

RARE EARTH AND SOME OTHER ELEMENTS IN  
ACID-RESIDUES OF UNEQUILIBRATED  
ORDINARY CHONDRITES

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**Abstract:** Some major, minor and trace elements including rare earth elements (REEs) were determined in bulk and acid (hydrochloric or nitric acid)-residual samples of unequilibrated ordinary chondrites (UOC's), Allan Hills (ALH-) 77299, ALH-78084, Yamato-74191, ALH-77011, ALH-78038, ALH-77252 and ALH-77278. REEs were analyzed by radiochemical neutron activation analysis (NAA), other elements by instrumental NAA.

The content of REEs was found to be higher and more variable in acid-residues of UOC's than in acid-residues of equilibrated ordinary chondrites (EOC's). Cl-normalized REE abundance patterns of acid-residues of UOC's show a gradual increase from La to Lu but their slopes are less steep than for EOC's. Both ALH-78038 and ALH-77011 show a weak negative Eu anomaly in the REE abundance patterns of acid-residues, which is again in sharp contrast to the large positive Eu anomaly in EOC's. Among the UOC's studies, paired meteorites, ALH-77011 and ALH-78038, are the most primitive UOC's.

The amount of REE and the Cl-normalized REE pattern for acid-residues of ALH-77252 are different from those of other UOC's and are essentially the same as those for EOC's, suggesting that this meteorite (or at least the specimen used in this work) is related to EOC's rather than UOC's.

## 1. Introduction

In equilibrated ordinary chondrites (EOC's) corresponding to petrologic types 4 to 6, Ca-phosphates (apatite and merrillite) are the host phase of rare earth elements (REEs) (VAN SCHMUS and RIBBE, 1969; MASON and GRAHAM, 1970; ALLEN and MASON, 1973; CURTIS and SCHMITT, 1979; EBIHARA and HONDA, 1983) and more than 80% of light and middle REEs are contained in Ca-phosphates (SHIMA and HONDA, 1967; EBIHARA and HONDA, 1984). The remaining REEs are partitioned into pyroxene and/or plagioclase. Chondrite-normalized abundance patterns of REEs in Ca-phosphates of EOC's are characterized by a large Eu depletion and a gradual decrease from La to Lu. However, Eu is enriched in plagioclase relative to the other REEs. These features can be explained in terms of an equilibrium distribution of REEs.

Unequilibrated ordinary chondrites (UOC's) corresponding to petrologic type 3 are characterized by several primitive features such as large variation of FeO in

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olivine and pyroxene and a low Ni content in troilite (*e.g.*, DODD *et al.*, 1967). Before the discovery of a large number of meteorites in Antarctica, studies of UOC's have been limited because of a shortage of specimens. A large number of UOC's from Antarctica are now available for study. The absolute contents of REE in UOC's are indistinguishable from those in EOC's: no fractionation among REEs and slight (40–60%) enrichment compared with C1 values (EBIHARA, 1984). The distribution of REEs in different phases of chondritic meteorites, however, may be expected to be quite different in UOC's compared to EOC's.

To study the distribution of REEs in UOC's from Antarctica, phase separation was performed using acid (hydrochloric or nitric acid). Considering that UOC's are assemblages of fine minerals and that sample amounts allowed to use are limited, mechanical separation using heavy liquids and an isodynamic separator is thought to be difficult, leaving a dissolution method for phase separation of UOC's.

In this study, some major, minor and trace elements were determined non-destructively by instrumental neutron activation analysis (NAA) for bulk and acid-residual samples. Furthermore, both bulk and acid-residues were analyzed for REEs by radiochemical NAA.

## 2. Experimental

### 2.1. Samples

A total of seven meteorites collected in Antarctica were studied in this work. Allan Hills (hereafter ALH-)77252 was previously classified as L3/L6 (L3 with L6) (YANAI, 1983) but is now listed as an L3 in the latest catalog issued by the National Institute of Polar Research (NIPR) (YANAI and KOJIMA, 1987). The rest are all type 3 UOC's. Sample names and additional information are listed in Table 1. ALH-77011 and ALH-78038 were paired by MCKINLEY *et al.* (1981), on the basis of their microscopic studies.

Several chips were loaned by NIPR for each meteorite. Appropriate amounts of these specimens were pulverized and ground in a clean agate mortar. Metal particles were not removed.

Table 1. Samples used in this work.

Meteorite	Subnumber	Class & type*	Degree of weathering**	Fraction (%) of acid-residues
ALH-77299	88	H3.7	A	35.1
ALH-78084	91	H3.9	B/C	35.1***
Y-74191	107	L3.6	A	37.8***
ALH-77011****	86	L3.5	C	39.2
ALH-78038	71	L3.4	C	38.2
ALH-77278	63	LL3.6	A	32.0
ALH-77252	71	L3(L4/L6)	B	36.0

\* SEARS *et al.* (1982) see text for ALH-77252.

\*\* A: minor, B: moderate, C: severe.

\*\*\* estimated values (see text).

\*\*\*\* ALH-78038 is tentatively paired with ALH-77011.

## 2.2. Phase separation by acid

About 50–200 mg of a powdered meteorite sample was taken into a clean glass vial (10 ml) with a screw cap. The sample was at first treated with 5–10 ml of 1 M hydrochloric acid or nitric acid at room temperature for 1 h. The residue was then treated several times with the same media under harder conditions (higher molality, up to 6 M, higher temperature, up to 120°C and/or longer contact time, *e.g.* overnight) until the acid-leachate became clean and colorless at the final treatment. The major phases dissolved in hydrochloric or nitric acid are metal, olivine and sulfide (mainly troilite), in which iron is a major constituent element. Calcium-phosphates, which are present in UOC's (RAMBALDI and RAJAN, 1982; YABUKI and EL GORESY, 1986) as well as in EOC's, can also be dissolved in acid. Being washed with distilled water several times, insoluble materials (referred to as acid-residues in this paper) were dried and weighed.

In some cases, silica gel appeared during acid-treatment. Because the content of silica could not be determined, silica-free acid-residues were carefully prepared to obtain REE abundance patterns for acid-residual phases. Acid residues consist mainly of pyroxene and plagioclase. Acid-residues may also contain some glass. Acid residual fractions are shown in Table 1. About 60–70% of the starting materials were dissolved after acid leaching. Values for ALH-78084 and Yamato (hereafter Y-)74191 are missing because of experimental problems. These blanks were filled by averaging values of the same petrologic types. Since values of the acid-residual fraction were fairly constant, no serious error may be involved in such an estimation.

## 2.3. Instrumental neutron activation analysis (INAA)

Major and minor elements were determined by INAA for both bulk and acid-residual samples. Some 5 to 30 mg of each sample was heat-sealed doubly in clean polyethylene bags. BCR-1, JB-1 and/or the Allende reference sample were used for reference standards (FLANAGAN, 1973; ANDO *et al.*, 1987; JAROSEWICH *et al.*, 1987). Samples were at first irradiated for 2 min at a neutron flux of  $1.5 \times 10^{12} \text{ cm}^{-2} \text{ s}^{-1}$  in a Triga-II reactor at the Institute for Atomic Energy of Rikkyo University or the Atomic Energy Research Laboratory of Musashi Institute of Technology. At this time pure reagents of SiO<sub>2</sub>, MgO and metallic Al were also irradiated to monitor the interference due to secondary reactions. After an appropriate cooling, gamma-ray intensities of short-lived nuclides such as <sup>27</sup>Mg, <sup>24</sup>Na, <sup>52</sup>V, <sup>28</sup>Al, <sup>56</sup>Mn, and <sup>49</sup>Ca were measured. All samples were again irradiated for 5–12 h at the same neutron flux in the reactor of Rikkyo University. After cooling for 4–5 days, gamma-ray counting started. Samples were repeatedly subjected to gamma-ray counting after appropriate cooling intervals. Relatively longer-lived nuclides such as, for example, <sup>153</sup>Sm, <sup>51</sup>Cr, <sup>198</sup>Au, <sup>192</sup>Ir, <sup>58</sup>Co, <sup>48</sup>Sc, <sup>60</sup>Co, <sup>59</sup>Fe, and <sup>24</sup>Na were detected.

## 2.4. Radiochemical neutron activation analysis (RNAA)

Bulk and acid-residual samples were analyzed for ten REEs, La, Ce, Nd, Sm, Eu, Gd, Tb, Tm, Yb, and Lu, by RNAA. Some 15–30 mg of each sample was sealed in a clean, synthesized quartz tubing. Standard samples were prepared from pure chemical reagents. All the samples were irradiated at a neutron flux of  $5 \times$

$10^{13} \text{ cm}^{-2} \text{ s}^{-1}$  for 12 h in the JRR-4 reactor of the Japan Atomic Energy Research Institute (JAERI). Analytical procedures applied were essentially the same as those described in EBIHARA (1987).

### 3. Results and Discussion

#### 3.1. Major and minor elements in bulk and acid-residual samples

INAA results for some major and minor elements in bulk and acid-residual samples are summarized in Table 2 and distributional budgets of these elements are given in Table 3. There seems to be a systematic error in Mg (and possibly Fe) data: most of the Mg data are smaller than the average literature values for the same group of meteorites. However, both Fe and Mg data, along with the Ca, Ni, Mn, Co and Sc data, for Y-74191 are in good agreement with those reported by HIRANO *et al.* (1980).

Table 2. Major and minor element compositions (in %, otherwise indicated) of bulk and acid-residual samples.

Meteorites	Fe	Mg	Al	Ca	Ni	Na	Cr	Mn	Co/ ppm	V/ ppm	Sc/ ppm	Ir/ ppb	Au/ ppb
<i>Bulk samples</i>													
ALH-77299	24.6	12.6	1.06	1.15	1.40	0.69	0.35	0.22	610	66	7.9	680	240
ALH-78084	22.6	13.1	1.00	1.22	1.42	0.58	0.35	0.22	630	60	7.9	590	188
Y-74191	19.3	14.9	1.18	1.37	1.03	0.68	0.40	0.27	410	75	8.9	360	134
ALH-77011	18.1	12.7	1.08	1.43	0.86	0.59	ND*	0.24	370	67	8.9	540	177
ALH-78038	18.8	13.7	1.20	1.33	1.10	0.51	ND	0.25	490	38**	7.8	380	159
ALH-77278	20.7	14.6	1.16	1.36	1.61	0.79	0.40	0.27	610	73	8.5	400	200
ALH-77252	20.7	12.9	1.06	1.02	0.95	0.61	0.42	0.24	470	67	8.7	460	123
<i>acid-residues</i>													
ALH-77299	5.6	12.9	2.2	2.1	BL***	1.26	ND	0.26	5.9	143	14.8	1030	88
ALH-78084	7.4	13.8	2.1	2.1	0.0196	1.19	0.68	0.28	13.1	118	14.8	270	165
Y-74191	6.9	13.8	2.1	2.3	0.0139	1.16	0.66	0.32	6.4	112	15.3	196	72
ALH-77011	4.7	12.9	1.91	2.3	BL	1.09	0.70	0.28	11.5	130	18.4	320	122
ALH-78038	4.7	12.3	1.78	2.2	BL	1.03	0.65	0.28	11.6	125	15.6	210	78
ALH-77278	7.0	11.1	2.7	2.7	BL	1.60	ND	0.24	8.2	170	15.1	570	22
ALH-77252	7.0	13.2	2.6	2.5	BL	1.48	ND	0.27	4.9	181	16.1	680	30

\* ND: not determined.

\*\* doubtful.

\*\*\* BL: below detection limit.

Table 3. Distributional fractions (in %) of major and minor elements in acid-residues.

Meteorites	Fe	Mg	Al	Ca	Ni	Na	Cr	Mn	Co	V	Sc
ALH-77299	8.0	37	73	64	—	64	—	41	0.34	76	66
ALH-78084	11.5	37	74	60	0.48	72	68	45	0.73	69	66
Y-74191	13.5	35	67	63	0.51	64	62	45	0.59	56	65
ALH-77011	10.2	40	69	63	—	72	—	46	1.22	76	81
ALH-78038	9.6	34	57	63	—	77	—	43	0.90	—	76
ALH-77278	10.8	24	74	64	—	65	—	28	0.43	75	57
ALH-77252	12.2	37	88	88	—	87	—	41	0.38	97	67

Both Ni and Co were found to have very low concentrations in acid-residues, suggesting that phase separation was performed satisfactorily in the sense that no acid-soluble phases remained undissolved in acid-residues. However, some 10% of Fe was still present in acid-residues. A part of this Fe can be explained by undissolved pyroxene. Considering that average ferrosilite contents (16.8% for ALH-78084 and 20.0% for ALH-77252, YANAI and KOJIMA, 1987), we cannot account for the Fe contents in acid-residues. It is, therefore, suggested that some Fe is present in the remaining phases (glass and/or Fe spinel?). Aluminum, Ca and Na are plagiophile elements and more than 60% of these elements were found to be present in acid-residues. In UOC's, plagioclase is less abundant than in EOC's. Most of these plagiophile elements must instead be present in glass.

Apparently there is a large discontinuity between ALH-77252 and the rest of UOC's in the distribution of plagiophile elements: Al, Ca, Na are more abundant in acid-residues of ALH-77252 than in those of the rest, suggesting that ALH-77252 is not related to UOC's. This possibility will be discussed in detail in the last section. Scandium was found to be more abundant in acid-residues of ALH-77011 (and ALH-78038) than in those in the remaining UOC's. REEs show a similar tendency as discussed in the following section.

Distributional fractions for refractory siderophiles, Ir and Au, are not shown in Table 3 although these values can be easily calculated based on Tables 1 and 2. They will, however, be discussed elsewhere.

### 3.2. REE distribution in acid-soluble and -residual phases of UOC's

REE contents in the bulk and acid-residual samples are summarized in Table 4 and their distributional budgets are given in Table 5.

Table 4. Rare earth elements (in ppb) in bulk and acid-residual samples.

Meteorites	La	Ce	Nd	Sm	Eu	Gd	Tb	Tm	Yb	Lu	lit.*
<i>Bulk samples</i>											
ALH-77299	355	889	596	205	80	295	58	37	234	37	(1)
ALH-78084	371	958	648	217	77	327	54	39	232	38	(2)
Y-74191	436	1120	809	272	110	401	67	47	285	47	(2)
ALH-77011	354	866	547	194	76	259	48	34	209	33	(1)
ALH-78038	281	812	506	185	74	262	50	34	202	33	(1)
ALH-77278	338	984	598	198	85	299	53	36	225	36	(1)
ALH-77252	408	1080	792	231	92	401	59	40	244	38	
<i>acid-residues</i>											
ALH-77299	310	602	692***	179	108	317	59	41	264	40	
ALH-78084	259	735	454	187	127	ND**	55	ND	283	46	
Y-74191	437	1180	888	310	130	ND	90	ND	412	66	
ALH-77011	510	1330	1010	327	91	467	85	54	376	54	
ALH-78038	368	989	629	250	82	352	61	44	305	46	
ALH-77278	281	754	541	187	155	264	58	45	267	44	
ALH-77252	109	234	230	95	134	152	31	29	198	32	

\* literature ((1): EBIHARA (1987); (2): EBIHARA (1984)).

\*\* ND: not determined.

\*\*\* doubtful.

Table 5. *Distributional fractions (in %) of REEs in acid-residual phases.*

Meteorites	La	Ce	Nd	Sm	Eu	Gd	Tb	Tm	Yb	Lu
ALH-77299	31	24	41*	31	47	38	36	39	40	38
ALH-78084	26	28	26	32	60	—	38	—	45	45
Y-74191	36	38	39	41	42	—	48	—	52	50
ALH-77011	57	60	72	66	47	71	70	63	71	64
ALH-78038	50	47	48	52	42	51	47	50	58	54
ALH-77278	27	25	29	30	58	28	35	40	38	39
ALH-77252	9.6	7.8	10.5	14.7	52	13.6	18.6	26	29	30

\* doubtful.

Except for ALH-77252, REEs were found to be more concentrated in acid-residues of UOC's than in those of EOC's, where only 20% of the bulk Sm was determined in acid-residual phases even for type 4 chondrite, Kesen (EBIHARA and HONDA, 1984). This value was found to be fairly constant for EOC's. On the contrary, the fraction of REEs in acid-residues seems to be rather variable among UOC's. For example, fractions of Sm allotted to acid-residues of UOC's vary from 30% to 66%. The only exception is Eu, whose fractions in acid-residues are rather constant (42–60%). Among REEs, heavier REEs are distributed in acid-residues more than lighter REEs.

Among the UOC's studied, Y-74191 (L3.6), ALH-77011 (L3.5) and ALH-78038 (L3.4) were found to be enriched in all REEs over the level of their bulk abundances (Table 4). REE contents in acid-residues of these three meteorites are apparently higher than those of ALH-78084 (H3.9), ALH-77299 (H3.7) and ALH-77278 (LL3.6). Considering that sub-numbers applied to UOC's are correlated with the degree of equilibrium in both mineralogy and chemistry (SEARS *et al.*, 1982; ANDERS and ZADNIK, 1985), REE contents in acid-residual phases (or REE budgets in acid-residues) seem to be closely correlated with the primitiveness for UOC's.

### 3.3. REE abundance pattern of acid residues of UOC's

REE abundance patterns of acid-residues are illustrated in Fig. 1. Cl chondrite values (ANDERS and EBIHARA, 1982) were used for normalization. For comparison, patterns for Kesen (H4) and St. Séverin (LL6) (M. EBIHARA, unpublished data) also are shown in Fig. 1.

Among the REEs, heavy REEs are relatively more enriched in acid-residues, causing a gradual increase towards Lu in their REE abundance patterns. The slopes of REE abundance patterns for the more metamorphosed chondrites (St. Séverin, for example) are steeper than those for the less metamorphosed meteorites (*e.g.* ALH-78038). Heavy REEs can migrate into pyroxene (one of major constituent minerals in acid-residues) more easily than light REEs, producing the observed slope in the REE abundance pattern for acid-residual phases of EOC's. Hence, relative enrichment of heavy REEs to light REEs also seems to be correlated with the primitiveness of chondrites. Apparently a nearly flat REE abundance pattern and a large distributional budget of REEs in acid-residues are suggested as the most primitive features for UOC's. Among UOC's studied, paired meteorites, ALH-77011 and ALH-78038,

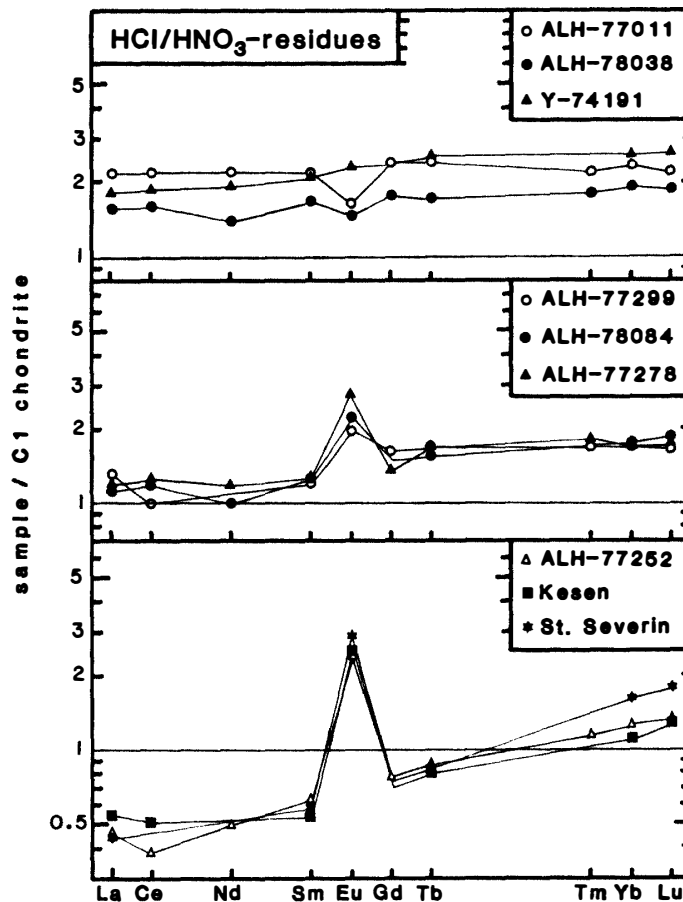


Fig. 1. C1 chondrite-normalized REE abundance pattern of acid-residues. Among the REEs, heavy REEs are relatively enriched in acid-residues. The slopes of the abundance patterns for the more metamorphosed meteorites are steeper than those for the less metamorphosed meteorites. Both positive and negative anomalies of Eu were found. Acid-residues of relatively primitive UOC's seem to have a negative Eu anomaly, while those of EOC's have a large positive anomaly of Eu. Relative enrichment of heavy REEs to light REEs as well as an Eu anomaly seem to be correlated with the primitiveness of chondrites. REEs must have been largely redistributed among UOC's according to the degree of thermal metamorphism.

belong to such a group of the most primitive UOC's.

Acid-residual phases of EOC's show a large positive anomaly of Eu in their abundance patterns of REEs. Such a positive anomaly of Eu is due to plagioclase, which is another constituent mineral in acid-residues. Among UOC's, a positive anomaly of Eu does not seem to be common for REE abundance patterns of acid-residues, because plagioclase is less abundant or absent in UOC's. ALH-78084, ALH-77299 and ALH-77278 show apparent positive anomalies, but these anomalies are small compared with those for EOC's. On the contrary, both ALH-78038 and ALH-77011 show a faint but definite negative anomaly of Eu in REE abundance patterns of acid-residues. Considering that these (paired) meteorites are assumed to be more primitive than ALH-77299 and ALH-77278, a negative anomaly of Eu is suggested to be one of the common features the most primitive UOC's possess. Con-

sidering that there is a large variation in REE patterns among UOC's, REEs must have been largely redistributed among UOC's according to the degree of thermal metamorphism the chondrites experienced, possibly on their parent body(ies).

### 3.4. ALH-78084 and ALH-77252

ALH-78084 was reclassified from H3 to H4 by SCOTT (1984) from the viewpoint of small deviation of olivine composition (Fa:  $18.7 \pm 1.7\%$ ). REE abundance pattern for acid-residual phases of ALH-78084, however, suggests that this meteorite is strongly related with UOC's rather than EOC's.

ALH-77252 was originally classified to be a half-breed of L3 and L6 (YANAI, 1983). Based on the compositional variations of olivine (Fa: 0.4–30.1%) and pyroxene (Fs: 4.8–37.6%), YANAI and KOJIMA (1987) later changed its classification to L3. On the other hand, KING *et al.* (1980) classified this meteorite as L4/L6 (L4 with L6 clasts) after revision of its earlier classification as L5-L6. SCOTT (1984) later described this meteorite as L3 and a regolith breccia containing solar-wind noble gases and equilibrated clasts. The REE budget of this meteorite is confirmed to be apparently distinct from that of other type 3 chondrites studied: distributional fractions of REEs into acid-residues of ALH-77252 are much smaller than those of type 3 chondrites. The Cl-normalized REE abundance pattern of the acid-residue of this meteorite is largely different from those of UOC's and is essentially the same as those for EOC's: low abundance with a steep slope and a large Eu anomaly. Thus, the REE distribution and their abundance pattern in acid-residues suggest that ALH-77252 is related to EOC's rather than to UOC's. It should be noted, however, that this conclusion cannot be necessarily applied to the whole specimen of ALH-77252 because the sample size used in this work was rather small (200 mg). Considering that this meteorite is somewhat brecciated (SCOTT, 1984), we have a possibility of having analyzed the equilibrated fragment in which REEs were redistributed by brecciation.

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