Supporting information

Rational Design of Molecular Crystals for Enhanced Charge Transfer Properties

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CONTENTS

1. **SEM image and EDS element mapping of BTBT-TCNQ bilayer structure sample.**

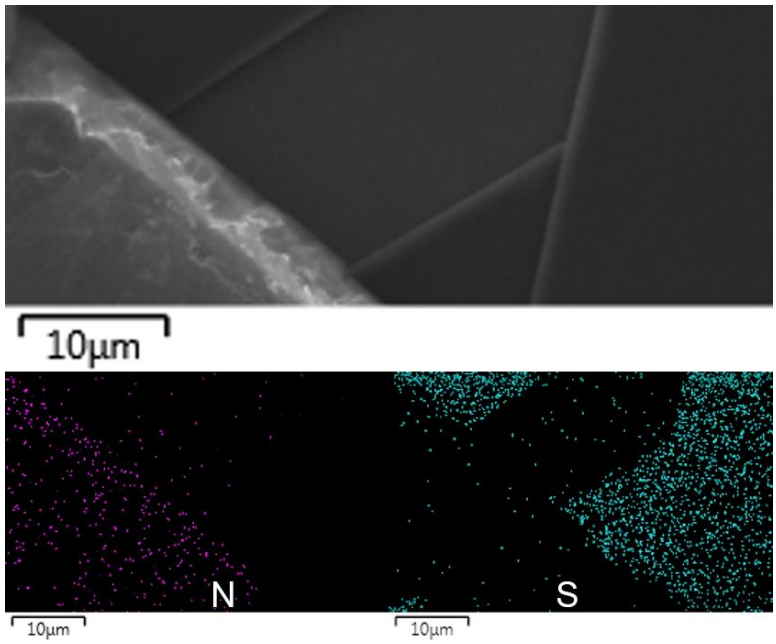

Figure S1 SEM image and EDS element map of BTBT-TCNQ sample, scale bar is 5um.

2. **XRD patterns of BTBT-TCNQ and C_{10}-BTBT-TCNQ bilayer structure.**

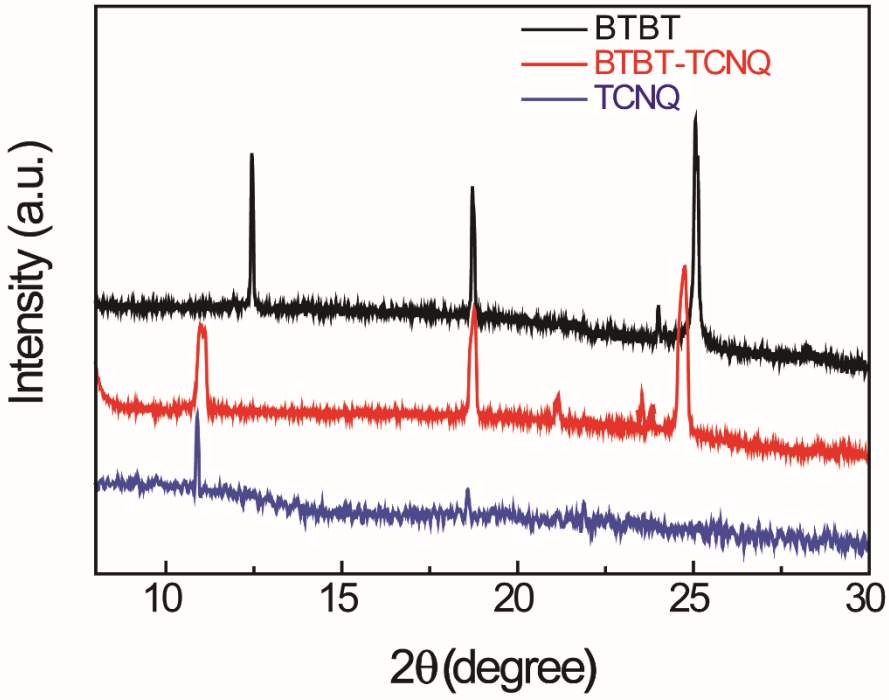

Figure S2 XRD patterns of BTBT, TCNQ and BTBT-TCNQ samples.

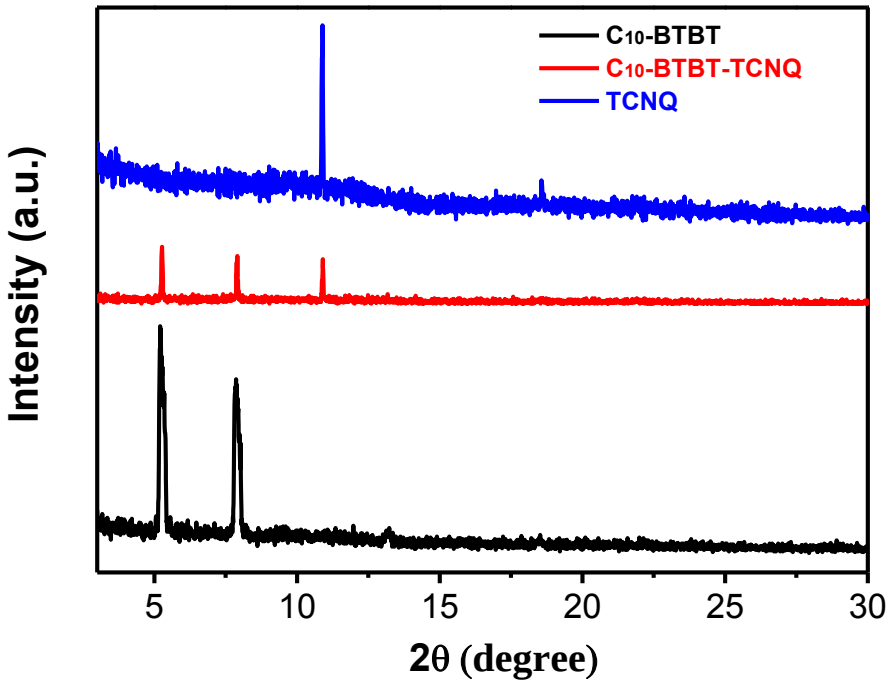

Figure S3 XRD patterns of C_{10}-BTBT, TCNQ and C_{10}-BTBT-TCNQ samples.

3. UV-Vis absorption spectra of BTBT-TCNQ and C_{10}-BTBT-TCNQ bilayer structure.

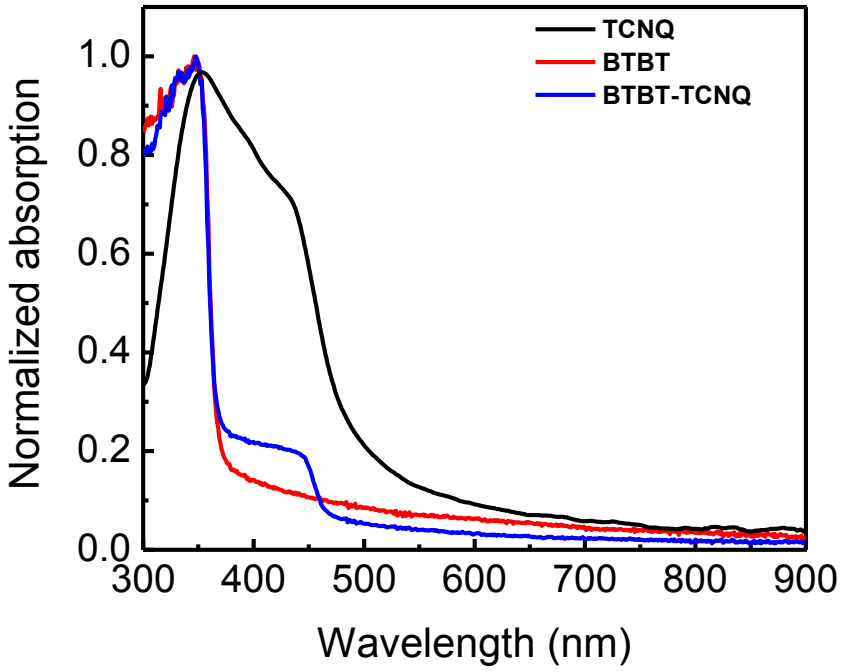

Figure S4. UV-Vis absorption of BTBT, TCNQ and BTBT-TCNQ samples.

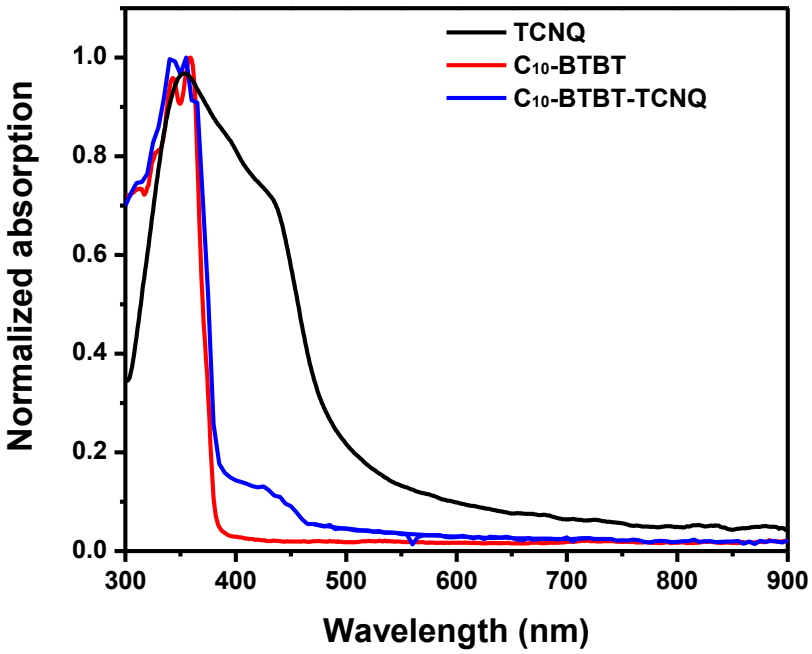

Figure S5 UV-Vis absorption of C_{10}-BTBT, TCNQ and C_{10}-BTBT-TCNQ samples.

4. **XRD pattern and UV-Vis spectrum of BTBT:TCNQ cocrystals.**

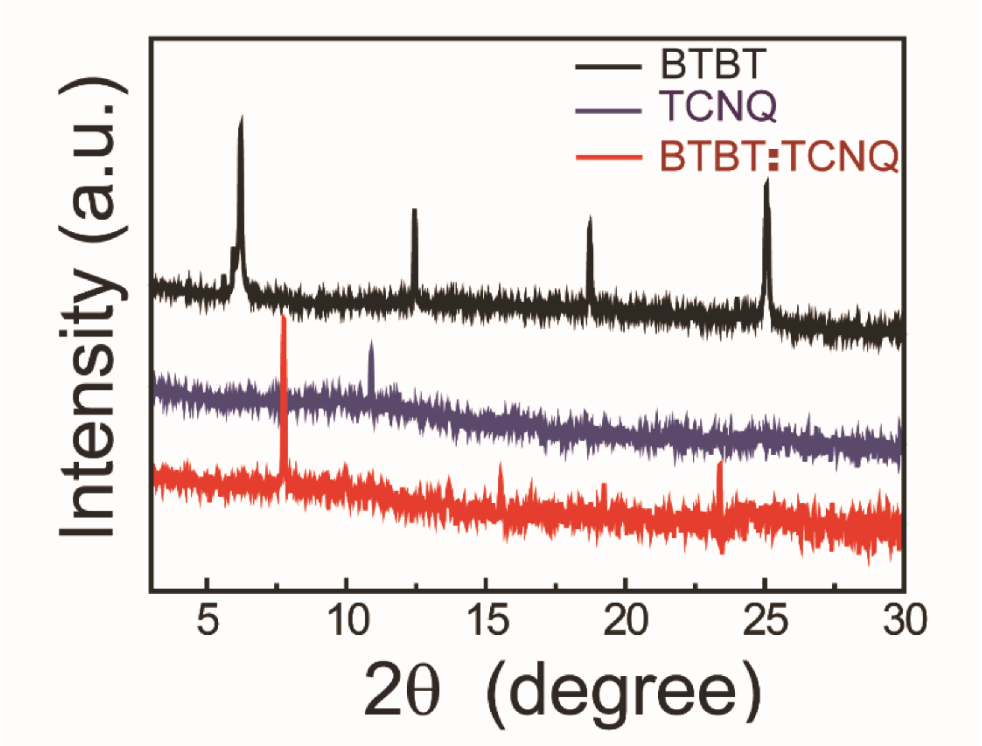

Figure S6 XRD pattern of BTBT:TCNQ cocrystals.

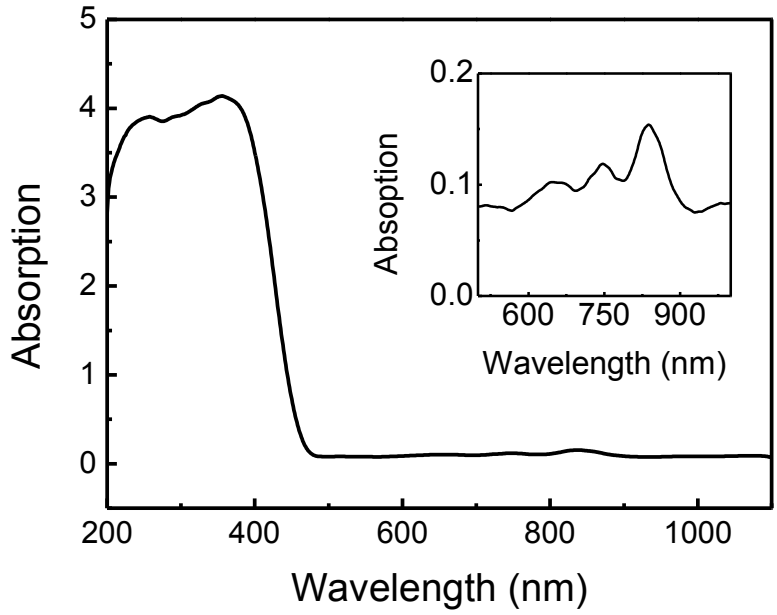

Figure S7 UV-Vis spectrum of BTBT:TCNQ cocrystals.

5. The measured current density-voltage (I-V) curves for two bilayer samples and BTBT:TCNQ cocrystals.

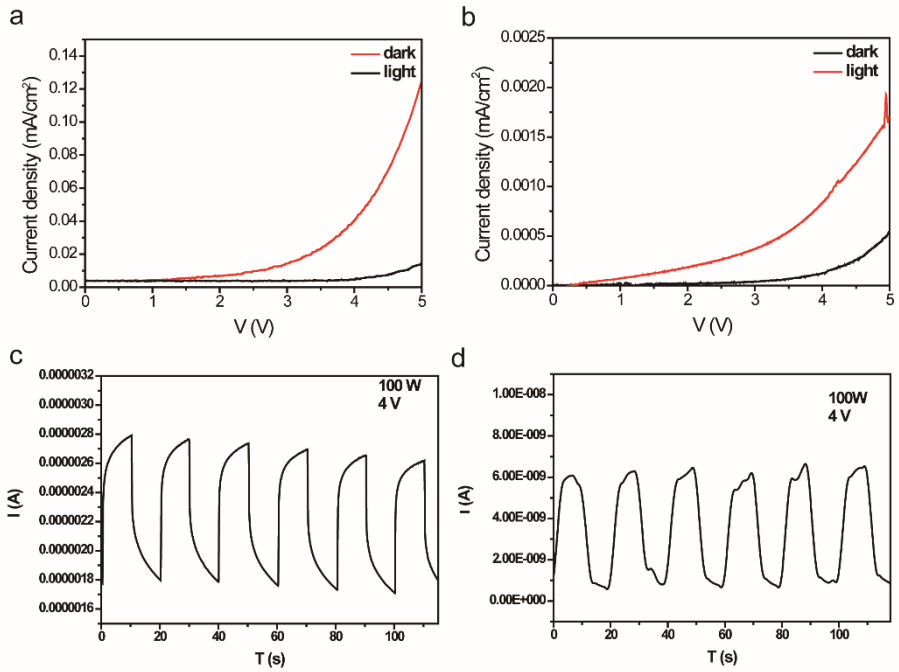

Figure S8 The measured current density-voltage (I-V) curves of (a, c) the BTBT-TCNQ bilayer and (b, d) C_{10}-BTBT-TCNQ bilayer under dark and light illumination.

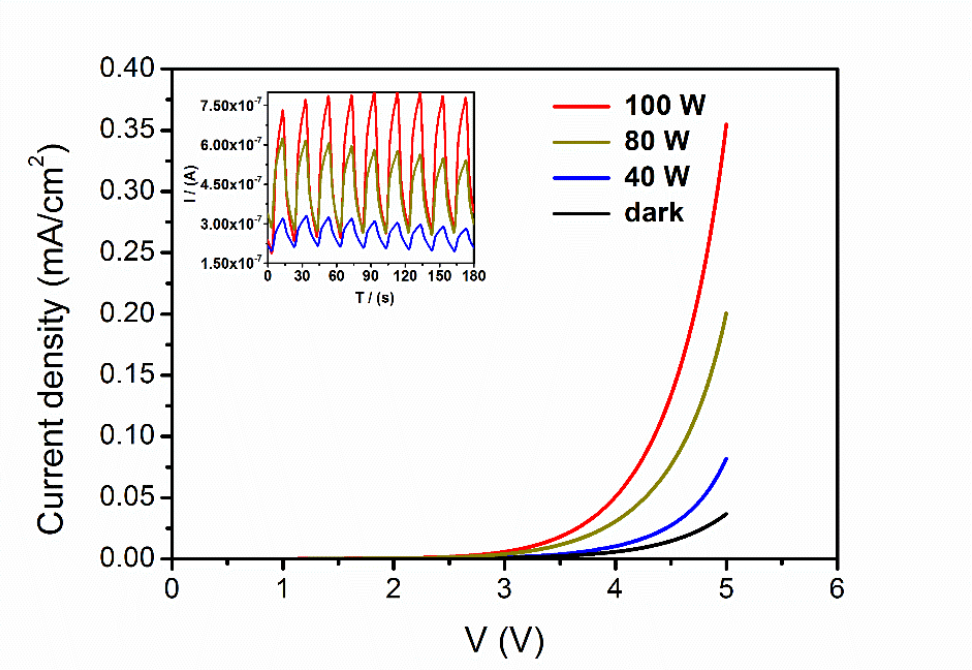

Figure S9 The measured current density-voltage (I-V) curves of the BTBT:TCNQ co-crystal under dark and light illumination.

6. The measured pyroelectric properties under external magnetic field with different intensity.

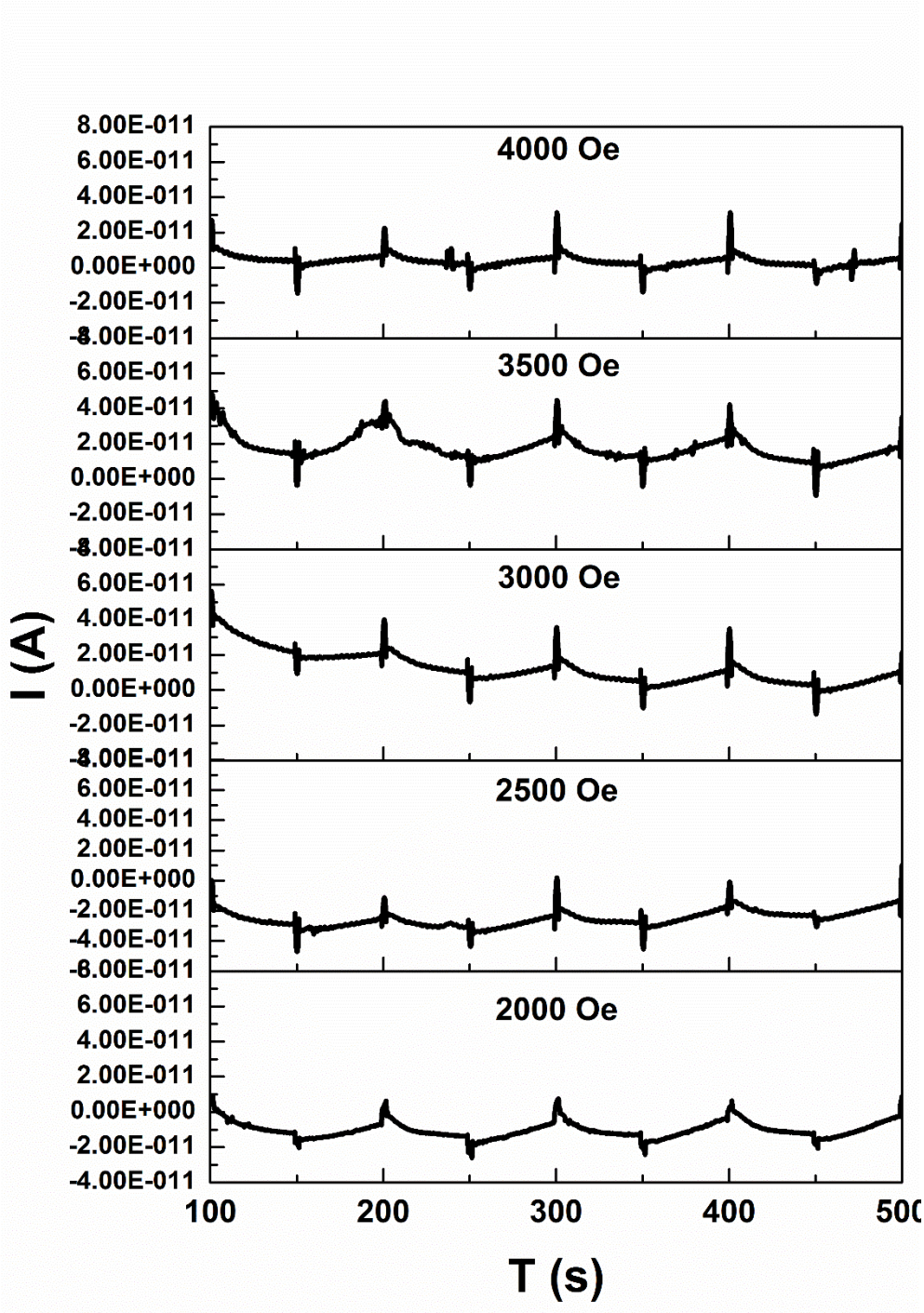

Figure S10 The measured pyroelectric current of the BTBT:TCNQ co-crystal under different intensity of external magnetic field.

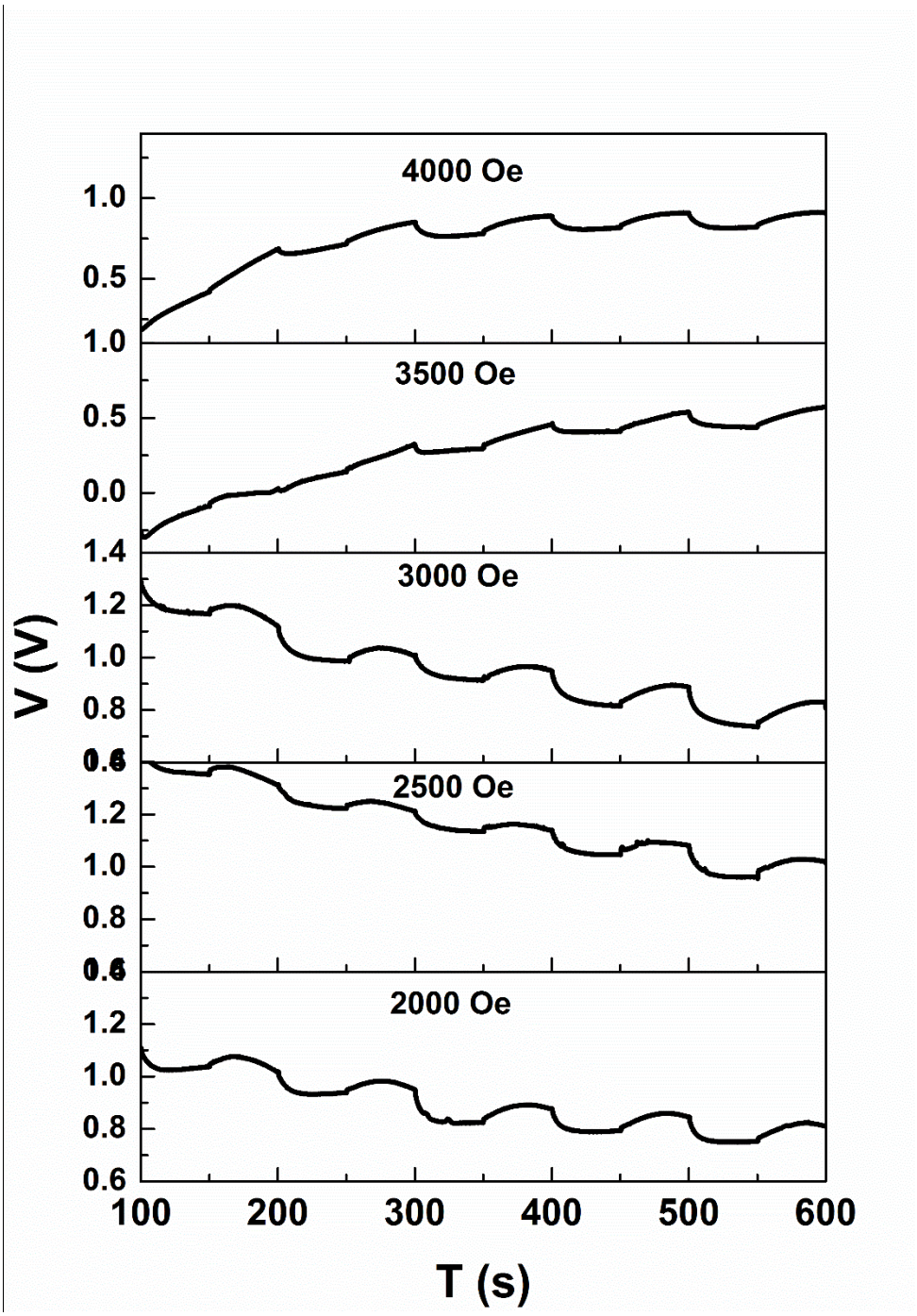

Figure S11 The measured pyroelectric voltage of the BTBT:TCNQ co-crystal under different intensity of external magnetic field.