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FT-IR SPECTROSCOPIC STUDIES OF MAJOR ORGANIC MATTER IN CARBONACEOUS CHONDRITES USING MICROSCOPIC TECHNIQUE AND COMPARISON WITH TERRESTRIAL KEROGEN

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Abstract: Infrared (IR) absorption spectra of major types of carbonaceous matter in four Antarctic carbonaceous chondrites (Y-74662, Y-791198, ALH-77307, Y-791717) and two non-Antarctic carbonaceous chondrites (Murchison, Allende) were taken using a microscopic FT-IR technique. Without any treatment for concentration of carbon, powdered samples of carbon-rich C2 chondrites such as Y-74662, Y-791198, and Murchison gave spectra of the carbonaceous matter with enough intensity by focusing the beam on carbonaceous aggregates. Carbon-poor C3 chondrites (ALH-77307, Y-791717, and Allende) necessitated carbon enrichment by acid treatment to give IR spectra of carbonaceous matter of adequate intensity. The IR spectra indicated the presence of similar functional groups in the major types of carbonaceous matter of all of the investigated chondrites. Some natural coals (typical terrestrial kerogen) show very similar IR spectra to the carbonaceous matter of carbonaceous chondrites.

1. Introduction

Investigations of carbonaceous components in carbonaceous chondrites are very important to understand not only a key step in the sequence of events leading to the origin of life but also presolar and solar nebula conditions. The structure of carbonaceous compounds changes very smoothly according to the environment, but retains some information about the environmental conditions before the change in the structure (KERRIDGE, 1993). The major components of the carbonaceous matter are in the form of a poorly characterized macromolecular complex resembling terrestrial kerogen (HAYATSU *et al.*, 1983). This kerogen-like compound may account for from about 70 percent to essentially all of the meteoritic carbon (HAYES, 1967; HAN *et al.*, 1969; MING *et al.*, 1989).

The findings of enrichment of ^{13}C (SWART *et al.*, 1983; KERRIDGE *et al.*, 1987; MING *et al.*, 1988; MING and ANDERS, 1988a, b) and deuterium (ROBERT and EPSTEIN, 1982; KERRIDGE, 1983; KERRIDGE *et al.*, 1987) in certain fractions of the carbonaceous matter and the ability of the carbon to serve as the host phase for isotopically anomalous noble gases (LEWIS *et al.*, 1975; OTT *et al.*, 1981; MING *et al.*, 1988; MING and ANDERS, 1988a, b) have raised the possibility of an extrasolar origin for at least part of the carbonaceous matter (LEWIS *et al.*, 1975; OTT *et al.*,

1981., ROBERT and EPSTEIN, 1982; MING and ANDERS, 1988b). Therefore, elucidation of the structure of the major types of carbonaceous matter is essential to understanding primordial carbon chemistry as well as the origin and chemical history of the carbonaceous chondrites.

Although a number of investigators have applied a variety of analytical methods in order to characterize the major carbonaceous matter in carbonaceous chondrites (SCHULZ and ELOFSON, 1965; HAYES and BIEMANN, 1968; SIMMONDS *et al.*, 1969; GREEN II *et al.*, 1971; BREGER *et al.*, 1972; LEVY *et al.*, 1973; BANDURSKI and NAGY, 1976; HAYATSU *et al.*, 1977, 1980; HOLZER and ORÓ, 1979; WHITTAKER *et al.*, 1980; SMITH and BUSECK, 1981, 1982; CRONIN *et al.*, 1987; MURAE *et al.*, 1987, 1990), the structure of the carbonaceous matter is not well understood, because no method is completely satisfactory for characterization of irregular carbon-containing structures in the solid state. Some results indicate the presence of structural resemblance between the major types of carbonaceous matter in Allende and poorly ordered graphite (GREEN II *et al.*, 1971; SMITH and BUSECK, 1981, 1982), nevertheless, other results suggest that an amorphous nature similar to that of terrestrial kerogen is the predominant character of the major types of carbonaceous matter of most of the carbonaceous chondrites (SCHULZ and ELOFSON, 1965; SIMMONDS *et al.*, 1969; BANDURSKI and NAGY, 1976; HAYATSU *et al.*, 1983).

The similarity of basic chemical characteristics among the major types of carbonaceous matter in Murchison carbonaceous chondrites (meteoritic organic polymer) and vitrinite macerals of low volatile bituminous coals or type III mature kerogen has been pointed out by HAYATSU *et al.* (1983) on the basis of chemical degradation experiments. However, no direct comparison between carbonaceous matter of carbonaceous chondrites and terrestrial kerogen has been carried out. In this paper, direct comparative studies of the chemical structures detectable by IR spectroscopy are described for carbonaceous chondrites of different classes and for coals of different types.

Although IR spectra give very useful information on the chemical structure of organic matter, especially on functional groups, there are few reports on IR spectra of carbonaceous matter in carbonaceous chondrites (SANDFORD and WALKER, 1985; MURAE *et al.*, 1990). This is due to the technical difficulty of determination of IR spectra of black powdered samples. The author determined IR spectra of acid-resistant residues from ALH-77307 using an FT-IR spectrometer equipped with a micro-sample cup diffuse reflectance attachment (MURAE *et al.*, 1990). Although the carbon concentration of the acid resistant residue was high (C: 45.5%), the intensity of the spectrum was very poor even after considerable spectral accumulation. This is probably due to 1) too small amount of sample, 2) small number of functional groups compared with molecular size of the high molecular organic compound, and/or 3) absorption and/or reflection by coexisting minerals.

Recent developments in FT-IR spectrometry using a microscope have enabled us to obtain information about the chemical structure of very small organic particles. The major types of carbonaceous matter, high molecular organic kerogen-

like compounds, usually aggregate with each other in meteorites. Therefore, if the concentration of carbon is enough, direct observation of IR spectra of the carbonaceous matter may be possible using a microscopic FT-IR technique. In this paper the author reports the results of microscopic FT-IR measurements of the carbonaceous matter in powdered meteorite samples before and/or after concentration of the kerogen-like matter by acid treatment. The results are compared with the IR spectra of several types of coal, typical forms of terrestrial kerogen, determined under the same measuring conditions.

2. Samples and Analytical Methods

The types of carbonaceous chondrites used in this work and their carbon contents are shown in Table 1. Most of the samples were remains of powdered meteorites samples prepared for previous work and stored in sealed vials (MURAE *et al.*, 1984, 1987, 1991). The acid resistant residues from Allende were prepared by the following procedure: soak in 20% HCl at room temperature and centrifugally separate the residues (repeated three times); wash with water and centrifugally separate; repeated digestion with 38% HF and centrifugally separate (three times); wash with water and then with methanol; and digest with a 2 : 1 mixture of benzene and methanol (three times). The acid resistant residues from Yamato (Y)-791717 were also prepared by the same procedures as those mentioned above for the residues from Allende. The preparation procedure of the acid resistant residues from Allan Hills (ALH)-77307 was described in a previous paper on spectroscopic studies of carbonaceous matter in carbonaceous chondrites (MURAE *et al.*, 1990).

Coal samples (Table 2) were selected from the collection of Professor A.

Table 1. Carbon contents of the acid untreated and the acid treated carbonaceous chondrite samples used in this work.

Carbonaceous chondrites	Class	Original carbon content*	Acid treatments	Carbon content after acid treatments
Allende	C3(V)	0.2%	HCl-HF	1.0%
Murchison	CM2	2.0%	—	
ALH-77307	C3(O)	0.7%	HCl-HF	45.57%
Y-791717	C3(O)	0.1%	HCl-HF	1.0%
Y-74662	CM2	1.9%	—	
Y-791198	CM2	2.2%	—	

* Cited from MURAE *et al.* (1991).

Table 2. Coal samples used in this study.

Coal field or coal mind	Place	Coal type	Maceral type	Spectrum
Deker	Montana (U.S.A.)	Subbituminus	Vitrinite	4a
Wasatch Plateau	Utach (U.S.A.)	Subbituminus	Vitrinite	4b
Blair Athol	Queensland (Australia)	Subbituminus	Fusinite	4c
Hosmer (Crow Nest)	British Columbia (Canada)	Bituminus	Fusinite	4d

AIHARA, Laboratory of Organic Earth Science, Department of Earth and Planetary Sciences, Faculty of Science, Kyushu University. Small chips of coal were powdered. The organic matter in the particles remaining between the sieves of 200 and 20 mesh were separated from minerals by gravity separation using a zinc chloride aqueous solution of 1.6 specific gravity. Floating particles were collected, washed with water and then dried at 60°C. The dried particles were powdered again into fine particles and used for measurement of IR spectra.

IR spectra were recorded with a Perkin Elmer 1600 FT-IR spectrometer equipped with a Spectra-Teck IR-Plan analytical microscope at a resolution of 4 cm^{-1} . The full field of view is 800 μm in diameter. The aperture blades were rotated and adjusted so that the aggregated particles were defined as a major object in the field of view. Depending on the sample, the field was altered between 40 $\mu\text{m} \times 40 \mu\text{m}$ and the full field of view.

Less than 1 mg of powdered sample was placed on an aluminum foil plate having a diameter of 6 mm and thickness of 240 μm (made of 16 foils, each foil having a thickness of 15 μm), covered with single aluminum foil having a thickness of 15 μm , and then pressed with a wrench using a dual-bolt pellet press. This operation makes a very thin spot of the sample on the surface of the aluminum foil. The pressed disk was uncovered and placed on the microscope stage. The microscope was operated in reflectance mode. Aperture blades were adjusted if it was necessary to limit the objectives. After collecting a sample spectrum, background acquisition was carried out on the same disk at a place where no sample was observed. The ratioed (background subtracted) spectrum was almost the same as that of a usual transmission absorption spectrum.

3. Results and Discussion

Recent developments in FT-IR spectroscopy have enabled us to measure spectra of black powdered materials. Although the diffuse reflectance method is very useful for measurement of such samples, selective measurement of an IR spectrum of organic matter mixed with other IR-opaque minerals in a powdered sample is difficult. If the different types of organic matter aggregate with each other, then selective measurement of the organic matter IR spectrum may be possible by application of microscopic methods. The concentration of carbon in Y-791717 (C3) is as low as 0.2%, and it is difficult to observe a spectrum of carbonaceous matter. However, in the carbon enriched sample prepared from the meteorite by acid treatment, we could find some parts which gave an IR spectrum of carbonaceous matter having considerable intensity.

Figure 1 shows IR spectra observed at different spots on a disk holding the residues (C: *ca.* 1%) prepared from Y-791717 (C: *ca.* 0.2%) with the acid treatment. The IR spectrum of major parts of the residues showed the feature such as 1a. Most types of organic matter have characteristic complex absorption bands in the so-called "fingerprint region" (1300–900 cm^{-1}) (SILVERSTEIN *et al.*, 1991). The absorptions at *ca.* 3500 (O-H stretching vibration) and *ca.* 1630 (O-H bending

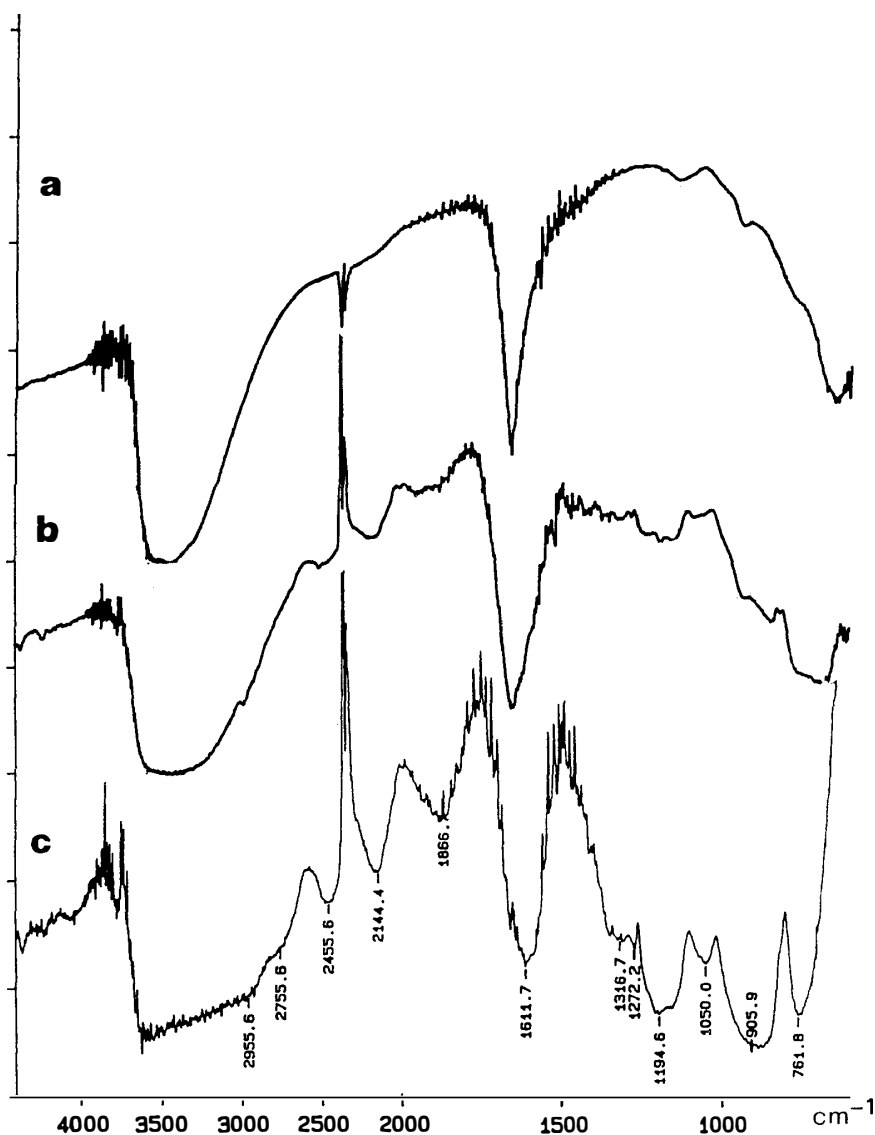


Fig. 1. IR spectra taken at three different spots on an aluminum plate holding a residue obtained from acid treatment of Y-791717. a: Spectrum recorded at major spot, b: spectrum recorded at the area containing few dark spots, c: spectrum obtained at a dark spot.

vibration) cm^{-1} of 1a may be due to minerals which contain loosely adsorbed water molecules (VAN DER MAREL and BEUTELSPACHER, 1976). The signals at *ca.* 2330 and 2360 cm^{-1} are due to carbon dioxide (all spectra in this work exhibit the signals caused by atmospheric background CO_2 in positive or negative form). In the spectrum 1b, some weak peaks due to absorption of organic matter overlap the major absorptions due to minerals observed in the spectrum 1a. These weak peaks are enhance in spectrum 1c to become major peaks. The absorption at *ca.* 1600 cm^{-1} (different from that at *ca.* 1700 cm^{-1} in spectra 1a and 1b) suggests the presence of aromatic rings. No peak due to carbon-hydrogen bonds is observed at around 3000 and 1400 cm^{-1} . This means that the size of the aromatic network of the organic

matter in this sample is probably large, and that the contribution of C-H bond is negligible.

The author has prepared a highly demineralized sample from C3 carbonaceous chondrite ALH-77307 (MURAE *et al.*, 1990). The carbon concentration of this sample is 45.5%. The original carbon content of ALH-77307 was 0.74%, so the carbon was enriched 60 times in the demineralized residue. Therefore, we may expect to find highly aggregated carbonaceous matter in that sample. Indeed, no search of the carbonaceous part was necessary to obtain the spectrum, and only a few accumulation scans (4 times was enough) gave a good quality spectrum (Fig. 2b).

Figure 2a shows the spectrum of carbonaceous matter in residues (C: *ca.* 1%) obtained from Allende (C: *ca.* 0.2%) by acid treatment. Fig. 2c, which is the same spectrum shown in Fig. 1c, is shown for comparison. These three IR spectra in Fig. 2 are closely similar to each other. This fact suggests that the major types of carbonaceous matter in these C3 carbonaceous chondrites contain very similar functional groups and the common basic chemical structure constructed with aromatic components (*vide infra*).

The C3 carbonaceous chondrites required the carbon to be concentrated by acid treatment to give IR spectra of the major types of carbonaceous matter with enough intensity. The concentration of carbon in C2 carbonaceous chondrites is one order higher than that in C3 carbonaceous chondrites in general. The distribution of carbon in C2 chondrites is not uniform; therefore, the major types of carbonaceous matter may aggregate each with other to form a mass where we can observe IR spectra with enough intensity. Although measurements of spectra of carbonaceous matter in the C3 chondrite ALH-77307 without enrichment of carbon by demineralization was unsuccessful, in the C2 chondrites the parts mainly composed by carbonaceous matter were found even in the acid-untreated powdered sample.

The IR spectra of carbonaceous matter in acid-untreated C2 chondrites Y-74662 (3a), Murchison (3b), and Y-791198 (3c) are shown in Fig. 3. The spectrum of the highly-carbon-enriched sample from ALH-77303 (3d: the same spectrum as 2b) is also shown in Fig. 3 for comparison. All of the spectra in Fig. 3 resemble each other. This means that the major types of carbonaceous matter in all of the carbonaceous chondrites investigated in this work have a common structure. And most of the IR active functional groups of the carbonaceous matter are same in all carbonaceous chondrites.

The resemblance of basic chemical structure between the major types of carbonaceous matter in Murchison carbonaceous chondrites (meteoritic organic polymer) and vitrinite macerals of low volatile bituminous coals or type III mature kerogen has been pointed out by HAYATSU *et al.* (1983) on the basis of chemical degradation experiments. No work that claimed that the insoluble carbonaceous matter in carbonaceous chondrites is amorphous in nature has indicated the chemical structure. The ¹³C NMR study by CRONIN *et al.*, (1987) showed similarity of the features of carbon spectra contained in the acid resistant residues from carbonaceous chondrites belonging to different types (C1, C2 and C3). Although

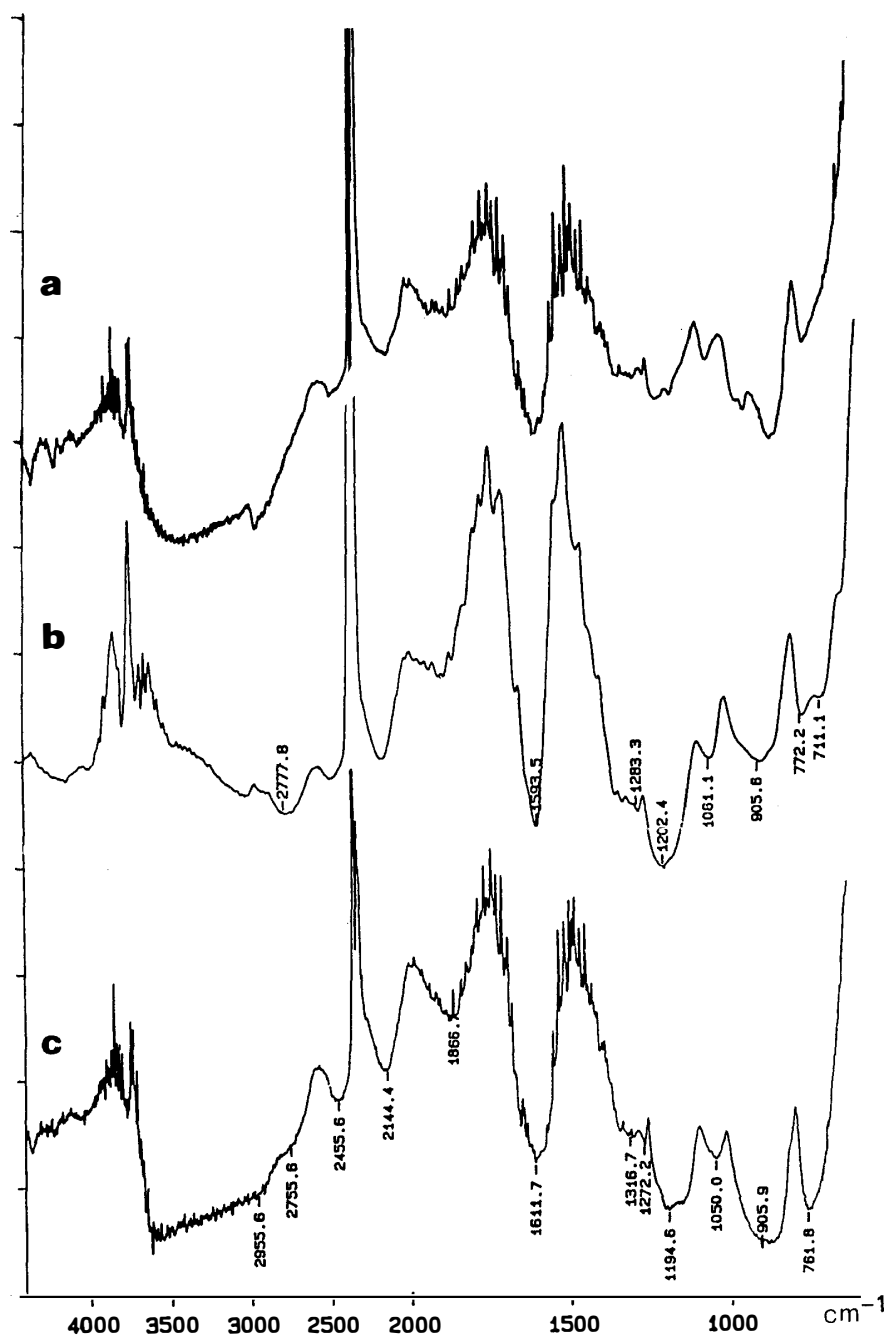


Fig. 2. IR spectra of carbonaceous matter in residues obtained by acid treatments of a: Allende (C: 1.0%), b: ALH-77307 (C: 45.5%), and c: Y-791717 (C: 1.0%; the same spectrum as in Fig. 1c).

this fact suggests that the major types of carbonaceous matter in different carbonaceous chondrites have common carbon skeletal structures, it is difficult to obtain information on the structures including the various functional groups.

The chemical structure of coal macerals is highly complicated. The coal macerals from different coal mines usually have different chemical structures, even if the coals are classified into the same class. The discrepancies are due to the

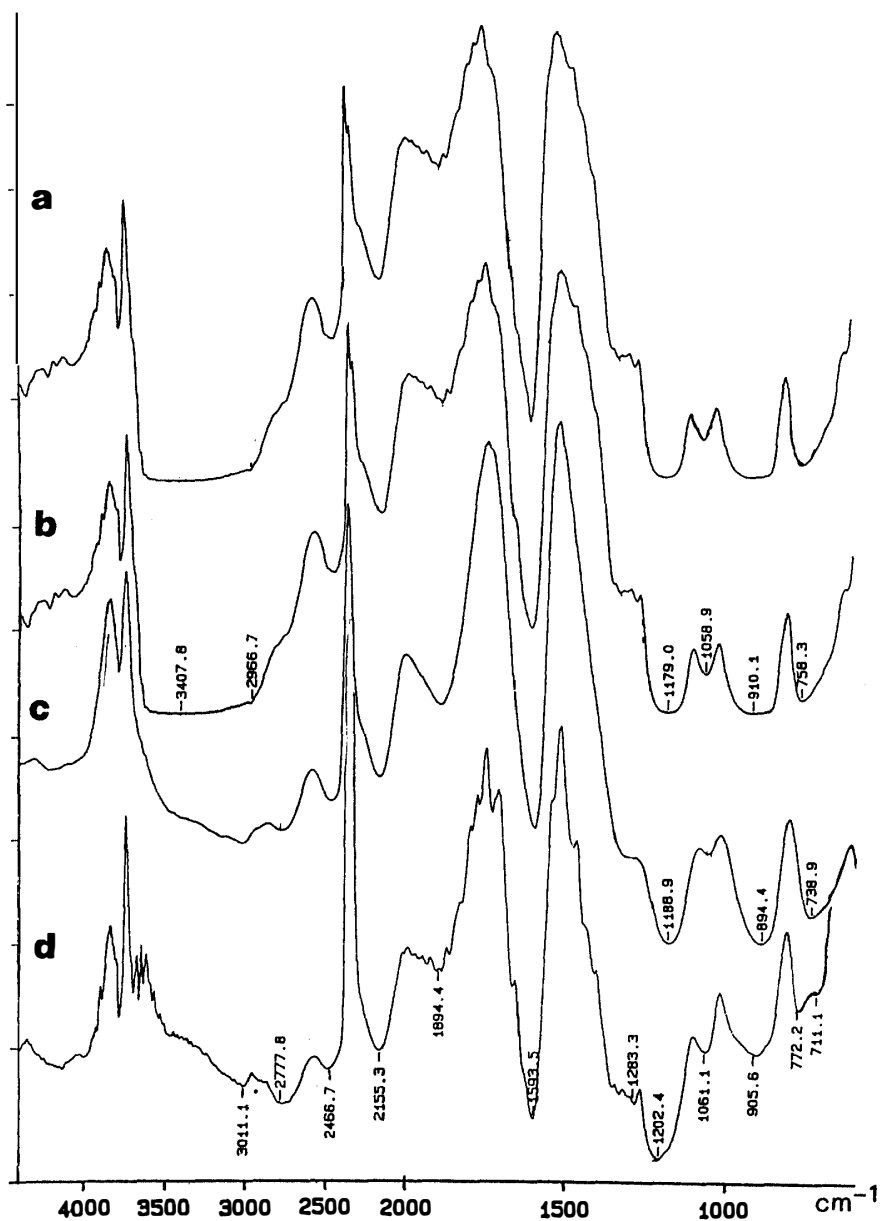


Fig. 3. IR spectra of carbonaceous matter in powdered samples without any mineral dissolution treatments of a: Y-74662 (C: 1.9%), b: Murchison (C: 2.0%), c: Y-791198 (C: 2.2%), and d: acid resistant residues (C: 45.5%) from ALH-77307 (the same spectrum as in Fig. 2b).

differences of original plants, maturation processes and maturation degrees. However, according to the progress of maturation of the coals, the structural variety diminishes, and the number of common structural factors increases, therefore, the chemical structures of mature coals resemble each other.

IR spectra reflect the difference of the chemical structure of the macerals very clearly, as shown in Fig. 4. The correct assignment in the "fingerprint" region between $1300\text{--}700\text{ cm}^{-1}$ is difficult on account of the various possibilities of structural arrangements of the organic molecule functions. There is an overlapping of the

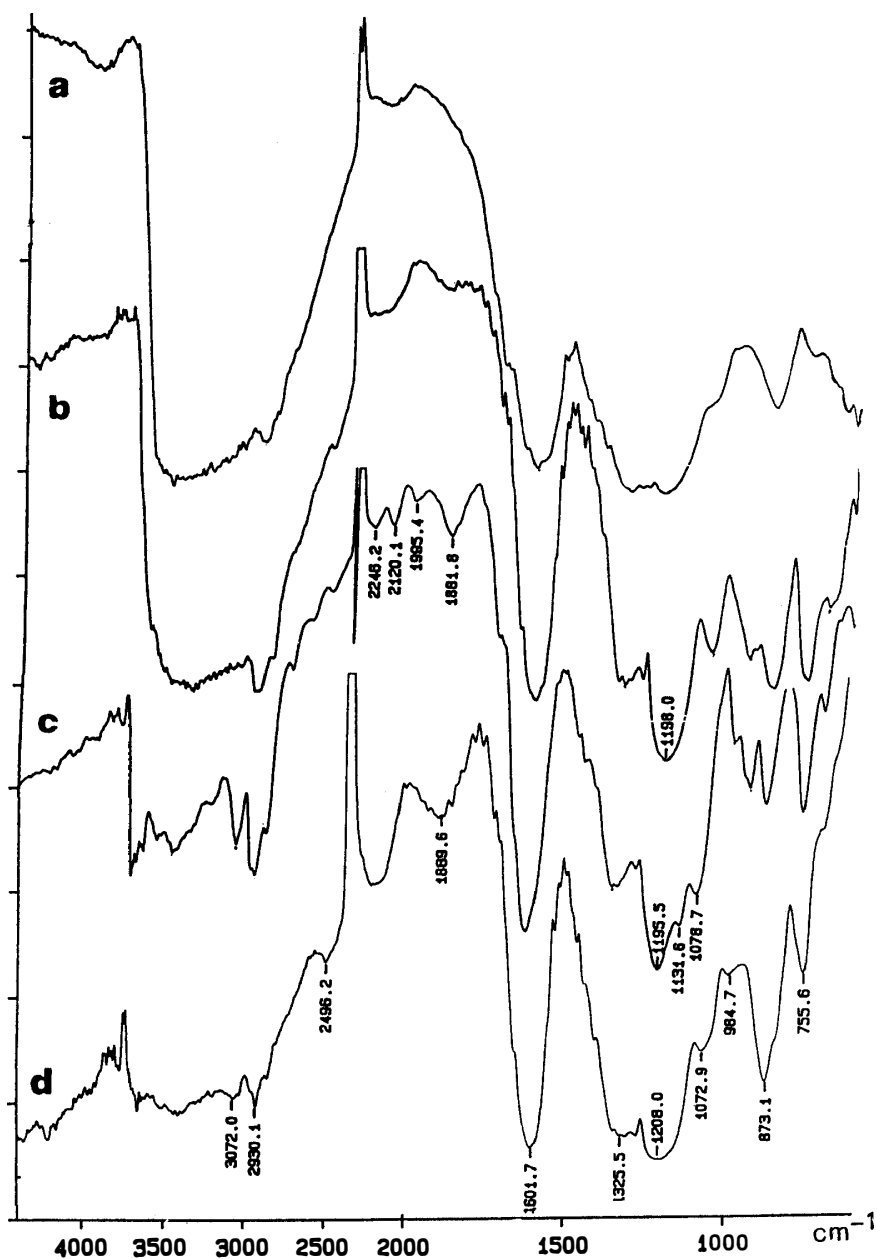


Fig. 4. IR spectra of coal macerals, a: Decker vitrinite, b: Wasatch Plateau vitrinite, c: Blair Athol fusinite, d: Hosmer fusinite.

bands with a large decrease in the wave numbers and the absorption intensities as the coalification process proceeds: especially for *ca.* 2950 cm^{-1} (νCH_3); *ca.* 2920 cm^{-1} (νCH_2); *ca.* 2850 cm^{-1} (νCH_2); *ca.* 1700 cm^{-1} ($\nu\text{C}=\text{O}$ carboxylic acids, ketones); *ca.* 1622 cm^{-1} ($\nu\text{C}=\text{C}$ phenyl nucleus); *ca.* 1420 cm^{-1} (νOH alcohols); and *ca.* $1160, 1058, 1030\text{ cm}^{-1}$ ($\nu\text{C}-\text{O}$ bands of stretching of etherial and hydroxylic groups) (VAN DER MAREL and BEUTELSPACHERE 1976). Three of the four types of coal whose IR spectra are shown in Fig. 4 (4a, b and c) are classified into subbituminous coal type and one (4d) into bituminous coal type. In terms of macerals, 4a and 4b belong to vitrinite and 4c and 4d to fusinite. Conspicuous

chemical-structural changes of macerals during the maturation process are decreases in the number of substituents by reactions such as decarboxylation, decreases in the number of C-H bonds (dehydrogenation), and increases of the number of aromatic rings (VAN KREVELEN, 1961; RIESSER, 1984). Usually the increases of the number of aromatic rings are accompanied with enlargements of the sizes of condensed polycyclic aromatic core structures and with diminution of the aliphatic alkyl chain connecting the polycyclic aromatic cores.

The present author has proposed a general chemical structure for the major types of carbonaceous matter in carbonaceous chondrites on the basis of the results of pyrolysis GC-MS experiments (MURAE *et al.*, 1987) and on the basis of spectroscopic investigations using mass (MS) spectra, nuclear magnetic resonance (NMR) spectra, and infrared (IR) spectra (MURAE *et al.*, 1990). The proposed structure is constructed with a condensed polycyclic aromatic core and substituents around the core. The size of the condensed polycyclic aromatic core is larger than those of polycyclic aromatic hydrocarbons (PAHs) which were suggested to be present in interstellar space (ALLAMANDOLA *et al.*, 1985) and in meteorites (HAHN *et al.*, 1988).

The IR spectra of a subbituminous coal (4b) and a fossil char (4d) and acid resistant residues derived from ALH-77307 (2b) very closely resemble each other to suggest that the IR-observable chemical structures of the major types of carbonaceous matter in the carbonaceous chondrites studied in this work are almost the same as those of some coal macerals (terrestrial kerogen) of relatively high maturity degree. Most of the strong absorption bands in the spectra of both terrestrial kerogens (4b and 4d) and meteoritic kerogen-like matter (2b) can be assigned to a phenyl nucleus: overtone at *ca.* 1900 cm^{-1} , $\nu\text{C}=\text{C}$ at *ca.* 1600 cm^{-1} , $\nu\text{C}-\text{H}$ (in plane) at *ca.* 1200 cm^{-1} , $\nu\text{C}-\text{H}$ (out-of-plane) at *ca.* 900 cm^{-1} . Although the terrestrial kerogens (4b and 4d) show absorption due to aliphatic C-H bonds, the meteoritic kerogen-like matter (2b) shows only absorption due to aromatic C-H bonds. The absorption due to aromatic C-H bonds is not observed in the spectra of meteoritic kerogen-like matter (3a and 3b). This fact along with the variations of the relative intensities of the absorption due to the aromatic skeleton may be interpreted by variations of aromatic nucleus size. These facts are compatible with the structure previously proposed for the major types of carbonaceous matter in carbonaceous chondrites by the author (MURAE *et al.*, 1987).

4. Summary

The major types of carbonaceous matter in C2 carbonaceous chondrites (Murchison, Y-74662, Y-791198) and C3 carbonaceous chondrites (Allende, ALH-77307, Y-791717) were examined by FT-IR spectroscopy using microscopic technique in diffraction mode. The C3 meteorites needed carbon enrichment to give the IR spectra of the carbonaceous matter. The IR spectra of the carbonaceous matter in C2 chondrites were obtained without any carbon enrichment. All of the spectra thus obtained closely resemble each other, suggesting that the major types of

carbonaceous matter in those carbonaceous chondrites have a common chemical structure with polycyclic aromatic rings at its core. The IR spectra measured by the same methods for some types of coal revealed that the structure of some mature terrestrial kerogen has structures similar to that of organic high molecular matter in the carbonaceous chondrites.

The structural similarity between the terrestrial kerogen in a limited maturation stage and kerogen-like matter in carbonaceous chondrites of different classes is revealed by direct comparison of IR spectra (this work) for kerogen-like matter in various Antarctic and non-Antarctic CM2, C3(V), and C3(O) carbonaceous chondrites and coal samples, by ^{13}C -NMR spectra (CRONIN *et al.*, 1987) for Orgueil (C1), Murchison (CM2), and Allende (C3(V)), and by chemical degradation experiments (HAYATSU *et al.*, 1983) for Murchison (CM2). The structural complexity of terrestrial kerogens originates to biological syntheses of the molecules and the following geological processes. The maturation processes simplify the structure of kerogens to yield graphite finally. Although the mechanisms which produce the complexity of the structures of kerogen-like matter in carbonaceous chondrites are not known, the similarity of their structures in carbonaceous chondrites belonging to different classes suggests that these types of carbonaceous matter may have been formed before the chondrites, and may not be affected so seriously by subsequent alteration processes.

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