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Noble Gases, Nitrogen and Carbon Isotopic Compositions of the Ghubara Meteorite, Revealed by Stepwise Combustion and Crushing Methods[1]

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Abstract—The Ghubara meteorite contains abundant trapped gases in voids of highly retentive phases that can be released by stepwise crushing and thermal degassing. Their composition is dominated by the solar wind component and by radiogenic argon. We favor a scenario in which a large impact event on L-chondrite asteroid 470 Ma ago caused release, mobilization, fractionation and redistribution of accumulated gases on the Ghubara parent body. The Ghubara breccia was formed at that event and occluded trapped gases into the voids. The uncommonly high $^{20}Ne/^{36}Ar$ ratios of the analysed samples compared to the solar composition is considered to be due to trapping of gases released from surrounding rocks that lost light noble gases preferentially over the heavy ones. The $^{4}He/^{20}Ne$ and $^{4}He/^{36}Ar$ ratios, being as usually lower than in solar wind, gradually increase during stepped crushing, indicating non equilibrium distribution of the gases between the voids of different sizes that can be caused by the dynamics of the shock metamorphism process. The neon isotopic composition released by stepwise crushing and combustion is a mixture of two components: solar dominating trapped and cosmogenic Ne. The former component is mainly degassed in the initial crushing steps opening the large inclusions/voids, while the relative contribution of the latter, likely released from galactic cosmic ray produced tracks, increases with progressive crushing. During stepwise combustion the same trend in the release of the Ne components with increasing temperature is observed. The nitrogen and carbon abundances as well as their isotopic compositions in Ghubara are usual for ordinary chondrites. Most of nitrogen is chemically bounded and associated with carbon. The delivery time of Ghubara from the parent body asteroid to the Earth calculated from its exposure age is 9–28 Ma.

Keywords: meteorites, L-chondrite parent body, noble gases, nitrogen, carbon, trapped extraterrestrial argon

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INTRODUCTION

Trapped argon with isotopic composition different from primordial/solar/terrestrial in samples of asteroidal origin was for the first time identified in L-chondrites (Korochantseva et al., 2007) and later in meteorites from other asteroids (e.g., Hopp et al., 2014; Trieloff et al., 2018). The results of Korochantseva et al. (2007) gave strong evidence that this component had been trapped following a large-scale impact event at ~470 Ma ago on the L-chondrite parent body. Its genesis was related to mixing and homogenization of the implanted solar argon and radiogenic ^{40}Ar mobilized and redistributed during thermal processes accompanying the shock episode. In attempt to specify the location of the trapped Ar and to check its relation to other light noble gases, nitrogen and carbon isotope compositions we carried out a combined noble gas, nitrogen, and carbon stepwise crushing and combustion for different lithologies (host, xenolith, impact melt inclusion) of the Ghubara meteorite. The application of stepwise crushing to the extraterrestrial materials is relatively uncommon mainly due to the low amounts of the released gases. As it was shown (Korochantseva et al., 2007) the Ghubara chondrite is a gas-rich meteorite in which $^{36}Ar_{trapped}$ accounts for >82% of the total ^{36}Ar release with the rest being of cosmogenic origin, this made it possible to assume the stepwise crushing technique to be an effective method of gas extraction. Here we report the first data obtained by this method for L-chondrites.

SAMPLE DESCRIPTION

Ghubara is a slightly weathered (W1) darkened L chondrite of petrological Type 5, indicating thermal metamorphism at temperature ~600°C (Dodd, 1981;

[1] The article is published in the original.

McSween et al., 1988). Its shock metamorphism level (S4) corresponds to the equilibrium shock pressures of ~30–35 GPa and the post-impact temperature up to 250–350°C (Stöffler et al., 1991). Ghubara is a regolith breccia (Ferko et al., 2002) comprised of chondrules, chondrule and mineral fragments, minor chondritic xenoliths and melt rock inclusions joined together by the re-crystallized matrix of fine-grained mineral and chondrule clasts. The xenolith represents a light-gray chondritic rock fragment (L5) of 1.5 cm^2 in size. The achondritic melt inclusion (1 × 0.4 cm) is composed of medium-grained olivine (50 vol %) and pyroxene (45 vol %) settled in the cryptocrystalline mesostasis of feldspathic composition. The compositions of silicates correspond to the host chondrite. When compared to the latter, the inclusion is depleted in FeNi metal and troilite (<0.1 vol %) that is a feature of some other melt rocks of the L-chondrite composition (e.g., Lorenz et al., 2018). The formation of the melt isclusion is related to the catastrophic impact on the L-chondrite parent body (Korochantseva et al., 2007). Detailed information on the petrography and mineralogy of the xenolith and impact melt inclusion is given by Korochantseva et al. (2007).

EXPERIMENTAL TECHNIQUES

The high sensitivity Finesse mass-spectrometer complex was used for analyses of Ghubara samples (host, xenolith and impact melt inclusion) at The Open University (Verchovsky et al., 1997). The samples of host chondrite (62.87 mg), xenolith (83.35 mg) and impact melt inclusion (42.05 mg) were stepwise crushed with cumulative number of strokes up to 4500. The samples of host (4.301 mg), xenolith (4.045 mg) and two samples of impact melt inclusion (#1 of 3.753 and #2 of 4.478 mg) wrapped in clean platinum foil were combusted in oxygen, supplied from CuO, in a double-walled quartz-ceramic furnace from 200 up to 1500°C with variable increments (up to 13 steps) for 30 minutes at each temperature step, followed by 15 min for oxygen resorption, before to transfer the gases produced to the clean-up section. He and Ne have been analysed on a quadrupole mass spectrometer, while N_2 and Ar on a magnetic sector mass spectrometer in static mode. Nitrogen was not analysed in the host and impact melt inclusion (#1) by combustion. The carbon measurements were carried out (on a separate magnetic sector mass spectrometer) only for combustion experiments because of very low amounts of carbon released by stepwise crushing.

The clean up procedure of gases released by combustion and crushing methods was identical (Verchovsky et al., 1998, 2002). First, the released gases were cryogenically separated using two cryotraps cooled with liquid nitrogen, one of which was filled with molecular sieve 5 Å and the other, made of glass, was empty. Argon and neon were purified using Ti–Al getters, and nitrogen—using a CuO furnace to ensure no CO was present. Carbon yields (recorded as ng of C) were calculated using pressure of CO_2 measured on a calibrated MKS Baratron™ capacitance manometer. Nitrogen and Ar yields were measured by peak height method via calibration of the mass spectrometer with known amounts of standard gases. Gases were transferred to different parts of the machine using a system of computer-controlled pneumatic valves.

In order to reduce the contributions from CO_2^{++} and $^{40}Ar^{++}$ on Ne masses (22 and 20), a low ionization voltage of ~40 V was used in the quadrupole ion source. Also, Ar presented in the system was cooled down on the molecular sieves, and the Ti–Al getter was open to the mass spectrometer chamber during Ne measurements.

The isotopic data are expressed using the delta (δ) notation, as parts per thousand (‰) deviations from standards (Vienna Peedee Belemnite (VPDB) for C, and terrestrial air (AIR) for N). The system blanks were monitored between sample analyses by putting an empty clean Pt foil bucket through the same stepped combustion procedure used for the sample analyses and collecting both abundance and isotopic data. Typical system blank levels for stepped combustion were <10 of C and <1 ng of N. Typical system blanks for ^4He were $<1 \times 10^{-8}$ cc, for ^{20}Ne were $<6.5 \times 10^{-10}$ cc, and for ^{40}Ar were $<8 \times 10^{-9}$ cc. For crushing analyses, the system blanks were monitored at several stages during the total sample crushing runs by stopping crushing and closing off the crushing tube for a length of time comparable to the next crushing step duration, and typically these were 0.4–1.2 ng of N, $<1 \times 10^{-10}$ for ^{20}Ne, $(0.3–1.6) \times 10^{-9}$ cc for ^4He, and for ^{40}Ar were $(04.–1.1) \times 10^{-8}$ cc.

Uncertainties of absolute concentrations of gases are 5–10%, and elemental ratios of noble gases are estimated to have an uncertainty of about 5%.

RESULTS AND DISCUSSION

Detailed data on the Ghubara samples are given in Tables 1–7. The bulk amounts and isotopic compositions for all samples are summarized in Table 8.

Noble Gas Release

The stepwise combustion of the Ghubara samples shows that the main release of Ne and Ar occurs at high temperatures (1200 and 1300°C; Fig. 1). In particular, in chondritic samples (host and xenolith) Ne and Ar are simultaneously degassed at 1200°C. The main release of ^4He in these specimens is observed as usual at lower temperatures due to its high diffusion rate (Fig. 1). However, the chondritic and impact melt samples are different in the degassing behavior of helium: the main release of ^4He in the impact melt material is observed at a higher temperature (1100°C)

Table 1. The results for noble gases (He, Ne and Ar) and N_2 obtained by stepwise crushing of the Ghubara host (62.87 mg)

Extractions[a]	^{4}He[b]	^{20}Ne[b]	^{21}Ne/^{22}Ne	^{20}Ne/^{22}Ne	^{40}Ar[b]	^{40}Ar/^{36}Ar	^{36}Ar[b]	^{36}Ar/^{38}Ar	N[c]	δ^{15}N	^{4}He/^{36}Ar	^{20}Ne/^{36}Ar	^{4}He/^{20}Ne
10	6533	299	0.0355(14)	12.74(11)	854	234(3)	3.65	5.25	181	18.6(6)	1792	82	22
40	8592	361	0.0352(13)	12.32(8)	958	227(3)	4.22	4.96	127	24.5(9)	2034	85	24
120	12513	630	0.0408(10)	13.31(8)	1090	222(3)	4.91	5.47	67	31.8(1.1)	2548	128	20
360	11333	371	0.0385(13)	12.49(10)	883	218(2)	4.04	5.34	48	39.3(1.6)	2802	92	31
1080	7347	151	0.0478(25)	12.77(16)	269	211(2)	1.27	n.a.	60	25.1(9)	5767	119	49
2100	2916	41	0.0689(53)	12.23(30)	56	179(2)	0.31	n.a.	110	5.9(3)	9391	133	71
3500	1945	29	0.1141(80)	11.60(31)	6	n.a.	n.a.	n.a.	84	19.7(7)	n.a.	n.a.	68
Total	51179	1882	0.0438(18)	12.76(10)	4115	223(3)	18.41	5.26	678	21.1(8)	2772	102	27

[a]Cumulative number of strokes.
[b] $\times 10^{-8}$ cm^3 STP/g.
[c] ppb.
The uncertainty in absolute concentrations of noble gases is 5–10%.
The numbers in parentheses refer to the last digits and are 1σ-uncertainties.
The uncertainties for ^{36}Ar/^{38}Ar were <0.01.
n.a.—not analysed.

Table 2. The results for noble gases (He, Ne and Ar) and N_2 obtained by stepwise crushing of the Ghubara xenolith (83.35 mg)

Extractions[a]	^{4}He[b]	^{20}Ne[b]	^{21}Ne/^{22}Ne	^{20}Ne/^{22}Ne	^{40}Ar[b]	^{40}Ar/^{36}Ar	^{36}Ar[b]	^{36}Ar/^{38}Ar	N[c]	δ^{15}N	^{4}He/^{36}Ar	^{20}Ne/^{36}Ar	^{4}He/^{20}Ne
6	4680	402	0.0340 (6)	12.52(4)	1503	231(24)	6.50	5.34*	846	4.0(2)	720	62	12
15	5292	496	0.0340(5)	12.32(4)	1208	208(22)	5.81	n.a.	176	4.9(4)	910	85	11
35	5918	584	0.0322(5)	12.31(4)	1148	194(20)	5.91	5.39*	251	6.2(4)	1001	99	10
80	6036	511	0.0332(5)	12.41(4)	920	186(1)	4.95	5.31(1)	104	7.0(6)	1219	103	12
170	5884	454	0.0312(6)	12.45(4)	777	185(1)	4.20	n.a.	62	6.2(7)	1402	108	13
350	1237	336	0.0292(4)	10.97(3)	543	177(1)	3.07	5.06(1)	56	19.1(2.0)	403	109	4
1050	1203	373	0.0311(5)	10.80(3)	608	172(1)	3.54	5.46(1)	52	13.1(1.5)	340	106	3
2200	1170	427	0.0301(4)	10.42(3)	738	169(1)	4.35	5.44(1)	42	33.3(4.2)	269	98	3
4500	1145	437	0.0316(5)	10.33(3)	776	162(1)	4.80	5.38(2)	26	94.9(13.8)	239	91	3
Total	32565	4019	0.0320(5)	11.62(4)	8221	191(10)	43.13	5.35(1)	1616	7.8(7)	755	93	8

[a]Cumulative number of strokes.
[b] $\times 10^{-8}$ cm^3 STP/g.
[c] ppb.
The uncertainty in absolute concentrations of noble gases is 5–10%.
The numbers in parentheses refer to the last digits and are 1σ-uncertainties.
* The uncertainties of these values were <0.01.
n.a.—not analysed.

than in the chondritic material (600–900°C). The high temperature release of Ar is typical of impact melts and shocked meteorites (e.g., Trieloff et al., 2018). The high temperature release of the light noble gases can probably be also related to the changes in the diffusion properties of minerals affected by shock metamorphism. During crushing noble gases and nitrogen are mostly released in the first steps. Afterwards the efficiency of gases release, defined as the amount of gas released per stroke, sharply decreases (Fig. 2).

Noble Gas Isotopic Compositions

The ^{20}Ne/^{22}Ne ratios of the studied Ghubara samples are close to solar with maximum value of 13.31 ± 0.08 measured in the host (Table 1). The literature data on Ghubara noble gases demonstrate that the ^{20}Ne/^{22}Ne ratio varies from 1.61 to 13.75 in different specimens (Schultz and Franke, 2004) indicating a significant heterogeneity of this breccia. The Ne isotopic compositions of all samples analysed here are

Table 3. The results for noble gases (He, Ne and Ar) and N_2 obtained by stepwise crushing of the Ghubara impact melt inclusion (42.05 mg)

Extractions[a]	$^{4}He^{b}$	$^{20}Ne^{b}$	$^{21}Ne/^{22}Ne$	$^{20}Ne/^{22}Ne$	$^{40}Ar^{b}$	$^{40}Ar/^{36}Ar$	$^{36}Ar^{b}$	$^{36}Ar/^{38}Ar$	N^{c}	$\delta^{15}N$	$^{4}He/^{36}Ar$	$^{20}Ne/^{36}Ar$	$^{4}He/^{20}Ne$
10	6288	614	0.0352(8)	12.54(6)	1511	378(31)	4.00	5.22(2)	642	0.3(2)	1573	153	10
30	5962	435	0.0365(8)	12.71(7)	817	300(22)	2.72	4.45(1)	143	4.1(5)	2190	160	14
120	10040	606	0.0368(8)	12.78(6)	1140	312(23)	3.65	4.76(3)	86	10.4(1.3)	2748	166	17
400	10818	514	0.0399(9)	12.74(6)	1055	319(2)	3.30	5.41(2)	67	19.2(2.5)	3274	155	21
1200	5700	167	0.0551(15)	12.02(8)	439	298(2)	1.47	5.00(7)	98	10.4(1.2)	3878	113	34
2400	1094	19	0.1334(31)	10.59(8)	89	282(2)	0.32	5.02(7)	188	4.2(7)	3464	60	58
Total	39902	2354	0.0421(9)	12.62(6)	5051	327(18)	15.46	4.97(3)	1222	3.9(6)	2580	152	17

[a]Cumulative number of strokes.
[b] $\times 10^{-8}$ cm^3 STP/g.
[c] ppb.
The uncertainty in absolute concentrations of noble gases is 5–10%.
The numbers in parentheses refer to the last digits and are 1σ-uncertainties.

Table 4. Stepwise combustion data for noble gases (He, Ne and Ar) in the Ghubara host (4.301 mg)

T, °C	$^{4}He^{a}$	$^{20}Ne^{a}$	$^{21}Ne/^{22}Ne$	$^{20}Ne/^{22}Ne$	$^{40}Ar^{a}$	$^{36}Ar^{a}$
200	n.a.	n.a.	n.a.	n.a.	258.0	0.94
300	54	n.a.	n.a.	n.a.	86.1	0.35
400	6	n.a.	n.a.	n.a.	9.4	b.d.
500	213	20	0.0426(134)	n.a.	1.3	b.d.
600	1958	80	0.0479(82)	12.13(48)	4.8	0.02
700	9283	144	0.0528(70)	12.34(44)	0.4	b.d.
800	18203	167	0.0677(66)	11.96(37)	b.d.	b.d.
900	18550	190	0.0685(68)	12.86(37)	b.d.	0.03
1000	10794	181	0.0574(58)	12.17(36)	29.8	0.25
1100	6935	337	0.0583(43)	12.00(25)	171.4	1.14
1200	4176	1403	0.0605(22)	12.10(14)	3846.8	15.02
1300	1123	120	0.0837(84)	11.99(41)	402.9	3.07
1400	n.a.	n.a.	0.0940(156)	n.a.	233.8	1.84
Total	71296	2642	0.0620(41)	12.15(24)	5044.6	22.66

[a] $\times 10^{-8}$ cm^3 STP/g.
The uncertainty in absolute concentrations of noble gases is 5–10%.
The numbers in parentheses refer to the last digits and are 1σ-uncertainties.
n.a.—not analysed.
b.d.—below detection.

shown on the neon 3-isotope diagram (Fig. 3a), which displays a trend reflecting a mixture between predominant solar/fractionated solar wind and minor cosmogenic Ne with a higher contribution of the latter at the end of crushing (Fig. 3b) and combustion (Fig. 4). Similar correlation and sequence of release of solar and cosmogenic Ne have been observed during stepwise etching and heating of Apollo samples (Wieler et al., 1986; Mortimer et al., 2016) and stepwise heating of lunar meteorites (Eugster et al., 1992, 1996), stepwise crushing of the Pesyanoe aubrite samples (Buikin et al., 2013, 2015) and the lunar meteorite Dhofar 1436 (Korochantseva et al., 2017a), indicating that cosmogenic Ne is more retentive component than solar Ne. Thus, we can conclude that the solar gases are mostly released from relatively large voids destroyed at the beginning, while cosmogenic nuclides are likely extracted from smaller ones at the end of crushing of the Ghubara samples. Several studies on mantle rock crushing show that cosmogenic nuclides located in the

Table 5. Stepwise combustion data for noble gases (He, Ne and Ar), nitrogen and carbon in the Ghubara xenolith (4.045 mg)

T, °C	$^4He^a$	$^{20}Ne^a$	$^{21}Ne/^{22}Ne$	$^{20}Ne/^{22}Ne$	$^{40}Ar^a$	$^{36}Ar^a$	C^b	$\delta^{13}C$	N^b	$\delta^{15}N$
400	7841	45	0.0386(59)	n.a.	870	2.21	755	–19.1(3)	17.3	10.8(1.8)
600	64900	352	0.0328(25)	12.26(12)	1258	2.58	329	–24.1(6)	17.3	10.7(4)
800	12075	828	0.0427(6)	12.52(3)	2274	2.08	1042	–8.1(3)	19.1	5.5(5)
1000	13994	395	0.0459(9)	12.56(5)	15700	0.54	6685	–6.7(4)	18.7	12.9(5)
1100	9327	476	0.0443(8)	12.46(4)	13598	2.67	7780	–6.3(6)	10.6	5.1(4)
1200	8937	830	0.0484(6)	12.22(3)	5356	13.95	4	2.4(5)	8.8	15.6(1.1)
1300	3140	219	0.0561(13)	12.16(6)	1019	4.60	5	–4.2(3)	7.3	17.5(1.4)
1400	504	70	0.0631(24)	11.36(9)	671	2.56	56	–24.8(2)	9.6	12.9(9)
1430	n.a.	6	0.0566(60)	n.a.	10	0.36	154	–18.6(3)	2.5	11.5(8)
1460	n.a.	15	0.0393(36)	n.a.	158	1.04	166	–17.7(1.8)	3.5	8.5(4)
Total	120716	3237	0.0460(9)	12.35(5)	40914	32.59	16976.6	–7.8(5)	114.6	10.6(8)

[a] $\times 10^{-8}$ cm^3 STP/g.
[b] ppm.
The uncertainty in absolute concentrations of noble gases is 5–10%.
The numbers in parentheses refer to the last digits and are 1σ-uncertainties.
n.a.—not analysed.

Table 6. Stepwise combustion data for noble gases (He, Ne and Ar) in the Ghubara impact melt inclusion (3.753 mg; first experiment)

T, °C	$^4He^a$	$^{20}Ne^a$	$^{21}Ne/^{22}Ne$	$^{20}Ne/^{22}Ne$	±	$^{40}Ar^a$	$^{36}Ar^a$
400	241	5	0.0263(140)	n.a.		221	1.08
600	2101	83	0.0389(23)	12.10(10)	0.10	163	0.56
800	10610	223	0.0745(15)	12.03(7)	0.07	325	0.49
1000	15781	214	0.0739(17)	11.54(6)	0.06	398	0.55
1100	11915	162	0.0678(19)	11.81(7)	0.07	687	1.70
1200	6102	245	0.0740(15)	11.94(5)	0.05	985	2.70
1300	733	198	0.0855(19)	11.44(5)	0.05	1023	3.56
1400	185	18	0.0488(47)	n.a.		741	2.78
Total	47667	1149	0.0743(17)	11.78(6)		4543	13.41

[a] $\times 10^{-8}$ cm^3 STP/g.
The uncertainty in absolute concentrations of noble gases is 5–10%.
The numbers in parentheses refer to the last digits and are 1σ-uncertainties.
n.a.—not analysed.

GCR-produced tracks can be extracted by crushing due to damage of the tracks along fractures (e.g., Yokochi et al., 2005; Moreira and Madureira, 2005). So, we believe that release of cosmogenic Ne at the end of the Ghubara samples crushing occurs the same way. It is possible that during stepwise combustion of the Ghubara samples the solar-like Ne is mainly released as a result of decrepitation that is supported by almost simultaneous degassing of Ne and Ar (Fig. 1), while cosmogenic Ne is released due to diffusion at higher temperatures.

The relative contributions of trapped ^{20}Ne in the Ghubara samples analysed by crushing and combustion are >98.9 and >96.9%, respectively. They are calculated using the endmember compositions of SW Ne with $^{20}Ne/^{22}Ne$ = 13.78 and $^{21}Ne/^{22}Ne$ = 0.0329 (Heber et al., 2009) and cosmogenic Ne with $^{20}Ne/^{22}Ne$ = 0.8 (Eugster and Michel, 1995) and $^{21}Ne/^{22}Ne$ = 0.86 (the average value for the $(^{21}Ne/^{22}Ne)_{cos}$ range, see Fig. 3a). $^{21}Ne_{cos}$ in the crushed host and impact melt inclusion samples accounts for 16.9–19.5% of the total amounts of the isotope. In contrast, 48.2–59.0% of the total cosmogenic ^{21}Ne (or 2.5–3.5 times higher than during crushing) is released during stepped combustion of the sample. The xenolith sample shows lower $^{21}Ne/^{22}Ne$ ratios compared to those obtained for the other samples by

Table 7. Stepwise combustion data for noble gases (He, Ne and Ar), nitrogen and carbon in the Ghubara impact melt inclusion (4.478 mg; second experiment)

Temp, °C	$^{4}He^{a}$	$^{20}Ne^{a}$	$^{21}Ne/^{22}Ne$	$^{20}Ne/^{22}Ne$	$^{40}Ar^{a}$	$^{36}Ar^{a}$	C^{b}	$\delta^{13}C$	N^{b}	$\delta^{15}N$
400	n.a.	n.a.	n.a.	n.a.	761	2.9	241	–23.7(3)	4.0	6.0 (5)
600	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	120	–23.9(4)	4.7	14.8(6)
800	8964	153	0.0706(40)	11.02(12)	186	0.15	18	–25.9(5)	1.9	–0.6(2.1)
1000	12881	170	0.0771(17)	11.63(6)	304	0.31	15	–23.4(5)	1.1	7.7(1.2)
1100	10746	146	0.0724(17)	12.06(7)	309	0.36	11	–19.5(6)	0.9	8.3(2.2)
1200	6616	237	0.0685(13)	12.21(6)	569	1.75	12	–23.5(1.4)	1.0	18.3(1.2)
1300	1231	219	0.0882(14)	11.83(5)	302	0.96	15	–24.2(5)	0.7	16.3(2.1)
1400	101	51	0.0820(33)	11.32(9)	138	0.66	33	–25.1(1.5)	1.0	15.8(1.5)
1500	13	15	0.0622(38)	n.a.	n.a.	0.24	84	–25.2(4)	1.5	11.7(6)
Total	40552	989	0.0768(20)	11.75(7)	2569	7.33	548	–24.1(5)	16.9	10.2(1.0)

[a] $\times 10^{-8}$ cm^3 STP/g.
[b] ppm.
The uncertainty in absolute concentrations of noble gases is 5–10%.
The numbers in parentheses refer to the last digits and are 1σ-uncertainties.
n.a.—not analysed.

Table 8. Summary results for the analysed Ghubara samples

Method	Sample	$^{4}He^{a}$	$^{20}Ne^{a}$	$^{40}Ar^{a}$	$^{36}Ar^{a}$	N, ppb	$\delta^{15}N$	C, ppm	$\delta^{13}C$	$^{4}He/^{20}Ne$	$^{20}Ne/^{36}Ar$	$^{4}He/^{36}Ar$
Stepwise crushing	Host	51179	1882	4115	18	678	21.1	b.d.	b.d.	27	102	2772
	Xenolith	32565	4019	8221	43	1616	7.8	b.d.	b.d.	8	93	755
	Impact melt inclusion	39902	2354	5051	15	1222	3.9	b.d.	b.d.	17	152	2580
Stepwise combustion	Host	71296	2642	5045	23	n.a.	n.a.	n.a.	n.a.	27	117	3146
	Xenolith	120716	3237	40914	33	114562	10.6	16977	–7.8	37	99	3704
	Impact met inclusion_1	47667	1149	4543	13	n.a.	n.a.	n.a.	n.a.	41	86	3554
	Impact met inclusion_2	40552	989	2569	7	16927	10.2	548	–24.1	41	135	5536

[a] $\times 10^{-8}$ cm^3 STP/g.
n.a.—not analysed.
b.d.—below detection.

the same methods. In particular, several crushing steps of the sample reveal $^{21}Ne/^{22}Ne < 0.0329$. Suggesting one of the endmembers to be Earth´s atmosphere Ne (instead of solar) with $^{21}Ne/^{22}Ne = 0.0290$ (Eberhardt et al., 1965), the relative contribution of $^{21}Ne_{cos}$ is comparable with other samples: 9.4% (crushing) and 54.5% (combustion).

The total amounts of ^{36}Ar extracted by crushing from voids/gas inclusions of host and impact melt are identical to the amounts of $^{36}Ar_{trapped}$ reported in Korochantseva et al. (2007). The $^{36}Ar/^{38}Ar$ ratios (measured only by crushing) are high (mainly >5). The $^{36}Ar_{cos}$ and $^{38}Ar_{cos}$ contents calculated using $(^{36}Ar/^{38}Ar)_{SW} = 5.47$ (Heber et al., 2009) and $(^{36}Ar/^{38}Ar)_{cos} = 0.65$ (Eugster et al., 1991) consist 0.3–1.4% and 2.5–10.4% of the total amounts of the isotopes in the Ghubara specimens, respectively. The $^{40}Ar/^{36}Ar$ ratio of all crushed samples gradually decreases with increasing number of strokes. Two of three crushed samples, host and xenolith, show identical $^{40}Ar/^{36}Ar$ ratio variations from 234 to 179 and from 231 to 162 with progressive crushing, respectively (Fig. 5). This range of the ratios completely coincides with the compositions of the trapped argon precisely determined using isochron method for Ghubara samples (Korochantseva et al., 2007). In particular, the Ar isotopic compositions in advanced crushing steps are similar to $(^{40}Ar/^{36}Ar)_{trapped} = 174–178$ for the Ghubara xenolith (Korochantseva et al., 2007). Hence, Ar released during crushing (except for the first few steps) seems to be dominated by trapped component of an asteroidal origin. The variations of the $^{40}Ar/^{36}Ar$ ratio in the crushing steps of the Ghubara samples are prob-

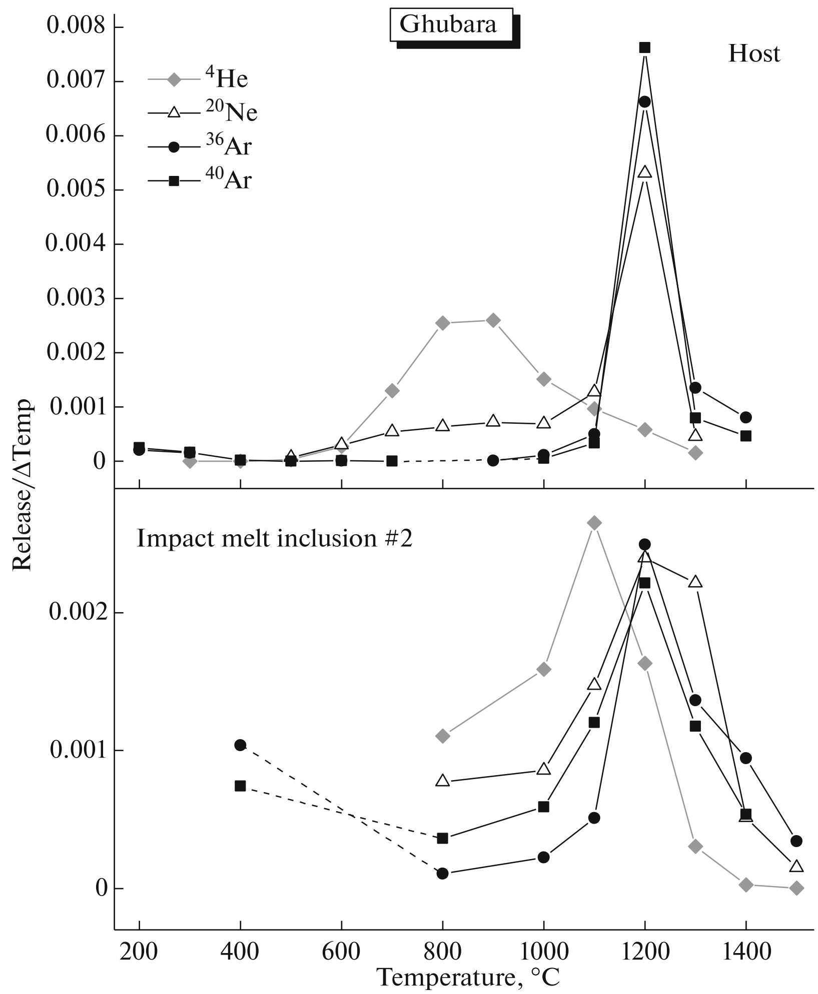

Fig. 1. ^{4}He, ^{20}Ne, ^{36}Ar and ^{40}Ar release patterns of the host material and impact melt inclusion of Ghubara. In both samples simultaneous main release of Ne and Ar are observed at 1200°C. The specimens are different in the degassing behavior of helium: The release of ^{4}He in the impact melt material occurs at a higher temperature than in the chondritic material.

ably related to the presence of more than one trapped Ar components (atmospheric and extraterrestrial). Atmospheric Ar incorporated during terrestrial residence of the meteorite in the Oman desert (e.g., Korochantseva et al., 2005) is likely to be released in the first steps. In comparison with the studied L-chondrite, the ^{40}Ar/^{36}Ar ratios of the crushed lunar meteorite Dhofar 1436 specimens are very stable ranging between 2–3 (except for just the first 1-2 extraction steps contaminated with atmospheric Ar; Korochantseva et al., 2017a). But Dhofar 1436 contains two order of magnitude higher amount of ^{36}Ar (predominantly of extraterrestrial origin) than Ghubara, therefore contamination of the lunar meteorite with atmospheric Ar is weakly pronounced. Ne in the third crushing step of the host sample displays the highest ^{20}Ne/^{22}Ne ratio of 13.31 and is associated with Ar having the ^{36}Ar/^{38}Ar ratio of 5.47 equal to the solar one (Heber et al., 2009). Its ^{40}Ar/^{36}Ar of 222 could be con-

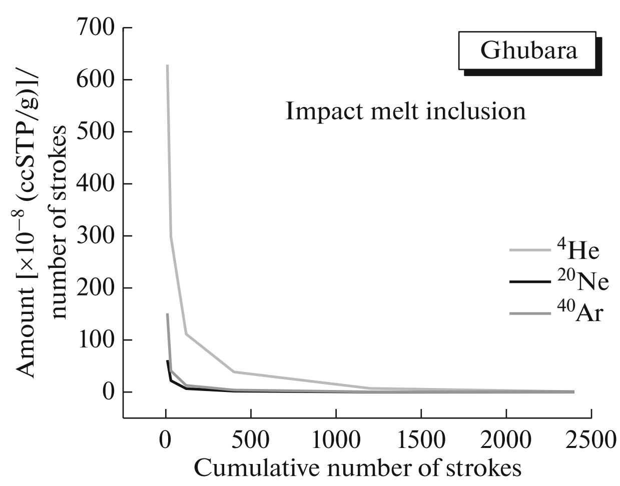

Fig. 2. The rate of noble gases release during Ghubara crushing.

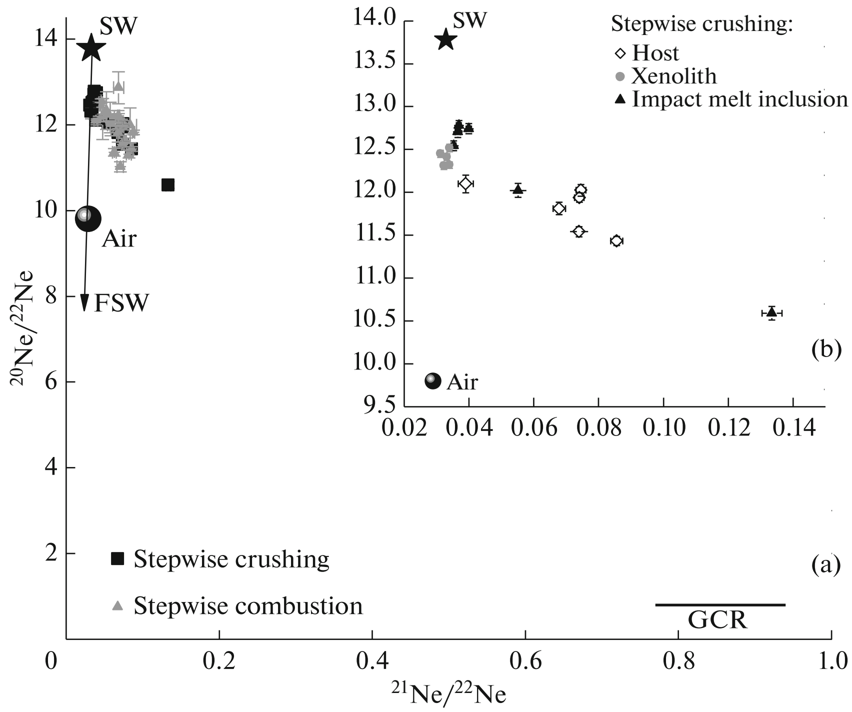

Fig. 3. Neon isotope variations during crushing and combustion analyses of the Ghubara samples. The solar wind (SW; Heber et al., 2009) and Earth's atmosphere (EA; Eberhardt et al., 1965) compositions are shown. The range for $(^{20}Ne/^{22}Ne)_c$ produced by galactic cosmic rays (GCR) is taken from Eugster and Michel (1995), and for $(^{21}Ne/^{22}Ne)_c$—according to Eugster (1988), which also includes the range evaluated using Leya and Masarik (2009) for L-chondrites of 0–85 cm radius—the size of the Ghubara meteoroid determined by Ferko et al. (2002). The depth-dependent implantation-fractionated solar wind (FSW) is plotted using Grimberg et al. (2006). (a) The Ne isotopic compositions of all samples analysed. (b) The results obtained by stepwise crushing. The four extraction steps (from 350 to 4500 strokes) of the Ghubara xenolith with unexplained nearly atmospheric Ne composition are omitted. The Ne isotopic compositions of the Ghubara samples can be explained as a mixture of solar and cosmogenic components. The advanced crushing steps display the lowest $^{20}Ne/^{22}Ne$ ratios and the highest contribution of the GCR-Ne component.

sidered as an upper limit of the $^{40}Ar/^{36}Ar$ ratio value for the trapped component in this Ghubara sample.

Noble Gas Elemental Ratios

The amounts of ^{36}Ar obtained by total extraction are (7–43) × 10^{-8} cm^3 STP/g and are comparable with those reported in literature (7–79) × 10^{-8} cm^3 STP/g (Schultz and Franke, 2004). However, our data for ^4He (326–1207) and ^{20}Ne (10–40) (Table 8) show higher concentrations than published values: ^4He (1–201) and ^{20}Ne (0.08–11) (Schultz and Franke, 2004), in units ×10^{-6} cm^3 STP/g. The $^4He/^{36}Ar$ and $^4He/^{20}Ne$ ratios are as usual strongly fractionated relative to the SW composition (Heber et al., 2009) showing deficit of He, but in much lesser extent than the previously measured (Schultz and Franke, 2004). They increase in the host and impact melt lithology by a factor of 2–6 in the advanced crushing steps. The analogous increase in the $^4He/^{20}Ne$ and $^4He/^{36}Ar$ ratios with progressive crushing is also reported for the lunar meteorite Dhofar 1436 (Korochantseva et al., 2017a) and seems to be controlled by very similar processes. A significant increase of the $^4He/^{20}Ne$ and $^4He/^{36}Ar$ ratios cannot be interpreted solely by admixing of the radiogenic or cosmogenic ^4He, which can be released during mechanical breakdown of the tacks containing in situ U,Th-derived ^4He (Scarsi, 2000; Matsumoto et al., 2002; Buikin et al., 2018) or of GCR-produced tracks (Yokochi et al., 2005; Moreira and Madureira, 2005). The production of radiogenic ^4He of 1.3 × 10^{-6} cm^3 STP/g from the in situ decay of U and Th with mean concentrations in L-chondrites of 13 and 43 ppb, respectively (Wasson and Kallemeyn, 1988), for 470 Ma and the contribution of cosmogenic noble gases to their total budget are too low. The increase of these ratios obviously reflects a truly different elemental ratio of the trapped gases between crush-accessible sites (inclusions and voids) varying in sizes. The $^{20}Ne/^{36}Ar$ ratios of the analyzed samples are very unusual in meteorites: the light noble gas is enriched relative to the heavy one compared to the SW composition. The $^{20}Ne/^{36}Ar$ ratios in the individual steps of the crushed material and the totals obtained by both techniques show rather similar values (Tables 1–8). The individual combustion steps

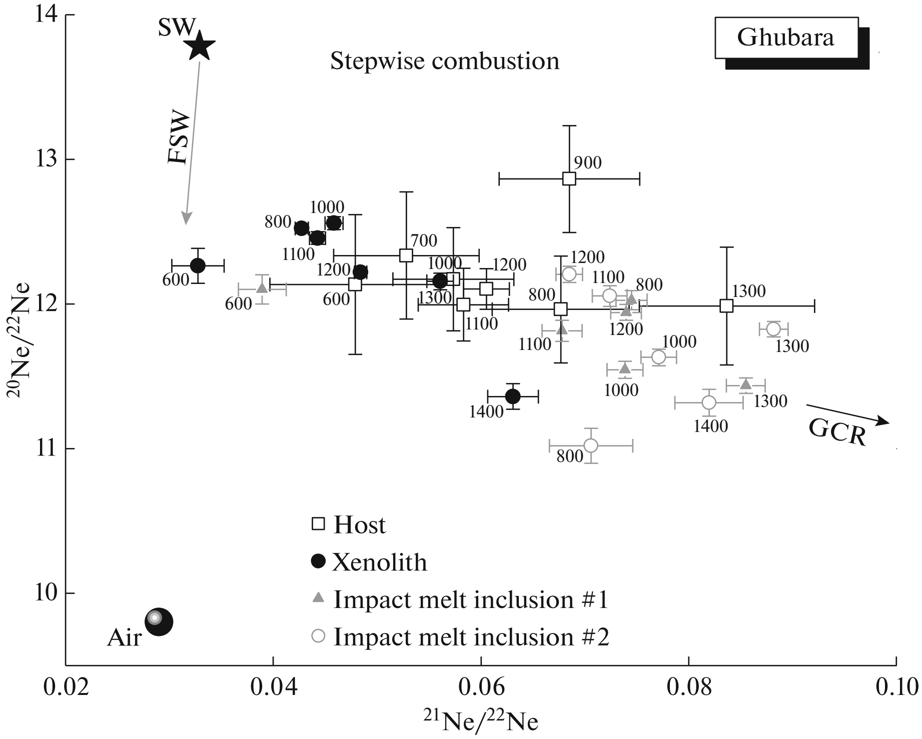

Fig. 4. Neon isotopic compositions for the stepwise combustion analyses of the Ghubara samples. The solar wind (SW; Heber et al., 2009) and Earth's atmosphere (EA; Eberhardt et al., 1965) compositions are shown. The depth-dependent implantation-fractionated solar wind (FSW) and galactic cosmic rays (GCR) compositions are also plotted (see figure caption of Fig. 3). Step temperatures in (°C) are shown next to each data point. At high temperature steps the contribution of cosmogenic component increases.

can be affected by diffusional fractionation upon stepwise heating extractions. But in the host sample Ne and Ar are released during stepped combustion almost simultaneously (Fig. 1), and the $^{20}Ne/^{36}Ar$ ratio at 1200°C remains identical to the totals. Along with that, the lower than solar $^{20}Ne/^{36}Ar$ ratios have been reported in all the Ghubara analyses before (Schultz and Franke, 2004). We however do not see any reasons to think that the measured high $^{20}Ne/^{36}Ar$ ratios observed in the present study are related to an analytical artifact. The variations of the noble gas elemental compositions and contents are rather related to the heterogeneity of the Ghubara breccia. Although the deficit of light noble gases is common for the stone material of the solar system, excess of light noble gases (particularly helium) is also observed as for instance in the terrestrial basalt glasses, due to their higher solubility during vesicle-melt partitioning (Jambon et al., 1986; Marty and Zimmermann, 1999; Buikin et al., 2017) and rarely in meteorites where the high $^{20}Ne/^{36}Ar$ ratios are not related to the high abundance of cosmogenic ^{20}Ne (Manuel and Kuroda, 1964; Gopalan and Rao, 1976; Pun et al., 1998). The bulk cosmogenic $^{21}Ne/^{38}Ar = 4–8$ (see section "Noble gas isotopic compositions" for details of calculations) in the gases released by crushing do not show a significant fractionation compared to their production rate ratios (7–9) evaluated using Leya and Masarik (2009) data for the L-chondrite composition and a meteoroid radius of 85 cm determined for Ghubara by Ferko et al. (2002). The "normal" $(^{21}Ne/^{38}Ar)_{cos}$ ratios and $^{21}Ne_{cos}$ concentrations identical to the taken from literature (see below) give an additional evidences for the correctness for the unusually high $(^{20}Ne/^{36}Ar)_{trapped}$ ratios.

The Origin of Trapped Noble Gases

Genesis of the trapped argon with isotopic composition different from primordial/solar/terrestrial in the samples of asteroidal origin is considered to be linked with the mobilization of solar, cosmogenic or radiogenic argon components during thermal processing leading to their redistribution into voids (Korochantseva et al., 2017b). Similar scenario for the origin of trapped noble gases has been suggested by Takaoka et al. (1996) based on the crushing experiments with enstatite chondrites. Inter alia, the presence of trapped gases in Ghubara is obviously related to the major impact event that resulted in the last total reset of K–Ar system 470 Ma ago for most L-chondrites (Korochantseva et al., 2007; Weirich et al., 2012; Yin et al., 2014) and induced extensive degassing of mate-

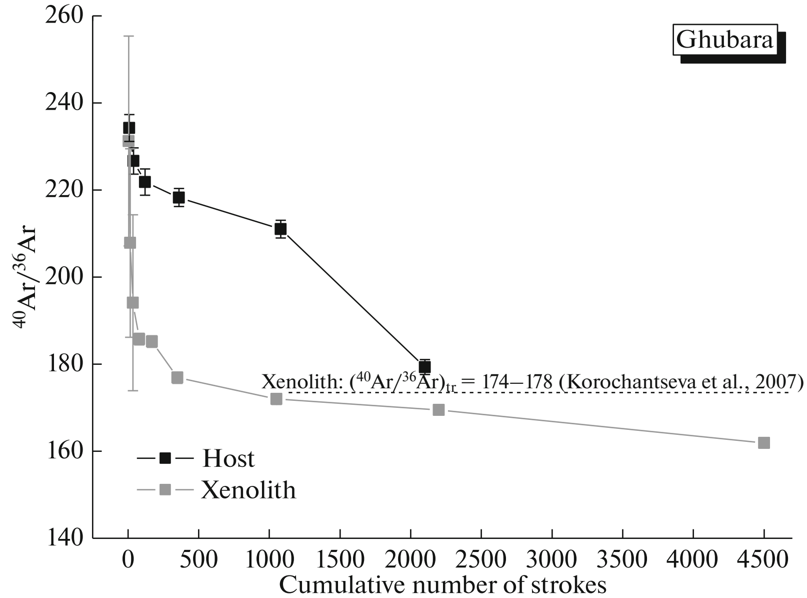

Fig. 5. The $^{40}Ar/^{36}Ar$ ratio variations in crushing steps for the Ghubara host and xenolith. The Ar isotopic compositions in the advanced crushing steps are similar to those precisely determined, using isochron method for the Ghubara xenolith (Korochantseva et al., 2007).

rial on the parent asteroid. The Ghubara breccia appears to have been formed at this event that lead to breakdown, fracturing, brecciation, melting and darkening of chondritic material buried under the hot ejecta blanket.

Two mechanisms of gas redistribution during thermal process seem to be possible: i) diffusional redistribution of accumulated gases within the rock, ii) trapping of gases released from surrounding rocks (e.g., gases released from deeper hot rocks are trapped by shallow cooling rocks into voids/fractures that have been immediately isolated). The latter mechanism could explain the enhanced Ne/Ar ratios and the higher light noble gas contents in the analysed Ghubara samples compared to those published earlier, if the neighboring rocks lost the light noble gases preferentially over the heavy ones. Helium could be subsequently lost during solar heating and/or late mild impacts resulted in reduction of the $^{4}He/^{20}Ne$ and $^{4}He/^{36}Ar$ ratios to the values below solar. Although He could escape in larger extent during the process of gas redistribution at 470 Ma event.

Cosmic Ray Exposure (CRE) Ages

The total concentrations of $^{21}Ne_{cos}$ in the combusted Ghubara samples are 3.3–6.4 ($\times 10^{-8}$ cm^3 STP/g; calculated using the solar and cosmogenic endmember compositions (see section "Noble gas isotopic compositions"). They are consistent with the $^{21}Ne_{cos}$ concentrations of 3.20–5.36 ($\times 10^{-8}$ cm^3 STP/g) measured by Ferko et al. (2002) who concluded that Ghubara likely experienced a simple exposure in an 85 cm meteoroid. The production rates evaluated using the model by Leya and Masarik (2009) for the L-chondrite composition and this meteoroid radius vary from 0.230 to 0.364. The CRE ages calculated using our $^{21}Ne_{cos}$ concentrations are in the range of 9–28 Ma and usual for L chondrites (Wieler et al., 2002). They are comparable with the average $^{10}Be/^{21}Ne$ and $^{26}Al/^{21}Ne$ exposure ages of 15–20 Ma reported by Ferko et al. (2002). A simple exposure during transition from the parent asteroid to the Earth is supported by the unfractionated (close to the production) $(^{21}Ne/^{38}Ar)_{cos}$ ratios observed in the crushing experiments.

Carbon and Nitrogen

The N and C abundances released by combustion are significantly higher in the chondritic material (xenolith) than in the impact melt inclusion (Table 8, Figs. 6, 7). The main release peak of carbon in the xenolith is observed at 800–1100°C (Fig. 7) and possibly linked with oxidation of graphite. The bulk $\delta^{13}C$ of –7.6 as well as the high N and C amounts of the xenolith (Table 8) are out of typical ranges for ordinary chondrites (Grady and Wright, 2002; Pillinger et al., 2013). The carbon isotopic composition of the impact melt inclusion is likely related to contamination by terrestrial organics ($\delta^{13}C$ of –25.9 to –19.5‰). The nitrogen compositions of the combusted samples are

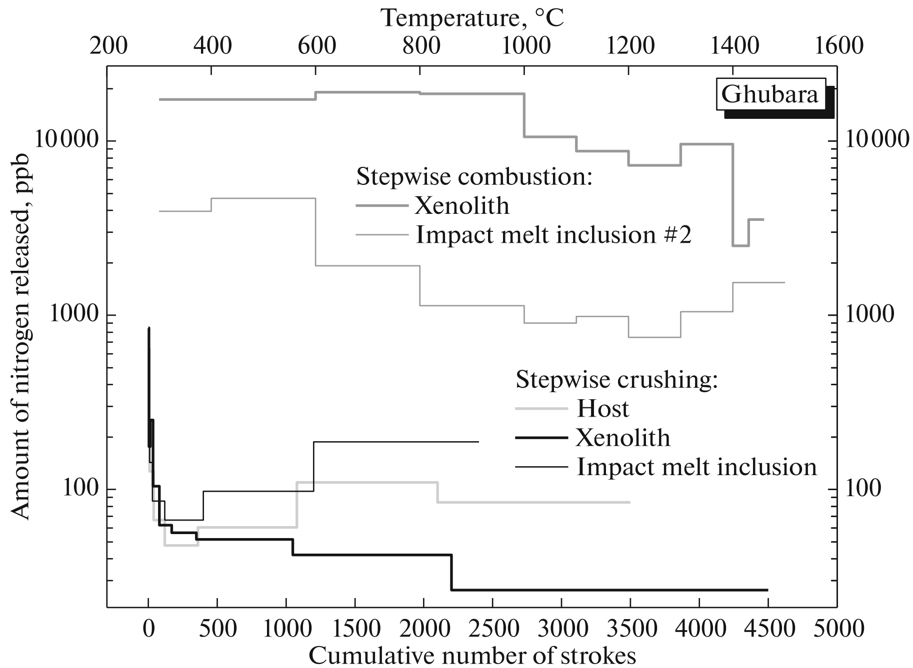

Fig. 6. Nitrogen abundance variations during crushing (bottom) and combustion (top) of the Ghubara samples. Combustion releases much more nitrogen than crushing and shows that the impact melt inclusion is depleted by nitrogen compared to the chondritic material (xenolith).

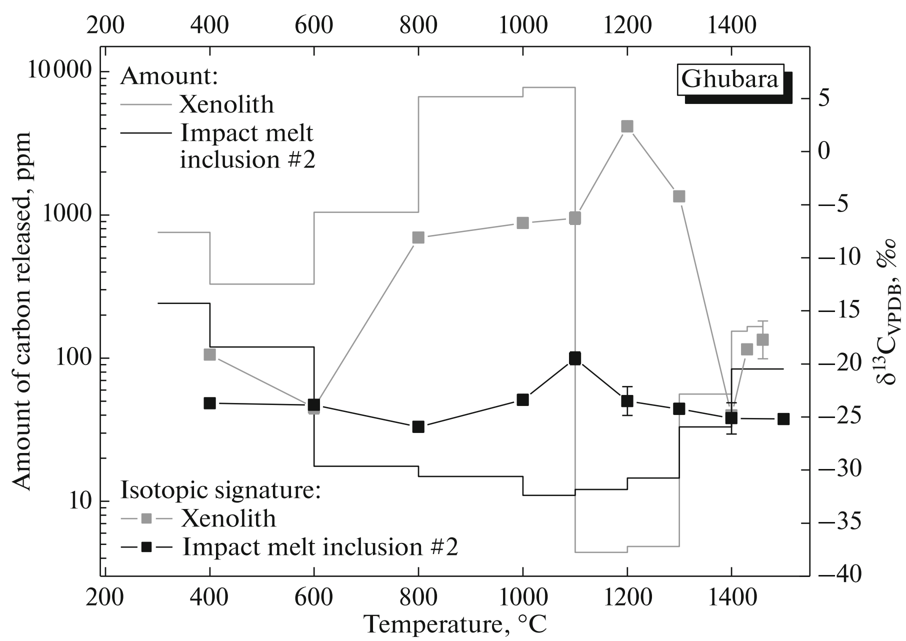

Fig. 7. Release patterns and isotope profiles of carbon analysed in the Ghubara xenolith and impact melt inclusion (experiment #2) by stepwise combustion.

similar with $\delta^{15}N$ ranging between –0.6 and 18.3‰ (Tables 5, 7; Fig. 8). The amounts of nitrogen released by crushing are very low relative to those released by combustion (Table 8). In particular, the amounts of nitrogen of the crushed xenolith and impact melt material accounts for only 1 and 7% of the amounts extracted by combustion of the respective samples, which assumes that most of nitrogen is chemically

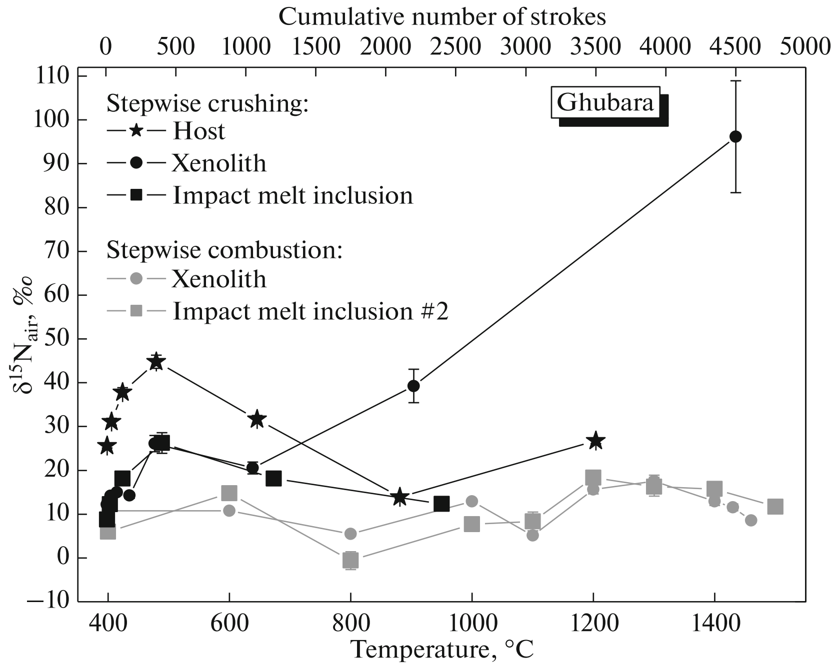

Fig. 8. Nitrogen isotope variations in Ghubara observed during stepped combustion and crushing. $\delta^{15}N$ profiles for the former are identical while the latter show isotopically heavier nitrogen.

bounded and associated with carbon. $\delta^{15}N$ of the crushed xenolith gradually increases from 4.0 to +94.9‰ (Fig. 8) that is perhaps related to a contribution of cosmogenic component in the final steps observed also in the Ne composition. The crushed host sample shows the heaviest bulk nitrogen composition. The nitrogen compositions of all analyses are usual for ordinary chondrites (Grady and Wright, 2003).

CONCLUSION

The trapped noble gases of extraterrestrial composition released by stepped crushing or heating from the Ghubara meteorite are related to voids of highly retentive phases, the products of shock metamorphism, unaffected by the subsequent less intensive degassing events. Their genesis is obviously linked to the major impact event that resulted in the last total reset of K-Ar system 470 Ma ago for most L-chondrites. The trapped gases are considered to be derived from mobilization and redistribution of different noble gas components (solar, cosmogenic, radiogenic) accumulated in the regolith material before the catastrophic event. The composition of trapped gases is dominated by the SW component, which was fractionated upon degassing from the parent regolith, and by mobilized radiogenic ^{40}Ar. The gases have been captured into voids in the formed breccia.

The abundance and isotopic composition of neon released by crushing can be explained by a mixture of trapped and cosmogenic gases accumulated during subsequent GCR irradiation. A gradual opening of the inclusions/voids of different sizes indicates that the relative contribution of cosmogenic Ne increases with progressive crushing. While trapped Ne is mostly released from relatively large voids, cosmogenic Ne is likely extracted from the GCR-produced tracks.

The noble gas elemental ratios of trapped gases are strongly fractionated relative to the SW composition. While the $^{4}He/^{20}Ne$ and $^{4}He/^{36}Ar$ ratios as usual show a deficit of helium, the $^{20}Ne/^{36}Ar$ ratios of the analyzed samples are very uncommon demonstrating an excess of ^{20}Ne relative to ^{36}Ar compared to the $(^{20}Ne/^{36}Ar)_{SW}$ ratio. The enhanced Ne/Ar ratios can be interpreted by trapping of gases released from surrounding rocks that lost the light noble gases preferentially over the heavy ones. Helium could be largely lost during the process of gas redistribution or the later events that resulted in the $^{4}He/^{20}Ne$ and $^{4}He/^{36}Ar$ ratios with usual helium shortage relative to the SW composition. The increase of these ratios with progressive crushing is apparently associated with distinct elemental ratio of the trapped gases present in the inclusions/voids of different sizes.

In the Ghubara meteorite most of nitrogen is chemically bounded and associated with carbon. The nitrogen and carbon abundances as well as their isotopic compositions in the host and impact melt inclusion are in the ranges for ordinary chondrites. The bulk $\delta^{13}C$, N and C amounts of the xenolith are atypical for ordinary chondrites. The melt inclusion is likely contaminated by terrestrial organics.

The cosmic ray exposure ages calculated using the total amounts of $^{21}Ne_{cos}$ for combusted samples are in the range of 9–28 Ma and likely correspond to the transition time of Ghubara from the parent asteroid to the Earth.

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