

Influence of flow velocity on denitrification in a pore-scale lab experiment

A. BOISSON*, AQUILINA AND O. BOUR

Géosciences Rennes, UMR 6118, 35000 Rennes, France

(*correspondence: alexandre.boisson@univ-rennes1.fr)

Assessing and understanding aquifers reactivity is an important issue worldwide. In these aquifers, a variety of flow velocities may rule the reactivity development, and the reaction efficiency. Only few studies assessing the influence of this physical parameter on the biochemical reactivity have been carried out yet.

We present an experimental setup, where denitrification occurs in 2mm diameter reactive tubes for different flow velocities which control the fluid residence-time and may impact on reaction kinetics. Conversely to most of the experiments, the carbon source is the tube itself and C is present in a homogeneous manner all along the tubes. This experiment shows reaction kinetics close to the ones observed at the field- or lab-scale with aquifer material (both water and rocks).

Results show that the setting up time of the reactivity (ruled by the bacterial biofilm development) is not highly influenced by the flow velocity [from 40 to 6 mm/min]. On the contrary, the stabilization and the maximum reaction rate depend on the flow velocity. The concentration profiles made along the flow lines exhibit a homogeneous reactivity along the tubes at the beginning of the experiments. The reactivity becomes localized and migrates towards the tube inflow with increasing time. This experiment provides useful information on the time scale needed for *in situ* reactivity assessments and for the building up of a reactivity model, in which the minimum concentration is controlled by fluid velocity whereas the initial reaction phase concentration is independent of fluid velocity.

The inhibitory effect of silicate on the Fe(II)-catalysed sequestration of U by Fe(III) oxides

D.D. BOLAND¹, R.N. COLLINS^{1,2}, T.E. PAYNE^{1,2}
AND T.D. WAITE^{1*}

¹School of Civil and Environmental Engineering, University of New South Wales, Sydney NSW 2052, Australia
(*correspondence: d.waite@unsw.edu.au)

²Institute for Environmental Research, Australian Nuclear Science and Technology Organisation, Menai, NSW 2234, Australia

It has recently been reported that the natural Fe (II)-catalysed transformation of Fe (III) minerals to more crystalline forms can result in the sequestration of uranium [1], thus potentially leading toward a solution to the universal and emotive problem of uranium contamination. While this process may reduce uranium migration, there is no clear knowledge of its viability in conditions which inhibit the transformation of iron oxides. Here we present XAS results of Fe (II)-catalysed transformations in systems containing 2-line ferrihydrite, silicate and uranium as U (VI).

The chemical environment of both co-precipitated and adsorbed U (VI) bound by 2-line ferrihydrite was initially identical, in both cases being associated with the iron oxides as a surface complex. Upon addition of aqueous Fe (II) in anoxic conditions, 2-line ferrihydrite with associated U (VI) transformed to goethite. *Ab initio* modelling of EXAFS data indicated that U (VI) associated with 2-line ferrihydrite was incorporated into the newly formed goethite mineral structure. In contrast, silicate-ferrihydrite only transformed to ferrihydrite with the associated U (VI) remaining in a form similar to its initial state. The adsorbed U (VI) did however become more resistant to reductive dissolution indicating at least a partial reduction in mobility.

These results demonstrate that the Fe (II)-catalysed crystallisation of iron oxides may not always induce uranium reduction or immobilisation in relevant environmental conditions. The precise mechanism of the inhibitory effect of silicate, with a focus on how to control conditions to reduce this effect, must be resolved before this process may be considered a reliable means of preventing sub-surface uranium transport.

[1] Nico, Stewart & Fendorf (2009) *Environ. Sci. Technol.* **43**, 7391–7396.