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Separation of Q-Noble gas carrier from bulk carbon in enstatite chondrites by stepped combustion

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SEPARATION OF Q-NOBLE GAS CARRIER FROM BULK CARBON IN ENSTATITE CHONDRITES BY STEPPED COMBUSTION. A. B. Verchovsky, M. A. Sephton, I. P. Wright and C. T. Pillinger. Planetary and Space Sciences Research Institute, The Open University, Milton Keynes, UK.

The nature of the planetary noble gas (PNG) carrier or Q-phase [1] in meteorites is still a subject of discussion. Failure to identify and isolate this carrier since the first successful concentration of it by a factor of ~200 in an Allende HF-HCl residue [1] and establishing it to be carbonaceous in nature [2], led eventually to the so-called "labyrinth" theory [3], according to which PNG initially found their way through a complex network of micropores on the surface of the carbon grains during adsorption. Following adsorption, the labyrinth channels could be plugged with organic molecules, preventing the release of PNG at elevated temperatures, or/and exchange with gases from other reservoirs such as the terrestrial atmosphere. In support of this theory a number of rather sophisticated experimental investigations on the sorption of noble gases on carbon grain surfaces have been undertaken [3-5], which have proved that the labyrinth mechanism can in principle be realised in natural conditions. However whether or not this is the case for PNG in meteorites remains uncertain. An important observation that fits the theory is the very effective removal of PNG from HF-HCl residues by treatment with oxidizing acids during which only a few percent of the total carbon is destroyed. Oxidation with pure oxygen should work in a similar way, although there have been no systematic investigations of the release of PNG and carbon from HF-HCl residues during stepped combustion.

In the present study we decided to fill this gap by analysing a number of HF-HCl residues separated from meteorites of different types and metamorphic history for noble gases, nitrogen and carbon using detailed stepped combustion. We analysed three carbonaceous chondrites (Orgueil (CI), Murchison (CM) and Renazzo (CR)), three enstatite chondrites (LEW87223 (E3), Indurich (EH4) and Yilmia (EL6)) and one ordinary chondrite Dhajala (H3.8).

Figure 1 shows a typical example of carbon and noble gases release from 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 oxidised. More precisely, when 80% of C is combusted only about 60% of planetary Ar and Xe are released. This release pattern is compatible with PNG being a volume, rather than surface-correlated component. A similar relationship

between C and PNG is also observed in most ureilites [6], in which PNG are volume-correlated, as they located mostly in shock-produced diamonds and therefore cannot be removed by treatment with HNO₃. Thus, oxidation with pure oxygen in a vacuum affects the release of PNG in carbonaceous chondrites in a way significantly different from what occurs during the treatment of the HF-HCl residues with nitric acid, and suggests that PNG are located not on the surface of any carbon grains but rather in a separate carrier (Q-phase). The specific chemical properties of this carrier remain, however, poorly understood.

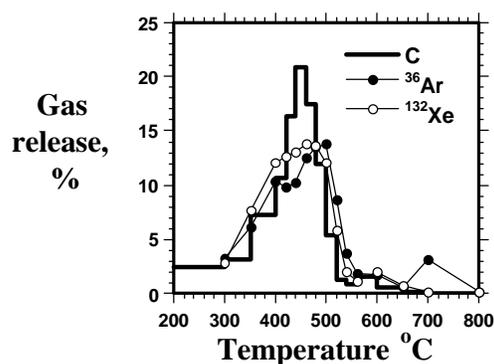


Figure 1. Ar, Xe and carbon (in the form of CO₂) release from HF-HCl residue from Murchison at stepped combustion. Isotopic compositions of the Ar and Xe within experimental errors (1-3%) correspond well to the planetary values.

A quite different and unexpected relationship between the release of PNG on one hand and carbon and nitrogen on the other is observed for enstatite chondrites (Fig. 2). In these meteorites, the release of most of the PNG and carbon are significantly separated from each other with the degree of separation depending on the metamorphic class of the meteorites. In all of the samples, the maximum PNG release occurs at the same temperature (about 500°C) while the release of carbon systematically shifts to higher temperatures (Fig. 2) with increasing thermal metamorphism from E3 (LEW87223) to EL6 (Yilmia). Compared to the carbonaceous chondrites discussed above, the peak of PNG release in enstatite chondrites seems to be a bit narrower, though the release maximum is observed at nearly the same temperature in both cases (Figs. 1 and 2).

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The simplest interpretation of the results is that there is a minor and presumably carbonaceous PNG carrier (Q-phase), which is present in all of the meteorites analysed in this study. It is effectively dissolved in oxidising acids (e. g. HNO_3) and oxidised with pure oxygen. During stepped

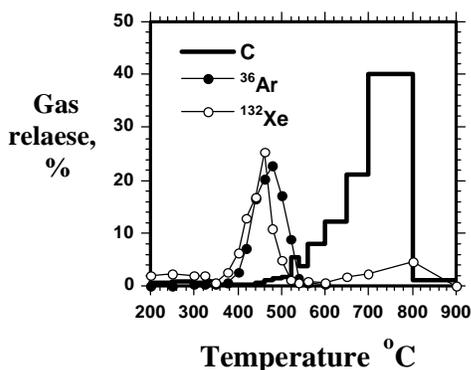


Figure 2. Release of Ar, Xe and C from HF-HCL residue from Yilmia enstatite chondrite at stepped combustion.

combustion experiments the majority of macromolecular carbon in carbonaceous chondrites (and perhaps in other primitive chondrites as well) overwhelms the PNG carrier as their combustion temperatures are nearly the same. In contrast transformation of the bulk of the macromolecular material into graphite during thermal metamorphism under reducing conditions in the enstatite chondrite parent bodies increases its combustion temperature and separates it from the thermally-resistant PNG carrier allowing the two components to be subsequently resolved during stepped combustion. The low limit of carbon (if any) associated with PNG can be determined from the results for Yilmia and amounts to <5% of the total carbon. The minimum ^{36}Ar concentration in the carrier (if it is carbonaceous) is $0.0015 \text{ cm}^3/\text{gC}$. This is the highest concentration of Ar observed so far in any 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 (Fig. 2), Xe is released slightly before Ar. This suggests a different distribution of Xe and Ar in the grains of their carrier, which could be a result of ion implantation, especially if the implantation range is comparable with the grain sizes. Similar differences for noble gas distributions are observed in presolar diamonds for which ion implantation seems to be the most likely

trapping mechanism [7]. The extremely high $^{36}\text{Ar}/\text{C}$ ratio in Yilmia also suggests ion implantation.

In Dhajala, PNG are also separated from most of carbon. Our results are similar to those obtained earlier [8]. This meteorite has a very low carbon content (0.015% [8]) which seems to be the result of a strong thermal metamorphism in an oxidising environment. However, in contrast to the carbonaceous chondrites, the $^{36}\text{Ar}/\text{C}$ ratio in Dhajala is an order of magnitude higher (but still a factor of 1.5 lower than in Yilmia), indicating that the PNG-carrier can survive oxidative degradation much better than the bulk organic matter.

Our results do not allow us to identify conclusively the PNG carrier. We have, however, established that its combustion temperature is almost the same in all types of chondrites and ureilites. If it is carbonaceous, it has properties distinct from those for the bulk carbon in the meteorites: it is not graphitised during strong thermal metamorphism under reducing conditions; it is susceptible to oxidation, but under such conditions, survives better than the bulk meteoritic carbon.

There have been some speculations about a possible candidate for the PNG carrier: fullerenes [9] and nanotubes [10] - both forms have cage-like structures, which can trap noble gases. The former, however, seems not to fit the observations for the PNG concentrations [11]. The latter appears to be a more attractive hypothesis though there is no direct evidence of the presence of nanotubes in meteorites so far. Another possible form of carbon which might be the PNG-carrier is diamondoid. Similar to fullerenes and nanotubes these hydrocarbon organic molecules have a cage-like structure and are highly resistant to thermal metamorphism and graphitisation in reducing environments.

In conclusion, 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 that will aid in its isolation and identification in the future.

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