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An attempt to separate Q from the Allende meteorite by physical methods

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Abstract—In order to characterize the planetary noble gas carrier Q, we separated a Q-rich floating fraction from the Allende meteorite into ten fractions by a combination of colloidal and density separations. All five noble gases in the separated fractions were analyzed by pyrolysis in 600 and 1600°C temperature steps. Half of Q in the floating fraction is concentrated in the fraction C1-8D with the density of $1.65 \pm 0.04 \text{ g/cm}^3$. All the separated fractions show similar isotopic ratios except for $^{40}\text{Ar}/^{36}\text{Ar}$ ratios. C1-8D has the lowest $^{38}\text{Ar}/^{36}\text{Ar}$ and $^{40}\text{Ar}/^{36}\text{Ar}$ ratios (0.18784 ± 0.00020 and 4.36 ± 0.15 , respectively) in the 1600°C fraction, confirming that the fraction is enriched in Q. Most grains in C1-8D are carbonaceous with small amounts of F and O. These results imply either that the density of Q is $1.65 \pm 0.04 \text{ g/cm}^3$ or that Q preferentially sticks to matter of that density. All the separates have similar Q to diamond ratios, indicating that Q and diamond are closely associated. Copyright © 2003 Elsevier Ltd

1. INTRODUCTION

Trapped noble gases, or so-called planetary noble gases, are commonly observed in primitive meteorites. It was, however, only after Lewis et al. (1975) dissolved the Allende meteorite and measured noble gases of the residues that it was realized that the noble gases reside in a very small portion of the meteorites. After removing silicates, Lewis et al. (1975) obtained a residue, which comprised ~0.5% of the meteorite, and found that the concentrations of all five noble gases in the residue were > 2 orders of magnitude higher than that of the bulk meteorite and that most of the trapped noble gases of the bulk meteorite were preserved in the residue. They further processed the residue with HNO_3 , which resulted in the weight loss of only 6% or so, and measured noble gases in the oxidized residue (~0.47% of the bulk). Concentrations of the light noble gases (He and Ne) were almost the same as those in the original residue, while a major part of the heavy noble gases (Ar, Kr and Xe) was gone. This implies that a very small portion of the meteorite carries most of the heavy noble gases and that it is readily destroyed by oxidants. They dubbed the carrier of the trapped noble gases Q for “quintessence.” Xenon in the further processed residue was enriched in both light (124 and 126) and heavy (134 and 136) isotopes (Xe-HL). Later, the carrier of Xe-HL and most of the lighter noble gases in the residue has been identified as diamond (Lewis et al., 1987). The discovery that only a very small portion of the meteorites contains most of the noble gases prompted the search for carriers of the anomalous noble gas components, which ultimately led to the discovery of presolar grains in meteorites and opened up a new field of astronomy (see Zinner, 1998).

Lewis et al. (1975) initially proposed that Q was an unknown CrFe-rich mineral. Later, carbonaceous matter became a more probable candidate (Phinney et al., 1976; Reynolds et al., 1978;

Ott et al., 1981). Indeed, the enrichment in the heavier noble gases in Q relative to solar is what is expected from adsorption and carbon is known to be very efficient in adsorbing gases (Wacker et al., 1985). Moreover, a very small mass of Q could be explained if Q gases were located on or very near the surface and only the surface of grains was etched. Wacker et al. (1985) simulated adsorption of Xe on carbon and suggested that the noble gases were trapped in a labyrinth of micropores on the surface of carbon grains. The main difficulty of this model is to explain the high release temperature of Q gases (1000–1400°C) by pyrolysis (Srinivasan et al., 1978) because adsorbed noble gases are released at much lower temperatures (Wacker et al., 1985). Recently, based on the similarity between the elemental noble gas abundance in Q and that in ureilites, Matsuda and Yoshida (2001) proposed that the Q gases were implanted into the surface of diamonds via glow discharge (plasma model).

As Q has never been isolated as a mineral phase, isotopic and elemental compositions of Q gases have been determined from the difference in noble gas abundances of residues before and after destroying Q by oxidants (Lewis et al., 1975; Huss et al., 1996). Huss et al. (1996) prepared HF-HCl resistant residues from 14 relatively unmetamorphosed chondrites from 7 chondrite classes. They found that Q gases, or P1 as they named it, were present in all meteorites they examined, indicating that Q gases were a ubiquitous component in the early solar system, and that the P1 abundance decreased with increasing metamorphic grade. Wieler et al. (1991, 1992) developed the closed-system stepped etching (CSSE) technique, where a sample is exposed to HNO_3 at increasing temperatures in a vacuum system and the released gases are measured at each step. With this technique, the Q gases are directly analyzed as a function of etching depth. Busemann et al. (2000) examined HF-HCl resistant residues of six chondrites with this technique and concluded that the isotopic compositions of Q gases were uniform among the different types of chondrites except for $^{20}\text{Ne}/^{22}\text{Ne}$. Elemental abundances provided a hint that Q consists of at least two carbonaceous carrier phases “Q₁” and “Q₂” with slightly distinct chemical properties.

As described above, isotopic and elemental abundances of Q

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gases have been studied in detail and are well determined, while Q, the phase itself, remains elusive. One thing that has been widely accepted is that Q is carbonaceous matter. Verchovsky et al. (2002) examined HF-HCl treated residues from three carbonaceous chondrites, one ordinary chondrite and three enstatite chondrites, by stepped combustion. They found that the peak release of ^{36}Ar was $\sim 450^\circ\text{C}$ for the residues of all types of chondrites, indicating that Q was combusted at this temperature in all types of chondrites and was not altered by the thermal metamorphism that the enstatite chondrites had experienced. However, the major release peaks of C of the enstatite chondrites were shifted to higher temperatures compared to those of carbonaceous chondrites ($400\text{--}450^\circ\text{C}$ for the carbonaceous chondrites and $600\text{--}800^\circ\text{C}$ for the enstatite chondrites). This indicates that Q is a very minor carbonaceous component in the meteorites (also see Schelhaas et al., 1990) and was hardly affected by thermal alteration, while the majority of carbon was altered, possibly graphitized (Verchovsky et al., 2002), leading to higher combustion temperatures. This implies that Q is a thermally rather stable compound compared with other forms of carbon in meteorites.

Until recently Q-rich fractions were always prepared by removing silicates with an HF-HCl mixture from bulk meteorites. Matsuda et al. (1999) succeeded for the first time to concentrate Q by only physical means. They disaggregated a chunk of the Allende meteorite with the freeze-thaw method and recovered a fraction that floated on the surface of distilled water. This "floating fraction" has isotopic and elemental noble gas compositions and concentrations that are very similar to those of HF-HCl resistant residues. One notable difference between floating fractions and chemically prepared residues is that $^{129}\text{Xe}/^{132}\text{Xe}$ ratios of the former (1.02–1.06) (Matsuda et al., 1999; Zaizen et al., 2000) are much lower than those of the latter (1.15) (Srinivasan et al., 1978). It has been explained by either the floating fractions being devoid of I-rich minerals or re-adsorption of ^{129}Xe during chemical separation (Srinivasan et al., 1978), but not during physical separation (Matsuda et al., 1999).

In this study we have made an attempt to further separate a floating fraction by only physical means to enrich and ultimately isolate Q. Preliminary results were reported by Amari et al. (2001).

2. EXPERIMENTAL PROCEDURES

2.1. Sample Preparation

We have chosen the Allende meteorite for this study for two reasons. First, since the Allende meteorite is the only meteorite that the physical separation method has been applied to, we have several sets of noble gas data of floating fractions to compare with our new results (Matsuda et al., 1999; Zaizen et al., 2000). In addition, C and N isotopic ratios of carbonaceous grains from the Allende floating fraction A1 have been analyzed (Amari and Matsuda, 1998). Second, among carbonaceous chondrites, which contain abundant trapped noble gases, the C concentration in the Allende meteorite (CV) is low (0.3%) (King et al., 1969) compared with those in C1 and CM carbonaceous chondrites (2–4%) (Wasson, 1985). In addition, most C in the meteorite is present as inorganic C rather than as organic polymer (Breger et al., 1972; Vis et al., 2002). Since Q is easily destroyed by oxidants but we are not able to use oxidants to remove organic matter that hinders a clear separation, the less organic matter that is present, the better the chances are to separate Q.

The freeze-thaw method was applied to disaggregate 3.846 g of the

Allende meteorite. The sample was put in distilled water, frozen for 90 min and ultrasonicated in a water bath for 40 min. After 141 cycles of freezing and ultrasonication, 2550 μg of material that floated on the surface of the water was recovered. Three hundred sixty and 230 μg of the floating fraction were named C1-6 and C1-7, respectively. Isotopic and elemental compositions of noble gases of C1-7 (Zaizen et al., 2000) were similar to those of previously separated floating fractions (Matsuda et al., 1999), indicating that C1-7 was enriched in Q and diamond. The bulk Allende in the water was further processed for 75 more cycles and yielded 70 μg of an additional floating fraction.

We combined C1-6 and the 70 μg of additional material and named it C1-8 (total of 430 μg) and processed it according to the outline in Figure 1. In the separation procedure of presolar grains (Amari et al., 1994), diamond is usually extracted after mild oxidation that removes reactive kerogen and before density separation that extracts graphite. Since we did not apply any chemicals, let alone oxidants to C1-8, the first step was an attempt to separate diamond. Diamond extraction is usually done by keeping diamond in suspension in a basic solution (colloidal separation) (Tang and Anders, 1988). Instead of pure basic solutions a mixture of isopropanol and NaOH was used. Since the sample floated on the surface of water, it was necessary to lower the surface tension of the sample so that the sample was suspended in the liquid. To determine the optimum proportion of isopropanol and NaOH solution, several mixtures of various proportions were tested on a chemically isolated diamond fraction from the Allende meteorite. In the end, a mixture of 20 vol.% isopropanol and 80 vol.% 0.003 N NaOH solution was used because stronger basic solution tended not to mix thoroughly with isopropanol. The colloidal suspension was later acidified with HCl to coagulate material in the supernatant, yielding C1-8A.

The residue C1-8B was separated into nine density fractions in a density gradient of sodium polytungstate (NaPT) ($\text{Na}_6[\text{H}_2\text{W}_{12}\text{O}_{40}]$) (Table 1). Approximately 0.5 wt. % of surfactant Brij 35 was added to NaPT to prevent coagulation. Most of the sample was concentrated in the density range between 1.5 and 1.8 g/cm^3 . The separation was repeated several times for the density range to achieve a clear separation. The sample formed six distinct layers (C1-8C–C1-8H) and densities of the separated fractions were determined by weighing 100 μl of the heavy liquid extracted with a micropipette, where the accuracy of the volume was estimated to be 0.8%. This was repeated a few times and averages of the measured densities were obtained (Table 1). It did not seem that much material was present in the density range between 1.1 and 1.6 g/cm^3 (C1-8J), and between 2.2 and 2.3 g/cm^3 (C1-8K). Nonetheless, these two fractions were processed in the same way as the rest of the separates.

Two small fractions named C1-8Ib and C1-8Kb were produced during washing out of NaPT from C1-8I and C1-8K. Their weights comprise only 3% of the sum of the separates (excluding C1-8Ib and C1-8Kb), while ^{132}Xe concentrations of C1-8Ib and C1-8Kb are only 12 and 20% of their parent fractions, indicating a significant part of the two fractions is a contaminant introduced during the washing. As a result, they together carried < 0.3% of the sum of ^{132}Xe of the separates (excluding C1-8Ib and C1-8Kb). Thus, for calculations of a mass balance and noble gas concentrations, we omitted these two minor fractions.

The total mass of all fractions is 431 μg if C1-8Ib and C1-8Kb are included, and 418 μg if they are excluded, implying at least 97% of the 430 μg of starting material was retained during the separation procedure.

2.2. Noble Gas Analysis

We used half of each fraction for noble gas analysis and saved the other half for other experiments and future study. Each fraction was suspended in isopropanol and transferred into a Pt container (15 mg in weight and 3 mm in diameter) by a syringe with a Teflon tube attached to the syringe needle. Then the Pt containers were placed on a hot plate at $\sim 80^\circ\text{C}$ to evaporate the isopropanol. Weights of the samples were obtained by a micro-balance as the difference of the weight after and before the sample loading. Weights of the fractions were calculated from the averages of five measurements with typical errors of 0.2 μg (Table 1).

Elemental and isotopic compositions of all noble gases were measured using the VG5400 noble gas mass spectrometer installed at

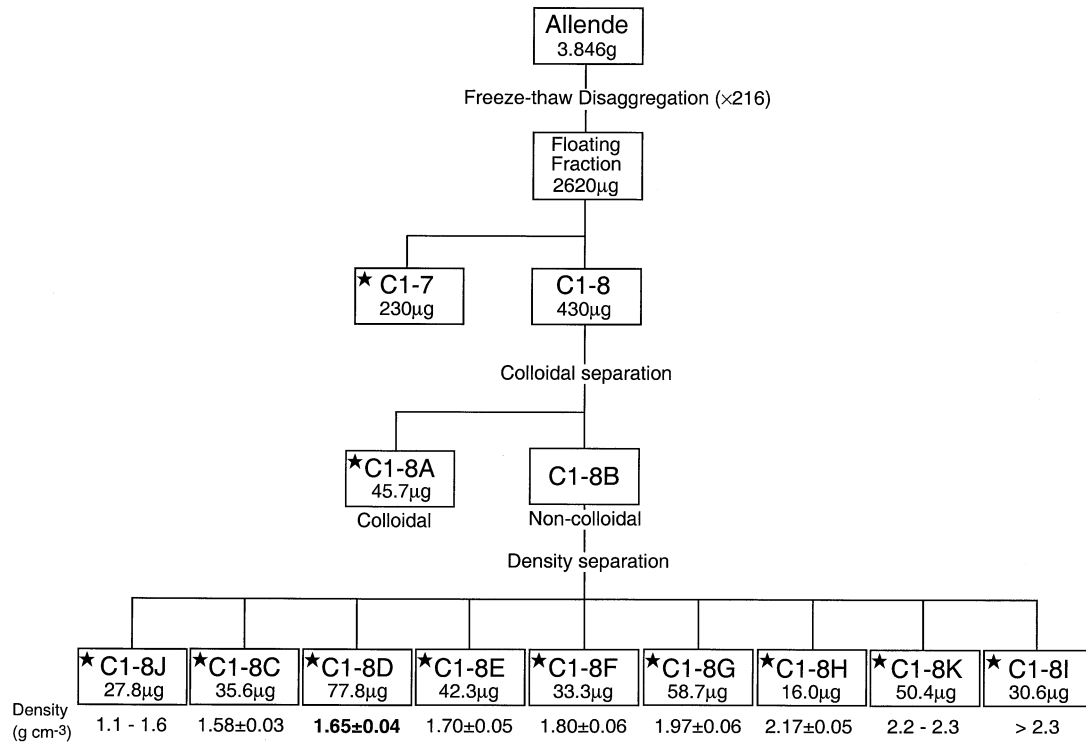


Fig. 1. Separation procedure of the floating fraction C1-8 from the Allende meteorite. Ten fractions were obtained by colloidal and density separations. Fractions marked by a star were analyzed for noble gases.

Osaka University. We extracted the gas by stepwise heating at 600 and 1600°C. A detailed description of gas purification and the experimental procedure is given by Wada and Matsuda (1998) and Zaizen et al. (2000). The extracted gases were purified in two stages by Ti-Zr getters and separated into five fractions using a cold trap composed of sintered stainless steel (Maruoka and Matsuda, 1995). Hot blanks of the two temperature steps were measured before analyzing each separate. The hot blanks of He, Ar, Kr and Xe showed no systematic variation during the measurement period while the Ne blank progressively improved as the analyses proceeded. Thus we used the average values of all hot blanks during the measurement period for He, Ar, Kr and Xe measurements and included the standard deviations as uncertainties into the calculation, and used the hot blank values, with uncertainties due to the

extrapolation to the time when gas was admitted to the mass spectrometer, measured just before the individual sample analyses for Ne. Typical hot blanks at 1600°C were 8.2×10^{-10} cm³ STP, 1.2 to 2.1×10^{-13} cm³ STP, 2.1×10^{-12} cm³ STP, 2.2×10^{-13} cm³ STP and 2.1×10^{-14} cm³ STP for ⁴He, ²²Ne, ³⁶Ar, ⁸⁴Kr and ¹³²Xe, respectively. Blanks at 600°C were about half of those at 1600°C.

⁴⁰Ar²⁺ and ²⁰Ne⁺ were partly separable in our mass spectrometer, and we chose the peak position of the flat peak of ²⁰Ne⁺ that was free from the interference of ⁴⁰Ar²⁺. The correction of CO₂²⁺ to ²²Ne⁺ ranged from 5% (C1-8D) to 70% (C1-8C), whereas that of ⁴⁰Ar₂⁺ to ⁸⁰Kr⁺ was < 0.2% (except for 0.6% for the 600°C fraction of C1-8E). The sensitivity and mass discrimination were determined by analyzing pipetted air except for the mass discrimination of He for which an

Table 1. Fractions separated from C1-8.

Fraction ^a	Density (g/cm ³) ^b	Weight (µg)	Weight fraction (%)	¹³² Xe fraction (%) ^c	Sample for noble gas analysis (µg)
C1-8A	Colloidal	45.7	10.93	11.4	22.8
C1-8C	1.58 ± 0.03	35.6	8.52	1.2	17.2
C1-8D	1.65 ± 0.04	77.8	18.61	48.3	36.3
C1-8E	1.70 ± 0.05	42.3	10.12	12.3	20.6
C1-8F	1.80 ± 0.06	33.3	7.97	7.7	15.9
C1-8G	1.97 ± 0.06	58.7	14.03	5.1	29.1
C1-8H	2.17 ± 0.05	16.0	3.82	0.7	8.3
C1-8I	>2.3	30.6	7.31	1.8	15.1
C1-8J	1.1-1.6	27.8	6.64	5.6	13.6
C1-8K	2.2-2.3	50.4	12.06	5.9	23.6

^a There are minor fractions C1-8Ib and C1-8Ib, weighting 6.1 and 6.3 µg, respectively. They are not included in mass balance and noble gas calculations.

^b The errors of the densities are standard deviations of the means of several measurements.

^c Values are relative to the sum of ¹³²Xe in all separated fractions.

Table 2. Helium, Ne and Ar in C1–8 separates, Q diamond and the air.

Sample	Temp (°C)	[⁴ He] ×10 ⁻⁶	³ He/ ⁴ He ×10 ⁻⁴	[²⁰ Ne] ×10 ⁻⁸	²⁰ Ne/ ²² Ne	²¹ Ne/ ²² Ne	[³⁶ Ar] ×10 ⁻⁸	³⁸ Ar/ ³⁶ Ar	⁴⁰ Ar/ ³⁶ Ar
C1–8A	600	16 ± 40		10 ± 26			114 ± 2	0.1819 ± 0.0026	22.5 ± 1.4
	1600	829 ± 56	1.55 ± 0.10	479 ± 25	8.60 ± 0.34	N.D.	1376 ± 27	0.18895 ± 0.00042	13.77 ± 0.55
	Total	845 ± 68		489 ± 36			1489 ± 27	0.18841 ± 0.00043	14.44 ± 0.52
C1–8C	600	13 ± 53		3 ± 46			43 ± 1	0.1676 ± 0.0090	41.9 ± 4.4
	1600	71 ± 69	1.31 ± 1.26	43 ± 27	8.9 ± 4.1	N.D.	169 ± 5	0.1909 ± 0.0037	79.0 ± 4.5
	Total	84 ± 86		46 ± 53			212 ± 5	0.1862 ± 0.0035	71.4 ± 3.7
C1–8D	600	23 ± 25		15 ± 15			206 ± 4	0.1862 ± 0.0012	14.50 ± 0.50
	1600	1868 ± 55	1.514 ± 0.030	1074 ± 29	8.63 ± 0.16	0.0351 ± 0.0011	3289 ± 64	0.18784 ± 0.00020	4.36 ± 0.15
	Total	1892 ± 61		1089 ± 32			3495 ± 64	0.18774 ± 0.00020	4.96 ± 0.15
C1–8E	600	14 ± 44		8 ± 20			103 ± 2	0.1797 ± 0.0032	26.4 ± 1.6
	1600	848 ± 61	1.50 ± 0.10	501 ± 25	8.69 ± 0.33	N.D.	1560 ± 31	0.18928 ± 0.00042	16.07 ± 0.54
	Total	862 ± 75		510 ± 32			1663 ± 31	0.18869 ± 0.00044	16.71 ± 0.51
C1–8F	600	13 ± 57		7 ± 68			80 ± 2	0.1760 ± 0.0053	79.0 ± 4.5
	1600	667 ± 76	1.39 ± 0.16	409 ± 26	8.88 ± 0.39	N.D.	1264 ± 25	0.18881 ± 0.00063	21.16 ± 0.84
	Total	680 ± 95		416 ± 73			1344 ± 25	0.18805 ± 0.00068	21.57 ± 0.81
C1–8G	600	5 ± 31		3 ± 9			33 ± 1	0.1926 ± 0.0067	38.1 ± 3.5
	1600	265 ± 41	1.46 ± 0.22	169 ± 12	8.66 ± 0.46	N.D.	521 ± 10	0.18933 ± 0.00074	38.6 ± 1.1
	Total	270 ± 52		171 ± 15			554 ± 10	0.18952 ± 0.00080	38.6 ± 1.0
C1–8H	600	20 ± 110		4 ± 40			25 ± 2		180.0 ± 6.8
	1600	90 ± 140	1.6 ± 2.7	121 ± 38	9.7 ± 2.1	N.D.	461 ± 12	0.1906 ± 0.0028	189.2 ± 2.0
	Total	110 ± 180		125 ± 55			486 ± 12		188.8 ± 1.9
C1–8I	600	12 ± 60		4 ± 17			46 ± 1	0.1695 ± 0.0098	34.5 ± 4.9
	1600	177 ± 78	1.97 ± 0.87	119 ± 19	8.1 ± 1.0	N.D.	373 ± 8	0.1929 ± 0.0019	94.1 ± 2.2
	Total	189 ± 99		122 ± 26			419 ± 9	0.1903 ± 0.0020	87.6 ± 2.0
C1–8J	600	5 ± 67		7 ± 24			76 ± 2	0.1757 ± 0.0066	23.8 ± 3.4
	1600	615 ± 88	1.30 ± 0.19	345 ± 23	8.37 ± 0.40	N.D.	1066 ± 21	0.18966 ± 0.00098	19.0 ± 1.2
	Total	620 ± 110		352 ± 33			1142 ± 21	0.1887 ± 0.0010	19.3 ± 1.1
C1–8K	600	5 ± 38		5 ± 14			53 ± 1	0.1777 ± 0.0053	38.0 ± 2.7
	1600	341 ± 51	1.29 ± 0.19	216 ± 12	8.44 ± 0.33	N.D.	684 ± 14	0.18958 ± 0.00069	42.30 ± 0.98
	Total	347 ± 64		220 ± 19			737 ± 14	0.18873 ± 0.00075	41.99 ± 0.93
C1–7	800	840	1.646 ± 0.105	580	8.76 ± 0.17	0.0383 ± 0.0008	1000	0.19053 ± 0.00087	5.09 ± 0.11
	1200	130	1.010 ± 0.098	78	9.00 ± 0.50		280	0.18920 ± 0.00087	1.52 ± 0.36
	1600			13	10.89 ± 4.3	0.0263 ± 0.0120	53	0.1890 ± 0.0015	50.84 ± 6.06
	Total	970	1.561 ± 0.091	670	8.82 ± 0.17		1400	0.19019 ± 0.00069	6.14 ± 0.27
Q ^a			1.23–1.59		10.05–10.70	0.0291–0.0321		0.18727 ± 0.00070	
P1 ^b								0.18762 ± 0.00106	
HL ^c			1.70 ± 0.10		8.50 ± 0.06	0.036 ± 0.001		0.22676 ± 0.00309	
Air			0.0140		9.80	0.0290		0.188	295.5

Gas concentrations are in the unit of cm³ STP/g. Errors are 1 σ . Significant digits of gas concentrations are shown such that their errors are given to two digits.

^a Helium and Ne isotopic ratios have a range. Busemann et al. (2000).

^b Huss et al. (1996).

^c In case of Ne, ratios indicate those of Ne-A2(= HL + P6). Huss and Lewis (1994).

artificially mixed standard gas HESJ was used (³He/⁴He = 20.63 ± 0.10 R_a, where R_a is the air value 1.399 × 10⁻⁶; Matsuda et al., 2002).

The obtained data are listed in Tables 2 to 4. Because of the small signals, we could not get meaningful values for ⁷⁸Kr/⁸⁴Kr, ¹²⁴Xe/¹³²Xe, and ¹²⁶Xe/¹³²Xe ratios.

3. RESULTS

3.1. Noble Gases

The main purpose of having 600°C temperature fractions was to obtain Q gases as pure as possible in the 1600°C temperature fractions, because air and other loosely bound components were expected to be released at the lower temperature. The major portion of noble gases was released in the 1600°C fractions in most cases. Since the errors in the isotopic ratios of the 600°C fractions are large due to the small gas amounts, we will focus on the isotopic ratios of the 1600°C

fractions. We use total extracted amounts of noble gases for elemental abundances.

3.1.1. Elemental abundances

To determine whether we lost a significant amount of Q during the separation, we calculated noble gas abundances of the starting sample C1-8 from masses and noble gas abundances of all separated fractions and compared it with noble gas abundances of the floating fractions A1, A1A, (Matsuda et al., 1999) and C1-7 (Zaizen et al., 2000). Both C1-7 and C1-8 were taken from the same floating fraction (see Fig. 1), making C1-7 an appropriate sample to compare with our starting sample C1-8.

Concentrations of ³⁶Ar, ⁸⁴Kr, and ¹³²Xe of the sum of the C1-8 separates (hereafter called C1-8) are 1.40 × 10⁻⁵, 9.85 × 10⁻⁸, and 1.37 × 10⁻⁷ cm³ STP/g, respectively, and are

Table 3. Krypton in C1–8 separates, Q and the air.

Sample	Temp (°C)	[⁸⁴ Kr] × 10 ⁻¹⁰	⁸⁰ Kr/ ⁸⁴ Kr	⁸² Kr/ ⁸⁴ Kr × 100	⁸³ Kr/ ⁸⁴ Kr	⁸⁶ Kr/ ⁸⁴ Kr
C1–8A	600	107 ± 14				
	1600	717 ± 47	3.67 ± 0.20	19.22 ± 0.87	19.75 ± 0.82	29.72 ± 0.98
	Total	825 ± 49				
C1–8C	600	74 ± 15				
	1600	118 ± 22	N.D.	18.8 ± 4.5	18.7 ± 4.5	23.3 ± 4.1
	Total	192 ± 27				
C1–8D	600	245 ± 17	4.86 ± 0.32	18.25 ± 0.94	19.3 ± 1.1	33.6 ± 1.5
	1600	2140 ± 130	3.84 ± 0.07	19.28 ± 0.28	19.80 ± 0.21	30.80 ± 0.35
	Total	2390 ± 130	3.94 ± 0.07	19.17 ± 0.27	19.75 ± 0.22	31.09 ± 0.35
C1–8E	600	135 ± 16	4.92 ± 0.86	17.5 ± 2.7	22.5 ± 2.1	33.5 ± 4.3
	1600	1091 ± 72	3.93 ± 0.16	18.88 ± 0.63	20.65 ± 0.51	30.21 ± 0.55
	Total	1226 ± 74	4.04 ± 0.17	18.73 ± 0.63	20.85 ± 0.51	30.57 ± 0.68
C1–8F	600	130 ± 20				
	1600	895 ± 65	4.22 ± 0.26	18.74 ± 0.74	20.54 ± 0.63	32.12 ± 0.97
	Total	1025 ± 67				
C1–8G	600	182 ± 24				
	1600	354 ± 27	4.57 ± 0.31	18.9 ± 1.2	21.5 ± 1.0	30.68 ± 0.99
	Total	536 ± 35				
C1–8H	600	114 ± 33				
	1600	299 ± 48				
	Total	413 ± 58				
C1–8I	600	88 ± 18				
	1600	210 ± 28				
	Total	298 ± 34				
C1–8J	600	114 ± 20				
	1600	640 ± 52	3.90 ± 0.35	18.48 ± 0.84	20.6 ± 1.1	30.5 ± 1.2
	Total	754 ± 56				
C1–8K	600	79 ± 13				
	1600	464 ± 35	4.41 ± 0.44	21.25 ± 0.76	21.5 ± 1.0	31.8 ± 1.2
	Total	544 ± 38				
C1–7	800	330	3.829 ± 0.048	20.43 ± 0.23	20.42 ± 0.20	31.81 ± 0.66
	1200	330	3.646 ± 0.061	19.83 ± 0.10	20.20 ± 0.09	30.94 ± 0.25
	1600	81	3.449 ± 0.157	19.79 ± 0.20	19.97 ± 0.31	31.28 ± 0.44
	Total	750	3.707 ± 0.039	20.10 ± 0.11	20.27 ± 0.11	31.37 ± 0.32
Q ^a			3.937 ± 0.007	20.18 ± 0.02	20.18 ± 0.02	30.95 ± 0.05
P1 ^b			3.97 ± 0.03	20.24 ± 0.03	20.21 ± 0.05	30.91 ± 0.09
Kr-HL ^c			3.05 ± 0.10	15.90	19.89 ± 0.10	36.23 ± 0.18
Air			3.9599	20.217	20.136	30.524

Gas concentrations are in the unit of cm³ STP/g. Errors are 1 σ . Significant digits of gas concentrations are shown such that their errors are given to two digits.

^a Busemann et al. (2000).

^b Huss et al. (1996).

^c Huss and Lewis (1994).

similar to those of C1-7 (1.40×10^{-5} , 7.50×10^{-8} , and 1.40×10^{-7} cm³ STP/g). Since Q is enriched in the heavier noble gases relative to solar, these results indicate that Q was not lost during the separation. Concentrations of ⁴He and ²⁰Ne of C1-8 (⁴He: 7.32×10^{-4} cm³ STP/g, ²⁰Ne: 4.32×10^{-6} cm³ STP/g) are 60 to 70% lower than those of C1-7 (⁴He: 9.70×10^{-4} cm³ STP/g, ²⁰Ne: 6.70×10^{-6} cm³ STP/g). Since diamond is enriched in the lighter noble gases relative to solar, the low concentrations of He and Ne can be interpreted by a substantial amount of diamond being lost during the separation. However, the data show that all separated fractions have similar Q/diamond ratios: ²⁰Ne/¹³²Xe ratios of all separates are essentially the same within the errors, with an average of 34 ± 7 . Their Xe isotopic ratios also indicate a uniform Q/diamond ratio in all separates. If diamond was preferentially lost during the separation, it had to be lost before the colloidal separation (that is, before the separation started) or we need to assume that diamond was lost from all separates such that Q/diamond ratios in

all separates are the same. Otherwise, we would have separates that were enriched in Q relative to diamond.

More likely, the difference of the concentrations of the lighter noble gases in C1-7 and C1-8 is due to the heterogeneity of the meteorite or a slight difference of conditions during the freeze-thaw disaggregation or a combination of both. Matsuda et al. (1999) also found that ¹³²Xe-HL/¹³²Xe-Q in A1A was 1.7 times higher than that in A1, where A1 and A1A were taken from the same suite of the floating fraction. ²⁰Ne/¹³²Xe ratios, which can be an indicator of diamond to Q ratios, of both floating fractions and HF-HCl resistant residues from the Allende meteorite, vary from 20 to 48 (Matsuda et al., 1999; Zaizen et al., 2000), and 30 to 40, respectively (Lewis et al., 1975; Srinivasan et al., 1978; Wieler et al., 1991).

As discussed in the following subsection, the separated fractions show similar ¹³⁴Xe/¹³²Xe and ¹³⁶Xe/¹³²Xe ratios, indicating that they have similar Q/diamond ratios. Thus, their ¹³²Xe concentrations can be used as an indicator of Q abun-

Table 4. Xenon in C1–8 separates, Q and the air.

Sample	Temp (°C)	[¹³² Xe] × 10 ⁻¹⁰	¹²⁸ Xe/ ¹³² Xe	¹²⁹ Xe/ ¹³² Xe	¹³⁰ Xe/ ¹³² Xe × 100	¹³¹ Xe/ ¹³² Xe	¹³⁴ Xe/ ¹³² Xe	¹³⁶ Xe/ ¹³² Xe
C1–8A	600	105 ± 10						
	1600	1320 ± 120	8.30 ± 0.16	102.75 ± 0.61	16.02 ± 0.18	81.80 ± 0.52	40.50 ± 0.58	33.34 ± 0.26
	Total	1430 ± 120						
C1–8C	600	50 ± 5						
	1600	135 ± 13	7.7 ± 1.1	101.7 ± 5.0	16.4 ± 1.0	78.4 ± 5.1	38.1 ± 2.0	34.6 ± 2.6
	Total	185 ± 14						
C1–8D	600	218 ± 20	8.08 ± 0.40	101.2 ± 1.6	15.49 ± 0.48	79.0 ± 1.2	36.86 ± 0.71	30.16 ± 0.72
	1600	3330 ± 300	8.23 ± 0.09	103.75 ± 0.38	16.12 ± 0.13	81.77 ± 0.30	39.18 ± 0.30	34.01 ± 0.16
	Total	3550 ± 300	8.22 ± 0.09	103.59 ± 0.37	16.08 ± 0.12	81.60 ± 0.30	39.04 ± 0.28	33.77 ± 0.16
C1–8E	600	107 ± 10						
	1600	1560 ± 140	8.11 ± 0.15	103.2 ± 1.1	16.31 ± 0.18	83.08 ± 0.85	39.13 ± 0.43	34.47 ± 0.50
	Total	1660 ± 140						
C1–8F	600	94 ± 9						
	1600	1220 ± 110	8.42 ± 0.31	106.7 ± 1.2	16.68 ± 0.23	80.7 ± 1.3	39.94 ± 0.64	33.81 ± 0.42
	Total	1320 ± 110						
C1–8G	600	36 ± 3						
	1600	465 ± 42	8.80 ± 0.34	104.5 ± 1.7	15.98 ± 0.37	82.7 ± 1.7	38.60 ± 0.64	35.89 ± 0.64
	Total	501 ± 42						
C1–8H	600	39 ± 5						
	1600	197 ± 21	7.5 ± 2.1	103.7 ± 5.8	14.9 ± 1.8	91.4 ± 6.5	41.2 ± 3.5	38.6 ± 3.0
	Total	236 ± 21						
C1–8I	600	43 ± 4						
	1600	299 ± 27	7.59 ± 0.71	104.3 ± 2.9	15.0 ± 1.1	80.7 ± 2.7	41.8 ± 1.5	35.5 ± 1.3
	Total	341 ± 27						
C1–8J	600	87 ± 9						
	1600	1070 ± 97	8.32 ± 0.30	104.3 ± 2.0	16.33 ± 0.50	81.6 ± 1.1	39.20 ± 0.91	34.23 ± 0.94
	Total	1156 ± 97						
C1–8K	600	53 ± 5						
	1600	613 ± 54	8.16 ± 0.20	104.6 ± 1.4	16.60 ± 0.52	79.0 ± 1.4	39.37 ± 0.59	35.6 ± 1.1
	Total	666 ± 54						
C1–7	800	1000	8.467 ± 0.083	104.32 ± 0.40	16.14 ± 0.14	82.38 ± 0.98	39.46 ± 0.14	33.99 ± 0.10
	1200	340	8.410 ± 0.087	107.95 ± 0.41	16.30 ± 0.14	81.85 ± 0.94	39.00 ± 0.16	33.44 ± 0.11
	1600	59	8.439 ± 0.152	117.87 ± 0.63	16.24 ± 0.19	81.86 ± 1.03	38.82 ± 0.21	33.12 ± 0.18
	Total	1400	8.452 ± 0.063	105.76 ± 0.31	16.18 ± 0.11	82.23 ± 0.74	39.32 ± 0.11	33.82 ± 0.08
Q ^a			8.22 ± 0.02	104.2 ± 0.2	16.19 ± 0.03	81.85 ± 0.09	37.80 ± 0.11	31.64 ± 0.08
P1 ^b			8.30 ± 0.03	104.0 ± 0.2	16.28 ± 0.04	82.30 ± 0.12	37.77 ± 0.11	31.61 ± 0.10
Xe-HL ^c			9.05 ± 0.06	105.6 ± 0.2	15.42 ± 0.03	84.57 ± 0.13	63.56 ± 0.13	69.91
Air			7.136	98.32	15.136	78.90	38.79	32.94

Gas concentrations are in the unit of cm³ STP/g. Errors are 1 σ . Significant digits of gas concentrations are shown such that their errors are given to two digits.

^a Busemann et al. (2000).

^b Huss et al. (1996).

^c Huss and Lewis (1994).

dances. We calculated relative abundances of ¹³²Xe in the separated fractions from their ¹³²Xe concentrations and weights (Table 1 and Fig. 2). Forty-eight percent of the Q in the parent floating fraction is concentrated in C1-8D with a very narrow density range of 1.65 ± 0.04 g/cm³, because it has the highest ¹³²Xe concentration and the largest mass among the separated fractions. Relative abundances of Q decrease with increasing density in the next three heavier-density fractions. Eleven percent of the Q is in the colloidal fraction C1-8A, which is expected to be enriched in diamond.

3.1.2. Xenon isotopic ratios

¹³⁴Xe/¹³²Xe and ¹³⁶Xe/¹³²Xe ratios in all separates are very similar to one another and to those of the floating fractions (Fig. 3a). Since errors in the Xe isotopic ratios of C1-8H are huge, it is not shown in Figure 3 to avoid clutter. In Figures 3 to 5, isotopic ratios of Q gases obtained by Wieler et al. (1991, 1992)

and Busemann et al. (2000) are indicated as Q and those by Huss et al. (1996) as P1. The separates plot close to Xe-Q and cluster around the mixing line between Xe-Q and Xe-HL. This indicates that Xe in all separated fractions is enriched in Xe-Q with small amount of Xe-HL, and that the Q/diamond ratios are similar in all separates as well as the floating fractions. Fraction C1-8A is slightly above the mixing line but is indistinguishable to the other fractions within 2 σ errors. It is somewhat surprising that fraction C1-8A, which was expected to be enriched in diamond, is not different from the other fractions. This will be discussed in section 4.2. In a ¹²⁹Xe/¹³²Xe vs. ¹³⁶Xe/¹³²Xe plot (Fig. 3b), all of the separates except for C1-8A and C1-8F are on the mixing line between Xe-Q and Xe-HL. C1-8A is slightly below the mixing line, whereas C1-8F lies slightly above the line, barely missing the line, if 2 σ errors are considered.

¹³⁰Xe/¹³²Xe and ¹³¹Xe/¹³²Xe ratios of the separates are also close to those of Q and there are no significant differences

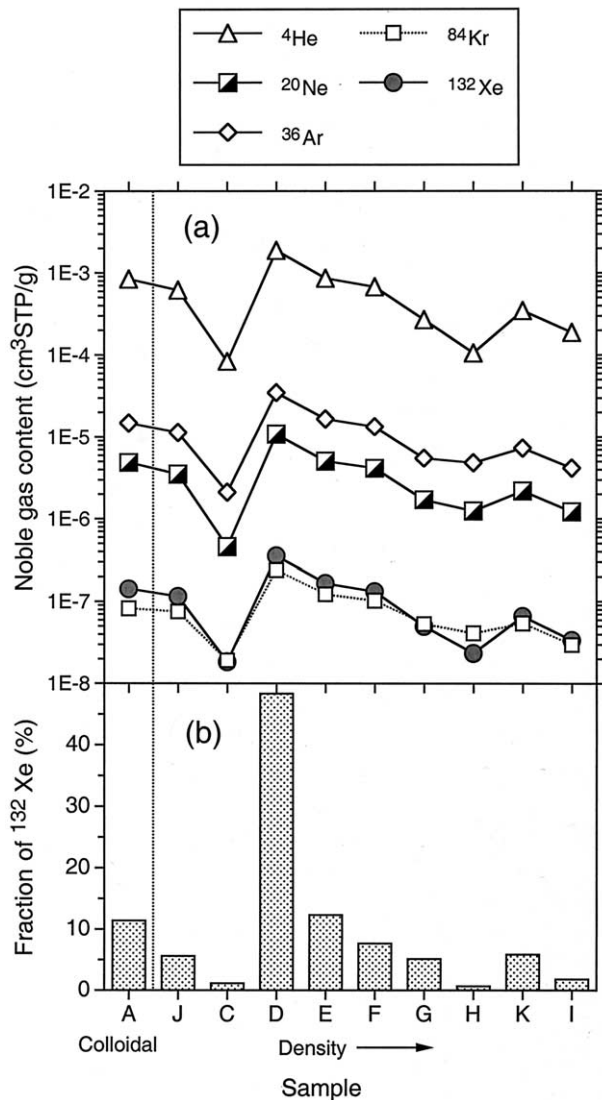


Fig. 2. (a) Noble gas concentrations of the C1-8 separates. (b) A histogram of abundances of ^{132}Xe in the C1-8 separates relative to the sum of ^{132}Xe in all separates. C1-8D with the density of $1.65 \pm 0.04 \text{ g/cm}^3$ carries half of the ^{132}Xe in C1-8. Note that the colloidal fraction C1-8A contains $> 10\%$ of the ^{132}Xe in C1-8, showing that it contains not only diamond but also Q. "C1-8" is omitted from the sample names.

among the C1-8 separates (Table 4). C1-8A and C1-8D, which have the smallest errors, are close to Xe-Q obtained by the closed-system stepped etching (CSSE) technique (Busemann et al., 2000) rather than that obtained by Huss et al. (1996).

3.1.3. Helium, Ne and Ar

$^{20}\text{Ne}/^{22}\text{Ne}$ ratios of all separated fractions are close to that of diamond (A2) (Fig. 4). Since diamond is enriched in He and Ne relative to Q, the light noble gases in the separates are dominated by those from presolar diamond. Again we do not see a difference among the separates including the colloidal fraction C1-8A. This confirms the conclusion, derived from the Xe isotopic ratios, that the fraction is not particularly enriched in diamond compared with the other separates. We were able to

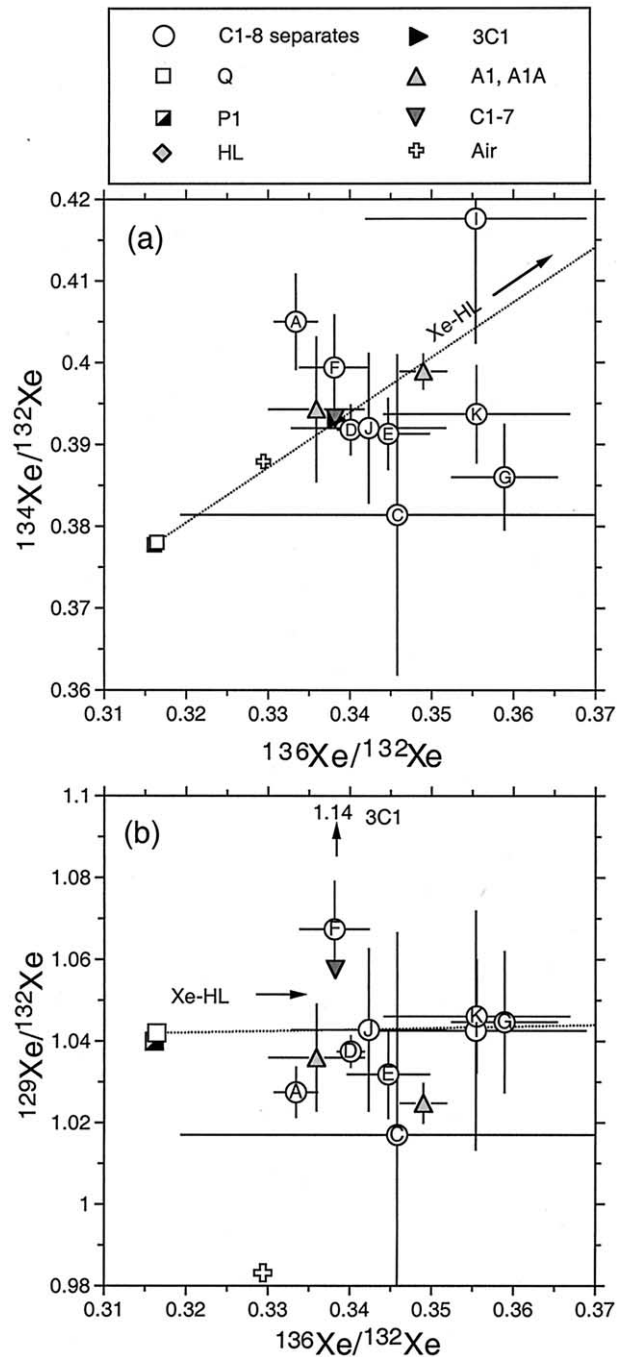


Fig. 3. (a) $^{134}\text{Xe}/^{132}\text{Xe}$ and $^{136}\text{Xe}/^{132}\text{Xe}$ ratios and (b) $^{129}\text{Xe}/^{132}\text{Xe}$ and $^{136}\text{Xe}/^{132}\text{Xe}$ ratios of the separates. In this figure and the subsequent figures, (1) errors are 1σ , (2) the ratios of the separates are those of the 1600°C temperature fractions, (3) plotted are total ratios of floating fractions, A1 and A1A (Matsuda et al., 1999) and C1-7 (Zaizen et al., 2000), and those of the HF-HCl resistant residue 3C1 (Lewis et al., 1975), (4) letters in the open circles indicate names of the separates. (5) Xe-Q determined by Wieler et al. (1991, 1992) and Busemann et al. (2000) is indicated as Q, whereas that by Huss et al. (1996) as P1. All C1-8 separates plot close to Q (P1). No systematic trend in the isotopic ratios is observed as a function of their densities. Mixing lines between Q (P1) and Xe-HL are shown as dotted lines. Ratios of fraction C1-8H are not shown because of their large errors.

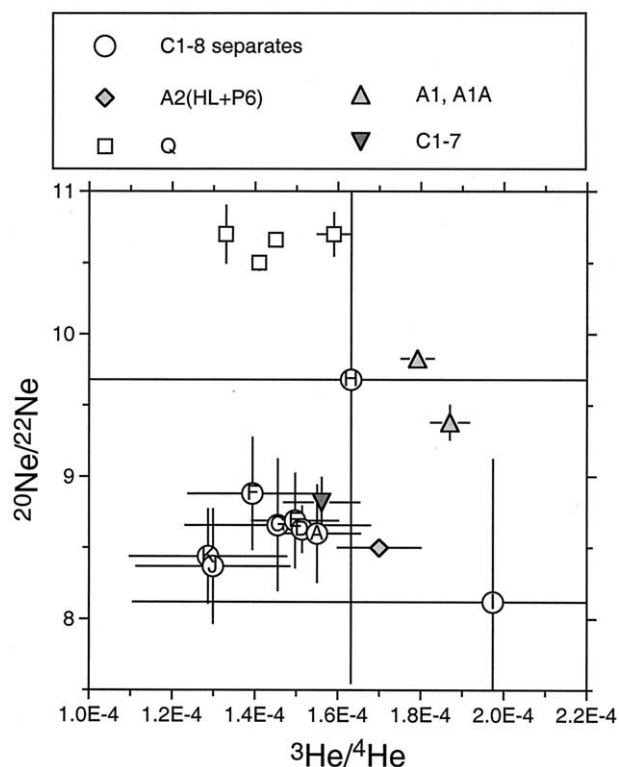


Fig. 4. $^{20}\text{Ne}/^{22}\text{Ne}$ ratios and $^3\text{He}/^4\text{He}$ ratios of the 1600°C temperature fractions in the C1-8 separates. For comparison, the ratios of the total fractions of A1, A1A (Matsuda et al., 1999) and C1-7 (Zaizen et al., 2000) are plotted. Q (Busemann et al., 2000) and A2 (Huss and Lewis, 1994) are also shown. Since diamond is enriched in lighter noble gases, the separates plot close to A2.

obtain a $^{21}\text{Ne}/^{22}\text{Ne}$ ratio only for C1-8D. The Ne isotopic ratios of C1-8D agree with those of Ne-A2 from presolar diamond (Table 2).

The $^3\text{He}/^4\text{He}$ ratio of C1-8, obtained from the masses and the $^3\text{He}/^4\text{He}$ ratios of the 1600°C fractions of all separates, is $1.49 \pm 0.07 \times 10^{-4}$, similar to the total ratio of C1-7, $1.561 \pm 0.091 \times 10^{-4}$, obtained by Zaizen et al. (2000). The ratio of C1-8 also agrees with the $^3\text{He}/^4\text{He}$ ratio of $1.59 \pm 0.04 \times 10^{-4}$ of an HF-HCl resistant residue from the Allende meteorite reported by Wieler et al. (1991). The $^3\text{He}/^4\text{He}$ ratios of the individual separates (except for C1-8H and C1-8I with large errors) are slightly lower than that of diamond, but in the same range as these of Q (Fig. 4). $^3\text{He}/^4\text{He}$ ratios of Q have a range of 1.23×10^{-4} to 1.59×10^{-4} , varying in different meteorites and in different extraction steps in a given meteorite (Wieler et al., 1991, 1992; Busemann et al., 2000).

One may wonder whether the $^3\text{He}/^4\text{He}$ ratios that are slightly lower than that of diamond can be due to a variation of $^3\text{He}/^4\text{He}$ ratios of Q itself. Wieler et al. (1991) observed that $^4\text{He}/^{20}\text{Ne}$ ratios of Q in the Allende meteorite progressively changed from 158 to 95 ($146-74$ for the second run) during the etching process. Since the total $^4\text{He}/^{20}\text{Ne}$ ratio of diamond from Allende (183; Table 7 in Wieler et al., 1991) is higher than these ratios and Ne in the separates is dominated by that of diamond as indicated in Figure 4, He in the separates should be dominated by diamond, but not Q.

Interestingly, Zaizen et al. (2000) observed a low $^3\text{He}/^4\text{He}$ ratio of $1.010 \pm 0.098 \times 10^{-4}$ in the 1200°C fraction in C1-7. They attributed the low ratio to the P3 component ($^3\text{He}/^4\text{He} < 1.35 \times 10^{-4}$). However, the major part of the He-P3 should

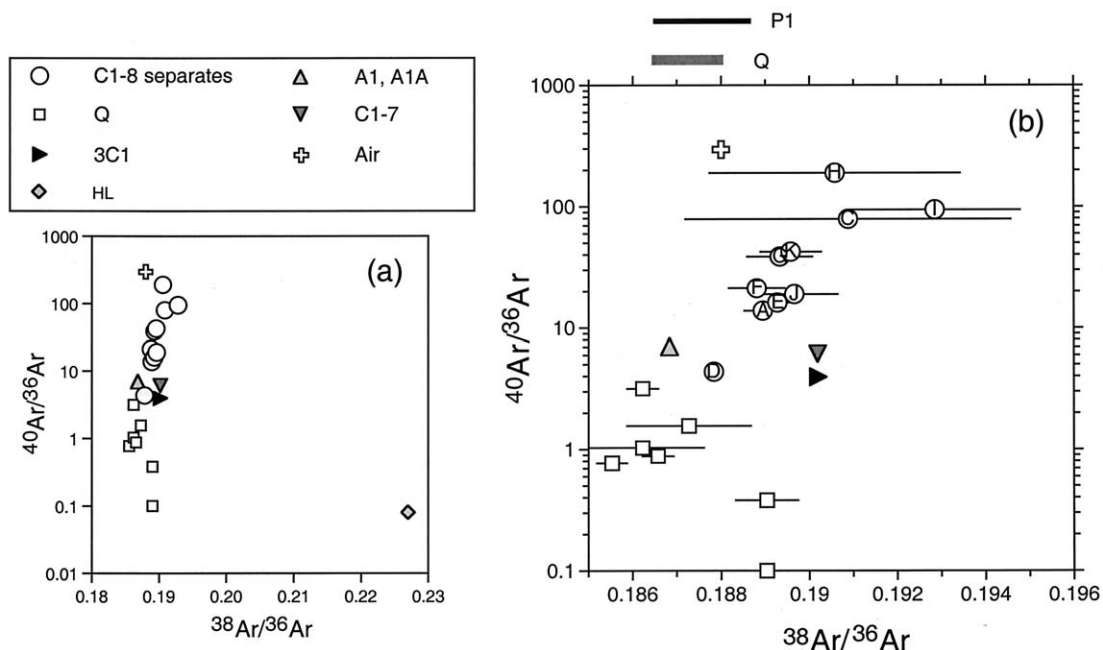


Fig. 5. Argon isotopic compositions of the 1600°C temperature fractions in the C1-8 separates. (b) is an enlargement of (a). C1-8D has the lowest $^{38}\text{Ar}/^{36}\text{Ar}$ and $^{40}\text{Ar}/^{36}\text{Ar}$ ratios, indicating that C1-8D is closest to the Q component. The $^{40}\text{Ar}/^{36}\text{Ar}$ ratios of Q are upper limits. The average $^{38}\text{Ar}/^{36}\text{Ar}$ ratios of Busemann et al. (2000) and Huss et al. (1996) are shown as thick lines.

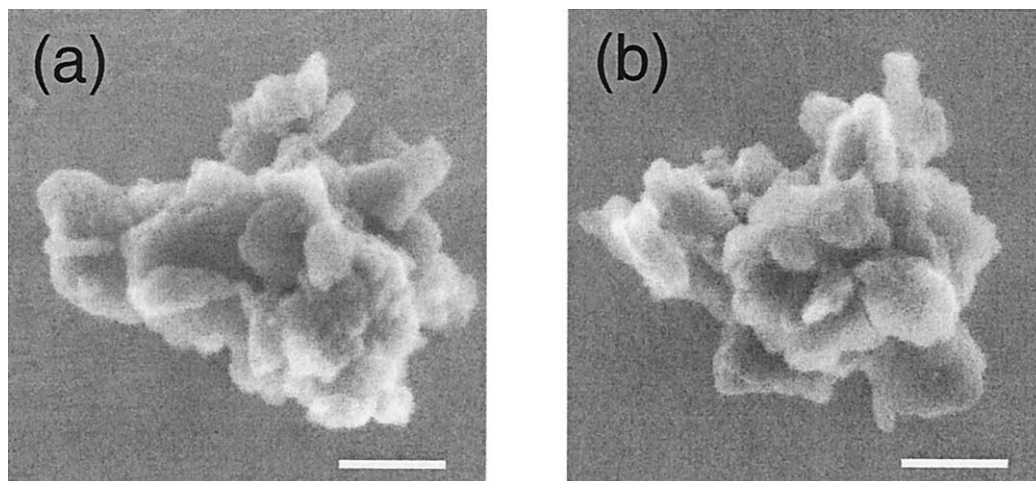


Fig. 6. Secondary electron images of carbonaceous grains in C1-8D. Many grains in this fraction look like aggregates of small grains. The scale bars are 1 μm .

have been released at the 600°C fraction because P3 is released between 200 and 900°C (Huss and Lewis, 1994).

Reynolds et al. (1978) also found a low $^3\text{He}/^4\text{He}$ ratio of $1.29 \pm 0.15 \times 10^{-4}$ in sample Allende A MII-1 that was prepared from a HF-HCl residue by colloidal extraction with methanol. (Noble gas isotopic ratios of the sample were similar to those of HF-HCl resistant residues.) With more than one observation of low $^3\text{He}/^4\text{He}$ ratios in the Allende samples, it may be worthwhile to reexamine $^3\text{He}/^4\text{He}$ ratios of Q and diamond as well as their $^4\text{He}/^{20}\text{Ne}$ ratios in the Allende meteorite.

The $^{38}\text{Ar}/^{36}\text{Ar}$ ratio of C1-8D is the lowest of these of the separates (Fig. 5). Within 2σ errors, however, the ratios of the separates (0.1878–0.1929) agree with those of Q and P1. On the other hand, $^{40}\text{Ar}/^{36}\text{Ar}$ ratios of the separates range from 4.4 (C1-8D) to 189 (C1-8H). $^{40}\text{Ar}/^{36}\text{Ar}$ ratios of Q in meteorites have been determined only as upper limits because ^{40}Ar is dominated by re-trapped radiogenic ^{40}Ar and atmospheric ^{40}Ar (Busemann et al., 2000). $^{40}\text{Ar}/^{36}\text{Ar}$ ratios of Q in various meteorites are as low as 0.1 and that of diamond is also low (0.08; Huss and Lewis, 1994).

The high $^{40}\text{Ar}/^{36}\text{Ar}$ ratios in the 1600°C fractions may result from meteoritic grains (other than Q and diamond) and/or terrestrial contamination during processing in the laboratory. In any case, the lowest $^{38}\text{Ar}/^{36}\text{Ar}$ and $^{40}\text{Ar}/^{36}\text{Ar}$ ratios of C1-8D indicate that this fraction is most enriched in Q among the separates.

3.2. Nature of Grains in C1-8D

Since fraction C1-8D is most abundant in Q among all C1-8 separates, we examined morphology, chemical compositions, and grain size of grains in the fraction. Grains in C1-8D were suspended in isopropanol, deposited onto Au foil, and examined for their chemical composition and grain size using the scanning electron microscope (JEOL 840A) at Washington University. Of 52 grains that were examined, grain sizes range from a little less than 1 to 4 μm , with an average diameter of 2 μm . Many grains appear as aggregates of smaller grains, having a flower-like shape (Fig. 6). It is not clear whether

aggregates formed in the laboratory or they were indigenous.

Energy dispersive X-ray (EDX) spectra taken at 10 kV using the Thermo Noran X-ray detector show that 51 out of 52 grains are carbonaceous and one grain is Cr-Fe oxide. Silicate grains have been observed in the floating fractions (Matsuda et al., 1999), but are absent in C1-8D, indicating that the density separation was successful. Fluorine and O peaks were observed in most carbonaceous grains. Average intensities of the F and O peaks relative to the C peak were 4 and 3%, respectively. Both elements were not detected when spectra were taken on the Au foil without grains. Amari and Matsuda (1998) noted that about a third of the carbonaceous grains in floating fraction A1 showed a F peak of an intensity comparable to the C peak at 10 kV. After the A1 grains were analyzed, a new X-ray detector with much higher sensitivity in lighter elements was installed. Thus it is not possible to directly compare the two results on F concentrations.

The presence of F in the grains is puzzling since we did not use hydrofluoric acid. A possible source of F is a Teflon beaker used during the freeze-thaw disaggregation. This will be discussed later.

4. DISCUSSION

4.1. Comparison with Previous Studies

4.1.1. Noble gas concentrations and mass yields

One of the advantages of preparing floating fractions rather than HF-HCl residues is that the rest of meteorite fragments can be saved for other studies. Yields of floating fractions depend on the number of cycles of the freeze-thaw disaggregation: the more cycling, the more floating material is recovered. In this study, after 216 cycles of the freeze-thaw disaggregation, 2620 μg of black material, or 0.068% of the starting material, floated on the surface, while after ~ 120 cycles Matsuda et al. (1999) recovered only 0.004% of the 15 g of Allende they started with. From the ^{132}Xe concentration of the Allende meteorite ($19 \times 10^{-10} \text{ cm}^3 \text{ STP/g}$; Lewis et al., 1975; Srinivasan et al., 1978; Matsuda et al., 1980), it is estimated that the floating fractions

of this study and by Matsuda et al. (1999) contain 5 and 0.4% of ^{132}Xe in the Allende meteorite, respectively. Thus, the majority of Q is still in the rest of the meteorite fragments. However, yields are not a useful indicator of the efficiency of the physical separation. Rather, as already discussed by Matsuda et al. (1999), it should be emphasized that the ^{132}Xe concentrations of the floating fractions (A1: 1.18×10^{-7} cm³ STP/g, A1A: 2.24×10^{-7} cm³ STP/g, and C1-7: 1.40×10^{-7} cm³ STP/g) (Matsuda et al., 1999; Zaizen et al., 2000) are equivalent to those of the HF-HCl resistant residues from the Allende meteorite (3C1: 3.03×10^{-7} cm³ STP/g, 1CS: 0.97×10^{-8} cm³ STP/g, BA: 1.96 and 2.21×10^{-7} cm³ STP/g) (Lewis et al., 1975; Srinivasan et al., 1978; Wieler et al., 1991), indicating that it is possible to enrich Q by physical separation as much as by chemical separation, without destroying the rest of the starting material.

The ^{132}Xe concentration of C1-8D is 2.5 times higher than that of C1-7 and the highest among all the C1-8 separates as well as those of previously studied HF-HCl residues from the Allende meteorite, among which 3C1 has the highest ^{132}Xe concentration (3×10^{-7} cm³ STP/g). 3C1 is known to contain chromite, whose abundance can be estimated from the analogous sample 4C1 (Table 2 in Lewis et al., 1975) to be 17 to 33 wt.% of the sample. (The two numbers result from the two different Cr concentrations in Table 2.) If chromite is removed from 3C1, its ^{132}Xe concentration is equivalent to that of C1-8D or higher. Although C1-8D is enriched in Q, the enrichment factor is not markedly different from those in the previous studies. We estimate that at most 5% of grains in C1-8D are Q, from a lower limit of ^{36}Ar concentration in Q ($\sim 6.8 \times 10^{-4}$ cm³ STP/g) obtained by Verchovsky et al. (2002).

An interesting aspect of this study is that half of the ^{132}Xe in the parent floating fraction is in C1-8D. The implications of the density of C1-8D will be discussed in section 4.5.

4.1.2. Density separation

Efforts have been made to characterize and isolate phase Q since its discovery (Lewis et al., 1975). Ott et al. (1981) separated three HF-HCl resistant residues of the Allende meteorite into several daughter fractions by physical and chemical methods. Fractions A-I-D (≤ 1.68 g/cm³) and A-I-C (1.68–1.73 g/cm³) that were separated by density from one of the HF-HCl residues comprised only 5.1 and 5.5% of the parent residue. The most abundant fraction was A-I-B2b with the density of 1.808 to 1.91 g/cm³, which comprised 67.8% of the residue. Starting from another HF-HCl resistant residue (A-II) that was treated slightly differently, Ott et al. (1981) obtained quite different yields of the fractions for the equivalent density range. Fraction A-II-E with a density of 1.808 to 1.91 g/cm³ comprised only 2.5% of the HF-HCl residue and 85% of the mass was in the fraction A-II-D with a density of 1.91 to 2.17 g/cm³. Their results of the two sets of density separation are different from the result in this study: the most abundant fraction in our study is C1-8D (1.65 ± 0.04 g/cm³) that comprises $\sim 19\%$ of the parent floating fraction. Fractions C1-8F (1.80 ± 0.06 g/cm³) and C1-8G (1.97 ± 0.06 g/cm³), whose density range is similar to that of A-I-B2b, are 22% of the parent sample by weight. The sum of C1-8G (1.97 ± 0.06 g/cm³) and C1-8H

(2.17 ± 0.05 g/cm³), whose density range is equivalent to that of A-II-D, comprise only 18%, not 85%.

This indicates how parent fractions have been processed critically affects the results of density separation. First, different treatments before density separation leave different kinds of minerals in the fractions that are to be separated by density. HF-HCl treated residues contain all forms of carbon and minerals that are resistant to HF, while in floating fractions carbon with high surface tension and minerals that attach to it are constituents. Since each mineral has its own stability range of pH against coagulation, the difference of mineral compositions is directly translated into the difference in stabilities against coagulation of the sample in solution at a given pH. In heavy liquids, where density separations are performed and pH cannot be defined (because there are not aqueous solutions), various minerals must have different affinities for a surfactant that is added to disperse the sample. Furthermore, even if mineral compositions are the same, surface of the minerals can be altered by chemicals: for example when carbonaceous matter is treated with oxidants, its surface becomes oxidized, changing the status of surface charges and resulting in different behavior in a solution (see subsection 4.2).

In broader terms, it seems that, in any kind of physical separation such as disaggregation, density separation, and size separation, the results critically depend on the kinds of minerals present and their surface conditions. Different results from various kinds of starting samples illustrate that separating Q is not an easy task.

4.2. Colloidal Separation

Xenon isotopic ratios of the separates indicate that Q/diamond ratios are very similar in all C1-8 separates and they are not markedly different from those of the floating fractions. Obviously we were not able to separate diamond by the colloidal separation.

The reason why diamond was not separated (into the colloidal fraction C1-8A) in our experiment may be due to the difference of the surface condition of diamonds because the sample in the preliminary experiment and C1-8 were prepared differently before the colloidal separation. The former was treated with boiling HClO_4 , a strong oxidant, while the latter was not subjected to any chemical reagents. It is a common procedure to oxidize samples (with milder oxidants such as H_2O_2 and $\text{Cr}_2\text{O}_7^{2-}$) before colloidal separation to reduce the amount of sticky kerogen that hinders any kind of separation. The absence of oxidation affected how the sample behaved in two ways. One is that organic matter, which would have been destroyed by an oxidant, was still in C1-8. It might have acted like glue and prevented diamond from entering the colloidal fraction. The other is that the surface of diamond in C1-8 was different from that of chemically processed diamond.

It has been proposed that diamond has weakly acidic groups such as $-\text{COOH}$ on the surface (Lewis et al., 1989) because it can be separated by changing pH of liquid. When the pH of a surrounding liquid increases, the following reaction proceeds and diamond becomes negatively charged, making diamond particles repulse one another and preventing them from coagulation in a suspension (Lewis et al., 1989).



When acids are added, the reaction moves to the left and diamond particles become neutral, forming aggregates. This is the reason why the colloidal separation, adding a basic solution to the sample suspension, is effective in extracting diamond.

The presence of carboxylic groups (–COOH) has been observed in infrared spectra of presolar diamond in carbonaceous chondrites (Lewis et al., 1989; Mutschke et al., 1995; Hill et al., 1997; Andersen et al., 1998; Braatz et al., 1998, 2000). Peaks at $\sim 1700 \text{ cm}^{-1}$ have been attributed to C=O bonds either in carboxylic groups (–COOH) (Hill et al., 1997; Braatz et al., 2000) or ester groups (Mutschke et al., 1995; Braatz et al., 2000). Braatz et al. (1998) observed an increase of the strength of the C=O peak at 1700 cm^{-1} compared to other peaks during the sample preparation and attributed it to an increasing oxidation of the sample by oxidizing chemicals.

In all cases where diamond-rich fractions were prepared by colloidal separation, samples had been oxidized by H_2O_2 and/or $\text{Cr}_2\text{O}_7^{2-}$ before the separation. Thus, the surface of diamond is very likely to have many carboxylic groups as a result of oxidation. In contrast, since we did not use any oxidants in our procedure, the surface of diamond was covered with fewer carboxylic groups compared to that of oxidized diamond.

4.3. Q and Diamond

All fractions have almost the same proportion of Q to diamond, indicating that they are closely associated. An effective bulk density of diamond was estimated to be 2.22 to 2.33 g/cm^3 (Lewis et al., 1987). It is not surprising, however, that diamond is distributed overall separates with a range of densities (from 1.1 to $> 2.3 \text{ g/cm}^3$). With its extremely small grain size (1.5–3 nm; Fraundorf et al., 1989; Daulton et al., 1996), diamond particles easily stick to other minerals. Andersen and Zinner (1993) suggested that Q gases were located on the surface of graphitized coatings on microdiamonds, thus Q and diamond were not physically separable. With this scenario, however, it is difficult to explain the fact that the release temperature of Xe-HL is lower (800–1000°C) than that of Q (1000–1400°C) in pyrolysis (Srinivasan et al., 1978).

There have been several observations that Q and diamond are unevenly distributed in meteoritic separates. $^{20}\text{Ne}/^{132}\text{Xe}$ ratios that are a good indicator of diamond/Q ratios vary among the floating fractions from the Allende meteorite. The ratios in the floating fractions range from 20.3 (A1) to 47.9 (C1-7). Matsuda et al. (1999) estimated that the $^{132}\text{Xe-HL}/^{132}\text{Xe-Q}$ ratio of A1A is 1.7 times higher than that of A1. From HF-HCl resistant residues of the Allende meteorite Ott et al. (1984) separated non-colloidal fractions that exhibited $^{132}\text{Xe-HL}/^{132}\text{Xe-Q}$ ratios 4 times lower than those of HF-HCl resistant residues. These results imply that diamond and Q do not always behave the same way during various stages of the separation and that it may be possible to separate Q and diamond without destroying Q. However, there are very few clues as to what conditions brought about the separation of these two phases.

4.4. Subphases in Q

It has been argued that Q is more than one phase with different susceptibilities to etching and amounts of noble gases. Gros and Anders (1977) progressively etched an Allende residue with HNO_3 and concluded that there were two Q-phases: “Q1” contained most of the heavy noble gases and was readily soluble in cold diluted HNO_3 , whereas “Q2” contained heavy noble gases with at least 1 order of magnitude lower concentrations than Q1 and was slowly dissolved in hot concentrated HNO_3 . From noble gas data of HF-HCl resistant residues from various meteorites as well as the data from Cold Bokkeveld (CM2), Busemann et al. (2000) have proposed that subphase Q1 has higher He/Xe and Ne/Xe ratios, releases noble gases by moderate etching, and contains the major part of Q gases, while subphase Q2 is more resistant to etching. They also concluded that the two phases react differently to aqueous and thermal alterations and that Q1 is more abundant in CM2 and CV3 (ox.) chondrites than in all other classes. Q1 and Q2 by Busemann et al. (2000) may correspond to “Q1” and “Q2” from Gros and Anders (1977), respectively.

If Q consists of more than one phase, it is not unreasonable to assume densities of subphases are slightly different from one another. In our experiment, about half of Q in the parent floating fraction is concentrated in C1-8D. Except for C1-8C, H and I that contained $< 2\%$ of Q, the fractions second most abundant in Q are C1-8E and C1-8A with slightly $> 10\%$ of Q. Q abundances in C1-8E, F, and G progressively decrease (Fig. 2). Busemann et al. (2000) observed that $^4\text{He}/^{20}\text{Ne}$ of Q1 was higher (170–240) than that of Q2 (50–80). $^4\text{He}/^{20}\text{Ne}$ ratios of the C1-8 separates range from 155 to 182 if we exclude that of C1-8H with a huge error (85 ± 154). The ratios of the separates are closer to that of Q1 and show no distinct difference among the separates. Since Q1 seems to carry most of the heavy noble gases (Gros and Anders, 1977; Busemann et al., 2000), if even a little amount Q1 exists in Q2-enriched separates, noble gases of Q1 would overwhelm those of Q2.

4.5. Carbonaceous Nature of Q

Half of Q in the parent floating fraction is concentrated in fraction C1-8D with a density of $1.65 \pm 0.04 \text{ g/cm}^3$, where most grains are carbonaceous. This implies either the density of C1-8D represents the density of Q, or that Q has a strong tendency to stick to the matter of that density.

A question arises as to what forms of carbon the grains in C1-8D consist of. The Allende meteorite contains $\sim 0.3\%$ of C (King et al., 1969), most of which is present as inorganic C rather than as organic polymer (Breger et al., 1972; Vis et al., 2002). Han et al. (1969) found that the abundance of organic matter that was soluble to solvents was a few orders of magnitude smaller in the Allende meteorite than in other carbonaceous chondrites. Smith and Buseck (1981a, 1981b) conducted HRTEM (High Resolution Transmission Electron Microscopy) studies on HF-HCl resistant residues and found that carbon in the Allende meteorite is predominantly a poorly crystalline graphite, structurally similar to “glassy” carbon that is a tangled aggregate of ribbon-shaped packets of graphite layer planes, most of the crystallites being < 10 graphitic layers. Lumpkin (1981) also examined HF-HCl resistant residues from Allende

by TEM and found that most of the 0.5 to 2 μm carbonaceous particles showed diffraction patterns characteristic of poorly ordered “graphitic” carbon or “turbostratic” carbon, having a structure composed of randomly stacked two-dimensional layers. He did not find any polymorphs of carbon such as diamond, lonsdaleite, and graphite.

Although the density of well-crystalline graphite is 2.2 g/cm^3 , that of poorly crystallized graphite varies depending on the degree of crystallization. Presolar graphite has a range of density (1.6–2.2 g/cm^3 ; Amari et al., 1994) and occurs in two morphologic forms, cauliflower and onion types (Hoppe et al., 1995). Bernatowicz et al. (1991, 1996) reported isotopic, chemical, structural, and crystallographic analyses of presolar graphite from the Murchison meteorite and found that the cauliflower-type grains that are more abundant in lower-density graphite fractions (KE3 and KFA1: 1.65–2.10 g/cm^3 ; Amari et al., 1994) consist entirely of turbostratic graphite (graphite with contorted layers with no long-range continuity). The density of C1-8D (1.65 \pm 0.04 g/cm^3) is in the low end of the density range of presolar graphite. Thus, at least part of the carbonaceous grains in C1-8D could be poorly crystalline graphite.

Organic matter in the Allende meteorite was investigated shortly after its fall. Levy et al. (1970) made a powder sample from the interior of the meteorite and heated it up to 460°C and analyzed released organic matter by gas chromatography. They found that many different aromatic hydrocarbons and several alkanes were present. Cronin et al. (1987) used ^{13}C NMR spectroscopy to analyze organic matter in a HF-HCl resistant residue from the meteorite and found that aliphatic groups and, to a lesser extent, aromatic groups were present. Densities of many kinds of organic compounds have a range of \sim 1.4 to 1.6 g/cm^3 , close to the density of C1-8D. Thus, part of C1-8D grains might be organic as well.

Observations suggest that not all the forms of carbon in the meteorite are Q. Verchovsky et al. (2002) have shown that Q in enstatite chondrites comprises a very small part of carbonaceous matter that is relatively unaffected by thermal alteration. In general, boiling points of hydrocarbons are much lower than those of polymorphs of carbon, indicating the former are more susceptible to thermal alterations than the latter. Thus, if the density of C1-8D represents the density of Q, Q is not likely to be organic matter, although we cannot completely rule out the possibility.

Another interpretation is that Q has a tendency to stick to material with the density of 1.65. In addition to organic matter and poorly crystalline graphite, which can be “attractors” of Q, there is one more possible, uninvited “attractor,” Teflon particles. During the freeze-thaw disaggregation, we used a beaker made of Teflon, whose density ranges from 1.7 to 2.15 g/cm^3 . Two hundred fourteen cycles of the process might have disaggregated small particles from the beaker and/or have abraded the surface of the beaker. Teflon particles might have stuck to carbonaceous Q and formed the aggregates as shown in Figure 6. (We cannot tell whether aggregates formed before the sample was processed in the laboratory or they are artifacts of the processing.) The detection of F peaks in the C1-8D grains does not contradict this scenario. We estimated the depth to be 2 μm from which F X-rays can be emitted from carbonaceous matter (1.65 g/cm^3) at the accelerating voltage of 10 kV. This is the same as the average grain size of C1-8D grains. Thus, even if

Teflon particles comprise a small portion of the particles we examined, we would not be able to tell whether the F peaks originated from a small region of the grains.

5. CONCLUSIONS

We separated a floating fraction, which is enriched in Q and diamond, into ten fractions by a combination of colloidal and density separations in an attempt to physically separate Q. Fraction C1-8D with the density of 1.65 \pm 0.04 g/cm^3 contains about half of Q in the parent floating fraction. Grains in C1-8D are mostly carbonaceous grains with a small amount of F and O. This observation implies either that the density of Q is 1.65 \pm 0.04 g/cm^3 or that Q tends to stick to material of that density. All separates as well as the floating fractions from Allende have similar Q/diamond ratios. Except for Ar, noble gas isotopic ratios of the separates are indistinguishable from each other.

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