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► **To cite this version:**

Stéphane Gosse, Sophie Schuller, Werner Verdier, Alain Cartalade, Paul Fossati. An integrated thermodynamic approach for phase separation and crystallization in nuclear glasses. MRS 2021 – 45th Scientific Basis for Nuclear Waste Management (SBNWM), Oct 2021, PARIS, France. cea-04673000

HAL Id: cea-04673000

<https://cea.hal.science/cea-04673000v1>

Submitted on 19 Aug 2024

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MRS 2021 – 45th Scientific Basis for Nuclear Waste Management (SBNWM)

An integrated thermodynamic approach for phase separation and crystallization in nuclear glasses

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Abstract

Radioactive waste vitrification has been carried out industrially in several countries for nearly 40 years. Research is ongoing to safely conditioning new types of wastes and develop innovative technologies. In these waste glasses, fission products and minor actinides are integrated in the vitreous network by reactions occurring in high temperature vitrification furnaces. During the continuous steps of the process, the state (solid, liquid, gas) of the precursors and waste evolve with time and temperature until a high-temperature homogeneous liquid is formed.

After cooling, the waste load can be *i*) solubilized in a homogeneous glass at the microscopic scale or *ii*) integrated in vitro-ceramic matrices where crystallized phases accommodate larger quantities of waste beyond their incorporation limit.

This work will focus on academic research carried out to understand the high temperature thermochemistry on insoluble molybdate phases in glass and glass-ceramic. Following a general introduction on French nuclear glass waste forms, some results based on a thermodynamic approach coupled with phase field modeling will be presented.

The high temperature thermodynamics of the calcium and sodium phases – mainly CaMoO_4 and Na_2MoO_4 – is described using the Calphad method and coupled with a phase field approach. The phase field model describe the local dynamics of the interface between liquid-liquid separated phases of the heterogeneous glass melt using a formalism coherent with its thermodynamic profile. This allows a tight coupling of the dynamics and the available thermodynamic data. The aim of this integrated methodology is to better explain and control the mechanisms of crystallization, phase separation and diffusion during simplified nuclear glass synthesis