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Synthesis by Hydrothermal Method of Complex Charge of Yttrium-Aluminum-Silicate Glass for Radiotherapy

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Abstract: A new method for the synthesis of glass charges from solutions has been developed. It has been approved that the hydrothermal complex glass charge of the yttrium-aluminum-silicate glass composition is melted at lower temperatures, and the glass melting time is also reduced. Physicochemical studies of the synthesized charge and glasses were carried out. The obtained glass can be used in radiotherapy to diagnose and treat liver cancer. The developed method for the synthesis of hydrothermal complex charge is cost effective and environmentally friendly.

Keywords: complex glass charge, yttrium-aluminum-silicate, radiotherapy, YAS glass

1. Introduction

The work presents some results of the development of a new method for synthesizing $Y_2O_3 - Al_2O_3 - SiO_2$ (YAS) composition glass charges. Selection of this glass is conditioned by its unique physical and mechanical properties: heat resistance, low coefficient of thermal expansion, reduced oxygen conductivity and thermal stability at high temperatures. The glasses based on $Y_2O_3 - Al_2O_3 - SiO_2$ are promising materials for the use in quantum generators, light beam modulators, hydrodynamic converters, heat resistant and dislocation-free materials and high-temperature coatings [1-2]. When irradiating $Y_2O_3 - Al_2O_3 - SiO_2$ glass microspheres with thermal neutrons, a short-lived isotope Y90 (half lifetime of 64.1 hr) is formed, which is a pure $2.28MeV$ γ-radiator [3-4]. Recently, glasses on the base of YAS systems have been used in the form of glass microspheres to treat oncological diseases because they are resistant to radiation, non-toxic, chemically resistant and practically water-insoluble (first dimming class of water resistance), which excludes the problem of washout of radioactive material [5-7]. YAS glass microspheres are injected into the hepatic artery, from where they are transferred to the diseased organ to destroy tumor by localized irradiation. Before injection into organism, microspheres are irradiated in a nuclear reactor.

YAS glasses are obtained by melting solid-phase components in platinum crucibles at temperatures of $1600^0 - 1650^0C$. A sol-gel method of synthesis has also been developed from salts of yttrium, aluminum and organic derivatives of silicon; however this method is very long and multistage [8]. The production of glass of this system requires new developments, since the existing traditional methods do not meet the demand of these glasses because of the high cost or not technological.

This paper presents the results of research of the development of a new synthesis method – synthesis of the hydrothermal complex glass charge of the yttrium-aluminum silicate glass composition in order to reduce the melting temperature and reduce the glass melting time.

2. Experimental Approaches

The essence of the method is as follows: the complex charge is synthesized directly from solutions of salts of the glass components. As for initial components sodium silicate obtained from rocks, nitrates and sulfates of yttrium and aluminum are used [9]. The sodium silicate solution at continued agitation is poured off to yttrium and aluminum salts mixture solution and hold for $30-40$ minutes at $80-85^0C\left(pH=7\right)$.

Calculations were performed according to the reaction for obtaining the given glass composition:

$$\mathrm{x}Y(NO_3)_3+yAl_2(SO_4)_3+3(\mathrm{x}+y)Na_2O.nSiO_2=\mathrm{x}Y_2O_3.yAl_2O_3.3n(\mathrm{x}+y)SiO_2+6\mathrm{x}NaNO_3+3yNa_2SO_4$$

Concentrations of initial solutions made $0.5-1.0M$. The obtained residual matter was filtered, washed to remove nitrate, sulfate and sodium ions and dried at 160^0-180^0C to $10-15\%$ humidity. Under co-precipitation of yttrium and aluminum silicates, homogeneous, fine, white amorphous powder - a solid solution of Y and Al hydrosilicates is obtained, which is the glass charge, ready for melting. The synthesized glass charge does not contain volatile components, except water. A comparative study of melting traditional and complex charges of YAS glass (composition in wt. %): $Y_2O_3-43,37;$ $Al_2O_3-22,09;$ $SiO_2-34,54)$ was carried out using Nabertherm LHT 08/17 electrical furnace.

3. Results and Discussions

A physical and chemical study of the synthesized charge and glass was conducted. X -ray phase analysis showed the amorphous structure of the charge and glass (Fig. 1).

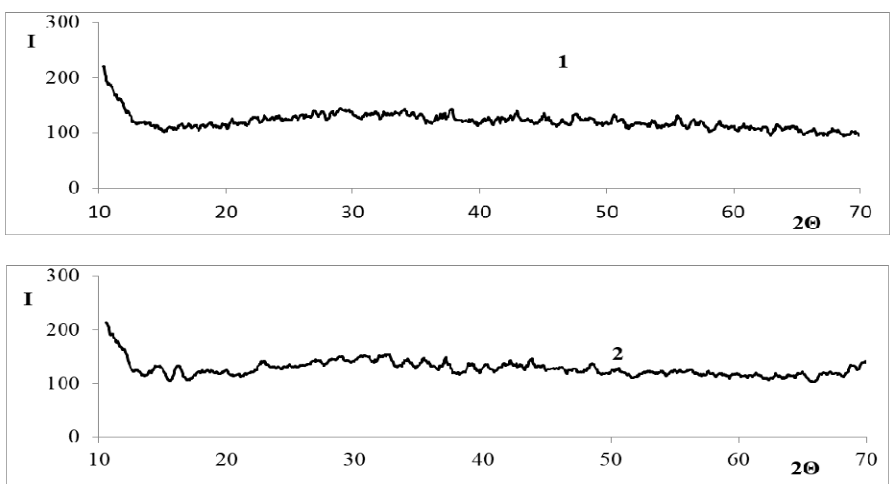

Figure1. X -ray patterns of the charge and YAS glass. 1 – hydrothermal complex charge, 2 – glass.

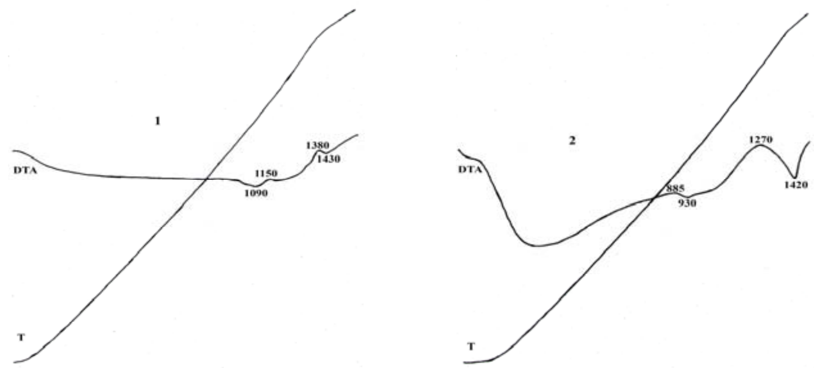

Figure 2. Thermograms of the hydrothermal charge – 1 and glass – 2.

Figure 2 shows the thermogravimetric analysis curves**.** The endothermic effect at 1030^0C is caused by the loss of structural water (OH group) of amorphous hydrosilicate and the formation of a crystalline phase $(\alpha - Y_2SiO_5)$: exothermic effect with a maximum at 1150^0C. The thermogram of glass shows that softening and deformation of glass begin at 885^0C and 930^0C and at 1270^0C crystallization is observed. Endo-effects at 1430^0C and 1420^0C are caused by melting of the charge and glass. IR spectra of the charge and glass samples are shown in Fig. 3. The broad absorption band with a maximum at $1100cm^{-1}$ due to the valence vibration of the $Si-O-Si$–bond typical to structural unit of silicon dioxide, is strongly shifted to the wave number of $1000cm^{-1}$. This indicates the effect of cations on the silicate framework due to the formation of $Al-O-Si-O-Y$ bonds. The appearance of a doublet in this region on the IR spectrum of glass can be explained by a change in the structure of silicates during glass melting; the valence vibration of the $Al-O-Si-$ and $Y-O-Si$ -bonds.

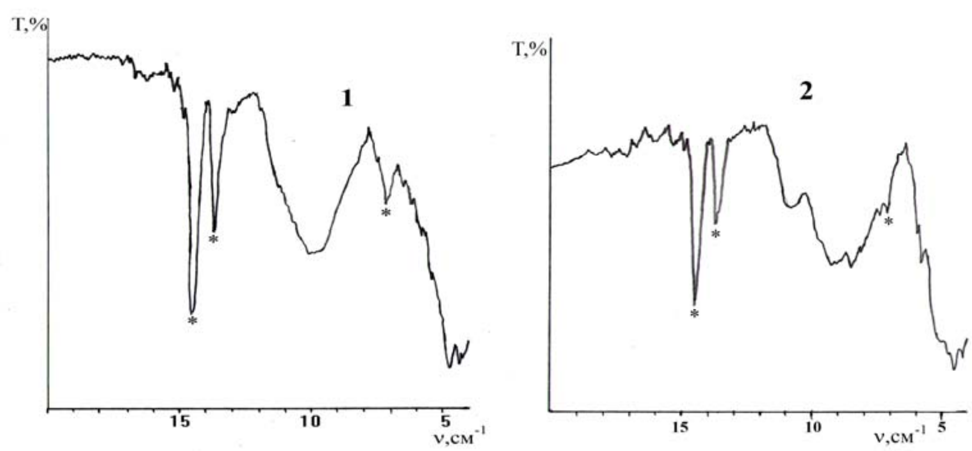

Figure 3. IR spectra of the hydrothermal charge – 1 and YAS glass samples – 2, * – petroleum jelly.

The melting characteristics of glass charge of the composition $Y_2O_3 - Al_2O_3 - SiO_2$ are studied. Melting curves of traditional and hydrothermal charges are shown in Fig. 4.

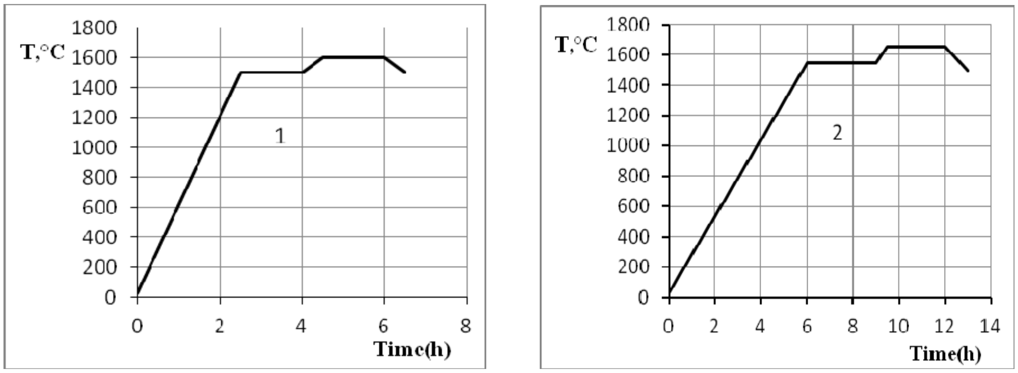

Figure 4. Temperature-time mode of YAS glass melting and producing: 1 – hydrothermal charge; 2 – traditional charge.

Comparison of the temperature and time regimes of melting YAS glass charge (Fig. 4) obtained by traditional and hydrothermal methods, testify the fact, that melting of hydrothermal charges take place much faster compared to traditional charges, because the silicate formation process is already completed during the liquid-phase synthesis of complex glass charge. Air emissions are water vapor, while nitrogen and sulfur oxides are not more than 0.1% of glass mass.

The coefficient of thermal expansion and the chemical resistance of glass are determined: YAS glass has the following characteristics. As for chemical stability, they fall into first dimming class; their temperature coefficient of linear expansion within the range of $20^0 - 300^0 C$ makes $(54.2)\times 10^{-7} grad^{-1}; tg - 875^0 C; t_d - 910^0 C; d - 3.45 g/cm^3$.

4. Conclusions

Thus, a method for the synthesis of a complex hydrothermal glass charge of the composition $Y_2O_3 - Al_2O_3 - SiO_2$ at low temperatures $(70^0 - 80^0 C)$ has been developed. The charge is a chemical compound yttrium-aluminum hydrosilicate. Hydrothermal method of synthesizing $Y_2O_3 - Al_2O_3 - SiO_2$ glass charges is capable to solve the following questions:

- Twofold reduce the glass melting time, reduce the melting temperature by $50^0 C$ due to the fact that homogenization and silicate formation take place at low temperatures during the charge synthesis.

- The use of cheap raw materials: the sodium silicate solution obtained from rocks.

- Harmful emissions into the atmosphere are excluded.

The developed method for synthesizing hydrothermal complex glass charges is cost-effective and ecologically clean. The obtained YAS glass can be used in radiotherapy to diagnose and treat liver cancer.

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