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## NITROGEN ISOTOPIC COMPOSITIONS OF SOME SOLAR-GAS-RICH CHONDRITES

#### Naoji SUGIURA, Kaoru KIYOTA and Shigeo ZASHU

Department of Earth and Planetary Physics, University of Tokyo, 11–16, Yayoi 2-chome, Bunkyo-ku, Tokyo 113

**Abstract:** Isotopically heavy nitrogen was detected in three solar-gas-rich (Weston(H4), ALHA77278(LL3.7) and Yamato-82133(H3)) and one solar-gas-poor (LEW86018(L3.1)) chondrites. Together with a previously studied solar-gas-rich chondrite, all solar-gas-rich chondrites that we examined have isotopically heavy nitrogen. Thus, we suggest that the solar nitrogen is isotopically heavy. The abundance of the heavy nitrogen is, however, not proportional to that of the solar Ne, suggesting complex processing on the surfaces of the parent bodies.

#### 1. Introduction

Determination of isotopic compositions of volatile elements in the solar wind is quite important, because the solar wind is regarded to represent the bulk solar composition which in turn represents the whole solar system. This is particularly true for nitrogen because the nucleosynthetic origin of <sup>15</sup>N is not well known (CLAYTON, 1983). The isotopic composition of solar wind nitrogen, detected in lunar soil samples is quite variable ranging from -200% to +200% (KERRIDGE, 1993). The cause of the variation is not well known. Solar-gas-rich meteorites are another valuable source of information on the solar wind composition. We reported previously (SUGIURA and ZASHU, 1994) that a solar-gas-rich chondrite (ALHA77216) has isotopically heavy nitrogen and suggested that it is solar nitrogen. It was recognized, however, that heavy nitrogen was enriched in the magnetic fractions, and the abundance relative to solar rare gases is higher than that in the lunar soil samples. Thus, the nitrogen (and rare gases) in ALHA77216 could not be unprocessed solar nitrogen, and there remained the possibilities that the heavy nitrogen could *e.g.*, be presolar or indigenous nitrogen.

Here, we report the presence of the isotopically heavy nitrogen in four ordinary chondrites three of which are solar-gas-rich, and compare them with the previous results on ALHA77216. We emphasize that the detection of isotopically heavy nitrogen in bulk primitive ordinary chondrites is rather rare. We have examined more than 20 bulk primitive ordinary chondrites and detected isotopically heavy nitrogen in seven chondrites (four reported here, ALHA77216, Yamato-74191 (considered to be due to presolar grains: SUGIURA and HASHIZUME, 1992) and Mezo Madaras (to be reported elsewhere)).

#### 2. Experimental

The chondrites we examined are Weston (H4), ALHA77278 (LL3.7), Yamato-82133

(H3) and LEW86018 (L3.1). The classifications are mainly based on SEARS *et al.* (1991). The former two chondrites are well known solar-gas-rich chondrites (*e.g.*, SCHULTZ and KRUSE, 1989). As will be shown later, the sensitivity and precision of our neon measurement is not very high, but we are fairly confident that Yamato-82133 is a solar-gas-rich chondrite. (Here, we define "solar-gas-rich" by the presence of solar Ne.) In an effort to detect solar Ne, two bulk samples of LEW86018 have been measured, but no solar Ne was detected. Therefore, by definition, this is not a solar-gas-rich meteorite. But it may be possible that a solar-gas-rich chondrite lost solar Ne by mild heating while retaining heavier gases because Ne diffuses more quickly than heavier gases. Since there are a couple of features which LEW86018 once had solar Ne which was lost subsequently, this chondrite is included in the present analyses.

In the case of Weston, magnetic and non-magnetic fractions were examined because it has been reported that metal in lunar soil contains isotopically heavy nitrogen (BECKER, 1989). The magnetic fraction was extracted with a hand magnet and still contains a large amount of silicates.

Since solar-gas-rich chondrites are quite heterogeneous, reproducibility of the results of duplicate measurements is not very good. The non-magnetic and magnetic fractions of Weston were prepared from a homogenized powder (typically 100 micrometer in size) sample. The rest of the samples were prepared from separate pieces which are typically 1 mm in size.

Nitrogen, neon and argon were extracted from samples of about 50 mg by stepped combustion. About 1 torr of oxygen was used for combustion. The temperature was increased from 200°C to 1200°C in steps of 100°C. The 1200°C combustion step was repeated to extract as much gas as possible. The gases obtained by the second extraction at 1200°C were always a small fraction of the amount released by the first one. Therefore, the two are combined and shown as a single value for 1200°C. Hot blanks (typically 0.2 ng for nitrogen,  $6 \times 10^{-12}$  ccSTP for <sup>22</sup>Ne, and  $8 \times 10^{-12}$  ccSTP for <sup>36</sup>Ar at 900°C) are always a small fraction of the amounts of extracted gases except for Ne at the highest temperatures. Blank corrections were made for all gases. The details of the gas extraction method and the mass spectrometry have been published elsewhere (HASHIZUME and SUGIURA, 1990; SUGIURA and HASHIZUME, 1992).

#### 3. Results

Total abundances of gases are summarized in Tables 1 and 2 and the results of stepped combustion are given in Tables 3 to 9.

#### 3.1. Comparison with previous studies and reproducibility

The results of rare gases in Weston and ALHA77278 can be compared with literature values (SCHULTZ and KRUSE, 1989). Our results of the bulk abundances of rare gases are not inconsistent with literature values if we take into account heterogeneities of gas rich chondrites and the uncertainty in the absolute sensitivity of our mass-spectrometry system (about 10%). The abundance ratio of <sup>22</sup>Ne to <sup>36</sup>Ar in the bulk Weston is about 3 which is consistent with the previous results found in the compilation of rare gases

Sample	Cosmo. <sup>21</sup> Ne	Solar <sup>22</sup> Ne	Trapped <sup>36</sup> Ar E-8 ccSTP/g	Cosmo. <sup>38</sup> Ar	<sup>40</sup> Ar
Weston bulk	9.65	42.87	17.36	1.18	3751
Weston nonmagnetic	11.82	40.56	15.17	0.50	3978
Weston magnetic	6.88	29.07	12.99	1.54	2968
ALHA77278	5.50	2.88	13.93	0.69	4939
Yamato-82133	0.94	0.27	10.30	0.098	11868
LEW86018 #1bulk	5.38	1.14	15.25	0.54	4664
LEW86018 #2bulk	5.96	0.50	21.38	0.53	3992

Table 1. Summary of rare gases.

Table 2. Summary of nitrogen.

Sample	Nitrogen (total)	$\delta^{15}\mathrm{N}_{ave}$ (total)	Nitrogen (700°C≤T)	δ¹⁵N <sub>ave</sub> (700°C≤T)	$\frac{\delta^{15}N_{max}}{(700^{\circ}C \le T \le 1100^{\circ}C)}$
	(ppm)	(‰)	(ppm)	(%0)	(%0)
Weston bulk	9.94	14.7	2.57	30.5	54.2
Weston nonmagnetic	2.16	9.7	0.39	14.0	18.3
Weston magnetic	5.70	15.6	1.61	33.0	71.1
ALHA77278 bulk	12.67	34.3	5.39	54.3	91.5
Yamato-82133 bulk	8.16	18.4	1.19	84.9	113.0
LEW86018 #1bulk	22.71	22.9	1 1.73	29.7	61.9
LEW86018 #2bulk	39.39	26.2	18.46	39.1	66.7

(SCHULTZ and KRUSE, 1989).

Weston is the only meteorite among the present samples that has been studied for their nitrogen isotopic composition (KUNG and CLAYTON, 1978; BECKER and PEPIN, 1991).. The present results of 9.94 ppm at  $\delta^{15}N=14.7\%$  on the bulk sample are not inconsistent with those reported by KUNG and CLAYTON (1978). The abundance (14 ppm) in a metal separate reported by BECKER and PEPIN (1991) is somewhat higher than that for the present magnetic fraction. This nitrogen is, however, probably mostly terrestrial contamination, because, as shown below for the present samples, it is mainly released at low temperatures and isotopically close to the atmospheric composition.

Since two bulk samples have been measured for LEW86018, reproducibility of the results can be examined. Generally speaking, the two data sets are consistent with each other. Therefore, the following discussion is mainly based on the bulk sample #1 and the data shown in the following figures are those obtained from the bulk sample #1, unless specified otherwise. An exception are the <sup>40</sup>Ar/<sup>36</sup>Ar ratios which are significantly different for the two bulk samples, well beyond the uncertainty of our measurement, suggesting that heterogeneity of the sample could be a major reason for the poor reproducibility. The neon data are not very reproducible mainly because of the poor data quality as discussed below.

#### 3.2. Neon

Ne in solar-gas-rich meteorites is considered to be mainly composed of solar Ne (SW and SEP), air Ne and cosmogenic Ne. Using appropriate isotopic compositions of

Temp.	Nitrogen	$\delta^{_{15}}N$	<sup>20</sup> Ne/ <sup>22</sup> Ne	<sup>21</sup> Ne/ <sup>22</sup> Ne	<sup>22</sup> Ne	<sup>38</sup> Ar/ <sup>36</sup> Ar	<sup>40</sup> Ar/ <sup>36</sup> Ar	<sup>36</sup> Ar
(°C)	(ppm)	(‰)		E-8	ccSTP/g		E-8	ccSTP/g
200	0.28	-3.9	12.47	0.050	0.22	0.186	213	0.31
	0.00	1.6	0.36	0.005	0.00	0.003	1	0.01
300	0.05	1.1	12.32	0.072	0.90	0.185	88	0.63
	0.00	6.0	0.19	0.004	0.01	0.002	1	0.01
400	1.28	-0.6	12.22	0.083	2.17	0.193	88	1.55
	0.00	1.4	0.15	0.003	0.02	0.002	0	0.01
500	2.38	10.7	11.74	0.987	3.32	0.205	201	2.18
	0.00	1.5	0.33	0.002	0.01	0.001	1	0.01
600	2.46	13.6	11.46	0.110	3.45	0.245	659	1.13
	0.00	1.5	0.16	0.002	0.01	0.002	3	0.01
700	1.24	13.2	11.21	0.121	5.31	0.238	418	1.48
	0.00	1.1	0.10	0.002	0.01	0.002	1	0.02
800	0.52	21.8	9.96	0.214	10.31	0.226	226	1.73
	0.00	1.5	0.05	0.002	0.02	0.001	1	0.01
900	0.58	15.4	9.64	0.232	12.15	0.232	96	2.62
	0.00	1.5	0.04	0.002	0.03	0.001	0	0.02
1000	0.48	20.6	9.90	0.194	7.36	0.272	108	2.48
	0.00	1.6	0.11	0.002	0.02	0.002	0	0.02
1100	0.17	54.2	8.80	0.266	3.61	0.295	229	1.49
	0.00	1.9	0.13	0.003	0.02	0.002	1	0.03
1200	0.50	67.3	6.96	0.390	5.02	0.299	174	2.55
	0.01	1.8	0.06	0.015	0.16	0.003	1	0.03
Total	9.94	14.7	9.99	0.203	53.82	0.244	207	18.14
	0.01				0.17			0.20

Table 3. Weston bulk (205.9 mg).

Tables 3-9;

Second row for each temperature shows 1 sigma error.

The errors involving weight of the sample and changes in sensitivity of the spectrometer (which are estimated to be about 10%) are not included in the errors of abundances.

these end-members (see SUGIURA and ZASHU, 1994 for details), the abundances of solar <sup>22</sup>Ne and cosmogenic <sup>21</sup>Ne were calculated (Table 1). Using production rates of cosmogenic Ne in chondrites (EUGSTER, 1988), cosmic ray exposure ages were calculated to be about 3 my for Yamato-82133, about 17 my for ALHA77278 and LEW86018 and 31 my for Weston, respectively. Our results on solar Ne in Yamato-82133, ALHA77278 and LEW86018 are not of good quality mainly because the abundances of the solar Ne are small (Fig. 1). An additional reason that the isotopic composition of solar Ne was not determined accurately is that planetary Ne may be present in these samples. It has been reported by SCHELHAAS et al. (1990) that ALHA77278 contains a fair amount of planetary Ne. In the case of LEW86018, because of the low petrologic grade (L3.1, SEARS et al., 1991) the presence of some planetary Ne is expected. The abundances of the solar <sup>22</sup>Ne given in Table 1 for these chondrites were estimated by subtracting the cosmogenic <sup>22</sup>Ne and hot blank <sup>22</sup>Ne from the observed value. Thus they are actually the abundances of trapped Ne which includes both solar and planetary Ne, in addition to some air Ne which could be important if hot blank corrections are not done accurately. On the Ne three isotope plot (not shown) several data points (with one sigma error bars) of Ne in

Temp.	Nitrogen	$\delta^{_{15}}N$	<sup>20</sup> Ne/ <sup>22</sup> Ne	<sup>21</sup> Ne/ <sup>22</sup> Ne	<sup>22</sup> Ne	<sup>38</sup> Ar/ <sup>36</sup> Ar	<sup>40</sup> Ar/ <sup>36</sup> Ar	<sup>36</sup> Ar
(°C)	(ppm)	(‰)		E-8	ccSTP/g		E-8	ccSTP/g
200	0.25	-3.8	12.77	0.029	0.24	0.179	123	0.21
	0.00	2.4	0.72	0.008	0.01	0.007	2	0.00
300	0.27	3.3	12.21	0.046	0.48	0.188	111	0.41
	0.00	2.4	0.53	0.008	0.02	0.005	1	0.01
400	0.47	9.9	11.58	0.089	1.62	0.190	129	1.795
	0.00	1.8	0.30	0.007	0.03	0.003	1	0.01
500	0.51	14.2	11.66	0.081	4.96	0.205	238	2.67
	0.00	1.7	0.15	0.003	0.05	0.002	1	0.02
600	0.27	13.7	11.12	0.143	2.64	0.239	641	1.03
	0.00	2.3	0.23	0.007	0.03	0.004	5	0.01
700	0.18	18.3	10.79	0.140	5.33	0.225	443	1.61
	0.00	3.0	0.15	0.005	0.05	0.003	3	0.01
800	0.09	2.8	9.35	0.251	11.81	0.209	320	1.43
	0.00	4.2	0.08	0.005	0.07	0.003	2	0.01
900	0.03	7.5	8.23	0.350	11.34	0.210	248	0.72
	0.00	7.0	0.07	0.006	0.07	0.004	2	0.01
1000	0.04	3.2	9.13	0.287	8.95	0.214	192	1.57
	0.00	6.7	0.09	0.006	0.06	0.003	1	0.01
1100	0.05	-16.0	9.23	0.255	6.20	0.219	169	3.55
	0.00	5.9	0.12	0.006	0.05	0.002	1	0.02
1200*	0.003	767.1	4.44	0.606	0.41	0.433	185	0.17
	0.00	366.0	0.47	0.052	0.02	0.013	3	0.01
Total	2.16	9.7	9.58	0.241	53.98	0.215	257	15.17
	0.01				0.07			0.04

Table 4. Weston nonmagnetic (37.6 mg).

\*First run at 1200°C was accidentally lost.

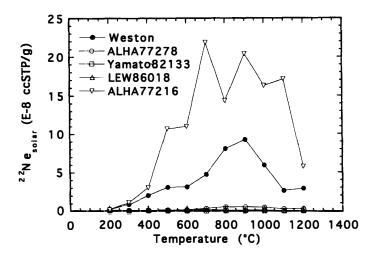


Fig. 1. Release patterns of solar neon from the present samples obtained by stepped combustion. ALHA77216 is also included for reference. The abundances of solar neon in ALHA77278 and Yamato-82133 are quite small compared with that in Weston. LEW86018 is probably a solar-gas-poor chondrite.

<sup>36</sup> Ar	<sup>40</sup> Ar/ <sup>36</sup> Ar	<sup>38</sup> Ar/ <sup>36</sup> Ar	<sup>22</sup> Ne	<sup>21</sup> Ne/ <sup>22</sup> Ne	<sup>20</sup> Ne/ <sup>22</sup> Ne	$\delta^{_{15}}N$	Nitrogen	Temp.
ccSTP/g	E-8		(‰)	(ppm)	(°C)			
0.10	167	0.173	0.09	0.036	13.82	3.5	0.83	200
0.00	3	0.009	0.01	0.014	1.24	1.6	0.00	
0.36	137	0.186	0.50	0.041	12.77	7.6	2.05	300
0.00	2	0.005	0.01	0.006	0.44	1.9	0.00	
1.31	163	0.191	1.33	0.090	12.08	6.2	0.46	400
0.01	1	0.003	0.02	0.006	0.26	2.2	0.00	
1.73	253	0.210	3.15	0.076	11.91	11.5	0.23	500
0.01	2	0.003	0.03	0.004	0.17	2.3	0.00	
0.98	711	0.280	2.28	0.131	11.21	14.4	0.29	600
0.01	5	0.004	0.03	0.006	0.19	2.1	0.00	
1.36	376	0.257	4.39	0.143	10.71	22.0	0.24	700
0.01	2	0.003	0.04	0.005	0.13	2.2	0.00	
1.55	164	0.265	7.44	0.235	9.29	17.9	0.21	800
0.01	1	0.003	0.17	0.009	0.23	2.4	0.00	
1.48	96	0.322	6.75	0.250	9.27	1.1	0.68	900
0.01	1	0.004	0.11	0.008	0.17	2.0	0.00	
1.72	94	0.356	5.20	0.216	9.64	6.1	0.29	1000
0.01	1	0.004	0.05	0.006	0.12	2.1	0.00	
1.77	171	0.344	3.69	0.290	8.34	71.1	0.16	1100
0.01	1	0.004	0.04	0.009	0.14	2.8	0.00	
1.66	110	0.323	2.06	0.393	7.18	149.1	0.25	1200
0.01	1	0.003	0.04	0.016	0.24	2.3	0.00	
14.02	212	0.283	36.88	0.210	9.79	15.6	5.70	Total
0.04			0.11				0.00	

Table 5. Weston nonmagnetic (48.0 mg).

Yamato-82133 are plotted above the air Ne-cosmogenic Ne mixing line. Therefore it is likely that a small amount of solar Ne is present in Yamato-82133. In the case of LEW86018 we are not sure if it contains solar Ne.

In the case of Weston, solar Ne is the dominant component, and the isotopic composition and the abundance can be determined fairly precisely. It can be seen (Fig. 1) that the abundance of solar Ne in Weston is by far the largest among the four samples although it is not as high as that in ALHA77216 (SUGIURA and ZASHU, 1994). The release patterns of the solar Ne from Weston and ALHA77216 are not grossly different, although it is released over a somewhat wider range of temperature from the ALHA77216. Solar Ne seems to be slightly enriched in the non-magnetic fraction compared with the magnetic fraction. (In the case of ALHA77216 both magnetic and non-magnetic fractions contain similar amounts of solar Ne.) The <sup>20</sup>Ne/<sup>22</sup>Ne ratio of the Weston is definitely higher than that of ALHA77216 (Fig. 2). It has been suggested that the high isotopic ratio (<sup>22</sup>Ne/<sup>20</sup>Ne)<sub>solar</sub> may mean that Weston had been exposed to the solar wind long ago (BECKER and PEPIN, 1991). Cosmogenic Ne in ordinary chondrites is released mainly at 800–900°C (Fig. 3). It is a bit peculiar that both cosmogenic Ne and solar Ne are released at similar temperatures from Weston. This is also true for ALHA77278 although this cannot be seen clearly in Fig. 1.

Temp.	Nitrogen	$\delta^{_{15}}N$	<sup>20</sup> Ne/ <sup>22</sup> Ne	<sup>21</sup> Ne/ <sup>22</sup> Ne	<sup>22</sup> Ne	<sup>38</sup> Ar/ <sup>36</sup> Ar	40Ar/36Ar	<sup>36</sup> Ar
(°C)	(ppm)	(‰)		E-8	E-8 ccSTP/g			
200	1.33	-6.3	13.04	0.069	0.04	0.190	389	0.22
	0.00	2.0	3.58	0.042	0.01	0.009	6	0.01
300	2.64	-2.3	8.45	0.114	0.07	0.199	924	0.31
	0.00	1.9	2.04	0.045	0.01	0.007	13	0.01
400	1.05	15.8	5.91	0.525	0.12	0.221	2362	0.22
	0.00	2.2	1.30	0.097	0.01	0.009	38	0.01
500	0.82	57.2	5.39	0.452	0.20	0.301	4007	0.19
	0.00	1.8	1.52	0.067	0.02	0.011	70	0.01
600	1.44	64.5	4.78	0.525	0.38	0.242	1701	0.55
	0.00	1.9	0.81	0.056	0.02	0.006	18	0.01
700	1.50	60.1	4.03	0.57	0.91	0.204	396	1.90
	0.00	1.8	0.54	0.040	0.04	0.003	2	0.02
800	1.83	47.5	2.88	0.672	2.15	0.213	230	1.68
	0.00	2.2	0.31	0.029	0.06	0.003	2	0.02
900	0.57	91.5	3.76	0.661	2.09	0.227	123	1.59
	0.00	2.3	0.48	0.036	0.08	0.003	1	0.01
1000	0.97	50.4	4.40	0.566	1.29	0.216	79	2.58
	0.00	1.7	1.12	0.058	0.10	0.002	0	0.02
1100	0.23	6.1	4.43	0.587	0.80	0.223	153	2.84
	0.00	2.6	2.38	0.115	0.13	0.002	1	0.02
1200	0.28	41.7	3.77	0.629	1.08	0.284	157	2.31
	0.00	2.4	3.06	0.156	0.23	0.003	1	0.02
Total	12.67	34.3	3.91	0.612	9.12	0.229	343	14.39
	0.01				0.31			0.13

Table 6. ALHA77278 bulk (25.8 mg).

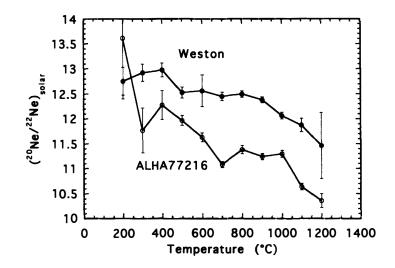


Fig. 2. Isotopic composition of solar neon obtained from Weston by stepped combustion is compared with that from ALHA77216.

## 3.3. Argon

The abundances of radiogenic <sup>40</sup>Ar (corrections were not made for trapped and cos-

Temp.	Nitrogen	$\delta^{_{15}}N$	<sup>20</sup> Ne/ <sup>22</sup> Ne	<sup>21</sup> Ne/ <sup>22</sup> Ne	<sup>22</sup> Ne	<sup>38</sup> Ar/ <sup>36</sup> Ar	<sup>40</sup> Ar/ <sup>36</sup> Ar	<sup>36</sup> Ar
(°C)	(ppm)	(‰)		E-8	ccSTP/g		E-8	ccSTP/g
200	2.63	1.7	nd	nd	nd	0.177	588	0.14
	0.00	1.8	nd	nd	nd	0.008	11	0.00
300	1.75	6.3	nd	nd	nd	0.177	1470	0.20
	0.00	1.6	nd	nd	nd	0.007	23	0.00
400	0.56	19.9	13.70	0.363	0.04	0.195	1899	0.36
	0.00	1.7	6.04	0.069	0.00	0.006	22	0.01
500	1.13	6.7	14.68	0.219	0.04	0.217	3490	0.19
	0.00	1.5	8.47	0.112	0.01	0.008	56	0.00
600	0.91	17.0	16.22	0.437	0.04	0.213	4069	0.18
	0.00	1.7	11.65	0.149	0.01	0.008	67	0.00
700	0.50	95.2	3.15	0.573	0.10	0.193	1314	1.01
	0.00	2.5	4.99	0.868	0.12	0.003	10	0.01
800	0.32	113.0	1.4	0.729	0.23	0.185	182	4.44
	0.00	2.9	0.49	0.197	0.04	0.002	1	0.02
900	0.17	99.9	1.75	0.833	0.36	0.189	487	1.19
	0.00	2.9	0.64	0.059	0.01	0.003	3	0.01
1000	0.05	19.5	5.12	0.826	0.20	0.199	2214	0.72
	0.00	5.1	3.18	0.080	0.01	0.004	19	0.01
1100	0.11	-9.7	11.64	0.766	0.15	0.208	2496	1.31
	0.00	3.6	7.24	0.104	0.01	0.003	16	0.01
1200	0.04	14.1	12.89	0.763	0.16	0.249	2935	0.63
	0.00	5.8	5.50	0.075	0.02	0.005	26	0.01
Total	8.16	18.4	6.20	0.399	1.34	0.195	1145	10.37
	0.00				0.13			0.03

Table 7. Yamato-82133 bulk (44.2 mg).

nd: not detected in significant amounts.

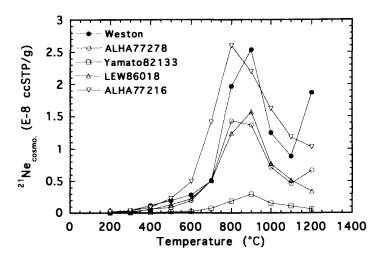


Fig. 3. Release patterns of cosmogenic <sup>21</sup>Ne. Most of the cosmogenic <sup>21</sup>Ne is released at 800–900°C by stepped combustion.

mogenic <sup>40</sup>Ar because their contributions are small), cosmogenic <sup>38</sup>Ar, and trapped <sup>36</sup>Ar are summarized in Table 1. An interesting feature common to the present solar-gas-rich

Temp.	Nitrogen	$\delta^{_{15}}N$	<sup>20</sup> Ne/ <sup>22</sup> Ne	<sup>21</sup> Ne/ <sup>22</sup> Ne	<sup>22</sup> Ne	<sup>38</sup> Ar/ <sup>36</sup> Ar	<sup>40</sup> Ar/ <sup>36</sup> Ar	<sup>36</sup> Ar
(°C)	(ppm)	(‰)		E-8	ccSTP/g		E-8	ccSTP/g
200	0.88	11.6	9.06	0.207	0.09	0.187	389	0.28
	0.00	1.9	1.57	0.056	0.01	0.007	6	0.01
300	1.82	4.1	6.18	0.238	0.10	0.192	904	0.40
	0.00	2.4	1.53	0.060	0.01	0.006	11	0.01
400	2.53	14.0	6.43	0.340	0.17	0.198	1531	0.67
	0.00	2.0	2.41	0.063	0.02	0.005	15	0.01
500	2.32	21.8	4.65	0.498	0.27	0.223	2977	0.58
	0.00	2.2	2.11	0.063	0.02	0.005	31	0.01
600	3.45	19.8	3.88	0.659	0.34	0.239	1656	0.55
	0.00	1.7	1.23	0.076	0.02	0.006	18	0.01
700	3.65	21.5	2.62	0.680	0.75	0.203	145	2.27
	0.00	2.3	0.51	0.052	0.03	0.003	1	0.02
800	3.52	20.5	1.59	0.827	1.50	0.201	28	2.94
	0.00	1.7	0.35	0.04	0.05	0.002	0	0.02
900	2.92	37.0	1.36	0.817	1.92	0.227	20	1.40
	0.00	1.9	0.38	0.044	0.07	0.004	0	0.02
1000	0.78	61.9	1.86	0.772	1.00	0.214	7	2.31
	0.00	2.0	1.03	0.088	0.09	0.003	0	0.02
1100	0.61	39.7	1.37	0.791	0.65	0.212	6	3.19
	0.00	1.8	2.02	0.165	0.11	0.002	0	0.03
1200	0.25	68.7	1.27	0.769	0.44	0.319	37	1.03
	0.00	2.8	5.26	0.387	0.21	0.006	0	0.04
Total	22.71	22.9	2.13	0.747	7.24	0.217	299	15.61
	0.01				0.27			0.15

Table 8. LEW86018 #1 bulk (22.9 mg).

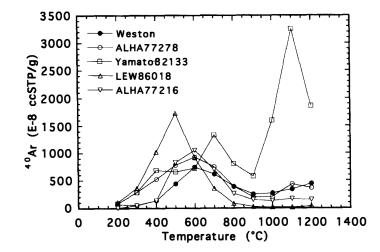


Fig. 4. Release patterns of radiogenic  ${}^{40}$ Ar. The release at low temperatures  $(T \sim 600^{\circ}C)$  seems to be characteristic to solar-gas-rich chondrites.

chondrites (except for Yamato-82133) is the release of  ${}^{40}$ Ar at relatively low (600°C) temperatures (Fig. 4). ( ${}^{40}$ Ar release by stepped combustion from non-gas-rich chondrites which will be published elsewhere often occurs at higher temperatures.) Such early re-

Temp.	Nitrogen	$\delta^{15}$ N	<sup>20</sup> Ne/ <sup>22</sup> Ne	<sup>21</sup> Ne/ <sup>22</sup> Ne	<sup>22</sup> Ne	<sup>38</sup> Ar/ <sup>36</sup> Ar	<sup>40</sup> Ar/ <sup>36</sup> Ar	<sup>36</sup> Ar
(°C)	(ppm)	(‰)		E-8	ccSTP/g		E-8	ccSTP/g
200	1.32	-8.0	5.61	0.175	0.02	0.189	393	0.45
	0.00	1.7	1.73	0.076	0.00	0.004	4	0.00
300	9.26	7.8	2.06	0.576	0.05	0.137	542	0.76
	0.01	1.7	0.80	0.143	0.01	0.003	4	0.01
400	2.48	-1.5	1.54	0.574	0.10	0.192	945	0.92
	0.00	1.5	0.75	0.206	0.03	0.003	6	0.01
500	3.14	26.5	1.32	0.756	0.17	0.219	2158	0.60
	0.00	1.3	0.53	0.077	0.01	0.004	17	0.01
600	4.72	36.0	1.27	0.751	0.40	0.224	985	0.77
	0.00	1.9	0.22	0.058	0.02	0.003	7	0.01
700	4.14	27.3	1.13	0.682	0.74	0.202	103	2.44
	0.00	1.5	0.30	0.126	0.12	0.002	1	0.01
800	8.25	36.5	1.04	0.851	1.67	0.218	101	1.47
	0.01	1.5	0.09	0.023	0.02	0.002	1	0.01
900	4.48	47.4	0.87	0.882	1.67	0.203	12	2.72
	0.00	1.5	0.12	0.026	0.03	0.002	0	0.01
1000	0.82	66.7	0.79	0.848	1.19	0.200	5	5.08
	0.00	2.8	0.23	0.045	0.05	0.001	0	0.02
1100	0.35	40.2	0.98	0.857	0.77	0.204	4	4.83
	0.00	2.2	0.46	0.056	0.04	0.001	0	0.02
1200	0.41	62.5	0.98	0.803	0.47	0.293	6	1.67
	0.00	1.6	1.34	0.120	0.06	0.003	0	0.01
Total	39.39	26.2	1.01	0.823	7.26	0.208	184	21.73
	0.02				0.15			0.05

Table 9. LEW86018 #2 bulk (62.2 mg)

lease of <sup>40</sup>Ar has also been observed for other solar-gas-rich chondrites, Feyetteville (WIELER *et al.*, 1989) and PCA91002 (SUGIURA and ZASHU, 1995). Since K containing minerals are not expected to be substantially oxidized at such low temperatures, the release is likely to be mainly due to diffusion. The dominance of low temperature release from solar-gas-rich chondrites is probably attributed to the small grain size of these chondrites as a result of gardening processes on the parent bodies.

The present solar-gas-rich chondrites have a fair amount of radiogenic <sup>40</sup>Ar, suggesting insignificant impact degassing from these chondrites. This is consistent with the presence of solar rare gases which could be degassed relatively easily by heating. A slight depletion of <sup>40</sup>Ar from Weston (also from ALHA77216) is likely to have been caused by localized heating associated with impact events which may be either comminution events to produce dark portions of solar-gas-rich chondrites or the final impact event which lithified the chondrite.

The release patterns of trapped (non-cosmogenic) <sup>36</sup>Ar are shown in Fig. 5. They are calculated by assuming the isotopic compositions of trapped Ar ( ${}^{38}$ Ar/ ${}^{36}$ Ar=0.187) and cosmogenic Ar ( ${}^{38}$ Ar/ ${}^{36}$ Ar=1.5). If the ratio of the abundances of solar Ne and solar Ar is constant (similar to that of solar abundances ~3) as observed in many solar-gas-rich meteorites, then the contribution of the solar Ar to the observed Ar contents is negligible for Yamato-82133, ALHA77278 and LEW86018 and the trapped Ar in these chondrites

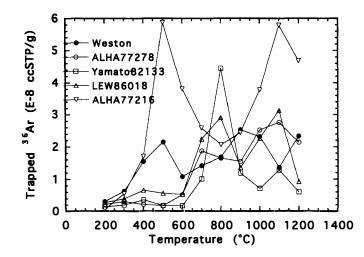


Fig. 5. Release patterns of trapped (solar + primordial) <sup>36</sup>Ar. The low and high temperature peaks from ALHA77216 is interpreted as due to release of SW and SEP neon, respectively. The 800°C peak from Yamato-82133 is characteristic to many primitive ordinary chondrites and considered to be due to primordial (presolar?) argon.

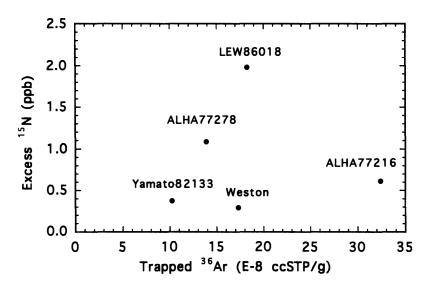


Fig. 6. Abundances of heavy nitrogen (excess <sup>15</sup>N) plotted against abundances of trapped <sup>36</sup>Ar. If both heavy nitrogen and trapped <sup>36</sup>Ar are of solar wind origin a good correlation is expected. In the case of LEW86018 average values of two bulk measurements are plotted.

is likely to be mostly primordial Ar. It is, however, possible that the solar Ne has been mostly lost by impact degassing while solar Ar has been retained due to its slower diffusion. If this is the case, then a part of the trapped Ar could be solar Ar. As shown in Fig. 6, abundances of heavy nitrogen (excess <sup>15</sup>N, see the next section for the definition) which is likely of solar wind origin are not proportional to the abundances of non-cosmogenic <sup>36</sup>Ar. If this is interpreted as a result of preferential loss of <sup>36</sup>Ar from LEW86018 and ALHA77278, then a lot of solar Ne must have been lost from these chondrites.

In the following we provide our interpretation of the trapped Ar release patterns (Fig. 5). The patterns are quite variable from one sample to another, but a couple of features can be recognized. The release pattern for Yamato-82133, which is characterized by a prominent peak at  $800^{\circ}$ C with an additional peak at  $1100^{\circ}$ C, is quite similar to that from the ALHA77214 (SUGIURA and HASHIZUME, 1992), which is a gas-poor chondrite. The Ar in ALHA77214 seems to be associated with isotopically light nitrogen which was suggested to be possibly carried by presolar grains. The same pattern has been observed for several primitive ordinary chondrites (unpublished data) and the carrier seems to be widespread among ordinary chondrites. Thus we consider that the peak in the Yamato-82133 release pattern is also due to this primordial (possibly presolar) Ar but not due to solar Ar. The 700-800°C Ar release peaks of LEW86018 and of ALHA77278 are also considered to be due to the same primordial Ar but not due to solar Ar. The peaks at 1100°C for these chondrites are larger than that of Yamato-82133. Since this is the temperature where ALHA77216 released Ar which is considered to be mostly SEP Ar (SUGIURA and ZASHU, 1994), it is possible that these peaks in the release profiles of LEW86018 and ALHA77278 are also due mostly to SEP Ar. It is, however, also possible that this peak is due to another kind of primordial Ar. SEP Ar, SW Ar and primordial Ar have slightly different isotopic compositions (BENKERT et al., 1993). Since cosmogenic Ar is also mixed in the extracted gases, it is not possible to distinguish these Ar according to the isotopic composition.

In the case of Weston (H4), solar Ar is considered to be the dominant component. But the <sup>36</sup>Ar release pattern for the bulk Weston is quite complicated with possibly three release peaks at 500°C, 900°C and above 1200°C. It is inferred from the data of ALHA77216 that the low temperature release peak is likely due to SW Ar. Stepped combustion results of many gas-poor ordinary chondrites show that primordial <sup>36</sup>Ar is mainly released at 1100–1200°C (except for those presumably due to presolar grains). Therefore, the high temperature release peak of Weston is likely due to trapped (non-solar) Ar. Since the light (gas-poor) portion of Weston contains a few times  $10^{-8}$  ccSTP/g of trapped Ar (SCHULTZ and KRUSE, 1989), this non-solar Ar quantitatively corresponds to the highest temperature release from the bulk sample. Then the release peak in the intermediate temperature range is considered to be due to SEP Ar. This interpretation on the release patterns of trapped <sup>36</sup>Ar might not be unique, but seems to be consistent with all available data. The nonmagnetic fraction of Weston contains slightly more non-cosmogenic <sup>36</sup>Ar than the magnetic fraction. The release pattern of <sup>36</sup>Ar from the nonmagnetic fraction shows three peaks similar to those of the bulk sample, while that from the magnetic fraction is rather flat. Thus the features of non-cosmogenic <sup>36</sup>Ar observed for the bulk Weston may be mostly attributed to the non-cosmogenic <sup>36</sup>Ar in silicates.

#### 3.4. Nitrogen

Nitrogen in chondrites consists of many components; 1) terrestrial contamination, 2) solar nitrogen, 3) isotopically anomalous, possibly presolar nitrogen, 4) isotopically nearly normal indigenous nitrogen, and 5) cosmogenic nitrogen. Since nitrogen has only two isotopes, it is not possible to separate these components unambiguously. However, by stepped combustion some of these components are released at different temperatures, making it possible to a certain degree to separate them into the five components. For

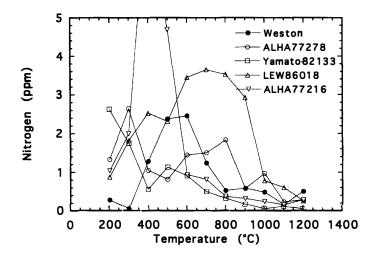


Fig. 7. Release patterns of nitrogen. Those samples which show large isotopic anomalies (Fig.6) tend to release rather small amounts of nitrogen in the intermediate temperature range.

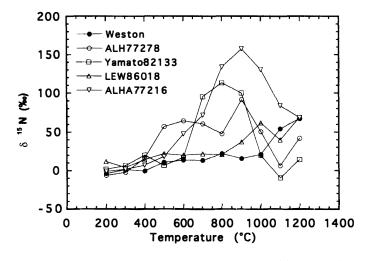


Fig. 8. Isotopic compositions of nitrogen released by stepped combustion. Heavy nitrogen is released mainly in the intermediate temperature range except for Weston.

instance, terrestrial contamination is released at lower temperatures, while cosmogenic nitrogen is released mostly at 1200°C. Release patterns of nitrogen are shown in Fig. 7, and the isotopic compositions are shown in Fig. 8.

The nitrogen released below 600°C is dominated by terrestrial organic nitrogen whose  $\delta^{15}N$  is about 15‰. According to a previous study on ALHA77216, isotopically heavy nitrogen which is considered to be solar nitrogen is mainly released from 700°C to 1000°C. The releases of these two components somewhat overlap. In Figs. 7 and 8 it is seen that the abundances of released nitrogen are rather small for ALHA77216 and Yamato-82133 in this intermediate temperature range between 700°C and 1000°C, and the maximum  $\delta^{15}N$  values are higher than 100‰. For the rest of the samples, in the same temperature range, the abundances of nitrogen are higher and the  $\delta^{15}N$  values are less than 100‰.

Such a negative correlation between the abundance and the  $\delta^{15}N$  value is also recognized in the slight changes in the abundance and the isotopic composition of nitrogen released from the present samples during the stepped combustion. For instance, an increase in the  $\delta^{15}N$  for LEW86018 up to 1000°C is associated with a rapid decrease in the abundance of nitrogen. Also, for ALHA77278 the maximum in the  $\delta^{15}N$  at 900°C corresponds to the minimum in the nitrogen abundance. Such anti-correlation between the abundance and the  $\delta^{15}N$  value can be explained as due to dilution of isotopically heavy (solar) nitrogen with isotopically nearly normal nitrogen which could be either indigenous to the chondrites or terrestrial contamination. At least qualitatively all the results (except for the Weston) in the intermediate temperature range are consistent with the above interpretation.

At 1100°C such anti-correlation between the nitrogen abundance and the isotopic composition fails. This is a temperature where a third component of nitrogen, that is isotopically anomalous, possibly a presolar component, is released by stepped combustion from primitive ordinary chondrites (unpublished data). The break down of the anti-correlation is probably due to the release of this third nitrogen component.

At 1200°C, there is  $\alpha$  significant contribution from the cosmogenic nitrogen, which causes an increase in the  $\delta^{15}N$  values. It is possible to estimate the contribution of the cosmogenic <sup>15</sup>N to the excess <sup>15</sup>N in these chondrites, based on the correlation between the abundances of the excess <sup>15</sup>N and the cosmogenic <sup>38</sup>Ar released from non-gas-rich chondrites at 1200°C (HASHIZUME, unpublished data). Excess <sup>15</sup>N (ppb) is defined by nitrogen abundance (ppm) × ( $\delta^{15}N/1000$ ) × ( $^{15}N/^{14}N$ )<sub>air</sub>, and is a measure of the abundance of isotopically heavy nitrogen. Figure 9 shows that there is a fair correlation between the abundances of these cosmogenic species for non-gas-rich chondrites, and also shows that the abundances of excess <sup>15</sup>N are higher for the solar-gas-rich chondrites are

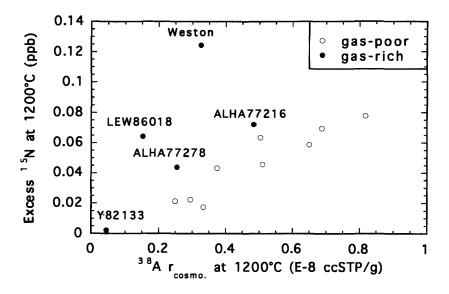


Fig. 9. Relationship between cosmogenic <sup>38</sup>Ar and excess <sup>15</sup>N released at 1200°C. The data for gas poor chondrites are taken from unpublished data of K. HASHIZUME. Release of non-cosmogenic heavy nitrogen from solar-gas-rich chondrites is suggested.

consistent with the idea that the isotopically heavy solar nitrogen is released at 1200°C from solar-gas-rich chondrites. This is particularly important for the interpretation of the Weston data, because isotopically heavy nitrogen is not detected by stepped combustion in the intermediate temperature range from 700°C up to 1000°C where isotopically heavy (solar) nitrogen is detected from the other solar-gas-rich chondrites. We are not sure why Weston behaves differently from the other solar-gas-rich chondrites, but based on Fig. 9 it is suggested that the Weston also contains isotopically heavy (solar) nitrogen which is somehow released at 1100–1200°C. The non-magnetic fraction of Weston contains a very small amount of nitrogen. The heavy nitrogen in Weston seems to be mostly carried by metals. Redistribution of nitrogen between metal and silicates seems to be required to explain this observation.

### 4. Discussion

All solar-gas-rich ordinary chondrites studied by us, three newly studied (excluding LEW86018) in addition to one previously studied seem to have isotopically heavy nitrogen. In addition to the chondrites described above, several solar-gas-rich chondrites have been examined for the nitrogen isotopic composition (KUNG and CLAYTON, 1978; GRADY and PILLINGER, 1988; MURTY and MARTI, 1990; SUGIURA and ZASHU, 1995). These previous studies showed that except for two chondrites (Feyettville and PCA91002), they all have isotopically heavy nitrogen, although in most cases the  $\delta^{15}N$  values are only marginally higher than the normal value. The R group chondrite PCA91002 is a solar-gas-rich chondrite. We reported previously (SUGIURA and ZASHU, 1995) that the (solar) nitrogen in this chondrite is considered to be isotopically heavy, but due to abundant indigenous nitrogen which is isotopically light, the measured isotopic composition which is a mixture of the solar and the indigenous nitrogen, is isotopically light. Since the details of nitrogen in this chondrite has been reported elsewhere, and since the presence of a large amount of indigenous light nitrogen prevented detailed analyses of the solar nitrogen in the chondrite, these data are not included in the present analysis. Detailed descriptions of the nitrogen in Feyettville have not been given, and the reason why it does not have isotopically heavy nitrogen is not known. In any case, the majority (seven out of eight) of solar-gas-rich chondrites seems to have isotopically heavy nitrogen which we suggest is solar nitrogen. In contrast, the probability of detecting isotopically heavy ( $\delta^{15}N > 50\%$ ) trapped nitrogen in gas-poor bulk ordinary chondrites is less than 10% (HASHIZUME and SUGIURA, 1995, and unpublished data on primitive ordinary chondrites). LEW86018 is counted as a gas-poor chondrite in this statistics.

We again note that solar Ne has not been positively detected in LEW86018. Therefore, there remains a possibility that the isotopically heavy nitrogen in this chondrite is not solar nitrogen. We list a couple of lines of circumstantial evidence which suggest that LEW86018 was a solar-gas-rich chondrite. 1) A small amount of Ne which seems to be in excess of the hot blank of the system is present in one of the samples, although the isotopic composition was not determined accurately. 2) <sup>40</sup>Ar is released at low temperatures which seems to be a feature common to solar-gas-rich chondrites. 3) The release pattern of trapped Ar is similar to that from another solar-gas-rich chondrite ALHA77278. These are the reasons, in addition to the presence of heavy nitrogen, that this chondrite

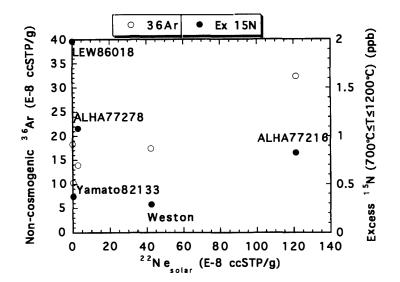


Fig. 10. Relationship between the abundances of solar Ne, non-cosmogenic Ar and excess <sup>15</sup>N for bulk samples. The non-cosmogenic Ar in Weston and ALHA77216 is mostly solar Ar, while that in the other chondrites is probably non-solar Ar. The excess <sup>15</sup>N in ALHA77216 is most probably due to solar nitrogen, while those in the other chondrites may be either due to solar nitrogen or due to indigenous nitrogen. In the case of LEW86018 average values of two bulk measurements are plotted for the nitrogen and argon, and the solar neon abundance is assumed to be zero.

is considered to have had solar Ne which was lost subsequently. However, whether LEW86018 was a solar-gas-rich or not, is not a crucial factor in the following discussion.

The combined data from this and the previous studies reveal that there are two major problems in the interpretation of isotopically heavy nitrogen in solar-gas-rich chondrites as solar nitrogen. Firstly, the release profile of heavy nitrogen from Weston which is a well known solar-gas-rich chondrite is quite different from the rest of solar-gas-rich chondrites. Secondly, there is no good correlation between the abundances of isotopically heavy nitrogen (excess <sup>15</sup>N) and solar rare gases (Fig. 10). Complicated history in the regolith of the parent bodies and also after the lithification seems to be required to explain these observations. In the following, we will explain what kind of processes are needed to explain these observations consistently.

#### 4.1. Why is Weston different from the other solar-gas-rich chondrites?

Weston is rather peculiar in the following points. (a) A large fraction of the solar Ne is released at ~900°C together with the cosmogenic Ne, suggesting that it might not be surface sited. (b) When compared with ALHA77216, solar rare gases are more concentrated in the Weston non-magnetic fraction than the magnetic fraction, while nitrogen is more concentrated in the Weston magnetic fraction than the non-magnetic fraction. In other words, solar wind species seem to be more fractionated in Weston than in ALHA77216. (c) No isotopically heavy nitrogen in Weston seems to be released until 1100°C, while it is released from about 700°C on from the other solar-gas-rich chon-

drites. The features (a) and (b) suggest that Weston is more thermally processed than ALHA77216. The feature (c) is not easy to explain but may be due to the presence of isotopically light indigenous nitrogen which counter-balances the isotopically heavy solar nitrogen at the lower temperatures. Electron microscopic observations suggest that Weston experienced rather mild shock heating which may or may not be consistent with the features (a) and (b), and that Weston may have experienced terrestrial weathering which has altered grain surfaces (ASHWORTH and HUTCHISON, 1975; ASHWORTH and BARBER, 1976). Feature (c) may somehow be related to this terrestrial weathering. The isotopic compositions of solar Ne in Weston and ALHA77216 are quite different (Fig. 2). This difference may also somehow be related to the difference in the release behavior of nitrogen between these chondrites.

# 4.2. Why is there no correlation between the abundances of solar rare gases and heavy nitrogen?

If the heavy nitrogen is of solar wind origin there should be good correlations between the abundance of heavy nitrogen (excess <sup>15</sup>N) and the abundances of solar rare gases. Figure 10 shows that this is not the case. In the case of trapped Ar in ALHA77278 and Yamato-82133, the presence of a large amount of primordial <sup>36</sup>Ar, as discussed above, is an obvious explanation for the lack of correlation between the abundances of solar Ne and Ar. In the case of heavy nitrogen it will be an extreme coincidence if isotopically heavy indigenous nitrogen (which has a similar isotopic composition and a release pattern to solar nitrogen) is present in these two chondrites. It seems more likely that the abundance of solar nitrogen has not been much changed since (or during) the formation of solar-gas-rich chondrites, while the abundances of solar rare gases have been reduced by various heating events which occurred to these two chondrites.

Let us examine if such an explanation is consistent with the other observations. The abundance ratio of solar Ne to excess <sup>15</sup>N is considered to indicate the degree of neon loss due to heating events. Thus, ALHA77278 and Yamato-82133 are considered to have experienced higher temperatures due to reheating than ALHA77216. The ratio of solar Ne to excess <sup>15</sup>N for Weston is similar to that of ALHA77216. However, as discussed above, heavy nitrogen in Weston is not detected in the intermediate temperature range, either due to a loss during terrestrial weathering or due to simultaneous release of light nitrogen. In either case, it is considered that the abundance of solar nitrogen in Weston was once much larger than that indicated in Fig. 10. Therefore, we think that Weston experienced higher temperatures than ALHA77216. In this context, LEW86018 is considered to be an extreme case where solar Ne was completely lost by heating events. Release profiles of trapped Ar from LEW86018 and ALHA77278 (only the high temperature release peak is attributed to solar Ar) shown in Fig. 5 are consistent with the above interpretation. The release profiles of <sup>40</sup>Ar, *i.e.* release at low temperatures, may seem to be in contradiction with the above interpretation but if the solar gases had been acquired at a very early stage of the parent body formation and then lost still early in the history of the solar system, then we may consider that <sup>40</sup>Ar was once lost together with solar rare gases and then accumulated afterwards. Thus the assumption that the heavy nitrogen in LEW86018 is of solar wind origin seems to be not inconsistent with the other observations. However, this is far from saying that the nitrogen in LEW86018 is of solar wind origin. In fact it is quite possible that the heavy nitrogen in LEW86018 is indigenous nitrogen which happens to be isotopically heavy.

#### 5. Conclusions

Nitrogen isotopic compositions in solar-gas-rich chondrites were found to be isotopically heavy. We think this is solar nitrogen. But there are two problems in this interpretation. Firstly, the release profile of heavy nitrogen from Weston which is a well known solar-gas-rich chondrite is quite different from the rest of solar-gas-rich chondrites. Secondly, the abundances of the isotopically heavy nitrogen are not proportional to the abundances of solar rare gases. We examined a working hypothesis that the abundance of heavy nitrogen best represents the total amount of implanted solar wind, while the abundances of solar rare gases have been reduced by heating events since or during the formation of solar-gas-rich chondrites. It was found that such a hypothesis is not inconsistent with various observations. But in the case of LEW86018 which actually does not contain solar rare gases, the heavy nitrogen could be explained as indigenous nitrogen.

It seems that the presence of heavy nitrogen in solar-gas-rich chondrites is well established but further studies are needed to establish the origin.

#### Acknowledgments

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