THEREDA – A thermodynamic database for increasing confidence in waste disposal

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Motivation and Implementation

For geochemical modeling of scenarios for the disposal of radioactive and (chemo)toxic waste, comprehensive and internally consistent thermodynamic data are required as well as sorption data for the surrounding host rocks. The use of different databases renders it difficult to compare results of geochemical modeling, due to incompleteness, inconsistencies, restricted ranges of variation (temperature, density, pressure), limitations in solution composition (ionic strength) and last but not least missing sorption data.

Features of the Thermodynamic Database THEREDA

THEREDA (THErmodynamic REference DAtabase, www.thereda.de) – a cooperative project of leading research institutes in Germany – adresses these issues, providing full documentation, transparency of all data and a detailed quality assurance scheme [1,2]. THEREDA contains data for the three ion-ion interaction models extendend Debye-Hückel, Specific Ion Interaction Theory, and Pitzer model. At present, two datasets has been released: the oceanic salt system (Na^+, K^+, Mg^{2+}, Ca^{2+}, Cl^-, SO_4^{2-}, H^+, and H_2O(l) within a temperature range of 273.15–523.15 K) and Am/Nd/Cm (Am(III), Nd(III), Cm(III), Na^+, Mg^{2+}, Ca^{2+}, Cl^-, H^+, H_2O(l) at 273.15 K). Tailored parameter files for various geochemical modeling codes are provided (PhreeqC, ChemApp, EQ3/6, Geochemist's Workbench). Documented benchmark calculations allow comparisons with other databases as well as between the different geochemical codes. All data, documentation and references are freely accessible via the projects homepage. Additionally, a user forum allows direct contact with the THEREDA members.

Integration of Sorption Data of RES³T

A holistic view of geochemical processes in the context of a safety analysis requires the inclusion of sorption calculations. A thermodynamically consistent treatment of these processes is only possible with surface complexation modeling (SCM). Respective data are already compiled in the RES³T database [3] (www.hzdr.de/res3t), providing competing entries for many systems. An integration into THEREDA thus not only requires a synchronization of data structures but also a rigorous review process, leading to uniform recommended data sets for each sorbent-sorptive system. This data review process is already in progress.

[1] Altmaier *et al.* (2011) *Report GRS-265*, 63 p. [2] Altmaier *et al.* (2008) *ATW* **53**, 249-253. [3] Brendler *et al.* (2003) *J. Contam. Hydrol*. **61**, 281–291.

Applications of Synthetic Uranium Reference Materials for Geochemistry Research

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Introduction

For many applications in geochemistry research isotope ratio measurements play a significant role. For instance, in geochronology isotope abundances of uranium and its daughter products thorium and lead are being used to determine the age and history of various samples of geological interest. For measuring the isotopic compositions of these elements by mass spectrometry, suitable isotope reference materials are needed in order to validate measurement procedures and to calibrate Faraday cup multi-collector and ion counting detector systems. IRMM is a well recognized provider for nuclear isotope reference materials to the nuclear industry and nuclear safeguards authorities, which are also being applied widely for geochemical applications.

Synthetic Uranium Reference Materials and their Applications

The preparation of several new synthetic uranium reference materials at IRMM during the recent five years has provided significant impacts on geochemical research. These synthetic isotope reference materials are prepared based on proven methods of purifying and gravimetrically mixing oxides or solutions from isotopically highly enriched uranium materials.

Firstly, the double spike IRMM-3636 [1] with a $^{233}U/^{236}U$ ratio of about 1:1 was prepared which allows an internal mass fractionation correction for high precision $^{235}U/^{238}U$ ratio measurements. The ^{234}U abundance of this double spike material is low enough to allow an accurate and precise correction of $^{234}U/^{238}U$ ratios, even for measurements of close to equilibrium uranium samples. This double spike has been used successfully for characterizing uranium isotopic ratios in various materials of geochemical interest [2] as well as natural consensus standards [3], i.e. NBL CRM 112A [4], and was found very valuable for U-Pb and U-Th geochronology.

Secondly, the application of the IRMM-074/1-10 series of isotopic of isotope reference materials [5] for linearity investigations of secondary electron multiplier (SEM) detectors will be discussed and new procedures introduced. This is of high relevance for U-Pb and U-Th geochronology by isotope mass spectrometric methods.

A general and historical overview about the preparation and certification of uranium isotope reference materials at IRMM will be given, including plans for future certification projects.

[1] Richter (2008), Int. Journal of Mass Spectrometry, **269**, 145–148

[2] Brennecka (2010), Earth and Plan. Science Letters, **291**, 228–233

[3] Condon (2010), Geoch. et Cosmoch. Acta, **74**, 7127–7143

[4] Richter (2010), Int. Journal of Mass Spectrometry, **295**, 94–97

[5] Richter (2009), Int. Journal of Mass Spectrometry, **281**, 115-125.