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Reexamination of purely physical separation of the phase enriched in noble gases from the Allende meteorite

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Abstract: We have prepared a new sample of the "floating fraction" of the Allende meteorite and have analyzed the elemental and isotopic compositions of the noble gases to examine the properties of the separation technique. The "floating fraction" is the black fine material, which floats on the water during freeze-thaw disaggregation. The floating fraction separated in this study was enriched in noble gases, and its isotopic compositions were very similar to those of chemical residues. We thus confirmed that the physical separation is an effective method to obtain a noble-gas-rich fraction than the chemical treatment. The only major difference between our floating fraction and those previously prepared was that the former had small excesses in ^{40}Ar and ^{129}Xe which were supposed to be due to the contamination from small amounts of silicate like sodalite. There were some variations in the Xe-Q/Xe-HL ratios among floating fractions, suggesting that Q- and HL-components might be separable by a physical process.

1. Introduction

Lewis *et al.* (1975) found that noble gases were concentrated in a very small fraction of the Allende meteorite. They dissolved the Allende CV3 chondrite with HCl, HF and CS_2 and obtained a residue, which comprises 0.5% of the bulk meteorite. The residue ("original residue") carried most of noble gases in the bulk meteorite. Therefore, the concentrations of noble gases in the original residue were higher than those in the bulk meteorite by more than two orders of magnitude. Then, they etched the original residue with oxidizing acids like HNO_3, which dissolved about 4 to 8% of the original residue (*i.e.*, only about 0.02 to 0.04% of the bulk meteorite), and obtained the "oxidized residue". The oxidized residue retained most of He and Ne, but lost most of heavy noble gases such as Ar, Kr and Xe. This means that the heavy noble gases were strongly enriched in the fraction removed by the oxidation procedure. Lewis *et al.* (1975) named this oxidizable fraction "phase Q" after "*quintessence*". The Q component is isotopically "normal" and dominates the Ar, Kr and Xe in primitive chondrites. In the oxidized residue of Allende, in terms of noble gas concentrations, diamond is the dominant phase, and carries the "HL" component, an isotopically "exotic" component which exhibits large excesses of light and heavy Xe isotopes and smaller excesses of heavy Ar and Kr isotopes. Q-gas is enriched in heavy noble gases, while HL-gas is enriched in light noble gases. In order to obtain these grains with chemical procedures, we have to dissolve more than 99% of sample.

Recently, Q-gas rich fractions were separated from the Allende meteorite by a purely physical method (Matsuda *et al.*, 1999). They applied the freeze-thaw disaggregation method to the Allende meteorite and found that fine black material floated on the water during the procedure. This material contained abundant noble gases with concentrations similar to the "original residue". This was the first time that Q-gas rich fractions had been separated from a bulk meteorite by purely physical method. The original residues had ^{129}Xe excess probably due to re-adsorption of ^{129}Xe from the dissolved iodine-rich minerals (Srinivasan *et al.*, 1978). On the other hand, the floating fractions by Matsuda *et al.* (1999) did not show the ^{129}Xe excess. Absence of the ^{129}Xe excess indicated that the physical separation should be free from the re-adsorption. Therefore, the physical separation is a better way to obtain the pure primordial noble gases than the chemical separation.

Matsuda *et al.* (1999) also reported that the Xe-Q/Xe-HL ratio in one floating fraction was different from that in the other. This observation indicated that phase Q and presolar diamonds (host phase of Xe-HL) can be separated by purely physical means, although Q and HL components are tightly coupled and could not previously be separated without using an oxidizing acid.

In this study, we rechecked the property of the physical separation technique, and examined whether noble gas compositions in floating fractions were uniform or not in comparison with the previous report (Matsuda *et al.*, 1999).

2. Sample preparation

We applied the freeze-thaw disaggregation method, which has been used to recover refractory inclusions in the Murchison CM2 meteorite (MacPherson *et al.*, 1983), to the Allende CV3 meteorite. 3.846 g of Allende was soaked in distilled water, and was frozen for 90 min and then put into an ultrasonic bath for 40 min. The water temperature in the ultrasonic bath increased to about 50°C. After the ultrasonic operation, the sample was frozen again. As this cycle was repeated, the meteorite gradually came to disintegrate by the expansion of water which soaked into the meteorite. The chondrules and inclusions sank quickly to the bottom of the beaker, whereas the matrix remained suspended in the water for a while after ultrasonic vibration. We decanted the water in which matrix materials were in suspention into another beaker every 30 cycles, and added fresh distilled water to the original beaker. The matrix material sank usually in a day or so. Some very fine black materials kept floating on the water. They are the materials named "floating fraction" in Matsuda *et al.* (1999). We scooped them up with aluminum foil, and dried it at 120°C for one hour. After 141 cycles of the freeze-thaw disggregation, 2.55 mg of a floating fraction has been obtained. Nine percent of the fraction (0.23

mg), labeled C1-7, was used for noble gas analysis.

3. Experimental

The sample C1-7 was loaded in a molybdenum crucible in the furnace of vacuum line. Noble gases were extracted from the sample by stepwise heating at 800°C, 1200°C and 1600°C with a tantalum heater. The extracted gas was purified by a Ti-Zr getter, which was heated at ~700°C for 15 min, and then was cooled down for 10 min. After the purification, the heavier noble gases were trapped on activated charcoal (CT-1) at liquid nitrogen temperature (−196°C, for 30 min).

Table 1. He, Ne and Ar in fractions separated from the Allende meteorite.

Sample	Temp. [°C]	[^{4}He]	^{3}He/^{4}He ×10^{-4}	[^{20}Ne]	^{20}Ne/^{22}Ne	^{21}Ne/^{22}Ne	[^{36}Ar]	^{38}Ar/^{36}Ar	^{40}Ar/^{36}Ar
C1-7	800	84000	1.646	580	8.76	0.0383	1000	0.19053	5.09
floating fraction			±.105		±.17	±.0008		±.00087	±.11
(this study)	1200	13000	1.010	78	9.00		280	0.18920	1.52
			±.098		±.50			±.00087	±.36
	1600			13	10.89	0.0263	53	0.18903	50.84
					±4.32	±.0120		±.00151	±6.06
	total	97000 [*6]	1.561 [*6]	670	8.82		1400	0.19019	6.14
			±.091		±.17			±.00069	±.27
A1 [*1]		34000	1.870	240	9.38	0.0448	1400	0.18841	
floating fraction			±.046		±.12	±.0047		±.00057	
A1A [*1]		81000	1.792	560	9.827	0.0541	2190	0.18683	7.02
floating fraction			±.039		±.057	±.0011		±.00036	±.53
bulk Allende [*2]		3100	34.	4.7	2.329	0.7852	22	0.2045	207.
					±.022	±.0088		±.0048	±3.
3C1 [*3]		189000		1085	9.44	0.04231	3715	0.1902	3.95
original residue					±.15	±.00075		±.0012	±.08
3C2 [*3]		169000		946	8.53	0.04346	390	0.1918	1.1
oxidized residue					±.17	±.00082		±.0027	±1.0
1CS [*4]				388	8.997	0.04239			
original residue					±.073	±.00044			
BA [*5]		105800	1.779	583	8.61	0.0513	1880	0.1887	1.84
original residue			±.016		±.08	±.0005		±.0004	±.02

Gas concentrations in 10^{8}cm^{3}STP/g.

*1 Matsuda *et al.* (1999). Only the total data were shown.

*2 Lewis *et al.* (1977).

*3 Lewis *et al.* (1975). Helium isotope was not measured.

*4 Srinivasan *et al.* (1978). There were no data for Helium and Argon.

*5 Wieler *et al.* (1991).

*6 This was calculated from the data of 800°C and 1200°C fractions.

The remaining gases were purified again by second Ti-Zr getter and exposed to a cold trap (CT-2) and a SAES getter. After this re-purification, the gases were introduced to the mass spectrometer for the Ne, and then He isotopic analyses. After analyses of Ne and He, the heavy gases (Ar, Kr and Xe) were released from CT-1 at ~100°C, and purified again by the Ti-Zr getter. The heavy noble gases were frozen onto another cold trap (CT-3) composed of a sintered stainless steel element (Maruoka and Matsuda, 1995). Argon was separated from Kr and Xe by using CT-3 kept at the liquid nitrogen temperature, which traps all Kr and Xe, and about half of Ar. After the analysis of Ar, Kr was separated from Xe by using CT-3 kept at −163°C, which could trap all Xe and releases all Kr. After Kr analysis,

Table 2. Kr in fractions separated from the Allende meteorite.

Sample	Temp.	[^{84}Kr]	^{78}Kr	^{80}Kr	^{82}Kr	^{83}Kr	^{86}Kr
	[°C]		/ ^{84}Kr = 100				
C1-7	800	330	0.68	3.829	20.43	20.42	31.81
floating fraction			±.14	±.048	±.23	±.20	±.66
(this study)	1200	330	0.59	3.646	19.83	20.20	30.94
			±.05	±.061	±.10	±.09	±.25
	1600	81	0.57	3.449	19.79	19.97	31.28
			±.16	±.157	±.20	±.31	±.44
	total	750	0.63	3.707	20.10	20.27	31.37
			±.07	±.039	±.11	±.11	±.32
A1 *1		1160					
floating fraction							
A1A *1		1970					
floating fraction							
bulk Allende *2		26.1	0.613	5.268	20.621	20.187	30.964
			±.039	±.015	±.039	±.048	±.033
3C1 *3		3827	0.603	3.969	20.153	20.209	31.043
original residue			±.006	±.037	±.097	±.085	±.082
3C2 *3		389	0.57	3.76	19.08	20.024	32.067
oxidized residue			±.033	±.063	±.015	±.099	±.095
1CS *4		1015	0.593	3.94	20.059	20.168	31.221
original residue			±.012	±.018	±.052	±.046	±.065
BA *5		1830	0.620	3.965	20.19	20.29	31.12
original residue			±.004	±.010	±.03	±.07	±.06

Gas concentrations in 10^{10}cm^{3}STP/g.

*1 Matsuda *et al.* (1999). There were no data for Krypton isotopic ratios.

*2 Srinivasan *et al.* (1978).

*3 Lewis *et al.* (1975).

*4 Srinivasan *et al.* (1978).

*5 Wieler *et al.* (1991).

Xe was released from CT-3 at −90°C and introduced to the mass spectrometer.

A hot blank, which was measured by the same procedure as that of a sample run but without a sample in the furnace, was measured before each run and used for a correction in order to obtain the noble gas composition indigenous to the sample.

Sensitivity and mass discrimination were determined by analyzing known amounts of atmospheric gases except He. Isotopic compositions of standard air used here (except He) were from Ozima and Podosek (1983). For He, an artificially prepared sample was used (^{3}He/^{4}He$=(2.862\pm0.049)\times10^{-5}$; Matsumoto and Wada, private communication).

Each noble gas fraction was analyzed with VG5400 (extended radius of 54 cm), a sector-type mass spectrometer, installed in Osaka University.

Table 3. Xe in fractions separated from the Allende meteorite.

Sample	Temp. [°C]	[^{132}Xe]	^{124}Xe	^{126}Xe	^{128}Xe	^{129}Xe	^{130}Xe	^{131}Xe	^{134}Xe	^{136}Xe
					/ ^{132}Xe = 100					
C1-7	800	1000	0.472	0.389	8.467	104.32	16.14	82.38	39.46	33.99
floating fraction			± .026	± .032	± .083	± .40	± .14	± .98	± .14	± .10
(this study)	1200	340	0.494	0.378	8.410	107.95	16.30	81.85	39.00	33.44
			± .030	± .036	± .087	± .41	± .14	± .94	± .16	± .11
	1600	59	0.409	0.370	8.439	117.87	16.24	81.86	38.82	33.12
			± .063	± .067	± .152	± .63	± .19	± 1.03	± .21	± .18
	total	1400	0.475	0.385	8.452	105.76	16.18	82.23	39.32	33.82
			± .020	± .025	± .063	± .31	± .11	± .74	± .11	± .08
A1 *1		1180			8.15	103.6	16.39	82.2	39.43	33.59
floating fraction					± .21	± 1.3	± .46	± 1.2	± .88	± .58
A1A *1		2240	0.47	0.418	8.054	102.48	15.99	81.65	39.89	34.90
floating fraction			± .013	± .014	± .065	± .48	± .11	± .17	± .21	± .28
bulk Allende *2		17	0.47	0.418	8.30	207.5	16.07	81.97	39.39	33.84
			± .004	± .004	± .04	± 1.1	± .04	± .14	± .10	± .14
3C1 *3		3033	0.4712	0.4109	8.152	113.88	16.09	81.83	39.31	33.83
original residue			± .0068	± .0044	± .043	± .76	± .12	± .66	± .20	± .21
3C2 *3		421	0.586	0.4565	8.55	110.33	15.92	82.94	47.25	46.11
oxidized residue			± .013	± .0094	± .067	± .77	± .12	± .62	± .30	± .31
1CS *4		971	0.4708	0.4142	8.225	114.81	16.131	81.85	39.677	34.385
original residue			± .0036	± .0025	± .027	± .25	± .042	± .14	± .059	± .049
BA *5		1960	0.475	0.414	8.288	114.4	16.17	81.73	39.20	33.77
original residue			± .005	± .004	± .022	± .4	± .06	± .26	± .09	± .09

Gas concentrations in 10^{10}cm^{3}STP/g.

*1 Matsuda *et al.* (1999). Only the total data were shown.

*2 Lewis *et al.* (1977).

*3 Lewis *et al.* (1975).

*4 Srinivasan *et al.* (1978).

*5 Wieler *et al.* (1991).

4. Results and discussion

The elemental abundances and isotopic ratios of noble gases are shown in Tables 1 to 3. We also list the data for bulk Allende (Lewis *et al.*, 1977), original residues 3C1 (Lewis *et al.*, 1975), 1CS (Srinivasan *et al.*, 1978) and BA (Wieler *et al.*, 1991), oxidized residue 3C2 (Lewis *et al.*, 1975), and floating fractions A1 and A1A (Matsuda *et al.*, 1999) of Allende for comparison.

4.1. Elemental abundance

The elemental concentrations are shown in Fig. 1. Noble gas concentrations in the samples obtained by the chemical or physical separation are higher than those of bulk Allende by around two orders of magnitude. C1-7 also has the concentrations clearly higher than those of the bulk meteorite, and its noble gas elemental abundance pattern is very similar to those of original residues just like the previous samples Al and A1A (Matsuda *et al.*, 1999). Therefore, we confirmed that elemental abundances of the noble gases in floating fractions were very similar to

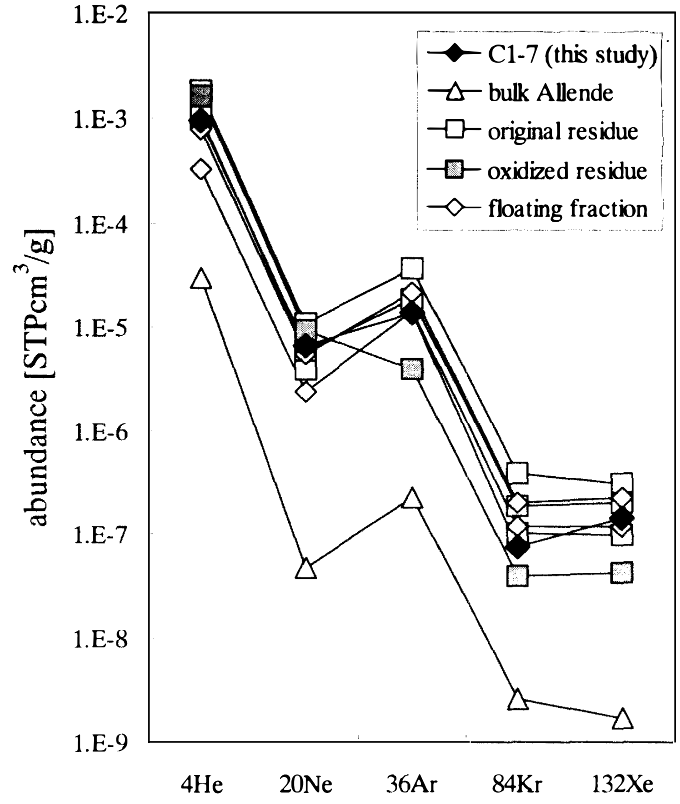

Fig. 1. Comparison of elemental concentrations of noble gases in the fractions separated from the Allende meteorite. The vertical axis is in a logarithmic scale. The data sources are as follows; bulk Allende (Lewis et al., 1977), 3C1 and 3C2 (Lewis et al., 1975), 1CS (Srinivasan et al., 1978), BA (Wieler et al., 1991) and A1 and A1A (Matsuda et al., 1999). The noble gases were as highly concentrated in the floating fractions as in the original residues.

those in the original residues. We collected 2.55 mg of a floating fraction in total from 3.846 g of the Allende bulk. This yield of ~0.066% is much lower than the 0.5–1% reported for original residues (Lewis *et al.*, 1975; Srinivasan *et al.*, 1978; Wieler *et al.*, 1991), but is much higher than the ~0.004% for the floating fractions reported by Matsuda *et al.* (1999). Thus, the recovery yield depends on the degree of decomposition of samples.

4.2. *Isotopic ratios*

^{3}He/^{4}He ratios are shown in Fig. 2. The total value of the ^{3}He/^{4}He ratio in C1-7 ($(1.561 \pm 0.092) \times 10^{-4}$) is nearly the same as those in He-Q ($(1.59 \pm 0.04) \times 10^{-4}$; Wieler *et al.*, 1991) and He-HL ($(1.70 \pm 0.10) \times 10^{-4}$; Huss and Lewis, 1994). Although the ^{3}He/^{4}He ratio in the bulk meteorite is much higher than these values due to the cosmogenic He (Lewis *et al.*, 1977), the cosmogenic He did not severely affect He in C1-7. The 800°C fraction has a somewhat higher ratio than the 1200°C fraction, due to the cosmogenic He. Therefore, the higher temperature fraction may give a better value for the ^{3}He/^{4}He ratio indigenous to the floating fraction. The obtained value of ($(1.010 \pm 0.098) \times 10^{-4}$) at 1200°C is much lower than the lowest value ($(1.516 \pm 0.029) \times 10^{-4}$) of 1700°C fraction of A1A, and probably due to the contribution of He-P3 ($< 1.39 \times 10^{-4}$; Huss and Lewis, 1994) in the presolar diamond (Matsuda *et al.*, 1999).

Figure 3 shows a ^{20}Ne/^{22}Ne *vs.* ^{21}Ne/^{22}Ne diagram. The data point of the 800°C

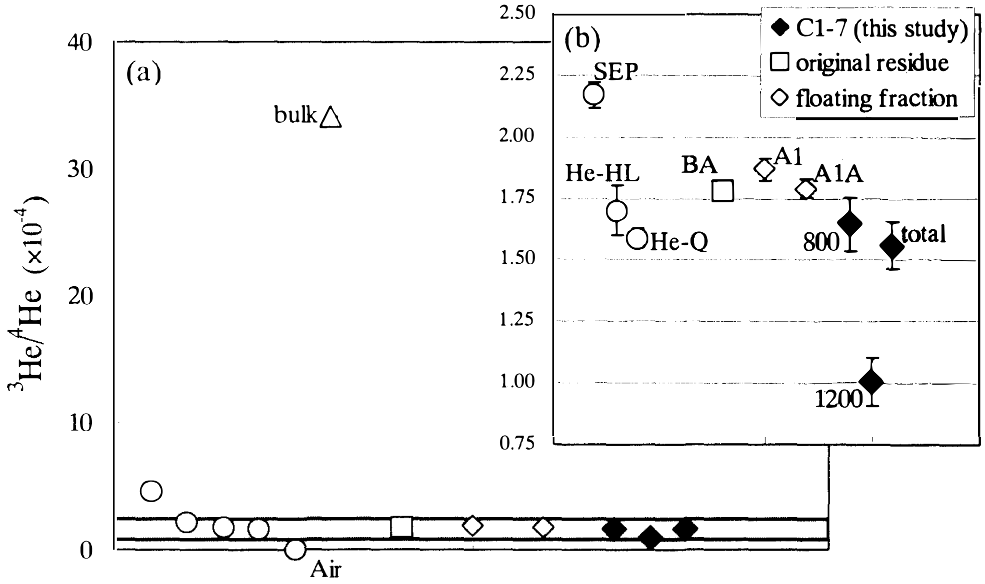

Fig. 2. *(a) A ^{3}He/^{4}He diagram and (b) its enlarged one of the rectangular portion. The data sources of separated samples are as same as those in Fig. 1. Others are as follows; Solar Wind and SEP (solar energetic particles) (Benkert et al., 1993), Atmosphere (Ozima and Podosek, 1983), He-HL (Huss and Lewis, 1994) and He-Q (Wieler et al., 1991). The assigned error includes 1σ error at the measurement. The 1600°C fraction of C1-7 is omitted because the amount of He gas extracted at 1600°C was small and cannot be measured. The ^{3}He/^{4}He ratio of C1-7 is similar to those of He-Q or He-HL. In this diagram, C1-7 is free from atmospheric component.*

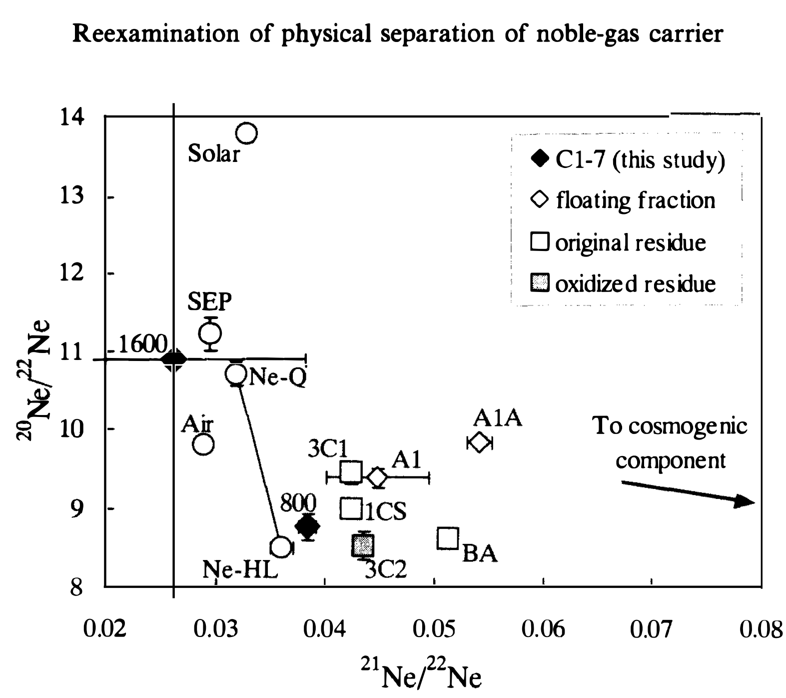

Fig. 3. *A $^{20}Ne/^{22}Ne$ vs. $^{21}Ne/^{22}Ne$ diagram. The numerical values near the data symbols show extraction temperatures (°C) for each step. Ne-HL is from Huss and Lewis (1994) and Ne-Q is from Wieler et al. (1991). The rest of the data sources are as same as those in Fig. 2. The assigned error includes 1σ error at the measurement and the conservative 100% error by the blank. We omitted the 1200°C fraction, for the amount of ^{21}Ne is too small to give the precise isotopic ratio. The 800°C fraction of C1-7 is similar to Ne-HL.*

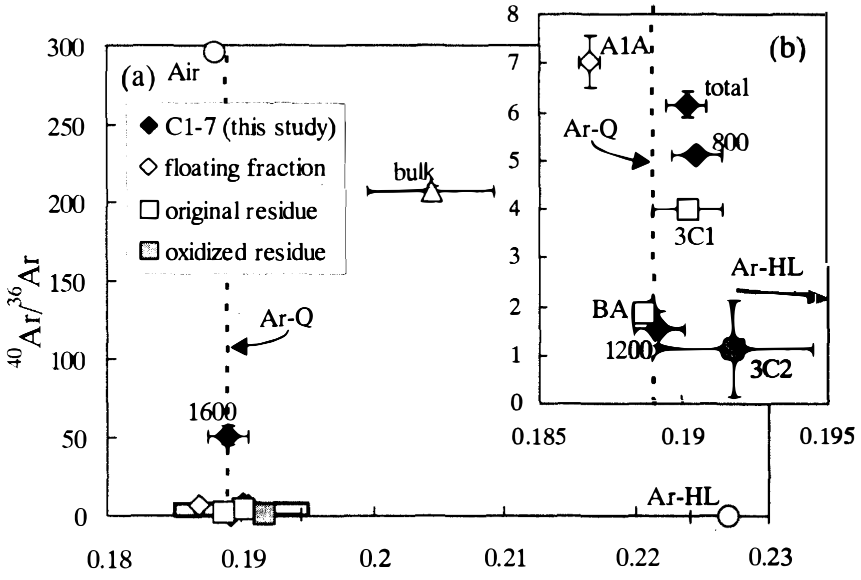

Fig. 4. *(a) A $^{40}Ar/^{36}Ar$ vs. $^{38}Ar/^{36}Ar$ diagram and (b) its enlarged one of the rectangular portion. The numerical values near the data symbols show extraction temperatures (°C) for each step. Argon-HL is from Huss and Lewis (1994) and the $^{38}Ar/^{36}Ar$ ratio of Ar-Q is shown by a dotted line (Wieler et al., 1991). The Ar-Q is at the lower side on this line (the $^{40}Ar/^{36}Ar$ ratio is probably below 0.1), although the exact $^{40}Ar/^{36}Ar$ ratio of Ar-Q is not determined (Huss et al., 1996). The rest of the data sources are as same as those in Fig. 2. The assigned error includes 1σ error at the measurement and the conservative 100% error by the blank.*

fraction of C1-7 is situated close to Ne-HL, showing a slight shift to the right side from the mixing line between Ne-Q and Ne-HL. The rightward shift suggests the addition of the cosmogenic Ne. Therefore, the data shows that Ne in C1-7 consists of mainly Ne-HL and small amounts of Ne-Q and cosmogenic Ne. Compared to neon observed in A1, A1A and chemical residues, our sample C1-7 has less cosmogenic effect judged from its low $^{21}Ne/^{22}Ne$ ratio than others.

A three-isotope plot of Ar is shown in Fig. 4. Although the $^{40}Ar/^{36}Ar$ ratio of Ar-Q is not well determined, it is supposed to be below 0.1 (Huss *et al.*, 1996). The low $^{40}Ar/^{36}Ar$ ratio in the total fraction of C1-7 seems to be compatible with that C1-7 contains Q or HL. The $^{38}Ar/^{36}Ar$ ratios of C1-7 are very close to that of Ar-Q rather than that of Ar-HL. The 1600°C fraction of C1-7 is higher on the diagram because of atmospheric contamination from the reaction between a molybdenum crucible and sample and/or a radiogenic component from the residual silicate in C1-7. As we will be shown later, the $^{129}Xe/^{132}Xe$ ratio of 1600°C fraction does not support the atmospheric contamination. Thus, contribution from the radiogenic component is more likely explanation. Figure 4 indicates that Ar-Q should be dominant in Ar of floating fraction C1-7.

Figure 5 shows a $^{86}Kr/^{84}Kr$ *vs.* $^{82}Kr/^{84}Kr$ diagram. This diagram also shows that Kr in C1-7 is almost identical to Kr-Q. The data points of the original residues are also located near that of Kr-Q, whereas that of oxidized residue shifts towards

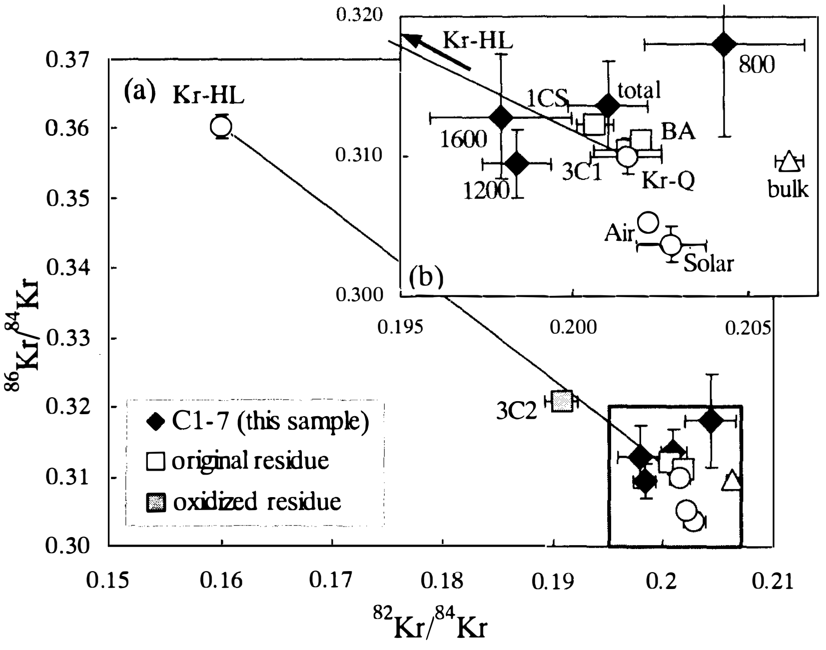

Fig. 5. (a) A $^{86}Kr/^{84}Kr$ vs. $^{82}Kr/^{84}Kr$ diagram and (b) its enlarged one of the rectangular portion. The numerical values near the data symbols show extraction temperatures (°C) for each step. The data sources are as follows; bulk Allende (Srinivasan et al., 1978), Solar Wind and SEP (Wieler and Baur, 1994) and Kr-Q (Wieler et al., 1992). The rest of the data sources are as same as those in Fig. 2. The assigned error includes 1σ error at the measurement and the conservative 100% error by the blank. Kr in C1-7 is almost identified with Kr-Q.

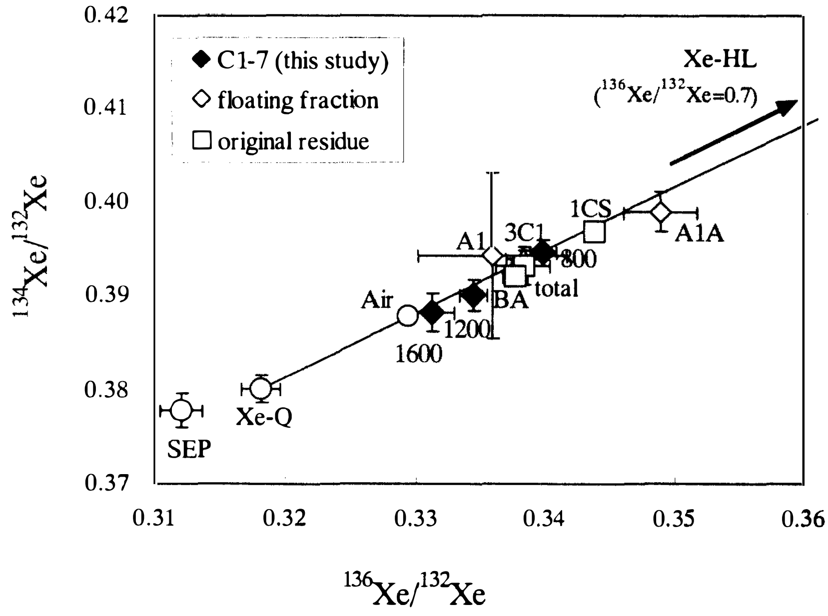

Fig. 6. A ^{134}Xe/^{132}Xe vs. ^{136}Xe/^{132}Xe diagram. The numerical values near the data symbols show extraction temperatures (°C) for each step in C1-7 measurements. As to A1 and A1A, only the total data were shown. Xe-HL and Xe-Q are from Huss and Lewis (1994). The rest of the data sources are as same as those in Fig. 2. The assigned error includes 1σ error at the measurement. All data of samples from Allende lie on the mixing line between Xe-Q and Xe-HL. The total fraction of C1-7 is very similar to that of original residues. The Xe-Q/Xe-HL ratio of C1-7 is different from that of A1A, suggesting that Q- and HL-components might be physically separable.

Kr-HL. The floating fractions are similar to the original residues also for the isotopic compositions of Kr.

Figure 6 shows a ^{134}Xe/^{132}Xe *vs.* ^{136}Xe/^{132}Xe diagram. The data points of C1-7 lies on the mixing line defined by Xe-Q and Xe-HL, close to the original residues. This also indicates that noble gases in the floating fraction C1-7 are very similar to those in the original residues. There is a systematic increase in the Xe-Q/Xe-HL ratios as the extraction temperature increases, which implies that the Q component is released at higher temperature than the HL component. This is compatible with the data of A1A, and also with previous experiments where Xe-HL degassed at 800°C to 1000°C whereas Xe-Q was released at 1000°C to 1200°C (*e.g.*, Srinivasan *et al.*, 1978; Huss *et al.*, 1996). The Xe-Q/Xe-HL ratio in the total fraction of C1-7 is close to that of A1, but is different from that of A1A. Thus, we could confirm the heterogeneity of the Q/HL ratios in the floating fractions.

Figure 7 shows a ^{129}Xe/^{132}Xe *vs.* ^{136}Xe/^{132}Xe diagram. It is interesting that Xe data points of the present sample C1-7 are different from those of the previous ones (A1 and A1A). All the temperature fractions of A1 and A1A lie on the mixing line between Xe-Q and Xe-HL, whereas those of C1-7 do not, and are situated at the upper side from the line with increasing the extraction temperatures, indicating the presence of ^{129}Xe excess. Xenon-129 was produced by the decay of the extinct nuclide ^{129}I (half life$=1.57\times10^{7}$ years). Original residues also had large ^{129}Xe

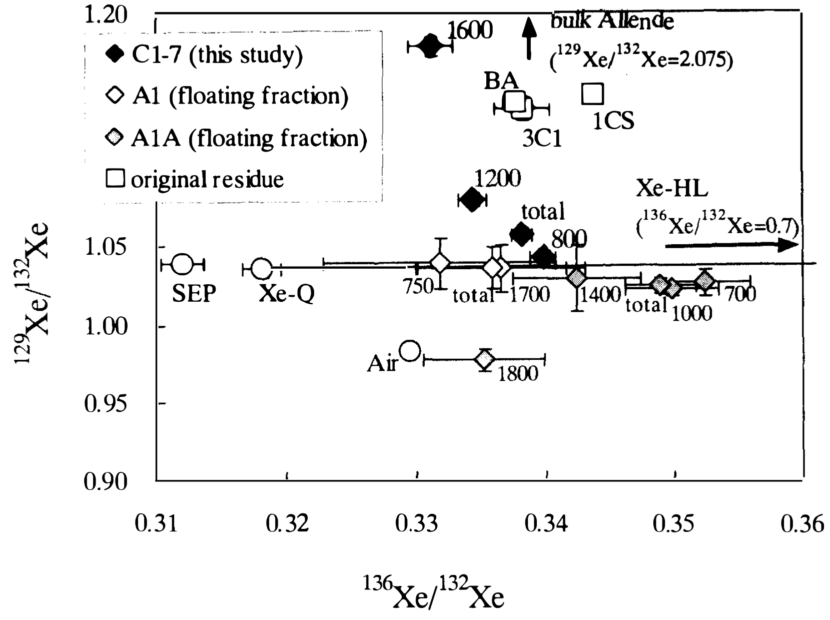

Fig. 7. A $^{129}Xe/^{132}Xe$ vs. $^{136}Xe/^{132}Xe$ diagram. The numerical values near the data symbols show extraction temperatures (°C) for each step. The data sources are as same as those in Fig. 6. The assigned error includes 1σ error at the measurement. The data of C1-7 are above the mixing line. The higher the extraction temperature is, the larger the ^{129}Xe excess becomes. The sample C1-7 has the ^{129}Xe excess, indicating the presence of the silicate phase.

excesses, which can be explained by the re-adsorption of ^{129}Xe on the surface of the residues from dissolved minerals like sodalite during the chemical treatment (Srinivasan *et al.*, 1978). The ^{129}Xe excess of the total fraction is much smaller than in the chemical residue. Re-adsorption of ^{129}Xe cannot be applied to the floating fraction because it was separated by the physical method. The ^{129}Xe excess in C1-7 is most likely due to the contamination of silicate phase like sodalite. This silicate phase could be also the source of radiogenic ^{40}Ar. This is because K and I can occupy the position of Na and Cl, respectively, which are the major constituent elements of sodalite.

5. Conclusions

We applied the freeze-thaw disaggregation to the Allende meteorite and obtained the black fine material, which floated on the water. The floating fraction contained noble gases with very similar elemental ratios and isotopic compositions to those of the original residue isolated by chemical treatment with HCl, HF, and CS_2 (Lewis *et al.*, 1975, 1977). This residue is very similar to the one recovered previously by Matsuda *et al.* (1999). Thus, we confirm that a physical separation method using freeze-thaw disaggregation is a useful method for isolation the gas-rich fraction of carbonaceous chondrites. There were differences between the present results and previous ones (Matsuda *et al.*, 1999). The He and Ne data in this study showed less cosmogenic signature compared to those in previous samples prepared in the same manner. Another difference is the presence of ^{40}Ar and ^{129}Xe excesses in our sample. This is probably due to the silicate phase like sodalite in our sample. As we stated above in the discussion of Xe isotopic ratios, we also confirmed that Xe-Q/Xe-HL ratios vary among the different floating fractions. This suggests that the two carriers, phase Q and presolar diamond, can be separated by some means, although it has never been accomplished.

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References

Benkert, J.-P., Baur, H., Signer, P. and Wieler, R. (1993): He, Ne, and Ar from the solar wind and solar energetic particles in lunar ilmenites and pyroxenes. J. Geophys. Res., **98**, 13147–13162.

Huss, G. R. and Lewis, R. S. (1994): Noble gases in presolar diamonds I: Three distinct components and their implications for diamond origins. Meteoritics, **29**, 791–810.

Huss, G. R., Lewis, R. S. and Hemkin, S. (1996): The "normal planetary" noble gas component in primitive chondrites: Compositions, carrier, and metamorphic history. Geochim. Cosmochim. Acta, **60**, 3311–3340.

Lewis, R. S., Srinivasan, B. and Anders, E. (1975): Host phase of a strange xenon component in Allende. Science, **190**, 1251–1262.

Lewis, R. S., Gros, J. and Anders, E. (1977): Isotopic anomalies of noble gases in meteorites and their origines 2. Separated minerals from Allende. J. Geophys. Res., **82**, 779–792.

MacPherson, G. J., Bar-Matthews, M., Tanaka, T., Olsen, E. and Grossman, L. (1983): Refractory inclusions in the Murchison meteorite. Geochim. Cosmochim. Acta, **47**, 823–840.

Maruoka, T. and Matsuda, J. (1995): Noble gas separation by adsorptive phenomena in mass spectrometry. J. Mass Spectrom. Soc. Jpn., **43**, 1–8.

Matsuda, J., Amari, S. and Nagao, K. (1999): Purely physical separation of a small fraction of the Allende meteorite that is highly enriched in noble gases. Meteorit. Planet. Sci., **34**, 129–136.

Ozima, M. and Podosek, F. A. (1983): Noble Gas Geochemistry. Cambridge, Cambridge Univ. Press, 367 p.

Srinivasan, B., Lewis, R. S. and Anders, E. (1978): Noble gases in the Allende and Abee meteorites and a gas-rich mineral fraction: investigation by stepwise heating. Geochim. Cosmochim. Acta, **42**, 183–198.

Wieler, R., Anders, E., Baur, H., Lewis, R. S. and Signer, P. (1991): Noble gases in "phase Q": Closed-system etching of an Allende residue. Geochim. Cosmochim. Acta, **55**, 1709–1722.

Wieler, R., Anders, E., Baur, H., Lewis, R. S. and Signer, P. (1992): Characterisation of Q-gases and other noble gas components in the Murchison meteorite. Geochim. Cosmochim. Acta, **56**, 2907–2921.

Wieler, R. and Baur, H. (1994): Krypton and xenon from the solar wind and solar energetic particles in two lunar ilmenites of different antiquity. Meteoritics, **29**, 570–580.

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