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Influence of the Composition and Structural Differences of Zirconium-Containing Sodium–Cesium Aluminoborosilicate Glasses on Their Heat and Water Resistance

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Abstract—The study deals with the effect exerted by adding various zirconium amounts on the heat resistance and chemical durability of aluminoborosilicate glass-containing host materials prepared by rapid cooling (quenching) of melts with different ratios of sodium and cesium. The specific features of the anionic structure of the glass-containing part of these materials were studied by vibrational spectroscopy. The Na/Cs ratio and zirconium additions influence the softening/glass transition point and the rate of dissolution of the major components on prolonged keeping in an aqueous medium. The results obtained are recommended for use in evaluation and correction of properties of borosilicate host materials for high-level waste immobilization.

Keywords: radioactive waste, vitrification, host materials, structure, heat resistance, chemical durability, vibrational spectroscopy, cesium, zirconium

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INTRODUCTION

One of the ways for high-level waste (HLW) immobilization is vitrification [1–3]. Vitrification technologies are widely used at the Mayak Production Association since 1987. For enterprises implementing such technologies, it is economically appropriate to dispose of vitrified HLW in a minimal volume with maximum possible incorporation of components at which the main characteristics of the glass matrix are still preserved. Within the HLW vitrification technology, there are regulatory limits and requirements aimed at ensuring the reliable long-term retention of radioactive elements. These requirements, in particular, concern the heat resistance and chemical durability of the host materials [4].

The base formulations of borosilicate host materials suggested for vitrification appreciably differ in the heat resistance and chemical durability depending on the ratio of the major components, composition of additional components, and external actions [5–9]. Therefore, it is topical to further optimize the composition of host materials for HLW immobilization and to study their properties, including the heat resistance and chemical durability [10, 11].

Previously we have studied the structure and properties of model host materials based on the Na_2O–$Cs(Rb)_2O$–B_2O_3–SiO_2–Al_2O_3 system and synthesized in the form of aluminoborosilicate glasses of the developed base composition with the addition of alkali, alkaline earth, and transition metal oxides as surrogates of radioactive waste components [12]. Within the

An increase in the cesium content of the materials accelerates the transfer of sodium, boron, and silicon into the aqueous solution. At higher zirconium content of the glass, the silicon solubility decreases, because the formation of a large amount of Zr–O–Si bonds decelerates their hydrolysis and the silicon washout from the glass structure.

High degree of zirconium binding in the glass structure excludes its participation in diffusion processes, which confirms the correctness of using Zr as a tracer in constructing the normalized concentration profiles of the dissolution of other glass elements in an aqueous medium [21].

The results obtained are of fundamental importance and are recommended for practical use in the development of regulations concerning installations for the vitrification of zirconium-containing HLW in borosilicate glass.

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CONFLICT OF INTEREST

The authors declare that they have no conflicts of interest.

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