

Establishment of a thermodynamic database for the solution chemistry and solubility of Eu(III) inorganic species

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Performance assessments of geological repositories for the underground disposal of high-level radioactive waste require a deep understanding of the phenomena influencing the mobility of radionuclides, e.g. sorption, redox immobilization, surface precipitation, incorporation, etc. Reliable thermodynamic databases (TDB) are required in order to generate speciation calculations, surface complexation and reactive transport models to predict the aforementioned mechanisms. In this work, the focus was set on europium (Eu), a lanthanide used for decades as a chemical analogue of trivalent actinides (Pu, Am). Indeed, a consolidated and internationally recognized Eu TDB does currently not exist.

Several reviews and reports [1-4] on the aqueous chemistry/geochemistry of europium were published, but had several drawbacks, for example:

→ Insufficient transparency about the selection procedure,

→ Lack of systematic screening to gather primary literature sources,

→ Too high reliance on the analogy with trivalent actinides,

→ For weak complexes such as chloride and nitrate, changes in the activity coefficients due to the replacement of up to 100 % of the background electrolyte anion by Cl⁻ or NO₃⁻ was either completely overlooked or, if recognized, not handled properly,

→ Too high reliance on the charge analogy for the estimation of missing ion interaction coefficients when the Specific ion Interaction theory was applied.

This study aims at significantly improving the situation by carefully addressing all aforementioned issues in order to provide a reliable, robust, and internally consistent TDB for europium. Recently, results of our critical evaluation for the chloride, sulphate, and phosphate ligands were published. The recommended complexation constants and solubility products for further inorganic ligands, e.g. hydroxide and carbonate, will also be presented [6].

[1] P.L. Brown, C. Ekberg, Hydrolysis of Metal Ions. Vol. 1, Wiley-VCH, Weinheim, **2016**.

[2] W. Hummel, et al., Nagra/PSI Chemical Thermodynamic

Data Base 01/01, Technical Report 02-16, **2002**.

[3] J.A. Rard, Chemical Reviews, 85(6) (**1985**) 555-582.

[4] K. Spahi, J. Bruno, A selected thermodynamic database for REE to be used in HLNW performance assessment exercises, SKB Technical Report, **1995**.

[5] N. Jordan et al., Coordination Chemistry Reviews, 473, 214608 (**2022**).

[6] N. Jordan et al., Coordination Chemistry Reviews, (**2023**) (in preparation).