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# Tin isotope analysis for an archaeological application

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[Introduction] Bronze products are composed of Cu, Sn, and Pb that were used before iron products become common. Major topics in archaeological science include archaeometry and provenance study. Additionally, to verify whether ancient bronze products experienced recycling is a significant problem because an existence of the recycle affects provenance studies.
It was suggested that Sn isotope are available to investigate recycling of bronze products (Budd et al., 1995). This prediction is based on the characters of Sn. That is, Sn behaves volatile element and has the largest numbers of isotopes. Therefore, ancient metallurgical processes could cause measurable Sn isotope fractionation. Copper and Pb isotopes could fractionate as well as Sn isotopes. But no significant Cu isotope fractionation during metallurgical processes has been detected (Mathur et al., 2009). Moreover, Pb isotopic composition varies largely in each region and they have been utilized to provenance studies of ancient bronze products (e.g. Yener et al., 19 91). Consequently, Pb isotope fractionation during metallurgical processes is predicted to be lower than regional variation (Mulliken and Harkins, 1921; Budd et al., 1995). Mathur et al. (2009) suggested that Cu isotopes could also contribute to provenance studies.
Tin isotope analysis for ancient bronze products by Gale (1997) showed that there is no significant isotopic difference between several kinds of bronze products and laboratory standard material. They concluded that these bronze samples did not experience recycling with metallurgical processes. However, Sn isotope fractionation during metallurgical processes has not been confirmed. Hence, there would be a possibility that Sn isotope fractionation in ancient bronze products is detected.
The previous studies assumed that cassiterite as source materials of ancient bronze products have uniform Sn isotopic composition, independent of regional differences. McNaughton and Rosman (1 991) analyzed eight samples of cassiterite, metallic Sn and laboratory standard by using thermal ionization mass spectrometer (TIMS). They presented that most of cassiterite samples have similar Sn isotope ratios to those of the standard. Clayton et al. (2002) conducted Sn isotopic analysis for standard material and cassiterite with Multicollector Inductively Coupled Plasma mass spectrometer (MC-ICP-MS). This research showed that the cassiterite was enriched in heavier Sn isotopes than standard.
[Objective] As just described, there has not been enough isotopic data for cassiterites and we need to expand data base. We are going to investigate variations of Sn isotopic composition for cassiterite in Japan. These cassiterite samples are provided by Dr. Ishihara at National Institute of Advanced Industrial Science and Technology (AIST).
[Experiment] Cassiterite was crushed in a stainless mortar and powdered in an agate mortar. About 1 mg of powdered sample was weighed into PFA vials. 0.3 ml of hydroiodic acid was added and heated overnight by using teflonbomb at 100 C. Subsequently, samples were dried at 80 C and leached in hydrochloric acid (about 0.6 ml). The supernatant solutions were collected in a bottle. The procedures were repeated until the sample is dissolved. Collected sample solutions were diluted in 2% nitric acid so that Sn concentration becomes 60ppb and Sb was added for external fractionation correction. The dilute solutions were injected into a MC-ICP-MS (the Micromass Isoprobe at ERI, the Univ. of Tokyo). Sn isotopic ratios of samples were evaluated as the deviation

from Sn isotopic ratios of standard (SPEX).
[Result] In the result of preliminary experiment, it was showed that cassiterite samples from Peru were enriched in heavier Sn isotopes than standard. Cassiterite samples deviated about 0.2 permil per mass. We will analyze Japanese cassiterites and we will discuss if there is isotopic heterogeneity in them.

Keywords: Tin, isotope, bronze