**Title: Synchrotron Microtomography Investigation of Nanoparticle Entrapment and Mobility in Porous Media**

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Study of the mobility of nanoparticles in sand columns (or core flooding) at laboratory scale provides a good basis for understanding processes important in environmental engineering and hydrocarbon recovery from subsurface resources. Both of these applications can involve multi-phase fluid flow in subsurface porous systems. In hydrology, column experiments are used, among others, to support the design of in-situ groundwater remediation processes including those involving mostly reactive nanoparticles. In oil industry, core flooding experiments are used to study enhanced oil recovery processes including the use of mostly non-reactive nanoparticles. Both these applications deal with large-scale processes; however, the fluid flow actually happens at pore-scale (mostly micro-meter scale). This makes small scale laboratory experiments of special importance.

Conventional column experiments use information such as the concentration of the species of interest (in this case, particles) at column inlet and outlet, pressure, etc. to indirectly infer the pore-scale processes that occur in a sample during a flow and/or particle transport experiment. X-ray computed micro-tomography (X-ray micro-CT) allows a greater degree of understanding with regards to pore-scale dynamics through a process of 4D imaging (3D images resolved in time).

This study aims to investigate the mobility and entrapment of zero-valent iron nanoparticles (nZVI) in porous media. Two types of sand-packed columns with different grain size distributions (coarse and fine) were studied. The experiments started by saturating the column with water and a subsequent injection of the nanoparticle suspension. A post water flushing was done to remove the mobile nanoparticles. Using an X-ray transparent flow cell allowed capturing a sequence of 3D images during the experiments. In our previous studies only one field of view was imaged at every injection step (1, 2). A distinctive aspect of this experiment is that we decided to image the column in three segments by moving the micro-CT stage in Z direction. The three segments are then stitched together in the post-processing step. This gives us the chance to investigate the nanoparticle mobility profile along the entire column length at each time step, without compromising on the image resolution.

The collected images are filtered, segmented, and analysed to show the degree of nanoparticle mobility in these different samples, and to allow calculation of flow properties (e.g. permeability) based on the images collected before and after the nanoparticle injections. The existence of both mobile and non-mobile nanoparticle clusters was imaged in this experiment. At pore-scale, the trapped nanoparticles are mainly observed to occupy the pore-throats, i.e. the narrowest parts of the flow pathways. The injected nanoparticle suspension phase is not miscible with the water phase (initially occupying the pore-space) at first contact. Our images show the formation of a water film covering the sand grains in presence of the nanoparticle suspension phase, making water the wetting phase. The results of this experiment shed light on the pore-scale mechanisms involved in nZVI entrapment in porous media. Comparison of our findings for the coarse and fine sand samples will assist in improving the efficiency of nanotechnology-based groundwater remediation processes.

References

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