

Supplementary Information

Controlled Potential Electrodeposition of a Microcrystalline Thin Film of The Charge Transfer Material Tetrathiafulvalene-Polyoxometalate of approximate composition $(\text{TTF})_{5.3}(\text{Bu}_4\text{N})_{0.7}[\text{P}_2\text{W}_{18}\text{O}_{62}] \cdot 3\text{H}_2\text{O}$

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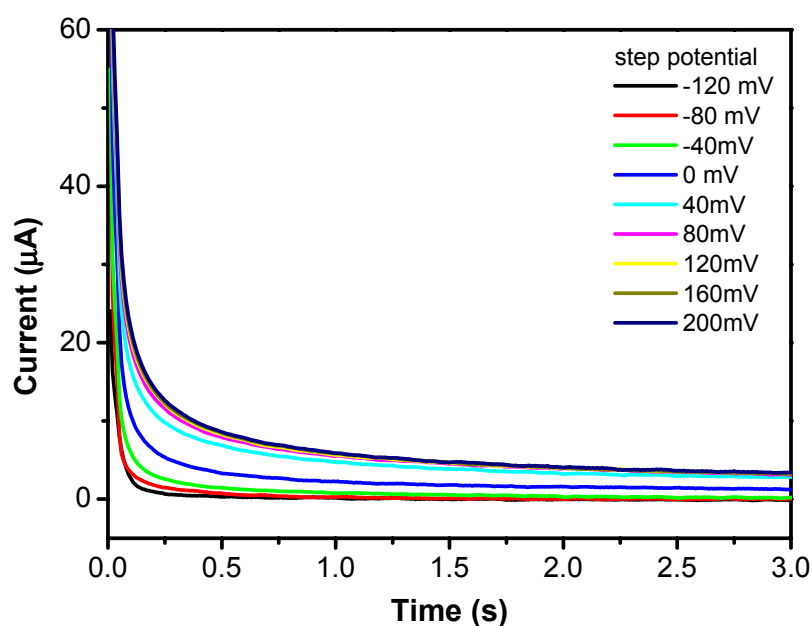


Figure S1. Chronoamperometric (*i-t*) transients obtained at a 3 mm GC electrode in acetonitrile (0.1 M $[\text{Bu}_4\text{N}][\text{PF}_6]$) for a mixture of TTF (0.3 mM) and $\text{K}_6\text{P}_2\text{W}_{18}\text{O}_{62}$ (0.1 mM) when the potential was stepped from an initial value (-0.5 V) to more positive potentials (-0.12 to 0.2 V with 40 mV increments).

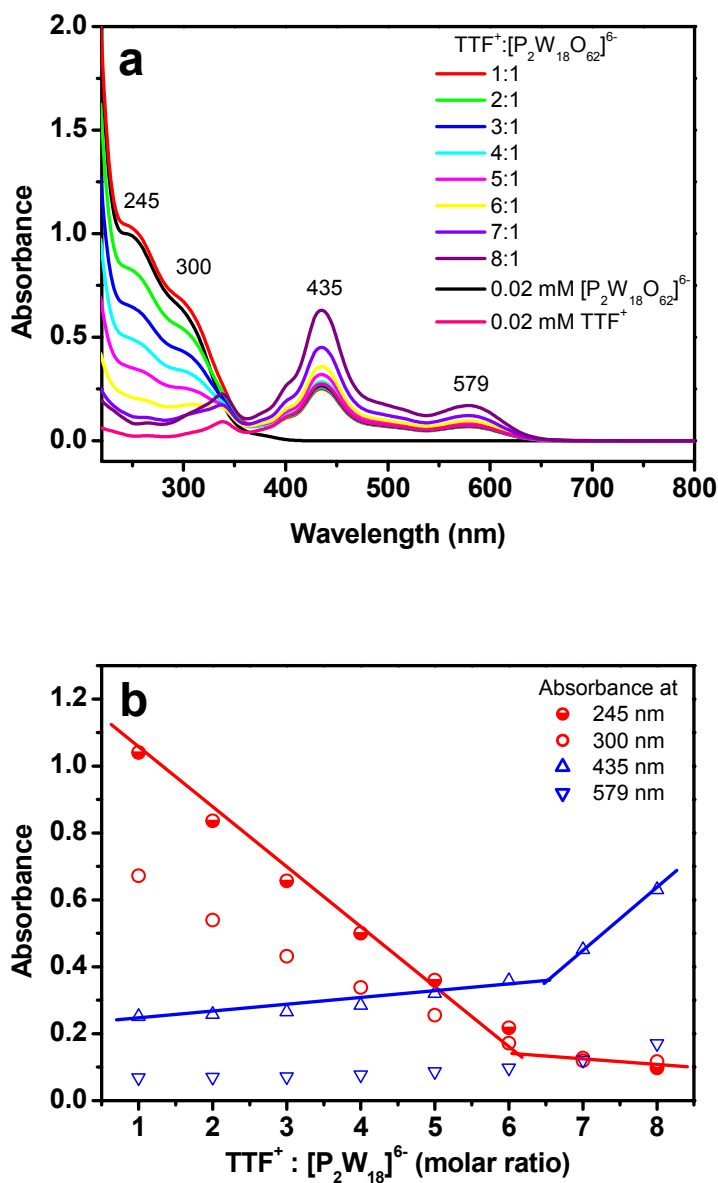


Figure S2. The UV-vis spectra (a) of the supernatants obtained from the addition of 2 mL [TTF][PF₆] in MeCN (0.1 M [Bu₄N][PF₆]) with varied concentrations (0.2, 0.4, 0.6, 0.8, 1.0, 1.2, 1.4, 1.6 mM) to 2 mL MeCN solution of 0.2 mM [P₂W₁₈O₆₂]⁶⁻ anion. The supernatants were collected 10 min after the addition of [TTF][PF₆], followed by five-fold dilution before the measurements. The absorptions at

characteristic wavelengths were plotted (b) as a function of the molar ratio of TTF⁺ to [P₂W₁₈O₆₂]⁶⁻. Fitted lines were shown to guide the eyes.

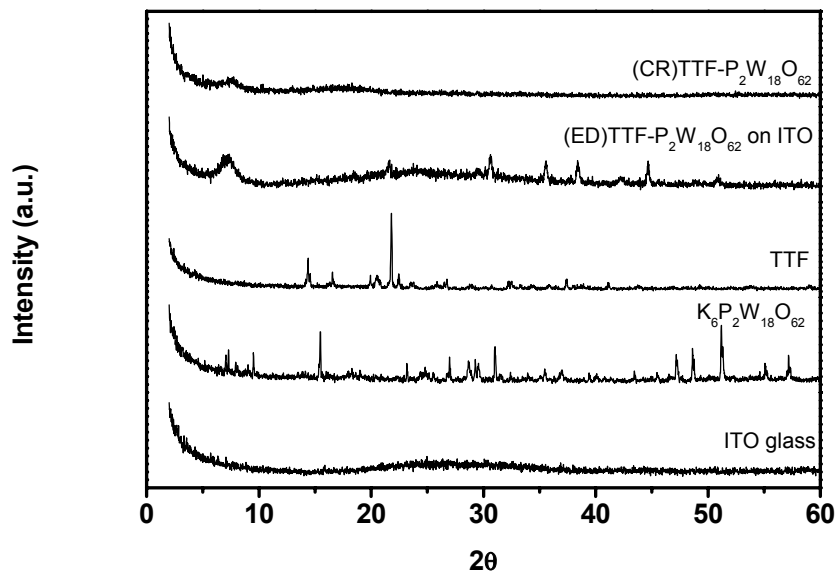


Figure S3. X-ray powder diffraction patterns of TTF-P₂W₁₈O₆₂ material produced chemically ((CR)TTF-P₂W₁₈O₆₂) or by electrodeposition ((ED)TTF-P₂W₁₈O₆₂) on ITO glass, compared to XRD of TTF, K₆P₂W₁₈O₆₂ and bare ITO glass.