# **Electronic Supplementary Information**

# Edge-on lamellar crystallization of oligo(3-methoxythiophene) in polyester matrix films and a gold tone development thereof

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#### Supplementary Note

#### Calculation of penetration depth of X-ray<sup>S1</sup>

The depth of penetration of the CuK $\alpha$  rays into Film 0 was calculated based on the linear attenuation coefficient ( $\mu_l$ ) of Film 0, determined as follows:

$$\mu_l = \rho \sum_i g_i(\mu_m)_i \tag{S1}$$

where  $g_i$  and  $(\mu_m)_i$  are the mass fraction and mass attenuation coefficient of the element *i*, respectively, and  $\rho$  is the density of O3MeOT. The summation included all of the constituent elements, namely C, H, O, S, and Cl. The  $g_i$  value of each element was determined by simple calculation, while the  $\mu_m$  values of each element were taken from Reference S1, and the  $\rho$  value of O3MeOT (1.54 g cm<sup>-3</sup>) was given in Section 3.5 in the text. These values were substituted in equation S1 to give a  $\mu_l$  value of 50.3 cm<sup>-1</sup>. The penetration depth of the CuK $\alpha$  rays incident perpendicular to the surface of Film 0 was taken as the depth at which the intensity of incident X-rays became 1/e. This was calculated as the reciprocal of  $\mu_l$ , which is 199 µm. However, as the incident angle of X-rays in this study was 1° relative to the film surface, thus the penetration depth was calculated as 199 µm × sin 1° = 3.5 µm.

#### **Supplementary Discussion**

#### Electric conductivity measurement system

Fig. S1 is a schematic diagram of electrical conductivity measurement of the films. The measurement probe (URSS probe, Mitsubishi Chemical Analytech.) comprised ring and circular electrodes, and was pressed in contact with the sample films. The probe is connected to a resistance meter (Hiresta-UX MCP-HT800, Mitsubishi Chemical Analytech.), and the bias voltage applied between the two electrodes was 10 V.



**Fig. S1.** Schematic diagram of the electric conductivity measurement system viewed from the (a) side and (b) top.

#### Elemental mapping analysis based on SEM

Fig. S2 shows the cross-sectional SEM image of Film 4 (a), the magnified EDX mapping image of near film surface for S (b), and the magnified mapping image of the film/glass plate interface for S (c). This sample was prepared under the same conditions as the one shown in Fig. 6 of the text. In support of the results in Fig. 6 of the text, the distribution of elemental S in Fig. S2 shows that O3MeOT is localized on the film surface and at the interface between the film and the glass plate.



**Fig. S2. (a)** SEM images of the cross section of Film 4. Magnified EDX mapping images of the (b) film surface and (c) film/glass plate interface.

#### EDX and XPS analyses of an O3MeOT/polyester(Vylon200) blend film

Polyester (PES; trade name: Vylon200®; number average molecular weight =  $1.7 \times 10^4$ )<sup>S2</sup> was kindly provided by TOYOBO Co., Ltd. PES (0.080 g) was dissolved in GBL (1.0 g) to give a 1 wt.% resin solution. O3MeOT (0.010 g) was added to the resin solution and stirred for complete dissolution. The coating solution was applied to a glass plate and the coating was dried for 6 h at 60°C in a hot air dryer (Yamato Scientific Co., Ltd., DN64). The sample surface was covered with a vacuum-evaporated aluminum layer.

The elemental composition and positional relationship (results of linear analysis) of Al, S and Cl were measured along the thickness of the blend film (Fig. S3). The Al profile (Fig. S3a) illustrates that position =  $1.3 \mu m$  corresponded to the film surface, while position =  $40.6 \mu m$  corresponded to the film/glass plate interface. The element S from the thiophene ring and Cl from the dopant were observed in the same positions in Fig. S3b and S3c, indicating that O3MeOT was highly abundant in



**Fig. S3.** Concentration profiles of (a) Al, (b) S, and (c) Cl along the thickness direction of a O3MeOT/polyester blend film with a blend ratio and thickness of 1/1 and 39  $\mu$ m, respectively.

the areas with a high signal intensity. This area was present discretely in the direction of the film thickness, and the signal intensities of S and Cl were higher at the film surface and the film/glass plate interface. These observations indicated that O3MeOT was localized at both the film surface and the interface.

Fig. S4 shows the XPS spectra of the S 2p (a) and Cl 2p (b) regions of the O3MeOT/Vylon200 blend film. For both the surface of the film and the interface with the glass plate, signals of S derived from the thiophene unit of O3MeOT and Cl derived from the dopant  $ClO_4^-$  are observed, indicating the presence of O3MeOT on the surface and at the interface, supporting the above SEM-EDX results.



**Fig. S4.** XPS spectra in the (a) S 2p and (b) Cl 2p regions for the O3MeOT/Vylon200 blend film. The solid line shows the signal from the film surface in contact with the glass plate, and the dotted line shows the signal from the film surface.

### **Supplementary References**

- S1 D. C. Creagh and J. H. Hubbell, in *International Tables for Crystallography*, ed. E. Prince, Wiiley, Chichester, 2011, vol. C, ch. 4.2.4, pp. 220–229.
- S2 https://www.toyobo-global.com/seihin/xi/vylon\_es/fra\_vylon\_cg/fra\_vylon\_cg.htm (accessed October 7, 2021).