

Effect of ionic strength on the mobility of radionuclides in the presence of natural organic matter: Testing the Linear Additive Model

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Interaction with natural organic complexants such as humic substances can be decisive for the mobility of radiotoxic metals in case of release from an underground repository. Depending on the geochemical surroundings, their migration can be both enhanced and retarded. Models must be able to describe such complex systems by few parameters. According to the Linear Additive Model [1], total metal adsorption in the presence of humic matter is calculated by linking parameters for adsorption of both components and for their interaction with each other. The applicability of this approach is, however, not unanimously accepted.

Clay rock is discussed as a potential host formation for a final repository, mainly because of its high adsorption capacity. This barrier function may, however, be subverted by screening or competition effects due to high salt contents of pore waters. Complexation of radionuclides with humic-like clay organics may cause an additional mobilisation.

In this study, the influence of electrolyte concentration (up to 4 M) on interactions within the system Tb(III) / fulvic acid (FA) / Opalinus clay was investigated for the major electrolyte constituents of pore waters: Na^+ , Mg^{2+} and Ca^{2+} . ^{160}Tb was employed as a tracer analogue of trivalent actinides. FA (humic matter) was radiolabelled with ^{14}C by an azo-coupling reaction. The sensitivity of radiotracer analysis allows experiments at low concentrations, in accordance with relevant scenarios. Complexation of Tb(III) with FA was investigated by means of ultrafiltration. For adsorption studies, clay suspensions were conditioned to a constant pH value of 5.0.

Effects of Na^+ , Mg^{2+} and Ca^{2+} proved to be very different. On the whole, interaction of Tb(III) with FA and clay is suppressed at increasing electrolyte contents, whereas interaction of FA with clay is promoted because electrostatic repulsion is screened. For this reason, mobilising effects of humic-like complexants are generally counteracted in saline systems; metals are solely mobilised due to common salt effects. For the bivalent electrolytes, these relationships can be quantitatively described by the Linear Additive Model.

[1] Zachara *et al* (1994) *GCA* **58**, 553-566.