

# A PRELIMINARY ISOTOPIC STUDY ON FOUR YAMATO DIOGENITES –Sm-Nd AND Rb-Sr SYSTEMATICS–

Noboru NAKAMURA

*Department of Earth Sciences, Faculty of Science, Kobe University,  
Rokkodai-cho, Nada-ku, Kobe 657*

**Abstract:** Precise Sm and Nd isotopic measurements have been performed on whole rocks of four unique Yamato diogenites (Yamato-74013, -74037, -74097 and -74136). Although the observed  $^{143}\text{Nd}/^{144}\text{Nd}$  ratios show some variations, ranging from 0.5126 to 0.5146, the data do not define an isochron. Rb-Sr measurements for mineral separates and whole rocks from the meteorites also do not yield a definite age information for these meteorites. The present results suggest that multiple young (possibly younger than 2.0 b.y.) metamorphic events may be responsible for the disturbance of the Rb-Sr and Sm-Nd systems of the meteorites.

## 1. Introduction

Several fragments (0.2–2 kg) of diogenitic meteorite were collected on the bare ice sheet near the Yamato Mountains, Antarctica, in 1974 (YANAI, 1978). According to the mineralogical and petrological studies (TAKEDA *et al.*, 1978; YABUKI *et al.*, 1978), the Yamato-74 diogenites are unique in their textures; unlike usual diogenites they are not brecciated and the pyroxene of the meteorites seems to have recrystallized, and trace amounts of plagioclase and tridymite occur in interstices of orthopyroxene grains.

From the close genetic relationships of diogenites with eucrites and howardites with respect to chemical, petrological and isotopic compositions (McCARTHY *et al.*, 1973; STOLPER, 1977; CLAYTON *et al.*, 1976), one can conceive the early origin of these meteorites. However, because diogenites are monomineralic and contain extremely low abundances of key elements for isotopic measurements, the radiometric age determination for diogenites has not been fully undertaken. To understand the evolution history of diogenites as well as eucrites and howardites, the age data on diogenites are urgently required.

The purpose of this paper is to present the first Sm-Nd and Rb-Sr isotopic data for the diogenites from Antarctica, and to give a brief discussion of an evolution history of the diogenitic meteorites.

## 2. Experimental

The specimens of Yamato-74013, -74037, -74097 and -74136 were provided by the National Institute of Polar Research, Japan. The sample processings of these meteorites in the field are described by YANAI (1978). Most of the analytical procedures in this work were carried out in a controlled clean laboratory at the U. S. Geological Survey, Denver in cooperation with Dr. M. TATSUMOTO. The chemical and mass spectrometric procedures were almost the same as those by NAKAMURA *et al.* (1976) with some modifications; because of the extremely low abundances of Sm and Nd in the samples, REE were recovered with  $\text{Fe}(\text{OH})_3$  coprecipitation as the first step using ammonium hydroxide. Other Sm-Nd chemistry was basically the same as our regular procedures. The isotope ratio measurements were also made with facilities of the U. S. Geological Survey, Denver. The mass spectrometric data acquisition systems have been changed recently to a digital controlling system which covers a large mass range. The isotopic measurements of NBS Nd and Sr standards have been carried out repeatedly during the course of this study. No significant isotopic ratio shift has been detected before and after the modifications. (The results of Nd isotopic measurements of NBS Nd standard will be published elsewhere.) For the Nd isotopic and Sm and Nd concentration measurements, 4–5 g of whole rock samples were decomposed. This sample size corresponds to  $\sim 50\text{--}120$  ng Nd for isotopic measurements. For Rb-Sr analysis,  $\sim 180$  mg of orthopyroxene and a similar amount of coarse-grained chromite were separated by hand-picking. Low density fractions ( $\rho \leq 2.85$  and  $2.85 \leq \rho \leq 3.3$  g/cm<sup>3</sup>) from Yamato-74013 were also collected using a heavy liquid.

## 3. Results and Discussion

### 3.1. Sm-Nd systematics

The blank effects on Sm and Nd concentrations were slightly higher than those of NAKAMURA *et al.* (1976), but still less than 0.5% of the total contents. The effects of the blanks on the isotopic measurements for the samples were negligible. The results of the Sm-Nd analyses are given in Table 1. The Sm concentrations in the four diogenites analyzed here are similar to or slightly less than that of the Shalka meteorite (SCHMITT *et al.*, 1963). Precise  $^{143}\text{Nd}/^{144}\text{Nd}$  values were obtained for three diogenites, Yamato-74013, -74097 and -74136, but the isotopic measurements for Yamato-74037 were not successful owing to an incomplete separation of Nd. The  $^{147}\text{Sm}/^{144}\text{Nd}$  ratios range from 0.209 to 0.266 while the  $^{143}\text{Nd}/^{144}\text{Nd}$  ratios range from 0.5126 to 0.5146, as shown in Table 1. If these four sets of data are plotted on the Sm-Nd evolution diagram, the data points do not form a line, within analytical uncertainties.

Table 1. Sm-Nd analytical results for Yamato diogenites.

Sample	Sm (ppm)	Nd (ppm)	$\frac{^{147}\text{Sm}^+}{^{144}\text{Nd}}$	$\frac{^{143}\text{Nd}^{*+}}{^{144}\text{Nd}}$
Yamato-74037	0.01395	0.04133	$0.2093 \pm 3$	$0.5126 \pm 3$
-74013	0.007007	0.01786	$0.2371 \pm 2$	$0.513157 \pm 42$
-74136	0.01067	0.02731	$0.2368 \pm 1$	$0.514350 \pm 30$
-74097	0.00980	0.02222	$0.2655 \pm 4$	$0.514649 \pm 23$

\* Ratios are normalized to  $^{150}\text{Nd}/^{144}\text{Nd}=0.236433$ .

+ Uncertainties correspond to the last figures and are  $2\sigma$  mean.

From morphological features, Yamato-74013, -74097 and -74136 meteorites have been suggested to belong to one meteorite fall (YANAI, 1978). The similarity in the major element compositions of orthopyroxenes from these meteorites also supports this suggestion (TAKEDA *et al.*, 1978). On the other hand, the REE abundances in these meteorites show some variations in the Sm/Nd ratios (MASUDA *et al.*, 1979). Therefore, if these meteorites have a common thermal history, one would expect to obtain a whole-rock isochron for the Sm-Nd systems of the meteorites. However, as mentioned above, in spite of the presence of some variations in  $^{143}\text{Nd}/^{144}\text{Nd}$  ratios, the data points do not define an isochron.

To investigate this problem, a  $^{143}\text{Nd}$  evolution diagram is shown in Fig. 1. If we assume the same initial ratio for diogenites as that for eucrites (LUGMAIR *et al.*, 1976; NAKAMURA *et al.*, 1977), the calculations of model ages yield 4.5 b.y. for Yamato-74097, 4.8 b.y. for Yamato-74136, 4.1 b.y. for Yamato-74136 and 4.2 b.y. for Yamato-74037. These results suggest that the Sm-Nd systems in the meteorites under consideration were partially reset by later events after formation of the parent body, although the model ages obtained here may not have any physical meaning.

From features seen in Fig. 1, it is considered that the two diogenites Yamato-74097 and -74136 experienced similar thermal events (possibly at  $\sim 1.6$  b.y. age) in the same parent body. However, the Nd isotopic compositions of the other two diogenites, Yamato-74013 and -74037, are distinctly different from Yamato-74136 and -74097. Such large differences in isotopic composition are not easily explained in terms of a single thermal event, but can be explained by multiple events, if we assume the same parent body for these diogenites. However, it is not clear from the present results whether these meteorites came from the same parent body or not.

As demonstrated by the case of the Pasamonte eucrite (UNRUH *et al.*, 1977), the Sm-Nd systems in meteorites are not easily reset by moderate meteoritic impacts.

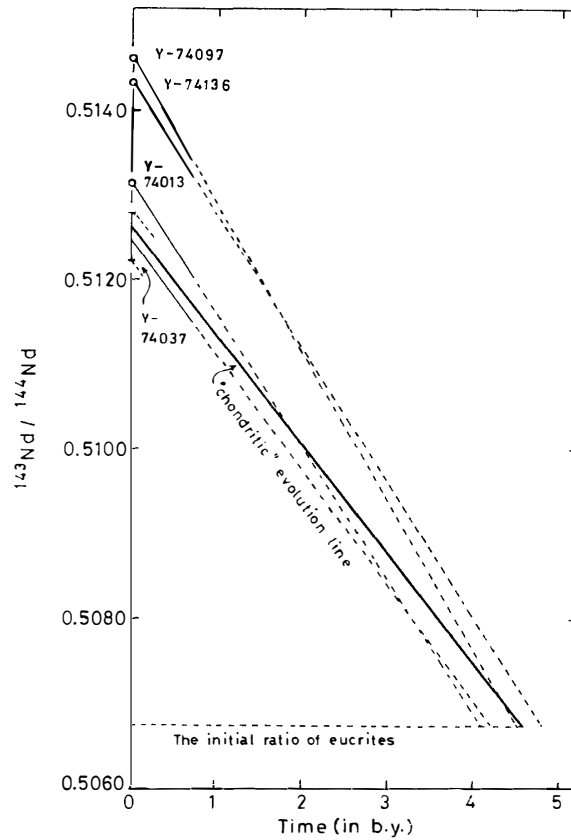


Fig. 1. The  $^{143}\text{Nd}$  evolution diagram for four Yamato diogenites. The close  $^{143}\text{Nd}$  evolution of Yamato-74097 and -74136 is suggested but other two meteorites, Yamato-74013 and -74037, show a different evolution trend.

However, the Sm-Nd system of plagioclase is most susceptible to such an event (LUGMAIR and SCHEININ, 1975). Such events are usually older than 4 b.y. On the other hand, as a rare case a young thermal event (at 1.3 b.y.) which reset a Sm-Nd system is also known for the Nakhla meteorite (NAKAMURA *et al.*, 1978).

### 3.2. Rb-Sr systematics

In Table 2, the Rb-Sr results for mineral separates of Yamato-74013 and whole rocks of the other two diogenites are presented. Although the blank levels in this work are almost the same as those of NAKAMURA *et al.* (1976), total amounts of Rb in the mineral separates ( $\rho \leq 2.85$  and  $2.85 \leq \rho \leq 3.3$  g/cm<sup>3</sup>) were too small to obtain accurate concentrations. Thus, the uncertainties in the Rb concentrations in these two density fractions and the hand-picked orthopyroxene and chromite are several per cent due to blank corrections. However, the Sr blanks for these samples were less than 0.5% except for chromite. The blank effect on chromite analysis is believed to be nearly two per cent. Therefore, the corresponding effects on the

Table 2. Rb-Sr results for the Yamato diogenites.

Sample	Rb (ppm)	Sr (ppm)	$\frac{87\text{Rb}}{86\text{Sr}}$	$\frac{87\text{Sr}^*}{86\text{Sr}}$
Yamato-74013				
$\rho < 2.85$	5.0	127.7	0.112	$0.69963 \pm 7$
$\rho = 2.85 - 3.3$	0.230	30.0	0.0220	$0.69963 \pm 3$
Opx	0.0194	0.545	0.103	$0.70102 \pm 5$
Chromite	0.0134	0.026	$1.51 \pm 15$	$0.70322 \pm 35$
Yamato-74037	0.0306	1.905	0.0465	$0.70025 \pm 5$
Yamato-74097	0.00330	0.2873	0.0107	$0.70293 \pm 7$

\* Uncertainties correspond to the last figures and are  $2\sigma$  mean.

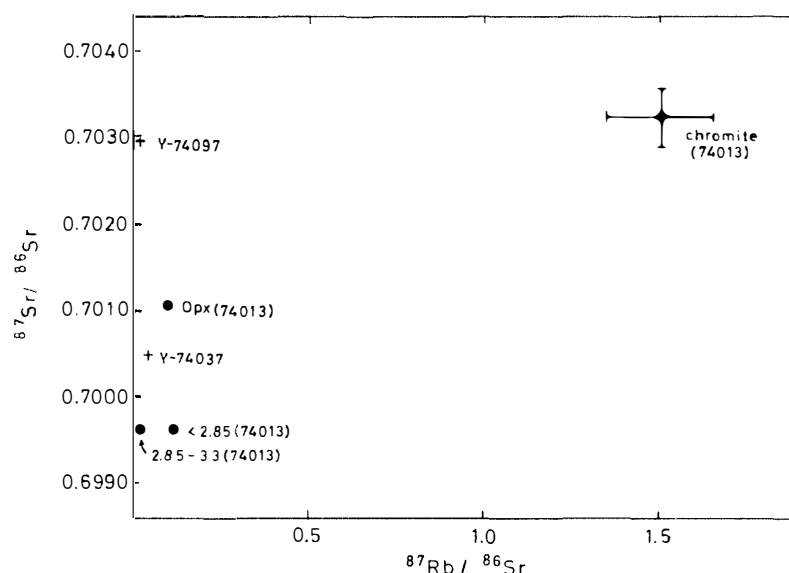


Fig. 2. Rb-Sr isochron diagram for Yamato diogenites. The solid circles indicate data points of mineral separates of Yamato-74013 and the crosses are for whole rocks of Yamato-74037 and -74097. (The Rb-Sr systems in Yamato diogenites are seriously disturbed by metamorphic events and therefore the data do not define an isochron.)

Sr isotopic ratios may be comparable with or less than the errors of mass spectrometric analyses, although the Sr isotopic ratios of the blanks are not determined in this work.

The data are plotted on a Rb-Sr evolution diagram in Fig. 2. Again, the data points are scattered in this diagram. Even if all conceivable factors which might cause experimental errors are taken into account, the isotopic variations cannot be explained solely in terms of experimental errors, but are considered to be real.

The concentrations of Rb and Sr in the low density fractions are quite high as compared with those in the orthopyroxene of Yamato-74013 and the whole rocks (Table 2). MASUDA *et al.* (1979) measured REE, major and minor element abundances in the acetone "float" fraction of Yamato-74010 (a similar type of diogenite). The present observation of high Rb and Sr concentrations in the low density fractions of Yamato-74013, is in general agreement with the elemental abundances in the acetone "float" fraction analyzed by MASUDA *et al.* (1979). The data suggest the presence of ultra-fine phase, enriched in elements with larger ionic radii. The presence of such a phase may result in a heterogeneous distribution of Rb and Sr in the whole-rock samples, like those found in Yamato-74037 and -74097, regardless of the homogeneity of major elements.

In Fig. 1, a model age for chromite of 2.0 b.y. has been calculated assuming the same initial  $^{87}\text{Sr}/^{86}\text{Sr}$  ratio as "BABI" (PAPANASTASSIOU and WASSERBURG, 1969). The age may give the upper limit for the time of thermal event which caused Rb-Sr re-equilibration. The tied lines between the data points of chromite and orthopyroxene and the low density fractions ( $\rho \leq 2.85$  and  $\rho = 2.85\text{--}3.3$  g/cm<sup>3</sup>) give the model ages of 1.1, 1.8 and 1.7 b.y., respectively. These young ages are rather similar to the  $^{40}\text{Ar}\text{--}^{39}\text{Ar}$  age obtained for Yamato-74097 meteorite (KANEOKA *et al.*, 1979). The  $^{40}\text{Ar}\text{--}^{39}\text{Ar}$  age spectrum of Yamato-74097 meteorite suggests that the K-Ar isotopic system in this meteorite was almost completely reset at  $\sim 1.1$  b.y. ago. It is considered that such a strong thermal event may have reset or partially reset the Rb-Sr system. Therefore, it is suggested that the model ages calculated above give the approximate time of metamorphic events for the Yamato diogenites.

The grain size of orthopyroxene in the Yamato diogenites is quite variable from portion to portion even on a one-centimeter scale (TAKEDA *et al.*, 1978; TAKEDA, personal communication, 1979). The heterogeneous distribution of trace elements could be a reflection of such a heterogeneity in texture. From the above discussion, it may be suggested that the observed variations in Rb/Sr and Sm/Nd ratios of whole rocks of Yamato diogenites are controlled mainly by the minor phases, probably the interstitial materials which might have formed during young metamorphic events.

Therefore, the disturbed Rb-Sr and Sm-Nd isotopic systems of Yamato diogenites may be the results of heterogeneous distribution of such minor phases.

Although the Yamato diogenites are finds, the specimens investigated in this work were quite fresh under binoculars and generally hard (TAKEDA *et al.*, 1978). Moreover, any chemical and petrological evidence for the Yamato diogenites which is suggestive of weathering effects in the bare ice has not been noted so far, though it might be prudent not to rule out the possibility that some of the chemical features observed here were brought about by terrestrial effects.

### 3.3. Conclusion

(1) In this work, the first Sm-Nd and Rb-Sr isotopic measurements for diogenites have been performed. Although a well-defined age was not obtained for the Yamato diogenites due to the isotopic disturbance, the high precision Nd isotopic measurements in this work demonstrate that the diogenites have significant variations in  $^{143}\text{Nd}/^{144}\text{Nd}$  ratio which would enable us to determine the age in an undisturbed system.

(2) The Rb-Sr and Sm-Nd systems of the Yamato diogenites have been partially reset by young multiple metamorphic events (possibly at 1.1~1.8 b.y. ago). It is difficult to deduce a definite time of the metamorphic events from the present results of isotopic analyses.

(3) From the present results and other information on diogenites, it is suggested that three major thermal events were responsible for the evolution of the parent body of diogenites; the first, the magmatic differentiation (4.55 b.y. ago), the second, the brecciation events as found for usual brecciated diogenites (possibly just after 4.55 b.y. ago) and the third, the youngest metamorphic events (~1.1–1.8 b.y. ago) which have been suggested for the Yamato diogenites in this work.

More detailed isotopic works are urgently required for the unique Yamato diogenites as well as the other types of diogenites to prove the model presented here.

### Acknowledgments

The author wishes to thank Prof. T. NAGATA and Dr. K. YANAI and the members of the field party of the Japanese Antarctic Research Expedition in 1974. He would like to express deepest gratitude to Dr. M. TATSUMOTO, U. S. Geological Survey, Denver for guiding him to geo-, cosmochemistry, helpful cooperation in this work and critical reading of the manuscript. He is also indebted to Prof. A. MASUDA, Kobe University for helpful cooperation and critical reading of the manuscript, and to Mr. D. COFFRANT and Dr. J. HAMET for assistance in data acquisition, and to Messrs. D. UNRUH and E. DAUGS for helping with mineral separation. He thanks Prof. H. TAKEDA, University of Tokyo, and Dr. M. MIYAMOTO, Kobe University, for discussion and valuable information on the samples during the course of this study. The author gratefully acknowledges the invaluable comments from Prof. M. HONDA and Dr. K. NOTSU. He is also indebted to Miss J. ASAKURA for typing the manuscript.

This work was supported, in part, by the Grant in Aid for Fundamental Scientific Research from the Ministry of Education.

## References

- CLAYTON, R. N., ONUMA, N. and MAYEDA, T. K. (1976): A classification of meteorites based on oxygen isotopes. *Earth Planet. Sci. Lett.*, **30**, 10–18.
- KANEOKA, I., OZIMA, M. and YANAGISAWA, M. (1979):  $^{40}\text{Ar}$ - $^{39}\text{Ar}$  age studies of four Yamato-74 meteorites. *Mem. Natl Inst. Polar Res., Spec. Issue*, **12**, 186–206.
- LUGMAIR, G. W. and SCHEININ, N. B. (1975): Sm-Nd systematics of the Stannern meteorite. *Meteoritics*, **10**, 447–448.
- LUGMAIR, G. W., MARTI, K., KURTZ, J. P. and SCHEININ, N. B. (1976): History and genesis of lunar troctolite 76535 or: how old is old? *Proc. Lunar Sci. Conf. 7th*, 2009–2033.
- MCCARTHY, T. S., ERLANK, A. J. and WILLIS, J. P. (1973): On the origin of eucrites and diogenites. *Earth Planet. Sci. Lett.*, **18**, 433–442.
- MASUDA, A., TANAKA, T., SHIMIZU, H., WAKISAKA, T. and NAKAMURA, N. (1979): Rare-earth geochemistry of Antarctic diogenites. *Mem. Natl Inst. Polar Res., Spec. Issue*, **15**, 177–188.
- NAKAMURA, N., TATSUMOTO, M., NUNES, P. D., UNRUH, D. M., SCHWAB, A. P. and WILDEMAN, T. R. (1976): 4.4 b.y.-old clast in Boulder 7, Apollo 17: A comprehensive chronological study U-Pb, Rb-Sr and Sm-Nd methods. *Proc. Lunar Sci. Conf. 7th*, 2309–2333.
- NAKAMURA, N., UNRUH, D. M., GENSHO, R. and TATSUMOTO, M. (1977): Evolution history of lunar mare basalts. Apollo 15 samples revisited. *Lunar Science VIII*. Houston, The Lunar Science Institute, 712–713.
- NAKAMURA, N., UNRUH, D. M. and TATSUMOTO, M. (1978): The young magmatic event in the Nakhla achondrite parent body. 4th Int. Conf. Geochronol. Cosmochronol. Isotope Geol., ed. by R. E. ZARTMAN. Snowmass-at-Aspen, U.S. Geological Survey, 305–306.
- PAPANASTASSIOU, D. A. and WASSERBURG, G. J. (1969): Initial strontium isotopic abundances and the resolution of small time differences in the formation of planetary objects. *Earth Planet. Sci. Lett.*, **5**, 361–376.
- SCHMITT, R. A., SMITH, R. H., LASCH, J. E., MOSEN, A. W., OLEHY, D. A. and VASILEVSKIS, J. (1963): Abundances of the fourteen rare-earth elements, scandium, and yttrium in meteoritic and terrestrial matter. *Geochim. Cosmochim. Acta*, **27**, 577–622.
- STOLPER, E. (1977): Experimental petrology of eucritic meteorites. *Geochim. Cosmochim. Acta*, **41**, 587–611.
- TAKEDA, H., MIYAMOTO, M., YANAI, K. and HARAMURA, H. (1978): A preliminary mineralogical examination of the Yamato-74 achondrites. *Mem. Natl Inst. Polar Res., Spec. Issue*, **8**, 170–184.
- UNRUH, D. M., NAKAMURA, N. and TATSUMOTO, M. (1977): History of the Pasamonte achondrite: Relative susceptibility of the Sm-Nd, Rb-Sr, and U-Pb systems to metamorphic events. *Earth Planet. Sci. Lett.*, **37**, 1–12.
- YANAI, K. (1978): Yamato-74 meteorites collection, Antarctica from November to December 1974. *Mem. Natl Inst. Polar Res., Spec. Issue*, **8**, 1–37.
- YABUKI, H., YAGI, K. and ONUMA, K. (1978): Petrological studies of Yamato-74 meteorites (1). *Mem. Natl Inst. Polar Res., Spec. Issue*, **8**, 142–155.

*(Received May 1, 1979; Revised manuscript received July 28, 1979)*