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LAL-ICPMS 法を用いた局所高精度 Pb 同位体分析による熱水性方鉛鉱の起源推定
High precision in situ Pb isotope analysis of galena by LAL-ICPMS technique

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Radiogenic Nd and Pb isotopic compositions of the fluids originated from subdcting Pacific and Philippine Sea plates have been characterized from isotopic trends observed among arctic volcanic rocks (Nakamura et al., 2008). Origin and evolution of the fluids that produced hydrothermal ore deposits may now be investigated by radiogenic isotopic compositions of ore deposits. In this study, we analyzed the micro scale isotopic variation of Pb in a hydrothermal galena to shed light on the macro scale dynamics of the fluids. To investigate the possibly small degree of isotopic changes within a galena sample, both high spatial resolution and high precision are required for the isotopic analysis. We employed the combination of laser ablation in liquid (LAL) micro sampling technique (Okabayashi and Hirata, 2011) and solution-based Pb isotopic analysis by MC-ICPMS technique to meet the analytical requirements. In the LAL micro sampling, laser-ablated sample particles are trapped in the liquid that placed above the sampling area. The trapped samples are then dissolved and introduced to the ICPMS as a solution. The advantage of the combined LAL-ICPMS technique over laser ablation (LA) ICPMS technique is the stable ion signals due to solution form, which allows high-precision isotope ratio measurement.

Sample analyzed in this study was a hydrothermal galena from Hosokura mine (Miyagi, Japan). A microscopic texture of the sample was observed in detail with FE-SEM-EDS system (JEOL JSM-6500F) prior to the isotopic analysis. A fs laser (IFRIT, Cyber Laser, Japan) with a wavelength of 780 nm (~200 fs pulse width) was used for the LAL micro sampling. Care was taken to avoid sampling of grain boundaries and inclusions. Typical special resolution was 150 micron in diameter and 30 micron in depth. The laser-sampled PbS (300-400ng Pb) trapped in Milli-Q water was dissolved in conc. HNO3, and adjusted to 200 ng/mL Pb solution in 0.15 M HNO3 for Pb isotopic analysis. Pb isotope ratios were determined with a MC-ICPMS, Neptune (Thermo Instruments, Bremen, Germany). An isotopic reference material of Tl (NIST-SRM 997) was added to the final sample solutions for the correction of mass discrimination of Pb in the instrument to have a concentration of 20 ppb Tl.

Galena occurs as discrete layers of ca. 1cm width in between layered CaF_2 as well as sub mm-sized inclusion within thick CaF_2 layer. Galena inclusion and layers were numbered from 1 to 3 according to its precipitation order. Grain size of the galena in each of the layer is several hundred microns to several millimeters. Euhedral quartz with a size of 10-100 micron occurs along the grain boundary of galena and as an inclusion within galena grains.

Small but significant Pb isotopic variation of sub-permil order was observed among and within the 3 galena layers. The analyzed samples clearly form a linear trend in the $^{208}Pb/^{207}Pb$ vs. $^{206}Pb/^{207}Pb$ diagram. The observed Pb isotopic trend indicates that the Pb isotopic composition of the fluid that produced the galena has slightly changed during galena precipitation. The Pb isotopic composition of the galena is consistent with mixing of a sediment component of the Pacific plate (Nakamura et al., 2008) with a deep fluid derived from Pacific Ocean plate (Nakamura et al., 2008) and/or the DMM. With the high-precision isotopic analysis as demonstrated in this study, LAL-ICPMS may have an important contribution to high-spatial-resolution geochemical studies in the future.

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