The secondary history of Sutter’s Mill CM carbonaceous chondrite based on water abundance and the structure of its organic matter from two clasts

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Abstract—Sutter’s Mill is a regolith breccia composed of both heavily altered clasts and more reduced xenoliths. Here, we present a detailed investigation of fragments of SM18 and SM51. We have characterized the water content and the mineralogy by infrared (IR) and thermogravimetric analysis (TGA) and the structure of the organic compounds by Raman spectroscopy, to characterize the secondary history of the clasts, including aqueous alteration and thermal metamorphism. The three methods used in this study suggest that SM18 was significantly heated. The amount of water contained in phyllosilicates derived by TGA is estimated to be approximately 3.2 wt%. This value is quite low compared with other CM chondrites that typically range from 6 to 12 wt%. The infrared transmission spectra of SM18 show that the mineralogy of the sample is dominated by a mixture of phyllosilicate and olivine. SM18 shows an intense peak at 11.2 μm indicative of olivine (Fig. 1). If we compare SM18 with other CM and metamorphosed CM chondrites, it shows one of the most intense olivine signatures, and therefore a lower proportion of phyllosilicate minerals. The Raman results tend to support a short-duration heating hypothesis. In the I_D/I_G versus FWHM-D diagram, SM18 appears to be unusual compared to most CM samples, and close to the metamorphosed CM chondrites Pecora Escarpment (PCA) 91008 and PCA 02012. In the case of SM51, infrared spectroscopy reveals that olivine is less abundant than in SM18 and the 10 μm silicate feature is more similar to that of moderately altered CM chondrites (like Murchison or Queen Alexandra Range [QUE] 97990). Raman spectroscopy does not clearly point to a heating event for SM51 in the I_D/I_G versus FWHM-D diagram. However, TGA analysis suggests that SM51 was slightly dehydrated as the amount of water contained in phyllosilicates is approximately 3.7 wt%, which is higher than SM18, but still lower than phyllosilicate water contents in weakly altered CM chondrites. Altogether, these results confirm that fragments with different secondary histories are present within the Sutter’s Mill fall. The dehydration that is clearly observed for SM18 is attributed to a short-duration heating based on the similarity of its Raman spectra to that of PCA 91008. Because of the brecciated nature of Sutter’s Mill and the presence of adjacent clasts with different thermal histories, impacts that can efficiently fragment and heat porous materials are the preferred heat source.
A few tons of extraterrestrial material falls on Earth every day (Engrand and Maurette 1998). However, only a tiny fraction of it is recovered and made available to the scientific community through national and international collections. Observations of meteors and recovery of the landed samples offer unique opportunities for planetary sciences. First, the quick recovery of extraterrestrial samples after their fall minimizes any subsequent modifications induced by terrestrial weathering. Furthermore, fresh falls provide a unique opportunity to study highly labile mineral and organic phases (Brown et al. 2000; Jenniskens et al. 2009, 2012; Haack et al. 2012). Moreover, when the meteor has been accurately observed in space and time, its trajectory can be precisely reconstructed to extract its orbit. A fall provides a tool to connect hand samples with the structure and population of minor bodies in the solar system. Rare massive meteorite falls give access to the structure of the meteoroid up to the metric scale, by studying the heterogeneity of the recovered fragments (i.e., polymict or monomict breccias, monolith).

Sutter’s Mill is an observed, relatively large meteorite fall that occurred on April 22, 2012 in the golden state of northern California near the Lotus-Coloma region. Real-time Doppler radar observations of its landing areas enabled its subsequent quick recovery in the spring of 2012. More than 70 samples, representing a total mass around 1 kg, were recovered within a month of the observed fall (Jenniskens et al. 2012). Orbit reconstruction reveals a high eccentricity with an aphelion close to Jupiter. The calculated Tisserand parameter ($T_J = 2.81 \pm 0.32$) may suggest a link with the Jupiter Family Comets, underlying the potential comet-asteroid continuum (Jenniskens et al. 2012).

Sutter’s Mill results from a complex formation history at the parent body surface. Mineralogy, petrography, and isotope chemistry of several distinct recovered fragments led to its classification as a regolith breccia composed of CM carbonaceous chondrite material and highly reduced xenolithic materials. Preliminary examination of some fragments also reveals distinct lithologies that have experienced variable degrees of secondary processing (Jenniskens et al. 2012).

Here, we extend the investigation of this rare material further by focusing on the characterization of the postaccretional history of two centimeter-sized fragments of Sutter’s Mill, SM18 and SM51. SM18 contains several millimeter-sized clasts, and SM51 is composed of several lithologies that have experienced variable degrees of aqueous alteration (Jenniskens et al. 2012). Although they were quickly recovered, the two samples were still subjected to terrestrial weathering, in particular through postfall heavy rain on April 25–26, 2012. To get a more precise characterization of the postaccretional history experienced by Sutter’s Mill, we performed a multianalytical characterization of raw pieces of SM18 and SM51. Aqueous alteration is assessed through the speciation and abundance of water and hydrated minerals, based on infrared spectroscopy and thermogravimetric analysis. The thermal history is constrained through the structural characterization of the included organic matter by Raman and infrared spectroscopy, respectively, on bulk and extracted insoluble organic matter. Results obtained on Sutter’s Mill are then compared to other primitive chondrites, previously characterized by the same analytical approaches (e.g., Beck et al. 2010, 2014; Quirico et al. 2012).

**IR SPECTROSCOPY OF BULK SAMPLES**

**Method**

For each meteorite, 30–50 mg of rock was ground dry in an agate mortar. Out of this mass, 1.0 mg was
weighed and mixed with 300 mg of commercial ultrapure KBr. This mixture was then compressed to 5000 psi to obtain a 13 mm diameter pellet with good optical quality. Prior to pellet generation, KBr was heated to 80 °C to minimize terrestrial water contamination. The IR spectra of the pellets were measured at room temperature, and after heating at two elevated temperatures, 150 °C and 300 °C, respectively over 2 h. This procedure removed any adsorbed mesopore water. In the case of the most hydrated samples, cracking of the pellets was observed that could lead to an increase in the diffuse background of the spectra, but did not significantly affect the quality of spectra.

Infrared spectra were measured with a Bruker Vertex 70v spectrometer at the Institut de Planétologie et d’Astrophysique de Grenoble. Spectra were acquired at a 2 cm⁻¹ spectral resolution in the 5000-400 cm⁻¹ range (2–25 μm). Spectra were measured on ground fragments of meteorites and therefore provided global information on the bulk meteorite mineralogy.

RESULTS

The 5–25 μm spectral range of silicate is dominated by SiO₂-related vibrations, either as stretching modes of Si–O within tetrahedra around 10 μm or as bending or deformation modes at higher wavelengths. In the case of –OH bearing minerals, additional absorptions are present around 3 and 6 μm due to –OH stretching, X–OH bending and combinations, and at higher wavelength due to –OH libration (within 12–16 μm).

The transmission spectra of SM18 and SM51 are presented in Fig. 1, together with transmission spectra of other CM chondrites and the CV chondrite Allende (Beck et al. 2014). The spectra are ordered according to the intensity of the 11.2 μm feature that is attributed to olivine. Variations in the intensity of this feature, with respect to the 10.0 μm feature attributed to phyllosilicates, are explained by variability in the amount of phyllosilicates with regard to olivine. This ratio estimates the extent of aqueous alteration, as it is correlated with the aqueous alteration scale of Rubin et al. (2007), as well as with the total phyllosilicate content (Howard et al. 2009, 2011) for the CM chondrites. The two fragments of Sutter’s Mill fall at different positions within this sequence. Fragment SM51 appears to be close to the ratio of the least altered CM chondrites, like Queen Alexandra Range (QUE) 97990 (petrologic type 2.6 according to Rubin et al. 2007). This suggests a primitive characteristic for this fragment. The SM51 spectra shows a broad band centered around 10.0 μm that can be explained by a poorly crystalline phyllosilicate (Beck et al. 2010). On the other hand, SM18 clearly shows an intense 11.2 μm olivine feature, more intense than in any “unmetamorphosed” CM chondrite. The attribution of the 11.2 μm absorption to abundant olivine is confirmed by the presence of another absorption around 16.5 μm. As discussed later, heating and recrystallization of olivine from phyllosilicates might explain such spectra from SM18.

THERMO-GRAVIMETRIC ANALYSIS

Method

Thermogravimetric analyses were performed with a TGA/SDTA 851e Mettler Toledo instrument. A fragment of the meteorite was grinded manually (around 50 mg) in a mortar and 10 mg of this powder was selected for experiments and deposited inside a 150 μL alumina crucible with a pierced cover under inert N₂ atmosphere maintained with a flow of 50 mL min⁻¹. The heating was performed from 25 °C to 1200 °C with a heating rate of 10 °C min⁻¹. The accuracy on mass measurements with our TGA is approximately ±1 μg. In our experimental conditions (i.e., 10 mg of sample and about 10% of mass loss), the TGA mass loss error was ±0.1%. Also, the sample mass weighting error was 0.1%. The temperature measurement during heating is accurate to ±0.25 °C.

Thermogravimetric analysis was used to quantitate the amount of hydrogen in the chondrites by mass loss of the sample during the heating. The first derivative (DTG) of the TG curves allow the peak temperature of mass loss to be determined and gives an indication of the carrier phase of the released volatile elements.

RESULTS

The mass loss curves of both fragments of Sutter’s Mill are shown in Fig. 2. They are compared with those from two other CM chondrites: Allan Hills (ALH) 84029, a highly altered chondrite (Alexander et al. 2013), and Wisconsin Range (WIS) 91600, a slightly thermally metamorphosed chondrite (Nakamura 2005). The TGA curves can be used to quantitate volatile elements, in particular the amount of H₂O and –OH. The values are reported in Table 1. The total mass losses of SM18 and SM51 are 6.7 wt% and 10.5 wt%, respectively, and can be further deconvolved in temperature range to indicate the different hosts of H₂O and –OH.

The first release from 25 °C to 200 °C corresponds to molecular water (adsorbed and in mesopore). In the case of SM51, the DTG curves (Fig. 3) present a single peak at approximately 80 °C, which is typical of adsorbed water. Differences are observed between SM18
and SM51 in this temperature range, but this weakly bonded H₂O is not relevant, as it most likely results from terrestrial contamination rather than parent body processes. The second temperature range that can be defined is from 200 °C to 400 °C, and corresponds to the release of water from (oxy) hydroxides. For both SM clasts, these minerals only account for a small mass loss (<0.5 wt%) and no significant peaks are present on the DTG curve. The third temperature range is from 400 °C to 770 °C, and is due to the hydroxyl groups (−OH) delivered by the decomposition of phyllosilicates. Significant differences are measured in the phyllosilicate range (from 400 °C to 770 °C) with a mass loss of 12.3 wt%, 7.4 wt%, 3.7 wt%, and 3.2 wt% for ALH 84029, WIS 91600, SM51, and SM18, respectively.

The low values found for Sutter’s Mill fragments clearly point to a dehydration event in comparison with other CMs. SM18 seems to be more depleted in phyllosilicate H₂O than SM51, but the difference observed is too low to be able to discuss differences in thermal history between the two fragments from TGA only. However, the DTG curves show clearly a difference of mineral host, with the major peak around 450 °C for SM18, in contrast to a small peak around 475 °C and a broad peak between 525 °C and 600 °C for SM51. These features probably point to a difference in the mineralogy of the phyllosilicates between the two fragments. The last temperature range from 770 °C to 900 °C corresponds to the release of CO₂ from carbonates and possibly SO₂ from sulfates. The mass loss in this temperature range is different for the two clasts with values of 1.3 wt% and 0.7 wt% for SM18 and SM51, respectively. However, important variations in mass loss within this temperature range were observed in a TGA repeatability study on CM chondrites, suggesting a very heterogeneous carbonate presence in chondrites from this chemical class (Garenne et al. 2012).

**RAMAN SPECTROCOPY OF ORGANIC MATTER**

**Method**

Raman spectroscopy was performed at Laboratoire de Géologie de Lyon (LGL: Université Claude Bernard—Ecole Normale Supérieure de Lyon, France). We used a LabRam Raman spectrometer (Horiba Jobin–Yvon) equipped with a 600 g mm⁻¹ grating and a Spectra Physics Ar⁺ laser that provided a 514 nm excitation wavelength. The laser beam was focused through a ×100 objective, leading to an approximately 0.9 μm circular spot. The typical power on the sample and acquisition time were 300 μW and 90 s, respectively. Spectra were acquired in the 490–2230 cm⁻¹ spectral range. Measurements were mostly performed on raw matrix grains to enhance heat dissipation with surrounding minerals. Raw matrix fragments were selected from bulk rocks under a binocular microscope, and then transferred to a glass slide. They were pressed by applying another glass slide, and the slide that contained the largest and flattest sample was used for Raman analysis.
Raman spectra were analyzed as follows. First, the fluorescence background was subtracted assuming a linear shape within the 800–2000 cm$^{-1}$ range and the peak intensity of the G band was set to 1 to normalize the whole set of data. We then applied a so-called LBWF fit to the selected spectra, consisting in a Lorentzian profile for the D-peak and a Breit-Wigner-Fano profile for the G-peak. Raman spectral parameters such as width at half maximum (FWHM-G, FWHM-D), peak position ($x_G$, $x_D$), and ratio of peak intensity ($I_D/I_G$) of the G and D bands were then obtained.

RESULTS

The Raman spectrum of SM51 displays broad G and D bands superimposed onto a fluorescence background of high intensity (Fig. 4). In contrast, the spectrum of SM18 is devoid of fluorescence, but still displays broad G and D bands. The spectral analysis confirms the preliminary visual observation (Fig. 5). SM51 plots along with unmetamorphosed type 2 chondrites. Its $I_D/I_G$ parameter is among the lowest of this main group, along with the MAC 88100 and ALH84029 CM chondrites, indicative of minimal thermal processing. In contrast, SM18 is characterized by higher $I_D/I_G$ and smaller FWHM-D (Fig. 5). These Raman characteristics are similar to those of the metamorphosed CM chondrites Pecora Escarpment (PCA) 91008, WIS 91600, or the ungrouped C2 Tagish Lake (Fig. 5) (Quirico et al. 2012). Raman spectroscopy therefore shows the presence of at least two lithologies in Sutter’s Mill: one thermally metamorphosed, through a short-duration process triggered by impact; and one with barely detectable effects of thermal process.

DISCUSSION

CM chondrites form the largest carbonaceous chondrite family ($n > 400$). They are usually recognized as being fragments of a similar lithology that experienced variable extents of aqueous alteration (McSween 1979; Zolensky et al. 1993; Rubin et al. 2007; Howard et al. 2009, 2010, 2011; Alexander et al. 2013; Rubin 2013). Differences in the alteration of chondrules, as well as in the chemistry of some specific secondary phases, are observed. Matrix metals are generally not preserved, and usually transformed to tochilinite (with the exception of some lithologies of the Paris meteorite; Leroux et al. 2013). Posthydration thermal metamorphism has been observed, and “metamorphosed-CMs” or “heated-CMs” have been described (Akai 1992; Nakamura 2005), with pronounced groupings in three oxygen isotope space (see for example fig. 4 in Jenniskens et al. 2012).
In general, three factors are likely to control the variability in water abundance within carbonaceous chondrites. The first factor is the chondrule-to-matrix ratio, as the fine-grained and porous matrix are more easily hydrolized than the larger and pore-free chondrules. The second factor is the aqueous alteration history, for which the controlling parameters were the chemistry of the fluid, its temperature, the fluid/rock ratio, and the duration of the process. Lastly, the posthydration thermal history can lead to the modification in the alteration phases, and ultimately to the total dessication of the sample. For the latter process, a parameter of importance was the temperature-time path that controlled the extent of destabilization of mineral phases.

The two methods used here for characterizing water abundance show that SM18 is clearly depleted in H$_2$O with respect to unheated CM chondrites. A low abundance of phyllosilicates with regard to olivine is found (Fig. 1), and the amount of H$_2$O determined by TGA is lower by a factor of about two with regard to a typical weakly altered CM like QUE 97990 (classified as petrologic type 2.6 in Rubin et al. 2007). The fragment SM51 shows a slightly higher abundance of phyllosilicates compared with SM18, but tends to be similar to samples that have been described as weakly heated CMs like WIS 91600 (type II heated CM in Nakamura [2005] classification). The thermogravimetric analysis of these fragments also points toward a heating event. All these results are in fair agreement with parallel TGA measurements obtained by Garvie (2013), and oxygen isotopes data obtained by Ziegler and Garvie (2013), that identified thermally metamorphosed clasts in Sutter’s Mill from their heavy oxygen isotope composition.

Raman spectroscopy of meteorite organics can be of great interest in reconstructing the thermal history of metamorphosed CM chondrites (Bonal et al. 2006, 2007; Quirico et al. 2012). The measured Raman spectra (Figs. 4 and 5) confirm that SM18 is clearly unusual compared with unheated CM and CR2 chondrites when looking at the FWHM-D versus $I_D/I_G$ diagram, which tends to point directly toward a heating event. Sample SM51 is located closer to nonheated CMs, but appears to be borderline, possibly due to mild heating. Studies on meteorites and coals reveal that Raman spectroscopy is insensitive to the first stages of carbonization, which consist mostly of compositional modifications with no or undetectable structural rearrangement (Quirico et al. 2003). Determining the degree of mild heating in SM51 would require the extraction of insoluble organic matter (through chemical demineralization) and characterization of its composition with nuclear magnetic resonance or infrared spectroscopy.
Here, we studied two fragments of clasts SM18 and SM51. Sutter’s Mill is described as a breccia, and important variations in clast mineralogy were reported from a number of analytical methods (Jenniskens et al. 2012). As is the case for all polymict breccia, sampling bias must be considered; it is virtually impossible to have a representative measurement unless the entire sample is analyzed. Therefore, we must note that our measurements are only sampling a part of the diverse thermal history experienced by the different clasts of Sutter’s Mill.

As has been proposed to explain the nature of metamorphosed CM chondrites (Nakamura 2005), three heat sources might have acted to modify Sutter’s Mill mineralogy and organic matter. The first possible source is radiogenic heating by short-lived radionuclides, similar to the process that affected type >3.0 chondrites. The time-scale for this kind of process is of the order of a few million years (Bouvier et al. 2007). The second is short-duration heating during impact metamorphism, produced by hypervelocity collisions between small bodies, which can lead to a fast temperature increase. The third process is radiative heating by sunlight absorption that happened for meteoroids orbiting close to the Sun.

The Raman spectra of SM18 suggests that the thermal history of this fragment was of the same type as that of the heated CM chondrite PCA 91008 (Fig. 5). For this latter sample, the study of its organic matter revealed a heating process dissimilar to long-duration radioactive heating (Yabuta et al. 2010; Orthous-Daunay et al. 2013), ruling out this process as a heat source for SM18, by inference. In other words, the Raman signature of SM18 macromolecular organic compounds is inconsistent with a long-duration thermal metamorphism as observed for ordinary chondrites. An alternative is radiative heating, which has been proposed to explain the formation of heated CM chondrites (Nakamura 2005), but was only tested recently in a quantitative approach. Simulations by Chaumard et al. (2012) revealed that the CV-CK relation could be explained by this kind of metamorphism. Radiative heating will be efficient for small meteoroids, with eccentric orbits whose perihelion is within a few tens of astronomical units. This is almost the case for the Sutter’s Mill meteoroid, with a perihelion of 0.456 AU, which might produce surface temperature in excess of 600 K (Chaumard et al. 2012). However, one would expect radiative heating to cause the formation of a metamorphosed crust, rather than the close coexistence of fragments with different thermal histories as observed for Sutter’s Mill (Jenniskens et al. 2012). As a consequence, impact-induced metamorphism seems to remain the only possible heat source to explain our observation on SM18 and possibly SM51.

The frequency of low-density objects (Britt and Consolmagno 2001; Carry 2013) among asteroids suggests that collision is a major geological process in the main belt. The relative velocity between main-belt asteroids (MBA) is, on average, about 5 km s⁻¹ (Bottke et al. 1994), and “encounters” between objects traveling at this velocity will generate shock waves. Upon shock-wave arrival, the target is compressed to high pressure along the Hugoniot curve, leading to an adiabatic increase in temperature. With anticipated high porosity on the parent body, additional energy dissipation will occur due to pore collapse (Ahrens et al. 1992; Meyers et al. 1999; Beck et al. 2007) that can be much more efficient as a heat source than adiabatic compression. In the case of CM chondrites, for which porosity can be high (Britt and Consolmagno 2000), energy dissipation by pore collapse will be efficient and partial melting might occur at relatively low shock pressures with respect to the nonporous Hugoniot. Evidence for this process has been observed during shock-wave experiments performed on Murchison (Tomeoka et al. 1999; Beck et al. 2011). In the later studies, partial melting was observed at 20 GPa, while the adiabatic compression for a nonporous chondrite would only raise the temperature of a few hundred K for such shock pressures (see the calculation for pure olivine and pyroxene in Malavergne et al. 2001).

Impact features are uncommon in CM chondrites with the exception of the presence of an impact-induced petrofabric (Rubin 2013). However, there are a number of meteorites that are now identified as heated CM chondrites (see for instance Alexander et al. 2012, 2013), which could have been produced by impact-induced thermal metamorphism. Our observations that clasts of Sutter’s Mill experienced some severe impact heating raise the question whether this impact event could have induced the disruption of the parent body, or if such thermal processing is achievable within a “normal” asteroid regolith formation scenario.

Significant knowledge on the mechanism of impact brecciation came from the lunar studies, where the word regolith was initially defined in the Planetary Sciences context (Shoemaker et al. 1967). However, a lunar style regolithization model cannot be applied directly to asteroidal bodies where the gravitational field is drastically reduced. For such bodies, the regolithization process will be a competition among shattering, compaction, ejection, and possible subsequent re-accretion. The strength of the asteroidal target will be of importance; as an example, for very porous asteroids, the formation of crater ejecta can be suppressed (Holsapple and Housen 2012). From the viewpoint of the impactor, the problem is multivariate as the distributions in impactor sizes, speeds,
porosities, and strengths will all play a role in controlling the maturation of the regolith. These distributions will also influence the mechanisms of heat dissipation in the target asteroid through compaction and possible burial below ejecta blankets. Shock-wave experiments can help in understanding some mineralogical and thermal effects of impacts (see for example Stöffler et al. 1991), but the textural and mechanical aspects are more difficult to reproduce due to limitation in the thickness of the shock front generated by such experiments.

The systematic presence of solar wind implanted gases in CM chondrites (Bischoff and Schultz 2004), including Sutter’s Mill, might point toward an origin within an asteroidal regolith. However, as explained by Bischoff et al. (2006), the catastrophic disruption and subsequent re-accretion of CM parent body(ies) could explain the systematic presence of breccias among CM chondrites and the presence of solar wind gases if some fraction of regolithic material was incorporated. Members of the old 24-Themis asteroid family (>1 Gyr, Marzari et al. 1995) show spectral similarities with carbonaceous chondrites and the presence of solar wind gases if some fraction of regolithic material was incorporated. We make the following conclusions from our study of two clasts, SM18 and SM51, from the Sutter’s Mill carbonaceous chondrite.

1. The infrared spectra and thermogravimetric analysis of powders from the two clasts show that they contain different phyllosilicate abundances.
2. The abundances of phyllosilicates are lower than are typically found in unheated CM chondrites.
3. The Raman spectra of SM51 is clearly distinct from unheated CM chondrites, and instead resembles the heated CM chondrite PCA 91008. This fragment did experience short-duration thermal metamorphism.
4. The Raman spectra of SM18 are borderline, but within the range of unheated CM chondrites. Mild thermal metamorphism cannot be excluded from these Raman spectra.
5. These data show that the two fragments coming from clasts SM18 and SM51 of Sutter’s Mill experienced a short-duration thermal metamorphic event. This event differed in intensity between the two fragments, which points toward an impact origin.
6. The presence of heated clasts in the Sutter’s Mill breccia might support the hypothesis that the CM parent body experienced a major impact event at some point of its history leading to the disruption of the parent body, following a possible scenario described in Bischoff et al. (2006).

**CONCLUSIONS**


**REFERENCES**


