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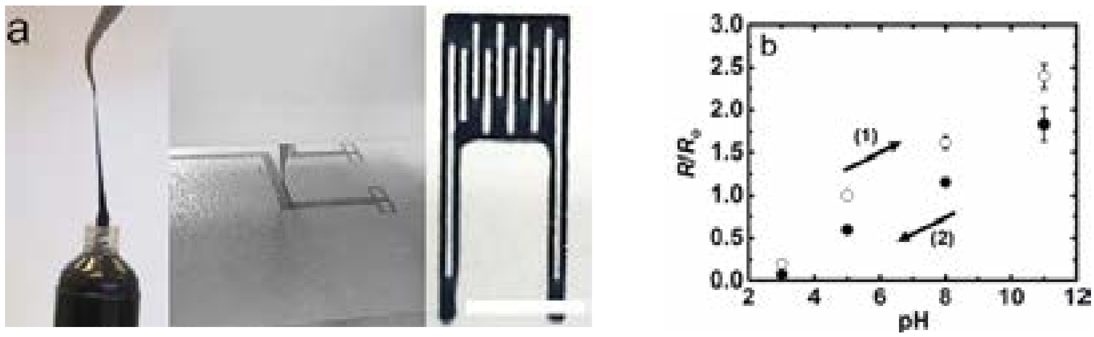
Printed, flexible pH sensor hydrogels for biomedical applications

Introduction: Current sensors for monitoring environmental signals, such as pH, are often made from rigid materials that are incompatible with soft biological tissues. The high stiffness of such materials sets practical limitations on the in situ utilisation of sensors under biological conditions. To this end, various innovative concepts have been introduced to develop more flexible sensors for wearable devices, textiles, bionics, soft electronics, and soft robotic systems. Yet, the majority of these methods are cumbersome, inconvenient, and often expensive. In this study, we demonstrated an elegant approach for rapid manufacturing of highly flexible pH-sensitive hydrogels based on poly(3,4-ethylenedioxy thiophene) (PEDOT) doped with negatively charged poly(styrenesulfonate) (PSS) and robust hydrophilic polyurethanes (HPU). PEDOT that is p-doped with PSS is a hole conductor material. Given the complex nature of PEDOT: PSS, its electronic characteristics are sensitive to pH. By optimising the composition of the PEDOT: PSS/HPU inks, we were able to utilise 3D printing for the fabrication of pH sensitive PEDOT: PSS/HPU hydrogel sensors in one step (Figure 1). Materials and Methods: The HPU used in this study was based on poly(ethylene oxide) soft chains (PEO) and cyclohexylisocyanate hard segments. Depending on the length of PEO blocks, the HPU macromolecules are soluble in certain concentrations of water and ethanol (EtOH) mixture but remain insoluble in pure water or EtOH. For ink formulation, first a 5w/v% HPU solution was made in a 95:5 EtOH:water solvent. To this solution, various amounts of the 1.1 wt% PEDOT: PSS aqueous dispersion (Sigma Aldrich) was added. The PEDOT: PSS/HPU conductive inks were then 3D printed using the EnvisionTEC 3D-Bioplotter® (Figure 1a). Results and Discussion: A linear and reversible correlation between the conductivity of fully swollen PEDOT: PSS/HPU hydrogel sensors and the pH was observed, which is attributed to the pH-sensitivity of the PEDOT:PSS element (Figure 1b). The electrical resistance of hydrogel sensors was not affected by extreme bending and twisting, and consecutive tensile cycling – up to 400 cycles of 50% extension. The 3D printed, hydrogel-based pH sensors remained functional after two months of storage in Milli-Q water. The inherently low modulus of hydrogels developed in this work (~103 kPa) is 102 to 104 times lower than most materials traditionally used in sensor technology. The combination of low modulus and the sub-millimetre thickness of hydrogel sensors have enabled us to create pH sensors with flexural stiffness at the order of 1 µN m. As a comparison, human skin has a flexural stiffness of ~102 µN m. Conclusions: Optimised formulations of PEDOT: PSS/HPU inks were developed to fabricate highly flexible pH sensors that linearly respond to pH in wet environments. Such printable inks that can produce biosensors using a one-step fabrication method eliminate the cumbersome and inconvenient processing of rigid materials that are used in the manufacturing of conventional pH sensors.



Biography

Fariba Dehghani leads a multidisciplinary bioengineering research team comprising engineers, scientists, clinicians and molecular biologists focused on developing technologies for nutritional food products and biomaterials, with particular emphasis on tissue engineering and regenerative medicine. She is one of the members of ARC College of Expert, the director of Australian Research Council for Australian Food Processing Industry in the 21st Century, as well as director of Centre of Excellence in Advanced Food Ergonomics in the Faculty of Engineering and Information Technologies at the University of Sydney.

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