

Kinetics of Thermal Decomposition of Organic Matter from Carbonaceous Chondrites and Influence of Minerals

Yoko Kebukawa (癸生川陽子)

Organic matter in meteorites consists of various forms of organic matter including mainly complex macromolecular materials called insoluble organic matter (IOM), and contains records of their parent body processes. Studying organic matter in meteorites is the key to understand how the solar system has evolved and the processes associated with this. In this thesis research, I first aim to elucidate spatial relationship of organic matter and inorganic phases, then effects of minerals on thermal transformation of organic matter. Finally, I evaluate thermal processing on chondrite parent bodies by kinetic studies of chondritic organic matter using Fourier transform infrared (FTIR) spectroscopy, which is a non-destructive technique for organic components and minerals.

First I checked organic contamination of carbonaceous chondrites for FTIR microspectroscopic analyses. Organic contamination was found on some carbonaceous chondrites containing abundant hydrous minerals by FTIR microspectroscopy on the samples pressed on Al plates. This contamination occurred within one day of storage, when the samples pressed on Al were stored within containers including silicone rubber mats, silicone grease or adhesive tape. Samples stored in glass and polystyrene containers did not show any contamination. Therefore, meteorite samples prepared for micro FTIR measurements should be stored in glass or polystyrene containers without silicone and sticky materials.

Second, I conducted mapping of chondrite by high resolution infrared imaging technique using near-field infrared (NFIR) microspectroscopy for microscopic and spatial characterization of chondritic organic matter. The NFIR spectroscopy has recently been developed to enable infrared spectral mapping beyond the optical diffraction limit of conventional FTIR microspectroscopy. NFIR spectral mapping measurements were conducted on the Bells (CM2) carbonaceous chondrite 300 nm thick sections on Al plates for several μm^2 areas. Heterogeneous distributions of organic functional groups as well as those of inorganic phase such as silicates (Si-O) were observed with less than 1 μm spatial resolution. Some slices of Bells showed ~ 1 μm aliphatic C-H rich areas which were considered to represent the organic rich areas. One of the organic rich portions in a slice of Bells may contain C-O bonds. Some of organic rich regions might correspond to organic globules which are abundantly distributed in the Bells meteorite. The NFIR imaging method can provide submicron

spatial distribution of organic functional groups and their association with minerals.

Third, I conducted heating experiments of IOM and bulk of Murchison (CM2) and Orgueil (CI1) meteorites to elucidate effects of mineral phases on thermal stability of organic matter under micro FTIR spectroscopy with a heating stage in an inert gas flow. Thermal stability of organic matter was lower in the presence of minerals than IOM for Murchison, while thermal stability was higher in the presence of minerals than IOM for Orgueil. Similarly, heating experiments were conducted using leonardite humic acid (LHA) with and without saponite, antigorite and olivine as analogs of carbonaceous chondrites, under Ar flow and air. The thermal stability of LHA is higher with saponite. On the other hand, antigorite and olivine accelerate the decrease of aliphatic fractions, and decelerates the decrease of aromatic fractions. These effects were dominant in oxidizing atmosphere. These results suggest that thermal stability of organic matter depend on coexisting minerals.

Finally, I conducted heating experiments of bulk and IOM of Murchison to estimate kinetic parameters using infrared bands. Samples are heated at 160-300°C isothermally in the heating stage under Ar gas flow and air for several hours. Decreases of aliphatic C–H peak heights in Ar flow are well fitted with Ginstling-Brounshtein three dimensional diffusion model, and the rate constants for decreases of aliphatic C–H were determined. Activation energies and frequency factors are estimated from these rate constants at different temperatures using Arrhenius equation. The activation energy and frequency factor for bulk Murchison under Ar flow are 72 ± 6 kJ/mol and 48 s^{-1} , respectively, while these for Murchison IOM under Ar flow are 107 ± 3 kJ/mol and $5.2 \times 10^4 \text{ s}^{-1}$, respectively. Activation energy values of aliphatic C–H decrease are larger for IOM than bulk of Murchison. Hence, the mineral assemblage of Murchison meteorite might have catalytic effect for the organic matter decomposition.

Kinetic heating experiments allow us to constrain the time and temperature range of thermal changes of organic matter in carbonaceous chondrites by using obtained kinetic parameters such as rates and activation energies. For example, aliphatic C–H decrease by half is within ~ 4 months and to zero within ~ 3 years at 100°C in the presence of Murchison mineral assemblage. Without minerals, aliphatic C–H decrease by half is within ~ 20 years and aliphatic C–H is lost within ~ 200 years at 100°C. Enhanced and additional kinetic studies of thermal processing of organic matter may further help sorting out the thermal history of the parent bodies. In addition, spatial distribution of organic matter with minerals observed by NFIR spectroscopy may help understanding associations and roles of minerals on the parent body processes.