Electronic Supplementary Material (ESI) for Materials Horizons.

Supporting Information

Mechanically Triggered Composite Stiffness Tuning Through Thermodynamic Relaxation (ST3R)

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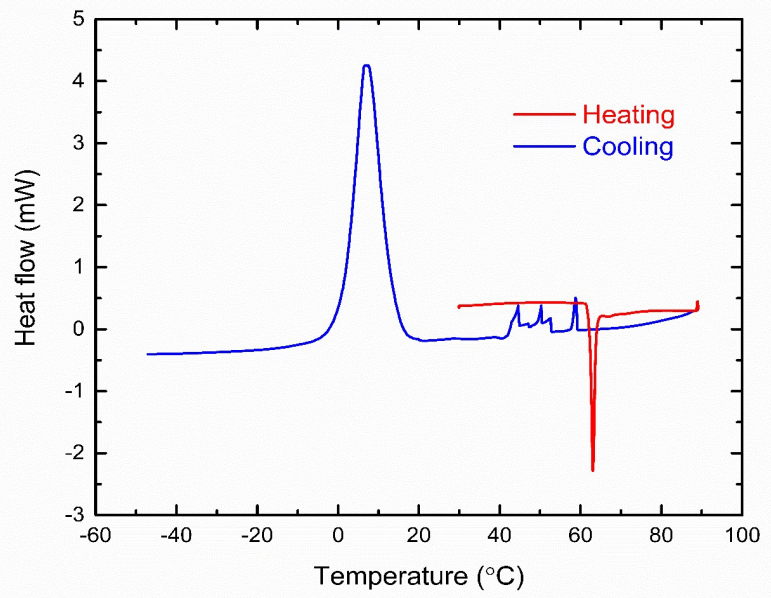

Figure S1. Differential scanning calorimetry (DSC) of Field's metal undercooled particles. Smaller freezing peaks observed between 40-60°C correspond to solidified particles, in agreement with the melting peak from the heating curve.

Estimation of heat dissipation during solidification

Heat conduction based on Fourier's law is given by:[1]

$$\frac{Q}{t} = \frac{kA(T_H - T_C)}{d}$$

Q is the heat transferred, t is time, k is thermal conductivity, A is the cross-sectional area, T is temperature and d is the length between the hot and cold end.

Considering a ST3R (50% loaded) test strip (4 x 0.5 x 0.1 cm) and assuming d as thickness (0.001 m), k for PDMS (0.2 W/m.K)[2] and A to be the total surface area between particles and matrix (0.03 m^2, estimated assuming 20 micron diamater particles), the time taken for latent heat dissipation is calculated. Differential scanning calorimetry of Field's metal identifies the latent heat of solidification to be 25 J/g and each test strip containts 0.8 g of particles. Thus, re-writing the equation for heat tranfer from particle (62°C) to surroundings (25°C):

$$t = \frac{Qd}{kA(T_H - T_C)}$$

The estimated dissipation time is 0.1 s. The overall change in temperature during phase change would be mostly unnoticeable and should have minimal effect on the mechanical properties of the composite.

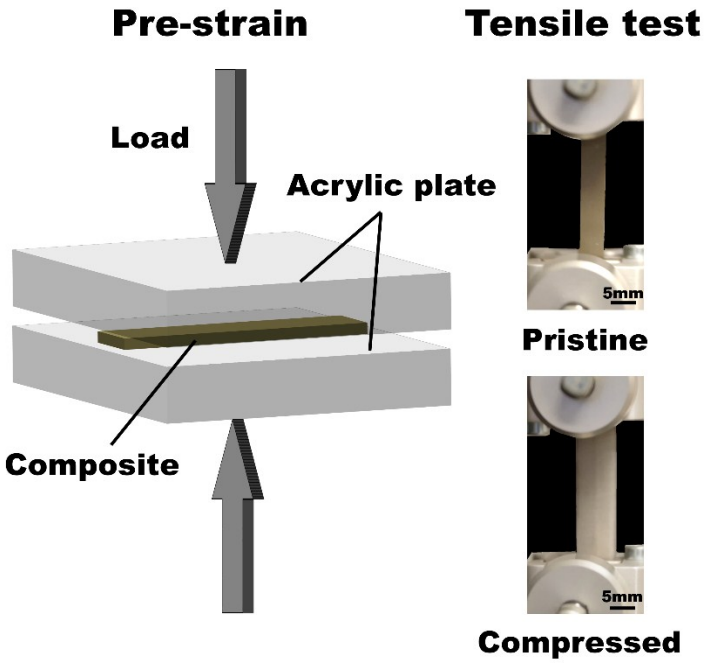

Figure S2. Schematic for pre-compresion of the composites. The composite is placed at the center of 2 arylic plates (5x10 cm). The setup is then compressed using a hydraulic press at the desired loading.

It was observed that the ST3 composites decrease in thickness and increases in width upon compression. In a typical sample, ϕ = 50% showed decrease in thickness from 1.23 to 0.93 mm and increase in width from 4.94 to 7.76 mm. Conversely, solid FM composite showed smaller changes in thicknesss (1.48 to 1.39 mm) and width (5.06 to 6.37 mm). The width of compressed samples are an average of 4 readings along the length.

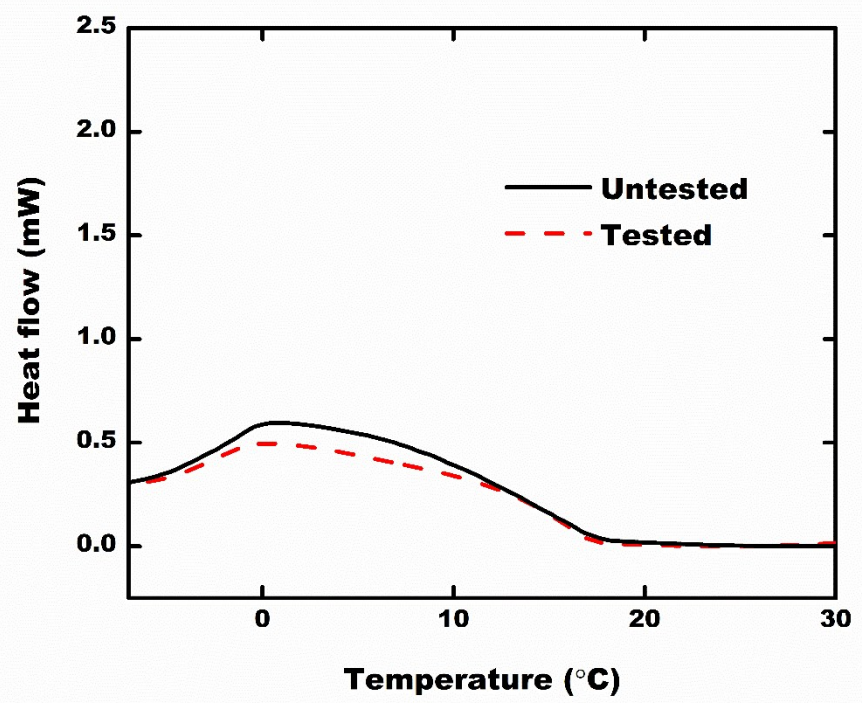

Figure S3. DSC cooling curve of FM undercooled composite before and after tensile testing up to 80% strain. 14% reduction in the solidification peak area was measured.

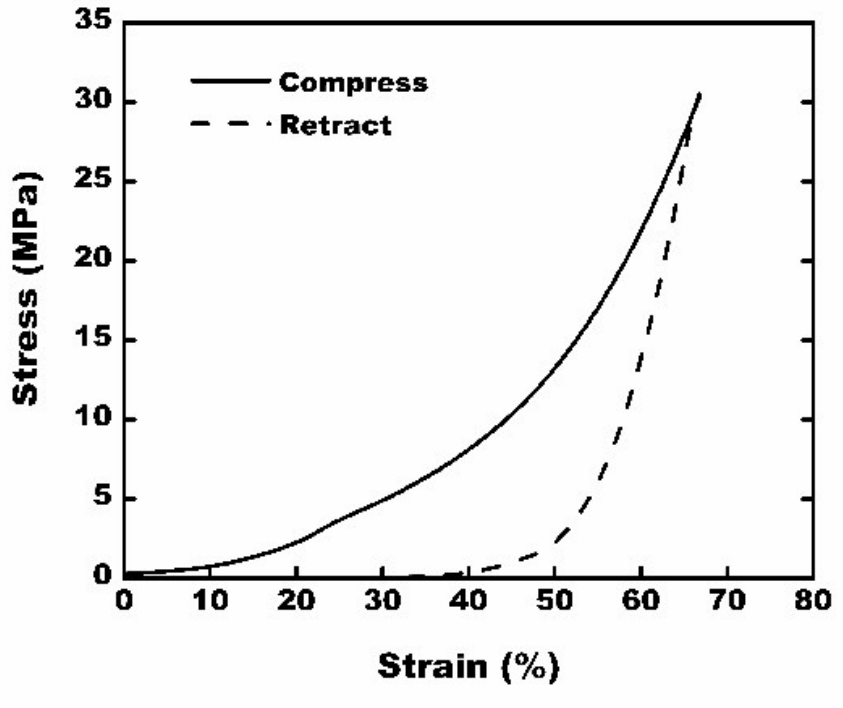

Figure S4. Compression test on ST3R (50%) rectangular strips. 68% strain resulted in 30% permanent deformation, thus, a spring-back of 38% was observed.

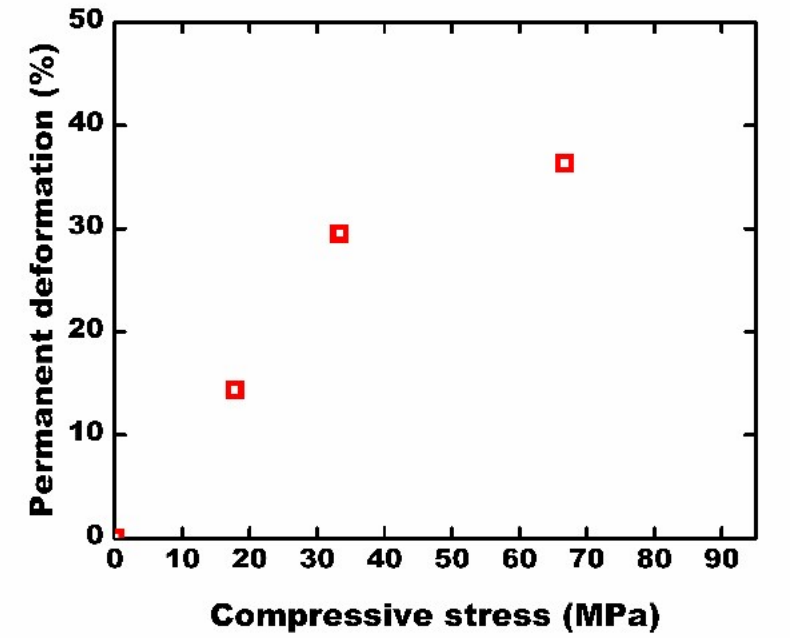

Figure S5. Permanent deformation (change in thickness) of ST3 composite as a result of uni-axial pre-compression to different prescbired compressive stresses.

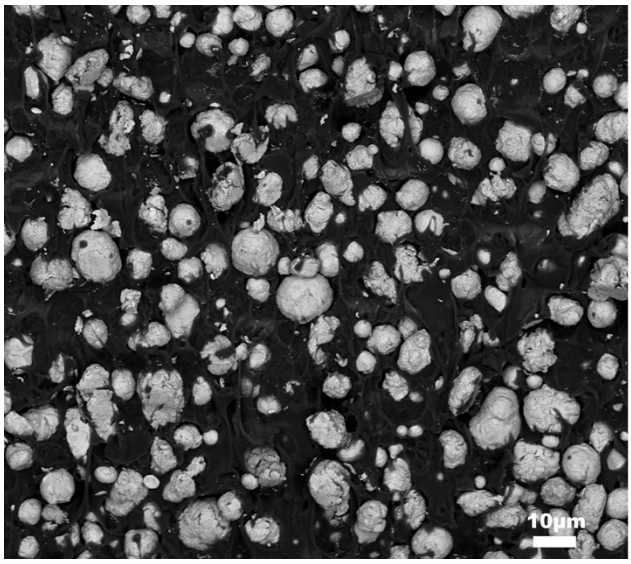

Figure S6. SEM micrograph of solid FM composite (ϕ = 50%) compressed at 33 MPa. The shape of the particles are not significantly changed as observed in the ST3R composite.

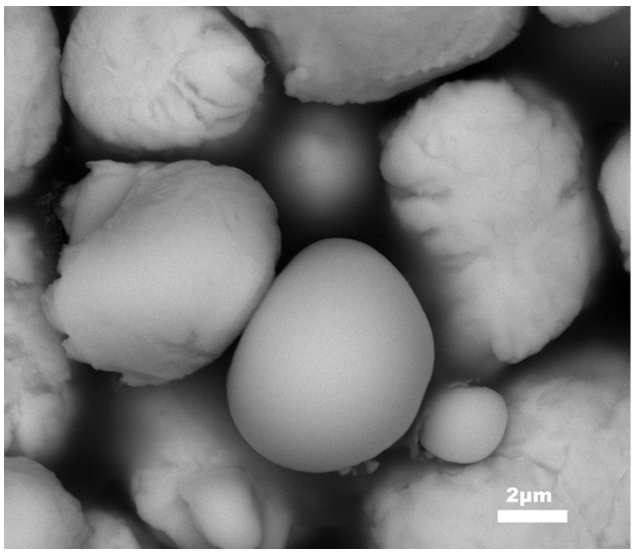

Figure S7. SEM micrograph of pristine ST3R composite at an accelerating voltage of 30 kV.

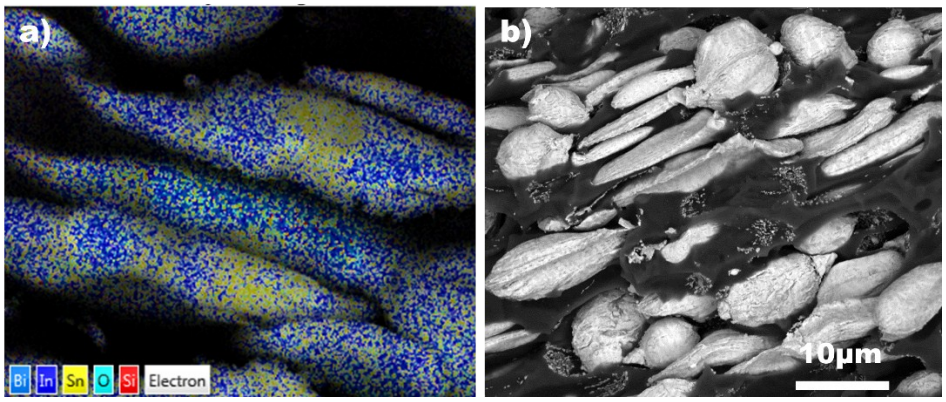

Figure S8. a) Energy dispersive x-ray spectroscopy dot map of compressed ST3R composite showing spinodal decomposition. b) SEM micrograph showing interconnects after compression.

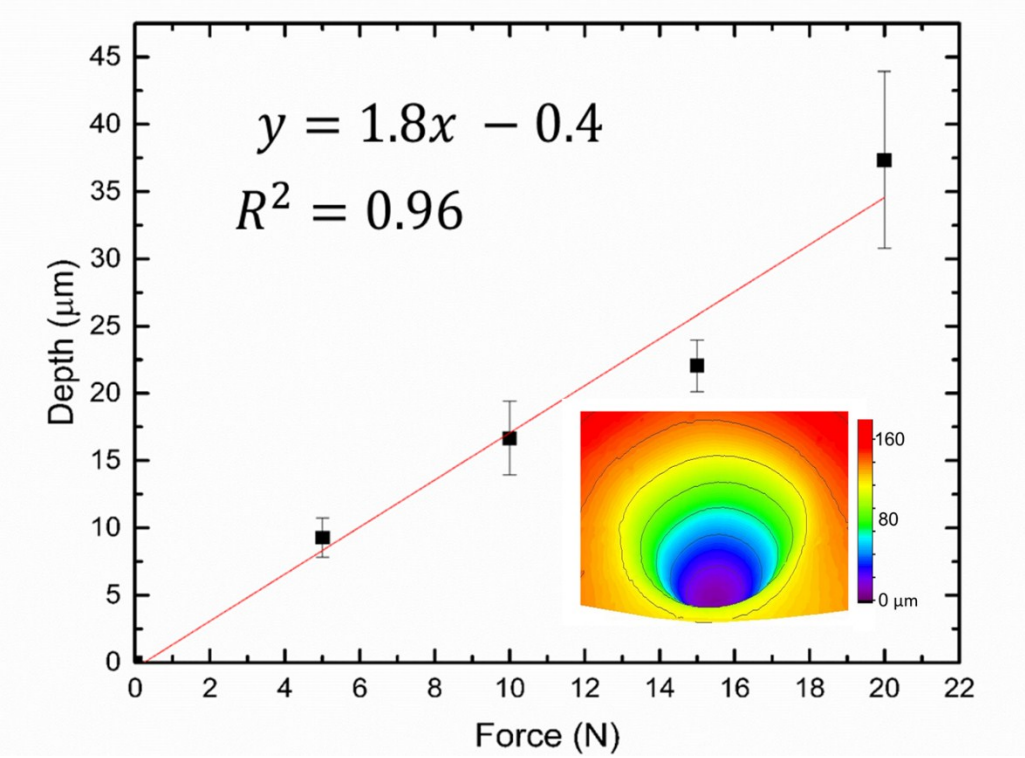

Figure S9. Well depth as a function of indentation force.

Indentations on the composites were performed using an Instron 5569 equiped with a conical tip in compression mode. Depth profiles of indentations are collected using an Alicona Infinite Focus Microscpe (IFM), a contactless surface profilometer.

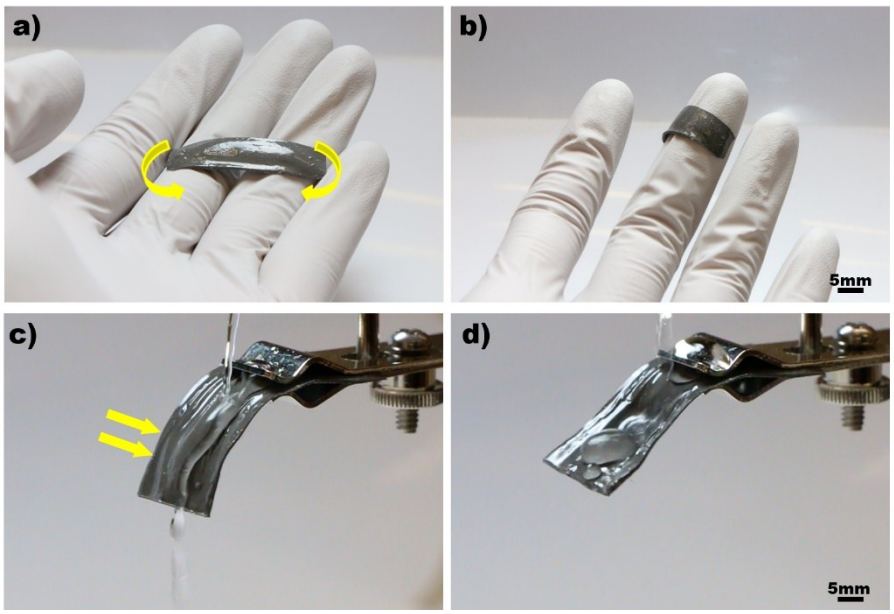

Figure S10. Additional demonstrations on shape reconfiguration. a-b) Folding the ST3 composite results in a gripping material. c-d) A flat composite is reconfigured into a spoon-like shape by embossing at a point midway along the length. This allows the composite to retain its new shape even under a stream of flowing liquid. The shape also holds the fluid akin to a spoon.

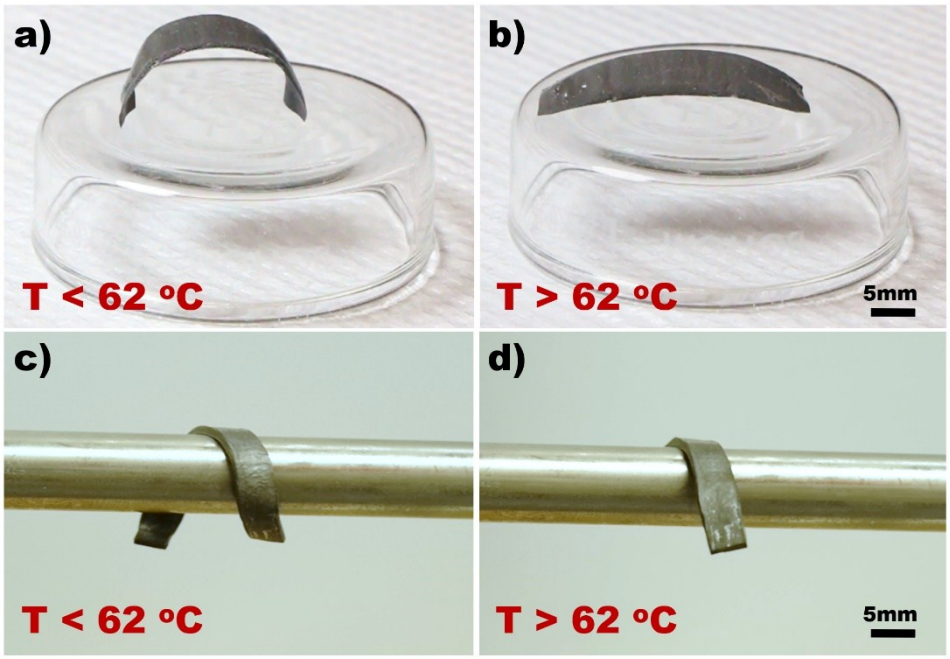

Figure 11. Shaped ST3 Elastomer matrix relaxes due to melting of the metal filler.

1. F. P. Incropera, D. P. DeWitt, T. L. Bergmann and A. S. Lavine, *Fundamentals of Heat and Mass Transfer*, John Wiley and Sons, Inc, 6th edn., 2007.
2. M. D. Bartlett, N. Kazem, M. J. Powell-Palm, X. Huang, W. Sun, J. A. Malen and C. Majidi, *Proceedings of the National Academy of Sciences*, 2017, 201616377.