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Investigation of Properties of Composite Ceramics

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Recycling of fibrous mineral wool waste represents a rather serious problem. The results of research presented in the study show that the suggested composite ceramic system out of low melting illite clay and fibrous mineral wool waste is a promising way for solving the problem of mineral wool waste utilization. The performed researches into the properties of the system under investigation showed that upon introduction of fibrous mineral wool waste into the composition of low melting clay, the temperature expansion interval decreased from 810 °C to 775 °C. It was found out that the free oxides CaO and MgO forming during decarbonization of clay, reacted actively with clay minerals and that crystallization of reaction products resulted in anorthite and diopside. The experimental investigations of fusibility showed that upon introduction of 30 % fibrous mineral wool waste into the composition of low melting clay, the melting start temperature of composite mixture increased from 1115 °C to 1135 °C.

Keywords: mineral wool waste, low melting illite clay, composite ceramics.

INTRODUCTION

In recent years a rather great attention was paid to utilization and recycling of waste with more than 40 % SiO_2 content. The data provided in the literature show that such waste may be introduced as an additive into the composition of various clays, as well as of concretes and mortars [1 – 8].

The sources of literature tackle at large the silicate systems SiO_2-CaO-MgO-Na_2O where content of SiO_2 makes more than 70 %. Furthermore, the waste of such glasses contains a great deal of Na_2O [1 – 5]. There was investigated the impact of glass waste from the system SiO_2-CaO-MgO-Na_2O on the process of sintering and the physical-mechanical parameters of the ceramic paste from low melting illite clay and there was explained the mechanism of sintering of ceramic paste [5]. The results of investigation show that along with melting of glass additive contained in the ceramic paste, diffusion of ions Na^+ takes place in the zone of contact between glass grind grain and clay. It was revealed that ions Na^+ react actively with clay minerals and correct melting properties of the system under investigation. Therefore, after introduction of glass grind of system SiO_2-CaO-MgO-Na_2O into the composition of low melting illite clay, formation of liquid phase occurs within lower interval of temperatures [5]. There was investigated an alumosilicate system, into the composition of which 30 % glass grind was introduced [4]. It appeared that during sintering of such a system, the glass-ceramic matrix was formed. The radiographic results showed that while sintering of glass-ceramic matrix, devitrite, cristobalite and volastonite crystallized [4].

The research was performed to the aim of elucidating the impact of glasses of system SiO_2-CaO-MgO-Na_2O and SiO_2-Ba_2O_5-Na_2O on clay containing a rather high percentage of kaolin (50 %) [9]. For research, grinds of glasses of typical composition, as well as those of boron-silicate glasses were used. Upon introduction of 30 % waste of such glass into the composition of clay, there was obtained sintered ceramics, which compression strength increased from 30 % to 40 % and temperature of sintering decreased to 1100 °C.

A possibility to utilize glass waste in the production of porcelain articles was considered [1 – 3]. The results of research showed that upon introduction of 10 % glass waste, the temperature of sintering decreased and the strength values increased [3].

Another type of waste is a waste from the production of mineral wool with considerably lower SiO_2 content [10 – 14]. It is a silicate system SiO_2-CaO-MgO-Al_2O_3, which consists of mineral wool fibre and small pieces of not crushed melt [13, 14]. The researches for solving the problem related to utilization of mineral wool waste are directed to development and improvement of technologies for briquetting of this waste and the results of these researches are patented [10 – 12]. In recent years the researches commenced as to peculiarities of the composite system made of low melting illite clays and fibrous mineral wool waste [6, 13, 14]. The composite system out of low melting illite clay, cement dust, dolomite and fibrous mineral wool waste was investigated [6]. It was determined that for prevention of formation of free oxides CaO and MgO during heating, it is necessary to decrease the content of dolomite. The research results tell us that a promising field for utilization of fibrous mineral wool waste is the production of ceramic composite articles, however, the larger researches should be performed first.

In the sources of literature the impact of silicate glasses of system SiO_2-CaO-MgO-Na_2O on properties of sintered ceramics is tackled at large. Nevertheless, the composite ceramics made of low melting illite clay and fibrous mineral wool waste of system SiO_2-CaO-MgO-Al_2O_3 lacks investigations. Therefore, large and exhaustive researches of this system are required to be able to utilize this waste in the production of composite ceramic articles. The aim of this study is to investigate properties of composite ceramic paste, as well as peculiarities of its components – illite clay and fibrous mineral wool waste.

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Table 1. Chemical composition of raw materials

Raw materials	Chemical composition, %								
	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	K_2O	Na_2O	TiO_2	Loss on ignition
Clay A	46.67	17.93	6.70	8.90	4.26	3.07	0.15	–	12.25
MW waste	42.13	18.30	5.81	16.15	13.94	0.27	1.46	2.02	1.40

TEST METHODS

For testing there was used low melting illite clay from the devonian finding-place (clay A) and mineral wool waste (waste MW). The chemical composition of raw materials is provided in Table 1. Clay A and fibrous mineral wool used for testing was dried at 105 °C ±5 °C in the not ventilated drying room. The organic content of mineral wool waste was 0.96 % and moisture – 15.6 %.

The study dealt with the modeling system of low melting illite clay (clay A) – fibrous mineral wool waste (MW waste). Three series of modeling mold mixtures were prepared for testing: composite modeling mold mixtures Series CMW20 (clay A 80 % and MW waste 20 %) and Series CMW30 (clay A 70 % and MW waste 30 %), and for comparative testing – modeling mold mixture Series CA out of low melting illite clay. The dimensions of specimens prepared for testing were (70×70×70) mm.

The heated specimens were analyzed rentgenographically by a diffractometer DRON-7, the phase composition was identified basing on the file of reference data ASTM. The thermographical tests were performed by a derivatograph F. Paulik, I. Paulik and I. Erdei Q-1500, temperature up to 1000 °C, testing medium – air, temperature raise rate 10 °C/min. The dilatometric tests were performed by a dilatometer Linseis L76, temperature raise rate 2 °C/min, temperature up to 1000 °C. The ultrasound tests of heated specimens were performed by an apparatus UK-14P. The experimental tests of fusibility for composite modeling mold mixtures were performed by a high temperature microscope MHO-2. The initial temperature of testing was 20 °C, temperature raise rate 5 °C/min. During testing the specific changes in shape of specimens were fixed.

RESULTS AND DISCUSSION

The low melting illite clay used for testing is characterized by minerals contained in the composition of clay, such as illite (hydromica), chlorite, quartz, feldspar, calcite, dolomite. The not heated fibrous mineral wool waste is characterized by diopside content.

The modeling mold mixture CA and composite modeling mold mixture CMW20 were tested dilatometrically. The analysis of test data showed that the property of deformation of modeling mold mixture CA might be described as an expansion obtained while specimen heating within interval of temperatures from 100 °C to 910 °C and as a contraction, which starts at 950 °C (Fig. 1). It was determined that the expansion temperature interval of specimen makes 810 °C. The comparative analysis of dilatometric data for modeling mold mixtures CA and CMW20 showed that the introduced fibrous mineral wool waste decreased the expansion temperature interval from 810 °C to 775 °C (Fig. 1).

The composite modeling system of clay A and mineral wool waste was tested by differential thermal analysis and X-ray diffraction (XRD), the same was done with the modeling mold mixture CMW30 (Fig. 2 and Fig. 3). Preparation of specimens for XRD tests included heating at temperatures 850 °C, 900 °C and 1000 °C.

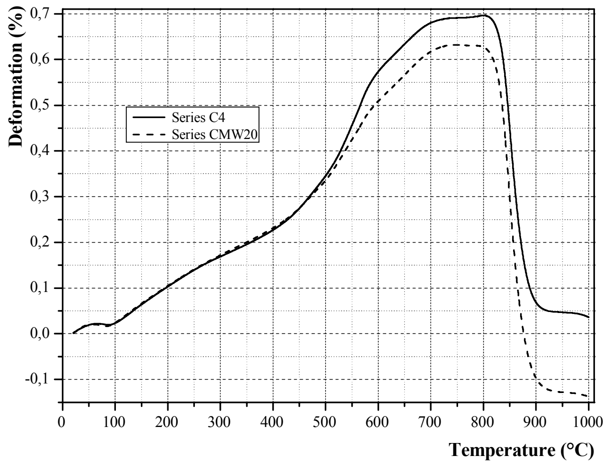

Fig. 1. Dilatometric data for modeling mold mixtures CA and CMW20

The results of thermographic tests show that in the course of heating the composite modeling mold mixture CMW30 underwent changes, which might be characterized by complicated processes of dehydration and decarbonization. From the DTA curve analysis it is obvious that the endothermic effect measured at temperature of 600 °C describes the end of dehydration of hydromicas contained in clay (Fig. 2). From XRD data of composite modeling mold mixture CMW30 heated at 850 °C we can see that formation of alumosilicate compounds of calcium and magnesium takes place during crystallization of clay minerals (Fig. 3).

The test results pointed to the fact that the endothermic effect fixed at 850 °C depicts the splitting of carbonates ($CaCO_3$ and $MgCO_3$) contained in clay and the formation of free oxides of calcium and magnesium (CaO and MgO) (Fig. 2).

According to the comparative analysis of XRD and thermographic data, the free oxides CaO and MgO react actively with the alumosilicate compounds contained in clay. It was determined that the endothermic effect fixed at temperature of 890 °C describes the reaction of free oxide CaO with minerals contained in clay. The XRD of specimens heated at 850 °C and 900 °C evidenced the crystallization of gehlenite and anorthite (Fig. 3). The comparative analysis of gehlenite peak intensity showed proofs that along with raise in temperature of heating, the intensity of peaks was decreasing. No gehlenite peaks were observed in the specimen heated at 1000 °C. It is clear from the analysis of provided test results that gehlenite, as a product of reaction of free oxide CaO and alumosilicate compounds contained in clay, does not build a stable crystalline structure. Furthermore, the content of this mineral is rather small. Upon examination of relationship of anorthite peak intensity and specimen heating temperature, one can see that gehlenite most likely transformed into anorthite (Fig. 3).

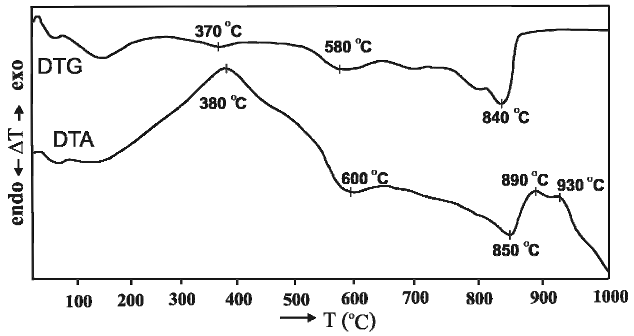

Fig. 2. Thermographic data for composite modeling mold mixture CMW30

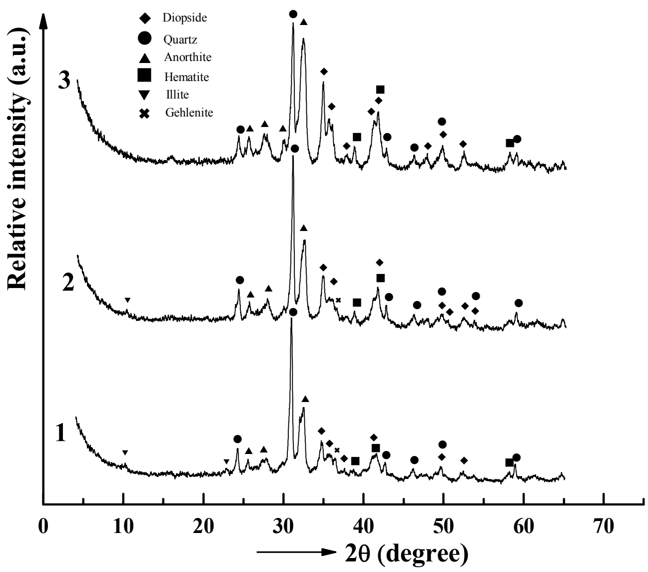

Fig. 3. XRD data for composite modeling mold mixture CMW30: 1 – heated at 850 °C; 2 – heated at 900 °C; 3 – heated at 1000 °C

The data of XRD tests show that the exothermic effect fixed at 930 °C characterizes the reaction of free oxide MgO, which formed during carbonate splitting, with clay-contained minerals. It was found out that with crystallization of products of this reaction, diopside crystallized (Fig. 2 and Fig. 3). By analysis of XRD data, it is obvious that the formation of diopside and its crystallization at 930 °C defines not only the processes going on in clay paste, but also the crystallization of this mineral in fibrous mineral wool waste. As to the results of ultrasound testing, the values of ultrasound spreading rate differed only very slightly from one another in the composite modeling mold mixtures CMW20 and CMW30 heated within interval of temperatures from 800 °C to 1000 °C (Fig. 4). The comparative analysis of ultrasound tests pointed to a decrease of ultrasound spreading rate in the specimen, due to decarbonization of clay within 850 °C – 900 °C and due to formation and crystallization products from the reaction of free oxides CaO and MgO with clay minerals, in case of the modeling mold mixture CA. True, that after heating of specimen at 850 °C, the ultrasound spreading rate decreased from 2761 m/s to 2209 m/s. It is also true that fibrous mineral wool waste compensates the decrease of ultrasound rate within this interval of temperatures (Fig. 4).

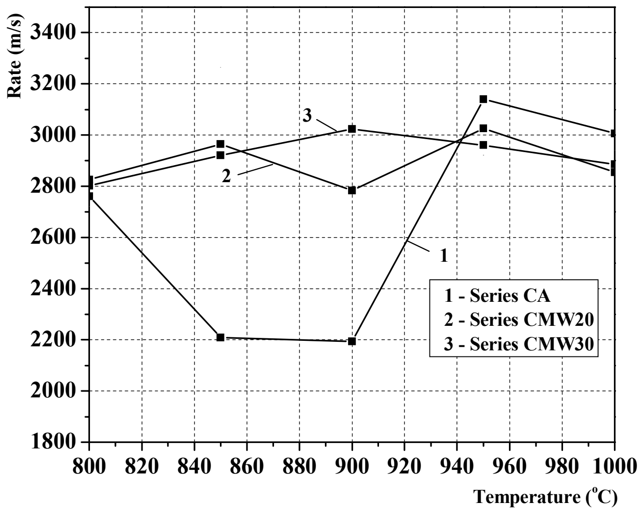

Fig. 4. Ultrasound data for modeling mold mixtures CA, CMW20 and CMW30

The experimental analysis of melting properties was performed in the composite modeling mold mixture CMW30 by a high temperature microscope MHO-2. The modeling mold mixture CA underwent comparative testing. During tests the changes occurring in the shape of tested specimens were fixed.

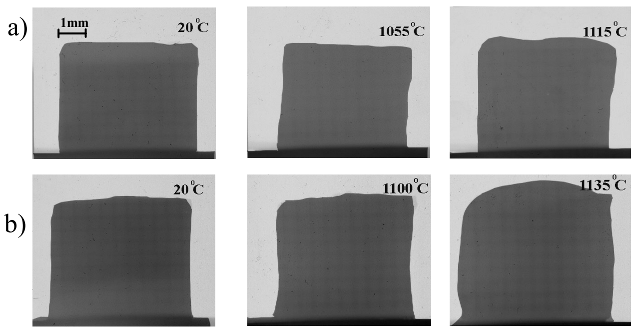

Fig. 5. Data of experimental test of melting properties for modeling mold mixtures CA (a) and CMW30 (b)

The resulted melting parameters of modeling mold mixtures are provided in Fig. 5. The analysis of test results proves that the specimen CA retains its regular shape within interval of 20 °C to 1055 °C. However, a considerable contraction of specimen may be observed and this contraction gives a signal of an active process where the liquid phase is appearing (Fig. 5, a). It was found that the tested specimen started melting at temperature of 1115 °C and that the process of melting may be described by specific changes in the shape of tested specimen.

By examining melting properties for the modeling mold mixture CMW30, it was revealed that within interval of temperatures from 20 °C to 1100 °C the tested specimen retained its regular shape (Fig. 5, b). The start of melting in the specimen was observed at 1135 °C. The comparative analysis of test data showed that upon introduction of 30 % fibrous mineral wool waste into the composition of clay, the melting start temperature increased from 1115 °C to 1135 °C.

CONCLUSIONS

The tests with composite modeling mold mixture containing fibrous mineral wool waste and low melting illite clay showed that upon introduction of 20 % fibrous mineral wool waste, the temperature expansion interval decreased from 810 °C down to 775 °C. According to thermographic and XRD data, the crystallization of diopside at 930 °C characterizes not only the processes going on in clay paste, but also the crystallization of this mineral in fibrous mineral wool waste. The analysis of ultrasound test results demonstrated that the processes of clay decarbonization in the modeling mold mixture of clay A within interval of temperatures from 850 °C to 900 °C, as well as the formation of reaction products from the reaction of free oxides CaO and MgO with clay minerals and their crystallization reduces the ultrasound spreading rate in the specimen. It was found out that in the composite modeling mold mixture the fibrous mineral wool waste compensates the decrease of ultrasound rate, therefore, within interval of temperatures from 800 °C to 950 °C, the ultrasound spreading rate changes but very slightly. The experimental tests of fusibility showed that the temperature of melting start in the composite modeling mold mixture increases from 1115 °C to 1135 °C.

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