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Carbon XANES analyses of Q-gas rich carbonaceous fractions obtained by freeze-thaw separation of Allende CV3 chondrite

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Introduction:

While Q gases, the planetary noble gases, from different chondritic meteorites have been elementally and isotopically studied, the carrier of Q gases, phase Q, is yet to be identified. Amari et al. (2003) [1] analyzed Q gases in the density-separated fractions of a floating fraction from the Allende CV3 meteorite which is similar to HCl-HF resistant residues regarding to noble gas compositions but obtained by the freeze-thaw disaggregation. They revealed that half of Q gases in the parent floating fraction was concentrated in the fraction with the density of ~1.65 g/cm3 (C1 -8D). In this study, we apply X-ray absorption near edge structure (XANES) spectroscopy using the scanning transmission X-ray microscope (STXM) that is an appropriate technique to probe sensitively the functional groups at high spatial resolution, in order to focus on the quantitative molecular characterization of the fraction C1-8D, Q-rich carbonaceous matter from Allende.

Experimental:

A floating fraction was recovered by the freeze-thaw disaggregation of the Allende and was separated into 9 density fractions by density separation after the colloidal separation. In this study, seven of the density fractions including C1-8D, a colloidal fraction, and an acid insoluble carbonaceous residue for comparison, were used. The samples were embedded in epoxy and were sectioned into 100-140 nm thickness using a diamond knife and an ultramicrotome. C-XANES of these samples was conducted using STXM at Beam line 5.3.2, at the Advanced Light Source, Lawrence Berkeley National Laboratory.

Results and Discussion:

In the C-XANES spectrum of the most Q gas-rich fraction C1-8D, the peak intensity of aromatic carbon is significantly lower than that of acid insoluble residue. The peak of 1s-sigma* exciton derived from highly conjugated sp2 carbon is characteristic in thermally metamorphosed chondritic organics [2], but is not seen in C1-8D. Moreover, two large peaks of carbon 1s-sigma* transitions that are derived from C-C, C-O, or C-F and C-C were originally detected in C1-8D at the energies exceeding the C 1s ionization threshold, ~292 and ~295.7 eV. This spectral pattern was clearly distinctive from that of the acid insoluble residue. The C-XANES spectrum of C1-8D is also distinctive from those of graphite, fullerene, carbon nanotube, and glassy carbon all of which have the developed peaks of 1s-sigma* exciton. The spectrum of C1-8D is also distinctive from that of pure diamond. When the C-XANES spectrum of C1-8D is compared to those of the other density fractions, the spectra show several clear differences, e.g., the lowest peak intensity of aromatic carbon and the most developed peak at 295.7 eV in C1-8D. These results indicate that there exists the electronic structural differences between C1-8D and the other density fractions. Therefore, concentration of Q gas may be related to kinds or abundances of specific chemical bondings in its carrier material. The result that only C1-8D is distinct from the other density fractions in the spectral characteristics while the others fractions are similar appears to be

consistent with the micro-Raman spectroscopic behaviors of these fractions [3]. They reported that only C1-8D has the lower ID/IG ratios while all other fractions have similar ID/IG. In general, the ID/IG is proportional to the sp2/sp3 carbon [4], and thus C1-8D may be rich in sp3 carbon. This is consistent with this study that aromatic carbon is less abundant and carbons associated with 1s-sigma* transition are dominant in C1-8D. Consequently, Q gas could be concentrated in sp 3 carbon-rich material.

References: [1] Amari S. et al. (2003) GCA 67, 4665. [2] Cody et al. (2008) EPSL 272, 446. [3] Matsuda J. et al. (2009) Geochem. J. 43, 323. [4] Ferrari A. C. and Robertson J. (2000) Phys. Rev. B61, 14095

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