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

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Uranium mining legacies and enhanced radioactivity in usable groundwater and surface water resources cause environmental hazards. To predict radionuclide migration, for installing effective water purification technology and as decision basis 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 equilibrium thermodynamic data and adequate speciation software. However, available thermodynamic data sets differ considerably with respect to number of uranium compounds and with regard to completeness and consistency. Hence, model results may vary widely depending on the used data set^[1]. In addition, data for radionuclides published in recent years have only been partially integrated into the databases.

The validity of the thermodynamic data for the environmental media affected by uranium mining legacies 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^[2] and is modified or supplemented as necessary e.g. for calcium and magnesium uranyl carbonates. The specific ion interaction theory^[3], which is sufficient for the considered low ionic strengths, is used to estimate activity coefficients. The success of this approach was validated by comparative experimental investigations and model calculations (PHREEQC^[4]) 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. The latter method is nonintrusive and therefore does not change the chemical composition of the investigated waters. This is very important, because any change of the system under study may also change the speciation. For all investigated systems at least the two species with the greatest expected relative abundance were detected. Moreover, all species with expected concentrations $> 1 \cdot 10^{-7}$ mol/L were observed. The present database is consistent and can be considered validated for the model systems and comparable waters, whereas for waters of very different composition an additional validation is necessary. The validity range of the database generally extends over a wide range of compositions, pH (2-12) and also to higher ionic strengths (up to 3 mol/L).

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