

## Mass-dependent stable isotope fractionation of Nd in terrestrial materials

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Every element suffers from isotope fractionation during a chemical reaction. Both equilibrium and kinetic chemical reactions fractionate isotopes of an element. Isotope fractionation induced variations of isotope composition in nature have been studied widely on light elements such as H, O, C, N and S. Large difference of relative mass between isotopes in these elements leads to large and easily measurable variations in their isotope compositions. No natural variations in isotope compositions of high-atomic-number elements are expected beyond analytical error because of the small relative mass differences between isotopes in these elements. However, recent advance in mass spectrometry have opened the door to high-precision stable isotope analysis of intermediate to high-atomic number elements. Stable isotopes of the newly introduced intermediate to high-atomic number elements are referred as non-traditional stable isotopes. Studies on the non-traditional stable isotopes have showed that mass-dependent fractionation induced variations of isotope compositions in nature are measurable in every elements investigated.

This study has focused on one of the rare earth elements (REEs), Nd, as a target for a new non-traditional stable isotope study. Rare earth elements are one of the most useful elements in geochemistry. Elemental fractionation pattern of REEs is widely used in geochemical studies to constrain the origin of geological materials. Moreover, abundance of the radiogenic nuclide  $^{143}\text{Nd}$ , product of the long-lived Sm-Nd radioactive decay system, provides chronological constraints of the sample. Combining stable isotopes with these two existing methodology, multiple information regarding the age, the origin and process will be obtained from a single element (or a single group of elements) to constrain the history of geological materials.

A combined double-spike TIMS technique is developed in this study for high-precision stable isotope analysis of Nd. The long-term reproducibility of the epsilon  $^{146}\text{Nd}$  (relative deviation of  $^{146}\text{Nd}/^{144}\text{Nd}$  in parts per  $10^4$  with reference to average of JNdi-1 measurements) values of JNdi-1 was  $\pm 0.2$  (2SD,  $n = 44$ ). Mass-independent fractionation can be detected by the developed technique if present. Various terrestrial materials were analyzed for Nd stable isotopes. The stable Nd isotope compositions of igneous rocks (including basalts, granites and rhyolites), Fe-Mn nodule and coral were epsilon  $^{146}\text{Nd} = -0.2 \pm 0.5$  (2SD,  $n = 7$ ),  $+0.2$  and  $+0.2$ , respectively, and agreed well within analytical error. REEs in Fe-Mn nodule and coral are of seawater origin. The consistency of the epsilon  $^{146}\text{Nd}$  values in Fe-Mn nodule and coral implies that stable isotope composition of Nd in these two materials is a good proxy of that of modern seawater. Large variation in Nd isotope composition was found among marine carbonate rocks: epsilon  $^{146}\text{Nd}$  values of 16 marine carbonate rocks range from  $-0.1$  to  $+2.6$ . Difference of the REE concentrations between carbonate rocks and organic calcite, precursor material of the carbonate rocks, suggests that REE was concentrated in carbonate rocks via inorganic chemical reaction between calcite and seawater during diagenesis. All the observed isotopic variation of Nd was mass-dependent. The possibility of mass-independent fractionation of REEs will be also discussed.