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HEAVY NITROGEN IN THE YAMATO-74191 AND THE HETEROGENEITY OF THE PRIMITIVE SOLAR NEBULA

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Abstract: A systematic search for presolar grains in ordinary chondrites was made by measuring isotopes of nitrogen. Only one chondrite (Yamato-74191) out of 20 ordinary chondrites showed the presence of very heavy nitrogen of probably presolar origin. The heavy nitrogen is not particularly concentrated in either magnetic nor non-magnetic fractions. The HF/HCl residue is enriched in nitrogen but the heavy nitrogen seems to have been lost by acid treatments. The carrier of the heavy nitrogen has not been identified. The release patterns of excess ^{15}N are similar to those of primordial Ar. Chondrites more primitive than the Y-74191 do not contain the heavy nitrogen, suggesting heterogeneous distribution of the carrier of the heavy nitrogen. Together with evidence for the heterogeneous distribution of presolar SiC in meteorites, it is concluded that exotic materials were not well mixed in the primitive solar nebula.

1. Introduction

Presolar grains (grains that predate the solar system) are important sources of information on the formation of interstellar dust, the environment of molecular clouds, the formation of the solar nebula, etc. Microdiamonds (LEWIS *et al.*, 1987), SiC (ZINNER *et al.*, 1989) and graphite (AMARI *et al.*, 1990) have been identified as presolar grains based on the anomalous isotopic composition of various elements. These minerals are chemically very stable, and thus had a higher probability of survival in the early solar system. They also are able to survive hard acid treatments in laboratories, to be concentrated in acid residues of primitive chondrites. These are probably the main reasons that they are the only identified presolar grains found so far. Are there any other presolar grains in primitive meteorites? The answer is yes, because there are several meteorites (a small group of mesosiderites, PROMBO and CLAYTON, 1985; polymict ureilites, GRADY and PILLINGER, 1988; and an anomalous chondrite, GRADY and PILLINGER, 1990) which are known to contain isotopically anomalous nitrogen, although the carrier mineral has not been identified. Since these meteorites are a rather rare breed it was not clear how common this particular type of presolar grain was in the early solar system.

We made a systematic search for presolar grains in ordinary chondrites by measuring nitrogen isotopes, to find out the distribution of the unidentified presolar grains. Among twenty chondrites, we found one L3 chondrite (Yamato-74191) that contains anomalous nitrogen. Implications of the rather rare occurrence of this presolar grain type in ordinary chondrites, together with the implications of heterogeneous distribution of other presolar grains are discussed.

2. Experiments

Nitrogen isotopes (masses 14 and 15) in chondrites were determined using a static mass-spectrometry system. The gas extraction and purification system is similar to that at Minnesota University (Frick and Pepin, 1981). A quadrupole mass spectrometer was used for the measurement of isotopes. The details of the system are described in Hashizume and Sugiura (1990). Nitrogen is measured as molecules at masses 28, 29 and 30. Interferences from CO and hydrocarbons are corrected by measuring mass 26, and making certain assumptions on the compositon of carbon and oxygen isotopes. Nitrogen isotopic composition can be determined with a precision of better than 3 permil (one sigma) for 3 ng of air nitrogen. For chondritic samples of 50 mg, which have of the order of 1 ppm of indigenous nitrogen, stepwise combustion in 100°C steps generates more than 3 ng of nitrogen at most steps. Hot blank nitrogen at the highest temperature step (1200°C) sometimes exceeds 3 ng, resulting in higher uncertainty in the isotopic ratio. A more serious problem is the contribution of cosmogenic nitrogen, which has a high 15/14 ratio. The production rate of cosmogenic nitrogen is approximately known from the measurements of lunar soils. Therefore, corrections can be made for cosmogenic nitrogen using data on the cosmic ray exposure age. The cosmic ray exposure age of the sample is estimated from the measurement of cosmogenic Ne and Ar by the same stepwise combustion experiments by which nitrogen is measured. The cosmogenic nitrogen tends to be released at the highest temperatures. Therefore, the uncertainty of the indigenous nitrogen isotopic ratio is somewhat larger at the highest temperatures for samples with long cosmic ray exposure ages.

3. Results

The results for most chondrites will be reported separately (Hashizume and Su-

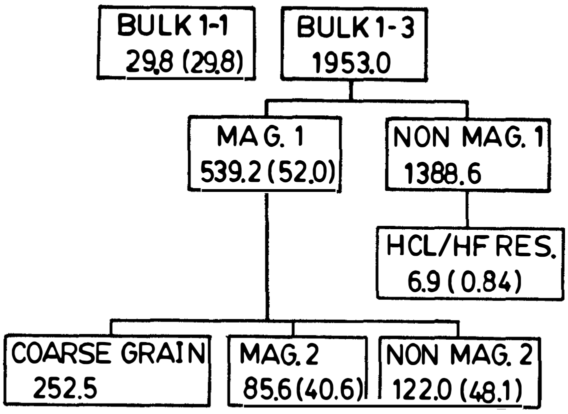

Fig. 1. Preparation of various samples from Y-74191. Weight of each sample is shown in the unit of mg. Weight (mg) of each sample analyzed for nitrogein is shown in parentheses.

Table 1a. Nitrogen, cosmogenic neon and primordial argon of the bulk sample of Y-74191 (29.8 mg).

Temperature °C	N_2 ppm	Delta-^{15}N permil	Cosmogenic Ne ccSTP/g	Primordial Ar ccSTP/g
200	1.82±0.00	16.0± 0.0	n.d.	1.5±0.1 E-09
200	3.84±0.01	17.5± 0.0	n.d.	5.7±0.1 E-09
400	4.42±0.01	23.4± 0.0	n.d.	1.1±0.0 E-08
500	3.70±0.01	26.8± 0.1	n.d.	8.6±0.2 E-09
600	1.90±0.01	56.7± 0.3	1.5±0.2 E-09	2.9±0.0 E-08
700	0.91±0.01	170.8± 2.3	4.3±0.3 E-09	8.0±0.0 E-08
800	0.63±0.02	180.3± 4.6	1.2±0.0 E-08	4.0±0.0 E-08
900	0.58±0.02	182.9± 6.6	9.9±0.5 E-09	1.7±0.1 E-08
1000	0.64±0.03	201.7± 8.6	4.5±0.7 E-09	3.6±0.1 E-08
1100	0.32±0.03	715.9± 77.5	4.1±0.7 E-09	0.2±0.1 E-08
1200a	0.11±0.04	750.5±280.6	2.6±1.0 E-09	4.0±0.1 E-08
1200b	0.33±0.04	35.4± 5.8	n.d.	1.2±1.6 E-09
Total	19.20±0.08	64.3± 0.27	3.8±0.2 E-09	3.6±0.0 E-09

n.d.: not detected.

Table 1b. Nitrogen, cosmogenic neon and primordial argon of the magnetic fraction of Y-74191 (40.6 mg).

Temperature °C	N_2 ppm	Delta-^{15}N permil	Cosmogenic Ne ccSTP/g	Primordial Ar ccSTP/g
200	0.61±0.00	10.8± 0.2	n.d.	1.8±0.1 E-09
300	1.10±0.01	0.8± 0.2	n.d.	3.2±0.1 E-09
400	4.17±0.01	17.7± 0.1	n.d.	5.7±0.2 E-09
500	1.75±0.01	35.6± 0.3	n.d.	4.4±0.2 E-09
600	1.04±0.02	99.7± 1.9	0.7±0.2 E-09	1.4±0.0 E-08
700	0.48±0.02	168.3± 8.6	1.6±0.2 E-09	2.0±0.0 E-08
800	1.93±0.02	147.5± 1.8	3.3±0.4 E-09	1.7±0.0 E-08
900	0.85±0.02	200.9± 5.7	3.1±0.4 E-09	2.4±0.0 E-08
1000	0.91±0.02	344.2± 8.0	3.3±0.7 E-09	7.9±0.0 E-08
1100	0.94±0.02	445.5± 9.9	3.1±0.8 E-09	12.4±0.1 E-08
1200a	0.80±0.03	433.2±14.9	0.3±1.0 E-10	4.6±0.1 E-08
1200b	0.25±0.02	344.7±31.5	n.d.	1.4±0.0 E-08
Total	14.83±0.06	131.8± 0.6	1.2±0.2 E-08	35.5±0.1 E-08

GIURA in preparation, 1991). Their nitrogen isotopic composition ranges from −30 to +60 permil, which could be produced by mass-dependent fractionation processes. A bulk sample of Y-74191 (L3.7) was found to have very heavy nitrogen. Thus a detailed study was made on various fractions of this chondrite. Figure 1 shows the scheme of the sample preparation. A bulk sample was gently crushed and a magnetic fraction (MAG.1) and a non-magnetic fraction (NONMAG. 1) were separated. The MAG. 1 sample still contained a lot of silicate grains. Thus, after separating some coarse grains which mostly consist of the chondrules and rims, the rest of MAG. 1 fraction was further crushed and separated into MAG. 2 and NONMAG. 2 fractions. A HF/HCl residue was prepared from the NONMAG. 1 fraction. The results for these samples (except for the MAG. 1 whose result is not shown because it is quite similar to that of the bulk sample) are summarized in Tables 1a–1d.

Table 1c. Nitrogen, cosmogenic neon and primordial argon of the non-magnetic fraction of Y-74191 (48.1 mg).

Temperature °C	N$_2$ ppm	Delta-^{15}N permil	Cosmogenic Ne ccSTP/g	Primordial Ar ccSTP/g
200	3.24±0.01	8.7± 0.0	n.d.	4.9±0.1 E-09
300	4.59±0.01	6.0± 0.0	n.d.	5.7±0.1 E-09
400	3.54±0.02	27.6± 0.2	n.d.	2.2±0.0 E-08
500	2.74±0.01	41.8± 0.3	4.7±1.2 E-10	1.6±0.0 E-08
600	3.33±0.02	66.2± 0.3	2.5±0.2 E-09	3.9±0.0 E-08
700	1.03±0.01	291.7± 4.0	8.7±0.3 E-09	9.3±0.0 E-08
800	0.63±0.01	358.3± 8.3	1.0±0.0 E-08	5.1±0.0 E-08
900	0.56±0.02	462.5± 12.8	9.7±0.4 E-09	2.9±0.0 E-08
1000a	0.61±0.02	564.0± 15.5	1.4±0.1 E-08	1.4±0.0 E-07
1100	0.66±0.02	592.7± 16.6	6.1±0.7 E-09	1.6±0.0 E-07
1200a	0.27±0.01	606.6± 31.7	2.8±1.0 E-09	7.0±0.0 E-08
1200b	0.03±0.01	782.6±327.5	0.2±8.0 E-10	7.7±0.3 E-09
Total	21.22±0.05	103.5± 0.2	5.3±0.2 E-08	65.5±0.1 E-08

Table 1d. Nitrogen, cosmogenic neon and primordial argon of HF/HCl residue of Y-74191 (0.84 mg).

Temperature °C	N$_2$ ppm	Delta-^{15}N permil	Cosmogenic Ne ccSTP/g	Primordial Ar ccSTP/g
200	1.1±0.2	57.7±11.6	n.d.	9.1±0.3 E-08
300	4.9±0.2	58.1± 2.7	n.d.	2.7±0.0 E-07
400	191.3±0.3	32.6± 0.0	n.d.	1.1±0.0 E-06
500	131.7±0.3	127.4± 0.2	n.d.	2.2±0.0 E-06
600	62.5±0.3	155.1± 0.7	n.d.	1.7±0.0 E-06
700	83.1±0.3	246.8± 0.9	n.d.	2.0±0.0 E-06
800	12.5±0.3	128.1± 3.5	n.d.	2.7±0.0 E-07
900	3.4±0.4	112.7±13.6	n.d.	3.6±0.2 E-08
1000	0.7±0.4	113.9±76.4	n.d.	6.7±2.2 E-09
1100	3.4±0.5	43.9± 0.5	n.d.	4.4±0.3 E-08
1200a	7.1±0.7	9.4± 1.8	n.d.	3.1±0.3 E-08
1200b	7.2±0.6	10.3± 1.9	n.d.	6.1±2.7 E-09
Total	508.8±1.4	109.9± 0.3	n.d.	772.4±1.8 E-08

The result of the stepwise combustion experiment for a bulk sample is shown in Fig. 2. A large amount of nitrogen is generated at lower temperatures (<700°C). Although it could be indigenous organic nitrogen because Y-74191 is an unequilibrated chondrite, the isotopic ratio is close to that of terrestrial materials. Therefore, this is probably terrestrial contamination, *i. e.* adsorbed air or organic materials. The isotopic ratio rises to 200 permil at 700°C and stays there till 1000°C. The isotopic ratio increases to >700 permil at >1000°C. The error bars in the figure are mostly due to the hot blanks, and the contribution of the cosmogenic nitrogen is not subtracted.

The results of the Ne and Ar isotopic measurements are also summarized in Table 1. Since the sample chamber is made of quartz glass, air neon could diffuse into the gas extraction line of our system. Therefore, the neon is mostly dominated

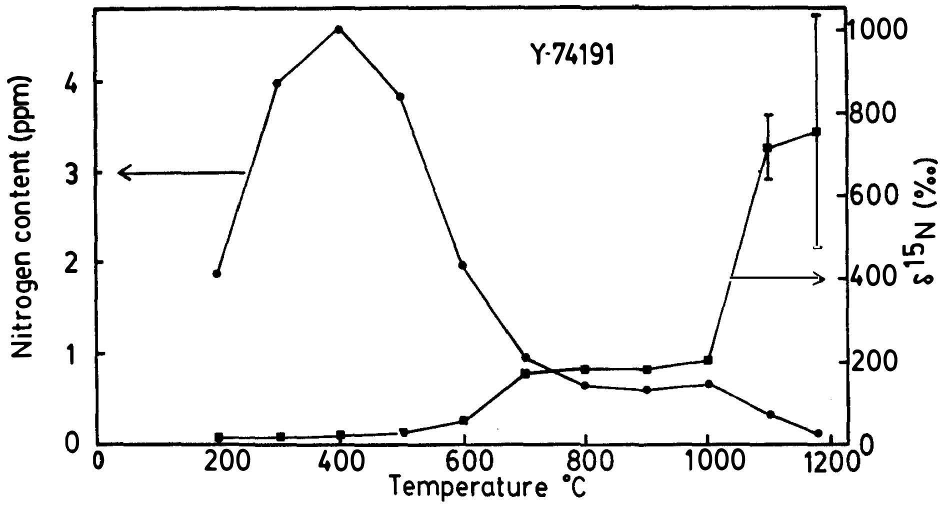

Fig. 2. Abundance and isotopic ratio of nitrogen released from the bulk sample of Y-74191 by stepwise combustion. The isotopic ratio is given in the unit of permil difference from that of the standard air.

by air neon. ^{21}Ne is, however, a rare isotope in primordial and air neon, so the contribution from the cosmogenic neon can be recognized. Using the production rate of cosmogenic neon 0.31×10^{-8} ccSTP/g/Ma for L chondrites (NISHIIZUMI, 1980), the cosmic ray exposure age is estimated.

The measured ^{36}Ar and ^{38}Ar consist of primordial and cosmogenic Ar. Assuming the 38/36 ratios of the primordial and cosmogenic argon, the amounts of cosmogenic and primordial Ar could be calculated. Using the production rate of cosmogenic Ar, the cosmic ray exposure age also could be estimated. This cosmic ray exposure age is not used for estimating the cosmogenic nitrogen, because the shielding effect for cosmogenic nitrogen is more similar to that of cosmogenic neon rather than cosmogenic argon. Published data (NAGAO and TAKAOKA, 1979) on Ne and Ar in Y-74191 agree quite well with the present results, suggesting that our system of rare gas measurement is reliable. The cosmic ray exposure age obtained from the present experiment is 12.3 Ma, which is somewhat higher than the literature age 6.5 Ma (calculated with a different production rate of cosmogenic neon) by NAGAO and TAKAOKA (1979). The production rate of cosmogenic nitrogen for lunar samples is 3.6 pg/g/Ma (BECKER *et al.*, 1976). Making corrections for the difference in composition and in the geometric factor between lunar and chondritic samples, the production rate in L chondrites is estimated to be 5.5 pg/g/Ma. Using this production rate, the amount of cosmogenic nitrogen in Y-74191 is estimated to be 67.7 pg/g. As shown in Fig. 6a, the excess ^{15}N (which is calculated by (delta ^{15}N)$\times$(nitrogen abundance)$\times$(^{15}N/^{14}N)$_{\mathrm{air}}$) in Y-74191 is much larger than the estimated cosmogenic nitrogen. The total excess ^{15}N at $T>700°\mathrm{C}$ is about 3 ng/g. Therefore, the heavy nitrogen is only partly due to cosmogenic nitrogen, and probably mostly due to presolar grains. (This conclusion is reinforced by the measurement of the magnetic fraction of Y-74191. The main component in the magnetic fraction, Fe does not produce much cosmogenic nitrogen, but highly anomalous nitrogen was likewise observed in the sample. See Fig. 3.)

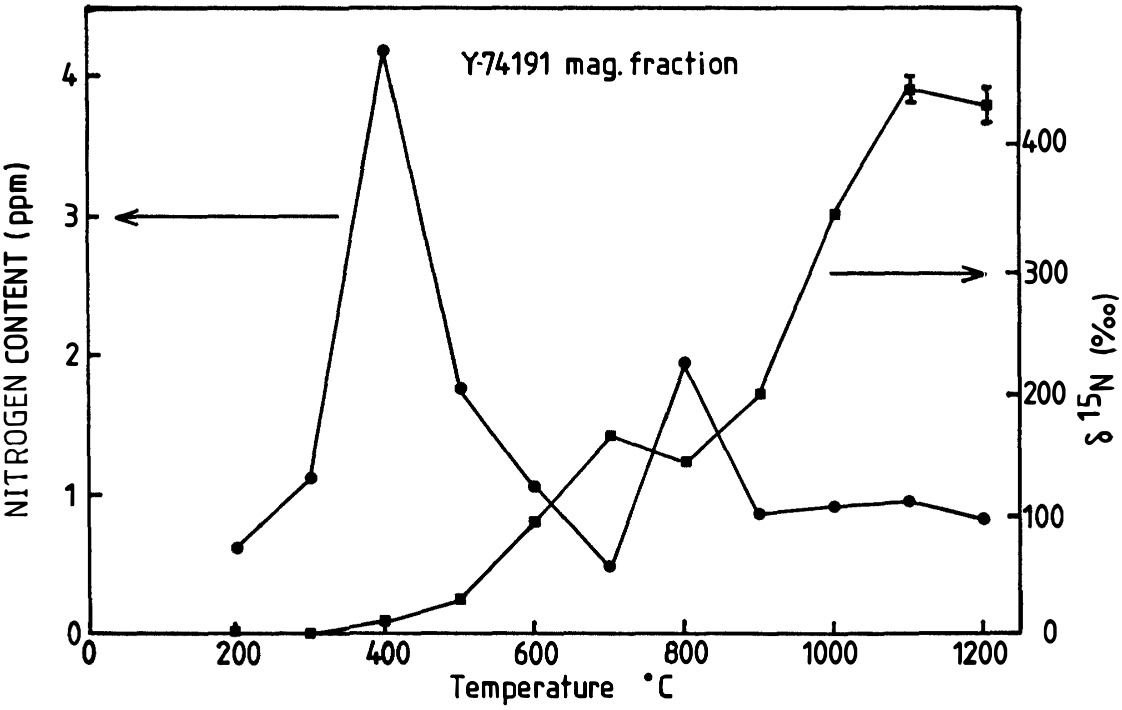

Fig. 3. *Abundance and isotopic ratio of nitrogen from the magnetic fraction of Y-74191 released by stepwise combustion.*

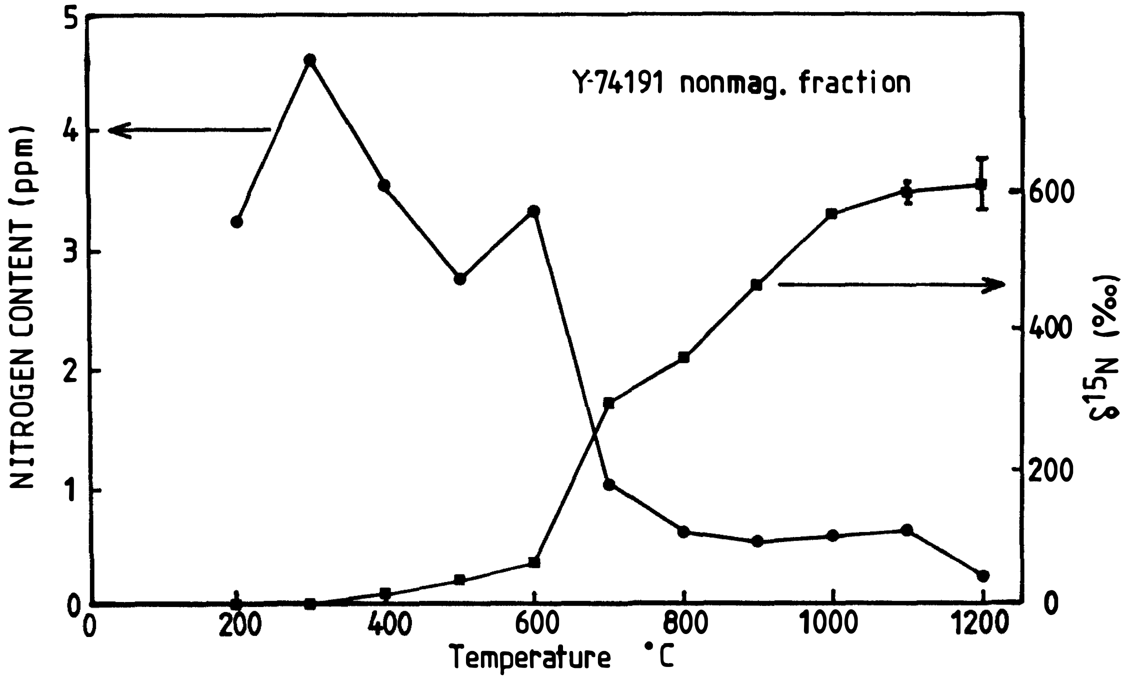

Fig. 4. *Abundance and isotopic ratio of nitrogen from the non-magnetic fraction of Y-74191 released by stepwise combustion.*

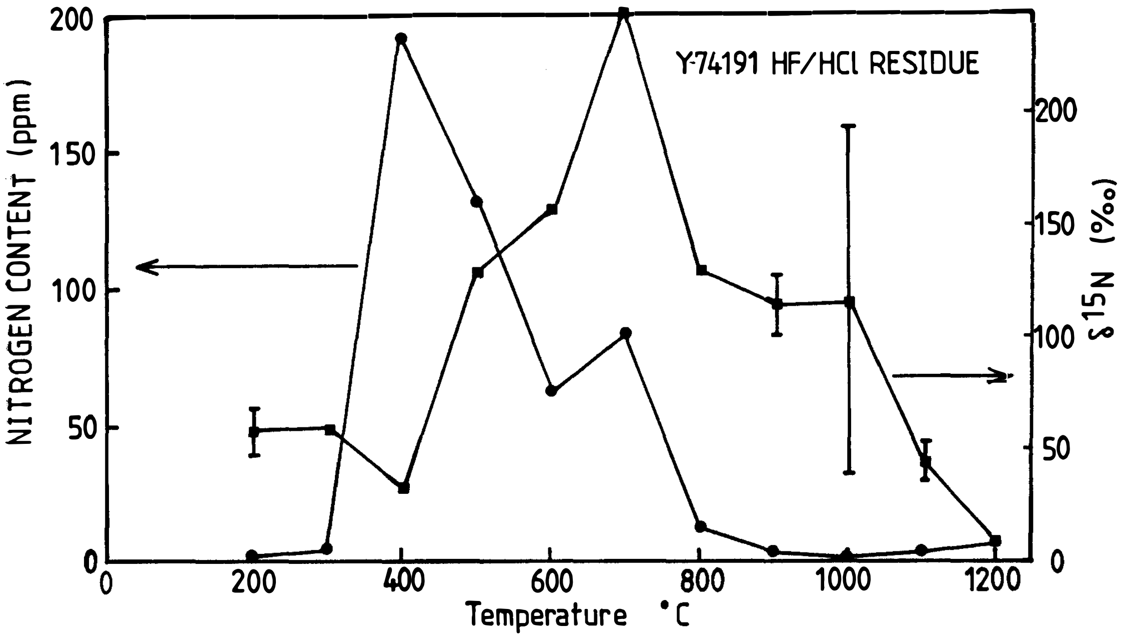

Fig. 5. Abundance and isotopic ratio of nitrogen from the HF/HCl residue of Y-74191 released by stepwise combustion.

To locate the carrier of the anomalous nitrogen, a magnetic fraction, a non-magnetic fraction and a HF/HCl residue of Y-74191 were examined. The result for the magnetic fraction is shown in Fig. 3. The sample is enriched with metallic iron but some silicate grains also exist in the sample. The patterns for the abundance and the isotopes are generally similar to those for the bulk sample. The highest δ^{15}N is, howerver, only 400 permil. The result for the non-magnetic fraction is shown in Fig. 4. Again the patterns are generally similar to those for the bulk sample. Slightly higher delta value at 700–1000°C and slightly lower maximum delta value (600 permil) are noted. It was expected that a higher maximum delta value would be obtained in one of these fractions. The fact that both samples showed lower maximum delta values suggests that some of the presolar grains may have been lost during the preparation of the samples. It is inferred that the carrier may be very fine particles which tend to be lost during the preparation. The result for the HF/HCl residue is shown in Fig. 5. The release pattern and the isotopic ratios are quite different from those for the bulk sample. Most of the nitrogen is now released at <700°C and the maximum delta value is only 250 permil. Nitrogen is highly concentrated in the residue (190 ppm). But the abundance, normalized to the bulk sample, is about 10% of the total nitrogen. The low maximum delta value and the lower release temperature suggest that the presolar carrier mineral of the nitrogen is soluble in HF/HCl. There is, however, a slim chance that these presolar grains are not soluble in HF/HCl, but are enclosed in minerals that are soluble in HF/HCl. The low maximum delta value is, in this case, interpreted as a result of cogeneration of normal nitrogen and anomalous nitrogen at <800°C. This possibility will be checked in the near future.

The results described above failed to identify the carrier mineral. In the course

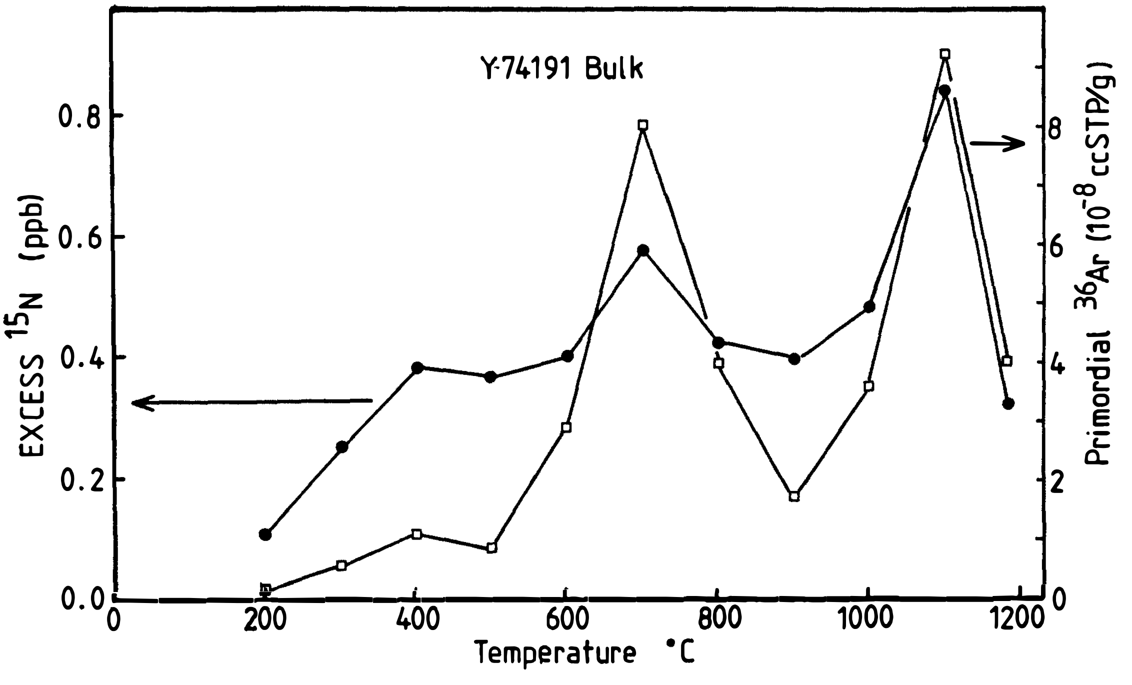

Fig. 6a.

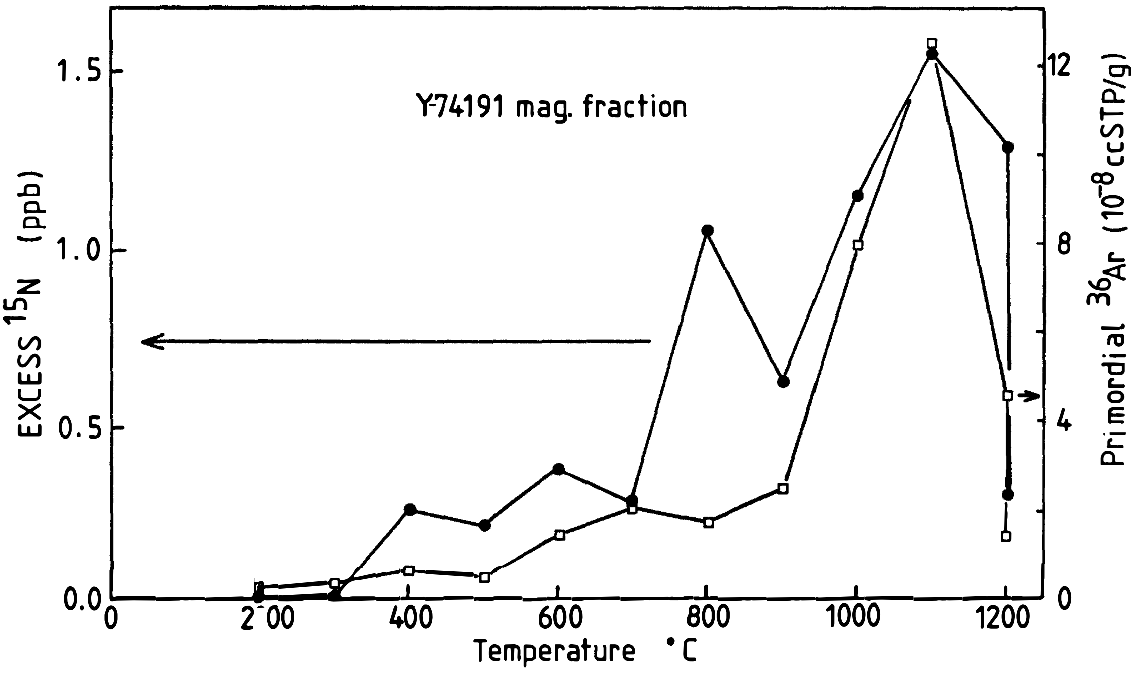

Fig. 6b.

Fig. 6. *Abundance of excess ^{15}N and primordial ^{36}Ar released from a) bulk sample, b) magnetic fraction, c) non-magnetic fraction and d) HF/HCl residue of Y-74191 released by stepwise combustion.*

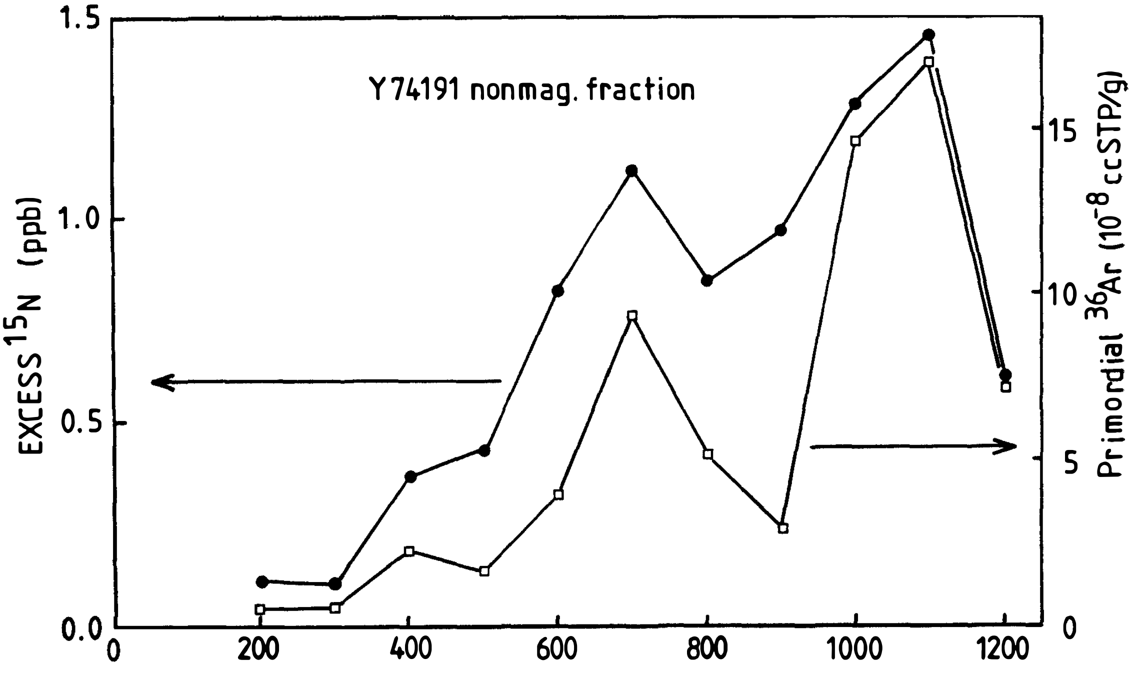
Y74191 nonmag. fraction
EXCESS ^{15}N (ppb)
1.5
1.0
0.5
0.0
0
200
400
600
800
1000
1200
Primordial ^{36}Ar (10^{-8} ccSTP/g)
15
10
5
0

Fig. 6c.

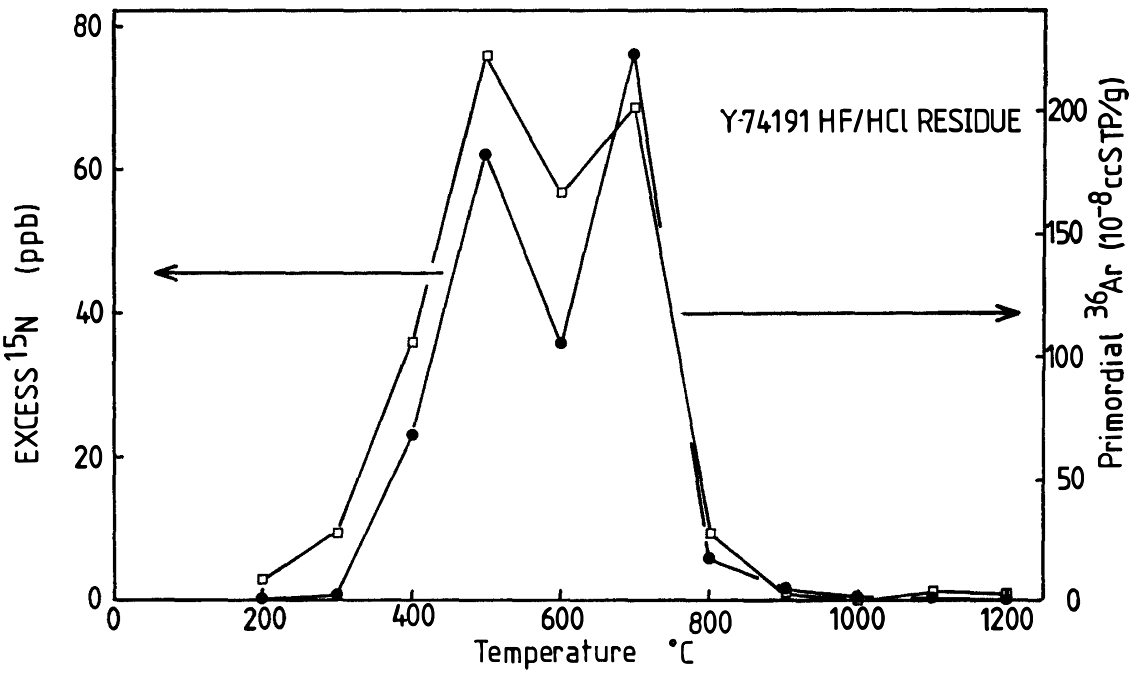
Y-74191 HF/HCl RESIDUE
EXCESS ^{15}N (ppb)
80
60
40
20
0
0
200
400
600
800
1000
1200
Temperature °C
Primordial ^{36}Ar (10^{-8}ccSTP/g)
200
150
100
50
0

Fig. 6d.

of the experiment, however, it was noted that the heavy nitrogen is associated with primordial ^{36}Ar. Release patterns of primordial ^{36}Ar and excess ^{15}N are compared in Figs. 6a–6d. The patterns are quite similar for all samples. The release pattern of primordial Ar in the bulk sample shows three peaks and the release pattern of excess nitrogen also shows peaks at the same temperature. Since the primordial Ar is mostly carried by the "Q" phase (*e. g.* SWINDLE, 1988), the "Q" phase may seem to be the carrier of the heavy nitrogen. However, other primitive chondrites which contain abundant primordial Ar do not have this heavy nitrogen. Also, Y-74191 is known to contain more ^{36}Ar (when normalized to other primordial rare gases) compared with normal chondrites (TAKAOKA and NAGAO, 1980a). Perhaps part of the ^{36}Ar in Y-74191 is also presolar in origin.

4. Comparison with Heavy Nitrogen in Other Meteorites

Heavy nitrogen in Bencubbin (mesosiderite), EET 83309 (polymict ureilites), ALH 85085 (anomalous chondrite) and Y-74191 (L3.7) is compared in Table 2. There are some common features among these occurrences of heavy nitrogen. More than two types of nitrogen seem to be present in all of these meteorites, as indicated by the delta value profile of stepwise combustion experiments (*e. g.* Fig. 2). A distinct plateau generally suggests a component. It is possible, however, that it is a mixture of an anomalous nitrogen with a normal nitrogen at a constant ratio. For instance, the isotopic ratio of the bulk sample (Fig. 2) is constant at 600–1000°C, suggesting the presence of a component while the isotopic ratio of the non-magnetic fraction

Table 2. Heavy nitrogen in meteorites.

	Bencubbin		EET 83309		ALH 85085			Y-74191	
Type	mesos.		ureilite		anomalous ch.			L3	
Component	N_α	N_β	N_I	N_{II}	N_A	N_B	N_C	N_a	N_b
T comb. (°C)*	900	600	300		250	550	<800	1100	700
	↓	↓	↓		↓	↓		↓	↓
	1050	800	600		500	800		1200	1000
^{15}N (%)	860	730	527		860	990	1497	750	200
Abundance (ppm)	50		49.6		190	50	<1	0.43	2.76
Silicate	3 : 1							N_a rich	
Metal	15 : 1				—	60 ppm		N_b rich	
HF/HCl residue					(HCl residue)				
Abundance (ppm)**	0	3	0	17.9	96	0	0	0	1.45
δ ^{15}N (‰)	—	1000	—	211	797	—	—	—	180
T comb. (°C)*	—	600	—	400	300	—	—	—	500
					↓				↓
					600				700
Reference	(1)		(2)		(3)			This study	

* Combustion temperature.

** Abundance normalized by whole rock.

(1) FRANCHI *et al.* (1986).

(2) GRADY and PILLINGER (1988).

(3) GRADY and PILLINGER (1990).

(Fig. 4) changes gradually, suggesting mixing at 600–1000°C. So the presence of two components (N_a and N_b for high and low temperature components, respectively) in Y-74191 is not well established. Similar uncertainty exists concerning the presence of multiple components in other meteorites. As discussed by GRADY and PILLINGER (1990), it is unlikely that, even if there are two components, they are independent presolar grains. Chances are very slim for two presolar components to coexist in a few samples but to be absent in most chondrites. (In the case of polymict ureilites (GRADY and PILLINGER, 1988), the moderately heavy nitrogen is enriched in carbonaceous clasts while the heaviest nitrogen is located somewhere else. So, two kinds of heavy nitrogen do coexist in meteorites. But the former nitrogen is probably a fragment of carbonaceous chondrite, which, strictly speaking, may be solar rather than presolar in origin.) It is more likely that two types of nitrogen are produced from the same presolar grains during some alteration events.

Another common feature of the heavy nitrogen is that the higher the release temperature, the higher the delta value. (Again, polymict ureilites are exceptions.) This may also indicate that the apparent lower temperature component is a mixture of the higher temperature component and normal nitrogen. The higher temperature component dissolves in HF/HCl (or HCl), which is also a common feature of the heavy nitrogen in these meteorites.

There are differences in the properties of the heavy nitrogen in these meteorites, in particular the abundance, the maximum delta value and the combustion temperature. The abundance of heavy nitrogen is very high in ALH 85085, five times higher than those in Bencubbin and in EET 83309, and nearly two orders of magnitude higher than that in Y-74191. ALH 85085 also has the highest delta-^{15}N value (1500 permil), while the other meteorites have delta-^{15}N values of several hundreds permil. Combustion temperatures are highest for Y-74191 ($>$700°C), followed by Bencubbin, ALH 85085, and polymict ureilites ($<$600°C).

None of these differences are, however, sufficient for arguing against a common origin of these apparent varieties of heavy nitrogen. The presence of protecting minerals and mixing with small amounts of normal nitrogen could explain the differences in the properties of nitrogen among these meteorites.

If the heavy nitrogen is presolar in origin, then it is expected that the carrier is associated with minerals which have anomalous isotopic (or elemental) compositions. Bencubbin has a rather anomalous oxygen isotopic composition (CLAYTON and MAYEDA, 1989), while the oxygen in ALH 85085 is similar to that in CR chondrites (CLAYTON and MAYEDA, 1989) and the oxygen in polymict ureilites is on the mixing line for the main group ureilites (CLAYTON and MAYEDA, 1988). Rare gases in Bencubbin (LEWIS, 1985) are similar to the sub-solar gases found in E chondrites, though this may be a result of Ne loss during a heating event. Rare gases in polymict ureilites and in Y-74191 are also similar to the sub-solar gases. Data on rare gases in ALH 85085 are not available. Isotopic composition of carbon has been measured for Bencubbin (FRANCHI *et al.*, 1986), ALH 85085 (GRADY and PILLINGER, 1990) and polymict ureilites (GRADY *et al.*, 1985). Carbon in these meteorites is isotopically nearly normal. In summary, heavy nitrogen in meteorites seems to be associated with a large amount of primordial Ar, but large isotopic anomalies in other elements have not been found so far.

It is certainly possible to postulate four independent sources of presolar nitrogen. The following discussion does not depend on the number of sources for the heavy nitrogen observed in meteorites. For the sake of simplicity, we assume that there is only one type of presolar nitrogen.

5. Is Y-74191 Unique?

Y-74191 is the only ordinary chondrite which contains anomalous nitrogen among the 20 chondrites we studied. There are a couple of chondrites among these 20 chondrites which are more primitive than Y-74191 whose petrologic type is L3.7. Since the abundance of primordial Ar is a good indicator of the petrologic type (*e. g.* SWINDLE, 1988), the abundances of primordial Ar and nitrogen are plotted (Fig. 7) for the primitive chondrites we studied. It can be seen that Y-74191 is not the most primitive sample among the chondrites. Then, why does it contain anomalous nitrogen? Since the carrier of the anomalous nitrogen was not destroyed by metamorphism in this type 3.7 chondrite, it should be preserved in more primitive chondrites

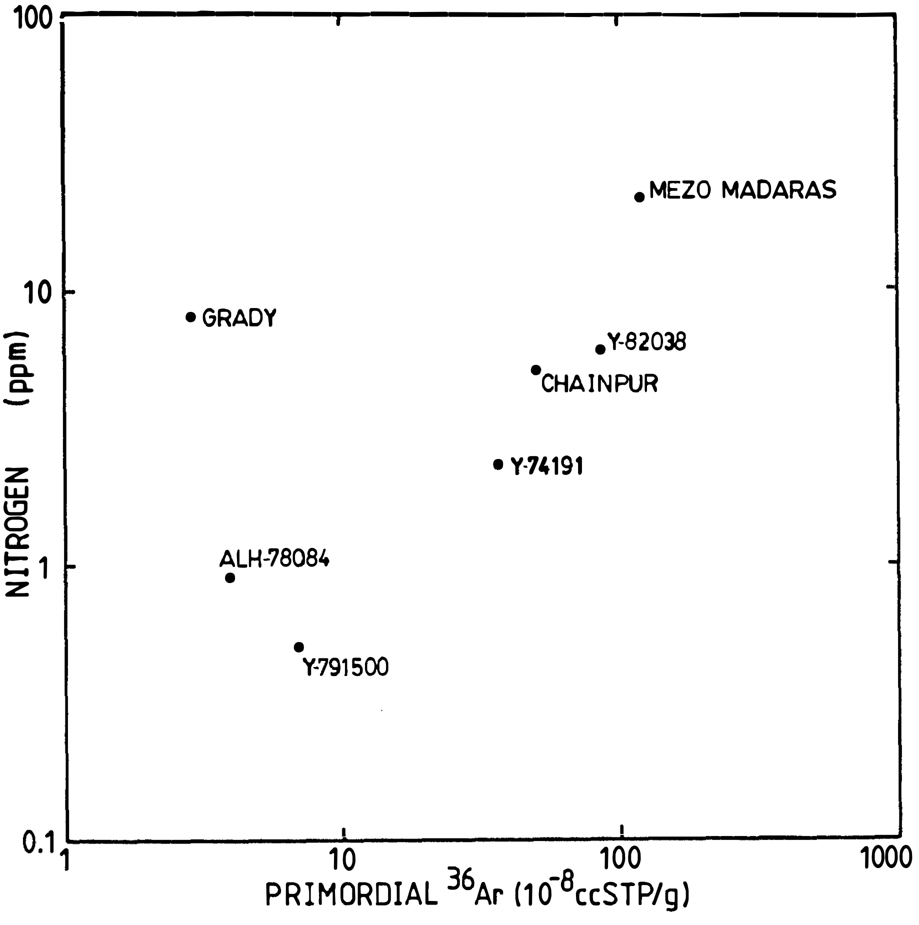

Fig. 7. Comparison of the amount of nitrogen (> 800°C fraction) and primordial ^{36}Ar in primitive ordinary chondrites. The data are taken from (HASHIZUME and SUGIURA in preparation, 1991) except for the Ar data for Chainpur (SCHULTZ and KRUSE, 1989).

if they had ever contained it. Therefore, the absence of anomalous nitrogen in other primitive chondrites probably means that the anomalous nitrogen was not incorporated in those chondrites. In other words, the anomalous nitrogen is probably distributed heterogeneously among primitive ordinary chondrites. This is an important conclusion that has many implications for various processes in the primitive solar nebula. So far the number of samples we examined is not large, and the statistical significance of this result is not firm. Certainly we need more data on the nitrogen in primitive ordinary chondrites. In the following discussion we assume that anomalous nitrogen is inhomogeneously distributed among primitive chondrites.

The next question we ask is if Y-74191 is a rare chondrite which contains anomalous nitrogen, whether it is a peculiar chondrite in any other respect. A detailed petrographic study of Y-74191 (IKEDA and TAKEDA, 1979) shows that it is not particularly unique as a type 3 chondrite. Rare gas studies show that it contains a somewhat higher amount of primordial Ar (TAKAOKA and NAGAO, 1980a) as already discussed. It also contains a fairly large amount of ^{80}Kr and radiogenic ^{129}Xe (NAGAO and TAKAOKA, 1979). The Kr could be explained as a result of reaction with epithermal neutrons (TAKAOKA and NAGAO, 1980b). The high abundance of ^{129}Xe means either a high concentration of iodine or early accretion of Y-74191. A slight anomaly in Mg isotopes of Y-74191 has been reported (NISHIMURA and OKANO, 1982). It is not known whether such an anomaly is common among primitive meteorites. ^{40}Ar-^{39}Ar dating of Y-74191 (KANEOKA, 1981) suggests that a mild (shock) reheating affected this chondrite, which is not so unusual for an L chondrite. The literature data, as a whole, indicate that Y-74191 is not particularly anomalous.

6. Distribution of the Heavy Nitrogen among and within Meteorites

The meteorites that were found to contain heavy nitrogen belong to rather rare classes. Bencubbin and Weatherford are rare mesosiderites (or iron-rich chondrites). Polymict ureilites are a minor group of ureilites, with no more than several members. ALH 85085 is a unique chondrite. Y-74191 is a first meteorite with heavy nitrogen which belongs to a major group of meteorites. Since the mesosiderites, polymict ureilites and ALH 85085 are breccias, it is possible that the heavy nitrogen was introduced to these meteorites from a projectile at the time of collision (GRADY and PILLINGER, 1990). Y-74191, being a primitive chondrite, is a primitive breccia by definition. Thus, we can categorically argue that the heavy nitrogen was introduced into Y-74191 as a foreign inclusion, too. However, this chondrite shows no clear features of brecciation or impact. It also is not a 'gas rich' chondrite. The formation process of this chondrite seems to have been a rather normal one for a primitive chondrite. Judging from nitrogen measurements on different pieces of Y-74191, the heavy nitrogen seems to be homogeneously distributed in the chondrite, at least on a 10 cm scale. The heavy nitrogen distribution within ALH 85085 is not well known because only a powdered bulk sample was studied (GRADY and PILLINGER, 1990). In the mesosiderites, heavy nitrogen is carried by both metallic and silicate clasts, which are the major phases in the meteorite. It is then tentatively concluded that the heavy nitrogen was not introduced into meteorites as a minor component at the time of brecciation. (Polymict ureilites are exceptions, where relatively heavy nitrogen, not the heaviest nitrogen, is concentrated in carbonaceous clasts, which were probably introduced by collision as a minor component of the breccia.) The heavy nitrogen is probably carried by micron-sized minerals which are homogeneously distributed in the meteorites. Identification of the carrier mineral will certainly help to confirm this tentative conclusion.

7. Distribution of the Other Presolar Grains among and within Meteorites

Among the identified presolar grains, graphite was just identified recently (AMARI *et al.*, 1990), and the distribution has not yet been studied in detail. Microdiamonds are homogeneously distributed among primitive chondrites (HUSS, 1990), though they are restricted to a less metamorphosed type (type <3.5). Only one type of microdiamonds has been found by SIMS (secondary ion mass spectrometer) judging from the carbon and nitrogen isotopes (ZINNER *et al.*, 1989). SiC is also found in many primitive chondrites whose petrologic type is lower than 3.5 (HUSS, 1990). Many types of SiC with various isotopic compositions of Si, C and N have been identified by SIMS. The presence of at least 5 groups of SiC has been suggested on the basis of clustering of Si-isotope compositions (ZINNER *et al.*, 1989). The CM chondrite Murchison contains all five groups of SiC. Interestingly, the L3 chondrite Krymka contains only two groups of SiC, and another L3 chondrite Inman contains only one group of SiC (ALEXANDER *et al.*, 1990). Such a heterogeneous distribution of SiC (and also nitrogen) is not easily explained and hence could provide a severe constraint on the formation of the primitive solar nebula. (It is possible (ANDERS, personal communication) that the distribution of SiC was determined by grain size sorting. Different grain size fractions of SiC seem to have different isotopic compositions of silicon, nitrogen and carbon. But it is not well known if grain size sorting has occurred in the solar nebula.) The distribution of these presolar grains within each meteorite has not yet been studied in detail. But different pieces of the same meteorite usually contain similar amounts of presolar grains, suggesting homogeneous distribution of fine presolar grains in a given meteorite. (In the case of Allende, which is a large meteorite, a slight difference in the concentration of nitrogen in microdiamonds among different pieces was suggested (RUSSELL and PILLINGER, 1990)).

It seems that presolar grains (SiC and the carrier of heavy nitrogen) are heterogeneously distributed among meteorites, but are nearly homogeneously distributed within each meteorite.

Oxygen isotope anomalies in meteorites are well known (CLAYTON, 1981). Both inter- and intra-meteorite anomalies have been observed. Calcium-aluminum rich inclusions are the main source of intra-meteorite anomalies. There is no systematic relation between any properties of meteorites and the isotopic composition of oxygen. Therefore, the origin of the inter-meteorite oxygen anomalies is not known. All we can say is that the primitive solar nebula was not homogeneous with respect to oxygen isotopes.

8. Recipe for Making Heterogeneous Distribution of Presolar Grains

The observation on the distribution of presolar grains suggests that they are carried by micron size grains and distributed homogeneously on a 10-cm scale, yet heterogeneously distributed among various meteorite parent bodies (CM and L chondrites) and also within a parent body (L chondrites). An important implication is that presolar grains must have been incorporated in 1 m to 10 km size bodies, before their accretion onto the meteorite parent bodies. Here we assume that heavy nitrogen was incorporated as part of the initial accretion process. A model in which heavy nitrogen is introduced by later impacts will be examined later. The lower limit of the size is set by the typical size of a meteorite, which is isotopically homogeneous. One meter is also the size of agglomerates expected to form in a turbulent nebula (WEIDENSCHILLING *et al.*, 1989). The upper limit of the size is set by the fact that two L3 chondrites are isotopically different. Since the size of the parent body of L chondrites is considered to be not larger than 100 km, and L3 occupies only a small fraction of the parent body, 10 km is probably not a bad estimate. Ten km is also the size of a planetesimal produced by gravitational instability. The body may have been produced in another solar system, in a molecular cloud or in the primitive solar nebula. Since theoretical arguments suggest the former two possibilities are small (DRAINE, 1985), we will concentrate on the formation of such bodies in the primitive solar nebula.

Let us start from the situation in the molecular cloud and follow what may have happened to the presolar grains until they finally ended up in meteorite parent bodies. Presolar grains were ejected from star such as novae, supernovae and red giants. In the case of SiC, the stars must have been rich in carbon relative to oxygen. The condensation sequence of solid grains around such carbon stars is quite different from that of the solar nebula (LARIMER and BARTHOLOMAY, 1979). Therefore, SiC must have been accompanied by a lot of exotic minerals which also contained many isotopically anomalous elements. At least five carbon-rich stars ejected SiC into the molecular cloud that was destined to form the solar system. In the case of the carrier of the heavy nitrogen, only one star made contribution (lacking the identity of the carrier and SIMS data this is just a guess). The mixing process in the molecular cloud is not well known theoretically. The observation on meteoritic SiC and heavy nitrogen suggests that the five types of SiC (and the heavy nitrogen) were not well mixed in the molecular cloud. Thus the regions that contained SiC were isolated from each other and the bulk chemical composition of the regions may have been quite different from that of the sloar system. The molecular cloud then collapsed and formed the primitive solar nebula. In the following, a static nebula is considered. (Alternatively, it may be considered as a time-dependent accretion disk, which will be examined later.) The regions that contained various kinds of SiC and heavy nitrogen were not well mixed initially in the primitive solar nebula. Then solid grains started to agglomerate and settle to the equatorial plane of the solar system. Depending on the presence or absence of turbulence, the agglomerated grains may have grown to cm-size or meter-size, respectively (*e.g.* WEIDENSCHILLING *et al.*, 1989). Mixing of the presolar grains must have occurred at this stage, because planetesimals are formed at the next stage and it becomes difficult to mix things intimately thereafter.

(Mixing could have occurred at an earlier stage if partial mixing could be preserved in the dust-rich layer.) All five kinds of SiC were mixed in the region which ended up in Murchison. Only one or two kinds of SiC were mixed in the region which ended up in L3 chondrites. When the density of the agglomerated grains in the dust-rich later reached a certain value, planetesimals were formed by gravitational instability. A meteorite parent body may consist of several tens of planetesimals. But if there were nebula gas, the relative speed of collision between planetesimals is relatively small and intimate mixing of materials from different planetesimals is not expected.

In the above scenario, we did not explain how chemical homogeneity was achieved in a parent body. In the case of CM chondirites (Murchison), solid grains may have been thoroughly mixed to produce a nearly solar composition. But in the case of L chondrites, apparently matter was only partly mixed. Considering that SiC must have been accompanied by many exotic isotopes and minerals, chemical homogeneity (also the identity of oxygen isotope) of L3 chondrites is not easily explained. One possibility is to postulate that the contribution of the SiC and the associates to the bulk chemical composition is small and most isotopic anomalies of other elements were erased by chondrule formation events. In this case isotope anomaly is restricted to very refractory elements and/or matrix materials which did not experience heating events. Another possibility is that chondrites are not really chemically homogeneous. Actually there are suggestions that some elements may be distributed heterogeneously in ordinary chondrites (KALLEMEYN *et al.*, 1989). It is also known that chemical composition of type 3 ordinary chondrites is somewhat variable, making classification (H, L or LL) difficult. There are not enough good chemical data and we are not sure if the apparent heterogeneity of the bulk chemical composition of chondrites is simply due to statistical error associated with a small sample size. ALH 85085 may be a good example of real chemical heterogeneity. The chemical composition of this chondrite is unlike any other groups of chondrites (GROSSMAN *et al.*, 1988). The origin of the anomalous chemistry may be traced back to the molecular cloud and further to the star which ejected the heavy nitrogen. This possibility can be checked experimentally, because isotope anomalies of some other elements are expected to be found in the meteorite. (It is unfortunate that the meteorite is very small.) In the case of Y-74191, judging from the abundance of the anomalous nitrogen compared with that in ALH 85085, the abundance of the exotic material is two orders of magnitude smaller than that in ALH 85085. So the anomaly in the bulk chemistry would be hard to detect. Yet it is worthwhile to examine if isotopic anomalies may be found in some other elements.

The sizes of the isotopically heterogeneous bodies are not well constrained at present. Detailed studies of the heavy nitrogen in L chondrites will be helpful to solve this question. If many L3 chondrites are found to contain heavy nitrogen at a fairly constant abundance, then a large (10 km) size body is preferred. If, in contrast, a number of L3 chondrites contain anomalous nitrogen, in a rather variable concentration, small size bodies are preferable. It is also important to examine if a chondrite is really homogeneous isotopically.

9. Accretion in a Time-Dependent Accretion Disk

If planetesimals were formed at an early stage of the formation of the primitive solar nebula, a slightly different scenario is proposed. In the evolving solar nebula, most of the material fall onto the sun. Differential motion between planetesimals and aggeregates of dust particles could develop in such a situation, causing aggregates of dust particles to fall onto the sun more rapidly than planetesimals. Then, a sequence of presolar grains on their way to the sun could accrete on planetesimals. Planetesimals would be covered by layers of exotic materials. Such a scenario is convenient for explaining the heterogeneous distribution of presolar grains, but there is a problem of maintaining chemical homogeneity of a parent body in the presence of isotopic heterogeneity. This is the same problem which we discussed in the previous section, and similar solutions, negligible contribution of exotic materials to the bulk composition or existence of chemical heterogeneity are suggested.

10. Impact with an Anomalous Projectile

Another way of explaining the heterogeneous distribution of presolar grains in meteorites is to postulate that the heavy nitrogen was introduced into parent bodies by collisions with exotic bodies. As argued by GRADY and PILLINGER (1990), heavy nitrogen has been found mostly in brecciated meteorites. Hence it is natural to consider that the heavy nitrogen was introduced to the parent bodies from the projectile. In the sense that materials are mixed mechanically, this scenario is similar to the scenario we discussed above. The main difference is the timing (after the accretion of parent bodies) of the impact event, and the presence of presumably larger and more solid bodies. If the relative velocity of collision is less than 3 km/s, then small fragments (clasts) of rocks are expected to be produced. However, as discussed earlier in this article, heavy nitrogen seems not to have been incorporated in meteorites (mesosiderites, polymict ureilites, and chondrites) as small fragments. If the impact velocity exceeds 3 km/s, melt and vapor phases are produced. If projectile materials mostly vaporized and recondensed quickly, fine particles with isotopic anomaly may be produced. However, it is very difficult to condense nitrogen in an oxidizing environment. Compositions of olivine in the meteorites with heavy nitrogen suggest that the conditions were not particularly reducing when they were formed. (The redox conditions actually were rather reducing, but not quite as reducing as in enstatite chondrites.) Absence of agglutinates and glassy spherules which are often associated with rapid cooling after high velocity impacts, also suggest that the heavy nitrogen was not introduced through high velocity impacts.

Overall, it is concluded that although many meteorites with heavy nitrogen are impact breccias, the heavy nitrogen is not a minor component introduced from a projectile.

11. Conclusion

Heavy nitrogen was found in an L3 (Y-74191) chondrite. It is due not to cosmogenic nitrogen but probably to presolar grains. The heavy nitrogen is similar to that found in rare mesosiderites, polymict ureilites and an anomalous chondrite. Since such heavy nitrogen was not found in less metamorphosed ordinary chondrites, the presolar nitrogen seems to be distributed heterogeneously in meteorites. Such heterogeneous distribution of presolar grains has also been indicated in the case of SiC. It is suggested that the heterogeneity is not due to the introduction of presolar grains as a minor component by a late stage collisions. More likely, these grains were incorporated into the parent bodies of meteorites during the initial stage of their formation. The heterogeneity was created by formation of bodies 1 m to 10 km in size before the final accretion of the parent bodies. The apparent absence of chemical heterogeneity in samples with isotopic anomalies is not easily explained. More detailed studies on the distribution of heavy nitrogen are needed. It is also important to find whether isotope anomalies in other elements are present in meteorites with heavy nitrogen.

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