

## **Electronic supplementary information for Photothermal COFs with donor-acceptor structure for friction reduction and antiwear**

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### **Section 1. Experimental details**

#### **Raw materials**

Tris(4-aminophenyl) amine was obtained from Shanghai Aladdin Biochemical Technology Co. Ltd, and methanol (AR99.5%) and tetrahydrofuran (AR99.5%) were obtained from Tianjin Fuyu Fine Chemical Co. Ltd. 3,6-bis(5-formylthiophen-2-yl)-2,5-bis(2-ethylhexyl)pyrrolo[3,4-c]pyrrole-1,4(2H,5H)-dione was obtained from Suzhou Nakai Technology Co. Glacial acetic acid (AR99.5%) and trichloromethane (AR99%) were purchased from Yantai Far East Fine Chemical Co. Ltd.

#### **1.1 Synthesis of COF**

32 mg of tris(4-aminophenyl) amine was dissolved in 105 mL of methanol. 3,6-bis(5-formylthiophen-2-yl)-2,5bis(2-ethylhexyl)pyrrolo[3,4-c]pyrrole-1,4(2H,5H)-dione of 93 mg was weighed and added to 105 mL of chloroform. The ratio of the above two raw materials was adjustable, and COF materials with different sizes were

obtained. After mixing the above two solutions, 15 mL of ice acetic acid was added dropwise to catalyze the reaction, which was placed on a magnetic stirrer for 5 min, and then left for 48 h. During the reaction process, the solution changed to dark purple color. The solution was centrifuged in a high-speed centrifuge at 7000 rpm for 12 min, washed with tetrahydrofuran, and continue centrifugation several times to remove any unreacted reagents. Final products were obtained by freeze drying.

### **1.2 Tribological property study**

The tribological properties of COF as a lubricant additive were tested in reciprocating mode using a CFT-I tester (ball-on-disk friction machine, BOD) produced by Lanzhou Zhongke Kaihua Science and Technology Development Co. Ltd. Tribological characterization of COF as a lubricant additive was tested in reciprocating mode using a disc-ball friction tester. The upper friction partner was GCr-15 steel ball ( $\Phi=6$  mm, AISI 52100,  $R_a=25$  nm), which was kept static. The lower friction partner was a GCr-15 steel block ( $\Phi=24$  mm,  $h=7.9$  mm, AISI 30400,  $R_a=10 \pm 2$  nm), which was held in place using a fixture and controlled by a DC servomotor in a reciprocating motion. The reciprocating distance was set at 2 mm, the load was changed from 20 N, 30 N, 40 N to 50 N, and the frequency was varying from 1 Hz, 2 Hz, 3 Hz to 5 Hz. COF material was ultrasonically dispersed into TMT-3 base oil. After the dispersion was completed, the mixed lubricant was dripped on the contact area of the two friction pairs to simulate the actual application working conditions. The wear and morphology of the scratches on the lower disc were analyzed using a 3D Contour Surface Analyzer (GTK-18-0433, Bruker, Germany). The surface morphology and elemental composition of the scratches were determined using a scanning electron microscope (Model EVO 10, Zeiss, Germany) and an energy dispersive spectrometer (EDS) (Xplore 15, Oxford, 186 UK), respectively.

### **1.3 Photothermal property and controlled viscosity**

The absorption and conversion properties of the COF material were measured by a fiber laser. The COF material was dispersed into base oil, and the mixed lubricating oil with required concentrations was prepared. 1 mL of the above mixed oil with different concentrations was taken and placed in a glass test tube. The power of the

NIR light was controlled to be 0.5-1.25 W/cm<sup>2</sup> by adjusting the current level so that it was directed to the mixed oil at the bottom of the glass test tube. The temperature of the composite lubricant was recorded at 30 second intervals and the viscosity of the base oil and the mixed oil at the corresponding temperatures were measured.

## Section 2. Figures S1–S9

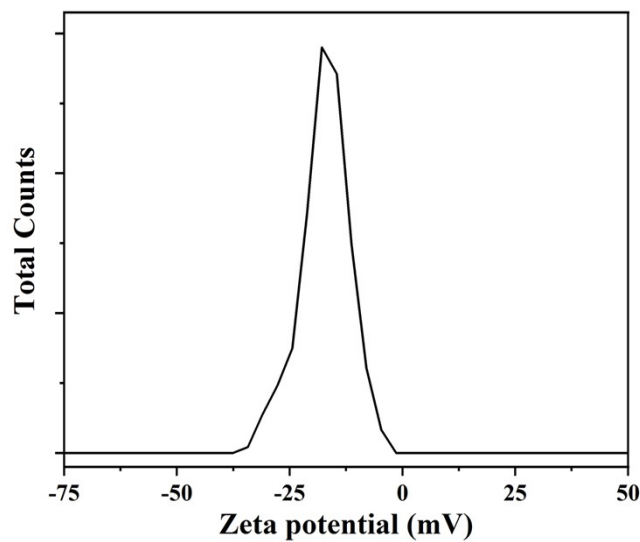


Fig. S1 Zeta potential of COF.

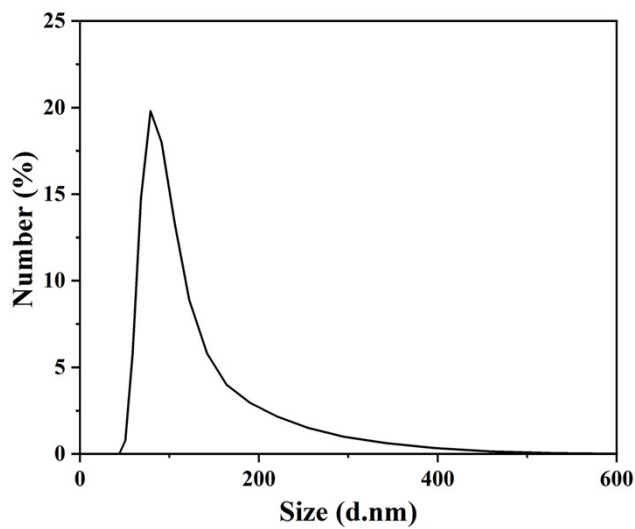
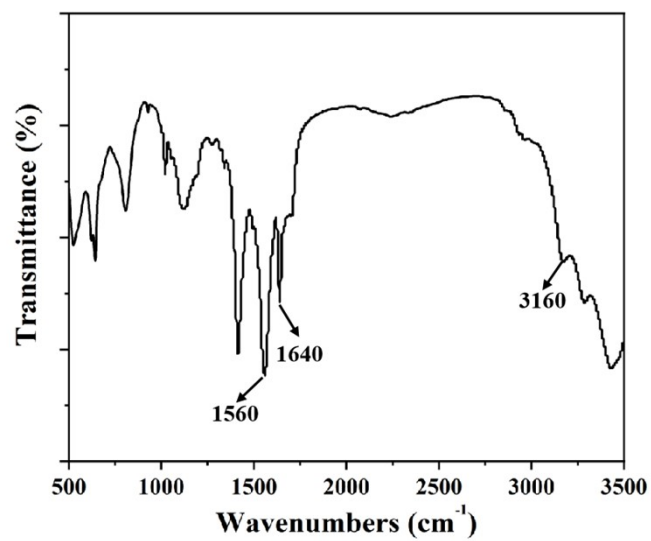
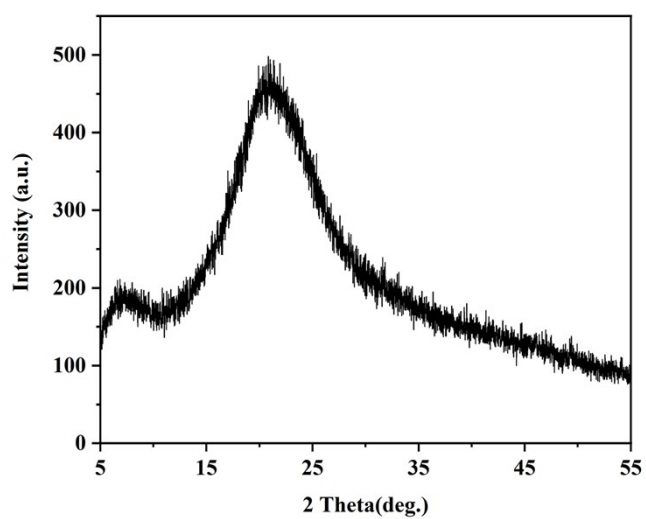


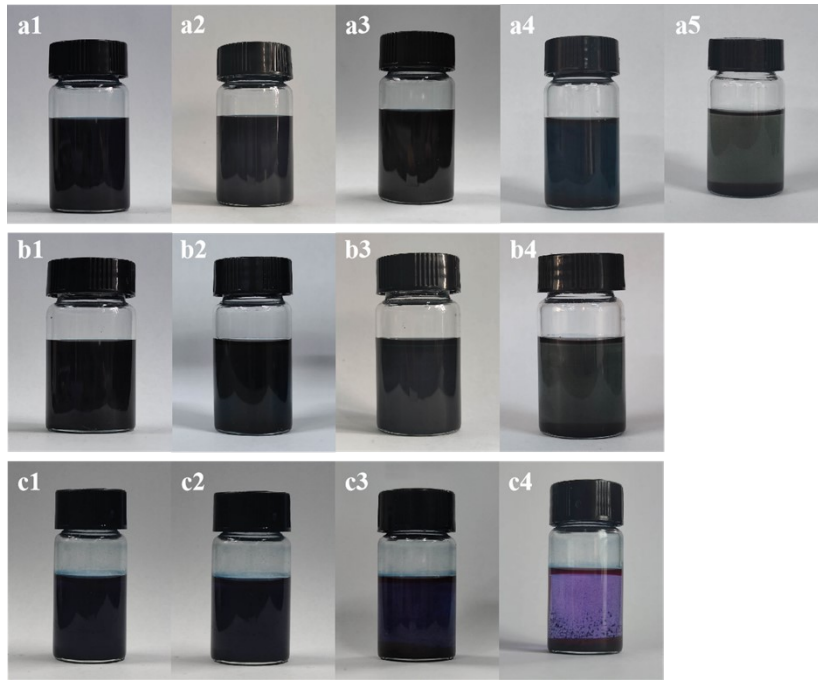
Fig. S2 Size distribution of COF.



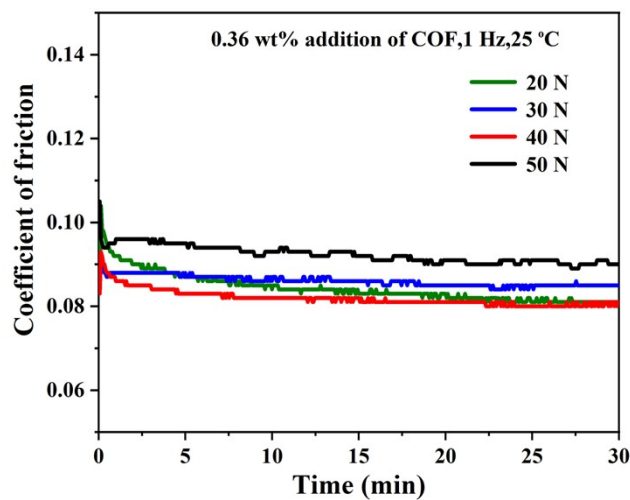
**Fig. S3** FTIR data of COF.



**Fig. S4** XRD pattern of COF.



**Fig. S5** (a1-a5) Optical images of 500 SN containing 0.125 wt% COF after ultrasound treatment for 10 min, and after 3 d, 7 d, 14 d and 30 d. (b1-b4) Optical images of 5W-30 containing 0.2 wt% COF after ultrasound treatment for 10 min, and after 3 d, 7 d, and 14 d. (c1-c4) Optical images of liquid paraffin containing 0.2 wt% COF after ultrasound treatment for 10 min, and after 1 d, 3 d, and 7 d.



**Fig. S6** Friction coefficient curves under different loads.

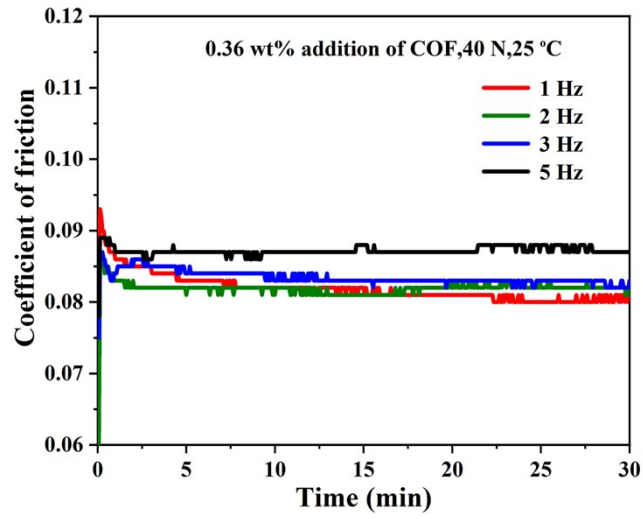


Fig. S7 Friction coefficient curves under the different frequency.

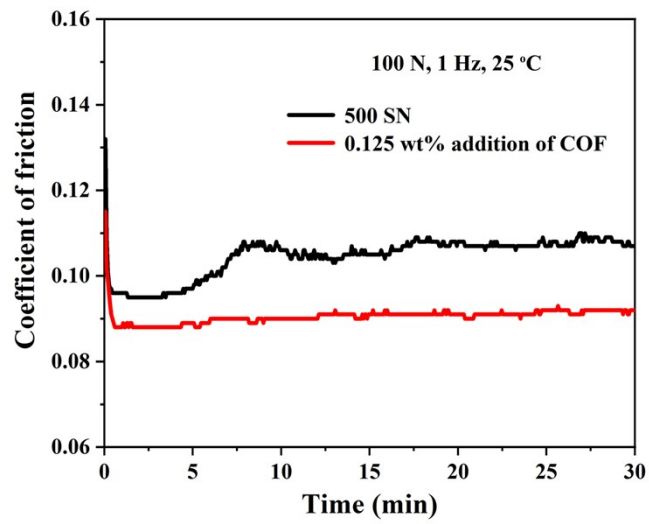
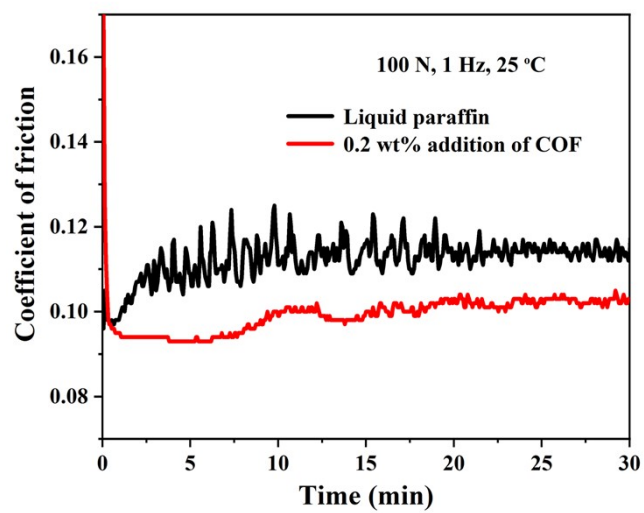


Fig. S8 Friction coefficient curves of 500 SN with 0.125 wt% addition of COF.



**Fig. S9** Friction coefficient curves of LP with 0.2 wt% addition of COF.