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フェムト秒LA-MC-ICP-MS による銅同位体比局所分析法の開発と鉱石試料への応用
Development of in-situ Cu isotope ratio measurement by femtosecond-LA-MC-ICP-MS and its applications to ore minerals

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A new method for determining copper isotope compositions of copper-rich minerals (native copper, cuprite, chalcocite, chalcopyrite, cubanite and malachite) using a femtosecond LA-MC-ICP-MS has been developed. The standard-sample-standard bracketing technique was applied to correct the instrumental mass fractionation. Matrix effects found in chalcocite, chalcopyrite, cubanite and malachite can be corrected using the matrix-matched calibration standard. The analytical precision ($<$0.14 ‰, 2σ) and accuracy were significantly improved compared with those of previous works using a nanosecond-LA-MC-ICP-MS.

The developed LA-MC-ICP-MS method was applied to the measurements of copper isotope ratios of minute copper ore minerals in igneous rocks (e.g., Horoman peridotite complex) and seafloor hydrothermal deposits (modern: Mariana Trough; ancient: Besshi-type and Kuroko-type volcanogenic massive sulfide deposits) in order to investigate variability of copper isotopic compositions in these samples.

The δ^{65}Cu (where δ^{65}Cu = [(^{65}Cu/^{63}Cu)$_{sample}$/ (^{65}Cu/^{63}Cu)$_{NIST-SRM976}$-1] $\times$ 1000) values of copper-rich sulfide minerals of the active seafloor hydrothermal deposits are significantly large (δ^{65}Cu = -0.7 to 4.0 ‰) compared to those of the ancient submarine hydrothermal deposits (δ^{65}Cu = -0.3 to 0.4 ‰) and the igneous rocks (δ^{65}Cu = -0.3 to 0.3 ‰). These large copper isotope variations in the modern active seafloor hydrothermal deposits are most likely explained in terms of a redox-controlled isotope fractionation during hydrothermal reworking or alteration of precipitated copper-rich minerals. These results also suggest that sub-seafloor and metamorphic recrystallization effects probably have reduced the original range of copper isotopes.

Secondary malachite (δ^{65}Cu = 2.6 to 3.0 ‰) and native copper (δ^{65}Cu = 1.4 to 1.7 ‰) in the Besshi-type deposit have heavier copper isotope values compared to precursor copper-rich minerals. These variations are mainly due to isotope fractionation during redox reactions (weathering) at low temperatures involving the preferential incorporation of heavy copper isotope in secondary Cu(II) solutions. Therefore, copper isotope geochemistry could be a useful tool for understanding geochemical processes of copper transport and deposition in ore-forming systems.

キーワード: 銅同位体比, フェムト秒 LA-MC-ICP-MS, 鉱石鉱物
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