

INVESTIGATION OF THE PHASE FORMATION OF PORCELAIN-FAIENCE COMPOSITE MATERIALS

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ABSTRACT.

The article presents the results of a radiographic and petrographic study of experimental samples from composite ceramic sanatoriums. It is shown that samples burned at an optimal temperature contain mainly crystalline phases - a granular mullite, needle mullite, quartz and cristobalite.

KEYWORDS: faience, porcelain, firing, properties, temperature, mullite, quartz, cristobalitis.

INTRODUCTION. In order to strengthen the economy of the Republic of Uzbekistan, it is necessary to develop sectors of the national economy, including the construction industry, energy, electrical engineering, mechanical engineering, oil refining, chemical, metallurgical industries, etc. The development of these industries is closely related to the use of various ceramic materials. Their development and rational use in the national economy will allow not only to strengthen the economic base, but also to enter the world market in many types of products. One of the important places in the infrastructure of the national economy of Uzbekistan is occupied by the composite materials industry. Currently, the production of composite materials, including the production of composite building materials, is experiencing a shortage of raw materials. In this aspect, many studies are devoted to the problem of finding new deposits of raw materials [1-4].

In recent years, rocks such as perlite, obsidian, and others, rich in alkaline oxides, have found wide application in the ceramic industry [3-4].

For the further development of the republic's economy, it is required to develop and create high-quality composite building ceramic materials and products. At present, the population's demand for composite building materials is also not fully met.

In this regard, one of the most important tasks is the scientific development and introduction into the practice of import-substituting masses of a new composition of composite building ceramic materials, including sanitary-technical porcelain and faience products, which have high quality indicators and economic efficiency.

OBJECTS AND RESEARCH METHODS. The objects of research were the developed masses of sanstroy-faience materials based on local mineral raw materials (glauconite clay, kaolin, pegmatite,

bentonite) and production waste (silica-containing). In this regard, glauconite and bentonite clays seem to be the most promising, however, the diversity of their chemical and mineralogical composition creates the need for individual studies in each specific case. To solve this problem, first of all, we carried out an X-ray phase analysis of experimental samples. The prototypes were fired at temperatures of 1000, 1100, 1200 and 1250 ° C according to the mode developed by us. These samples were subjected to grinding and then X-ray phase analysis. To decipher the X-ray diffraction patterns, we used the Giller tables of interplanar distances [5], identification was carried out according to the reference books of L.N. Mirkin et al. [6] and V.N. Mikheev [7].

RESEARCH RESULTS AND DISCUSSION. As a result of the physicochemical processes occurring during firing, new crystalline phases are formed, due to which the materials under study acquire the necessary physical and technical properties. By regulating the firing modes, it is necessary to create the necessary conditions for the full-fledged course of physicochemical processes and the formation of new crystalline phases that favor an increase in the physical and technical properties of the material obtained. In this regard, it is especially important to establish the phase composition of the materials under study.

Figures 1, 2 show X-ray diffraction patterns of prototypes fired at different temperatures from the most optimal mass compositions.

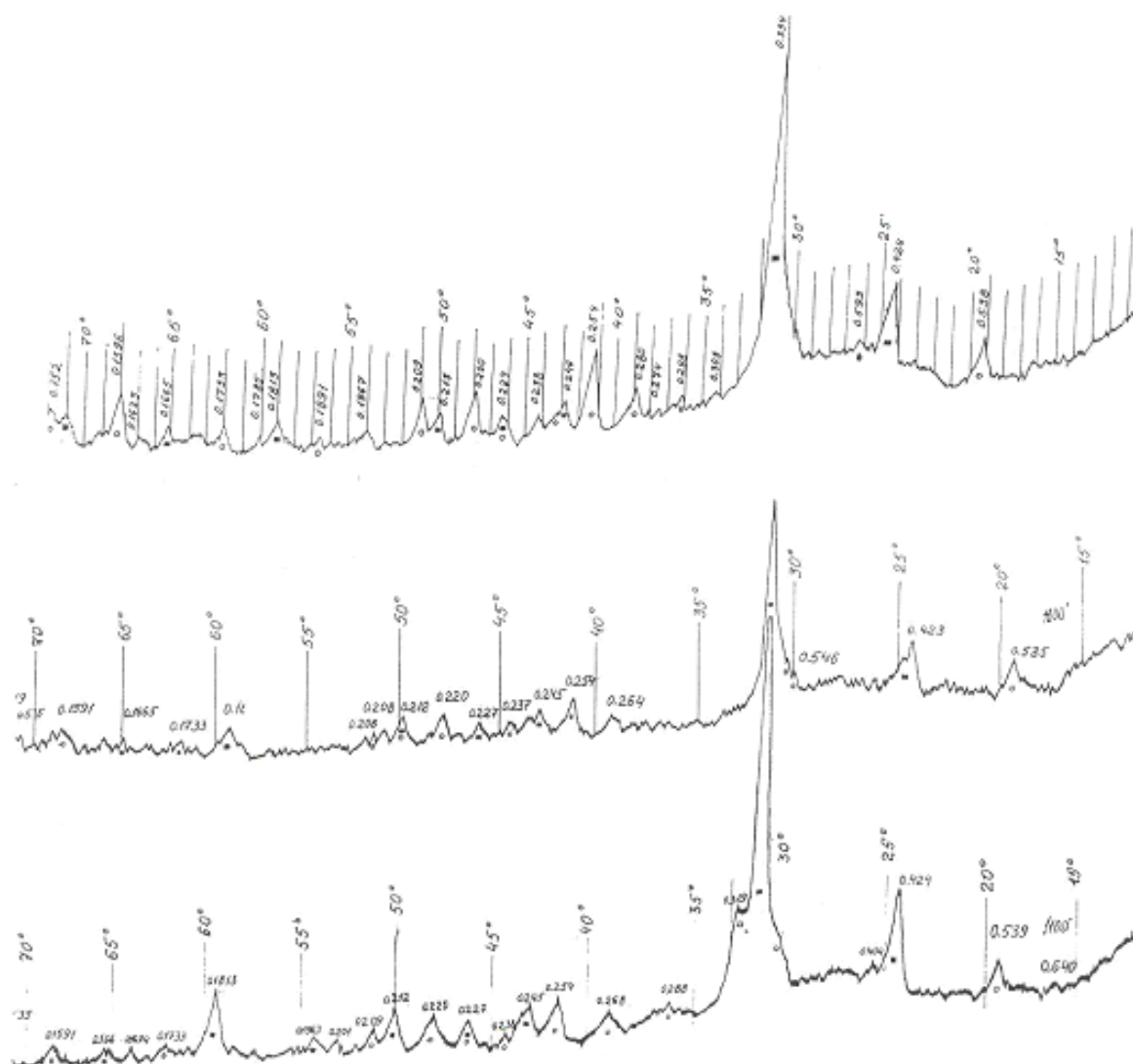


Figure 1. X-ray diffraction patterns of samples from mass C-1, fired at different temperatures, 0C: a) 1100; b) 1200; c) 1250

In samples from the composition of mass C-1, fired at 1000 ° C, the structure changes, contains β -quartz with interplanar distances $d/n = 0.423; 0.334; 0.244; 0.223; 0.212; 0.197; 0.181; 0.166; 0.157$ nm, feldspar with interplanar spacing $d/n = 0.318; 0.294; 0.254$ nm and as a product of dehydration of clay-forming minerals metakaolinite with interplanar distances $d/n = 0.403; 0.375; 0.367; 0.348$ nm, etc.

In specimens fired at 1100 ° C (Fig. 1a), the crystal lattice of feldspar is completely destroyed, it contains β -quartz with interplanar distances $d/n = 0.242; 0.334; 0.245; 0.227; 0.212; 0.196; 0.181; 0.166$ nm. Along with it, mullite grains are formed with characteristic interplanar distances $d/n = 0.539; 0.288; 0.268; 0.254; 0.238; 0.227; 0.220; 0.209; 0.159$ nm, as well as isomorphic feldspar with $d/n = 0.319; 0.254$ nm, etc.

As can be seen from Fig. 1b, in the samples fired at 1200 ° C, the crystal lattice of feldspar is completely destroyed, the content of β -quartz in it decreases, and the content of mullite increases. Quartz is characterized by interplanar distances $d/n = 0.423; 0.332; 0.254; 0.245; 0.227; 0.212; 0.181$ nm, mullite has interplanar distances $d/n = 0.535; 0.423; 0.346; 0.288; 0.254; 0.220; 0.212$ nm.

In specimens fired at 1250°C (Fig. 1 c), the amount of β - quartz with interplanar spacing $d/n = 0.424$ decreases; $0.334; 0.245; 0.227; 0.313$ nm and the amount of mullite with characteristic interplanar distances $d/n = 0.538$ increases; $0.346; 0.288; 0.254; 0.242; 0.227; 0.220; 0.209; 0.189; 0.173$ nm.

Thus, in the process of firing prototypes from mass C-1, after noticeably strong processes of dehydration, decomposition of carbonates, burnout of organic impurities at a temperature of 1100 ° C, processes of new formations of β - quartz, feldspar and, as a product of dehydration, metakaolinite occur little noticeably.

With a further increase in the firing temperature, the feldspar crystal lattice is completely destroyed, a glassy melt is formed, which fills the space between solid particles, the quartz content decreases due to its partial melting in the silica-feldspar melt, the formation of mullite occurs to a final temperature of 1250 ° C.

Figure 2 shows X-ray diffraction patterns of prototypes from mass C-2, fired at different temperatures. In samples from the compositions of mass C-2, fired at 1000 ° C, the structure of the sample partially changes, contains β -quartz with interplanar spacing $d/n = 0.424; 0.334; 0.245; 0.227; 0.212; 0.197; 0.181; 0.166; 0.153$ nm, cristobalite with $d/n = 0.406; 0.285; 0.249; 0.244; 0.212; 0.202; 0.197$ nm, dehydration products of clay-forming minerals, metakaolinite with interplanar spacing $d/n = 0.718; 0.443; 0.358; 0.254; 0.244; 0.233; 0.202; 0.195$ nm.

Samples from mass C-2, fired at 1100 ° C (Fig. 2 a), contain large amounts of β -quartz with interplanar spacing $d/n = 0.424; 0.334; 0.245; 0.227; 0.212; 0.197; 0.181; 0.166; 0.153$ nm, as well as cristobalite with interplanar spacing $d/n = 0.404; 0.318; 0.286; 0.249; 0.244; 0.202; 0.152$ nm. It is noted that the crystal lattice of feldspar is completely destroyed, the formation of mullite begins with interplanar distances $d/n = 0.538; 0.334; 0.286; 0.269; 0.254; 0.245$ nm

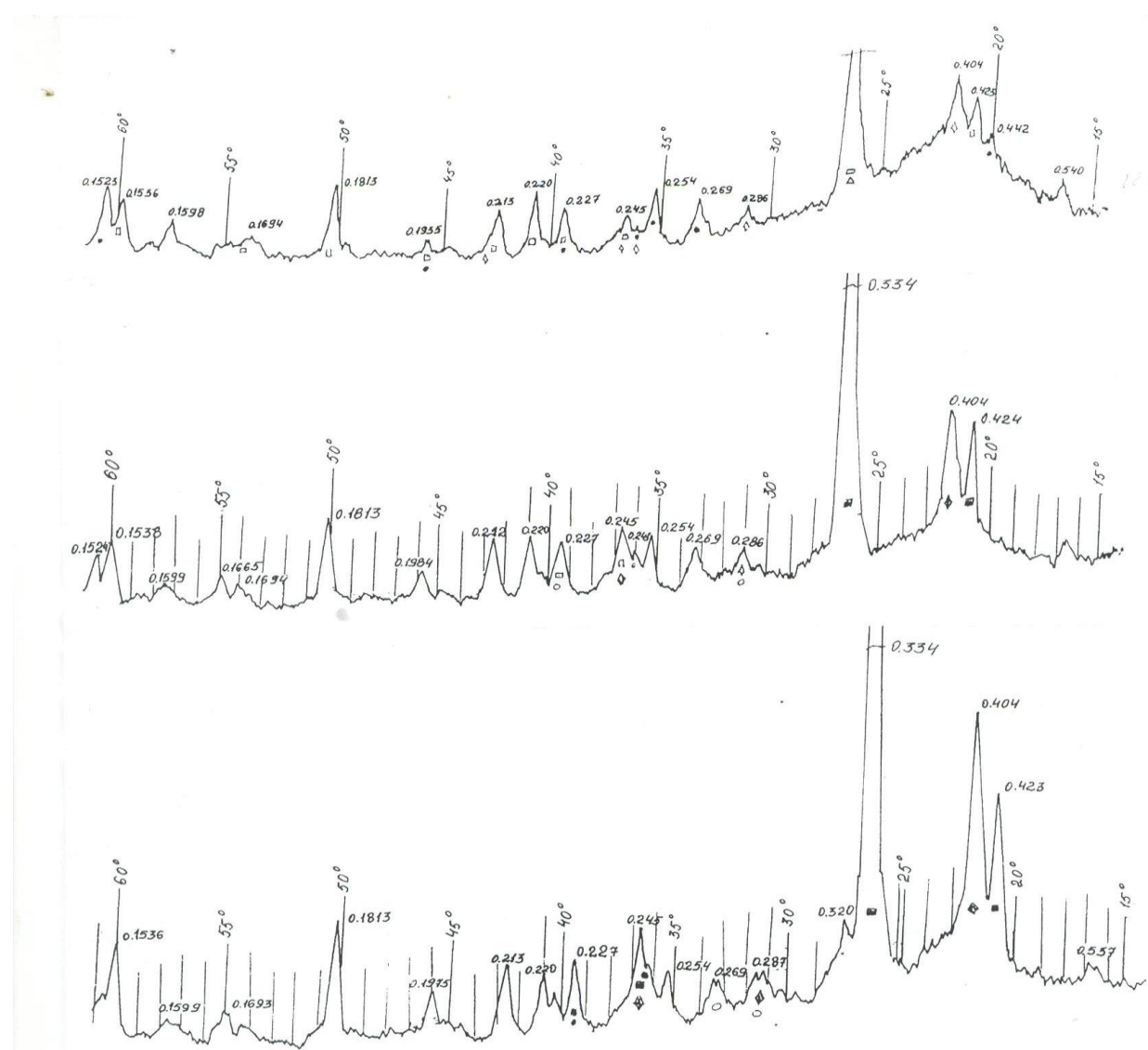


Fig. 2. X-ray diffraction patterns of samples from mass C-2, fired at different temperatures, 0C: a) 1100; b) 1200; c) 1250

In samples from mass C-2 (Fig. 2 b), fired at 1200 ° C, the amount of β -quartz decreases and the amount of mullite increases. It is noted that β -quartz is characterized by interplanar distances $d/n = 0.424; 0.334; 0.245; 0.227; 0.223; 0.213; 0.197; 0.181; 0.166; 0.152$ nm and mullite with $d/n = 0.539; 0.334; 0.286; 0.268; 0.254; 0.245; 0.220; 0.213; 0.188; 0.169; 0.166; 0.159; 0.152$ nm, cristobalite with $d/n = 0.424; 0.286; 0.249; 0.245$ nm.

In samples from mass C-2 (Fig.2c), fired at 1250 ° C, there is a sharp increase in the amount of mullite compared to heating to a temperature of 1200 ° C ($d/n = 0.537; 0.334; 0.254; 0.220; 0.212; 0.188; 0.169; 1.159$ nm), the amount of β -quartz ($d/n = 0.424; 0.334; 0.245; 0.227; 0.212; 0.181; 0.153$ nm) decreases, cristobalite ($d/n = 0.404; 0.314; 0.286; 0.249$ nm) increases.

From the above, we can say that during the firing of samples from the mass C-2, at a temperature of 10000C, the formation of quartz and cristobalite crystalline phases and dehydrated clay-forming minerals is observed. At a temperature of 11000C, the destruction of the crystal lattice of feldspar occurs, and the formation of mullite begins. At a temperature of 12000C, the quartz content decreases, the mullite content increases, and cristobalite crystals appear. At a temperature of 12500C, a sharp increase in the mullite content occurs, the amount of quartz decreases, and the content of cristobalite increases.

Results of petrographic research of prototypes. Figures 3, 4 and 5 show micrographs of C-1 prototypes fired at temperatures of 1100, 1200 and 1250 ° C.

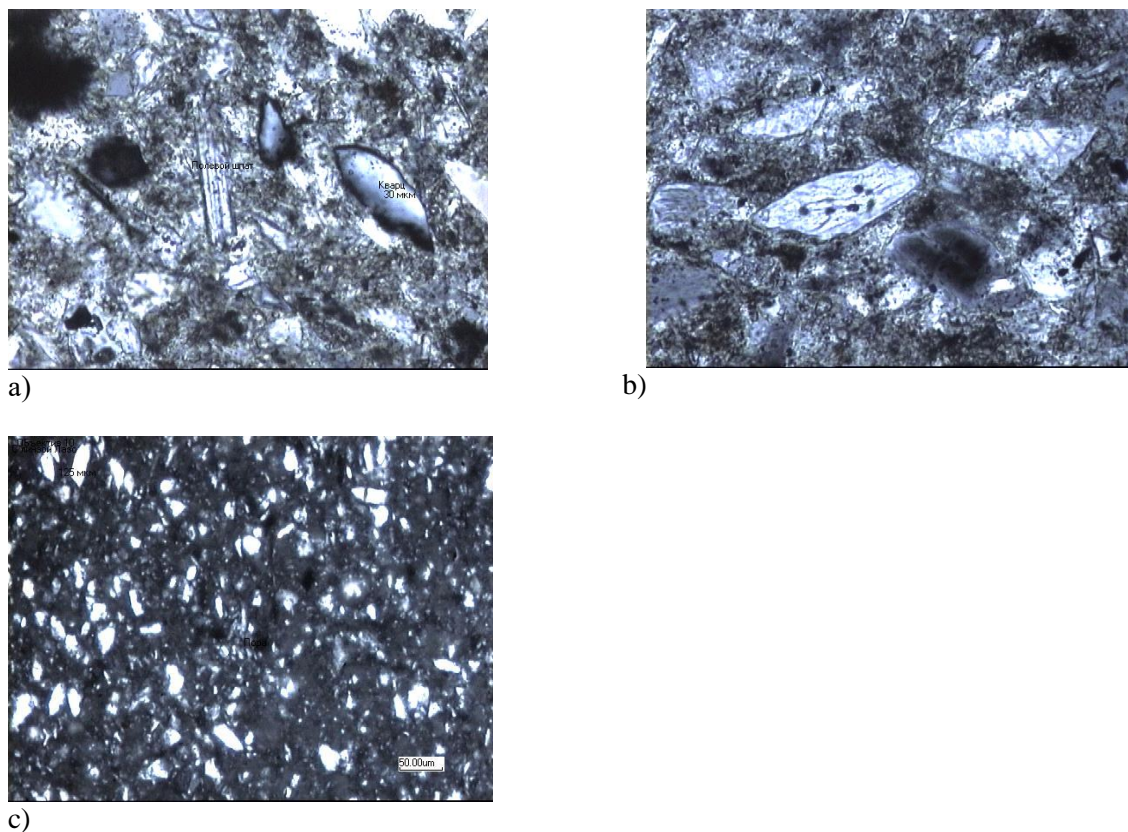


Fig. 3 Micrograph of prototypes from mass S-1, fired at temperatures of 1100, 1200, and 12500C

The structure of the sample (Fig. 3a) is heterogeneous, coarse-grained, and there are significant pores. Contains metakaolinite, quartz with $N_e = 1.552$; $N_o = 1.540$. The size of quartz grains ranges from 30 to 40 microns, and also contains feldspar grains with a size of 26-38 microns. It should be noted that metakaolinite is formed due to dehydration of kaolinite. Traces of mica are rarely found in the bulk.

It can be seen from Figure 3b (fired at a temperature of 12000C) that the structure of the sample is inhomogeneous, fine-grained, there are pores with a size of 30-35 microns. The bulk contains a glassy phase, which is formed due to the melting of low-melting components.

The content of the vitreous phase is sufficient. In the samples, the appearance of mullite grains from metakaolinite products with a grain size of 2-3 microns, quartz grains 25-35 microns in size, is observed in an insignificant amount of cristobalite formation. In the studied samples, quartz grains have $N_e = 1.552$; $N_o = 1.540$. Part of the quartz at high temperatures transforms into α -cristobalite, which has $N_e = 1.484$; $N_o = 1.487$. Quartz grains have an irregular shape, around the quartz grains there is a fusion border with a size of 2-3 microns.

The structure of the sample (Fig. 3c) is heterogeneous, fine-grained, there are pores of a rounded shape, their size reaches 20-25 μm , and their content decreases. At this temperature, the bulk consists of a vitreous phase, granular mullite, cristobalite and quartz. In the groundmass, mullite grains are distributed, there are separate areas of accumulations of fine-grained mullite with a grain size of 2-3 microns. The vitreous mass is distributed throughout the volume. The content of quartz is slightly reduced in comparison with the sample fired at a temperature of 1200 ° C.

The content of grains of cristobalite is more common, it is established by determining the indicators of light refraction of quartz and cristobalite. Quartz grains have an irregular fragmentary shape, the average size of quartz grains is 20-23 microns.

The results of a petrographic study of prototypes from mass C-2, fired at temperatures of 1100, 1200 and 1250 ° C are shown in Figure 4.

The structure of the sample (Fig. 4a) is non-uniform, coarse-grained, there are round pores with a size of 30-45 μm . Contains metakaolinite, feldspar, quartz, mica and traces of biotite. Metakaolinite is a dehydration product of kaolinite. A glassy phase is distributed in the bulk; it cements the existing crystalline phases, it is formed due to the melting of low-melting components. Comparing the results obtained with the previous results, we can say that this sample is the most porous.

A micrograph of a prototype from mass C-2, fired at a temperature of 1200 ° C, are shown in Figure 4b. The structure of the sample is inhomogeneous, medium-grained, contains rounded pores with a size of 25-30 microns. The bulk contains a glassy phase, which is formed due to the melting of low-melting components. The amount of the glassy phase is increased, the crystalline phases are cemented by the glass phase.

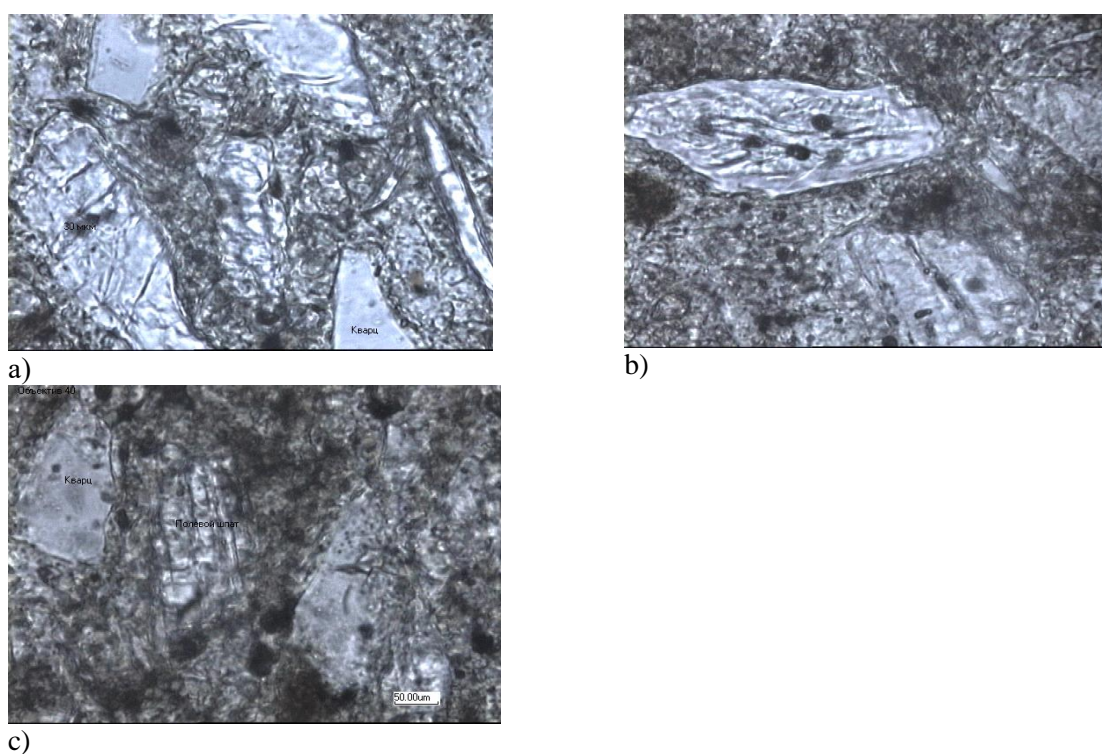


Fig. 4. Micrograph of prototypes from mass C-2, fired at temperatures of 1100, 1200 and 12500C

As crystalline phases, mullite grains with a size of 2-3 microns are observed, formed from the decomposition products of metakaolinite, its amount at this temperature is insignificant. An increase in the content of the glassy phase is observed in the bulk. In the sample under study, quartz grains are observed that have an irregular shape; around the quartz grains there is a fusion border with a thickness of 2-4 microns, which, under the influence of high temperatures, turns into a-cristobalite, having $N_e = 1.484$; $N_o = 1.487$.

The structure of sample C-2 (fired at a temperature of 1250 ° C) is heterogeneous, rather fine-grained, and contains round pores with a size of 20-30 microns. The content of pores decreases due to filling them with a glassy phase, the material becomes relatively dense.

The bulk consists of crystals of mullite, quartz, cristobalite. There are separate areas of accumulations of fine-grained mullite with a grain size of 2-3 microns. The glassy phase fills the space between the crystalline phases. The quartz content decreases, the fusion border around the quartz is expanded to 8-

10 microns, the cristobalite content increases. Cristobalite has a refractive index of $n_e = 1.484$; $n_o = 1.487$. Quartz is characterized by $n_e = 1.552$; $n_o = 1.540$.

CONCLUSION. Thus, based on the X-ray phase analyzes of prototypes fired at different temperatures, we can say that the nature of the phase formation process occurs similarly to classic earthenware products. There are significant differences, which include the following: with an increase in the firing temperature, the dehydration of clay minerals first occurs, and then the destruction of the crystalline phases is in direct proportion to the increase in the firing temperature, the crystalline phase of β -quartz partially transforms into cristobalite, which is confirmed by the appearance of intense peak lines characteristic of cristobalite, and partially dissolves in the feldspar melt. In addition, the amount of mullite increases with an increase in the content of clay components.

REFERENCE:

1. Maslennikova G.N., Alekseeva N.N., Moroz I.Kh. Structure formation of porcelain based on complex raw materials from the Far East. Improvement of technology and quality of industrial goods: Sat. scientific. work. DGU. –Vladivostok, 1995. –p. 31-32.
2. Moroz I.I., Koms kaya M.S., Sivchikova M.G. Handbook of porcelain and faience industry: - M., Light industry, 1976. T1.
3. Magidovich V.I. Feldspar raw materials, its genetic types and evaluation principles. -M., 1964. -S. 52.
4. Belinskaya G.V., Vydrik G.A. "Technology of electrovacuum and radiotechnical ceramics". M., Energiya, 1977, 56 p.
5. Giller L.Ya. Tables of interplanar distances. - Moscow: Nedra, 1966, Vol. 2, -264 p.
6. Mirkina L.N., Gorshkov V.S., Timashev V.V., Saveliev V.G. Methods of physical and chemical analysis of binders. –M.: Higher school, 1981. –334p.
7. Mikheev V.N. Radiometric determinant of minerals. - Moscow: Gosgeoltekhizdat, 1957, 868 p.