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In situ analysis of Fe and S-isotope composition of hydrothermal chimneys by laser ablation MC-ICPMS

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We have developed a technique for the accurate and precise determination of ^{34}S/^{32}S, ^{56}Fe/^{54}Fe, and ^{57}Fe/^{54}Fe isotope ratios in S- and Fe-bearing minerals using a combination of solution and laser ablation analysis. Fe- and S-isotope ratios were measured by high-resolution MC-ICPMS (ThermoElectron Neptune), enabling us to resolve major isobaric interferences on S-isotopes and Fe-isotopes from O_2^+, ArN^+, and ArO^+. A New Wave UP213 laser was used as the ablation source with He as the sample carrier gas. Fe- and S-isotope ratios were calibrated against aqueous standards prepared in appropriate matrix composition. This approach allows the use of “sample-standard bracketing technique” as well as internal normalization technique where the instrumental mass bias is corrected by simultaneously measuring Ni of known isotope composition. The long-term reproducibility of S- and Fe-isotope compositions was typically 0.2 ‰ and 0.1‰, respectively.

We applied this technique to assess the fine scale variations of δ^{56}Fe and δ^{34}S values in inactive black smoker chimneys and massive sulfides recovered from the East Pacific Rise at 9-10˚N. Consistent with bulk conventional analysis, δ^{34}S and δ^{57}Fe values of euhedral and collomorph pyrite range from 2.9 to 4.3‰ (relative to V-CDT) and -1.5 to -3.0‰ (relative to IRMM-14), respectively. Goethite encrustation associated with massive sulfides have generally δ^{57}Fe values similar to those of pyrite, whereas 2-line ferrihydrite from inactive chimneys display δ^{57}Fe values ranging from -7.2‰ to -1.5‰. These values are both lower and higher than δ^{57}Fe values of associated pyrite

and suggest the occurrence of different generations of Fe-oxyhydroxide precipitation. We interpret the Fe-isotope signatures of goethite and ferrihydrite as reflecting primarily different Fe oxidizing environments related to: (1) direct oxidation from Fe from pyrite in massive sulfide under oxygenated seawater conditions, and (2) Fe oxidation from late stage Fe-Si-rich hydrothermal fluids in chimney environments.

Seafloor hydrothermal systems at mid-ocean ridges encompass fundamental processes controlling the exchange of heat and chemical species between seawater and ocean crust, and support diverse and unique biological communities capable of using dissolved chemical species and minerals for energy metabolism. This new analytical capability coupled to spatially-resolved elemental and mineralogical data (synchrotron radiation XRF, XRD, and XAS) is expected to provide further constrain on: (1) the complex interactions between hydrothermal fluids, seawater, and sulfide minerals over a wide range of temperature and redox conditions, and (2) potential biosignatures of Fe-oxidizing and Fe-S-reducing bacteria in chimney environments.