Photomechanical gel: Fabrication of photothermal bi-layered actuators by adhesion of PEDOT/PSS and thermoresponsive gels

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Introduction: Poly(*N*-isopropylacrylamide) (PNIPAAm) hydrogels are well-known thermoresponsive materials which show a volume phase transition upon being heated at a temperature corresponding to the lower critical solution temperature (LCST). The mechanical bending of PNIPAAm hydrogels in response to external stimuli is important for artificial muscles and switching devices.^[1,2,3] Recently, we prepared bi-layered soft actuators by adhesion of PNIPAAm hydrogels.^[4,5] In this study, we prepared photothermal actuators by adhesion of PNIPAAm hydrogels and poly(3.4-ethylenedioxythiophene)/poly(sodium 4-styrenesulfonate) (PEDOT/PSS) films, which bent in an arc by laser irradiation (**Fig. 1**).

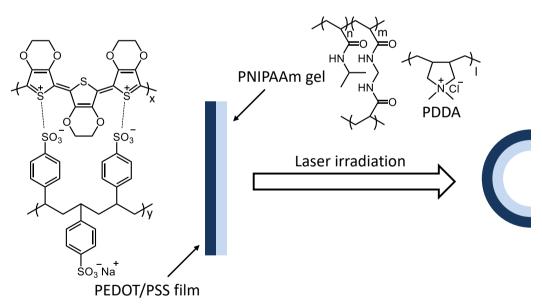


Figure 1. Schematic illustration and a chemical structure of photothermal bi-layered actuators, which bend in an arc in response to laser irradiation.

Experimental: Bi-layered actuators were prepared by adhesion of poly(3.4ethylenedioxythiophene)/poly(sodium 4-styrenesulfonate) (PEDOT/PSS) films and cationic polymers incorporated PNIPAAm hydrogels. The PNIPAAm hydrogels were prepared by photopolymerization using NIPAAm as a monomer, N,N'-methylenebis(acrylamide) (MBAAm) as a crosslinker, irgacure 2959 as a photo-initiator, and poly(diallyldimethylammonium chloride) (PDDA) as a cationic polymer. Thus, two layers were adhered by the electrophoretic adhesion.^[4,5] **Results and Discussion:** It is well-known that PEDOT/PSS shows high conductivity and sulfonate ion of PSS is dopant. Because of an excess amount of PSS, the PEDOT/PSS films are negatively charged. Therefore, anionic PEDOT/PSS films are able to adhere on cationic hydrogels. Adhesion of cationic PNIPAAm hydrogels and anionic PEDOT/PSS films was carried out *via* electrophoresis. During electrophoresis, cationic and anionic polymers move to a cathode and an anode, respectively, and then form a polyion complex at the contact interface between the two materials for adhesion.^[4,5] In this study, we prepared bi-layered actuators consisting of PNIPAAm and PEDOT/PSS by electrophoretic adhesion (**Fig. 1**).

When CW-808 nm laser (9.0 W cm⁻²) was irradiated to the bi-layer, it bent toward the PNIPAAm layer (**Fig. 2a, b**). PEDOT/PSS releases thermal energy from photoexcitation of materials, so-called photothermal effect, by laser irradiation. As shown in **Fig. 2c-f**, temperature of bi-layered actuators changed from 19.5°C to 32.2°C by laser irradiation. Because LCST of PNIPAAm hydrogels is ca. 32°C, the PNIPAAm hydrogels shrunk by laser irradiation, and then bi-layered actuators changed their shape (**Fig. 2a, b**). After turning off laser irradiation, the shape of bi-layered actuators went back to the original shape in water (**Fig. 2a**).

In conclusions, laser-driven hydrogel actuators were prepared by adhesion of thermoresponsive hydrogels and conductive polymer films. We believe our novel, photothermal bi-layered actuators will further advance the development of functional gels for intelligent soft-actuator systems.

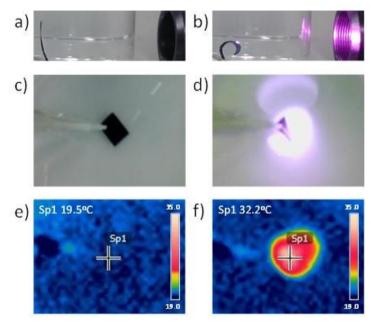


Figure 2. (a, b) Bending motion of photothermal bi-layered actuators prepared by adhesion of PEDOT/PSS and PNIPAAm hydrogels. Optical (c, d) and thermal (e, f) images of adhered gels. (a, c, e) and (b, d, f) indicates without and with laser irradiation, respectively.

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References:

[1] Z. Hu et al., Science, 1995, 269, 525.

[2] T. Asoh et al., Adv. Mater., 2008, 20, 2080.

[3] T. Asoh et al., Chem. Commun., 2009, 3548.

[4] T. Asoh et al., Chem. Commun., 2010, 46, 7793.

[5] T. Asoh, *Polym. J.*, **2016**, *48*, 1095.