Supporting Information Available

In Figure S1, FT-IR spectra of pure PU and PU/PANi hybrid nanofibrous bundle are presented. The spectra of PU/PANi hybrid nanofibrous bundle showed peaks around 1570 cm^{-1} and 1490 cm^{-1} which were attributed to quinoid and benzenoid C=C vibrations respectively. The peaks around 1300 cm^{-1} and at 1252 cm^{-1} were due to C-N stretching of PANi. The out of plane N-H bending also occur in this region. The sharp band observed at 1143 cm^{-1} was attributed to the charged defect. The peak at 1720 cm^{-1} was due is the carbonyl stretching of PU. This peak is shifted to lower frequency as compared to the spectra of PU which was attributed to the possible H-bonding interaction between –NH of PANi and C=O group of PU. In addition to this, the FT-IR spectra of PU showed peak around 1531, 1462, 1367 and 1254 cm^{-1} which were attributed to aromatic C-C stretching, -CH_3 asymmetric deformation, amide bond and phenyl –O stretching respectively. The band corresponds to N-C-N stretching are observed around 1063 cm^{-1}.

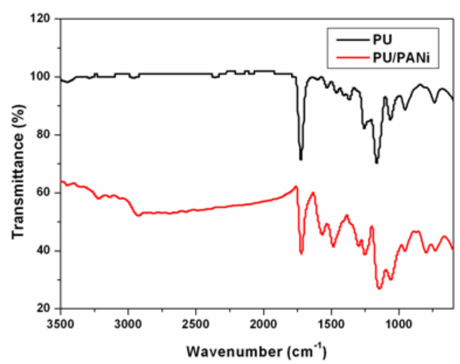

Figure S1. FT-IR shows peaks of pure PU and PU/PANi hybrid nanofibrous bundle.

There is considerable curve deviation at higher scan rates. The calculated diffusion coefficient values support the increase in redox peak current with scan rate as observed in the CV curve. The redox peaks is indicative of Faradaic capacitance. The voltammetric current is directly proportional to the scan rate indicating capacitive behaviors.

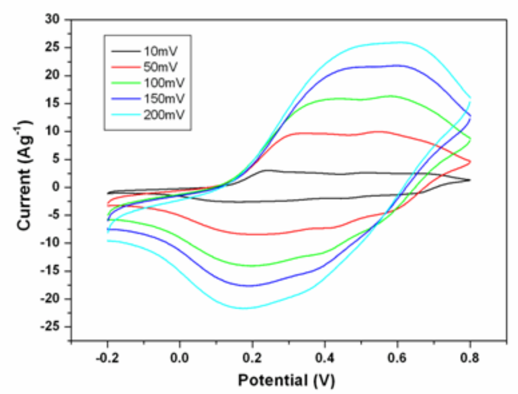

Figure S2. Cyclic voltammogram of the PU/PANi hybrid nanofibrous bundle recorded at different scan rates.

Figure S3 shows long term cycling upon switching the potential between -0.2 V and 0.8 V gave a stable actuation profile. Efficient cyclability up to 100 cycles the work per cycle corresponding is above 75 %.

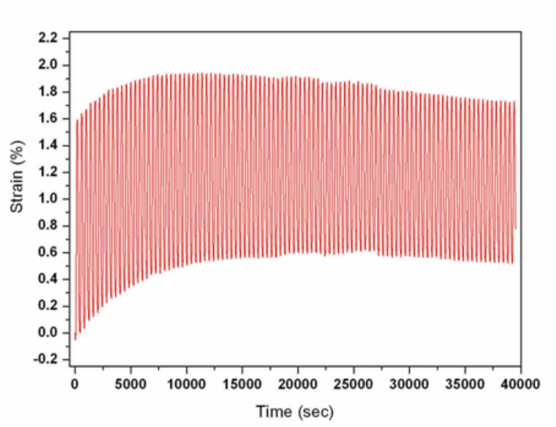

Figure S3. Actuation characteristics of PU/PANi hybrid nanofibrous bundle.

Electrochemical actuation profiles at different applied stresses. The PU/PANi hybrid nanofibrous bundle could be stably actuated without significant creep up to an applied load of 11 mN (2.263 MPa) beyond which significant creep behavior appears.

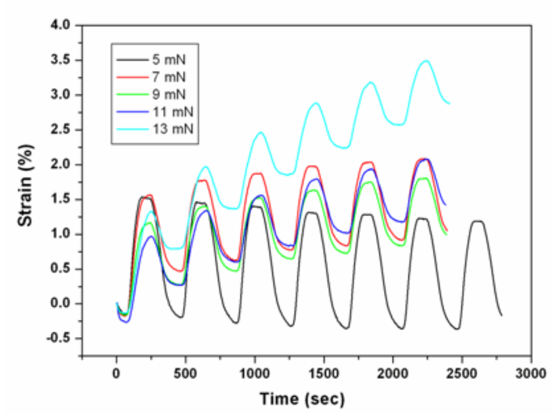

Figure S4. Electrochemical actuation profiles at different applied stress in 1 M MSA (scan rate: 5 mV/sec).

Figure S5 shows stress-strain behavior of nanofibrous bundles tested in the dry state. The hybrid PANi/PU nanofibrous bundle showed a tensile strength of 35 MPa and a linear elastic behavior up to the point a fracture. The elongation at break was observed to be about 90%. The elastic modulus was approximately 37 MPa. Similar testing of the bare PU nanofibrous bundles gave a slightly lower modulus (24 MPa) and strength (25 MPa), but similar elongation at break.

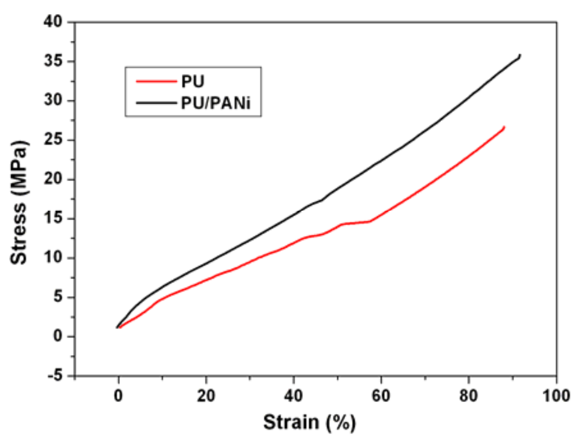

Figure S5. Stress and strain curve of PU/PANi hybrid nanofibrous bundle.

Given that the bundles show iso-strain behaviour where the PANi coating and PU core are strained equally when a stress is applied, then the mechanical behavior can be followed using the standard “rule of mixtures” approximation. The hybrid material modulus will be:

$$E_{Hybrid} = V_{PANi} E_{PANi} + V_{PU} E_{PU}$$

from which the PANi modulus (E_{PANi}) can be calculated to be 40 MPa.