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Noble gas measurement in individual micrometeorites using laser gas-extraction system

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Abstract: All noble gases in 12 individual Antarctic micrometeorites, series F96C and F96D, taken from a water tank at the Dome Fuji Station, were measured by using Nd-YAG CW laser-extraction system. According to the results of light noble gases, the micrometeorites can be separated into two groups, "gas-rich" and "gas-poor". Samples in the gas-rich group have SEP-like $^{3}He/^{4}He$ ratios and Ne isotopic ratios between SEP- and Solar wind-Ne, which are consistent with previous reports on micrometeorites or IDPs. On the other hand, samples in the gas-poor group have very low $^{3}He/^{4}He$ ratios and $^{20}Ne/^{22}Ne$ ratios lower than the atmospheric values. The $^{40}Ar/^{36}Ar$ ratios are also separated into two groups, *i.e.*, samples with lower $^{40}Ar/^{36}Ar$ belong to the gas-rich group. Cosmogenic noble gases were undetected in all the samples and cosmic-ray exposure ages may be shorter than 4.5 Ma considering error limits of Ne data. Heavy noble gas elemental compositions are chondritic. There was no correlation between the concentration of volatile elements, sulfur, and that of He and Ne.

1. Introduction

Antarctic micrometeorites (AMMs) survived from severe heating during atmospheric entry, and they can provide us with new information about the solar system. In fact, cosmic dust particles are abundant in space and might be originated from various source materials, some of which may differ from parent bodies of meteorites. Noble gas analysis has been applied to small particles collected from deep-sea sediments and the stratosphere to obtain evidence of extraterrestrial origin and to investigate the temperature at the atmospheric entry (*e.g.*, Merrihue, 1964; Nier *et al.*, 1990). Several investigators have attempted to measure noble gases on single particles using high sensitive noble gas mass spectrometers. Nevertheless, only a few have been reported of finding light noble gases in individual particles collected from Antarctica and Greenland (*e.g.*, Maurette *et al.*, 1991). Hence noble gas data of single AMMs are required to clarify their extraterrestrial origin, source materials, irradiation histories by solar and galactic cosmic rays of these particles,

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and to clarify the relationship between the AMMs and the particles collected from other places such as the stratosphere and the deep-sea.

As a part of consortium study on the Antarctic micrometeorites collected by the Japanese Antarctic Research Expedition (Nakamura *et al.*, 1999), we have measured all noble gases in individual micrometeorites. We present here the results of noble gas data for 12 individual micrometeorites and discuss their origin in the solar system.

2. Samples

Samples studied in this work were hand picked from precipitated fine particles recovered from a water tank at the Dome Fuji Station. These AMMs were found in the recent fallen snow around the station located at the top of a moraine at 3810 m above sea level in Queen Maud Land at 77°19′ south latitude, 39°42′ east longitude (Nakamura *et al.*, 1999). All micrometeorite samples measured in this work have been identified as extraterrestrial materials using low-vacuum scanning electron microscope with energy dispersive spectrometer (LV-SEM/EDS) at Kyushu University under the criteria: (1) large peaks at Mg, Si, and Fe, (2) small peaks at Al and Ca, (3) traces of Cr and Ni, (4) variable height of a peak at S, and (5) absence of other elements, especially K (Nakamura *et al.*, 1999). Their major element compositions in the criteria are similar to those of matrix from Allende and Murchison. In the present work, samples at about 1 μg and diameters ranging 100–200 μm were selected in order to measure all noble gases in individual particles. Only one sample (F96CK005) was a totally melted spherule and the others were unmelted to partially melted, irregular shaped particles. The database of AMMs including these samples is published on the World Wide Web [URL: http://dust.cc.gakushuin.ac.jp] via the Internet.

3. Experimental procedures

Individual micrometeorites were carefully picked with Taxal tweezers and each dropped into a platinum crucible and settled in a crucible holder made of stainless steel. The platinum crucibles with 2.0 and 1.8 mm outside and inside diameters, respectively, were made from platinum tube annealed in a high temperature furnace. Thirty-seven platinum crucibles can be set in the sample holder. After all samples were set, the sample holder was put in the ultra-high vacuum chamber connected to a noble gas purification line. For noble gas extraction, individual samples were heated using a slightly defocused Nd-YAG continuous wave laser beam by increasing the output power (Fig. 1). The range of output power of YAG laser is 2.5–14.5 W. The heating time is about 10 min. The advantage of laser heating is that only one sample is heated thus reducing the blank level. The sample can be observed through the microscope equipped with the laser-system, and also displayed on a CRT monitor through a CCD camera. Since only the sample emits red colored light during the laser heating, it is easy to confirm the position of the sample by the CCD

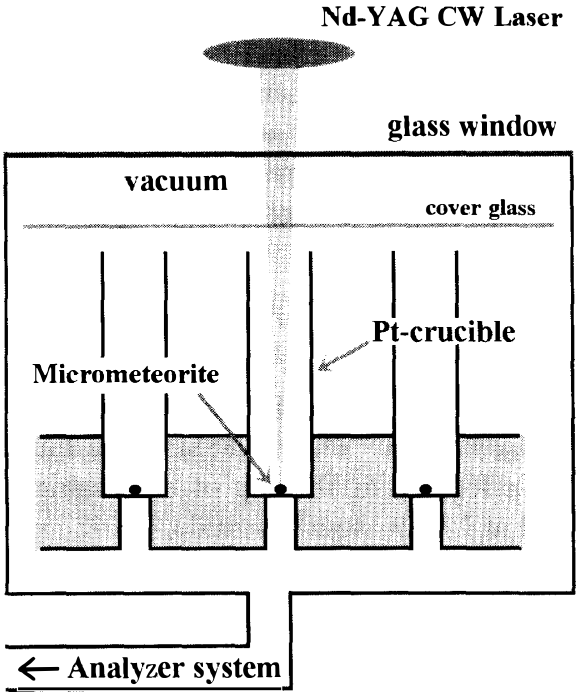

Fig. 1. A schematic drawing of sample chamber. Each micrometeorites is put in a Pt-crucible, its inner diameter is 1.8mm. Nd-YAG laser beam is focussed on a sample to evaporate by heating. Cover glass prevents the evaporating materials from a deposition onto glass window during heating. Evolved gas is introduced into analyzer system, followed by purification, separation among noble gas elements, and mass spectrometry.

camera. Extracted gases were purified by two Ti-Zr getters and noble gases except for He were trapped on a cryogenically cooled trap at 15 K. Ne, Ar, Kr and Xe were released successively at 45, 100, 135 and 250 K and the isotopic composition of each gas was analyzed on a modified VG-5400 mass spectrometer (MS-III) at the Laboratory for Earthquake Chemistry, University of Tokyo. All noble gas elements were measured by an ion counting collector. Sensitivities for all noble gases and mass discrimination effects were calibrated by measurement of atmospheric noble gases, and a helium standard gas with $^3He/^4He = 1.71 \times 10^{-4}$ prepared by mixing pure ^3He and ^4He gases in our laboratory. In neon analysis, mass interferences of Ar^{++} and CO_2^{++} were corrected. Blank corrections were carried out for all samples, but released amounts of noble gases of some gas-poor particles are comparable to the blank levels, for which blank correction was difficult and are shown in parentheses (Tables 3–5). The average value of six blank measurements was taken as blank value for the blank correction.

4. Results and discussion

Main features for AMMs in this study, weight, size, type of AMM, S/Si ratio, color, transparency and luster are presented in Table 1. Range of sample sizes is 80–250 μm in diameter and their weights are 0.4–7.1 μg, except for one spherule F96CK 005 (21.5 μg). Optical characteristics of these samples were determined using transmitted and reflected light through binocular microscopes based on a classification scheme of cosmic dust catalog published by NASA JSC (Nakamura *et al.*, 1999).

Table 1. List of AMMs F96 series studied in this work.

Sample[1)]	Weight (μg)	Size (μm)	Type[2)]	S/Si[3)]	Color	Trans.[4)]	Luster[5)]
F96CK005	21.5	250×250	Sp	0	black	O	D
F96DK002	7.1	200×150	Mm	0.5	black	O	D
F96DK014	3.0	200×120	Mm	0.9	dark gray	O	D
F96DK016	1.0	130×80	Mm	0.1	black	O	D
F96DK019	2.7	120×100	Mm	0.8	black	O	D
F96DK026	1.1	130×100	Mm	0	black	O	D
F96DK029	1.0	170×120	Mm	0	black	O	D
F96DK036	0.9	100×80	Mm	0.2	dark gray	O	D
F96DK044	1.1	150×110	Mm	0	dark gray	O	SM
F96DK051	0.4	130×100	Mm	0.3	black	O	D
F96DK052	1.4	150×150	Mm	0	black	O	SM
F96DK060	1.0	160×100	Mm	0.8	black	O	D

1) ID number of AMMs expresses collection site and years, identities of precipitated material, and facilities performed the initial examination; F96AK001 was collected at Dome Fuji (F) in 1996 (96), contained in a magnetic fraction in 960423-1 precipitated material (A), and processed in Kyushu University (K).
2) Types 'Sp' and 'Mm' stand for spherule and micrometeorite, respectively.
3) Sulfur/Silicon ratios were determined by LV-SEM/EDS and normalized to Murchison matrix.
4) Transparency (abbreviated Trans) is determined by optical microscopy to be opaque (O).
5) Luster is classified to Dull (D), and Submetalic (SM).

4.1. Amounts and concentrations of all noble gases

Amounts of all noble gases released from the individual AMMs and calculated concentrations are shown in Table 2. The amounts of released noble gases from the samples are very small and variable among the samples. Blank levels are comparable to those of some samples. There is no correlation between the weight of AMMs and noble gas concentration, which may reflect variable amounts of implanted gases of solar origin and different degrees of heating during atmospheric entry. F96CK005 has the lowest concentrations of light noble gases helium and neon. This particle is a spherule and its morphology indicates that the particle became molten during atmospheric entry and lost its noble gases during this event.

4.2. He

The result of He analysis is shown in Table 3. $^3He/^4He$ ratios higher than in the atmospheric value (1.4×10^{-6}) were observed in most micrometeorites. Absolute amounts of helium released from the samples were generally very small, and the smallest amount of 6.4×10^{-12} cm^3 STP ^4He was observed in F96DK044. The highest concentration of ^4He for F96DK026 (3×10^{-3} cm^3 STP/g) is, however, lower than the high concentrations ($\sim10^{-1}$ cm^3 STP/g) reported for interplanetary

Table 2. Measured noble gas amounts released from the individual AMMs, and calculated amounts for unit mass (1 g).

Sample	Weight (μ g)	$^{4}He^{1)}$	$^{20}Ne^{1)}$	$^{36}Ar^{1)}$	$^{40}Ar^{1)}$	$^{84}Kr^{1)}$	$^{132}Xe^{1)}$	$^{4}He^{2)}$	$^{20}Ne^{2)}$	$^{36}Ar^{2)}$	$^{40}Ar^{2)}$	$^{84}Kr^{2)}$	$^{132}Xe^{2)}$
		(10^{-12} cm^{3}STP)						(10^{-6} cm^{3}STP/g)					
F96CK005	21.5	29.4	1.56	0.34	110	0.0059	0.0053	1.1	0.042	0.005	2.1	0.00023	0.00024
F96DK002	7.1$^{4)}$	60.2	40.4	4.2	142	0.046	0.035	7.7	5.6	0.55	11	0.0063	0.0049
F96DK014	3.0$^{4)}$	104	5.22	0.80	110	0.0036	0.021	33	1.5	0.19	15	0.0009	0.0068
F96DK016	1.0$^{4)}$	54.7	0.65	0.42	130	0.0092	0.011	49	---$^{3)}$	0.19	64	0.0084	0.011
F96DK019	2.7$^{4)}$	35.7	21.2	1.4	77.9	0.0046	0.0058	11	7.6	0.44	4.4	0.0014	0.0021
F96DK026	1.1$^{4)}$	3660	14.5	2.4	157	0.0059	0.011	3300	13	2.0	82	0.0045	0.010
F96DK029	1.0$^{4)}$	494	5.81	3.3	204	0.0028	0.0053	490	5.1	3.0	138	0.0019	0.0051
F96DK036	0.9$^{4)}$	196	7.25	3.6	157	0.016	0.014	210	7.3	3.8	101	0.016	0.016
F96DK044	1.1$^{4)}$	6.4	2.02	0.30	81.4	0.0014	0.0002	0.95	1.2	---$^{3)}$	---$^{3)}$	---$^{3)}$	---$^{3)}$
F96DK051	0.4$^{4)}$	19.9	0.52	0.28	69.0	0.0025	0.0036	36	---$^{3)}$	---$^{3)}$	---$^{3)}$	0.0041	0.0085
F96DK052	1.4$^{4)}$	129	3.55	1.52	185	0.015	0.012	88	2.1	0.92	85	0.010	0.0087
F96DK060	1.0$^{4)}$	13.4	0.60	0.36	88.5	0.0038	0.013	8.1	---$^{3)}$	0.12	22	0.0029	0.013
blank$^{5)}$	---	5.4	0.66	0.23	66.1	0.0009	0.0002	---	---	---	---	---	---

1) Measured abundances without blank corrections. They have about 5-20% uncertainties.
2) Blank corrections were carried out.
3) Concentrations cannot be determined due to low abundance of noble gases comparable to blank values.
4) Weights have 10-30 percent errors. They were measured with a precise balance which is able to measure 0.1 μ g.
5) Average values of six measurements.

dust particles (IDPs) collected in the stratosphere (*e.g.*, Rajan *et al.*, 1977; Nier and Schlutter, 1990). It is thought that the IDPs were implanted with a large amount of solar gases in space and were not heated enough to degas during atmospheric entry. The lower concentration of helium in the micrometeorites measured in this study compared to that of IDPs may be due to smaller surface/weight ratio of the micrometeorites to that of the IDPs. The IDPs collected in the stratosphere are generally irregular shaped and aggregates of smaller grains, while the typical AMMs are not aggregates of smaller grains. Farley *et al.* (1997) have concluded that the extraterrestrial He in seafloor sediments is surface-correlated.

In Fig. 2, $^{3}He/^{4}He$ ratios are plotted against measured amounts of ^{4}He. Samples with relatively high helium concentrations have isotopic ratios similar to that of Solar Energetic Particles (SEP). This is evidence that these micrometeorites are exposed to solar wind or solar flare in space and might have lost low energy solar wind component during atmospheric entry. These values resemble the case of magnetic separates of deep-sea sediments (*e.g.*, Fukumoto *et al.*, 1986; Matsuda *et al.*, 1990).

On the other hand, samples with low concentrations of He show lower $^{3}He/^{4}He$ ratios. This trend is different from current reports for IDPs collected in the stratosphere (Nier and Schlutter, 1993; Pepin and Schlutter, 1998; Kehm *et al.*, 1999), in which $^{3}He/^{4}He$ ratios are generally higher than SEP- or Solar-He in samples with low ^{4}He concentrations. This result might show a difference between IDPs and the AMMs studied in this work, though origin of the He with low $^{3}He/^{4}He$ ratio is not clear at present.

Table 3. Released amounts of He and isotopic ratios.

Sample	not corrected for blank			corrected for blank[1)]		
	^{3}He	^{4}He	^{3}He/^{4}He	^{3}He	^{4}He	^{3}He/^{4}He
	(10^{-12}cm^{3}STP)		(10^{-4})	(10^{-12}cm^{3}STP)		(10^{-4})
F96CK005	nd	29.4	---	---	24.0	---
F96DK002	0.012	60.2	1.98 ±0.03	0.012	54.8	2.17 ±0.03
F96DK014	0.027	104	2.63 ±0.53	0.027	98.6	2.77 ±0.56
F96DK016	0.00036	54.7	0.07 ±0.17	0.00035	49.4	0.07 ±0.19
F96DK019	0.0039	35.7	1.1 ±1.2	0.0038	30.4	1.3 ±1.4
F96DK026	0.944	3660	2.58 ±0.21	0.943	3654	2.58 ±0.21
F96DK029	0.086	494	1.73 ±0.32	0.086	488.9	1.75 ±0.32
F96DK036	0.046	196	2.36 ±0.68	0.046	190.6	2.43 ±0.70
F96DK044	(0.0001)	6.4	(0.17) (±0.39)	---[2)]	---[2)]	---[2)]
F96DK051	0.0009	19.9	0.45 ±0.60	0.0009	14.6	0.6 ±0.8
F96DK052	0.021	129	1.61 ±0.45	0.021	123.3	1.68 ±0.47
F96DK060	0.0017	13.4	1.3 ±1.6	0.0017	8.1	2.2 ±2.7
blank[3)]	nd	5.4 ±1.5	---	---	---	---

1) Atmospheric isotopic ratio was assumed.
2) cannot be corrected for blank due to small amounts of He.
3) Average value of six measurements.
All errors are one sigma

4.3. Ne

Isotopic compositions of Ne for the individual micrometeorites are shown in Table 4 and Fig. 3. The correlation of isotopic ratios of Ne and the content of ^{20}Ne are also shown in Fig. 4. Based on the observed isotopic ratios, samples can be divided into two groups as indicated in Fig. 3, where they are plotted separately in the upper and lower areas. SEP- and SW-Ne are observed only in the group with high concentrations of Ne, and this corresponds to the group characterized by high ^{3}He/^{4}He (SEP-like He) (Fig. 5). The SEP-like Ne isotopic ratios observed in this work resemble the results of the AMMs recovered from Antarctic blue ice (Maurette *et al.*, 1991) and the unmelted particles from Greenland sediment

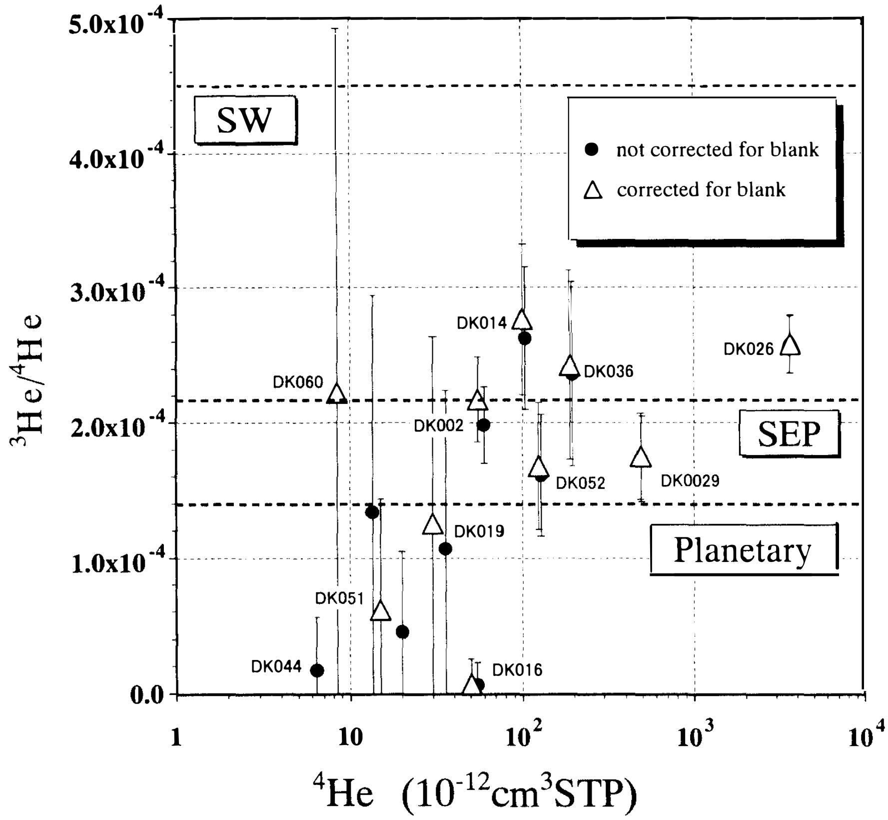

Fig. 2. Measured amounts of He and $^3He/^4He$ ratios for individual AMMs are plotted. There is a tendency that the samples with large amounts of He have high $^3He/^4He$ ratios. Error bars are one sigma. It was assumed that the blank has atmospheric composition. Solar wind (SW) and Solar Energetic Particles (SEP)-He are from Benkert et al. (1993) and Planetary-He from Reynolds et al. (1978).

(Olinger *et al.*, 1990). In the case of deep-sea sediments, SEP-like $^{20}Ne/^{22}Ne$ ratios were observed in magnetic fraction (*e.g.*, Amari and Ozima, 1988). Kehm *et al.* (1998) reported of Ne compositions reflecting a mixture between SW- and SEP-Ne in single IDPs. This may reflect that IDPs were less heated than AMMs and still contain low energy solar wind in their extreme surface layer.

The other group with low concentrations of Ne has low $^{20}Ne/^{22}Ne$ ratios. It is difficult to determine the isotopic ratios for such small amounts of Ne, resulted in relatively large uncertainties for the isotopic ratios as indicated in Fig. 3. Hence, we can only show the values before blank correction in Table 4 and Figs. 3–5. However, it is evident that they lack in solar-type He and Ne. Accurate determination of Ne isotopic composition in this group may lead to interesting findings, such as original materials before solar gas implantation.

Spallation-produced Ne was not observed in all "gas-rich" samples within experimental error limits. Ne measurements of deep-sea sediments and most of IDPs also indicate a negligible contribution of cosmic-ray spallation Ne (*e.g.*, Fukumoto *et al.*, 1986; Matsuda *et al.*, 1990; Kehm *et al.*, 1998). Hence we can give only an upper limit for cosmic-ray exposure ages of these samples with solar-type

Table 4. Contents and isotopic ratios of Ne in micrometeorites.

Sample	not corrected for blank					corrected for blank[1)]	
	^{20}Ne	^{21}Ne	^{22}Ne	$^{20}Ne/^{22}Ne$	$^{21}Ne/^{22}Ne$	$^{20}Ne/^{22}Ne$	$^{21}Ne/^{22}Ne$
	($10^{-12}cm^3STP$)						
F96CK005	1.56	(0.0061)	(0.189)	(8.3) (±1.5)	(0.032) (±0.021)	---[2)]	---[2)]
F96DK002	40.4	0.118	3.32	12.18 ±0.11	0.036 ±0.004	12.23 ±0.12	0.035 ±0.005
F96DK014	5.22	0.013	0.475	10.99 ±0.92	0.028 ±0.007	11.2 ±1.1	0.021 ±0.010
F96DK016	0.65	(0.00188)	(0.0773)	(8.4) (±1.4)	(0.024) (±0.017)	---[2)]	---[2)]
F96DK019	21.2	0.0514	1.73	12.26 ±0.31	0.030 ±0.004	12.36 ±0.33	0.028 ±0.004
F96DK026	14.5	0.0353	1.26	11.46 ±0.33	0.028 ±0.008	11.56 ±0.36	0.026 ±0.008
F96DK029	5.81	0.0165	0.521	11.2 ±1.1	0.032 ±0.006	11.4 ±1.3	0.027 ±0.008
F96DK036	7.25	0.0201	0.595	12.2 ±0.58	0.034 ±0.008	12.5 ±0.7	0.030 ±0.009
F96DK044	2.02	(0.0102)	(0.225)	(9.0) (±1.3)	(0.045) (±0.030)	(8.8) (±2.0)	(0.033) (±0.048)
F96DK051	0.52	(0.00319)	(0.0592)	(8.8) (±1.1)	(0.054) (±0.030)	---[2)]	---[2)]
F96DK052	3.55	0.0094	0.317	11.2 ±1.1	0.030 ±0.010	11.6 ±1.4	0.020 ±0.014
F96DK060	0.60	(0.0037)	(0.0753)	(8.0) (±1.6)	(0.049) (±0.025)	---[2)]	---[2)]
blank[3)]	0.66 ±0.56	0.005 ±0.004	0.07 ±0.06	9.3 ±0.8	0.071 ±0.016	---	---

1) Blank corrections were carried out for the samples with more than five times Ne gases than blank level.
2) cannot be corrected for blank due to small amounts of Ne.
3) Average values of six measurements.
All errors are one sigma

Ne. We do not consider the samples with low He, Ne concentrations due to large experimental uncertainties. If error limits for the Ne isotopic ratios are taken into consideration, an upper limit of cosmogenic ^{21}Ne concentration is estimated to be 4.9×10^{-9} cm^3 STP/g for the sample F96DK036. Reedy (2000) has shown that ^{21}Ne production rate by SEP becomes high in very small chondritic objects such as cosmic dust (*i.e.*, 1.07×10^{-8} cm^3 STP/g/Ma at 1AU), while the production rate by galactic cosmic-rays (GCR) was calculated to be as small as 0.08×10^{-8} cm^3 STP/g/Ma at 1 AU in small particles (0 cm particles in Graf *et al.*, 1990). If we assumed

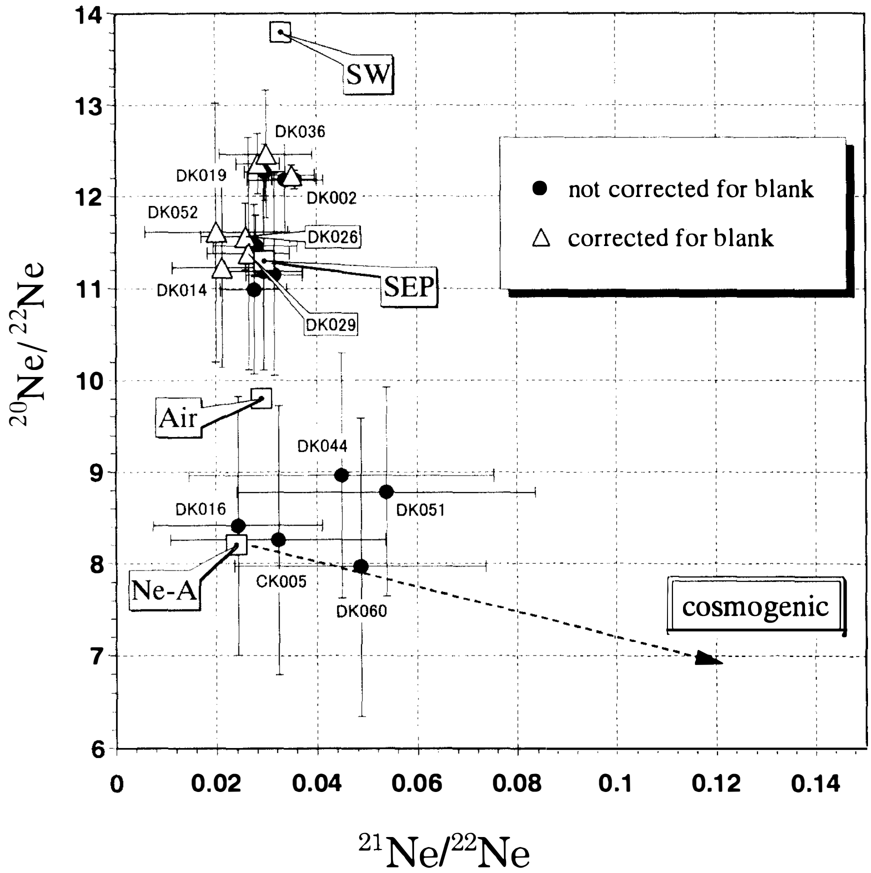

Fig. 3. Ne three isotope plot. Blank corrected values for the samples with low concentrations of Ne can not be plotted in this figure due to large ambiguity. Samples with high concentrations of Ne are distributed between SW and SEP. Error bars are one sigma. Solar wind (SW) and Solar Energetic Particles (SEP)-Ne are from Benkert et al. (1993), Ne-A from Black and Pepin (1969) and Air-Ne from Eberhardt et al. (1965).

that the production rate by SEP is inversely proportional to the square of distance from the sun, and SEP contribution was limited within the range from 1 to 3 AU, cosmic-ray exposure age is calculated to be 4.5 Ma for F96DK036 in upper limit. Since the cosmogenic ^{21}Ne concentration should be lower than the estimation described above, the exposure age would be much shorter than 4.5 Ma.

4.4. *Ar*

Absolute amounts of Ar and isotopic ratios of Ar in AMMs are shown in Table 5 and Fig. 6 is a three-isotope plot of Ar. The $^{40}Ar/^{36}Ar$ ratios show that more than half of the samples have values much lower than atmospheric ratio (296). Since terrestrial materials do not have such low isotopic ratios, the low $^{40}Ar/^{36}Ar$ ratios indicate the extraterrestrial origin for these samples.

AMMs with low $^{40}Ar/^{36}Ar$ have relatively high concentrations of Ar as in the case of Ne. Ar isotopic ratios in this group may be explained as a mixture of several components such as atmospheric, SEP-, SW-, and Planetary-Ar.

The observed $^{40}Ar/^{36}Ar$ ratios are higher than the value of SEP- and SW-Ar,

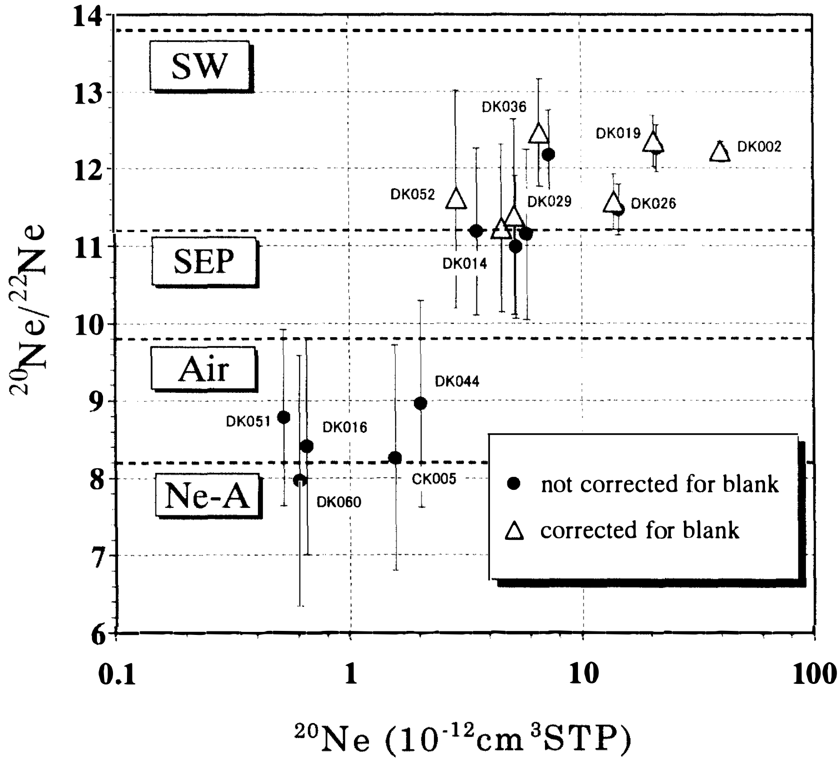

Fig. 4. Measured amounts and isotopic ratios of Ne for individual AMMs are plotted. These samples are clearly separated into two groups. Error bars are one sigma.

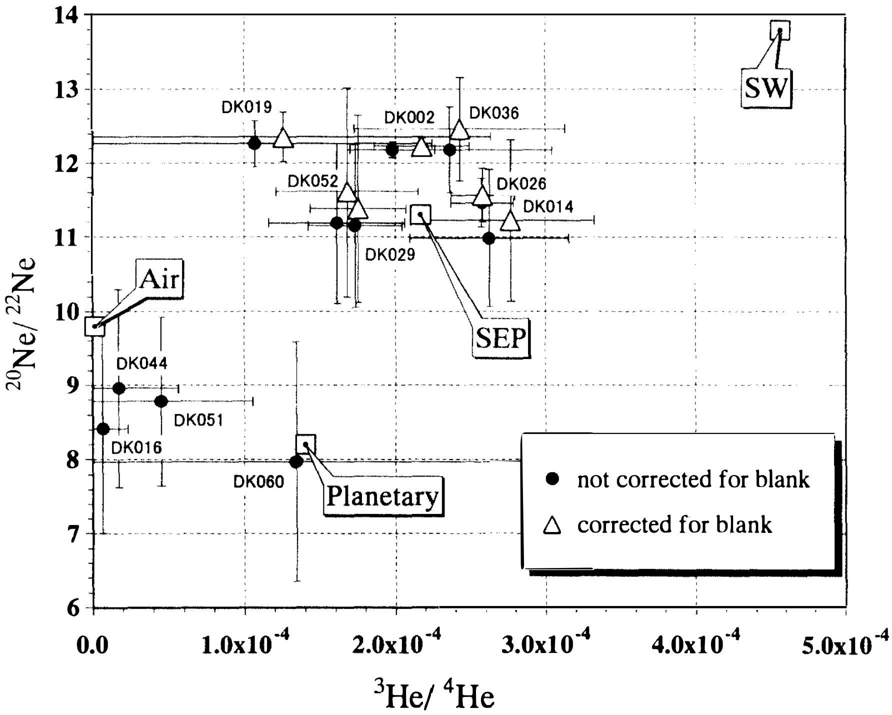

Fig. 5. Relationship of $^{3}He/^{4}He$ ratio and $^{20}Ne/^{22}Ne$ ratio. Most AMMs with SEP-like He have also SEP-like Ne. This shows that solar-He and Ne were implanted into AMMs simultaneously. On the other hand, AMMs with low $^{3}He/^{4}He$ have not SEP-like Ne.

Table 5. Contents and isotopic ratios of Ar in micrometeorites.

Sample	not corrected for blank ^{36}Ar	^{38}Ar (${10^{-12}}cm^3STP$)	^{40}Ar	$^{38}Ar/^{36}Ar$	$^{40}Ar/^{36}Ar$	corrected for blank[1] $^{38}Ar/^{36}Ar$	$^{40}Ar/^{36}Ar$
F96CK005	0.34	0.069	110	0.200 ±0.006	320.4 ±5.9	0.218 ±0.028	399 ±31
F96DK002	4.2	0.80	142	0.191 ±0.003	34.0 ±0.2	0.191 ±0.004	19.2 ±2.5
F96DK014	0.80	0.16	110	0.195 ±0.004	137.0 ±2.6	0.196 ±0.006	77.0 ±14.4
F96DK016	0.42	0.079	130	0.186 ±0.024	306.3 ±6.7	0.180 ±0.053	335 ±16
F96DK019	1.4	0.28	77.9	0.198 ±0.004	54.9 ±1.1	0.199 ±0.005	10.0 ±8.7
F96DK026	2.4	0.47	157	0.195 ±0.003	65.1 ±1.0	0.195 ±0.004	41.7 ±4.3
F96DK029	3.3	0.60	204	0.184 ±0.006	62.8 ±0.8	0.184 ±0.007	45.8 ±3.1
F96DK036	3.6	0.69	157	0.189 ±0.003	43.4 ±0.3	0.189 ±0.003	26.9 ±2.8
F96DK044	0.30	(0.053)	81.4	(0.180) (±0.012)	275.4 ±6.2	---[2]	---[2]
F96DK051	0.28	(0.052)	69.0	(0.188) (±0.011)	248.6 ±4.3	---[2]	---[2]
F96DK052	1.5	0.29	185	0.191 ±0.005	121.8 ±2.2	0.191 ±0.006	91.5 ±6.2
F96DK060	0.36	0.067	88.5	0.187 ±0.014	247.7 ±4.1	0.176 ±0.045	181 ±42
blank[3]	0.23 ±0.035	0.045 ±0.007	66.1 ±9.9	0.192 ±0.008	283.1 ±3.1	---	---

1) Blank corrections were carried out for the samples with more than 1.4 times Ar gases than blank level.
2) cannot be corrected for blank due to small amounts of Ar.
3) Average value of six measurements.
All errors are one sigma.

for which $^{40}Ar/^{36}Ar$ is assumed to be <1. Though possible contamination of our samples by atmospheric Ar can not be eliminated, the ^{40}Ar in our gas-rich samples would be *in-situ* produced radiogenic ^{40}Ar since the measured concentrations of ^{40}Ar are in the range for chondrites (10^{-6}–10^{-5} cm^3 STP/g). This assumption is supported by the elemental composition of heavy noble gases shown in Fig. 7, where the gas-rich samples are plotted in the area represented by most chondrites. Apart from these samples, effect of atmospheric noble gas contamination for the gas-poor samples is difficult to be eliminated.

Kehm *et al.* (1998) reported Ar isotopic ratios of IDPs which are similar to our results. Since $^{38}Ar/^{36}Ar$ ratios for the components SW, Planetary, SEP and

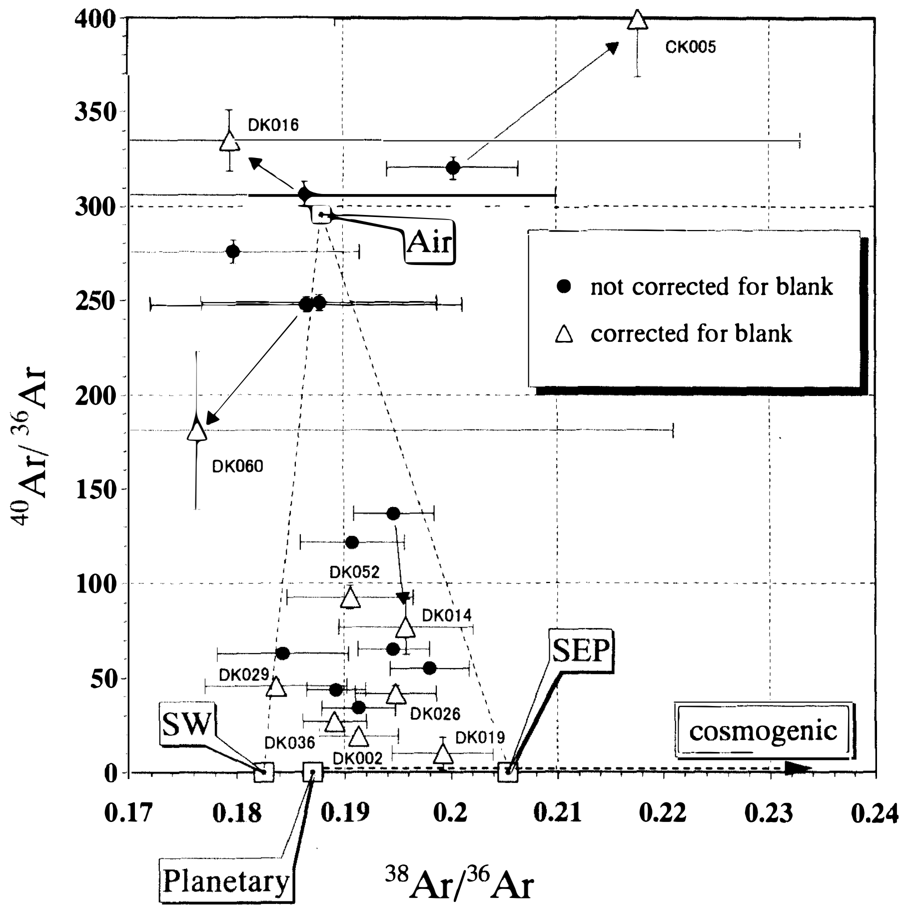

Fig. 6. Ar three isotope plot. Error bars are one sigma. Solar wind (SW) and Solar Energetic Particles (SEP)-Ar are from Benkert et al. (1993), Planetary-Ar from Reynolds et al. (1978) and Air-Ar from Nier (1950).

atmosphere are in the narrow range as shown in Fig. 6, it is difficult to determine which component is dominant in these samples. No obvious contribution of spallogenic Ar has been observed in these samples, which is consistent with He and Ne data.

On the other hand, the group with high $^{40}Ar/^{36}Ar$ ratio corresponds to the samples with low contents of Ar. This trend is similar to that shown by Ne. Only one spherule sample F96CK005 has slightly higher $^{40}Ar/^{36}Ar$ and $^{38}Ar/^{36}Ar$ than the atmospheric values. The isotopic ratios might reflect the residual Ar gases trapped in the sample, though most gases had escaped by heating during atmospheric entry. From the excess ^{38}Ar, we can estimate a lower limit of cosmic-ray exposure ages of about 0.6 Ma for this spherule using production rate proposed for ordinary chondrites (Eugster, 1988). Original material of this spherule might be a relatively large fragment of meteorites with cosmic-ray exposure age of more than 0.6 Ma. A possible source of this spherule is a fragment from ordinary chondrites of high petrologic type, since their $^{40}Ar/^{36}Ar$ ratios are generally higher than the atmospheric one and the cosmic-ray exposure ages of most chondrite are longer than 1 Ma.

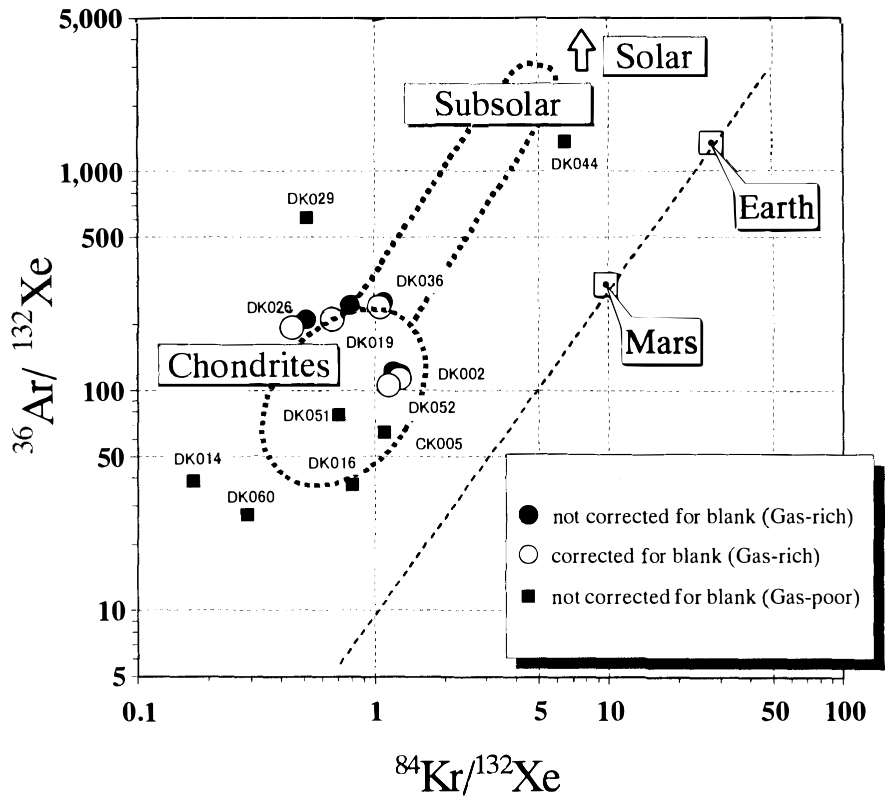

Fig. 7. Plot of heavy noble gas elemental compositions. Samples with relatively high concentrations of heavy noble gases have chondritic noble gas compositions. For some gas-poor samples, blank correction introduces large uncertainties due to the low concentrations of heavy noble gases.

4.5. *Heavy noble gas*

Amounts of Kr and Xe released from individual particles and their isotopic ratios are shown in Table 6. $^{129}Xe/^{132}Xe$ ratios do not show large excesses in ^{129}Xe, and are similar to those of carbonaceous chondrites (*e.g.*, Nagao *et al.*, 1984) though the experimental errors are still large due to extremely small amounts of Xe ($<3\times10^{-14}$ cm^3 STP).

Elemental compositions of heavy noble gases are presented in Fig. 7, where $^{36}Ar/^{132}Xe$ ratios are plotted against $^{84}Kr/^{132}Xe$. The gas-rich samples show chondritic heavy noble gas compositions, which are clearly different from the atmospheric composition. For the samples of the gas-poor group, accurate determination of heavy noble gas concentrations was difficult, and not plotted in the figure. This result indicates that the micrometeorites originally had chondritic noble gases, and light noble gases of solar origin were implanted into these particles in space.

4.6. *Sulfur*

Depletions in the volatile elements S and Zn have been proposed as indicators of heating level during atmospheric entry (Greshake *et al.*, 1994). Sulfur tends to evaporate at a lower temperature than zinc, in the heating simulation of Orgueil meteorite fragments of $\sim$100 μm in size, ninety percent of sulfur is lost at 1000°C/

Table 6. Released amounts of Kr and Xe and isotopic ratios.

sample	^{84}Kr	^{132}Xe	^{82}Kr	^{83}Kr	^{86}Kr	^{129}Xe	^{130}Xe	^{131}Xe	^{134}Xe	^{136}Xe
	($10^{-15}cm^3STP$)		^{84}Kr = 100			^{132}Xe = 100				
F96CK005	5.9	5.3	20	21	30	114	23	98	54	43
			±4	±5	±8	±29	±10	±32	±17	±12
F96DK002	46	35	20	18	31	102	15	80	39	36
			±4	±2	±3	±10	±3	±8	±4	±6
F96DK014	3.6	21	19	18	29	98	17	83	39	27
			±5	±8	±9	±12	±5	±13	±6	±7
F96DK016	9.2	11	22	19	34	101	16	65	43	28
			±6	±3	±5	±17	±4	±18	±12	±11
F96DK019	4.6	5.8	23	21	33	118	19	127	59	44
			±8	±8	±8	±17	±6	±22	±13	±11
F96DK026	5.9	11	19	18	28	96	20	72	37	31
			±5	±8	±5	±19	±6	±13	±9	±8
F96DK029	2.8	5.3	23	20	31	106	17	84	37	35
			±5	±4	±5	±8	±2	±10	±3	±3
F96DK036	16	14	20	18	30	99	16	78	35	31
			±6	±3	±3	±16	±5	±19	±10	±13
F96DK044	1.4	0.2	nd	nd	nd	nd	nd	nd	nd	nd
F96DK051	2.5	3.6	nd	nd	nd	nd	nd	nd	nd	nd
F96DK052	15	12	21	21	29	91	14	71	40	30
			±5	±5	±5	±27	±3	±19	±8	±9
F96DK060	3.8	13	24	21	29	108	19	90	42	34
			±9	±10	±10	±21	±4	±24	±10	±7
blank	0.9[1)]	0.2[1)]	42[2)]	48[2)]	82[2)]	30[2)]	8[2)]	63[2)]	29[2)]	34[2)]
	±1.9	±0.5	±34	±25	±29	±27	±7	±33	±10	±14

1) Average value of six measurements.
2) Result of the first experiment. Isotopic ratios could not be determined in other five experiments due to small amount of heavy noble gases.
All data are not corrected for blank.
All errors are one sigma
nd: not determined due to the low contents of gases.

20 s, on the other hand, zinc is hardly depleted in the same condition (Greshake *et al.*, 1994, 1996). It has been shown for IDPs that the concentrations of light noble gases correlate positively with those of volatile element zinc (Kehm *et al.*, 1998). However, such a correlation is not observed in the present study as is shown by the plot for sulfur/silicon ratios versus He and Ne concentrations (Figs. 8a and b). Flynn *et al.* (1993) showed no correlation between zinc and sulfur contents for IDPs, and the inferiority of sulfur as an indicator of entry heating. Our result for AMMs is consistent with their conclusion. Lack of positive correlation between sulfur and light noble gases may be explained by the fact that sulfur concentrations reflect only the mineralogical characteristics of AMMs.

4.7. *Elemental abundance pattern*

Figure 9 shows elemental abundance patterns for typical AMMs studied in this

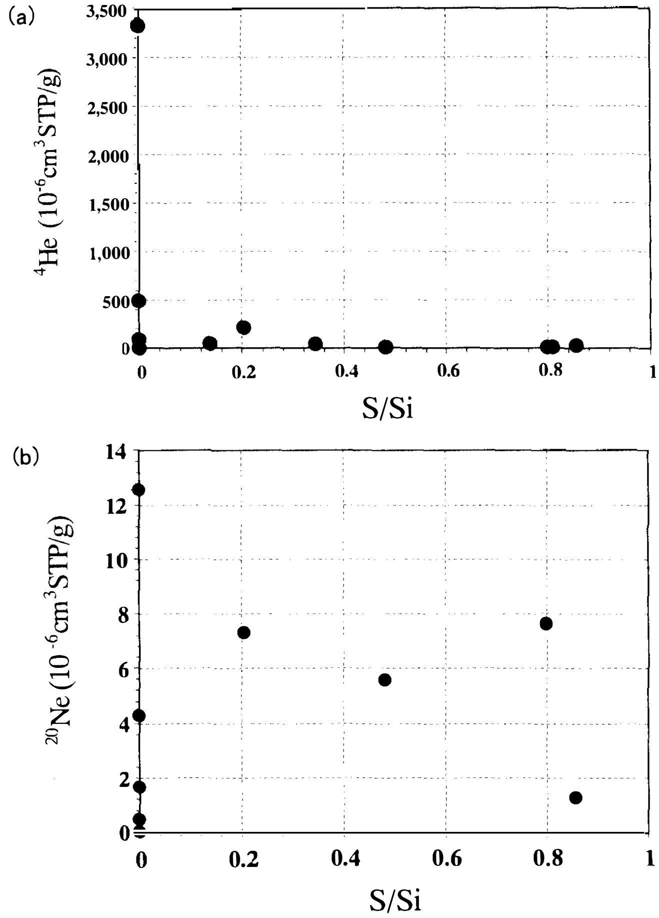

Fig. 8. Sulfur contents normalized for Murchison matrix versus concentrations of ^{4}He and ^{20}Ne. There are no correlations between light noble gas concentrations and contents of sulfur which is one of the volatile elements. It is impossible to evaluate the heating history of micrometeorites during atmospheric entry from the information of sulfur concentrations.

work. Compared to lunar soil or planetary noble gases, the patterns for the micrometeorites have a large diversity, though the patterns have some remarkable features. The heavy noble gas patterns are similar to the planetary-type pattern. On the other hand, the pattern for light noble gases of F96DK026, in which the noble gases of solar origin are abundant, resembles the solar-type pattern as observed in Lunar soil 12001 (Eberhardt *et al.*, 1972). However, most samples have unique light noble gas patterns which are unlike those of both planetary and solar. These might be due to some effects such as implantation of solar noble gases, different trapped components and different degrees of depletion of noble gases during atmospheric entry.

4.8. *Origin of AMMs*

Lack of cosmogenic noble gases in micrometeorites show a very important fact that AMMs had not experienced long term cosmic-ray irradiation. Hence, the noble gas data obtained in the present work clearly indicates that the gas-rich micrometeorites were small objects in space, and not fragments produced from break up of

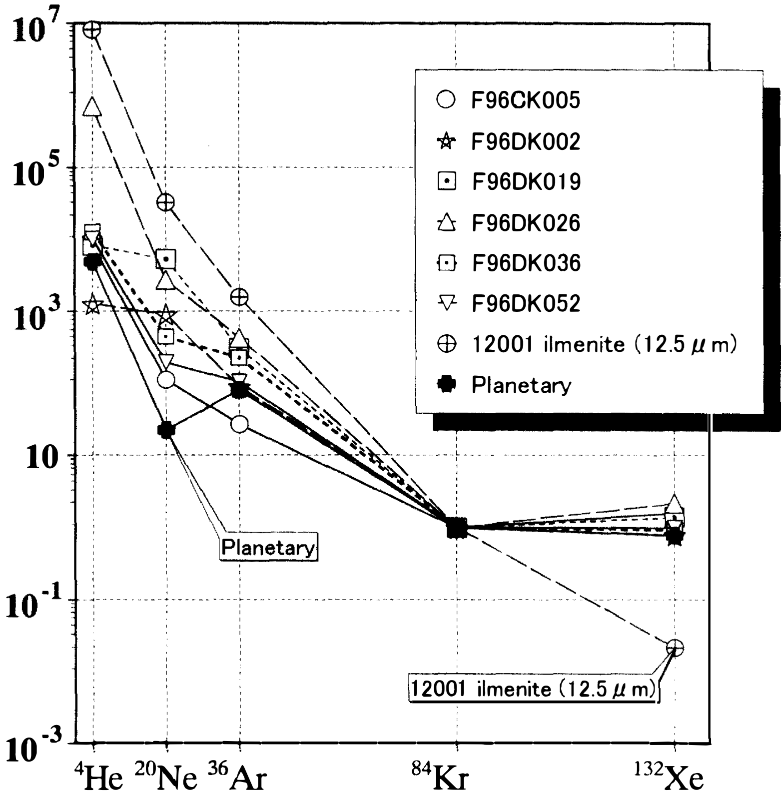

Fig. 9. Elemental abundance patterns of all noble gases in typical AMMs normalized to ^{84}Kr. The patterns for heavy noble gases are very similar to the Planetary one. Lunar soil 12001 ilmenite is from Eberhardt et al. (1972) and Planetary from Mazor et al. (1970).

meteorites which fell in Antarctica. If these AMMs were fragments produced by break-up of meteorite which fell on the earth, they would generally have spallogenic noble gases, since normal size meteorites commonly have spallogenic noble gas isotopes such as ^{3}He, ^{21}Ne and ^{38}Ar, and meteorites with abundant solar noble gases are rare. This information limits the possible candidates for the origin of AMMs.

One appropriate candidate for the source material of micrometeorite is fragments in Asteroidal belt. Infrared Astronomical Satellite (IRAS) observation showed the asteroidal dust bands which could be a significant contributor to the remainder of the zodiacal signal (*e.g.*, Dermott *et al.*, 1984). Such small particles will fall into the sun from the original orbit due to the Poynting-Robertson effect (Wyatt and Whipple, 1950), and part of the particles would be captured by the earth. If we assume that the micrometeorites came from Asteroidal belt, the Poynting-Robertson effect suggests a very short time before these micrometeorites reach the orbit of the earth. For example, in case of the particles, 100 μm in diameter and 1 g/cm^3 in density located at 2 AU, it takes only 0.15 Ma before they drop to the earth's orbit. This short period is not long enough to accumulate detectable amounts of cosmogenic noble gases in the particles due to their small sizes and the solar gas implantation. Hence, the undetectable amounts of cosmogenic nuclides in the AMMs favor the asteroid origin. However, if this is the case, the particles should have been derived from deep interior of parent asteroids, otherwise we would have observed some samples which came from the surface of asteroids where abundant cosmogenic noble gases were accumulated due to weak shielding to cosmic-rays. From the photographic observations of meteorite falls and spectral investigations of reflected light from asteroidal objects, most meteorites are thought to come from the Asteroidal belt. Among the meteorites, only a few stony meteorites with exposure ages shorter than 1Ma have been discovered. The remarkable difference between the meteorites and AMMs with respect to the concentrations of cosmogenic noble gases might be an important point indicating the origin of AMMs.

Another possible source of AMMs is dust grains ($>50\,\mu$m) in Kuiper belt (Liou *et al.*, 1996). However, it takes a longer period for the particles to reach the earth's orbit from Kuiper belt (30–40 AU) than that from Asteroidal belt (2–3 AU). AMMs measured in this study have irradiation ages shorter than 4.5 Ma. If typical $100\,\mu$m size AMMs came from Kuiper belt, it takes about 50 Ma, which eliminates the possibility of Kuiper belt as the origin of AMMs in the present work.

One of the most appropriate candidates for the origin of micrometeorites may be short period comets from Kuiper belt. Messenger and Walker (1998) showed the possibility that the cluster IDPs that were collected during June–July 1991 came from the comet Schwassmann-Wachmann 3. A large amount of extraterrestrial materials might be supplied to the inner solar system from Kuiper belt by the mediation of short period comets. This assumption may be the best to explain the result of extremely short cosmic-ray exposure ages of AMMs. Since surface layer will be ablated by solar radiation in the vicinity of perihelion of every revolution, signs of the cosmic-ray irradiation probably were erased from the surface. Short period comets have a revolution cycle less than 200 years, and have plenty opportunities to approach the sun scattering large amounts of particles. The dusts in tail of comet have very large variation of grain size (Fulle *et al.*, 1990). Light noble gases of solar origin are implanted into the micrometeorites during a relatively short period, since implantation of solar wind is saturated in only a few decades (*e.g.*, Hudson *et al.*, 1981; Rajan *et al.*, 1977). Though comet particles are supposed to enter the atmosphere at higher velocities than the particles of asteroidal origin. For example, Comet Halley whose perihelia is 0.6 AU has atmospheric entry velocity of 65 km/s (Flynn, 1989). But in the case of Comet Kopff with small inclination and perihelia greater than 1.5 AU, it has low entry velocity of 12.6 km/s (Flynn, 1989). If such comets are the source of AMMs and IDPs, the particles might have survived the heating during atmospheric entry and kept noble gas signature of solar origin. Hence, short period comets from Kuiper belt seem to harmonize with the noble gas data observed in AMMs.

At present, it is difficult to clarify the origin of AMMs, and they are possibly the mixture of different origins. However, in any case, AMMs are interesting particles which escaped severe heating during atmospheric entry, providing us with new insights into the origin and evolution of our solar system.

5. Conclusions

1) Abundant solar-type He and Ne (dominantly solar component) were detected in more than half of the samples, which indicate that the AMMs orbited in space as small particles and solar noble gases were implanted into them.

2) Ar gases of extraterrestrial origin were detected in many samples. However, it is difficult to determine which component of Ar, *e.g.*, SEP, solar or planetary, is relevant to the observed one.

3) Spallogenic noble gases are negligible in all samples, which indicates short cosmic-ray exposure ages (<4.5 Ma) for the samples.

4) Chondritic heavy noble gas components were detected, which indicates that the AMMs originally had chondritic noble gases.

5) The noble gas data suggest that the AMMs are not fragments produced from break-up of meteorites which fell on the earth. If these AMMs were fragments of meteorites of normal size, they would generally have cosmogenic noble gases and solar noble gases would not have been abundant in them.

6) We could not find out any correlation between noble gas contents and concentrations of sulfur which is one of the volatile elements.

7) The best candidate of source material of AMMs seems to be the short period comets from Kuiper belt. This conjecture seems to be in harmony with noble gas data.

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