

Allende 隕石から分離した富 Q ガス、貧 Q ガス炭素物質の X 線吸収分光学的比較 Carbon-XANES spectroscopic comparison of Q-gas rich and depleted fractions from Allende meteorite

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Introduction:

Although the planetary noble gases enriched in the heavy noble gases, Q gases, have been frequently studied, phase Q itself is yet to be identified. Matsuda et al. (2010) has reported that TEM observations did not show clear difference between Q-rich- and depleted- carbonaceous materials, while the Raman spectroscopic differences were observed between the two. They concluded that release of Q gases is not accompanied by mass loss but resulted from rearrangement of carbon structure during removal of Q (oxidation). Our previous study has revealed that the physically separated Q-rich density fraction may be rich in diamond-related sp^3 carbon. Thus, the release of Q gases and/or phase Q could be related to carbon functional group chemistry rather than carbonaceous morphology. This study focuses on the XANES analyses of Q-rich- and depleted- materials from Allende meteorite to refine carbon chemistry that is likely associated to the release of Q gases. Moreover, Q-rich fractions obtained via the suspension and physical separation from Allende, respectively, are analyzed for comparison.

Experimental:

An Allende meteorite was treated with HF-HCl for preparing an acid resistant carbonaceous residue. During the removal of elemental S from the residue with CS₂, One-seventh of the total fraction of the residue suspended in the supernatant was recovered (AMD3). The rest of the fraction (AMD1), which is rich in Q gases, was further treated with Na₂Cr₂O₇ to remove Q gases, yielding the oxidized residue, AMD2. Apart from AMD samples, a Q-rich floating fraction (G1), was obtained by the freeze-thaw disaggregation of Allende meteorite in stainless beaker. All the samples were embedded in sulfur and ultramicrotomed. Carbon-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 C-XANES spectra of AMD1 and 2, aromatic and aliphatic carbons are much lower in AMD2 than those in AMD1. Carbonyl carbon is slightly higher in AMD2 than that in AMD1. On the other hand, the peak intensities of 1s- σ^* exciton derived from graphene, are not changed between AMD1 and 2. The differences in molecular features between AMD1 and 2 may not be directly reflected by phase Q, but more likely indicate the changes in major organic macromolecule by oxidation. Nonetheless, the depletion of aliphatic and production of carbonyls probably influence the structural rearrangement related with the release of Q-gases. No quantitative change of 1s- σ^* exciton intensity before and after oxidation indicates that phase Q is unlikely related to graphene.

Only one clear difference between C-XANES spectra of AMD1 and 3 is a peak at 287.43 eV in AMD 3 is remarkably developed compared to AMD1. A peak around at this energy is generally assigned to aliphatic carbon, or alternatively, another sp^3 carbon such as -C-CF of fluorinated diamond (Yu et al. 2003). The peak may have an important relation to Q-richness, since noble gas concentrations in AMD3 are 2-4 times higher than those of AMD1 (Amari et al. 2003).

G1 showed a quite different C-XANES spectrum from those of AMD1 and 3. While aromatic carbon is low, two unidentified peaks are detected. One of the two peaks is possibly derived from diamonds or their related compounds. Such spectral features are very similar to those in the Q-rich density fraction. Additionally, C-XANES spectra of different regions of G1 are heterogeneous. These spectra are not exactly the same as those of individual density fractions in our previous study. It may be because the individual density fractions show more detailed spectral features that were covered in bulk fractions, or might have been partially contaminated with Teflon beaker used in the past treatment by Matsuda et al. (1999). To summarize, it is further suggested that some sp^3 carbon may be related to the release of Q gases and/or phase Q.

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