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Determination of mineral-specific distribution coefficients of 133Ba from thin sections of granitic rock using filmless electronic autoradiography

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The final disposal of spent nuclear fuel will be performed in Finland in a geological repository in crystalline granitic rock at a depth of 400 meters [1]. The processes affecting the transport of radionuclides from the spent nuclear fuel to the biosphere need to be carefully taken into account when considering the overall long-term safety of the repository [2]. Consequently, it is necessary to study the sorption and diffusion properties of safety relevant radionuclides from laboratory scale all the way up to in-situ scale in the different barriers of the repository, for instance, the bedrock. In this study the mineral-specific sorption of 133Ba was studied in thin sections of veined gneiss and pegmatitic granite obtained from the in-situ site in Olkiluoto using filmless electronic autoradiography (i.e. the BeaverTM). The thin section samples were equilibrated with 1 mL of groundwater simulant made to resemble the fracture groundwater in the Olkiluoto site after which it was replaced with 1 mL of the same groundwater simulant with added 133Ba [3]. After a week, the groundwater simulant was pipetted from the thin section samples and it was measured with gamma spectrometry.

The spatial distribution of barium activity in thin sections was measured with two autoradiography methods; digital autoradiography using IP plate technique (Fuji 5100) and the BeaverTM, which is based on a gas detector incorporated by micromesh Parallel Ionization Multiplier (PIM) [4]. The BeaverTM allows real-time counting of charged particle emission from the analyzed surface with high sensitivity and linearity. Methylmethacrylate standards of 133Ba were made in order to determine the counting efficiency to derive activities of the measured count rates.

The distribution coefficients of 133Ba for main minerals were determined from the thin sections with the BeaverTM measurements and from the activity decrease in solution with gamma spectrometry. The mineral-specific distribution coefficients of 133Ba were found to decrease in the order: biotite > plagioclase > potassium feldspar > quartz, which was in good agreement with previous results [5]. The BeaverTM has proved to be an efficient method for quantifying heterogeneous sorption of sorbing radionuclides on rock materials. In addition, sorption experiments on thin sections provide a way to upscale distribution coefficients from crushed to intact rock. The sorption data obtained in this study will be utilized in the heterogeneous diffusion modelling as well as in the interpretation of results from the in-situ through diffusion experiments.

Primary authors: Mrs MUURI, Eveliina (Department of Chemistry, University of Helsinki); Ms HEZCKO, Vilma (Department of Chemistry, University of Helsinki); Dr DUVAL, Samuel (Ai4r); Dr SIITARI-KAUPPI, Marja (Department of Chemistry, University of Helsinki)

Presenter: Mrs MUURI, Eveliina (Department of Chemistry, University of Helsinki)

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