Time-resolved X-ray diffraction study for phase behavior of the binary mixtures of leuco dyes and long-chain alkyl type developers

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Introduction

In order to reduce the consumption of paper, the development of the re-writable paper has been continued intensively for more than ten years. Coloring/de-coloring mechanism of the mixture of leuco dyes and long-chain alkyl type developers is keen interesting to understand the mechanism of re-writing process. To clarify this mechanism, the synchrotron radiation X-ray diffraction study has been done. Here, the difference of the transition rate from the coloring to the de-coloring stage between using two types of long-chain alkyl type developers, PU18 and PA21, will be reported.

Materials and Methods

Materials: A special kind of dye, leuco dye ODB2, and long-chain alkyl type developers, PU18 and PA21, were synthesized. According to the SAXS studies of developers, both samples have lamellar structures whose repeat distances of PU18 and PA21 were 6.28 nm at room temperature and 5.43 nm at 110 °C, and 6.70 nm for all temperatures, respectively. When ODB2 and PU18 or PA21 aggregate each other, coloring occurs. In opposite, ODB2 and developers separate each other, decoloring occurs. It is noticed that ODB2 with PA21 de-colored faster than ODB2 with PU18. The reason of this difference will be explained below.

<u>Method</u>s: SAXS-WAXS simultaneous measurement was performed at BL-15A and BL-9C. The timeresolved X-ray diffraction study was performed in every 10 sec. Pelleted samples were put into the LINKAM holder, and controlled the temperature from 20 to 110 °C with the rate of 2 °C/min.

Results and Discussion

Fig.1 shows the results of the time-resolved SAXS-WAXS measurement with heating for ODB2-PU18 (Fig.1(a)) and ODB2-PA21 (Fig.(b)). The diffraction peaks at 4.20 and 4.53 nm in Fig.1 (a) and (b), respectively, mean that ODB2 and developers aggregate in the coloring stage and form the lamellar structure whose hydrocarbon chains of developers form the interdigitated structure. The repeat distance of these lamellar structures changed more than 6 nm; meaning that ODB2 and developers split each other and developers formed the lamellar without dye. Comparing the SAXS results, the diffraction peak at 6.96 nm in Fig.1(a) shifted to 5.54 nm at more than 90 $^{\circ}$ C; it means the molecular

axis of acyl chains constructed the lamellar of PU18 tilt from the right angle of the lamellar plain. On the other hand, the peak at 6.96 nm in Fig.1(b) didn't changed. It means that there are two steps of decoloring process in ODB2-PU18, while only one step for ODB2-PA21. This difference in molecular level must be reflected to the changing rate from the coloring to the decoloring stage.



Fig. 1 SAXS-WAXS results of a dye - developers system. (a) ODB2-PU18, (b) ODB2-PA21.

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