

NITROGEN ISOTOPE STUDIES OF THE BENCUBBIN METEORITE.

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The Bencubbin polymict breccia consists of recrystallised kamacite and silicates of an unusual achondritic nature welded together by a shock-melted metal/silicate matrix. The meteorite also contains a variety of chondritic xenoliths and shows evidence for intensive shock (1) and remelting of the host silicates. The formation locale and conditions are open to speculation. Kallemeyn et al. (2) propose a low velocity impact into a carbonaceous chondrite type regolith followed by melting, metal/silicate separation and magma flow. Newsom and Drake (3), on the other hand, consider that the chemical characteristics of the metal were imparted during condensation and as such should be considered as primary in origin.

Oxygen isotopic compositions for the host silicates and xenoliths in Bencubbin (and Weatherford, a very similar meteorite) are quite distinct (4) and more recently it has been recognised that these meteorites show enrichments in ^{15}N far beyond any previously studied extraterrestrial sample (5-7). Whole rock $\delta^{15}\text{N}$ values (+923 and +973‰) are greater than any values measured for the major constituents of metal and silicate (+829 to 489‰ respectively) (5,7), therefore, the nitrogen isotope anomaly would appear to be associated with a minor phase. Quantitative and isotopic determinations of metal and silicate using stepped combustion suggest a common host for the heavy nitrogen and are thus consistent with its occurrence within adhering matrix material (6).

Previous studies demonstrate variability in the isotopic composition of the nitrogen throughout Bencubbin which suggests mixing of two components - one highly enriched in ^{15}N , the other composed of isotopically light (or normal) nitrogen. The purpose of the present work is to establish an upper limit to ^{15}N enrichment, to elucidate the location, distribution and identification of the carrier phase and to recognise any light nitrogen component. Noble gases in Bencubbin show no unusual compositions (although there may be a sub-solar component (8)) and the vast majority of the carbon in Bencubbin displays no isotopic irregularities (6). Therefore, in order to discover how the ^{15}N excess was produced (candidates include nucleosynthesis, photochemical reactions, extreme mass dependant isotopic fractionation, etc.) relationships with other elements must be considered.

A stepped combustion of a hand-picked matrix sample supports the idea of a matrix location for heavy nitrogen by providing a high temperature fraction with a $\delta^{15}\text{N}$ similar to the highest bulk measurement, albeit with a lower nitrogen abundance. This conclusion is also favoured by laser microprobe studies. Unfortunately, due to the low concentrations present, the isotope data are subject to large errors, nevertheless a concentration of heavy nitrogen was observed in the matrix relative to the metal and silicate clasts. It also appears that the unusual chondritic clasts may be eliminated as a source for the heavy nitrogen since both of the specimens so far analysed (a carbonaceous and an ordinary chondrite) show only modest ^{15}N enrichments with bulk $\delta^{15}\text{N}$ values of +209 and +284‰ respectively.

The most successful experiments to identify the source of the N enrichment have involved chemical treatment of a 2g sample of metal clasts and matrix with various acids. The parent sample was first treated with 6M HCl (2hr at 70°C) which resulted in a 92.1% weight loss. A stepped

combustion on the residue yielded 671 ppm N with a bulk $\delta^{15}\text{N}$ of +939‰. The calculated nitrogen concentration of 53 ppm in the initial sample is within the range of whole rock values (5) and implies that most, if not all, of the heavy nitrogen in Bencubbin is in an HCl-resistant carrier. The stepped combustion data show two distinct releases of gas, the main one from 300 to 500°C with a $\delta^{15}\text{N}$ of about +984‰ and a distinct secondary release accounting for 10% of the gas from 525 to 550°C with a $\delta^{15}\text{N}$ of +975‰. More than 96% of the total nitrogen was released below 600°C which contrasts markedly with the undissolved sample experiments where most of the isotopically heavy nitrogen is liberated between 900 and 1100°C (6). We therefore believe that the heavy nitrogen host phase is shielded by an HCl-soluble material, presumably metallic iron.

A portion of the HCl residue was treated with HF/HCl for 72 hr at room temperature whereupon a further weight loss of 99.6% occurred to leave a very fine, powdery, black residue. A stepped combustion analysis yielded 6000 ppm N with a bulk $\delta^{15}\text{N}$ of +857‰ which represents just under 2 ppm N in the parent metal/matrix sample. The release profile shows that a substantial proportion of the heavy nitrogen liberated from the HCl residue between 300 and 500°C has been removed by the HF treatment. The minor release previously observed at 550°C in the HCl residue corresponds with the major release in the HF/HCl residue which has a $\delta^{15}\text{N}$ value of +903‰. Once again very little nitrogen was released above 700°C although a maximum $\delta^{15}\text{N}$ of +1168‰ was observed at 725°C. Another aliquot of the HF/HCl resistant residue was combusted and measured for carbon. The sample contained only 2.16 wt% C with the major release from 300 to 400°C ($\delta^{13}\text{C}$ value of +1‰). The C/N ratio for the bulk residue is 4; over the temperature range of the main release of the nitrogen the C/N ratio falls to 0.1.

The above experiments have identified the existence of two heavy nitrogen carriers, both stable in HCl but the minor one also resistant to HF. Neither seem to be carbonaceous in nature. The low temperature of nitrogen release during combustion argues strongly against a silicate but coincides quite closely with that obtained for various sulphides including troilite (9). Newsom and Drake (3) and Hutchison (this volume) report that troilite in Bencubbin is extremely chromium rich (up to 30 wt%). Such high concentrations of chromium might make the mineral acid stable similar to daubreelite (FeCr_2S_4) which has been recognised in Bencubbin (10). The possibility that sulphide minerals are implicated as heavy nitrogen carriers is fascinating since presolar Cr and Ti sulphides might be produced in supernovae (11). The hot CNO cycle which would accompany a star evolved to the silicon burning stage can produce $^{15}\text{N}/^{14}\text{N}$ ratios which are very large (12).

Refs:(1) Jain and Lipschutz (1973) Nature, 242, 26. (2) Kallemeyn et al (1978) GCA, 42, 507. (3) Newsom and Drake (1979) GCA, 43, 689. (4) Clayton and Mayeda (1978) GCA, 42, 325. (5) Prombo and Clayton (1985) LPS XVI, 679. (6) Franchi et al (1985) Meteoritics, 20 in pr. (7) Prombo and Clayton (1985) Science, 230, 935. (8) Lewis et al (1985) Meteoritics, in pr. (9) Burgess et al (1985) LPS XVI, 99. (10) Ramdohr (1973) The opaque minerals in stoney meteorites. (11) Clayton and Ramadurai (1977) Nature, 265, 427. (12) Audouze and Vauclair (1980) An introduction to nuclear astrophysics. We thank Dr. R. Hutchison for samples and very helpful discussions.