Supplementary Information

Fabrication of Tunable Photothermal Actuator via In-Situ Oxidative Polymerization of Polydopamine Nanoparticles in Hydrogel Bilayers

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[#]Current Address: KU-KIST Graduate School of Converging Science and Technology, Korea University, 145 Anam-ro, Seongbuk-gu, Seoul 02841, Republic of Korea S1. Comparison of thermal stability of agarose and agarose/alginate double network hydrogels.



Figure S1. Photograph of agarose and agarose/alginate double-network hydrogel in an aqueous medium. Both gels were immersed in 90 °C water for 5 min. For visualization, plasmonic gold nanoparticles were introduced in the hydrogel matrix.

S2. Scanning electron microscope images of polydopamine nanoparticles (PDA NPs).



Figure S2. Scanning electron microscope images of PDA NPs synthesized through (a) batch polymerization and (b) in-situ polymerization in agarose/alginate double-network hydrogel matrix. Scale bars indicate 1 μm.

S3. Comparison for in-situ oxidative polymerization of PDA NPs in agarose gel and agarose/alginate double network gel (DN gel).



Figure S3. (a) Photographs agarose gel (left) and agarose/alginate DN gel (right) before (top) and after (bottom) PDA NP incorporation under the same polymerization condition. (b) Transmittance of PDA NP incorporated agarose gel (red) and agarose/alginate DN gel (green).



S4. Absorbance of PDA NP dispersion with varying PDA NP concentration.

Figure S4. Absorption spectra of PDA NP dispersion with varying PDA NP concentration (C_{PDA NP}).



S5. Delamination of bilayer actuator due to poor adhesion between layers.

Figure S5. (a) Schematic for alternative fabrication process of PDA NP-incorporated bilayer actuator. PDA NPs were introduced prior to photopolymerization of PNIPAm layer. (b) Photograph showing delamination of PNIPAm layer from agarose/alginate layer due to weak bonding between bilayers.



S6. Transmittance of PDA NP incorporated hydrogel bilayer panels.

Figure S6. Photograph (inset) and transmittance spectra of four bilayer panels of pinwheel-shaped actuator.

S7. Micropatterning of DN gels by controlling the polymerization cycles of PDA NPs. For local patterning of PDA NPs in agarose/alginate DN gels, a shadow mask made of poly(dimethylsiloxane) was introduced. Briefly, the entire agarose/alginate DN gel was immersed in a 10 wt% dopamine hydrochloride solution for an hour. Subsequently, a shadow mask having four square holes was put on the top of agarose/alginate DN gel. Then, 10 mM NaOH solution was selectively dropped into the square hole to proceed with the reaction for an hour. After repeating the polymerization process described above 1, 2, 3, and 4 times for each hole, the shadow mask was removed.



Figure S7. Schematics (top) and photograph (bottom) of micropatterned agarose/alginate DN gel by PDA NPs. Polymerizations were carried out 1, 2, 3 and 4 times for each hole (refer to the numbers in the figure).