

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

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Arsenic (As) is a toxic element, and elevated levels of geogenic As in drinking water threaten the health of several hundred million people worldwide. 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 filters.

Two 250- μm thick microchannels 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, the corrosion of ZVI and the formation and transformation of Fe minerals was followed with optical microscopy. After operation, the microchannels were examined with micro-focused X-ray fluorescence spectrometry ($\mu\text{-XRF}$), X-ray absorption spectroscopy ($\mu\text{-XAS}$), chemical imaging and full-field XAS to gain insights into the distribution of Fe, As and other elements and of Fe minerals.

Optical microscopy-based time-lapse movies provide new insights into the highly dynamic mineral phase evolution in the pore space during filter operation. In combination with 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. Upstream P removal leads to downstream Ca-carbonate precipitation, which in turn promotes 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 relevant for the optimization of ZVI-based drinking water treatment, but also for 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. Further advances in the study of geochemical processes at the pore scale can be achieved by combining micromodel experiments with in-situ synchrotron X-ray spectroscopies.

Reference: Wielinski, J., Jimenez-Martinez, J., Göttlicher, J., Steininger, R., Mangold, S., Hug, S. J., Berg, M., Voegelin, A.,