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論 文 要 旨

Isotopic evolution of the early Solar System inferred from nucleosynthetic isotope anomalies in refractory materials from CV chondrites

（CV コンドライトの難揮発性物質に見られる核合成起源の同位体異常から推察される初期太陽系の同位体組成進化）

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Carbonaceous chondrites contain refractory materials represented by calcium-aluminum-rich inclusions (CAIs). CAIs are one of the oldest rocks in the Solar System that condensed from nebular gas with a solar composition, which possibly record the isotopic composition of the early Solar System gas. They have remarkable isotopic compositions different from those of the Solar System for a variety of elements, which is called nucleosynthetic isotope anomalies. The isotopic anomalies in CAIs are important for tracing nucleosynthesis events that supplied elements into the Solar System. Previous studies on the isotopic compositions of refractory materials have focused primarily on coarse-grained CAIs (CGs). However, CGs have undergone melting condensation from the nebular gas, such that some of the initial information at the time of their condensation has been lost. On the other hand, fine-grained CAIs (FGs) evaded the melting and are expected to reflect the isotopic heterogeneity of gas reservoirs where they condensed. The

isotopic analysis of FGs has been gaining attention in recent years, and diverse isotopic compositions different from those of CGs have been reported. However, the number of isotopic data for FGs hitherto obtained is limited, so that the cause of the diversity in isotopic compositions for FGs is unknown. In addition, few isotopic anomalies have been compared for each element in CAIs, and no isotopic evolution model has been presented to explain the isotopic anomalies for multiple elements.

In this doctoral thesis, we constrained the formation process of refractory materials and the isotopic evolution of their formation reservoir by performing high-precision isotopic analysis of multiple elements. This doctoral dissertation consists of six chapters.

Chapter 1 is a general introduction and review of recent findings on isotopic heterogeneity in the early Solar System, nucleosynthesis, and the chemistry of refractory materials in carbonaceous chondrites.

In Chapter 2, we investigated the distribution of rare-earth elements (REEs) in FGs. The REE abundance is important for understanding the condensation and evaporation processes of FGs. The elemental imaging technique using multi-spot femtosecond laser ablation ICP-MS (msfsLA-ICP-MS) was applied to FGs for the first time in the world, and the REE distribution and abundance patterns for each mineral were clarified. Unzoned FGs have homogeneous REE abundance patterns, while zoned FGs have different REE patterns. This indicates that msfsLA-ICP-MS can be used to rapidly describe the REE distribution of FGs, which was previously difficult, and that unzoned and zoned FGs evolved through different formation processes.

In Chapter 3, high-precision Sr isotope analysis of FGs was performed. The variability of Sr isotopic anomalies increased with decreasing sample size due to the nugget effect. The variability of Sr isotopic anomalies also correlates with the depletion of heavy REEs, excluding Tm and Yb, in the REE pattern of FGs. This suggests that the influx of the *p*-nuclide, ^{84}Sr, into the protostar disk gas was associated with reheating events that fractionated REEs. Our results suggest that FGs with low and high degrees of Sr isotopic anomalies may have formed repeatedly at different location or time.

In Chapter 4, we performed high-precision Cr-Ti isotopic analysis of CGs and FGs. In previous studies, the Cr isotopic data for CAIs, especially for FGs, was limited. This study is the first to report and discuss Cr isotopic compositions with emphasis on FGs. Unlike relatively homogeneous Cr-Ti isotopic compositions of CGs determined in previous studies, our new results on FGs show more variable Cr-Ti isotopic compositions. The more variable Ti isotopic composition in FGs than CGs implies that the formation of FGs occurred at multiple locations or times. Our results also indicate that the contribution of FGs and CGs cannot explain the negative correlation of Cr and Ti isotopic compositions in carbonaceous chondrites.

In Chapter 5, we performed high-precision Ca isotope analyses for the four types of refractory materials, including CAIs. The analyses, which were performed with 1–2 orders of magnitude higher precision than previous studies, revealed a negative correlation between $\mu^{43}Ca$ and $\mu^{48}Ca$ values (μ indicates 10^6 times relative deviation from a standard material). This indicates that there are two types of carriers for anomalous Ca isotope compositions in the refractory materials, one with high $\mu^{43}Ca$-low $\mu^{48}Ca$ and the other with vice versa, and that the two were heterogeneously distributed along the heliocentric distance in the early Solar System.

Chapter 6 synthesized the data for Ca-Ti-Cr-Sr isotopic compositions obtained in Chapters 3–5 and discussed the isotopic evolution of the early Solar System during CAI formation. The correlations observed in some isotopic compositions indicate that the isotopic heterogeneity in the CAI formation field can be explained by the incomplete mixing of three gas reservoirs with different isotopic compositions. Furthermore, the diversity of either Sr or Ti isotopic compositions indicates differences in the timing or location of FG formation. Additional geochronological data, such as Al-Mg, are desired to elucidate which isotopic composition of Sr or Ti reflects the differences in time or space.

Based on previous studies and the CAI data in this thesis, we propose that the isotopic compositions of refractory materials in meteorites, CCs, and non-carbonaceous chondrites (NCs) can be explained by a two-component mixture of components from supernovae and interstellar medium. In addition, the origin of each type of CAI is discussed in terms of the characteristics of their isotopic compositions. As a future perspective, partial sampling techniques and isotopic analyses of smaller sample amounts will reveal the chemical evolution of the disk with higher resolution.