

LABORATORY STUDY ON THE FORMATION OF PAH CLUSTERS AND THEIR UV IRRADIATION EFFECTS USING ANTHRACENE.

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Introduction: PAHs (Polycyclic Aromatic Hydrocarbons) are most plausible carriers of unidentified infrared (UIR) bands observed from many different objects in various stages both young and evolved stars such as Herbig Ae/Be stars, HII regions, reflection nebulae, diffuse interstellar medium, post-asymptotic giant branch stars (AGB) and planetary nebulae. Since it has been reported that the UIR bands observed in those objects show systematically different spectral features [1,2], the physical environments and chemical evolution in those kinds of objects can be derived from the details of band positions, relative intensities and widths of UIR bands through a series of PAHs studies. In addition to the theoretical and observational studies, therefore, laboratory synthesis experiments of PAHs and their clusters are necessary to understand the formation and subsequent growth to the PAH clusters and their alteration by UV irradiation. In this experimental series, we elucidate a correlation between cluster size and IR features of produced anthracene clusters. Anthracene has a molecular structure constructed by three benzenes in straight. It has two solo C-H bonds and two quartet C-H bonds. UV was irradiated to the anthracene clusters and the IR spectral changes were observed.

Synthesis of anthracene clusters: Anthracene clusters were produced by evaporation of commercial powder from on an electrically heated molybdenum boat in He gas of 30, 80 and 150 Torr and collected on a glass plate placed 5 cm above the boat for measurements of spectra and on a standard Cu transmission electron microscope (TEM) grid supported with carbon holey film for observation of the size and the structure. As a result of TEM observation, we found that the size of the anthracene clusters was widely distributed from 50 nm to 5 μ m and the electron diffraction pattern showed Bragg reflection spots attributed to anthracene crystal. With increasing the He gas pressure, the size of the anthracene clusters becomes larger. A typical TEM image of anthracene clusters produced in He gas of 80 Torr has been shown in Fig. 1. The diffraction pattern from anthracene crystal was immediately getting weak under electron beam for 10^{-3} - 10^{-2} A/cm², which is 10 times weaker compared with typical intensity during common observation, with acceleration voltage at 100 keV and finally disappeared

within about 15 seconds, i.e., the crystal structure of anthracene was easily broken. Although the crystal structure of anthracene clusters is very sensitive to an electron beam, the external geometry has been kept after electron beam irradiation.

IR spectra of anthracene clusters: The produced anthracene clusters were embedded in KBr pellets by mixing directly on the glass plate. The mid-IR spectra of anthracene clusters produced in He gas of 30, 80 and 150 Torr showed many significant features including 11.31- and 13.77- μ m bands, which are attributed to the vibration of aromatic solo and quartet C-H out-of-plane bending mode, respectively, as shown in Fig. 2(a)-(c), respectively. The intensity of the peaks in the spectra from anthracene clusters produced in higher pressure is larger, because the amount of produced samples is larger in higher pressure. In these spectra, only 12- μ m band was newly shown up as increase gas pressure. In other words, it can be considered that the 12- μ m band was appeared by larger anthracene clusters. This 12- μ m band was attributed to very small carbonaceous grains with 350-600 C atoms [3] based on quantum chemical calculations and supportive observational results in the real astrophysical environments are firstly given by Buss et al. (1993) around the carbon rich objects in transition from the

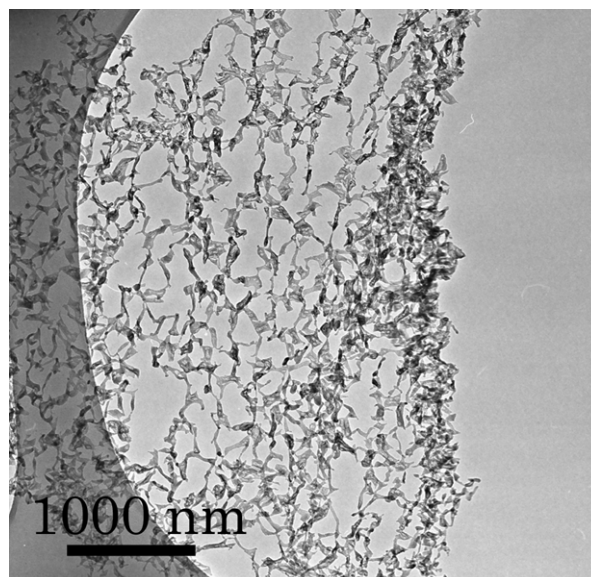


Fig. 1. Typical TEM image of anthracene clusters produced in He gas of 80 Torr. The film, which is seen in left side, is holey carbon film on TEM grid.

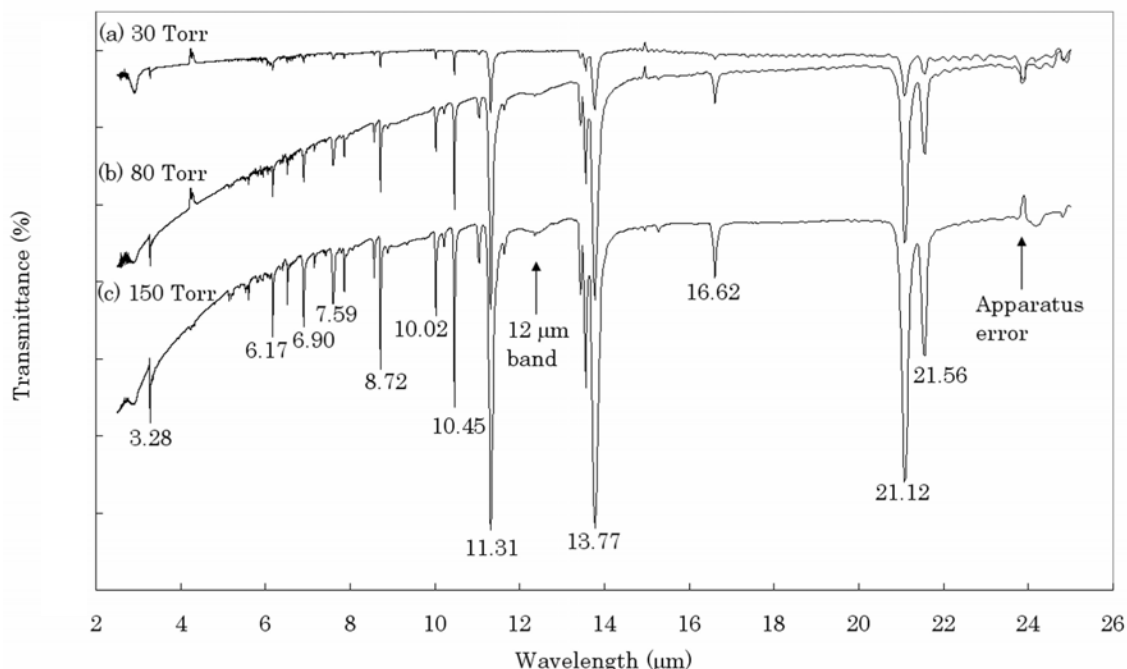


Fig. 2. Mid-IR spectra of anthracene clusters embedded in KBr pellets produced in He gas of 30, 80 and 150 Torr, respectively, were measured with a Fourier-transform IR spectrometer (Horiba Inc. FT210). The energy resolution used for this work was 2 cm^{-1} . The numbers show corresponding peak position. It has seen that the 12- μm bands were grown as increase of the gas pressure. These spectra have been shifted.

AGB to the planetary nebula stage of evolution [4]. Recently, Rapacioli et al. (2005) have found the growth of this 12-14 μm plateau in the photodissociation regions with the increasing distance from the UV source [5]. They have given an interpretation to their results that 12-14 μm plateau is suggested to be carried by carbonaceous very small grains or PAH clusters, which are easily photoevaporated into free-flying PAHs. Our results that the 12- μm band appears and grows as anthracene clusters grow have given an important evidence, for the first time in the laboratory experiments, such that the 12-14 μm plateau does originate in PAH clusters although our anthracene clusters have much more C atoms. This result may suggest that the origin of the plateau at 12-14 μm is not required PAH clusters constructed by large or mixture molecules.

Influence of UV irradiation: The anthracene clusters produced in He gas of 80 Torr were irradiated by UV at 254 and 365 nm for 15, 40 and 60 hours. The intensities at 254 and 365 nm of UV lamp is 61 and 74 mW/cm^2 , respectively. In the case of irradiation at 254 nm, almost all features shown in Fig. 1(b) such as at 8.72, 10.02, 10.45, 11.31, 13.77, 21.12 and 21.56 μm were gradually decreased as increase of the irradiation time. In contrast, many different bands at 8.19, 8.62, 9.70, 10.59, 12.27 and 14.64 μm were

newly appeared and the band at 16.62 μm was shifted to 16.72 μm and became intense. In the case of irradiation at 365 nm, those peaks were similarly changed as 254 nm irradiation whereas the changing was very fast compared with 254 nm. The features were completely disappeared by irradiation at 365 nm for only 15 hours, although initial bands had been remained after 60 hours of 254 nm irradiation. Namely, the wavelength at 365 nm was more affective to anthracene clusters than 254 nm in spite of weaker energy. Anthracene has roughly two absorption features at 210-290 and 310-440 nm in the range of 200 to 800 nm. The irradiated two different wavelengths correspond to the central wavelength of their absorption features. This result implies that the alteration of PAHs by radiation of UV is depending on the wavelength. Similar experiments have been performed to understand the growth mechanisms and optical properties of PAHs and their clusters using other PAHs, such as naphthalene, naphthalene, pyrene, chrysene and coronene.

References: [1] Peeters E. et al. (2002) *A&A*, 390, 1089-1113. [2] van Diedenhoven B. et al. (2004) *ApJ*, 611, 928-939. [3] Allamandola L. J. et al. (1989) *ApJS*, 71, 733-775. [4] Buss R. H. et al. (1993) *ApJ*, 415, 250-257. [5] Rapacioli M. et al. (2005) *A&A*, 429, 193-204.