## Supercapactive polymer hydrogel electrode via pure PEDOT:PSS hydrogel printed on laserinduced graphene for DC stimulation applications

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Bioelectronic interfaces rely on intimate contact with tissue to establish efficient signal exchange between electrodes and the biological target structure. Mechanically soft and ion conducting materials are beneficial in this context, in particular for semi-dry interfaces such as electrodes contacting skin. Typical natural (e.g. alginate, collagen, agar) and synthetic (e.g. polyacrylamide, polyethylene glycol, polyvinyl alcohol) hydrogels used for biomedical implementation are not inherently electronically and/or ionically conductive, wherefore development of conducting hydrogel systems is highly relevant. We demonstrate here how a supercapacitive polymer hydrogel whose network is entirely comprised of conducting polymer poly(3,4ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS) nanofibril interconnections can be combined with laser-induced graphene (LIG) on a flexible substrate (i.e. polyimide). This results in a bioelectronic interface which easily conform and connect to skin and can support both recording and electrical stimulation. The PEDOT:PSS hydrogel precursor can be printed (i.e. direct ink writing) or dip-coated onto laser pyrolyzed polyimide (e.g. Kapton) film. The LIG and PEDOT:PSS hydrogel electrode was shown to have a high charge storage capacity, electrochemical stability (retains more than 85% of the original charge storage capacity over 500 cycles) and ability to deliver long term biologically-relevant electric fields (10-1000 mV/mm). In short, via affordable technology, we were able to make devices that outperform previous technology based on noble metals. Thereby we pave the way for disposable bioelectronics as well as DC stimulation of cells and tissues at levels relevant for regenerative therapy.



**Fig. 1)** Flexible laser-induced graphene electrodes with improved charge storage capacity (CSC) and steady electrochemical stability via pure PEDOT:PSS hydrogel functionalization. (a) Process of how polyimide (i.e. Kapton) film are transformed to supercapacitive hydrogel electrodes. (b) Cyclic voltammetry for the titration of PEDOT:PSS hydrogel precursor spot-deposition shows that the conducting polymer hydrogel is better than both bare LIG and LIG with electropolymerized PEDOT/PSS, in terms of CSC. (c) Electrochemical stressing via 500 cycles of CV shows that LIG/PEDOT:PSS hydrogel loses less than 15% of it's original CSC