Electronic Supplementary Information

Slippage- and load-induced changes in the crystalline orientation of oligo(3-methoxythiophene) powder to develop a gold-tone luster

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Grinding of the 3-methoxythiophene oligomer in a mortar

Fig. S1 shows a photographic image of the 3MeOT oligomer ground in a mortar, wherein it can be seen that even a weak manual force can produce a gold-toned luster.

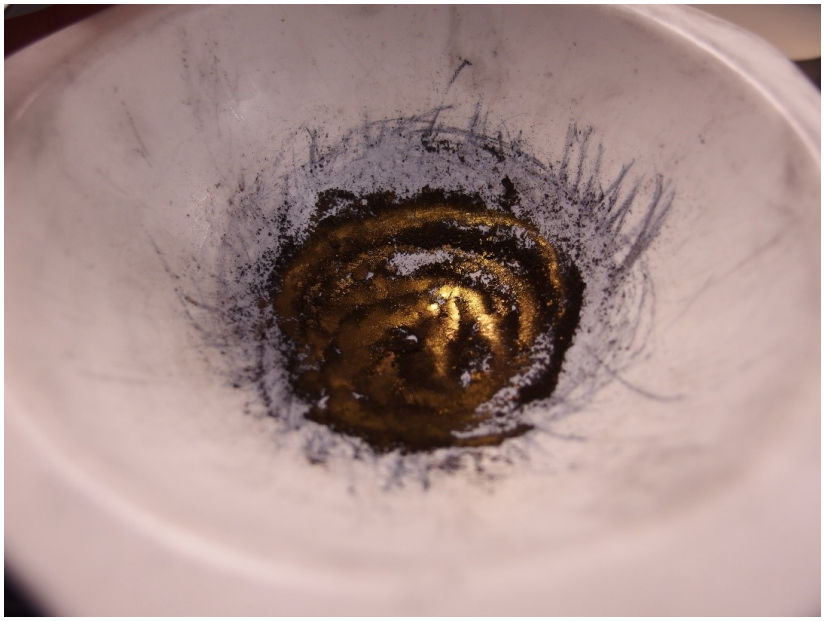

Fig. S1. Photographic image showing the development of a gold tone upon the grinding of a blackish brown 3-methoxythiophene oligomer powder in a mortar.

Observation of the surface morphology of the 3-methoxythiophene oligomer tablets

Fig. S2 shows laser microscopy images of tablets-0.05t, 0.5t, 1t, 2t, 4t, 8t, and 10t. Table 1 in the main text gave the root-mean-square roughness, R_q, of each tablet, which indicates that an increase in the applied pressure resulted in a smaller R_q value. The images shown in Figure S2 provide visual support for this result.

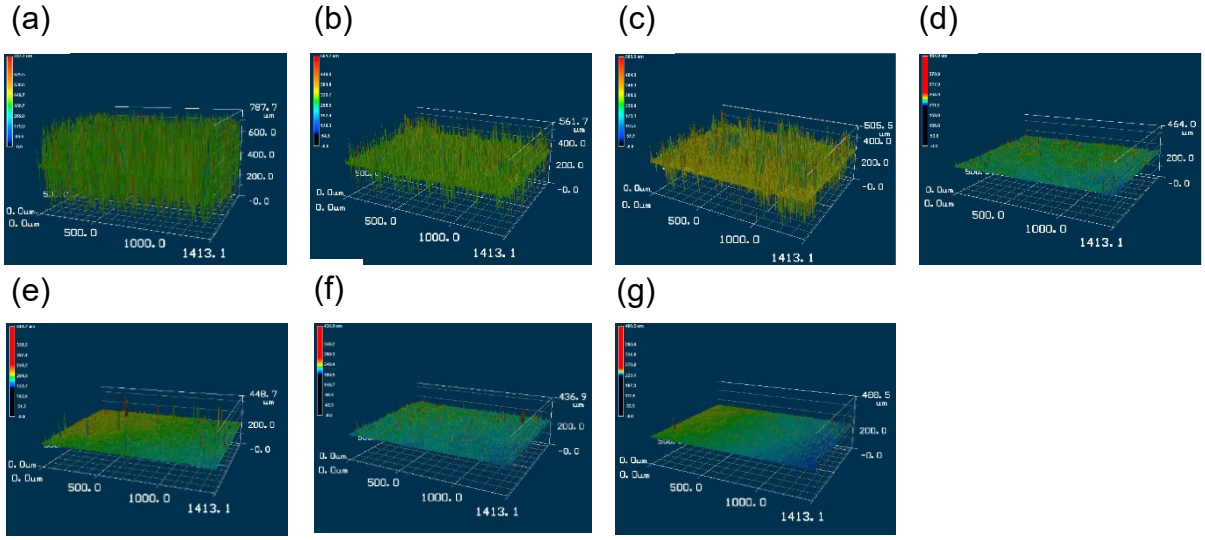

Fig. S2. Surface topography images of tablets-0.05t (a), 0.5t (b), 1t (c), 2t (d), 4t (e), 8t (f), and 10t (g), as measured by a laser microscope.

Variable-angle spectral reflectance measurements

To confirm that the gold tone of O3MeOT layer 2 was not derived from the structural color, we carried out variable-angle spectral reflectance measurements. Thus, **Fig. S3** presents a schematic representation of the goniometric system used for measuring the spectral reflectance. The incident light angles θ_i with respect to the normal of the film surface were 15, 30, 45, and

60°. The detection angle θ_r of the reflected light was set between −80 and 80° at 5° intervals. Based on the result shown in Figure 5a in the main text, O3MeOT layer 2 was considered to possess a strong specular reflection component; therefore, the measurements were performed at 1° intervals throughout the 10° range before and after the value of the specular reflection angle. In these measurements, a barium sulfate plate was used as the reference material, and thus, the measured reflection intensity of the O3MeOT film represents the relative reflection intensity compared to that of barium sulfate.

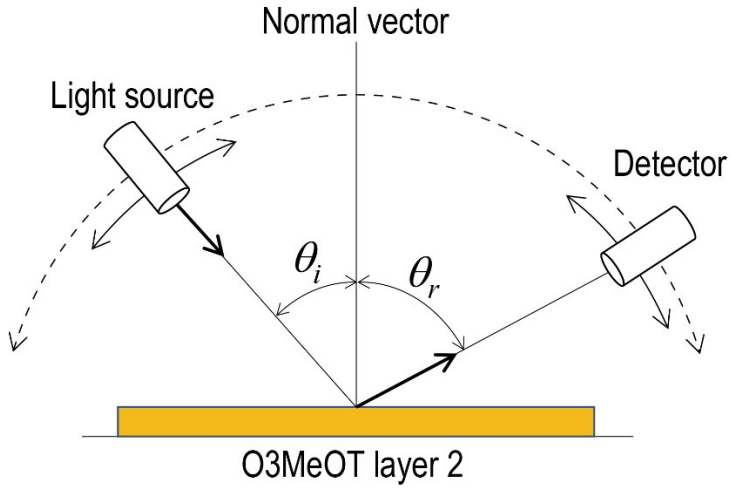

Fig. S3. A schematic representation of the instrumental setup used for the variable-angle spectral reflectance measurements of O3MeOT layer 2.

Fig. S4 shows the θ_i and θ_r dependences of the reflection spectra of O3MeOT layer 2. Initially, θ_i was fixed and the specular reflection spectra were recorded while θ_r was changed. Subsequently, θ_i was changed and the spectra were measured in the same way to obtain the spectra shown in Figure S4. For any incidence angle θ_i, the reflectance reached its maximum when the relationship $\theta_i = \theta_r$ was satisfied (i.e., the specular reflection condition), and no light reflection occurred in cases where $\theta_r > \theta_i + 4°$ or $\theta_r < \theta_i - 4°$. It is important to note that the shape of the specular reflection spectrum is approximately the same regardless of θ_i, i.e., the

reflection color of the film does not change with θ_i. In the case of many materials that possess a structural color, it is known that they exhibit different colors depending on the value of θ_i.[1–5] In other words, the reflection spectrum shows a θ_i -dependence. These results therefore confirm that the lustrous color of O3MeOT layer 2, which exhibits no viewing angle dependence, is not based on the color development mechanism of structural color. In other metal-free organic glossy materials, similar variable-angle spectral reflectance measurements have been performed to confirm that their gloss is not based on the structural color.[6, 7]

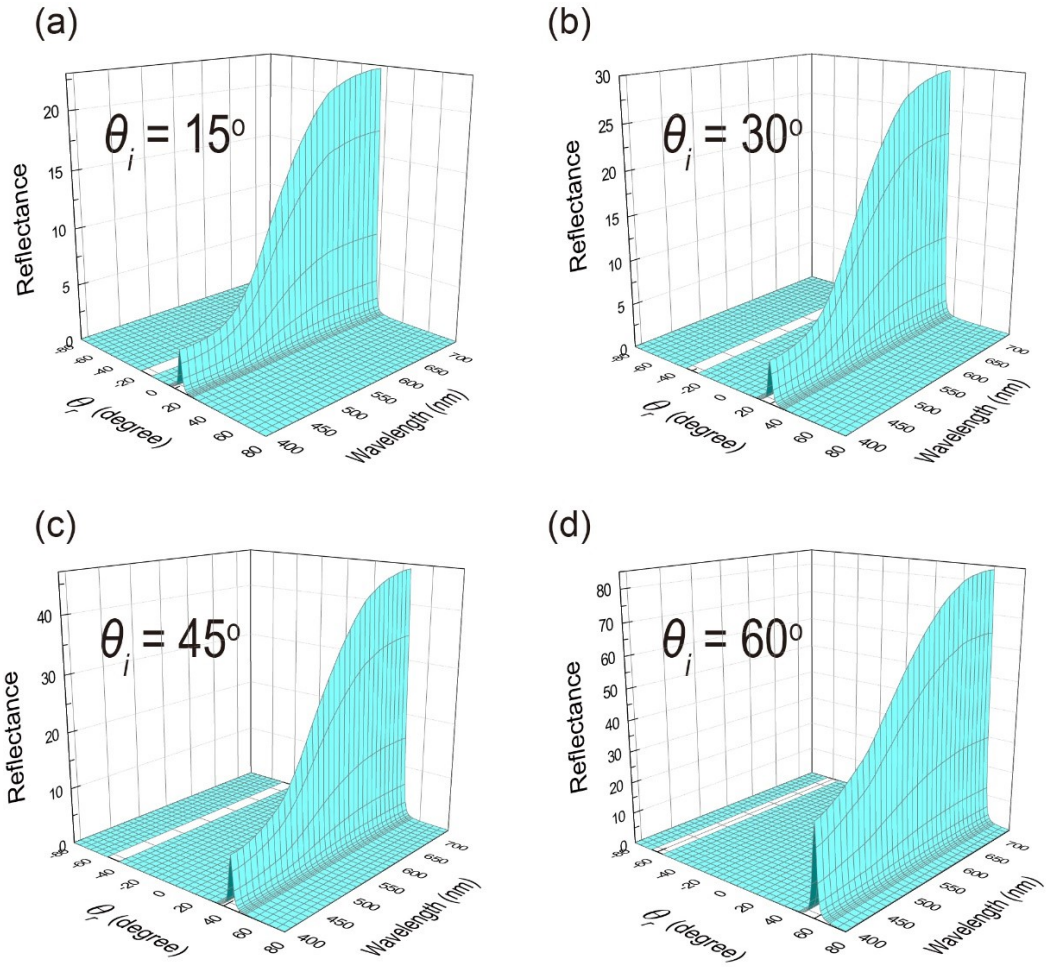

Fig. S4. Reflection spectra of O3MeOT layer 2 measured as a function of the detection angle, θ_r, where the incident light angle, θ_i was set at (a) 15°, (b) 30°, (c) 45°, and (d) 60°.

Supplementary References

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