

Oxygen isotope evidence regarding the formation of spinel-bearing chondrules

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Received 2 November 1998; revised version received 9 March 1999; accepted 11 March 1999

Abstract

Oxygen isotope compositions of individual mineral grains in two spinel-bearing chondrules have been determined by secondary ion mass spectrometry. O isotope ratios of spinel grains in a plagioclase-rich chondrule were extremely ¹⁶O-rich ($\delta^{18}\text{O} \sim \delta^{17}\text{O} \sim -35\text{‰}$) indicating close relationship to spinels in Ca–Al-rich inclusions (CAIs), whereas O isotope ratios of a spinel grain in barred-olivine chondrule were less enriched in ¹⁶O ($\delta^{18}\text{O} \sim \delta^{17}\text{O} \sim -10\text{‰}$). The degree of ¹⁶O-excesses among minerals within each chondrule was parallel to the crystallization sequences. These measurements suggest that some CAI-spinel grains were incorporated into the chondrule precursors, and that the O isotopic variation among minerals may result in incomplete exchange of O isotopes between ¹⁶O-rich chondrule melt and ¹⁶O-poor ambient nebular gas. The mesostasis having an O-isotopic composition near the terrestrial fractionation line may have been somewhat affected by the aqueous alteration in the parent body. © 1999 Elsevier Science B.V. All rights reserved.

Keywords: meteorites; chondrules; calcium-aluminium inclusions; spinel group; oxygen; isotopes; secondary ion mass spectroscopy

1. Introduction

Chondrule- and CAI-forming events are widely believed to have occurred in the solar nebula although relations among them are poorly understood [1]. CAIs have larger ¹⁶O-excesses and larger quantities of short-lived nuclides such as ²⁶Al than chondrules [2,3]. This evidence favors the view that chondrules and CAIs might be formed by different processes, at times [2] and/or places far removed from one another [1,4].

In CV chondrites, the oxygen isotope ratios of chondrules are quite different from those of CAIs; the ¹⁶O-rich component is extremely enriched in CAIs, whereas less enriched in chondrules [3]. One of the ¹⁶O-rich end-members among CAI minerals is spinel which shows ~4% enrichment of ¹⁶O relative to the standard mean ocean water (SMOW). Therefore, O isotope compositions offer good tracers for discussing relationships between chondrules and CAIs. Especially, O isotopes of rare of spinel grains in chondrules may provide key evidence [2,5,6]. However, it is unclear whether CAI-like spinels observed in chondrules are genetically related to CAI-spinels. We made in-situ O isotope measurements of individual mineral grains within two spinel-bearing

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chondrules (A1-2b-1 and AL95-2-1-CDL.1) from the Allende CV carbonaceous chondrite.

2. Experimental procedure

Back-scattered electron imaging and quantitative analyses were performed on the JEOL JSM-5400 scanning electron microscope (SEM) equipped with the Oxford LINK energy dispersive system (EDS). In each mineral phase, the concentrations of major elements were determined using the EDS. The quantitative analyses were carried out at an accelerating voltage of 20 kV with 0.3 nA probe current with counting times of 50 s. Conventional ZAF methods were used to convert X-ray intensity to concentration.

Oxygen isotopic measurements were performed with the TiTech Cameca ims 1270 SIMS instrument using a ~ 5 μm diameter Cs^+ beam and an electron flood gun to compensate for electrostatic charging on Au-coated sample surface. The mass resolution power was set to ~ 6000 to resolve ^{17}O from the interference of ^{16}OH . Measurements were made by magnetic peak jumping through the following mass sequences; the tail of $^{16}\text{O}^-$, $^{16}\text{O}^-$, $^{17}\text{O}^-$, $^{16}\text{OH}^-$, and $^{18}\text{O}^-$. Measurements of the tail of ^{16}O and ^{16}OH were performed for correction of the ^{16}OH contribution on ^{17}O peaks. This correction is always less than 0.5%. Secondary ion signals were detected with an electron multiplier. Data were corrected for deadtime (21.2 ± 0.8 ns) and the instrumental mass fractionation by utilizing a terrestrial spinel standard [7]:

$$\delta^{17 \text{ or } 18}\text{O}_{\text{SMOW}} = \left(\frac{{}^{17 \text{ or } 18}\text{O}/{}^{16}\text{O}_{\text{sample}}}{{}^{17 \text{ or } 18}\text{O}/{}^{16}\text{O}_{\text{SMOW}}} - 1 \right) \times 1000 [\text{‰}]$$

where SMOW indicates standard mean ocean water value.

The critical parameter to be determined from the O isotope measurements is the magnitude of deviation of the data points from a reference line, taken as the fractionation line for the terrestrial materials. The terrestrial fractionation (TF) line is defined as: $\delta^{17}\text{O} = 0.52\delta^{18}\text{O}$. For chondrules, an excess in ^{17}O can be defined as follows: $\Delta^{17}\text{O} = \delta^{17}\text{O} - 0.52\delta^{18}\text{O}$. The magnitude of the ^{16}O -excess of chondrule minerals can be simply shown by the $\Delta^{17}\text{O}$ values.

3. Results and discussion

The spinel-bearing chondrule, A1-2b-1 (abbreviated to A1 hereafter), is a low-FeO type-I barred-olivine chondrule (BOC) surrounded a porphyritic olivine rim (Fig. 1). A coarse, subhedral spinel grain is embedded in the A1. The composition of the core of the spinel grain is the almost pure MgAl_2O_4 , whereas the rim is rich in FeO (~ 7.4 wt%, 4–60 μm width) and Cr_2O_3 (~ 3.2 wt%, ~ 30 μm width). These chemical compositions and petrography of the A1 resemble those of another Allende chondrule, the R-11 BOC [5], except that the spinel in A1 is not associated with platinum-group metal nuggets but with Ni-rich Fe metal nuggets.

The another spinel-bearing chondrule, AL95-2-1-CDL.1 (abbreviated to CDL hereafter), can be classified as an independent (members of a set having significantly different textures and/or compositions [8]) and a consorting (one subequal to the other in size and shape [8]) compound chondrule (Fig. 2). The chondrule consists of two (left and right) spheroidal chondrules. Several spinel grains of pure MgAl_2O_4 have been observed in the right-hand chondrule of Fig. 2, which can be categorized as a plagioclase-rich chondrule, or a plagioclase–olivine inclusion (POI) of [6]. Olivine and spinel grains in the plagioclase-rich chondrule are poikilitically enclosed by an anorthite-rich assemblage. The spinel grains exhibit a corroded or serrated surface. According to its texture and the chemical composition determined by a broad beam technique of electron microprobe, this plagioclase-rich chondrule may be categorized in the group-2 of [6]. However, in crystallization experiments, the group-2 melt does not coexist with spinel in equilibrium [6]. These features suggest that the spinel grains observed are a relict phase. No spinel grains have been observed in the left-hand chondrule which consists of a porphyritic olivine chondrule (POC) core enveloped by porphyritic pyroxene chondrule (PPC).

The O isotopic compositions are shown in Fig. 3, and the oxygen isotopic data of A1 and CDL are listed in Table 1 and Table 2, respectively. In the spinel grain in A1 the O isotopic composition enriched in ^{16}O ($\Delta^{17}\text{O} \approx -8\text{‰}$), close to the coexisting barred-olivine ($\Delta^{17}\text{O} \approx -6\text{‰}$). Because the spinel and the olivine grains have similar O isotopic

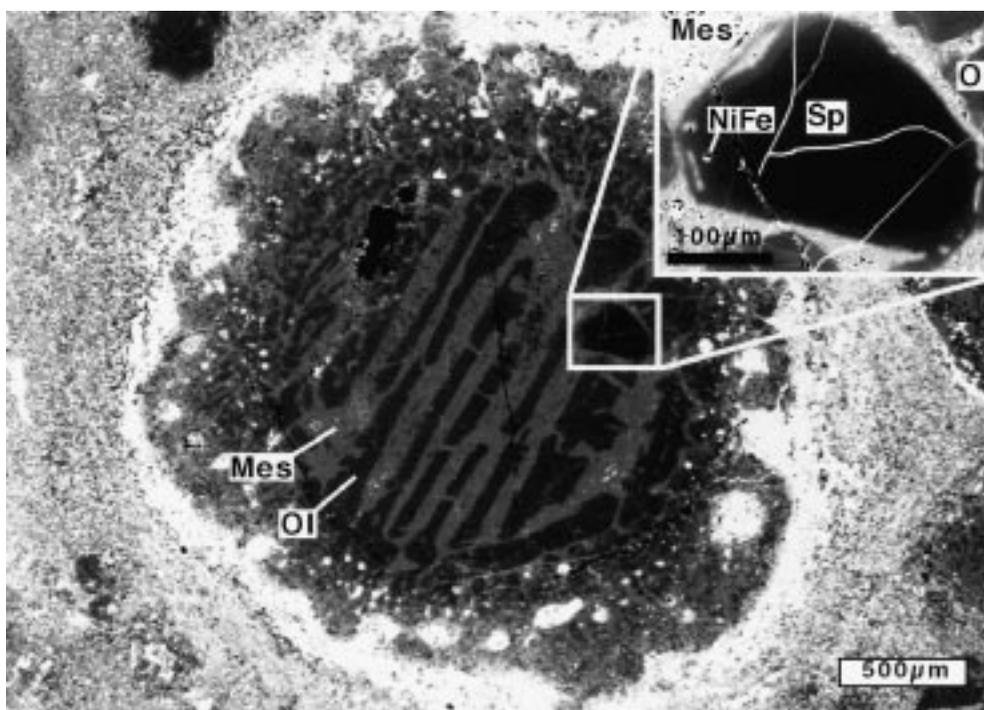


Fig. 1. Back-scattered electron (BSE) image of A1-2b-1 barred-olivine chondrule in the Allende chondrite. This chondrule mainly consists of olivine, mesostasis, spherules of Fe–Ni metal or sulfide. The core part (~ 1.7 mm diameter) is surrounded by the porphyritic olivine rim (200–500 μm width). Olivine bars in the core part are forsteritic (F_{98–100}). The mesostasis is dominated by a Ca-feldspathic phase; the fassaitic phase and Ca–Fe-rich silicate assemblages are surrounded by the feldspathic phase. A coarse subhedral spinel grain is embedded in the core part. *Sp*: spinel, *Ol*: olivine, *Mes*: mesostasis, *NiFe*: Ni–Fe metal nugget.

compositions, we conclude that the spinel grain was a primary phase during the chondrule crystallization; its subhedral texture of spinel supports the in-situ crystallization. Similar ^{16}O -excesses have been observed in individual olivine grains [9,10] and in olivine grains of a POC [11] from the Allende meteorite. This evidence suggests that the spinel grain in A1 is not related to CAI spinels in spite of the similarity of chemical composition.

The spinel grains in CDL showed extreme O isotopic composition enriched in ^{16}O ($\Delta^{17}\text{O} \approx -17\text{‰}$), close to the CAI spinel value ($\approx -20\text{‰}$). Such a large ^{16}O -excess has not previously been observed in chondrules. Similarities of the ^{16}O -excess and of the chemical composition demonstrate that the origin of this spinel grain is linked to those of the spinel grains in CAIs.

The O isotopic compositions of olivine, pyroxenes, anorthite and nepheline in the plagioclase-rich chondrule are less enriched in ^{16}O than those of

spinel grains. All data plot along the carbonaceous chondrite anhydrous minerals (CCAM) line. The range in O isotopic compositions in this plagioclase-rich chondrule is greater than observed in past analyses of individual olivine grains [9,10] and chondrules [3,11]. The observed disequilibrium in O isotopes indicates that the spinel grains are relict phases which have survived the chondrule-forming event [6], and suggest that the spinel grains were related to the CAI forming event.

Laboratory studies of the group-2 melt show that olivine is the first phase to crystallize, followed by pyroxenes and anorthite [6]. Nepheline exists as matrix surrounding other phases or partially replacements of anorthite grains. From the textural observation, nepheline is apparently the secondary phase as a result of the alteration of anorthite. Similar occurrence of nepheline in Allende chondrules has been observed by [12]. The crystallization sequence is parallel to the degree of ^{16}O -enrichment we observed

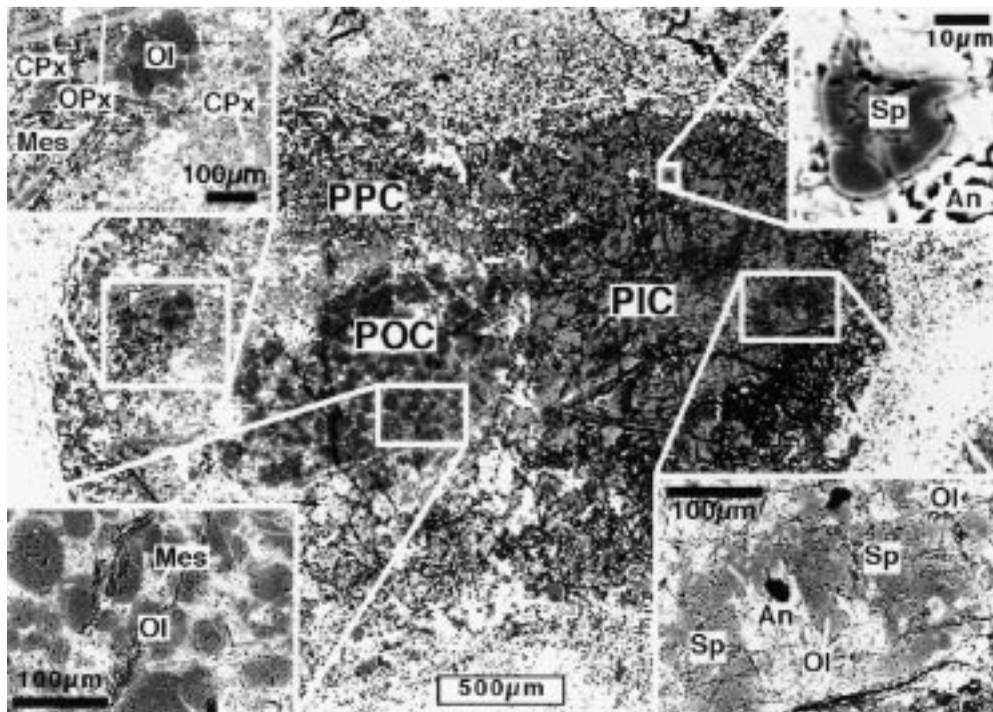


Fig. 2. Back-scattered electron (BSE) image of AL95-2-1-CDL.1 compound chondrule from the Allende chondrite. The porphyritic olivine chondrule (*POC*) mainly consists of porphyritic olivine grains (Fo_{98}) and interstitial mesostasis. The enveloping porphyritic pyroxene chondrule (*PPC*) consists of abundant Ca-rich pyroxene lath and porphyritic Ca-poor pyroxene. Olivine (Fo_{85}) is minor phase. The interstitial mesostasis bears Na and Cl. The plagioclase-rich chondrule (*PIC*) has a sub-ophitic texture and consists of Ca-rich pyroxene (small Ca-poor pyroxene cores exist), anorthite (An_{88-100}), olivine (Fo_{90-98}), spinel and nepheline. *Sp*: spinel, *An*: anorthite, *OI*: olivine, *OPx*: Ca-poor pyroxene, *CPx*: Ca-rich pyroxene, *Mes*: mesostasis.

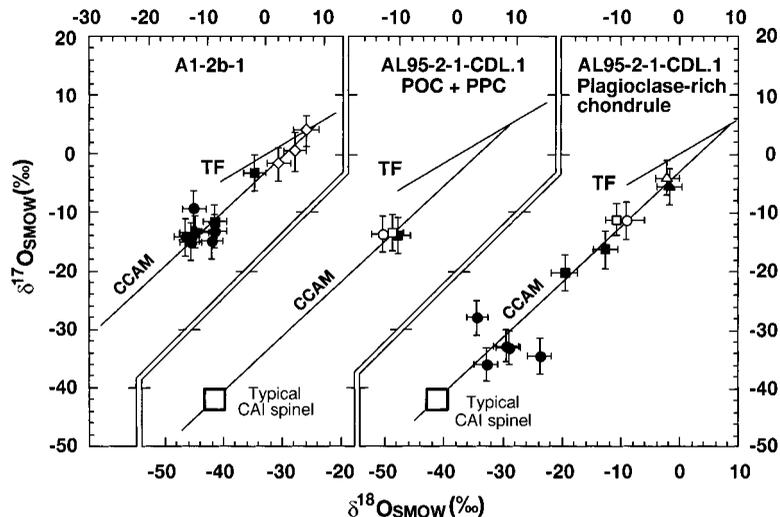


Fig. 3. Oxygen isotopic compositions of each phase in the AL95-2-1-CDL.1 and the A1-2b-1 chondrules. All errors are $1\sigma_{\text{mean}}$. Typical CAI spinel value [3] is also plotted. *TF*: terrestrial fractionation line [3], *CCAM*: carbonaceous chondrite anhydrous minerals line [3]. Solid circle: spinel, solid square: olivine, open circle: Ca-rich pyroxene, open square: Ca-poor pyroxene, solid triangle: anorthite, open triangle: nepheline, open diamond: mesostasis.

Table 1
Oxygen isotopic data for Allende barred-olivine chondrule A1-2b-1

Analysis	$\delta^{17}\text{O}_{\text{SMOW}}$ (‰)	σ_{mean}	$\delta^{18}\text{O}_{\text{SMOW}}$ (‰)	σ_{mean}	$\Delta^{17}\text{O}$ (‰)
Spinel					
SpC01	-13.2	2.8	-8.3	2.1	-8.9
SpC02	-14.2	3.2	-13.2	2.0	-7.3
SpC03	-15.0	3.1	-12.3	1.9	-8.6
SpR01	-9.4	3.0	-11.7	2.0	-3.3
SpR05	-14.9	3.0	-8.8	2.0	-10.3
Average ^a	-13.3	1.0	-10.8	1.0	-7.7
Olivine					
O106(FO ₁₀₀) ^b	-3.2	3.2	-1.3	1.8	-2.5
O110(FO ₉₉)	-11.5	2.9	-8.1	2.1	-7.3
O111(FO ₁₀₀)	-13.3	2.7	-11.6	2.5	-7.3
Average ^a	-9.4	3.1	-7.0	3.0	-5.7
Mesostasis					
Meso01	-1.7	2.8	2.8	2.0	-3.2
Meso02	0.4	3.3	5.6	1.9	-2.5
Meso06	4.0	2.6	7.5	2.0	0.1
Average ^a	0.9	1.7	5.3	1.3	-1.9

Standard errors (σ_{mean}) were calculated from standard deviation of a measurement cycle and the cycle numbers.

^a Average of each mineral and the standard error of the mean were calculated from the values of each spot.

^b Forsteritic content.

among minerals of the plagioclase-rich chondrule, i.e., olivine is the most ¹⁶O-enriched phase ($\Delta^{17}\text{O} \approx -10\text{‰}$) among the silicate phases, whereas anorthite or nepheline is the least ¹⁶O-enriched phase ($\Delta^{17}\text{O} \approx -5\text{‰}$).

The similar relationship of O isotope ratio among minerals is also observed in A1, i.e., minerals which crystallized early, such as spinel, are more enriched in ¹⁶O than mesostasis. The $\Delta^{17}\text{O}$ values of olivine grains in A1 varies from $\sim -7\text{‰}$ (the spinel value) to $\sim -3\text{‰}$ (near the mesostasis value). The O isotopic compositions of mesostasis in A1 show the values near the TF line. The values of mesostasis may not preserve the O isotope ratio of chondrule melt at solidification, but result from O isotopic exchange during aqueous alteration in the parent body because the Allende meteorite shows evidence of the aqueous alteration [13,14].

The evidence among primary minerals in the chondrules implies that the chondrule melt droplets were originally enriched in ¹⁶O, and the degree of

Table 2
Oxygen isotopic data for compound chondrule AL95-2-1-CDL.1

Analysis	$\delta^{17}\text{O}_{\text{SMOW}}$ (‰)	σ_{mean}	$\delta^{18}\text{O}_{\text{SMOW}}$ (‰)	σ_{mean}	$\Delta^{17}\text{O}$ (‰)
AL95-2-1-CDL.1 (Plagioclase-rich chondrule)					
<i>Spinel</i>					
Sp01	-33.0	2.9	-28.8	2.0	-18.1
Sp02	-35.8	2.9	-32.5	2.0	-18.8
Sp03	-32.6	2.7	-29.1	2.1	-17.5
Sp04	-34.3	3.1	-23.6	2.0	-22.0
Sp05	-27.9	3.0	-34.1	1.8	-10.1
Average ^a	-32.7	1.3	-29.6	1.8	-17.3
<i>Olivine</i>					
O101(FO ₉₇) ^b	-20.2	3.1	-19.3	2.3	-10.2
O102(FO ₉₆)	-16.3	3.2	-12.4	2.2	-9.8
Average ^a	-18.2	2.0	-15.9	3.5	-10.0
<i>Ca-rich pyroxene</i>					
ssCPx04	-11.3	3.2	-8.8	2.0	-6.7
<i>Ca-poor pyroxene</i>					
ssOPx01	-11.2	2.7	-10.4	1.9	-5.8
<i>Anorthite</i>					
An04	-5.6	3.0	-1.5	2.1	-4.8
<i>Nepheline</i>					
Ne04	-4.0	3.0	-2.0	2.0	-3.0
AL95-2-1-CDL.1 (POC-PPC)					
<i>Olivine</i>					
psO101(FO ₉₈) ^b	-14.2	3.1	-10.8	2.1	-8.6
<i>Ca-rich pyroxene</i>					
psCPx01	-14.0	3.0	-13.2	2.1	-7.1
<i>Ca-poor pyroxene</i>					
psOPx01	-13.7	3.0	-11.6	1.9	-7.7

Standard errors (σ_{mean}) were calculated from standard deviation of a measurement cycle and the cycle numbers.

^a Average of each mineral and the standard error of the mean were calculated from the values of each spot.

^b Forsteritic content.

enrichment gradually decreased during the solidification, although O isotopic composition of mesostasis may have been affected by aqueous alteration in the parent body. The most negative $\Delta^{17}\text{O}$ value of chondrule initial melt that may be reached is -20‰ according to a recent discovery of extreme ¹⁶O-rich olivine around a CAI rim [15] and in amoeboid olivine aggregates in a carbonaceous chondrite [16].

Oxygen isotopic exchange reaction between chondrule melt and the surrounding nebular gas,

in which O isotopic composition was less enriched in ^{16}O [3], is one mechanism that has been proposed to change the O isotope ratio of chondrules. This argument is also consistent with the observations between O isotope ratio of bulk chondrules and the petrographic and chemical features [17,18]. Since chondrules may have been formed by pulse heating events and the cooling rate of chondrule melt was in the range of 10–2000°C/hr [19,20], it is necessary to evaluate the degree of the O isotopic exchange of molten chondrule within a period of hours. As rate-limiting processes for the exchange reaction, O diffusion rate in the chondrule melt and surface reaction rate of O isotopic exchange should be considered. If the principal rate-limiting mechanism was diffusion in the melt, homogenization time scale of O isotope in a molten chondrule, typically mm-size, can be roughly calculated using O self diffusion [21,22]. The homogenization time scale by diffusion is estimated to the range of several minutes to several hours even at peak temperatures of flash-melted chondrules (1500–1900°C) [20,23]. Therefore, complete equilibrium between chondrule melt and the ambient gas is not obvious during chondrule cooling time scale even if the surface reaction rate was enough fast. Results of oxygen isotope exchange experiments between chondrule composition melt and ambient gas support this argument [24]. Thus, incomplete O isotopic exchange by nebular gas under rate-limiting processes such as diffusion in melt and/or surface reaction is plausible to explain the variations of O isotope ratios observed among the minerals within chondrules. The O isotope ratios observed in chondrules in this study distribute along the CCAM line defined by CAI minerals, indicating that ^{16}O -rich dust and ^{16}O -poor gas continued together in the solar nebula from the CAI-forming period until the chondrule-forming period.

4. Conclusion

The discovery of spinel grains having the O isotopic composition of both of CAI- and chondrule-related spinel grains in individual chondrules in the same carbonaceous chondrite indicates that the similarity of chemical composition of major elements is not always sufficient evidence to infer the origin of

spinel grains. The existence of the ^{16}O -rich spinel provides direct evidence that CAI minerals incorporated into precursor materials of chondrules and some of them survived chondrule-forming events. The variations of O isotope ratios among silicate minerals in chondrules may indicate that O isotope exchange reaction has occurred between chondrule melt and the ambient nebular gas, although aqueous alteration in the parent body may have disturbed the O isotopic composition of mesostasis.

Acknowledgements

We thank G.L. Kim for technical assistance with the SIMS analyses. We thank J.T. Wasson and an anonymous reviewer for constructive comments and for English correction. Supported by Kagaku-gijutsu-cho and Monbu-sho grants. [CL]

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