Sorption on to Samples of Permo-Triassic and Carboniferous Sedimentary Rock Samples from Boreholes 1, 2, and 10 at Sellafield

Type: NDA/Nirex Reports

Abstract:

As part of the Nirex Safety Assessment Research Programme (NSARP), a large number of batch-sorption experiments have already been carried out to study the sorption of uranium(VI), plutonium, thorium and radium. This work has concentrated on assessing the influence of organic degradation products on sorption under both near-field (pH-l2) and far-field (pH-8) conditions. Further experiments have now been carried out to study (i) uranium(VI) sorption onto a sample of basal breccia from the Brockram, (ii) selenium sorption onto a sample of sandstone from the St Bees Sandstone Formation, (iii) uranium(VI), plutonium, thorium and radium sorption onto a sample of limestone from the Carboniferous Limestone Group and (iv) radium sorption onto a sample of sandstone from the St Bees Sandstone Formation. All the rock samples were from the Sellafield site. These experiments were all performed under far-field conditions. For all the experiments except those studying sorption onto basal breccia, synthetic equilibrated groundwaters corresponding to the appropriate geological horizon were used. Since no groundwater composition was available for the basal breccia, in these experiments two different synthetic groundwaters were used appropriate for other horizons at Sellafield. Batch-sorption experiments studying uranium(VI) sorption onto the basal breccia gave RD values of 70 cm3g-1 from both tuff and sandstone groundwaters. The tuff groundwater water has sodium, calcium and chloride concentrations two orders of magnitude greater than the sandstone groundwater. However, the carbonate levels in the two groundwaters are very similar. Therefore, these experiments have demonstrated that, at a constant carbonate level, the ionic strength of the groundwater is not an important variable for uranium (VI) sorption. The batch-sorption technique has also been used to study the effect of redox potential on selenium sorption onto the sandstone. Under strongly-reducing conditions, (-390 mV versus standard hydrogen electrode) RD values of ~104 - >106 cm^3g^{-1} were obtained, compared to ~500 cm^3g^{-1} under non-reducing conditions (+360 mV to +490 mV versus standard hydrogen electrode). All these RD values are unexpectedly high for sorption of the predicted species, the anions, HSe^- and $HSeO_3^-/SeO_3^{2-}$. Through-diffusion experiments conducted under non-reducing conditions gave significantly lower RD values, ~ 4 cm^3g^{-1}. The lower RD values obtained using the through-diffusion methodology have been rationalised in terms of (i) restricted accessibility of selenite ions to small-scale pores within the sandstone sample and/or (ii) rock-catalysed oxidation of strongly sorbing selenite to weakly sorbing selenate in the through-diffusion experiments, due to the large rock to water ratio when compared to batch-sorption experiments. Batch-sorption studies of uranium(VI), plutonium, thorium and radium onto the limestone indicate that sorption onto the limestone is significantly weaker than that observed for other rock formations, RD values being <1 cm^3g^{-1}, ~400 cm^3g^{-1}, ~200 cm^3g^{-1} and <7cm^3g^{-1} for uranium(VI), plutonium, thorium and radium sorption respectively. The radium batch- sorption results were confirmed by through-diffusion experiments, which also showed no significant radium sorption. In addition to determining the extent of sorption from synthetic equilibrated groundwater, radium sorption onto the limestone was also measured from solutions containing a range of initial calcium concentrations ($1.0\ 10^{-2}$ -$5.0\ 10^{-2}$ M). These experiments were performed in order to determine whether competition with groundwater ions for sorption sites has an important influence on radium sorption, since there is a range of groundwater compositions at Sellafield and it is important to understand how sorption may vary across the Sellafield site. However, the extent of radium sorption in these experiments was so low that it was not possible to determine whether competition with groundwater ions for sorption sites has an important influence on radium sorption onto limestone. The batch-sorption technique was also used to measure radium sorption onto the sandstone from solutions of high ionic strength, sodium chloride solutions of 3-4.SM. These experiments were designed to complement earlier data investigating the effect of ionic strength on sorption. RD values of ~30 cm^3g^{-1} and <8 cm^3g^{-1} were measured for sodium chloride concentrations of 3M and 4.5M respectively. When compared with data obtained from previous work there is a clear trend towards lower RD values as the ionic strength is increased. Overall, these results have demonstrated that when selecting sorption parameters for use in performance assessment modelling, (i) the redox potential needs to be considered for selenium sorption and (ii) the ionic strength of the groundwater needs to be considered for radium sorption. However, for uranium(VI) sorption, the ionic strength of the groundwater is not an important variable. This report was prepared by AEA Technology under contract to United Kingdom Nirex Limited. The main technical work reported here was carried out in the period April 1993 to March 1994 and the report is based upon, and solely refers to, information available at that time. Thus, for example, where the report refers to `current knowledge' this should be read to mean `knowledge existing at the time the work was carried out'. This work forms part of the Nirex Safety Assessment Research Programme.

More Details

Report Number:	NSS/R383	Detailed	
Complexity:	No ISBN Number for this document	Berry, J.A., Bond, K.A., Brownsword, M., Green, A., Linklater, C.M.	
ISBN Number:	No Contractor for this document	2001	
Authors:	385362	6763935	
Contractor:			
Publication Year:			
PCDocs Reference Number:			
Livelink Reference Number:			