Supplementary Information

4D Printed Biocompatible Magnetic Nanocomposite Toward Deployable Constructs

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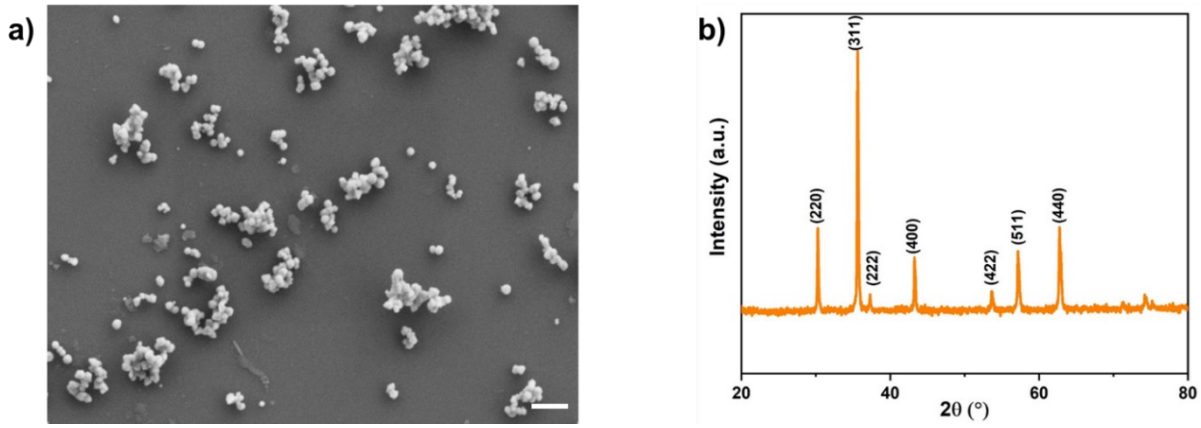

Fig S1: a) SEM and b) XRD of Fe_3O_4 nanoparticles. (Scale: 1 µm)

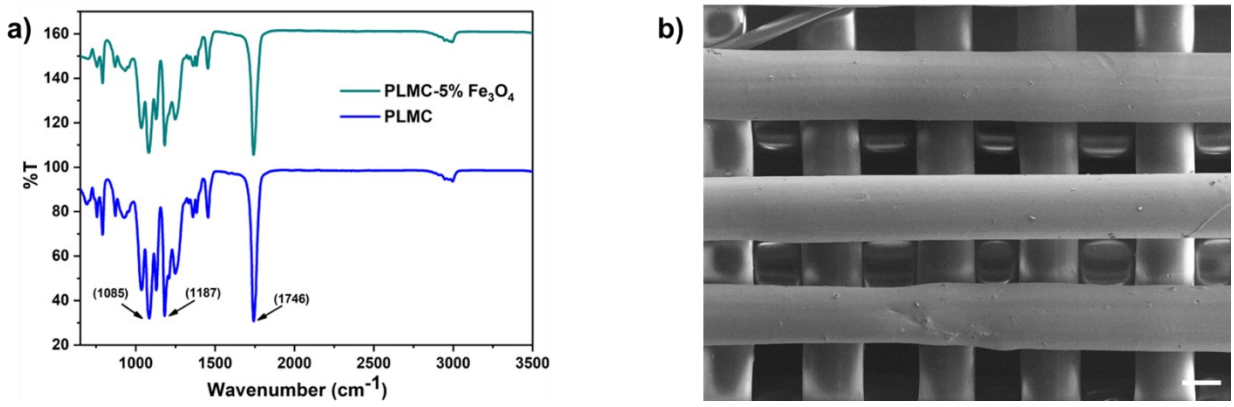

Fig S2: a) FTIR spectra of PLMC and composite, b) SEM of 3D printed PLMC-5% Fe_3O_4 composite. (Scale: 100 µm)

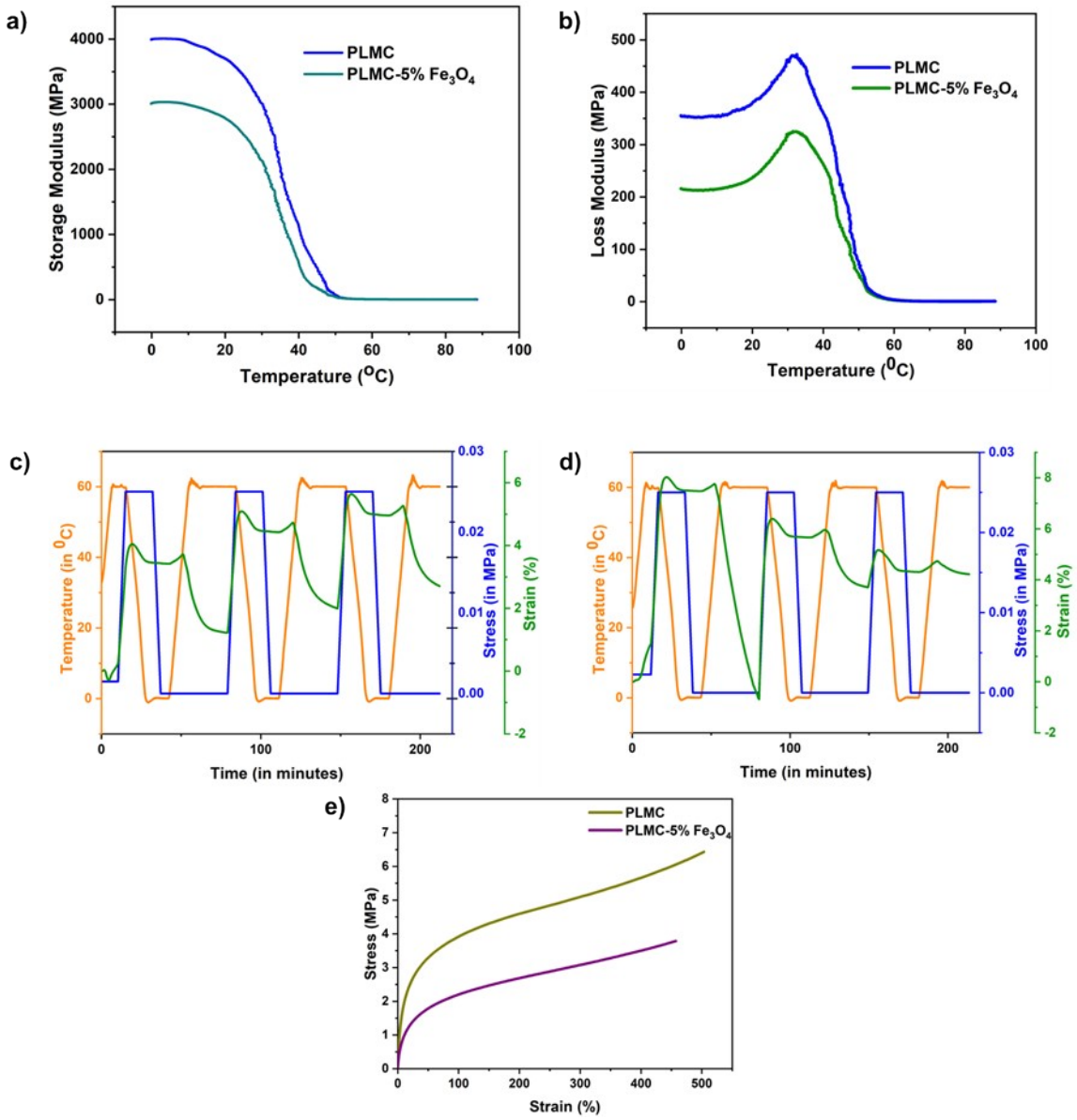

Figure S3: a) Storage modulus of PLMC and composites versus temperature, b) Loss modulus of PLMC and composites versus temperature, c) Cyclic shape memory testing of PLMC, d) Cyclic shape memory testing of PLMC composites, e) Uniaxial stress-strain profiles of PLMC and composites at 37°C.

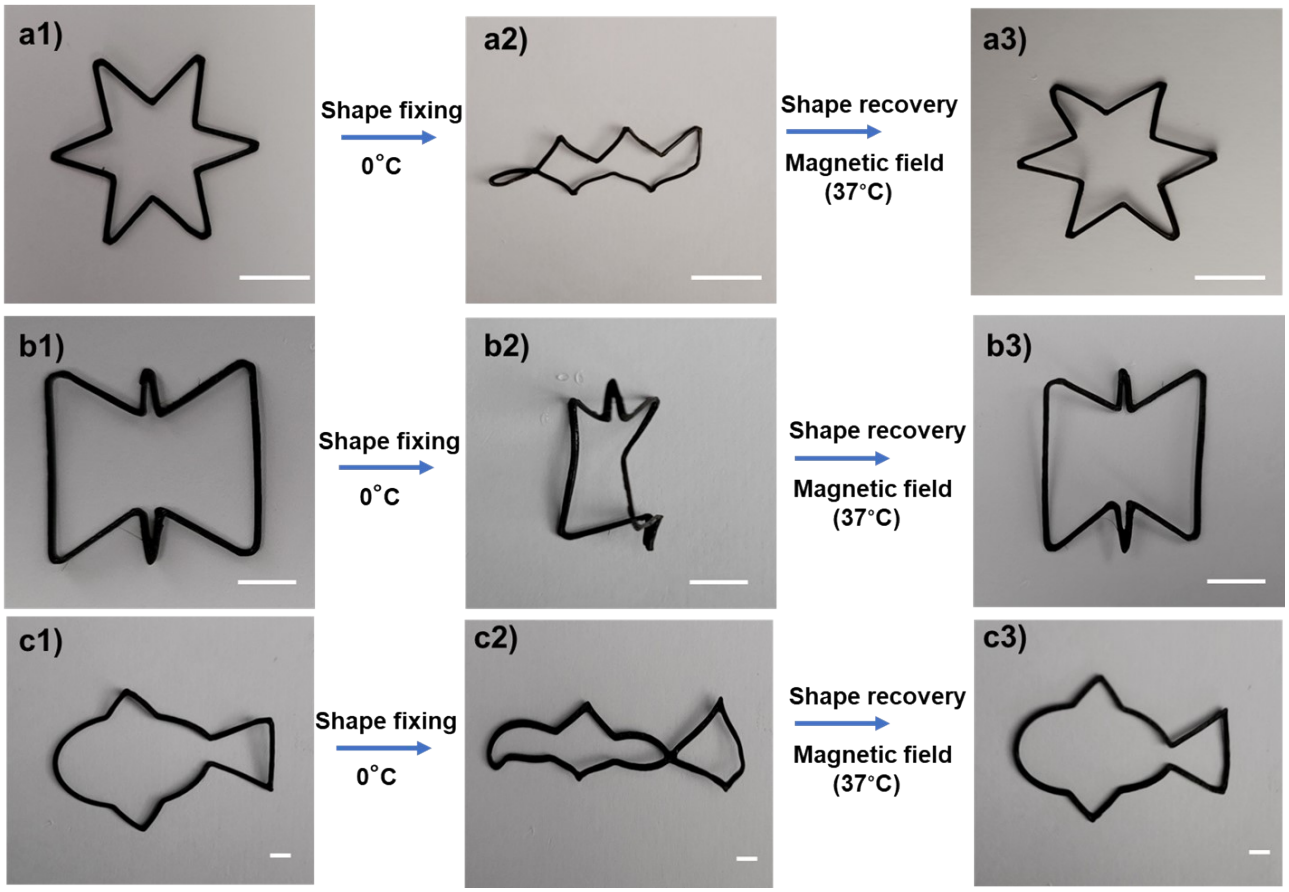

Figure S4: Shape recovery of 3D printed PLMC-5% Fe_3O_4 2D shapes from pre-programmed temporary shapes to original shapes under alternating magnetic field; a1) As printed star shape, a2) Deformed and fixed ($<T_g$) star, a3) Recovered ($>T_g$) star, b1) As printed butterfly shape, b2) Deformed and fixed ($<T_g$) butterfly, b3) Recovered ($>T_g$) butterfly, c1) As-printed fish shape, c2) Deformed and fixed ($<T_g$) fish, c3) Recovered ($<T_g$) fish (Scale bar is 10 mm).

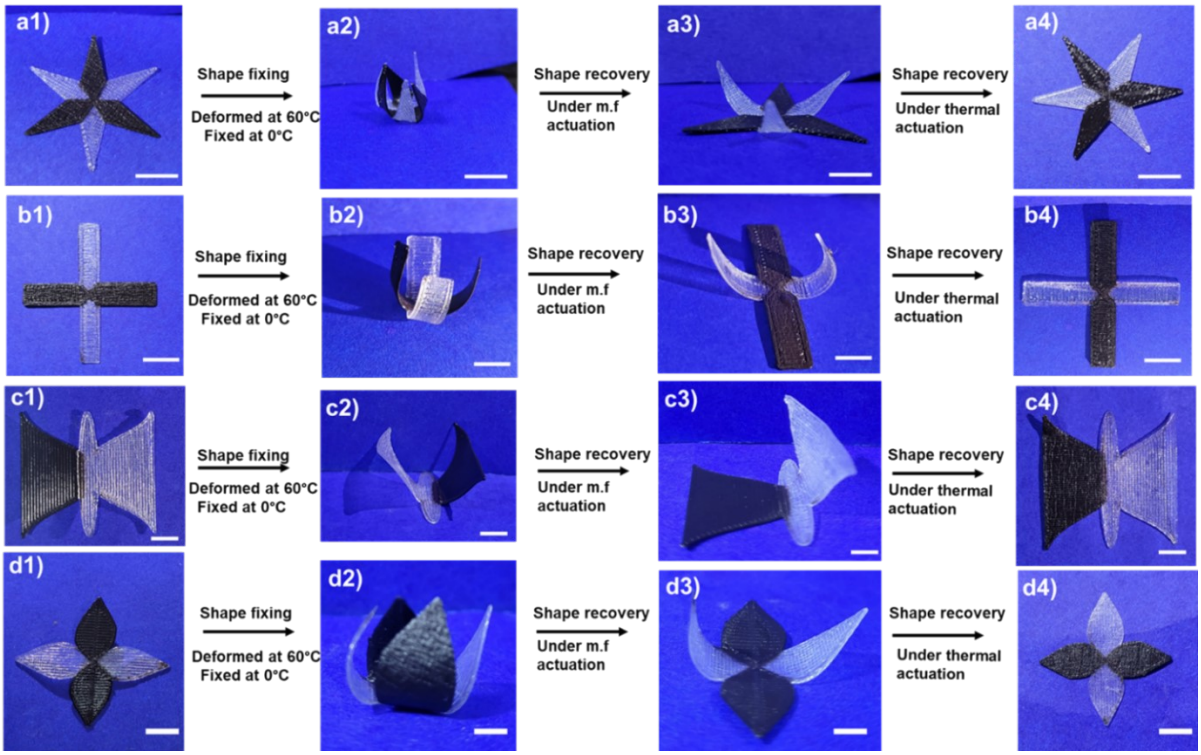

Figure S5: Shape recovery of 3D printed PLMC and PLMC-5% Fe_3O_4 composite dual structures sequentially and selectively actuated under alternating magnetic field and then heat; a1) Dual-printed star, a2) Deformed star, a3) Partially recovered star by indirect heating, a4) Fully recovered star by direct heating, b1) Dual-printed cross, b2) Deformed cross, b3) Partially recovered cross by indirect heating, b4) Fully recovered cross by direct heating, c1) Dual-printed butterfly, c2) Deformed butterfly, c3) Partially recovered butterfly by indirect heating, c4) Fully recovered butterfly by direct heating, d1) Dual-printed petal, d2) Deformed petal, d3) Partially recovered petal by indirect heating, d4) Fully recovered petal by direct heating (Scale:10 mm)

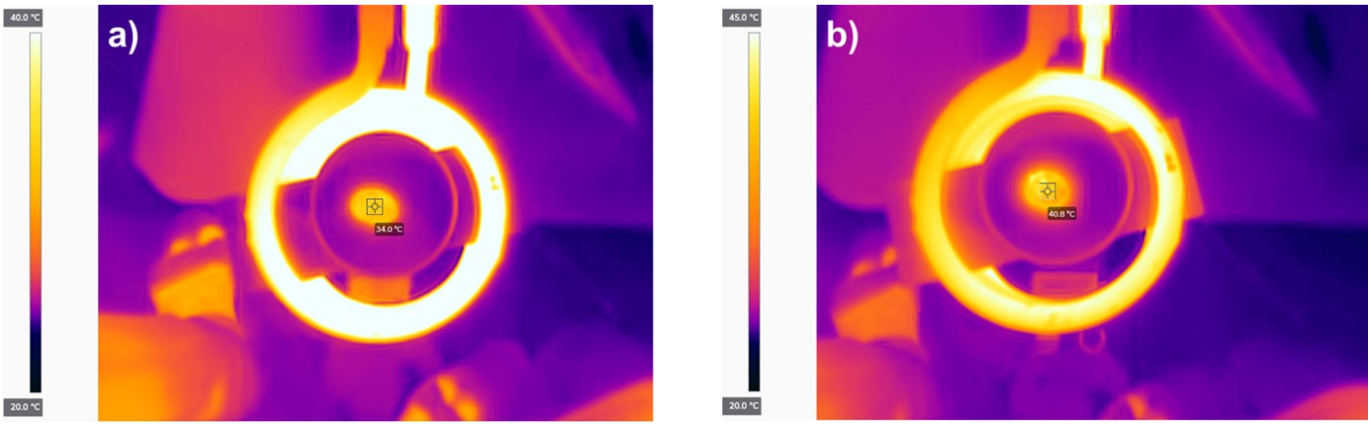

Figure S6: Thermal images captured during shape recovery of composite scaffold inside an alternating magnetic field; a) image taken at t=5 s, b) image taken at t=13 s after complete recovery.

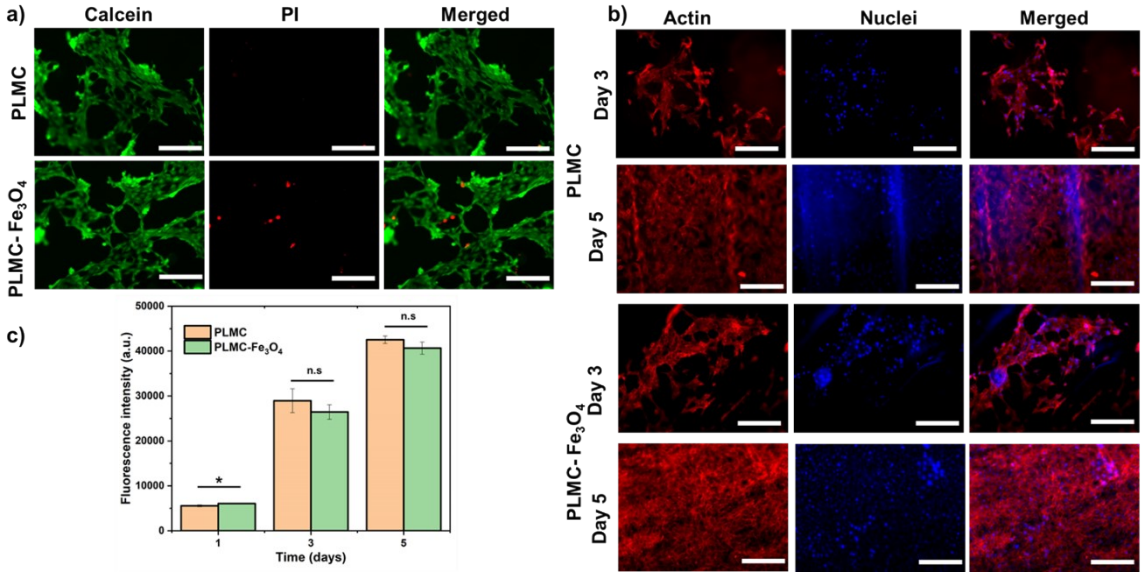

Figure S7: *In vitro* biological characterization of PLMC and its composite. a) Live-dead assay, b) Cytoskeletal staining, and c) Alamar blue assay. * denotes $p<0.05$, n.s is non-significant. (Scale bar is 100 µm)

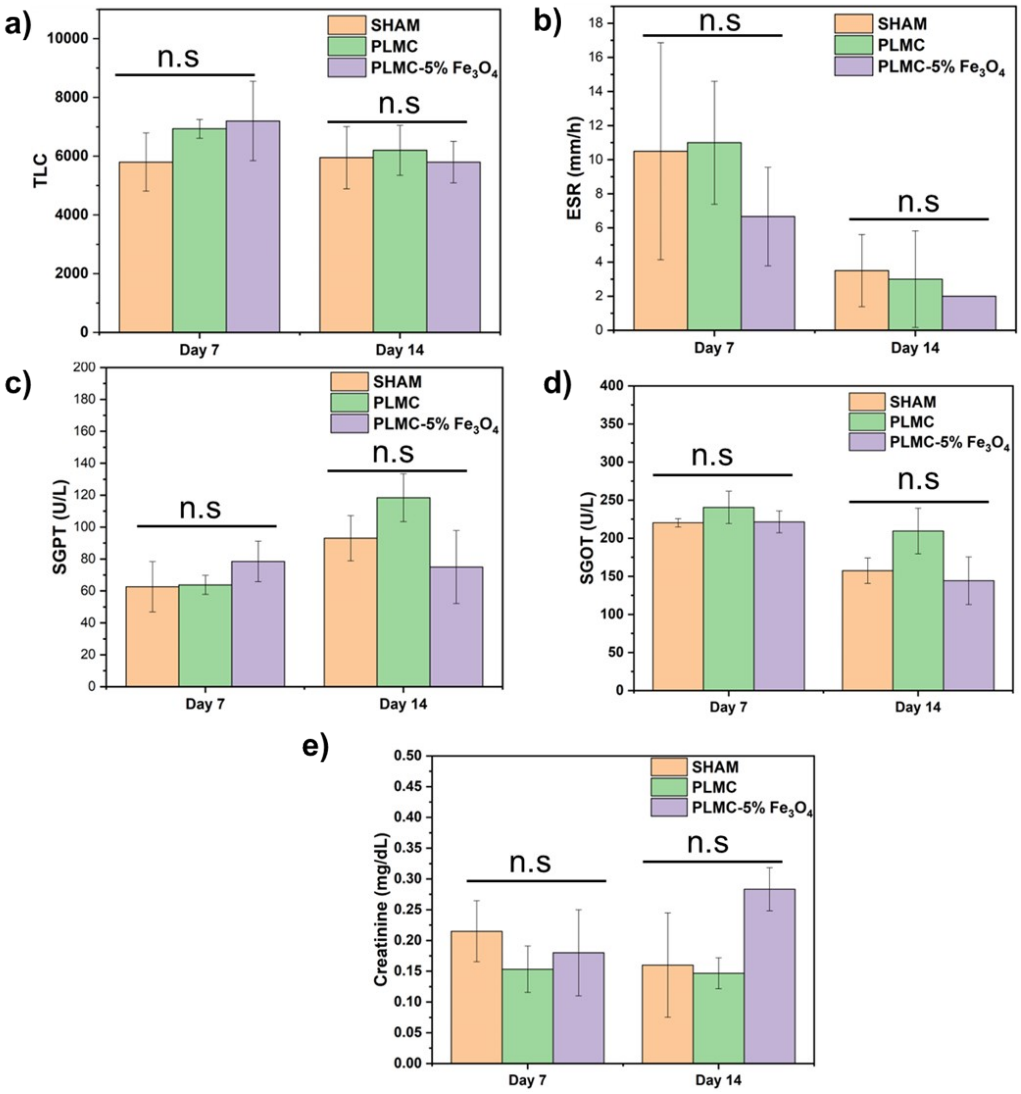

Figure S8: Plots showing data of different blood parameters a) TLC (Total leucocyte count), b) ESR (Erythrocyte sedimentation rate), c) SGPT (serum glutamic pyruvic transaminase), d) SGOT (serum glutamic-oxaloacetic transaminase), e) Serum creatinine levels assessed from SHAM, PLMC, and PLMC-Fe_3O_4 composite groups at day 7 and day 14 confirming minimal response of the materials to elicit allergy, inflammation, hepatotoxicity, and nephrotoxicity, respectively (n.s inidicates non-sigificance, one-way ANOVA).

Table S1: Comparison of this study with reported inductive heating SMP systems for biomedical applications:

Material system	Filler loading (wt%)	T_g (from DSC) (in °C)	T_g (from DMA) (in °C)	Printed structures	Proposed application	Comment	Reference
PLA/ Fe_3O_4	15	65.35	90	Porous glass-sponge like scaffolds	Tracheal scaffold	Though the composites were printed in various porous designs, high temperature of 65°C limits *in vivo* deployment.	[1]
PCL-based PUA (polyurethane acrylate)-hybrid Fe_3O_4	9	43.8 (transition temperature corresponds to melting)	-58.1	n/a	n/a	Recovery extent is slightly less (96%), no specific application proposed.	[2]
PCL-dimethacrylate-Fe_3O_4	5	74 (maximum temperature attained)	n/a	Honeycomb, spider web, etc.	Soft robotics	High surface temperatures (74°C) limit biomedical applications.	[3]
PLMC/PTMC blend (50:50)/ Fe_3O_4	20	47.22	51.86, 63.73	Flower, petal, etc.	n/a	Both the T_gs being high is a limiting factor, along with no clear justification of triple SMP in biomedicine.	[4]
PLA/ Fe_3O_4	15	64	n/a	Spinal bone, flower, etc.	n/a	High temperatures combined with higher filler loading restricts usage.	[5]
PLMC- Fe_3O_4	5	30	51	Butterfly, fish, Bone scaffolds	Deployable bone scaffold	Physiologically relevant temperatures, low filler loading and demonstration of scaffold deployment in free as well as restrictive conditions make for promising candidates.	This study

References:

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Video captions:

V1: Shape recovery of a deformed 2D star under alternating magnetic field (4X)

V2: Shape recovery of a deformed 2D butterfly under alternating magnetic field (4X)

V3: Shape recovery of a deformed 2D fish under alternating magnetic field (4X)

V4: Shape recovery of a deformed 3D petal under an alternating magnetic field

V5: Shape recovery of a deformed 3D butterfly under an alternating magnetic field

V6: Shape recovery of a deformed 3D fish under an alternating magnetic field

V7: Shape recovery of a deformed 3D porous scaffold under an alternating magnetic field

V8: Restrictive shape recovery of deformed shape placed inside a tube under an alternating magnetic field

V9: Magnetic guidance of deformed shape inside a glass tube with bar magnets

V10: Partial shape recovery of a dual-printed star under an alternating magnetic field

V11: Partial shape recovery of a dual-printed cross under an alternating magnetic field

V12: Partial shape recovery of a dual-printed butterfly under an alternating magnetic field

V13: Partial shape recovery of a dual-printed petal under an alternating magnetic field