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# INFRARED DIFFUSE REFLECTANCE SPECTRA OF SEVERAL THERMALLY METAMORPHOSED CARBONACEOUS CHONDRITES

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Abstract: Infrared diffuse reflectance spectra  $(2.53-25 \,\mu\text{m})$  were measured for several Antarctic carbonaceous chondrites. Thermally metamorphosed carbonaceous chondrites with CI-CM affinities (Belgica(B)-7904, Yamato(Y)-793321, Y-82162, and Y-86720), which were recently found in the Antarctic collection, show weaker absorption bands near 3  $\mu$ m due to hydrous minerals, compared with the Murchison (CM) carbonaceous chondrite. These results show that hydrous minerals are dehydrated as a result of thermal metamorphism, consistent with the results of chemical, mineralogical, and petrologic studies. B-7904, Y-793321, and Y-86720 show weaker absorption bands near  $6.9 \,\mu m$  due to carbonates than those of Murchison. For Y-82162, the 1450 cm<sup>-1</sup>(6.9  $\mu$ m) band is relatively strong unlike the other thermally metamorphosed carbonaceous chondrites despite the weak 3  $\mu$ m band of all the thermally metamorphosed ones measured. The shape of the spectral curves near 3 µm of the thermally metamorphosed carbonaceous chondrites resembles those of Antarctic ordinary chondrites affected by terrestrial weathering, implying that major absorption features near 3  $\mu$ m of the thermally metamorphosed carbonaceous chondrites may be due to secondary hydrous minerals produced by terrestrial weathering.

#### 1. Introduction

As part of the consortium study, infrared diffuse reflectance spectra (2.53– 25  $\mu$ m) of several thermally metamorphosed Antarctic carbonaceous chondrites were measured. Although most CI and CM carbonaceous chondrites have not been heated over a few hundred °C, several carbonaceous chondrites with CI-CM affinities, which show thermal metamorphic features, were recently found in the Antarctic meteorite collection (KOJIMA *et al.*, 1984; KOJIMA and YANAI, 1987). These carbonaceous chondrites (Belgica(B)-7904, Yamato(Y)-793321, Y-82162, and Y-86720) show unusual properties and have been intensively investigated to explain the thermal metamorphism and differences from usual CI and CM carbonaceous chondrites (*e.g.*, AKAI, 1988, 1990a, b; KALLEMEYN, 1988; TOMEOKA *et al.*, 1989a, b; TOMEOKA, 1990; ZOLENSKY *et al.*, 1989a, b, 1991; PAUL and LIPSCHUTZ, 1990; IKEDA, 1991).

As many investigators have taken a great interest in CI and CM carbonaceous chondrites because of their primitiveness among extraterrestrial materials (e.g., KERRIDGE and BUNCH, 1979; McSWEEN, 1979), the thermal metamorphism of

carbonaceous chondrites may give important information on the early evolution of planetary bodies by comparison with thermal metamorphism of ordinary chondrites.

In this paper, infrared diffuse reflectance spectra of thermally metamorphosed Antarctic carbonaceous chondrites are shown to study the effects of thermal metamorphism on the spectra by comparison with the results of the Murchison (CM) and Y-791198 carbonaceous chondrites.

### 2. Samples and Experimental Techniques

Samples of Antarctic carbonaceous chondrites were supplied by the National Institute of Polar Research and consisted of some small chips. These chips were ground in a corundum mortar and passed through a 100  $\mu$ m sieve. Each powder sample was dried in a desiccator for more than 48 hours to remove any adsorbed water from the grain surfaces. A specimen weighing approximately 20 mg was taken from each powder sample and used for the spectral measurements.

Diffuse reflectance spectra (biconical reflectance) were measured in dry-air surroundings by the use of a Fourier transform infrared spectrophotometer (JASCO, FT/IR-3) equipped with a diffuse reflectance attachment. Dry air with  $-60^{\circ}$ C dew point was passed into the spectrophotometer. Each powder sample was placed in the hollow space of a sample holder 1.5 mm in depth. After setting the sample in the spectrophotometer, the sample was left in dry-air surroundings for 1 hour before measuring the sample reflectance. Spectra were taken over the range from 3950 cm<sup>-1</sup> (2.53  $\mu$ m) to 400 cm<sup>-1</sup> (25  $\mu$ m) at a resolution of 4 cm<sup>-1</sup>. Scans were integrated 1000 times to enhance the signal-to-noise ratio. An aluminum-coated mirror was used as standard.

In order to calculate the integrated intensity of absorption bands near 3  $\mu$ m, a background curve approximating to a straight line obtained by the least squares fitting was determined by using reflectances from 3950 to 3800 cm<sup>-1</sup> and from 2600 to 2500 cm<sup>-1</sup>. Normalized reflectances at the reflectance of 3800 cm<sup>-1</sup> were numerically integrated from 3800 to 2600 cm<sup>-1</sup> to obtain the integrated intensity. It is generally thought that the integrated intensity of absorption bands in reflectance spectra is not in direct proportion to the concentration of a substance unlike that in absorption spectra. Therefore, the integrated intensity was also calculated using the spectra after the transformation by the Kubelka-Munk function, although there still remain some theoretical problems in the Kubelka-Munk function (WENDLANDT and HECHT, 1966; CLARK and ROUSH, 1984; KINOSHITA and MIYAMOTO, 1990).

#### 3. Results and Discussion

#### 3.1. Absorption bands near 3 jum

Figure 1 shows infrared diffuse reflectance spectra of several thermally metamorphosed carbonaceous chondrites in the spectral range from 2.53  $\mu$ m (3950 cm<sup>-1</sup>) to 25  $\mu$ m (400 cm<sup>-1</sup>) in comparison with those of the Murchison (CM) and Allende (CV) carbonaceous chondrites. Sharp absorption at 2350 cm<sup>-1</sup> is caused by atmospheric CO<sub>2</sub>. Figure 2 shows the absorption bands around 3  $\mu$ m caused by hydrates



Fig. 1. Midinfrared diffuse reflectance spectra of some carbonaceous chondrites. The spectra are offset for clarity.

Fig. 2. Absorption bands near 3 µm of some carbonaceous chondrites. The spectra are offset for clarity.

and/or hydroxyl ions. Table 1 summarizes the integrated intensity of absorption bands near 3  $\mu$ m.

B-7904, Y-793321, Y-82162, and Y-86720 carbonaceous chondrites show weaker absorption bands near 3  $\mu$ m compared with the Murchison (CM) carbonaceous chondrite (Fig. 2, Table 1). The integrated intensities of absorption bands near 3  $\mu$ m of these meteorites are significantly smaller than that of Murchison (Table 1). This result means that these Antarctic carbonaceous chondrites contain smaller amounts of hydrous minerals in spite of their classification into CI or CM group. This is due to the fact that the integrated intensity of the absorption bands near 3  $\mu$ m is related to the amount of hydrous minerals (*e.g.*, MIYAMOTO, 1991a).

The spectrum of Y-791198 shows strong absorption bands near 3  $\mu$ m similar to that of Murchison (CM) (Fig. 2, Table 1).

Allende (CV) shows no absorption bands near 3  $\mu$ m (Fig. 2; Table 1), indicating that there are little hydrous minerals.

B-7904 shows unusually low  $H_2O$  content compared with other C2 carbonaceous chondrites (HARAMURA *et al.*, 1983). Phyllosilicates are considerably dehydrated

Meteorites	Group	Integrated iuntensity*	Integrated intensity**
B-7904	CM <sup>+</sup>	58	63
Y-791198	СМ	271	394
Y-793321	$CM^+$	118	132
Y-82162	CI <sup>+</sup>	117	122
Y-86720	CM <sup>+</sup>	102	113
Murchison	СМ	286	459
Allende	CV	8	8

Table 1. The integrated intensity of absortion bands near 3 µm of some carbonaceous chondrites.

+ Thermally metamorphosed.

\* Calculated using reflectance.

\*\* Calculated after transformation by the Kubelka-Munk function.

and almost completely transformed to olivine (AKAI, 1988; TOMEOKA, 1990; ZOLENSKY *et al.*, 1989b). Trace element study by PAUL and LIPSCHUTZ (1990) also shows thermal metamorphic characteristics of this meteorite. B-7904 is classified into CM group (KOJIMA *et al.*, 1984; AKAI, 1988; KALLEMEYN, 1988; TOMEOKA, 1990, PAUL and LIPSHUTZ, 1990), despite its oxygen isotopic composition which places it with CI (MAYEDA *et al.*, 1987; CLAYTON and MAYEDA, 1989).

Y-86720 also has low  $H_2O$  content and phyllosilicates are dehydrated and almost completely transformed to olivine or an intermediate phase between serpentine and olivine (TOMEOKA *et al.*, 1989b; ZOLENSKY *et al.*, 1989b; AKAI, 1990a). According to TOMEOKA *et al.* (1989b), Y-86720 is a CM chondrite that has experienced extensive aqueous alteration and subsequently mild thermal metamorphism after accretion to the parent body. Its petrological and mineralogical characteristics are similar to B-7904. Both B-7904 and Y-86720 are genetically related, having been derived from a common source. These results are consistent with those obtained by trace element chemistry (KALLEMEYN, 1988; PAUL and LIPSCHUTZ, 1990).

Y-793321 is classified into CM group (KOJIMA et al., 1984; MAYEDA et al., 1987) and contains an intermediate phase in the transformation from serpentine to olivine, showing thermal metamorphic features (AKAI, 1988).

Y-82162 is the first CI chondrite from Antarctica (KOJIMA and YANAI, 1987; MAYEDA et al., 1987; KALLEMEYN, 1988; TOMEOKA et al., 1989a). Comprehensive reviews of characteristics of this meteorite and detailed mineralogical and petrologic studies are given in IKEDA (1991). Y-82162 has a distinctly lower H<sub>2</sub>O content than other CI chondrites (TOMEOKA et al., 1989a) and the Y-82162 matrix contains abundant fine grains of olivine, which contrasts with non-Antarctic CI chondrites (TOMEOKA et al., 1989a; AKAI, 1990a). These observations suggest that matrix phyllosilicates are dehydrated and altered to olivine by thermal metamorphism (AKAI, 1988; TOMEOKA et al., 1989a; ZOLENSKY et al., 1989a; AKAI, 1990a). Y-82162 may have been derived from a different source from that of non-Antarctic CI chondrites (TOMEOKA et al., 1989a), consistent with the results of trace elements chemistry (KALLEMEYN, 1988; PAUL and LIPSCHUTZ, 1990). Y-791198 is a CM chondrite similar to Murchison, and is different from Y-793321 and B-7904 (KOJIMA *et al.*, 1984; AKAI and KANNO, 1986).

In short, petrologic and mineralogical studies show that B-7904, Y-793321, Y-82162, and Y-86720 are thermally metamorphosed to various degrees and Y-791198 resembles CM chondrites (*e.g.*, Murchison). Thermally metamorphosed carbonaceous chondrites show weak absorption bands near 3  $\mu$ m (Fig. 2; Table 1), suggesting that most hydrous minerals in these carbonaceous chondrites were dehydrated by thermal metamorphism. Spectral absorption features near 3  $\mu$ m of these Antarctic carbonaceous chondrites are consistent with the results of mineral-ogical and petrologic studies.

Figure 3 compares the spectral features near 3  $\mu$ m of the thermally metamorphosed carbonaceous chondrites with those of Antarctic ordinary chondrites which contain secondary hydrous minerals produced by terrestrial weathering (MIYAMOTO, 1991a). Allan Hills (ALH)-77299 (H3) shows typical absorption features near 3  $\mu$ m caused by secondary hydrous minerals among Antarctic ordinary chondrites. Y-75097 (L6) is one of the least weathered Antarctic ordinary chondrites and Y-75029 (H3) is one of the most weathered ones (MIYAMOTO, 1991a). The similarity in shape of the spectral curves near 3  $\mu$ m of the thermally metamorphosed carbonaceous chondrites to those of Y-75097 (L6) and ALH-77299 (H3) affected by terrestrial weathering implies that major absorption intensities near 3  $\mu$ m of the thermally metamorphosed carbonaceous chondrites may be due to secondary hydrous minerals produced by terrestrial weathering. Also, the wave-



Fig. 3. Comparison of absorption features near 3 µm of carbonaceous chondrites with those of Antarctic ordinary chondrites. The spectra are offset for clarity.

length position of reflectance minima near 3400 cm<sup>-1</sup> (2.9  $\mu$ m) of the thermally metamorphosed carbonaceous chondrites is slightly shifted toward longer wavelengths from that (3560 cm<sup>-1</sup>(2.81  $\mu$ m)) of Murchison (Fig. 3). However, weak (shoulder) absorption bands near 3550 cm<sup>-1</sup>(2.82  $\mu$ m) similar to a reflectance minimum of the Murchison spectra can be detected in the spectra of the thermally metamorphosed carbonaceous chondrites (Figs. 2 and 3). It suggests that primary hydrous minerals still remain in them. For severely weathered ordinary chondrites (*e.g.*, Y-75029 (H3)), absorption bands near 3200 cm<sup>-1</sup>(3.1  $\mu$ m) are noticeable (MIYAMOTO, 1991a) and are much different from a reflectance minimum (3560 cm<sup>-1</sup>(2.81  $\mu$ m)) of Murchison (Fig. 3). Anyway, we need to pay much attention to secondary minerals produced by terrestrial weathering in distinction from primary preterrestrial hydrous minerals in the thermally metamorphosed carbonaceous chondrites.

## 3.2. Absorption bands near 7 jum

Figure 4 shows the results of diffuse reflectance spectra near 7  $\mu$ m. Although the spectrum of Murchison shows absorption bands near 6.9  $\mu$ m (1450 cm<sup>-1</sup>), probably due to primary calcite (SANDFORD, 1986; MIYAMOTO, 1987), the spectra of B-7904, Y-793321, and Y-86720, which are thermally metamorphosed, display no absorption bands near 6.9  $\mu$ m (Fig. 4). This suggests that most carbonates



Fig. 4. Absorption bands near 7  $\mu$ m of some carbonaceous chondrites. The arrow indicates the 1450 cm<sup>-1</sup> (6.9  $\mu$ m) band. The spectra are offset for clarity.

were decomposed during heating events.

The spectrum of Y-791198 shows absorption bands near 6.9  $\mu$ m (1450 cm<sup>-1</sup>) similar to Murchison. This result is in line with the spectral features of 3  $\mu$ m similar to those of Murchison.

Although the presence of carbonates is reported in B-7904 (TOMEOKA, 1990; ZOLENSKY *et al.*, 1989b), Y-793321 (KOJIMA *et al.*, 1984), and Y-86720 (ZOLENSKY *et al.*, 1989b; TOMEOKA *et al.*, 1989b; AKAI, 1990a), no significant absorption bands near 7  $\mu$ m are detected.

Although Y-82162 shows weak absorption bands near 3  $\mu$ m as a result of dehydration (Fig. 2), absorption bands near 7  $\mu$ m are seen in the spectrum (Fig. 4), that is, Y-82162 contains a relatively large amount of carbonates compared with the other thermally metamorphosed carbonaceous chondrites. In fact, carbonates are detected in Y-82162 (TOMEOKA *et al.*, 1989a; ZOLENSKY *et al.*, 1989a; AKAI, 1990a). Y-82162 shows relatively broad absorption bands near 7  $\mu$ m compared with those of Y-791198 and Murchison, that is, the spectrum of Y-82162 shows the reflectance minimum around 1510 cm<sup>-1</sup> (6.6  $\mu$ m) in addition to that around 1450 cm<sup>-1</sup> (6.9  $\mu$ m), which is probably caused by calcite and is also seen in the spectra of Murchison and Y-791198. The 1510 cm<sup>-1</sup> (6.6  $\mu$ m) band of Y-82162 may be caused by mineral(s) produced by terrestrial weathering, because severely weathered chondrites show faint absorption bands near 1510 cm<sup>-1</sup> (MIYAMOTO, 1991a). We need further study to identify the mineral(s).

In short, a salient difference between Y-82162 and the other thermally metamorphosed carbonaceous chondrites (B-7904, Y-793321, and Y-86720) is the relatively strong absorption bands near 1450 cm<sup>-1</sup> (6.9  $\mu$ m) of Y-82162 despite the weak absorption bands near 3  $\mu$ m of all the thermally metamorphosed carbonaceous chondrites measured. This result suggests that metamorphic temperature of Y-82162 was high enough to decompose hydrous minerals, but was not enough to decompose carbonates, or that Y-82162 experienced later alteration process to produce carbonates after heating events which decomposed hydrous minerals. We prefer the latter possibility, because our heating experiments show that both the 3 and 6.9  $\mu$ m bands disappear by heating at 500°C but these bands remain by heating at 450°C (MIYAMOTO, 1990). We cannot, however, exclude the possibility that the 1450 cm<sup>-1</sup> (6.9  $\mu$ m) band of Y-82162 is due to other material(s) or mineral(s). For example, hydrocarbon shows an absorption band at 1450 cm<sup>-1</sup> (CLOUTIS, 1990). Visible-Near IR measurement of Y-82162 may give different features from the other carbonaceous chondrites. Actually, unpublished Vis-NIR data show a very similar spectrum to tar sand (hydrocarbons) (CLOUTIS, 1990; HIROI, personal communication, 1991).

#### 3.3. Thermal metamorphic temperature

Several workers have discussed thermal metamorphic temperatures of these thermally metamorphosed carbonaceous chondrites. PAUL and LIPSCHUTZ (1990) proposed the  $600-700^{\circ}$ C temperature range, at relative temperatures B-7904< Y-82162<Y-86720 by comparison of labile trace element contents in these meteorites with those in Murchison samples heated in the laboratory (MATZA and LIPSCHUTZ, 1977).

AKAI (1990a, b) also estimated metamorphic temperatures of 400–800°C, at relative temperatures Y-793321<Y-82162<Y-86720 on the basis of mineralogical observations of thermally metamorphosed Antarctic carbonaceous chondrites by comparison with the Murchison, serpentine, and saponite samples heated in air or in a vacuum at various temperatures. The relative metamorphic temperature for B-7904 is different from that by PAUL and LIPSCHUTZ (1990). AKAI (1990a) reported that B-7904 may have experienced the highest temperature among these four meteorites because no phyllosilicate structures were found and many irregularly shaped olivines were observed in B-7904.

ZOLENSKY *et al.* (1991) also performed heating experiments of Murchison in an anoxic  $(10^{-1} \text{ bars H}_2)$  at the 400–800°C range for one week. They reported that from the preservational state of phyllosilicates, B-7904, Y-82162, and Y-86720 did not locally experience temperatures higher than 60°C, and more probably 400 to 500°C. These temperatures are lower by several hundred degrees than those suggested by AKAI (1990a, b), from experiments carried out in a vacuum, and are also below those estimated by PAUL and LIPSCHUTZ (1990).

MIYAMOTO (1991b) reported that both the 3 and 6.9  $\mu$ m bands disappear in the specta of the Murchison samples heated above 500°C, suggesting that metamorphic temperature is  $\geq$ 500°C for thermally metamorphosed Antarctic carbonaceous chondrites, because they show weaker absorption bands near 3 and 6.9  $\mu$ m compared with Murchison (Fig. 2).

Assuming that the integrated intensities of the thermally metamorphosed carbonaceous chondrites are related to the amount of primary hydrous phases, that is, metamorphic temperatures, the thermal metamorphism becomes stronger in the order, Y-793321<Y-82162<Y-86720<B-7904. B-7904 shows a significantly weaker integrated intensity compared with the other three meteorites (Table 1), apparently consistent with the result by AKAI (1990a, b). However, these results are not reliable, because the integrated intensities of the 3  $\mu$ m bands of thermally metamorphosed carbonaceous chondrites are probably dependent on secondary hydrous minerals produced by terrestrial weathering.

Thermal metamorphic temperatures of meteorites give important information on the early evolution of meteorite parent body(ies). For example, thermal metamorphic temperatures of ordinary chondrites placed some conditions on the size, structures, and thermal evolution of the parent body internally heated by the energy of <sup>26</sup>Al (MIYAMOTO *et al.*, 1981). Model calculations for thermal metamorphism of these carbonaceous chondrites similar to ordinary chondrites showed that the metamorphic temperatures proposed for the thermally metamorphosed Antarctic carbonaceous chondrites can be satisfied when the <sup>26</sup>Al/<sup>27</sup>Al ratio is  $5 \times 10^{-6}$ , which is a similar value to the ordinary chondrite parent body, for the bulk Al contents of CI and CM chondrites (MIYAMOTO, 1991b). This result implies that thermal metamorphism is a common phenomenon at the early evolution of planetary bodies. We need further studies to precisely determine thermal metamorphic temperatures of thermally metamorphosed carbonaceous chondrites.

In the case of internal heating model, the greater portion of the internally heated body experiences maximum attainable temperatures (MIYAMOTO, 1991b).

Therefore, carbonaceous chondrites which have not experienced intense thermal metamorphism must have originated from the surface portion, and thermally metamorphosed ones from the inner portion of the parent body. It is, therefore, important to study the location of thermally metamorphosed carbonaceous chondrites in their parent body(ies) compared with that of CI or CM chondrites to test the internal heating model.

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