Workshop on

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Uranium speciation in biological media : A selection of thermodynamic data from OECD/NEA , IUPAC and NIST databases

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The knowledge and understanding of transfer mechanisms for radiotoxic elements in human body requires information on their chemical speciation, which may involve free or complexed soluble ionic or neutral species, adsorbed species, precipitates such as (hydr)oxides or colloids.
Uranium is an actinide for which the biokinetics of the main compounds (oxides, fluorides, nitrates) have been studied after inhalation and ingestion (ICRP 69), but whose speciation in biological media is not yet very well known. In aqueous media such as blood, cellular liquids, saliva, gastric juice, uranium usually described at the oxidation state +VI, as the uranyl ion, UO_2^{2+}, is mainly complexed with carbonate, citrate and transferrin. However the presence of precipitated uranium phosphates in lung macrophages and kidney cells has occasionally been reported and may indicate the existence of uranium at the oxidation state +IV under the considered conditions, as typically observed in environmental conditions.
This work presents a basic thermodynamic database BASSIST (Base Applied to Speciation in Solution at Interface and SolubiliTies) selected and used by a French working group (GT 32) for investigating solution, precipitation and sorption reactions. This database was completed from OECD / NEA, IUPAC and NIST databases, as well as from CEA original data. The main ligands of interest include carbonates, phosphates, citrates, fluorides, chlorides, some amino acids and proteins; information on competitive ions such as calcium, potassium, sodium and magnesium is also supplied.
Three examples of modelling, using CHESS software (Chemical Equilibrium with Species and Surfaces) illustrate our theoretical approach. They are related to the speciation of uranium (IV) and uranium (VI) in three aqueous media simulating blood serum (pH~7,4), saliva (pH from 5,6 to 7,6) and gastric juices (pH from 1,5 to 4,0), respectively. In particular, speciation diagrams as a function of pH are shown to be useful to explain the early precipitation of uranium phosphates under certain biological conditions. The influence of redox potential on species distribution and kinetics aspects are also discussed.
The main interest of our theoretical approach is to allow the biologist to select relevant experimental conditions, to help in the interpretation of *in vivo* or *in vitro* experiments, and more generally to show to which extent modelling can be used in a predictive manner.–