

COMPOSITIONS OF REE, K, Rb, Sr, Ba, Mg, Ca, Fe, AND Sr ISOTOPES IN ANTARCTIC “UNIQUE” METEORITES

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Abstract: The abundances of Mg, Ca, Fe, K, Rb, Sr, Ba, and REE in five Antarctic “unique” meteorites, acapulcoite-lodranite type, Yamato (Y)-74063, Y-74357, Y-8002, and Allan Hills (ALH)-78230, and winonaite type, Y-75300 were determined by isotope dilution mass spectrometry. The Sr isotopic compositions of Y-74063, Y-75300, Y-8002, and ALH-78230 were also measured to investigate the whole-rock Rb-Sr isotopic systematics.

In terms of the REE abundance pattern, these meteorites are grouped as follows; (1) Y-74063 and ALH-78230 with chondritic REE patterns, (2) Y-74357 with light REE depletion plus alkali and alkaline earth element depletion, and (3) Y-75300 and Y-8002 with middle REE depletion (V-shaped REE pattern) plus a positive Eu anomaly). The mineralogy of these meteorites is closely related to the above REE classification. The ⁸⁷Rb–⁸⁷Sr age determined from ‘whole-rock samples’ for Y-74063, Y-75300, Y-8002, and ALH-78230 is consistent with their formation of 4.5 Ga, albeit with a large uncertainty due to the blank correction.

Using the mineral/liquid partition coefficients of trace elements, the petrogenetic model calculations were performed. The results give indication on the origin of these “unique” meteorites. Y-74063 and ALH-78230 could be formed through a small degree of partial melting (less than a few%) from a chondritic starting material. Y-74357 could be formed by a larger degree (12%) of partial melting. On the other hand, the V-shaped REE pattern of the third group (Y-75300 and Y-8002) could not be explained by a simple partial melting process. The V-shaped REE pattern may be explained only by assuming the solid state equilibration within a reservoir with chondritic composition. It is suggested that Y-75300 and Y-8002 might be derived from parental materials depleted in phosphate and clinopyroxene.

1. Introduction

Most of chondritic meteorites have been classified into nine chemical groups based on chemical composition and oxidizing state (see a review by SEARS and DODD, 1988).

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There are numbers of unclassified chondritic meteorites which show similar chemical and petrological characteristics to each other. The Mg/(Mg + Fe) ratios in olivines and pyroxenes in these meteorites are between those in enstatite chondrites and H chondrites. It means that they were formed under the redox condition between those for enstatite and H chondrites. The petrological texture is highly recrystallized, *i.e.*, granoblastic texture, so that some of them were called type 7 chondrites (YANAI and KOJIMA, 1991). They were also called primitive achondrites (PRINZ *et al.*, 1983), since some of these meteorites show the mineral compositions slightly fractionated from typical chondritic composition. Nevertheless they are quite different from any other known achondrites. Oxygen isotopic compositions of these meteorites are also different from other types of meteorites (CLAYTON *et al.*, 1984; MAYEDA *et al.*, 1987; MAYEDA and CLAYTON, 1989). Because of such uniqueness, now we call them "unique" meteorites.

Several non-Antarctic "unique" meteorites were previously reported; Acapulco, Winona, Mt. Morris (Wisconsin), Pontlyfni (BILD and WASSON, 1976; BILD, 1977; GRAHAM *et al.*, 1977; PALME *et al.*, 1981). The silicate inclusions in IAB irons and Lodran are also classified into the same group (PRINZ *et al.*, 1983). On the basis of the petrology and oxygen isotopic composition, they were further classified into two groups (MAYEDA and CLAYTON, 1989; NAGAHARA *et al.*, 1990), and named acapulcoite and winonaite.

This study has an aim to investigate the genesis of the "unique" meteorites on the basis of the abundances of lithophile elements and ^{87}Rb - ^{87}Sr systematics. The REE abundances are especially important to constrain the melting processes in the parent body. Previously, both unfractionated and fractionated REE patterns were reported in several "unique" meteorites (BILD, 1977; FUKUOKA *et al.*, 1978; FUKUOKA and SCHMITT, 1978; PRINZ *et al.*, 1980; PALME *et al.*, 1981; SCHULTZ *et al.*, 1982; KALLEMEYN and WASSON, 1985). One of the interesting features of REE fractionation in these meteorites may be depletion of the middle REE (hereafter called V-shaped REE pattern) with a positive Eu anomaly. The origin of this pattern has not been well explained yet.

In this paper, as part of the consortium study for the Antarctic "unique" meteorites, the results of chemical analyses of five Antarctic "unique" meteorites are presented, and petrogenetic implications are further discussed.

2. Experimental Procedures

2.1. Sample preparation

Y-74063

A fragment (Y-74063,54) weighing 1.509 g was supplied to us for the purpose of the ^{87}Rb - ^{86}Sr systematics. It was broken into several pieces, and a major part of the sample was set aside for detailed Rb-Sr study. For the whole-rock analysis, about 120 mg piece was taken and ground into powder using an agate mortar. An aliquot of powder weighing 38.6 mg was used for the analyses.

Y-75300 and Y-8002

Since the original mass of the meteorites is less than a few grams, only 10 mg-sized samples (Y-75300,53 and Y-8002,53) consisting of numbers of small fragments were available for the present study. They were ground into powder and used for the analyses.

Because of such small sample sizes, it is questionable to assume that the results represent “whole-rock analyses” of the meteorites. KIMURA *et al.* (1991) reported that Y-75300 consists of coarse- and fine-grained lithologies with different mineral proportions. The sample used in this study was obtained from the coarse-grained portion (K. YANAI, private communication).

Y-74357 and ALH-78230

The samples (Y-74357,72 and ALH-78230,56) were supplied to us through Dr. T. FUKUOKA, after INAA analyses of major and trace elements (FUKUOKA and KIMURA, 1990). Each of them consists of several small chips, of which total weight was about 100 mg. Two-thirds of each sample was taken and ground into powder, and an aliquot of 30–40 mg of the powder was used for the analyses.

2.2. Chemical procedures and mass spectrometry

Abundances of major and trace elements (Mg, Fe, Ca, K, Rb, Sr, Ba, and REE) were determined by isotopic dilution mass spectrometry. The procedure was the same as that described in YAMAMOTO and NAKAMURA (1990). In this work, except for Y-74357, isotopic analysis of Sr was performed for the same sample solution used for isotopic dilution analysis. For the samples weighing more than 30 mg, about a half of sample solution was used for the abundance determination, and the other half was used for the Sr isotopic analysis. For the samples less than 10 mg, the abundances of REE and the isotopic compositions of Sr were obtained from the same split of the solution through cation exchange column chemistry. The blank correction was applied to each sample according to that used in YAMAMOTO and NAKAMURA (1990). Except for K and Rb, the blank levels are less than 1 % of the analytical results. Even though it is as large as 30 % of the analyzed amounts of Rb in some of the samples, the general feature of the abundance pattern does not change significantly by the blank correction. However, such a large blank correction seriously affects the Rb-Sr systematics, as will be described in the next section.

The chemical separation of Sr from major elements was carried out by using cation exchange resin (AG 50W-12X, 200–400 mesh) contained in a small quartz column. The Sr isotopic compositions were measured by either VG 354 or MAT 261 single collector thermal ionization mass spectrometers. The samples were loaded on a W-filament with $\text{H}_3\text{PO}_4\text{-Ta}_2\text{O}_5$ activator. The precision of $^{87}\text{Sr}/^{86}\text{Sr}$ ratios was considered to be better than 0.01 % for the samples containing more than 30 ng Sr.

3. Results and Discussion

3.1. Chemistry

3.1.1. General features

The results of chemical analyses are shown in Table 1. The CI-normalized abundances of trace elements are shown in Figs. 1a, b, and c. It is pointed out that the meteorites are more or less chemically fractionated from chondritic abundances, both among alkali metals and among REE.

It is remarkable that the Rb abundances of all the “unique” meteorites analyzed are much less than CI chondritic abundance; as low as 0.1 times CI. The depletion of

Table 1. Elemental abundances in Antarctic "unique" meteorites.

Sample	Y-74063	Y-75300	Y-8002	Y-74357	ALH-78230
Classification*	Acapulcoite	Winonaite	Acapulcoite	Acapulcoite	Acapulcoite
Weight dissolved (mg)	38.59	9.56	8.24	38.59	33.77
Mg (%)	15.1	10.4	8.4	18.5	13.4
Fe (%)	20.0	22.4	3.5	27.9	24.7
Ca (%)	1.43	0.439	0.90	0.91	1.48
K (ppm)	580	180	330	35	540
Rb (ppm)	0.24	0.28	0.19	0.11	0.38
Sr (ppm)	10.9	6.5	15	0.9	9.6
Ba (ppm)	3	n.a. [†]	n.a.	n.a.	2
La (ppm)	0.397	0.137	0.313	0.0666	0.274
Ce (ppm)	1.01	0.207	0.645	0.201	0.738
Nd (ppm)	0.780	0.0708	0.410	0.194	0.591
Sm (ppm)	0.259	0.0187	0.153	0.0766	0.206
Eu (ppm)	0.0940	0.0569	0.130	0.0141	0.0808
Gd (ppm)	0.357	0.0361	0.284	0.135	0.307
Dy (ppm)	0.451	0.0986	0.468	0.188	0.389
Er (ppm)	0.298	0.112	0.371	0.133	0.264
Yb (ppm)	0.303	0.147	0.418	0.139	0.272
Lu (ppm)	0.0457	0.0232	0.0653	0.0236	0.0411

* Classified according to NAGAHARA *et al.* (1990).

[†] "n.a." denotes not analyzed.

Rb in a similar range was also found from one non-Antarctic "unique" meteorite, Acapulco (PALME *et al.*, 1981), which is an "observed fall". Therefore, the Rb depletion observed here may not be due to the weathering in Antarctic ice, but a common feature for "unique" meteorites, which may indicate some specific petrogenetic meaning.

The REE abundances of these meteorites are variable; 0.1–3 times CI. The CI-normalized REE patterns for these meteorites are divided into three groups; (1) nearly flat REE patterns for Y-74063 and ALH-78230, (2) depletion of light REE with a negative Eu anomaly for Y-74357, (3) V-shaped REE patterns with a large positive Eu anomaly for Y-75300 and Y-8002.

3.1.2. The least fractionated REE pattern: Y-74063 and ALH-78230

The CI-normalized abundances of trace elements in Y-74063 and ALH-78230 are shown in Fig. 1a. Mostly, the abundances of the elements are from 1 to 2 times those in CI, which are similar to the range of H chondrites. Therefore, these meteorites are chemically less fractionated from the chondritic composition. An exception is the Rb abundances; as low as 0.1 times that in CI chondrite, though the abundances of K are chondritic. The abundances of Na in the same meteorites obtained by FUKUOKA and KIMURA (1990) are also chondritic. Hence, only Rb is significantly depleted among alkali metals. The depletion of Rb observed here is similar to those observed for Acapulco (PALME *et al.*, 1981) and ALH-77081 (SCHULTZ *et al.*, 1982), of which other lithophile elements are also less fractionated.

The absolute abundances of heavy REE are correlated with Mg abundance. For

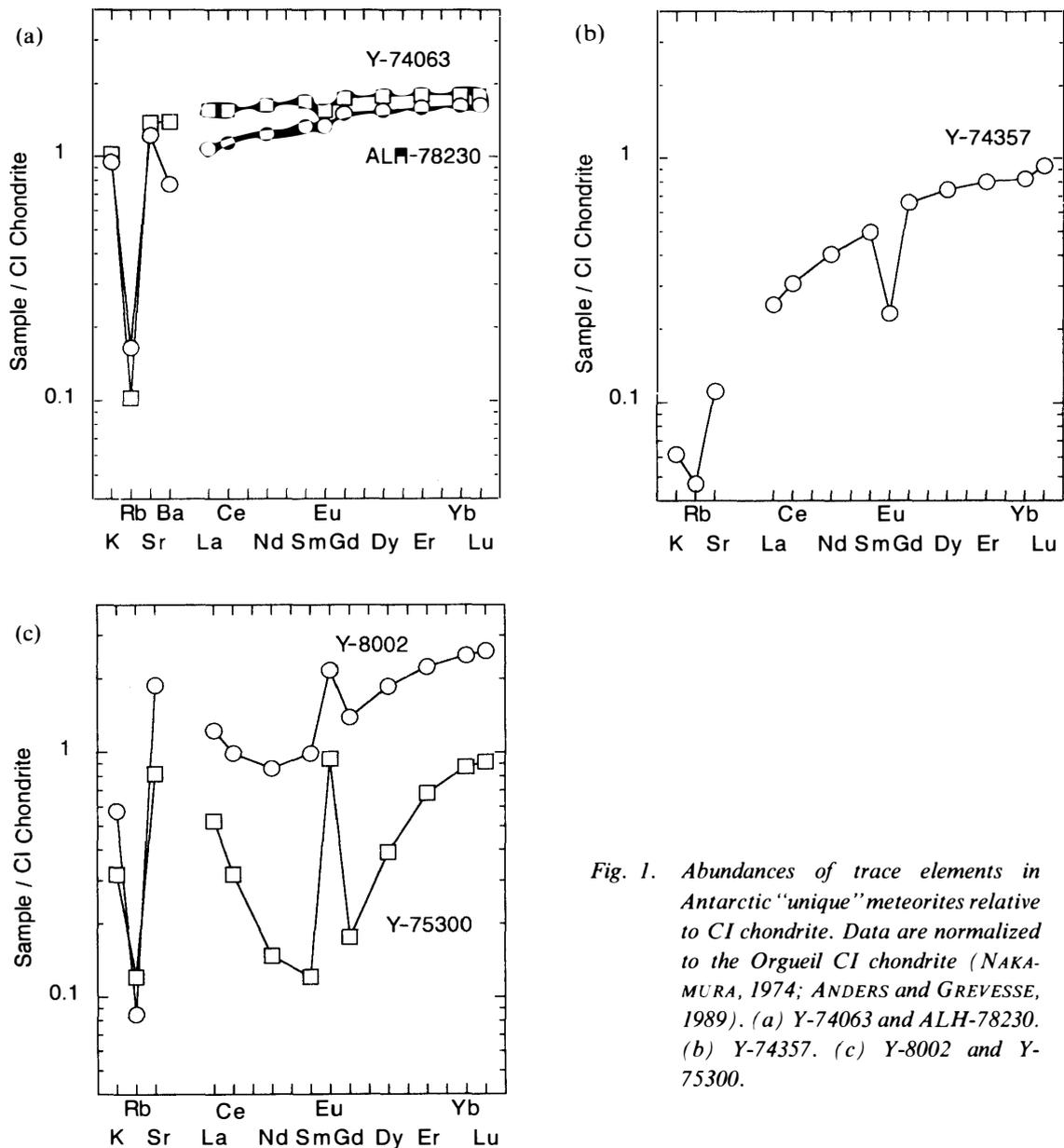


Fig. 1. Abundances of trace elements in Antarctic "unique" meteorites relative to CI chondrite. Data are normalized to the Orgueil CI chondrite (NAKAMURA, 1974; ANDERS and GREVESSE, 1989). (a) Y-74063 and ALH-78230. (b) Y-74357. (c) Y-8002 and Y-75300.

example, the CI-normalized Lu/Mg ratios are 1.16 and 1.14 for Y-74063 and ALH-78230, respectively. It means that REE abundances in silicate portions of both meteorites are essentially the same. Nevertheless there is a small but systematic depletion of lighter REE; CI-normalized La/Lu abundances are 0.87 and 0.67 for Y-74063 and ALH-78230, respectively. It is known that REE in bulk ordinary chondrites show only a small degree of fractionation (NAKAMURA, 1974; KALLEMEYN *et al.*, 1985). Although there is a possible effect of heterogeneity on the REE abundances in the present samples, the REE fractionation in these meteorites are clearly discriminated from those observed for the ordinary chondrites. Therefore, it is suggested that the light REE depletion in Y-74063 and ALH-78230 are real and may have some petrogenetic implications.

3.1.3. Light REE depletion with a negative Eu anomaly: Y-74357

As shown in Fig. 1b, lithophile trace elements in Y-74357 are depleted relative to CI chondrite. The depletion is significantly larger for alkali metals, alkaline earth elements, and lighter REE, as low as 0.05 times those in CI chondrite. The depletion of Eu is much larger than other REE, and hence a negative Eu anomaly occurs. This pattern is quite similar to that obtained from Lodran (FUKUOKA *et al.*, 1978).

The relative abundance of Eu to Sr in Y-74357 is distinctly larger than others; CI-normalized abundance ratio of Eu/Sr in Y-74357 is 2.0, while those in other four meteorites are 1.1 with variations less than 2%. It is thus possible that the ratio of trivalent to divalent Eu in Y-74357 is larger than others. This appears to be consistent with the fact that the Fs content in orthopyroxene of 13.8 mole% in Y-74357 is larger than those of ≤ 11 mole% in other "unique" meteorites. It is suggested that Y-74357 was formed in a more oxidizing condition than the other "unique" meteorites.

3.1.4. V-shaped REE pattern with a positive Eu anomaly: Y-75300 and Y-8002

The abundances of alkali metals for Y-75300 and Y-8002 are lower than those in CI chondrite (Fig. 1c), but in a similar range of those for Y-74063 and ALH-78230. The CI chondrite-normalized REE patterns clearly show the V-shaped pattern with a positive Eu anomaly. The minimums of the V-shape are at Nd for Y-8002 and at Sm for Y-75300. The V-shape is deeper in Y-75300 than in Y-8002.

The V-shaped pattern with a positive Eu anomaly was observed from several winonaites and silicate inclusions in IAB irons (FUKUOKA and SCHMITT, 1978; KALLEMEYN and WASSON, 1985). It should be pointed out that a similar V-shaped REE pattern has been also found for a chondrule in the equilibrated chondrite, Bjurböle (NAKAMURA *et al.*, 1989), and for the large clasts from metamorphosed L chondrites (NAKAMURA *et al.*, 1984; WARREN and KALLEMEYN, 1987). Including the "unique" meteorites, all these materials are thermally metamorphosed and related to chondritic meteorites. Hence the V-shaped REE pattern may have some relation to the solid state equilibration processes for the chondritic materials.

3.2. The ^{87}Rb - ^{87}Sr systematics

The results of the Rb and Sr analyses are presented in Table 2. The Rb-Sr isochron diagram for "whole-rock samples" of Y-74063, Y-75300, Y-8002, and ALH-78230 is shown in Fig. 2. The uncertainties of the $^{87}\text{Rb}/^{86}\text{Sr}$ ratios are large because of a large blank correction. All the data points plot close to the 4.5 Ga reference isochron which was obtained for a number of the bulk chondrites (MINSTER *et al.*, 1982). Therefore, the present results are consistent with the early formation of these meteorites. Our earlier suggestion for the late disturbance on the Rb-Sr systems of the meteorites (YAMAMOTO *et al.*, 1990, 1991) is found to be misleading due to the blank problem.

3.3. Relation between chemistry and mineralogy

Petrology and mineralogy of these "unique" meteorites have been studied extensively in the consortium study (YANAI and KOJIMA, 1991; HIROI and TAKEDA, 1991; NAGAHARA *et al.*, 1990; KIMURA *et al.*, 1991, 1992; NAGAHARA, 1992). The modal mineralogy of the "unique" meteorites is summarized in Table 3. Because of the large grain size, heterogeneity in modal mineralogy among different thin sections seems to

Table 2. Rb-Sr analytical results for Antarctic "unique" meteorites[†].

Sample	Weight (mg)	⁸⁷ Rb/ ⁸⁶ Sr ^{††}	⁸⁷ Sr/ ⁸⁶ Sr
Y-74063	17.29	0.0624 ± 24	0.703257 ± 10 ^{†††}
Y-75300	5.92	0.124 ± 8	0.706824 ± 40 ^{†††}
Y-8002	3.91	0.038 ± 5	0.701120 ± 54 ^{†††}
ALH-78230	12.02	0.111 ± 3	0.706312 ± 15 [§]
NBS 987 STD	VG 354 (ICRR) MAT 261 (KSU)		0.710124 [#] ± 10 0.710209 [§] ± 21

[†] Errors correspond to the last digits.

^{††} Calculated from Table 1. Errors include those due to blank correction.

^{†††} VG 354 mass spectrometer (Institute for Cosmic-Ray Research, University of Tokyo) was used. The discrimination between two machines was adjusted to MAT 261 mass spectrometer. Errors are $2\sigma_m$.

[§] MAT 261 mass spectrometer (Kyoto Sangyo University) was used. Errors are $2\sigma_m$.

[#] Weighted mean for 19 analyses.

[§] Weighted mean for 2 analyses.

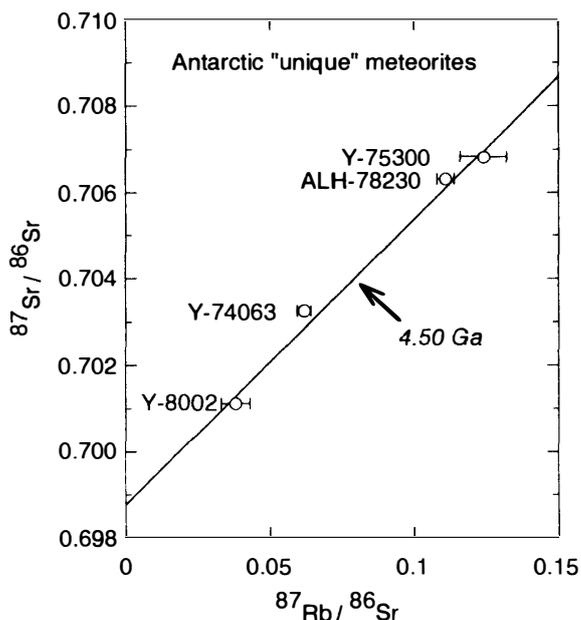


Fig. 2. The whole-rock ⁸⁷Rb-⁸⁷Sr system in Antarctic "unique" meteorites. The solid line indicates a 4.50 billion years reference isochron with the initial Sr isotopic ratio of low-Rb, Ca, Al-rich inclusions from the Allende, meteorite ($(^{87}\text{Sr}/^{86}\text{Sr})_{\text{ALL}} = 0.69877$; GRAY *et al.*, 1973).

be present. Especially, the difference between two thin sections from Y-74357 is significantly large. Therefore, we tried to calculate norm mineral composition of the sample analyzed in this work using the major element data. In the calculation, it is assumed that the chemical compositions of constituent minerals are the same as those obtained by YANAI *et al.* (1984). The result is also shown in Table 3.

It is remarkable that the meteorites in each chemical group, which is based on the abundances of the trace elements, show similar modal mineralogy to each other. The first group with almost flat REE patterns has nearly chondritic mineral proportions, *i.e.*, abundant olivine and orthopyroxene with minor amounts of clinopyroxene, plagioclase, and phosphate. The second group with light REE depletion contains olivine and

Table 3. Mineral proportions in Antarctic "unique" meteorites¹.

Meteorite	REE pattern	Olivine	Opx	Cpx	Plagioclase	Phosphate	Sil + Phos ² bulk	Fa ³	Reference
Y-74063	chondritic	29.0	55.2	7.4	7.0	1.4 ⁴	0.75	11	(1)
ALH-78230	chondritic	32	56	4	8	<1	0.79	10	(2)
Y-74357	LREE depleted	78.7	17.2	4.0	—	0.1		Fs = 14	(3)
		90	7	3	—	—			(2)
		57.1	36.8	6.1	—	—	0.74		This work ⁵
Y-8002	V-shaped	28.0	58.9	2.7	10.4	—	0.95	2	(3)
		36.5	52.5	1.5	9.5	—			(4)
Y-75300	V-shaped	22.1	63.8	—	14.1	—	0.70	4	(1) Coarse
		21.5	69.7	—	8.8	—			(1) Fine
H chondrite		48.8	33.9	5.4	11.1	0.8	0.56	17	(5)

¹ Expressed by weight %, 100%, for total silicates and phosphates. Recalculated from modal proportion of minerals. The densities of minerals used are as follows: olivine, Opx, and Cpx = 3.3 g/cm³, plagioclase = 2.7 g/cm³, phosphate = 3.1 g/cm³, troilite = 4.2 g/cm³, and metal = 8.0 g/cm³.

² The weight ratios of sum of silicates and phosphates to bulk meteorites.

³ Fayalite content in olivine. For Y-74357, ferrosilite content in Opx is shown, since its olivine was reduced and has smaller Fa than equilibrium value (HIROI and TAKEDA, 1991).

⁴ Assuming volume % of phosphate to be 1.2%, twice as large as that in H chondrites, according to the analysis of phosphorus by YANAI and KOJIMA (1991).

⁵ Estimated abundances based on the chemical analyses of Mg, Ca, and Fe, shown in Table 1. See text. References: (1) KIMURA *et al.* (1991), (2) HIROI and TAKEDA (1991), (3) YANAI *et al.* (1984), (4) NAGAHARA (1992), (5) MASON (1965).

pyroxenes, but lacks plagioclase and phosphate. The same holds true for the mineral composition of Lodran; it consists of equal amounts of olivine, pyroxene and metal (BILD and WASSON, 1976). The third group with V-shaped REE pattern is depleted in clinopyroxene and phosphate relatively to chondritic composition. Therefore, it is strongly suggested that the observed trace element features (typically fractionated REE) were produced by the petrogenetic processes which established the modal mineral proportions of the meteorites. Hence, a rough genetic relation may be considered among these meteorites; the first group represents the least fractionated "unique" meteorites, from which the second group was chemically and mineralogically fractionated. The close relation between chemistry and mineralogy indicates that relatively simple petrogenetic processes prevailed during the formation of the meteorites under consideration.

4. Model Calculation

4.1. Principles of the calculation

In order to investigate the origin of the "unique" meteorites quantitatively, model calculations for partition of trace elements are discussed. Two kinds of simple models are considered; one is single stage partial melting model, and another is solid state equilibrium partition model. First we consider the single stage partial melting model. In this model, the "unique" meteorites are assumed to represent residues of partial melting in a chondritic parent body. As a result, trace element abundances in the

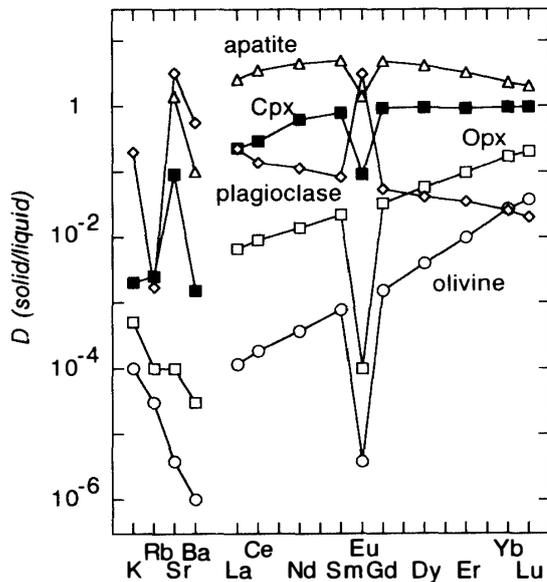


Fig. 3. Partition coefficients between minerals and liquid used in the model calculation. References: Phosphate; Sr and REE from WATSON and GREEN (1981) and 0.1 times that of Sr is assumed for Ba; Clinopyroxene; Sr and REE from KUEHNER *et al.* (1989) and K, Rb and Ba from SHIMIZU (1974). Plagioclase; Sr, Ba, and REE from DRAKE and WEILL (1975), K from MCKAY and WEILL (1977), and Rb from MCKAY and WEILL (1976). Olivine and orthopyroxene; Sr, Ba, and REE from COLSON *et al.* (1988) and for K and Rb extrapolated values according to the data for mono-valent cations from MATSUI *et al.* (1977) were used.

“unique” meteorites of the third group cannot be simply explained by the partial melting process. Then, for the second step, solid state partition model will be introduced. In this model, it is assumed that a material consisting of nonchondritic mineralogy is enclosed by the large chondritic (solid) reservoir, and they are in chemical equilibrium. In both models, chemical composition of the starting material (or reservoir in the latter model) is assumed to be the same as those of metal-sulfide-free portion of the ordinary chondrites; abundances of the lithophile trace elements are 2.5 times CI.

We employ the partition coefficients between minerals and liquid of the five minerals, *i.e.*, olivine, orthopyroxene, clinopyroxene, plagioclase, and phosphate (Fig. 3). It is known that mineral/liquid partition coefficients depend on temperature, pressure, and chemical composition. Experimentally determined partition coefficients are mostly obtained at higher temperatures compared to the solidus temperature of chondritic materials, using the non-chondritic materials. Thus, there may be some limitation of applying the available data to the melting processes of the chondritic materials.

As for the temperature dependence of the partition coefficients, the values corresponding to $T=1150^{\circ}\text{C}$ were used, if available. This temperature is in the range of those indicated by mineral thermometers for the “unique” meteorites (PALME *et al.*, 1981; NAGAHARA, 1992). For the pressure dependence, data under nearly 1 atm were used, since the pressure of the interior of the meteorite parent bodies may be much lower than that in the earth interior. Except for phosphate, we used partition coefficients which were obtained for the mineral composition similar to those in the “unique” meteorites; forsterite for olivine, enstatite for orthopyroxene, diopside for clinopyroxene, and albitic plagioclase. Many of these data are available more recently, especially for REE. Particularly, data for olivine and orthopyroxene (COLSON *et al.*, 1988) are given in the form of the numerical function of temperature, pressure, and Fa or Fs contents.

4.2. Partial melting model

Here we consider the single-stage partial melting model. Concentration of elements

in liquid C_l and solids C_s are obtained by the following equations;

$$C_l = C_o / (f + D(1 - f)), \quad (1)$$

$$C_s = D \cdot C_l, \quad (2)$$

where C_o is the concentration in starting material, f is the degree of partial melting, and D is the solid/liquid partition coefficient. If the weight fraction and partition coefficient of i -th mineral are x_i and D_i , respectively, D is written as

$$D = \sum(x_i \cdot D_i), \quad (3)$$

where the sum is over all the minerals in the solid. Since the "unique" meteorites are assumed to be the residue of the partial melting, x_i is taken to be the mineral proportion of each meteorite shown in Table 3. The results of calculations are described below in detail.

4.2.1. Rb depletion together with almost flat REE pattern

Figure 4 shows the patterns calculated for Y-74063 and ALH-78230. The degree of partial melting is estimated to fit the analyzed data; 1% for Y-74063, and 3% for ALH-78230. In the case of such low degrees of partial melting, only Rb is significantly depleted while the other elements show limited fractionation from chondritic abundances. The partition coefficients of Rb between minerals and liquid are an order of magnitude smaller than those for other elements, so that Rb depletes first from the partially melted chondritic source. Hence the depletion of heavy alkali metals without depletion of other elements may result from such a low degree of partial melting.

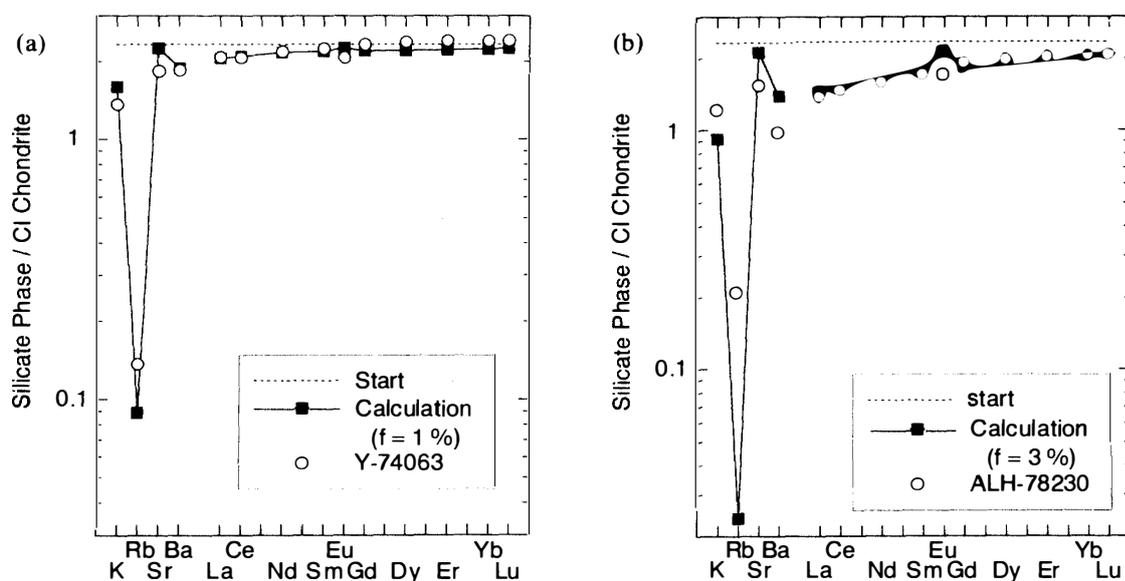


Fig. 4. Partial melting model for the solid with nearly chondritic compositions. (a) The calculated pattern was obtained for the mineral composition of Y-74063. The degree of partial melting is 1%. (b) The calculated pattern was obtained for the mineral composition of ALH-78230. The degree of partial melting is 3%. Abundances in silicate phase are shown relative to CI chondrite. Filled symbols indicate the model partial melting residue, and open symbols indicate the analytical data for the "unique" meteorites. The dashed line indicates the starting material.

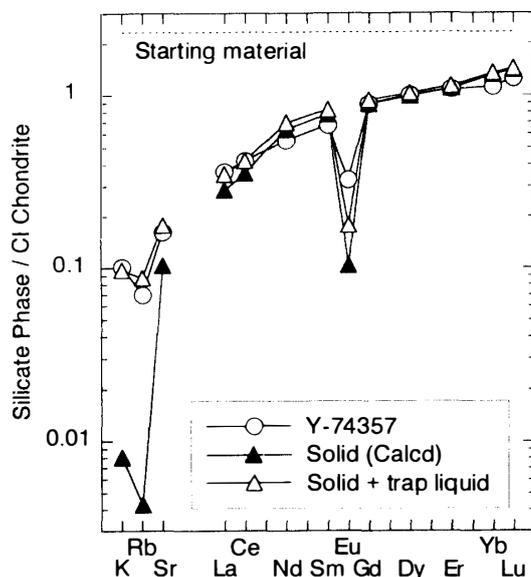


Fig. 5. Partial melting model for the solid with Y-74357 mineral composition. The degree of partial melting is 12%. Filled triangles indicate the model partial melting residue. Open triangles indicate the addition of 0.4% liquid into the residue as a trap liquid.

4.2.2. Light REE depletion

The results of calculation for Y-74357 are shown in Fig. 5. The light REE are depleted because the partition coefficients of light REE in olivine and pyroxenes are low. In view of REE abundances, the result of $f = 12\%$ is most consistent with the analyzed data. This is comparable to the amount of plagioclase in chondritic meteorites ($\sim 10\%$). The melting temperature of plagioclase is lower than other Mg-rich silicates, when the composition of plagioclase is albitic like in other “unique” meteorites. Therefore, the meteorite might have lost plagioclase from the chondritic source by partial melting and segregation of liquid.

The calculation shows extreme depletion of alkali metals. It is an order of magnitude lower than the analyzed data. By adding a small amount of liquid ($< 1\%$) to the solid, however, the higher concentration of alkali metals in the meteorite can easily be explained without affecting REE abundances (Fig. 5). Such a process may be possible when a small amount of liquid was trapped between the solid grains during the segregation of liquid from solid.

4.2.3. V-shaped REE patterns: not produced by partial melting

The V-shaped REE patterns in “unique” meteorites were previously obtained by several authors (BILD, 1977; FUKUOKA and SCHMITT, 1978; PRINZ *et al.*, 1980; KALLEMEYN and WASSON, 1985), and they were explained by partial melting processes. We made a number of model calculations on the partial melting process by applying the various parameters which might have prevailed in the melting processes. However, as typically shown in Fig. 6, our model calculation shows that the observed V-shaped REE patterns could not be produced by any partial melting processes using the typical REE partition coefficients. Similar difficulties were seen in modeling the V-shaped REE pattern of a clast from the Y-75097 (L6) chondrite (WARREN and KALLEMEYN, 1989). In the next section, we will introduce solid state partition model, as more realistic one for solving the problem.

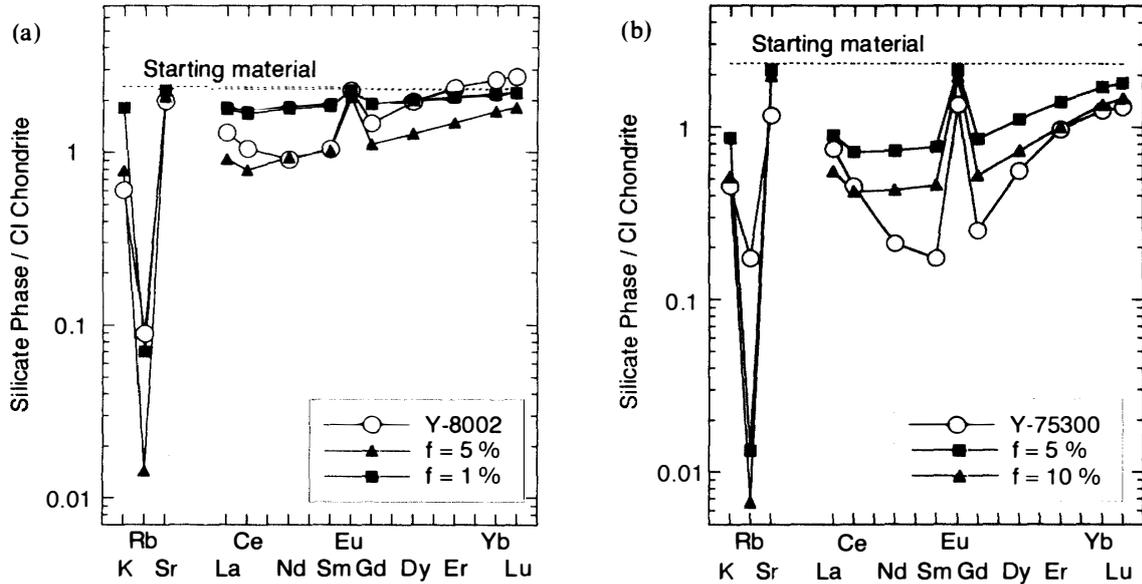


Fig. 6. Partial melting model for the solid with Y-8002 and Y-75300 mineral compositions. (a) The calculated pattern for the mineral composition of Y-8002. (b) The calculated pattern for the mineral composition of Y-75300. Filled symbols indicate the model partial melting residue. Open symbols indicate the analytical data for the "unique" meteorites.

4.3. Solid state partition

4.3.1. Equilibrium concentration in minerals in chondritic reservoir

Before turning to a detail calculation for V-shaped pattern, the concentration in minerals under solid state equilibrium should be considered. The mineral composition of the chondritic reservoir is assumed to be the same as that of Y-74063, the least fractionated unique meteorite (NAGAHARA *et al.*, 1990).

Since the mineral/liquid partition coefficients are indices of the relative compatibility of specific elements with minerals, they may also be applied to the problems of partitioning elements among minerals under the solid state. As discussed by CURTIS and SCHMITT (1979), the distribution of elements in minerals of equilibrated chondrite is closely related to the mineral/liquid partition coefficients; the concentrations of elements in mineral separates of L6 chondrites were proportional to the mineral/liquid partition coefficients. Thus, for each element, we employ the following relation between the concentration in mineral-A, C_A , and the total concentration of the chondritic reservoir, C_o ;

$$C_A/C_o = D_A / \sum(D_i \cdot x_i). \quad (4)$$

In eq. (4), D_A and D_i are the mineral/liquid partition coefficients of an element for mineral-A and mineral- i , respectively, x_i is the weight fraction of mineral- i , and the summation is over the five minerals in the reservoir. Replacing eq. (4) will give the concentration in mineral A;

$$C_A = C_o \cdot D_A / \sum(D_i \cdot x_i). \quad (5)$$

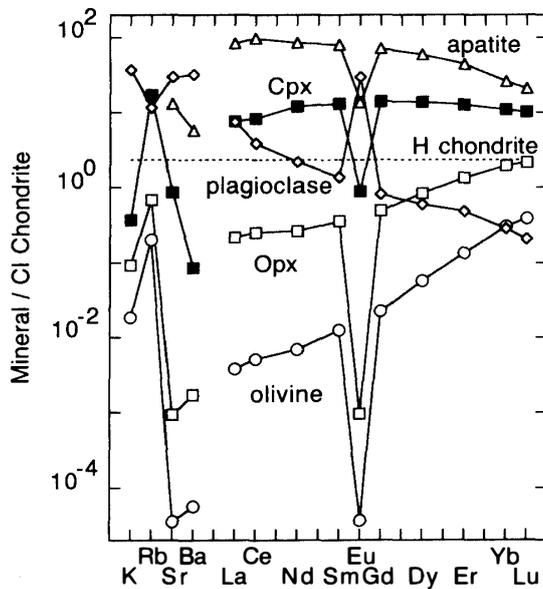


Fig. 7. Equilibrium concentrations of elements in minerals of the unfractionated unique meteorite. The mineral proportion is assumed to be the same as that of Y-74063.

The equilibrium concentrations of five major minerals were obtained by eq. (5) and are shown in Fig. 7.

4.3.2. V-shaped REE pattern

We assumed that Y-75300 and Y-8002 had existed in the chondritic reservoir. When the system reaches chemical equilibrium, the concentrations of trace elements in each mineral are the same as those obtained by eq. (5). Then the equilibrium concentration of an element in the material with the specific mineral assemblage, C , will be

$$C = \sum(C_j \cdot y_j) = C_o \cdot \sum(D_j \cdot y_j) / \sum(D_i \cdot x_i), \quad (6)$$

where y_j is the weight fraction of mineral- j in the meteorites with specific mineral assemblage, Y-75300 and Y-8002 (Table 3). The results are shown in Fig. 8. Now, the V-shaped pattern is clearly seen, though the calculated patterns are less pronounced than the analyzed ones. The absolute REE abundances in Y-8002 are significantly higher than the calculated one. For this discrepancy, we will give a more detail examination later. The REE pattern in Y-75300 is generally consistent with the calculated pattern while the observed pattern is even more fractionated. In spite of many trials, such an extreme fractionation was not obtained by using the present data set of partition coefficients.

The solid state equilibration model could explain the V-shaped REE pattern, but could not explain the depletion of heavy alkali metals. Calculated abundances of alkali metals are similar to or even more enriched than chondritic abundances. It is because these meteorites are enriched in plagioclase component relative to the chondritic composition. It should be pointed out that the degree of alkali element depletion in these meteorites is in a similar range to that in Y-74063, which might be produced by a small degree (less than 1 %) of partial melting. We may recall that a small degree of partial melting does not change too much the elemental abundances of other elements, as discussed in the previous section. Therefore, it is suggested that prior to the equil-

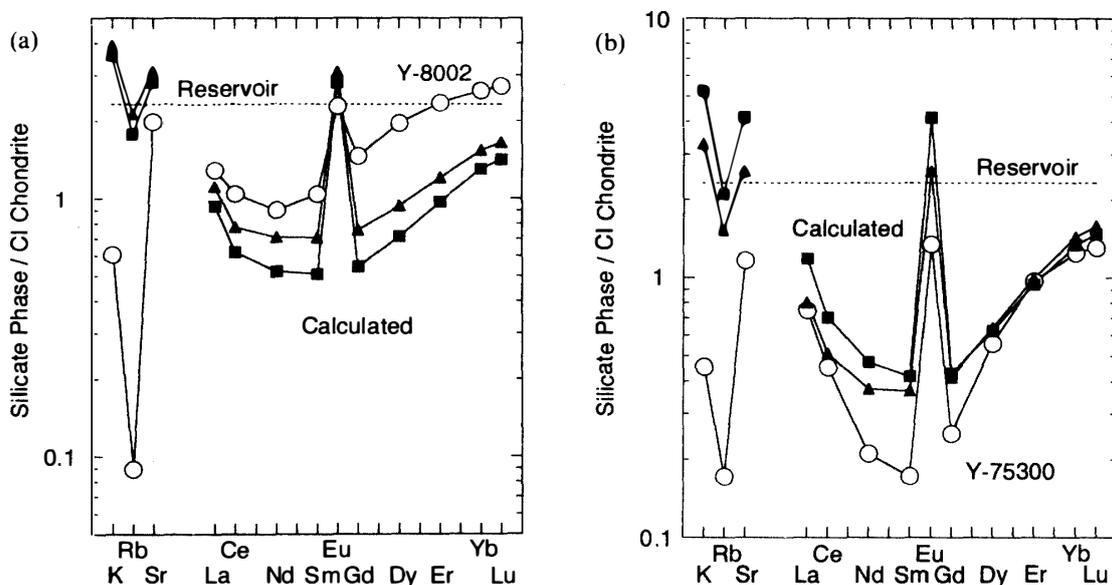


Fig. 8. Model calculations for the abundances of elements in V-shaped meteorites. (a) The calculated pattern for the mineral composition of Y-8002. (b) The calculated pattern for the mineral composition of Y-75300.

ibration with the chondritic reservoir, these meteorites had lost a significant amount of Rb by a small degree of partial melting.

Let us now return to the problem of the REE abundance in Y-8002. It is clear from eq. (4) that the equilibrated concentrations in minerals are affected by mineral composition of the reservoir, x_i . Among the minerals under consideration, phosphate is the most important, because it has the largest partition coefficients of REE. In Y-8002, however, phosphate has never been observed, instead phosphide (shreibersite) is present (YANAI *et al.*, 1984; NAGAHARA, 1992). Further, the Fe/(Fe + Mg) ratios in olivine and pyroxene are the lowest of all the acapulcoite meteorites, as shown in Table 3. These facts indicate reducing condition during the formation of this meteorite. Therefore it is likely that P was present as phosphide but not as oxidized form of phosphate during the chemical equilibration.

The calculation assuming that no phosphate had existed in the reservoir, *i.e.*, $x_{\text{phosphate}} = 0$, was further performed in a similar way. The results for Y-8002 are shown in Fig. 9a. The calculated REE abundances are similar to the analyzed data, indicating the reduced condition during the equilibration of elements. However, the pattern of fractionation in the analyzed sample is somewhat enriched in heavy REE. It is possible that the heterogeneity of the sample affected the analytical results, because of the small sample size. As an example, the model calculation for the mineral composition enriched in orthopyroxene is shown in Fig. 9b. The result indicates that a little enrichment of heavy REE will be explained if the sample analyzed in this study was enriched in orthopyroxene.

4.3.3. Search for a complementary REE pattern

The REE pattern which is complementary to the V-shaped pattern was not observed in this work. Since the meteorites with the V-shaped pattern are depleted in phosphate

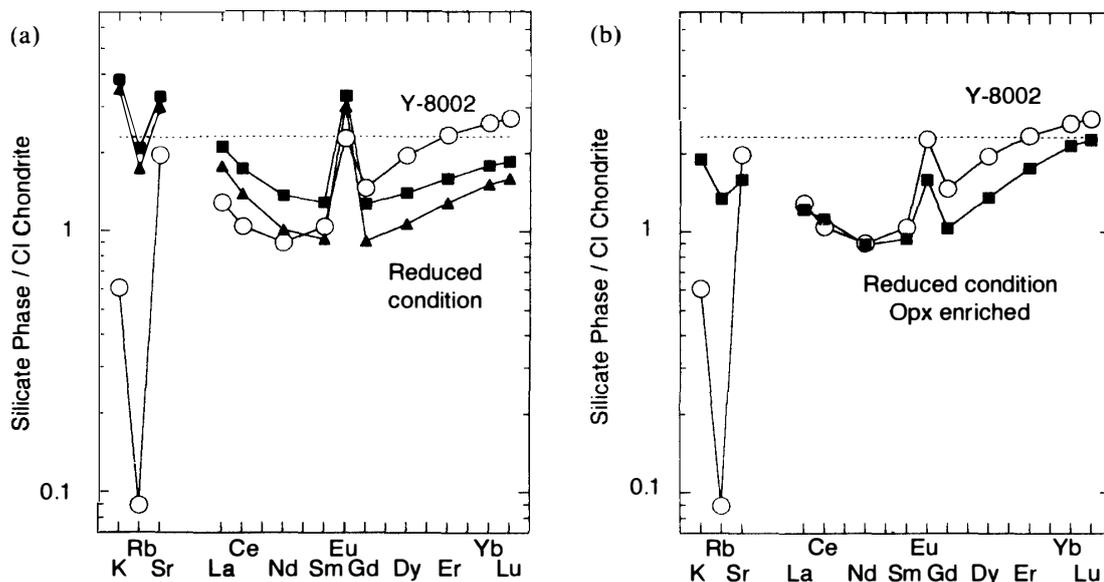


Fig. 9. Model calculations for the abundances of elements in V-shaped meteorites under the reduced condition. (a) The calculated pattern for the mineral composition of Y-8002. (b) The calculated pattern for the mineral composition enriched in orthopyroxene. Mineral composition is as follows; 5% olivine, 89% Opx, 1% Cpx, and 5% plagioclase.

and clinopyroxene, the inverse pattern is expected for the meteorites which are enriched in phosphate or clinopyroxene. Such examples have been obtained from Winona by PRINZ *et al.* (1980). They analyzed two different specimens from Winona, and obtained both V-shaped and the complementary patterns. The REE pattern of the latter is upward convex with maximum at Sm and Gd, and shows a negative Eu anomaly. This sample contains phosphate three times larger amounts than the former sample. Their observation may also be explained by the solid state equilibration process.

4.4. Implications for the formation of "unique" meteorites

4.4.1. Source and thermal processes

The oxygen isotopic study indicates that the "unique" meteorites (acapulcoites-lodranites and winonaites) may be derived from two parent bodies. The petrological studies also indicate that the "unique" meteorites were formed by relatively simple thermal processes (extensive thermal metamorphism and/or partial melting) from a "chondritic" source (NAGAHARA, 1992; KIMURA *et al.*, 1992). The results of trace element model calculations obtained in this work are found to, in general, indicate further constraints on their thermal processes.

As demonstrated above, the presence of both alkali (typically Rb depletion) and REE fractionations for the "unique" meteorites requires at least two-stage evolution from a chondritic source. The first stage, very small degree (<1%) of partial melting and loss of the melt from the source. This stage does not affect REE distribution but only for alkalis, particularly Rb, which is also noted for non-Antarctic "unique" meteorite Acapulco which has a light-REE enriched pattern (PALME *et al.*, 1981). The second thermal process substantially established the REE distributions in the meteorites.

The REE fractionations of the four "unique" meteorites, Y-8002, -74063, ALH-78230, and Y-74357 (acapulcoites-lodranites sub-group) are interpreted as an increasing order of metamorphism (solid state equilibration) to partial melting. Y-8002 may have formed by solid state thermal equilibration (metamorphism). Y-74357 may have formed by more extensive partial melting.

From mineralogical evidence (such as highest Mg/(Mg + Fe) ratios of olivine and pyroxene, and the highest An content of plagioclase), NAGAHARA (1992) suggested that Y-8002 formed by the highest degree of melting among these meteorites from a common chondritic precursor. The trace element data, however, do not seem to support this interpretation. For example, the minor loss of melt formed by extensive partial melting of chondritic source is considered to yield "relatively chondritic" trace element pattern for the meteorite source, which is not consistent with the Rb depletion and the V-shaped pattern for Y-8002. We interpret the different chemical and mineralogical features for the meteorite to be due to the differences in source material and formation condition.

As noted above and often being exercised previously (WARREN and KALLEMEYN, 1989), the V-shaped REE pattern requires specific petrogenetic considerations. Y-75300 (winonaite) has the similar mineral assemblage and the more conspicuous V-shaped REE pattern compared with Y-8002 (acapulcoite). Including these two meteorites, some winonaite samples with V-shaped REE patterns are interpreted to have formed by extensive thermal metamorphism in a "heterogeneous" chondritic source. The trace element implications as expressed here seem to be similar, in general, to the interpretation for winonaite by KIMURA *et al.* (1992).

The solid state equilibrium model for the meteorites with the V-shaped REE pattern indicates that the parent body of these meteorites was heterogeneous in mineral compositions. The scale of the heterogeneity was at least several centimeters, *i.e.*, the size of the meteorites. Nevertheless, mineralogy and chemistry of the parent body was chondritic. This heterogeneity is exemplified by chondrules and surrounding matrices in chondrites, in the sense that olivine, orthopyroxene, and plagioclase are enriched in chondrules, relatively to matrix minerals. If the oxygen isotopic composition of each clast was heterogeneous, just like between a clast in equilibrated ordinary chondrite and its host (MAYEDA *et al.*, 1987), the differences in oxygen isotopic composition among the "unique" meteorites may be explainable.

4.4.2. Formation of Y-74357 and Lodran

Y-74357 is similar to Lodran in many respects; (1) both meteorites consist of olivine, pyroxenes, and Fe-Ni metal, (2) the lithophile elements are depleted to similar degrees, (3) Fs content in pyroxene in both meteorites is 14 (BILD and WASSON, 1976; HIROI and TAKEDA, 1991), which is slightly higher than Fs contents in pyroxenes in other acapulcoite type meteorites (Fs = 11). Further, the Eu anomaly in Lodran (FUKUOKA *et al.*, 1978) is in a similar range to that in Y-74357. These observations suggest that the both meteorites were formed in environments with similar oxygen fugacity and by similar solid-melt partitioning processes. Y-74357 and Lodran may have been derived from the same parent body which is different from that of other "unique" meteorites.

4.4.3. Time of fractionation of the "unique" meteorites

The time of formation of the "unique" meteorites seems to be old, though it was not precisely resolved by the present study. However, there are many lines of evidence

that these meteorites were formed very early in the history of the solar system. The existence of extinct ^{146}Sm at the time of formation of Acapulco was confirmed by the anomaly of ^{142}Nd (PRINZHOFER *et al.*, 1992). KANEOKA *et al.* (1991) obtained ^{40}Ar – ^{39}Ar plateau ages of 4.55 Ga from Y-74063 and ALH-78230, albeit the latter was disturbed the K-Ar system by a later thermal event around 400–500 Ma. Therefore, it is likely that the chemical fractionation processes discussed above may have occurred as a primary process, rather than as a metamorphism of equilibrated chondrites.

5. Conclusions

In order to understand the origin of the “unique” meteorites, chemical and Sr isotopic analyses of several Antarctic “unique” meteorites were performed. The lithophile trace element abundances and modal mineralogy of the meteorites were closely related, so that it was possible to divide them into three groups; (1) Y-74063 and ALH-78230 show nearly chondritic chemical and mineralogical compositions, (2) Y-74357 shows light REE depletion with a negative Eu anomaly and it lacks plagioclase component. (3) Y-75300 and Y-8002 show V-shaped REE patterns with a positive Eu anomaly, and they are depleted in clinopyroxene and phosphate. The close relation between mineralogy and chemistry may reflect the simple processes for the formation of these meteorites. The Rb–Sr isotopic systematics for “whole-rock samples” of these meteorites are consistent with their formation of 4.5 Ga, albeit with a large uncertainty due to the blank correction.

The model calculations for the partition of trace element were performed to constrain the process of chemical and mineralogical fractionation. The first and the third groups may be formed through the small degree (\leq a few %) of partial melting. Such partial melting will produce Rb depletion, but will not fractionate other elements too much. The third group may have been derived from large clast-like materials depleted in phosphate and clinopyroxene. Following the partial melting, the meteorites in this group might have been equilibrated with chondritic reservoir in a solid state. The solid state equilibration between the clast-like materials and a chondritic reservoir can explain unique V-shaped REE pattern.

The second group, Y-74357, is similar to Lodran. The meteorite may have been derived from a solid residue of a partial melting. The degree of partial melting is estimated to be 12%. It is possible that the plagioclase was completely melted and was lost from the solid source region during the partial melting. The variation of Eu/Sr ratios among the ‘unique’ meteorites suggests that redox conditions during the formation of Y-74357 could be more oxidizing than those of other “unique” meteorites did. Therefore, it seems likely that Y-74357 and Lodran could be formed in a parent body different from other “unique” meteorites.

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