ANALYZING THE THERMAL METAMORPHISM PROCESS OF CARBONACEOUS CHONDRITES USING RAMAN SPECTROSCOPY. A.G. Dall'Asén[1], R. Kayastha[1], A.R. Stokke[1], R. Paul[1], B.C. Bromley[2], and S.J. Kenyon[3]. [1]Department of Physics and Astronomy, Minnesota State University-Mankato, Mankato, MN 56001, USA. E-mail: analia.dallasen@mnsu.edu. [2]Department of Physics and Astronomy, University of Utah, 115 South 1500 East, Salt Lake City, UT 84112, USA. [3]Smithsonian Astrophysical Observatory, 60 Garden St, Cambridge, MA 02138, USA.

Introduction: Carbonaceous chondritic meteorites are some of the most primitive materials in our solar system. They did not experience melting or other processes on their parent bodies (e.g. asteroids and comets) during their initial formation, and thus, they preserve information of physical and chemical mechanisms in the solar nebula, which can unveil evidence about the origin of the planets and their components. However, most carbonaceous chondrites are exposed to secondary processes on their parent bodies, such as thermal metamorphism and aqueous alteration, modifying the primary properties of the carbonaceous chondritic constituents. Hence, it is important to analyze the modifications they have experienced induced by these secondary processes.

In this work, we study the thermal metamorphism of different carbonaceous chondrites (Allende, Bali, Moss, Murray and Nogoyá) examining their carbon composition in different locations of these samples by Raman spectroscopy [1-4]. We analyze the Raman spectra of carbon allotropes to obtain specific parameters that we use for thermal metamorphism mathematical models [5-6]. In addition, we correlate the Raman results with those acquired using SEM/EDS (Scanning Electron Microscopy/Energy Dispersive X-ray Spectroscopy) [2-4].

Samples, Experimental Methods and Data Analysis: Five carbonaceous chondritic fragments were studied: Allende (CV3, 5.19 g, fell in Pueblito de Allende, Mexico, in 1969), Bali (CV3, 0.0662 g, fell in Nana-Mambere, Central Africa Republic, in 1907), Moss (CO3.6, 1.283 g, fell in Ostfold, Sweden, in 2006), Murray (CM2, 3.366 g, fell in Kentucky, USA, in 1950), and Nogoyá (CM2, 0.0578 g, fell in Nogoyá, Argentina in 1879). No sample preparation was required for none of the experimental techniques employed and all the samples were studied using similar experimental conditions.

The topographical features of the samples were first analyzed using optical microscopy. Low- and high-resolution Raman spectroscopy measurements were carried out to study their structural and mineralogical composition. These measurements were performed at room temperature using a custom-built micro-Raman spectroscopy system with a 532-nm excitation source, a laser spot of ~3 μm and power of ~5 mW on the sample. Raman spectra were taken from numerous spots of selected inclusions and surrounding matrix using a 1-s integration time and different number of accumulations (from 5 to 100) to obtain higher signal-to-noise ratios. The spectrum peak parameters, such as the center position ω and full width half-maximum (FWHM) Γ, were analyzed fitting Gaussian and Lorentzian functions using commercial software packages to identify the materials found in the samples. In addition, the peak parameters of the main Raman bands of graphitic and amorphous carbon (D- and G-bands) for those spectra with high Raman signal-to-noise ratios were used to analyze their thermal metamorphism. In particular, the D-band FWHM Γ_D was used to apply two mathematical models (quadratic [5] and linear [6]) to determine their peak metamorphic temperature (PMT). Finally, the morphological structure and elemental composition of the samples were performed and analyzed by a SEM/EDS system (JEOL JSM 6510LV, Thermo-Noran) using secondary-electron (SE) and backscattered-electron (BSE) detections, and the data analysis software equipped with the system.

Results and Discussion: Figure 1 shows two representative Raman spectra of graphitic carbon and amorphous carbon, with the characteristic D- and G-bands, obtained from Moss and Nogoyá, respectively, that have been used to analyze the thermal metamorphic process of these samples. Figure 2 shows a photograph of the Nogoyá fragment, SEM (SE and BSE) images and an EDS map of the carbon structure associated with the Raman spectrum shown in Figure 1b.

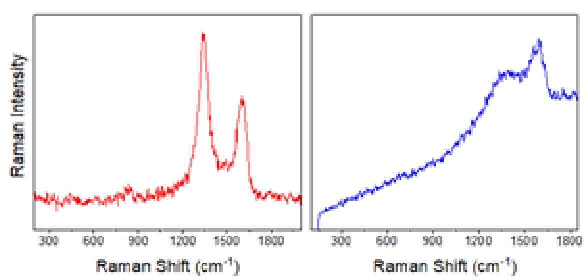

Figure 1. Representative Raman spectra of (a) graphitic carbon found in Moss; and (b) amorphous carbon obtained from Nogoyá.

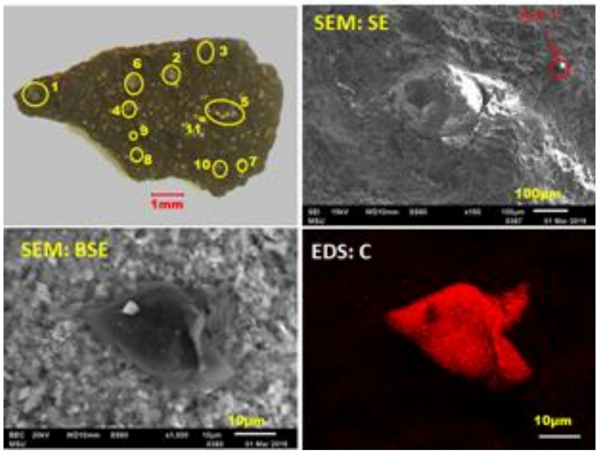

Figure 2. Nogoyá fragment: (a) Photograph of the sample showing the selected regions (inclusion and matrix) marked with yellow circles. (b) SE-SEM image of region 8 at 140X, (c) BSE-SEM image of region 8 spot 1 at 1800X, and (d) EDS map image of region 8 spot 1 at 1800X showing a carbon structure associated with the Raman spectrum in Fig. 1b.

Table 1 shows the main parameters obtained from the Raman D- and G-bands of the carbon spots found in the samples and their PMT values calculated with two mathematical models using the D-band FWHM Γ_D [5-6].

	Allende	Bali	Moss	Murray	Nogoyá
ω_D (cm^{-1})	1351	1347	1340	1348	1360
ω_G (cm^{-1})	1603	1602	1594	1585	1579
Γ_D (cm^{-1})	63	80	68	95	267
Γ_G (cm^{-1})	58	51	71	84	143
PMT Quadratic (°C)	648 ± 20	583 ± 10	630 ± 10	531 ± 50	286 ± 30
PMT Linear (°C)	620 ± 40	505 ± 20	590 ± 10	400 ± 112	-

Table 1. Main Raman peak parameters of D- and G-bands (ω_D, ω_G, Γ_D and Γ_G) of the carbon structures found in Allende, Bali, Moss, Murray and Nogoyá, and their PMT values using two mathematical models: quadratic [5] and linear [6].

The PMT values obtained for Allende, even though some of them do not fall within other values given in the literature considering the petrologic type [e.g. 7] or using the same mathematical models [5-6], are comparable to them. In the case of Murray, the PMT values obtained in this work for both models are higher than those ones reported in the literature [5-6]. This can be due to the relatively low Raman signal-to-noise ratios that in general has been observed for this sample [e.g. 6], which may result in different peak parameter values obtained in other works. For Nogoyá, only the quadratic model is reported here and is in good agreement with those ones reported in another work using the same model [8]. Due to the amorphous nature of the carbon found, which gives broad Γ_D values, the linear model resulted in a non-reliable PMT value. For Moss, the obtained values are comparable with the range given considering its petrologic type [7]. For Bali and Moss, to the best of the authors' knowledge, PMTs values have not been reported using Raman-based mathematical models, and thus, the present work adds to the research that has been conducted on thermal metamorphism of carbonaceous chondritic meteorites using Raman spectroscopy.

Finally, for both models, the PMT values obtained in this work indicated that Allende had relatively high degree of graphitization and its parent body experienced relatively more thermal metamorphism, while Nogoyá had the lowest degree of graphitization and its parent body was the least thermal altered.

References: [1] Dall'Asén A. G. et al. (2017) *Spectrosc. Lett*. 50:417-425. [2] Dall'Asén A. G. (2018) *49th LPSC*:2571. [3] Dall'Asén A. G. (2019) *50th LPSC*:2897. [4] Kayastha R. (2019) *NCUR Proceed. 2019*: 350-358. [5] Busemann H. et al. (2007) *Meteoritics & Planet. Sci.* 42:1387–1416. [6] Homma Y. et al. (2015) *J. Mineral. Petrol. Sci*. 110:276-282. [7] Huss et al. (2006) *Meteorites and the Early Solar System II*. Tucson: The Univeristy of Arizona Press. pp. 567-586. [8] Chan et al. (2017) *Geochim. et Cosmochim. Acta* 201:392-409.