Background analysis of actinide and ⁹⁹Tc tracers in the frame of an in-situ bentonite diffusion experiment at the Grimsel Test Site

FRANCESCA QUINTO 1 , INGO BLECHSCHMIDT 2 , THOMAS FAESTERMANN 3 , KARIN HAIN 4 , DOMINIK KOLL 5 , GUNTHER KORSCHINEK 3 , STEPHANIE KRAFT 1 , JOHANNA PITTERS 4 , MARKUS PLASCHKE 1 , GEORG RUGEL 6 , THORSTEN SCHÄFER 7 , PETER STEIER 4 AND HORST GECKEIS 1

¹Karlsruhe Institute of Technology (KIT), Institute for Nuclear Waste Disposal (INE)

Presenting Author: francesca.quinto@kit.edu

A bentonite engineered barrier will be used in deep geological repositories in granitic host rock to retard the transport of radionuclides possibly released from corroded canisters to the geo- and biosphere. Thus, it is relevant to investigate the diffusion of radionuclides through bentonite and their consequent migration through water-conductive features in granitic rock.

Such scenario in the case of glacial melt water intrusion has been simulated at the Grimsel Test Site with the Long-Term insitu Test (LIT). A packer-system containing bentonite rings spiked with radionuclide tracers was emplaced in the crystalline rock in contact with a water-conducting shear zone [1]. The bentonite pore-water mixing with Grimsel groundwater was collected at sampling points 5.6 cm from the bentonite for ca. 4.5 years. In these samples, we have determined the concentration of the actinide tracers ²³³U, ²³⁷Np, ²⁴²Pu, ²⁴¹Am and ⁹⁹Tc. The expected ultra-trace radionuclide concentrations could only be addressed via the high analytical sensitivity of Accelerator Mass Spectrometry. Actinide tracers were analyzed at the 3 MV tandem accelerator of VERA, while 99Tc at the 14 MV tandem accelerator of TUM, enabling quantification at the level of 25 at/g and 0.5 fg/g, respectively [2, 3]. In order to identify radionuclides originating from the LIT, an extensive analysis of the background originating from previous in-situ radionuclide tracer tests [2] was undertaken. First results suggest, that ²⁴²Pu release from the bentonite could not be detected. Concentrations of ²³³U, ²³⁷Np and ²⁴¹Am were found slightly above background for samples collected within 600 days. The levels of 99Tc were always above background indicating the possible release of ca. 8.5×10^{-3} % of the total ⁹⁹Tc within the experimental time frame. Such early appearance of tracers is unexpected. Further analysis is ongoing in order to identify the possible transport mechanism.

References:

- [1] NAB 14-48. GTS Phase VI CFM Phase 3. February 2015
- [2] Quinto et al., (2017) Anal. Chem. 2017, 89, 7182-7189
- [3] Quinto et al., (2019) Anal. Chem. 2019, 91, 4585-4591

Acknowledgments: German Federal Ministry for Economic Affairs and Energy, BMWi, grant number 02E11759B & international consortium of the CFM project

²NAGRA (National Cooperative for the Disposal of Radioactive Waste)

³Technical University of Munich (TUM), Physics Department

⁴University of Vienna, Faculty of Physics, VERA Laboratory

⁵The Australian National University, Research School of Physics and Engineering, Department of Nuclear Physics

⁶Helmholtz-Zentrum Dresden-Rossendorf, Accelerator Mass Spectrometry and Isotope Research

⁷Friedrich Schiller University Jena