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Redox behavior of ruthenium in borosilicate glass melts for nuclear waste vitrification

Cloé Laurin^{1*}, Elise Régnier¹, Stéphane Gossé², Michael Toplis³ and Olivier Pinet¹

¹CEA, DES, ISEC, DPME, Univ. Montpellier, Marcoule, Bagnols-sur-Cèze, France

²CEA, DES, ISAS, Univ. Paris-Saclay, Saclay, Gif-sur-Yvette, France

³Observatoire Midi-Pyrénées, Univ. Toulouse III Paul Sabatier, Toulouse, France

*cloe.laurin@cea.fr (corresponding author)

Most of radionuclides are soluble in high-level waste borosilicate glass and are integrated in the vitreous network. However, Ruthenium and Palladium are poorly soluble and form RuO₂ particles and Pd-Te beads (Fig. 1a) during the vitrification process. Ruthenium is highly conductive and forms needle-shaped particles that may change the physical properties of the glass melt. Moreover, particles containing Ru⁰ are sometimes observed in nuclear glasses (Fig. 1b). These particles are then bigger and denser, and Ru⁰ is more conductive than RuO₂. Therefore, the unexpected reduction of RuO₂ in Ru⁰ may have a significant impact on the physical properties of the melt and hinder the vitrification process.

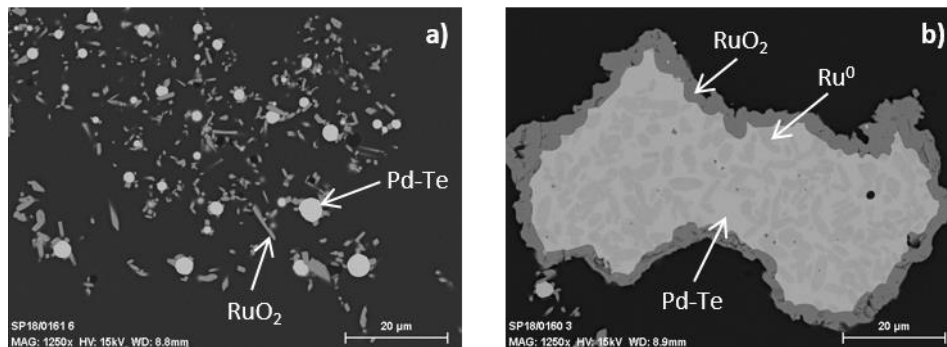


Fig. 1: SEM pictures of (a) RuO₂ needles and Pd-Te beads and (b) a Ru⁰-containing particle in a simulated nuclear glass

Hence, the objective of this study is to better understand and predict the redox behavior of ruthenium in nuclear borosilicate glasses. A dual experimental and modelling approach is adopted: experimental results are compared to thermodynamic calculations by a Calphad model.

RuO₂ is mixed with a simplified borosilicate glass and heat-treated at different temperatures. The reduction of RuO₂ into Ru⁰ (observed after cooling) in this glass is in agreement with thermodynamic calculations [1]: when the oxygen pressure (p(O₂)) of the glass melt at the considered temperature is equal to the calculated equilibrium p(O₂) between RuO₂ and Ru⁰, both ruthenium forms coexist. At higher oxygen pressure, only RuO₂ is observed. By adding Pd and Te to the glass melt, Ru⁰ is observed even when the oxygen pressure of the glass corresponds to the RuO₂ thermodynamic domain. The solubility of Ru in Pd-Te liquid must be responsible for the reduction of RuO₂ into Ru⁰ and needs further study.

References –

[1] Gossé S *et al.* Ruthenium Thermodynamics in Nuclear Waste Glasses. MRS Proceedings. 2011;1369:mrss11-1369-xx02-07. doi:10.1557/opl.2012.508