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

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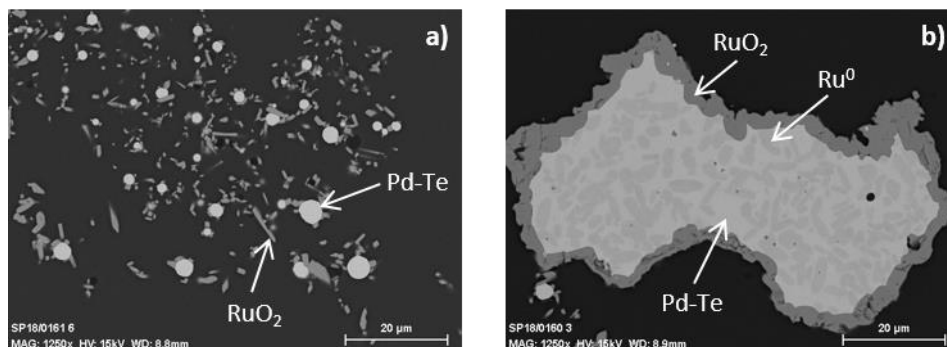
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Most of radionuclides are soluble in high-level waste borosilicate glass and are integrated in the vitreous network. However, Ruthenium and Palladium are poorly soluble and form RuO<sub>2</sub> particles and Pd-Te beads (Fig. 1a) during the vitrification process. Ruthenium is highly conductive and forms needle-shaped particles that may change the physical properties of the glass melt. Moreover, particles containing Ru<sup>0</sup> are sometimes observed in nuclear glasses (Fig. 1b). These particles are then bigger and denser, and Ru<sup>0</sup> is more conductive than RuO<sub>2</sub>. Therefore, the unexpected reduction of RuO<sub>2</sub> in Ru<sup>0</sup> may have a significant impact on the physical properties of the melt and hinder the vitrification process.



**Fig. 1:** SEM pictures of (a) RuO<sub>2</sub> needles and Pd-Te beads and (b) a Ru<sup>0</sup>-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<sub>2</sub> is mixed with a simplified borosilicate glass and heat-treated at different temperatures. The reduction of RuO<sub>2</sub> into Ru<sup>0</sup> (observed after cooling) in this glass is in agreement with thermodynamic calculations [1]: when the oxygen pressure (p(O<sub>2</sub>)) of the glass melt at the considered temperature is equal to the calculated equilibrium p(O<sub>2</sub>) between RuO<sub>2</sub> and Ru<sup>0</sup>, both ruthenium forms coexist. At higher oxygen pressure, only RuO<sub>2</sub> is observed. By adding Pd and Te to the glass melt, Ru<sup>0</sup> is observed even when the oxygen pressure of the glass corresponds to the RuO<sub>2</sub> thermodynamic domain. The solubility of Ru in Pd-Te liquid must be responsible for the reduction of RuO<sub>2</sub> into Ru<sup>0</sup> 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