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Neodymium isotope heterogeneity of ordinary and carbonaceous chondrites and the origin of non-chondritic ^{142}Nd compositions in the Earth

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ABSTRACT

We present high-precision Nd isotope compositions for ordinary and carbonaceous chondrites determined using thermal ionization mass spectrometry with dynamic and multistatic methods. The ordinary chondrites had uniform and non-terrestrial μ^{142}Nd, μ^{148}Nd, and μ^{150}Nd values, with data that plot along the mixing line between *s*-process and terrestrial components in μ^{150}Nd versus μ^{148}Nd and μ^{142}Nd versus $\mu^{148,150}$Nd diagrams. In contrast, the carbonaceous chondrites were characterized by larger anomalies in their μ^{142}Nd, μ^{148}Nd, and μ^{150}Nd values compared to ordinary chondrites. Importantly, the data for carbonaceous chondrites plot along the *s*-process and terrestrial mixing line in a μ^{150}Nd versus μ^{148}Nd diagram, whereas they have systematically lower μ^{142}Nd values than the *s*-process and terrestrial mixing line in μ^{142}Nd versus $\mu^{148,150}$Nd diagrams. This shift likely results from the incorporation of calcium- and aluminum-rich inclusions (CAIs), indicating that the Nd isotopic variability in the ordinary chondrites and CAI-free carbonaceous chondrites was caused solely by the heterogeneous distribution of *s*-process nuclides. The isotopic variation most likely results from nebular thermal processing that caused selective destruction of *s*-process-depleted (or *r*-process-enriched) dust grains in the inner Solar System where the parent bodies of ordinary chondrites formed, whereas such grains were preserved in the region of carbonaceous chondrite parent body formation. The Nd isotope dichotomy between ordinary and bulk aliquots of carbonaceous chondrites can be related to the presence of Jupiter, which may have separated two isotopically distinct reservoirs that were present in the solar nebula. After correcting for *s*-process anomalies and CAI contributions to the Nd isotopes observed in the chondrites, we obtained a μ^{142}Nd value (-2.4 ± 4.8 ppm) that was indistinguishable from the terrestrial value. Our results corroborate the interpretation that a missing reservoir (e.g., a hidden enriched reservoir, erosional loss of crust) is not required to explain the observed differences in ^{142}Nd/^{144}Nd ratios between chondrites and terrestrial materials.

1. Introduction

Nucleosynthetic isotope anomalies in bulk chondrites and differentiated meteorites, which are characterized by isotopic deviations from terrestrial materials, have become a well-studied phenomenon for several refractory elements including Ti, Cr, Sr, Mo, and Ba (e.g., Bermingham et al., 2016; Burkhardt et al., 2011; Dauphas et al., 2004; Moynier et al., 2012; Trinquier et al., 2007, 2009; Yokoyama et al., 2015). These findings point to the existence of planetary-scale isotopic heterogeneities in refractory heavy elements as a result of the heterogeneous distribution of presolar grains in the protosolar nebula before the onset of planetesimal formation. In contrast, some elements exhibit uniform isotope compositions across different meteorite groups, including Te (Fehr et al., 2006; Fukami and Yokoyama, 2017), Os (Yokoyama et al., 2007, 2010), and Hf (Sprung et al., 2010). Such inconsistencies in isotope distributions are critical for understanding the physical and chemical processes that occurred in the solar nebula and/or in planetary bodies.

High-precision Nd isotope analyses of bulk chondrites have been the focus of recent interest in the cosmochemistry community (Andreasen and Sharma, 2006; Boyet and Carlson, 2005; Burkhardt et al., 2016; Carlson et al., 2007; Gannoun et al., 2011). A pioneering study on high-precision Nd isotope analyses with thermal ionization mass spectrometry (TIMS) found that bulk chondrites possess ^{142}Nd/^{144}Nd ratios that are approximately

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20 ppm lower than those in terrestrial rocks (Boyet and Carlson, 2005). The cause of this anomaly was interpreted to be Sm–Nd fractionation via early differentiation of the terrestrial mantle.

Variations in $^{142}Nd/^{144}Nd$ ratios have been reported for three types of chondrites (enstatite, ordinary, and carbonaceous chondrites) in subsequent studies (Carlson et al., 2007; Gannoun et al., 2011). Because the $^{142}Nd/^{144}Nd$ ratios in chondrites roughly correlate with the ratios of non-radiogenic Nd isotopes (e.g., $^{148}Nd/^{144}Nd$, $^{150}Nd/^{144}Nd$), it has been suggested that the isotopic variation in Nd isotopes results from nucleosynthetic isotopic heterogeneities in the early Solar System. Importantly, these studies argued that the isotopic composition of terrestrial materials cannot be reproduced by the heterogeneous distribution of isotopically anomalous grains in the solar nebula. Dickin (2016) pointed out that the regression line obtained from plotted data for the three chondrites types does not pass the terrestrial composition on a $^{142}Nd/^{144}Nd$ versus $^{148}Nd/^{144}Nd$ diagram.

Recently, two studies reported intriguing results from high-precision Sm–Nd isotope measurements on bulk aliquots of chondrites, as well as calcium- and aluminum-rich inclusions (CAIs) and their mineral fractions. Burkhardt et al. (2016) found that non-radiogenic isotopes in chondrites and CAIs from the carbonaceous chondrite Allende (CV3) display nucleosynthetic anomalies. The authors argued that the Sm–Nd isotope anomalies in the chondrites could be explained by a deficit in *s*-process components relative to the Earth, although the anomalies in the bulk Allende material required an additional anomalous component. In contrast, Bouvier and Boyet (2016) found that the bulk CAIs and their mineral fractions from three carbonaceous chondrites had non-radiogenic Nd isotope compositions similar to that of a terrestrial standard, excluding CAIs from NWA 2364 (CV3). In addition, the authors argued that the CAIs without nucleosynthetic isotope anomalies shared a $^{146}Sm–^{142}Nd$ isotopic evolution that is consistent with that of the Earth. In spite of the contrasting results obtained in the two studies, both studies reached the conclusion that the bulk silicate Earth has a chondritic Sm/Nd ratio as well as chondritic abundances of other refractory elements, negating the need for a hidden reservoir model or collisional erosion scenario to explain the elevated $^{142}Nd/^{144}Nd$ ratio of the Earth's mantle relative to chondrites.

Bulk carbonaceous chondrites generally show the largest nucleosynthetic isotope anomalies for refractory heavy elements (e.g., Cr, Sr, Mo, Ru). In addition, the strength of the isotope anomalies for some elements (e.g., Cr, Sr) is variable across different groups of carbonaceous chondrites. Therefore, carbonaceous chondrites are well suited to preserving information about the distribution of isotopically anomalous carriers in the early Solar System. However, high-precision Nd isotope analyses of bulk carbonaceous chondrites, especially for presolar-grain-bearing unequilibrated chondrites, have been hindered due to analytical difficulties in achieving complete sample dissolution with acid digestion. The importance of complete sample digestion in nucleosynthetic isotope anomaly studies on unequilibrated chondrites has been demonstrated in several previous works (Sr: Yokoyama et al., 2015; Ba: Bermingham et al., 2016; Os: Yokoyama et al., 2007, 2010). In this study, we performed high-precision Nd isotope analyses on various types of ordinary and carbonaceous chondrites using TIMS with an improved dynamic multi-collection method (Fukai et al., 2017). Notably, samples containing acid-resistant presolar grains were processed by a new acid digestion technique that achieved complete dissolution of presolar grains (Yokoyama et al., 2015). With the new high-precision Nd isotope data for ordinary and carbonaceous chondrites, we revisit the origin of nucleosynthetic Nd isotope variability in chondrites and terrestrial materials.

2. Experimental procedures

We investigated three terrestrial samples (JB-3, basalt; JA-2, andesite; JR-2, rhyolite), six carbonaceous chondrites (Tagish Lake, C2-ungrouped; Murchison, CM2; Dho 1432, CR2; Allende, CV3; DaG 190, CO3; NWA 2090, CO3), and nine ordinary chondrites (Adrian, H4; Kesen, H4; Forest City, H5; Saratov, L4; Etter, L5; Modoc (1905), L6; Hamlet, LL4; Tuxtuac, LL5; Saint-Séverin, LL6). The meteorite chips were cleaned with distilled acetone and H_2O for 30 min, and then powdered using either an agate mortar and pestle or a sample homogenizer (Multi-beads shocker®, Yasui Kikai, Japan), with which the rock chips were pulverized in polyethylene bottles at 2500 rpm for 900-s.

The concentrations of Nd and Sm in the meteorite samples were determined using the isotope dilution method with a quadrupole-type ICP-MS (X-Series 2, Thermo Fisher Scientific, USA) following the procedure described in Kagami and Yokoyama (2016). In brief, aliquots (∼5%) of meteorite sample solutions prepared by acid digestion for Nd isotope analysis (see below) were transferred to Teflon vessels to which ^{145}Nd- and ^{149}Sm-enriched spikes were added. The vessels were tightly capped and heated at 100 °C overnight to ensure isotopic equilibrium. Then, the solutions were dried and dissolved in 3 M HNO_3 (0.5 mL). Subsequently, the sample solutions were passed through a 100–150-μm TRU resin (Eichrom) to remove major elements. Finally, the $^{145}Nd/^{146}Nd$ and $^{147}Sm/^{149}Sm$ ratios of the sample solutions were determined simultaneously with ICP-MS.

The analytical protocol for the determination of Nd isotope compositions in meteorites and terrestrial rocks consisted of sample digestion, chemical separation, and mass spectrometry, the details of which are described in Yokoyama et al. (2015), Kagami and Yokoyama (2016), and Fukai et al. (2017), respectively. Here we outline the analytical protocol and modifications from the previous methods that were made in the present study.

Ordinary and carbonaceous chondrites with a petrologic grade of less than 4 are known to contain isotopically anomalous acid-resistant presolar grains such as SiC and graphite. These meteorite samples (∼0.5 g) were placed in a polytetrafluoroethylene (PTFE) insert (50 mL vol.) in a high-pressure digestion system, DAB-2 (Berghof, Germany), together with 6.8 mL 30 M HF (Kanto Chemical, AAS grade with distillation), 4.8 mL 16 M HNO_3 (Mitsubishi Chemical, EL grade with distillation), and 9.6 mL 96% H_2SO_4 (Kanto Chemical, Ultrapur-grade). The insert was then placed in a stainless steel jacket and heated at 250 °C for 48 h under high pressure. The effectiveness of this approach in decomposing refractory presolar grains was confirmed by Yokoyama et al. (2015). After sample digestion, the PTFE insert was placed on a hotplate at 220 °C in order to completely evaporate the HF and HNO_3. The solution was transferred to a quartz glass beaker and dried at 310 °C in order to evaporate the remaining H_2SO_4. The residue in the PTFE insert was dissolved in 6 M HCl and transferred to a glass beaker, then dried at 150 °C. Finally, the dried sulfate-containing residue was dissolved in 20 mL of 1 M HCl for chemical separation (see below). Chondrite samples with a petrologic grade of 4 or greater were digested using the normal procedures described in Yokoyama et al. (2017). In brief, the powdered samples (up to 2 g) were placed in Teflon vessels with a 1:1:1 mixture of 30 M HF, 12 M $HClO_4$ (TAMA Chemical, AA-100 grade), and 16 M HNO_3 in proportions of 1 mL of each acid per 0.1 g of sample. The sample solutions were heated at 120 °C for 12 h, at 165 °C for 20 h, and at 195 °C until dry. To completely decompose insoluble fluorides, the same amount of 12 M $HClO_4$ was added to the dried sample and heated at 165 °C for 20 h and at 195 °C until dry. The dried solutions were finally dissolved in 20 mL of 1 M HCl for chemical separation.