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題目(和文)	動的光重合による液晶の三次元配向パターニング
Title(English)	Three-Dimensional Alignment Patterning of Liquid Crystals Directed by Scanning Wave Photopolymerization
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The purpose of this study is development of a photochemical method to directly induce three-dimensional molecular alignment of LCs in polymeric systems. This would be key to developing highly functional materials for electronics, photonics, and soft robotics. Here, the author investigated alignment behavior of molecules in densely crosslinked LC polymer films and serendipitously discovered the induction of out-of-plane alignment by photopolymerization using spatiotemporal light scanning. The films with complex manner were used as an alignment layer, enabling to fabricate an actuator utilizing three-dimensional molecular alignment of the film.

In Chapter 2, alignment control of mesogenic moieties in densely crosslinked LC polymer films with various molar ratios of an oxetanyl anisotropic monomer and crosslinker was demonstrated. This experiment revealed that a driving force for the unidirectional molecular alignment of SWaP is valid for the crosslinker LC polymer films at high crosslinker concentrations and regulate the alignment direction depending on the crosslinker concentration by its mass flow regime. SWaP facilely fabricated the molecularly aligned crosslinked LC polymer films with high thermal stability.

In Chapter 3, the author serendipitously discovered out-of-plane molecular alignment and explored the three-dimensional alignment of molecules and higher-order structures by SWaP. The LC moieties in the photopolymerizable sample formed the homeotropic alignment on the glass substrate even before photoirradiation. In this situation, 1D light scanning with the constant exposure energy induced the tilt alignment with different angles in the films. By GI-WAXD measurement, it was found that not only the LC moieties but also higher-order structures were tilted along the light scanning direction. Furthermore, the author estimated induced tilt angles from experimentally obtained birefringence, and fabricated three-dimensional molecular alignment patterns.

In Chapter 4, the author experimentally and theoretically analyzed that the molecular diffusion caused by concentration gradient exclusively ruled the molecular alignment and investigated how the tilted molecular alignment was generated by SWaP-triggered mass diffusion. The experimental alignment length almost corresponded to the theoretical model based on Fick's law of diffusion. Furthermore, the author developed a basic model of thermodynamic force which triggers the molecular diffusion by SWaP. The concentration gradient was presumed by the polymer conversion experimentally measured by FT-IR spectroscopy, leading to the evaluation of thermodynamic force. Using the obtained diffusion coefficient, the diffusion velocity was estimated, and the tilt angles and tilt direction were rationalized.

In Chapter 5, the author examined the ability of molecularly aligned polymer films fabricated by SWaP as an alignment layer, and prepared a thick free-standing film enough to act as an actuator. As the test system, a low-molecular-weight LC was injected into the cell prepared by adhering a pair of glass substrate coated with the fabricated polymer film. As a result, the injected LCs were aligned along the same direction as the prepared polymer film by cooperative effect of LCs, indicating that the molecularly aligned polymer film fabricated by SWaP acts as the alignment layer. Using this alignment layer, the author successfully fabricated a thick free-standing crosslinked polymer film with three-dimensional molecular alignment by injecting the photopolymerizable LCs and subsequent photopolymerization. This thick free-standing film exhibited thermal actuation and bent at its alignment boundary due to the difference between expansion at the homeotropic surface and shrinkage at the homogeneous surface.

In Chapter 6, the author summarized the obtained results. In this study, the author extended the versatility of SWaP through the investigation of arbitrary control of complex

molecular alignment patterns with both in-plane and out-of-plane directors. The driving force of the molecular alignment in SWaP, which is the light-triggered mass flow, allows us to manipulate the alignment directors of anisotropic molecules and higher-order structures. The differences of tilt angle of the alignment were elucidated by estimating the diffusion length and velocity resulting from the mass flow based on the theoretical model. By utilizing such manipulation of alignment directors, the author fabricated the polymer film with 3D molecular alignment patterning, which was difficult to be achieved by the conventional alignment method. Furthermore, the author applied the polymer film to an alignment layer and fabricated a thermal film actuator. Thus, it was proved that SWaP also has great potential to control complex molecular alignment patterns and generate functional materials.