

Development of a new method for the measurement of Si-isotopic compositions with SIMS and Application to CAIs

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Calcium-Aluminum-rich Inclusions (CAIs) in Primitive meteorites are the oldest materials in the solar system, and they can tell us about the early stage of the solar system. Although terrestrial materials and most materials in the solar system have the uniform Si-isotopic ratios ($\delta^{30}\text{Si} \sim 0$ permil), bulk CAIs have both negative and positive δ -values with a range of ~ 10 permil along the mass-dependent fractionation line (Clayton R.N. et al., 1985). When some materials are evaporated in vacuum, evaporative residue becomes isotopically heavier. So it is generally assumed that isotopically heavy materials are evaporative residue and isotopically light materials are condensates. Since some CAIs are isotopically heavy and some are light, it is known that CAIs formed through the process of both condensation and evaporation repeatedly.

However, only bulk data of Si-isotopic fractionation of CAIs have so far been reported. Bulk datum means an average composition of one CAI, so that we do not know the differences among texture and minerals inside the CAI.

Secondary Ion Mass Spectrometry (SIMS) is a method with which we can measure the isotopic composition in an area of several tens μm in diameter of a sample. The local analysis of Si-isotopic ratios inside CAIs may tell us isotopic heterogeneity derived from degree and timing of condensation and/or evaporation, partial melting and alteration.

However, data of Si-isotopic fractionation measured with SIMS have not been reported yet. The isotopic ratios measured with SIMS are not equal to the true isotopic ratios of the samples because of Instrumental Mass Fractionation (IMF). Since IMF is also mass-dependent and the level of IMF of Si isotope is very large, the precise analysis of Si-isotopic fractionation of meteoritic samples are difficult. IMF seems to occur in many processes of SIMS instrument, and is so complicated that quantitative data have not been obtained.

So, in this study the instrumental properties of SIMS were investigated using several terrestrial mineral samples whose Si-isotopic ratios were known, for the purpose of improvement of measurement precision and for the precise understanding of level of IMF.

Measurement condition that provides adequate precision was found by adjusting many parameters. After this improvement, several mineral samples were measured and IMF for the minerals were determined. The level of IMF depends on the chemical composition of the samples even for minerals in the solid solution. Quantitative comparison of IMF and the chemical composition of the samples showed an interesting feature that the level of IMF is related to Ca- or Mg-content.

Si-isotopic ratios in three types of CAIs (type A, B1 and B2, respectively) were investigated. After the correction of IMF, Si-isotopic ratios of three CAIs were found to be isotopically heavy. And also, there was a tendency that Si-isotopic ratios of the mantle in a type B1 CAI was heavier than that of core. However, the improved precision was not enough for detailed study of Si-isotopic variation among and/or within CAIs.

Variations of the bulk Si-isotopic ratios of CAIs are larger than the precision achieved in this study. So, the systematic measurements of more types of CAIs and further improvement of precision will tell us new information about the formation of CAIs and the early solar system.