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Supporting Information for Manuscript Entitled with

Conductive Microcapsules for Self-healing Electric Circuits

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Experimental Section

Raw Materials**:** Urea (99%, Showa, Japan) and formaldehyde (37%, Echo, Taiwan) are the primary chemical reagents for the synthesis of shell wall material of microcapsules. Resorcinol (98%, Acros, USA), ammonium chloride (99.5%, Showa, Japan) and triethanolamine (99%, Sigma-Aldrich, USA) were used as additives for synthesis. Eicosane (99%, Acros, USA) was used as the core material for the synthesized microcapsules. The surfactant used for encapsulation was poly(ethylene-alt-maleic anhydride) (EMA; 99%, Sigma-Aldrich, USA). Gum arabic (99%, Parchem, USA) was used as the dispersant and silver nitrate ($AgNO_3$; 99%, Solar Technology, Taiwan), glucose (99.5%, Sigma-Aldrich, USA) and ammonia solution ($NH_4OH_{(aq)}$; 30%, Showa, Japan) were used as reagents for the surface coating of as-synthesized microcapsules. All chemical reagents mentioned above were used as-received.

Synthesis of Poly(urea-co-formaldehyde) (PUF) Microcapsules**:** The technique for encapsulation was in-situ polymerization. As the first step, the additives of 0.25 g resorcinol and 0.25 g ammonium chloride were mixed with 2.5 g urea and dissolved in 100 ml de-ionized water. This aqueous solution was then warmed to 40 ^oC and the pH was adjusted to 3.5 by the addition of triethanolamine. The melted 5.5 ml eicosane was dropped into the aqueous solution and emulsified using a homogenizer (T25, IKA, Germany) with a speed of 13,000 rpm for 5 min. After homogenization, 6.33 g formaldehyde was instantly added and the emulsion was heated to 55 ^oC and stirred on a hot-plate for an additional 2 hrs. After the reaction, microcapsules were removed from the suspension and washed repeatedly using de-ionized water.

Surface Coating on PUF Microcapsules: 0.30 g of the microcapsules were re-dispersed into 70 ml de-ionized water by using 0.35 g gum arabic as the dispersant. After stirring for 30 min, the suspension of microcapsules was mixed with an aqueous solution of 0.35 g $AgNO_3$ and 0.6 ml $NH_4OH_{(aq)}$ for another 30 min. The suspension was set under constant 4 ^{o}C and slowly mixed with 20 ml aqueous solution of 0.40 g glucose and 0.17 g NaOH. The mixture was kept stirring at 400 rpm for 2 hrs for the reduction of Ag^+ on the surface of microcapsules.

Characterizations: The microstructures were characterized by field emission scanning electron microscopy (FE-SEM; S-470, Hitachi, Tokyo, Japan). The surface chemistry of microcapsules was characterized through the electroacoustic method (ZetaProbe, Colloidal Dynamics Inc., MA, USA) by measuring the zeta-potential of the aqueous suspensions of microcapsules with a solid loading of 0.5 wt%. The thermal properties of microcapsules were analyzed by thermogravimetric analysis (TG; Q50, TA instruments Ltd., Crawley, UK). Silver based circuits were prepared by blending 20 vol% of various microcapsules with conductive silver paste (product no. 16031) that was purchased from TED PELLA, Inc. The silver paste blend was casted by a micrometer-controlled doctor blade on a regular glass and dried under room temperature for 24 hrs, and then the electrical properties were measured on a power supplier (PST-3202, GW Instek, Taiwan).

Additional Results

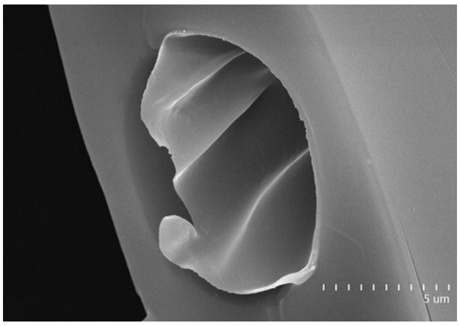

Figure S1 Cross-sectional SEM image of a broken PUF-C20 microcapsule embedded in resin (core eicosane has been dissolved by solvent), showing a shell thickness of about 50-100 nm.

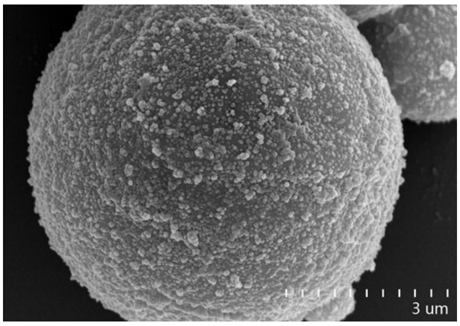

Figure S2 High resolution SEM image, showing the size of the surface-coated Ag nanoparticles is about 50 nm.

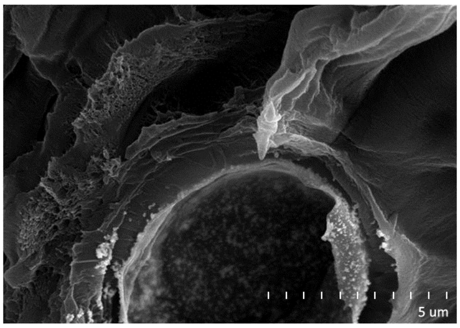

Figure S3 Cross-sectional SEM image of a broken Ag@*m*PUF-C20 microcapsule embedded in resin, showing the thickness of the shell with coated Ag nanoparticles is about 150 nm.

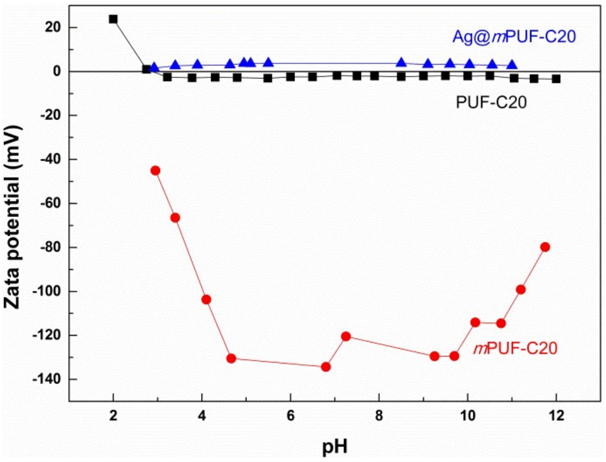

Figure S4 Zeta potentials of various microcapsules in aqueous suspensions with a solid loading of 0.5 wt%. The zeta potentials of PUF-C20 are primarily within 0 mV and -5 mV and are insensitive to the pH. With the addition of 2.5 wt% (based upon the weight of PUF-C20) gum arabic, the zeta potentials of the gum arabic adsorbed PUF-C20 (*m*PUF-C20) significantly shifted to large magnitude negative values. After reduction of Ag on the surface of *m*PUF-C20, the zeta potential shifted to + 3 mV, with low positive values resembling the characteristic surface chemistry of metallic Ag.

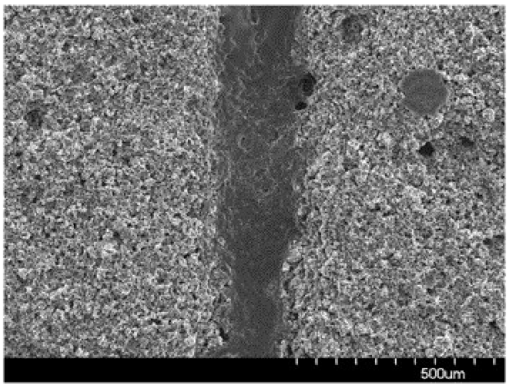

Figure S5 SEM image of the recovered zone viewed from the top surface of the cast Ag circuit. The solid loading of the PUF-C20 microcapsule embedded in the Ag matrix was 20 vol%. The valley of the crack was filled with the core material, eicosane.

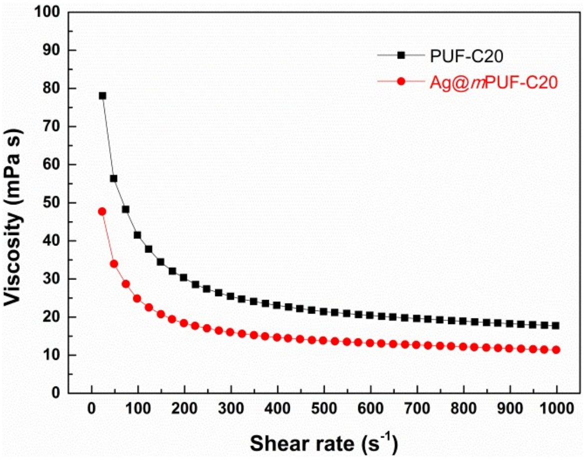

Figure S6 Viscosities as the functions of shear rates for the two Ag pastes with respective additions of 20 vol% PUF-C20 and Ag@*m*PUF-C20. By further analyzing the two rheological curves based on power law, the flow indices (*n*) which indicates the degree of deviation from the well-dispersed Newtonian behavior (*n* = 1) were 0.85 and 0.89 for the two Ag pastes with containing 20 vol% PUF-C20 and Ag@*m*PUF-C20, respectively. This result suggests that the agglomeration of PUF-C20 microcapsules is more severe than that of Ag@mPUF-C20 microcapsules in the Ag paste.