Determination of microscopic interactions between plutonium and humic substances

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Large amount of plutonium has been introduced into the environment as a result of nuclear weapons testing, and nuclear power-plant accidents. Contaminated areas, which need a particular survey, have become a very interesting place to study and understand the plutonium behaviour in the environment. Until few years ago, it was admitted that plutonium introduced into subsurface environment is relatively immobile, owing to its low solubility in ground water and strong sorption onto rocks. However, studies of contaminated areas show that humic substances, which are ubiquitous in environment, can alter the speciation of metal ion, e.g., plutonium, and thus their migration. These humic substances are major components of the natural organic matter in soil and water as well as in geological organic deposits such as lake sediments, peats and brown coals. They are complex heterogeneous mixtures of polydispersed supra-molecules formed by biochemical and chemical reactions during the decay and transformation of plant and microbial remains.

The knowledge of the impact of humic substances on the plutonium migration is required to assess their transport in natural systems. However, due to the complex and heterogeneous nature of humic substances, there are a lot of difficulties in the description of microscopic interactions.

The aim of this PhD thesis is to evaluate as precisely as possible interactions between plutonium and humic substances. This work is divided in two parts: on the one hand humic substances will be separated to identify each component, on the other hand the speciation of plutonium with characterized humic substances will be studied.

In the first part of this study, a new way of humic substances separation had been developed using an electrophoresis capillary apparatus. This efficient separation technique allows to simplify the characterisation of the complex heterogeneous mixtures without changing speciation of the chemical system.

In the second part, new methods are developed to study the speciation of plutonium with humic substances using two kinds of mass spectrometers: an ICP-MS and a high resolution mass spectrometer using various ionisation devices (ESI, APCI, DART, APPI) in order to determine all active molecules for the complexation.

This study will lead to evaluate which parameters are responsible for complexation of plutonium by the humic substances.