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ISOTOPIC COMPOSITION OF NITROGEN IN THE PCA 91002 R GROUP CHONDRITE

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Abstract: Nitrogen isotopic composition of an R-group chondrite PCA 91002 was measured. A delta ^{15}N value as low as −60 permil was found in some combustion steps. Since this is a gas-rich meteorite, interpretation of the result is not straightforward, but with the aid of results on an H_2O_2-treated sample, we conclude that isotopically light nitrogen is indigenous to the meteorite.

1. Introduction

Pecora Escarpment (PCA) 91002 is an R (Rumuruti) group chondrite (RUBIN and KALLEMEYN, 1994). The R group (formerly the Carlisle Lakes group) is a newly identified group which is characterized by a distinct oxygen isotopic composition (enriched in ^{17}O) and highly oxidized mineralogy (RUBIN and KALLEMEYN, 1989). Since it has been recognized that nitrogen isotopic composition is quite distinct for many groups of meteorites (KUNG and CLAYTON, 1978), and hence may serve as a useful criterion for identifying meteorite groups, the nitrogen isotopic composition of this new chondrite group is of great interest. (Nitrogen isotopic composition has not been reported yet for the R group chondrites.) Furthermore, isotopic compositions of nitrogen and oxygen are useful tools in revealing the relationship between source materials of chondrites, since isotopic compositions cannot be changed easily by solar system processes. In particular, if an extreme isotopic composition is obtained, it may serve as an end member of source materials which were mixed and processed to form various chondrites. Bearing these purposes in mind, isotopic composition and abundance of nitrogen in PCA 91002 were measured.

2. Experiments

Nitrogen was extracted by stepped combustion, and nitrogen isotopic composition and abundance were measured with a static mass spectrometer. Details of the experimental method have been published elsewhere (HASHIZUME and SUGIURA, 1990) and recent improvement of the system has been described in SUGIURA and ZASHU (1994). Neon and argon were also measured to obtain information on cosmogenic nitrogen and also on solar nitrogen (if it is present). A bulk sample (81.15 mg), a coarse grained (≈100 μm) HCl residue (6.98 mg), a fine grained HCl residue (0.93 mg) and a H_2O_2-treated sample (65.79 mg) were measured. The HCl residues were prepared from a 50.06 mg fragment, while the bulk sample and H_2O_2-treated sample were prepared from separate fragments. This caused a problem, as will be seen later, because this chondrite turned out to be a gas rich chondrite and is very heterogeneous with respect to the rare gas contents. But we were not able to foresee this problem. The HCl treatment was done with 2N HCl for about 3 hours to find out if the nitrogen isotopic anomalies are carried by acid-resistant presolar grains. The fine grained fraction, which did not settle after a couple of minutes of suspension, was centrifuged and collected separately. The H_2O_2 treatment was done at 80°C for 1 hour, mainly for removing terrestrial contamination. This treatment also attacks surface layers of various minerals, partially removing implanted solar wind species. Weight loss by the H_2O_2 treatment is negligible.

The hot blank of ^{22}Ne was about 2×10^{-11}ccstp at 1200°C, which is about 1% of the measured amounts at that temperature for most samples. The hot blanks were negligible at lower temperatures. Blank corrections for nitrogen (0.1 ng) are at most 10% of the smallest amount of nitrogen extracted by stepped heating, and negligible for most temperature steps. Blank corrections for ^{36}Ar are of the order of 1×10^{-12} ccstp/g, which is less than 1% of the measured amounts extracted by stepped heating.

3. Results

The results of stepped combustion are given in Tables 1–4 and a summary is given in Table 5. The data for residues are not normalized back to the whole rock values. The concentrations and the isotopic ratios have been corrected for blanks. From the isotopic composition of neon (^{20}Ne/^{22}Ne≈13) and the high ratios of ^{20}Ne/^{36}Ar, this chondrite is considered to be a gas-rich chondrite enriched in solar type noble gases. Assuming appropriate end-members' isotopic compositions (see SUGIURA and ZASHU, 1994 for details), the neon was deconvolved into solar neon and cosmogenic neon.

Figure 1 shows the ^{20}Ne/^{22}Ne isotopic ratio for the solar Ne and Fig. 2 shows the release patterns of the solar ^{22}Ne. Because of the smaller sample size, the results for the HCl-treated samples have larger errors. Generally speaking, (^{20}Ne/^{22}Ne) trapped in this chondrite is rather high compared with many gas rich meteorites. Another gas rich meteorite, ALHA77216, which was measured with the same instrument, gives a ratio close to 11.3 (the nominal value for SEP neon) at temperatures equal to and higher than 700°C (SUGIURA and ZASHU, 1994). Compared with this value, the ratio for PCA 91002 is definitely high. It seems that this is due to a higher proportion of SW neon (^{20}Ne/^{22}Ne=13.7: BENKERT *et al.*, 1993) in this chondrite.

The chondrite is quite heterogeneous on the basis of variable solar Ne abundances in the samples (Fig. 2 and Table 5). The abundance and the release pattern of the solar Ne in the H_2O_2-treated sample are not consistent with those of the bulk sample and must be due to the heterogeneity of the meteorite. (Note that the weight loss by the H_2O_2 treatment is negligible, yet the concentration of ^{22}Ne is higher in the sample.) Excepting this sample, two release peaks are observed in Fig. 2. It is tempting to consider that these peaks correspond to SW and SEP neon. But, the isotopic composition of the trapped neon is not much different for these peaks. Therefore the difference in the release temperature may be due to different

Table 1. Stepped combustion of the bulk sample of PCA 91002.

Temp (°C)	Nitrogen (ppm)		Delta ^{15}N (permil)		^{20}Ne/^{22}Ne		^{21}Ne/^{22}Ne		^{22}Ne (E-8ccstp/g)		^{38}Ar/^{36}Ar		^{40}Ar/^{36}Ar		^{36}Ar E-8ccstp/g	
200	18.30	0.02	−63.8	1.4	6.27	1.858	0.674	0.196	0.02	0.00	0.186	0.009	388.5	7.7	0.06	0.00
300	35.12	0.04	−58.1	1.2	6.64	0.892	0.337	0.060	0.07	0.01	0.148	0.006	373.2	5.4	0.11	0.00
400	2.55	0.00	−29.0	1.6	8.71	0.671	0.337	0.038	0.20	0.01	0.196	0.005	373.4	3.7	0.25	0.00
500	1.13	0.00	−35.4	1.4	12.08	0.143	0.129	0.004	2.72	0.02	0.224	0.003	1111.6	7.7	0.55	0.00
600	0.46	0.00	−30.9	1.6	11.86	0.081	0.162	0.003	6.75	0.03	0.223	0.003	1182.6	6.8	0.96	0.01
700	0.50	0.00	−27.2	1.6	8.51	0.078	0.386	0.006	5.84	0.03	0.200	0.002	467.9	2.0	2.44	0.01
800	0.30	0.00	−36.3	1.9	7.89	0.086	0.412	0.008	3.10	0.02	0.220	0.003	337.5	1.9	0.97	0.01
900	0.32	0.00	−34.0	1.9	9.69	0.091	0.283	0.005	3.96	0.03	0.235	0.003	97.8	0.5	1.06	0.01
1000	0.16	0.00	−29.0	2.0	9.82	0.093	0.254	0.005	4.28	0.03	0.239	0.003	59.7	0.3	0.96	0.01
1100	0.36	0.00	−45.5	1.8	8.78	0.093	0.313	0.006	6.07	0.05	0.218	0.001	24.7	0.1	3.98	0.02
1200	0.40	0.00	−55.8	1.7	4.30	0.099	0.653	0.013	2.85	0.03	0.294	0.002	15.1	0.1	2.43	0.01
Total	59.60	0.04	−57.3		9.35		0.308		35.86	0.09	0.231		266.9		13.77	0.03

Second columns of data show one sigma errors.

Table 2. Stepped combustion of the HCl treated sample (coarse grain) of PCA 91002.

Temp (°C)	Nitrogen (ppm)		Delta ^{15}N (permil)		^{20}Ne/^{22}Ne		^{21}Ne/^{22}Ne		^{2}Ne (E-8ccstp/g)		^{38}Ar/^{36}Ar		^{40}Ar/^{36}Ar		^{36}Ar E-8ccstp/g	
200	0.22	0.01	−20.4	6.0	7.64	2.23	0.118	0.054	0.20	0.03	0.207	0.020	442.4	16.9	0.19	0.01
300	0.53	0.01	−17.5	3.8	10.52	1.04	0.231	0.041	0.74	0.05	0.207	0.013	376.8	9.1	0.45	0.01
400	4.20	0.01	−2.4	2.0	11.26	0.51	0.168	0.018	2.38	0.08	0.242	0.008	400.5	5.8	1.26	0.02
500	7.74	0.01	3.3	2.0	9.18	0.70	0.229	0.031	1.18	0.06	0.252	0.006	879.0	9.3	2.40	0.03
600	3.87	0.01	6.4	1.8	7.47	0.86	0.336	0.048	1.05	0.06	0.269	0.006	1489.5	15.2	2.62	0.03
700	1.13	0.01	6.8	2.5	6.07	0.40	0.441	0.032	2.64	0.08	0.365	0.011	3040.1	49.6	0.98	0.02
800	0.50	0.01	−0.8	3.9	5.92	0.28	0.530	0.031	3.65	0.10	0.350	0.018	1262.5	32.7	0.40	0.01
900	0.28	0.01	4.6	5.3	5.04	0.51	0.667	0.056	1.92	0.08	0.387	0.025	499.1	17.2	0.24	0.02
1000	0.32	0.01	6.7	4.9	4.24	0.76	0.723	0.080	1.30	0.08	0.559	0.026	246.5	7.1	0.34	0.02
1100	0.22	0.01	8.8	5.9	4.65	1.53	0.639	0.115	0.74	0.09	0.643	0.022	121.9	2.7	0.56	0.02
1200	0.63	0.01	17.4	3.5	6.09	2.69	0.379	0.098	0.79	0.13	1.067	0.020	39.6	0.6	1.42	0.03
Total	19.64	0.03	2.6		6.98		0.442		16.60	0.27	0.406		975.7		10.85	0.13

Table 3. Stepped combustion of the HCl treated (fine grain) sample of PCA 91002.

Temp (°C)	Nitrogen (ppm)		Delta ^{15}N (permil)		$^{20}Ne/^{22}Ne$		$^{21}Ne/^{22}Ne$		^{22}Ne (E-8ccstp/g)		$^{38}Ar/^{36}Ar$		$^{40}Ar/^{36}Ar$		^{36}Ar (E-8ccstp/g)	
200	1.44	0.07	−23.7	6.5	17.64	9.49	0.057	0.073	0.44	0.17	0.155	0.020	97.6	4.1	1.39	0.17
300	6.70	0.05	−5.2	2.9	10.23	1.37	0.036	0.017	2.99	0.27	0.183	0.012	67.8	1.6	3.64	0.16
400	337.64	0.34	2.9	1.8	10.79	1.94	0.084	0.028	5.35	0.81	0.197	0.007	86.3	1.3	9.09	0.19
500	201.98	0.21	7.3	1.8	12.67	2.43	0.032	0.020	2.17	0.29	0.190	0.004	127.6	1.2	24.57	0.26
600	39.48	0.07	5.9	1.6	11.93	7.98	0.047	0.053	1.18	0.63	0.199	0.006	182.8	2.1	14.93	0.21
700	14.99	0.07	8.2	1.9	9.99	1.47	0.218	0.046	4.52	0.39	0.197	0.008	151.5	2.3	8.70	0.21
800	2.56	0.05	11.2	4.6	11.01	1.11	0.163	0.027	8.34	0.51	0.214	0.024	117.4	5.3	1.22	0.12
900	1.53	0.05	12.0	6.1	11.10	3.32	0.067	0.031	3.24	0.54	0.273	0.030	53.7	2.8	0.98	0.12
1000	0.95	0.06	8.1	8.0	11.05	11.06	0.065	0.074	1.20	0.65	0.294	0.028	20.2	1.0	1.20	0.13
1100	1.52	0.06	18.4	6.2	10.15	15.09	0.072	0.102	1.07	0.81	0.361	0.027	5.7	0.3	1.59	0.13
1200	1.67	0.08	9.2	5.9	10.15	10.00	0.029	0.040	2.01	1.01	0.622	0.050	39.0	2.1	0.96	0.13
Total	610.46	0.44	4.6		10.92		0.107		32.51	2.01	0.206		126.2		68.25	1.45

First extraction at 1200°C was lost accidentally.

Table 4. Stepped combustion of the H_2O_2-treated sample of PCA 91002.

Temp (°C)	Nitrogen (ppm)		Delta ^{15}N (permil)		$^{20}Ne/^{22}Ne$		$^{21}Ne/^{22}Ne$		^{22}Ne (E-8ccstp/g)		$^{38}Ar/^{36}Ar$		$^{40}Ar/^{36}Ar$		^{36}Ar (E-8ccstp/g)	
200	0.03	0.00	−38.3	4.9	11.54	0.63	0.068	0.012	0.19	0.01	0.162	0.006	197.8	2.9	0.13	0.00
300	0.24	0.00	−21.0	1.8	10.69	0.47	0.166	0.016	0.30	0.01	0.170	0.005	207.3	2.5	0.20	0.00
400	0.14	0.00	−25.1	2.5	10.78	0.25	0.177	0.010	0.85	0.01	0.191	0.004	282.8	2.3	0.47	0.00
500	0.38	0.00	−21.3	1.4	12.10	0.14	0.130	0.004	2.91	0.03	0.191	0.002	459.0	2.7	1.06	0.00
600	0.66	0.00	−20.6	1.5	11.26	0.08	0.173	0.003	8.46	0.04	0.196	0.002	475.5	2.2	2.24	0.01
700	0.31	0.00	−50.4	1.6	8.92	0.05	0.310	0.004	12.88	0.06	0.197	0.002	195.4	0.8	3.18	0.01
800	0.34	0.00	−58.7	2.1	9.74	0.06	0.261	0.003	11.72	0.05	0.208	0.002	70.5	0.3	3.52	0.01
900	0.28	0.00	−61.8	1.7	10.94	0.07	0.164	0.003	9.71	0.05	0.217	0.002	37.5	0.2	3.00	0.01
1000	0.16	0.00	−58.0	2.3	10.54	0.08	0.189	0.003	7.03	0.04	0.228	0.002	18.8	0.1	2.71	0.01
1100	0.06	0.00	−43.9	3.6	10.07	0.15	0.213	0.006	2.85	0.03	0.347	0.002	12.3	0.1	1.90	0.01
1200	0.07	0.00	68.0	4.1	7.33	0.61	0.370	0.026	0.62	0.02	0.359	0.003	9.8	0.0	2.12	0.01
Total	2.68	0.00	−34.2		10.22		0.223		57.51	0.12	0.226		137.8		20.53	0.03

Table 5. Summary of nitrogen, neon and argon measurements.

Sample	Weight (mg)	N_2 (ppm)	Delta ^{15}N* (permil)	Excess ^{15}N (ppb)	Cosmog. ^{21}Ne (E-8ccstp/g)	Solar ^{22}Ne (E-8ccstp/g)
Bulk	81.51	59.60	−63.8	−12.64	10.33	24.1
HCl, coarse	6.8	19.64	−20.4	0.19	7.09	8.6
HCl, fine	0.93	610.46	−23.7	10.47	2.59	29.6
H_2O_2	65.79	2.68	−61.8	−0.34	11.52	44.4

	Cosmog. ^{36}Ar (E-8ccstp/g)	Non-cosmo. ^{36}Ar (E-8ccstp/g)
Bulk	0.69	13.3
HCl, coarse	2.71	9.0
HCl, fine	1.50	67.2
H_2O_2	0.92	19.9

*Lowest value observed by stepped combustion.

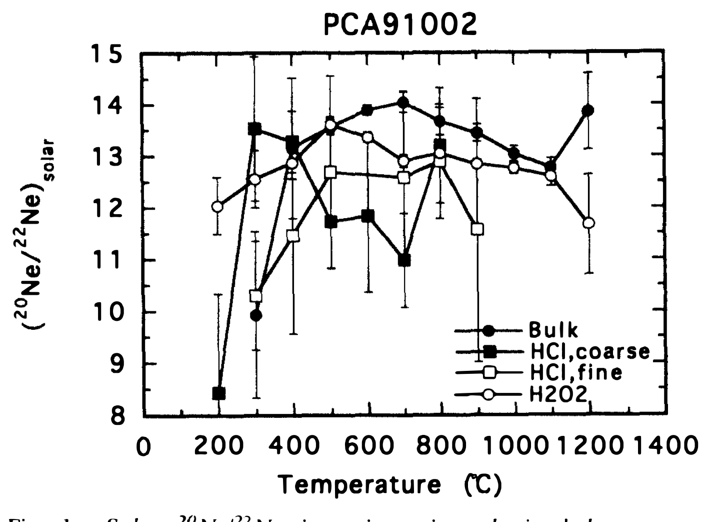

Fig. 1. Solar $^{20}Ne/^{22}Ne$ isotopic ratios obtained by stepped combustion. Data points with very large errors are omitted.

retentivities of minerals.

The release of cosmogenic ^{21}Ne is shown in Fig. 3. The pattern which shows a peak at 700°C is a typical pattern obtained for chondrites by stepped combustion. Some neon is released from the bulk sample at higher temperatures. This is due to the coarser grain size of this sample. (The other samples are powdered samples.) The higher abundance of cosmogenic Ne in the H_2O_2-treated sample is attributed to the heterogeneity of this chondrite.

Figure 4 shows $^{38}Ar/^{36}Ar$ ratios obtained by stepped combustion. The ratio is

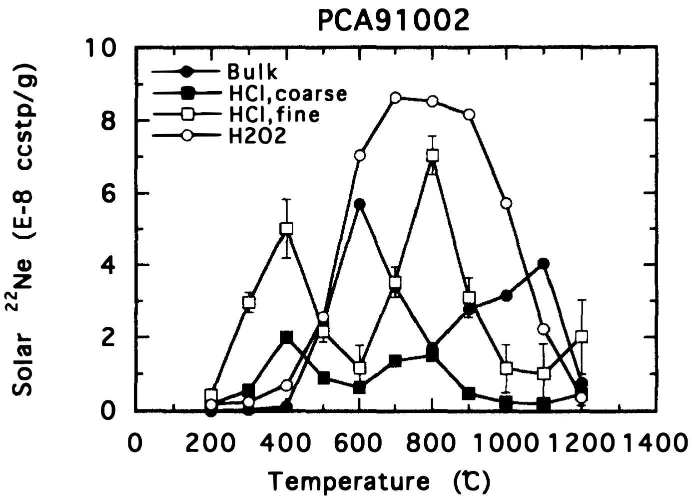

Fig. 2. Abundances of solar ^{22}Ne released by stepped combustion.

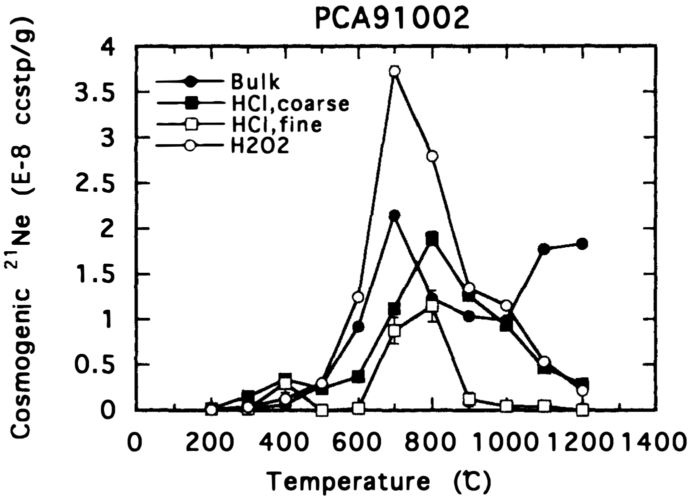

Fig. 3. Release patterns of cosmogenic ^{21}Ne.

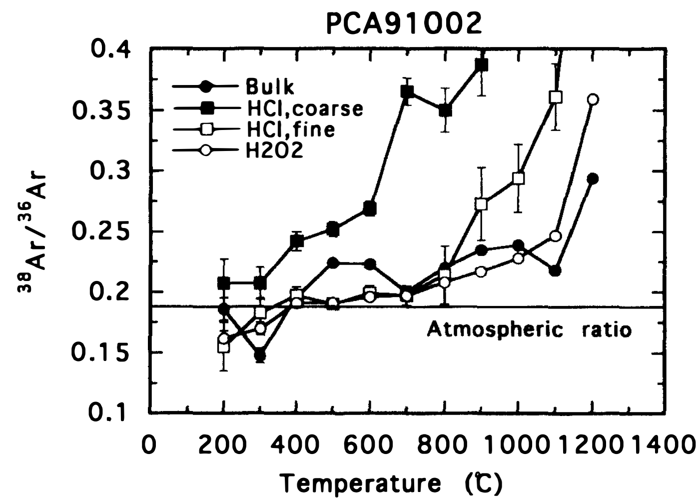

Fig. 4. $^{38}Ar/^{36}Ar$ isotopic ratios obtained by stepped combustion.

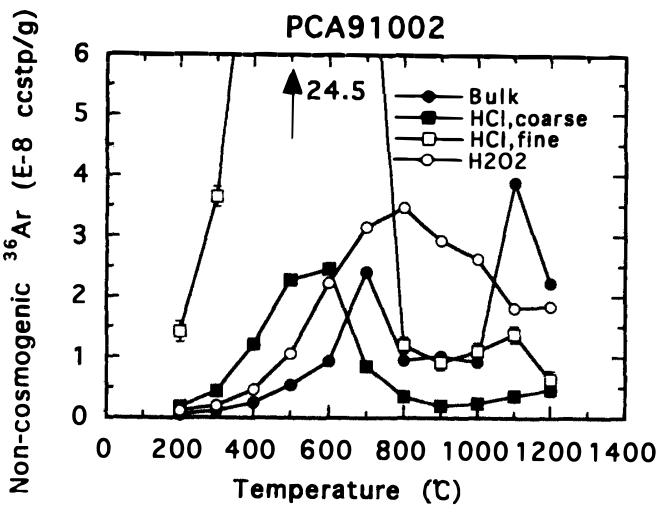

Fig. 5. Abundances of non-cosmogenic ^{36}Ar released by stepped combustion.

lower than the atmospheric ratio at 300°C for the bulk sample and the H_2O_2-treated sample. It has been shown that the ratio of the SW argon (0.182) is lower than the atmospheric ratio (0.187) (Benkert *et al.*, 1993). Since the abundance of Ar gas at that temperature is rather small, we are not sure if the observed value is significantly lower than the SW ratio. But, together with the high $^{20}Ne/^{22}Ne$ ratio, this indicates that this chondrite is rich in the SW gases relative to the SEP gases.

Figure 5 shows the release profiles of non-cosmogenic ^{36}Ar. The fine-grained HCl residue is rich in argon. Since it is not enriched in solar neon, the argon is probably mostly primordial (so called Q Ar which can be concentrated in HCl/HF residues) rather than solar Ar. (Judging from the isotopic ratio, most of the argon can not be due to air contamination.) The Ar release patterns for the bulk and the H_2O_2-treated samples are similar to those of the solar Ne release patterns, in that the bulk sample shows bimodal release while the H_2O_2-treated sample shows a broad single peak. Therefore, it is natural to consider that this Ar is mostly solar Ar, although there may be some contribution of primordial Ar at the highest temperatures (1100° to 1200°C).

Release patterns of nitrogen, the nitrogen isotopic compositions, and the excess ^{15}N (defined by delta^{15}N × nitrogen abundance × $(^{15}N/^{14}N)_{air}$) are shown in Figs. 6, 7 and 8, respectively. Isotopically light nitrogen is observed in the bulk and the H_2O_2-treated samples. The HCl treatment seems to remove most of the isotopically light nitrogen, leaving (or adding some) terrestrial nitrogen contamination with a delta ^{15}N of about 10 permil. Since a fairly constant value of delta ^{15}N of −60 permil is observed at various temperatures for the bulk and the H_2O_2-treated samples, this is considered to be the isotopic composition of the indigenous nitrogen. Then higher values of delta ^{15}N (−20 to −30 permil) in the bulk and the H_2O_2-treated samples are explained as a result of addition of isotopically heavier components. In the case of the bulk sample, we interpret that the heavier component is low temperature combustible material (delta ^{15}N is 10 to 15 permil), possibly organic in nature at 400°–600°C, and solar wind

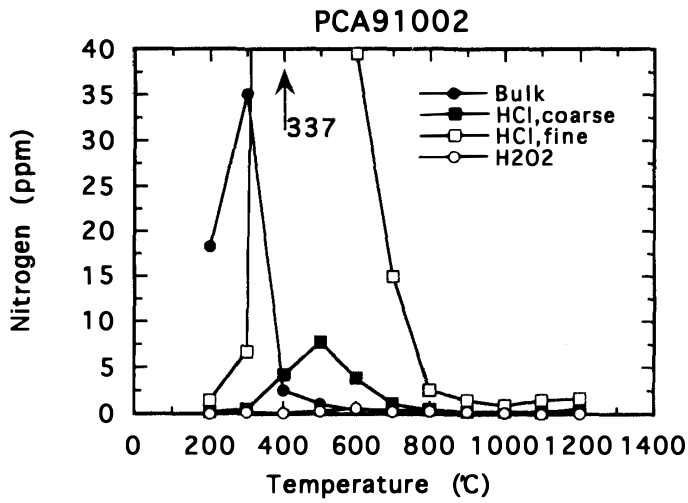

Fig. 6. Abundances of nitrogen released by stepped combustion. The abundant nitrogen observed in the fine-grained HCl residue sample could be terrestrial contamination, judging from the isotopic composition (see Fig. 7).

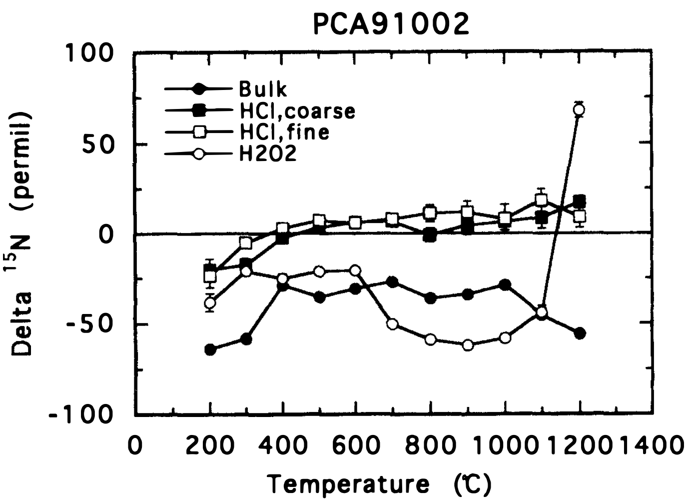

Fig. 7. Isotopic composition of nitrogen released by stepped combustion.

nitrogen (delta ^{15}N is 170 permil or higher) at 700°–1100°C (see Fig. 7 which shows that the bulk sample releases isotopically heavier nitrogen compared with the H_2O_2 treated sample in this temperature range). It is not possible to definitely prove this interpretation but this is consistent with our interpretation of nitrogen measurements of many ordinary chondrites (HASHIZUME, 1993) and a detailed study of a gas rich chondrite ALHA77216 (SUGIURA and ZASHU, 1994) which suggested that nitrogen in the solar wind is isotopically heavy.

As shown by the latter study, solar nitrogen is substantially removed by H_2O_2 treatment. The lower values of excess ^{15}N for the H_2O_2-treated sample (Fig. 8) as compared with the bulk sample over the temperature range from 700°C to 1000°C is

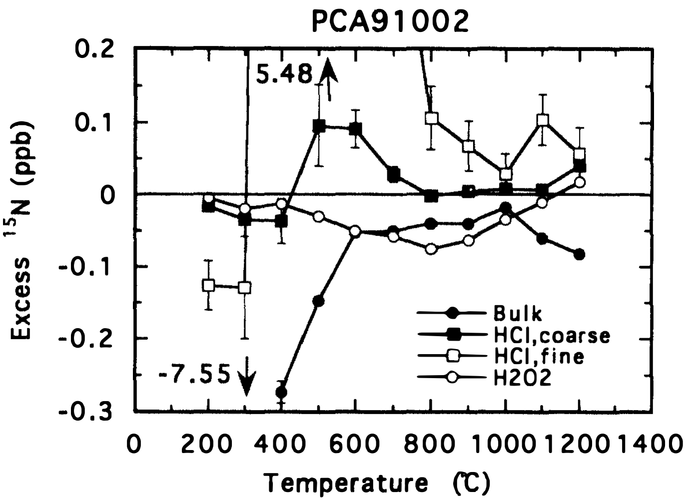

Fig. 8. Excess ^{15}N released by stepped combustion. The difference between the bulk and the H_2O_2-treated samples over 700°–1000°C, suggests that isotopically heavy solar nitrogen may be present in the bulk sample.

consistent with the interpretation that the solar nitrogen is removed by the H_2O_2 treatment. The abundant isotopically light nitrogen (delta ^{15}N is about −60 permil) released at low temperatures from the bulk sample is probably extra-terrestrial organic nitrogen, as it is removed by the H_2O_2 treatment (see Figs. 7 and 8). We do not know what the carrier of the light nitrogen which releases nitrogen at high temperatures is. In the case of ordinary chondrites, metal grains are such carriers (HASHIZUME, 1993), but in the case of R chondrites, the abundance of metal is quite small (RUBIN and KALLEMEYN, 1989). Further studies are necessary to identify this carrier.

The delta ^{15}N value for the H_2O_2-treated sample is slightly higher than −60 permil at low temperatures. We think that this is mainly due to mixing of a small amount of terrestrial contamination, coupled with removal of the majority of the indigenous nitrogen. Delta ^{15}N at 1200°C for the H_2O_2-treated sample is strongly affected by the cosmogenic nitrogen, because the treatment removed most of the indigenous nitrogen. The nearly constant delta ^{15}N value of −60 permil observed at various temperatures for the bulk and the H_2O_2-treated samples, suggests that the nitrogen is widely distributed among various minerals in this chondrite. This is important because it suggests that this is not a presolar component which is often observed in primitive ordinary chondrites.

To examine if the isotopically light nitrogen (delta ^{15}N=−60 permil) is solar nitrogen, the abundances of solar Ne and excess ^{15}N are compared in Fig. 9. (It has not yet been widely accepted that the solar nitrogen in meteorites is isotopically heavy.) There is no good correlation between these values, suggesting that the light nitrogen is not solar nitrogen. It is not clear why solar Ne is not more efficiently removed by H_2O_2 treatment compared with nitrogen. The tendency for Ne to be more stably retained than nitrogen against H_2O_2 treatment was observed for ALHA77216 (SUGIURA and ZASHU, 1994), and remains a puzzle.

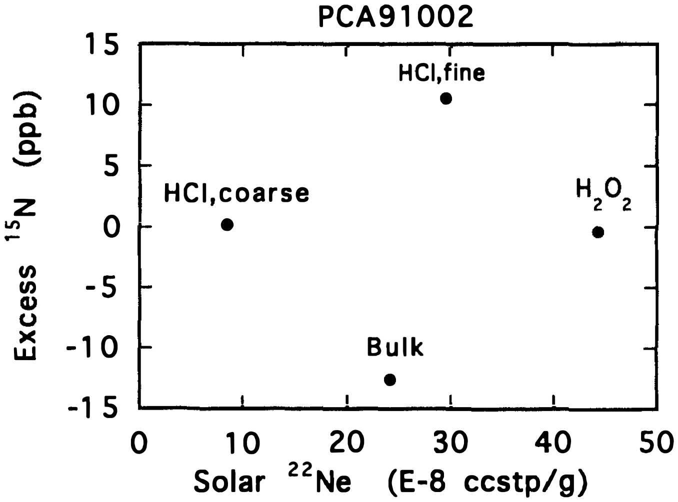

Fig. 9. Excess ^{15}N plotted against solar ^{22}Ne. The HCl fine sample may have a larger amount of terrestrial contamination, and thus may be disregarded. Absence of correlation suggests that the light nitrogen is not of solar nitrogen.

Table 6. Comparison with other gas-rich samples.

Sample	Excess ^{15}N ppb	Temp. range (°C)	Solar ^{22}Ne E-8ccstp/g	Non-cosmog. ^{36}Ar E-8ccstp/g	References
PCA 91002	0.16	(700–1100)	24.1	13.3	This study
ALHA77216	0.61	(700–1200)	121.8	32.5	SUGIURA and ZASHU, 1994
Apollo 67701	17.02	(600–800)	17750	76800	FRICK *et al.*, 1988

To further confirm our interpretation that the nitrogen isotopic composition (−30 permil) of the bulk sample over the temperature range 700°C–1100°C is a result of mixing of solar nitrogen (+170 permil) and indigenous nitrogen (−60 permil), we compared the abundances of the isotopically heavy nitrogen (+170 permil), solar Ne and non-cosmogenic Ar in PCA 91002 with those in another gas-rich chondrite and a lunar sample (Table 6). For this calculation, it was assumed that 1) the solar nitrogen is released from PCA 91002 from 700°C to 1100°C, 2) the indigenous nitrogen isotopic composition is −60 permil, and 3) the difference between this value and the observed value for the bulk sample is due to mixing of isotopically heavy (+170 permil) solar nitrogen. The result shows that the amount of heavy nitrogen relative to rare gases in PCA 91002 is somewhat higher than that in the lunar soil sample but is similar to that in the gas rich chondrite ALHA77216. Thus, our interpretation that the isotopically heavy nitrogen in the PCA 91002 is the solar nitrogen is not inconsistent quantitatively with previous observations.

4. Discussion

The PCA 91002 chondrite contains isotopically light nitrogen (−60 permil) which seems to be distributed among various minerals including organic materials. The bulk nitrogen isotopic composition (−57.3 permil, Table 1) is the lightest value ever observed for whole-rock chondrites, although lighter values have been observed in some temperature steps of various chondrites (*e.g.* GRADY *et al.*, 1986). Previously, the lightest average nitrogen isotopic composition was about −30 permil for the enstatite chondrite group (GRADY *et al.*, 1986). The bulk nitrogen isotopic composition of some iron meteorites is as low as −100 permil (FRANCHI *et al.*, 1993; PROMBO and CLAYTON, 1993) and that of Acapulcoites is as low as −150 permil (STURGEON and MARTI, 1991; KIM and MARTI, 1994). Thus there was a large gap between the lowest delta ^{15}N value for chondrites and that for more evolved meteorites. Now the gap is partially filled by the PCA 91002. The present result on PCA 91002 suggests that source materials for evolved meteorites are similar to those in primitive meteorites.

Figure 10 shows the oxygen-nitrogen isotope systematics of various meteorite groups. Assuming that the nitrogen isotopic composition of PCA 91002 is representative of the R group meteorites, the R group is located near the upper-left corner of the diagram. Such a location for a newly defined chondrite group has an important implication; our collection of chondrites on the Earth may be a rather limited collection of a much broader spectrum of asteroids at the main asteroidal belt. The presence of iron meteorites and Acapulcoites whose nitrogen isotopic compositions

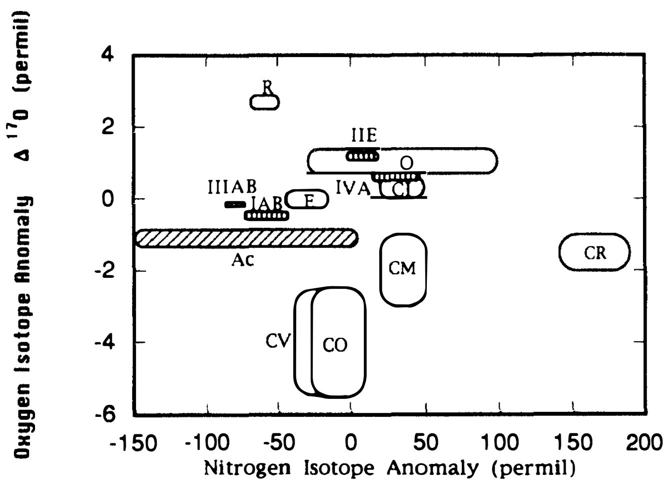

Fig. 10. Nitrogen–oxygen isotope diagram for chondrites. This figure is not meant to be rigorous, since the data quality for nitrogen isotopes is rather variable from one population to another. Some of the very anomalous samples (e.g. ALH 85085) are not included. The data are taken from the following literature: CLAYTON, 1993; HASHIZUME, 1993; KERRIDGE, 1985; KUNG and CLAYTON, 1978; GRADY et al., 1986, 1993; KIM and MARTI, 1994; CLAYTON et al., 1986, 1992. Evolved meteorites are shown by the hatched area. Ac stands for Acapulcoites and includes Lodranites.

extend down to -150 permil suggests that chondrite parent bodies with delta ^{15}N values as low as -150 permil still exist among the main belt asteroids.

It seems that there is no reason why the oxygen isotopic composition of chondrites should be limited to the range observed for the present collection of chondrites. In fact it is expected that the $\Delta^{17}O$ value of the solar nebula gas could be as high as 8.6 permil (CLAYTON and MAYEDA, 1984). Therefore, we expect that chondrite data points will extend at least to -150 permil for delta ^{15}N and to $+8.6$ permil for the $\Delta^{17}O$, which means that there is a lot of room for discovery of new groups of chondrites.

There seems to be no significant trend between nitrogen and oxygen isotopic compositions (Fig. 10). (Note that on such a diagram, a mixing line is not necessarily a straight line, if the nitrogen concentrations of the end members are different.) There is no doubt that the solar nebula was quite heterogeneous in composition. Yet it may be a result of mixing of a few end members. One of the aims of studying isotopes of meteorites is to find out such end members and to reveal their provenance. Several presolar grains which have been found in primitive chondrites (*e.g.* LEWIS *et al.*, 1987), are in a sense such end members. But they are rather minor in abundance and therefore they probably do not contribute significantly to the isotopic composition of bulk chondrites. Our study will continue to find major end members which became building blocks of meteorites and planets.

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