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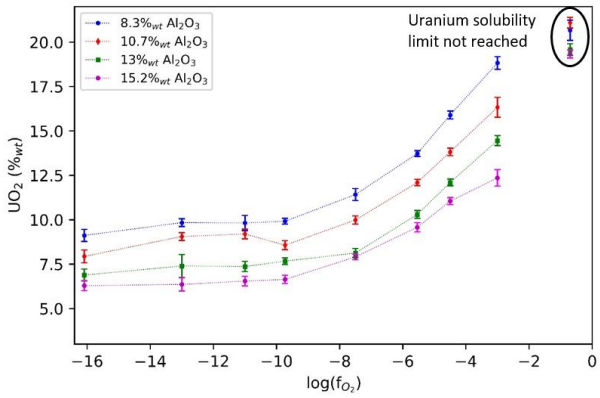
The immobilization of highly radioactive waste using vitrification process has been in application since many decades [1]. This process can reduce the potential for radionuclides migration or dispersion [2] by chemical incorporation into the structure of a glass matrix in order to trap radionuclides. This process is now under investigation for conditioning Intermediate Level Waste – Long Life (ILW-LL) using aluminosilicate glasses as host matrices. The vitrification must be able to maintain, within a certain range of composition and redox potential, the actinides solubility. It is known that uranium chemistry is complex in glass-forming systems due to the presence of many oxidation states (VI, V, IV), each of them having a specific solubility in melts.
The aim of this work is to determine and model uranium solubility and its oxidation states as a function of the glass melt composition and oxidoreduction potential (oxygen fugacity, from $10^{-0.7}$ atm to 10^{-16} atm). The ternary SiO_2-Al_2O_3-CaO (CAS) and SiO_2-Al_2O_3-MgO (MAS) systems are studied. Different compositions are synthesized by modifying the Al content for each ternary system. Uranium is added in excess in order to reach an equilibrium between the melt and uranium oxide crystals.



(a) (b)

Figure 1. Total uranium content reported as weight% UO_2 as the function of oxygen fugacities for CAS system. Elaboration temperature was 1400°C. Error bars represent the standard deviation based on reproducibility of 30 EDS analyses for each samples.

This presentation will give results on the behaviour of uranium over a large redox range. The results of uranium solubility (SEM/EDS) as a function of oxygen fugacity as well as a function of Al2O3 content will be presented (example on Figure 1). Analyses of uranium oxide crystals (XRD) were carried out. The proportion of uranium oxidation states was determined by XANES while their different structures were analysed by EXAFS. Hypotheses on the differences in behaviour between the two systems CAS and MAS will be formulated.

References

[1] P. Colombo; *Current Opinion in Solid State and Materials Science*, *7(3),* 2003, 225-239.
[2] M.I. Ojovan; An introduction to nuclear waste immobilisation, *Elsevier* 2019