

Introduction

Brooks and Taylor[1] studied the structure and molecular arrangement of carbonaceous mesophase spherules which were formed in pitches during heat treatment at 350-450°C by the use of polarized-light microscopy and electron diffraction method.

On the other hand, new type mesophase spherules having a molecular arrangement different from that proposed by Brooks and Taylor have been found[2] when pitches are heat treated with a small amount of carbon black (APF or GPF). Fig.1 shows the molecular arrangement of these spherules.

The behavior of Brooks-Taylor type spherules in a magnetic field was reported[3-4] and it was proved that these spherules were aligned with their polar diameter perpendicular to the magnetic field direction. In above report, molecular arrangement of Brooks-Taylor type spherules remained unchanged in a magnetic field up to 10K gauss.

We have studied the behavior of the new type mesophase spherules in various magnetic field strengths and it was found that the molecular arrangement of the new type spherules formed by addition of APF could be transformed into that of the Brooks-Taylor type spherules when a magnetic field of over 3K gauss was applied during heat treatment. The mechanism of this phenomenon was investigated.

Experimental

About 5wt% of carbon black (APF or GPF) was added into quinoline-soluble coal tar pitch and mixed homogeneously at 430°C under stirring. The mixture thus obtained was heat treated at 430°C for a certain period at various magnetic field strengths with sample rotation (also cooled in the field). The mesophase in pitch was examined in both parallel and perpendicular directions to the magnetic field direction using a reflected polarized-light microscopy with crossed polarizers and a gypsum plate.

Results and Discussion

When the pitch is mixed with GPF and heat treated in 8K gauss magnetic field, the new type mesophase formed with their polar diameter parallel to the direction of the magnetic field. Above phenomenon is proved by the polarized-light microscope

observation that there are many spherules which have same extinction contour and pleochroism with stage rotation especially on the face parallel to the magnetic direction as shown in Fig.2. And the molecular arrangement of the spherules remains that of the new type ones in a magnetic field up to 12K gauss.

When APF is used, the new type mesophase spherules are also formed in the pitch at a field strength less than 2K gauss, but the Brooks-Taylor type spherules are also found when the field strength is increased above 3K gauss, as shown in Table 1.

As mentioned above, the addition of GPF to the pitch leads to the formation of the new type mesophase spherules irrespective of the field strength applied, while APF produces in the pitch both the Brooks-Taylor type and new type spherules depending on the field strength. Among several attempts using carbon blacks, the new type spherules are found only when APF and GPF are employed. Those carbon blacks are always attached to the surface of mesophase spherules regardless of the magnetic field strength. Carbon blacks other than APF and GPF are not attached to the surface of mesophase spherules and do not produce the new type spherules. There is no appreciable difference between APF and GPF with respect to particle size, manufacturing procedure, properties and microstructure. However, those carbon blacks have different influence on the formation process of mesophase spherules in the magnetic field, as is evident in Table 1. It can be considered that carbon blacks such as APF and GPF give a strong effect on the molecular arrangement of mesophase spherules especially by the fine crystal face existing on exterior of these carbon blacks.

It is of particular interest to know at what stage of mesophase formation the magnetic field influences on the molecular arrangement of mesophase spherules. After new type spherules were formed in the pitch containing APF under the influence of the magnetic field of 2K gauss at 430°C and cooled down to room temperature. The pitch containing the new type spherules were heat treated again in the magnetic field of 8K gauss at 430°C for 30 min. The mesophase spherules obtained by this treatment were also the new type ones. From this fact, the new type mesophase spherules formed once in the pitch cannot be transformed into the Brooks-Taylor type spherules even though the

magnetic field strength is increased above 3K gauss. This indicates that the field strength is not enough to transform the new type spherules into the Brooks-Taylor type ones and that the interaction between APF and mesophase constituent molecules is extremely strong. It can be considered that the molecules in the pitch matrix are aligned parallel to the magnetic field direction when the magnetic field is applied. In this case the magnetic field prevents the molecules surrounding the APF carbon black from being oriented parallel to the APF surface. Therefore the interaction between APF and those molecules is weakened in the presence of the magnetic field and then the Brooks-Taylor type spherules which are more stable than the new type ones are formed.

References

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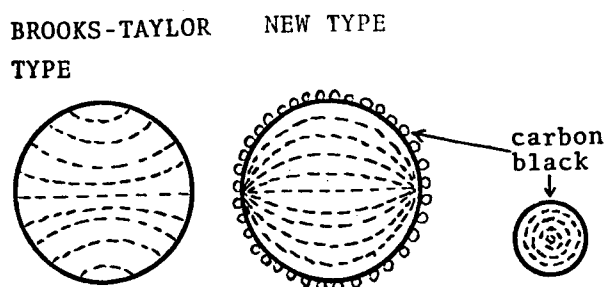


Fig. 1. Molecular arrangement of the Brooks-Taylor type mesophase spherules and the new type one

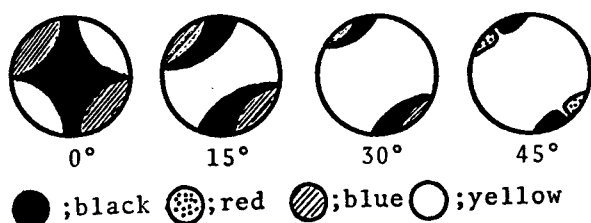


Fig. 2. Extinction contour and pleochroism of the new type mesophase spherules with stage rotation (crossed polarizers with a gypsum plate).

Table 1. Influence of a magnetic field on the molecular arrangement of mesophase spherules.

Strength of magnetic field (K gauss)	APF added	GPF added
0	N.T.	N.T.
1	N.T.	N.T.
2	N.T.	N.T.
3	B-T.T.	N.T.
4	B-T.T.	—
6	B-T.T.	—
8	B-T.T.	N.T.
10	—	N.T.
12	—	N.T.

N.T.; New type mesophase spherules
B-T.T.; Brooks-Taylor type mesophase spherules

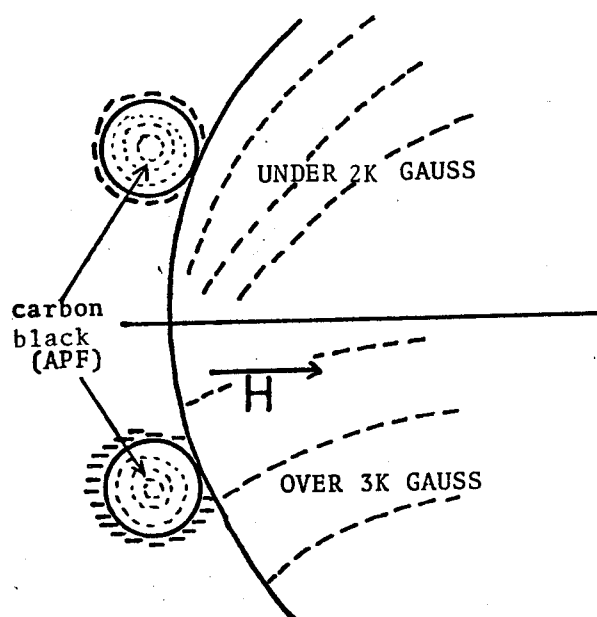


Fig. 3. Schematic view of mesophase spherules with carbon blacks (APF) in various magnetic field strengths.