

Release of colloidal pyrite during shale-water interaction

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During hydraulic fracturing, a commonly used technique to extract oil and gas from shales, large volumes of water are injected into the subsurface. Crucially, the injected fluid can interact with minerals present within the shales, releasing contaminants into fluid phase. Pyrite, which is common in organic-rich shales, is a potential source of toxic elements, including arsenic and lead, and it is generally thought that for these elements to become mobilized, pyrite must first dissolve. In this study, we use direct observations with atomic force microscopy to show that during fluid-rock interaction, the dissolution of carbonate minerals in a range of different shales leads to the physical detachment, and mobilization, of embedded pyrite grains. To simulate the way fluid interacts with a fractured shale surface, we also reacted rock samples in a flow-through cell, and used environmental scanning electron microscopy to compare the surfaces before and after interaction with fluid. In experiments carried out over a range of pH, salinity, and temperature we found that in all cases pyrite particles became detached from the surface of the shale. Grain detachment was usually associated with the dissolution of surrounding calcite. On average, the amount of pyrite detached was equivalent to $6.5 \times 10^{-11} \text{ mol m}^{-2} \text{ s}^{-1}$, which is over an order of magnitude greater than the rate of pyrite oxidation expected under similar conditions. This result suggests that mechanical detachment of pyrite grains could be the dominant mode by which arsenic and other inorganic elements are mobilized in the subsurface during hydraulic fracturing. Developing reactive transport models that can account for the release of colloidal particles in shale systems therefore represents a critical future goal.