Molecular-based study of the electric double layer in hydrothermal systems

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Overview

The main goal of this molecular dynamics study is to characterize the electric double layer on the microscopic level, which would provide rigorous links between specific microscopic details of the water/oxide interactions and its macroscopic manifestation and help the interpretation of ongoing experimental results (titration (Ridley, Machesky et al. 1999)., neutron scattering, X-ray diffraction (Fenter, Cheng et al. 2000)). Here we model the TiO2 surface on the atomistic level, starting from the ab initio obtained structures by Kubicki et al. Both hydroxylated and non-hydroxylated surfaces are studied. Aqueous solutions of RbCl and SrCl2 have been our main system of interest. The axial profiles of structural and electrostatic properties and the lateral distribution of water and ionic species has been analyzed to interpret surface adsorption and the structure of the interfacial layers. The dynamic behavior has also been studied in dependence on the distance from the surface.

References

Fenter, P., L. Cheng, et al. (2000) Journal of Colloid and Interface Science 225: 154-165.

Ridley, M. K., M. L. Machesky, et al. (1999) Geochimica et Cosmochimica Acta 63: 3087-3096.

Kubicki, J. et al (2002) Unpublished.

Geochemical and hydrodynamic controls on the accumulation of nuclear reprocessing wastes in coastal sediments

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Over the past five decades, authorised low-level discharges from coastal nuclear facilities have released a significant quantity of artificial radionuclides into the marine environment. In NW Europe, the majority of the total discharge has come from nuclear reprocessing activities at Sellafield in the UK and COGEMA-La Hague in France. At the Sellafield site, a significant inventory of historical discharges has been trapped in local coastal and estuarine sediments, and notably in offshore fine sediment deposits, and much research has been focussed on understanding the burial and reworking of long-lived radionuclides in these deposits. In contrast, there are few high-resolution published data on the vertical distribution of radionuclides in fine-grained estuarine sediments near to, and downstream of, COGEMA-La Hague. This is despite the presence of large areas of mudflat and saltmarsh in the estuaries of the Normandy coast, environments that are important sinks for radionuclides around the Sellafield facility and in other areas. This paper therefore examines the vertical distribution of a range of anthropogenic radionuclides in dated sediment cores from two estuaries, one adjacent to, the other downstream of, the COGEMA-La Hague pipeline (Havre de Carteret and Baie de Somme).

While the sediments examined are clearly labelled with COGEMA-La Hague-derived radionuclides, and retain a record of temporal changes in the COGEMA-La Hague discharges, the radionuclide activities observed are extremely low, particularly in comparison with salt marsh sediments near to the Sellafield facility. For example ^{241}Am in sediment from Barneville-Carteret, within 40km of the La Hague discharge point, has a maximum ^{241}Am activity of 14 Bq/kg, compared with estuarine sediments close to Sellafield (at Ravenglass, Cumbria) which can show ^{241}Am activities of more than 28,000 Bq/kg. The differences in uptake seen between French and UK sites reflect in part the mineralogy of the suspended and deposited sediment along with local hydrodynamic conditions. Given the low activities observed in this study for coastal sediments, it seems likely that the bulk of the particle-reactive radionuclides discharged from COGEMA La Hague are widely dispersed at low acitivities in the coarser-grained bottom sediments of the English Channel, rather than being concentrated in carbonate-rich coastal fine-grained sediments.