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## Nanoparticle heterogeneous adsorption in porous media

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Colloidal particles released by various chemical and industrial processes penetrate soils and groundwater, and transport themselves other contaminants like heavy metals or PCBs. Thus, an accurate description of the transport and retention of these particles is required to prevent and manage environmental contamination, like the pollution of drinking water supplies. Literature stands that colloid sorption (also referred to as attachment) is the main process controlling transport in the case of strong particle-medium interactions and absence of straining events. Sorption mechanisms have been widely studied using micromodels (2D systems or pore scale studies [2,3]) or indirect observation (Breakthrough curves [6]), completed by simulations to account for the complexity of real media like soils [7]. We propose here to tackle the usually neglected impact of the system's heterogeneity, in direct 3D visualizations at local and global scales.

Glass beads at random close packing are chosen as a model porous medium for their large pore throat size distribution (down to 0 at contact points). We inject in the system suspensions of iron nanoparticles (NPs), with a strong attraction interaction with the medium, which leads to heterogeneous adsorption and coverage of the beads. Coverage is followed by confocal imaging in time and at multiple scales; from local singular geometries (pore scale, tens of  $\mu$ m) to global average behaviours (sample's scale, cm).

We show that for a given NPs-medium affinity, the dynamics of adsorption strongly depend on the accessibility of the surface, in accordance with the flow streamlines. We identify configurations (regions of highest confinement, i.e. contact points between beads) where the adsorption is fully diffusion limited. At early stages of adsorption, these regions can account for up to 10% of the surface of the medium and therefore impact the supposedly known dynamics at global scale.

To go further, we link local observations and global description of the dynamics to discuss the impact of the pore size heterogeneity, at various flow rates (Darcy velocities in the range 0.05 to 9 mm/s). We show the impact of the flow on the heterogeneity of the deposition of the NPs through 1D coverage profiles over time and over the whole sample. Finally, we propose a simple model describing the full beads coverage dynamics over space and time. We can define and decouple a geometric (characteristic deposition length) and a kinetic (characteristic exploration time) components for a complete understanding of the adsorption mechanisms.

Figure –(Left) Confocal sections of a glass beads pack partially saturated with NPs. From a clean medium, progressive adsorption of nanoparticles at the beads surface is followed by fluorescence. Local and global studies (Right) highlight variations of the dynamics due to the heterogeneity of the medium: confined areas (blue) are covered later and slower than areas directly accessible to the flow.

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