

Mineralogical Transitions during Early Diagenesis in Incubation Experiments – A Perspective from Iron Speciation and Iron and Sulfur Isotope Ratios

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Iron speciation in sedimentary rocks is one of the most widely used tools to determine oxic, anoxic ferruginous (Fe^{2+} -containing), or euxinic (HS^- -containing) depositional conditions. However, dissolution and precipitation of iron mineral phases can occur during early diagenesis, which may alter the original, environmental, iron speciation signatures. In particular, the exposure of iron-bearing sediments to hydrogen sulfide produced through microbial sulfate reduction may convert highly reactive iron to iron sulfide minerals. We present batch incubation experiments of sediments with high iron content (4-5 wt%) to study the evolution of iron speciation signatures of sediments when exposed to varying rates of microbial sulfate reduction. We present dissolved Fe^{2+} , HS^- and SO_4^{2-} concentrations, as well as the sulfur isotopic composition ($\delta^{34}\text{S}$) of both sulfate in solution and solid phase pyrite-sulfur, combined with iron speciation analyses throughout the incubations, varying the rate and pacing of the addition of organic carbon and sulfate to the vials. As expected, our results suggest that the presence of iron (oxyhydr)oxides in the sediment and excess hydrogen sulfide generated by microbial sulfate reduction results in enhanced sulfidation of Fe (oxyhydr)oxides. We suggest that the rate of transition is limited by the slow dissolution of iron (oxyhydr)oxide phases relative to the precipitation of monosulfide minerals and ultimately pyrite. Our study highlights progressive sulfidation of highly reactive Fe phases during burial of sediments through the zone of microbial sulfate reduction in marine and marginal marine settings, and we explore the application of coupled iron and sulfur isotope ratios to track the transition of iron mineral phases during early diagenesis.