Uranium(VI) complexation with lactate and citrate in dependence on temperature (7-65°C)

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After disposal in nuclear waste repositories the chemical and migration behavior of actinides depends on many factors. It is estimated that maximum temperatures in the near field of a repository could reach 300°C in dependence on the waste forms [1] and the host rock [2]. Thus, for the long-term safety assessment, knowledge of the interaction of actinides such as uranium with inorganic and organic ligands at elevated temperatures is required. The amount of organic matter in a repository can be separated in humic substances and in low molecular weight organic substances. A not negligible component of low molecular weight organic substances is the group of carboxylic acid. For example, citric acid is used in nuclear reprocessing [3] and acetic, lactic and formic acid were identified in rock extracts and pore water of Opalinus Clay [4]. Reliable experimental data on the complexation of U(VI) in solution at elevated temperatures are still needed.

Therefore, we studied the U(VI) complexation by lactic acid (pH 3) and citric acid (pH 0-10) in the temperature range from 7 to 65°C. Species distribution and complex formation constants were determined by means of UV-Vis and time-resolved laser-induced fluorescence spectroscopy. In the U(VI) lactate system, we identified the formation of 1:1- and 1:2-complexes. In the presence of citrate, we could characterize five U(VI) complexes in dependence on pH value. The complex formation between U(VI) and these both ligands was found to be endothermic and entropy-driven. The complex stability constants of the U(VI) complexes increase with increasing temperature. This could lead to an increased mobility of U(VI) at higher temperatures.

[1] Rao, Jiang, Zanonato, Di Bernardo, Bismondo & Garnov (2002) *Radiochimica Acta* **90**, 581–588. [2] Warwick, Hall, Zhu, Dimmock, Robbins, Carlsen & Lassen (1997) *Chemosphere* **35**, 2471–2477. [3] Dodge & Francis (1997) *Environmental Science & Technology* **31**, 3062–3067. [4] Courdouan, Christl, Meylan, Wersin & Kretzschmar (2007) *Applied Geochemistry* **22**, 2926–2939.

Early differentiation of terrestrial planets: The relative importance of big impactors and small impactors

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Planetary accretion arises through discrete events but is often treated as if those events were so frequent and large in number as to produce a continuum (a mass flux and characetristic time interval of accretion). Although there have also been published models that study the effects of discrete large impacts, insufficent attention has been paid thus far to the differences that arise for delivering material through a large number of small bodies and delivering the same mass (at the same epoch) as a small number of large bodies (e.g. Earth or Moon-sized giant impactors). The nature of the impacts is different in respect of the depth to which melting takes place, the extent to which equilibrium is establised between core forming materials and mantle materials, and the pressure range of equilibration. As a result, two accretion scenarios that involve identical sequences (timing and size) of imapct events can have different outcomes (different Hf/W inferred chronologies, different mantle siderophile inventories, different initial core T, etc) according as to the physical models one uses to describe the outcome of different sized impacts. Contrary to what some of the current literature implies, this is *not* merely the issue of the timing of the last giant impact since multiple giant impacts can be responsible for a large fraction of the mass delivery. It is also true that one cannot ignore the background 'drizzle' of smaller bodies (implied by the dynamic friction in the Nice models for example). I will describe semi-quantitatively the range of outcomes according as to the range of possible accretion histories for the terrestrial planets in general but with particular emphasis on Earth. This will also require an update of our current understanding of the mechanisms of core formation and the nature of melting curves and magma oceans in the deep mantle.