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Abstract

In the frame of the development of nuclear waste vitrification processes, special attention is paid to solid waste containing metals contaminated with actinides, and in particular with uranium (production waste, decommissioning waste, for example). Whether to decontaminate the metal phase or to vitrify the actinides, the objective is to solubilize the actinides in the vitreous phase. Nevertheless, oxidation-reduction reactions between the glass and the metallic phase can modify the composition of the melt. The vitrification must be able to maintain, within a certain range of compositions and redox potential, the actinides solubility. It is known that uranium solubility is highly dependent on its oxidation states (VI, V, IV) [1]. Indeed, Uranium VI is the most soluble oxidation state while uranium IV is the least soluble.

The aim of this work is to determine and model uranium solubility and its oxidation states as a function of the glass melt oxidoreduction potential and composition. To do so, two parameters; which are oxygen fugacity and glass composition; are evaluated. In this work, the ternary SiO_2-Al_2O_3-CaO and SiO_2-Al_2O_3-MgO systems are studied. For each one, different compositions are synthesized while keeping the ratio Si/Ca and Si/Mg constant. Uranium is introduced into the glass in its most soluble form, i.e. U(VI). The redox of the glass is then modified in a muffle furnace at 1400°C with a CO/CO_2 gas mixture up to the equilibrium. The quenched samples were then analyzed by SEM and EDS to determine the solubility of uranium.

This paper presents our results on the uranium solubility in aluminosilicate glasses. The evolution of the uranium solubility as a function of the oxygen fugacity and for different glass compositions is exposed.

[1] Schreiber, H. D. (1983). The chemistry of uranium in glass-forming aluminosilicate melts. *Journal of the Less Common Metals*, *91*(1), 129-147.