GROWTH AND DEVELOPMENT OF ANISOTROPIC PHASE IN PITCH AND ULTRASONIC PROPERTIES

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Introduction

Mesophase pitch is one of the most important intermediates and precursors for high performance carbon products, and the molecular aggregation state of pitch strongly affects the quality of carbon products. We proposed a new approach for investigating the process and/or rate of growth and development of the mesophase (optically anisotropic phase) with heating by means of ultrasonic measurements[1, 2].

Experimental

Two kinds of coal-tar pitch were used in this study: mesophase pitch and hydrogenated pitch. Mesophase pitch (C) was prepared by heat-treatment in nitrogen gas from a hydrogenated pitch. Hydrogenated pitch (HC) was prepared from QI-free pitch, which was obtained by removing the primary quinoline-insoluble components in coal-tar by filtration followed by vacuum distillation. The characteristics of the pitch samples are summarized in Table 1.

Table 1. Characteristics for coal-tar pitch samples

	SP[a)]	BI[b)]	QI[c)]	AP[d)]
Hydrogenated pitch(HC)	76.6	14.0	0.1	0
Mesophase pitch(C)	311.2	89.8	75.1	95
C-BS	-	(0)	(0)	<0
C-BI	-	(100)	(100)	>90

[a]Softening point(℃); [b]Benzene-insoluble fraction(wt%); [c]Quinoline-insoluble fraction(wt%); [d]Percent of anisotropy(%).

Figure 1 shows a schematic diagram of the ultrasonic apparatus that we have newly designed. The propagation intensity (A) was measured under N_2 gas atmosphere as a function of temperature. The value of -lnA is proportional to the attenuation coefficient when the propagation distance does not change.

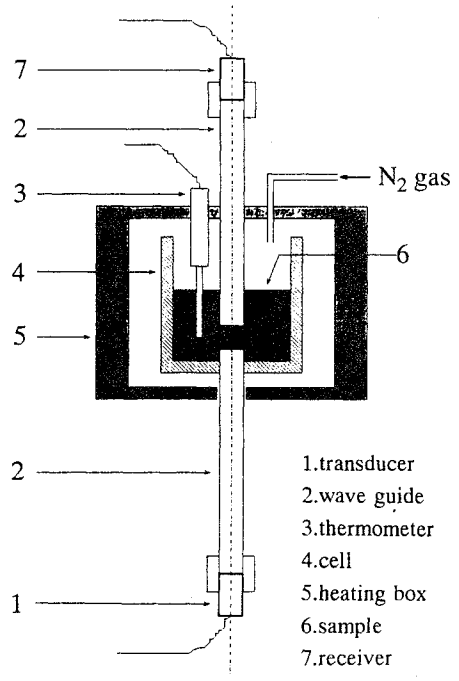

Fig. 1. Schematic diagram of a main part of the ultrasonic apparatus.

Results and Discussion

Ultrasonic measurements were carried out at temperatures ranging from room temperature to 420℃. Before the measurement the pitch was premelted above the softening point and cooled down to room temperature. The variation of -lnA

with temperature and the separated peaks for mesophase pitch are shown in the bottom of Figure 2. Two distinct peaks were seen around 250℃ and 385℃.

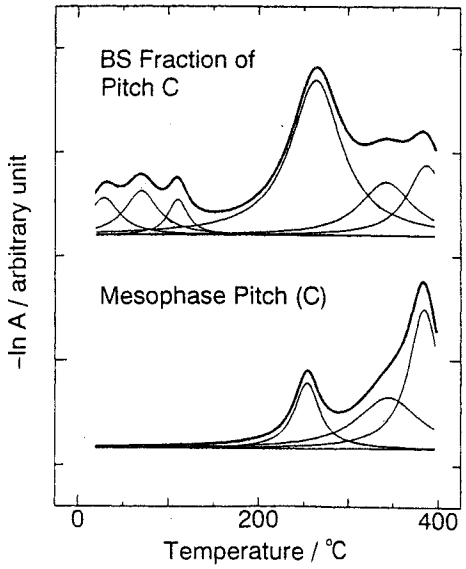

Fig. 2. The variation of -ln A with temperature and separated peaks for mesophase pitch (C) and benzene-soluble fraction of pitch C

We fractionated the mesophase pitch into soluble (C-BS) and insoluble components (C-BI) by benzene. Ultrasonic measurement was carried out for both samples. However, for the BI fraction, it was impossible to conduct ultrasonic measurement because the BI fraction did not melt. For the BS fraction, only one peak was observed around 250℃ (see the top of the figure). Hence, the peak around 250℃ seen for the mesophase pitch is ascribable to the BS component. The observation of the mesophase was carried out using a polarizing optical microscope. For the BI fraction, 90% of the field of vision was occupied by the mesophase and 10% by the isotropic phase. On the contrary, for the BS fraction, 10% by the mesophase and 90% by the isotropic phase. The higher peak (at 385℃) might be ascribable to the mesophase. Thus, it may be suggested that ultrasonic measurement allows us to monitor the appearance and development of the mesophase in situ when pitch is heated.

The mesophase, as is well known, is produced by heating coal tar pitch between 350℃ and 500℃ under nitrogen atmosphere. We measured the attenuation as a function of time when hydrogenated pitch was held at 380 ℃ . A mesophase of 5% appeared after heating for 7 hours, suggesting the development of an aromatic structure. However, the variation of the attenuation with heating time was quite small. The wave packets which were measured at an interval of ca. 5～10 min. during heat-treatment were transformed to the frequency spectra by Fourier Transformation. Figure 3 shows a contour plot of -lnR; R is a relative amplitude at a given frequency. In the measurement, a transducer with resonant frequency near 150kHz was used.

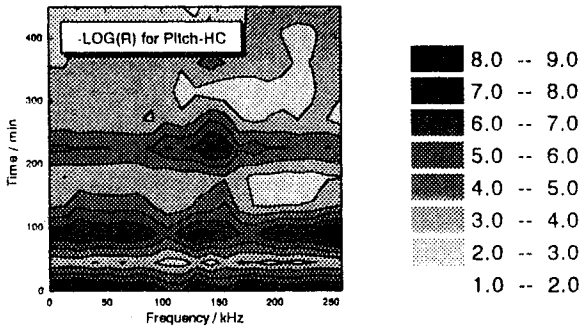

Fig 3. Contour plot of -lnR for pitch-HC.

The higher level or dark portion denotes the occurrence of greater attenuation, indicating that some phase transition(or structural change) may occur. Thus, this plot may be helpful for monitoring the growth and development of the mesophase.

References

1. A. Tanaka, H. Matsumoto, C. Yamaguchi, and K. Tokumitsu, *Carbon* 32, 1137 (1994).
2 A. Tanaka, C. Yamaguchi, J. Mondori, and K. Tokumitsu, *Reports on Progress in Polymer Physics, Japan*, Vol. 39, 453 (1996).