LABORATORY-CREATED CHONDRITIC MATRIX: TEM STUDY OF NANOPHASE FE-SULFIDES AND MAGNETITE EMBEDDED IN FE-RICH AMORPHOUS SILICATES. N. M. Abreu[1] and J. A. Nuth III[2], [1]Penn State University, DuBois Campus (nma12@psu.edu), [2]Astrochemistry Laboratory – Goddard Space Flight Center.

Introduction: Two observational approaches are commonly applied to studying the solar nebula. The first is to directly study the materials that formed from primordial gas and dust (chondrites, chondritic IDPs, and comets). The second approach is to study young stellar objects (YSOs) and their disks. Recently, exceptionally pristine meteorites [e.g., 1,2,3] and cometary samples [e.g., 4] have become available. In these samples, amorphous silicates and nanocrystalline magnetite are common: Nanophase Fe-sulfides are ubiquitous. IR observations of YSOs suggest that nanophase Fe-sulfides were also important components of the solar nebula and of disks around young stars [e.g., 5].

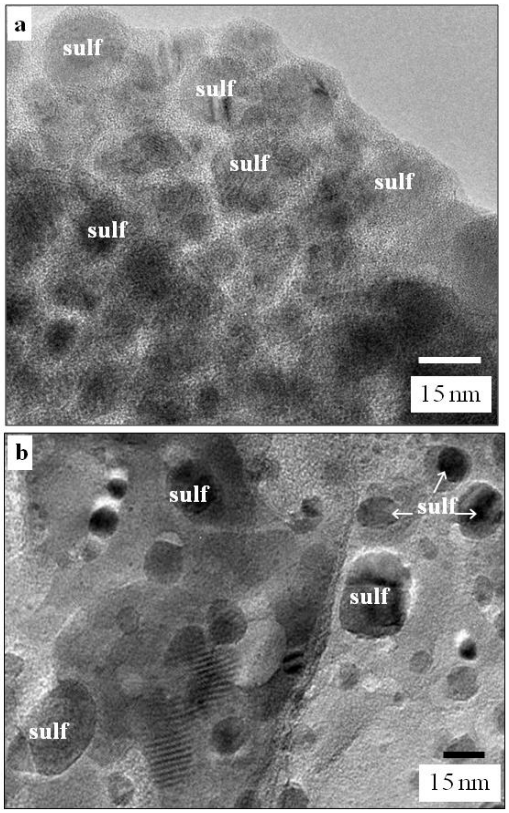

Fig. 1: TEM of typical region from (a) Run 1, showing Fe-sulfides (sulf) surrounded by amorphous silicates (b) rim in CR 3.0 QUE 99177, with Fe-sulfides in amorphous silicates.

We suggested multiple scenarios in which amorphous silicates and nanosulfides found in type 3.0 carbonaceous chondrites may have formed [3]. One of these mechanisms involves direct disequilibrium condensation, as a parcel of nebular material cools. We have tested this hypothesis using a dust generator and characterizing the resulting materials via high-resolution transmission electron microscopy (HRTEM) and EDX. Here we focus on comparing these samples to type 3.0 carbonaceous chondrites. However, some aspects can be extrapolated to other nebular materials.

Results: Samples were produced by vaporizing solid S in a furnace tube into which a H_2, Fe-carbonyl, silane, and O flame was generated. The furnace was operated at a range of temperatures (175-340°C) and the flame at two Fe:Si:O (50:20:40 and 50:20:20) ratios. The furnace gases cooled rapidly as they flowed into a room temperature chamber, condensed and were deposited as thin films and powders on an aluminum collector. Initial observations were performed by grinding the resulting materials and suspending them in isopropyl. TEM analysis of these samples was not successful at identifying any crystalline materials due to the large thickness of the particulates in suspension [6].

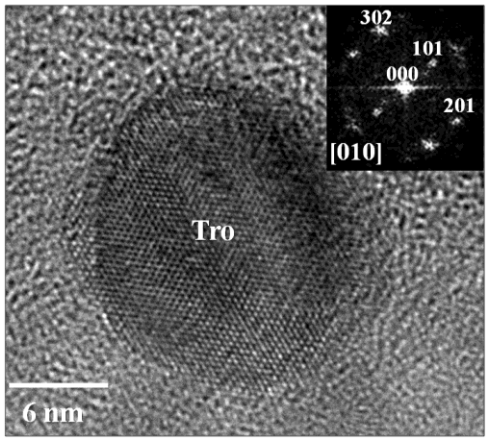

Fig. 2: HRTEM of sulfide (Tro) surrounded by amorphous silicate materials in Run 1. Inlaid Fourier transform (FFT) can be indexed as the [00$\bar{1}$] zone axis of distorted troilite.

We prepared two new samples from each of the same experimental products by mixing them with a C-based epoxy that cured at room temperature. This epoxy mixture was pressed between two pieces of Si, and ion-milled to electron transparency at low temperatures (< 260 K). TEM images show that the Si substrate is clearly distinguishable from the samples, as it is completely featureless. All samples consistently show abundant nanocrystalline phases embedded in amorphous Si-, O-, Fe-, S- phases whose compositions varied widely at the nm-scale. No crystalline silicates were identified. Due to the extremely fine-grained nature of these materials, we were rarely able to obtain single phase (stoichiometric) analyses from individual grains due to beam overlap with the surrounding amorphous material.

Run 1. Rounded, nanocrystalline (<20 nm diameter) Fe-sulfides and less abundant rounded to subrounded Fe-oxide crystals (40-60 nm diameter) were found embedded in amorphous silicates (**Fig. 1a**). The abundance of crystalline phases varies significantly at the micron scale. Fe-sulfides were identified as troilite and less commonly pyrrhotite based on their compositions and indexing of FFT patterns (**Fig. 2**). Rare, rounded, mottled Fe oxides have been identified as magnetite based on composition, characteristic 2.5 Å d-spacings, and indexing of FFT patterns (**Fig. 3**). Surrounding all crystals, there is amorphous materials, containing Si- (0-80 wt%), O- (0-30 wt%), Fe- (5-80 wt%) and S- (10-60 wt%).

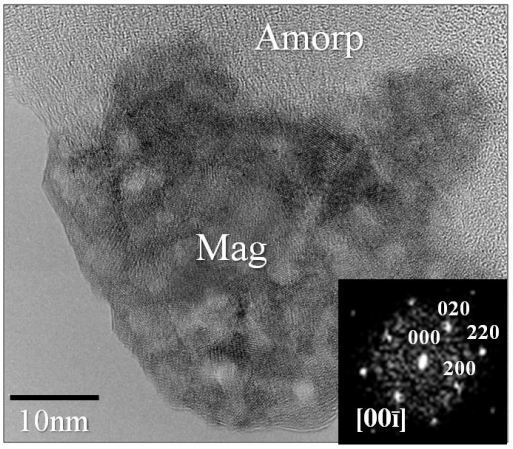

Fig. 3: HRTEM of iron oxide (Mag) embedded in amorphous (Amorp) silicate in Run 1. FFT can be indexed as the [00ī] zone axis of magnetite.

Run 2. Although this run produced materials that are texturally similar to those observed in Run 1, there are some significant differences. For example, no magnetite crystals were identified. This could be attributed to more reducing conditions (50% less O) in the dust generator flame in comparison with Run 1.

Discussion: We have succeeded in simultaneously preparing nanophase Fe-sulfides, amorphous silicates, and nano-magnetites under disequilibrium conditions by a single mechanism. Prior experiments have produced troilite and pyrrhotite directly from a nebular–like gas under disequilibrium conditions [7] and from Fe-S smokes [8]. Amorphous silicates have been commonly produced by the method outlined above [e.g. 9]. However, it was not clear if crystalline and amorphous phases could be produced by the same process or if the products from these experiments would resemble natural samples. Our results show that rapid cooling in a gas containing Si, O, Fe, and S results in the phases observed in the matrices of type 3.0 chondrites.

Texturally, samples from both experimental runs described above resemble the matrices of type 3.0 carbonaceous chondrites (**Fig. 1**). Nanocrystalline phases produced occur in similar relative abundances and sizes as they do in type 3.0 chondrites. Fe-sulfides are the dominant phase and rare, larger magnetite crystals may also occur [e.g. 1,2,3].

Flash heating and cooling events that may potentially volatilize and mix Si, O, Fe, and S are thought to be common in the solar nebula, namely chondrule forming events. If the matrices of pristine chondrites indeed formed by direct disequilibrium condensation of nebular gas, then it is possible that these materials are a by-product of chondrule formation as suggested by [1] and [3]. [3] argued that the residual gases from chondrule formation that eventually condense into chondritic matrices would be significantly enriched in Fe and S with respect to solar abundances.

Although these are very promising results, the composition, relative elemental fugacities, specific pressure and temperature under which our experimental samples were produced are different from those expected in nebular environments. The most important compositional departure from the natural sulfides is the absence of Ni, which is not used in our experiments due its extremely hazardous nature. Furthermore, the resulting amorphous silicate materials are generally much more S-rich than amorphous materials in chondrites [< 2 wt% - 3], due to the extreme S-fugacities in the dust generator chamber. Mineralogically, these experiments failed to reproduce the rare crystalline silicates observed in type 3.0 carbonaceous chondrites [1,2,3]. However, it is possible that these silicates may have formed by the subsequent annealing of amorphous silicates as suggested by [3].

Conclusion: We produced matrix-like materials by disequilibrium condensation of gases that may resemble the remnant gases produced by chondrule forming events. These experiments may also be relevant to the chemistry following the high velocity impacts of iron sulfide grains into aerogel.

References: [1] Brearley A. J. (1993) *GCA, 57*, 1521-1550. [2] Greshake A. (1997) *GCA, 61*, 437-452. [3] Abreu N. M. & Brearley A. J. *doi:10.1016/ j.gca.2009.11.009*. [4] Zolesky *et al.* (2006) *Science, 314*, 1735-1739. [5] Keller *et al.* (2002) *Nature, 417*, 148-150. [6] Abreu N. M. & Nuth J. A. (2009). [7] Stephen & Kothari (1978) *The Moon and Planets, 19*, 139-152. [8] Kaito & Nagata (1992) *Proc. NIPR Symp. Antarct. Meteorit., 5*, 318-326. [9] Nuth J. A. & Donn B. (1983) *Proc. 13th Lunar Planet. Sci. Conf.; J. Geophys. Res. 88*, A847-A852.

Acknowledgments: This work was funded by the DuBois Educational Foundation and American Astronomical Society and carried out at the Astrochemistry Lab (NASA) and Material Research Institute – Penn State University.