Antarct. Meteorite Res., **13**, 311–321, 2000

Solar-wind derived light noble gases in micrometeorites collected at the Dome Fuji Station: Characterization by stepped pyrolysis

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Abstract: Noble gases in micrometeorite-bearing particles were characterized by the total-melting and stepped-heating analyses, in order to determine average compositions of light noble gases of micrometeorites collected at the Dome Fuji Station. He and Ne are dominated by solar-wind derived noble gases and the concentrations are comparable to the highest ones detected so far in carbonaceous chondrites. Cosmogenic gases are a very minor component in micrometeorites, suggesting short exposure to solar and galactic cosmic rays. The high ratio of solar to cosmogenic gases suggests that the micrometeorites had been small particles in the interplanetary space to have large surface areas to be exposed to solar winds. The micrometeorites are supposed to have fallen on Antarctica in the recent fifty years with snow around the Dome Fuji Station (T. Nakamura *et al.*, Antarct. Meteorite Res., **12**, 183, 1999a), and hence they are particles generated in the modern solar system and came to the Earth after short periods of exposure to solar winds and galactic cosmic rays.

1. Introduction

Micrometeorites are extraterrestrial particles with typical size range from 50 to 400 μm, recovered from Antarctic and Greenland ice fields and northern Canada (Maurette *et al.*, 1986, 1987, 1991, 1992, 1994; Cresswell and Herd, 1992; Taylor *et al.*, 1998; Yada and Kojima, 1999). They represent the major fraction of the material accreting to the Earth. It is known that a large fraction of micrometeorites is similar in mineralogy, mineral chemistry, and bulk elemental abundance to the constituents of hydrous carbonaceous chondrites, although there are differences in some features such as higher C/O and pyroxene/olivine ratios, absence of carbonate, and depletion of Ca, Ni, S, and Na in micrometeorites (*e.g.*, Maurette *et al.*, 1991, 1994; Steele, 1992; Kurat *et al.*, 1994; Greshake *et al.*, 1996; Genge *et al.*, 1997; Yano and Noguchi, 1998; England and Maurette, 1998).

Micrometeorites are severely heated upon atmospheric entry and the peak temperature reached increases with an increase of particle size, density, and entry velocity. Computer simulations and radar detection of meteor trails show that most micrometeorites larger than 100 μm vaporize on atmospheric entry (Hughes, 1978; Flynn and McKay, 1990). Micrometeorites that can survive the heating without melting are those with entry velocities near earth escape velocity (Flynn *et al.*, 1993). But even those escaping melting suffer thermal alteration at subsolidus temperature such as decomposition of hydrous minerals and loss of volatile elements S, Zn, Ga and Ge (*e.g.*, Kurat *et al.*, 1994; Greshake *et al.*, 1996, 1998; Nozaki *et al.*, 1999). Noble gases are elements that are subjected to be lost during heating in the atmosphere, but many micrometeorites are found to retain extraterrestrial noble gases (Olinger *et al.*, 1990; Nier *et al.*, 1990; Maurette *et al.*, 1991). According to these previous results, light noble gases in individual micrometeorites are dominated by solar-wind derived gases, suggesting that they were exposed to solar winds as micrometeoroids.

In the present investigation, concentration measurements along with isotopic ratio determinations of light noble gases in micrometeorite-bearing particles that were collected at the Dome Fuji Station in Antarctica were carried out in order to know average signatures of noble gases of the Dome-Fuji micrometeorites.

2. Sample preparation and experimental methods

Samples are a set of magnetic fine particles less than 300 μm in size with high sedimentation rates in the water, having been separated from F960901 precipitated material (F3 fraction in Nakamura *et al.*, 1999a). Scanning electron microscope analysis with an energy dispersive X-ray detector indicated that the set of particles is the richest in micrometeorites compared with other sets of particles (Nakamura *et al.*, 1999a). Based on the terrestrial accretion rates of micrometeorites (Taylor *et al.*, 1998) and the snow-fall rates around the Dome Fuji Station, 100 mg of the sample particles should contain 1.2 mg micrometeorites (Nakamura *et al.*, 1999a). Constituents other than micrometeorites are dominated by iron or iron oxides that are artificial contaminants probably welding evaporate generated by some activities around the station. In the present investigation we further separated the sample particles into three fractions: particles ranging in size less than 70 μm, those from 70 to 200 μm, and those from 200 to 300 μm. Noble gas analyses were done for the following two sets of particles: 20.0 mg of 70–200 μm particles, which are expected to contain 0.247 mg of micrometeorites, and 16.1 mg of $<70\,\mu$m particles, which would contain 0.199 mg of micrometeorites. Typical weight of a single micrometeorite with $\sim$100 μm in size is 1 μg (*e.g.*, Osawa *et al.*, 2000) and thus, in a rough estimate, each set of sample particles contains 200 micrometeorites.

Noble gas analyses were made by a MM5400 mass spectrometer at Kyushu University equipped with a Ta furnace and a stainless steel purification line "Jack and the beanstalk". He and Ne of the 70–200 μm-sized particles were analyzed by total melting at 1700°C, while He, Ne, and Ar in the $<70\,\mu$m-sized particles are characterized by stepped pyrolysis at 400, 700, 1000, 1300, and 1700°C. Heating duration at each temperature is 25 min that include temperature rising periods of a furnace, thus the real heating duration is shorter. Detailed examination of heating profiles using thermocouples indicates that the real heating duration at the designated temperature is approximately 15 min. The extracted gases were purified by two sets of Ti getters, separated into He-Ne and Ar fractions using charcoal traps, and analyzed by the daily collector for gases more than 10^{-10} cc STP or the ion counting system for gases less than that. Blank measurements were done in between sample analyses and typical blank level at 1700°C is 6×10^{-10}, 4×10^{-12}, and 2×10^{-9} cc STP for ^{4}He, ^{20}Ne, and ^{40}Ar, respectively. Blank correction was made only for the 1700°C temperature step of an analysis of the $<70\,\mu$m particles.

3. Results and discussion

3.1. *Solar winds in micrometeorites*

Herium and Ne concentrations and isotopic ratios of a set of particles with diameter from 70 to 200 μm are shown in Table 1. ^{3}He/^{4}He, ^{20}Ne/^{22}Ne, and ^{21}Ne/^{22}Ne ratios are very close to those of solar flare (SF), 0.000260, 11.6, and 0.030, respectively, reported by Rao *et al.* (1991). This suggests that micrometeorites in diameter from 70 to 200 μm retain high energy particles emitted from the Sun. Concentrations of solar-wind derived He and Ne are comparable to the highest ones detected so far in carbonaceous chondrites by spot analyses on meteorite thin plates using a laser probe with approximately 100 μm diameter (Nakamura *et al.*, 1999c).

Stepped hating analysis of a set of particles less than 70 μm in diameter indicates that micrometeorites in this size range are also dominated by solar-type

Table 1. He and Ne composition of a set of micrometeorite-bearing particles from 70 to 200 microns in diameter separated from F960901.

^{4}He[10^{-4} cc/g]*	^{3}He/^{4}He	^{20}Ne[10^{-6} cc/g]*	^{20}Ne/^{22}Ne	^{21}Ne/^{22}Ne
15.0	0.000267±0.000005	47.1	11.759±0.118	0.036±0.002

* Concentrations are determined by deviding absolute amounts of gases by an assumed weight of micrometeorites (0.24 mg).

Table 2. He and Ne composition of a set of micrometeorite-bearing particles less than 70 microns in diameter separated from F960901.

	^{4}He[10^{-4} cc/g]*	^{3}He/^{4}He	^{20}Ne[10^{-6} cc/g]*	^{20}Ne/^{22}Ne	^{21}Ne/^{22}Ne	^{36}Ar[10^{-6} cc/g]*	^{38}Ar/^{36}Ar	^{40}Ar/^{36}Ar
400°C	8.2	0.00023 ±0.00005	2.0	11.29 ± 0.17	0.032 ±0.0021	1.2	0.190 ±0.002	252.4 ± 0.5
700°C	12.4	0.00025 ±0.00004	8.6	12.28 ± 0.14	0.032 ±0.0021	1.3	0.189 ±0.002	168.0 ± 0.4
1000°C	3.0	0.00025 ±0.00007	20.8	11.90 ± 0.11	0.031 ±0.0016	1.9	0.192 ±0.002	80.3 ± 0.1
1300°C	0.1	0.00029 ±0.00006	5.2	11.51 ± 0.17	0.032 ±0.0023	1.1	0.193 ±0.002	152.8 ± 0.3
1700°C	0.03	0.00030 ±0.00127	0.2	10.39 ± 0.43	0.027 ±0.0074	0.3	0.187 ±0.003	286.5 ± 0.5
Total	23.7	0.00024	36.7	11.89	0.032	5.8	0.191	129.9

* Concentrations are determined by dividing absolute amounts of gases by an assumed weight of micrometeorites (0.19 mg).

light noble gases (Table 2). Stepped pyrolysis at increasing temperature is an effective method to separate noble-gas components by differences in retentivity between components. Figure 1 shows variations of He and Ne isotopic ratios during the stepped pyrolysis, which indicates that He and Ne in the $<70\,\mu$m-sized micrometeorites consist in a large part of solar energetic particles (SEP) and minor amounts of solar winds (SW). The highest SW contribution is observed in the 700°C fraction where SW-Ne/SEP-Ne ratio is approximately 0.7 based on the $^{20}Ne/^{22}Ne$ isotopic ratio. For the other temperature fractions, SW-Ne/SEP-Ne ratios are smaller than 0.4. The SEP contribution in micrometeorites apparently larger than that in lunar fines (Srinivasan *et al.*, 1972) which in turn is dominated by Ne with SW-Ne/SEP-Ne ratios higher than 0.7 (Fig. 1). He isotopic ratio also shows higher contribution of SEP in micrometeorites than in lunar fines (Fig. 1).

The low contribution of SW relative to SEP in micrometeorites suggests preferential loss of SW component. Several processes can be listed as the possible mechanisms for the loss of SW such as leaching or erosion of a surface layer while the micrometeorites are in the Antarctic environment, or erosion or abrasion during collection of micrometeorites. But we believe that the most probable process responsible for the loss is frictional heating upon atmospheric entry. Assuming that micrometeorites were exposed to solar wind as small particles as suggested by Ne

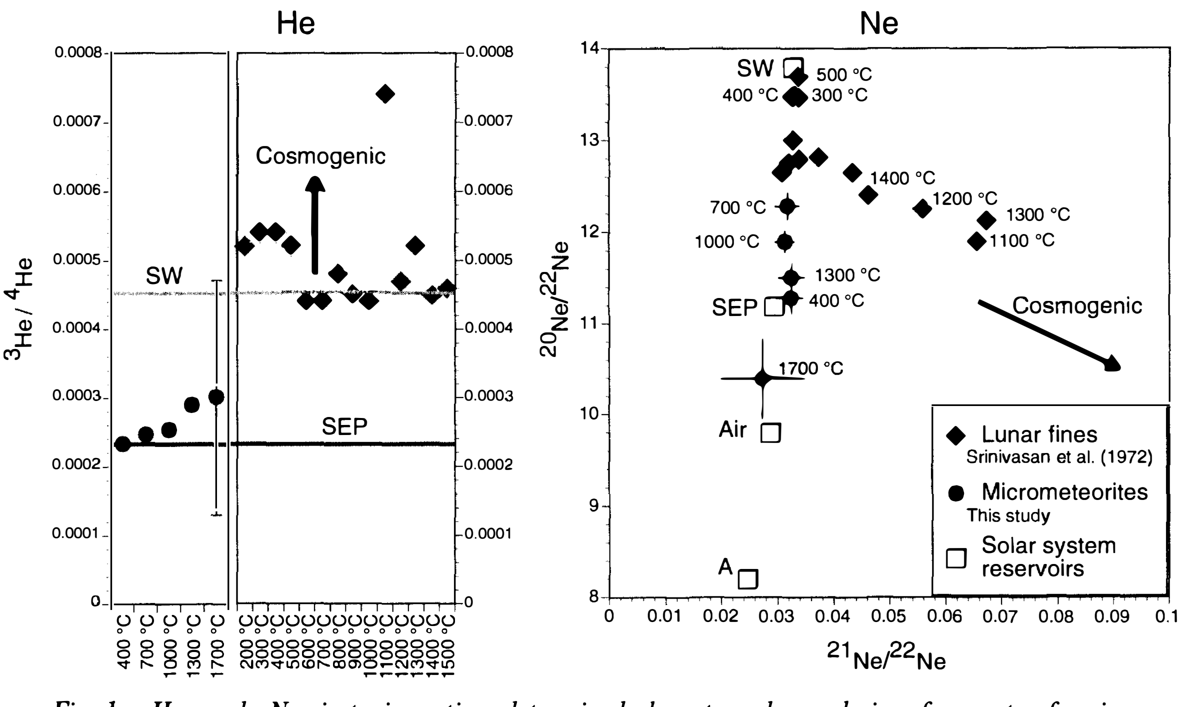

Fig. 1. He and Ne isotopic ratios determined by stepped pyrolysis of a set of micro-meteorite-bearing particles less than 70 µm in diameter, showing He and Ne in micrometeorites consist mainly of SW and SEP gases. Also shown are the data of lunar fines (Srinivasan et al., 1972). Plots of lunar fines are sifted towards cosmogenic components at high temperatures, but those of micrometeorites are not, indicating a very small contribution of cosmogenic gases in micrometeorites. SW and SEP values are from Benkert et al. (1993) and Ne-A is from Pepin (1967).

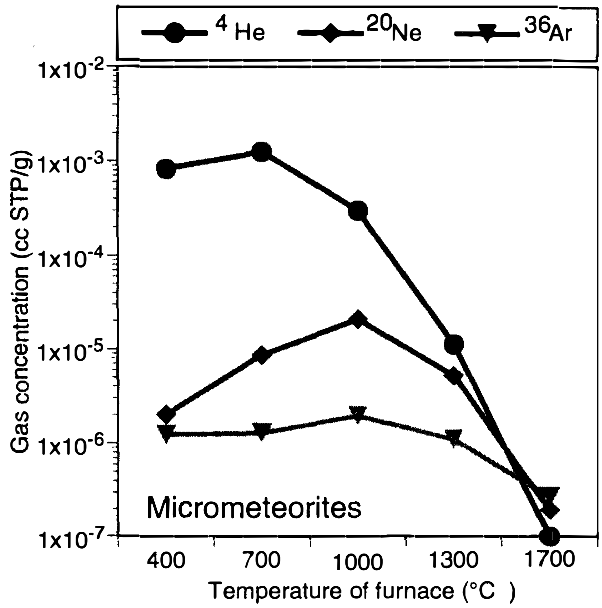

Fig. 2. Release patterns of ^{4}He, ^{20}Ne, and ^{36}Ar from micrometeorite-bearing particles less than 70 μm in diameter.

analyses of individual micrometeorites (Olinger *et al.*, 1990; Nier *et al.*, 1990; Maurette *et al.*, 1991; Osawa *et al.*, 2000), SW gases were implanted in the uppermost surface layers of the particles, while SEP gases were distributed throughout the particles. In space, micrometeorites might have had a high SW-Ne/SEP-Ne ratio similar to the incoming flux ratio from the Sun (>14.2 from Wieler *et al.*, 1986). During the atmospheric entry the temperature of the surface layers of micrometeorites were preferentially elevated due to short duration of heating (*e.g.*, Love and Brownlee, 1991). In fact, micrometeorites showing evidence of melting only in the surface portions commonly occur (*e.g.*, Imae *et al.*, 1999). SW gases were easily lost from the hot surfaces, resulting in the low SW-Ne/SEP-Ne ratios in the micrometeorites reached to the Earth.

The loss of SW is also confirmed by release patterns of stepped heating analysis. If SW gases were retained in micrometeorites, the largest releases should be observed at low temperatures, because during exposure to solar winds the low-energy, low-retentive SW gases would have been much abundant in micrometeorites relative to high energy SEP gases. However, the largest releases of ^{4}He, ^{20}Ne, and ^{36}Ar are observed in the middle temperature fractions of 700 and 1000°C, respectively (Fig. 2). It indicates that the low-retentive SW gases have been lost from micrometeorites, being consistent with a tendency derived from He and Ne isotopic ratios. The degrees of gas loss can be estimated from elemental ratios: $^{4}He/^{20}Ne$ ratio of micrometeorites is 65 that is far below compared with that of contemporary solar winds recorded in Al-foils of satellites (570 from Geiss, 1973). No loss of ^{20}Ne during the atmospheric heating corresponds to 89% loss of ^{4}He from micrometeorites, but ^{20}Ne was also lost during the heating as deduced from Ne isotopic ratios, thus the loss of ^{4}He is higher than 89%.

3.2. Cosmogenic gases in micrometeorites

Cosmogenic Ne in solar-gas rich chondritic meteorites and lunar fines is released mainly at temperatures higher than 1000°C in stepped heating analysis as shown in Fig. 1. Most solar gases were already released at temperatures lower than 1000°C and cosmogenic gases, which are sited throughout the minerals, are increasingly released at high temperatures. Ne compositions in high temperature steps of the Dome-Fuji micrometeorites are, however, plotted close to a mixing line between SW and SEP components (Fig. 1), indicating little contribution of cosmogenic gases. Reedy (2000) has shown that very small chondritic objects such as cosmic dust have high ^{21}Ne production rates, *i.e.* 1.07×10^{-8} cc STP/g Ma at 1AU, induced by solar protons of SEP. ^{21}Ne production rates induced by galactic cosmic rays (GCR) in such small particles was calculated to be 0.08×10^{-8} cc STP/g Ma at 1AU (^{21}Ne production rate in zero-cm particles in Graf *et al.*, 1990) which is an order of magnitude lower than that by SEP. When we take 1.15×10^{-8} cc STP/g Ma as a total ^{21}Ne production rate in micrometeorites, cosmic ray exposure ages are calculated to be 0.4 ± 0.5 and 2.2 ± 1.0 Ma for sets of micrometeorites, $<70\,\mu$m and 70–200 μm in diameter, respectively. The longer exposure age for 70–200 μm-sized micrometeorites than $<70\,\mu$m-sized ones is consistent with the predictions from the Poynting-Robertson effect that cosmic-dust lifetime in the solar system is proportional to diameters of dust particles in space.

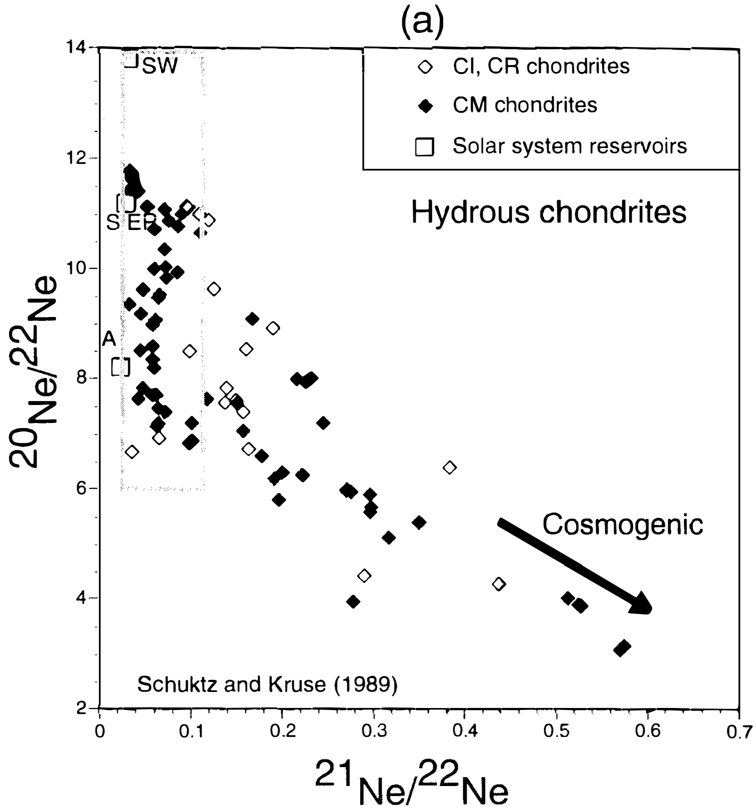

Fig. 3a. Ne isotopic ratios of the bulk CM, CR, and CI chondrites (data from Schultz and Kruse, 1989). The area enclosed by gray lines is enlarged in (b).

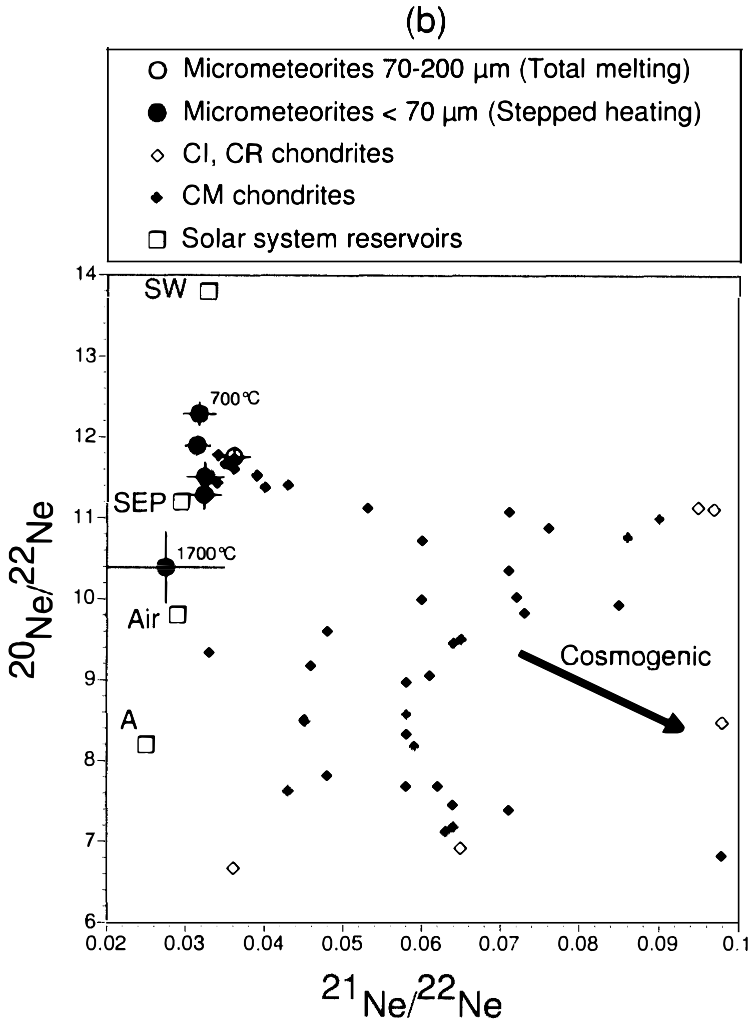

Fig. 3b. Ne isotopic ratios of the Dome-Fuji micrometeorites and hydrous carbonaceous chondrites.

3.3. Terrestrial noble gas abundance

Terrestrial artificial and natural particles are main constituents of micrometeorite-bearing particles analyzed in the present investigation. Thus noble gases measured contain terrestrial noble gases. In this section we calculate terrestrial He and Ne abundance in the measured concentrations. If all measured ^{40}Ar in the analysis of $<70\,\mu m$ particles is assumed to be terrestrial (Table 2) and the terrestrial gases have atmospheric $^{4}He/^{20}Ne$ and $^{20}Ne/^{40}Ar$ elemental ratios, 0.02 and 4% of total ^{4}He and ^{20}Ne are terrestrial. These proportions are obtained by neglecting the presence of extraterrestrial ^{40}Ar, therefore these values are upper limits of terrestrial ^{4}He and ^{20}Ne abundance in the measured concentrations. The upper limits are small enough not to disturb any discussion stated above, thus we report He and Ne concentrations and isotopic ratios without any correction for terrestrial noble gases, except for blank correction at 1700°C (Tables 1 and 2). However, terrestrial gases appear to occupy a major fraction of total ^{36}Ar, 54% at most, thus ^{36}Ar abundance shown in Fig. 2 is corrected one for terrestrial ^{36}Ar.

3.4. Comparison to hydrous carbonaceous chondrites

Previous studies have shown that a large fraction of micrometeorites is basically similar in mineralogy, mineral chemistry, and bulk elemental abundance to hydrous carbonaceous chondrites (*e.g.*, Maurette *et al.*, 1991, 1994; Steele, 1992; Kurat *et al.*, 1994; Greshake *et al.*, 1996; Genge *et al.*, 1997; Yano and Noguchi, 1998; England and Maurette, 1998). But no studies compare noble-gas composition of micrometeorites with those of hydrous carbonaceous chondrites. The distribution of Ne isotopic ratios of bulk hydrous carbonaceous chondrites of CM, CR, and CI types indicates that Ne in these chondrites have a wide range of isotopic ratio (Fig. 3a; data from Schultz and Kruze, 1989). On the other hand, Ne in micrometeorites has isotopic ratios indicative of an ultimate enrichment of solar-type Ne (Fig. 3b). The difference strongly indicates that micrometeorites are not particles produced by break-up of hydrous carbonaceous chondrites during the atmospheric heating, since a set of such particles must have Ne isotopic ratio close to an average value of hydrous carbonaceous chondrites that is enriched in cosmogenic gases (Figs. 3a and 4). The high ratio of solar/cosmogenic gas in micrometeorites suggests that they had been small particles in the interplanetary space to have wide surface areas to be exposed to solar winds (Fig. 4), which is consistent

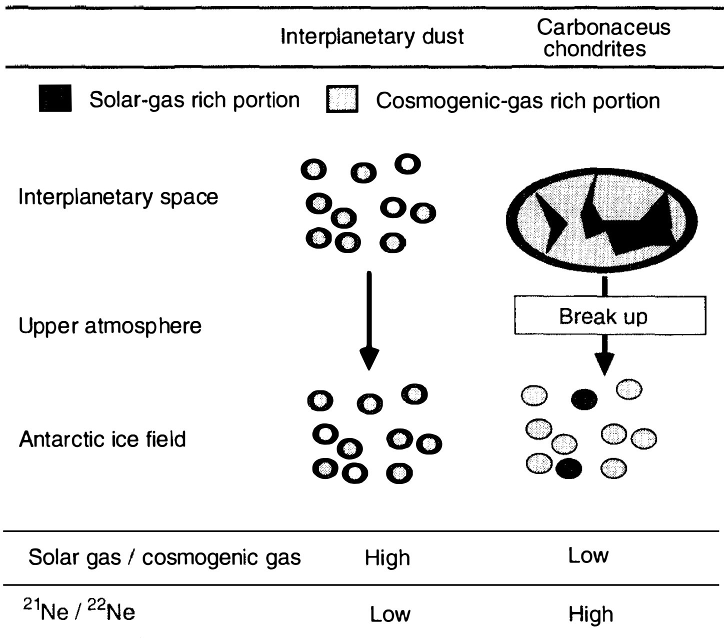

Fig. 4. A schematic illustration showing two possible processes of micrometeorite formation. In the case of interplanetary dust, micrometeorites are small particles in space and are exposed to solar winds and cosmic rays, resulting in a high ratio of solar/cosmogenic gas and a low $^{21}Ne/^{22}Ne$ ratio. On the other hand, in the case of carbonaceous chondrites, micrometeorites are particles produced in the atmosphere by disruption of hydrated carbonaceous chondrites in which solar-type noble gases, that were implanted by precompaction irradiation, were heterogeneously distributed (Nakamura et al., 1999b, c). A set of micrometeorites generated in the process should have Ne isotopic ratio similar to that of an average value of hydrated carbonaceous chondrites. But the observed Ne isotopic ratios of micrometeorites are in favor of the case of interplanetary dust.

with the interpretation derived from Ne analysis of individual micrometeorites (Olinger *et al.*, 1990; Nier *et al.*, 1990; Maurette *et al.*, 1991; Osawa *et al.*, 2000).

In summary, micrometeorites collected around the Dome Fuji Station are those having fallen to the Earth in recent fifty years (Nakamura *et al.*, 1999a). Short duration of exposure to SEP and GCR indicates that the micrometeorites had been buried inside of the parental objects at depth more than a few meters to which GCR could not reach, and that they have emerged as small particles in the modern solar system and come to the Earth after short traverse in space. The longer exposure age of 70–200 μm-sized micrometeorites than $<70\,\mu$m-sized ones is consistent with the predictions from the Poynting-Robertson effect.

Acknowledgments

We are indebted to Drs. T. Noguchi, N. Imae, and K. Terada for arrangements of a consortium study on the Dome-Fuji micrometeorites, and to Drs. K. Nagao and R. Okazaki for technical supports during an installation of the noble-gas mass spectrometer. Thanks are also due to Drs. Y. N. Miura and G. J. Flynn for reviewing the manuscript and Messrs. W. Nozaki and K. Shimada for assistance and support during the course of this study. This work has been supported by the Grant-in-aid of the Japan Ministry of Education, Science and Culture to TN (No. 11740303) and to NT and TN (No. 09304055).

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(Received August 25, 1999; Revised manuscript received December 13, 1999)