Degree of phase transformations in the conditions
of polythermal synthesis of $Sr_{2-x}Ba_xFeMoO_{6-\delta}$

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Abstract

The sequence of phase transformations during the crystallization of $SrBaFeMoO_{6-\delta}$ by the solid-phase technique from a stoichiometric mixture of simple oxides $SrCO_3$ + $BaCO_3$ + $0.5Fe_2O_3$ + $MoO_3$was studied. It has been established that the synthesis of barium – strontium ferromolybdate proceeds through a series of sequential - parallel stages. It was found that to minimize the effect of intermediate reaction products, it is necessary to use combined synthesis modes. As a result of using combined synthesis modes for annealing for 20 hours and T = 1443 K in vacuum of 10^{-5}Torr at the pressure of residual oxygen gas 10^{-8} Pa, it was possible to obtain a single-phase barium – strontium ferromolybdate compound with superstructural ordering of iron and molybdenum cations.

Keywords: magnetic metal oxide compound, double perovskite, polythermal synthesis, differential thermal analysis, thermogravimetricanalysis, crystallization rate, sequence and degree of phase transformations.

Introduction

Solid solutions of double perovskites with the general formula $Sr_{2-x}Ba_xFeMoO_{6-\delta}$, which have high chemical stability in a reducing atmosphere, high Curie temperatures (380 –420 K), and a significant degree of spin polarization of conduction electrons (~100%), as well as low values of the controlling magnetic fields (B<0.5T) are of great interest to specialists working in the field of spintronics [1–6]. Interest in such materials is due to the fact that these objects have unique and extremely important for practical applications magnetic and magneto-transport properties, and in some parameters, they surpass the known manganite-based systems. Nevertheless, the values of magnetoresistance (MR) and other important physical characteristics of $Sr_{2-x}Ba_xFeMoO_{6-\delta}$ used in microelectronics may differ between researchers, which, apparently, is associated with features of sample preparation techniques [7–12].

Whileanalysing the accumulated data obtained in[13,14], the multi-stage crystallization process of $Sr_{2-x}Ba_xFeMoO_{6-\delta}$was established, which is due to the complexity of phase transformations, low kinetics of phase formation, and weak mobility of Fe^{3+} and Mo^{5+} cations [15,16]. The other research works contain more information on the preparation of $Sr_{2-x}Ba_xFeMoO_{6-\delta}$ by the mechanico-chemical method with a high-temperature synthesis in a reducing gas environment[16–21]. At the same time, rigorous correlations between the functional characteristics of materials and their production conditions are practically absent in the performed studies. However, the formation of a single-phase $Sr_{2-x}Ba_xFeMoO_{6-\delta}$ compound under conditions of control over the defect formation processes and, accordingly, compound with reproducible physico-chemical properties needs analysis of the phase transformations occurring in the batchas well as studies of the kinetics of the degree of conversion of double perovskite during its crystallization. Therefore, lately, the attention of researchers has been drawn to deeper and more detailed approaches to the synthesis of double perovskites associated with the study of the sequence of phase transformations during their crystallization [11,13–19]. In this regard, investigations aimed at studying high-temperature phase transformations and determining the composition of intermediate crystalline phases in the synthesis of $Sr_{2-x}Ba_xFeMoO_{6-\delta}$ are of particular importance. The main aim of this study is toestablish a correlation between the rate of phase transformations and the degree of phase transformation of barium – strontium ferromolybdate, which will allow for a controlled change in the phase composition of the synthesized ceramic with reproducible physico-chemical properties.

Experimental

Strontium carbonate ($SrCO_3$), barium carbonate ($BaCO_3$), ferric oxide (Fe_2O_3), and molybdenum trioxide (MoO_3) were used as initial reagents for studying the sequence of phase transformations in obtained by the solid-phase technique compounds of variable composition $Sr_{2-x}Ba_xFeMoO_{6-\delta}$. Mixing and grinding of the stoichiometric mixture of the starting reagents was carried out in a ball mill "PM 100" Retsch GmbH in ethanol for 15 hours. The resulting powder was pressed into tablets 10 mm in diameter and 4–5 mm thick. Annealing was carried out in polythermal mode at temperatures of 573–1473 K in vacuum of 10^{-5} Torr at the pressure of residual oxygen gas 10^{-8} Pa and at different heating rates of 0.7: 1.4 and 2.5 deg / min, followed by quenching at room temperature. An important factor for the formation of superstructural ordering of Fe / Mo cations in the compound is the selection of the synthesis medium in order to control oxygen deficiency. Based on other research works on the development of double perovskite synthesis modes, it was found that if the pressure of the residual oxygen gas exceeds 10^{-8} Pa, it is impossible to obtain a single-phase compound [21]. For this reason, we do not consider this case in this study.

The phase composition and structure were determined using D8 Discover X-ray diffractometer (Bruker AXS GmbH) with a LynxEye (0D mode) detector Cu Kα (λ= 1.54 Å) radiation. X-ray generator voltage and current was 40 kV and 40 mA, respectively. Couple 2θ / θ scans were performed in the range of $20 - 80$ ° with a step size 0.033 °, time per step of 0.3 s. Processing of the resultant diffractograms was performed with ICSD–PDF2 (Release 2000) database and PowderCell, FullProf software by the Rietveld technique.

The thermal behavior was investigated using differential thermal analysis (DTA) and thermogravimetric analysis (TGA) by means of a differential scanning calorimeter (DSC) SetaramLabsys TG – DSC16 set up in an argon stream at a heating rate of 1.4 deg / min.

Results and Discussion

According to the differential thermal analysis, practically no changes occur in the DTA curves, when the sample consisting of the starting reagents in the stoichiometric ratio $SrCO_3$ + $BaCO_3$ + $0.5Fe_2O_3$ + MoO_3 is heated from 300 to 453 K. Nevertheless, according to the TGA data, a slight decrease in its mass is observed ($\Delta m/m_0 < 2\%$), that can be explained by the chemical processes taking place in the batchand associated evolution of gaseous reaction products (Fig. 1 and 2).

When heated to higher temperatures, thermal processes intensify and five well pronounced endothermic effects are observed in the temperature range 573–1473 K (Fig. 1). The first endothermic effect, starting at a temperature of 453 K and reaching a minimum atT = 825 K, is

caused by a significant decrease ($\Delta m/m_0 \sim 8\%$) in the mass of the sample according to thermogravimetric analysis, and is most likely due to the evolution of gaseous reaction products CO_2 and O_2 (Fig. 2 a, b).

This assumption is indicated by XRD data (Fig. 3). So, in the temperature range corresponding to the first endothermic effect, crystallization of the $BaMoO_4$ compound in the $BaCO_3 - MoO_3$ system is observed, proceeding with the release of carbon dioxide according to the reaction equation:

$$BaCO_3 + MoO_3 = BaMoO_4 + CO_2\uparrow \qquad (1)$$
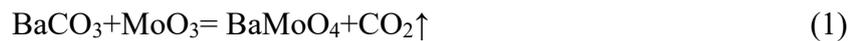
At the same time, according to the TGA data, in the temperature range 753–873 K (Fig. 2a), a sharp growth in the rate of decrease in the mass of the sample was noted, indicating the occurrence of parallel chemical processes with different amounts of gaseous reaction products released and absorbed. When studying the phase composition of the sample, the appearance of a strontium ferrite compound in the temperature range 453 K $<T\leq743$ K and a $SrBaFeMoO_{6-\delta}$ solid solution in the temperature range 743 K $<T\leq873$ K was detected. In this case, the chemical reaction with the formation of strontium ferrite occurs simultaneously with the absorption of oxygen and the release of carbon dioxide (2). The formation of a solid solution of strontium barium ferromolybdate is taking place with the release of carbon dioxide and oxygen (3):

$$SrCO_3 + 0.5Fe_2O_3 + 0.25O_2 = SrFeO_3 + CO_2\uparrow \qquad (2)$$

$$SrCO_3 + BaCO_3 + 0.5Fe_2O_3 + MoO_3 = SrBaFeMoO_{6-\delta} + 2CO_2\uparrow + (0.5+\delta)/2O_2\uparrow \qquad (3)$$

The second insignificant endothermic effect in the mixture of the starting reagents $SrCO_3 + BaCO_3 + 0.5Fe_2O_3 + MoO_3$ with a minimum at T = 927 K is practically not accompanied by a change in the mass of thebatchand, accordingly, the release of gaseous reaction products. According to the XRD data, no significant changes in the phase composition of the samples were detected.

In the temperature range corresponding to the third endothermic effect in the mixture of the starting reagents $SrCO_3 + BaCO_3 + 0.5Fe_2O_3 + MoO_3$ with a minimum at T = 1016 K, a sharp increase in the mass loss of the sample is observed. In this case, the amount of double perovskite increases faster than barium molybdate. Since at T> 1043 K the $SrFeO_3$ compound is absent in the mixture, and the concentration of the solid solution increases $SrBaFeMoO_{6-\delta}$, the endothermic effect is probably due to the following chemical reaction with the evolution of oxygen:

$$SrFeO_3 + BaMoO_4 = SrBaFeMoO_{6-\delta} + (1+\delta)/2O_2\uparrow \qquad (4)$$
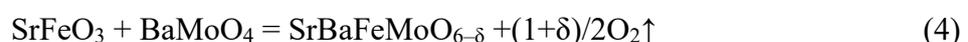
As it follows from the equation (4), the surface of strontium ferrite acts as the active centers for the appearance of nuclei of the new $SrBaFeMoO_{6-\delta}$phase, on which the dissociation process takes place, accompanied by the disappearance of $SrFeO_3$ during the growth of$SrBaFeMoO_{6-\delta}$. It was found that at room temperature the $SrFeO_3$, $SrCO_3$ and $BaCO_3$compounds practically disappear, and the $BaFeO_3$ phase appears. In this case, the amount of $BaMoO_4$ increases and reaches 54.5% of the total phase composition of the sample, which indicates the further course of chemical processes according to equation (1). Based on the foregoing, the formation of barium ferrite can be represented as follows:

$$SrFeO_3 + BaCO_3 = BaFeO_3 + SrO + CO_2 \uparrow \qquad (5)$$
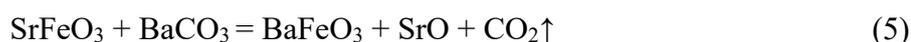
According to expression (5), the resulting strontium oxide most likely dissolves in the barium – strontium ferromolybdate matrix, since no other compounds are formed at temperatures $T \geq 1043$ K.

With a further increase in temperature, an insignificant fourth endothermic peak is observed with a minimum at $T = 1183$ K. In this case, a slight increase in the$SrBaFeMoO_{6-\delta}$phase and a decrease in the content of $BaMoO_4$ and $BaFeO_3$ phases were found in the quenched sample at $T = 1183$ K.

With an increase in temperature to $T = 1341$ K in the region where the fifth endothermic effect was recorded, the main reflection of the $BaFeO_3$ phase significantly decreases and its content in the sample is no more than 0.5%. In this case, the interface between the solid phases, where the mutual diffusion of chemical elements occurs, moves deep into the intermediate $BaMoO_4$ phase. In this case, decrease in the growth rate of $SrBaFeMoO_{6-\delta}$for the degree of conversion $\alpha \geq 70\%$ is due to an increase in the thickness of the interface between the solid phases. If the formed product layer has a low mobility of cations and anions, then the heterogeneous reaction from the adsorption – chemical transitions changes to the diffusion mode, which, in turn, is indicated by the above results. Samples heated to temperatures of 1418 K and 1473 K according to the XRD data are qualitatively identical and differ only in their quantitative composition. So, the composition of the sample heated to T=1418 K has a quantitative phase ratio: $SrBaFeMoO_{6-\delta}$- 75.8%, $BaMoO_4$ - 24.2%. With a further increase in temperature to 1473 K, the composition changes in the direction of increasing the content of double perovskite to $SrBaFeMoO_{6-\delta}$to 83.3% and decreasing barium molybdate to $BaMoO_4$ - 16.7%.

In the temperature range 1219–1473 K, the sample mass practically does not change, while the thermal effect is present and the sample remains multiphase. This indicates the difficulty of solid-state reactions with the formation of a solid solution of barium – strontium ferromolybdate.

When considering the dynamics of phase transformations, it was found that $BaCO_3$, $SrCO_3$, $BaMoO_4$, and $BaFeO_3$ are the main concomitant compounds during crystallization of the solid solution of $SrBaFeMoO_{6-\delta}$ double perovskite. When analysing the phase composition of the mixture, consisting of a mixture of the initial stoichiometric reagents: $SrCO_3 + BaCO_3 + 0.5Fe_2O_3 + MoO_3$, it was noticed that with increasing temperature, complex compounds $BaMoO_4$, $SrFeO_3$, and then $SrBaFeMoO_{6-\delta}$ appear almost simultaneously. This circumstance indicates that $BaMoO_4$ and $SrFeO_3$ compounds are structure-forming for solid solution of barium-strontium ferromolybdate. With a subsequent increase in temperature to 1043 K, the formation of new $BaFeO_3$ compound and the disappearance of $SrFeO_3$ were detected. In this case, the amount of the double perovskite increases faster than that of the barium molybdate.

Thus, at the initial stage of the interaction, the formed solid solution of barium-strontium ferromolybdate is enriched in iron and its composition during the reaction changes toward an increase in the molybdenum content. Since the complex oxides $BaMoO_4$ and $BaFeO_3$ are accompanying in the initial batchduring its annealing in almost the entire temperature range of the synthesis of $SrBaFeMoO_{6-\delta}$, it is important to work out the synthesis modes for a single-phase compound in which they dissolve as quickly as possible.

It is noted that with an increase in the heating rate, a decrease in the amplitude values $\max\alpha_{(\vartheta=const)}$) is observed for both $BaFeO_3$ and $BaMoO_4$ compounds. In this case, the values $\max\alpha=f(T)_{\vartheta=const}$ shift with an increase in the heating rate toward higher temperatures, which indicates the presence of kinetic difficulties during their crystallization (Fig. 4).

The presence of more significant kinetic difficulties in the formation of $BaFeO_3$ is indicated by temperature data at which the amplitude values of the derivative of the degree of conversion $(d\alpha/dt)_{max}$, depending on the heating rate, are 293–373 K higher than for the $BaMoO_4$ compound. The values of $(d\alpha/dt)_{max}$, depending on the heating rate, are 0.09–0.17 higher for $BaFeO_3$ than for $BaMoO_4$. This indicates a higher rate of chemical processes with the formation of barium ferrite than barium molybdate (Fig. 5).

When considering the decomposition rates of double oxides, it was noted that the largest minimum values $(d\alpha/dt)_{min}=-0.408$ are observed for $BaMoO_4$ at T=1331 K and $\vartheta=1.4$ deg/min, and for $BaFeO_3$ $(d\alpha/dt)_{min}=-0.375$ at T=1181 K and $\vartheta=1.4$ deg/min(Figs. 4, 5).

Based on the above data, it follows that in order to reduce the phase formation processes and increase the decomposition rate of intermediate products of the $BaFeO_3$ and $BaMoO_4$ reaction during crystallization of a barium – strontium ferromolybdate solid solution, the dynamics of phase transformations should be taken into account and combined heating conditions should be applied. So, in the low-temperature region, where the formation and growth of double oxides takes place, the

rate of temperature rise should be used at maximum, and in the high-temperature region, where dissolution of side compounds is observed, the rate of temperature rise should be low.

To establish the temperature range between the low-temperature and high-temperature synthesis regions, we consider the correlation dependences $\alpha=f(T)_{\vartheta=const}$ and $d\alpha/dt=f(T)_{\vartheta=const}$ for the $SrBaFeMoO_{6-\delta}$ compound with fixed heating rates of 0.7; 1.4; 2.5 deg / min(Fig. 6 a, b). According to the analysis of $\alpha=f(T)_{\vartheta=const}$, it was found that with an increase in the rate of temperature rise, the maximum $SrBaFeMoO_{6-\delta}$ conversion degree shifts toward higher temperatures and does not reach the maximum values of 100% at T = 1443 K. In this case, for temperatures above T≅1123 K, a slowdown in the growth rate of barium – strontium ferromolybdate is observed, reaching $min|d\alpha/dt=f(T)_{\vartheta=const}|$ at $1173 \leq T \leq 1243$ K and $0.7 \leq \vartheta \leq 2.5$ deg / min. It is noted that with a decrease in the heating rate, a shift in the degree of conversion is observed in $min|d\alpha/dt=f(T)_{\vartheta=const}|$ towards lower temperatures. With a further increase in the degree of conversion, the second maxima of the rate of change in the degree of conversion are observed. Moreover, the quantities $max|d\alpha/dt=f(T)_{\vartheta=const}|$ with an increase in the rate of temperature increase shift toward higher temperatures. The detected phased change in the growth rate of $SrBaFeMoO_{6-\delta}$ with the presence of two maxima of a function of the form $d\alpha/dt=f(T)_{\vartheta=const}$ and the presence of $min|d\alpha/dt=f(T)_{\vartheta=const}|$ in the temperature range 1123–1323 K, is most likely due to a decrease in the coefficients of chemical diffusion of reagents into the reaction zone. Since we are dealing with series-parallel reactions occurring simultaneously with the formation of various products and, above all, $BaFeO_3$ and $BaMoO_4$, the rate of the processes will be determined by a slower stage. Since the appearance of the $BaFeO_3$ compound with its subsequent decomposition was detected in this temperature range, it can be assumed that the volumetric diffusion of reagents through the layer of the reaction product is the limiting stage in the crystallization of $SrBaFeMoO_{6-\delta}$, which leads to an increase in the diffusion range of the reagents. Therefore, to increase the growth rate of $SrBaFeMoO_{6-\delta}$, it is necessary to reduce the diffusion path of the starting reagents to the reaction zone by eliminating intermediate reaction products during a crystallization of the barium – strontium ferromolybdate.

In this case, the revealed fact that $BaMoO_4$ reacts more quickly and precisely in the temperature range of the second maximum is most likely associated with the implementation of such a crystallization mechanism of $SrBaFeMoO_{6-\delta}$ at which kinetic difficulties are minimized and, accordingly, the growth rate of barium – strontium ferromolybdate increases. In this case, the rate of the entire transformation is determined by the rate of interaction of the reagents at the interface with $SrBaFeMoO_{6-\delta}$ grains.

Based on the above data, to obtain a single-phase solid solution, the combined heating modes were optimized:

- at the first stage, preliminary synthesis was performed at T = 1323 K and a heating rate of 2.5 deg / min for 15 hours. This temperature was chosen due to the fact that $\min|d\alpha/dt=f(T)_{\vartheta=const}|$ for compounds $BaFeO_3$ and $BaMoO_4$, as well as the second maximum $\max|d\alpha/dt=f(T)_{\vartheta=const}|$ for solid solution$SrBaFeMoO_{6-\delta}$were observed;

- at the second stage, to increase the reactivity of the mixture and the diffusion mobility of the reactants, the formed layer of the reaction product was crushed, homogenized, and the mixture was highly dispersed by thin vibro-grinding in alcohol for 2 hours;

- at the third stage, in order to decompose the intermediate phases $BaFeO_3$ and $BaMoO_4$ as quickly as possible and achieve the degree of conversion α = 100% for $SrBaFeMoO_{6-\delta}$, the synthesis was carried out at T = 1323 K and a heating rate of 2.5 deg/min for 5 hours, followed by heating to T = 1443 K at ϑ = 0.7 deg/min, since at this speed the maximum values of the degree of the barium – strontium ferromolybdate conversion were achieved.

As a result of using combined synthesis modes for annealing for 20 hours and T = 1443 K in vacuum of 10^{-5}Torr at the pressure of residual oxygen gas 10^{-8} Pa, it was possible to obtain a single-phase barium – strontium ferromolybdate compound with superstructural ordering of iron and molybdenum cations (Fig. 7).

Conclusions

Based on the study of the sequence of phase transformations during crystallization of the $SrBaFeMoO_{6-\delta}$solid solution and the establishment of correlations between the synthesis modes and the degree of phase transformations of the synthesized compounds, using a stoichiometric mixtureof simple oxides $SrCO_3$ + $BaCO_3$ + $0.5Fe_2O_3$ + MoO_3, the multistage crystallization of the compound is determined, which is due to the complexity of the phase transformations in concern with the sequentially parallel chemical reactions and low kinetics of phase formation.

It was found that in order to minimize the effect of intermediate reaction products at which kinetic difficulties are minimized and, accordingly, the growth rate of single-phase barium – strontium ferromolybdate increases with the presence of superstructural ordering of iron and molybdenum cations, it is necessary to use combined synthesis modes in it. An important factor for the formation of superstructural ordering of Fe / Mo cations in the compound is the selection of the synthesis medium in order to control oxygen deficiency.

As a result of using the combined synthesis modes for annealing for 20 hours and T = 1443 K in vacuum at the pressure of residual oxygen gas 10^{-8} Pa, it was possible to obtain a single-phase barium – strontium ferromolybdate compound with superstructural ordering of iron and molybdenum cations.

Acknowledgments

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Figure Captions

Figure 1. Temperature dependence of the derivative of thermal effects in a mixture ofSrCO$_3$ + BaCO$_3$ + 0.5Fe$_2$O$_3$ + MoO$_3$ powder annealed in vacuum at the pressure of residual oxygen gas 10^{-8} Pa at a heating rate of 1.4 deg/min

Figure 2. Temperature dependence of the change in the normalized mass of the mixture of oxides (SrCO$_3$ + BaCO$_3$ + 0.5Fe$_2$O$_3$ + MoO$_3$) powder (a) and its derivative (b), annealedin vacuum at the pressure of residual oxygen gas 10^{-8} Pa at a heating rate of 1.4 deg / min

Figure 3. X-ray diffraction patterns of samples synthesized in vacuum at the pressure of residual oxygen gas 10^{-8} Pa andat heating rate of 1.4 deg / min, obtained at differentannealing temperatures

Figure 4. Temperature dependences of the degree of conversion (a) and the growth rate (b) of the BaMoO$_4$ compound in samples synthesized at different heating rates

Figure 5. Temperature dependences of the degree of conversion (a) and growth rate (b) of the BaFeO$_3$ compound in samples synthesized at different heating rates

Figure 6. Dependence of the rate of change of the degree of conversion on the degree of conversion of the SrBaFeMoO$_{6-\delta}$ compound in a sample synthesized under polythermal conditions in vacuum at the pressure of residual oxygen gas 10^{-8} Pafrom a stoichiometric oxidesmixture (SrCO$_3$ + BaCO$_3$ + 0.5Fe$_2$O$_3$ + MoO$_3$)

Figure 7. X-ray diffraction pattern of the SrBaFeMoO$_{6-\delta}$ sample synthesized under combined annealing conditions and quenched at room temperature. The inset shows the microstructure and EDX analysis of the obtained sample (color online)
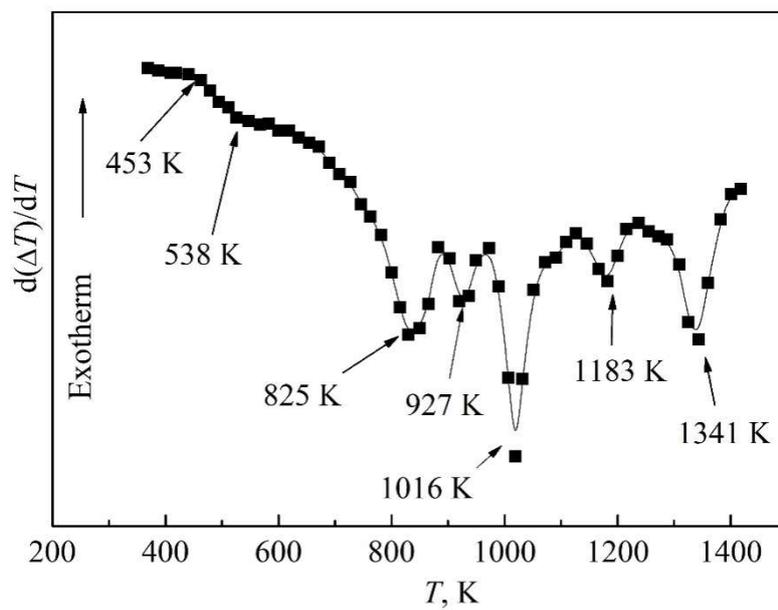
Figure 1
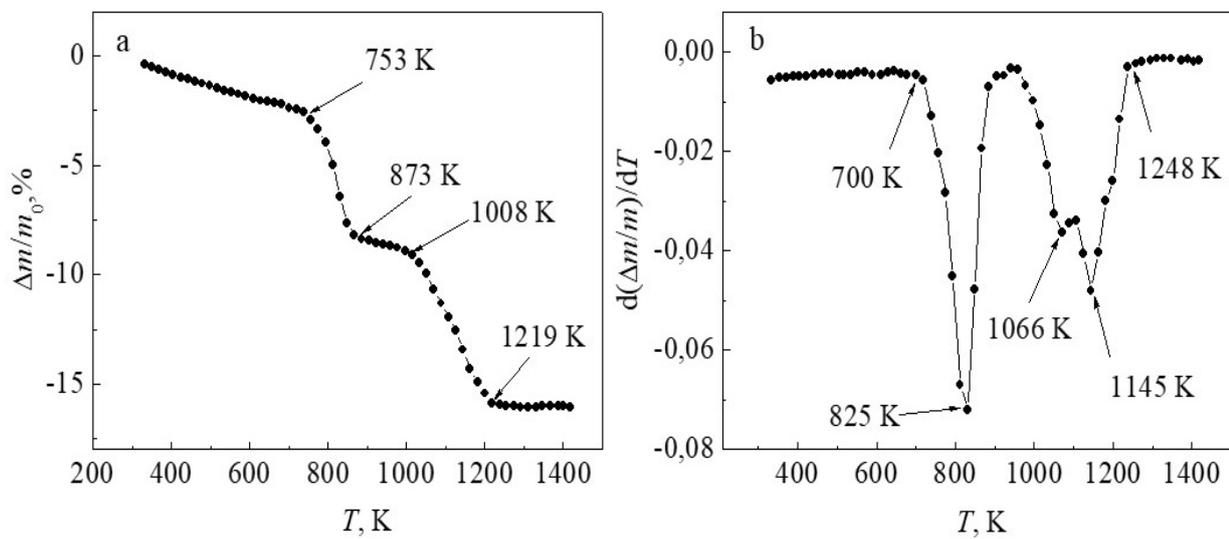
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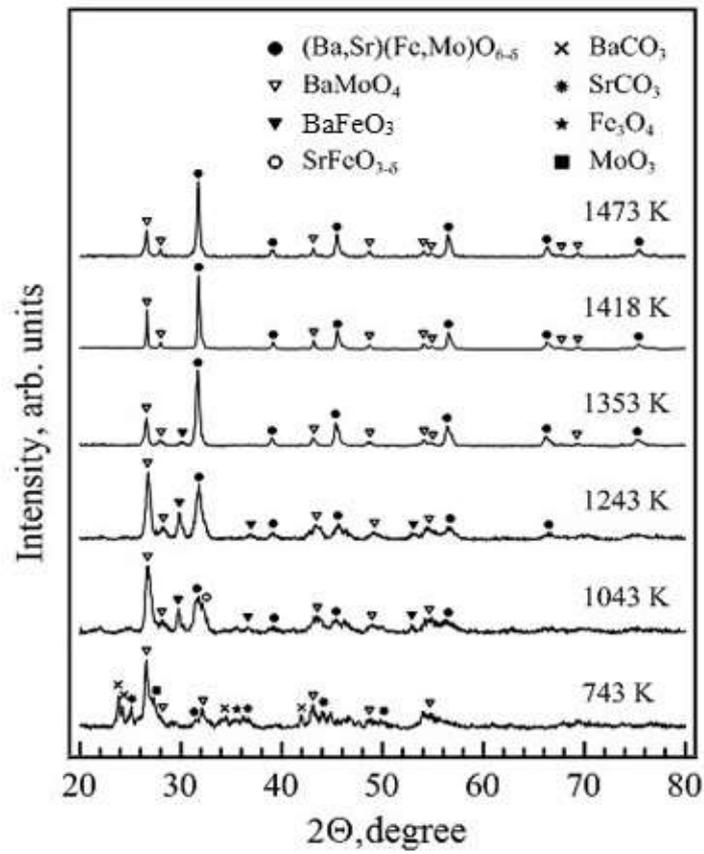
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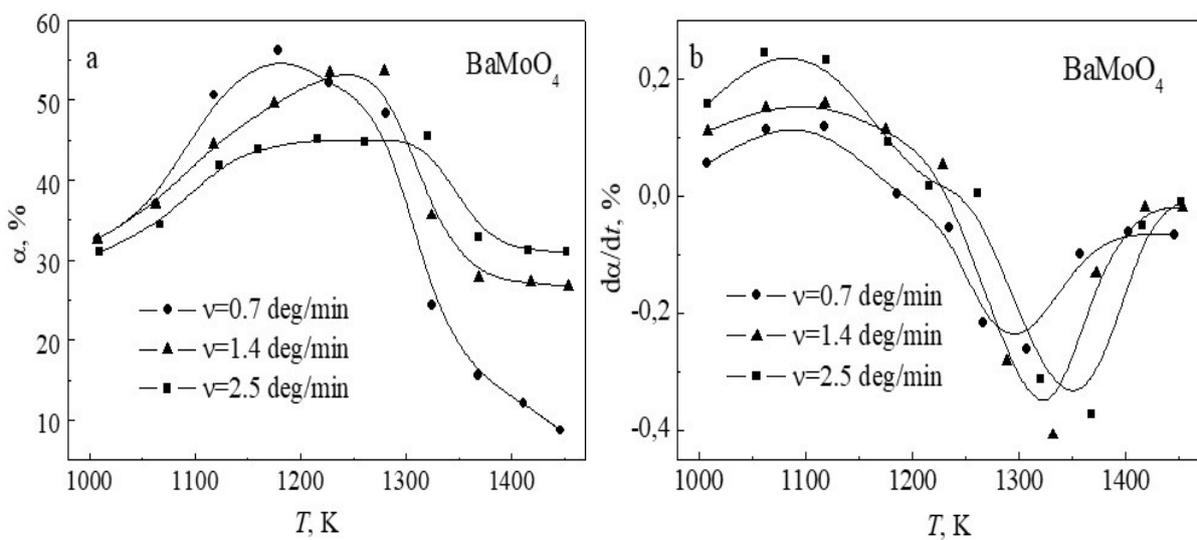
Figure 4
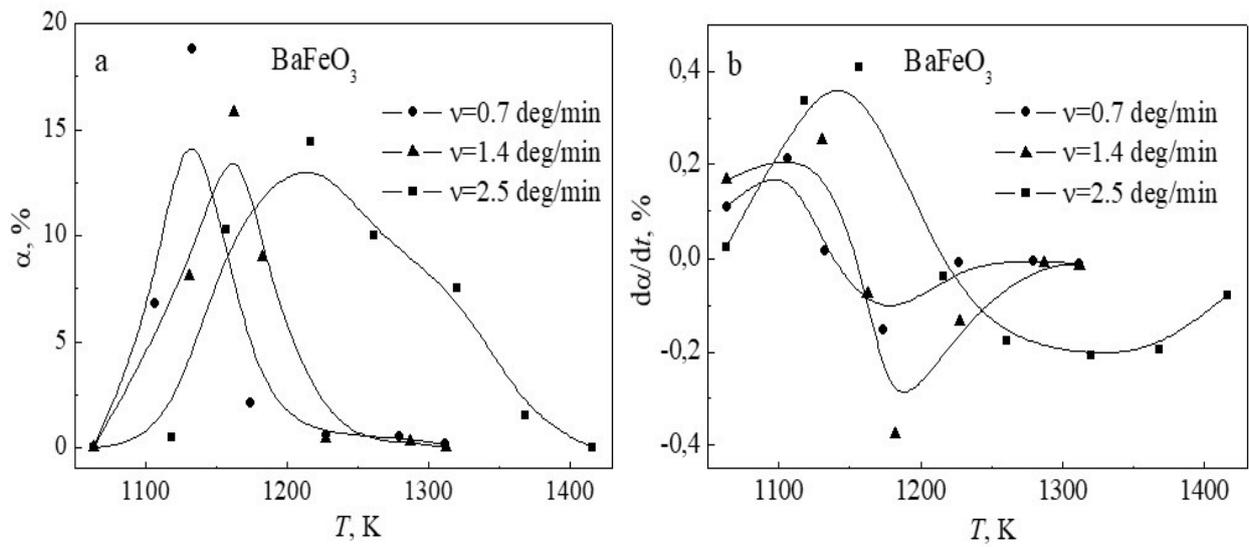
Figure 5

Figure 6

Figure 7