

論文 / 著書情報
Article / Book Information

題目(和文)	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
Title(English)	Isotopic evolution of the early Solar System inferred from nucleosynthetic isotope anomalies in refractory materials from CV chondrites
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出典(和文)	学位:博士(理学), 学位授与機関:東京工業大学, 報告番号:甲第12651号, 授与年月日:2024年3月26日, 学位の種別:課程博士, 審査員:横山 哲也,中本 泰史,石川 晃,奥住 聡,癸生川 陽子
Citation(English)	Degree:Doctor (Science), Conferring organization: Tokyo Institute of Technology, Report number:甲第12651号, Conferred date:2024/3/26, Degree Type:Course doctor, Examiner:,,,,
学位種別(和文)	博士論文
Category(English)	Doctoral Thesis
種別(和文)	要約
Type(English)	Outline

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

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

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

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

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

48 In Chapter 3, high-precision Sr isotope analysis of FGs was performed. The
49 variability of Sr isotopic anomalies increased with decreasing sample size due to the
50 nugget effect. The variability of Sr isotopic anomalies also correlates with the depletion
51 of heavy REEs, excluding Tm and Yb, in the REE pattern of FGs. This suggests that the
52 influx of the *p*-nuclide, ⁸⁴Sr, into the protostar disk gas was associated with reheating

53 events that fractionated REEs. Our results suggest that FGs with low and high degrees of
54 Sr isotopic anomalies may have formed repeatedly at different location or time.

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

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

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

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

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