ラマンスペクトルを基にした反応速度論から探る隕石有機物の熱履歴
Kinetic approaches for the thermal history of organic matter in meteorites based on Raman spectra

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CI, CM, and CR chondrites experienced aqueous alteration in their parent bodies, and contain organic carbon up to a few wt%. More than 70 wt% of it exists in the form of insoluble organic matter (IOM) (e.g., Pizzarello et al., 2006). Organic matter (OM) is susceptible to heating, and changes their structures, reflecting aqueous alteration and/or thermal metamorphisms. Thus, it attracts attention as the index of the parent body processes.

Kebukawa et al. (2010) estimated kinetic parameters for thermal degradation of aliphatic C-H through in-situ heating experiments of IOM and bulk of the Murchison meteorite (CM2) using the Fourier transform infrared micro-spectroscopy (microFTIR) combined with heating stage. They revealed that the peak intensity of aliphatic C-H is a good index of low-temperature and/or short-duration heating in the parent bodies. However, microFTIR is not enough for estimating thermal process of OM under high-temperature and long-duration heating which IR signals of aliphatic C-H are unlikely to be detected. Raman spectroscopy is a complementary tool to IR spectroscopy, since their spectral features of OM in the chondrites reflect thermal changes in aromatic structure in the complex OM (Quirico et al. 2003; Bonal et al., 2006, 2007; Busemann et al., 2007). When chondritic OM is heated, its aromatic structure will be more ordered to graphitic structure. This process is an irreversible, and results in changes of disordered (D_1-: ~1350 cm^{-1}) and graphite bands (G: ~1590 cm^{-1}) in Ramn spectra. Therefore, D_1- and G-bands were often used as indicators of degree of thermal metamorphism, but understanding of kinetic behaviors of the Raman spectral parameters is still limited. Thus, in order to evaluate thermal history of OM in chondrites under high-temperature and/or long-duration heating, we performed a series of heating experiments of the Murchison meteorite powders and kinetic analyses of their Raman spectral parameters.

The Murchison meteorite powders were heated under low oxygen condition in a vacuum furnace for 3-48 h at 600-900°C. The low-oxygen condition was maintained by a gas mixture of N_2 and H_2 (99:1, v/v). The samples after heating were analyzed using Raman spectrometer with a 532 nm laser (RAMANtouch; Nanophoton). The Raman spectra of all the samples show D_1- and G-bands. We determined 5 Raman spectral parameters: the full width at half maximum (FWHM) and the peak positions of D_1- and G-bands, and the ratio of the peak intensity of D_1- to G-band. The changes in these Raman parameters with time at each temperature will be fitted with algebraic kinetic equations deduced by possible physical mechanisms, and estimate reaction rate constant. Then the apparent activation energies and frequency factors will be calculated by the reaction rates using the Arrhenius equation. Finally, we will establish the Raman parameters as functions of heating temperature and time. Combining previously reported kinetics of aliphatic C-H (Kebukawa et al. 2010), it may allow us to calculate precise time and temperature of thermal processes for chondrites.

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