STRUCTURAL CHARACTERIZATION OF URANIUM AND LANTHANIDE LOADED BOROSILICATE GLASS MATRIX

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Spent nuclear fuel remains highly radioactive after reprocessing due to actinides and fission products, which require safe containment within a durable, long-lived inert medium over extended timescales. Borosilicate glass is a potential candidate for this purpose, thus understanding the effects caused by the combined presence of uranium and actinides within these matrices is of great importance. For the structural studies lanthanides were chosen as chemical surrogates for actinides. The model glass matrix composition, $55SiO_2 \cdot 10B_2O_3 \cdot 25Na_2O \cdot 5BaO \cdot 5ZrO_2$ (mol%), was loaded with 20 wt.% UO_3 and 10 wt.% of lanthanide oxides (CeO_2, Nd_2O_3, and Eu_2O_3). An additional composition was also tested in which all oxides were simultaneously incorporated at 10 wt.% each. Neutron diffraction in combination with Reverse Monte Carlo simulation confirmed that the basic glass structure is comprised of tetrahedral SiO4, and mixed BO_3/BO_4 units. The presence of Ce^{III} and Nd^{III} ions causes a decrease in the quantity of non-bridging oxygens, as indicated by increased Si-O and B-O coordination numbers. The samples show a comparable trend reducing the number of non-bridging oxygens and promoting isomerization from BO_3 to BO_4. X-ray absorption spectroscopy indicated that uranium exists in the forms of U^{V} and U^{VI}, while europium and neodymium are present as Eu^{III} and Nd^{III}, with cerium mainly as Ce^{III}. X-ray photoelectron spectroscopy indicated distinct oxidation states, revealing a depth-dependent variation in the U^{IV}/U^{VI} ratio. While all compositions showed increased dissolution of Si, B, and Na, the composition with 10 wt.% of each oxide exhibited an enhanced structural stability. The studied borosilicate glass matrix shows a strong capacity to incorporate high concentrations of actinides and uranium, suggesting its viability for use in high-level nuclear waste disposal.
Details of the structural characteristics, the glass stability and incorporation ability of the ions will be presented.

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