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単層酸化グラフェン基板上の PVDF 薄膜の自己組織化
Self Assembly of Ultrathin PVDF on Monolayer Graphene Oxide

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1. 背景と研究目的

Ferroelectric (FE) materials possess spontaneous polarizations arising due to the presence of non-centrosymmetric inversion symmetry. In our work, for obtaining high piezoelectric constant and stability of polarization, the orientation along with crystallinity of the FE dipoles in PVDF-TrFE was manipulated by fabricating PVDF-TrFE nano lamellae (PVDF-NL) on the defective monolayer graphene oxide (mGO). Graphene oxide (GO) which is a 2D polar nanomaterial was utilized since they can interact with the polymer to enhance the FE polarization stability through self-assembly as established in our earlier reports[1]. We then characterize the ultrathin films using X-ray diffraction to determine the crystal orientation.

2. 実験内容

PVDF-TrFE films were spin coated on a mGO film and annealed at 140 °C (PVDF-NL). For comparison PVDF-TrFE was spin coated and annealed on bare Si wafers (PVDF-Si).The crystallinity and the crystal plane orientation in the PVDF samples were determined by the two dimensional grazing incidence X-ray diffraction measurements (2D-GIXRD) employing a synchrotron X-ray source (beamline BL8S1, Aichi Synchrotron Radiation Center in Japan). In the 2D-GIXRD measurement, synchrotron X-ray radiation (λ = 1.35 Å, beam dimension of 0.2 x 0.8 mm and energy of 9.16 keV) was shined on the sample (1 x 1 cm sample mounded on a glass slide) at a grazing angle of 0.5° (which produced a beam foot print of 22.9 mm) and the diffracted rays were collected using a 2D X-ray detector (PILATUS 100K, dimension of 83.7 x 33.5 mm).

3. 結果および考察

We expect that PVDF molecular chains form the edge-on lamellae (chains growing parallel to the substrate) instead of the flat-on lamellae (chains growing perpendicular to the substrate) in PVDF-NL due to the presence of GO. The orientation of the PVDF crystal can be determined by considering the diffraction of (200)/(110) planes of orthorhombic PVDF from the 2D-GIXRD measurements by employing a 2D detector. 2D diffraction patterns were obtained for all samples with varying thickness values (10 nm, 20 nm, 90 nm). The GIXRD measurements for the ultrathin PVDF-NL samples reveal preferential crystal orientation (the scattering vector q_{200} is perpendicularly oriented) as shown in **Fig. 1a**. Samples with low thickness exhibit weak diffraction corresponding to the (200) plane. For the case of PVDF-Si, continuous Debye ring formation was observed (**Fig. 1b**) revealing the presence of randomly oriented crystals (as scattering vectors q_{110} and q_{200} are oriented in random directions) where the molecular chains grow in random directions w.r.t substrate (mixture of edge-on and face-on lamellae). Similarly, continuous rings with lesser diffraction intensities were observed for other thickness values in PVDF-Si (**Fig 1b**).

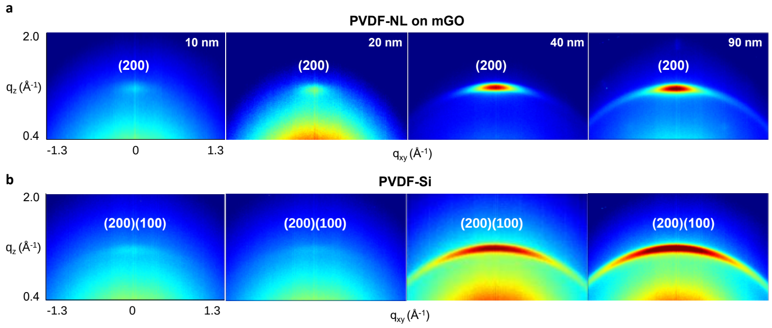

Fig. 1: 2D-GIXRD patterns of (a) PVDF-NL and (b) PVDF-Si for various thickness values.

4. 参考文献

[1] P. Viswanath, K. K. H. De Silva, H. H. Huang, M. Yoshimura, *App. Surf. Sci.* **2020**, *532*, 147188.