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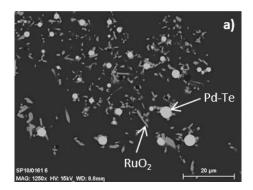
Redox behavior of ruthenium in borosilicate glass melts for nuclear waste vitrification

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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 RuO_2 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 Ru^0 are sometimes observed in nuclear glasses (Fig. 1b). These particles are then bigger and denser, and Ru^0 is more conductive than RuO_2 . Therefore, the unexpected reduction of RuO_2 in Ru^0 may have a significant impact on the physical properties of the melt and hinder the vitrification process.



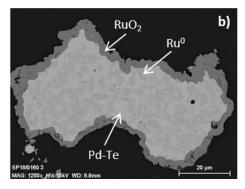


Fig. 1: SEM pictures of (a) RuO₂ needles and Pd-Te beads and (b) a Ru⁰-containing particle in a simulated nuclear glass

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.

 RuO_2 is mixed with a simplified borosilicate glass and heat-treated at different temperatures. The reduction of RuO_2 into Ru^0 (observed after cooling) in this glass is in agreement with thermodynamic calculations [1]: when the oxygen pressure $(p(O_2))$ of the glass melt at the considered temperature is equal to the calculated equilibrium $p(O_2)$ between RuO_2 and Ru^0 , both ruthenium forms coexist. At higher oxygen pressure, only RuO_2 is observed. By adding Pd and Te to the glass melt, Ru^0 is observed even when the oxygen pressure of the glass corresponds to the RuO_2 thermodynamic domain. The solubility of Ru in Pd-Te liquid must be responsible for the reduction of RuO_2 into Ru^0 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