## **Electronic Supplementary Information (ESI)**

## Highly Stretchable Coaxial P3HT Electrospun Fibers with Enhanced Reversibility

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**Fig. S1** a) Schematic of the rotating collector and inset showing the paper cut with fiber mat; b) custom-made tensile testing set up.

Fig. S1a shows that fiber mats were collected by placing a paper frame on a rotating collector. The gap between the paper frame's parallel strips determined the electrospun fiber mat's gauge length ( $l_0$ ). The gauge region of the fiber mat had a length of 5 to 10 mm, a width of 10 mm, and a thickness of 5 to 10  $\mu$ m. As shown in Fig. S1b, the tensile test setup supported the paper-cut samples with two pairs of rectangular aluminum slabs. Fiber mats were stretched to failure at a low tensile speed of 0.25 mm s<sup>-1</sup> and speed was controlled by a NI LabVIEW program.



Fig. S2 Optical micrographs of two fibers crossing connecting two electrodes.



Fig. S3 Optical micrographs of coaxial fibers of 10% PMMA (core) and 3% P3HT (shell).



**Fig. S4** Optical micrographs of coaxial fibers showing the progression to bead-free fibers with a higher concentration of BR. a) Beaded fibers of 3%BR-3%P3HT; b) Beaded fibers of 6%BR-3%P3HT; c) Bead-free fibers of 10% BR-3%P3HT.



Fig. S5 Effects of P3HT flow rate (shell side) on fiber formation: a) discontinuous fibers, b)

uniform fibers, and c) beaded fibers. Here, BR was at the core.



Fig. S6 Core-shell P3HT\_BR electrospun fibers showing the cylindrical shape.



Fig. S7 Cross-sectional view of multiple coaxial fibers obtained using TEM. The image contrast was adjusted to differentiate the core and shell layers.



**Fig. S8** Electrical conductivity measurement of a single fiber. Here, A, I, V, L, R<sub>1</sub>, and R<sub>2</sub> are the fiber cross-sectional area, current, voltage, length, overall radius, and core radius of the fiber, respectively.

The cross-section of P3HT\_BR fiber was measured using TEM, and the conductivity of a single fiber was determined using the above equations.



Fig. S9 I-V responses of fibers with different flexible core polymers.



1035% strain

Fig. S10 (a-f) Mechanical stretching of fibers; g) The crack onset at 1035% strain. The scale bar represents 10 mm.