Supporting Information

## Organic Single Crystal Surface-induced Polymerization of Conducting Polypyrrole

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Figure S1. Polypyrrole produced under different polymerization temperatures. a)– d), SEM images (scale bar: 10  $\mu$ m) of polypyrrole synthesized under different temperatures: a), 0 °C; b), 5 °C; c), 10 °C; and d), 15 °C.



**Figure S2. FT-IR spectra of synthesized PPy samples.** a), b), c) and d) PPy synthesized at 0, 5, 10 and 15 °C in the presence of KSBA, respectively; e) and f) PPy synthesized at 0 and 15 °C without KSBA, respectively.



Figure S3. N1s core level spectra of x-ray photoelectron spectroscopy. a), b) PPy synthesized at 0 and 15 °C in the presence of KSBA, respectively; c) PPy synthesized at 0 °C without KSBA. The area ratio of  $(=N^+- + -NH^+-)/N_{total}$  indicating the doping level of the samples is 0.25, 0.24, and 0.25 in a)–c), respectively, and the respective average electrical conductivity is 412, 174, and 75 Scm<sup>-1</sup>. This indicates that the samples have similar doping levels irrespective of the polymerization conditions.



**Figure S4. Qualitative measurement of conjugation length**. **a)** Transmission Fourier transform infrared (FT-IR) spectra of PPy samples polymerized in the presence of KSBA at a given temperature. The 1560 and 1480 cm<sup>-1</sup> bands are aligned vertically in these spectra; **b)** Plot of the conductivity versus the ratio of the integrated absorption intensities of the 1560 and 1480 cm<sup>-1</sup> bands for the samples in Figure S2a. According to Tian and Zerbi, the conjugation length is inversely proportional to the ratio of the peak areas at 1560 and 1480 cm<sup>-1</sup>, indicative of antisymmetric ring stretching mode and symmetric mode in the Py ring, respectively. It is clear that the log conductivity of our synthesized PPy is linearly related to  $A_{1560}/A_{1480}$ .



Figure S5. SEM image of the KSBA crystals at polymerization t = 60 min. White arrows indicate the fragments of the KSBA crystals by continuous dissolution and mechanical stirring. Therefore, the PPy structure patterned after the pre-existing KSBA crystal is easily separated from the KSBA crystal.