

Separation of planetary noble gas carrier from bulk carbon in enstatite chondrites during stepped combustion

A.B. Verchovsky*, M.A. Sephton, I.P. Wright, C.T. Pillinger

Planetary and Space Sciences Research Institute, Open University, Walton Hall, Milton Keynes MK7 6AA, UK

Received 17 October 2001; received in revised form 6 March 2002

Abstract

The exact location of planetary noble gases in meteorites remains unknown but it is inferred to be closely associated with, if not precisely some portion of, the macromolecular organic material in carbonaceous chondrites [1]. Herein we show that for enstatite chondrites the major carbonaceous component is not the carrier of the gases. This may also be true for other chondritic groups. Rather, these gases are all contained in a minor combustible constituent. This situation has all the hallmarks of a Russian matryoshka doll problem, as was witnessed previously in the study of meteorites prior to the understanding of the presence of presolar grains. A possible conclusion, which is in line with previous suggestions [2,3], is that planetary noble gases are also presolar and located in a new, and as yet unidentified, form of presolar material. © 2002 Elsevier Science B.V. All rights reserved.

Keywords: meteorites; carbon; nitrogen; noble gases; isotopes; combustion

1. Introduction

The noble gas component known as ‘planetary’ (PNG, for planetary noble gases), which is also referred to as ‘Q’ [1] or P1 [4], is usually one of the most abundant primordial noble gas constituents found in primitive meteorites. The origin of PNG is still not completely understood [5–7], but it is generally accepted that they were acquired during the birth of the Solar System (e.g. [8–10]) or possibly earlier [3]. The PNG are present in variable amounts in almost all types of chondritic

meteorites [11–14] and some ureilites [15–17], and are closely associated with meteoritic carbon [1,18–20]. Isotopically, the PNG are similar to, but nonetheless characteristically different from, those in the Sun [9], which we will refer to as SNG (for solar noble gases). Compared to the SNG, the PNG are strongly and systematically depleted in the light elements [7]. The most obvious isotopic differences between PNG and SNG are observed for Ne and Xe. The differences suggest that PNG could have formed from SNG as a result of isotope and element fractionation [9]. At present, the most generally accepted incorporation mechanism is the sorption of PNG onto the surfaces of carbon grains at low temperatures [21,22]. It is thought that this material was then sampled by the accreting meteorite parent bodies. It is not possible to exclude, however, the possi-

* Corresponding author. Tel.: +44-1908-65-50-08;
Fax: +44-1908-65-59-10.

E-mail address: a.verchovsky@open.ac.uk
(A.B. Verchovsky).

bility that trapping of PNG into their carrier took place outside of, and before formation of, the Solar System [2,3]. In this scenario, presolar carbon-containing PNG was inherited by the early Solar System and then incorporated into the parent bodies of the primitive meteorites. The fact that noble gas components with isotopic compositions close to those in the Sun appear to be common galactic components (e.g. P3 [23] or N-noble gases [24] in presolar diamonds and SiC respectively), and the observation that the most abundant carbon phase in meteorites, the macromolecular material, is present in meteorites in rather constant ratios with respect to presolar grains [25], support this proposal.

The most important features of PNG are: (i) they are concentrated in acid (HF-HCl) resistant residues, which account for only a small fraction of the total mass of meteorites and consist mostly of carbon phases and oxide minerals [1]; (ii) the carrier of the noble gases is closely associated with meteoritic carbon and is combustible in an atmosphere of oxygen [1,18–20,26]; (iii) the gases are effectively removed from their carrier(s) by treatment with oxidizing acids and reagents (HNO₃, H₂O₂, HClO₄, etc. [1,27]). The first two features point to a carbon phase as a carrier for PNG, while the interpretation of the third characteristic is not straightforward. Although oxidizing chemicals remove most of PNG, they destroy only a few percent of the total carbon.

Attempts to identify the carrier of the PNG (Q-phase, according to [1]) in meteorites have so far been unsuccessful. Perhaps the most satisfactory explanation [28], to date, is that there is no specific PNG carrier; rather the close association of PNG with carbon is the result of the surface adsorption of noble gases onto any carbonaceous material having a sufficiently high surface area. The interpretation is consistent with the chemical extraction procedures, as oxidizing acids would destroy the surfaces of carbon grains resulting in the release of surface-sited noble gases.

An important analytical observation that places some constraints on the nature of the PNG carrier, is that during stepped heating (i.e. vacuum pyrolysis) the noble gases are released at temperatures of 1200–1600°C, whilst under stepped gas-

phase oxidation the gases are evolved at 400–550°C. To explain the high release temperature of PNG during pyrolysis, Zadnik et al. [28] developed the ‘labyrinth’ hypothesis where it was proposed that during initial trapping, noble gases found their way through a complex network of micropores on the surface of the carbon grains. Following adsorption, the labyrinth channels were plugged with organic molecules. This would have had the effect of preventing the release of PNG until the carrier was subjected to elevated temperatures (either on the parent body, or during laboratory heating experiments). While the PNG were trapped within the labyrinth, isotopic exchange with gases from other reservoirs, such as the terrestrial atmosphere, was also precluded. This explanation, based on a number of rather sophisticated experiments [22,28,29], and relying upon the observations mentioned above, also suggests that oxidation in an atmosphere of pure oxygen will release PNG in the same way as the oxidizing acids. However, when the PNG adsorption and labyrinth mechanisms were originally formulated, no direct measurements of the simultaneous release of CO₂ and PNG during stepped combustion were available.

The combustion experiments which had been done at that time included bulk sample (effectively one-step) combustion [20] and stepped combustion experiments in which noble gases and CO₂ were released from separate aliquots of the same sample in different experiments [19]. The first experiment where both yields of carbon and noble gases were determined simultaneously was performed later by Schelhaas et al. [13] in which the amount of CO₂ was measured with low precision using a Pirani gauge and the stepped combustion experiment was carried out with rather poor temperature resolution. Despite that, data from an HF-HCl residue of Dhajala (H3.8) showed that most of PNG were released before the main carbon release, which seemed to be in keeping with the labyrinth theory (though complete release of the noble gases required destruction of about 50% of the carbon which seems to be too high for the surface-sited noble gases).

In the present study, we undertook a detailed examination of the relationship between PNG and

carbon in meteorites of different types through the use of high resolution stepped combustion experiments. A primary purpose of the investigation was to establish conclusively whether or not oxidation with pure O₂ gas would confirm the surface siting of PNG on carbonaceous components. To this end, seven meteorites (Murchison, Orgueil, Renazzo, Dhajala, LEW87223, Indarch and Yilmia) were analysed. By choosing chondrites of different chemical groups and petrological types it was considered that some of the subtleties of parent body processing (e.g. metamorphism and aqueous alteration) and the resulting structural transformations of the putative carbon carrier materials could also be studied.

2. Experimental

HF-HCl resistant residues from each meteorite were measured by precise, static-mode mass spectrometry to determine simultaneously the concentrations and isotopic compositions of noble gases, nitrogen, and carbon (in the form of CO₂). The residue from Dhajala was prepared as in [13] and provided for the study by U. Ott. The preparation of the residues from Orgueil, Murchison, Renazzo and Indarch are described in [30] and the residues from LEW87223 and Yilmia were prepared in a similar way. The preparation of the Murchison residue is described in [31] (K procedure until post HF-HCl step). SiC was removed with sodium polytungstate density separation and any free organic compounds were removed by solvent extrac-

tion (93:7 DCM/MeOH). This residue was also subjected to aqueous alteration at 330°C using the standard hydrous pyrolysis technique for meteorite samples [32]. The gases of interest were liberated by stepped combustion. Isotopic compositions of noble gases and nitrogen and amounts of carbon were measured on the Finesse instrument [33,34]. Where sufficient gas was present, isotopic compositions could be determined to a precision of ±3–5% for argon and xenon and ±1–3‰ for nitrogen. Measured ³⁸Ar/³⁶Ar ratios were close to 0.19 for all the samples and the ⁴⁰Ar/³⁶Ar ratio varied from 2 to 10 indicating a mixture of ³⁶Ar from the samples with a small amount of atmospheric Ar from the blank. Isotopic compositions of carbon were measured in all samples using stepped combustion on another static-mode mass spectrometer (MS 86 [35]); since carbon yields were also measured on this system it was possible to compare directly the release profiles from MS 86 and Finesse. Both Finesse and MS 86 are completely automated and provide a highly reproducible protocol for high resolution multi-step analysis. The typical sample weight used for noble gas and nitrogen analyses on Finesse was about 1–3 mg, while 0.2–0.5 mg was used for carbon isotope analyses on MS 86.

3. Results

A summary of the results obtained from the stepped combustion of the HF-HCl residues from different types of meteorites analysed herein

Table 1

Summary of results from stepped combustion experiments of HF-HCl residues of the meteorites analysed herein (nm, not measured)

Sample	Type	C (%)	δ ¹³ C ^a (‰)	N/C (g/g)	δ ¹⁵ N (‰)	³⁶ Ar/C (cm ³ /g)	¹³² Xe/C (cm ³ /g × 10 ⁻⁷)
Orgueil	CI	9.1	-18.5	0.020	20.6	0.000109	15
Murchison	CM2	9.5	-22	0.029	-25.4	0.000120	12
Renazzo	CR2	40	-19 ^b	0.037	157	0.000073	nm
Dhajala	H3.8	4.5	nm	0.011	4.5	0.000460	82
LEW87223	E3	20	-7	0.005	7.3	0.000031	3.9
Indarch	EH4	32	-10.6	0.004	-7.5	0.000043	3.3
Yilmia	EL6	18	-3.7	0.006	-26.5	0.000032	0.27

^a Obtained on MS 86.

^b Data from Alexander et al. [25].

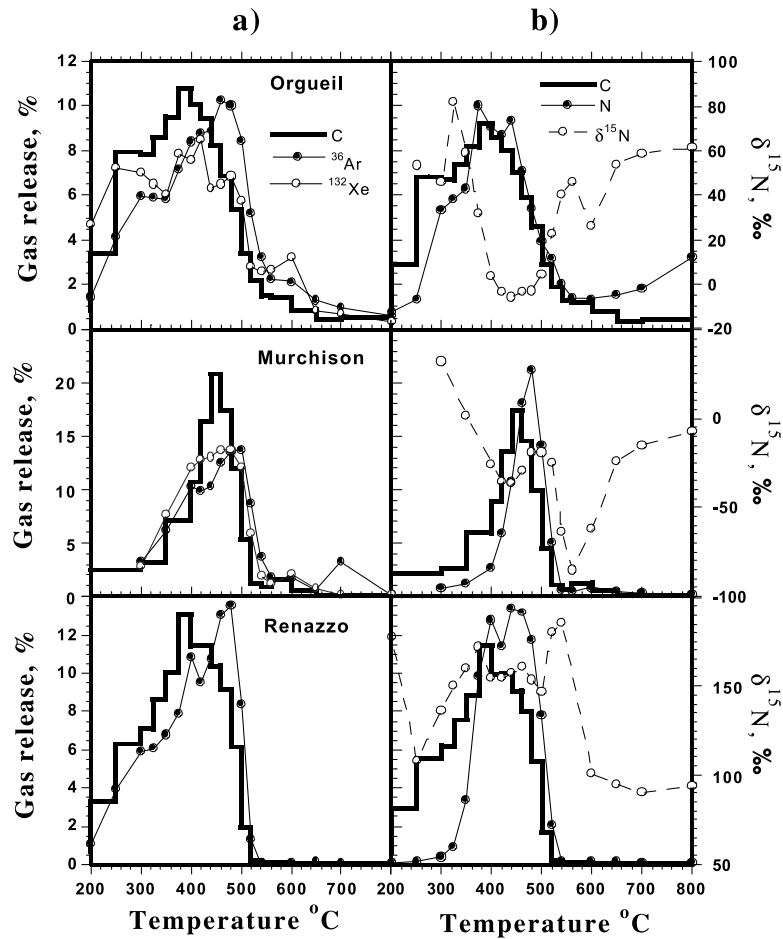


Fig. 1. Release of carbon, ³⁶Ar and ¹³²Xe (a) and carbon, nitrogen and nitrogen isotope profile (b) from HF-HCl residues of three carbonaceous chondrites: CI (Orgueil), CM2 (Murchison) and CR2 (Renazzo). The temperature range of noble gases and nitrogen release are similar to each other and to that for most of carbon, suggesting noble gases to be rather volume- than surface-correlated components. The maximum of noble gas release is observed at about 500°C.

is given in Table 1 (for more details see Appendix). Three of these meteorites (Orgueil, Murchison and Renazzo) are carbonaceous chondrites that contain primitive macromolecular material and have been subjected to aqueous alteration. The CI chondrite Orgueil is the most extensively altered, Murchison (CM2) less so, whilst Renazzo (CR2) is considered to contain the least aqueously altered carbon [25]. However, according to the abundances of P3 noble gases in diamonds, Renazzo was subject to moderate thermal metamorphism, up to 350°C [36]. The enstatite meteorites (LEW87223, Indarch and Yilmia) have experienced thermal, rather than aqueous, processing

at elevated temperatures in a chemically reducing environment. The carbonaceous material in the ordinary chondrite Dhajala has experienced thermal metamorphism, but with a certain degree of oxidation [37].

Xe isotopic compositions in all the temperature fractions of every sample correspond to P1 within the error limits. We could not observe Xe with increased ¹³⁶Xe/¹³²Xe ratios releasing from presolar diamonds of carbonaceous chondrites because of the poor precision at the low amounts of Xe detected, but we did observe a clear signal from the diamonds in the form of a ⁴He release, which peaks at higher temperature than those for all

other gases (Appendix). Such a release pattern of noble gases during stepped combustion was first observed in [19] before the ^4He carrier was identified as presolar diamonds.

Fig. 1 shows the release patterns of carbon, nitrogen and noble gases obtained by stepped combustion from the three carbonaceous chondrites. As can be seen, carbon and noble gases are released in the same temperature range, so that the complete release of noble gases occurs only after most of the carbon has been oxidized. More precisely, at the point where 80% of C has been combusted only about 60% of planetary Ar and Xe have been released. This release pattern is compatible with PNG being a volume-, rather than surface-correlated component. The similarity between the release of PNG and nitrogen (Fig. 1b), which appears to be a structural constituent of organic macromolecular material in these meteorites [38], supports this conclusion. Thus, the results obtained for carbonaceous chondrites indicate that oxidation of the PNG carrier with pure O_2 gas affects the release of the gases in a way that suggests the PNG are located not on the surface of any carbon grains, but are distributed more or less uniformly within the grains, or associated only with some of them (Q-phase), which have combustion temperatures indistinguishable from the bulk carbon.

A quite different relationship between the release of PNG on the one hand and carbon and nitrogen on the other, is observed for enstatite chondrites (Fig. 2). In these meteorites, the releases of most of the PNG and carbon are significantly separated from each other during stepped combustion with the resolution depending on the metamorphic class of the meteorites. In all of the samples (carbonaceous and enstatite chondrites) the maximum PNG release occurs at the same temperature (about 500°C) while for the enstatite chondrites the release of carbon systematically shifts to higher temperatures (Fig. 2a) with increasing thermal metamorphism from E3 (LEW87223) to EL6 (Yilmia). Compared to the carbonaceous chondrites discussed above, the profiles of the PNG release in enstatite chondrites occur over a narrower temperature range (Figs. 1a and 2a). The simplest interpretation of the re-

sults is that there is a minor, and presumably carbonaceous, PNG carrier, which is present in all meteorites, and which is effectively dissolved in oxidizing acids (e.g. HNO_3) and oxidized with pure O_2 gas. It would appear that this component, which we now feel confident to rechristen 'Q' (i.e. to denote its distinction as a truly separate material) behaves differently during parent body processing from the bulk of chondritic macromolecular carbon. In those meteorites where this processing has been relatively extensive (i.e. high petrological types, enstatite chondrites, etc.) a combination of effects has acted to alter macromolecular material, but not Q. In contrast, where minimal processing has taken place (e.g. low petrologic types, carbonaceous chondrites, etc.) stepped combustion is not effective at separation because the oxidation temperatures of macromolecular material and Q are similar which, if care is not exercised, gives the illusion that they are one and the same.

Stepped combustion results for the ordinary chondrite Dhajala were generally similar to those for the enstatite chondrites; most of the noble gases in it are associated with a small amount of carbon. However, in contrast to the enstatite chondrites, the release pattern of carbon from Dhajala is more complicated and consists of at least three peaks. It appears that, in this relatively highly metamorphosed meteorite, only a part of the total carbon (released above 500°C) was graphitized. Most of the carbon in Dhajala appears to have been lost during metamorphism under oxidizing conditions resulting in a very low carbon content of 0.07 wt% C. This result represents an upper limit of carbon content in this meteorite as, at this level, laboratory contamination may be significant. In [13] the estimated carbon concentration in Dhajala is 0.015 wt%. Thus, both results confirm that the Dhajala meteorite has an anomalously low carbon content. For noble gases, we analysed the same sample of Dhajala HF-HCl residue (R10) that was obtained in [13], and our results are in a good agreement with the previous data. The PNG are released during stepped combustion in the same temperature range as observed for other meteorites, although in contrast to the carbonaceous chondrites, the

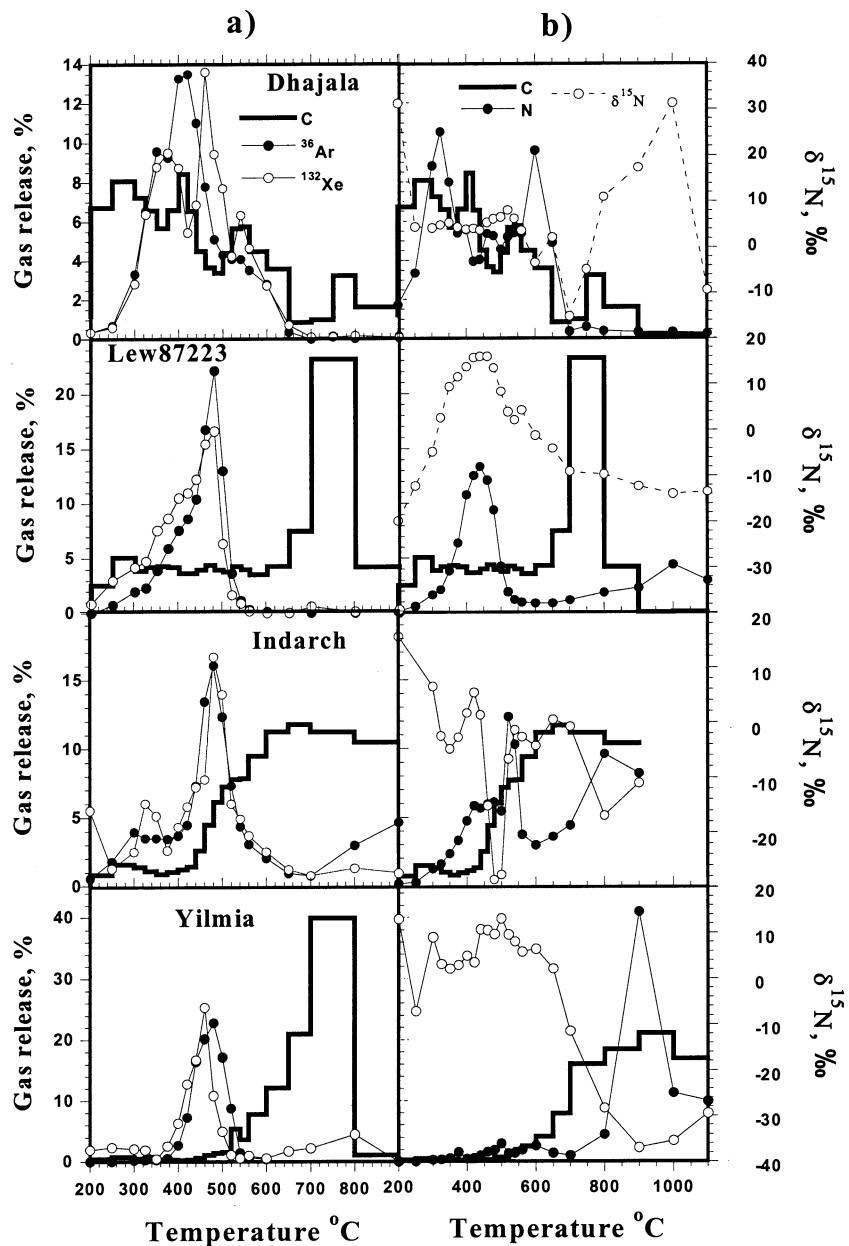


Fig. 2. Release of carbon, ^{36}Ar and ^{132}Xe (a) and carbon, nitrogen and nitrogen isotope profile (b) from HF-HCl residues of ordinary chondrite H3.8 (Dhajala) and three enstatite chondrites: E3 (LEW87223), EH4 (Indarch) and EL6 (Yilmia). Release of most of the carbon in all these samples is separated from that for noble gases. Enstatite chondrites show a clear trend of degree of separation of noble gases from carbon with increasing petrological type of the meteorites. Release temperature of the noble gases in these samples is similar to that observed for carbonaceous chondrites. Note that noble gases and nitrogen in Yilmia were measured in two different experiments.

$^{36}\text{Ar}/\text{C}$ ratio in Dhajala is higher by a factor of 4 (Table 1), indicating that the PNG carrier is better able to survive oxidative degradation than the bulk organic matter. It is important to note that oxidation of carbon under the conditions prevailing in meteorite parent bodies does not involve pure oxygen, but occurs as a result of chemical reactions, which might, for instance, involve minerals such as magnetite [30].

Nitrogen concentrations in the enstatite chondrites and Dhajala are lower than in carbonaceous chondrites, and there is no obvious association of N release with either carbon or noble gases. Compared to carbonaceous chondrites the isotopic composition of the N is relatively ^{15}N -depleted and there is a clear trend in enstatite chondrites for N to become isotopically lighter with increasing levels of metamorphism. It is important to note here that the PNG carrier is not associated with significant amounts of nitrogen as seemed to be the case for the carbonaceous chondrites. If there is some N associated with PNG it is quite difficult to establish its amount and isotopic composition from the data obtained so far. Strictly speaking this will only be possible after physical separation of the PNG carrier. Even with the best separation achieved in the present work, there is still a significant interference from combustible and N-containing carbon phases. Therefore the previous [39–41] and current attempts to identify Q-nitrogen remain unconvincing.

The relatively high amounts of light nitrogen ($\delta^{15}\text{N}$: $-10 \div -40\%$) observed in the enstatite chondrites at high temperatures ($>900^\circ\text{C}$, Appendix) appear to be associated with the combustion of silicon nitride, which has earlier been identified [42] in this type of meteorites.

In more detail the nitrogen and carbon data for the HF-HCl residues will be discussed elsewhere [43].

4. Discussion

Within enstatite chondrite parent bodies transformation of macromolecular organic material into graphite during thermal metamorphism

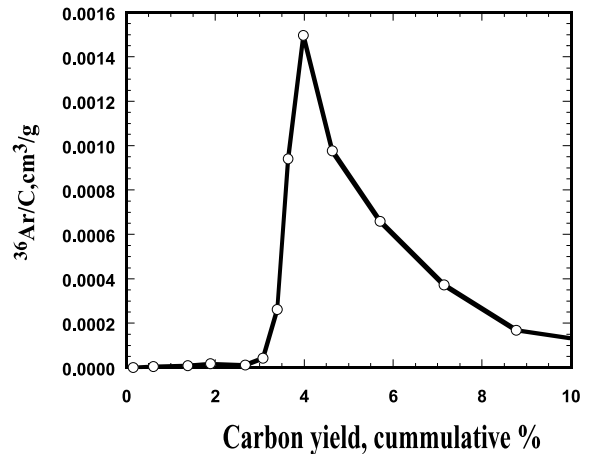


Fig. 3. Variations of ^{36}Ar concentrations during stepped combustion of the Yilmia HF-HCl residue.

under reducing conditions increases its combustion temperature. This ultimately allows the separation of graphite from Q, the PNG carrier, during laboratory stepped combustion extraction 4.5 billion years later. The total $^{36}\text{Ar}/\text{C}$ ratio in the enstatite chondrites is a factor of ~ 3 lower than in carbonaceous chondrites due to, perhaps, partial gas losses from Q and/or its partial disintegration. From this simple observation we must conclude that under reducing conditions, Q is relatively stable. We know from laboratory experiments herein and elsewhere that Q is not stable under oxidizing conditions. These characteristic responses to reducing and oxidizing conditions represent important clues as to the nature of Q.

The upper limit of carbon (if any) associated with PNG can be determined from the results for Yilmia (Fig. 3) and amounts to $\sim 5\%$ of the total carbon. The corresponding concentration of ^{36}Ar in the carrier (if it is carbonaceous) is $\sim 6.8 \times 10^{-4} \text{ cm}^3 \text{ g}^{-1} \text{ C}$ (i.e. cm^3 per gram of carbon). Obviously, this represents only a lower limit for the ^{36}Ar concentration in the carrier because, as can be seen (Fig. 3), the $^{36}\text{Ar}/\text{C}$ ratios are extremely variable during stepped combustion suggesting that ^{36}Ar is, in fact, associated only with a fraction of the carbon released in the temperature range where most of ^{36}Ar is released. Therefore, the best estimate for a lower limit of

^{36}Ar concentration in Q can be obtained from the maximum $^{36}\text{Ar}/\text{C}$ observed for Yilmia ($1.5 \times 10^{-3} \text{ cm}^3 \text{ g}^{-1} \text{ C}$; Fig. 3). This is one of the highest concentrations of Ar detected in Solar System materials.

Planetary Xe and Ar are released from their host phase during stepped combustion in a similar way, though in detail there are some differences. In Yilmia, for example, Xe is released before Ar. This might indicate that Q is not a single carrier, but represents a number of carriers with different relative Ar and Xe concentrations [44]. The same effect can also be observed if Xe and Ar in the grains of their carrier have been acquired as a result of ion implantation. Similar differences for noble gas distributions are observed in presolar diamonds for which ion implantation seems to be the most likely trapping mechanism [45]. The extremely high $^{36}\text{Ar}/\text{C}$ ratio in Yilmia ($1.5 \times 10^{-3} \text{ cm}^3 \text{ g}^{-1} \text{ C}$), which is much higher than in presolar diamonds ($0.06 \times 10^{-3} \text{ cm}^3 \text{ g}^{-1} \text{ C}$ [45]), tends to suggest ion implantation as a mechanism of incorporation.

Our present results do not allow the conclusive identification of Q, the PNG carrier. We have, however, confirmed that it is combustible. Also, we have found that for the meteorites analysed so far, the combustion temperature of Q is almost the same; we postulate that this will be true for all chondritic meteorites as well. The release temperature of Xe observed by Frick and Pepin in their stepped combustion experiment of Allende HF-HCl residues [19] is also in a good agreement with our results. It allows us to conclude that if Q is carbonaceous, it has properties distinct from those of the bulk carbon in the meteorites; for instance, during parent body processing it is not graphitized during thermal metamorphism under reducing conditions. Furthermore, Q is susceptible to oxidation, but under such conditions, survives better than the bulk of meteoritic carbon in the parent bodies.

Enstatite chondrites contain evidence that, in some ways, suggests a relationship with ureilites, including similar $^{36}\text{Ar}/^{132}\text{Xe}$ ratios and carbon isotopic compositions [46]. Comparing the stepped combustion results for the enstatite chondrites obtained in the present work, with similar

published data for the ureilites, appears to strengthen these links. For instance, the release of PNG at a temperature much lower than the bulk carbon as seen in the enstatite chondrite Yilmia (Fig. 2) has also been observed (for Ar at least) for the diamond-free ureilite ALH78079 [39]. Furthermore, it appears that the nature of the PNG carrier is responding to the thermal history of these highly metamorphosed meteorites, i.e. the higher the petrological type the greater the resistance of the PNG carrier to oxidizing acids. For example, in the metamorphosed enstatite chondrite Abee (EH5), only 37% of total P1 Xe is lost during chromic acid treatment compared to almost 100% for enstatite chondrites of lower petrological type [14]. In the more highly metamorphosed ureilite ALH78019, the PNG cannot be released by treatment with HNO_3 [46]. These observations allow us to suggest that PNG in unshocked ureilites are located in the same carrier as in enstatite chondrites (and chondritic meteorites in general). Similar conclusions have been made earlier on the basis of isotope similarities of PNG in ureilites and chondrites [13,20]. It is likely that the higher resistance of the PNG carrier to oxidizing acid etching in ureilites (and in enstatite chondrites of high petrological types) is not connected with diffusion of the PNG from the surfaces to interiors of carbon grains [41]. Alternatively, we suggest that it is a result of increasing metamorphism strengthening the PNG carrier under reducing conditions that occurred in both enstatite chondrite and ureilite parent bodies. A similar conclusion was reached earlier by Huss et al. [14].

There has been some speculation about the possible candidates for the PNG carrier, e.g. fullerenes [47] and nanotubes [48]. Both forms have cage-like structures, which can trap noble gases. The former, however, seem not to fit the observations of noble gas concentrations found in fullerenes extracted from the carbonaceous chondrite Allende [49]. A nanotube carrier appears to be an attractive hypothesis, but so far there is no direct evidence of the presence of nanotubes in meteorites. Other forms of carbon that we should also consider here are diamondoids, which, like fullerenes and nanotubes, have cage-like struc-

tures. Diamondoids (diamond-like carbon [50]) are highly resistant to thermal metamorphism and graphitization in a reducing environment, yet their structure leaves them susceptible to oxidation.

Something which may ultimately constrain the nature and origin of Q is the mode of incorporation of the PNG. Fullerenes and nanotubes open up the possibility that the PNG were occluded contemporaneously with grain growth. On the other hand, we have already alluded to the fact that the PNG may have been added by ion implantation. Clearly this possibility needs to be explored in further work.

5. Conclusions

Though we have not unequivocally identified the PNG carrier in meteorites, we have established key physical and chemical properties that help constrain its nature and will ultimately aid in its separation and identification (although the separation method is not, as yet, obvious). On the basis of the results herein we feel confident in asserting that it will eventually be possible to separate Q. Furthermore, we can hypothesize that under appropriate conditions (i.e. those that

mimic metamorphism on primitive meteorite parent bodies) it should be possible to induce changes in the bulk macromolecular organic carbon that will allow resolution of Q with its PNG, from the bulk of the carbon, during a subsequent stepped combustion. On a more speculative level the concentrations of PNG in Q, coupled with the relative scarcity of the latter, clearly bring into question the accepted origin of the PNG (i.e. as a Solar System-wide process). Rather, it now seems entirely possible that, as has been suggested earlier [2,3], PNG could be presolar.

Acknowledgements

We are grateful to Drs. U. Ott and R.S. Lewis for their critical reviews. The readability of the paper is much improved following the correct implementation of the King's English. We thank U. Ott for the sample of Dhajala. Financial support for this work was from the Particle Physics and Astronomy Research Council.[*BW*]

Appendix. C, N and noble gases in HF–HCl residues from chondritic meteorites

T °C	Dhajjala 0.894 mg				LEW87223 0.745 mg				Indarch 1.255 mg				Yilmia 1 0.638 mg				Yilmia 2 1.542 mg								
	C µg	⁴ He 10 ⁻¹⁰ cm ³	³⁶ Ar 10 ⁻¹⁰ cm ³	¹³² Xe 10 ⁻¹² cm ³	N ng	δ ¹⁵ N ‰	C µg	⁴ He 10 ⁻¹⁰ cm ³	³⁶ Ar 10 ⁻¹⁰ cm ³	¹³² Xe 10 ⁻¹² cm ³	N ng	δ ¹⁵ N ‰	C µg	⁴ He 10 ⁻¹⁰ cm ³	³⁶ Ar 10 ⁻¹⁰ cm ³	¹³² Xe 10 ⁻¹² cm ³	N ng	δ ¹⁵ N ‰	C µg	⁴ He 10 ⁻¹⁰ cm ³	³⁶ Ar 10 ⁻¹⁰ cm ³	¹³² Xe 10 ⁻¹² cm ³	N ng	δ ¹⁵ N ‰	
200	0.49	17.6	0.7	0.9	7.7	31.4	0.47	<0.1	0.3	1.4	-19.7	0.77	<0.1	1.0	0.7	4.0	15.7	0.39	<0.1	0.13	0.19	0.1	1.3	13.1	
250	2.70	2.0	1.3	1.7	15.0	4.5	3.43	0.3	1.5	4.5	-12.2	3.17	17.8	3.1	2.1	5.8	25.8	1.13	<0.1	0.17	0.71	0.4	1.5	-7.1	
300	3.24	5.8	6.2	9.2	39.4	4.3	6.79	0.8	2.2	11.6	-4.7	6.18	23.1	6.8	2.7	20.5	6.6	1.96	0.2	0.15	1.44	0.1	4.0	9.0	
325	2.90	7.9	12.0	21.1	47.2	4.8	5.19	1.0	2.5	14.8	2.7	5.43	13.4	6.0	9.0	26.3	-2.4	1.27	0.2	0.13	1.10	0.2	5.3	3.1	
350	2.64	20.1	17.7	29.2	35.7	5.4	5.69	1.6	4.0	26.3	9.4	4.22	20.0	6.1	7.2	38.0	-4.8	1.97	0.2	0.03	0.87	0.4	9.3	2.1	
375	2.28	21.6	17.0	31.6	24.1	4.4	5.79	2.4	4.6	44.0	11.5	3.37	47.0	6.0	2.8	53.2	-2.7	1.01	0.4	0.18	0.81	0.7	26.1	2.9	
400	2.65	24.1	24.5	28.9	24.9	3.9	5.63	3.1	5.7	74.4	13.7	4.03	59.4	6.4	6.4	75.2	1.7	0.80	2.1	0.44	0.33	1.9	8.1	4.9	
420	3.38	25.9	24.9	18.0	17.7	4.1	4.91	3.5	2.6	86.5	15.8	4.69	33.5	7.8	8.3	92.3	5.4	0.60	5.7	0.88	0.28	1.9	9.5	3.6	
440	2.63	22.1	20.3	22.7	18.1	3.7	4.93	4.3	6.6	91.9	16.0	5.48	69.3	12.7	10.4	89.3	1.4	0.87	13.0	1.15	0.42	3.6	16.3	10.7	
460	1.81	17.4	14.3	45.3	24.0	5.4	5.31	6.8	8.3	83.7	16.0	10.05	177.9	23.4	10.0	93.9	-15.1	1.63	15.9	1.75	0.54	5.5	26.4	10.5	
480	1.47	15.2	9.4	31.3	23.6	6.2	5.85	32.2	9.1	9.0	64.9	13.5	17.32	257.9	27.8	22.8	96.3	-28.7	2.72	17.9	0.75	0.72	9.3	31.4	9.7
500	1.37	14.1	7.9	25.5	20.4	6.6	5.30	25.9	5.3	3.3	29.7	8.4	23.69	199.7	21.4	19.9	86.0	-27.8	3.63	13.5	0.34	2.07	13.7	48.9	13.1
520	1.77	14.9	7.5	13.9	23.3	8.1	5.06	10.2	1.5	0.8	13.4	4.0	28.10	74.9	12.7	8.7	192.2	-6.6	4.09	6.9	<0.03	2.04	14.5	22.2	9.6
540	2.27	14.8	7.5	20.9	24.2	6.3	5.70	<0.1	0.5	0.3	8.4	2.2	30.11	26	7.6	7.0	161.3	-1.4	13.46	1.2	<0.03	3.31	14.2	23.6	8.0
560	2.31	13.6	6.5	15.3	24.0	3.6	5.35	0.1	<0.1	6.9	4.3	30.29	0.7	5.3	5.2	59.9	-2.6	9.17	0.5	0.07	5.41	9.7	31.5	5.9	
600	1.80	4.3	5.2	8.9	43.0	-3.4	4.75	0.1	<0.1	6.2	-1.2	36.68	5.9	3.5	3.4	47.8	-4.3	19.73	0.3	0.04	6.87	3.2	43.0	6.5	
650	1.44	1.7	0.7	2.1	22.0	2.2	5.74	<0.1	<0.1	6.1	-4.0	43.50	0.8	1.6	1.6	57.2	0.5	30.59	<0.1	0.12	11.39	9.2	23.7	2.1	
700	0.34	14.3	<0.1	0.1	1.7	-15.0	9.99	<0.1	0.2	8.1	-9.0	45.45	0.3	1.3	1.0	70.3	-0.7	52.80	0.1	0.16	21.82	3.5	16.4	-11.5	
750	0.40	14.9	0.3	0.1	2.7	-4.7																			
800	1.30	10.5	<0.1	0.3	1.8	11.0	30.85	<0.1	<0.1	13.0	-9.6	43.44	193.0	5.2	1.2	150.4	-16.9	100.61	<0.1	0.31	44.16	0.1	73.7	-28.3	
900	0.65	<0.1	0.3	<0.1	1.5	17.3	5.63	<0.1	<0.1	16.0	-12.3	40.56	76.3	8.0	0.9	128.6	-11.0	2.70	0.1	<0.03	50.87	0.2	682.7	-36.9	
1000	0.09	0.1	<0.1	<0.1	1.6	31.3	0.13	0.3	<0.1	30.6	-13.9	16.00	28					0.16	0.1	<0.03	58.08	0.1	187.7	-35.5	
1100	0.07	<0.1	<0.1	0.1	1.2	-9.3	0.17	<0.1	<0.1	20.7	-13.5							0.02	0.1	<0.03	46.73	0.1	165.4	-29.4	
1200	0.06	0.2	0.1	0.2	1.1	-12.8	0.16	<0.1	<0.1	6.4	-32.0							0.15	<0.1	<0.03	15.69	0.1	48.7	-33.1	
1300	0.08	3.4	<0.1	<0.1	1.7	-6.8	0.32	0.1	0.1	20.7	-32.5							0.30	<0.1	<0.03	0.19	0.2	47.7	-34.6	
1400																									
total	40.13	286.6	184.3	327.4	447.7	4.5	133.2	84.8	40.9	51.9	690.2	7.3	402.52	1324.7	173.6	131.3	1548.5	-7.5	251.79	78.6	6.81	276.28	92.7	1667.3	-26.5

T °C	Orgueil 0.206 mg					Murchison 1.287 mg					Renazzo 1.203 mg						
	C µg	⁴ He 10 ⁻¹⁰ cm ³	³⁶ Ar 10 ⁻¹⁰ cm ³	¹³² Xe 10 ⁻¹² cm ³	N ng	δ ¹⁵ N ‰	C µg	⁴ He 10 ⁻¹⁰ cm ³	³⁶ Ar 10 ⁻¹⁰ cm ³	¹³² Xe 10 ⁻¹² cm ³	N ng	δ ¹⁵ N ‰	C µg	⁴ He 10 ⁻¹⁰ cm ³	³⁶ Ar 10 ⁻¹⁰ cm ³	N ng	δ ¹⁵ N ‰
200	0.16	10.2	0.29	1.35	3.2		3.29	3.0	3.6	3.1	177.6	3.29	3.0	3.6	3.1	177.6	
250	0.63	22.7	0.84	2.09	5.7	53.5	15.76	9.0	13.7	23.7	107.9	15.76	9.0	13.7	23.7	107.9	
300	1.49	25.0	1.21	2.05	23.2	45.9	30.27	54.1	20.7	74.4	136.0	30.27	54.1	20.7	74.4	136.0	
325	1.47	46.5	0.36	3.25	8.5	81.9	33.95	66.4	21.1	190.1	150.4	33.95	66.4	21.1	190.1	150.4	
350	1.62	48.5	1.19	1.76	27.6	59.2	41.36	103.3	23.7	684.8	159.8	41.36	103.3	23.7	684.8	159.8	
375	1.78	19.8	1.47	2.29	43.9	32.2	3.33	10.4	8.0	9.9	44.6	3.33	10.4	8.0	9.9	44.6	
400	2.02	17.7	1.71	2.20	39.5	4.0	7.75	38.5	13.3	15.3	99.1	7.75	38.5	13.3	15.3	99.1	
420	1.89	16.9	1.80	2.46	38.0	-3.6	11.58	30.0	12.8	15.8	218.9	11.58	30.0	12.8	15.8	218.9	
440	1.77	20.2	1.81	1.84	40.7	-5.8	17.61	39.2	13.3	16.5	399.9	17.61	39.2	13.3	16.5	399.9	
460	1.54	36.4	2.10	1.89	31.0	-3.4	22.51	54.3	16.1	17.4	673.4	22.51	54.3	16.1	17.4	673.4	
480	1.29	69.4	2.05	1.99	23.6	-2.9	18.87	86.2	17.5	17.3	789.8	18.87	86.2	17.5	17.3	789.8	
500	1.01	103.2	1.73	1.68	17.2	4.7	12.91	122.5	17.8	15.2	528.0	12.91	122.5	17.8	15.2	528.0	
520	0.64	49.8	1.06	0.81	13.7	22.9	5.78	166.6	11.2	7.7	189.1	5.78	166.6	11.2	7.7	189.1	
540	0.41	25.2	0.65	0.76	8.8	40.4	1.37	234.7	4.7	2.4	22.5	1.37	234.7	4.7	2.4	22.5	
560	0.28	2.8	0.45	0.77	5.9	46.3	0.94	283.3	2.3	1.2	17.4	0.94	283.3	2.3	1.2	17.4	
600	0.27	7.4	0.43	0.93	5.8	26.6	1.73	642.0	2.2	1.7	31.4	1.73	642.0	2.2	1.7	31.4	
650	0.17	10.3	0.27	0.25	6.6	54.0	0.62	159.7	0.7	0.7	18.2	0.62	159.7	0.7	0.7	18.2	
700	0.08	0.0	0.19	0.20	8.0	59.0	0.10	3.9	4.1	0.3	9.2	0.10	3.9	4.1	0.3	9.2	
800	0.10	1.4	0.12	0.09	14.0	61.6	0.06	7.3	0.2	0.4	7.4	0.06	7.3	0.2	0.4	7.4	
900	0.02	0.0	0.27	0.00	1.7	55.9	0.03	9.2	0.3	0.1	7.0	0.03	9.2	0.3	0.1	7.0	
1000	0.02	0.0	0.08	0.00	1.5	46.2	0.04	6.4	0.3	0.2	7.6	0.04	6.4	0.3	0.2	7.6	
1100	0.02	0.0	0.04	0.19	1.2	-13.2	0.03	14.0	0.3	0.2	8.4	0.03	14.0	0.3	0.2	8.4	
1200	0.02	0.0	0.08	0.00	1.1	-0.2		23.0	129.3	3.3	8.1		23.0	129.3	3.3	8.1	
1300	0.02	3.8	0.09	0.13	1.4	-11.2											
1400	0.04	10.9	0.12	0.06	2.9	2.6											
total	18.75	548.0	20.41	29.04	374.9	20.6	107.95	1931.2	129.4	129.3	3107.7	-25.4	479.45	5079.4	348.6	17870.1	157.0

References

- [1] R.S. Lewis, B. Srinivasan, E. Anders, Host phase of a strange xenon component in Allende, *Science* 190 (1975) 1251–1262.
- [2] U. Ott, R. Mack, S. Chang, Noble gas rich separates from the Allende meteorite, *Geochim. Cosmochim. Acta* 45 (1981) 1751–1788.
- [3] G.R. Huss, E.C. Alexander Jr., On the presolar origin of the ‘normal planetary’ noble gas component in meteorites, *Proc. Lun. Planet. Sci. Conf. 17th Part II, J. Geophys. Res. Suppl.* 92 (1987) E710–E716.
- [4] M. Tang, E. Anders, Isotopic anomalies of Ne, Xe and C in meteorites. III. Local and exotic noble gas components and their interrelations, *Geochim. Cosmochim. Acta* 52 (1988) 1245–1254.
- [5] R.O. Pepin, Origin of noble gases in the terrestrial planets, *Annu. Rev. Earth Planet. Sci.* 20 (1992) 389–430.
- [6] M. Ozima, R. Wieler, B. Marty, F.A. Podosek, Comparative studies of solar, Q-gases and terrestrial noble gases, and implications on the evolution of the solar nebula, *Geochim. Cosmochim. Acta* 62 (1998) 301–314.
- [7] R. Wieler, ‘Q-gases’ as ‘local’ primordial noble gas component in primitive meteorites, in: J. Matsuda (Ed.), *Noble Gas Geochemistry and Cosmochemistry*, Terra Scientific Publishing Company, Tokyo, 1994, pp. 31–41.
- [8] M. Ozima, F.A. Podosek, *Noble Gas Geochemistry*, Cambridge University Press, Cambridge, 1983, 367 pp.
- [9] R.O. Pepin, On the origin and early evolution of terrestrial planet atmospheres and meteoritic volatiles, *Icarus* 92 (1991) 2–79.
- [10] L. Alaerts, R.S. Lewis, E. Anders, Primordial noble gases in chondrites: the abundance pattern was established in the solar nebular, *Science* 198 (1980) 927–930.
- [11] L. Alaerts, R.S. Lewis, E. Anders, Isotopic anomalies of noble gases in meteorites and their origins. III. LL-chondrites, *Geochim. Cosmochim. Acta* 43 (1979) 1399–1416.
- [12] J.-I. Matsuda, R.S. Lewis, H. Takahashi, E. Anders, Isotopic anomalies in meteorites and their origins, VII. CV3 carbonaceous chondrites, *Geochim. Cosmochim. Acta* 44 (1980) 1861–1874.
- [13] N. Schelhaas, U. Ott, F. Begemann, Trapped noble gases in unequilibrated ordinary chondrites, *Geochim. Cosmochim. Acta* 54 (1990) 2869–2882.
- [14] G.R. Huss, R.S. Lewis, S. Hemkin, The ‘normal planetary’ noble gas component in primitive chondrites: compositions, carrier, and metamorphic history, *Geochim. Cosmochim. Acta* 60 (1996) 3311–3340.
- [15] H.W. Weber, F. Begemann, H. Hintenberger, Primordial gases in graphite-diamond-kamacite inclusions from Haverø ureilite, *Earth Planet. Sci. Lett.* 29 (1976) 81–90.
- [16] R. Gobel, U. Ott, F. Begemann, On trapped noble gases in ureilites, *J. Geophys. Res.* 83 (1978) 855–867.
- [17] T. Yamamoto, K. Hashizume, J.-I. Matsuda, T. Kase, Multiple nitrogen isotopic components coexisting in ureilites, *Meteorit. Planet. Sci.* 33 (1998) 857–870.
- [18] U. Frick, S. Chang, Elimination of chromite and novel sulphides as important carriers of noble gases in carbonaceous meteorites, *Meteoritics* 13 (1978) 465–470.
- [19] U. Frick, R.O. Pepin, On the distribution of noble gases in Allende: a differential oxidation study, *Earth Planet. Sci. Lett.* 56 (1981) 45–63.
- [20] U. Ott, J. Kronenbitter, J. Flores, S. Chang, Colloidally separated samples from Allende residues: noble gases, carbon and an ESCA-study, *Geochim. Cosmochim. Acta* 48 (1984) 267–280.
- [21] F.P. Fanale, W.A. Cannon, Physical adsorption of rare gas on terrigenous sediments, *Earth Planet. Sci. Lett.* 11 (1971) 362–368.
- [22] J.F. Wacker, Laboratory simulation of meteoritic noble gases. III. Sorption of neon, argon, krypton and xenon on carbon. Elemental fractionation, *Geochim. Cosmochim. Acta* 53 (1989) 1421–1433.
- [23] G.R. Huss, R.S. Lewis, Noble gases in presolar diamonds I: three distinct components and their implications for diamond origins, *Meteoritics* 29 (1994) 791–810.
- [24] R.S. Lewis, S. Amari, E. Anders, Interstellar grains in meteorites. II. SiC and its noble gases, *Geochim. Cosmochim. Acta* 58 (1994) 471–494.
- [25] C.M.O’D. Alexander, S.S. Russell, J.W. Arden, R.D. Ash, M.M. Grady, C.T. Pillinger, The origin of chondritic macromolecular organic matter: a carbon and nitrogen isotope study, *Meteorit. Planet. Sci.* 33 (1998) 603–622.
- [26] R.S. Lewis, J. Gros, E. Anders, Isotopic anomalies of noble gases in meteorites and their origins 2. Separated minerals from Allende, *J. Geophys. Res.* 82 (1977) 779–792.
- [27] J.H. Reynolds, U. Frick, J.M. Neil, D.L. Phinney, Rare-gas-rich separates from carbonaceous chondrites, *Geochim. Cosmochim. Acta* 42 (1978) 1775–1797.
- [28] M.G. Zadnik, J.F. Wacker, R.S. Lewis, Laboratory simulation of meteoritic noble gases. II. Sorption of xenon on carbon: etching and heating experiments, *Geochim. Cosmochim. Acta* 49 (1985) 1049–1059.
- [29] J.F. Wacker, M.G. Zadnik, E. Anders, Laboratory simulation of meteoritic noble gases. I. Sorption of xenon on carbon: trapping experiments, *Geochim. Cosmochim. Acta* 49 (1985) 1035–1048.
- [30] C.M.O’D. Alexander, J.W. Arden, R.D. Ash, C.T. Pillinger, Presolar components in the ordinary chondrites, *Earth Planet. Sci. Lett.* 99 (1990) 220–229.
- [31] S. Amari, R.S. Lewis, E. Anders, Interstellar grains in meteorites: I. Isolation of SiC, graphite, and diamond: size distribution of SiC and graphite, *Geochim. Cosmochim. Acta* 58 (1994) 459–470.
- [32] M.A. Sephton, C.T. Pillinger, I. Gilmour, Small-scale hydrous pyrolysis of macromolecular material in meteorites, *Planet. Space Sci.* 47 (1999) 181–187.
- [33] I.P. Wright, S.R. Boyd, I.A. Franchi, C.T. Pillinger, High-precision determination of nitrogen stable isotope ratios at the sub-nanomole level, *J. Phys. E* 21 (1988) 865–875.
- [34] A.B. Verchovsky, A.V. Fisenko, L.F. Semjonova, C.T. Pillinger, Heterogeneous distribution of Xe-HL within

- presolar diamonds, *Meteorit. Planet. Sci.* 32 (1997) A131–132.
- [35] I.P. Wright, C.T. Pillinger, Carbon isotopic analysis of small samples by use of stepped-heating extraction and static mass spectrometry, *US Geol. Bull.* 7890 (1989) 9–34.
- [36] G.R. Huss, A.P. Meshik, C.M. Hohenberg, Abundances of presolar grains in Renazzo and Axtell: implications for their thermal histories, *Lunar Planet. Sci. Conf. XXXI*, 2000, CDROM, abstr. No. 1467.
- [37] R.N. Clayton, K.T. Mayeda, J.L. Gooding, K. Keil, E.J. Olsen, Redox processes in chondrules and chondrites, *Lun. Planet. Sci. XII* (1981) 154–156.
- [38] M.A. Sephton, I. Gilmour, Macromolecular organic materials in carbonaceous chondrites: a review of their sources and their role in the origin of life on the Early Earth, in: I. Gilmour, C. Koberl (Eds.), *Impacts and the Early Earth*, Lecture Notes in Earth Sciences, Vol. 91, Springer, New York, 2000, pp. 27–49.
- [39] S.V.S. Murthy, V.K. Rai, U. Ott, Nitrogen and noble gases in the diamond-free ureilite Allan Hills 78019, *Meteorit. Planet. Sci.* 34 (1999) A85.
- [40] S.V.S. Murthy, Isotopic composition of nitrogen in ‘phase Q’, *Earth Planet. Sci. Lett.* 141 (1996) 307–313.
- [41] K. Hasizume, Y.T. Nakaoka, Q nitrogen in CO₃ chondrites, *Meteorit. Planet. Sci.* 33 (1998) A65.
- [42] M.R. Lee, S.S. Russell, J.W. Arden, C.T. Pillinger, Nierite (Si₃N₄), a new mineral from ordinary and enstatite chondrites, *Meteoritics* 30 (1995) 387–398.
- [43] M.A. Sephton, A.B. Verchovsky, P.A. Bland, I. Gilmour, M.M. Grady, I.P. Wright, Investigating the variations in carbon and nitrogen isotopes in carbonaceous chondrite meteorites, *Geochim. Cosmochim. Acta* (2002) submitted.
- [44] H. Busemann, H. Baur, R. Wieler, Primordial noble gases in ‘phase Q’ in carbonaceous and ordinary chondrites studied by closed-system stepped etching, *Meteorit. Planet. Sci.* 35 (2000) 949–973.
- [45] A.B. Verchovsky, A.V. Fisenko, L.F. Semjonova, I.P. Wright, M.R. Lee, C.T. Pillinger, C, N, and noble gas isotopes in grain size separates of presolar diamonds from Efremovka, *Science* 281 (1998) 1165–1168.
- [46] J.F. Wacker, Noble gases in the diamond-free ureilite, ALHA 78019: the roles of shock and nebular processes, *Geochim. Cosmochim. Acta* 50 (1986) 633–642.
- [47] D. Heymann, Buckminsterfullerene, its siblings, and soot: carriers of trapped inert gases in meteorites? *Proc. Lunar Planet. Sci. Conf. 17th Part I*, *J. Geophys. Res.* 91 (1986) E135–E138.
- [48] D. Heymann, R.D. Vis, A novel idea about the nature of phase Q, *Lunar Planet. Sci. Conf. XXIX*, 1998, CDROM, abstr. No. 1098.
- [49] L. Becker, R.J. Poreda, T.E. Bunch, Fullerenes: an extra-terrestrial carbon carrier phase for noble gases, *Proc. Natl. Acad. Sci. USA* 97 (2000) 2979–2983.
- [50] R.C. Fort, *Adamantane: the Chemistry of Diamond Molecules*, Marcel Dekker, New York, 1976.