

## **Speciation of Uranium: Validation of a thermodynamic database by modelling techniques and comparative experiments**

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To predict radionuclide migration, for installing effective water purification technology and as basis for decisions for remediation strategies of the contaminated sites a profound knowledge of uranium speciation in these waters is essential. Predictive modelling of uranium speciation in natural waters uses adequate speciation software and equilibrium thermodynamic data whose validity is of utmost importance. Therefore, a comprehensive and consistent database was established according to current knowledge. The uranium data included in the database is based on the NEA TDB<sup>[1]</sup> and is modified or supplemented as necessary. The success of this approach was validated by comparative experimental investigations and model calculations (PHREEQC<sup>[2]</sup>) for several model systems. The waters differ in pH (2.7 – 9.8), uranium concentration ( $10^{-9}$  -  $10^{-4}$  mol/L) and ionic strength (0.002 - 0.2 mol/L). We used chemical extraction experiments, ESI-Orbitrap-MS and time-resolved laser-induced fluorescence spectroscopy (TRLFS) to measure the uranium speciation. For all investigated systems at least the two species with the greatest expected relative abundance were detected. The database is consistent and can be considered validated for the model systems and comparable waters.

[1] R. Guillaumont et al., Update on the Chemical Thermodynamics of U, Am, Np, Pu and Tc Elsevir, Amsterdam, 2003.

[2] D. L. Parkhurst and C. A. J. Appelo, in Water-Resources Investigations Report, 1999.