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Supporting Information

Design-encoded Dual Shape-morphing and Shape-memory in 4D Printed Polymer Parts Toward Cellularized Vascular Grafts

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1. Results



Figure S1: Thermal and thermomechanical characterization of PLMC. a) Second heating curve of PLMC in DSC; b) Thermogram showing thermal degradation profile of PLMC in TGA; c) Estimation of T_g through tan δ profile in DMA; d) Shape memory property quantification in DMA. e) Cyclic shape memory property quantification in DMA.



Figure S2: Stress-strain profile of as-printed PLMC bars (60×6×0.4 mm³) in longitudinal (L) and transverse (T) infill stackings.



Figure S3: Deformation of flat sheets into final shapes with respect to the encoding direction in the bottom layer



Figure S4: Low magnification of HUVECs over PLMC tubes. a) Live-dead staining showing high viability of HUVECs; b) Actin-nuclear staining highlighting high coverage of HUVECs forming monolayers at day 5 (Scale: 200 μm)



Figure S5: Dynamic mechanical testing of 3D printed PLMC bars in uniaxial tension mode at 37°C. a) Stress-strain profile; b) Cyclic stress-strain profiles up to fifteen consecutive cycles; c) Strain profile and d) Strain recovery profile in creep test performed under loading stress of 1 MPa.



Figure S6: Dual shape morphing and shape recovery in different as-printed PLMC structures with varied infill angles. Sequential a) Bending and b) Twisting deformation of a butterfly, flattening and fixing, and recovery to the original bent/ twisted shape. i) As-printed butterfly

with a) TTLL and b) (45°, -45°) design encoding; ii) Programmable a) Bending and b) Twisting deformation due to relaxation of residual stresses at 80°C; iii) External force aided flattening of the morphed shapes into original shapes; iv) Shape recovery of the flat shapes at 37°C into original a) bent and b) twisted butterfly shapes. Sequential c) Bending and d)

Twisting deformation of a cross shape, flattening and fixing, and recovery to the original bent/ twisted shape. i) As-printed cross with c) TTLL and d) (45°, -45°) design encoding; ii) Programmable c) Bending and d) Twisting deformation due to relaxation of residual stresses at 80°C; iii) External force aided flattening of the morphed shapes into original shapes; iv) Shape recovery of the flat shapes at 37°C into original a) bent and b) twisted cross shapes.

Video Captions:

V1: Complete bending of rectangular flat sheet (40×12×0.4 mm³) with TTLL design encoding into hollow tubes

V2: Complete bending of square flat sheet $(30 \times 30 \times 0.4 \text{ mm}^3)$ with TTLL design encoding into hollow tubes

V3: Bending deformation of cross shape with TTLL design encoding

V4: Twisting deformation of cross shape with (45°, -45°) design encoding

V5: Bending deformation of butterfly shape with TTLL design encoding

V6: Twisting deformation of butterfly shape with (45°, -45°) design encoding

V7: Bending of a Venus flytrap shape with TTLL design encoding

V8: Simultaneous curling and twisting deformations in a single flat sheet with TTLL and (45°,

-45°) design encoding

References:

 Choudhury, S.; Joshi, A.; Dasgupta, D.; Ghosh, A.; Asthana, S.; Chatterjee, K., 4D Printed Biocompatible Magnetic Composite for Minimally Invasive Deployable Structures. 2022 <u>https://doi.org/10.26434/chemrxiv-2022-dv25j</u>