SOLUBILITY AND AQUEOUS SPECIATION OF Ln(III) AND An(III) IN DILUTE TO CONCENTRATED AQUEOUS NITRATE AND SULFATE SYSTEMS: PITZER AND SIT MODELS AT 25 °C

PEDRO FELIPE DOS SANTOS1, XAVIER GAONA1, ARNAULT LASSIN2, KRASSIMIR GARBEV1, ANDREJ SKERENCAK-FRECH1, STÉPHANE TOUZELET2, YOHANN CARTIGNY3, MARCUS ALTMAIER1 AND BENOIT MADÉ4

1Karlsruhe Institute of Technology
2BRGM
3Univ Rouen Normandie
4Andra

Presenting Author: pedro.santos@kit.edu

Taking into account the similarities in ionic radii and equal charge, Eu(III) is commonly considered to be an inactive and redox-stable analogue of trivalent actinides (An) expected in nuclear waste. Thus, thermodynamic and activity models capable of describing the solubility and ionic interactions of Eu(III) in dilute to concentrated solutions of nitrate and sulfate can provide relevant information for the description of An(III) under relevant conditions for waste disposal [1]. In this work, comprehensive solubility experiments for the Eu3+-Na+-Mg2+-SO42−-NO3− aqueous systems were combined with advanced spectroscopic techniques (TRLFS) and a multimethod approach for solid phase characterization in order to derive complete thermodynamic and (SIT, Pitzer) activity models for these systems.

Diluted to concentrated aqueous solutions containing Na2SO4, NaNO3, MgSO4 and Mg(NO3)2 were prepared and contacted with the binary, well characterized solid phases Eu3+(SO4)3·8H2O(cr) and Eu(NO3)3·6H2O(cr). Batch undersaturation solubility samples under air conditions (3.3 < pH < 5.8) were continuously stirred, and the concentration of Eu(III) in the liquid phase was monitored by ICP-OES until attaining equilibrium conditions. Solid phases were characterized (XRD, Raman and TG-DTA) after terminating the solubility experiments. As reported for other lanthanides, the formation of the double salt Na2Eu2(SO4)4·2H2O (cr) was observed above ~0.01 M Na2SO4.

The new solubility and spectroscopic experimental data obtained in this study for the Eu3+-Na+-Mg2+-SO42−-NO3− aqueous system, in combination with previously reported solubility and isopiestic data, are considered to derive the thermodynamic and (SIT, Pitzer) activity models for these systems. The implemented optimization routine combines PEST, a parameter estimation software, PhreeqC [2] or PhreeSCALE [1] for chemical equilibria calculations, and the dedicated database. The ThermoChimie database is considered as basis for the development of the SIT models [3].

Acknowledgements: this work was partially funded by ANDRA.

References