Fate of Cobalt and Nickel during diagenetic pyrite formation

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As iron-sulfide mineral phases are important sedimentary sinks for naturally occurring or contaminant metals, it is important to know the fate of these metals during mineral transformations by diagenesis into more stable phases, such as pyrite (FeS₂). We studied nickel (Ni) and cobalt (Co) incorporation into freshly-precipitated mackinawite (FeS), and after experimental diagenesis to pyrite (FeS2) using S⁰ as an oxidant at 65°C. Metal incorporation was quantified on bulk digests using ICP-OES or ICP-AES. Bulk mineralogy was characterized with micro X-ray diffraction, documenting transformation of mackinawite to pyrite. Grain mounts were prepared anoxically, and transported to the Stanford Synchrotron Radiation Lightsource (SSRL). There we used micro X-ray Fluorescence (µXRF) to map the distribution of Co and Ni, and make chemical speciation image maps through the iron (Fe) and sulfur (S) K absorption edges. Micro X-ray absorption near edge spectroscopy ($\mu XANES$) was used with multi-energy XRF maps to determine the distribution of distinct S and Fe oxidation states and mineralogy. Bulk data preliminarily indicate that Co was lost from the solid phase into solution during transformation from mackinawite to pyrite. We also observed heterogeneities in Ni and Co within particles using µXRF that are not indicated by bulk analyses. Sulfur within the diagenetically-produced pyrite samples was also heterogenous in oxidation and coordination state. Our results reveal how Co and Ni are accommodated into iron sulfides, and what structural or chemical factors control their mobility during diagenesis and alteration.