Spatially and temporally resolved mineral phase evolution and arsenic retention in microfluidic models of zerovalent iron-based water treatment

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Arsenic (As) is a toxic element, and elevated levels of geogenic As in drinking water pose a threat to the health of several hundred million people worldwide. In this study, we used microfluidics in combination with optical microscopy and X-ray spectroscopy to investigate zerovalent iron (ZVI) corrosion, secondary iron (Fe) phase formation, and As retention processes at the pore scale in ZVI-based water treatment filters.

Two 250 μm thick microchannels filled with single ZVI and quartz grain layers were operated intermittently (12 h flow/12 h no-flow) with synthetic groundwater (pH 7.5; 570 μg/L As(III)) over 13 and 49 days. During operation, the corrosion of ZVI and the formation and transformation of Fe mineral phases was followed with spatiotemporal resolution using optical microscopy. After operation, the microchannel was resin-embedded for analysis by synchrotron-based micro-focused X-ray fluorescence spectrometry (µ-XRF) and X-ray absorption spectroscopy (µ-XAS), chemical imaging and fullfield XAS to gain insights into the spatial distribution of Fe, As and other elements, the distribution of Fe mineral phases, and the redox speciation of As.

At the start of filter operation, lepidocrocite (Lp) and carbonate green rust (GRC) were the dominant secondary Fe-phases. They underwent cyclic transformation over intermittent periods of water flow and stagnation. During no-flow, lepidocrocite partially transformed into GRC and into small fractions of magnetite, kinetically limited by Fe(II) diffusion or by decreasing corrosion rates. When water flow was resumed, GRC rapidly and nearly completely transformed back into lepidocrocite. Longer filter operation combined with a prolonged no-flow period accelerated magnetite formation. Phosphate adsorption onto Fe-phases allowed for spatially separated downstream precipitation of calcium carbonate and, consequently, accelerated anoxic ZVI corrosion. Arsenic was retained on Fe-coated quartz grains and in zones of cyclic Lp-GRC transformation, as both As(V) and As(III). Our results suggest that intermittent filter operation and the resulting redox cycles promote the formation of denser secondary Fe-solids and thereby ensure prolonged filter performance.

Methodologically, this work highlights the potential of spatially and temporally resolved studies on micromodels to offer new insights into geochemical processes at the pore scale under conditions of kinetic and transport limitations.

REFERENCE