

Supplementary Information

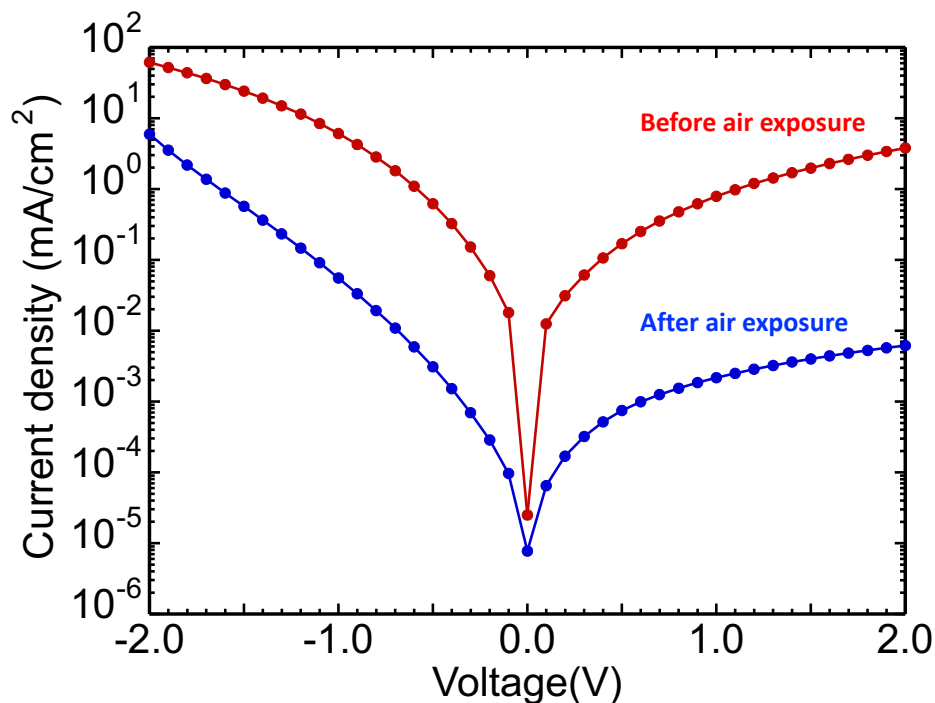
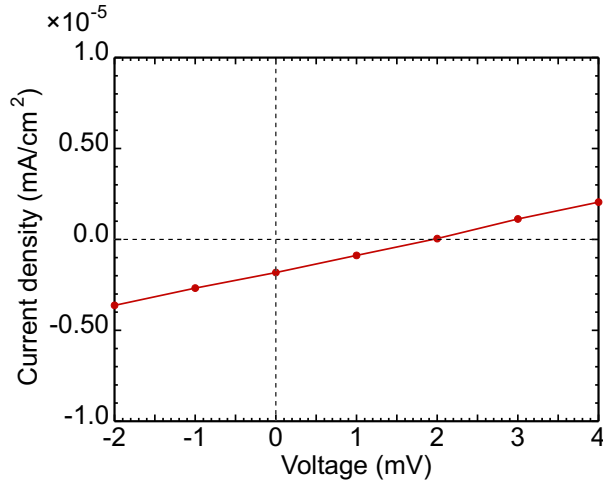


Figure S1: Influence of air exposure for current density-voltage (J - V) characteristics of CuPc/F₁₆CuPC-based device. The device is composed of ITO (100nm)/CuPc (30 nm)/F₁₆CuPC (70 nm)/Al (100 nm). The red and blue lines correspond to before and after air exposure, respectively. CuPc/F₁₆CuPC bilayer devices provided unstable J - V characteristics occasionally since these molecules formed polycrystalline textures in their thin films formed by vacuum evaporation. Therefore, there is a discrepancy between the J - V characteristics of **Figs. 1(c)** and **S1**. However, such instability has been significantly overcome by forming a C₆₀/BCP layer on top of the CuPc/F₁₆CuPC layers, resulting in highly reproducible J - V and thermoelectric properties.

(a)



(b)

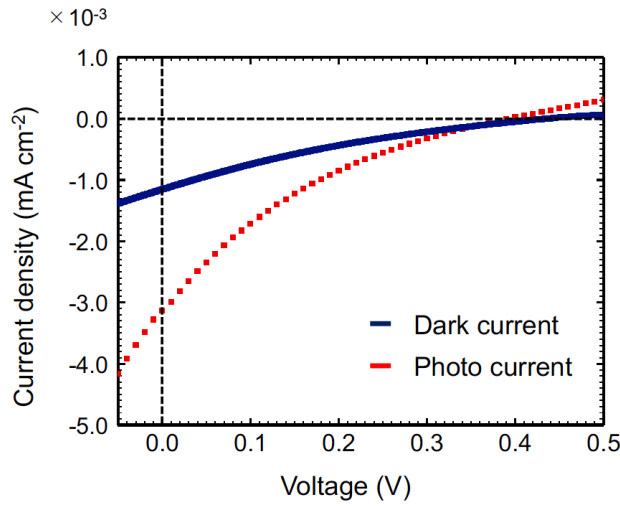


Figure S2: (a) Current density-voltage (J - V) characteristics of CuPc/C₆₀/BCP-based device. J - V characteristics of ITO (100 nm)/CuPc (20 nm)/C₆₀ (40 nm)/BCP (10 nm)/Al (100 nm) device near the origin, demonstrating the existence of a very small-scale thermoelectric property. The measurement circuit was zero-corrected before the measurements were taken. **(b)** J - V characteristics of ITO (100 nm)/CuPc (110 nm)/F₁₆CuPc (45 nm)/C₆₀ (40 nm)/BCP (20 nm)/Al (100 nm) device under dark and light irradiation condition (AM1.5).

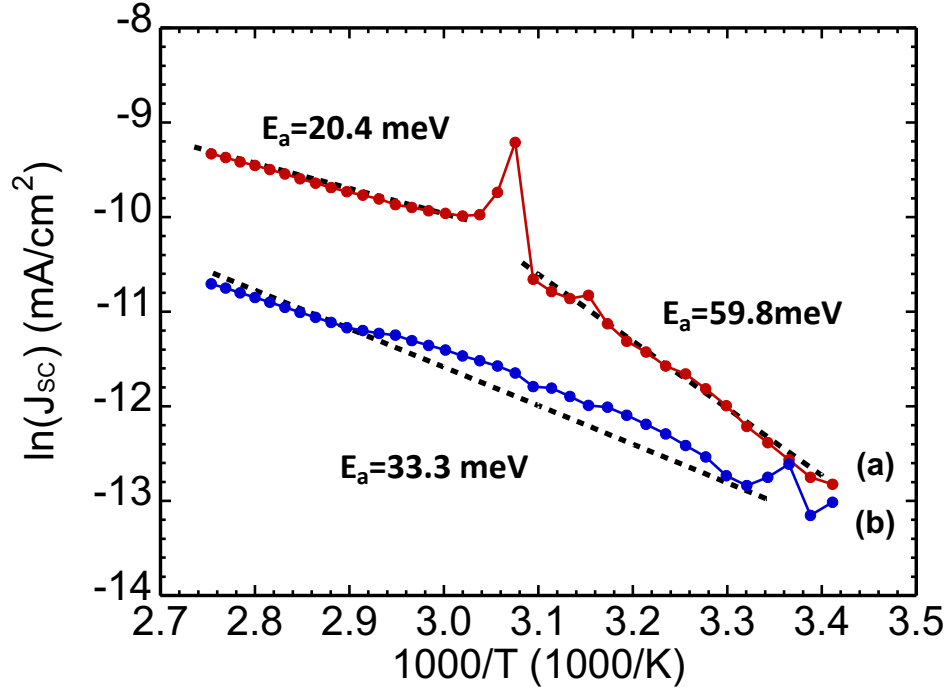


Figure S3: Temperature dependence of thermoelectric characteristics. a, $\ln(J_{sc})$ vs. $1000/T$ characteristics of the ITO (100 nm)/CuPc (30 nm)/F₁₆CuPc (20 nm)/C₆₀ (40 nm)/BCP (20 nm)/Al (100 nm) device. Two activation energies of $E_a=59.8$ meV (low-temperature region) and $E_a=20$ meV (high-temperature region) were observed. **b,** $\ln(J_{sc})$ vs. $1000/T$ characteristics of the ITO (100 nm)/CuPc (30 nm)/F₁₆CuPc (70 nm)/Al (100 nm) device. There is a slight deviation between the fitted and experimental data as a first approximation, resulting in $E_a=33.3$ meV.

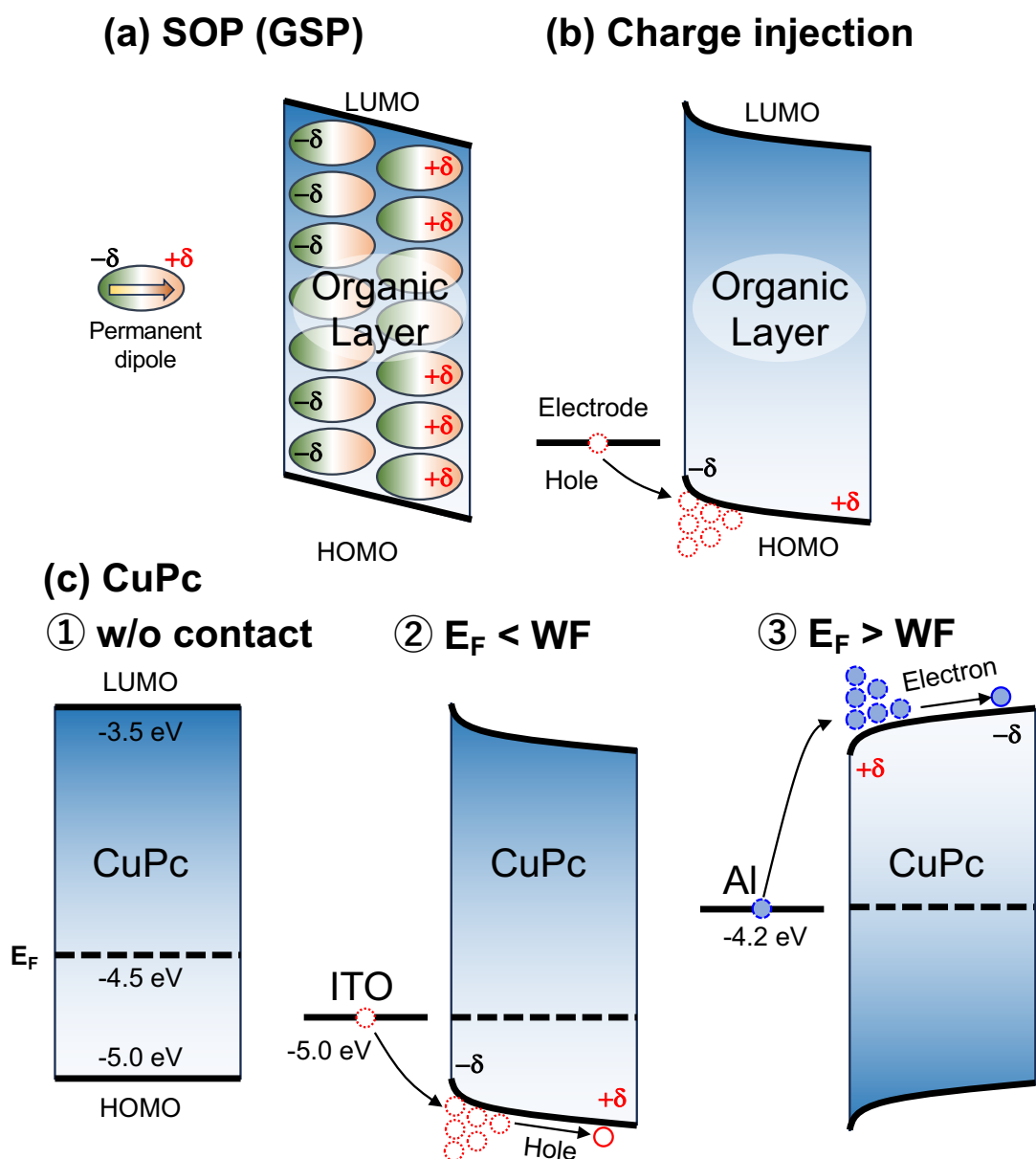


Figure S4: Schematic energy diagrams of the surface potential changes induced by SPO and charge injection. a, SOP (GSP) and b, charge injection and successive accumulation. c, ① Energy diagram of the CuPc layer without electrode contact, ② change in energy potential of the CuPc layer on the ITO electrode with $E_F < WF_{ITO}$ (the ITO work function), and ③ change in energy potential of the CuPc layer on the Al electrode with $E_F > WF_{Al}$ (the Al work function).

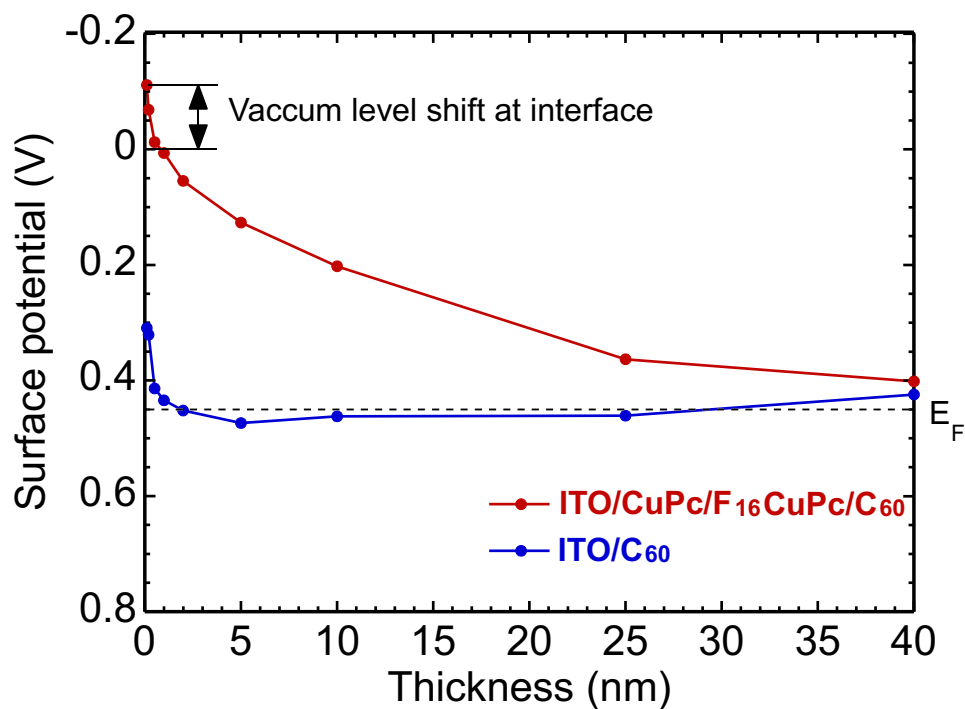


Figure S5: C₆₀ thickness dependence of the surface potential. The red and blue lines correspond to ITO (100 nm)/CuPc (40 nm)/F₁₆CuPc (40 nm)/C₆₀ (*X* nm) and ITO (100 nm)/C₆₀ (*X* nm) structures, respectively.

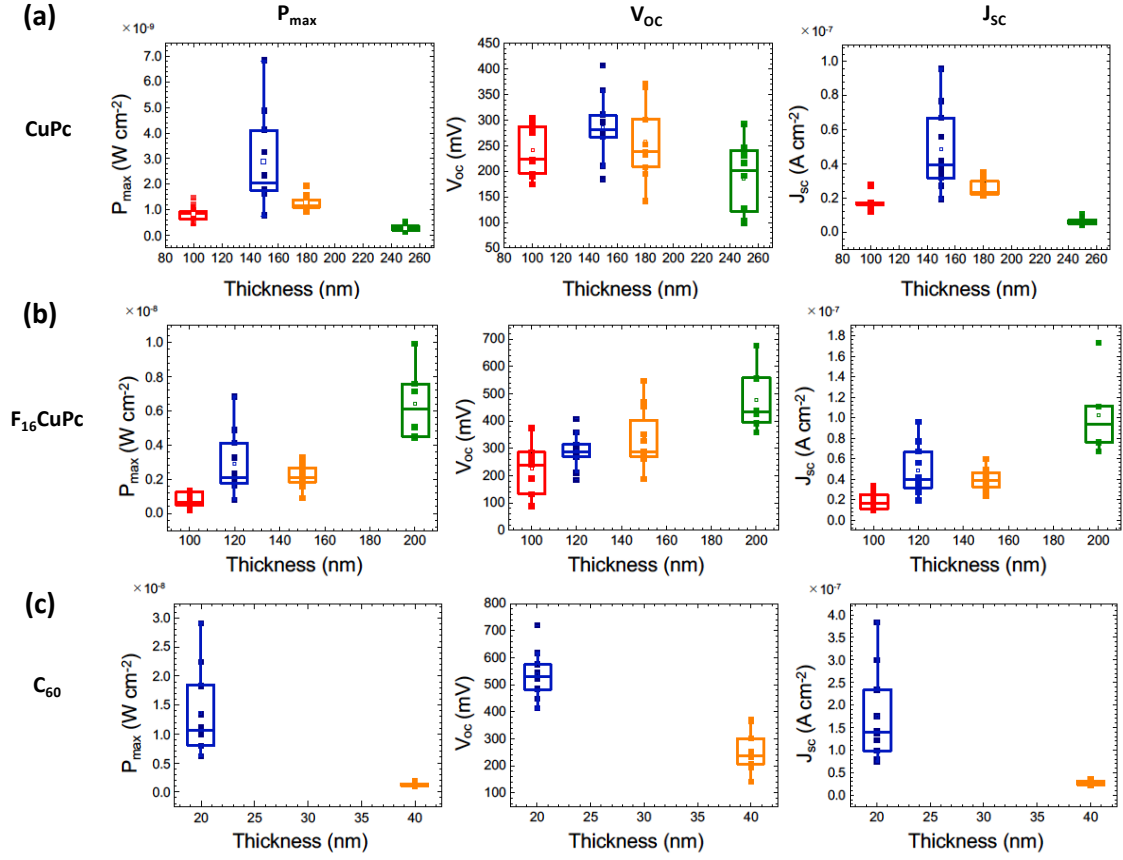


Figure S6: Thickness dependences of thermoelectric characteristics. **a**, CuPc thickness dependance of P_{max} , V_{OC} , and J_{SC} change in ITO (100 nm)/CuPc (X nm)/F₁₆CuPc (120 nm)/C₆₀ (40 nm)/BCP (20 nm)/Al (100 nm), **b**, F₁₆CuPc thickness dependance of P_{max} , V_{OC} , and J_{SC} in ITO (100 nm)/CuPc (150 nm)/F₁₆CuPc (X nm)/C₆₀ (40 nm)/BCP (20 nm)/Al (100 nm), and **c**, C₆₀ thickness dependance of P_{max} , V_{OC} , and J_{SC} in ITO (100 nm)/CuPc (180 nm)/F₁₆CuPc (120 nm)/C₆₀ (X nm)/BCP (20 nm)/Al (100 nm).