Fluid-mineral-organic interactions and contaminant release in shale reservoirs

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The extraction of oil and gas resources from shale reservoirs using hydraulic fracturing has increased significantly in recent years. During hydraulic fracturing, large volumes of oxic fluid are injected into subsurface shale formations. This drives fluid-mineral-organic interactions that can release contaminants and alter porosity. We use a combination of experiments, three-dimensional imaging, and modelling to track the fate of contaminants and changes in porosity during hydraulic fracturing.

Experiments with a variety of shales and kerogen isolates revealed that contaminants such as U and Pb were released both from shale minerals and kerogen upon exposure to fracturing fluids. During the formation of Fe-(oxy)hydroxides these contaminants were removed from solution. The rate at which Fe was oxidized to form these oxy(hydroxide) minerals was more rapid than expected at the acidic fluid pH. We found that the rate of Fe(II) oxidation was accelerated in the presence of bitumen. A rate law for Fe(II) oxidation was developed. This rate law will help better predict the formation of Fe-(oxy)hydroxides which both occlude porosity and sequester contaminants. Geochemical modelling coupled with imaging also revealed that the distribution of highly reactive phases like pyrite in the shale matrix is a strong control on fluid composition. Our results highlight the myriad of geochemical interactions that may control porosity evolution and the release and transport of contaminants. This illuminates opportunities for tailoring fracturing fluid compositions to minimize environmental degradation, and identifying conditions under which contaminants are most likely to be mobilized.