Modelisation of liquid nuclear waste vitrification : focus on the chemical processes

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Abstract

Vitrification process is a technology commonly used for the treatment and the immobilisation of high-level radioactive waste resulting from spent fuels. In France, the waste are conditioned in glass matrix which provide a stable and sustainable confinement. A nitric solution of the waste is first calcined and the residual calcine is then added to glass melt in a cold crucible by induction processes (solid feeding). Another way of waste vitrification could be to introduce the waste solution directly at the glass melt surface (liquid feeding) in order to avoid the calcination step. Simulation studies1-3 have enabled to propose a 3D model of nuclear waste vitrification in cold crucible by solid feeding, taking into account the fluid mechanics, the induction heating, the thermal and the chemical aspects. However, no modeling of vitrification by liquid feeding has been proposed yet. Such modeling could bring new information on the evolution of the chemical processes and help to predict and optimise the waste vitrification by liquid feeding.

In this context, this study aims to bring new constrain to the 3D modeling on the thermal and chemical reactions occuring during vitrification process by liquid feeding. In situ experiments in temperature have been performed (ESEM and XRD) in order to study the microstructure and to characterise the phases. This first experimental approach has allowed to identify the steps of phase transformation in the liquid in interaction with glass frit (nitrate melting, nitrate decomposition, glass softening, melting and phase dissolution), from the ambiant temperature to 1200∘C. Ex situ experiments in furnace have also been performed to characterise the evolution of glass composition. Element concentrations have been analysed for different temperature (from 800 to 1200∘C) at different dwell time (2, 5, 30, 120 and 480 minutes). The evolution of concentrations allow to determine the liquid dissolution kinetics occuring during vitrification by liquid feeding. The results will pave the way to a global 3D model of liquid radioactive waste vitrification.

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