Transportation of Carbonaceous Mesophase
Spherules in Pitch to the Anode

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A pitch containing carbonaceous mesophase spherules was treated at 300 °C for 10 min in an electric field stronger than 600 V/cm. The spherules are transported to the anode without coalescence and aligned with aromatic lamellae parallel to the direction of electric field.

Carbonaceous mesophase spherules formed during carbonization of coal tar or petroleum pitches consist of stacked aromatic molecules, exhibiting optical, diamagnetic, and electric anisotropies.  $^{1-3}$  It has been reported that lamellae in mesophase spherules are aligned parallel to the magnetic field direction  $^{4-6}$  and that the layer planes in the magnetic field orientate flatter than expected by the familiar Brooks-and-Tayor model in which layer planes stack perpendicular to the spherule surface.  $^{7-9}$  Irrespective of electric anisotropies of carbonaceous mesophase,  $^{3}$  little information has been obtained about effects of electric field on the behavior of carbonaceous mesophase spherules. This study reports transportation and orientation behavior of carbonaceous mesophase spherules dispersed in isotropic phase of pitch in an electric field, demonstrating an important role of columbic interaction of carbonaceous mesophase spherules.

A pitch containing mesophase spherules was prepared from liquefied Wandoan coal supplied by Sumitomo Metal Mining, Co., Ltd. The distillation residue (8 g) of the coal liquid was heated in a Pyrex reactor at  $480\,^{\circ}\text{C}$  for 3 h by bubbling nitrogen through the residue. The yields of the carbonized product thus obtained were 30 and 10 wt% based on the residue

and the coal liquid, respectively. The H/C atomic ratio and softening point of the carbonized product were 0.66 and 120 °C, respectively. The pulverized carbonized product (200 mg) was charged in a Pyrex reactor (I.D. 6 mm) and Cu electrodes, which were fixed with a silicon rubber stopper, were inserted in the reactor. The cathode was a wire (0.65 mm¢), while the anode was a cylindrical type (I.D. 4 mm). After a prescribed DC voltage (up to 1000 V) was applied by using a DC power supply (Sansya Denki Seisakusyo, Co., Model KSA4-2000-0.1), the reactor was heated at 300 °C for 10 min with a heating rate of 10 °C/min under a stream of nitrogen. Then, application of the DC voltage was stopped and the reactor was cooled by immersing in an iced water. The treated product in the reactor was mounted in epoxy in situ, polished transversely, and it was observed by a Nikon polarized light microscope in conjunction with a gypsum compensator.

Figure 1a shows polarized light photomicrograph of mesophase spherules dispersed pitch before the electric field treatment. The mesophase spherules with about 10  $\mu m$  diameter dispersed randomly in the isotropic region, occupying about 10% of the total view area. On the other hand, the mesophase spherules were transported to the anode by the application of DC

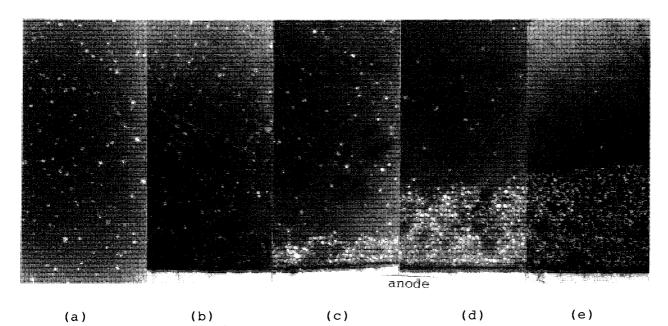


Fig. 1. Polarized light photomicrographs of mesophase spherules dispersed pitch before and after electric field treatments. (a) before treatment. After treatment at (b) 100 V, (c) 200 V, (d) 500 V, and (e) 1000 V. These pictures were taken from the region surrounded by dotted line in Fig. 2 (a).

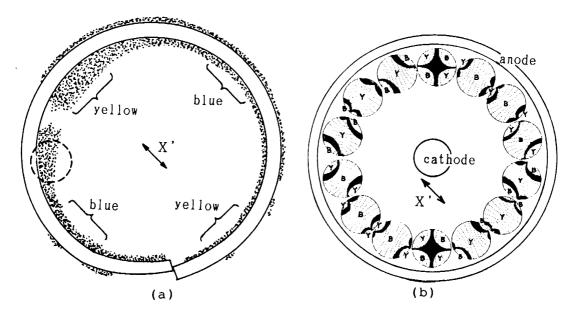


Fig. 2. Sketch of polarized light photomicrograph of collected mesophase spherules (a) and schematic model of aligned spherules (b).

voltage stronger than 200 V (Fig. 1b-e). Number of the transported spherules increased with an increase in voltage and almost all of the spherules were transported at 1000 V. The electric field treatment resulted in no coalescence of spherules. Thus, mesophase spherule may consist of electrically polarizable material whose negative charge plays dominant role as compared to the positive charge as far as the transportation phenomenon is concerned.

In order to determine a direction of the lamellae in the transported spherules, a transverse view of the whole carbonized product treated at 1000 V is sketched in Fig. 2a, where X' denotes a direction of amplitude of waves of fast polarized light produced by the gypsum compensator. The spherules with yellow interference color were observed in regions parallel

to the direction, whereas the spherules with blue interference color were observed in regions perpendicular to it, indicating that the lamellae in the spherules may orientate parallel to direction of electric field as schematically shown in Fig. 2b. Such orientation of the lamellae implies that the equator of the mesophase

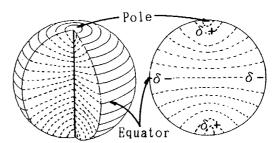


Fig. 3. Schematic model of mesophase spherule.

spherule is more negatively charged than the pole, presumably because aromatic lamellae, which have mobile  $\pi$  electrons, are aligned more widely and arranged flatter at the equator than the pole if the spherules are composed of similar lamellae structure to the familiar Brooks-and-Taylor model (Fig. 3). The findings of no coalescence of transported spherules and the presence of isotropic region between the collected spherules (Fig. 1) may be originated from columbic repulsion between the spherules because they are aligned as to face negatively polarized equators with each other or positively polarized poles with each other (see Fig. 2b).

This study demonstrates the important aspect of the mesophase spherules as an electrically polarizable material. The electric field treatment described above may be useful as a new method to collect mesophase spherules, which could be used as materials, such as filler of liquid chromatography 10) and activated carbon with high surface area. 11)

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