

Antarct. Meteorite Res., **13**, 170–176, 2000

Neon isotopic composition of carbon residues from the Canyon Diablo iron meteorite

Miwa Namba¹, Teruyuki Maruoka¹, Sachiko Amari²
and Jun-ichi Matsuda^{1*}

¹*Department of Earth and Space Science, Graduate School of Science,
Osaka University, Machikaneyama, Toyonaka 560-0043*

²*Laboratory for Space Sciences and the Physics Department, Washington
University, St. Louis, MO 63130-4899, U.S.A.*

Abstract: We analyzed noble gases in acid residues extracted from the Canyon Diablo iron meteorite by density and colloidal separation. The results do not indicate any significant variation among the samples although spallogenic noble gases were effectively removed by the treatment. The isotopic compositions of the heavy noble gases are close to those of the terrestrial atmosphere, suggesting that the samples were seriously contaminated by atmospheric noble gases during the treatment. However, the Ne isotopic ratios fall between atmospheric Ne and planetary Ne, suggesting that the primordial component of the noble gases in carbonaceous material in Canyon Diablo is planetary rather than solar.

1. Introduction

Elemental and isotopic abundances of noble gases in iron meteorites have been extensively studied in order to investigate their origin and evolution. After Paneth *et al.* (1952) reported the presence of spallogenic ³He in iron meteorites, there have been many reports on the spallation component of noble gases in iron meteorites (e.g., Voshage and Feldmann, 1978). Light noble gases (helium, neon and argon) in iron meteorites are characterized by a huge amount of cosmic-ray produced nuclides and their analyses provide us the information on exposure ages, the location of iron meteorites inside the parent bodies, the preatmospheric size of the meteorites etc. On the other hand, primordial components of noble gases are masked by a large amount of such spallogenic components, and have not been well characterized. An “atmosphere-like” trapped component was reported in iron meteorites (Hennecke and Manuel, 1977) and in acid residues of iron meteorites (Murty *et al.*, 1983). If the atmosphere-like component is really present in iron meteorites and it is indigenous, it may give us a clue to understand the evolution of the terrestrial atmosphere. However, it is not clear that the effect of adsorption of terrestrial atmosphere could be totally excluded (Fisher, 1981; Matsuda *et al.*, 1996).

* Corresponding author (E-mail: matsuda@ess.sci.osaka-u.ac.jp).

The purpose of this work is to investigate the primordial component in IAB iron meteorites and to study the carrier and the origin of noble gases in carbonaceous material of the Canyon Diablo IAB iron meteorite. First, we chemically removed metal and silicates to obtain carbonaceous material in the meteorite, and then separated it into various kinds of carbon by density separation. We chose Canyon Diablo as the sample because it has been well studied and a wealth of data on the meteorite are available to compare our result with them.

2. Separation by chemical procedures

A flow chart of our chemical procedure is shown in Fig. 1. The Canyon Diablo iron meteorite, 73.67 g of a chunk, was treated with 12N HCl to dissolve the metal phase. As the residual fraction might contain silicates, it was demineralized with 10N HF-1N HCl, treated with 1N HCl to remove any reaction products of fluorides, and finally treated with aqua regia to dissolve any residual metal. We named the residue "Can4an".

Graphite was identified in Can4an by X-ray diffraction analysis. However, TG-DTA (Thermogravimetric-Differential Thermal Analyzer) analysis (Hirata, personal. comm.) pointed out that the sample was also enriched in hydrocarbon, presumably produced during the acid treatments. In order to remove the hydrocar-

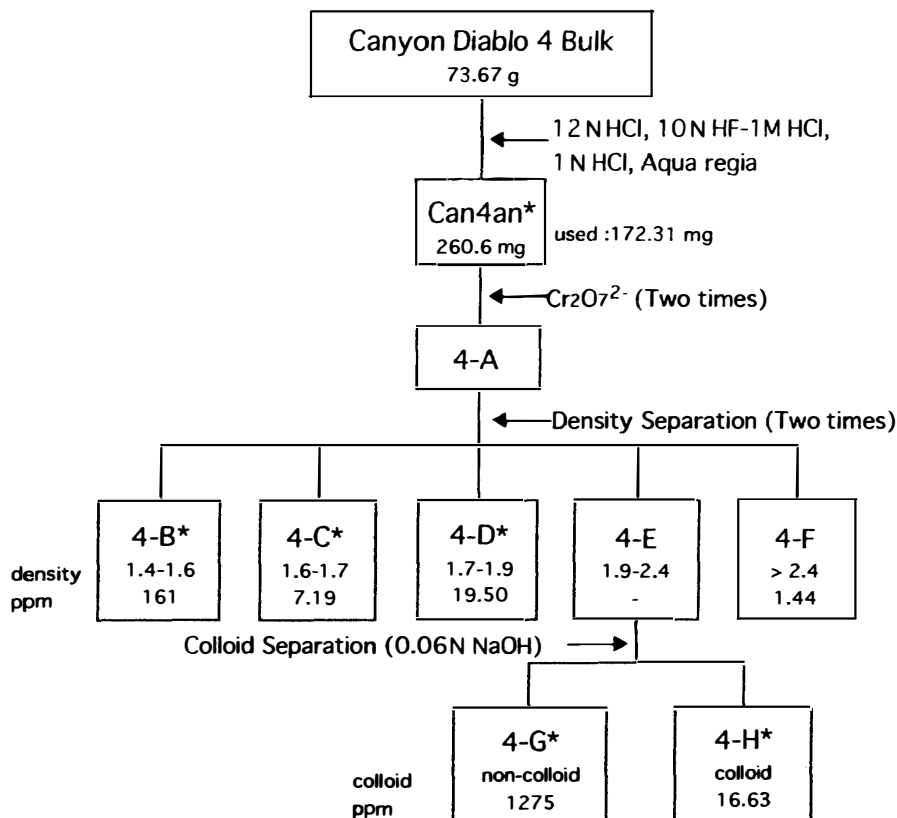


Fig. 1. Flow chart of the chemical treatments. Fractions marked by an asterisk (*) were analyzed for noble gases.

bon, the residue was twice oxidized with $\text{Cr}_2\text{O}_7^{2-}$ at 85°C . We designated the remaining oxidized fraction "4-A". The sample 4-A was separated into five density fractions (4-B, 4-C, 4-D, 4-E, and 4-F) in a stepped density gradient of sodium polytungstate ($3\text{Na}_2\text{WO}_4 \cdot 9\text{WO}_3 \cdot \text{H}_2\text{O}$) with about 0.5% by weight of Brij 35 surfactant.

4-E comprises about 87% of the mass of 4-A. Thus, we further separated the fraction into two fractions: it was suspended in 0.06N NaOH and was centrifuged ten times to separate microdiamonds, yielding the non-colloidal fraction 4-G and the colloid fraction 4-H.

3. Noble gas analyses and results

Analyses of the samples were performed with the mass spectrometer VG5400 connected to an all-metal extraction system at Osaka University. The analytical procedure is described in detail in our previous papers (*e.g.*, Matsuda *et al.*, 1989; Maruoka and Matsuda, 1995). Discrimination factors and sensitivities of the mass spectrometer were monitored by measuring known amounts of an air standard. Hot blanks were measured before the sample measurement for each temperature step.

Results of the noble gas analyses are listed in Table 1, together with the data of a graphite nodule from Canyon Diablo (Alexander and Manuel, 1967; Bennett and Manuel, 1967) and the atmosphere. The tabulated values have been corrected for hot blanks. In cases where the hot blank exceeds more than 40% of the measured values, the elemental abundances are expressed as upper limits and the blank correction was not carried out for isotopic ratios. However, both isotopic ratios and elemental abundances in total fractions have been calculated from the blank-corrected values if the blank correction was less than 40% for the total fractions. Isotopic analysis was not performed for He. Isotopic compositions of Kr and Xe in the samples were very close to those of the atmosphere. In addition to atmospheric Ar we observe a small amount of radiogenic ^{40}Ar (Table 1). The isotopic ratios of these noble gases show no systematic variations among the samples. Thus, we only list the elemental abundances of all noble gases and the isotopic data of Ne and Ar.

The amounts of noble gases in the fractions 4-C, 4-D and 4-H are shown by upper limits in many temperature steps. This is because their sample sizes were about 1/50 of those of Can4an or 4-G, and their gas amounts were very small. The amount of gases released at 800°C was much larger than that at 2000°C in Can4an. Similar trends were observed, especially for Ar in all samples except for 4-G and 4-H. All noble gases in 4-G and Kr in 4-H were mostly released at 1600°C .

4. Implications from isotopic ratios of noble gases

Isotopic compositions of Ar, Kr and Xe were very close to those in the terrestrial atmosphere. This is probably due to the adsorption of atmospheric noble gases on the surface of carbon material during the chemical treatment, which is supported by the fact that these noble gases were mainly released at the lowest

Table 1. Noble gases in acid residue in Canyon Diablo.

Sample	Weight (g)	Tem. (°C)	[⁴ He]	[²⁰ Ne]	²⁰ Ne/ ²² Ne	²¹ Ne/ ²² Ne	[³⁶ Ar]	³⁸ Ar/ ³⁶ Ar	⁴⁰ Ar/ ³⁶ Ar	[⁸⁴ Kr]	[¹³² Xe]
Can4an	0.03483	800	620	0.47	7.8	0.033	1.4	0.1876	310	3.3	1.5
					±1.9	±0.010		±0.0014	+13		
		1600	0.17	0.030	9.1	0.0318	0.22	0.1868	310	1.0	0.16
					±1.4	±0.0054		±0.0015	+13		
	2000	-	0.014	8.1	0.0256	0.064	0.1854	324	0.25	0.065	
				±1.0	±0.0069		±0.0020	+14			
	total	620	0.52	7.9	0.0329	1.7	0.1874	310	4.6	1.7	
				±1.8	±0.0098		±0.0012	±11			
4-B	0.00473	800	32	<0.091	7.52	0.046	1.2	0.1895	318	<0.69	0.35
					±0.15	±0.029		±0.0017	±13		
		2000	-	0.16	10.9	0.0171	0.27	0.1841	325	1.1	<0.16
				±2.0	±0.0084		±0.0031	±14			
	total	32	0.21	10.9	0.025	1.4	0.1885	319	1.4	0.40	
				±2.0	±0.010		±0.0015	±11			
4-C	0.00016	800	<390	<1.6	6.45	0.0174	6.0	0.1883	311	<14	<6.9
					±0.15	±0.0033		±0.0029	±13		
		2000	-	3.5	9.58	0.025	<9.8	0.1902	294	<33	<5.8
				±1.67	±0.012		±0.0081	±13			
	total	<390	3.6	-	-	10	0.1874	311	<47	<13	
							±0.0079	±15			
4-D	0.00035	800	<170	<0.98	7.32	0.0146	4.2	0.199	309	<7.8	2.5
					±0.13	±0.0040		±0.015	±13		
		2000	<190	1.6	9.4	0.0203	3.7	0.1862	315	<16	<2.8
				±1.6	±0.0074		±0.0026	±14			
	total	<360	2.0	10.0	0.0192	7.8	0.1928	312	<23	3.8	
				±1.9	±0.0087		±0.0083	±11			
4-G	0.02533	800	-	0.14	9.54	0.0302	0.41	0.1860	310.5	1.4	0.31
					±0.51	±0.0034		±0.0016	±2.8		
		1600	-	0.18	9.61	0.0238	0.52	0.1963	312.0	2.5	0.53
				±0.46	±0.0020		±0.0015	±3.0			
	2000	-	0.11	10.3	0.0310	<0.24	0.1870	310.4	<0.49	0.074	
				±0.8	±0.0050		±0.0015	±2.8			
	total	-	0.42	9.75	0.0277	1.1	0.1862	311.8	4.2	0.92	
				±0.32	±0.0019		±0.0010	±2.0			
4-H	0.00067	800	-	<0.58	5.88	0.0171	6.0	0.1847	318.9	5.6	<1.9
					±0.10	±0.0011		±0.0023	±6.2		
		1600	-	0.78	9.1	0.024	<2.2	0.1907	305.0	19	<4.4
				±2.5	±0.012		±0.0092	±3.0			
	2000	-	0.85	7.19	0.027	<3.3	0.1874	306.7	<7.0	<2.3	
				±0.60	±0.014		±0.0015	±3.0			
	total	-	1.9	7.80	0.026	8.0	0.1850	320.9	24	4.0	
				±0.94	±0.010		±0.0019	±6.1			
graphite nodule*			94	1.8	1.03	0.774	-	-	-	24	7.6
Terrestrial air					9.8	0.029		0.1880	295.5		

The symbol "<" shows upper limit. No blank correction was made for the isotopic ratios when the elemental abundance was given by upper limit.

Gas concentrations are in the units of 10⁻⁸cm³STP/g for He, Ne and Ar, and 10⁻¹⁰cm³STP/g for Kr and Xe.

* Helium and Ne are from Bennet and Manuel (1967), Kr and Xe are from Alexander and Manuel (1967).

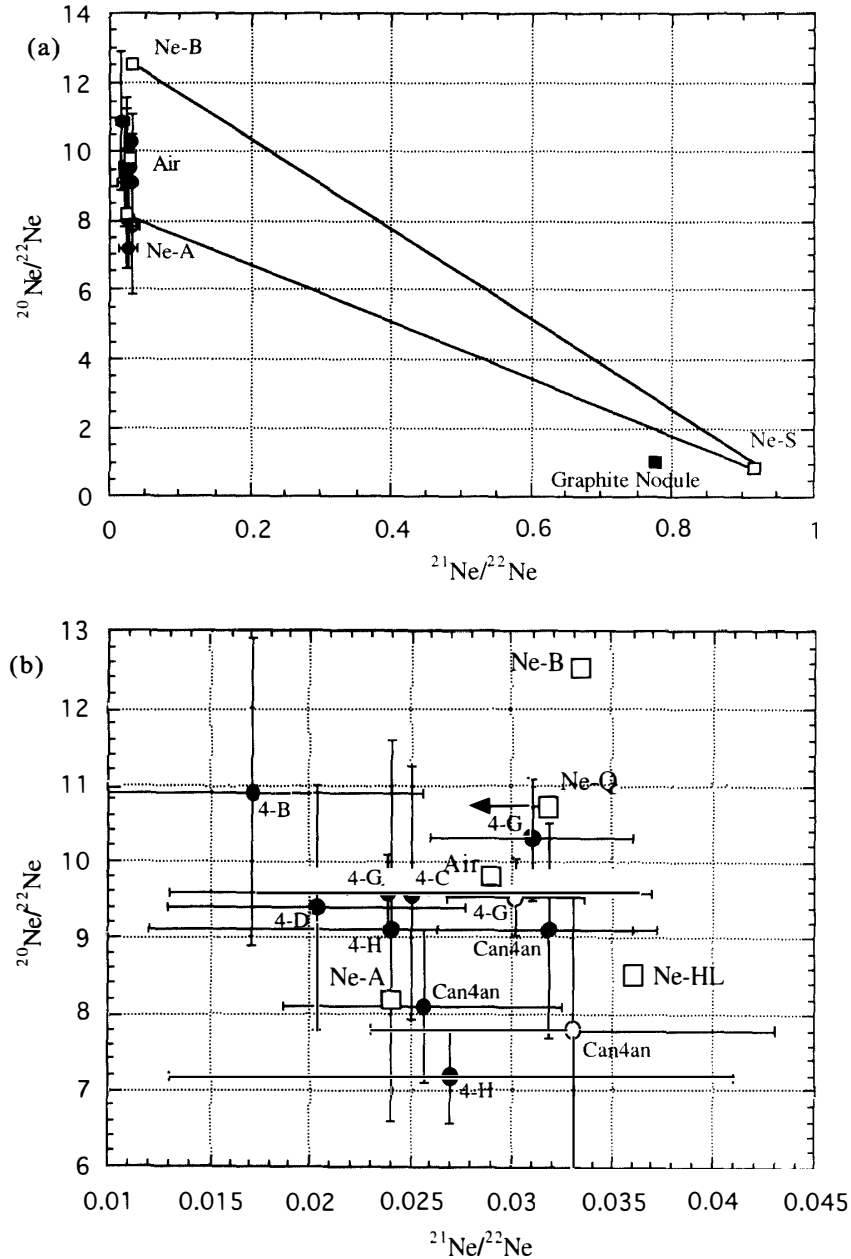


Fig. 2. (a) Neon three-isotope plot of $^{20}\text{Ne}/^{22}\text{Ne}$ vs. $^{21}\text{Ne}/^{22}\text{Ne}$ and (b) its enlarged diagram. Only the data with blank corrections were plotted with open circles (800°C) and closed circles (1600 and 2000°C). The closed square is a graphite nodule from Canyon Diablo (Bennet and Manuel, 1967). Ne-A is planetary Ne (Black and Pepin, 1969) and Ne-B is solar Ne (Black, 1972). Spallogenic Ne (Ne-S) is from Mazor et al. (1970). Ne-Q (Wieler et al., 1991) and Ne-HL (Ne-A2 in Huss and Lewis (1994)) are plotted only in (b). The $^{21}\text{Ne}/^{22}\text{Ne}$ ratio in Ne-Q has been obtained only as the upper limit.

temperature fractions. It seems that the adsorbed noble gases were so predominant that it masked primordial components of heavy noble gases in our samples.

However, several interesting trends have emerged from the Ne isotopic data. We have plotted our Ne data in the Ne three-isotope diagram (Fig. 2). The

spallogenic component is absent in our fractions since all the data points fall on the far left side in Fig. 2a. Spallogenic components are predominant in the light noble gases (He, Ne and Ar) in bulk iron meteorites. The presence of the spallogenic component of Ne has been reported in a graphite nodule from the Canyon Diablo iron meteorite (Bennet and Manuel, 1967), and also in graphite samples chemically separated from the El Taco iron meteorite (Mathew and Begemann, 1995). In this work, the spallogenic component has been effectively removed by our chemical treatment.

No systematic variations among different sample fractions have been observed, partly due to large errors of the data (Fig. 2b). However, it should be noted that the data points are not located between the atmosphere and Ne-B (solar component) (Black, 1972), but fall between the atmosphere and Ne-A (planetary component) (Black and Pepin, 1969). Our results suggest that the primordial noble gas component of Canyon Diablo is planetary rather than solar. It is believed that the planetary component is not a single component but that it consists of various components such as Ne-A1 (Alaerts *et al.*, 1980), Ne-Q and Ne-HL. As indicated in Fig. 2b, only an upper limit has been obtained for the $^{21}\text{Ne}/^{22}\text{Ne}$ ratio of Ne-Q, because of the addition of cosmogenic ^{21}Ne (Wieler *et al.*, 1991). Neon-HL (Ne-A2 in Huss and Lewis, 1994) is carried by presolar diamonds. Thus, the primordial Ne in our samples seem to be the mixture of these components, suggesting that the carbon material in Canyon Diablo formed under the circumstance where Q and presolar diamonds were present and that there has been no effect of solar wind irradiation. However, air contamination in the samples is still very large, overwhelming the primordial component in the samples. Further experiments should be necessary to obtain clear evidence to identify the primordial component in the graphite.

Acknowledgments

We would like to thank Prof. K. Nagao for his useful comments.

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(Received August 4, 1999; Revised manuscript received December 13, 1999)