Identification of the noble gas carrier Q in meteorites: Raman spectroscopic study

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Noble gases are very good tracers to examine the origin and the evolution of our solar system. As the noble gases are chemically inert, it is interesting to examine the chemical state where noble gases are trapped in meteorite. For such a purpose, Lewis et al. (1975) dissolved the Allende meteorite with HCl/HF. They obtained about 0.5% residue (chemical residue) where almost all noble gases are present. They dissolved this chemical residue with red-fuming HNO_3, and got about 0.46-0.48% of the bulk Allende (oxidized residue) and found that almost all heavy noble gases with normal isotopic ratios were disappeared during this process. The isotopic ratios of Xe in the oxidized residue are isotopically different from those in the normal solar system, leading to the subsequent major research field of presolar grains. The research of presolar grains is very interesting, but another important discovery in this paper is that normal component of primordial heavy noble gases are in the very tiny portion (0.02-0.04%) of the bulk Allende. They could not identify this fraction and named it as 'Q' from the word 'quintessense'. The subsequent study shows that Q is a carbonaceous material, but we do not know the precise crystal structure of Q.

Raman spectroscopy was particularly effective to examine the crystallinity of graphite and its metamorphic grade. Thus we made the Raman spectroscopic measurement of the chemical residue and the oxidized residue of Allende that were newly prepared. The amount of 4.9154g of the Allende bulk was dissolved with 10MHF+1MHCl and 6MHCl alternatively at the room temperature. We repeated several cycles of this procedure until the yellow color of the supernatant vanished. Finally we washed the obtained residue with acetone and CS_2 to dissolve out the elemental sulfur. Through this procedure we obtained the colloidal and non-colloidal fraction. This non-colloidal fraction is AMD1. We further treated AMD1 with 0.5N $Na_2Cr_20_7$+2N H_2SO_4 for 10 hours, and obtained the oxidized residue AMD2. This is the common procedure to obtain the residue and the oxidized residue.

We also made measurement of several fractions of C1-8 that were obtained by colloidal and density separation in Amari et al. (2003). Matsuda et al. (1999) firstly found that the black material that floated on the water surface during the freeze-thaw method was similar to the chemical residue. Amari et al. (2003) further applied the colloidal and density separation to this 'floating fraction' C1-8 and found that half of Q in this fraction was concentrated in the fraction C1-8D with the density of $1.65\pm0.04 g/cm^3$. We made measurement of Raman spectroscopy of C1-8D ($1.65\pm0.04 g/cm^3$ in density) and also other different density fractions; C1-8J ($1.1-1.6 g/cm^3$), C1-8G ($1.97\pm0.06 g/cm^3$) and C1-8K ($2.2-2.3 g/cm^3$).

Raman spectra were obtained with a Raman microscope (Kaiser HoloLab 5000 of Kaiser Optical Systems Inc.) in Tokyo Medical and Dental University using 532nm YAG laser (5-6mW at the sample surface). We performed measurements of five to eight spots in each sample. The typical Raman spectrum of graphitic carbon (G band of about $1580 cm^{-1}$ and D band of about $1350 cm^{-1}$) was obtained in our all samples, which is common in the carbon material in carbonaceous meteorites. The peak positions, intensity and FWHM (full-width at half-maximum) of G and D bands were obtained by applying the Lorentzian fitting curve for individual peaks.

We compared these Raman spectroscopic features of the acid residue (containing Q), oxidized residue (not containing Q) and the density separated floating fractions (being enriched and depleted in Q) of the Allende meteorite. As a result, we have identified the Raman feature of the phase Q, and concluded that Q is the graphitic carbon with the crystal size of about 50-60 angstrom.