摩擦転写法により作製した高分子薄膜トランジスタの電荷輸送異方性に及ぼす誘電体界面の影響
Effect of dielectric interface on charge transport anisotropy in polymer thin-film transistors fabricated by friction transfer method

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Introduction: Conjugated polymers (CPs) offer adequate freedom to improve the charge transport in their thin films by tuning their chemical structure or processing techniques aiming towards enhancing their crystallinity. Exploiting quasi 1-dimensional nature of CPs is one of the best possible approaches to improve the carrier transport by uniaxially orienting them along the channel of an organic field-effect transistor (OFET). Various solution-based approaches in order to orient the CPs leading to high-performance OFETs has been demonstrated, however, the limitations like need of good solubility of the CPs, surface wettability, pre-aggregation in solution, etc., are the bottleneck to generalize them. Moreover, inevitability of halogenated solvents for film fabrication may also lead to serious health and environmental hazards during large scale production. Friction transfer is a solvent-free processing technique to fabricate large-area oriented polymer thin films and can effectively be utilized to overcome aforementioned challenges. In this work, role of dielectric surface to control the anisotropic charge transport in the OFETs has been extensively investigated and considering the optical and electrical characteristics, a possible transport mechanism has been proposed.

Experimental: OFETs were fabricated in bottom-gate top-contact device architecture as shown in the Fig. 1(b) inset. As active layer, Poly[2,5-bis(3-tetradecylthiophen-2-yl)thieno[3,2-b]thiophene] (PBTTT) was friction transferred on bare or silane-treated oxide surfaces followed by annealing below and above its liquid crystalline (LC) temperature in Argon atmosphere. Finally, the source and drain electrodes were thermally evaporated. The thin films were characterized by polarized UV-Visible electronic absorption spectroscopy, angle-dependent Raman spectroscopy, and X-ray diffraction (XRD) measurements.

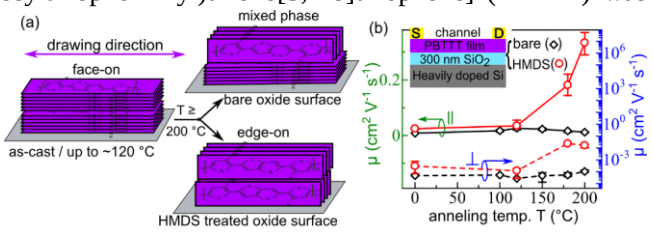

Figure 1. Schematic representation for conformational transformation in friction-transferred PBTTT after annealing on different surfaces (a) and its effect on corresponding charge transport anisotropy (b). Inset of (b) represents the schematic for OFET architecture

Results and Discussion: Performance of OFETs depends on macromolecular orientation and conformation, especially near the dielectric interface. In this work, highly oriented thin films of PBTTT were fabricated by optimizing the casting and annealing conditions, resulting in significant optical anisotropy with dichroic ratio (DR) >25. It was noticed that annealing on bare and silane-treated oxide surfaces, led to a similar trend in DR incensement; however, a drastic difference in electrical anisotropy was recorded, as shown in the Fig. 1. On bare SiO_2 optimum field effect mobility (μ) of 0.035 $cm^2 V^{-1} s^{-1}$ along the backbone orientation was obtained for the thin films annealed to 120 °C, whereas it reached up to 0.39 $cm^2 V^{-1} s^{-1}$ on silane-treated SiO_2 after annealing at 200 °C. In order to probe the interface dependent variation in μ, systematic XRD measurements of the thin films annealed to various temperatures above and below LC temperature were conducted. Thin films as-prepared or annealed below LC temperature exhibited only (h00) peaks and no (0h0) peaks in in-plane mode of grazing incidence XRD (GIXD) pattern, confirmig face-on oriented macromolecules. However, annealing beyond LC temperature on bare oxide surface, (h00) peaks appeared both in in-plane and out-of-plane mode corresponding to partial edge-on transformation in the thin film, imparting high resistance path to charge transport. The (h00) peak didn't completely disappear even after annealing at higher temperature for a longer time, which was attributed to the chemical interaction between thiophene rings and reactive groups on the bare oxide surface. Hence, on the bare oxide surface, with increase in annealing temperature μ decreased. Contrary to this, annealing on silane-treated oxide surface led to a gradual increase in μ, which occurred due to almost complete conformational transformation from face-on to edge-on, as evidenced by GIXD results. Further, considering the optical and electrical characteristics of the friction transferred PBTTT films, a transport mechanism was proposed, which will be discussed in detail during the presentation.