Electronic Supplementary Information

Super Strong Dopamine Hydrogels with Shape Memory and Bioinspired Actuating Behaviours Modulated by Solvent Exchange

Jiahe Huang,[a] Jiexin Liao,[a] Tao Wang,*[a] Weixiang Sun[a] and Zhen Tong*[ab]

[a.] Research Institute of Materials Science, South China University of Technology, Guangzhou 510640, China. E-mail: fetwang@scut.edu.cn, mcztong@scut.edu.cn. Tel: (86)-20-87112886; Fax: (86)-20-87110273

[b.] State Key Laboratory of Luminescent Materials and Devices, South China University of Technology, Guangzhou 510640, China.

Synthesis of Dopamine methacrylamide (DMA): The reaction was conducted in a 500 mL three-neck flask by adding 200 mL of distilled water, 20 g of sodium borate and 8 g of sodium bicarbonate. The solution was degassed by bubbling nitrogen for 20 min, and then 10 g of dopamine hydrochloride was added into the solution. A solution containing 9.4 mL of methacrylic anhydride (94%, Sigma) in 50 mL of tetrahydrofuran (THF) was prepared separately and added dropwise into the aqueous solution containing dopamine hydrochloride. The pH of the solution was tuned and kept at moderately basic (pH 8 or above) using 1 M NaOH, and checked by pH indication paper. The reaction solution was stirred for 14 hours at room temperature under nitrogen bubbling. After the reaction was finished, the solution was vacuum filtered to remove the salts. Then, the solution was acidified to pH = 2 with 6 M HCl and washed with 200 mL of ethyl acetate. The extracted clean brown organic layer in ethyl acetate was dried over $MgSO_4$ and reduced to 50 mL by a rotary evaporator, followed by precipitating twice in 500 mL of hexane. The precipitate was dried in vacuum overnight to yield the final gray powder. ^{1}H-NMR was conducted in d-DMSO using Bruker AVANCE III 600 MHz NMR spectrometer and proved that DMA was successfully synthesized and purified (Figure S1).

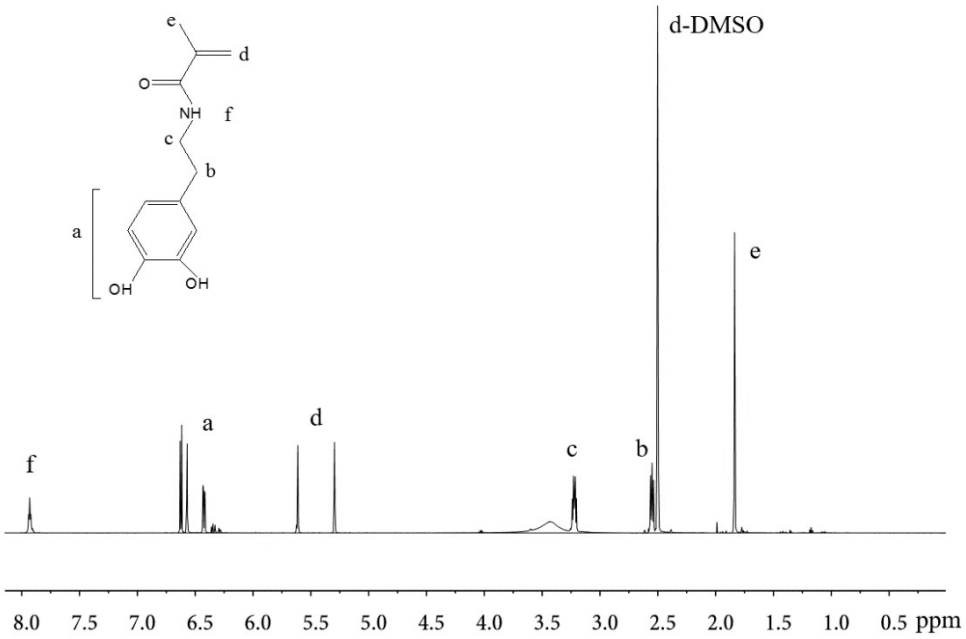

Figure S1. ^{1}H-NMR spectrum of DMA in d-DMSO.

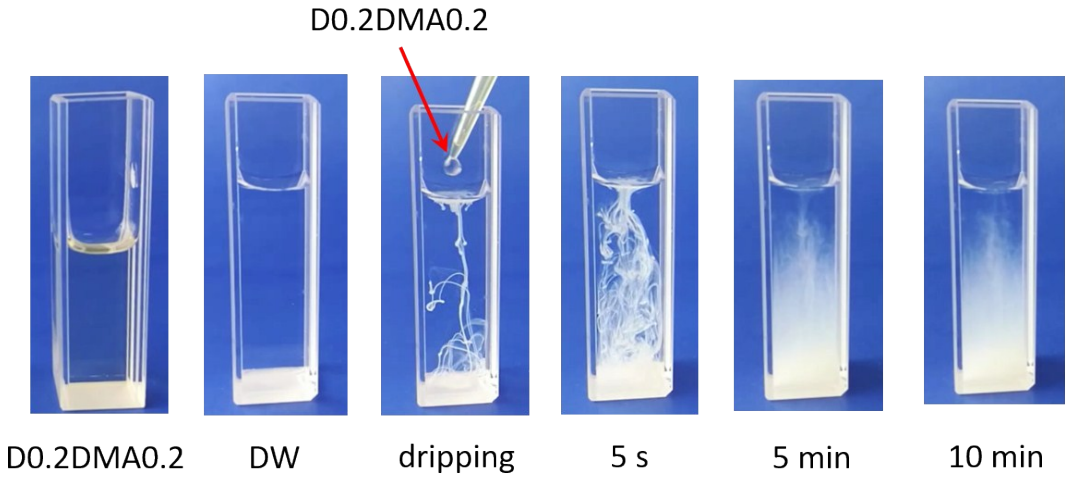

Figure S2. Photos of dripping 20 μL D0.2DMA0.2 solution into 3 mL deionized water.

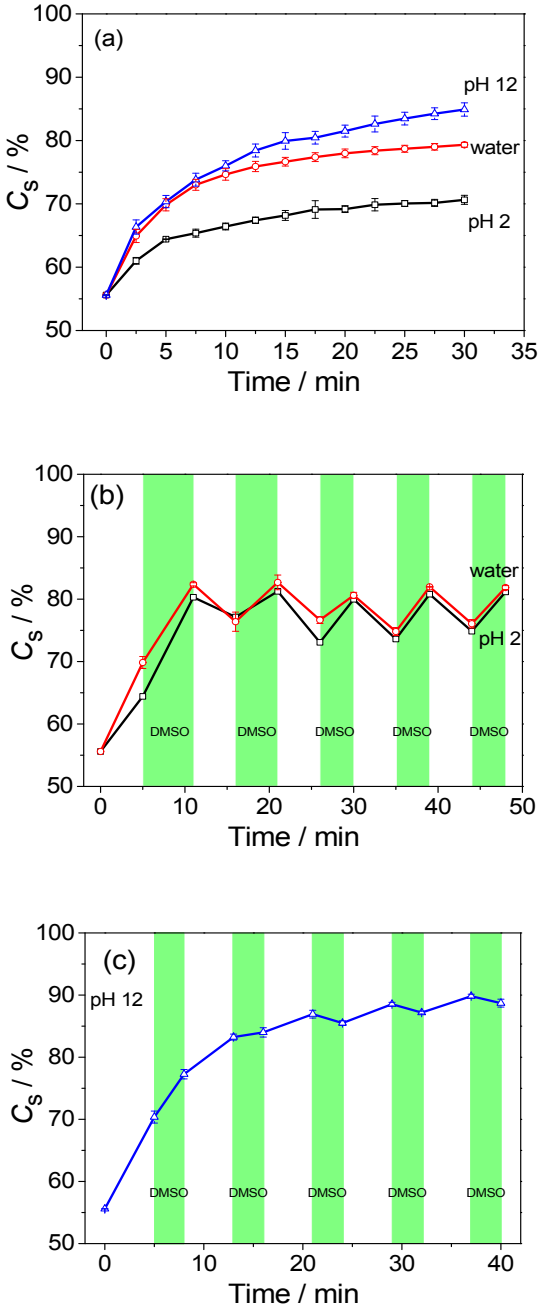

Figure S3. Solvent content C_s of the D2B0.4DMA2 hydrogel after different treatments: (a) immersed in solutions of indicated pH and deionized water for specified time; (b) alternately immersing in solution of pH = 2 or water for 5 min and in DMSO (green region) till the hydrogel becomes transparent; (c) alternately immersing in solution of pH = 12 for 5 min and in DMSO (green region) until the hydrogel becomes transparent.

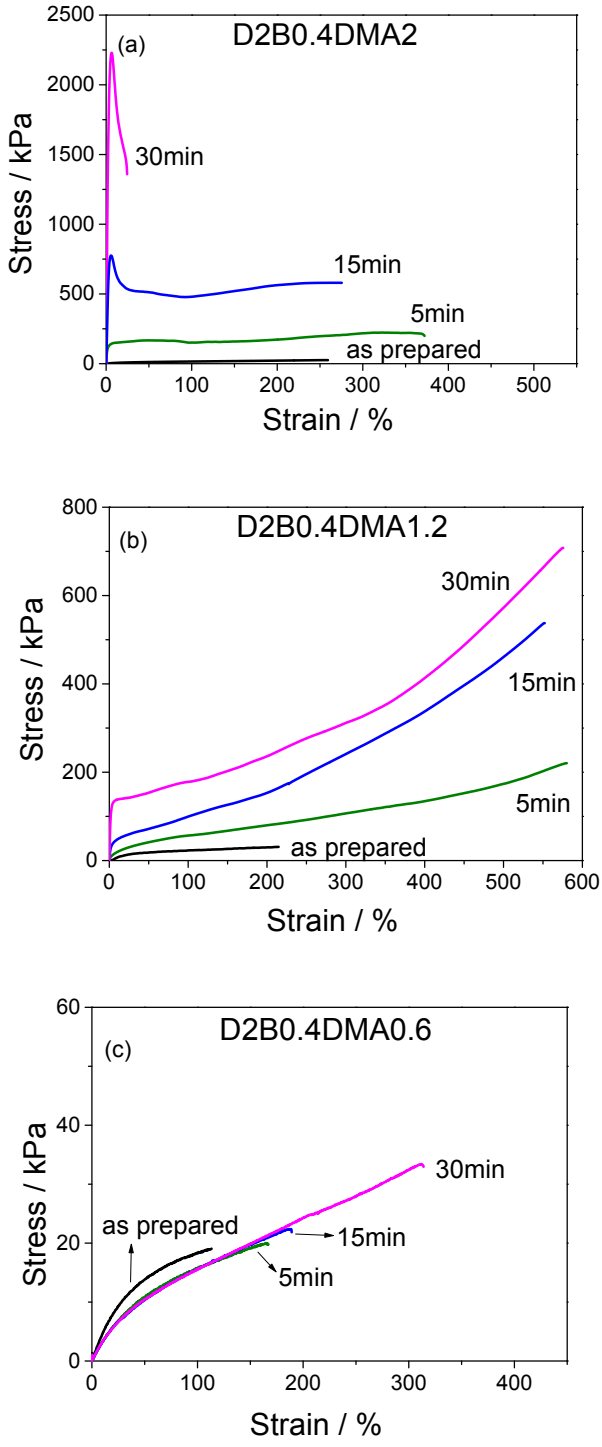

Figure S4. Tensile stress-strain curves for the D2B0.4DMA*m* hydrogels at states of as-prepared and immersed in water for the indicated times containing different DMA contents, (a) D2B0.4DMA2, (b) D2B0.4DMA1.2, and (c) D2B0.4DMA0.6.

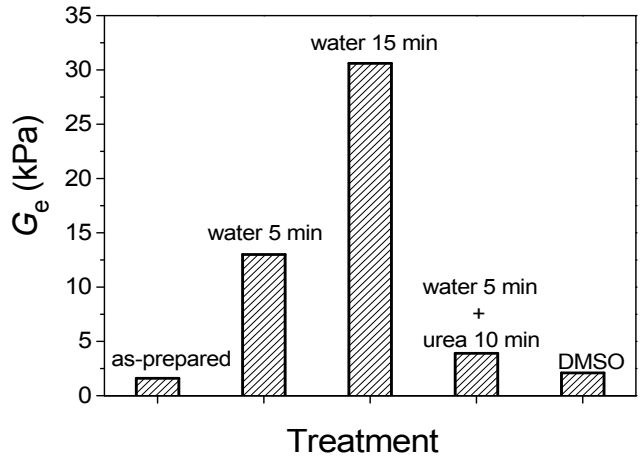

Figure S5. Equilibrium shear modulus G_e, taken from the plateau G' value at low frequency ω, of the hydrogel after specified treatments, and "water 5 min + urea 10 min" means immersed in urea solution (3 mol/L) for 10 min after immersed in water for 5 min.

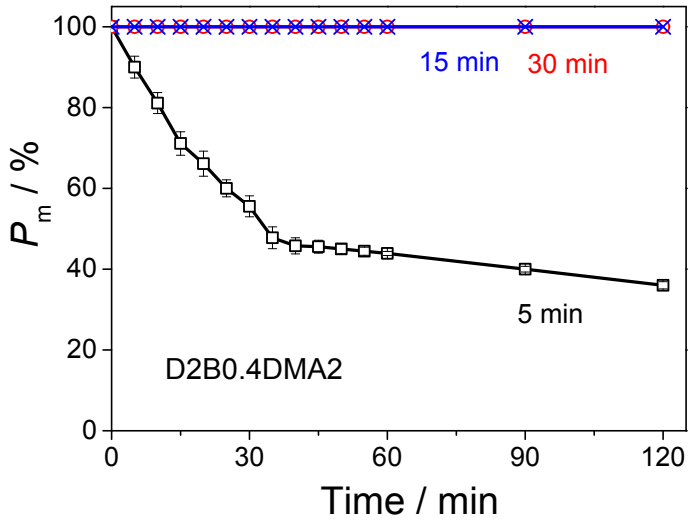

Figure S6. Shape maintaining percentage P_m of the D2B0.4DMA2 hydrogel after immersing in water for 5 min (black square), 15 min (blue cross) and 30 min (red circle) as a function of maintaining time in the atmosphere.

Supporting Movie

M1. Reversible whitening of D2B.04DMA2 hydrogel upon immersing in solution of pH = 2 and DMSO.

M2. Reversible whitening of D2B.04DMA2 hydrogel upon immersing in water and DMSO.

M3. Reversible whitening of D2B.04DMA2 hydrogel upon immersing in solution of pH = 12 and DMSO.

M4. Shape fixing and shape recovery process of the D2B.04DMA2 hydrogel.

M5. DMSO triggered retraction of the D2B.04DMA2 hydrogel tentacle.

M6. DMSO triggered weight lifting of the D2B.04DMA2 hydrogel arm.