Supporting Information

Ultratough Nacre-inspired Epoxy-Graphene Composites with Shape Memory Property

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Figure S1. Characterization of GO sheets. (a) SEM image of GO scattered on the surface of silicon. (b) The normal distribution of size of GO sheets.



Figure S2. The large-scale lamellar microstructures of a cross-section of scaffolds. (a) GO scaffold. (b) After heating to 800 °C, the space between the layers narrows in the rGO scaffold.



Figure S3. XPS of scaffolds before and after reduction. (a) GO scaffold contains C-C, C-O and C(O)O groups. (b) After thermal reduction, there is only C-C group in XPS curves.



Figure S4. The rGO scaffold shows anisotropic structure. (a) Lamellar structure of crosssection and (b) longitudinal section, (c) a large lamella on the side.



Figure S5. TGA curves of rGO, EP, and E-G-II. The residue of rGO is \sim 84.7 wt% and the curve of E-G-II is close to that of EP at 800 °C.



Figure S6. The fracture surfaces of EP and E-G-H, and nacre-like E-G composites. (a) The surface of EP is flat. (b) There are many stripes on the surface of E-G-H. (c) The layered structure of E-G-I and (d) aligned lamellar structure of E-G-II composite.



Figure S7. The stress-strain curves of (a) EP and (b) E-G-II perpendicular to the lamellar direction without notch upon three-point bending.



Figure S8. The electrical conductivity of E-G composites. The E-G composites show anisotropic conductivity. The conductivity of E-G-II composite is much higher than that of E-G-I composite.



Figure S9. The E-G-II composite in the perpendicular direction also can be fixed into various shapes such as a (a) circle, (b) twist and recover to their original shapes by heating or current.



Figure S10. (a) DSC curve and (b) DMA curves of E-G-II composite.



Figure S11. The consecutive shape memory cycles of (a) EP and (b) E-G-II composite.

| Nanofiller | Abbreviation |
|--|--------------------------|
| Epoxy-graphene oxide | E-GO ⁵³ |
| Epoxy-(3-glycidoxypropyl) trimethoxysilane graphene oxide | E-GPTS-GO ⁵⁴ |
| Epoxy-silane-functionalized graphene oxide | E-s-GO ⁵⁵ |
| Epoxy-reduction graphene oxide | E-RGO ⁵⁶ |
| Epoxy-graphene nanoplatelets | E-GPL ⁵⁷ |
| Epoxy-graphene platelets | E-GP ⁵⁸ |
| Epoxy-graphene platelets-polyoxypropylene | E-G-J230 ⁵⁹ |
| Epoxy-amine functionalized expanded graphene nanoplatelets | E-EGNP ⁶⁰ |
| Epoxy-graphene foam | E-GF ⁶¹ |
| Epoxy-carbon nanofibers | E-CNFs ⁶² |
| Epoxy-pristine carbon nanofibers | E-P-CNFs ⁶³ |
| Epoxy-polydopamine carbon nanofibers | E-D-CNFs ⁶³ |
| Epoxy-multiwalled carbon nanotube fibers | E-MWCNT ⁶⁴ |
| Epoxy-clay | E-clay ⁶⁵ |
| Epoxy-jeffamine XJT502 modified clay | E-Xjt-clay ⁶⁶ |

Table S1. Corresponding abbreviations of specific nanofillers