Separation of Q from carbon in CR meteorites during stepped combustion

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Introduction: The nature of the planetary noble gas carrier (Q) in meteorites remains uncertain. It is known that it is likely to be carbonaceous, but represents only a small fraction of the total macromolecular material. Q is oxidizable with nitric and other oxidizing acids. It seems to be partly destroyed by pyridine and may have an organic structure. Previously, we have shown that during parent body thermal metamorphism Q is less affected than the majority of other carbonaceous materials [1]. If organic matter is graphitized, as has happened in the enstatite chondrite parent bodies, Q remains unaffected. In the present study we have found that Q is also separable from the majority of carbon in type 2 and 3 CR chondrites during stepped combustion. It is possible that this is because Q has become encased within the matrix, in contrast to other carbon phases, during parent body metamorphism.

Samples and analysis: We have analysed 12 whole rock CR meteorites of all petrological groups: CR1 (GRO 95577 and Al Rais); CR2 (EET 92042, EET 87770, EET 26259, PCA 91082, GRA 95229, MAC 87320, Acfer 059, and El Djouf) and CR3 (NWA 1152 and SAH 00182). Their C, N, Ar and (for 4 of them) Xe isotopes have been measured simultaneously using our mass spectrometric complex Finesse [1, 2]. The gases have been released by stepped combustion in the temperature range from 200 to 1400°C with 100°C increments.

Results: The concentration of $^{36}$Ar in the samples varies by more than an order of magnitude from 2.4 x $10^{-7}$ to $5 \times 10^{-6}$ cc/g. There is no other carrier, apart from Q, that can provide such high concentrations of $^{36}$Ar in the bulk meteorites including nanodiamonds. This follows from the known concentration of $^{38}$Ar in nanodiamonds [3] and the concentration of nanodiamonds in CR meteorites [4].

Independent of petrologic type, most C and N in the samples is released in the temperature range from 300 to 800°C with a peak release at about 500°C (Figs. 1, 2). Variations of $\delta^{13}$N and $\delta^{15}$C are within the range found in the same or similar samples earlier [5, 6]. In contrast to C and N, the release pattern of $^{36}$Ar strongly depends on the petrologic type: in CR1s it is broad and almost similar to C and N (Fig. 1), in CR2s it is narrow (between 900 and 1100°C) with a peak at 1000°C, and in CR3s it tends to have a 100°C higher release temperature than in CR2s (Fig. 2). In one case (NWA 1152), Ar has a bimodal release with peaks at 800 and 1100°C. Xe, apart from a peak coinciding with Ar release at 1000°C, also has a higher T peak at 1200°C (Fig. 2). $^{36}$Ar/$^{132}$Xe ratios for the total release of the gases are close to that expected for the Q component.

N isotopic composition is heavy for all CR1 and CR2 samples ($\delta^{15}$N=+100 to +180‰), but light for CR3s (-2 to -3‰). However for all CR2 and CR3 samples, $\delta^{15}$N has an excursion to the light N composition (from +60 to -150‰) that almost perfectly coincides with the peak of Ar release (Fig. 2). This is not observed for CR1s (Fig. 1).

Discussion: The most important observation is the separation of carbon and $^{36}$Ar during stepped combustion of CR2 and CR3 samples. Most low-temperature carbonaceous material in the bulk samples of all petrological types can be easily accessed and oxidized by the molecular oxygen used in the experiments. In the CR1 samples, Q and other carbonaceous materials are equally accessible for oxygen. For CR2 and CR3 samples, the situation is significantly different. Up to 800-900°C, oxygen does not react with Q, possibly because the phase is somehow isolated from its direct access. One of the explanations for this could be that the Q grains are encased within the matrix which remains stable at T<900°C. At T>900°C, the matrix opens up and oxygen reacts with Q grains releasing...
noble gases. Ar can be released from Q without oxygen at such a high temperatures. However in this case its release pattern is broader [7]. Therefore we believe that oxygen plays a role during Ar and Xe release. The results for oxidation of HF/HCl residue of Renazzo (CR2) [1] can be regarded as an argument in favor of the above explanation: in this case, Q noble gases are released at low temperatures simultaneously with the majority of carbon. The results here suggest that very little carbon is associated with Q-Ar. The highest $^{36}$Ar/C ratio of 0.1 cc/g is obtained for PCA 91082 at 1000°C. This is probably the highest concentration of Ar observed so far in Solar System objects.

Figure 2. C, N, $^{36}$Ar and $^{132}$Xe release patterns and $\delta^{15}$N profile.

The simplest explanation for such a peculiar correlation between release of $^{36}$Ar and an isotopically light N component (Fig. 2) is that the light N is a part of Q. However, we need to exclude that it is not from nanodiamonds, which have $\delta^{15}$N $\approx$ 350‰. On the three-isotope diagram (Fig. 3) the point corresponding to the maximum $^{36}$Ar release at 1000°C for El Djouf with $\delta^{15}$N $=$ -140‰ is not on the mixing line between average nanodiamond composition and the rest of the data points for the other temperature steps. If we consider only a few steps preceding and following that with the lowest $\delta^{15}$N, then one can conclude that Q nitrogen is light ($\delta^{15}$N $< -$140‰) and has a high N/C ratio $\sim$ 0.1.

Separation of C and $^{36}$Ar in type 2 and 3 CR meteorites during stepped combustion suggests a physical separation of the Q carrier from the majority of carbon. For that, the sample NWA 1152 was combusted up to 700°C at which temperature almost all C is oxidized but only a fraction of Q gases are released. After that, the sample was removed from the furnace and treated with HF/HCl in order to dissolve all metal and silicate phases. Since Q is usually resistant to these acids we expected to obtain the residue enriched in Q and depleted in carbon. Surprisingly, almost all Q was also destroyed by the treatment. Less than 1% of the original $^{36}$Ar remained in the residue and that was released at low temperature (peak at 500°C). We suggest that heating the sample to 700°C prior to HF/HCl treatment somehow affects the Q carrier in such a way that it becomes soluble in the acids.

**Conclusions.** Q is a separate phase with different properties from most carbonaceous materials in CR3 chondrites. During metamorphism in CR parent bodies, Q becomes encased in the matrix preventing it from being oxidized in the laboratory and allowing it to be efficiently separated from other carbonaceous phases during stepped combustion. The extremely high $^{36}$Ar/C ratio (up to 0.1 cc/g) in some of CR2 and CR3 meteorites suggests that Q may not be carbonaceous at all. Apart from noble gases, Q contains isotopically light N ($\delta^{15}$N$<-140‰$), that may be related to solar nitrogen.

**References:**