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The Effect of Heating Radiation on the Synthesis and Crystallization of Cordierite Composition Glasses

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Abstract

The results on crystallization of glasses of the cordierite composition, synthesized under the influence of concentrated radiant flux of different densities, are presented. Synthesis was carried out using a solar furnace or a solar simulator, wherein Xenon lamps of 10 kW power serve as a heat source. We studied glasses of the following stoichiometric composition $2MgO: 2Al_2O_3: 5SiO_2$ without a catalyst and with TiO_2 as a catalyst. The initial raw materials were MgO, Al_2O_3 and quartz-kaolinite-pyrophyllite rock as a main source of SiO_2. The natures of phase transitions in the samples obtained are studied using the X-ray analysis (DRON-UM-1) and the differential-thermal method (Derivatograph Q-1500 D). The absorption spectra are obtained on spectrophotometer SF-56. A comparison of the phase composition of the crystallized samples shows that the crystallization of μ-cordierite and the transition of μ-cordierite to α-cordierite in glasses, synthesized using a Xenon lamp, occurs at lower temperatures than those synthesized using solar radiation, provided the same conditions of synthesis and annealing. Besides of this, in glasses containing TiO_2, the content of Ti^{3+} increases, and a decay of the concomitant phase, magnesium-aluminum-titanate, is activated at annealing temperatures above 1200°C. The differences in the character of the phase formation affect the activity of glass powders to sintering.

It is found that peculiarities of the spectral composition of a Xenon lamp and the Sun affect the nature of the glass crystallization process. A presence of a significant proportion of extreme ultraviolet radiation initiates the crystallization process by the photo-activation mechanism and has the same effect as a rise of the glass crystallization temperature or an increase of the catalyst concentration.

Keywords: Cordierite; Glass; Crystallization; Radiation

Introduction

Peculiarities of crystallization of glasses in the system $MgO: Al_2O_3: SiO_2$ have been studied in many researches. Particular attention has been given to the crystallization of glasses of the cordierite composition ($2MgO: 2Al_2O_3: 5SiO_2$), since various glass-ceramic materials with an optimum combination of dielectric and thermo-mechanical properties are produced on its basis. The most common method of production of cordierite glass-ceramic materials is glass melting in electric or induction furnaces followed by subsequent crystallization. The crystallization of cordierite occurs through the formation of a sequence of intermediate phases. The temperature intervals of the formation and stability of the phases are changed depending on the conditions of synthesis and crystallization of glasses, as well as the type and concentration of catalysts. The effect of such factors, as temperature, time, gas atmosphere and the chemical composition of raw materials, on the properties of these materials has been studied in details [1-6].

In our study, the synthesis of glass is carried out under the influence of concentrated radiant flux from different types of sources. These conditions differ from traditional synthesis methods and can have a significant impact on the crystallization of glasses and the properties of subsequent glass-ceramic materials. However, studying the effect of short-wave radiation on the course of the crystallization of cordierite glasses has much less attention in the literature. The authors of [7,8] using a solar furnace for sintering the powder mixtures to obtain cordierite.

At the same time, in work [9], it is shown that the largest acceleration of the crystallization process in production of lithium-silica-alumina glasses is achieved with use of Xenon arc lamps of different power. The authors believe that since at high temperatures the radiant heat transfer is realized in the glass, the presence of the radiant component of heat transfer initiates the crystallization process by means of the photo-activation mechanism. In paper [10], the effect of ultraviolet radiation on a formation of glass-ceramics in photosensitive glasses is studied. In our conditions, radiation exposure occurs during glass melting, which may also have some influence on the process of the crystallization and phase formation during subsequent heat treatment.

The present work is devoted to the study of the crystallization of glasses of the cordierite composition, synthesized under the action of concentrated radiant flux, and some properties of glass-ceramics, obtained on their basis.

Materials and Methods

The initial components of the glass batch is magnesium oxide MgO (puriss), alumina Al_2O_3 (puriss), as well as natural minerals as a source of SiO_2. These minerals have different compositions, and they contain in addition to basic oxides of silicon, aluminum and magnesium on the average of 4.54 wt% of impurities (Fe_2O_3, Na_2O, K_2O). In this study, a quartz-pyrophyllite-kaolinite rock is used as a silica-containing material (Table 1). Table 2 shows the batch composition for synthesis of a glass of the following stoichiometric composition ($2MgO: 2Al_2O_3: 5SiO_2$) wt %.

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Chemical composition, wt.%	SiO_2	Al_2O_3	Fe_2O_3	TiO_2	CaO	MgO	K_2O	Na_2O
	77.96	20.82	0.36	0.10	0.31	0.10	0.20	0.15
Mineralogical composition, wt.%	Silica (SiO_2) - 25, Kaolinite ($Al_4[Si_4O_{10}][OH]_8$) - 25, Pyrophyllite ($Al_2[Si_4O_{10}](OH)_2$) -50							

Table 1: Chemical and mineralogical composition of the quartz-kaolinite-pyrophyllite rock.

Components	Component Content, wt.%
Magnesium oxide	14.51
Aluminium oxide	22.29
Quartz-kaolinite-pyrophyllite	63.2

Table 2: The batch composition for production of the glass of the composition 13, 78 MgO: 34.86 Al_2O_3: 51.36 SiO_2 (wt.%).

Siliceous raw materials (quartz-kaolinite-pyrophyllite rock) are pulverized preliminarily to particles of the size less than 100 microns. Aluminium oxide is burned annealed at 1450-1500ºC for the transition into a stable α-form. The initial components are weighed according to the calculations. Then the mixture is stirred with simultaneous additional grinding in a planetary ball mill to achieve a grind fineness of 5-10 microns. The resulting powders are molded for subsequent glass melting.

The glass synthesis is performed on a solar furnace (The Big Solar Furnace, power 1 MW, Parkent town, Uzbekistan), as well as on a solar simulator, in which focused radiation of two Xenon lamps of 10 kW power is served as a source of heating. Synthesis of glasses is carried out at two flux densities of focused radiation: 1) the flux density of 250 W/cm^2 and 2) the flux density of 600 W/cm^2. Cooling the molten glass is carried out by quenching in water. This method of melt quenching enables to obtain fragile granules, which could subsequently be readily milled for further preparation of glass-ceramics using the ceramic technology. Crystallization of glasses is carried out in the one-stage mode in an electric furnace in the temperature range 850-1100°C. Samples are either fragments of 2-3 mm granules or powders of fineness of 40-80 microns. To determine the ability of powders to sintering, a powder is compressed into tablets with a diameter of 20 mm and a thickness of 5 mm, and then tablets are annealed at temperatures of 900-1350°C. The water absorption of the samples is selected as a characteristic of the degree of sintering. Water absorption was calculated using the following relationship:

$$\mathbf{W\ (\%) = [(m_2 - m_1) / m_1] * 100}$$

where,

m_1 = mass of the dry sample, in g

m_2 = mass of the water saturation sample, in g

We investigate glasses of the stoichiometric composition, as well as those supplemented with 12 wt% titanium dioxide (over 100%) as a crystallization catalyst.

The X-ray analysis, the differential-thermal method and optical methods are used to analyze the synthesized materials. The X-ray analysis is performed on the diffractometer DRON-UM 1 (Cu K_α radiation, and Ni-filter).

Derivatograph Q-1500 D is used for the differential thermal analysis (DTA). The heating rate used is 15°C/min. Measurements are carried out up to a temperature of 1200°C. Absorption spectra are studied with SF-56 spectrophotometer in the wavelength range of 100–1000 nm. The resolution of the device is 0.3 nm.

Results and Discussions

Previously, our research [11,12] has shown that during the synthesis of glasses of the cordierite composition under the influence of concentrated radiant flux, the crystallization of α-cordierite occurs through the formation of solid solutions with the structure of high-temperature quartz (quartz-O) and μ-cordierite. On the DTA curves, obtained on the crushed granular samples, the presence of diffuse exothermic peaks in the temperature ranges 700-900°C and 900-1000°C and most pronounced at 1040-1050°C are observed (Figure 1). According to the X-ray analysis, the peaks correspond to the crystallization of the aforementioned phases. In addition, as can be seen from derivatograms, the temperature of exoeffects and their profiles are changed depending on the density of radiant flux and the type of the radiation source.

The X-ray phase analysis of the coarse-grained samples shows (Figure 2) that during the synthesis of glass of the cordierite composition in the absence of catalysts with the use of a Xenon lamp, the crystallization process of μ-cordierite in coarse-grained samples starts at 880°C (density of the radiant flux is 250 W/cm^2).

With a significant overheating of the melt, which corresponds to the flux density of 600 W/cm^2, the amount of crystallizing μ-cordierite increased sharply, while at 980°C the only crystalline phase is α-cordierite, regardless of the flux density. For traditional methods of synthesis of glasses of the cordierite composition, the rate of crystallization of μ-cordierite is extremely small. At a temperature of 850°C, it takes at least 150 hours for μ-cordierite to begin to crystallize [13]. This process is activated under the action of radiant flux.

During the synthesis of glass in the solar furnace, the crystallization begins at higher (by ~ 20°C) temperatures. At the annealing temperature of 880-900°C (flux density of 250 W/cm^2), there are no crystalline phases on the difractograms and at the annealing temperature of 980-990°C, μ-cordierite is the main phase component. For glasses synthesized under high radiant flux density (600 W/cm^2) during annealing at 880–900°C, the crystallization just begins and μ-cordierite remains at the annealing temperature 990°C. To complete the transition μ → α, it is necessary to increase the annealing temperature above 1000°C regardless of the degree of overheating of the melt. A sharper rise of the branch of exothermic effects, a narrow profile and an increase of the peak area, as well as the decrease of corresponding temperature in the DTA curves indicate an increase of the rate of the crystallization process with increasing of flux density, while using the Xenon lamp.

By utilizing powder samples, the phase composition is not changed, but the ratio of the crystallizing phases is changed, showing an increase of μ-cordierite amount. In the DTA curves, the high-temperature exothermic peaks have more distinct forms indicating the surface crystallization of the glass in the absence of catalysts (Figure 3) [14].

A similar effect of radiation was observed for glasses with additions of titanium dioxide as a catalyst. It should be noted that the color of titanium-containing glasses changes from black to yellow. Black glasses are obtained in synthesis in radiation of a Xenon lamp, while yellow glasses are obtained in synthesis in the solar furnace.

On diffractograms of crystallized glasses, in addition to the μ- and

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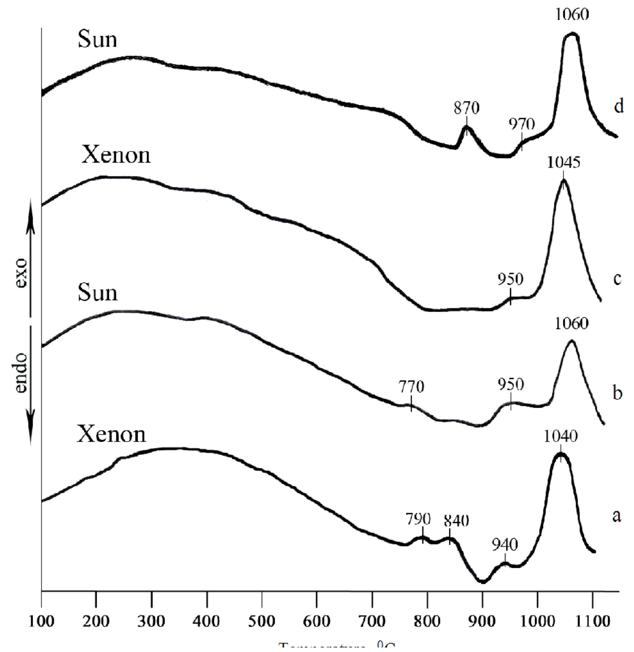

Figure 1: DTA curve fragments of 2-3 mm granules; a,b) flux density 250 W/cm^2, c,d) flux density 600 W/cm^2.

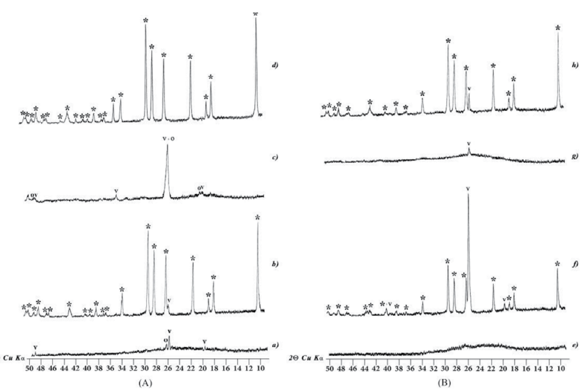

Figure 2: X-Ray diffraction patterns of cordierite composition glass samples: A) heating sources is Xenon lamp, B) heating sources is Sun; a,e) flux density 250 W/cm2, annealing temperature 880-900°C; b,f) flux density 250 W/cm2, treatment temperature 980-990°C; c,g) flux density 600 W/cm2, annealing temperature 880-900°C; d,h) flux density 600 W/cm2, annealing temperature 980-990°C.

α-cordierites, the presence of magnesium-aluminum-titanate (a solid solution $nAl_2TiO_5 \cdot mMgTi_2O_5$) is observed, and its amount increases with increase of content of titanium dioxide [12].

It is found that the synthesis of titanium-containing glasses under the action of radiation from a Xenon lamp in comparison with the solar radiation leads to a decrease of temperature of the corresponding phase transformations during subsequent crystallization from a μ-cordierite to α-cordierite, and at higher temperatures to accelerating of disintegration of magnesium-aluminum-titanate and formation of rutile. It is necessary to note that the phase composition of crystallized glasses with 12 wt % TiO_2, synthesized using the solar furnace, corresponds to the glass composition with 5-10 wt % TiO_2, crystallized at the same temperatures, but synthesized using a Xenon lamp.

Effect of radiation on the phase composition of the crystallized glasses is revealed in the ability of the glass powders, containing 12 wt % TiO_2, to sintering. A comparison of these results with those obtained from the X-ray analysis shows that the degree of sintering depends on the phase composition of the material. An appreciable reduction of water absorption is found only for the samples in which a transition from a metastable modification of cordierite to a stable α-form has been completed. Further intensification of the sintering process occurs with the decomposition of magnesium-aluminum-titanate. It is found that the glass powders, synthesized by using a Xenon lamp, are sintered better than those synthesized in the solar furnace (Figure 4), having the same conditions of synthesis and heat treatment.

Analysis of the absorption spectra shows peculiarities due to the influence of different contents of titanium dioxide, as well as the nature of ionizing radiation.

According to the previous works [15], the absorption band at 400-600 nm corresponds to Ti^{3+}. As shown in Figure 5, the intensity of the absorption band for glasses, obtained in the synthesis using a Xenon lamp, increases with increasing content of TiO_2, which may indicate an increase of the concentration of Ti^{3+}. In this case, the optical density of the glass is increased as well. This fact, along with a shift of the absorption band edge to longer wavelengths, can mean the system disordering. At the same time, this absorption band is practically absent in the cordierite glass (12% TiO_2), synthesized in the solar furnace. The nature of the curve of the latter spectrum is more like the absorption spectrum of a glass with less TiO_2, synthesized using a Xenon lamp.

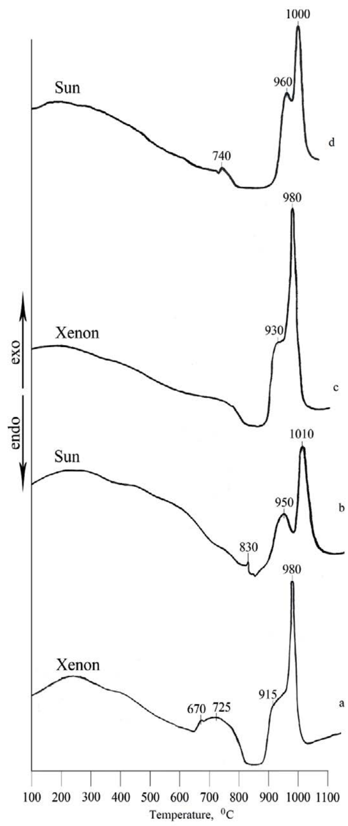

Figure 3: DTA curve or powders of fineness of 40-80 µm; a,b) flux density 250 W/cm^2, c,d) flux density 600 W/cm^2.

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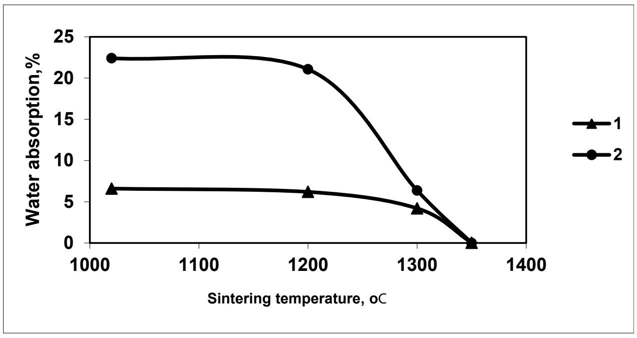

Figure 4: The dependence of the degree of sintering of cordierite glass composition with the addition of 12 wt % TiO_2 on synthesis conditions, the melt holding time is 5 minutes: 1) of a Xenon lamp; 2) the solar radiation.

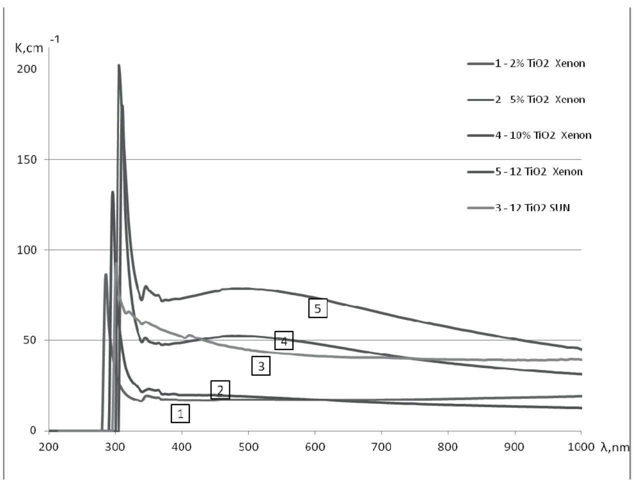

Figure 5: Absorption spectra of cordierite glasses doped by TiO_2 syntheses by irradiation of: Xenon lamp (curves 1,2,4,5), Sun (curve 3).

All absorption spectra have peaks in the region 350-370 nm, and the intensity of peaks remains constant for all of the samples. The origin of the peaks is difficult to identify unambiguously. According to [15], the absorbance at 350-370 nm, along with the absorption band of 400-700 nm may be due to the presence of Ti^{3+}. However, in this case, a correlation of the intensity of these peaks with percentage of TiO_2 should be observed. More likely, the peaks are associated with Fe^{3+} [16] due to the presence of Fe_2O_3 in the initial silica-containing materials. At the same time, there is a possibility of the formation of complex defects of FTi type, where F is the center near the titanium ion that replaces Al^{3+}.

It is known [17-21] that UV radiation has a photo-catalytic influence on a material, which results in a change in the material properties. In our case, this influence occurs at the stage of melting and glass synthesis that leads to the formation of structure imperfections, both on the surface and in the bulk of the melt. One of the effects of such an influence is the formation of anionic vacancies (in this case oxygen vacancies). In the presence of the transition d-elements in a glass, this process results in the possibility of formation of low-valent cations due to the capture of a free electron, for example: $e^- + Ti^{4+} \rightarrow Ti^{3+}$ [22]. In a cordierite glass without a catalyst, a formation of paramagnetic defects is due to the presence of trace contaminants [23], in particular Fe_2O_3, because of the use of mineral raw materials as a source of SiO_2. Introduction of TiO_2, as a catalyst, results in an increase of the concentration of such defects, including the formation of different complexes based Ti^{3+} (Ti^{3+}-Fe^{3+}, Ti^{3+}-Fe^{2+}, Ti^{3+}-Al^{3+} and so on). Changing the glass structure induces inhomogeneity, promoting segregation. A subsequent thermal treatment of such glasses leads to a bulk fine-grained crystallization with a release of the titanium-containing phase, and, consequently, to improvement of properties of the glass-ceramics.

Thus, the mentioned differences in the rate of crystallization and the nature of phase formation of the glasses studied are quite justified if we take into account the features of the spectral composition of the Xenon lamp and the sun. In particular, the spectrum of the Xenon lamp have a large proportion of extreme ultraviolet radiation in the wavelength region of less than 0.3 micron, which increases the crystallization process due to the photo-activation mechanism [24].

Conclusion

It is found that the crystallization of a glass of the cordierite composition, synthesized by the action of concentrated radiant flux depends on the type of a heat source, along with other known factors. Short-wave radiation activates the crystallization of μ-cordierite, the phase transitions (μ-cordierite to α-cordierite, decay the concomitant phase magnesium-aluminum-titanate in glasses containing TiO_2) and it has the same effect on the processes of crystallization as a rise of the temperature of the glass crystallization or an increase of catalyst concentration.

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