Supplementary Information

Ionic conductivities of bulk prepared NBR/PEGDM were determined to estimate the ionic conductivities of the electrospun fiber mats (Table S1). The high ionic conductivities of the bulk films correlate well with the materials good electroactivities. The lower ionic conductivity of the bulk prepared films in water is also in good agreement with the poorer electroactivity in water and with the poorer average poor size variation upon actuation. The lower ionic conductivity in water is probably a leading contribution to the poorer actuation, although there are other factors that would contribute as well, such as the materials hydrophobic nature preventing good swelling and the ion sizes (Cl^-) used water compared to the $TFSI^-$ ion.

Table S1. Sample properties and preparation methods of bulk NBR/PEGMA (100/56) IPN's films.

Photo-initiator conc. (vs NBR, %)	UV curing time	Extrac-tible content (%)	Ionic cond-uctivity in LiTFSi PC solution (S/cm)	Ionic cond-uctivity in NaCl water solution (S/cm)
20	200		1.9×10^{-4}	4.4×10^{-7}
10	200		9.4×10^{-4}	4.7×10^{-7}
5	300	12	2.8×10^{-3}	1.0×10^{-4}
10*	300	4	2.3×10^{-3}	7.9×10^{-7}

* Sample was prepared with 5% BPO vs NBR and cured in an oven for 16 hrs at 80 ° C.

Various NBR and PEGDM concentration for electrospinning were investigated. An NBR concentration of 10 % was found to be optimal while excessive beading occurred at 5 % and below (figure S1). The maximum PEGDM was limited due to it being a liquid at room temperature which would result in coalesced fibers if the PEGDM concentration was too high (Figure S2 A and B)

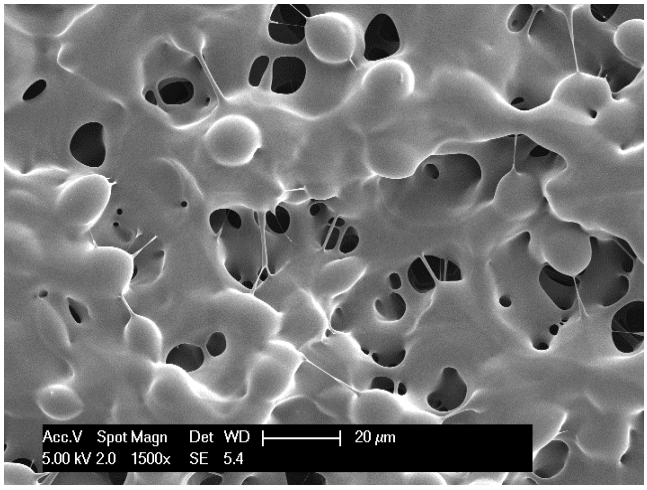

Figure S1 Electrospun NBR/PEGMA Elastomeric Fibres, eletrospun from 5 %wt/vol. NBR solutions and 56 %wt/wt (vs NBR) PEGDM with 10 %wt/wt. (vs. NBR) photoinitiator and 0 %wt/wt (vs NBR) BPO.

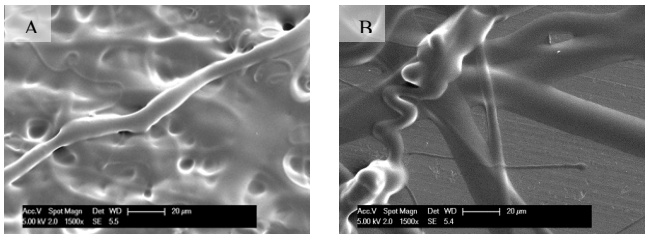

Figure S2 Electrospun NBR/PEGMA Elastomeric Fibres, eletrospun from 10 %wt/vol. NBR solutions and 150 %wt/wt (vs NBR) PEGDM with 20 %wt/wt. (vs. NBR) photoinitiator and 0 and 10 %wt/wt (vs NBR) BPO for a) and b) respectively, in chloroform (NBR:PEGM ratio; 100:150) at × 1500 magnification.

SEM EDX analysis was used to determine how PEDOT was distributed through the electrospun fibers (Figures S3, S4, S5 and S6). As expected, there was a gradient of PEDOT throughout the fibers with a crust on the exterior which rapidly lowered in concentration as it approached the center of the fibers.

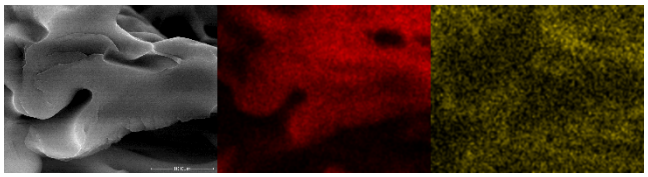

Figure S3 SEM image (left) and SEM EDX image showing elemental mapping for carbon (centre) and sulfur (right) at 8000 times magnification of sample 5A

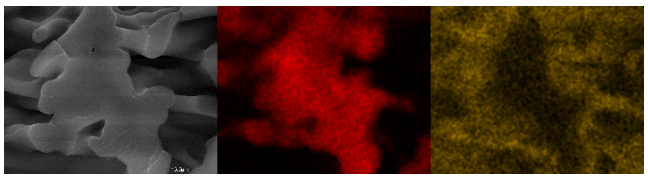

Figure S4 SEM image (left) and SEM EDX image showing elemental mapping for carbon (centre) and sulfur (right) at 8000 times magnification of sample 5C.

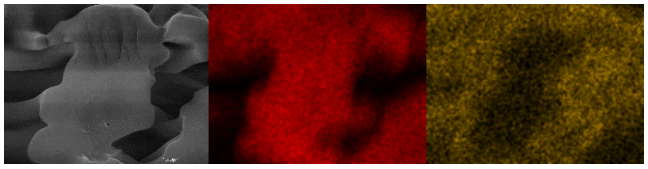

Figure S5 SEM image (left) and SEM EDX image showing elemental mapping for carbon (centre) and sulfur (right) at 8000 times magnification of sample 5D.

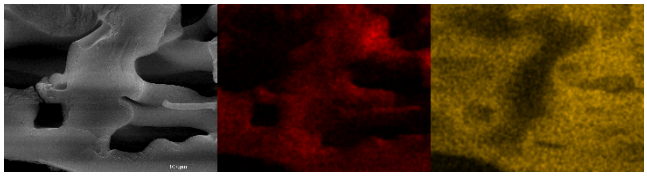

Figure S6 SEM image (left) and SEM EDX image showing elemental mapping for carbon (centre) and sulfur (right) at 8000 times magnification of with 60 min PEDOT polymerisation time.

Confocal microscopy with *in situ* square wave voltammetry was used to visualize and quantify the average pore size variation. This was done in both 1 M LiTFSI PC solution and PBS in reflectance (Figure S7) and topography mode (Figure S8). While the results in PBS were lower (5 % compared to 25 %) the average pore size variation was still significant (Figure S9).

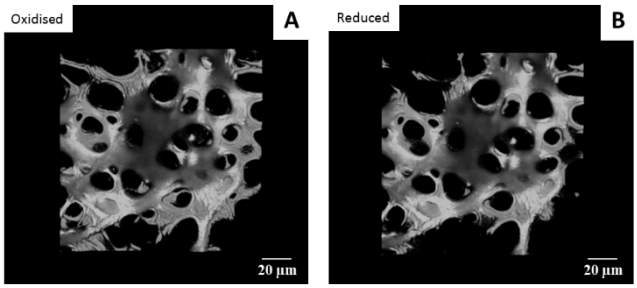

Figure S7 Images of a 3 µm slice (depth) of an electroactive, elastomeric, fibrous mats in 1 M PBS solution, in reflective mode, in an oxidized state (A) and in a reduced state (B).

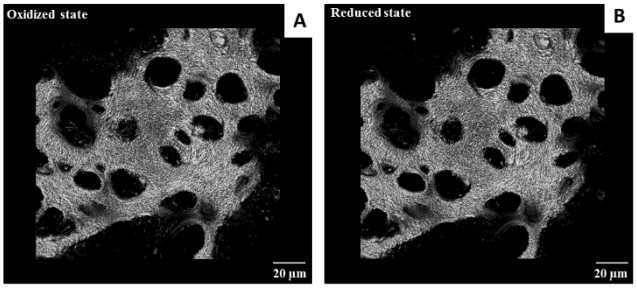

Figure S8 Images of a 3 µm slice (depth) of an electroactive, elastomeric, fibrous mats in 1 M PBS solution, in topography mode, in an oxidized state (A) and in a reduced state (B).

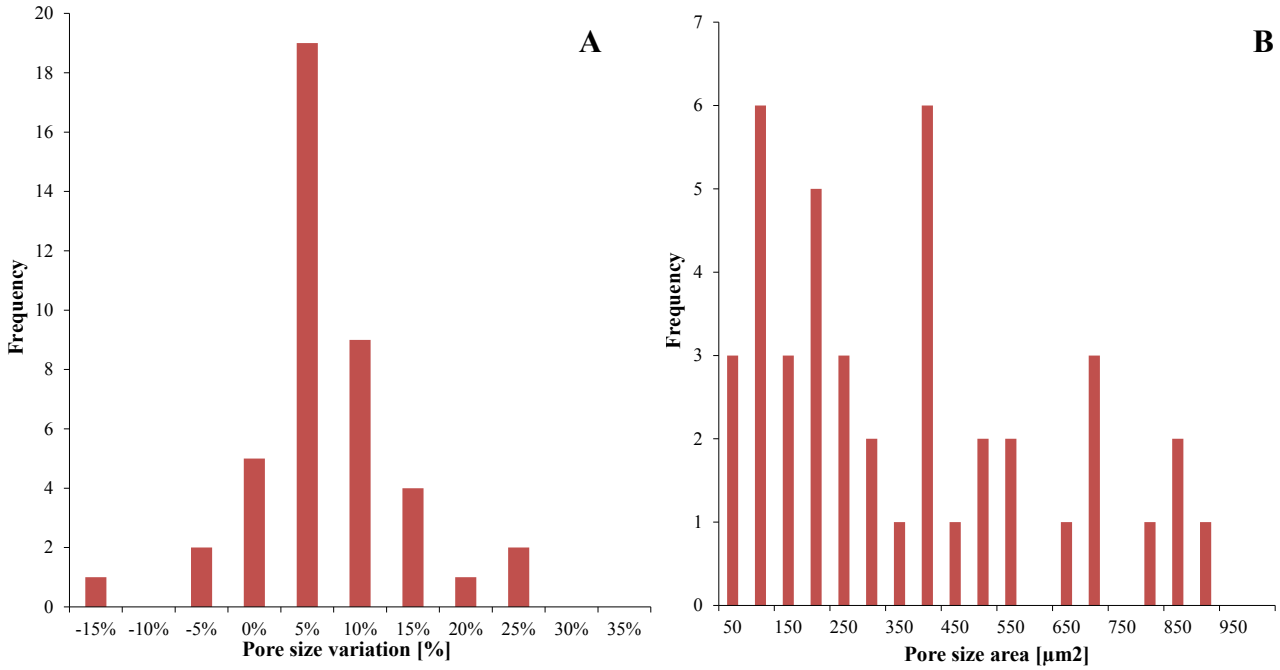

Figure S9 Graphs of pore size area distribution in the oxidized state (A) and pore size variation from oxidized to reduced state (B) obtained from the analysis of 45 pores in PBS solution

S10: The supplementary movie file shows a video of the average pore size variation was taken using confocal microscopy with *in situ* square wave voltammetry. This video shows a rapid and reversible average pore size variation of 25 % in 1 M LiTFSI PC solution.

Caption to supplementary movie: Confocal Microscope video showing pore size variation upon oxidation and reduction (+0.6 V and -0.5V) in 1 M LiTFSI PC.