Contribution ID: **920** Type: **Verbal**

The use of 152Eu(III) towards the elucidation of Pu(III) behavior with phosphates

Tuesday, 17 May 2022 11:40 (20 minutes)

Phosphate species from natural (e.g. phosphate mineral like apatite) as well anthropogenic (e.g. fertilizers) sources are ubiquitous in the environment [1]. Phosphoric acid is also present in the PUREX process as ultimate degradation product of tributylphosphate [2]. Phosphate species are expected to strongly interact with plutonium ions, plutonium being one of the main environmental pollutants associated with nuclear power generation and nuclear weapon testing [3]. In order to improve the models of Pu chemical behavior in the presence of phosphate species, it is necessary to collect fundamental data on interaction of Pu3+, Pu4+, PuO2+, and PuO22+ ions with these inorganic ligands. However, due to difficulties in handling Pu (redox properties, radiotoxicity), the aim is first to develop an experimental protocol using analogs in the same oxidation state[4]. The present work focuses on the determination of, on one hand, the speciation of Eu(III) at ultra-trace scale using 152Eu in the presence of H2PO4- and, on the other hand, of the solubility product of Nd-based rhabdophane.
The partition of 152Eu in the system HDEHP/Toluene/NaCl/HCl/NaH2PO4 has been studied at constant ionic strength and temperature. The mean composition of the formed complexes and their associated stability constants have been deduced from the variations of the distribution ratio of 152Eu as function of phosphate concentration.
Concerning the solubility product of PuPO4, stable rhabdophane Nd0.99Eu0.01PO4·nH2O and the 152Eu doped one have been synthesized. The stable solid has been characterized by XRD, TGA and SEM, and its dissolution has been proved to be congruent. Dissolution experiments have been then conducted on the doped phosphate compounds reducing the amount of solid (from 50 to 2 mg). The process was monitored by γ-spectrometry. The same behavior was observed in all the studied systems: the equilibrium is reached in about ten days and the solubility product obtained in these conditions agrees with literature data [5]. This validates the use of a small amount of PuPO4 in future dissolution experiments.

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Session Classification: Actinoids and Transactinoids

Track Classification: Chemistry of Actinide and Trans-actinide Elements