

**NUCLEOSYNTHETIC NEODYMIUM ISOTOPE ANOMALIES IN CARBONACEOUS AND ORDINARY CHONDRITES.** R. Fukai<sup>1</sup> and T. Yokoyama<sup>1</sup> <sup>1</sup>Department of Earth and Planetary Sciences, Tokyo Institute of Technology, Japan (fukai.r.aa@m.titech.ac.jp).

**Introduction:** A variety of isotope anomalies have been discovered in bulk chondrites and differentiated meteorites (e.g., Cr, Sr, Mo [1-3]). These results point to the existence of planetary-scale isotope heterogeneities for refractory heavy elements, which are most likely due to the heterogeneous distribution of presolar grains (e.g., SiC, graphite) in the protosolar nebula before the onset of planetesimal formation. By contrast, some elements exhibit uniform isotope compositions across different meteorite groups (e.g., Te, Os, Hf [4-6]). Such inconsistencies regarding the isotope distribution are critical to understanding the processes occurred in the solar nebula and/or in planetary bodies.

High precision Nd isotope analyses in meteorites have been the center of interest in recent cosmochemistry community [7-10]. One of the most remarkable results is that chondrites possess  $^{142}\text{Nd}/^{144}\text{Nd}$  ratios ~20 ppm lower than those in terrestrial rocks [7]. The anomaly was interpreted to be caused by the Sm-Nd fractionation via early differentiation of the terrestrial mantle. On the other hand, variations in stable Nd isotopes (e.g.,  $^{148,150}\text{Nd}/^{144}\text{Nd}$ ) have been documented in chondrites [9]. Although the authors concluded that the observed variation was due to incomplete digestion of presolar grain-bearing samples, the existence of Nd isotope anomalies in bulk aliquots of chondrites remains unclear unless high precision Nd isotope data with complete sample digestion become available.

In this study, we revisit high precision Nd isotope analysis of chondrites coupled with a new sample digestion technique that confirms complete dissolution of acid resistant presolar grains. We also develop a modified dynamic multicollecion method using TIMS to improve the analytical reproducibilities. Finally, we discuss the extent of Nd isotopic heterogeneities across different types of chondrites.

**Experimental:** We investigated one terrestrial basalt (JB-3), five carbonaceous chondrites (Tagish Lake, C2-ung; Dhofar 1432, CR2; Allende, CV3; NWA 2090, CO3; Dar al Gani 190/082, CO3), eight ordinary chondrites (Kesen, H4; Forest city, H5; Etter, H6; Saratov, L4; Modoc (1905), L6; Hamlet, LL4; Tuxtuac, LL5; Saint-Séverin, LL6). The meteorite chips were cleaned with acetone and H<sub>2</sub>O, then powdered using an agate mortar and pestle. The ordinary chondrites with a petrologic grade greater than 3.6 were dissolved by a conventional acid digestion method using HNO<sub>3</sub> + HF + HClO<sub>4</sub> [11]. For carbonaceous chondrites, each sample was digested using a

high-pressure digestion system (DAB-2, Berghof) with HF + HNO<sub>3</sub> + H<sub>2</sub>SO<sub>4</sub> to completely dissolve acid resistant presolar grains [2].

The Nd isotope compositions were measured by TIMS (Triton-plus, Tokyo Tech). In previous studies, Nd isotope compositions of bulk meteorites have been commonly measured in the static method with TIMS, which may be affected by the time-related deterioration of Faraday cups [12]. In contrast, the “multi-static” [13] or “dynamic” methods can reduce the effect of cup deterioration by acquiring Nd isotopes with multiple lines of different cup configurations within a single analytical cycle. In this study, we developed a modified “dynamic” method in which  $^{142,148,150}\text{Nd}/^{144}\text{Nd}$  ratios were obtained with a 2-line cup configuration. In addition, we corrected the effect of the time difference between two lines within a cycle. The results were obtained by averaging 360 ratios with 2σ rejection.

To test the analytical reproducibility in the dynamic method, we repeatedly analyzed a standard sample (JNdi-1) for eight months (April to November, 2015). The long-term reproducibilities obtained in the dynamic method were 4.2 ppm, 6.6 ppm and 9.7 ppm for  $^{142}\text{Nd}/^{144}\text{Nd}$ ,  $^{148}\text{Nd}/^{144}\text{Nd}$ ,  $^{150}\text{Nd}/^{144}\text{Nd}$  (n = 35), which are 2–11 times superior to the static and multistatic method. (The Nd isotope ratios are reported in the μNd notation which is parts per 10<sup>6</sup> relative deviation from the standard, JNdi-1)

**Results and Discussion:** Fig. 1 shows the  $\mu^{142}\text{Nd}-\mu^{148}\text{Nd}$  and  $\mu^{142}\text{Nd}-\mu^{150}\text{Nd}$  plots for meteorite samples analyzed in this study. Repeated analyses of the terrestrial basalt (triangles: n = 6) are all plotted within the range of analytical uncertainties for JNdi-1, confirming the accuracy of our analytical technique. Ordinary chondrites (circles) show uniform isotope anomalies for  $\mu^{142}\text{Nd}$  (−12 ± 5 ppm),  $\mu^{148}\text{Nd}$  (10 ± 8 ppm) and  $\mu^{150}\text{Nd}$  (20 ± 12 ppm). Although the  $\mu^{142}\text{Nd}$  values for ordinary chondrites obtained in this study are generally consistent with those of previous studies (small circles), positive anomalies in  $\mu^{148}\text{Nd}$  and  $\mu^{150}\text{Nd}$  were not recognized in previous studies. Because ordinary chondrites with petrologic grade greater than 3.6 would not contain isotopically anomalous acid-resistant presolar grains, Nd isotope anomalies observed are not caused by the incomplete sample digestion. Rather, the discrepancies are simply the issue of analytical precision.

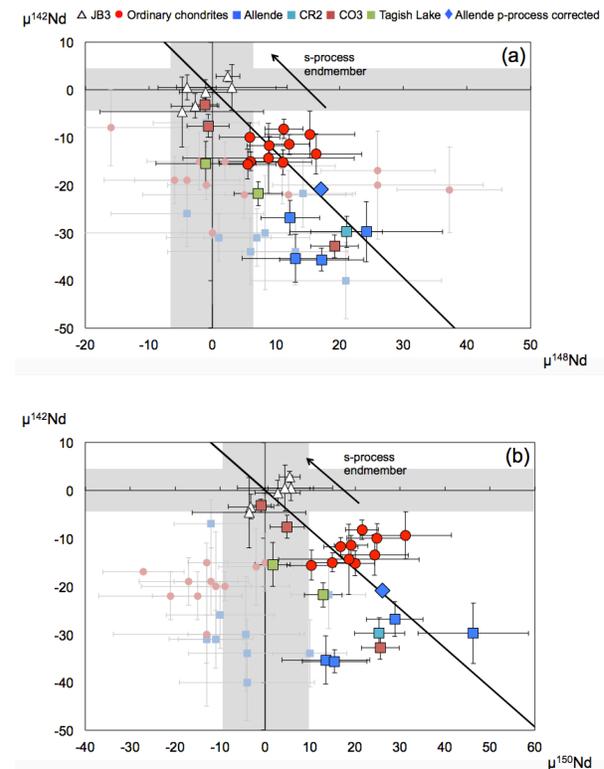
In contrast to ordinary chondrites, carbonaceous chondrites (squares) show variable Nd isotope anomalies exceeding analytical uncertainties. Again, the vari-

ation is not caused by incomplete sample digestion because the samples were decomposed using a high-pressure digestion system with HF + HNO<sub>3</sub> + H<sub>2</sub>SO<sub>4</sub>. As presented in Fig. 1, individual carbonaceous chondrites are categorized into three groups as a function of  $\mu^{142}\text{Nd}$ ; NWA 2090 (−5 ppm), Tagish Lake (−20 ppm), and Allende, DaG 190/082, and Dhofar 1432 (−30 ppm). Of these meteorites, two CO chondrites have distinctive  $\mu\text{Nd}$  values. This discrepancy could be due to the effect of secondary processes for NWA 2090 including thermal metamorphism in the parent body and terrestrial weathering, because this meteorite have a  $^{143}\text{Nd}/^{144}\text{Nd}$  ratio (0.51230) markedly lower than the other carbonaceous chondrites analyzed in this study ( $0.51261 \pm 0.00005$ ). Gannoun et al. [10] argued that the  $\mu\text{Nd}$  values in chondrites which are not plotted on the  $^{147}\text{Sm}$ – $^{143}\text{Nd}$  isochron of bulk chondrites could be disturbed by such secondary processes. Therefore, we exclude this meteorite from the following discussion.

The bold lines with negative slopes in Fig. 1 represent the mixing line between the terrestrial composition and the putative *s*-process endmember calculated from [14]. The data points for ordinary chondrites are generally plotted on this mixing line. This means that the isotope anomalies in ordinary chondrites are induced by the heterogeneous distribution of *s*-process nuclides in early Solar System. By contrast, most of the carbonaceous chondrites deviate from the mixing line towards the direction with lower  $\mu^{142}\text{Nd}$  values. We presume that the offset from the mixing line is caused by the heterogeneous distribution of *p*-nuclides in the early Solar System, because a part of  $^{142}\text{Nd}$  was produced by the *p*-process nucleosynthesis and  $\alpha$  decay of a pure *p*-nuclide  $^{146}\text{Sm}$ . Previous studies reported deficits of a pure *p*-nuclide  $^{144}\text{Sm}$  in carbonaceous chondrites compared to the terrestrial component, whereas ordinary chondrites showed no anomalies in  $\mu^{144}\text{Sm}$  [8-9]. Andreasen and Sharma [8] calculated that the extent of  $^{144}\text{Sm}$  deficits in Allende ( $\mu^{144}\text{Sm} = -120$  ppm) corresponded to the shift of −11 ppm in  $\mu^{142}\text{Nd}$  relative to ordinary chondrites. With this correction, the mean  $\mu\text{Nd}$  values of our multiple measurements for Allende are plotted on the terrestrial vs *s*-process mixing line (blue diamond; Fig. 1).

Our results indicate that negative  $\mu^{142}\text{Nd}$  values observed in chondrites simply reflect the heterogeneous distribution of *s*-process nuclides for ordinary chondrites and *s*- plus *p*-process nuclides for carbonaceous chondrites. Although the Earth and parent bodies of chondrites do not share building blocks with a common Nd isotopic composition, the excess  $^{142}\text{Nd}$  signature of the Earth would not necessarily require the existence of a hidden reservoir with a subchondritic

Sm/Nd ratio deep in the Earth's mantle as proposed previously [7,9].



**Fig. 1** Plots of  $\mu^{142}\text{Nd}$ ,  $\mu^{148}\text{Nd}$  and  $\mu^{150}\text{Nd}$  values in chondrites and terrestrial samples. Error bars of individual data points are 2SE of single measurements. The gray zones are uncertainties (2SD) of the standard analyzed by the dynamic method. Data obtained in previous studies (Red; Ordinary chondrites, Blue; Allende [7-10]) are also plotted for comparison as small symbols.

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