Investigating mechanisms to understand variability in radium isotopic activity ratios in natural environments

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Introduction

Radium isotopes are used as tracers in multitude of hydrogeological applications[1]. However, a complex mileu of geochemical and physical processes occurring in subsurface environment result in spatial and temporal variability in radium isotopic activities, including alpha-recoil, a primary mechanism of mobilization for multiple isotopes [3]. Numerous factors affect recoil processes, including spatial distribution of parent radionuclides within native materials, differences in nuclide recoil length in host matrix and physical structure of the rock strata.

Results

Here, we present an experimental method to measure radium recoil co-efficient (fraction of nuclide escaping grain-nuclide boundary) and analyze its variations as a function of pore structure and parent nuclide distribution within host matrices, including Marcellus shale rock (U= 19 ppm and Th=8.2 ppm) and Granite (U=3.33 %wt, Th=0.75% by wt, Serrie-Copper Pegmatite). Columns are packed with granulated rock matrices of varying surface area (40,000-60,000 cm^2/g) and subjected to low salinity solution in a close loop. Our data illustrates there is a ^{224}Ra recoil co-efficient for shale nearly 150% higher than granite. Experimental trends are related to mineralogical and microstructural analysis of host matrices using XRD, XAS and autoradiography. Our study elucidates the contribution of alpha recoil on the observed radium disequilibrium in subsurface environment. Further, we illustrate the effects of chemical and physical heterogeneity on the rate of recoil process, which may inform models predicting the fate and transport of radionuclides in subsurface environments.

Reference

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