Direct measurement of the flow field with GeoPET as the starting point for reactive transport modelling

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Introduction: Reactive transport simulations rely on appropriate conceptions of the flow field, in particular the distribution of the effective volume and the velocity distribution. These are frequently based on computational fluid dynamics conducted on structural µCT-images with the highest possible resolution. Highest resolution implies small sample volume or extremely large models. In contrast to these intensive computational methods, we strive to apply experimental methods that are able to analyze the flow field data directly. In addition, such approaches bridge the scale to macroscopic dimensions and improve the size of the reactive transport model. This approach is an application of positron emission tomography on geological materials (GeoPET).

Shortly after its clinical introduction, positron emission tomography was tentatively applied non-medical objects, including rocks [1, 2]. The capabilities of such experiments were limited by the low spatial and temporal resolution of this imaging method. Meanwhile, fast high-resolution scanners make possible positron emission process tomography with frame rates of about 1 minute and a spatial resolution of 1 mm, yielding quantitative movies of the tracer distribution in soils and rocks [3].

Spatially-resolved effective volume and velocity are experimental data derived from the spatiotemporal pattern of the concentration of the labelled compound, which is the most important variable in reactive transport modelling (RTM). The concentration is measured with molecular sensitivity and directly upscaled to the integration volume of 1 µL (i.e., the spatial resolution equivalent of the PET method). This spatial resolution is sufficient for models on the decimeter scale of drill cores.

The derivation of both the effective porosity and the velocity field from a series of tracer concentration maps is challenging. Measuring errors interfere with simple tracing algorithms and thus we developed a robust algorithm that takes into account the stochastic nature of the data. With this treatment it is now possible to calculate the spatial velocity distribution and the effective volume distribution with statistical confidence.

Such coupled porosity/ velocity parameter sets are the starting point of our reactive transport computations on the millimeter scale. Such “dynamic models” have been created and run with the coupled COMSOL Multiphysics® - Phreeqc – software system iCP [4]. Because of the limited model size and complexity, the expenses for development and computation are very small. Moreover, the step from experimental data to modelling is made at the stage of fluid dynamics, what is at a later phase of the reactive transport workflow than in common practice in digital rock physics, where fluid dynamics are simulated on the basis of structural images. The motivation is to minimize the deviation from reality. The readily manageable size of the models facilitates to study the impact of more sketchy scenarios, like the impact of interface properties or the balance of transport and kinetics.

Experimental Methods: Our GeoPET method applies a high-resolution PET-scanner (ClearPET by Elysia-raytest) in our dedicated radioisotope laboratory. Appropriate positron-emitting radionuclides are produced with an on-site cyclotron. These are injected into samples with typical dimensions of drill cores. PET then provides quantitative spatiotemporal records of the tracer concentration. Such quantitative GeoPET images are exceptionally sensitive to displacements of pico molar tracer quantities detected within 1 mm grids on laboratory/drill core scale. However, in spite of high selectivity and sensitivity, the effects of noise and detectability have to be considered.

We developed a robust algorithm for deriving flow process parameters in each voxel from the PET-records. The effective volume is derived from the measured maximum activity concentration; the flow vector field is computed from its temporal variations and the relation to the nearest neighbors. Apparent spatially intermittent flow, which occurs when the tracer concentration underruns the detection threshold, is cured with a maximum-likelihood procedure. The algorithm was validated on simple synthetic samples and successfully tested on fractured natural rocks.

The complete workflow is illustrated with leaching experiments on a fractured sandstone sample from a Kupferschiefer copper mine (Fig. 1). From the observed frames of the fluid flow process in the drill core (diameter: 70 mm, length: 100 mm) the effective porosity and velocity field is extracted by our robust parameterization algorithm. These 3D-fields were then imported into COMSOL Multiphysics®. Three dimensional RTM by means of iCP 1.3 (an interface coupling of the finite element based code COMSOL Multiphysics® with the geochemical code PhreeqC[4]) was performed to predict the leaching process and solute transport through the core sample by using kinetic mineral dissolution and precipitation data. The reactive transport model results are compared to and the model was iteratively refined using the analytical results of the column leaching experiments.

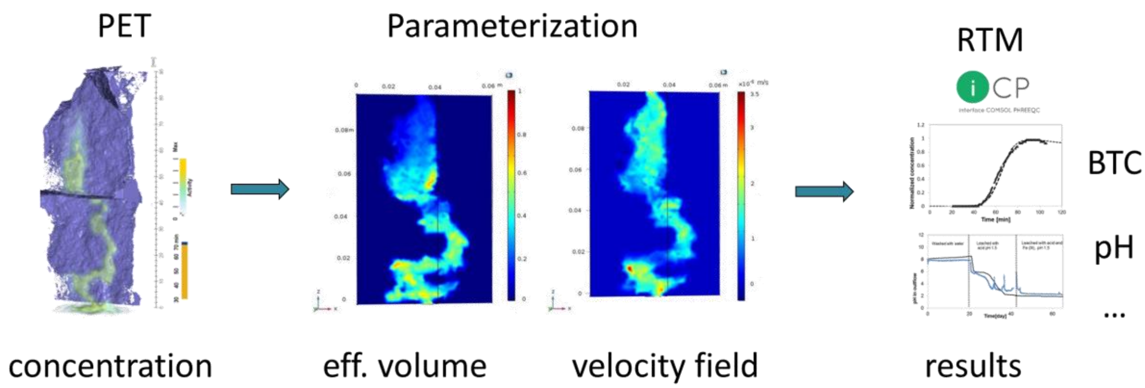

Figure 1: Workflow of the RTM-procedure based upon fluid dynamical parameters derived from process tomography with PET. From left to right: Experimental determination of tracer concentration with PET (here shown as PET/µCT overlay), parameterization of effective volume and velocity vector field, iCP-computation based on these fluid dynamical parameters.

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