SIMS MEASUREMENT OF MAGNESIUM ISOTOPIC RATIOS IN CHONDRITES

Hiroshi NISHIMURA and Jun OKANO

Institute of Geological Sciences, College of General Education, Osaka University, 1-1, Machikaneyama-cho, Toyonaka 560

Abstract: Isotopic ratios of $^{25}Mg/^{24}Mg$ and $^{26}Mg/^{24}Mg$ have been measured for Mg-rich and Al-poor portions of Yamato, Plainview, Bruderheim, Leedey, Potter, Murchison, Leoville, Allende and Bondoc Peninsula meteorites with a Hitachi IMA 2A ion microprobe mass analyzer. Terrestrial samples of forsterite, biotite and anorthite have been also analyzed. In the three isotope plot, the data for ordinary chondrites of petrological types 5 and 6, an achondrite, a stony-iron meteorite and terrestrial samples are along the normal mass fractionation line through the reference point. On the contrary, the data for ordinary chondrites of petrological type 4 and carbonaceous chondrites fall on a straight line with the slope of unity with a few exceptions. This line can be explained to be the mixing of original magnesium and practically pure ^{24}Mg. This can be an evidence for the two component hypothesis of the primordial solar nebula.

1. Introduction

Isotopic abundance anomalies in primitive meteorite specimens are important to the understanding of the origin of the primordial solar nebula. The excess of ^{26}Mg found in several Al-rich inclusions in the Allende and Leoville carbonaceous chondrites (BRADLEY *et al.*, 1978; ESAT *et al.*, 1978; GRAY and COMPSTON, 1974; HUTCHEON *et al.*, 1977, 1978, 1979; LEE *et al.*, 1976, 1977a, b; LORIN *et al.*, 1977; NISHIMURA and OKANO, 1980; SHIMIZU *et al.*, 1978; WASSERBURG *et al.*, 1977a, b) and the excess of ^{16}O in an ordinary and some carbonaceous chondrites (CLAYTON *et al.*, 1973, 1977; CLAYTON and MAYEDA, 1977, 1978) lead us to a view that the primordial solar nebula consists of two or more components (CLAYTON *et al.*, 1973, 1977; SCHRAMM, 1978).

Excess ^{26}Mg correlating with Al concentration is thought to be due to the *in situ* decay of the extinct ^{26}Al (half life$=7.2\times10^5$ y). This ^{26}Al and the excess ^{16}O are both considered to be synthesized during explosive carbon burning process in a supernova explosion (ARNETT, 1969; ARNETT and TRURAN, 1969; CAMERON and TRURAN, 1977), and to be injected into the primordial solar nebula (CAMERON, 1978; CAMERON and TRURAN, 1977; SCHRAMM, 1978).

The isotopes of ^{24}Mg and ^{20}Ne would be produced together with ^{26}Al and ^{16}O during the explosive carbon burning (ARNETT, 1969), and these isotopes would have been injected into the solar nebula at the same time. Therefore, it may be possible to find the excess of ^{24}Mg in some primitive chondrites.

In order to find the remnant of the injected ^{24}Mg, we have measured magnesium isotopic ratios for Mg-rich and Al-poor portions of several chondrite samples, where the contribution of ^{26}Al is expected to be negligibly small. An ion microprobe mass analyzer was used to measure the isotopic ratios for the localized portions of the specimens.

Preliminary results are described in the following.

2. Experimental

The isotopic analysis has been carried out by a Hitachi IMA 2A ion microprobe mass spectrometer. 17 keV O_2^+ ions were used as primaries. The diameter and the current of the primary ion beam on the sample surface were 70–100 μm and $2–4\times10^{-7}$ A, respectively. The ultimate pressure in the sample chamber was about 3×10^{-5} Pa and the pressure under the isotopic ratio measurement was $4–8\times10^{-5}$ Pa.

Meteorite samples used are listed in Table 1 together with their classification. A terrestrial forsterite from Ehime Pref., Japan, a terrestrial biotite in granodiorite from Hyogo Pref., Japan, and a terrestrial anorthite from Miyakejima Island, Japan were also analyzed.

Most of these samples were spalled and the fresh surfaces were used in the point-to-point analysis. The specimens of Bruderheim, a few specimens of Allende and a terrestrial forsterite were polished and the flat surfaces were used.

In order to avoid the charge-up due to the positive ion bombardment, electron spray was applied to the sample surfaces. Mass scannings were repeated 50 to 90 times for one probed area over the range of $^{24}Mg^+$ to $^{26}Mg^+$. The record of mass spectra was automatically controlled by a microcomputer. An example of the mass spectra taken for the sample of Allende by using this controlling system is shown in

Table 1. Samples used for Mg isotopic analysis and their classification.

Sample	Classification	Sample	Classification
Ordinary chondrite		Carbonaceous chondrite	
Yamato-74155	H4†	Murchison	C2*
Yamato-74495	H4†	Leoville	C3*
Plainview	H5*	Allende	C3*
Yamato-74640	H6†	Achondrite	
Yamato-74190	L5-6†	Yamato-74159	EUC†
Bruderheim	L6*	Stony-Iron	
Leedey	L6*	Bondoc Peninsula	MES*
Potter	L6*		

EUC: eucrite, MES: mesosiderite.
† YANAI (1979), * WASSON (1974).

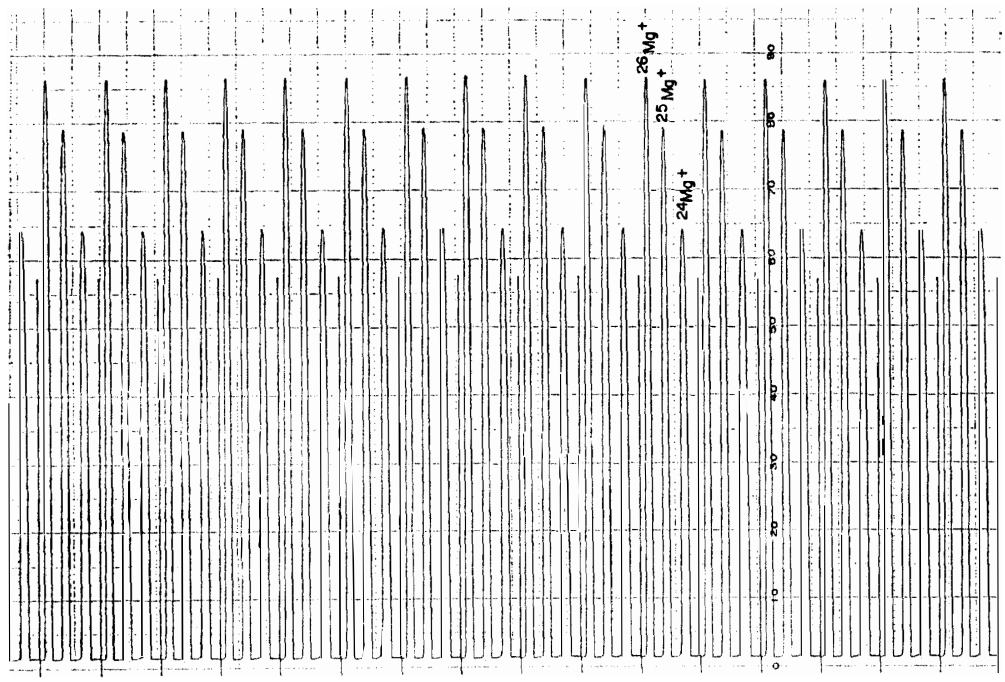

Fig. 1. Mass spectra repeated for the mass range of ^{24}Mg, ^{25}Mg and ^{26}Mg. Sample is the matrix of the Allende carbonaceous chondrite. The sensitivities for ^{25}Mg and ^{26}Mg are 10 times higher than the one for ^{24}Mg.

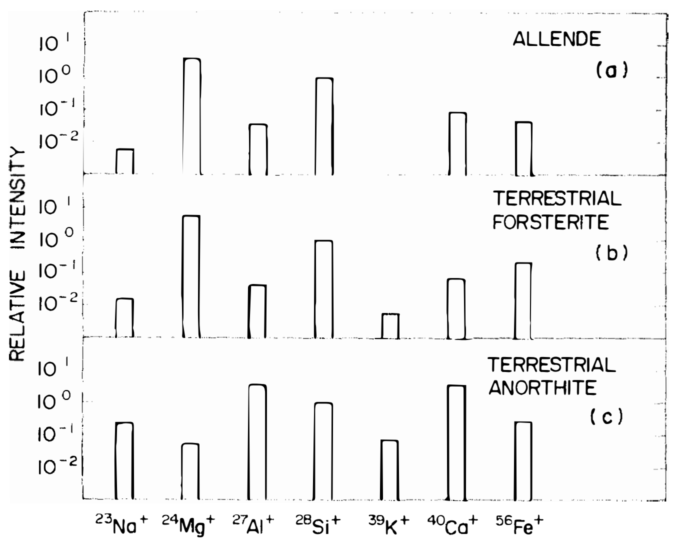

Fig. 2. Relative intensities of secondary ions for Allende (a), terrestrial forsterite (b) and terrestrial anorthite (c). By comparing these patterns, olivine-rich portions in meteorite specimens can be found.

Fig. 1. Intensities of $^{23}Na^+$, $^{24}Mg^+$, $^{27}Al^+$, $^{28}Si^+$, $^{39}K^+$, $^{40}Ca^+$ and $^{56}Fe^+$ were also recorded for each probed portion. Relative intensities of these secondary ions for Allende matrix, terrestrial forsterite and terrestrial anorthite are illustrated in Figs. 2a, 2b and 2c, respectively. By comparing these patterns with each other, olivine-rich portions in the meteorite specimens could be found. Especially, $^{23}Na^+/^{24}Mg^+$ and $^{27}Al^+/^{24}Mg^+$ ratios could be used as fine measures to localize the ion probe on the olivine-rich area.

3. Examination of Interferences

The interferences of molecular and doubly-charged ions to the peaks at mass numbers of 24, 25 and 26 have been extensively examined. Possible overlapping ionic species are listed in the second column of Table 2. In the third column of the table, are shown the maximum contribution of each overlapping ionic species to the subject peak.

The interferences of doubly-charged ions of Cr, Ti and Ca were estimated from the intensities of $^{53}Cr^{2+}$, $^{47}Ti^{2+}$ and $^{40}Ca^{2+}$. The estimation for the contributions of $^{12}C_2^+$, $^{12}C^{13}C^+$, $^{12}C_2H^+$, $^{13}C_2^+$, $^{12}C^{13}CH^+$, $^{12}C_2H_2^+$ and $^{12}C^{14}N^+$ was done in the following way. The ratios of $^{12}C_2^+/^{12}C^+$, $^{12}CN^+/^{12}C^+$ and $^{12}CH^+/^{12}C^+$ were at first evaluated to be about 0.032, 0.0055 and 0.021, respectively, for the samples of calcite and graphite. Using these values, the intensities at mass numbers of 12, 12.5, 13, 24, 25 and 26, and the

Table 2. Interferences of molecular and doubly-charged ions with the mass peaks of 24, 25 and 26.

Subject ionic species	Overlapping ionic species	Contribution to the subject peak
$^{24}Mg^+$	$^{12}C_2^+$	$<5.4\times10^{-6}$
	$^{23}NaH^+$	$<1\ \times10^{-6}$
	$^{48}Ca^{2+}$	$<4.1\times10^{-5}$
	$^{48}Ti^{2+}$	$<6.1\times10^{-6}$
$^{25}Mg^+$	$^{12}C^{13}C^+$, $^{12}C_2H^+$, $^{23}NaH_2^+$	$<1\ \times10^{-6}$
	$^{50}Ti^{2+}$	$<3.5\times10^{-6}$
	$^{50}Cr^{2+}$	$<9.4\times10^{-5}$
	$^{24}MgH^+$	$<1.0\times10^{-3}$
$^{26}Mg^+$	$^{13}C_2^+$, $^{12}C^{13}CH^+$, $^{12}C_2H_2^+$	$<1\ \times10^{-6}$
	$^{12}C^{14}N^+$	$<3.7\times10^{-6}$
	$^{52}Cr^{2+}$	$<8.5\times10^{-5}$
	$^{25}MgH^+$	$<1.3\times10^{-4}$

diatomic ion formation ratio*, we could evaluate the maximum contributions as shown in Table 2. The contribution of $^{23}NaH^+$ was found to be negligibly small based on the intensities at mass numbers of 23, 24 and 25 for a terrestrial anorthite sample. The contributions of $^{24}MgH^+$ and $^{25}MgH^+$ were estimated from the intensity ratios of mass numbers 25 and 26 to 24 for a terrestrial forsterite sample, and based on the simultaneous equations set up for the ratios of $^{25}Mg/^{24}Mg$ and $^{26}Mg/^{24}Mg$.

4. Results and Discussion

The isotopic ratios of $^{25}Mg/^{24}Mg$ and $^{26}Mg/^{24}Mg$ were calculated from the intensities of mass 24, 25 and 26. The deviations of these ratios from the reference values (SCHRAMM *et al.*, 1970), Δ_{25} and Δ_{26}, were calculated according to eq. (1).

$$\Delta_m=\left[\frac{(^{m}Mg/^{24}Mg)_{meas}}{(^{m}Mg/^{24}Mg)_{ref}}-1\right]\times 1000\ , \quad (m=25,\ 26) \qquad (1)$$

where $(^{25}Mg/^{24}Mg)_{ref}=0.12663$ and $(^{26}Mg/^{24}Mg)_{ref}=0.139805$ from SCHRAMM *et al.* (1970).

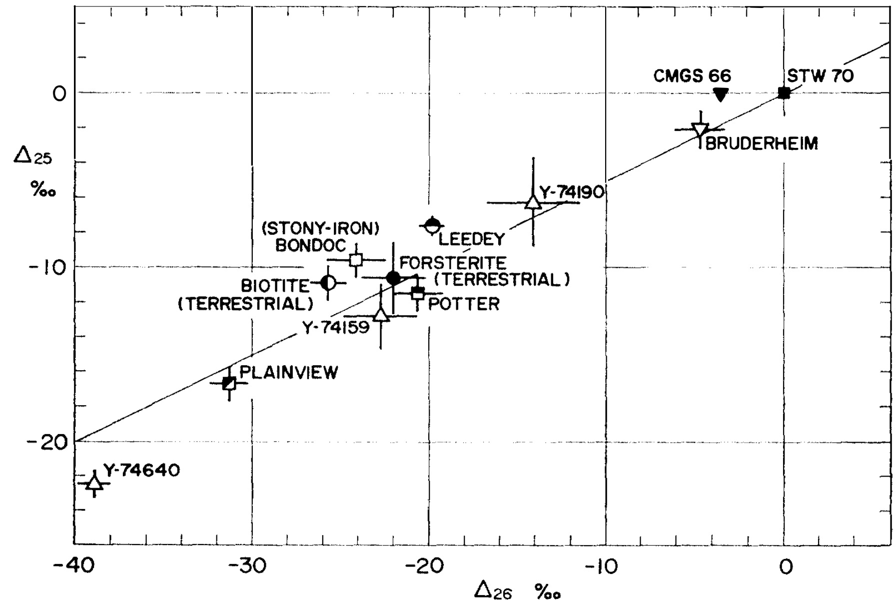

Fig. 3. Three isotope plots of magnesium for the samples of ordinary chondrites belonging to petrological types 5 and 6, an achondrite, a stony-iron meteorite, a terrestrial forsterite and a terrestrial biotite. The isotopic ratios at reference point (STW 70) are $(^{25}Mg/^{24}Mg)_{ref}=0.12663$ and $(^{26}Mg/^{24}Mg)_{ref}=0.139805$. A normal mass fractionation line through the reference point is illustrated. The definition of Δ_{25} and Δ_{26} appears in the text. STW 70: SCHRAMM et al. (1970), CMGS 66: CATANZARO et al. (1966).

* The diatomic ion formation ratio was calculated as $^{12}C_2^+$: $^{12}C^{13}C^+$: $^{13}C_2^+$=100: 2.2: 0.013 from the arithmetic combination of isotopic abundances of carbon.

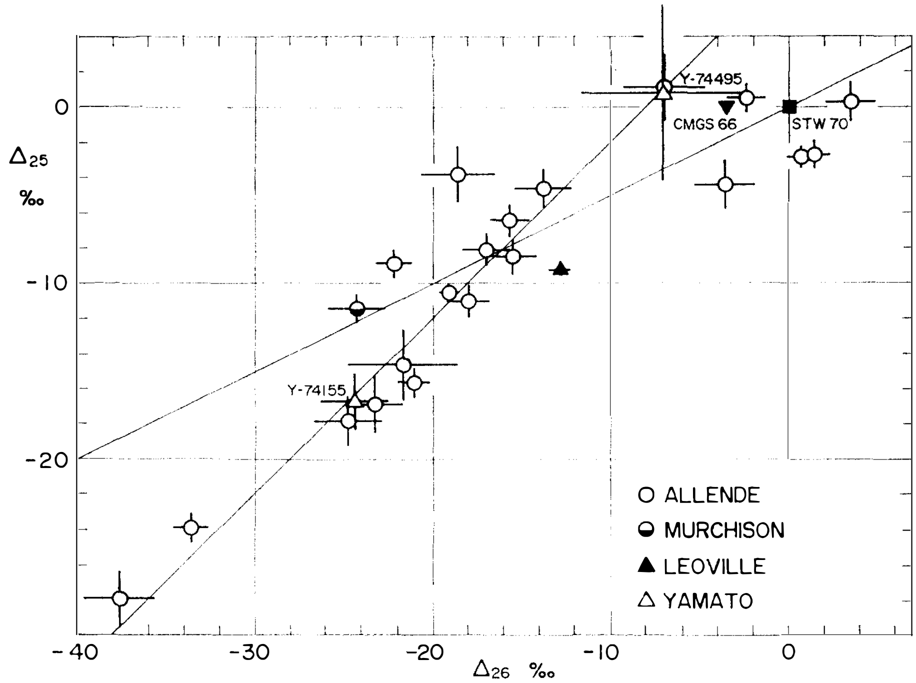

Fig. 4. Three isotope plots of magnesium for the samples of ordinary chondrite belonging to petrological type 4 and carbonaceous chondrites. The isotopic ratios at reference point (STW 70) are $(^{25}Mg/^{24}Mg)_{ref}=0.12663$, and $(^{26}Mg/^{24}Mg)_{ref}=0.139805$. A normal mass fractionation line through the reference point and a straight line with the slope of unity through most plotted points are illustrated. The definition of Δ_{25} and Δ_{26} appears in the text. STW 70: SCHRAMM et al. (1970), CMGS 66: CATANZARO et al. (1966).

The three isotope plots for each sample using Δ values are shown in Figs. 3 and 4. The serial numbers of the Yamato meteorites are put aside the plotted points. In Fig. 3, are shown the data for ordinary chondrites belonging to petrological types 5 and 6, an achondrite, a stony-iron meteorite and terrestrial samples. In Fig. 4, the data for ordinary chondrites of petrological type 4, and carbonaceous chondrites are plotted. The reference values from SCHRAMM *et al.* (1970) and CATANZARO *et al.* (1966) are also plotted in both the figures. The error bars illustrated in the figures represent twice of the standard deviation of the mean value.

In Fig. 3, most of the data are plotted near the normal mass fractionation line through the reference point with the slope of 1/2. On the contrary, most data plotted in Fig. 4 fall along a straight line with the slope of unity with a few exceptions.

The line with the slope of unity appearing in Fig. 4 may be explained as the mixing of two kinds of magnesium of different origins. One of them is magnesium with original isotopic composition in the primordial solar nebula. The other is of almost pure ^{24}Mg.

This practically pure ^{24}Mg could be produced at the same time as the production of ^{16}O and ^{26}Al, according to the theoretical consideration by ARNETT (1969). In other words, the anomaly of ^{24}Mg shown in Fig. 4 can be an evidence for two (or more) component hypothesis of the primordial solar nebula (CLAYTON *et al.*, 1973, 1977; SCHRAMM, 1978).

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