Spontaneously Induced Pattern Formation of Liquid Crystal Orientation by Gradient Photopolymerization

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Since liquid crystals show the birefringence, liquid crystalline materials have been applied to various optical materials such as diffractive gratings, lens, reflective devices. The key to those materials is the controlled pattern of molecular orientation, which bring about the distribution of refractive index. Thus, technologies for controlling molecular orientation in one- or multi dimensions have been desired. The current methods achieve the control of one-dimensional molecular orientation by applying uniform external fields along one direction such as surface rubbing treatment and mechanical stress. Recently, light-driven orientation control methods achieve the control of two-dimensional molecular orientation[1,2]. However, to date, the arbitrary control of molecular orientation with a periodicity below a few micrometers over large area is still challenging. Previously, we have found that the periodic molecular orientation patterns were spontaneously formed by photopolymerization with intensity-gradient patterned light (gradient photopolymerization). In this study, we discussed the mechanism of the formation of molecular orientation pattern.

The chemical structures of a monomer mixture used for gradient photopolymerization are shown in Figure 1. We irradiated monomer mixture in handmaid grass cell (thickness: 4 μm) with UV light (λ = 365 nm) using a photomask. Light intensity was continuously changed by a photomask using black gradation pattern. In resultant polymer film, the stripe patterns were observed with polarized optical microscopy (POM), as shown in Figure 2A. White line shows the spatial light intensity profile in observation area. To evaluate the molecular orientation in the stripe pattern, we performed the POM observation with a retardation plate (R = 137 nm), as shown in Figure 2B. Blue dotted arrow shows the optical axis of a retardation plate. In the bright area of stripe pattern, liquid-crystalline molecules (mesogen) oriented perpendicular to optical axis. In the dark area, mesogen oriented parallel or perpendicular to optical axis. Thus, periodic molecular orientation pattern was induced by gradient photopolymerization, as shown in Figure 2C.

1) K. Ichimura, *Chem. Rev.* **2000**, *100*, 1847.
2) A. Shishido, *et al.*, *Sci. Adv.* **2017**, *3*, e1701610.

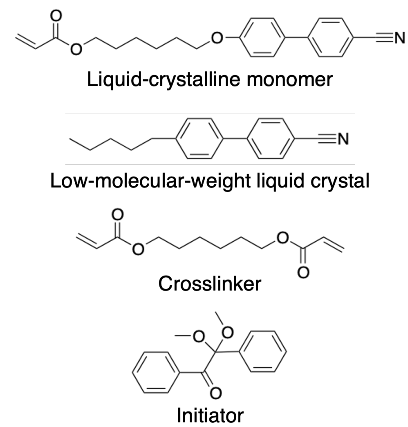

Figure 1. Chemical structures of a monomer mixture used in this study

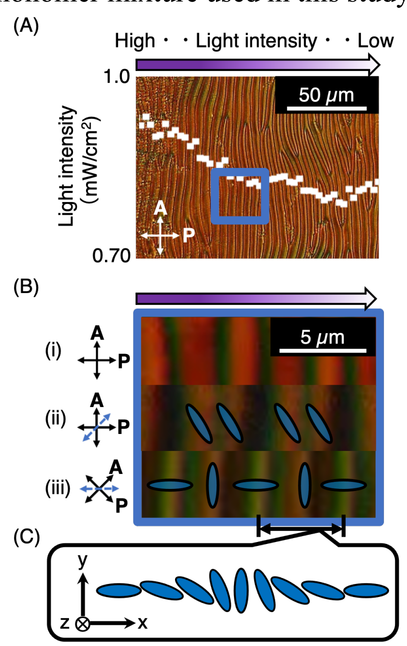

Figure 2. (A) POM observation of polymer film. The spatial profile of light intensity is plotted in the figure. (B) Enlarged view (i) without a retardation plate with offset 0°, (ii) with a retardation plate with offset 0°, (iii) with offset 45°. (C) Schematic illustration of molecular orientation pattern in the polymer film