

## KINETIC ESTIMATION OF THE THERMAL HISTORY OF ORGANIC MATTER IN CHONDRITES USING RAMAN SPECTROSCOPY.

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One of the characteristics represent CI, CM, and CR chondrites is to contain organic carbon up to a few wt%. Organic matter (OM) in chondrites above 70 wt% exists in the form of insoluble organic matter (IOM), which is complex and macromolecular compounds [e.g., 1]. IOM consists of carbon skeletons as polycyclic aromatic hydrocarbons (PAHs) with aliphatic side chains, carboxyl, hydroxyl, and carbonyl groups [2]. OM is a good index of the parent body processes because the final molecular structures of OM in chondrites depend on the degrees of the parent body processes. OM changes its structure reflecting aqueous alteration and/or thermal metamorphism. In this study, we utilized the property of OM which is susceptible to heating in order to estimate the thermal history of chondrites.

Kinetic approaches are an effective way to predict the thermal history of OM in chondrites and its parent bodies [3,4]. Kebukawa et al. [4] obtained the kinetics of degradation of aliphatic C-H through in-situ heating experiments of bulk of the Murchison meteorite and IOM using the Fourier transform infrared micro-spectroscopy (microFTIR). According to them, low-temperature and/or short-duration heating in chondrites' parent bodies can be traced by the peak intensity of aliphatic C-H. However, this method is insufficient under high-temperature and/or long-duration heating, since IR signals of aliphatic C-H are unlikely to be detected.

To overcome the problem, we applied Raman spectroscopy, which is a complementary tool to IR spectroscopy. Raman spectral features of OM in the chondrites reflect thermal changes not in aliphatic C-H but in aromatic structure in the complex OM ([5-8]). While heating, chondritic OM changes into more ordered graphitic structure. The irreversible process appears as the alteration in disordered (D<sub>1</sub>: ~1350 cm<sup>-1</sup>) and graphite (G: ~1590 cm<sup>-1</sup>) bands of Raman spectra. That is, Raman spectroscopy is a promising tool for assessing thermal processes of OM in the chondrites under high-temperature and/or long-duration heating. Although D<sub>1</sub>- and G-bands could be used as indicators of degree of thermal metamorphism, researches in kinetics using the two Raman spectral parameters are relatively limited. We performed a series of heating experiments of the Murchison meteorite powders with relatively higher temperature and longer duration compared to Kebukawa et al. [4], to analyze Raman spectral parameters for kinetic estimation of the thermal history of OM in chondrites.

We heated the Murchison meteorite powders under low-oxygen condition for 3-48 h at 600-900°C in a vacuum gas replacement furnace. The low-oxygen condition was certainly sustained by purging a gas mixture of N<sub>2</sub> and H<sub>2</sub> (99:1, v/v). After heating, the samples were analyzed using a Raman spectrometer with a 532 nm laser (RAMANtouch; Nanophoton). D<sub>1</sub>- and G-bands were obtained in the Raman spectra of all the samples, and we determined 5 Raman spectral parameters: the full width at half maximum (FWHM) and the peak positions of D<sub>1</sub>- and G-bands, and the ratio of the peak intensities of D<sub>1</sub>- to G-band. These Raman spectral parameters variously changed with time at each temperature. We will fit these data with algebraic kinetic equations deduced by possible physical mechanisms, and estimate reaction rate constants. Then, based on the Arrhenius equation, the apparent activation energies and frequency factors will be calculated by the reaction rates. In the end, we will be able to use the Raman spectral parameters as functions of heating temperature and time. Combining kinetics of aliphatic C-H previously reported in Kebukawa et al. [3], it may enable us to calculate time and temperature of thermal processes in chondrites' parent bodies.

**References:** [1] Pizzarello S. et al. (2006) In *Meteorites and the early solar system II* edited by Lauretta D. S., Leshin L. A., and McSween H. Y. Jr. Tucson, AZ: The university of Arizona Press. pp. 625–651. [2] Hayatsu R. et al. (1977) *Geochimica et Cosmochimica Acta* 41:1325–1339. [3] Cody G. D. et al. (2008) *Earth and Planetary Science Letters* 272: 446-455. [4] Kebukawa Y. et al. (2010) *Meteoritics and Planetary Science* 45:99–113. [4] Quirico E. (2003) *Meteoritics and Planetary Science* 38:795–811. [5] Bonal L. et al. (2006) *Geochimica et Cosmochimica Acta* 70:1849–1863. [6] Bonal L. et al. (2007) *Geochimica et Cosmochimica Acta* 71:1605–1623. [7] Busemann H. et al. (2007) *Meteoritics and Planetary Science* 42:1387–1416.