

## Electronic Supplementary Information for

### A conductive stretchable PEDOT-elastomer hybrid with versatile processing and properties

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#### Experimental

##### Composite preparation

###### PGS synthesis and curing

PGS pre-polymer was synthesised using a polycondensation reaction between equimolar amounts of glycerol (99%, Sigma-Aldrich) and sebacic acid (99%, Sigma-Aldrich). Materials were mixed at room temperature and then reacted at 130 °C for 24 h under nitrogen gas purged at a flow rate of  $\sim 130 \text{ cm}^3 \text{ min}^{-1}$ . The produced pre-polymer was dissolved in tetrahydrofuran (THF, 99.9%, Sigma-Aldrich) and then cast on glass slides. Following THF evaporation under ambient conditions, the PGS was cured under vacuum at 130 °C for either 48, 60 or 72 h. Composites fabricated will be designated according to their PGS cure time (i.e. 48h-PGS-PEDOT). The vacuum was released after cooling to room temperature and samples were soaked in water for 24 h and then peeled off the glass slides.

###### Vapour phase polymerization of PEDOT

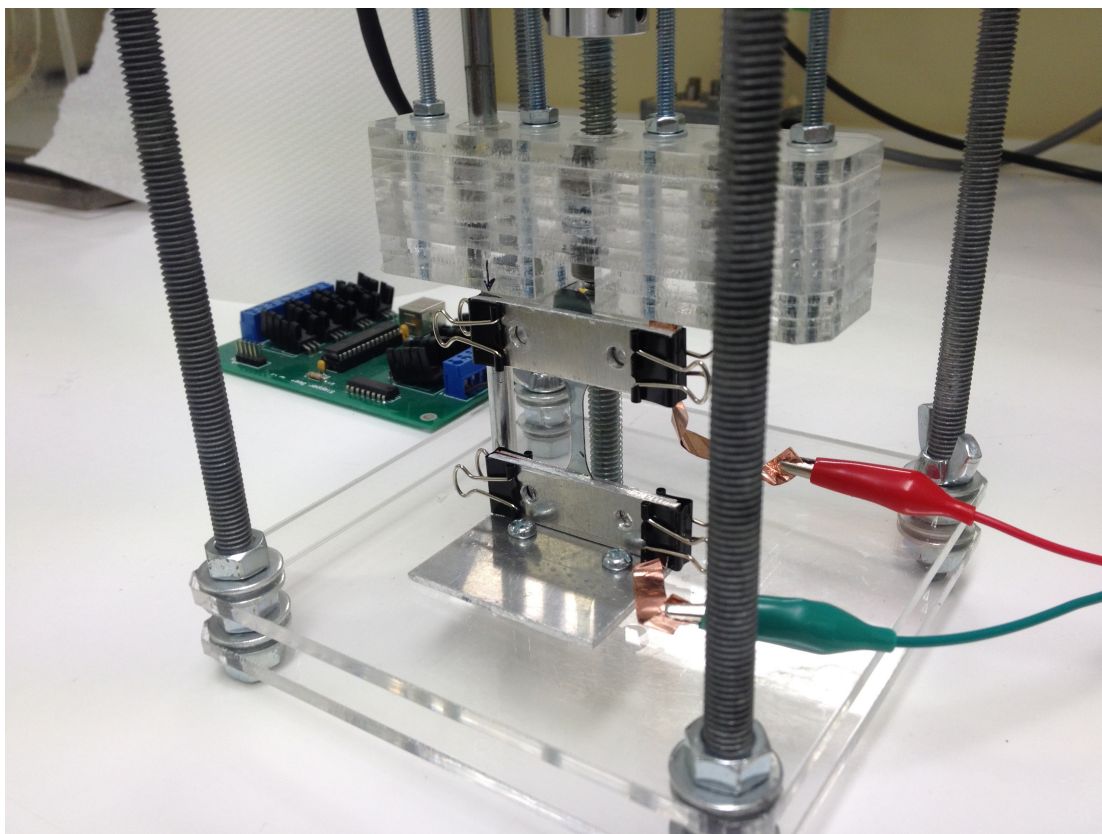
An oxidant solution for VPP of PEDOT was prepared by creating a solution of 1 ml : 23  $\mu\text{l}$  of 40% Fe(III)tosylate (Fe(III)TOS) in butanol (Yacoo Chemicals Ltd.) and pyridine (Ajax Chemicals). The oxidant solution was spincoated (1500 rpm for 30 seconds) onto PGS substrates immediately after deposition. Samples were then transferred to the polymerisation chamber containing 3,4-Ethylenedioxythiophene (EDOT) monomer (97%, Sigma-Aldrich). Samples were polymerised at 70 °C for 60 minutes. Following this, the samples were then removed and washed twice in deionised water for 30 minutes and then once more overnight. PGS films were then cut into dog-bone shaped specimens of 65 x 3.23 x t mm (length x width x thickness, t  $\approx 0.4$  mm) with a gauge length of 12.5 mm

###### Composite characterisation

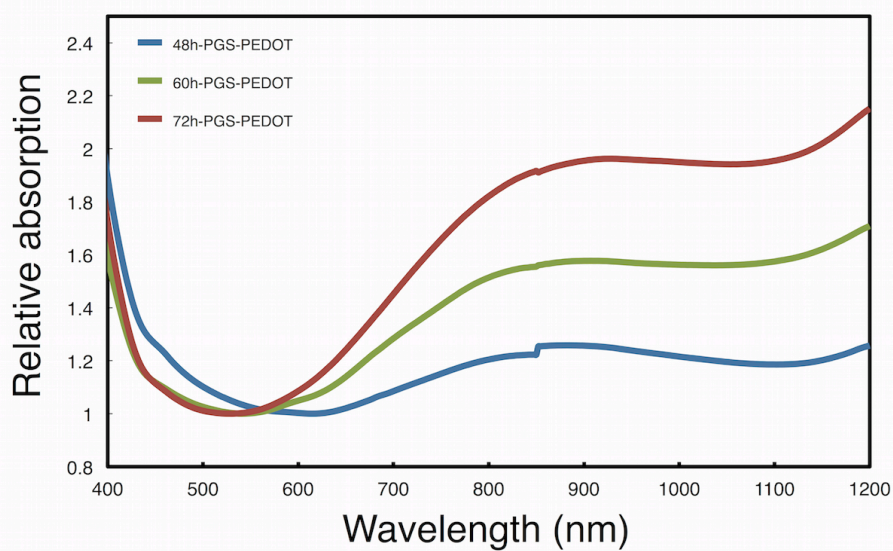
The surfaces of the samples before, after and during straining to 25% were analysed by optical microscopy. UV-Vis absorption spectra of the polymer composites was obtained using a Jasco V-670 Spectrophotometer. Samples for scanning electron microscopy (SEM) analysis were frozen in liquid nitrogen and fractured to allow for cross-section analyses of PEDOT integration into PGS. SEM and energy-dispersive X-ray analysis (EDX) were performed on gold coated samples using a JOEL 7001F Field Emission Gun Scanning Electron Microscope at 5 and 15kV, respectively.

Resistance measurements were made using a Jandel four-point probe.

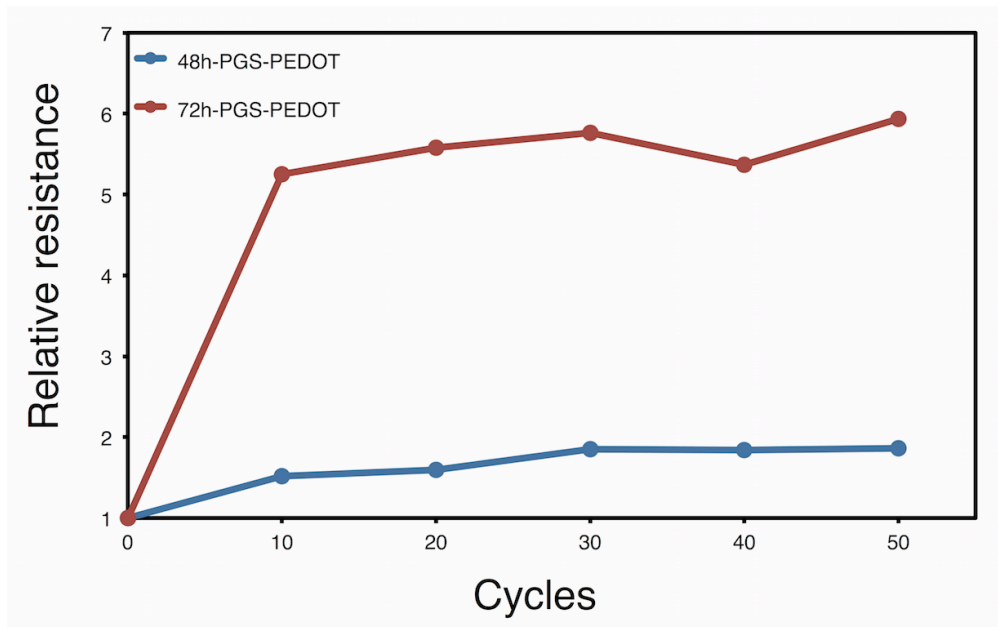
Electromechanical properties were investigated by applying a uniaxial strain with a custom-built device in conjunction with a VMP3 multichannel potentiostat.



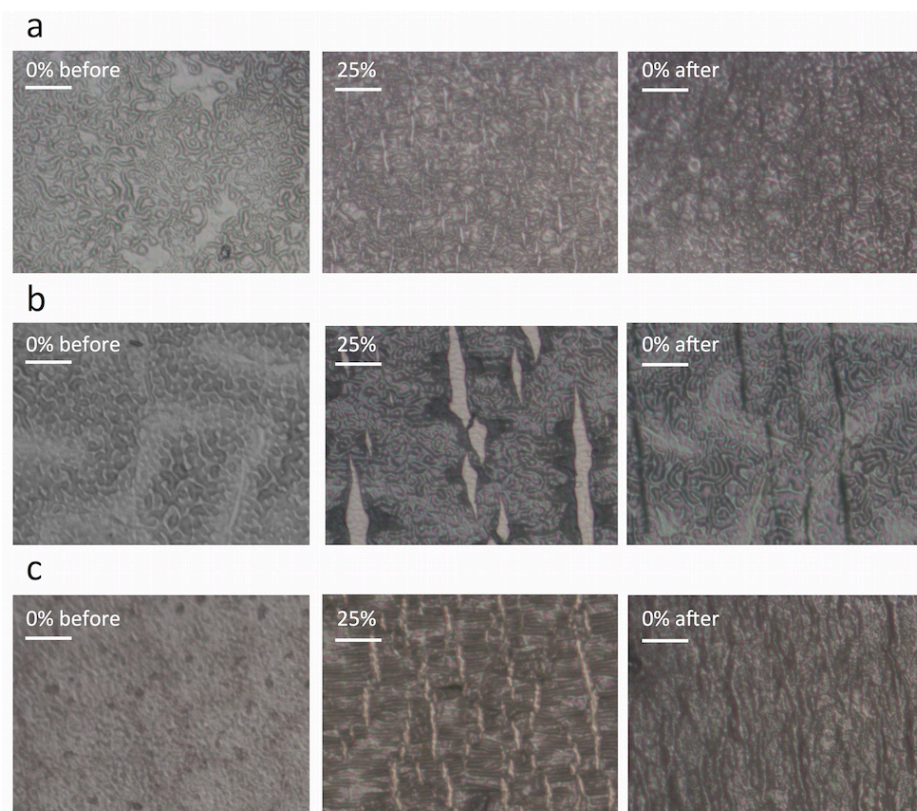
**Figure S1** Custom-built device for electromechanical investigation PGS-PEDOT composites



**Figure S2** UV-Vis absorption spectra of PGS-PEDOT hybrids with increasing PGS cure time



**Figure S3** Relative change in electrical resistance (measured at 0% strain) during 50 cycles of stretching to 25%



**Figure S4** Optical microscope images of (a) 48h-PGS-PEDOT, (b) 60h-PGS-PEDOT and (c) 72h-PGS-PEDOT before, during and after elongation to 25% original length. Scale bars represent 25  $\mu\text{m}$