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## Supporting Information

# TTF-TCNQ Complex: An Organic Charge-Transfer System with Extraordinary Electromagnetic Response Behaviors

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## **1. Experimental section**

### 1.1 Materials

TTF and TCNQ were purchased from Shang Hai Aladdin Company, and anhydrous acetonitrile was supplied by Alfa chemical Company. Other chemicals were used directly without further purification.

1.2 The Synthesis of TTF-TCNQ complex (Scheme 1)

50 mg of TTF was dissolved in 20 mL of acetonitrile under stirring to furnish solution A. 50 mg of TCNQ was dissolved in 20 mL of acetonitrile under stirring to

provide solution B. At 60 °C, solution A was dripped into solution B under stirring for 30 minutes, and then gradually cooled to room temperature. The solid was obtained by filtration, washed with acetonitrile and dried under vacuum to furnish the target TTF-TCNQ complex.

#### 1.3 Characterization

Fourier transform infrared (FT-IR) spectra was recorded on a Nicolet IS-10 spectrometer with ATR attachment. The X-ray diffraction (XRD) patterns was measured on a Bruker D8 ADVAVCE with Cu K $\alpha$  radiation (0.154 nm) at 40 kV and 40 mA. The operating 2  $\theta$  angle ranges from 5 to 80°. X-ray Photoelectron Spectroscopy (XPS) spectra was recorded on ESCALABTM 250Xi apparatus. The morphology features of TTF-TCNQ were observed by a JEOLJSM-6380LV scanning electron microscope (SEM) working at 30 kV. The high-resolution images were obtained by a field emission transmission electron microscope (TEM) (Tecnai G<sup>2</sup> F20UTwin, FEI). The complex permittivity ( $\varepsilon_r$ ) and permeability ( $\mu_r$ ) were measured by a vector network analyzer (VNA, N5242A PNA-X, Agilent). The measured samples were mixing with wax at 85 °C, then pressed into toroidal-shaped samples (outer diameter: 7.00 mm, inner diameter: 3.04 mm).

## 2. Supporting figures



Figure S1.  $\mu'$  (a),  $\mu''$  (b), tan $\delta_{\mu}$  (c) curves of TTF-TCNQ/wax composites with different filler

loading ratio, hysteresis loop of TTF-TCNQ (d).



Figure S2. Cole-cole plots of TTF-TCNQ with different filler loading ratio, 20 wt% (a), 45 wt%

(b).