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Behaviour of Cs in Grimsel granodiorite: Sorption on main minerals and crushed rock

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INTRODUCTION

Estimating the sorption of caesium on mineral surfaces is important in risk assessment in the deep geological repositories. It is commonly considered that, in case of a leakage, the most important ways of retaining the caesium from the flowing fracture water are matrix diffusion into pore spaces and sorption onto the mineral surfaces. The aim of this study was to determine the distribution coefficients of caesium in different minerals with varying concentrations to support the interpretation of the caesium diffusion and sorption results in the upscaling of the experiments. The work is part of the Long-Term Diffusion (LTD) experiment (Fig.1) into Phase III (2014 – 2018) executed by the National Cooperative for the Disposal of Radioactive Waste, Switzerland.

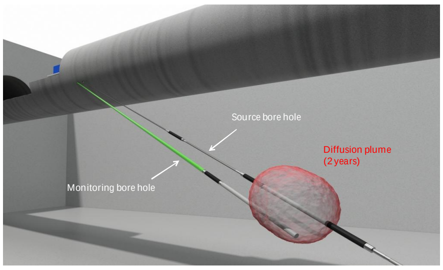

Fig 1. Experimental layout of Phase III of the in-situ test.

EXPERIMENTAL

The sorption of caesium onto quartz, potassium feldspar, plagioclase, biotite and granodiorite was investigated in a variable caesium concentration from $1.0 \cdot 10^{-9}$ M to $1.0 \cdot 10^{-3}$ M. All experiments we re conducted in room temperature and in the Grimsel groundwater simulant. Crushed minerals having grain size below 0.3 mm were first immersed into the groundwater simulant in liquid scintillation bottles after which the bottles were agitated for two weeks. The concentrated caesium solution was added to the samples after which they were again agitated for the ion exchange process to occur and reach completion. ^{134}Cs was used as tracer. The distribution coefficients are given as

$$K_d = \frac{c_l}{c_s}$$

where c_l is the caesium concentration in liquid and c_s is the caesium concentration in solid. The sorption of caesium on biotite was modelled with PHREEQC.

RESULTS

The distribution coefficients obtained from the batch experiments were largest on biotite (0.32 m^3/kg in 10^{-9} M caesium concentration) and plagioclase (0.15 m^3/kg in 10^{-9} M caesium concentration). The distribution coefficient of 0.038 m3/kg in 10^{-9} M caesium concentration was obtained on potassium feldspar. Furthermore, the sorption of caesium on quartz was found to be negligibly small in all investigated concentrations. Finally, caesium sorption behaviour in crushed granodiorite followed the trend of one of its most abundant mineral, plagioclase with distribution coefficient of 0.11 m^3/kg in 10^{-8} M caesium concentration. The results are presented in Fig 2.

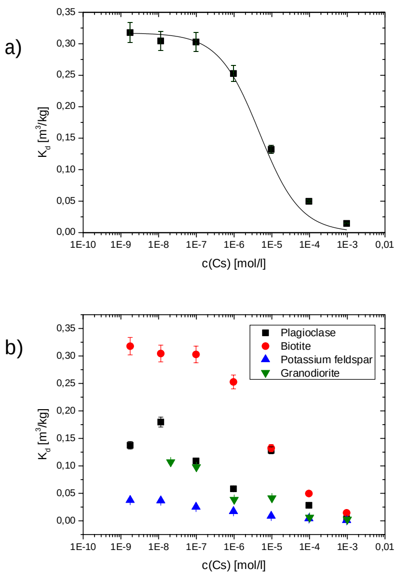

Fig 2. a) The sorption results of caesium on biotite with PHREEQC-modelling. b) The sorption results of caesium on plagioclase, biotite, potassium feldspar and granodiorite.

The sorption of caesium on biotite was found to behave differently in different concentrations. At low caesium concentrations (<10^{-6} M) caesium is sorbed mainly on the selective frayed edge sites of biotite. Once these sites are fully occupied caesium sorbs additionally to the Type II and Planar sites. Hence, at high caesium concentrations (>10^{-6} M) sorption is thus significantly reduced. The crystalline structure of biotite is presented in Fig 3.

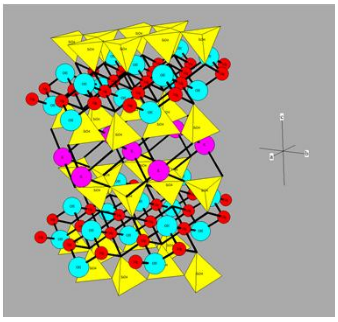

Fig 3. The crystalline structure of biotite.

CONCLUSIONS

The results from the batch sorption experiments were similar to and in good accordance with previous studies. The sorption results of crushed granodiorite were slightly bigger than the sorption results obtained from the in-situ –experiments conducted in the Grimsel test site. It is thus clear from these experiments that in order to fully understand the sorption and diffusion of radionuclides in rock it is necessary to take the heterogeneity of the rock and the in-situ conditions into account. The obtained sorption isotherms of caesium on different minerals will be used in the diffusion modelling which takes the minerals and the texture heterogeneity into account to further study the behaviour of caesium in Grimsel granodiorite.

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