Electron backscatter diffraction study of iron-sulfide nodules in sediments off SW Taiwan: Implications for puzzling early diagenetic formation of iron sulfides

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Iron sulfides play a major role in biogeochemical C-O-S cycles and metal sequestration in sedimentary environments. Current understanding of the early diagenetic formation and transformation of iron-sulfide minerals in marine sediments heavily relies on results from synthesis works in part due to small crystallite size and large reactivity of metastable iron sulfides in natural specimens.

Millimeter-sized iron-sulfide nodules occurred in the sediments of an active cold-seep site with the sulfate-methane transition (SMT) zone at ca. 4.5 mbsf in the SE flank of the Good Weather Ridge off SW Taiwan. High-resolution FESEM-EDS-EBSD investigation of selected nodules at 265-270 cmbsf revealed a complex growth sequence of iron-sulfides in which partially dissolved pyrite framboids were mantled by massive greigite aggregates having corroded edges surrounded by mackinawite crystals up to 2 μm in size. The mackinawite was locally engulfed by later formed platy or blocky pyrrhotite. Precipitation of smaller platy crystals of pyrrhotite occured in cavities within the nodules as well. Such nodules experienced later-stage sulfidization that was driven by anaerobic oxidation of methane in the past SMT zone and characterized by proliferative pyritization along mackinawite and/or greigite grain boundaries and vein-filling pyrite crystallization along fractures. Iron-sulfide nodules at depth exhibit similar features except pyrite mineralization.

The presence of complex assemblages of metastable iron sulfides is attributed to non-steady state diagenesis controlled by fluctuations of methane flux and pore water chemistry and shielding of nodular growth. The result implies that reductive early diagenesis could induce dissolution of pyrite replaced by greigite, formation of mackinawite after greigite, and neoformation of pyrrhotite in marine sediments, and that early diagenetic pyritization of greigite and mackinawite could occur via diffusive dissolution and crystallization along grain boundaries. This study adds to the increasing body of evidence for new formation modes of early diagenetic iron sulfides in marine sediments.