

Supplementary Materials

Title: A general strategy of 3D printing thermosets for diverse applications

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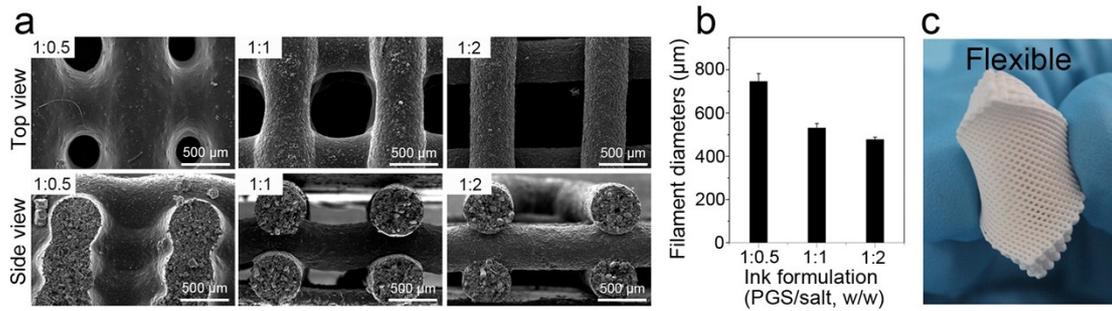
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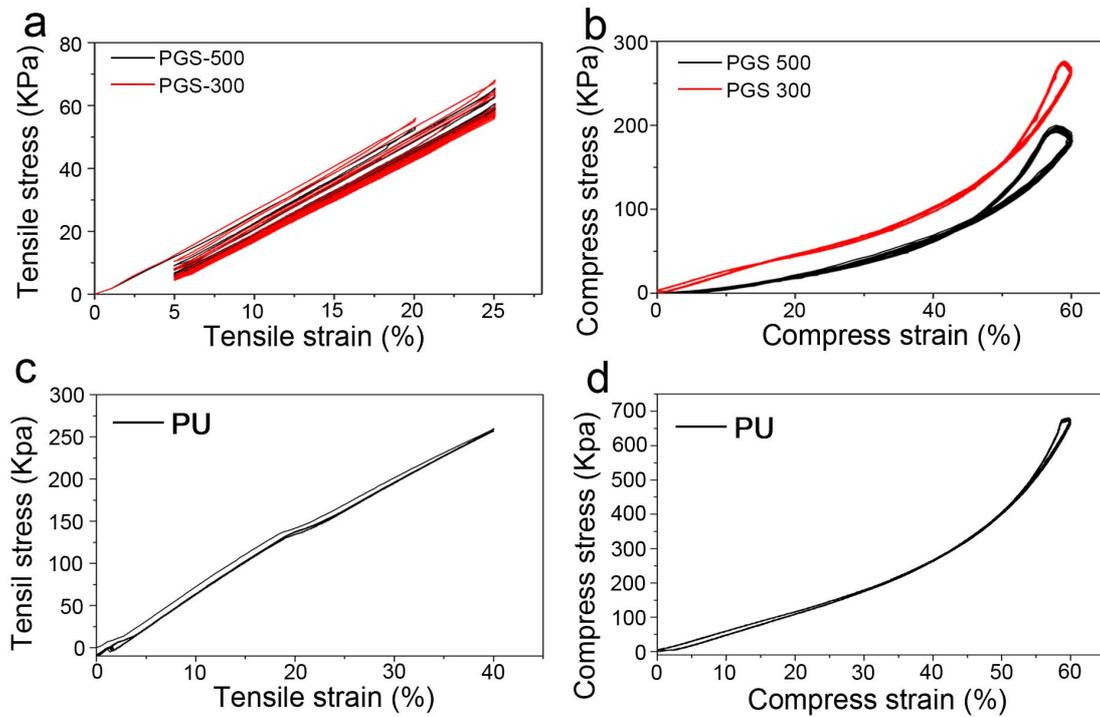
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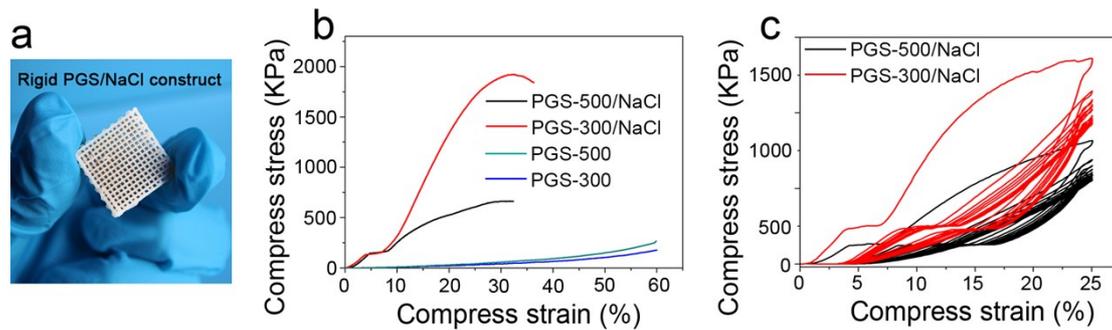
Keywords: 3D printing; thermoset; sensor and actuator; tissue engineering scaffold; cardiac patch.



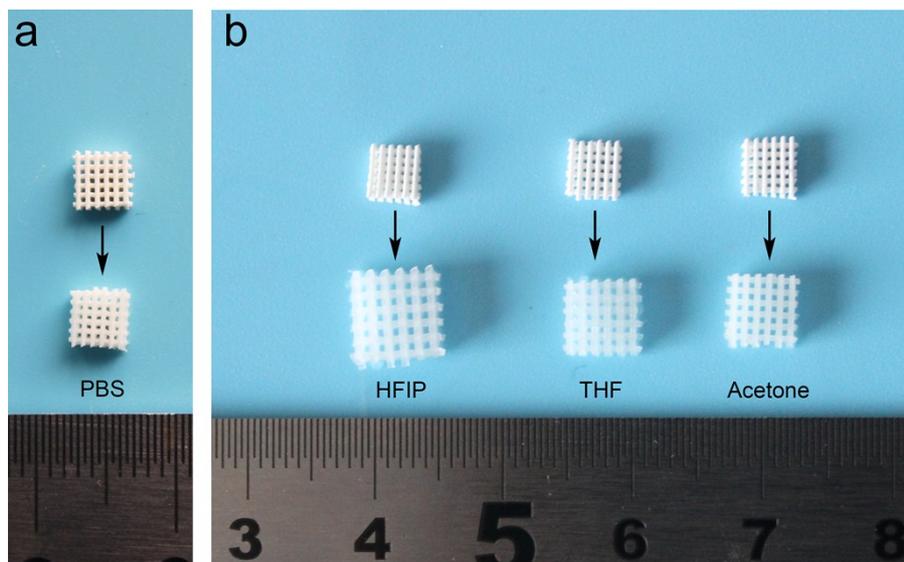
Supplementary Figure 1 a,b, SEMs (**a**, Scale bar, 500 µm) and filament diameters (**b**) of cured PGS-500 constructs with different formulations (PGS/salt, w/w) before salt leaching. **c**, Multilayered PGS scaffolds showed good flexibility after leaching process.



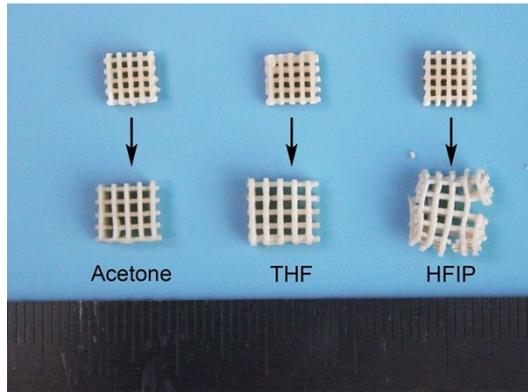
Supplementary Figure 2. Cyclic tensile (**a,c**) and compression (**b,d**) tests for 10 cycles of 3D printed thermoset PGS and PU constructs, respectively revealed their good elasticity and anti-fatigue property.



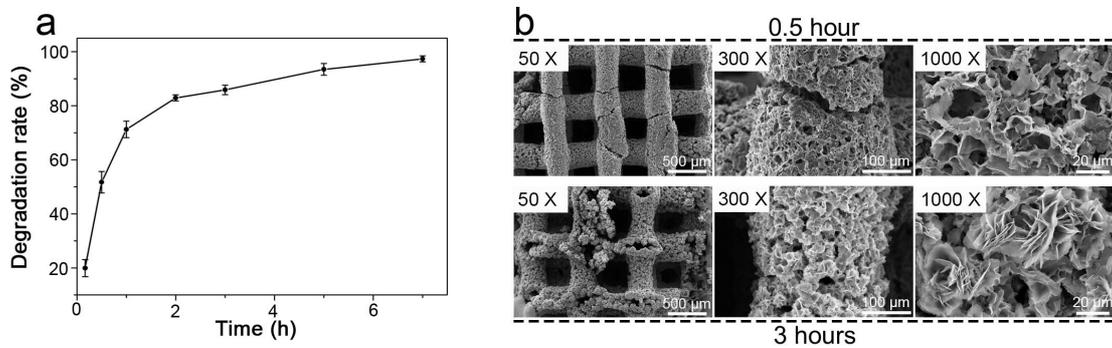
Supplementary Figure 3. **a**, Photograph of rigid multilayered PGS/NaCl construct. **b**, Typical stress vs. strain curves of uniaxial compression tests of 3D printed thermoset PGS constructs before and after removal of NaCl particles. **c**, Cyclic compression tests for 10 cycles of 3D printed thermoset PGS constructs containing NaCl particles.



Supplementary Figure 4. Swelling properties of PGS constructs in different solvents. **(a)** Negligible swell in phosphate-buffered saline (PBS) **(b)** The average volume swelling degrees in polar organic solvents were $1212 \pm 131\%$ (HFIP), $768 \pm 67\%$ (THF) and $511 \pm 57\%$ (acetone).



Supplementary Figure 5. PGS-300/NaCl composite constructs swelling in acetone, THF and HFIP.



Supplementary Figure 6. *In vitro* degradation of 3D printed PGS constructs in a lipase aqueous solution. The mass loss of PGS constructs with degradation time (**a**) and SEMs of PGS constructs after half hour and 3 hours degradation (**b**, scale bar, 500 μm, 50 μm and 10 μm).

Supplementary Movie S1

This Movie shows good flexibility of 3 D printed multilayered PGS scaffold.

Supplementary Movie S2

This Movie shows the cyclic compressive test of a 3D printed PGS construct. The 3D printed PGS construct has good elasticity and anti-fatigue property under dynamic deformation.

Supplementary Movie S3

This Movie shows the vapomechanically responsive sensor, which could sense the direction of vapor flow with a dramatic shape deformation and recovered in air.

Supplementary Movie S4

This Movie shows the “artificial flower” swelling behavior of soft actuator, while droplets of HFIP was added on the loose layer. The actuator was responsive to solvent and closed the petals, then steadily blossomed with desorption in a ventilation environment.

Supplementary Movie S5

This Movie shows the “artificial octopus” swelling behavior of soft actuator, while droplets of HFIP was added on the dense layer. The actuator was responsive to solvent and stood up, then steadily lay down with desorption in a ventilation environment.