

Supporting Information

A Skin Inspired Bio-smart Composite with Water Responsive Shape

Memory Ability

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Table S1 Formula of nonionic waterborne polyurethane dispersions

	Dosage (molar)						Solid content (%)
	PTMG-1K	PTMG-2K	N120	NPG	IPDI	EDA	
Nonionic waterborne PU	0.050	0.025	0.020	0.019	0.200	0.072	45.00%

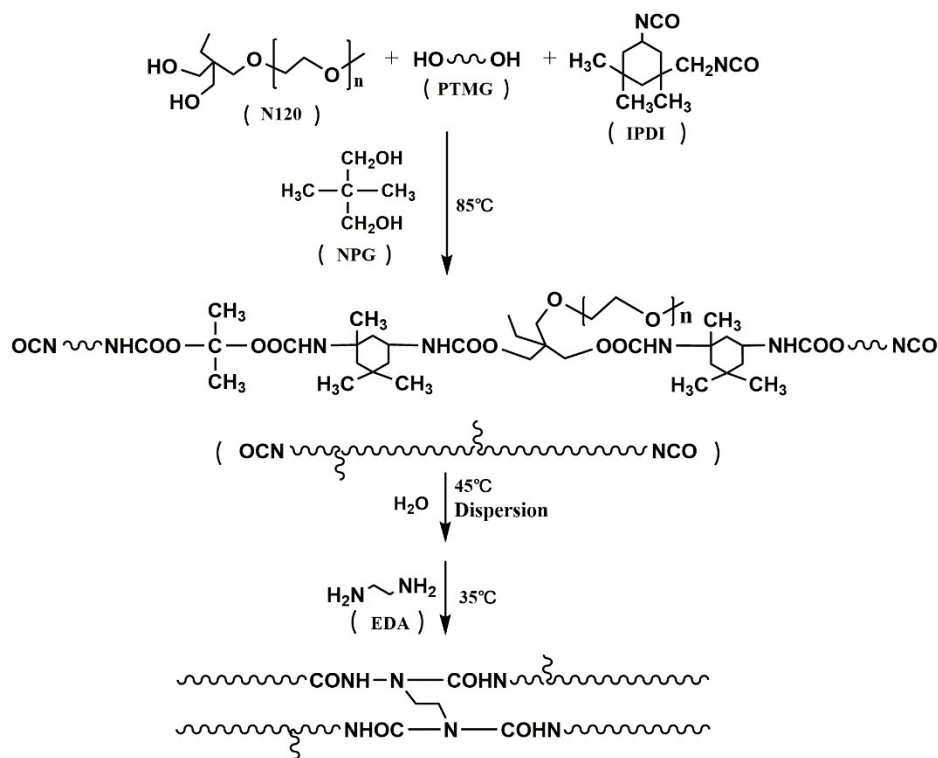


Figure S1. Preparation process and molecular structure of nonionic waterborne polyurethane dispersions

The formulation and molecular structure of nonionic waterborne polyurethane (PU) is given in Table 1 and Figure 1, respectively. The polymerization was carried out in a flask equipped with a mechanical stirrer, thermometer, reflux condenser and additional funnels under a constant temperature oil bath. Polytetramethylene glycol (PTMG), nonionic hydrophilic monomer (N120) were added to the flask. After mixing evenly, Isophorone diisocyanate (IPDI) was added followed addition of 0.05 wt % of dibutyltin dilaurate drop by drop. The mixtures were reacted at 85 °C for 3 h to obtain polyurethane prepolymer. Neopentyl glycol (NPG) was then added for chain extension for another 2 h. Then, the temperature was cooled down to 40~45 °C. Followed by rapidly addition of quantitative distilled water w at a stirring agitation speed of 2000 rpm for 10 min. Ethylenediamine (EDA) was added dropwise and chain extension reaction at 35 °C lasted for 1.5 h. The final obtained nonionic waterborne PU dispersion had ~45 wt.% solid content and pH value of 7.

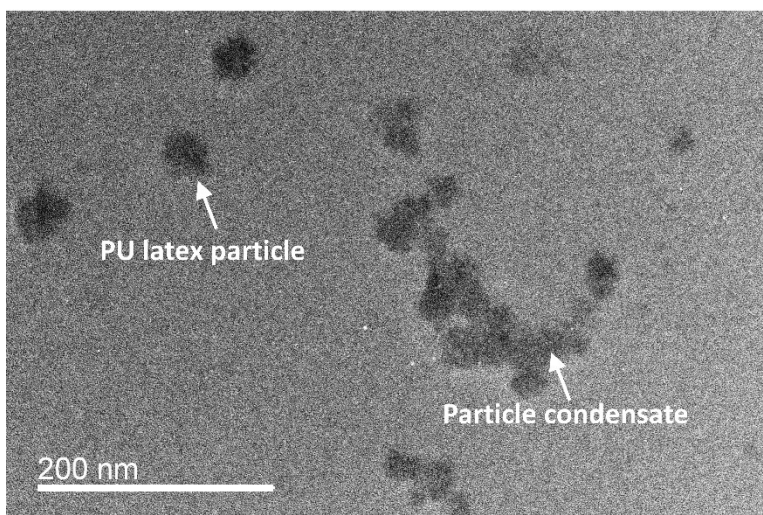


Figure SS2. TEM image of nonionic waterborne polyurethane dispersions

Nonionic waterborne polyurethane dispersions were diluted to a concentration of 1 wt% and were drop on a copper mesh. After drying, the copper mesh was observed by transmission electron microscopy (JOEL, Peabody, MA, USA) operated at an accelerating voltage of 200 kV.

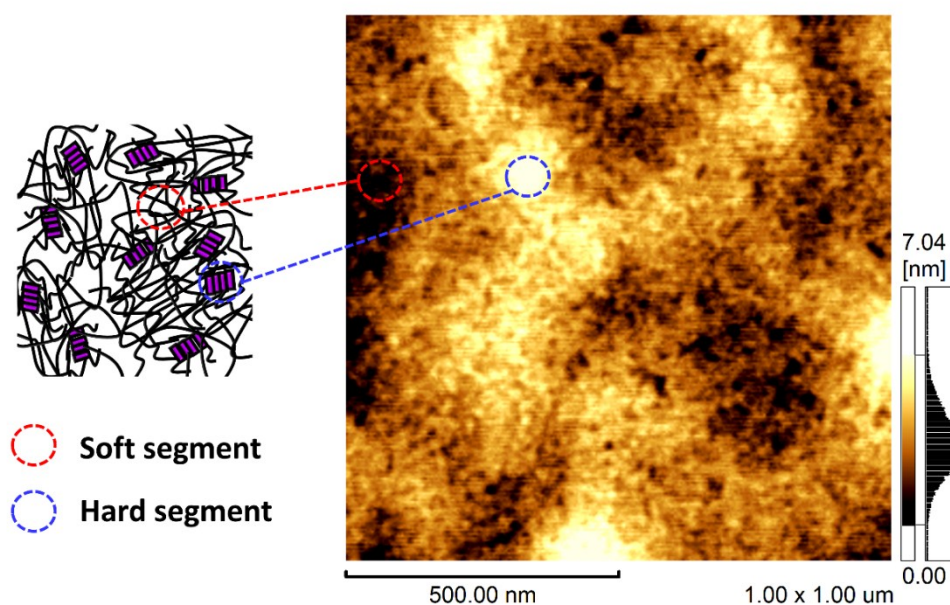


Figure S3. AFM microphase separation structure of PU

Atom force microscopy (AFM) was performed on the surface of the coatings with a scanning area of $5\ \mu\text{m} \times 5\ \mu\text{m}$ using the SPA-400 Atomic force microscope (Seiko Instruments Inc. Japan) in tapping mode. Figure 3 shows the phase images of cured PU matrix where dark and bright areas are owing to soft segments and hard segments respectively.

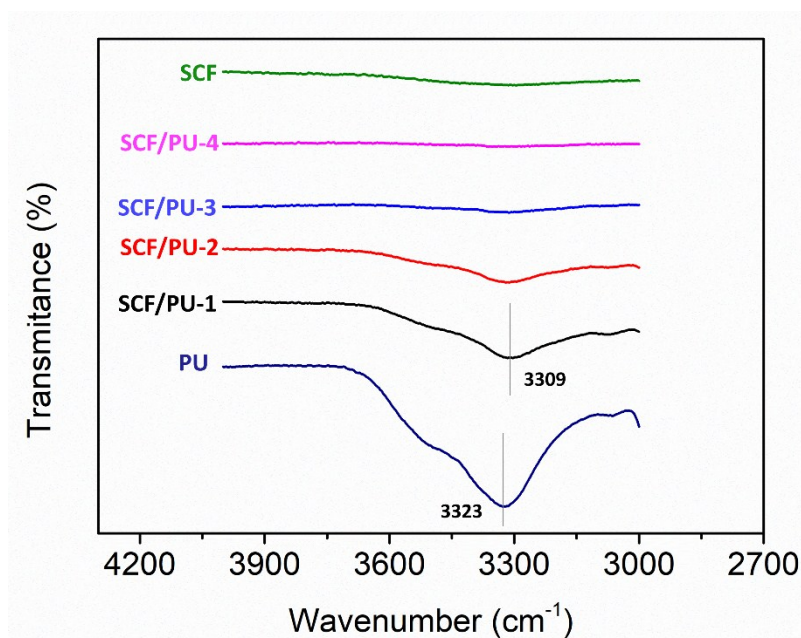


Figure S4. Magnification of FTIR spectra in N-H stretching of SCF/PU composites and PU.

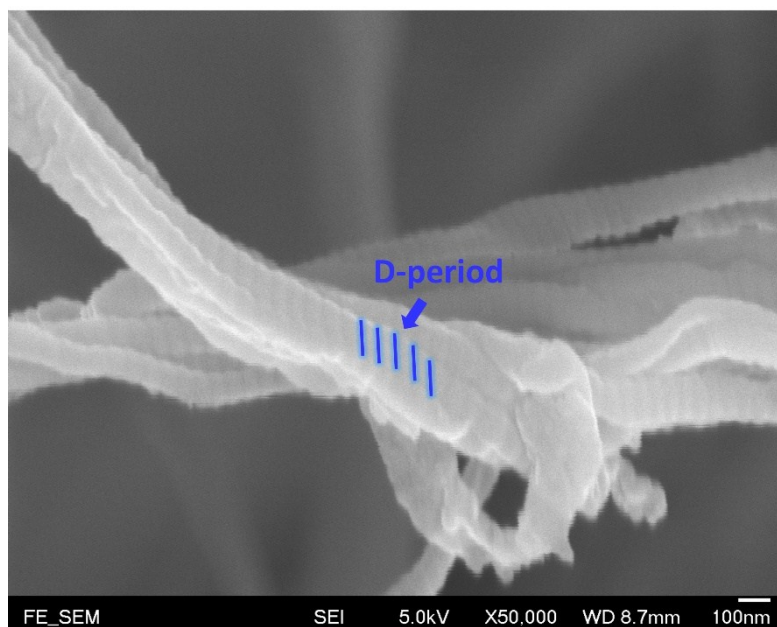


Figure S5. SEM image of collagen fibril which illustrate the typical D-period structure in collagen.

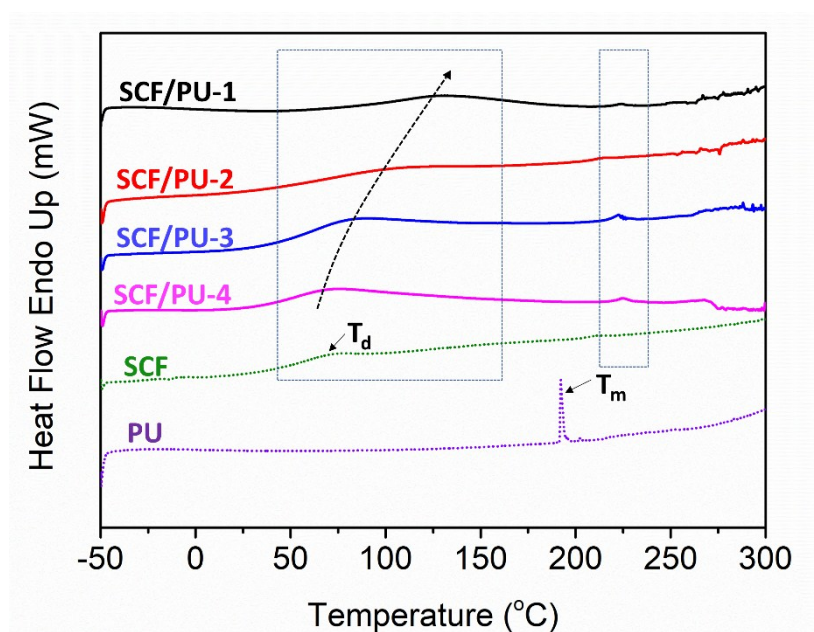


Figure S6. DSC results of SCF/PU composites and PU.

Differential Scanning Calorimetric (DSC) was performed DSC 8000 instrument (PerkinElmer, USA) from $-50\text{ }^{\circ}\text{C}$ to $300\text{ }^{\circ}\text{C}$ by a heating rate of $10\text{ }^{\circ}\text{C}/\text{min}$. As shown in Figure S6, characteristic peak of SCF at $65\text{ }^{\circ}\text{C}$ are attributed to denaturation temperature (T_d) of collagen molecules. This endotherm originates from the transition of the triple helix structures of collagen molecules into randomly coiled structures, which involves the breakage of intra- and inter- molecular hydrogen bonds. PU exhibits

a T_m of hard segments at 192 °C. It is noticed that there appeared new small peak at around 220 °C for SCF/PU composite, which may due to the existence of PU matrix.

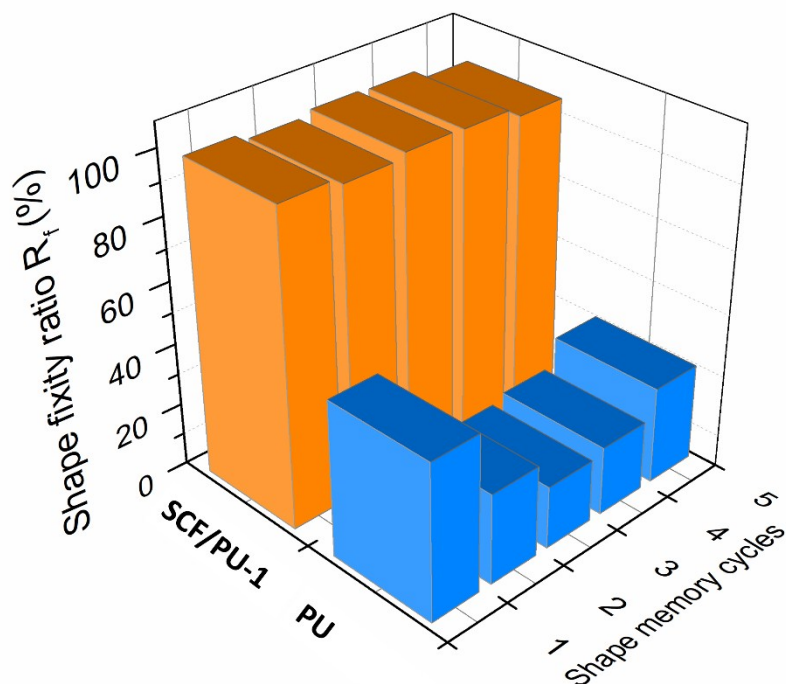


Figure S7. Shape fixing ratio (R_f) for SCF/PU-1 and PU during five shape memory programming cycles

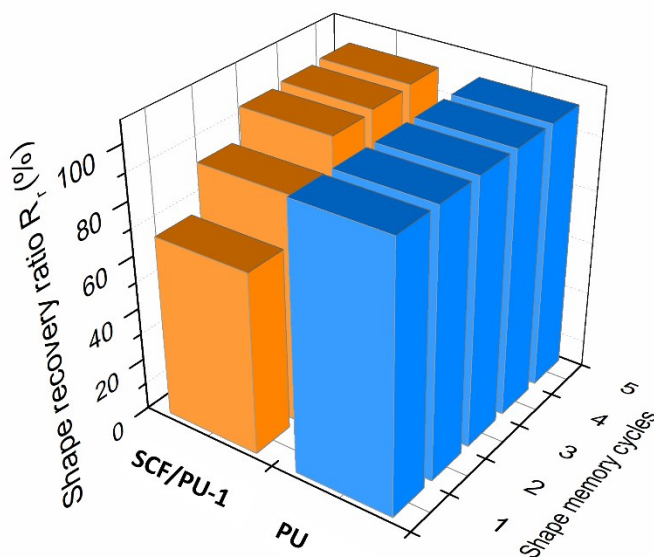


Figure S8. shape recovery ratio (R_r) for SCF/PU-1 and PU during five shape memory programming cycles

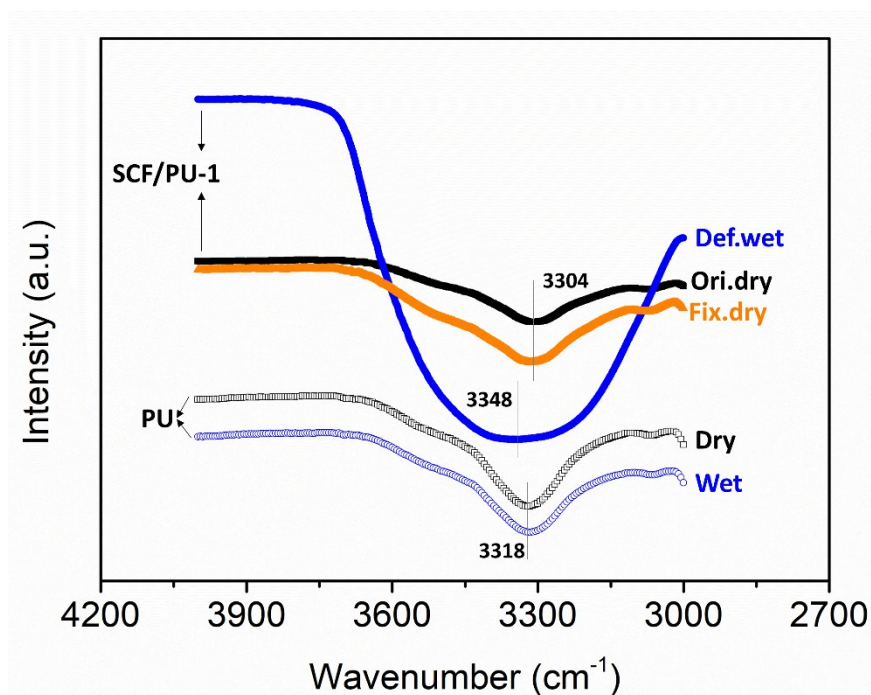


Figure S9. FTIR spectra in N-H stretching of SCF/PU-1 in shape memory programming steps and PU under dry and wet state (Ori.dry: original dry; Def.wet: deformation in wet state; Fix. dry: fixation in dry state)

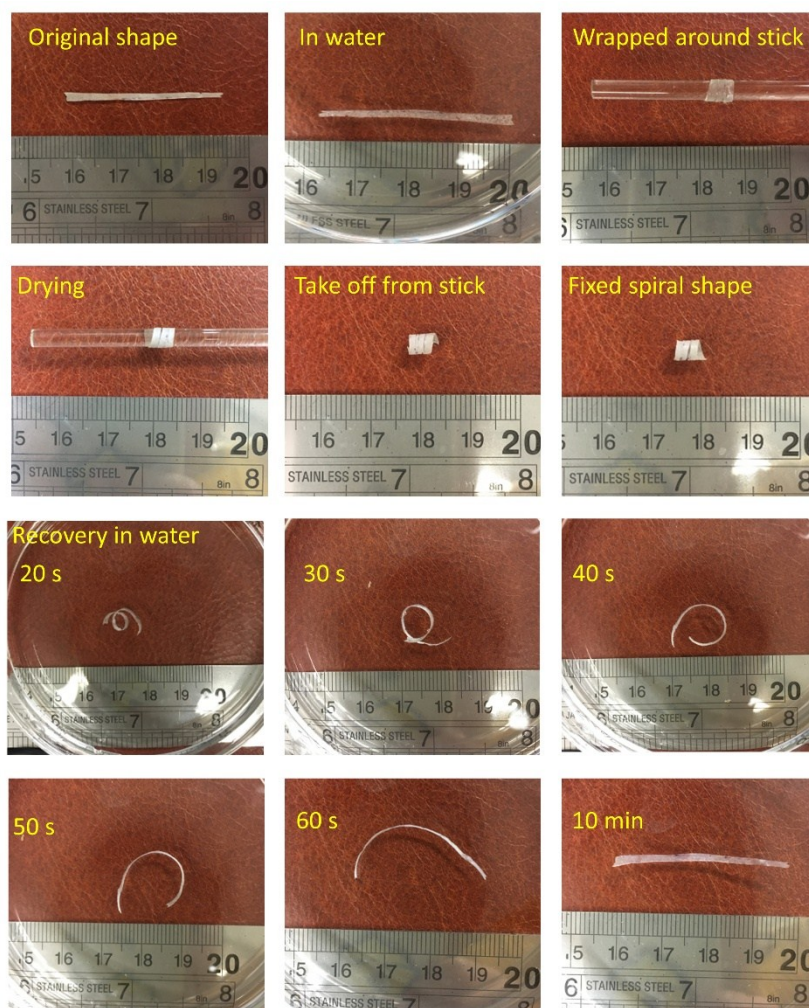


Figure S10. Photos for water-responsive shape memory ability of SCF/PU composite

Figure S10 shows the shape memory tunability of SCN/PU by photo. Helix shape of SCN/PU in wet state is almost completely fixed after drying. By immersing in water, the fixed shape gradually returns to its original straight shape.