SULFUR-RICH CARBONACEOUS NANOGLOBULES IN THE TAGISH LAKE METEORITE

L.A.J. Garvie[1] and P.R. Buseck[1,2], [1]Department of Geological Sciences, Arizona State University, Tempe, Arizona 85287-1404, lgarvie@asu.edu, [2]Department of Chemistry and Biochemistry, Arizona State University, Tempe, Arizona, 85287

Introduction: Understanding the distribution, chemistry, and structure of C in carbonaceous chondrites (CC) is fundamental to revealing information on early solar chemical evolution. The Tagish Lake CC has generated much interest partly because of its pristine nature and high C content [1,2,3]. Unique to the Tagish Lake CC are hollow carbonaceous nanoglobules [4]. These particles are morphologically similar to materials produced by UV pyrolysis of interstellar ice analogues, and thus these hollow organic globules have been interpreted as extremely primitive organic materials that formed before or during the formation of the solar system [4].

Using electron energy-loss spectroscopy (EELS) we determined the composition of carbonaceous nanoglobules in Tagish Lake. This spectroscopic method together with transmission electron microscopy (TEM) provides unique chemical and crystal-chemical information at high spatial resolution, free of spectral interferences from surrounding materials.

Materials and Methods: A few crumb-sized pieces of pristine Tagish Lake were kindly donated by Sandra Pizzarello of ASU. The pristine material was from the unaltered interior, free of fusion crust collected 2 to 3 days after the fall. For comparison, we also examined carbonaceous materials from Murray, Mighei, and Murchison CM chondrites.

Samples were prepared by crushing a ca. 0.5 mm piece of sample on a clean glass slide and dry spreading the powder onto a Cu TEM grid coated with lacey-C. In this way, the samples were kept as pristine as possible. No water or organics were used in the sample preparation.

High-resolution transmission electron microscopy (HRTEM) was undertaken with a 002B Topcon operated at 200 keV with a LaB_6 filament. EELS spectra were acquired with a Gatan 766 DigiPEELS attached to a Philips 400ST operated at 100 keV with a cold field-emission gun. Spectra were acquired with an energy resolution of ca. 0.8 eV with operating currents of 1 to 2 nA, from regions ca. 10 nm in diameter.

Results: Carbonaceous material was recognized by its large C K edge and distinctive spectral shape. We observed three morphologically distinct carbonaceous materials: flakes, rounded particles with a hollow core, and rounded single and agglomerated particles without hollow cores.

The carbonaceous nanoglobules are typically <200 nm in diameter (fig. 1a), although larger clusters occur (fig. 1b). Several of the spheres exhibit a central region with lower brightness in the TEM images (fig. 1a), consistent with previous studies and indicative of a hollow core [4]. The spherical hollow nanoglobules are relatively rare compared to the solid ones. A few carbonaceous flakes were found.

A total of 34 nanoglobules were analyzed by EELS, and all are rich in S (fig. 2), with lesser amounts of N and O. No other elements were detected.

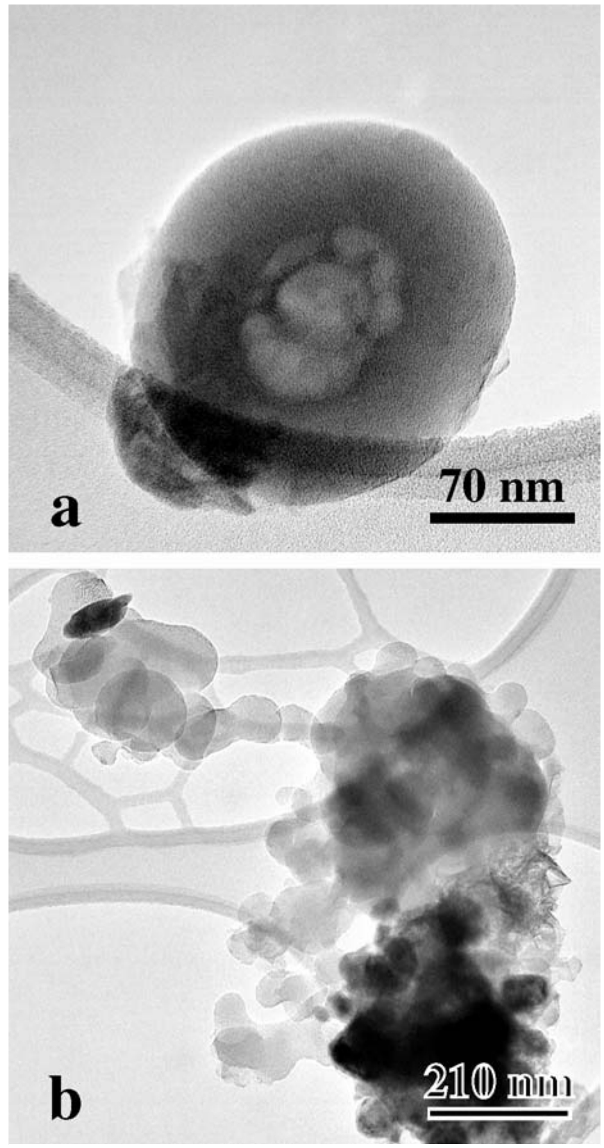

Fig. 1. a) Single carbonaceous nanoglobule. b) Cluster of rounded carbonaceous particles. The dark material at the bottom right of b) consists of magnetite and phyllosilicates. Spider-like fibers belong to the support substrate.

The C to S ratios range from 8:1 to 23:1. N and O are low, with C:N and C:O ratios less than 12:1 and 14:1, respectively. A typical S-rich nanoglobule has the composition $CS_{0.125}N_{0.08}O_{0.07}$. In contrast, the C flakes are free of S but contain N and O. A typical flake has the composition $CN_{0.002}O_{0.001}$.

All C K edge spectra exhibit similar shapes, with a sharp rise in intensity and a maximum at 285 eV (fig.

2). This peak is indicative of π bonding, whereas the broad intense peak centered around 300 eV reflects C-C σ bonding [5]. All C K edges of the nanoglobules are similar to each other. There are few spectral differences between the C K edges from the S-rich and S-poor nanoglobules. The shape of the C K edge from the nanoglobules are consistent with a lack of long-range order. The C K edge of the C flakes are similar in shape to that of graphite, although the spectral features are less distinct, reflecting some loss of long-range order.

Further information on the nanoglobule structure is evident from the HRTEM images. Three types of features were observed (fig. 3): amorphous C without lattice fringes, C with nanometer-sized domains with fringe spacings of 3.4 Å indicative of poorly-graphitized C, and a streaked structure. Of the 34 particles investigated, only three revealed the streaking. The individual streaks have widths of ca. 10Å and they are surrounded by regions free of streaking, i.e., it is not an artifact.

Although not shown, we found nanoglobules in all the CM chondrites investigated. They differed however from the Tagish Lake material in being free of S and N and lacking a hollow core.

Insoluble S-rich carbonaceous materials have not been observed in CCs, although soluble sulphonic acids were detected in the Tagish Lake meteorite [2]. Soluble and insoluble C_mS_n compounds can be produced by photopolymerization and radiolysis of CS_2, which is common in both solar and extra-solar environments [6,7]. It has been suggested that radiolysis and photolysis of CS_2 contribute C-S compounds to the circumstellar and interstellar media as well as to comets and atmospheres of the giant planets [6]. The presence of S-bearing carbonaceous nanoglobules in Tagish Lake adds a new dimension to this intriguing meteorite and provides support for the possible presolar origin of the nanoglobules. It also raises the possibility of cometary material as a potential source.

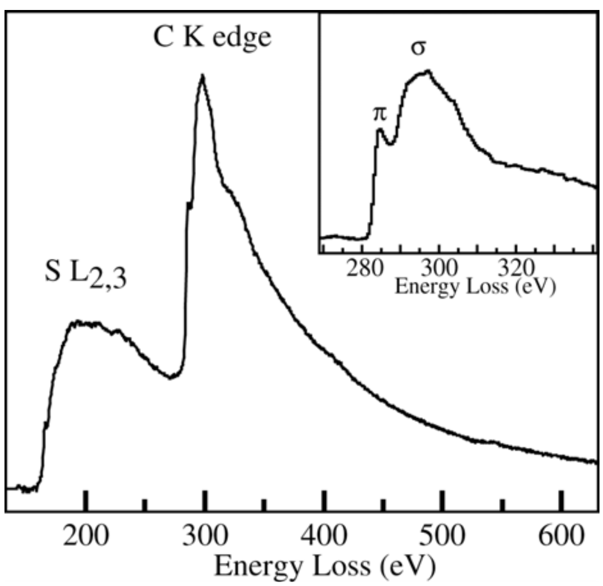

Fig. 2. EELS spectrum of a spherical nanoglobule. The background has been subtracted from beneath the S $L_{2,3}$ edge. The inset shows the C K edge at higher energy resolution.

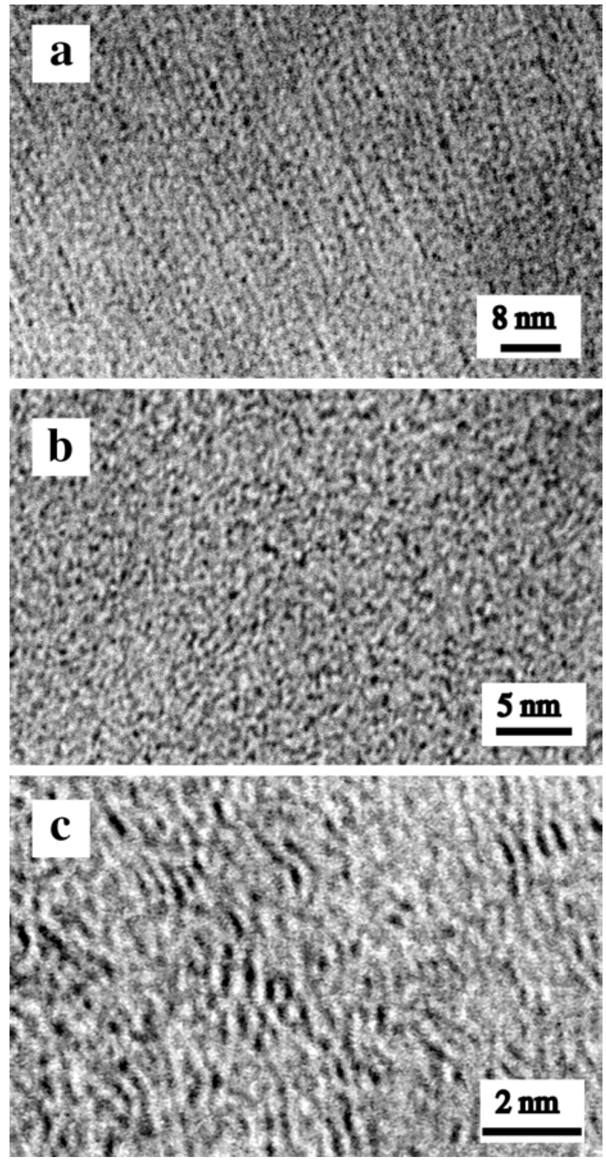

Fig. 3. images of nanoglobules. a) streaked texture, b) amorphous C, c) poorly-graphitized C.

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