Rheology of partially crystallized simulated nuclear glass melts

Elise Regnier*[1], Jeanini Jiusti[2], Norma Pereira Machado[3], Muriel Neyret[2], and François Faure[4]

[1]CEA, DES, ISEC, DPME, Univ Montpellier, Marcoule – CEA, DES, ISEC – France
[2]CEA, DES, ISEC, DPME, Univ Montpellier, Marcoule – CEA, DES, ISEC, CEA Marcoule 30207 Bagnols sur Ceze – France
[3]CEA, DES, ISEC, DPME, Univ Montpellier, Marcoule – CEA, DES, ISEC, CEA Marcoule 30207 Bagnols sur Ceze – France
[4]Centre de Recherches Pétrographiques et Géochimiques – Institut National des Sciences de l'Univers, Université de Lorraine, Centre National de la Recherche Scientifique – France

Abstract

In France, high-level wastes (HLW) resulting from the reprocessing of spent Uranium OXide fuel are conditioned in sodium alumino-borosilicate glasses. The formulation of such glass results from a compromise between the waste-loading (waste mass to final glass mass ratio), the technological feasibility of the glass at an industrial scale, and its long-term behavior. To date, the compositions of the alumino-borosilicate glasses are formulated to avoid any crystallization in the melt. However, the tolerance of crystals in the glass melt could allow to reach higher waste loadings, which would decrease the number of glass-containers and therefore the cost of the long-term storage.

Previous studies have shown that the presence of particles alters some properties of the glass melt, particularly its rheology. This is the case of platinoids elements, which are poorly soluble in the glass matrix (solubility limit generally $<$ 100 weight ppm), and which precipitate as RuO2 needles and PdTe beads. Hanotin et al. (1) observed that less than 1 vol% of platinoids particles leads to a very shear thinning behavior. Unlike platinoids particles, for which the volume fraction is constant during glass melting, the fraction of crystals considered in this study depends on temperature and time. Therefore, the study of the effect of crystals on the rheology of the melt has to take into account the time-temperature dependency of the crystal fraction. In addition, crystals morphologies are also likely to change with temperature (2). Two types of crystals observed in nuclear glasses due to exceeding solubility limits were chosen for this study: 1) cerianite, which generally shows a cubic morphology and slow crystallization kinetics 2) apatite, which generally has an acicular shape and fast crystallization kinetics (2). The morphology of crystal is expected to impact the rheology behavior of the melt (3).

In this work, we first evaluate the crystallization properties of both crystal types at high temperatures (around 1100◦C) in both static and dynamic conditions (absence / presence of flow). In static conditions, thermal treatments are performed in a tubular furnace. For the experiments in dynamic conditions, a stress-imposed rheometer with a multiblade rotor is used. The stirring is controlled by changing the imposed stress – the viscosity is acquired

*Speaker

simultaneously. Post-mortem analyses are carried out for each condition in order to assess the microstructure of the frozen melt. For the studied cerianite fractions (up to _˜1 vol%), virtually no change in the viscosity is observed. For the apatite case, however, a very shear thinning behavior is observed, similar to that observed for platinoids particles in previous studies (figure 1). Rheological models are tested to describe the behavior of the suspension. These findings will be useful to simulate the behavior of the glass melt during the vitrification process. References

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