

Integrated experimental and modeling assessment of potential effects of gas leakages on groundwater composition

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Storing renewably produced energy is one of the major challenges for the energy systems of the upcoming decades. Power-to-gas technologies coupled to geological storage of compressed air, methane, and hydrogen offer a comparatively safe and cost-efficient way for large-scale energy storage. However, the stored gases can potentially escape from their geological reservoir and may thus affect protected natural goods such as groundwater. The geochemical reactions responsible for these composition changes are usually investigated separately in experiments and numerical models.

Here we present the outcomes of an integrated experimental and modeling approach through the example of a compressed air leakage scenario. A main consequence of the presence of oxygen to be assessed in an aquifer is pyrite oxidation, well known from acid mine drainage sites. However, in contrast to acid mine drainage sites exhibiting unsaturated sediments and fed by meteoric low-carbonate water, aquifers such as in Northern Germany contain a considerable amount of solid and dissolved inorganic carbon species potentially buffering pH changes. High pressure flow-through column experiments representing an intrusion of compressed air into an aquifer were carried out to quantify pyrite oxidation kinetics and to incorporate the observations into a descriptive reaction model. Surface passivation was found to decrease the reactivity of pyrite by more than 90% after a few months of experimental run time. We propose that the carbonate buffer system enables the precipitation of a passivating mineral layer on the pyrite surface reducing the overall reaction rate significantly. Consequently, an established rate law from the literature was extended by a reactive surface passivation term[1].

This improved reaction rate equation was incorporated into a 3D numerical model using OpenGeoSys with parameters representing similarly typical aquifer conditions the experiments had characterized. These boundaries include pyrite content, oxygen dissolution kinetics, groundwater composition including the carbonate buffer, and diffusive and advective transport parameters. The results of site-scale multiphase reactive transport modeling revealed the expected spatial distribution of redox-sensitive species such as oxygen, pyrite, and sulfate in an aquifer following a leakage. The changes in concentration of sulfate, dissolved oxygen, and H⁺ observed in the lab-scale experiments were qualitatively reproduced by the models applying the same boundary conditions on a site-scale.

This integrated study acknowledged that the combination of experiments and models is a powerful tool to prognose the geochemical consequences of gas leakage on site scale. However, it is yet unknown how the passivation would be effected if the carbonate buffer depleted on the long term and under what circumstances a transition from the passivating pyrite oxidation process to the non-passivating process observed for instance in acid mine drainage setups occurs. These restrictions mark the limits of validity of our experimental and modeling concept. This conclusion suggests the feasibility of the presented integrated approach also when evaluating comparable scenarios on methane and hydrogen storage based on experimental results gathered similarly[2].

[1]Berta et al. *Environ Earth Sci* (2016) 75:1175, DOI 10.1007/s12665-016-5985-7.

[2]Berta et al. *First Break* (2015) 33,93-95, ISSN 1365-2397.

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