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Visualisation and Quantification of Transport in Barrier Rocks with Positron Emission Tomography

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In tight barrier rocks laboratory observation of radionuclide transport and determination of transport parameters is a demanding and interminable task, because of slow rates, small concentrations, and intricate chemical interactions. The validity of results from common laboratory methods, like flow- and diffusion experiments on small samples, is limited by the heterogeneity of the pathways and adherent upscaling issues, because homogeneous conditions have to be presumed for these input-output investigations. But nano-pores or micro-fractures could be present, which would provide pathways for heterogeneous transport processes. Transport properties of these pathways are most influential boundary conditions for reactions between fluid components and crystal surfaces.

We propose Positron Emission Tomography (GEO-PET) as an appropriate method for direct observation of heterogeneous transport of radiotracers in tight material on the laboratory scale. With high-resolution PET scanners, which are common instruments of biomedical research ("small animal PET"), it is possible to determine the spatio-temporal distribution of the tracer activity with a resolution of almost 1 mm during about three periods of the tracer half-life (half-lives of some applicable PET tracers: ^{18}F: 1.8 h, ^{124}I: 4.2 days, ^{58}Co: 70.8 days). The PET tracer is applied as ion in solution or as marker for compounds, like colloids.

The most considerable difference between PET applications on geomaterial compared to biological tissue is the stronger attenuation and scattering of radiation because of the higher density of rock material. After travelling the positron attenuation length in dense material (about 1 mm), the positron annihilates in contact with an electron, transmitting two photons with 511 keV, propagating in antiparallel direction. The sample size of geomaterial is limited by the attenuation length of these photons. By applying an appropriate attenuation correction it is possible to investigate transport processes in rock cores with diameters up to 10 cm. Then at least 20% of the initial annihilation events are recorded as coincidences. However, one single photon of the annihilation radiation may be recorded while the other is absorbed; therefore, the signal to noise ratio is degraded by attenuation. Other sources of noise are scattered events, and the loss of one coinciding photon due to gaps between the detectors and other detection probability reasons. Also, the ratio of random coincidences increases with the noise level and impairs the image quality of the tomographic reconstruction. The reduction of these reconstruction artefacts by enhanced data correction methods is an important requirement for the development of the GEO-PET method. An other problem is the development of special methods for the quantitative evaluation of the extensive spatio-temporal data sets.

We present results from high-resolution PET for tomographic process observation during transport of colloids and conservative tracers in macroscopic samples of clays, saline rocks, and granites (diameter 5 to 10 cm, length 5 to 20 cm). In most cases we observed localized zones of transport, even in a homogenized compressed clay sample. This reflects the non-representative sample volume, which probably is not achievable for any laboratory method. However, at least the PET tomograms reveal these deviations from representativeness. Up to now, break-through-curve parameters can be determined from spatially resolved tracer concentration measurements at distinct regions of the sample, without mandatory penetration of the complete sample extension. A multiscale model-based inversion scheme for continuous scale-dependent parameter determination is currently developed.