

Supporting Information for

**PEDOT:PSS/Polyacrylamide Nanoweb – Highly Reliable Soft Conductors
with Swelling-Resistance**

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Resistivity-elastic modulus map

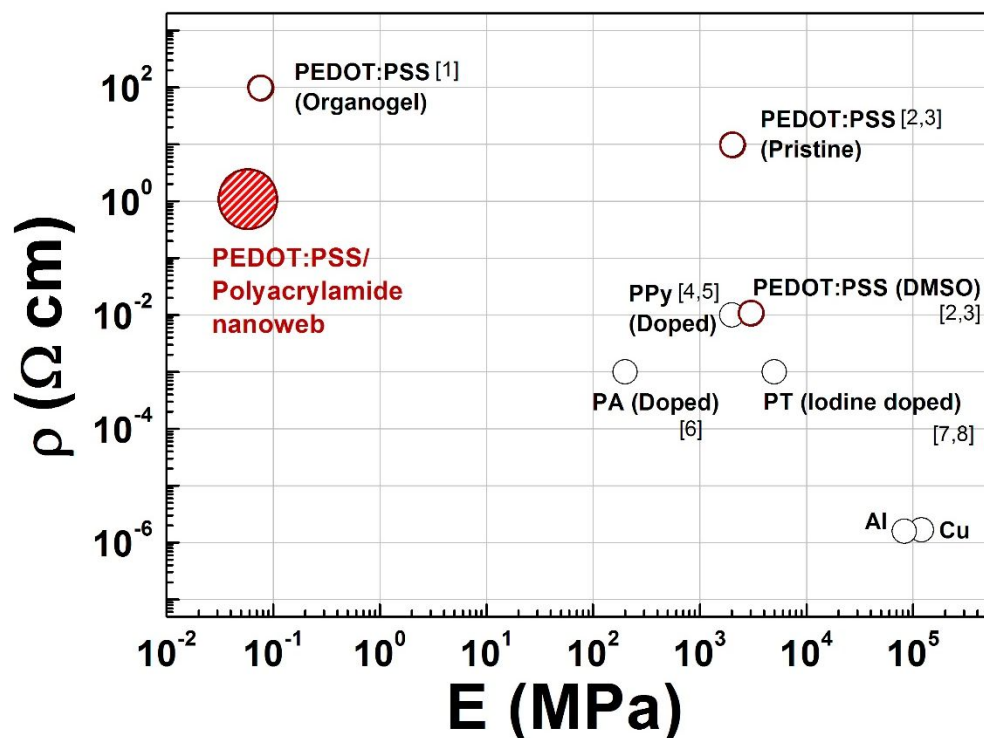


Figure S1. Resistivity-elastic modulus map. PEDOT:PSS organogel ¹, PEDOT:PSS (Pristine) ^{2, 3}, PEDOT:PSS (DMSO) ^{2, 3}, Polypyrrole (PPY) (doped) ^{4, 5}, Polyacetylene (PA) (doped) ⁶, Polythiophene (PT) (iodine doped) ^{7, 8}

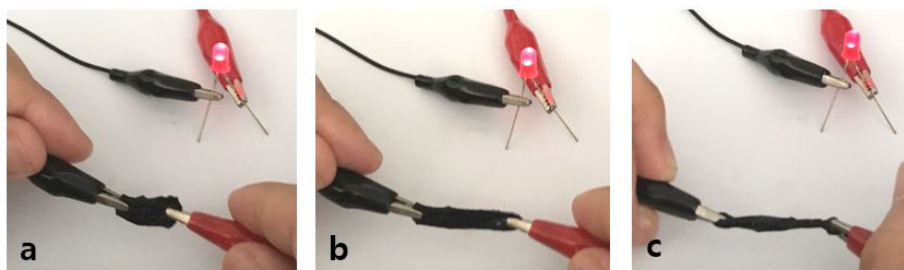


Figure S2. LED circuit with interconnects containing PEDOT:PSS/Polyacrylamide nanoweb. LED operation with the nanoweb interconnection (a) released, (b) stretched (90%), (c) twisted (720°) in the stretched state (110%).

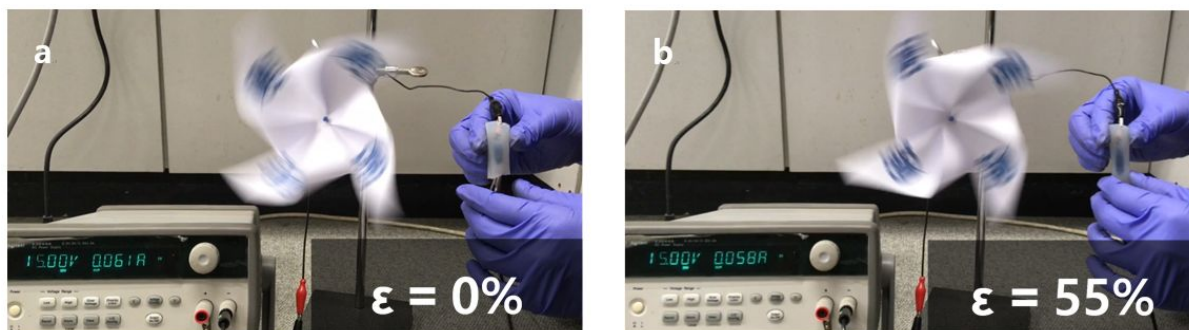


Figure S3. Propeller circuit with interconnects containing PEDOT:PSS/Polyacrylamide nanoweb. The nanoweb was passivated by ecoflex. Propeller operation with the nanoweb interconnection (a) released and (b) stretched (55%). The current reduced 5% compared to initial current.

We fabricated stretchable circuit consisting of PEDOT:PSS/Polyacrylamide nanoweb for LED and propeller. As shown in figure S2, the LED circuit was interconnected with PEDOT:PSS/Polyacrylamide nanoweb containing 20wt% PEDOT:PSS. The constant and stable light was emitted from the LED though the nanoweb composing LED circuit was stretched and twisted. Also, as shown in figure S3, propeller was connected in series with nanoweb passivated by Ecoflex. When the nanoweb was stretched with 55% strain, the current just reduced about 5% and uniform rotating rate was shown.

We confirmed the strain-insensitive current flowing into deformed nanoweb. The inherent stretchability and strain-insensitivity of nanoweb interconnection make it possible to operate electronic devices normally in deformation without rupture. It can be used as electrical interconnects to cover arbitrary curvilinear surface or three-dimensional structure in complex deformation.

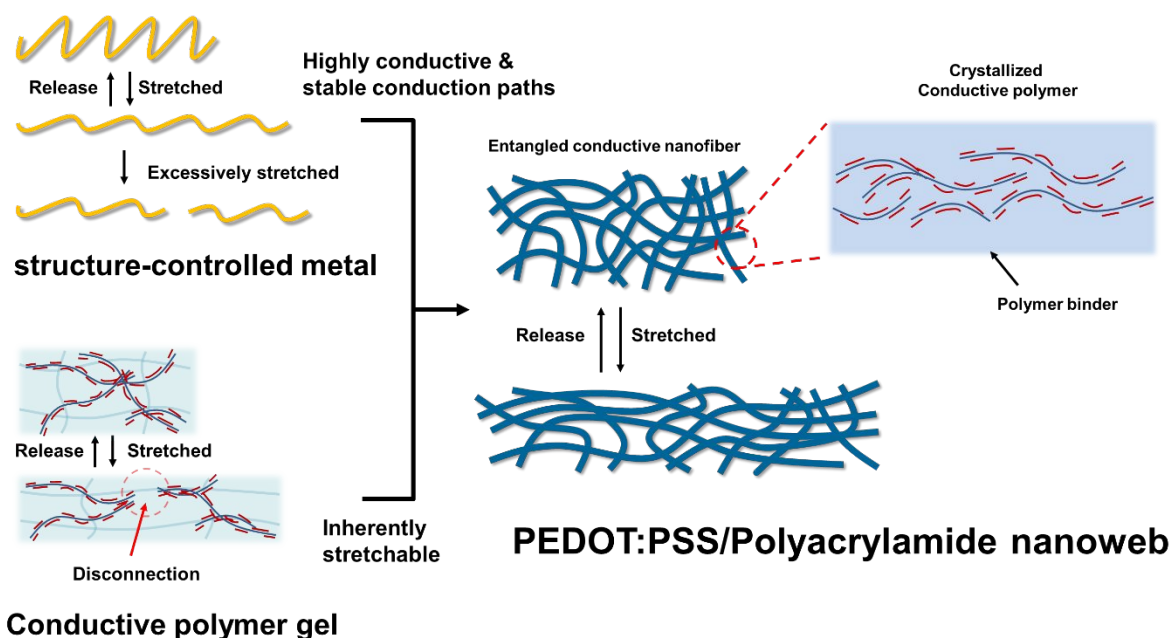


Figure S4. The concept of PEDOT:PSS/Polyacrylamide nanoweb.

The idea of PEDOT:PSS/Polyacrylamide nanoweb was inspired from two conventional stretchable conductors; structure-controlled metal and conductive polymer gel. Structure-controlled metal is the metallic stretchable conductor which obtains stretchability by designing structures such as wavy, buckled shape, or nanowire. It can be stretched due to structural alignment, not inherent deformation, which keeps the metallic conduction paths stable and highly conductive without rupture. However, there are limits of stretchable range by fully aligned. Over the range, the metallic conduction paths are inherently stretched and can be damaged.

Conductive polymer gel is the inherently stretchable conductor composed of gel as a container and conductive polymer as a conducting part. Gel is a cross-linked polymer network containing solvent, which exhibits no flow. It is intrinsically stretchable and soft. Conductive polymer is the conjugated polymer that has electrical conductivity, typically due to unpaired π electron. By inserting conductive polymer inside gel, electrically conductive and inherently stretchable material

can be fabricated. However, there are mechanical mismatch between conductive polymer and gel, which makes conduction paths of conductive polymer unstable.

From these, we devised the new stretchable conductor that has inherent stretchability and stable conduction paths by designing the materials composing conductive polymer gel. As transforming the microstructure of conductive polymer gel into nanofiber, the conduction paths can keep connection stable inside nanofiber, which improves electrical stability and conductivity. Also, it has inherent stretchability due to soft components, which make it possible to be deformed more than structural alignment.

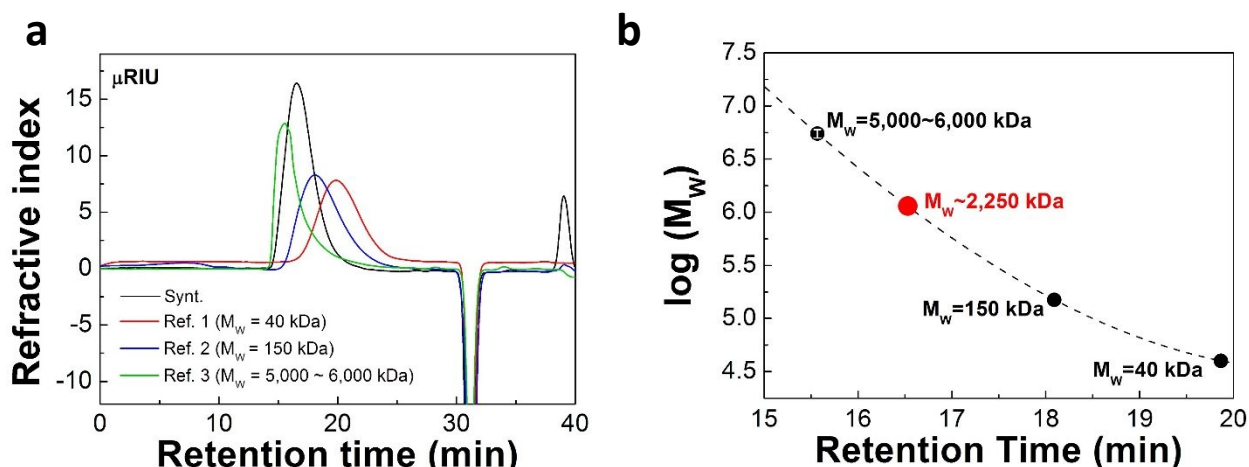


Figure S5. (a) Gel permeation chromatography profile for synthesized polyacrylamide and references. (b) Calibration of GPC profile. The weight average molecular weight of synthesized one is estimated around 2,250 kDa.

Gel permeation chromatography (GPC) is a kind of size exclusion chromatography (SEC), which separated analytes according to size that is proportional to molecular weight. To confirm the molecular weight of synthesized one, the polyacrylamides known molecular weight, 40 kDa, 150 kDa, and 5,000 ~ 6,000 kDa, were also analyzed as references. Unfortunately, it was hard to get uniform polymer in high molecular weight, especially over 5,000 kDa. Therefore, the weight

average molecular weight of synthesized polyacrylamide was approximately calculated. Normally, Mark–Houwink equation, $[\eta]=KM^a$, is used to calibrate GPC data. $[\eta]$ is intrinsic viscosity, M is molecular weight, K and a are Mark–Houwink parameters. Figure S5 shows the calibration curve of GPC and it was approximately 2,250 kDa and polydispersity index is 9.37.

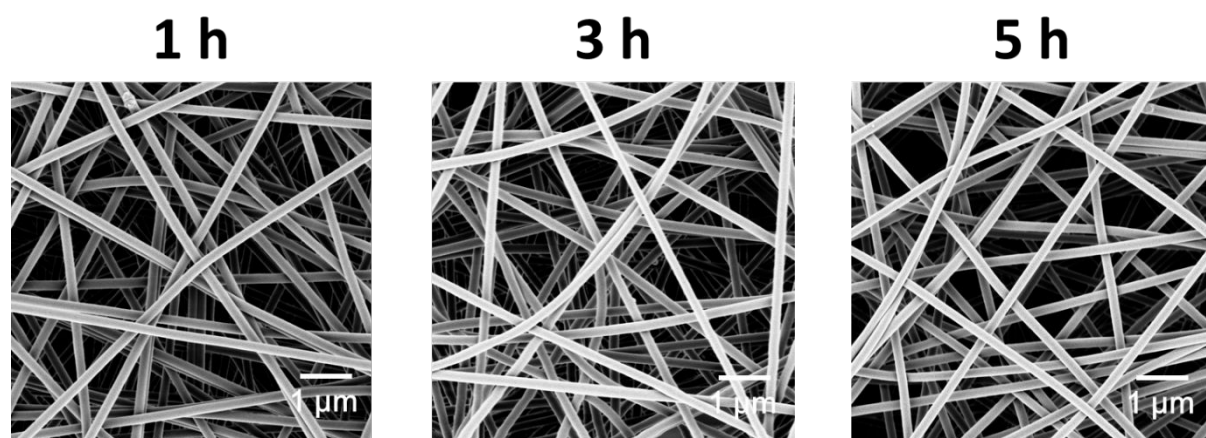


Figure S6. The microstructure of polyacrylamide according to thermal treated time, 1h, 3h, and 5h. All samples were coated by Pt and the images obtained by FESEM.

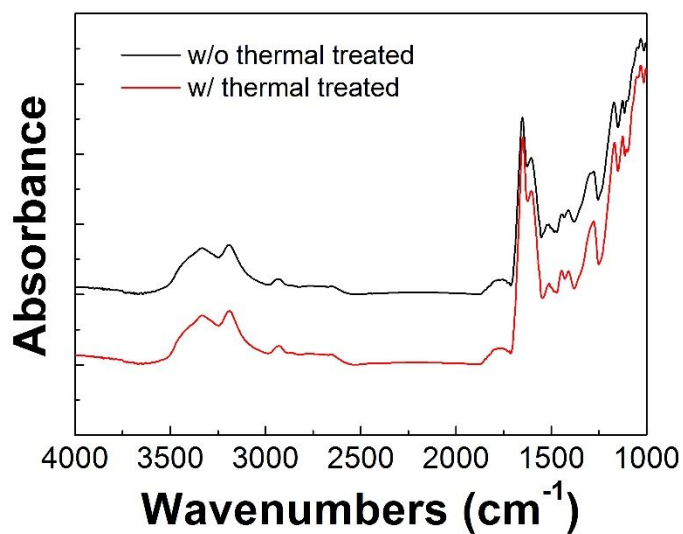


Figure S7. FT-IR analysis of PEDOT:PSS/Polyacrylamide nanofiber before and after heat treatment. After the heat treatment, no new peak was observed and only the peak near 1650 cm^{-1} indicating the C = O bond of acrylamide was elevated.

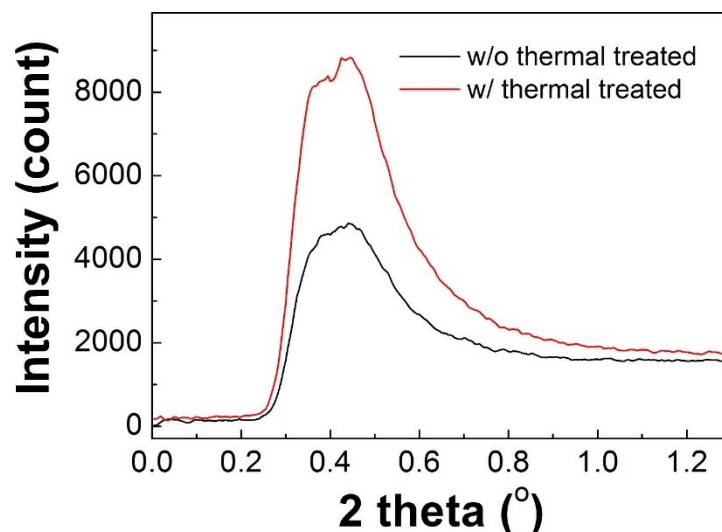


Figure S8. The SAXS profile of polyacrylamide nanofibers before and after thermal treatment. The x-ray beam was more scattered due to thermal-treated polyacrylamide nanofibers than not thermal-treated, which proves that thermal-treatment made nanofibers densified.

Small-angle X-ray scattering (SAXS) is an analysis method quantifying density differences of samples which have anisotropy in the nanometer range. It measures the scattering of X-rays by samples at very low angles, typically $0.1\sim 10^\circ$. In this small angular range, the scattering signals have the distances of partially ordered materials, the size and shape of macromolecules, and so on. In previous researches, it was reported that the more dense and crystallized polymers were, the larger x-ray scattering signals were.⁹ As shown in figure S6, the microstructure of polyacrylamide nanofibers were almost not changed following thermal treatment. The diameter of polyacrylamide nanofibers were kept around 200 nm. Therefore, it is considered that the stabilization after heat treatment is due to the change of the molecular structure, not the external change of the nanofiber.

Figure S7 shows the FT-IR analysis results. FT-IR is a typical analytical tool that can predict the molecular structure by confirming the molecular bond type by infrared spectroscopy. As a result of the analysis, no new peak was observed after heat treatment, so it is presumed that new molecular bonds did not appear. Also, a peak at 1650 cm^{-1} was observed to increase. This peak is predicted as the peak associated with $\text{C}=\text{O}$ of the acrylamide. It is presumed that the reason for the increase of the peak is due to the dense spacing between the polyacrylamides.

In figure S8, SAXS data of unheated and heated was shown. The scattering intensity of heated polyacrylamide nanofiber was larger than unheated, which means heated one is more densified. Therefore, it is confirmed that the stability of the nanoweb is improved due to the denser polyacrylamide nanofibers after heat treatment.

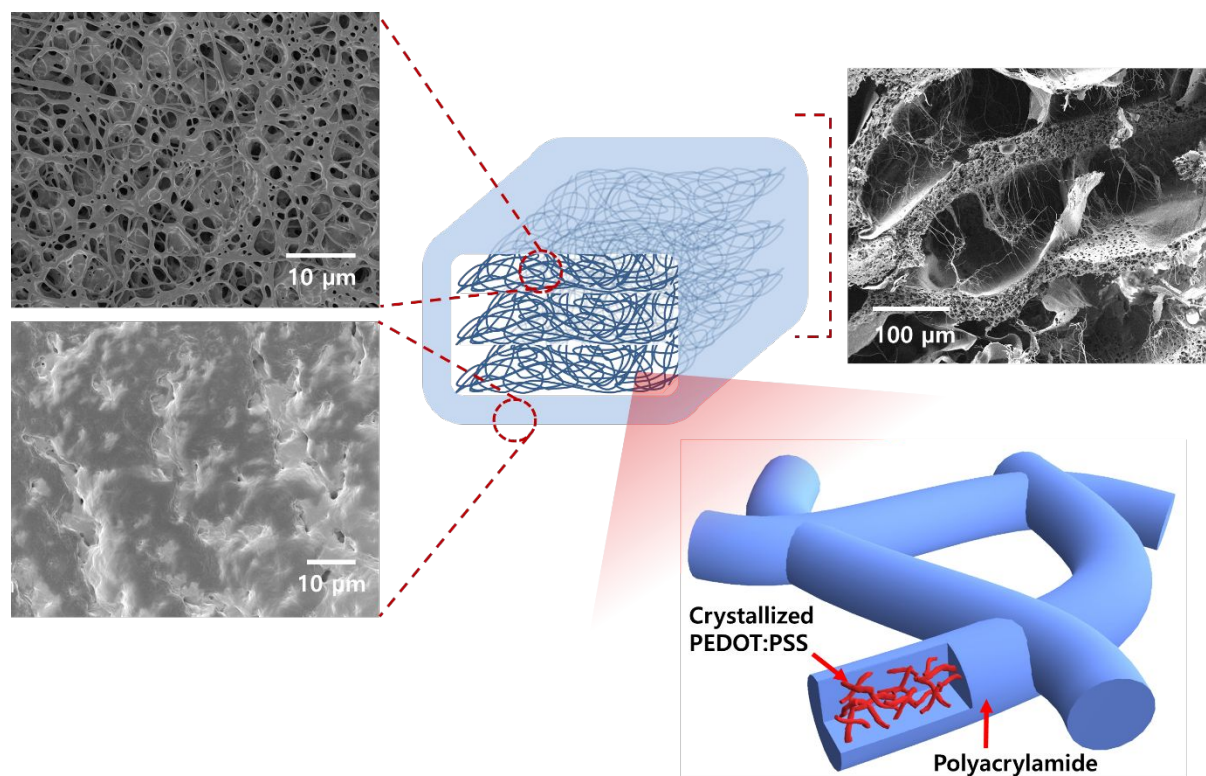


Figure S9. The entire morphology of PEDOT:PSS/Polyacrylamide nanoweb.

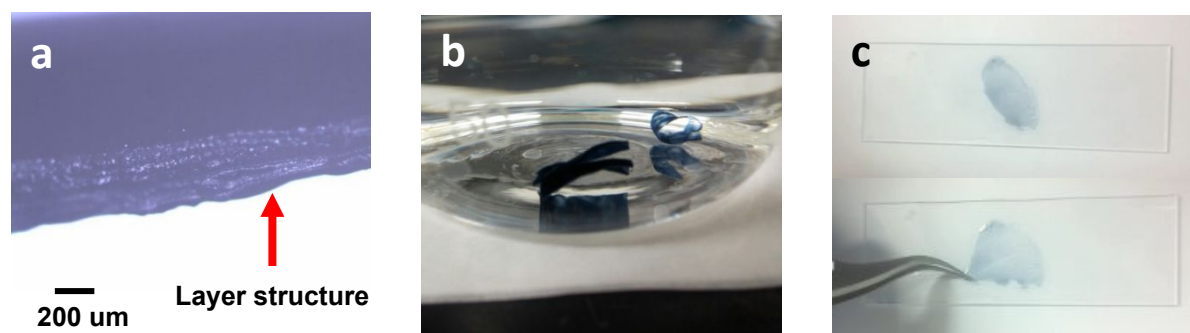


Figure S10. The Layer structure of PEDOT:PSS/Polyacrylamide nanoweb. (a) Optical image of layer structure. (b) Separation of layers by removing passivation (c) One layer extracted from nanoweb.

Figure S9 shows the entire morphology of PEDOT:PSS/Polyacrylamide nanoweb. The PEDOT:PSS/Polyacrylamide nanoweb is passivated by surface membranes formed from slightly agglomerated nanofibers. Also, inside the nanoweb, it has a layer structure because PEDOT:PSS/Polyacrylamide nanofibers were fabricated by electrospinning. The nanofibers were

piled up in layers during electrospinning and the layer structure remained even after solvents treatment due to densification of polyacrylamide by thermal treatment. The average thickness of layers is about 40 μm and each layer is also passivated by thin membranes. The web-like structure formed by tangled and adhered nanofibers was kept inside each layer.

As shown in figure S10, the PEDOT:PSS/polyacrylamide nanoweb layers was also observed by optical microscopy (fig. S10 a). Each layer can be separated after removing passivation. (fig. S10, c) One layer is so thin that it is optically transparent, but it still maintains inherent stretchability. Therefore, it can be used as a transparent conductor with inherent softness.

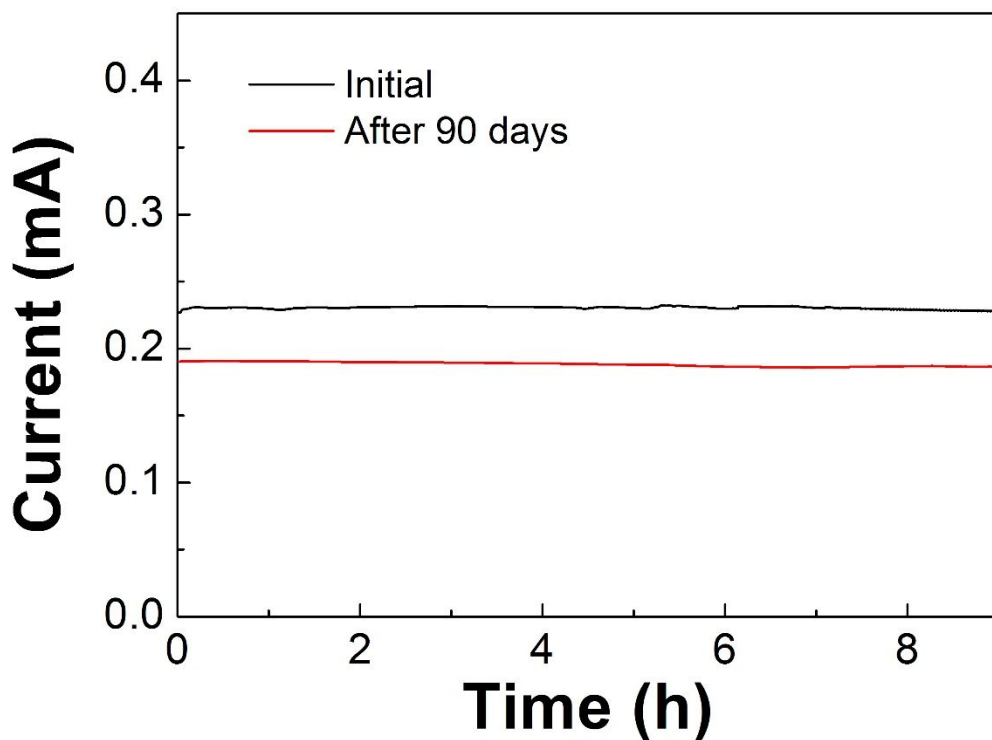


Figure S11. Electrical stability test before and after 90 days in deionized water. Applied voltage was 0.1V and the sample size is 10(w) \times 30(l) \times 0.5(t) mm. Current reduced just 17% compared with initial current.

Figure S11 shows the change of current with time before and after 90 days in water. As shown in the graph, during the initial 9 hours, the current was almost unchanged. In addition, it was confirmed that current noise is hardly generated even in underwater environment, and thus the stability in water is also excellent. As a result of measuring the current after 90 days in the water, the electric current is reduced by about 17% compared to the initial electric current, so that the electric property is not greatly lost for a long time and has excellent water stability. This can be a very important feature in water environment systems such as living bodies.

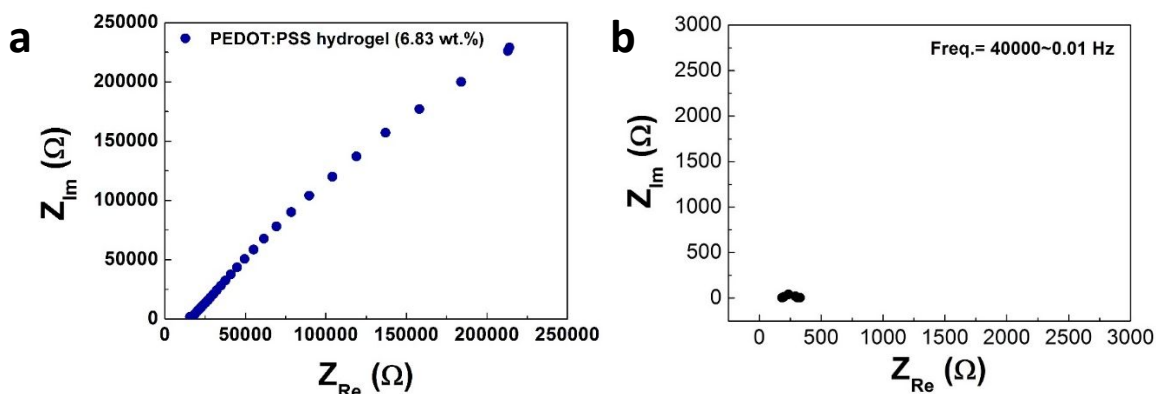


Figure S12. Impedance curves of (a) PEDOT:PSS hydrogel and (b) PEDOT:PSS/Polyacrylamide nanoweb (20wt%) with a frequency range from 40,000 to 0.1 Hz at 10 mV.

We conducted impedance measurement of the PEDOT:PSS hydrogel and nanoweb (20wt%). The frequency range was from 40 kHz to 0.1 Hz with 10 mV amplitude at 0 V offset (ZIVE MP2A, Wonatech). The impedance curves are shown above Figure S12, which are measured two different samples. For the PEDOT:PSS hydrogel, it shows large increase of the capacitive reactance when the frequency decreased, which means a capacitive behavior of the hydrogel is observed by the migration of ions. Similar behaviors are also observed for the other hydrogel based conductors.^{10,11} However, for the PEDOT:PSS nanoweb, the resistive impedance is not much changed with a large variation of the frequency, and moreover, the capacitive reactance shows very negligible change at even lower frequency. It is typical impedance behavior of electronic conductors. Therefore, the

conduction of the PEDOT:PSS nanoweb dominantly occurred by electronic migration of PEDOT:PSS polymeric conduction path.

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