Bioinspired Hydrogels for Transfer Printing of Ultracompliant Medical Electronics

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Statement of Purpose: Flexible electronics have the potential to serve as compliant biosensors for applications such as brain-machine interfaces. However, elastomers such as polydimethylsiloxane ($E \sim 1$ MPa) are still much stiffer compared to many exitable tissues found within the human body such as muscle and brain (E \sim 1 kPa). The mechanical mismatch could contribute to the cell and tissue damage after implantation, which can comprimise the integrtiy and reduce device performance. The resulting cellular reaction and inflammatory response could prevent the long-time, stable operations of the electronic implants. Hydrogels are structural and mechanical similar to living human tissues and now widely adopted in many biomedical applications. Hydrogel-based electronic devices will afford additional advantage over elastomer-based devices for biomedical applications by enabling the idea of electronics that are either compressible or injectable. The large volumetric change upon dehydrating the hydrogel permits injecting or even swallowing the dried hydrogel electronics without the need of surgeries to implant the devices. The hydrogel electronics will self-deploy in the aqueous environments of human tissues and become functional. Therefore, hydrogels are promising to be utilized as substrates for next-generation biomedical electronic devices. Some attempts have been made to form conductive patterns on ultra-compliant hvdrogel substrates via electropolymerization. Here we propose a different method to pattern electronic devices onto hydrogel substrates, which involves transfer printing pre-fabricated electronically active microstructures onto the adhesion-promoted hydrogel surfaces. Such fabrication strategy has the potential to achieve hydrogel-based ultra-compliant functional electronic devices while fully utilizing through the well-developed thin film patterning technology.

Methods: Catechol-bearing dopamine methacrylamide (DMA) was be used to prepare water-stable adhesive copolymers. In this study, 2-hydroxyethyl methacrylate (HEMA) and DMA monomers were copolymerized through ultraviolent curing to prepare the catecholbearing poly(HEMA-co-DMA) hydrogels, which are similar to commercial soft contact lens. Gold microelectrode arrays were pre-fabricated on a silicon dioxide donor substrate surface through standard photolithographic process. The fully swollen poly(HEMA-co-DMA) hydrogels were conformably laminated onto the donor substrate surfaces and slowly delaminated after 5 min to transfer the metallic microstructures from the donor substrates onto the poly(HEMA-co-DMA) hydrogel surfaces.

Results: Conductive gold microstructures with square shapes (feature size 200 μ m × 200 μ m, spacing 50 μ m) fabricated on silicon dioxide substrates by photolithography (**Figure 1a**) can be transferred onto the catechol-bearing swollen poly(HEMA-co-DMA) hydrogel surfaces (Figure 1b). The metallic surfaces

clearly become wrinkled after being transferred but remain cracking-free (Figure 1c). The wrinkling could result from the surface roughness of the hydrogel and also the deformation of the hydrogel during the kinetically peeling back process. Wrinkling has been demonstrated as an effective way to increase the stretchability of brittle thin film material. Figure 1d shows the current-voltage measurements performed on multiple microelectrodes in a two-probe manner. The I-V response is linear, which indicates good electrical conductivity of the metallic microelectrodes patterned onto the hydrogel surfaces. The resistance for the microelectrodes on hydrogel substrates is 19.0 \pm 6.7 Ω , which is higher compared to the initial resistance 9.9 \pm 0.9 Ω exhibited on silicon dioxide substrates, which could be caused by the inbuilt strain in the wrinkled surfaces.



Figure 1. Optical images of lithographically fabricated gold metallic microstructure arrays (a) pre-patterned on silicon dioxide wafers and (b-c) transfer printed onto catechol-bearing adhesive poly(HEMA-co-DMA) hydrogel surfaces. (d) Current-voltage measurements indicate good conductivity of the gold microelectrodes transferred on hydrogel surfaces.

Conclusions: The current study demonstrates that conductive metallic microstructures can be patterned onto catechol-bearing adhesive hydrogel substrates via transfer printing. This fabrication strategy can produce electronic microstructures that are integrated with ultracompliant hydrogel networks. Future work will focus on (1) utilizing the microelectrode patterned hydrogels as an ultracompliant, fully hydrated platform to study the in vitro cell cultivation under electrical stimulation; (2) testing the electrical performance of metallic microelectrodes patterned on hydrogel through surfaces hydration/dehydration cycles.

References: (Kim DH. Science. 2011; 333: 838-843.); (Hassler C. J. Polym. Sci., Part B: Polym. Phys. 2011; 49: 18-33.); (Sekine S. J. Am. Chem. Soc. 2010; 132: 13174-13175.); (Glass P. Langmuir. 2009; 25: 6607-6612.);