Evaluation of Raman Parameters of Organic Matter: The Comparative Study for Thermal Records in Carbonaceous Chondrites.

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Introduction

Raman spectrometry is a non-destructive, effective tool to assess the thermal records of carbonaceous materials. Many carbonaceous chondrites show the two first-order Raman bands of polyaromatic carbon materials, namely the D ("disordered") and G ("graphite") bands (e.g.,[1]). The D band is characteristic of a disordered carbon, whereas the G band is characteristic of an aromatic carbon (graphite type or polyaromatic solid). It has been shown previously that D and G bands (at ~1,350 and ~ 1,600 cm^{-1}, respectively) in Raman spectra of organic matter in chondrite matrices vary with thermal metamorphic recrystallization of the host meteorite. The full width, half-maximum (FWHM) of the D band decreases, and the intensity ratio I_D/I_G increases with thermal maturity of the organic matter (e.g., [2]-[4]). However, it is also suggested that the absolute values of Raman parameters vary depending on the experimental conditions, including the laser excitation wavelength, sample conditions, and fitting procedures [3]. In this study, we examined the Raman characteristics of primitive carbonaceous chondrites with variable aqueous alteration and thermal metamorphism and compared the Raman parameters to the values reported in previous studies.

Samples and methods

Five carbonaceous chondrites, Murchison (CM2), Tagish Lake (C2-ungr), ALH(ALHA)77307 (CO3.0), Efremovka (CVred), and Allende (CVox), were examined in this study. Raman spectra of randomly selected matrix areas on polished thin sections were collected using a JASCO NRS-1000 Raman microspectrometer at NIPR equipped using 532 nm excitation. Measurements are carried out in similar settings described in [2]. Spectra were acquired from polished thin sections under atmospheric conditions using an incident power of 11 mW in the spectral region 1000–1800 cm^{-1}. The first-order carbon bands were fitted with two components, a Lorentzian profile for D band, and Breit-Wigner-Fano (BWF) profile for the G band, with Origin 2020b software.

Results and discussion

Raman parameters of the observed D and G bands of carbon in the matrix of the carbonaceous chondrites (the coefficient of determination R^2>0.90) are shown in Fig.1. We find the Raman spectra of two aqueously altered carbonaceous samples Tagish Lake and Murchison show variable intensities of fluorescence background, which results in a fitting error (low value of R^2) for some cases. Raman parameters obtained from all samples in this study except Efremovka show a large scatter. Wide D-bands in Tagish Lake, Murchison, and ALH77307 are characteristic of highly disordered carbonaceous materials, and the decrease in the bandwidth of the D-bands and the increase of I_D/I_G ratios due to thermal metamorphism are observed in Efremovka and Allende.

Parameters from previous studies of (i) raw matrix grains of Tagish Lake, ALH77307, Efremovka, and Allende ([4]-[6]; 514 nm laser excitation wavelength) and (ii) insoluble organic matter (IOM) grains extracted from Murchison, Tagish Lake, ALH77307, and Allende ([3]; 532 nm laser excitation wavelength) are also plotted in Fig.1. The I_D/I_G ratios that we determined from matrix areas of polished thin sections of Tagish Lake and Efremovka are higher than the values determined from raw matrix grains ([4],[6]). On the other hand, both FWHM-D and I_D/I_G of Murchison are lower than the measurements from extracted IOM ([3]). The observed discrepancies may be due in part to the difference in band fitting method; Busemann's study uses Lorentzian profiles for both D and G bands, whereas Quirico and Bonal's studies use Lorentzian profile for D band and BWF profile for G band, as discussed by [3]. The discrepancy between Raman measurements from a thin section and from extracted IOM of Murchison (Fig. 1) may be due in part to the brecciated nature of Murchison [7]. Our thin section analyses of Raman parameters in ALH77307 and Allende overlap with previous analyses of raw matrix grains and extracted IOM and our values for Tagish Lake overlap with previous analyses of Tagish Lake IOM (Fig. 1).

In summary, our analyses of Raman parameters of FWHM-D and I_D/I_G in this study mostly overlap with the reported values in the studies of raw matrix grains and extracted IOM, though there are some discrepancies. In spite of the discrepancies, the metamorphic trends of D and G bands observed in this study are the same as determined from previous studies. Tagish Lake, Murchison and ALH77307 all have high FWHM-D and/or low I_D/I_G indicating relatively minor thermal metamorphism; FWHM-D decreases, and in our analyses I_D/I_G increases in Efremovka, indicating higher metamorphic grade; and the low

FWHM-D and high I_D/I_G detected in all analyses of Allende indicate still higher thermal maturity (Fig. 1). This trend is a good agreement with petrologic observations (e.g., [8]). More detailed examination for the Raman parameters focusing on the aqueously altered (C2) chondrites are in progress.

References

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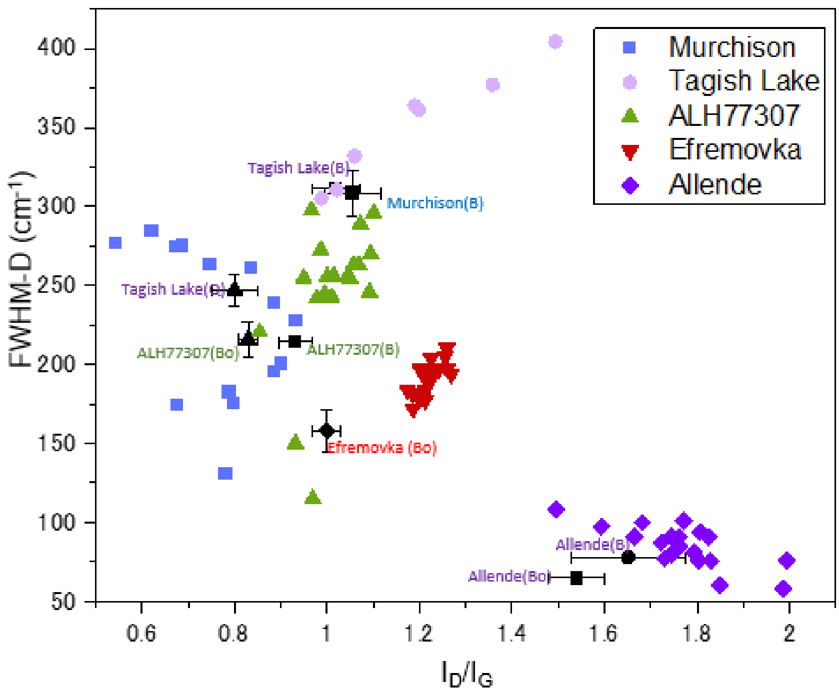

Fig.1. Raman parameters of carbonaceous chondrites in this study. Analyses shown in black are the averaged parameters with 1σ standard deviation from raw matrix grains (Tagish Lake (Q), ALH77307 (Bo), Efremovka (Bo), and Allende (Bo), ref. [4]-[6]) and extracted IOM (Tagish Lake (B), Murchison (B), ALH770307(B), and Allende (B), ref.[3]).