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Supporting Information

A mechanically and electrically self-healing graphite composite dough for stencil-printable stretchable conductors

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Experimental Section

Materials Synthesis: Graphite flakes (size < 45 μm) and branched polyethylenimine (bPEI, Mw ≈ 25,000, density = 1.030 g cm^{-3} at 25 °C) were purchased from Sigma-Aldrich (UK). 3.5 g of bPEI was dissolved in 20 mL ethanol with gentle mechanical stirring and then 6.5 g of graphite was added. The stirring was kept until the complete evaporation of the solvent. The graphite/bPEI mixture was transferred into a grinding bowl. The graphite/bPEI composite dough was obtained after manually grinding for 30 min until a uniform and compact mixture was formed.

Materials Characterization: The microstructure of the graphite/bPEI composite dough was investigated by scanning electron microscopy (SEM) (Inspect F, FEI), imaged using a 10 keV acceleration voltage. Rheological measurements of bPEI and graphite/bPEI composite dough were carried out on an ARES-G2 rheometer (TA Instruments, Delaware) with a parallel-plate fixture (25 mm diameter) and a dynamic frequency sweep mode. The storage and loss moduli as a function of shear rate (ranging from 0.1 to 100 rad s^{-1}) were measured at room temperature with 1-mm thick samples and a constant shear amplitude of 1%to keep the strain in the linear viscoelastic region. The electrical properties of the sample were monitored in two-point via a benchtop multimeter (Agilent 34401A, Keysight Technologies Inc.). For the conductivity test, a cubic sample (1 cm × 1 cm × 1 cm) was used and contacted to the circuit via aluminum electrodes. Tensile and compression tests were carried out using a Lloyd universal testing machine (Ametek Inc.). Samples for the tensile tests were cut into a rectangular shape (50 mm × 2 mm × 2 mm). A 10 N load cell was used with a speed of 50 mm min^{-1} and the distance between the sample grips was 30 mm. Five specimens were tested for each measurement. The graphite/bPEI/thermoplastic polyurethane (TPU) conductor was constructed through stencil print by using a 100 μm-thick Teflon sheet with a designed open (e.g. rectangle) as a mask. Briefly, the mask was placed on a TPU sheet, and graphite/bPEI was filled into the designed open. The leftover was scratched with a scalpel to make the surface of graphite/bPEI parallel to the mask. The graphite/bPEI/TPU conductor with the designed element was obtained after removing the mask. The volume percentage of graphite was calculated using the following equation (1).

$$vol\% = (w_{graphite}/\rho_{graphite})/(w_{graphite}/\rho_{graphite} + w_{bPEI}/\rho_{bPEI}) \times 100\% \quad (1)$$

where w is the weight of the component and ρ is the density. $\rho_{graphite} = 2.26$ g cm^{-3}.[1]

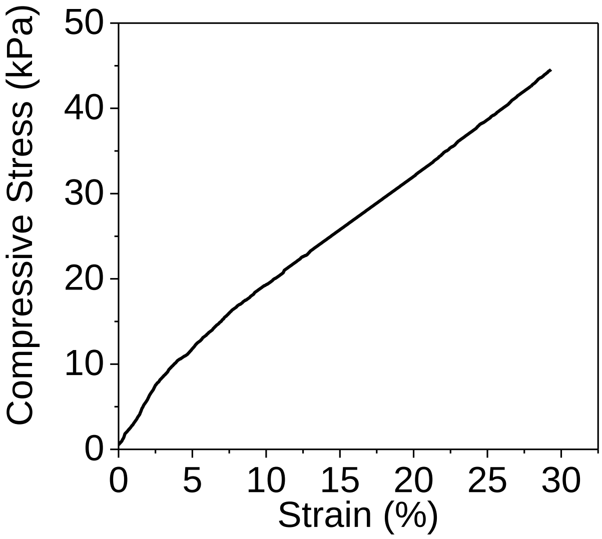

Fig. S1. Typical compressive stress-strain curve of the graphite/bPEI composite dough at room temperature.

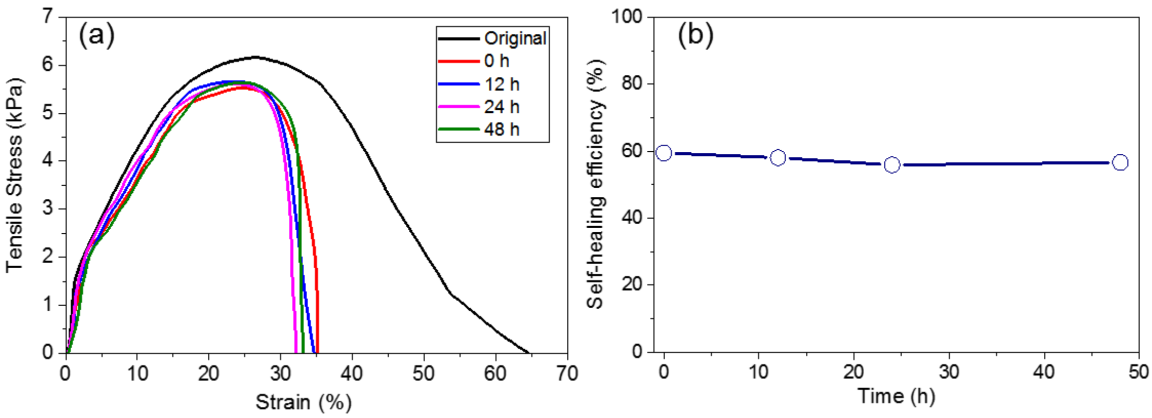

Fig. S2. a) Typical tensile stress-strain curves of original and healed samples. b) The healing efficiency of the graphite/bPEI composite dough as a function of the time period to keep the two freshly fractured surfaces apart before the self-healing process takes place. To heal the samples, the two fractured halves were pressed together by applying a gentle pressure of ~1 kPa for 2 s, and then allowed to stand still for over 8 s for further healing.

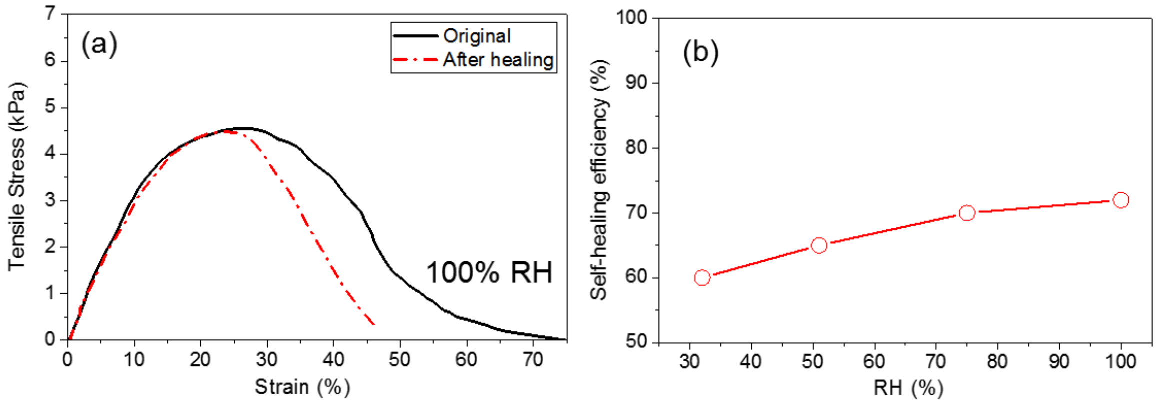

Fig. S3. The humidity dependence of self-healing. (a) Typical tensile stress-strain curves of original and healed samples at room temperature (20 ^{o}C) under 100% relative humidity (RH) and (b) the healing efficiency of the graphite/bPEI composite dough as a function of relative humidity. To heal the samples, the two fractured halves were pressed together by applying a gentle pressure of ~1 kPa for 2 s, and then allowed to stand still for over 8 s for further healing. To investigate the influence of humidity, the sample was left in constant RH at 20 ^{o}C for 16 h prior to each test: $MgCl_2$ saturated solution (SS) for 32% RH, Mg $(NO_3)_2$ SS for 51% RH, NaCl SS for 75% RH, and deionized water for 100% RH (saturated water vapor).[2]

References

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2. J. F. Young, J. Appl. Chem., 1967, **17**, 241-245.