



Bidirectional reflectance spectroscopy of carbonaceous chondrites: Implications for water quantification and primary composition



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ARTICLE INFO

Article history:

Received 21 April 2015

Revised 2 September 2015

Accepted 2 September 2015

Available online 12 September 2015

Keywords:

Meteorites

Asteroids

IR spectroscopy

Mineralogy

ABSTRACT

In this study, we measured bidirectional reflectance spectra (0.5–4.0 μm) of 24 CMs, five CRs, one CI, one CV, and one C2 carbonaceous chondrites. These meteorites are known to have experienced an important variability in their relative degrees of aqueous alteration degree (Rubin et al. [2007]. *Geochim. Cosmochim. Acta* 71, 2361–2382; Howard et al. [2009]. *Geochim. Cosmochim. Acta* 73, 4576–4589; Howard et al. [2011]. *Geochim. Cosmochim. Acta* 75, 2735–2751; Alexander et al. [2013]. *Geochim. Cosmochim. Acta* 123, 244–260). These measurements were performed on meteorite powders inside an environmental cell under a primary vacuum and heated at 60 °C in order to minimize adsorbed terrestrial water. This protocol allows controlling of atmospheric conditions (i.e. humidity) in order to avoid contamination by terrestrial water. We discuss various spectral metrics (e.g. reflectance, band depth, single-scattering albedo, ...) in the light of recent bulk composition characterization (Howard et al. [2009]. *Geochim. Cosmochim. Acta* 73, 4576–4589; Howard et al. [2015]. *Geochim. Cosmochim. Acta* 149, 206–222; Alexander et al. [2012]. *Science* 337, 721; Beck et al. [2014]. *Icarus* 229, 263–277; Garenne et al. [2014]. *Geochim. Cosmochim. Acta* 137, 93–112). This study reveals variability of reflectance among meteorite groups. The reflectance is not correlated with carbon or hydrogen abundance neither with measured grain size distribution. We suggest that it is rather controlled by the nature of accreted components, in particular the initial matrix/chondrule proportion. Band depth, integrated band depth, mean optical path length, normalized optical path length, effective single-particle absorption thickness were calculated on the so called 3- μm band for reflectance spectra and for single scattering albedo spectra. They were compared with hydrated phase proportions from previous study on the same meteorites by thermogravimetric analyses and infrared spectroscopy in transmission. We find that normalized optical path length (NOPL) is the most appropriate to quantify water abundance, with an absolute error of about 5 wt.%. These datasets also reveal a variability of the band shape between 2.8 and 2.9 μm , which is interpreted as reflecting variation in the chemical composition and structure of phyllosilicates. This chemical variation could also be used to quantify the aqueous alteration degree between meteorite groups. The combination of reflectance at 2 μm and the depth of 3- μm band can be combined, to classify carbonaceous chondrites in reflectance in term of primary composition (e.g. matrix/chondrule ratio, carbon content) and secondary processes (e.g. aqueous alteration, thermal metamorphism). This could be used to decipher the nature of aqueous alteration in C-complex asteroids.

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1. Introduction

Asteroids contain refractory materials that are expected to have only been marginally affected by chemical processes since the

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formation of the Solar System. They can be considered as geological antiques that are expected to preserve precious clues to the birth of our planetary system. The main-belt is spectroscopically diverse suggesting that a variety of mineralogical compositions is present. Large spectroscopic surveys combined with dynamical studies have led to the classification of asteroid families (taxons), some of which have related meteorite groups (Tholen, 1984; Bus and Binzel, 2002; Gaffey et al., 1993; Vilas, 1994; Burbine, 1998; McSween et al., 2002; DeMeo and Carry, 2014). However, in the case of the darkest objects (the C-complex, and the D-types from DeMeo et al. (2009)) associations to specific meteorite groups can be difficult, because of the lack of diagnostic absorptions in the visible and near-infrared (VNIR).

Major spectroscopic surveys are limited to the VNIR and typically lack the 3- μm feature, which is the most diagnostic for -OH/H₂O bearing phases (water-ice, hydrated hydroxylated minerals, alcohol function). Such “water” related absorptions have been observed on asteroidal surfaces (Lebofsky et al., 1981; Larson et al., 1983; Jones et al., 1990; Rivkin et al., 2002), but observations in this region are usually limited to large objects and the effort of building a taxonomy based on this spectral range is ongoing (Takir and Emery, 2012). At present, most observations at 3- μm have been interpreted by the presence of -OH bearing mineral phase, and a few of them by the presence of water ice (mostly outer main belt objects, Campins et al., 2010; Rivkin and Emery, 2010; Takir and Emery, 2012; see also Beck et al. (2011) for an alternative – goethite). Across the main belt, 3- μm absorption bands have been found for different asteroid classes with variations in band shape and band depth (Takir and Emery, 2012; Rivkin et al., 2003). Some other asteroid classes do not present detectable 3- μm absorption features.

Some meteorite groups also show clear evidence of hydration in the form of secondary minerals formed under low-temperature, by precipitation from an aqueous fluid. This is particularly evident for some carbonaceous chondrite classes, including the CI group. The CI group is an extreme case of aqueously altered meteorites since almost all primary minerals were transformed to secondary phase, including phyllosilicates (a mixture of clays and serpentine; Tomeoka and Buseck, 1988). This aqueous alteration event was heterogeneous across the different carbonaceous chondrite families and within a given group. From the nature and amounts of secondary mineral phases, alteration sequences have been discussed (e.g. McSween, 1979; Bunch and Chang, 1980; Tomeoka and Buseck, 1985; Zolensky and McSween, 1988; Takir et al., 2013). Recently, aqueous alteration scales have been constructed based on petrography and crystal chemistry (Rubin et al., 2007; Harju and Rubin, 2013), phyllosilicate abundance (Howard et al., 2009, 2011, 2015; Beck et al., 2014; Garenne et al., 2014) and C and H isotopic analyses (Alexander et al., 2012, 2013).

Our objective here is to compare the spectral metrics of aqueously altered carbonaceous chondrites in reflectance, with an emphasis on the 3- μm region for comparison with remote observations of small bodies. The reflectance spectra were measured using a set of 23 CMs, 5 CRs from Antarctica and 4 chondrite falls (Orgueil (CI), Murchison (CM), Allende (CV) and Tagish Lake (C2)). Spectra were obtained under vacuum and moderate temperature to minimize contamination by adsorbed water, and with a high photometric accuracy (<0.25%). These same specific samples of each meteorite were previously studied by infrared spectroscopy in transmission (Beck et al., 2014) and with thermogravimetric analyses (Garenne et al., 2014) to evaluate modal mineralogy and water abundance. Many of these meteorites also had modal abundances determined by X-ray diffraction, although on different powder aliquots. Based on these two studies, we calculate various spectral metrics related to the 3- μm band and try to identify the most reliable way to quantify hydrogen on these dark

meteorites and improve our understanding of the variability of the 3- μm band on low albedo asteroids.

2. Methodology, samples and analytical procedures

2.1. Meteorite samples studied

Reflectance spectroscopy was performed on 24 CMs, 5 CRs, 1 CI and 1 CV and one C2 chondrites (Table 1). Here we focused in particular on CM, CR and CI chondrites since they have been described in several articles as being significantly altered meteorites groups. These three groups have very distinct petrographical properties, different matrix proportion, carbon content, abundances of opaque phases, metal, secondary mineral phases, and water content (Brearley, 2006; Weisberg and Huber, 2007; Bunch and Chang, 1980; Zolensky and McSween, 1988; Krot et al., 2007). Their mineralogy and alteration degree have been studied previously (Bunch and Chang, 1980; Zolensky and McSween, 1988; Tomeoka et al., 1989; Browning et al., 1996; Rubin et al., 2007; Howard et al., 2009, 2011, 2015; Barber, 1981; Tomeoka and Buseck, 1985; Cloutis et al., 2011a,b, 2012, 2012b; Takir et al., 2013). The CM chondrites we analyzed were selected to span the full range of aqueous alteration from 2.0 to 2.6 defined by Rubin et al. (2007) and phyllosilicates range from 65 vol.% to 87.5 vol.% (Howard et al., 2011). One is a fall chondrite (Murchison) while some others was subjected to thermal events, like ALH 84033 and EET 83355. The CR chondrites studied cover a large range of aqueous alteration intensity from highly altered to primitive meteorites. Most of them are classified as CR2 except for GRO 95577, which is a CR1. EET 92159 has a primitive mineralogy (Abreu and Brearley, 2010) while RBT 04133 and GRA 06100 are spectrally different in the infrared (Beck et al., 2014). In the case of GRA 06100, the meteorite has been heated and shocked (Abreu and Stanek, 2009; Alexander et al., 2013).

Around 1 g of each meteorite was crushed and some part was used for the three methods; around 10 mg for thermogravimetric analyses (TGA), 1 mg for infrared spectroscopy in transmission and 700 mg for reflectance spectroscopy (Beck et al., 2014; Garenne et al., 2014).

2.2. H₂O/OH abundance determination

Three different methods were used to characterize the nature of aqueous alteration in our suite of chondrites: thermogravimetric analyses (TGA) (Garenne et al., 2014), infrared (IR) spectroscopy in transmission (Beck et al., 2014) and reflectance spectroscopy. TGA measurements were used to quantify water abundance, to identify the mineral host of water and the level of matrix aqueous alteration (Garenne et al., 2014). Infrared spectroscopy was used to identify the mineralogy from the silicate vibration modes and to quantify (-OH) abundance based on the 3- μm band (Beck et al., 2014). This paper presents the reflectance spectra of the same sample aliquots studied by TGA and transmission IR methods. This approach was used to minimize the chemical and mineralogical variations due to the heterogeneity of the meteorites (most of them are regolith breccia; Bischoff et al., 2006), and to avoid possible nugget effects. This heterogeneity was measured and quantified on one CM chondrite by thermogravimetric analyses and it is estimated at 0.95 wt% on H₂O quantification (for a sample containing an average of 5.3 wt% of H₂O in phyllosilicates, 6 measurements) (Garenne et al., 2014). The IR spectroscopy and TGA results allow us to constrain our reflectance spectra and to find the best spectral metrics to quantify the water abundance with the reflectance spectroscopy. In addition, the hydrogen content (H₂O/OH contributions) estimated by Alexander et al. (2012, 2013) from bulk H and C measurements were used to trace the impact of parent body process on reflectance spectra features.

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