HIGH-PRESSURE REAL GLASS CERAMICS FOR IODINE NUCLEAR WASTE IMMOBILIZATION: PRELIMINARY EXPERIMENTAL RESULTS

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Abstract

Several different matrices have been considered for the durable immobilization of iodine radioisotopes. Whereas aluminoborosilicate glasses represent the preferred storage form for nuclear waste it is difficulty applicable for iodine owing to its strong volatility. However, a recent approach proposed to use high-pressure synthesis conditions to incorporate a large iodine quantity into the structure of glasses.
Nowadays, there is a growing interest into developing glass ceramics matrices for various technological application in which nuclear waste immobilization is one of them. In the present work, we have successfully synthesised high-pressure I-bearing glass ceramics obtained from the partial crystallization of a parental glass.

The starting composition is a standard aluminoborosilicate glasses previously investigated in the system SiO2-Al2O3-B2O3-CaO-Na2O. The pressure conditions were 1.0 and 1.5 GPa and the experimental charges were loaded with either I2 or I2O5. The thermal history followed the classical melting-nucleation-crystallization protocol. The glass transition temperature (Tg) of the parental glass (with and without I) was determined by Differential Scanning Calorimetry (DSC) at ˜520◦C and the nucleation temperature (Tn) was assumed to be 50◦C above Tg. The crystallization temperature (Tc) was determined by DSC at 790◦C. A 24h duration was employed for Tn and Tc.

The recovered samples were characterized by high-resolution Scanning Electron Microscopy with Energy Dispersive Spectroscopy (SEM EDS) and X-Ray Diffaction (XRD). Results obtained from SEM EDS show the co-existence of a glass matrix enriched with I (up to 0.8 mol.% I) and I-bearing crystalline phase. The XRD revealed that iodosodalite (Na8Al6Si6O24I2) and nepheline (NaAlSiO4) are present in all the crystallized samples. Traces of NaPt3O4 have also been identified.

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For the samples equilibrated with I2 fluid, the structure of the iodosodalite crystalline phase is entirely resolved with the Rietveld refinement. For those samples, the cell parameter for iodosodalite is about 9.008 Å and the β cages of the iodosodalite cubic structure are fully occupied by I- species. Conversely, for the samples equilibrated with I2O5 fluid phase, the structure of the iodosodalite is incompletely resolved with the Rietveld refinement suggesting that the β cages is possibly partly filled with another iodine form such as iodate, IO3-.
We combined the SEM image analyses and the Rietveld refinement of the XRD diagrams on several selected samples to determine the mass proportion of each crystalline phase coexisting with I-bearing glass matrix: iodosodalite and nepheline are present in equivalent mass proportion, for up to 80% of the experimental charge volume. Those preliminary results indicate that glass ceramics could represent a serious candidate for the durable immobilization of iodine radioisotopes. Although our work represents the first evidence for real I-bearing glass ceramics; further experimental work is required to prevent from the nepheline crystallization that is detrimental for the matrix durability and to increase the proportion of iodosodalite for higher nuclear waste load.

Keywords: glass ceramics, high, pressure, iodine radioisotopes immobilization