



HAL
open science

Thermodynamic models for phase separation and crystallization in nuclear glasses

Stéphane Gosse, Paul Fossati, Sophie Schuller

► **To cite this version:**

Stéphane Gosse, Paul Fossati, Sophie Schuller. Thermodynamic models for phase separation and crystallization in nuclear glasses. Sunglass 2023, Sep 2023, Nîmes, France. cea-04673048

HAL Id: cea-04673048

<https://cea.hal.science/cea-04673048v1>

Submitted on 19 Aug 2024

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

Thermodynamic models for phase separation and crystallization in nuclear glasses

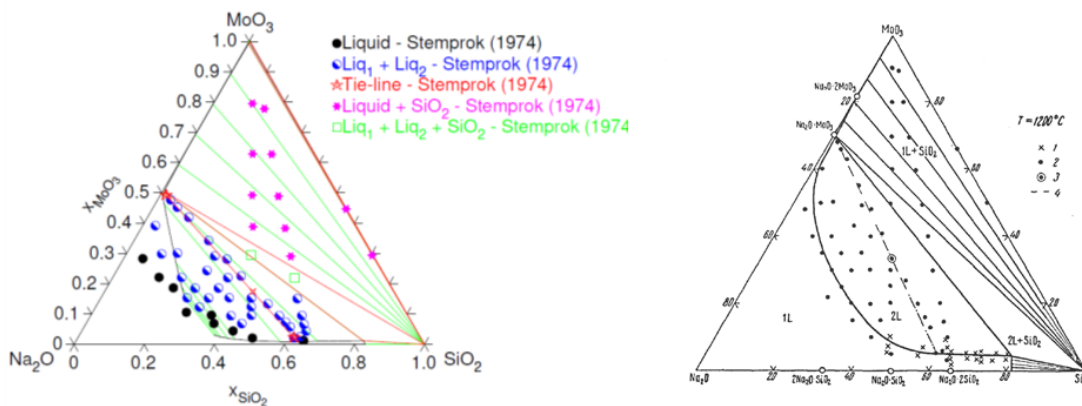
S. Gossé¹, P. Fossati¹, N. Dupin², A. Deshkar¹, S. Schuller³

¹ Université Paris-Saclay, CEA, Service de Recherche en Corrosion et Comportement des Matériaux, 91191, Gif-sur-Yvette, France

² Calcul Thermodynamique, Rue de l'avenir, 63670, Orcet, France

³ CEA, DES, ISEC, DPME, Univ Montpellier, Marcoule, France

stephane.gosse@cea.fr*



Radioactive waste vitrification has been carried out industrially in several countries for several decades. Meanwhile, research is still ongoing in France to safely conditioning new types of nuclear wastes and develop innovative technologies in hot pot or cold crucible. In these nuclear waste glasses, fission products and minor actinides are integrated in the vitreous network during high temperature reactions in vitrification furnaces. During the steps of the process, the solid, liquid and gaseous states of the glass precursor and waste evolve continuously with time and temperature until a high-temperature homogeneous liquid is formed. After cooling, the waste remains integrated at the molecular scale in a homogeneous glass. However, some fission product elements have limited solubility in the glass matrix and precipitate as aggregates or crystals in the glass. This study focuses on the fundamental research conducted to better describe the thermodynamics of these complex systems. Its objective is to better anticipate the behavior of typical insoluble phases in nuclear glasses such as platinoid precipitates and complex molybdates that can form beyond their solubility limits.

To predict the chemical interactions and the optimal conditions of these processes, a specific database, including the description of a simplified B₂O₃-Na₂O-SiO₂ glass melt, has been implemented using the Calphad method. This calculation tool allows to describe the phase equilibria and the phase separation phenomena between either platinoids or molybdates (Fig. 1) – mainly CaMoO₄ and Na₂MoO₄ – and the glass melt. In parallel to a direct comparison between thermodynamic calculations and experimental results, this database is also used for a phase field approach. The objective of this coupling is to provide a consistent formalism between the thermodynamic properties of the chemical system and the local dynamics of the interface between the separate liquid-liquid phases of a heterogeneous glass.

Reference: [1] M. Stempok et V. Jan, Homogenni Skla V Systemech," Silikaty 1, 19 (1974).