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FLUORESCENT ORGANIC MATTER IN CARBONACEOUS CHONDRITES

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ABSTRACT

Fluorescent organic matter in carbonaceous chondrites was investigated using a microscope equipped with a fluorescence spectrophotometer. Fluorescent particles were observed in powdered CM2 carbonaceous chondrites (Y-74662, Y-7791198, and Murchison) without carbon enrichment by acid treatments. Although it was difficult to find fluorescent particles in powdered sample of C3 chondrites (ALH-77307, Y-791717, and Allende) without acid treatments, many fluorescent particles were observed after carbon enrichment by acid treatments. Fluorescence of coronene and shock-altered graphite were observed using the same microscope and the same conditions as those for carbonaceous chondrites.

INTRODUCTION

The presence of fluorescent carbonaceous matter in carbonaceous chondrites has been noticed more than 35 years ago (Briggs, 1961; Claus and Nagy, 1961). Briggs detected three types of fluorescent organic compounds in the hot-water extract of powdered Mokoia meteorite (Briggs, 1961). Two of them are strongly fluorescent (yellow) in the short ultra-violet light. These strongly fluorescent compounds were separated by paper chromatography. The fluorescence of both these compounds is unaffected by pH. The fluorescent properties of these compounds suggest some conjugated ring system; while the water-solubility of all samples indicate the presence of polar carboxyl and hydroxyl groups. The third compound was weakly fluorescent.

Claus and Nagy (1961) reported the existence of many particles showing fluorescence (yellow) in ultraviolet light in crumbled grains of Orgueil and Ivuna. The fluorescent particles did not show birefringence in polarized light. The fluorescent particles were absent in noncarbonaceous chondrites, Holbrook and Bruderheim (Claus and Nagy, 1961). Fich *et al.* (1962) claimed that the fluorescent particles are troilite globules, sulfur droplets, or hydrocarbon droplets.

Acid treatments using hydrofluoric acid and hydrochloric acid did not seriously affect the morphology of these particles (Nagy *et al.*, 1962). A significant portion of these fluorescent particles showed relatively high density (densities between 2.3 and 2.4). Certain organic solvents appear to affect, but not to dissolve, these particles, eliminating the possibility that they are composed of sulfur, hydrocarbons, or of aggregates of low molecular weight organic compounds.

Alpern and Benkheiri (1973) reported that auto-fluorescence of organic constituents in Orgueil was obtained by excitation in blue light on polished, non-cemented and non-etched sections. The organic constituents were mostly circular bodies of one micron or less, regularly distributed in the phylitic matrix. And they suggested the possibility that the organic constituents might have resulted from the condensation of hydrocarbons on inorganic particles.

Deamer (1985) reported the observation of numerous yellow fluorescent particles in the micrometer size ranging from limit of resolution to ~10 μm as well as larger structures with deep red fluorescence in a Murchison meteorite sample immediately after fracture, and after gentle smoothing by a diamond-coated polishing disk. The yellow fluorescent particles were substantially reduced in number following a brief chloroform rinse, and these presumably represent the *in site* appearance of the organic components of the Murchison chondrite.

In the Nagy's report, the fluorescent particles in carbonaceous chondrites were considered to be the remains of microorganisms. However, other investigators (Briggs, 1961; Fich et al., 1962; Alpern and Benkheiri, 1973; Deamer, 1985) reported the fluorescent particles as organic components. Macro-size polycyclic aromatic sheet bearing edge defects was proposed as the structure of a major part of carbonaceous compounds in carbonaceous chondrites (Murae, 1997). Polycyclic aromatic hydrocarbons (PAHs) showing auto-fluorescence, such as naphthalene and phenanthrene, may be contained in the defect parts as substituents. If so, fluorescent particles will be probably found in most of carbonaceous chondrites.

EXPERIMENTALS

The samples of carbonaceous chondrites investigated in this work are four Antarctic meteorites and two non-Antarctic meteorites. Two of the Antarctic meteorites (Y-791198 and Y-74662) and one of non-Antarctic meteorite (Murchison) are classified into type CM2, and other meteorites (Y-791717, ALH-77307, and Allende) are classified into type C3. The meteorites were crushed roughly and washed with hexane. The washed ramps were powdered into fine particles, which were extracted with benzene-methanol (2:1) for 12 hrs using a Soxhlet extractor. Partial mineral dissolution of C3 carbonaceous chondrites (Y-77307, Y-791717, and Allende) was carried out by soaking in 20% hydrochloric acid at room temperature for 3 days, washing with water and centrifugal separation, repeated digestion with 38% hydrofluoric acid, and centrifugal separation. Washing with distilled water was followed by drying under air stream.

Less than 1mg of powdered sample was placed on an aluminum foil plate having a 6mm diameter and 240μm thickness (made of 16 foils, each foil having a thickness of 15 μm). The plate was covered with a single aluminum foil having a thickness of 15 μm, and then pressed with a wrench using a dual-blot pellet press. The pressed disk was uncovered and placed on a microscope stage. The microscope (Zeiss, MPM 400 microscope photometer) was equipped with plan-neofluar (x40) as an objective, and operated with reflectance mode. The fluorescence was observed under excitation with UV light from a mercury lump through a blue filter. The fluorescence spectra were recorded using a Zeiss lambda scan analysis program. Fine needle crystals of coronene (re-crystallized from toluene) were used as a typical example of PAHs. The carbonaceous matter recovered after shock experiments using a rail-gun was used as an example of macro-sized PAH like compounds bearing edge-defect structures (Murae, 1997).

RESULTS AND DISCUSSION

By the movement of the microscope, we could find fluorescent particles in all powdered samples of CM2 chondrites without any acid treatments. The number of the fluorescent particles was lower than reported by Claus and Nagy (1961) and Deamer (1985). Typical example of fluorescent particles in the powdered sample of Y-74662 is shown in figures 1**a** and 1**b**. Figure 1**a** shows a fluorescent particle observed under UV light. And figure 1**b** shows the view of the same part as figure 1**a** under polarized white light. The fluorescent particle seams to be an aggregate of smaller round fluorescent particles. The corresponding particle was difficult to find under white light. In the case of Murchison, a more yellowish fluorescent particle was observed (figure 1**c**). The shape of this particle was barely recognized as a translucent object

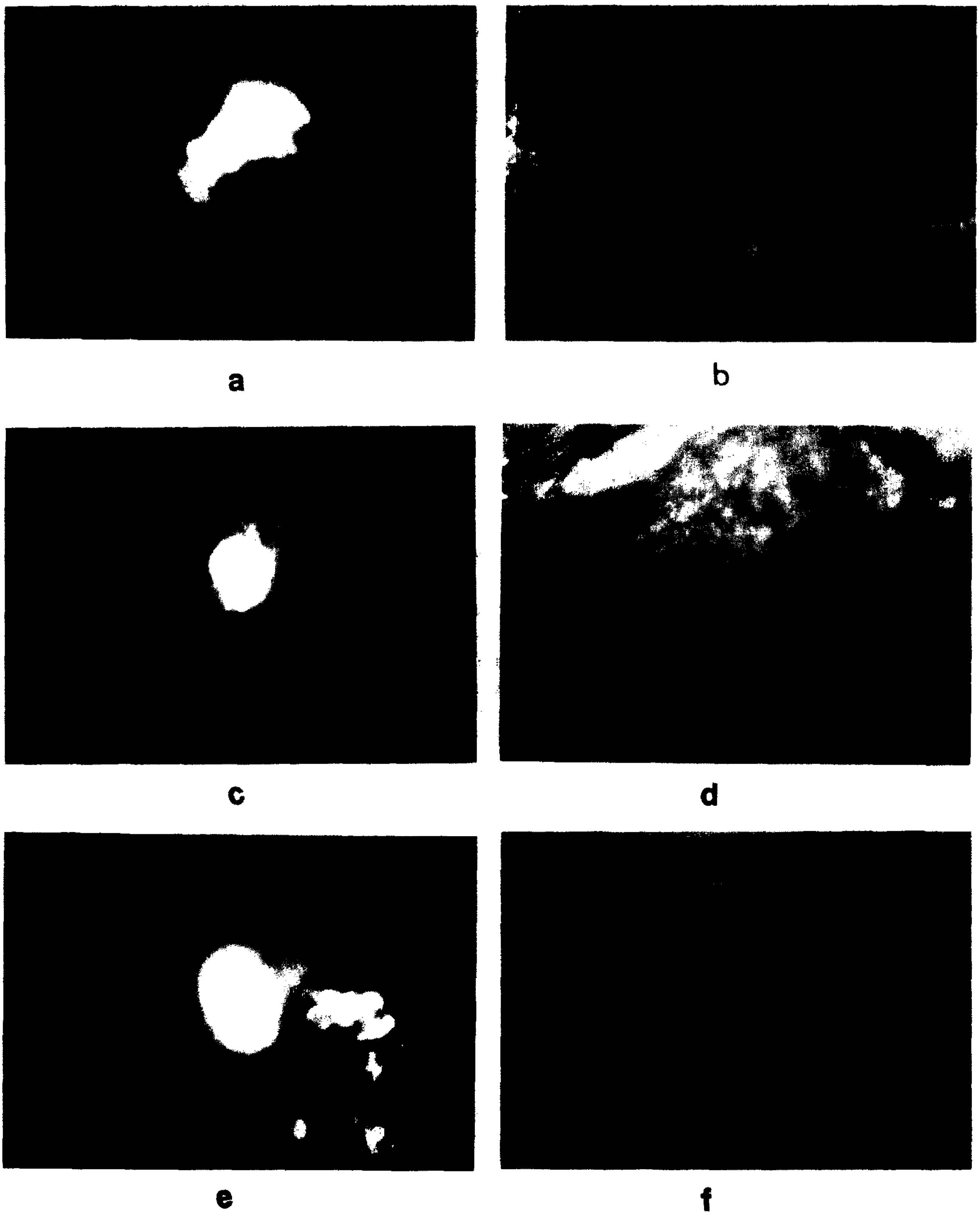


Fig. 1. Fluorescent particles in CM2 carbonaceous chondrites; **a** and **b**: Y-74662, **c** and **d**: Murchison, **e** and **f**: Y-791198. The photos **a**, **c**, and **e** were taken under UV light, and the photos **b**, **d**, and **f** show the same part observed under polarized white light, respectively.

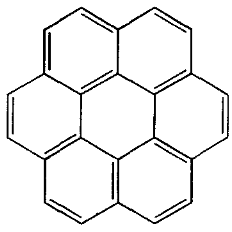
under polarized white light (figure 1**d**). A clearer shape (figure 1**e**) was observed under polarized white light for isolated fluorescent particles (figure 1 **f**) in the case of Y-791198.

The finding of fluorescent particles was very difficult in the samples of C3 chondrites without carbon enrichment by acid treatments. After acid treatments, a number of small fluorescent particles were observed for all of C3 chondrites. In the acid resistant residues from ALH-77307, we could find two types of fluorescent particles. One small particle and its aggregate (figure 2**a**) that may be observed as dark spots under polarized white light (figure. 2**b**). The other is a faint fluorescent irregular-shape particle (figure 2**c**) whose observation under polarized white light is difficult (figure 2**d**). The fluorescent matter observed in the acid resistant residues from Allende seams to be a mixture of very faint fluorescent part s and aggregates of relatively bright fluorescent particles (figure 2**e**), which may be recognizable under polarized white light (figure 2**f**).

The fluorescent compounds reported by Briggs (1961) was suggested to be the compounds constructed with some conjugated ring system with polar carboxyl, hydroxide, and amine groups. In our case, the fluorescent particles remained after solvent extraction; therefore these compounds may not be the same as those reported by Briggs (1961). Although the fluorescent particles that Deamer (1985) reported are soluble in chloroform, the fluorescent particles that we observed are insoluble in chloroform. The properties of the organized elements reported by Nagy *et al.* (1962) are closely related to those of macromolecular organic compounds that we have proposed as a major part of carbonaceous matter in carbonaceous chondrites. Nagy *et al.* (1962), however, reported the fluorescent particles as microfossils of extraterrestrial algae.

In order to confirm the fluorescent properties of polycyclic aromatic compounds under the same conditions, we prepared the samples of crystallized and powdered coronene (**I**), phenanthrene (**II**), and shock-altered graphite. The crystallized coronene was observed as fluorescent needles (figure 3**a**), and the intensity of fluorescence of the overlapping parts of crystals was very strong. The fluorescence intensity of a thin film of phenanthrene (figure 3**b**) is also high at the thick part. The coronene pressed on aluminum plate in the same way as that for meteorite powder was examined under the same microscopic conditions. Similar fluorescent pattern to the aggregated fluorescent particles in meteorites is observed under UV light (figure 3**c**), though a clear pattern can not be observed under polarized white light (figure 3**d**). Figure 3**e** indicates the presence of aggregated small fluorescent particles in shock-altered graphite (Murae, 1997). The aggregated feature of the particles may correspond to the pattern observed under polarized white light (figure 3**f**).

Fitch et al. (1962) observed a fluorescence maximum at 440nm in chloroform solution of a fluorescent extract from the Orgueil carbonaceous chondrite and reported that the fluorescent particles are super-cooled droplets of hydrocarbon. We have measured the fluorescent spectra of the samples of Y-74662, Murchison, acid-treated Allende and Y-791717, coronene (I), and phenanthrene (II) (figure 4) using microscopic technique.



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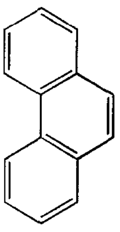

II

The fluorescence spectrum of fluorescent particles in Y-74662 chondrite (figure 4**a**) resembled that observed in the acid-resistant residue from Allende (figure 4**d**) showing an extinction maximum at about 480nm with broad tailing toward longer wavelength. The spectral feature of phenanthrene (**II**) (figure 4**f**) shows some similarity to that of Y-74662 (figure 4**a**) and acid treated Y-791717 (figure 4**d**). The peak top of the spectrum of a fluorescent particle showing pale orange color in Murchison chondrite (figure 4**b**) is shifted to the region of longer wavelength than other particles. The spectral feature of the particle (figure 4**b**) resembles that of coronene (**I**) (figure 4**e**). The spectral feature of the acid resistant residue from Allende (figure 4**c**) may indicate the mixing of the particles as observed in Y-74662 (or acid-resistant residue from Y-791717) with similar particles to those observed in the Murchison meteorite.

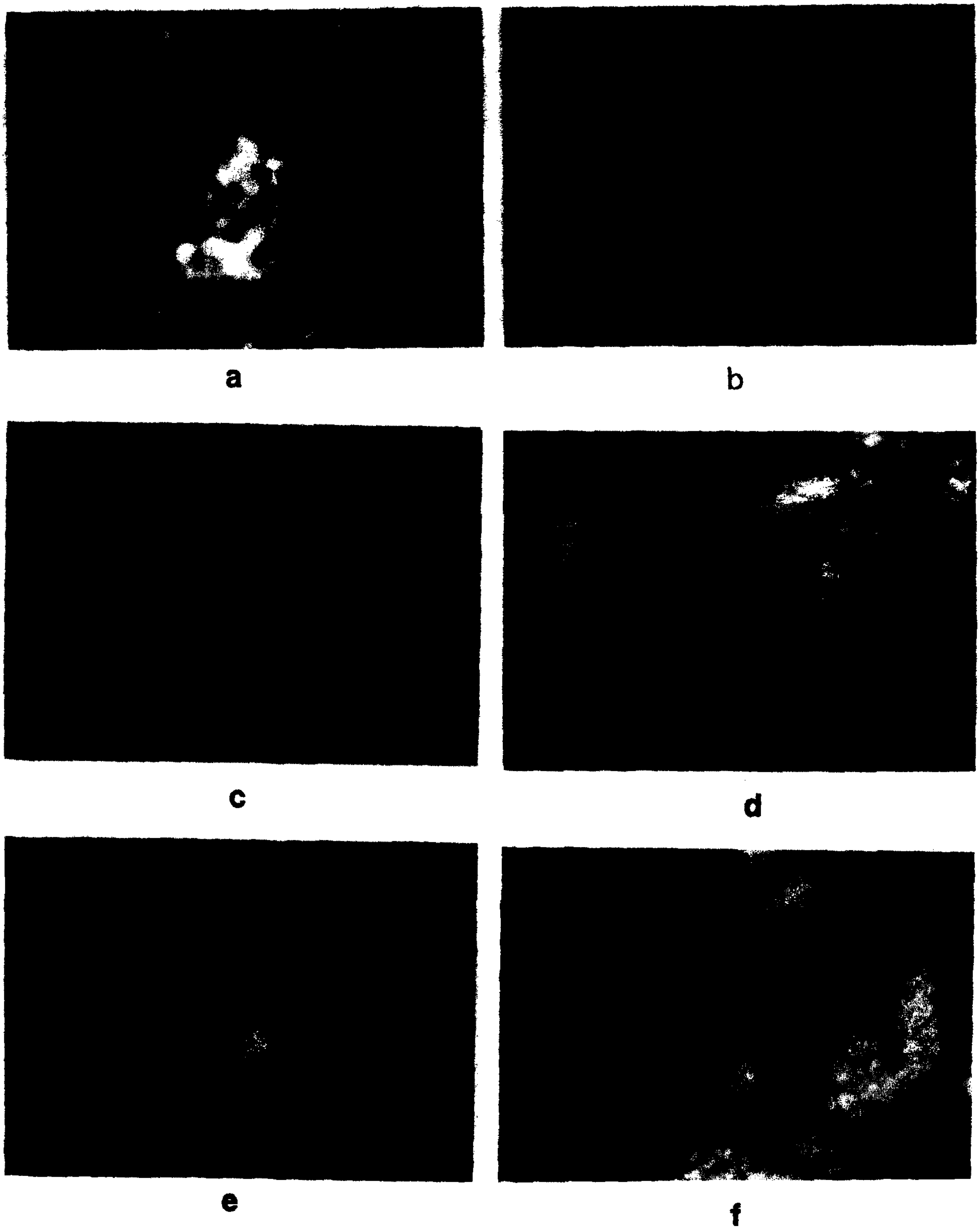

Fig. 2. Fluorescent particles in acid-resistant residues from C3 carbonaceous chondrites, **a**, **b**, **c**, and **d**: ALH-77307, **e** and **f**: Allende. The photos **a**, **c**, and **e** were taken under UV light, and the photos **b**, **d**, and **f** show the same part observed under polarized white light, respectively.

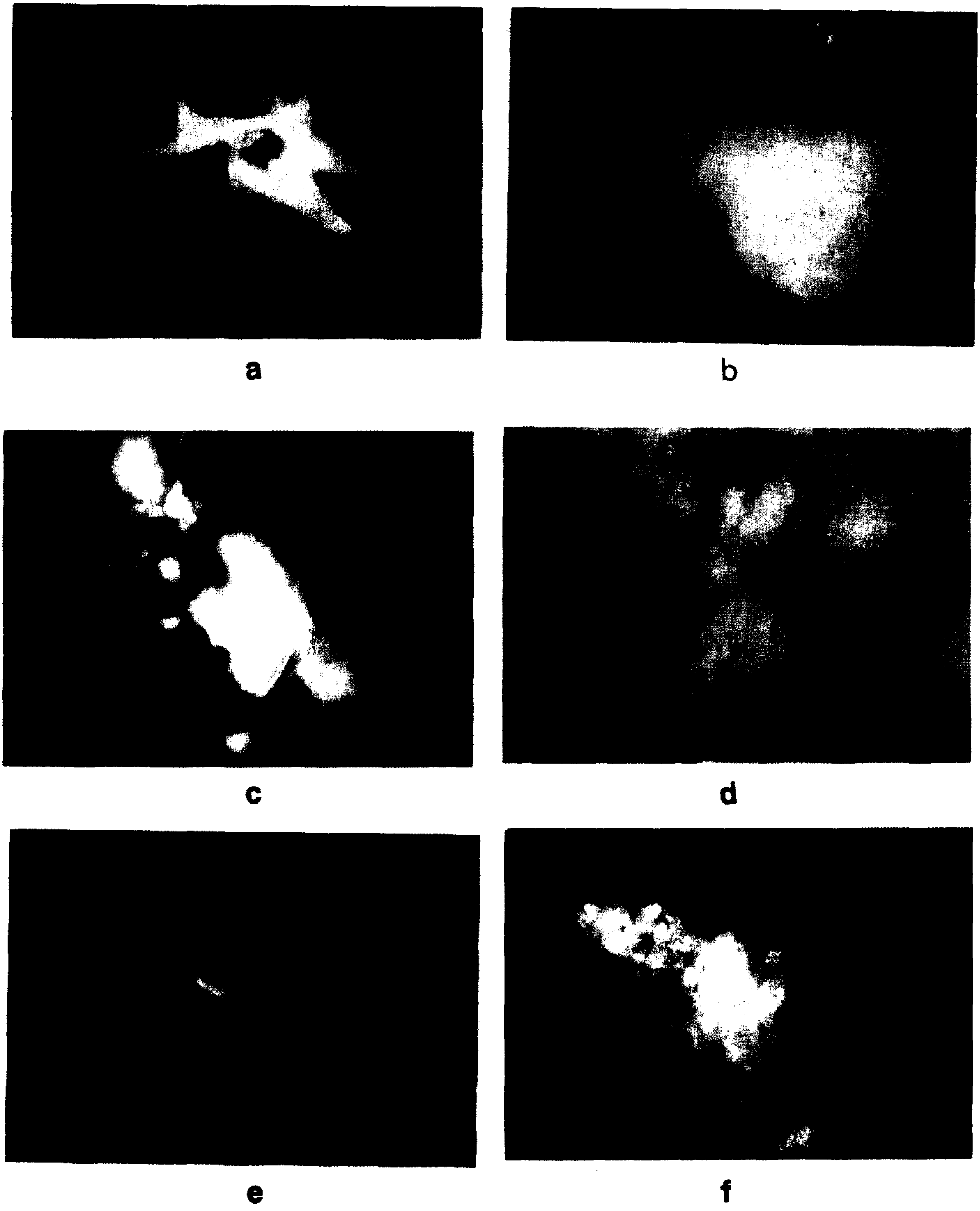

Fig. 3. Reference samples for fluorescent particles; **a**, **c**, and **d**: coronene, **b**: phenanthrene, **e** and **f**: shock-altered graphite. The photos **a**, **b**, **c**, and **e** were taken under UV light, and the photos **d**, and **f** show the same part as those of **c** and **e** observed under polarized white light, respectively.

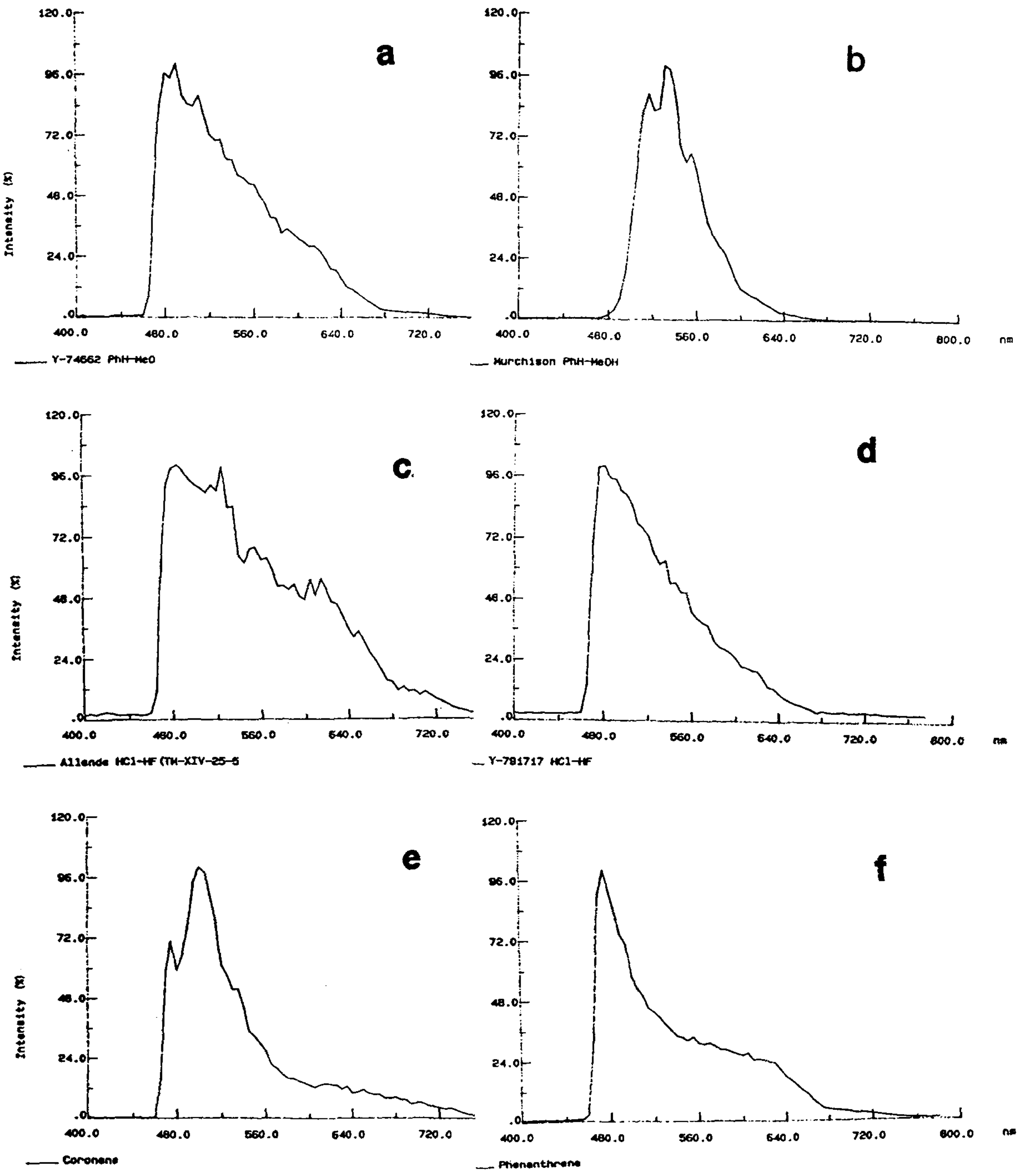

Fig. 4. Microscopic fluorescence spectra of fluorescent particles in **a**: Y-74662, **b**: Murchison, **c**: acid-resistant residues from Allende, **d**: acid-resistant residues from Y-791717, **e**: coronene, and **f**: phenanthrene.

We have proposed a model for the structure of high-molecular carbonaceous compounds that is a major part of carbonaceous matter in carbonaceous chondrites (Murae, 1997). The model is constructed with large polycyclic aromatic rings as main part along with some edge defects. The edge defects probably contain the aromatic rings showing fluorescent character.

SUMMARY

(1) Fluorescent particles were observed in all of the samples of powdered CM2 carbonaceous chondrites (Y-74662, Y-791198, and Murchison). (2) Although the observation of fluorescent particles in C3 carbonaceous chondrites (ALH-77307, Y-791717, Allende) was difficult, we could find fluorescent particles in acid-resistant residues remaining after acid treatments, followed by washing with water and organic solvents. (3) The features of fluorescence spectra showed some resemblance to those of coronene and phenanthrene. (4) The fluorescent properties of the fluorescent particles in carbonaceous chondrites may originate in the edge defects of high-molecular organic matter which accounts for a major part of carbonaceous matter in chondrites.

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