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Influence of Metal Component on Caking of Metal-Ceramic Composites.

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ABSTRACT

The article deals with participation of metallic aluminum in caking of metal-ceramic composites based on kaolinite-montmorillonite clays. The article shows that chemical modification of clays and thermomechanical modification of aluminum component along with simultaneous dispersion made it possible to increase aluminum content in the composite up to 20 %. Degree of chemical transformation of components at different thermal processing stages was assessed according to the X-ray phase analysis data. The beginning and termination of processes proceeding in mixtures during heat-treatment were defined. It was discovered that caking of composites under study is mostly influenced by the size of refractory phase particles and aluminum content in the mixture. Due to that the melt amount may be increased and the melt viscosity may be decreased. That is why more metal and, consequently, more liquid phase in the system make it possible to intensify the process at lower temperatures. It was determined that increase of metal component amount leads to some decrease of caking degree index in the series with T = const.

Keywords: caking, clays, aluminum, influence, properties

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INTRODUCTION

Production of high-strength materials is an up-and-coming approach to the solution of a wide range of problems in construction materials science. A way of solving this problem is to create composites in which the joint functioning of various constituents produces an effect equal to production of new material with properties different in amount and quality from the properties of each component [1 – 10].

Technologies of composite construction materials production develop in several directions [11 – 30]. Composites produced on the basis of metal and non-metal components are one of such promising composite construction materials.

We suggest a technology for production of ceramic composite materials with high content of metal component using the method of semi-dry pressing with subsequent drying and heat-treatment [31 – 36].

METHOD

Materials based on ceramic matrix and aluminum filler were prepared by way of mixing modified components with subsequent pressing, drying and heat-treatment. The quality of molding depended on humidity of the filler and pressure of pressing. When humidity decreased and pressure increased, stratification of the samples could be observed due to elastic deformations, emerging after releasing the pressure and its removal from the mould. Optimal results were received at humidity of 6 – 7 % and pressure 2 – 6 MPa.

Main part

One of the reasons of ceramic products break down at high temperatures is fissuring. Therefore introduction of metal component makes it possible to receive an interesting combination of the most important performance characteristics – high strength (including range of high temperatures), fatigue strength etc. Physicochemical processes and structural changes taking place in ceramic mixtures in the presence of metal during their production, modification and thermal processing affect the structure and properties of the produced material. Principal merits of such composite materials are connected with high operation temperatures (which is typical for ceramics) along with simultaneous significant increase of strength properties.

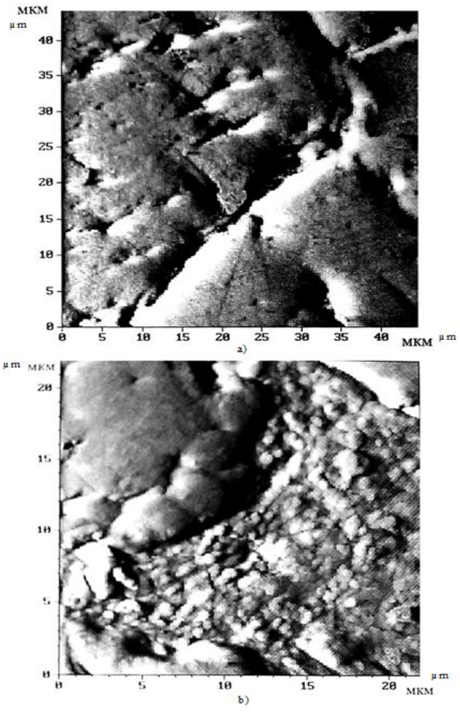


Figure 1 : Clay component particles surface a) Glukhovtsy kaolin; b) Krasnaya Yaruga clay.

Montmorillonite and kaolinite clays were used as a clay component in the process of new metal-ceramic composites development. Since the clay component particles surface has a defect structure (Figure 1), it is possible to complete their crystal structure with generating active centers, on which another material can be anchored afterwards.

Aluminum was chosen as a metal component, since it is relatively inexpensive and has good plasticity and yielding, and low melting temperature (660°C). The developed composite differs fundamentally from those known before, since physicochemical reactions proceeding between aluminum and non-metal component during the heat treatment of the composite make it possible to produce material with targeted performance characteristics.

One of the major problems which occurred in the process of composite material production with this method was to achieve compatibility of hydrophilic clays and hydrophobic metal. It was necessary to complete two tasks: provide strong connection between components and prevent aluminum melting at heat treatment stage, since the latter oxidizes in the process of caking almost in any environment, and further sample consolidation depends among other factors on phase transformations of newly formed oxide. Clay surface activation by mechanical processing, thermal and chemical modification was conducted for provision of clays compatibility with metal component and development of single-phase structure of the produced composite [37]. Surfactants in the amount of 0,1-1% were added into clay in order to reduce hydrophobicity. Chemical modification of clays by ions Al^{3+} from water solutions and thermomechanical modification of aluminum component with simultaneous dispersion made it possible to increase the content of aluminum in the composite to 20 %, avoiding at the same time metals melting and structure remolding by layers of unstable aluminum oxide, reducing the temperature of liquid phase formation by 80 - 120° C, and also displacing humps of viscosity curves to the area of lower temperatures. Introduction of small amounts of additives, which modify chemically both metal and clay, had a significant influence on interfacial energy reduction, and promoted a strong connection between chemically different particles through intermediate layer.

Physicochemical processes, which proceed during heating of aluminum and clay component to 700 °C, were studied with the help of differential thermal and X-ray phase analysis. Simultaneous complex record of curves DTA, DTG, TG and T helped to define the beginning and termination of processes proceeding in mixtures. Degree of chemical transformation of components at different thermal processing stages was assessed according to the X-ray phase analysis data.

Powders of aluminum, encapsulated in clay component, ingredients of which are given in the table 1, were prepared from Glukhovtsy kaolin, Krasnaya Yaruga clay and aluminum.

Table 1: Ingredients of powders of aluminum, encapsulated in clay component

Mixture No.	Content in mixture, % weight		
	Glukhovtsy kaolin	Krasnaya Yaruga clay	Aluminum
1	50	–	50
2	–	50	50

According to the results of differential thermal analysis of powders (pic. 2) curve DTA of mixture 1 has one endothermic effect with minimum at 525 and one exothermic effect with maximum at 690°C, and curve TG indicates weight reduction at a temperature of 525 °C, then small weight increase at a temperature of 690°C. It shows that within the temperature range 400-600 °C kaolinite dehydrates with the formation of metakaolinite, and within the temperature range 670-710 °C moderate oxidation of aluminum with the formation of aluminum oxide takes place.

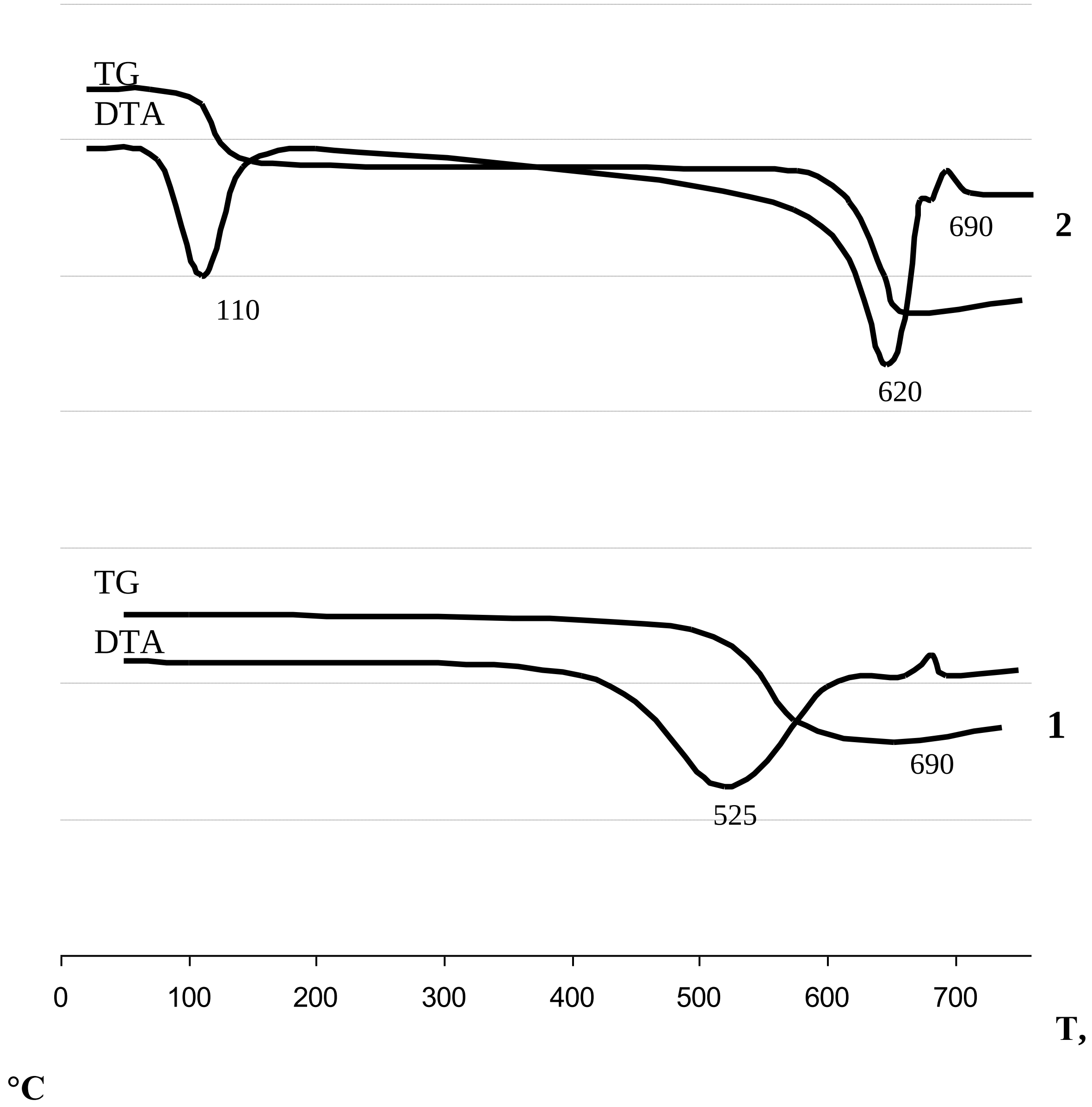


Figure 2: Differential thermal analysis of powders of aluminum, encapsulated in clay component

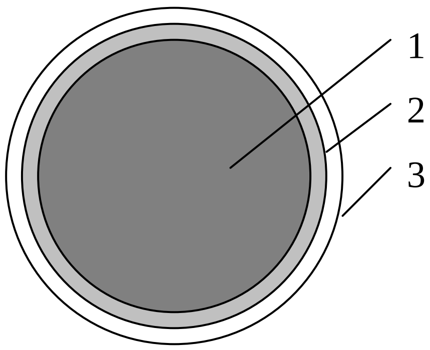
Figures 1 and 2 correspond to mixture indexes in table 1.

It is also confirmed by curves of X-ray phase analysis, which fix reflexes corresponding to phases of metakaolinite (at 1.55, 2.20, 2.66, 3.52, 3.97 A), quartz (at 3.63 A), aluminum (at 0.87, 0.92, 2.01 A) and aluminum oxide (at 1.92, 2.39, 2. 84, 4.24 A).

Curve DTA (pic. 2) of mixture 2 has two endothermic effects with minimums at 110, 620 and one exothermic effect at 690°C, and curve TG indicates weight reduction at temperatures of 110 and 620 °C, then small weight increase at a temperature of 690°C. Therefore, one can assume that in the presence of first two endothermic effects clay dehydration takes place, and the last effect correspond to oxidation of aluminum.

It is also confirmed by curves of X-ray phase analysis, which fix reflexes corresponding to phases of metakaolinite (at 2.20, 3.52, 3.97 A), aluminosilicate spinel of $Al_2Si_4O_{11}$ content (at 1.50, 4.42, 7.30 A), quartz (at 2.12, 3.39, 3.63, A), aluminum (at 0.87, 0.92, 2.01 A) and aluminum oxide (at 1.92, 2.39, 2. 84, 4.24 A).

Therefore, structure of the produced aluminum powder, encapsulated in clay component, is represented by aluminum, covered with films of aluminum oxide and dehydrated clay component (pic. 3), which is represented by metakaolinite and dehydrated clay for mixtures 1 and 2 (table 1) respectively.

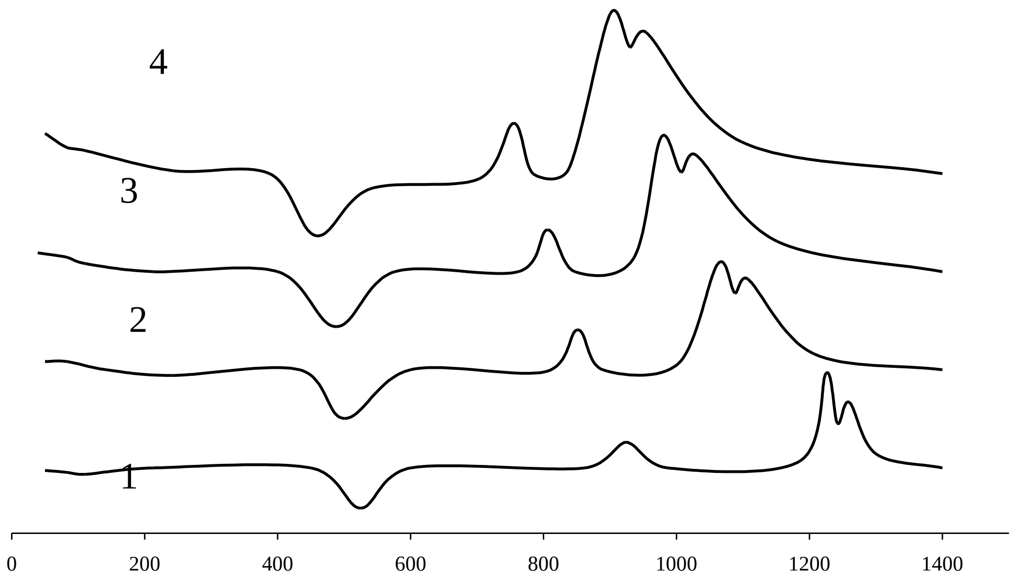


Pic 3: Structure of powder of aluminum, encapsulated in clay component: 1 – aluminum; 2 – aluminum oxide film; 3 - dehydrated clayish component film.

Thermomechanical dispersion of aluminum in clay component made it possible to produce homogeneous structure of composite during further caking.

The importance of study of processes, proceeding during caking of metal-ceramic composite, is defined, on the one hand, by the necessity of producing of chemical bonds between clay component and metallic aluminum and, on the other hand, by the necessity of assessment of composite technological strength.

Main signs of the process of caking of metal-ceramic composite are simultaneous increase of density and mechanical strength of the material, and also change of specific properties of the material depending on the temperature, such as water absorption, porosity, apparent density and heat setting.



a) **T, °C**

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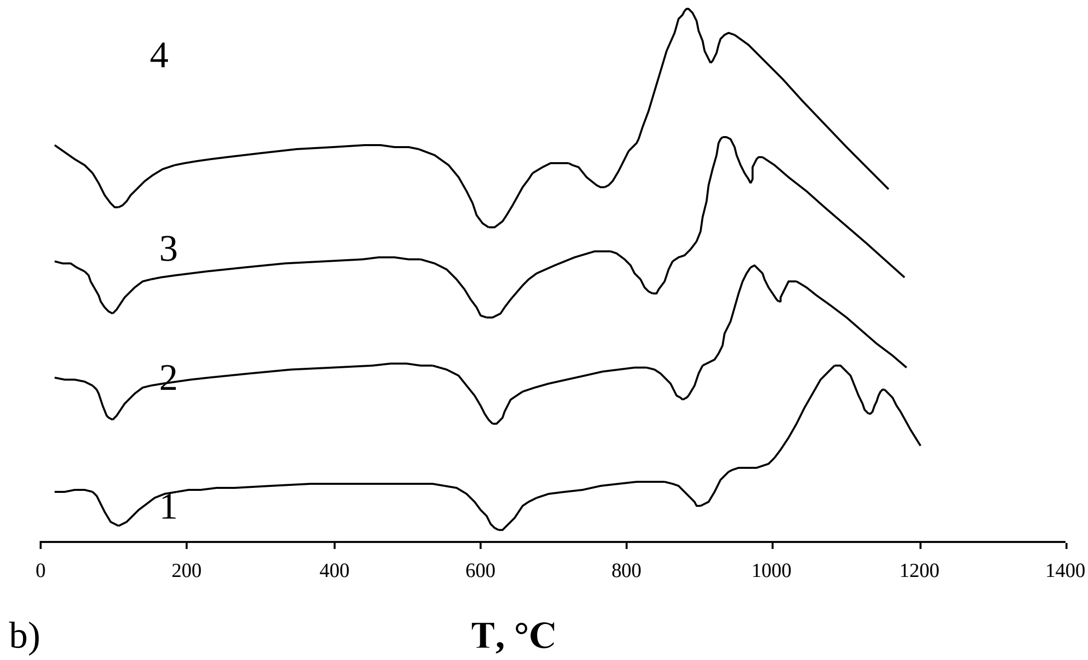


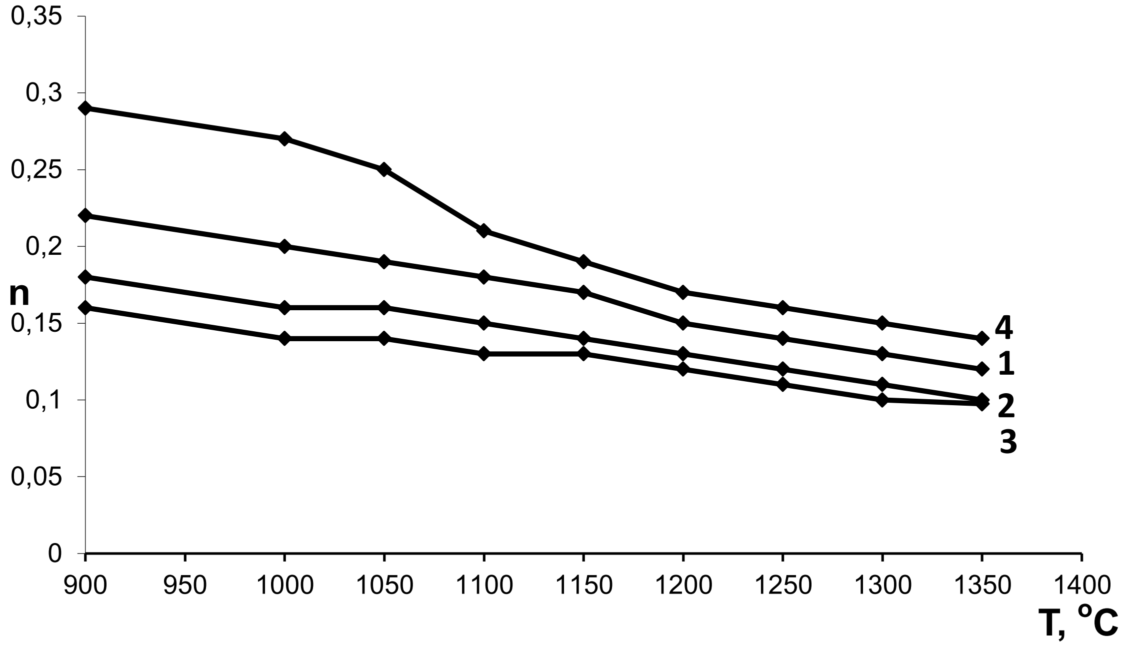
Pic 4: Differential thermal analysis of composites on the basis of kaolinite (a) and montmorillonite (b) clays with the content of aluminum filler, %: 1) – 0; 2) – 5; 3) – 10; 4) – 20

During heat treatment of modified ceramic mixtures containing aluminum filler complex physicochemical transformations take place. As a result of thermal breakdown of minerals and partial oxidation of aluminum, free oxides appear. At the same time intermediate finely divided phase appears - solid solution. Within the temperature range from 600°C and higher part of aluminum oxidizes with the formation of aluminum oxide, which is involved in structure formation of composite (by means of mutual dissolution and following chemical reactions). Physical adherence occurs between remaining unoxidized aluminum and modified matrix surface. Within the temperature range from 700 to 1350°C the process of caking proceeds with the involvement of liquid phase reacting with the solid. Molten aluminum in the mixture contributes to melt amount increase and melt viscosity decrease, as well as acceleration of lattice diffusion and crystal processes. Formation of aluminosilicates proceeds actively even at a temperature of 900 °C, that is clear while comparing the curves of differential thermal analysis (pic. 4).

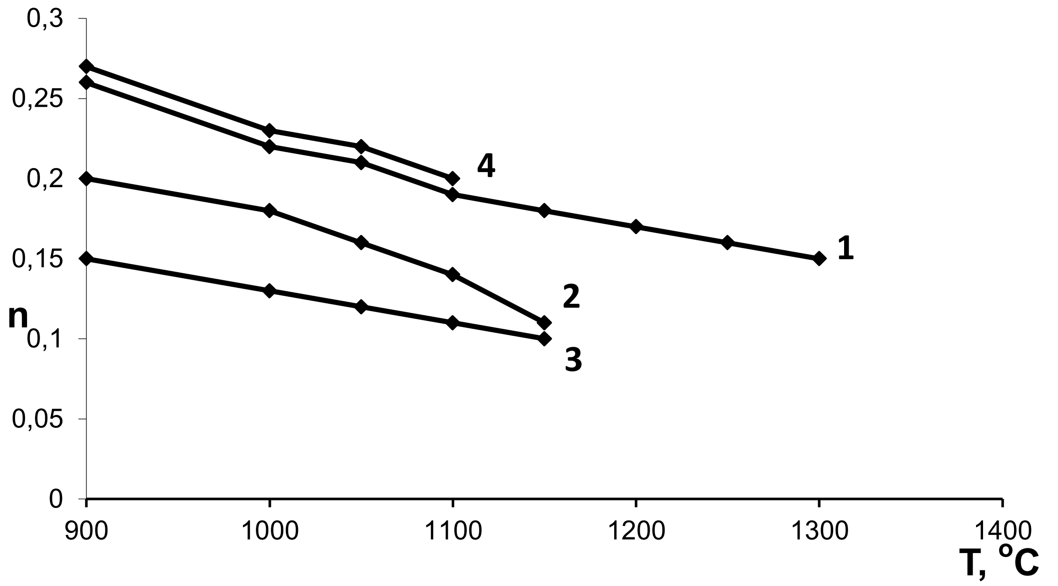
In the process of caking of metal-ceramic composite a number of physical transformations occur, which can include: lattice diffusion, plastic flow, surface diffusion and evaporation-condensation. This is the way surface diffusion, evaporation and condensation contribute to pore spheroidizing, increase of contacts between particles that leads to material strengthening.

Caking is a three-stage process, which includes rearrangement of particles of solid phase, dissolution-sedimentation and formation of a solid frame. These processes are not usually detached in time and often proceed simultaneously, influencing each other. Dissolution-sedimentation can be controlled both by chemical reaction kinetics at the boundary of the interface of solid and liquid phase and by diffusion of reaction products through the boundary layer.

Basic mechanism of metal-ceramic materials caking is the process of dissolution-sedimentation, which is controlled by diffusion. It is confirmed by comparison of caking degree indexes and data, which is given for this process in the literature. Mutual dissolution of solid phase in liquid one is confirmed by the fact that thick material can be produced on the basis of composites, containing up to 20% of aluminum.



a)



b)

Pic 5: Dependence of caking degree index on the temperature of heat treatment of metal-ceramic binder on the basis of kaolinite (a) and montmorillonite (b) clays with the content of aluminum filler, %: 1) – 0; 2) – 10; 3) – 20; 4) – 30%

One of the factors, which have a considerable influence on the caking process, is the amount of eutectic melt in the material during heat treatment. Decrease of caking index with temperature rise is observed in materials containing from 5 to 20% of aluminum (pic. 5), that is obviously because of motive force reduction.

CONCLUSIONS

All caking indexes fall within the limits, typical for the process of dissolution-sedimentation, which is controlled by diffusion, consequently, it is a basic process, limiting caking of composite speed. Increase of metal component amount leads to some decrease of caking degree index in the rows with *T* = const. It shows that more aluminum and, consequently, more liquid phase in the system make it possible to intensify the process at lower temperatures. At the same time thickening and setting almost terminate around the temperature of 1250 °C for composites on the basis of kaolinite clays and around 1050 °C for composites, which are produced from montmorillonite clays. Caking degree indexes for observed temperature range shows the similarity of mechanisms of caking of materials containing 5-20% of metal component.

RESUME

Thus, the research shows that caking of composites under study is mostly influenced by the size of refractory phase particles and aluminum content in the mixture. Due to that the melt amount may be increased and the melt viscosity may be decreased.

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