic, while CuO and ZnO amphoteric oxides. The metal atoms, forming the modifying oxides have electronic structure, characterized by the number of 3*d* electrons.

We studied the system of series *F*, *M*, *C*, and *Z*, obtained by partial equimolar substitution of MgO in the system of the composition $2MgO \cdot 2Al_2O_3 \cdot 5SiO_2$ by a modifying oxide (FeO, MnO, CuO, and ZnO, respectively), as well as of series *I*, whose composition corresponded to the stoichiometric content of oxides in magnesium-aluminosilicate $2MgO \cdot 2Al_2O_3 \cdot 5SiO_2$ (cordierite). The content of modifying agent in the systems of series *I*, *M*, *F*, *C* and *Z* were, respectively, 0, 5.9, 6.0, 6.6 and 6.7 wt %.

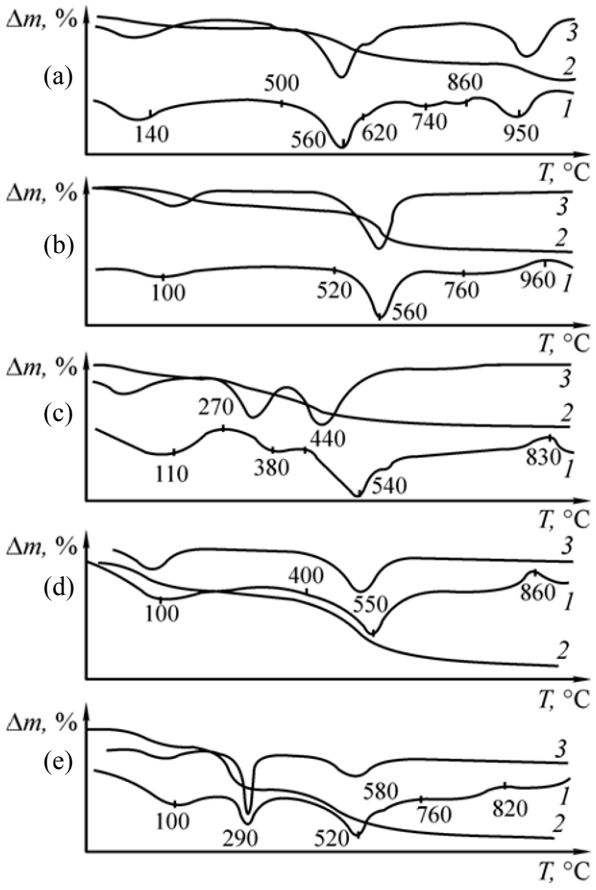

Fig. 1. Derivatograms of the reaction mixtures of compounds: (a) *I*, (b) *F*-1, (c) *M*-1, (d) *C*-1, and (e) Z-1. *T* is temperature, °C, Δ*m* is weight loss, %.

Based on data obtained by complex thermal analysis, X-ray diffraction (XRD) and infrared spectroscopy, it was found that the modifying additives show intensifying effect on the processes of phase formation in these systems. It is seen from the DTA curves (Figs. 1b–1e) that the exothermic effects corresponding to the phase formation are observed at the temperatures 820–960°C (1093–1233 K). In the DTA mass curve of the initial composition *I* (Fig. 1a) no thermal effect associated with the formation of crystalline phase in the temperature range 820–960°C (1093–1233 K) and below occurs.

As can be seen from Table 1, the physicochemical (apparent density $\rho_{apparent}$, kg m^{-3} and limit of mechanical strength of the static three-point bending $\sigma_{bending}$, MPa), thermal (thermal linear expansion coefficient, TLE, K^{-1}) and electrical (ρ_V, Ω cm) properties of the materials studied are improved compared with the unmodified material. The limit of mechanical strength $\sigma_{bending}$ of the samples of series *C* and volume resistivity ρ_V of the material samples of series *F* and *C* being slightly lower compared with other samples fall in the range of the characteristics of material with initial composition *I*, but their synthesis temperature is lower by 200–350 K. The TLE of the modified ceramic compositions falls in the series of modifying oxides in the direction: ZnO–FeO–MnO. The decrease in TLE of the ceramic compositions can be attributed to a greater degree of covalent bonding [3] in the oxides ZnO, FeO and MnO in comparison with the oxide MgO. Covalence of the CuO bond is also higher than that of MgO. However, the TLE of the samples of series *C* is much larger compared with initial composition *I*: the features of the composition and structure of both crystalline and glassy components of the ceramic material of *C* series probably affect the TFE magnitude more significantly.

Using the methods of XRD and IR spectroscopy and the data of electronic probe energy-dispersive X-ray fluorescence analysis (EDX) obtained with a JEOL JED-2201 instrument the regularities of the processes of phase formation of aluminates and aluminosilicates solid solutions in these systems were revealed. In the systems modified with oxides of the basic chemical nature (MnO, FeO), as in the case of magnesium-aluminosilicate system $2MgO{\cdot}2Al_2O_3{\cdot}5SiO_2$, the

Table 1. Physicochemical, thermal (TLE), and electrical (ρ_V) properties of ceramic materials of the composition MgO(MnO, FeO, CuO, ZnO)–Al_2O_3–SiO_2

Index of composition	Synthesis temperature, K	TLE×10^6, K^{-1}	ρ_V, Ω cm	$\sigma_{bending}$, MPa	$\rho_{apparent}\times10^{-3}$, kg m^{-3}
I	1623–1723	1.0–3.0	10^9–10^{10}	50.0	1.85–2.10
M	1323–1423	1.2–2.1	$(0.7–1.2)\cdot10^{11}$	58.3–59.9	2.25–2.37
F	1423–1523	1.9–2.3	$2.9\cdot10^{10}$	90.2–91.5	2.51–2.74
C	1273–1373	5.2–6.6	$(0.2–1.3)\cdot10^{10}$	39.5–49.3	2.34–2.41
Z	1423–1523	2.2–2.5	$(0.7–2.4)\cdot10^{11}$	68.2–79.2	2.32–2.34

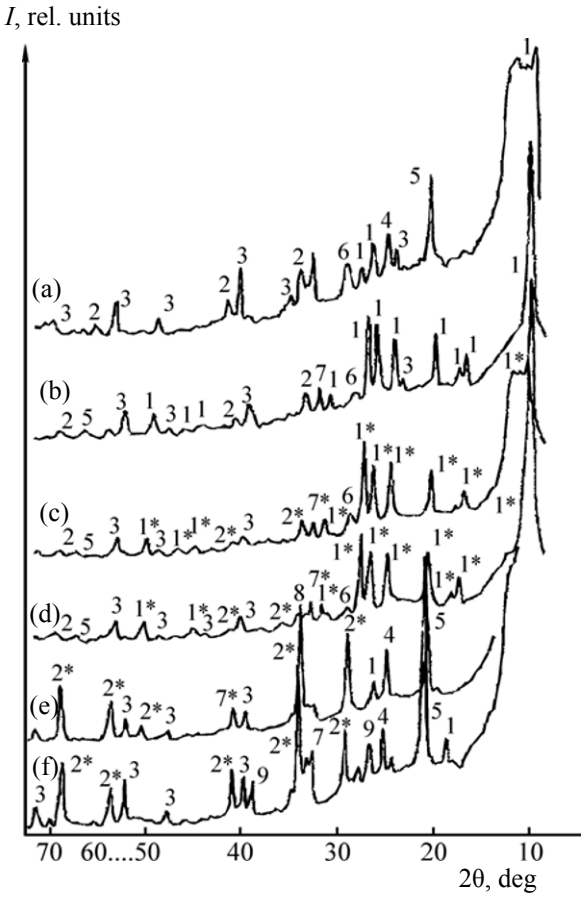

Fig. 2. Diffractograms of the samples of compositions *I*, *M*-1, *F*-1, *C*-1 and *Z* -1 synthesized at different temperatures (*T*, K): (a) *I*, *T* = 1473 K, (b) *I*, *T* = 1623 K , (c) *M*-1, *T* = 1423 K, (d) *F*-1, *T* = 1473 K, (e) *Z*-1, *T* = 1473 K, and (f) *C*-1, *T* = 1373 K. (1) $Mg_2Al_4Si_5O_{18}$, (1*) $Mg_{2-y}R_yAl_4Si_5O_{18}$, (2) $MgAl_2O_4$, (2*) $Mg_{2-x}R_xAl_2O_4$, (3) α-Al_2O_3, (4) α-quartz SiO_2, (5) β-cristobalite SiO_2, (6) $MgSiO_3$, (7) Mg_2SiO_4, (7*) R_2SiO_4, and (8) $MgFe_2O_4$.

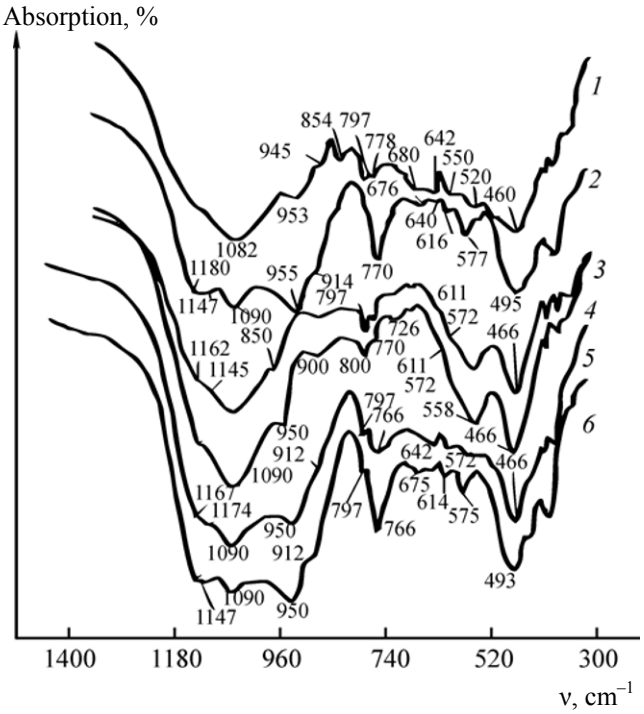

Fig. 3. IR spectra of the samples of series *I*, *F*, and *M* synthesized at different temperatures (*T*, K): (*1*) *I*, *T* = 1373 K, (*2*) *I*, *T* = 1623 K, (*3*) *F*-1, *T* = 1423 K, (*4*) *F*-1, *T* = 1423 K, (*5*) *M*-1, *T* = 1373 K, and (*6*) *M*-1, *T* = 1473 K.

compounds of aluminosilicate series are formed, the solid solution $Mg_{2-y}R_yAl_4Si_5O_{18}$ ($0.5 < y < 1.5$), that by X-ray data are close to the magnesium-aluminosilicates of high- and low-temperature modifications, where R is Mn(II) or Fe(II).This is evidenced by the appearance in the diffraction patterns (Fig. 2c and 2d) of the samples of series *M* and *F* synthesized at temperatures 1423 and 1473 K, of intense X-ray line 1* (interplanar distance *d* = 0.850–0.851, 0.490–0.491, 0.410, 0.337–0.338, 0.314–0.315, 0.304–0.305, 0.264–0.265, 0.233–0.234, 0.214, 0.188 nm) and in the infrared spectra (Fig. 3, curves *3*–*5*), of absorption bands in the frequency range 1167–1162, 1147–1145, 950, 900–912, 726–766, 611–614 , 558–575 cm^{-1}, characteristic of silicates with a ring structure of the silicon-oxygen group $[AlSi_5O_{18}]$ or $[Si_6O_{18}]$. In the material of *I* series the low-temperature modification $Mg_2Al_4Si_5O_{18}$ (cordierite) only at 1473 K begins to form, as evidenced by the appearance of poorly defined X-ray lines (*d* = 0.845, 0.313, 3.039 nm) in the diffraction pattern (Fig. 2a). The most intense X-ray lines of the $Mg_2Al_4Si_5O_{18}$ low- and high-temperature modifications appear in the diffraction patterns of samples of series *I* (*d* = 0.845, 0.491, 0.409, 0.337, 0.313, 3.039, 2.640, 2.340, 2.173, 1.875, 1.630 nm) when the synthesis temperature is 1623-1723 K (Fig. 2b).

The qualitative phase composition of the materials modified with oxides of amphoteric nature (ZnO, CuO) is characterized by the presence of silicate and aluminate solid solutions $Mg_{1-x}R_xAl_2O_4$ ($0.25 < x < 0.75$), where R is Cu(II) and Zn, as evidenced by the appearance in the diffraction patterns of samples of series *C* and *Z* (Fig. 2e and 2f) the characteristic *X*-ray lines 2* (*d* = 0.2436–0.2437, 0.2857–0.2857, 0.2020, 0.1649, 0.1554–0.1556, 0.1428–0.1429), 7 and 7*. The IR spectra of samples of series *C* and *Z* (Fig. 4) exhibits a strong absorption band characteristic of bound octahedral groups $[AlO_6]$ of aluminates (frequency range 500–680 cm^{-1}) and the tetrahedra $[SiO_4]$ in the compounds of the orthosilicate structural type

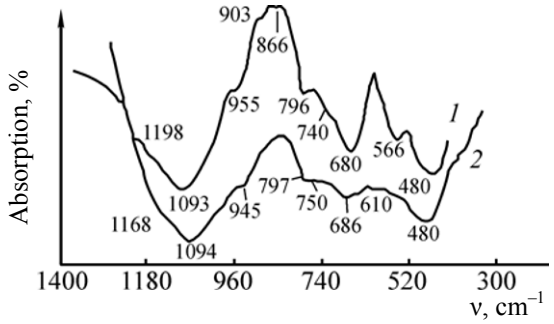

Fig. 4. IR spectra of the samples of series *C* and *Z* synthesized at different temperatures (*T*, K): (*1*) Z, *T* = 1473 K and (*2*) *C*, *T* = 1373 K.

(frequency range 800–900 cm^{-1}). In the samples of the materials of *M* and *F* series the solid solutions of aluminates and silicates of various compositions are formed as primary reaction products, and the intensity of the X-ray lines in the diffraction patterns (Fig. 2c and 2d) and the absorption bands in the IR spectra (Fig. 3, curves *3*–*5*) are much lower.

Based on the data obtained by quantitative X-ray diffraction, we determined the dependence of the degree of transformation on the duration of the reaction of formation of solid solutions of aluminates and aluminosilicates in the systems studied, as shown in Fig. 5. The phase formation at all temperatures proceeded during the first 30-45 minutes. The highest rate of interaction is typical for the compositions of *M* and *F* series modified with oxides of elements with a smaller number of valence 3*d*-electrons, and, respectively, with the less "effect of reverse screening" by 3*d*-orbital and the energy of bonding of valence 3*d*-electrons with the core.

As a result of complex studies carried out for all the compositions we obtained the reactions equations of phase formation of the aluminate and aluminosilicate solid solutions in the systems $MgO–RO–Al_2O_3–SiO_2$.

For a mathematical treatment of the data on the kinetics of phase formation and for the selection a model of the mechanism of the processes we used a series of published kinetic equations [4–9]. Using the Fisher's dispersion relation [10] in the analysis of obtained dependences of the degree of conversion F(α) on the reaction duration τ for each equation shows that the hypothesis of linearity can be accepted over the entire temperature range only for the diffusion models of the anti-Ginstling–Braunstein [Eq. (1)] and anti-Jander [Eq. (2)] physicochemical interaction:

$$k_{AGB}\tau = 1 + 2/3\alpha - (1 + \alpha)^{2/3}, \quad (1)$$

$$k_{AJ}\tau = [(1 + \alpha)^{1/3} - 1]^2. \quad (2)$$

Here k_{AGB} and k_{AJ} are the constants of anti-Ginstling–Braunstein and anti-Jander equations, respectively; τ is the duration of isothermal keeping; α is a degree of transformation.

We determined the dispersion of the adequacy [10] of experimental data and the selected kinetic models, that enabled us to compare experimental data with those obtained as a result of calculation (Fig. 5). Kinetics of formation of solid solutions of aluminates and aluminosilicates of low-temperature modification in the modified magnesium-aluminosilicate systems

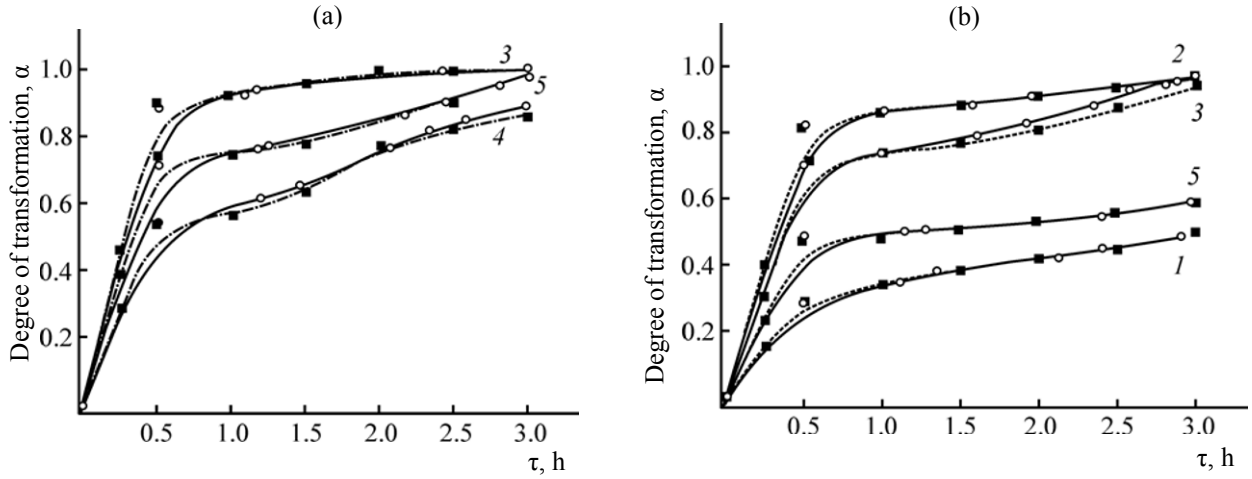

Fig. 5. Degree of transformation vs. duration of reaction during the formation of (a) $Mg_{1-x}R_xAl_2O_4$ and (b) $Mg_{2-y}R_yAl_4Si_5O_{18}$ at 1473 K for the samples of the series (*1*) *I*, (*2*) *M*, (*3*) *F*, and (*5*) *Z* and at 1273 K for the samples of the series (*4*) *C*. Dotted line and marked by squares are experimental data, solid lines are obtained by calculation.

are best described by the anti-Ginstling–Braunstein equation (1). The rate of reaction of the formation of high-temperature modification of the compounds of aluminosilicate series mainly obeys the anti-Jander equation (2). The Eqs. (1) and (2) are based on the same assumptions that the limiting stage of the formation of a new phase in the reactions of powder mixtures consisting of spherical particles is the diffusion of reactants through the product layer. These diffusion models differ by the law of changes in time or by the rate of change in the degree of conversion during the reaction [6]. In the system MgO–FeO–Al_2O_3–SiO_2 the formation of aluminates and aluminosilicates of low-temperature modification, unlike all the studied systems occurs in accordance with the diffusion model (2), and is characterized by low activation energy (Table 2). Formation of the aluminosilicates of high-temperature modification in the samples of the material of *F* series is also consistent with the diffusion model (2), but proceeds with a lower rate. This is due to the decrease in the amount of melt: the latter intensifies significantly the diffusion process due to its active participation in the reaction of formation of compounds $Mg_{2-y}Fe_yAl_4Si_5O_{18}$ of the low-temperature modification.

As a result of mathematical processing of experimental data using the selected kinetic models we obtained the values of the constants of kinetic equations (Fig. 6) and activation energies (Fig. 7) of the reactions listed in Table 2. In Figs. 6 and 7 as an example are shown the graphical dependences $F(\alpha) = f(\tau)$ and $\ln k = f(1/T)$ for one of the systems under study.

The result of the intensification of the reactions of phase formation in the modified systems is the increase in the values of the formation constants of aluminates and aluminosilicates compared with the similar constants for the original unmodified system MgO–Al_2O_3–SiO_2 (Table 2).

Using the values of ΔH_T^0 of studied phase formation processes obtained experimentally by differential scanning calorimetry (DSC) we calculated the standard heats of formation of $Mg_{1-x}R_xAl_2O_4$ and $Mg_{2-y}R_yAl_4Si_5O_{18}$ (Table 3) using the known procedure [11–14] and the reference data [6, 11, 15–18].

Comparison of the values of standard heats of formation of aluminates RAl_2O_4 from reference literature and the calculated by the method specified in the study of systems RO–Al_2O_3–SiO_2 [R = Mg, Mn(II), Fe(II), Cu(II), or Zn]) [19] (Table 4) confirms the validity of the method and allows us to consider that the results of the calculation are reasonably accurate.

Table 2. Constants of kinetic equations and activation energy of the reactions of formation of solid solutions $Mg_{1-x}R_xAl_2O_4$ and $Mg_{2-y}R_yAl_4Si_5O_{18}$ of the low- and high-temperature modifications

Index of composition	Temperature of calcination, K	E_a, kJ mol^{-1}	$k\times10^2$, h^{-1}
$Mg_{1-x}R_xAl_2O_4$			
F	1423	150±20	2.45±0.50[a]
	1450		3.16±0.50[a]
	1473		3.89±1.00[a]
C	1173	279±15	0.26±0.01[b]
	1223		0.93±0.15[b]
	1273		2.41±0.44[b]
Z	1423	282±13	2.04±0.50[b]
	1450		3.18±0.50[b]
	1473		3.26±1.00[b]
$Mg_{2-y}R_yAl_4Si_5O_{18}$ of the low-temperature modification			
I	1423	254±40	0.42±0.09[b]
	1450		0.63±0.08[b]
	1473		0.91±0.02[b]
F	1423	140±8	2.21±0.50[a]
	1450		2.73±0.06[a]
	1473		3.24±0.80[a]
Z	1423	140±16	1.01±0.50[b]
	1450		1.26±0.07[b]
	1473		1.47±0.40[b]
$Mg_{2-y}R_yAl_4Si_5O_{18}$ of the high-temperature modification			
I	1473	811±90	0.38±0.08[a]
	1500		1.23±0.09[a]
	1523		3.42±0.90[a]
M	1373	331±60	0.60±0.25[b]
	1423		3.78±0.80[b]
	1473		2.30±0.80[b]
F	1473	1040±75	0.25±0.09[a]
	1500		1.13±0.05[a]
	1523		2.57±0.35[a]

[a] k_{AGB} (anti-Gistling-Braunstein). [b] k_{AJ} (anti-Jander).

Our studies have established intensifying effect of modifying oxides in the processes of phase formation in the systems MgO–RO–Al_2O_3–SiO_2 [R = Mn(II), Fe(II), Cu(II), and Zn] resulting in a decrease in synthesis temperature of ceramic materials by 100–250 K with better principal characteristics compared with cordierite [20], and to obtain the kinetic characteristics

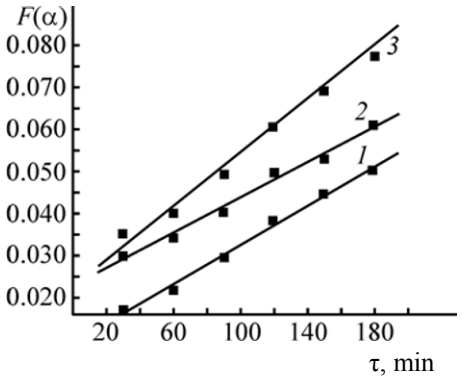

Fig. 6. Dependence of the function of conversion degree $F(\alpha)$ on the duration of reaction (τ) at the formation of solid solution $Mg_{0.45}Zn_{0.45}Al_2O_4$ in samples of *Z* series at different temperatures.

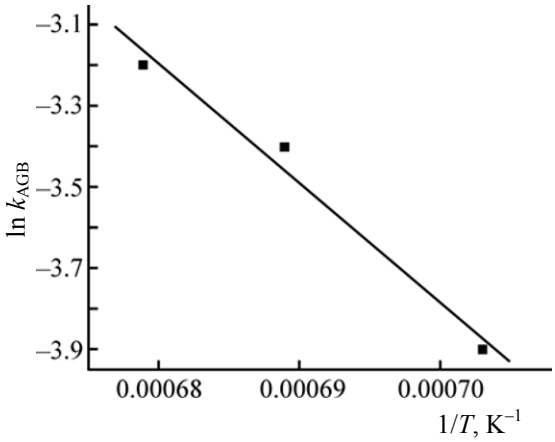

Fig. 7.Temperature dependence of ln k_{AGB} (anti-Ginstling–Braunstein) for the samples of *Z* series.

of the investigated processes and determine the standard heats of formation of solid solutions of aluminates and aluminosilicates that are absent in the literature.

EXPERIMENTAL

Synthesis of the samples was carried out by semidry pressing of powder mass with subsequent high-temperature processing of the blanks (1173–1723 K) in an electric furnace along the known method of producing ceramic materials [21]. As the starting materials were used the natural (mineral) raw components (refractory clay Veselovskaya "Granitic-Vesco," technical alumina GK-2 grade, talc Onotskoe) and chemically pure compounds of pure and chemically pure grade (manganese(II) carbonate, iron(II) oxide, copper(II) oxide, and zinc oxide). The properties of the synthesized ceramic materials listed in Table 1 were defined according to the methods traditionally used to determine the physicochemical [GOST 473.3-81 and GOST 24409-83], termal [GOST 24409-83] and electrical [GOST 24409-83] properties of ceramics.

Complex thermal analysis [recording of dilatometric (DTA), thermogravimetric (TG) and differential thermogravimetric (DTG) curves] of the experimental compositions were performed on a Paulic-Paulic-Erdey derivatograph in temperatures ranging from 20 to 1200°C (heating rate 10°C min^{-1}, the sample weight within 1 g). Diffraction patterns of the experimental samples were obtained using a Bruker D8 Advance X-ray diffractometer (Germany) and a DRON-3 installation (CuK_α radiation, λ = 0.15417 nm). Recording rate 1.2 deg min^{-1} in the range of angles 2θ = 10° to 70°. To identify the crystalline phases the U.S. database JCPDS was used. IR absorption spectra of

Table 3. Generalized calulated values of standard heat of formation of the aluminate and aluminosilicate solid solutions

Index of composition	Formula	Thermal effect of the phase forming reaction $-\Delta H^0_T$, J g^{-1}	Calculated standard heat of formation $-\Delta H^0_{f,298}$, kJ mol^{-1}	Reference data of $-\Delta H^0_{f,298}$, kJ mol^{-1} [5, 10]
M	$Mg_{0.5}Mn_{0.5}Al_2O_4$	7.15	2259	–
F	$Mg_{0.755}Fe_{0.245}Al_2O_4$	8.50	2209	–
M	$Mg_{1.12}Mn_{0.88}Al_4Si_5O_{18}$	39.73	9066	–
F	$Mg_{1.51}Fe_{0.49}Al_4Si_5O_{18}$	8.40	9909	–
I	$Mg_2Al_4Si_5O_{18}$	40.28	8944	9117.0±23.0
Z	$Mg_{0.55}Zn_{0.45}Al_2O_4$	13.03	2090	–
C	$Mg_{0.5}Cu_{0.5}Al_2O_4$	5.54	2301	–
I	$MgAl_2O_4$	23.06	2486	2313.3±2.1

Table 4. Calculated and reference values of standard heat of formation of aluminates RAl_2O_4 in the systems $RO–Al_2O_3–SiO_2$ [R = Mg, Mn(II), Fe(II), Cu(II), or Zn]

Formula	Estimated value of $-\Delta H^0_{f,298}$, kJ mol^{-1} [5]	Reference value of $-\Delta H^0_{f,298}$, kJ mol^{-1} [5]	Deviation of calculated from reference values, %
$MgAl_2O_4$	2486	2313	7.4
$MnAl_2O_4$	2080	2098	0.9
$FeAl_2O_4$	2018	1982	1.8
$CuAl_2O_4$	2029	1808	12.2
$ZnAl_2O_4$	2073	2067	0.3

test samples were recorded in the frequency range 400-1300 cm^{-1} at room temperature on a "Nexus" IR Spectrometer of "Terma" (USA) for the mixtures pelletized with KBr. For deciphering the spectra they were compared with the spectra of known substances [22–24].

The process of interaction in the system studied was performed using quantitative X-ray analysis under isothermal conditions. Isothermal calcination of the samples was carried out in an electric furnace SNOL 7.2/1300 with controlled temperatures from 1173 to 1523 K in steps of 50 K for a period from 0.5 to 3.0 h every 30 min. The accuracy in maintaining the temperature during the experiment was within 0.5 K. In determining the rate of the phase formation by XRD, the conditions for obtaining radiographs of all samples were identical: DRON-3 diffractometer (CuK_α radiation, λ = 0.15417 nm). The experiments were twice repeated. The conversion α was taken as the ratio of amount of the product formed at time τ in the current experiment to the amount of products formed under the condition of the reaction completion. The process of reaction was suggested completed when in the diffraction patterns of the samples the intensity of X-ray peaks of reaction products remained unchanged at increasing the duration and temperature of isothermal maintenance. Mathematical processing of experimental data obtained during the kinetic studies was carried out using a specially designed application software.

Determination of the change in enthalpy of phase formation processes in the investigated reaction mixtures was carried out by differential scanning calorimetry (DSC) using a DSC TGA / DSC 1 HT/319 STAR^e system of METTLER TOLEDO (Germany) and the original software STAR^e SW 9.20.

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