NOBLE GAS TRAPPING DURING CONDENSATION: A LABORATORY STUDY, B.K. Kothare, K. Marti, S. Niemeyer, S. Regnier and J.R. Stephens, Univ. of California, San Diego, Chemistry Department, La Jolla, California 92093.

Although the origin of planetary-type rare gases in meteorites is still poorly understood, a major step forward has been the isolation of a carbon-rich trace phase in carbonaceous chondrites which carries most of the rare gases [1,2]. Lewis et al. [1] argued that a metallic sulfide carries most of the rare gases, but Reynolds et al. [2] preferred a carbonaceous gas-carrier. The mechanism responsible for the fractionated elemental rare-gas pattern also remains a mystery. Frick and Chang [3] found that carbonaceous residues from two terrestrial cherts show nearly the same fractionation pattern (with respect to atmospheric composition). Elementally strongly-fractionated rare gases of distinct isotopic composition have also been observed in the carbon-rich phase from ureilites [4,5]. Thus, trapping of rare gases in carbon phases may be responsible for such elemental fractionations. But interpretation of elemental and isotopic signatures in terms of formation conditions (e.g. pressure, composition, degree of ionization) is not yet possible. Our approach is to study trapping of rare gases during condensation of grains from a hot gas produced in the laboratory.

The hot gas is produced by vaporizing a portion of a solid target by a laser pulse in an atmosphere of various mixtures of H2, N2, He and Ar [6]. The vapor jets away from the target surface into the ambient gas, cools by expansion and collision with the ambient gas, and then nucleates into droplets. Within milliseconds, grain growth is nearly finished and chains begin to form. These are collected by allowing the chains to settle onto Al foil, SEM buttons and TEM grids.

A TEM micrograph of a representative carbon condensate is shown in Figure 1. The condensate consists of chains of submicron particles, typical of all our condensates. The electron diffraction pattern for carbon condensate shows only diffuse rings, suggesting poorly crystalline grains. The diffraction pattern for pyroxene condensate produced in a H2-He-Ar atmosphere shows broad rings, indicating a glassy condensate, and a very weak pattern of Fe-Si alloy which is attributed to loss of oxygen during condensation.

Table 1 gives the rare-gas contents of condensates of C produced in different atmospheres, the carbon target itself, and a condensate of pyroxene in a H2-He-Ar atmosphere. Total gas pressure is one atmosphere, and in each case the mixture contains approximately equal amounts of each constituent. Sample weights are uncertain by a factor of two, due to an inadequate weighing procedure. Data are corrected for machine blanks and Al foil blanks. Carbon condensates (but not carbon target) were degassed at ~800C for several weeks prior to analysis. Analysis of two carbon condensates in atmospheres nominally void of rare gases was used as a baseline for comparison. The carbon 1 and 2 samples, condensed in a H2-He-Ar atmosphere, do not show any He detectable above the blank level of ~1 x 10^-8 cm^3 STP. But Ar is enriched by orders-of-magnitude in both these samples, with strikingly large concentrations (see Table 1).

All carbon samples contain significant amounts of Kr and Xe, regardless of the composition of the ambient atmosphere. All the samples in Table 1 give similar 86Kr/132Xe ratios, with an average value of 5.0 ± 0.9. This ratio is much lower than in air, consistent with an adsorbed atmospheric component. There are several reasonable sources for this component, and in particular, we note that the carbon target itself has about the same concentrations of Kr and Xe as the carbon condensates.

* Present address: Dept. of Terrestrial Magnetism, Washington, D. C. 20015

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This adsorbed component, however, is not responsible for Ar trapped in the carbon 1 and 2 samples. The $\frac{\text{Ar}}{\text{Xe}}$ ratio in carbon 2 is enhanced by a factor of 3 relative to atmosphere, whereas "blank" carbon samples and the carbon target have $\frac{\text{Ar}}{\text{Xe}}$ ratios well below atmospheric ratios, as expected for adsorption. Clearly, the $^{40}\text{Ar}$ in the carbon 1 and 2 samples is due to trapping of Ar during condensation and cooling of carbon grains in the Ar-rich atmosphere. Ar isotopic mass fractionation was not observed within an upper limit of 0.8%/amu.

Amounts of He and Ar in the pyroxene condensate are comparable to the blank, and thus only upper limits can be placed on the gas concentrations.

Conditions in our experiments are quite different from those that prevailed during formation of chondritic materials. In particular, the time scale for growth of grains in these experiments is probably many orders-of-magnitude shorter than in the solar nebula. Because of these differences, and in the absence of detailed understanding of rare-gas trapping in both cases, it is premature to interpret meteorite rare-gas abundances in terms of these results. Yet a few comparisons at this point are instructive. Laboratory produced carbon condensates contain little He, the lightest rare gas, while Ar, a heavy rare gas, is readily trapped. Planetary rare gases in meteorites also show depletion of He relative to Ar. We also found that carbon condensate traps orders-of-magnitude more Ar than pyroxene condensate. Although this single comparison does not rule out other mineral phases as equally good rare-gas trappers, it does lend support to the hypothesis of a carbonaceous rare-gas carrier in meteorites [2]. The Ar distribution coefficient (i.e. cm$^3$ STP Ar/g C/atm Ar) estimated for meteoritic carbonaceous residues is roughly $10^5$-$10^7$ times greater than in our carbon condensates. It is possible that this difference may be due to the vastly different rates of grain growth. Furthermore, the pressure dependence of our results has not yet been determined. Regardless of the difficulties in interpreting these preliminary results, this initial study demonstrates that this approach can offer new insights to questions concerning trapping of rare gases.

Acknowledgments: This research has been supported by NASA grants.

References:
Table 1: Rare-gas contents of carbon and pyroxene condensates.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Weights (µg)</th>
<th>$^{40}$Ar* ($\times 10^{-8}$ cm$^3$ STP)</th>
<th>$^{84}$Kr ($\times 10^{-12}$ cm$^3$ STP)</th>
<th>$^{132}$Xe ($\times 10^{-12}$ cm$^3$ STP)</th>
<th>$^{40}$Ar ($\times 10^{-6}$ cm$^3$ STP/g)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Carbon in $H_2$</td>
<td>40</td>
<td>&lt; 0.5</td>
<td>3.6</td>
<td>0.8</td>
<td></td>
</tr>
<tr>
<td>Carbon in $H_2$, $H_2$</td>
<td>40</td>
<td>&lt; 1.0</td>
<td>4.9</td>
<td>1.3</td>
<td></td>
</tr>
<tr>
<td>Carbon 1 in $H_2$, He, Ar</td>
<td>4 ± 3</td>
<td>28 ± 3</td>
<td>10.8</td>
<td>1.7</td>
<td>0.07 ± 0.05</td>
</tr>
<tr>
<td>Carbon 2 in $H_2$, He, Ar</td>
<td>40 ± 20</td>
<td>630 ± 30</td>
<td>24.3</td>
<td>5.4</td>
<td>0.16 ± 0.08</td>
</tr>
<tr>
<td>Carbon target</td>
<td>40 ± 20</td>
<td>13 ± 1</td>
<td>31.6</td>
<td>5.4</td>
<td>0.003 ± 0.002</td>
</tr>
<tr>
<td>Pyroxene in $H_2$, He, Ar</td>
<td>95 - 200</td>
<td>&lt; 2</td>
<td>3.8</td>
<td>0.8</td>
<td>&lt; 2 x 10^{-4}</td>
</tr>
</tbody>
</table>

* Absolute $^{40}$Ar amounts uncertain by an additional ± 10%.

Figure 1: TEM micrograph of carbon condensate produced in 1 atmosphere Ar. The chain-like structure is typical of all condensates. Carbon condensate produced in $H_2$ has a smaller grain size. Grains are spherical in the pyroxene condensate. Scale bar is 100nm.