OXYGEN ISOTOPE STUDY OF TSUKUBA CHONDRITE, SOME HED METEORITES AND ALLENDE CHONDRULES

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Abstract: Laser fluorination technique coupled with stable isotope mass spectrometry using O₂ is successfully employed to determine the δ^{17} O and δ^{18} O values of different types of meteorites. The oxygen isotope data are reported for the recent fall Tsukuba meteorite confirming it to be an H-type ordinary chondrite. Three diogenites Tatahouine, Y-791199 and Y-791000 are analyzed for oxygen isotopic signatures. Y-791199 and Y-791000 are regarded as cumulate eucrites by many researchers on the basis of petrogenesis. The oxygen isotopes of Tatahouine and Y-791199 are consistent with the diogenite group data. Y-791000 is showing lighter oxygen isotopic composition which is unique among the other diogenites. It may indicate that the parental source for some of cumulate eucrites is different from the non-cumulate ones. The slope 1/2 trend of oxygen isotopes observed in various Allende chondrules (mostly small porphyritic type) signifies that aqueous alteration might be responsible for scattering of data along a nearly mass dependant trend. These chondrules are plotted on a $\delta^{17}O$ - $\delta^{18}O$ diagram with a slope of 0.52 having $\Delta^{17}O = -4.24 \pm 0.27$. The range of $\delta^{18}O$ and $\delta^{17}O$ data observed in these chondrules is from -7.26 to -0.19‰ and -7.98 to -4.04‰ respectively. The variability in the isotopic composition which caused the scatter on the terrestrial trend implies that on the parent body some low temperature hydrothermal alteration took place.

1. Introduction

Oxygen isotopes play a unique role in determining the precursor materials, formation processes and genetic relationships among different types of meteorites. Oxygen isotope abundances in the solar system show a particular trend with respect to each parent body. It is one of the unique elements having three stable isotopes ¹⁶O, ¹⁷O and ¹⁸O which can undergo both mass dependent and mass independent fractionation processes (CLAYTON, 1993; THIEMENS and HEIDENREICH, 1983; THIEMENS, 1996). Oxygen isotope analysis can put constraints on conditions of formation and the various alteration processes associated with the parent bodies.

The laser fluorination technique coupled with stable isotope mass spectrometry provides a rapid and precise means of determining oxygen isotope ratios in silicate minerals. It is appropriate on account of low blanks and quantitative oxygen yields for the

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analysis of very small samples and/or refractory minerals (SHARP, 1990; MATTEY *et al.*, 1994). In the case of meteorites this technique is far better for its need of a small sample size and ease in getting complete reactions through the irradiation of almost all types of refractory minerals. In the present paper, we report δ^{17} O and δ^{18} O values of Tsukuba, Densmore and Gladstone (H6 chondrites), Y-791000, Y-791199 and Tatahouine (diogenites), silicate phase of Esquel (pallasite) and 15 chondrules and 2 CAI fractions from the carbonaceous chondrite Allende CV3. The data plot on the distinct areas in the δ^{17} O- δ^{18} O diagram for each meteorite or chondrule group. The parent body processes are discussed for each meteorite group, based on the measured oxygen isotopic compositions.

2. Experimental

2.1. Sample treatment

In the case of ordinary chondrites and diogenites whole rock chips were used for the analysis. Care was taken to remove any weathered parts. The olivine grains in the Esquel pallasite embedded in metal matrix were separated and the surface layer was removed by gentle polishing with diamond paste. The olivine was analyzed as single grain. Various types of chondrules were separated from 1.5 g of Allende CV3 carbonaceous chondrite using an edged tool. One side of the chondrule was well polished for quantitative analysis by electron microprobe. An acetone-soluble adhesive used to fix the chondrules in the polishing process was removed repeatedly in boiled acetone to perform oxygen isotope analysis of the same chondrule. Observations and quantitative analysis of the interior of the chondrules Ch-1 to Ch-5 and Ch-15 were performed with an electron microprobe analyzer (JEOL 733 super probe) using wavelength dispersive X-ray spectrometers (WDS) and the ZAF correction method. These were classified on the basis of scheme given by GOODING and KEIL (1981). The chondrules Ch-6 to Ch-14 were not identified by EPMA to avoid the weight loss for oxygen isotope analysis. WDS analysis was done at 15 kV acceleration voltage and 10 nA beam current. Natural and synthetic standards with compositions similar to minerals in the chondrules were used for EPMA calibration.

2.2. Oxygen isotope analysis

Oxygen isotope determination was performed with a mass spectrometer (modified VG-SIRA12) connected on-line to the 12W CO₂ laser extraction system. The calibration of the system was done on the basis of VSMOW-SLAP scaling (JABEEN and KUSAKABE, 1997). For this purpose international and laboratory reference water samples VSMOW, SLAP, GISP and MSA6 were decomposed with BrF₅ and the released oxygen (O₂) was measured against the working standard (pure O₂ gas; O₂ > 99.999%) to determine the δ^{17} O and δ^{18} O values. The conditions of the laser assembly (beam diameter, beam power, irradiation duration and holder size etc.) were optimized to extract oxygen from the sample.

0.5-2 mg of the samples (predried at 110° C) were loaded in a Ni holder with 9 holes of 1 mm diameter and 1.5 mm depth. The sample chamber was pumped under high

vacuum overnight with continuous heating at 105°C. The sample assembly was pretreated with 100 mbar BrF₅ for three hours to remove any adsorbed water. Each sample was irradiated with CO₂ laser under 300 mbar BrF₅ atmosphere while changing the laser beam diameter and power from defocused to focused and minimum to maximum, respectively. A single reaction was complete within 5 min. No sample scattering was observed. Oxygen yields were typically between 95–100%. The liberated oxygen was cryogenically purified by trapping in a molecular sieve 5A cold finger, and introduced directly into the micro-inlet system of the mass spectrometer for the determination of δ^{17} O and δ^{18} O values. The present data are reported on the VSMOW-SLAP normalized scale for ¹⁸O/¹⁶O and ¹⁷O/¹⁶O ratios. The analytical accuracy is about ±0.1‰ for δ^{17} O, δ^{18} O and Δ^{17} O values.

3. Results and Discussion

3.1. Terrestrial fractionation line (TF)

As the meteorites contain different types of minerals, a number of samples with different mineralogy and rock types were analyzed as standards before the unknown sample analysis. The ranges of δ^{17} O and δ^{18} O values are from -2.63 to 15.14 and -5.23



Fig. 1. Terrestrial fractionation line (filled triangles) with a slope of 0.505. Data with open circles are from CLAYTON and MAYEDA (1983).

| Sample | Weight | $\delta^{18}O$ | δ17Ο | σ_1^{18} | σ_1^{17} | ∆ ¹⁷ O | n |
|-----------------|------------|---------------------|--------|-----------------|-----------------|-------------------|---|
| (Type) | mg | %0 | %0 | %0 | %0 | | |
| RG-35 | 2.12-2.48 | 26.83 | 13.55 | 0.11 | 0.05 | 0.00 | 2 |
| (Quartzite) | | '(27.0) | | | | | |
| Col.12 | 2.03-3.55 | -5.23 | -2.63 | 0.49 | 0.39 | 0.01 | 2 |
| (Granodiorite) | | ² (-5.6) | | | | | |
| No. 16 | 1.66-1.71 | 9.03 | 4.61 | 0.03 | 0.08 | 0.05 | 2 |
| (Shale) | | ³ (9.36) | | | | | |
| No. 19 | 1.47-1.91 | 0.02 | 0.04 | 0.03 | 0.00 | 0.03 | 2 |
| (Dacite Tuff) | | ³ (0.04) | | | | | |
| No. 44 | 2.15-2.90 | 3.93 | 2.00 | 0.11 | 0.01 | 0.02 | 2 |
| (Andesite tuff) | | ³ (4.60) | | | | | |
| NBS-28 | 1.25 -2.85 | ⁴9.31 | 4.86 | 0.08 | 0.09 | 0.16 | 7 |
| (Quartz) | | ⁵ (9.23) | (4.80) | | | | |
| | | ⁶ (9.58) | | | | | |
| Olivine | 0.91-2.21 | 4.70 | 2.44 | 0.18 | 0.16 | 0.07 | 5 |
| (San Carlos) | | 7(4.88) | | | | | |
| Spinel | 1.78-2.22 | 24.09 | 12.21 | 0.05 | 0.03 | 0.04 | 4 |
| (Burma) | | ⁸ (22.3) | (11.6) | | | | |
| | | ⁹ (22.3) | (10.1) | | | | |
| Corrundum#1 | 1.55-1.60 | 23.71 | 12.02 | 0.24 | 0.02 | 0.05 | 2 |
| (Burma) | | | | | | | |
| Corrundum#3 | 1.89-3.43 | 18.32 | 9.24 | 0.05 | 0.08 | -0.01 | 2 |
| (Burma) | | | | | | | |

Table 1. Oxygen isotope data of various terrestrial samples.

Reference values: ¹Matsuhisa *et al.*, 1982, ²Matsuhisa, 1978, ³M. Noto (p.c.), ⁴ Jabeen and Kusakabe, 1997, ⁵Clayton and Mayeda, 1983, ⁶Gonfiantini *et al.*, 1995, ⁷Mattey *et al.*, 1994, ⁸Simon *et al.*, 1994, ⁹Weinbruch *et al.*, 1993.

 $\Delta^{17}O = \delta^{17}O - 0.505\delta^{18}O$. *n* means number of analysis. σ_1^{18} and σ_1^{17} are standard deviations (±1 σ) of the $\delta^{18}O$ and $\delta^{17}O$ values respectively.

*Standard deviation $1\sigma = \sqrt{\frac{\sum (x_i - \bar{x})^2}{n}}$.

to 30.8‰ respectively (details will be published elsewhere). The terrestrial fractionation line based on these data is shown in Fig. 1. All the data lie excellently on a single trend with a slope of 0.505 and R=0.99975. The details of some of these data, for which some reference values were known, is given in Table 1. The data are in good agreement with the reported oxygen isotope data from different laboratories. The sample Col-12 is an hydrothermally altered granodiorite with a remarkable δ^{18} O depletion so that it could be isotopically heterogeneous (MATSUHISA, 1978). In case of Burma spinel the data plots on the heavier side as compared to literature data (SIMON *et al.*, 1994; WEINBRUCH *et al.*, 1993). It attribute to the isotopic heterogeneity as Burma spinel used in this study was a mixture of three fractions–spinel, corrundum#1 and corrundum#3 (H. HIYAGON, pers. commun.). Two fractions spinel and corrundum#1 are very similar in oxygen isotopic compositions while corrundum#3 is about 5‰ lighter in oxygen isotopes.

| Meteorite | Weight | $\delta^{_{18}}\mathrm{O}$ | $\delta^{17}O$ | $\Delta^{17}O$ | O ₂ Yield | n |
|--------------|---------------|----------------------------|----------------|----------------|----------------------------|---|
| | mg | %0 | $\%_{0}$ | | μ mol.mg ⁻¹ | |
| Ordinary Cho | ndrite (H6 Ty | (pes) | | | | |
| Tsukuba* | 1.33 | 4.65 | 2.96 | 0.61 | 11.85 | 2 |
| | 2.35 | 4.79 | 2.94 | 0.52 | 8.96 | |
| Average | | 4.72 | 2.95 | 0.57 | 10.41 | |
| Densmore | n.d. | 3.88 | 2.84 | 0.88 | n.d. | 4 |
| | | 3.98 | 3.00 | 0.99 | | |
| | | 3.90 | 2.82 | 0.85 | | |
| | | 4.01 | 2.86 | 0.83 | | |
| Average | | 3.94 | 2.88 | 0.89 | | |
| Gladstone | n.d. | 3.75 | 2.45 | 0.56 | n.d. | 2 |
| | | 3.84 | 2.58 | 0.64 | | |
| Average | | 3.80 | 2.52 | 0.60 | | |

Table 2. Oxygen isotope data of ordinary chondrites.

 $\Delta^{17}O = \delta^{17}O - 0.505 \ \delta^{18}O$ is departure of a data point from Terrestrial Fractionation line (TF). 'n' stands for individual number of analysis. n.d. means not determined. *Described in YONEDA *et al.* (1996).



Fig. 2. The three isotope plot for Tsukuba, Densmore and Gladstone (filled triangles) plotted within data by CLAYTON et al. (1991) (open legends and crosses). TF means Terrestrial Fractionation line.

3.2. Ordinary chondrites (H6 type)

The oxygen isotope data of Densmore, Gladstone and the recent fall Tsukuba (No.1(a) in OKUYAMA-KUSUNOSE *et al.*, 1996, No. 1 in YONEDA *et al.*, 1996) are reported in Table 2. The three isotope plot (δ^{17} O- δ^{18} O) shows the data points for these meteorites in Fig. 2. The variations in δ^{18} O and δ^{17} O values are 0.92 and 0.43‰ respectively. The Tsukuba chondrite shows the highest δ^{18} O value among the H6 type ordinary chondrites. The Δ^{17} O values for Tsukuba and Gladstone are very similar and the oxygen isotope data falls towards the lower end of the H-chondrite group (see Fig. 2 and Table 2 for details). The oxygen isotope data from Tsukuba and Gladstone also fall close to the array defined by IIE irons silicates (Fig. 2). CLAYTON and MAYEDA (1978, 1996) suggested the genetic relationship between H chondrites and 0.59±0.07 for II-E). Our results on Tsukuba (Δ^{17} O=0.57±0.06) and Gladstone (Δ^{17} O=0.60±0.06) meteorites may represent the example of their genetic link.

3.3. Achondrites

HED meteorites are derived from the parent body which underwent global melting, isotopic homogenization and subsequent differentiation. On the basis of REE patterns

| Meteorite | Weight | δ ¹⁸ Ο | $\delta^{17}O$ | $\Delta^{17}O$ | O ₂ Yield | n |
|------------|--------|-------------------|----------------|----------------|----------------------------|---|
| | mg | %0 | %0 | | μ mol.mg ⁻¹ | |
| Diogenites | | | | | | |
| Y-791000 | 1.94 | 1.91 | 0.79 | -0.17 | 13.18 | 3 |
| | 2.19 | 2.42 | 1.03 | -0.19 | 13.16 | |
| | 1.31 | 2.34 | 0.98 | -0.20 | 13.54 | |
| Average | | 2.22 | 0.93 | -0.19 | 13.29 | |
| Y-791199 | 2.42 | 3.52 | 1.51 | -0.27 | 13.75 | 2 |
| | 2.76 | 3.56 | 1.63 | -0.17 | 13.60 | |
| Average | | 3.54 | 1.57 | -0.22 | 13.68 | |
| Ref. | | 3.56 | 1.49 | -0.22 | | |
| Tatahouine | 4.26 | 3.58 | 1.45 | -0.36 | 13.34 | 3 |
| | 2.97 | 3.19 | 1.50 | -0.11 | 13.27 | |
| | 2.09 | 3.38 | 1.52 | -0.19 | 13.32 | |
| Average | | 3.38 | 1.49 | -0.22 | 13.31 | |
| Ref. | | 3.41 | 1.42 | -0.34 | | |
| Pallasite | | | | | | |
| Esquel | 2.42 | 2.95 | 1.34 | -0.15 | 12.82 | 2 |
| ас. | 2.62 | 3.00 | 1.46 | -0.06 | 13.05 | |
| Average | | 2.98 | 1.40 | -0.10 | 12.94 | |
| Ref. | | 2.82 | 1.22 | -0.24 | | |
| | | | | | | |

Table 3. Oxygen isotope data of HED meteorites.

Reference: CLAYTON and MAYEDA (1996). Δ^{17} O values of CLAYTON and MAYEDA (1996) were recalculated using Δ^{17} O= δ^{17} O -0.505 δ^{18} O. Other details are same as in Table 2.



Fig. 3. $\delta^{17}O-\delta^{18}O$ plot for individual runs of Y-791199, Tatahouine, Y-791000 and silicate phase of Esquel pallasite (filled legends). The data with open legends are from CLAYTON and MAYEDA (1996).

(CONSOLMAGNO and DRAKE, 1977) and spectroscopic studies (BINZEL and XU, 1993) have shown that the asteroid Vesta is the candidate of HED parent body. The small isotopic variations in oxygen isotopes of these meteorites is also consistent with their derivation from a single parent body (CLAYTON and MAYEDA, 1996). However, there is a debate regarding the parental source for cumulate and noncumulate eucrites (Consolmagno and DRAKE, 1977; DELANEY and PRINZ, 1984; STOLPER, 1977; MA and SCHMIDT, 1979; HSU and CROZAZ, 1997). The oxygen isotope data for the individual analysis of three diogenites and Esquel pallasite is given in Table 3 and presented in Fig 3. Y-791199 and Tatahouine fall within the range of oxygen isotopic compositions observed for the diogenites (CLAYTON and MAYEDA, 1996). Y-791000 plot outside of this range and fall among the range of chromites from the IIIAB irons (Fig. 3). This achondrite is described as a special diogenite by TAKEDA and MORI (1985). The oxygen yield is not very different for the three diogenites which implies that they possess a very similar chemical composition (Table 3). Y-791199 and Y-791000 are both cumulate eucrites but the oxygen isotopic compositions differ by more than 1%. Such an oxygen isotope data of cumulates can be described by two possible ways: (1) the fractionation of plagioclase at relatively low temperature, and/or, (2) not formed in a same fractional crystallization sequence as the non cumulate ones. On the basis of bulk rock analysis, such processes cannot be explained very clearly. Many authors, such as MA and SCHMIDT (1979) and HSU and CROZEZ (1997), have regarded the cumulate eucrites to belong to a different source as noncumulate eucrites. On the basis of our data of oxygen isotopes, Y-791199 and Y-791000 as cumulate eucrites, may belong to a parental source which might be different than the other HED meteorites.

The isotopic composition of Esquel pallasite is consistent with the δ^{17} O and δ^{18} O data range determined for the pallasite group (CLAYTON and MAYEDA, 1996; see Table 3). Therefore, our data support the idea that the pallasites belong to the same parent body from which the HED meteorites came to our Earth.

3.4. Allende CV3

It is very interesting to note that the δ^{17} O and δ^{18} O values of porphyritic type small chondrules from Allende CV3 lie on three isotope plot with a slope of 1/2 parallel to, but below the terrestrial line (Fig. 4). This is the first time that a nearly single mass fractionation trend was seen among bulk oxygen isotopic compositions of porphyritic chondrules. The plot of δ^{18} O and δ^{17} O values for such chondrules shows a span of about 7% and 4‰, respectively. This data covers the range from -0.19 to -7.26‰ for δ^{18} O and from -7.98 to -4.04 ‰ for δ^{17} O values with Δ^{17} O=-4.24±0.27. CI and CM chondrites,



Fig. 4. Oxygen isotope data of Allende CV3 small chondrules- porphyritic olivines (PO), porphyritic olivine pyroxenes (POP) and glassy chondrules (GC) falling with slope 1/2 on three isotope plot (filled circles). The barred olivine (BO) falls off this trend (filled triangle). Calcium Aluminium rich inclusions (CAI) make a separate group below the chondrules (filled diamonds). The data (open legends) are taken from CLAYTON et al. (1983). DI and ESP mean dark inclusions and Eagle Station Pallasites, respectively. CCAM stands for Carbonaceous Chondrites Anhydrous Mineral line.

such as Orguel and Murchison, contain hydrous silicates which might form at low temperature and caused the oxygen isotopic mass fractionation (CLAYTON and MAYEDA, 1984). The recent petrologic studies also clearly demonstrate that the Allende parent body had undergone aqueous alteration followed by thermal metamorphism (BREARLEY, 1997; Колма and Томеока, 1996; Buchanan et al., 1997). Occurrence of phyllosilicates such as serpentinite, magnetite, talc, biopyriboles and Fe rich olivines in such chondrules is good evidence for the above mentioned processes. In case of Murchison, a δ^{18} O range of about 40% is reported, implying that the temperature of alteration was between 0°C and 25°C, based on the serpentine-calcite oxygen isotopic fractionation. For Allende chondrules in this study, data plot along a mass fractionation line, which indicates that similar low temperature processes took place on the Allende body. As the range of data is not so wide compared to that in Murchison, the temperature of alteration must be higher on the Allende parent body. On the basis of this and previous studies (CLAYTON et al., 1983; SAXTON et al., 1996; CHOI et al., 1997), it is evident that nearly all the data reported so far for Allende chondrules plot to the left side of the CCAM line. It may explain that if aqueous alteration is the cause for such a large span in delta values, then the initial water reservoir must be lighter in oxygen isotopes. The oxygen isotope data from the bulk analysis of small porphyritic chondrules are plotted within the data of magnetite and



Fig. 5. Oxygen isotope data of bulk chondrules (filled circles) from Allende CV3 compared with data of magnetite (open triangles) and olivines (open circles) from CHOI et al. (1997) and magnesian olivines (shaded area) from SAXTON et al. (1996).

olivine grains in these chondrules given by CHOI et al. (1997) (Fig. 5). These magnetite grains showing a nearly mass dependent trend for oxygen isotopes is inferred to be formed on the parent body by the oxidation of metal with water while the olivine data plot near the CCAM line and is thus not in isotopic equilibrium with the magnetite. In case of our data, the deviation in Δ^{17} O values (-4.24±0.27) is somewhat greater than the analytical error, which explain that some oxygen isotopic disequilibrium is there in these chondrules. However, the data lies with a slope of 0.52 on the three isotope plot which indicate that the secondary processes like aqueous alteration outweighs the primary signatures in these small chondrules. Such a variation in Δ^{17} O values can arise from the presence of both the primary and alteration signatures. As the detailed EPMA study was not performed, it is difficult to explain exactly what minerals are responsible for such alteration signatures. Some data regarding very magnesian olivines from Allende by SAXTON et al. (1996), also fall among our data (Fig. 5). In the case of SIMS (CHOI et al., 1997; SAXTON et al., 1996) the data errors are much larger, so that we cannot get a well defined trend for oxygen isotopes. At this point, the present study is most accurate so far in reporting the oxygen isotopes where the errors plot just inside the data legends. SAXTON et al. (1996), have explained that Fo-rich olivines from Allende (CC) and Julesburg (UOC) belong to a common population. In such a case we should think of a mixing behavior between chondrule line extending down from ordinary chondrites and the CCAM line.



Fig. 6. Oxygen isotope plot for Allende chondrules (filled circles) and ordinary chondrites (open circles) showing the mixing behavior with CCAM. OC stands for ordinary chondrite line.

| Chondrule | Weight mg | δ ¹⁸ Ο ‰ | δ ¹⁷ Ο %ο | $\Delta^{17}O$ | O ₂ Yield µmol.mg ⁻¹ |
|------------|--------------|------------------------|-------------------------|----------------|---|
| Ch-1*(PO) | 0.94 | -3.38 | -5.91 | -4.20 | 13.3 |
| Ch-2*(PO) | 1.60 | -2.40 | -5.63 | -4.42 | 14.1 |
| Ch-3*(PO) | 0.51 | -4.74 | -6.34 | -3.95 | 12.0 |
| Ch-4*(POP) | 0.61 | -3.08 | -6.09 | -4.53 | 11.8 |
| Ch-5*(GC) | 1.37 | -0.86 | -4.04 | -3.61 | 11.4 |
| Ch-6 | 0.69 | -2.27 | -5.42 | -4.27 | 11.7 |
| Ch-7 | 1.54 | -2.63 | -5.65 | -4.32 | 12.0 |
| Ch-8 | 1.99 | -5.61 | -6.78 | -3.95 | 13.4 |
| Ch-9 | 2.35 | -0.19 | -4.41 | -4.31 | 13.9 |
| Ch-10 | 0.42 | -1.38 | -4.73 | -4.03 | 10.7 |
| Ch-11 | 1.79 | -7.26 | -7.98 | -4.31 | 11.7 |
| Ch-12 | 0.99 | -2.24 | -5.59 | -4.46 | 13.2 |
| Ch-13 | 2.58 | -4.50 | -6.85 | -4.58 | 12.0 |
| Ch-14 | 1.02 | -1.74 | -5.28 | -4.40 | 12.5 |
| Average | | -3.02 | -5.76 | -4.24 | |
| σ_1 | | 1.94 | 1.03 | 0.27 | |
| Ch-15*(BO) | 1.18 | 2.57 | -1.68 | -2.98 | 12.6 |
| CAI-1 | 1.60 | -13.34 | -16.09 | -9.35 | 12.2 |
| CAI-2 | 0.81 | -11.75 | -14.08 | -8.15 | 10.6 |

Table 4. Oxygen isotope data of chondrules from Allende CV3 chondrite.

Abbreviations: PO- Porphyritic Olivine, POP- Porphyritic Olivine Pyroxene, BO- Barred Olivine, GC-Glassy chondrules, Ch-6 to Ch-14- Not Identified, CAI-1 & 2- Calcium Aluminium rich Inclusions. *Classified according to scheme by GOODING and KEIL (1981). Other details are same as in Table 2.

To constrain this, the ordinary chondrule line was extrapolated towards the ¹⁶O rich side (see Fig. 6). The Allende small chondrule data lies in between the OC line and CCAM line. However, there can be some points against such behavior; firstly, with how much confidence we can extend ordinary chondrite line to ¹⁶O rich side, secondly, there must be two ¹⁶O initial reservoirs, and lastly the end members responsible for such mixing from both sides are not known, because of the bulk data. In any case, the nearly slope 1/2 line shown by these chondrules is more difficult to explain on the basis of such a mixing model.

In favor of aqueous activity is the behavior of Na as can be seen in Table 5. The amount of Na₂O ranges from 2.20 to 17.46 wt% within these chondrules. Many workers have reported enrichment of elements such as, Na, K, Au, As, Sb, Br, Ba and Hg in Allende dark inclusions (PALME *et al.*, 1989; KOJIMA and TOMEOKA, 1996; BUCHANAN *et al.*, 1997). It shows that mesostasis within small chondrules is much affected by alteration processes.

The isotopic compositions of two fine-grained CAI fractions, barred olivine chondrule and porphyritic chondrules are shown on the oxygen isotope plot (Fig. 4). The data of the barred olivine chondrule is falling off the trend shown by porphyritic type chon-

| Oxides | Ch-1 | Ch-2 | Ch-3 | Ch-4 | Ch-5 | Ch-15 |
|--------------------------------|-------|-------|-------|-------|-------|-------|
| SiO ₂ | 41.57 | 41.25 | 41.24 | 41.45 | n.d. | 38.61 |
| TiO ₂ | 0.82 | 0.20 | 0.72 | 0.16 | n.d. | 0.16 |
| Al_2O_3 | 17.91 | 21.30 | 17.84 | 32.54 | n.d. | 23.71 |
| FeO | 0.95 | 0.93 | 0.25 | 0.65 | n.d. | 0.88 |
| MnO | 0.11 | 0.08 | 0.09 | 0.03 | n.d. | 0.03 |
| MgO | 4.75 | 3.98 | 5.27 | 2.80 | n.d. | 2.08 |
| CaO | 29.03 | 28.96 | 29.64 | 4.31 | n.d. | 23.71 |
| Na ₂ O | 2.93 | 2.79 | 2.20 | 17.46 | n.d. | 6.46 |
| K ₂ O | 0.00 | 0.00 | 0.01 | 0.02 | n.d. | 0.68 |
| Cr ₂ O ₃ | 0.46 | 0.46 | 0.34 | 0.04 | n.d. | 0.03 |
| NiO | 0.01 | 0.01 | 0.04 | 0.01 | n.d. | 0.00 |
| SO ₃ | 0.07 | 0.12 | 0.00 | 0.03 | n.d. | 0.01 |
| Total | 98.6 | 100.1 | 97.6 | 99.5 | | 96.4 |
| Fa | 0.5-5 | 0.5-3 | 0.5 | 0.5-5 | 0.5-7 | n.d. |
| | | | | | | |

 Table 5.
 Chemical composition of mesostatis glass and Fa

 content of olivine in Allende chondrules.

Ch-1 to Ch-5, Ch-15 stands for the same chondrules as in Table 4. n.d. means not determined.

drules. At this moment we have no good explanation for this type of chondrules because of only one analysis. A slope of ~0.9 is obtained by plotting CAI with different chondrule types, porphyritic and barred, but we infer that a regression should not be made for CAI and chondrules together. Both of these might have formed by different processes and in different environments, they may preserve different isotopic signatures. Moreover, they represent different regions on the three isotope plot (Fig. 4). The age data have shown that the chondrules were formed several million years after the formation of CAIs (SWINDLE *et al.*, 1996; SCOTT *et al.*, 1996). More work is a need to have a better understanding.

4. Conclusions

The oxygen isotope studies of different meteorite groups present a comprehensive view of the secondary processes on the parent bodies. The oxygen isotopic compositions of ordinary chondrites (Densmore, Gladstone and Tsukuba) support them to be H-type chondrites. In case of achondrites, the extraordinary behavior of one diogenite (Y-791000), regarded as a cumulate eucrite, implies that the parental source for this cumulate is different from the other HED diogenites. The data from small type Allende porphyritic chondrules defining a slope 1/2 on oxygen isotope plot may indicate that aqueous alteration is responsible for such behavior. There may have been redistribution of oxygen isotopes through isotopic exchange reactions between constituent minerals, which caused the isotopic compositions to move along the terrestrial trend. The bulk analysis of such small chondrules explains that alteration processes have added much to their isotopic behaviour which is preserved within after accretion. The clear constraints can only be made by analysis of large type chondrules and CAIs with well defined mineralogy.

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