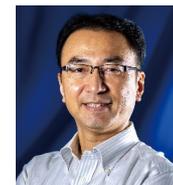


# Ultra-highly oriented thin-film coating based on rodlike-preaggregate formation of $\pi$ -conjugated polymer in solution

Division of Electrical, Electronic and Infocommunications Engineering, Graduate School of Engineering

Associate Professor Akihiko Fujii

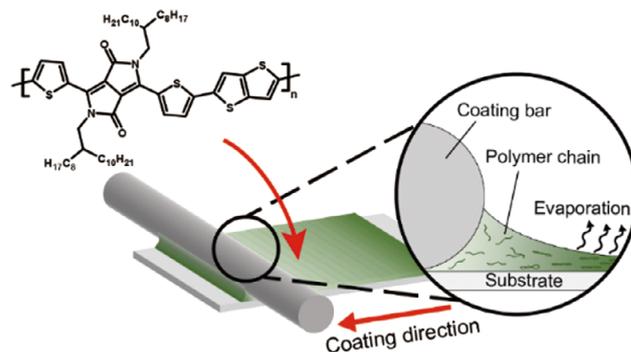


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## Abstract

A ultra-highly oriented thin film of a donor– acceptor conjugated polymer, poly[2,5-(2-octyldodecyl)-3,6-diketopyrrolopyrrole-*alt*-5,5-(2,5-di(thien-2-yl)thieno[3,2-*b*]thiophene)] (PDPP-DTT), which tends to form a rod-shaped aggregate in solution, was fabricated by bar-coating method. When the coating speed was 20  $\mu\text{m/s}$  or less, a very highly oriented thin film with quite high 2D orientation parameter  $S \approx 0.9$  was obtained. The alignment direction of the polymer main chain strongly depended on the solution concentration. When the solution concentrations were 1 g/L and 3 g/L, the main chains were uniformly aligned parallel and perpendicular to the coating direction, respectively. The molecular orientation mechanism was proposed by taking the aggregation phenomenon in the solution into account.

new insights about the dynamics of the film structure formation including molecular conformation and orientation.



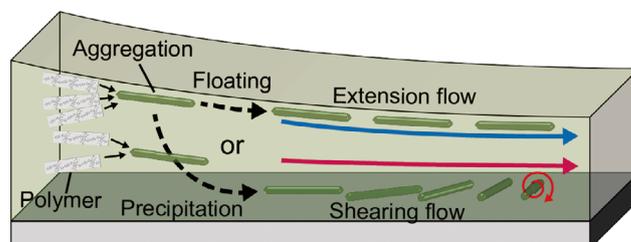
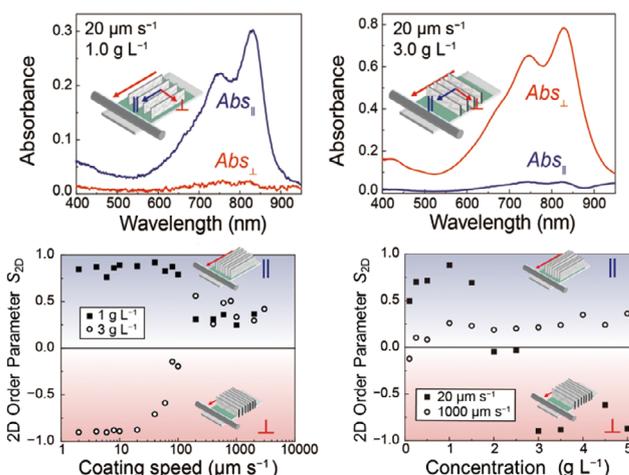
## Background & Results

By the recent progress of material development, the high solubility and processability of organic semiconducting materials have been realized, and the thin film fabrication by some solution processes for the printed electronics are enabled. For developing organic semiconductor devices with excellent carrier transport properties, the direct fabrication of highly oriented thin films from the solutions onto substrates is practically required. Herein, We focused on PDPP-DTT, which is a candidate solution-processed material and demonstrates the carrier mobility exceeding 10  $\text{cm}^2/\text{Vs}$ , and selected a bar-coating technique as a uniaxial solution process to fabricate highly oriented thin films.

The order parameter of molecular orientation as well as film thickness could be controlled by the sweep rate, and reached 0.9, which is the highest value for  $\pi$ -conjugated polymers among one-step coating methods. Besides, the direction of the main chain was selectively oriented parallel or perpendicular to the sweep direction, which depended upon the solution concentration under the low sweep rate like 20  $\mu\text{m/s}$ . Such unique orientation phenomena should be related to the nematic liquid crystallinity of PDPP-DTT aggregates formed under the slow coating process. The orientation dynamics was interpreted by the existence of rodlike PDPP-DTT preaggregates in the solution and their behavior due to the extension flow and shearing flow. Furthermore, the hole mobility depending upon the orientation direction, namely, the electrical anisotropy was demonstrated.

## Significance of the research and Future perspective

In the PDPP-DTT solution, the small rodlike structure of the polymer chains forms the preaggregate, and the size of the preaggregate becomes larger as the concentration increased. The ultra-high molecular orientation and its direction must be strongly related with the rodlike preaggregate formation in the coating solution. Such a thin-film coating process based on rodlike preaggregate formation would be applicable to other  $\pi$ -conjugated polymers, and provide



## Patent

Yabuuchi, Yuta; Minowa, Yu; Fujii, Akihiko et al. Direction-Selectable Ultra-Highly Oriented State of Donor-Acceptor Conjugated Polymer Induced by Slow Bar-Coating Process. *Advanced Electronic Materials*. 2021, 7, 2100313-1-8, doi: 10.1002/aeml.202100313

## Treatise

Yabuuchi, Yuta; Minowa, Yu; Fujii, Akihiko et al. Dynamics of Preaggregation and Film Formation of Donor-Acceptor  $\pi$ -Conjugated Polymers. *ACS Materials Letters*. 2022, 4, p.205-211, doi: 10.1021/acsmaterialslett.1c00734

Minowa, Yu; Yabuuchi, Yuta; Fujii, Akihiko et al. Fast-Coating Process Based on Elongated Rodlike Preaggregate for Highly Oriented Thin Film of Donor-Acceptor Conjugated Polymer. *ACS Applied Materials & Interfaces*. 2022, 14, p.50112–50119, doi: 10.1021/acsmi.2c13516

## URL

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## Keyword

$\pi$ -conjugated polymer, organic semiconductor, molecular orientation, printed electronics