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 spatially resolved 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, each filled with a single layer of quartz and ZVI grains were operated intermittently (12 h flow/12 h no-flow) with synthetic groundwater over 13 and 49 days. During operation, we followed the corrosion of ZVI and the formation and transformation of Fe mineral phases with optical microscopy. After operation, the microchannels were resin-embedded for analysis by synchrotron-based micro-focused X-ray fluorescence spectrometry (µ-XRF), X-ray absorption spectroscopy (µ-XAS), chemical imaging and full-field 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.

Optical microscopy-based time-lapse movies provide novel insights into the highly dynamic mineral phase evolution in the filter pore space during operation. In combination with the synchrotron data collected after operation, the results show that intermittent filter operation leads to cyclic phase transformations between green rust and lepidocrocite and gradual formation of magnetite close to ZVI grains as longer-term host for As. The results also show that upstream P removal leads to downstream Ca-carbonate precipitation, which in turn can promote anoxic ZVI corrosion. The study thus highlights the complex spatiotemporal coupling of various geochemical processes in the pore space. The results from this work are not only of interest with respect to the optimization of ZVI-based drinking water treatment, but also with respect to the use of ZVI in groundwater remediation.

Methodologically, this work shows that spatially and temporally resolved studies in micromodels can offer unprecedented insights into geochemical processes at the pore scale under conditions of kinetic and transport limitations. In the future, further advances in the study of geochemical processes at the pore scale can be achieved by combining micromodel experiments with in-situ spatially resolved synchrotron X-ray spectroscopies.

References