SEARCH FOR THE EXTRA-TERRESTRIAL MATERIALS IN DEEP SEA SEDIMENTS

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Abstract: In order to characterize He carrier in deep sea sediments, we magnetically separated the deep sea sediments collected at three different sites, and measured the $^3He/^4He$ ratio and the He content in each fraction. The $^3He/^4He$ ratios ($1.4–2.4 \times 10^{-4}$) and the He content (^3He: $1.6–10.7 \times 10^{-9}$ cm^3 STP/g) in the magnetic fractions were high, which reveals that the He carrier was concentrated in the magnetic fractions. The ferromagnetic property of the He carrier may be due to magnetite which had been produced by decomposition of poorly crystallized silicates in the He carrier during their atmospheric entry. Stepwise heating experiment was applied to one of the magnetic fractions. The $^3He/^4He$ ratios were almost the same (2×10^{-4}) in each temperature fraction and the single activation energy, 17 kcal/mol was obtained. The result strongly suggests that the observed He is of a single component. Thermomagnetic analyses gave us the information that the major part of the magnetic fractions is terrestrial titanomagnetite. The amount of ^{53}Mn in the magnetic fractions was measured by the RNAA method. However, the large amount of ^{55}Mn allowed us only to estimate the upper limits of the amount of ^{53}Mn.

1. Introduction

The $^3He/^4He$ ratios in extra-terrestrial materials are in general significantly higher than those in terrestrial materials, the ratio being larger than 1×10^{-4}. For example, the ratio of He-A which is regarded to represent the primordial He trapped in meteorites is about 1.4×10^{-4}, and the solar wind has the ratio of 4×10^{-4} (*e.g.* OZIMA and PODOSEK, 1983). The $^3He/^4He$ ratios in iron meteorites (which abound in cosmogenic He) are even much higher ($\sim 10^{-1}$) (SCHULTZ *et al.*, 1971). On the other hand, the $^3He/^4He$ isotopic ratios in terrestrial materials vary from 10^{-5} to 10^{-8}. For example, the $^3He/^4He$ ratios of MORB (Mid-Oceanic Ridge Basalts) which are considered to have been derived from the depleted mantle are in the range of $1.1–1.4 \times 10^{-4}$ (*e.g.* OZIMA and PODOSEK, 1983). Phenocrysts from the Haleakala Volcano in Hawaii have the $^3He/^4He$ ratios of $2–5 \times 10^{-5}$ (KANEOKA and TAKAOKA, 1980). This is considered that the phenocrysts have been derived from the undepleted mantle. Excess ^3He compared with air has been observed in deep sea water (CLARKE *et al.*, 1969). It is considered that the juvenile ^3He has been transported from the earth's interior.

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As mentioned above, the primordial materials in the earth have high ^{3}He/^{4}He ratios. On the other hand, the rocks which abound in U and Th have much larger amounts of ^{4}He, which makes the ^{3}He/^{4}He ratios much lower. Helium isotopic ratios in deep sea sediments have been measured by several investigators (MERRIHUE, 1964; KRYLOV et al., 1973; OZIMA et al., 1984) and it is now well established that deep sea sediments are characterized by considerably high ^{3}He/^{4}He ratios. OZIMA et al. (1984) suggested that these high ratios are caused by the incorporation of the extra-terrestrial materials, such as interplanetary dust particles (IDPs), in sediments. Since the high ^{3}He/^{4}He ratio is more pronounced in the magnetic fraction of deep sea sediments (MERRIHUE, 1964), we made careful magnetic separation of some deep sea sediments and tried to characterize the extra-terrestrial materials in these magnetic fractions.

2. Experimental and Results

2.1. Sample descriptions and preparations

Deep sea sediments from three different sites in the Pacific Ocean were analyzed. That is (1) the piston cored sediment (KOBAYASHI et al., 1980); KH75-3-5-2 (26°00.8'N, 150°00.0'E, uncorrected depth=5850 m, core length=907 cm), which is sampled by the R. V. HAKUHO MARU. For the analyses 662–711 cm intervals were used. (2) the box-cored sediment which is siliceous fossil-rich clay (MIZUNO et al., 1981); GH79-1-1476 (9°49.96'N, 167°17.68'W, corrected depth=5151 m); and (3) the box-cored sediment which is described as very dark brown siliceous and calcareous fossil-rich mud (MIZUNO et al., 1982); GH80-1-1627 (5°27.32'S, 163°46.01'W, corrected depth =4995 m). A hand magnet was used to separate magnetic fractions from the pulverized samples suspended in distilled water. In the case of KH75-3-5-2, we first separated the sediments into non-magnetic and magnetic fractions. After that, by a few hand magnets, three different grades of magnetic fractions were further separated from the magnetic fraction according to the intensity of their magnetization, i.e. M1, M2, M3, where the smaller figure corresponds to the stronger magnetization. Their weight fractions were about 0.22%, 0.09% and 0.27%, respectively. M1 was relatively abundant in particles smaller than 10 μm, compared with M2 and M3. The two box-cored samples (GH79-1-1476, GH80-1-1627) were separated into only two fractions—i.e. magnetic and non-magnetic fractions. In the case of the magnetic fraction of GH80-1-1627 (GH80M), the size of majority of the particles was a few microns, but in the magnetic fraction of GH79-1-1476 (GH79M), the particle size ranged from 50 to 100 μm, with few particles smaller than 10 μm.

2.2. Noble gas analyses

Helium isotopic ratios and the amount of He and ^{20}Ne were measured (Table 1), with a single-focusing, 60° deflection, 6'' radius mass spectrometer with a double collector. Experimental procedures for the gas extraction and the mass spectrometric analyses were essentially the same as those described previously (OZIMA et al., 1983). For KH75-3-5-2, the ^{3}He/^{4}He ratio in the non-magnetic fraction is slightly lower than that in the bulk sediment, though it is still much higher than ordinary terrestrial rocks. There are two possibilities; one is that not only magnetic minerals but also

Table 1.   Helium isotopic ratios and the amount of He and ^{20}Ne.

Sample[1]	Weight (mg)	$^3He/^4He$ (10^{-4})	^3He $(10^{-10}$ cm³ STP/g)	^4He $(10^{-8}$ cm³ STP/g)	^{20}Ne $(10^{-8}$ cm³ STP/g)
KH75-3-5-2					
Interval: 662–711 cm					
Bulk	101.6				
total		0.659	0.258	0.392	
		±0.132	±0.085	±0.050	
NM	104.5				
total		0.516	0.208	0.402	
		±0.085	±0.058	±0.047	
M1	42.25				
total		2.18	36.10	16.55	5.18
		±0.24	±6.16	±0.98	±0.61
M1	55.85				
520°C		1.99	7.72	3.88	
		±0.13	±1.28	±0.40	
690°C		2.05	10.74	5.25	
		±0.11	±1.62	±0.51	
770°C		1.91	8.11	4.25	
		±0.14	±1.51	±0.49	
860°C		1.90	5.71	3.01	
		±0.13	±1.04	±0.34	
950°C		1.76	3.37	1.91	
		±0.15	±0.73	±0.25	
1030°C		1.12	0.321	0.285	
		±0.46	±0.266	±0.120	
1110°C		3.59[2]	0.185[2]	0.141[2]	
M2	26.3				
total		1.87	5.78	3.10	
		±0.32	±1.68	±0.37	
M3	55.5				
total		1.75	2.15	1.23	
		±0.28	±0.57	±0.13	
GH79-1-1476					
M	41.91				
total		1.38	16.21	11.79	5.97
		±0.07	±2.52	±1.27	±3.17
GH80-1-1627					
M	30.53				
total		2.37	107.3	45.26	43.50
		±0.08	±13.4	±4.07	±8.55

[1] NM: non-magnetic fraction, M: magnetic fraction, M1, M2 and M3 (see the text).

[2] These values give only the upper limits because of the very small amount of the extracted gas.

* Errors represent 1σ including the analytical error in the mass spectrometry and 50% of the hot blank.

non-magnetic minerals in extra-terrestrial materials have high He content.   Another is that only the magnetic minerals are the He carrier.   Since the separation process could not succeed in separating all the magnetic minerals, some portion of the magnetic minerals must still remain in the non-magnetic fraction.   This is further dis-

cussed in the next section.

For KH75-3-5-2, the strongest magnetic fraction, M1 has the highest $^3He/^4He$ ratio and also the highest ^3He content.   From the weight fraction and the ^3He content, we can estimate the volume fraction of gas of each fraction occupied, which reveals that about 30% of ^3He in the bulk sediment was attributable to M1.   It is obvious that the He carrier is concentrated in the magnetic fraction.   Other magnetic fractions of box-cored sediments also have high isotopic ratio and ^3He content.   The isotopic ratios of three magnetic fractions are slightly different from each other.
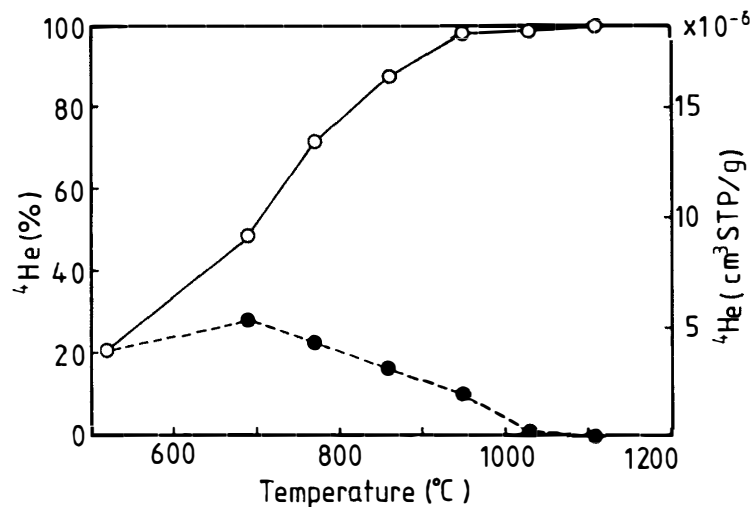

Fig. 1.   Helium thermal release pattern.   Solid line and open circle: cumulative value, dotted line and closed circle: value for each temperature step.
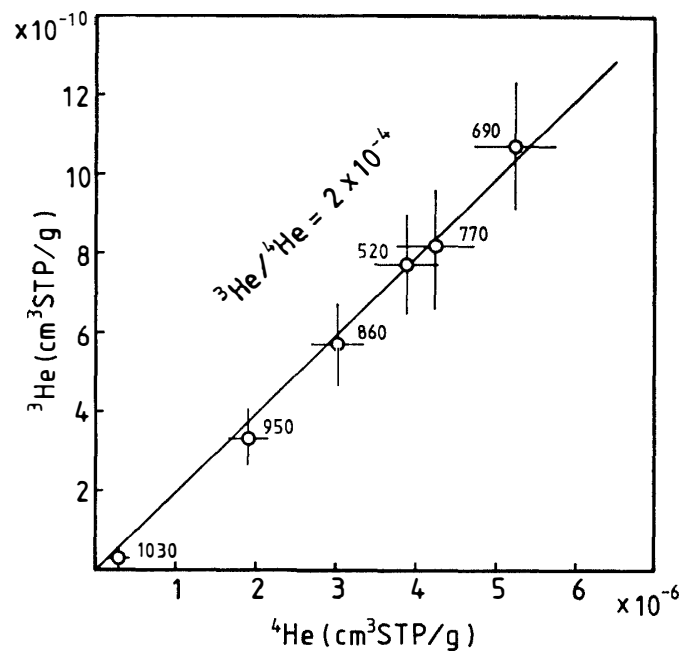

Fig. 2. $^3He/^4He$ correlation diagram.   The amount of ^3He is plotted against the amount of ^4He. Numerical figures attached to each data point indicate temperature (°C).   Error bars represent 1σ.
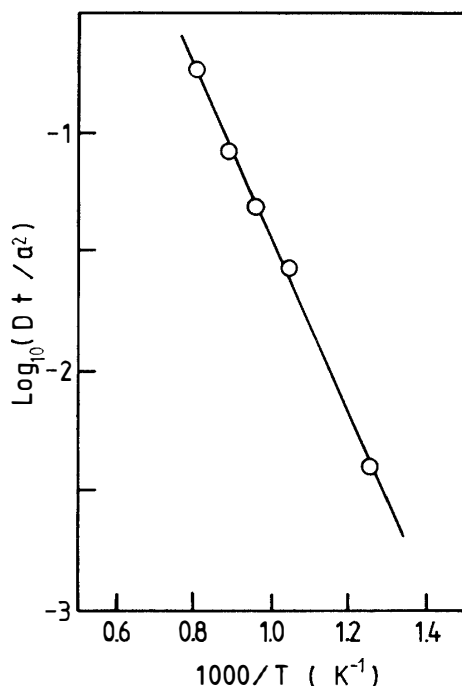
Fig. 3. Arrhenius plot for stepwise heating. The slope yields the activation energy of 17 kcal/mol. D: diffusion coefficient, t: time interval for each temperature step (1h), a: radius of a particle.

The ^{3}He/^{20}Ne ratio ranges from 0.03 to 0.07. They are not so much different either from the planetary ratio (0.03) or the solar wind He implanted in lunar soils (0.09) (e.g. Ozima and Podosek, 1983).

Stepwise heating was applied to M1. The temperature fractions are 520, 690, 770, 860, 950, 1030, 1130 and 1700°C. In Fig. 1 the released fraction of ^{4}He is plotted against temperature. In the highest temperature fraction (1700°C), the released He was indistinguishable from the hot blank. The experimental results show that 10% of the total ^{4}He still remains after 4 steps with 1 hour heating for each. As shown in Table 1, the ^{3}He/^{4}He ratios are almost the same (2×10^{-4}) for most of the temperature fractions. Figure 2 shows the released He has a constant ratio of ^{3}He/^{4}He, and this experimental result strongly suggests that the He is not of mixed components but of a single component. The straight line in the Arrhenius plot (Fig. 3) gives the activation energy of E=17 kcal/mol for the He degassing, or He diffusion. It should be noted that degassing of different components is characterized by different outgassing mechanisms or by different activation energies, supporting the above conclusion that the He in M1 is of a single component.

2.3. Thermomagnetic analyses

Thermomagnetic analyses were made on M1 and GH80M in a vacuum of about 10^{-3} Pa and at a magnetic field H=0.45T. The samples were heated up to 670°C at a heating rate of 6°C/min. We observed almost reversible thermomagnetic curves both on heating and cooling (Fig. 4). The Curie temperature was determined to be about 540°C for M1 and 575°C for GH80M, and the magnetization is 39 Am²/kg and 7 Am²/kg, respectively. From the thermomagnetic curves and the magnetic intensities and also from the chemical analyses by atomic absorption spectroscopy (Table 2), we may conclude that the ferromagnetic separates are essentially magnetite (with a
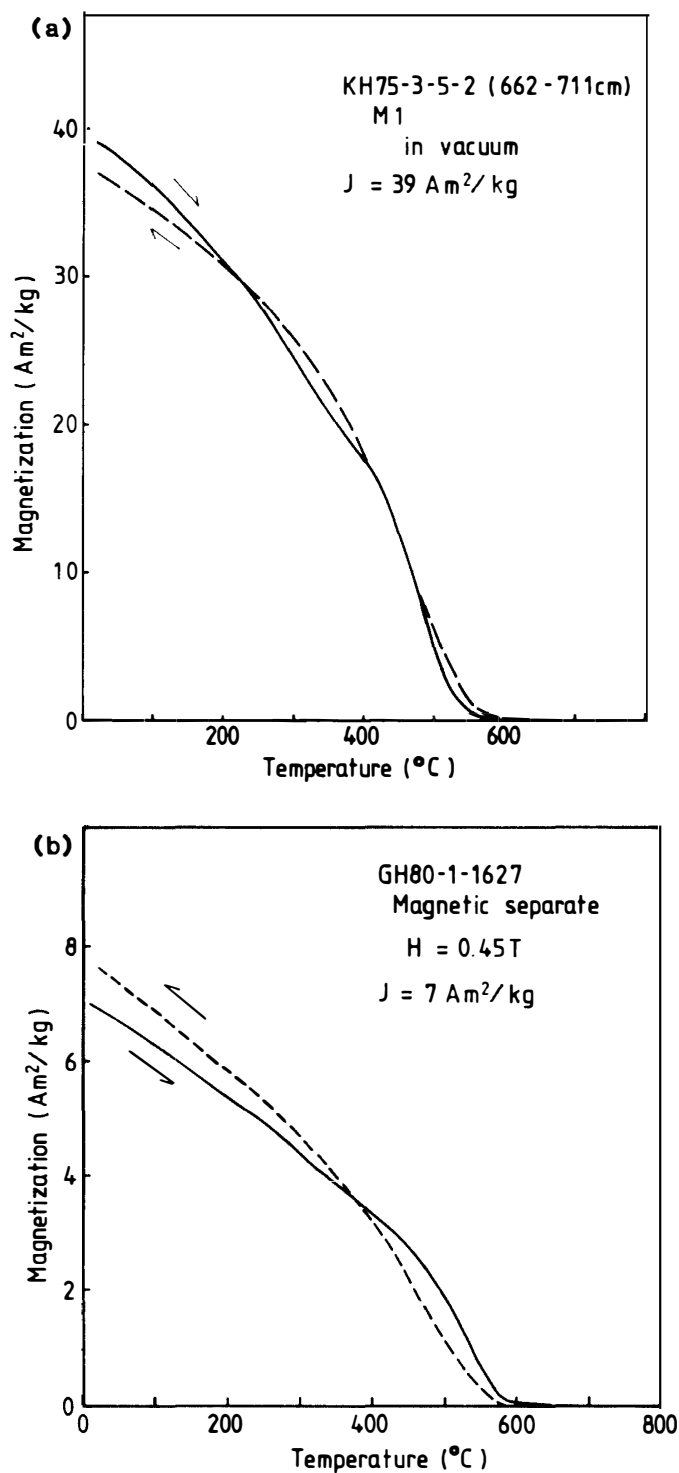
Fig. 4.   Thermomagnetic analysis.   a) for M1 and b) for GH80M respectively.   Solid line: heating curve, dashed line: cooling curve.

small amount of ulvöspinel in M1).   Since it is very common to observe nearly stoichiometric magnetite in deep sea sediments (LØVLIE, 1974), the majority of the ferromagnetic grains are likely to be of terrestrial origin.   It seems that the thermomagnetic curve for the carrier of exotic He is completely masked.

Table 2. The amount of ^{53}Mn and chemical composition.

| Sample | Sample description | | | | Irradiation sample counted | | After irradia-tion ^{54}Mn (cpm) | After correc-tion ^{54}Mn (cpm) | ^{53}Mn (dpm/kg sample) |
	weight (mg)	Fe (%)	Mn (%)	Ni (ppm)	Fe (μg)	Mn (μg)			
M1	33.31	31.17	0.433	4329	0.0923 ± 0.0088	120.15	1.122 ± 0.021	$\leqq 0.0564$	$\leqq 35.31$
GH79M	188.03	22.42	0.243	548	0.4287 ± 0.0179	361.43	3.341 ± 0.054	$\leqq 0.1144$	$\leqq 12.69$
GH80M	97.28	13.90	0.394	288	0.3528 ± 0.0107	313.59	2.869 ± 0.053	$\leqq 0.0733$	$\leqq 15.73$

2.4. ^{53}Mn measurement

Since the magnetic fractions must contain a significant amount of Fe, which would produce cosmogenic nuclide ^{53}Mn, we attempted to measure ^{53}Mn by means of an RNAA method. ^{53}Mn is a cosmic ray induced nuclide produced from iron bombarded by cosmic rays. The half life of ^{53}Mn is 3.7×10^6 years (e.g. HONDA and IMAMURA, 1971). ^{53}Mn is determined via ^{54}Mn which is produced from ^{53}Mn by thermal neutron irradiation. This measurement is interfered by ^{54}Fe and ^{55}Mn which also produce ^{53}Mn by $^{54}Fe(n, p)$ and $^{55}Mn(n, 2n)$. So, prior to the neutron irradiation we must do the chemical separation to reduce the interfering effect of ^{54}Fe and other elements. The samples were decomposed with 6N-HCl, 50% HF and HNO_3 on a hot plate. The residues were dissolved in 6N-HCl. Then a small amount of aliquots were taken in order to determine the concentration of Mn and Fe in the samples by atomic absorption spectrometry. By anion exchange, alkaline earth and other impurities were removed. After that, treatment with TTA (thenoyltrifluoro-acetone) was applied in order to remove Ti. The samples were dried, then the residues were dissolved in aqua regia to decompose organic compounds and then dissolved in HCl. Finally, the residues were put in aluminum foils and dried on a hot plate to change Mn to Mn_3O_4. Samples were irradiated for 11 days with thermal neutron flux of about 2×10^{13} n/cm²·s. After the irradiation, the samples were dissolved in dilute HNO_3 with a few drops of H_2O_2. Then the carrier for Fe, Co was added in each sample. In the anion exchange, the Fe fractions were collected and counted for the purpose of the correction of $^{54}Fe(n, p)^{54}Mn$. A Ge(Li) detector was used for the counting of ^{54}Mn and ^{59}Fe. The experimental procedure was described in detail elsewhere (IMAMURA et al., 1973). The results are shown in Table 2. The proportion of fast neutron to thermal neutron in the reactor was rather high. So the amount of ^{54}Mn produced from ^{55}Mn by fast neutron irradiation is large. Moreover, the contents of ^{55}Mn in the samples were considerably high (Table 2). Therefore, the countings of ^{54}Mn produced from ^{53}Mn were nearly zero after ^{55}Mn correction. So, we can estimate only the upper limits of ^{53}Mn.

3. Discussion

3.1. $^3He/^4He$ ratio

It is observed that the IDPs have high ^4He content, which can be most reasonably

attributed to solar wind implantation (RAJAN *et al.*, 1977). However, the isotopic ratio of the solar wind is 4.0×10^{-4} and that of the solar wind implanted lunar soil is 3.7×10^{-4} (*e.g.* OZIMA and PODOSEK, 1983). As stated in the previous section, the result of the stepwise heating experiment strongly suggests that the observed He is of a single component and it does not seem to be a mixing origin of solar wind He with planetary He and/or radiogenic He.

It is reported that the ^{3}He/^{4}He ratio of lunar soil which seems to have trapped the ancient solar wind was lower than the present value (PEPIN, 1980; KERRIDGE, 1980), the value being below 3×10^{-4}. However, the two box-cored samples are near the top of the sea bottom, which means that the sediments are of a recent age. Hence it is impossible to attribute the observed ratio to the ancient solar wind. Also, the difference in the ^{3}He/^{4}He ratio between the two box-cored samples which are contemporaneous makes it difficult to attribute the observed ^{3}He/^{4}He ratio to a temporal variation of the He isotopic ratio in the solar wind.

As the second possibility, we may suppose that the present solar wind He has experienced some fractionation process. If we assume that about 99% of the total He which IDPs originally had was lost, the solar wind ratio can be reduced to the observed value. When IDPs enter the atmosphere, they are heated. The maximum temperature reached during their atmospheric entry without melting has been estimated (FRAUNDORF, 1980). Since total melting of IDPs should result in total loss of He from the grain, the estimation was done under this condition. For example, in case of 10 μm-size particles with density 3 g/cm^3, half of them are heated above 800°C. The duration of the maximum temperature is typically 10 seconds (BROWNLEE *et al.*, 1977). For these IDPs, we can calculate the loss of He during their entry into the atmosphere. About 10% of the total He will be lost. Such amount of He loss can hardly cause the observed isotopic fractionation. For smaller grains, the maximum temperature is lower and the He loss would be even less, hence less isotopic fractionation. On the other hand, larger grains would be completely melted during their atmospheric entry, thus losing the total amount of He. Consequently, it is difficult to attribute the difference in the ^{3}He/^{4}He ratio between the magnetic fraction and the solar wind to isotopic fractionation due to He loss during the atmospheric heating. Also, it is observed that the ^{3}He/^{20}Ne ratio is not correlated with the ^{3}He/^{4}He ratio, but they are similar to those of solar wind implanted lunar soils. This fact makes the supposed large He diffusion loss rather unlikely. For such a large He loss which caused the isotopic fractionation must have also produced elemental fractionation.

Alternatively, a diffusion loss could occur when IDPs existed at the sea-bottom. However, the magnetic fraction of the box-cored sediment (GH79-1-1476), which was deposited rather lately, has the lowest ^{3}He/^{4}He ratio. If the diffusion in deep sea is dominant, the piston-cored samples which must be much older than the box-cored samples should show a lower ^{3}He/^{4}He ratio than the box-cored samples, which is contrary to the observation.

3.2. Why is the He carrier concentrated in the magnetic fractions?

Another problem is that the He carrier is concentrated in the magnetic fractions. If we assume that IDPs are the He carrier, it seems strange that magnetic fractions

have such high $^3He/^4He$ ratio and He content. As the majority of IDPs have a CI chondritic elemental abundance, magnetite and/or other magnetic minerals are not likely to be a major phase by analogy of CI chondrites. The amount of magnetite in CI chondrite is about 8 wt% (HYMAN et al., 1979). There are several direct observations of IDPs, e.g., by X-ray powder diffractions of IDPs (BROWNLEE, 1978), where most patterns show the existence of magnetite and pyrrhotite. It is supposed that magnetite may have been produced by atmospheric heating. FRAUNDORF (1981) concluded that the magnetite precipitation is a thermometer of the atmospheric entry heating process. By X-ray diffraction of chondritic IDPs, the following patterns are common; a) no lines, b) FeS only, c) FeS+magnetite and d) FeS+magnetite+olivine; this sequence is also considered to correspond to the heating sequence. Finely divided magnetite and olivine are produced by decomposition of poorly crystalized silicate materials (FRAUNDORF et al., 1982). Since poorly crystallized silicates are metastable, they have a tendency to change easily into other stable minerals with a little thermal stimulus, that is, magnetite and olivine. Therefore, a large part of IDPs which have experienced heating must contain magnetite. Magnetite formation transformed chondritic IDPs into ferromagnetic particles without changing the CI chondritic abundance. That is, the amount of magnetite could be larger than that inferred from CI chondrite. In sediments, magnetic particles of terrestrial origin are much less abundant than the non-magnetic particles. On the contrary, from the above arguments the portion of magnetic IDPs can be considered to be larger than non-magnetic IDPs and the enrichment of He carrier in magnetic fractions is reasonable.

4. Conclusions

(1) The carrier of exotic He in deep sea sediments is concentrated in magnetic fractions. The magnetization of the He carrier may be due to magnetite which had been produced by decomposition of poorly crystallized silicate in IDPs during their atmospheric entry.

(2) The $^3He/^4He$ ratio of the magnetic fractions ranges from $1.4–2.4 \times 10^{-4}$. Stepwise heating experiment was applied to one of the magnetic fractions. The result strongly suggests that the observed He is of a single component. Several possibilities are discussed to account for the observed $^3He/^4He$ ratio, although each has some difficulties.

Acknowledgments

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