Modelling Radionuclide Transport in the Far Field of a Radioactive Waste Repository

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Introduction

In the UK long-term storage of radioactive waste in a deep underground geological disposal facility (GDF) has received significant attention. A large proportion of the overburden rock in the UK is sandstone, so an understanding of the sorption and transport behaviour of radionuclides in this type of material is essential to establish the long-term effects of deep burial of radioactive wastes.

This project examines simple chemically homogeneous materials such as quartz to establish the components of the sorption process, working towards more physically and chemically complex simulated rock systems. The physical and chemical heterogeneities of a rock system at different scales need to be modelled to account for parameters such as porosity. Our ultimate aim is to model the sorption and transport behaviour of various radionuclides with sandstone and other geological materials.

Transport modelling through the use of values derived from column experiments allows an understanding of scaling, taking into consideration physical and chemical components such as spatial variation and chemical retardation.

The scaling of the retardation factor of a chemical species is related to the variability of the sorption coefficients in the rock and mineral. Column experiments are used to establish these values to allow for challenges of upscaling to be overcome.

The K1-D model, developed at Manchester, a flexible research code which allows the inclusion of kinetic data alongside transport and chemical equilibria, will be used to model the kinetic parameters of the transport process determined through our column experiments.

Sorption studies

In addition to column experiments, static sorption studies (Figure 4) are being conducted to allow correlation between fixed volume and mass systems and simple flow systems. An example of an isotherm on the mineral jasper red is given in figure 1.

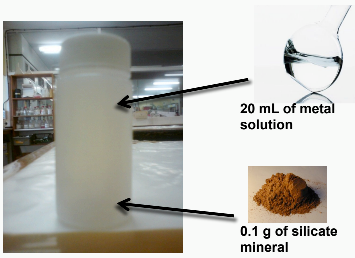

Figure 1: Sorption experiment setup

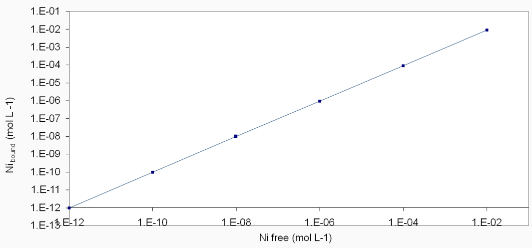

Figure 2: Nickel isotherm on jasper red.

We will be able to model the kinetic parameters of the transport process determined through our experiments as well as the stability constants of the speciated metals.

Column Experiments

The movement of Eu, Th, Cs, and Ni radionuclides will be examined in columns of *ca*. 6 cm packed with SiO_2 (figure 3). This silicate has a homogeneous extended structure and will allow initiation of the study. It is intended that our study will use (in increasing order of chemical and structural complexity): SiO_2, quartz, metal-doped quartzes, quartz/iron oxide, sand, synthetic sandstone and finally an intact sandstone core .

For the radionuclides under investigation we will examine:

- Effect of column packing – heterogeneity of the silicate system
- Flow rate of the radionuclide solution
- Column length
- Humic acid (HA) containing groundwater, to simulate the natural organic material present in soil
- Anthropogenic organic ligands (EDTA or cellulosic decomposition products) to chelate the metal ions

HA is a naturally occurring colloid with a high affinity for cations.
Water-soluble metal-HA complexes are important transport agents through the geosphere. For radionuclides in geological nuclear waste repositories, speciation with HA and mobility of these complexes are important phenomena to be understood. Where HA is used in our experiments, the column will be pre-equilibrated with HA, to achieve a uniform concentration along the column.

Figure 3: Operational column with fraction collector attached

Columns will be run by upflood / downflood techniques. Upflood will involve loading the column with metal ions, which are dissolved in aqueous $NaClO_4$. Downflood will involve pumping groundwater through the column. In some experiments, HA will be dissolved in the aqueous phase and used in both the upflood and downflood steps.

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