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Increasing the breakdown strength of dielectric actuators by using Cu/Cu_xO /silicone dielectric elastomer

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Experimental section

Chemicals

Cu particles were purchased from Ningbo Guangbo New Nanomaterials Stock Co., Ltd, China. Tetrahydrofuran and NaCl were purchased from Sinopharm Chemical Reagent, China. Polydimethylsiloxane (PDMS) (RTV-3483), Curing agent (RTV-3083), and compliant electrode material (Molykote HP-800 Grease) were purchased from Dow Corning Corporation.

Materials and general measurements

The crystal structure and composition were characterized by powder XRD using a German Bruker AXS D8 ADVANCE X-ray diffractometer. X-ray photoelectron spectroscopy (XPS) analyses were carried out with a Thermo ESCALAB 250 XPS system with Al K_α radiation. The binding energies were corrected by referencing the C 1s peak to achieve charge neutralization. Scanning electronic microscopy (SEM) images of the Cu, Cu/Cu_xO, and the fractured surface of the composites were taken on a Japan Hitachi S-4800 field emission SEM. The microstructure of Cu/Cu_xO core-shell particles was characterized by high resolution canning electronic microscopy (HRTEM) (American FEI Tecnai G2 F30 S-TWIN field-emission transmission electron microscopy).

The elastic modulus and dielectric properties of elastomers were characterized by dynamic mechanical analyzer analyzer (DMA, Q800, TA, USA) and a broadband dielectric analyzer (Novocontrol Technologies, Germany), respectively. Differential Scanning Calorimetry (DSC, Q2000, TA, USA) experiments were performed from -90°C to 50 °C at a heating and cooling speed of 10 °C/min. Thermogravimetric analyses (SDT 2960 thermal analyzer) were performed from 30 to 800 °C at a heating rate of 10 °C/min under N_2 atmosphere. The rheological measurements were carried out on a

HAAKE RheoStress 6000 rheometer. Dynamic oscillatory strain sweeps were measured at *f* = 1 Hz. Frequency sweeps of DEs with 1% strain amplitude were from 0.01 to 1000 rad/s at 25 °C. Both sides of dielectric elastomers were coated with circular, compliant electrodes (Molykote HP-800 Grease). The actuated strain of dielectric elastomer was immediately obtained when electric field was applied by an intelligent DC high voltage generator. The change of pixels of the picture during deformation was captured by a commercial camera. The crosslinking density was measured by using the Magnetic Resonance Crosslink Density Spectrometer (MicroMR-CL, Niumag, China). Testing conditions: the resonance frequency was 21.8000MHz; the intensity of magnet was 0.52T; the diameter of magnet coil was 10mm; The testing temperature was 80℃.

Preparation of Cu/Cu$_x$O Particles

In a typical procedure for the synthesis of Cu/Cu$_x$O core-shell particles, 1.0 g Cu and 9.2 g of NaCl were mixed and ground homogeneously in a carnelian mortar, then transferred to a corundum crucible. The crucible was heated in air at 375 ºC for 5 mim, then air-cooled to room temperature. Finally, black Cu/Cu$_x$O core-shell particles were obtained by washed carefully with distilled water and absolute alcohol to remove the salt. In order to study the oxidation process of Cu paritcles, the temperature and time length for the thermal treatment were varied when preparing the Cu/Cu$_x$O core-shell particles.

Preparation of Cu/Cu$_x$O/PDMS Films

Initially, Cu/Cu$_x$O core-shell particles with mass fraction ranging from 0 to 30 wt% were mixed with 18 g PDMS by stirring for 20 min at room temperature. Tetrahydrofuran was used as solvent. Subsequently, 0.9 g curing agent was blended with the above solution to obtain a uniform solution. The above suspension was poured into a polytetrafluoroethene (PTFE) mould and dried in an oven at 45 ºC for one day. The thickness of composite film was adjusted to be about 1mm for test of mechanical properties. It is also necessary to prepare a 0.12 mm thick film for breakdown strength testing. Cu/PDMS films were prepared in the same procedure.

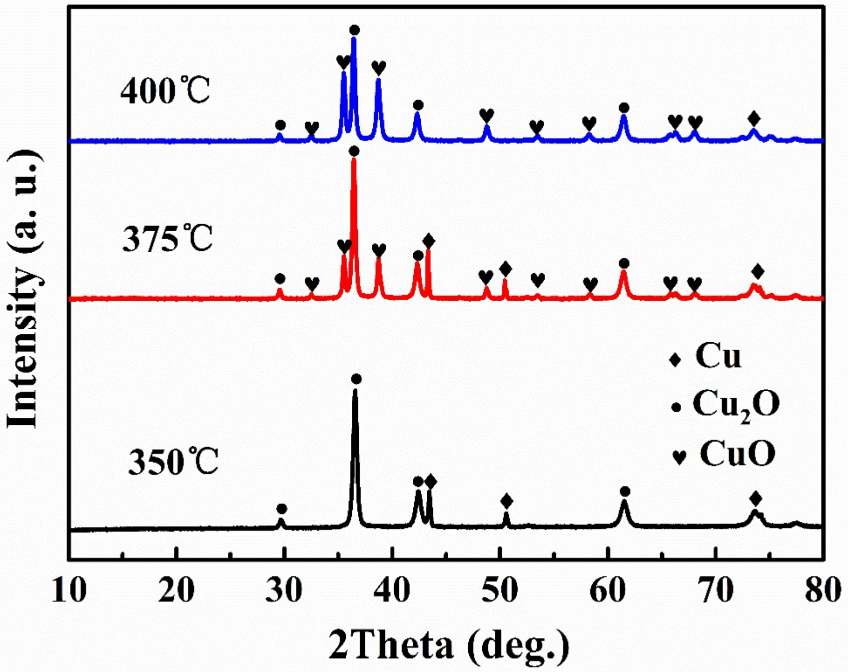

Figure S1. XRD patterns of the products of the thermal treatment of the precursor with low temperature molten salt method at 350 ºC, 375 ºC, 400 ºC for 5 min.

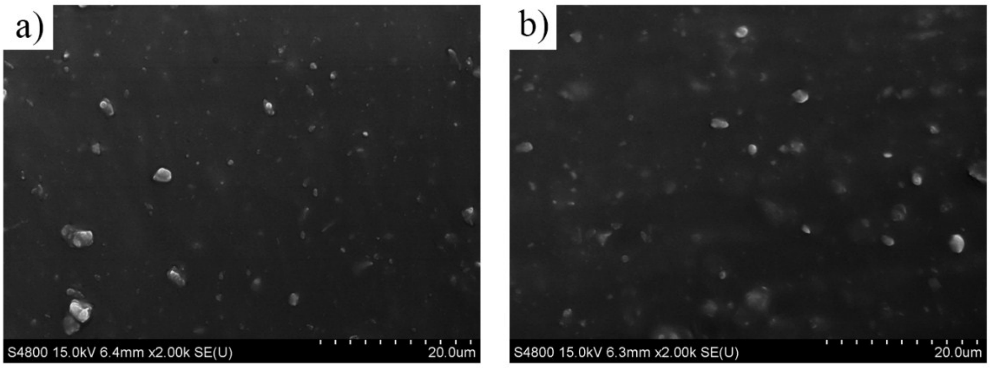

Figure S2. FESEM micrographs of the fractured surfaces of (a) Cu/PDMS film and (b)

$Cu/Cu_xO/PDMS$ film (15 wt%).

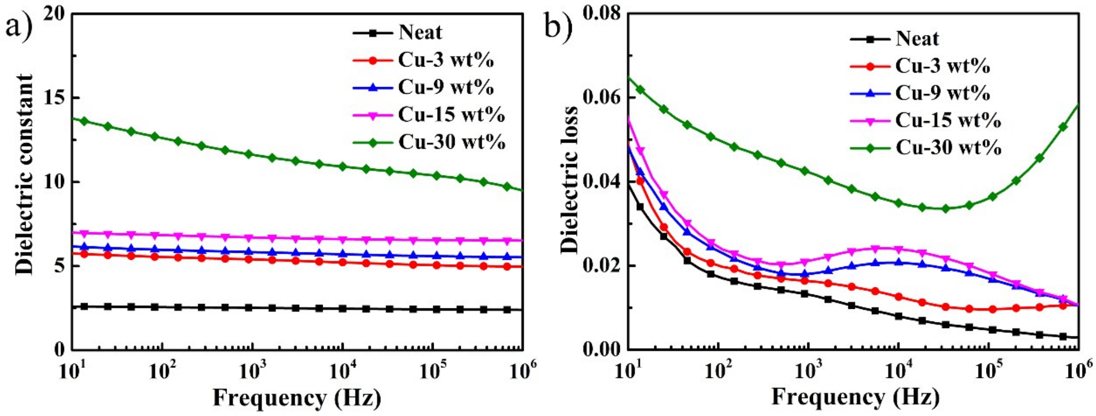

Figure S3. Frequency dependence of dielectric constant a) and dielectric loss b) of Cu/PDMS film in various weight fractions.

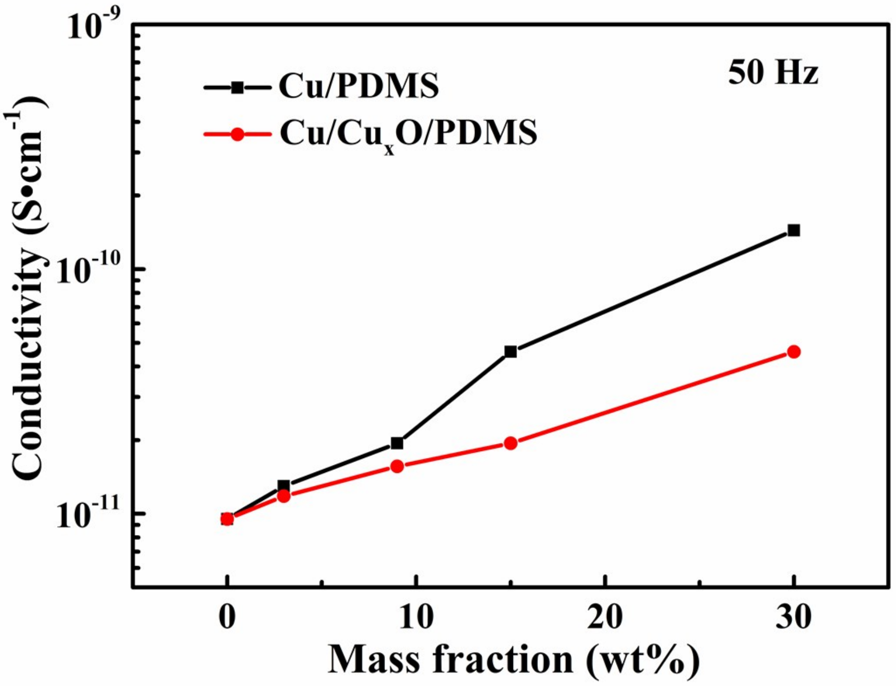

Figure S4. Dependence of the conductivity at 50 Hz of Cu/PDMS film and $Cu/Cu_xO/PDMS$ film on different mass fraction.

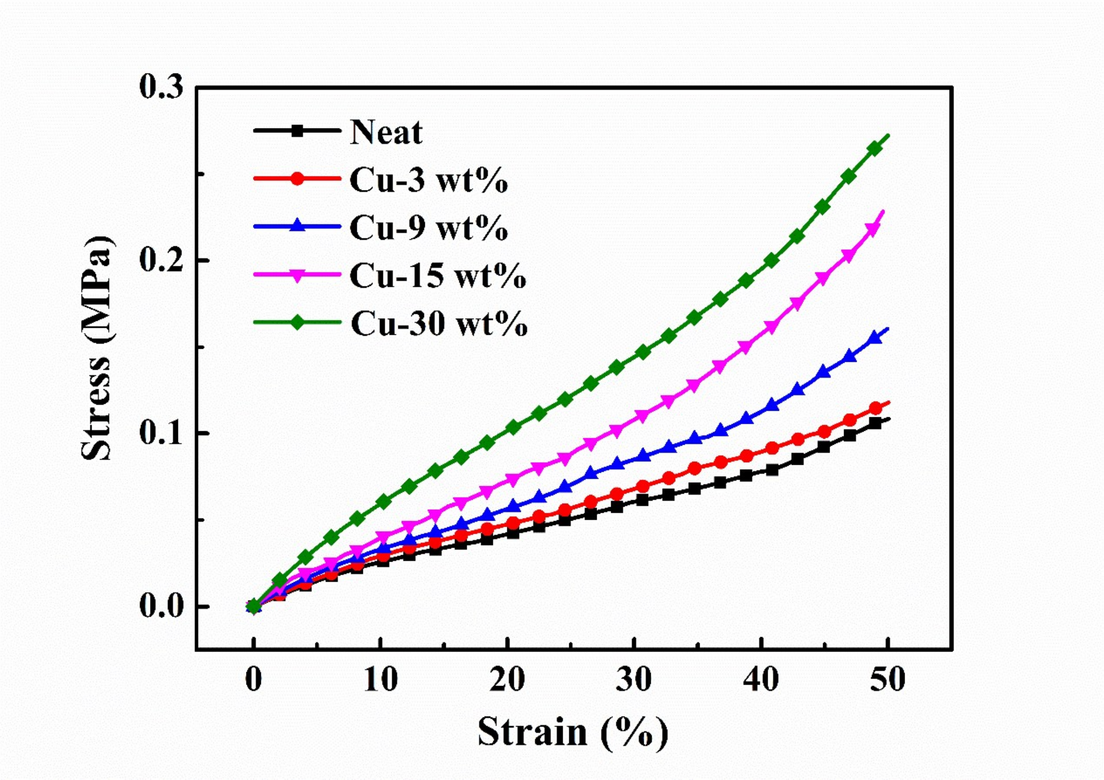

Figure S5. Stress-strain curves of Cu/PDMS film in various weight fractions.

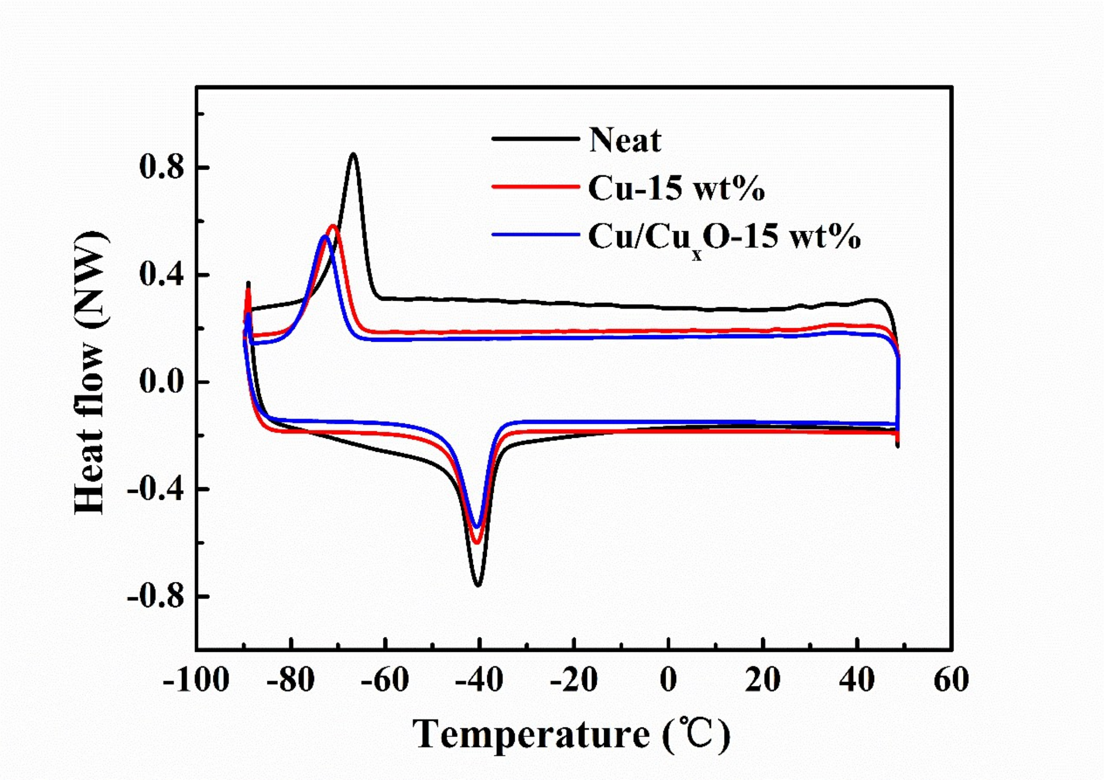

Figure S6. The DSC curves of pure silicone elastomer, Cu/PDMS and Cu/Cu$_x$O/PDMS film (15 wt%).

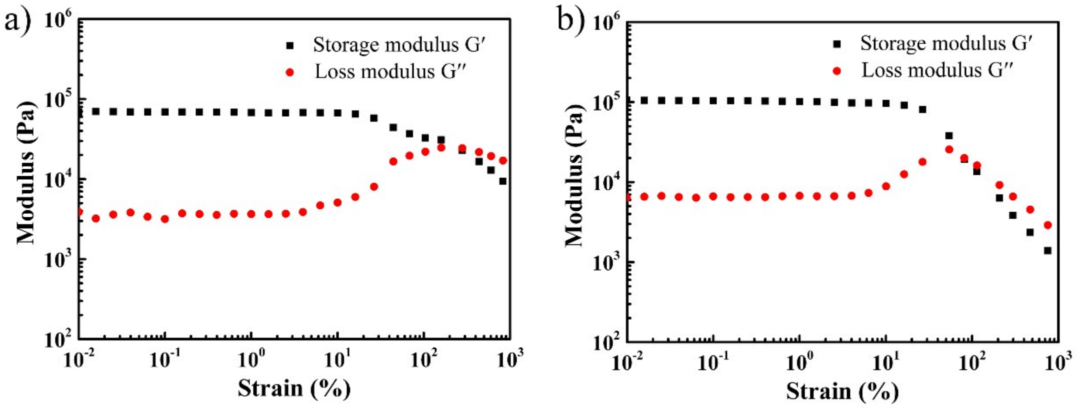

Figure S7. Rheological studies. (a) Dynamic oscillatory strain sweep of Cu/PDMS film (15 wt%) at *f* = 1 Hz. (b) Dynamic oscillatory strain sweep of Cu/Cu$_x$O/PDMS film (15 wt%) at *f* = 1 Hz.

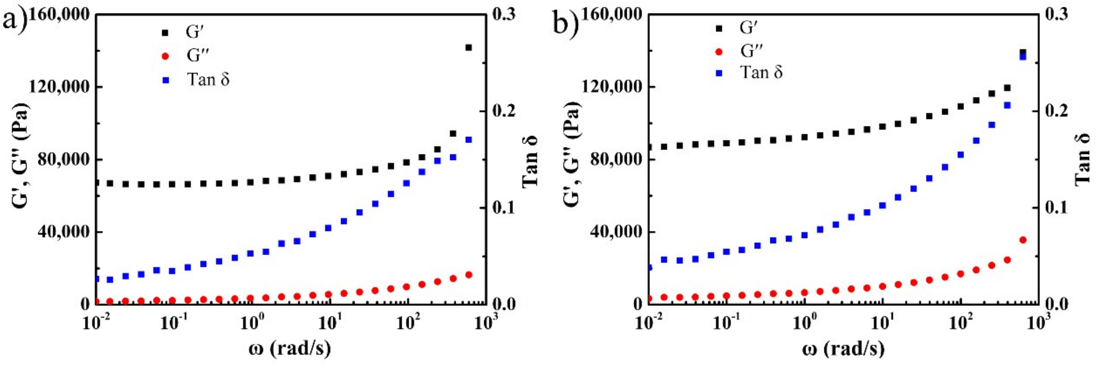

Figure S8. The rheological test. (a) Frequency sweep of Cu/PDMS film (15 wt%) with 1% strain amplitude. (b) Frequency sweep of Cu/Cu$_x$O/PDMS film (15 wt%) with 1% strain amplitude. All measurements were performed at 25 ℃.

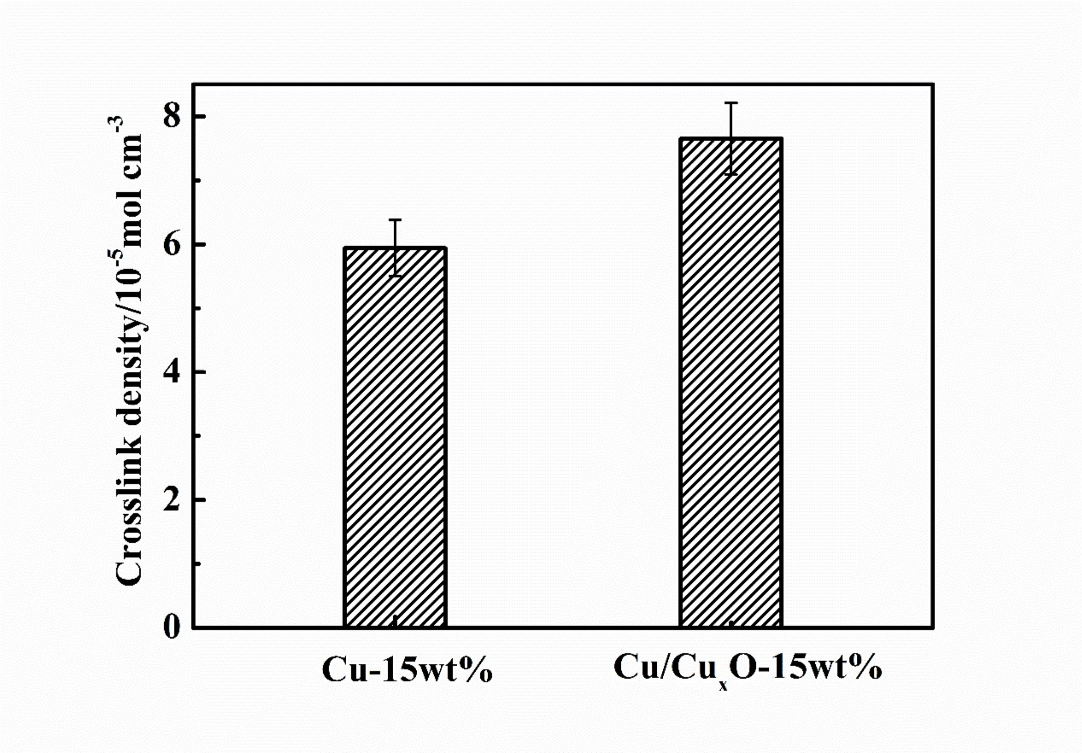

Figure S9. The crosslink densities of Cu/PDMS film and Cu/Cu$_x$O/PDMS film (15 wt%).

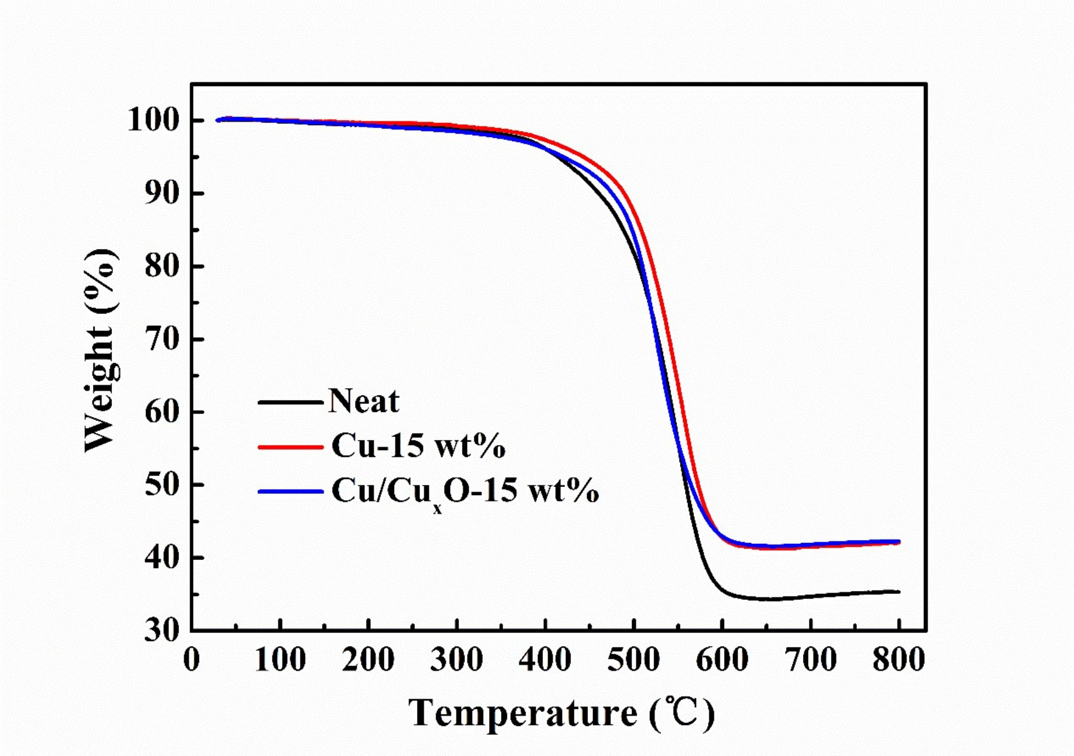

Figure S10. TGA curves of pure silicone elastomer, Cu/PDMS film and Cu/Cu$_x$O/PDMS film (15 wt%).

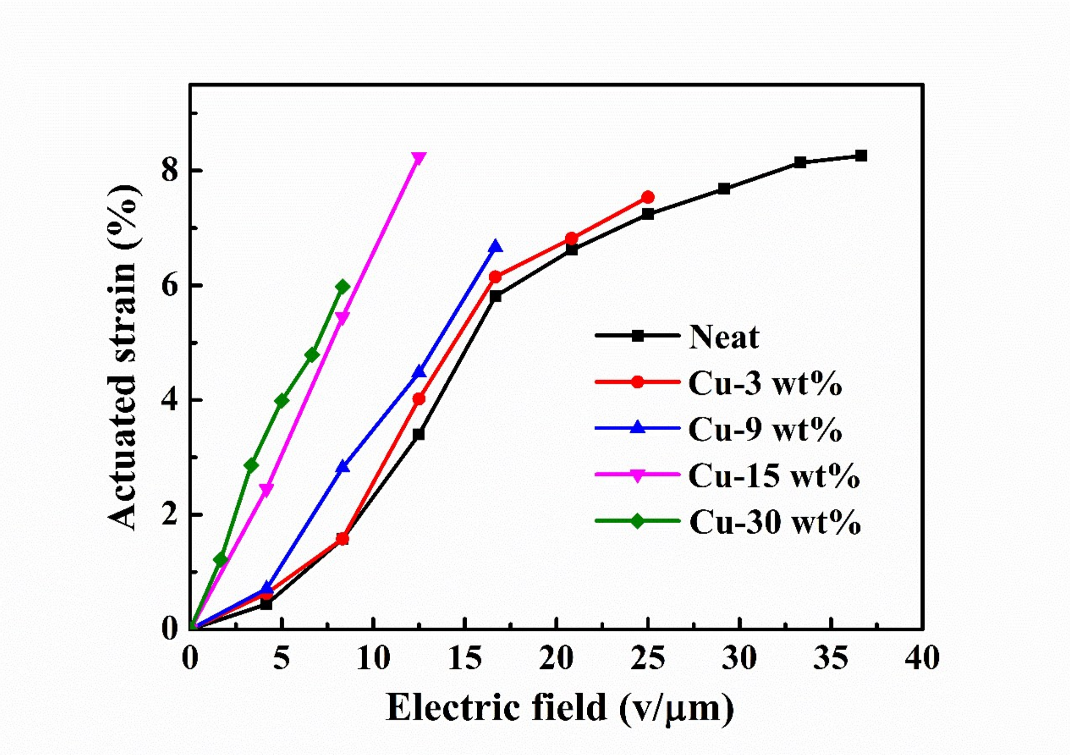

Figure S11. Actuation strain of pure silicone elastomer, Cu/PDMS film with various weight fractions in the electric field range of 0 V/μm -40 V/μm.

Table S1. The relational information of the synthesis and characterization of the pure silicone elastomer, Cu/PDMS and Cu/Cu_xO/PDMS film.

Composites	Y(MPa) 1-20%	Dielectric constant ε_r at 50 Hz	Actuated strain (%) at 12.5 V/μm	Breakdown strength (V/μm)	Maximal actuated strain (%)
Pure silicone	0.20	2.57	3.40	36.67	8.26
Cu -3 wt %	0.23	5.60	4.02	25.00	7.54
Cu-9 wt %	0.27	6.02	4.48	16.67	6.67
Cu-15 wt %	0.35	6.88	8.24	12.50	8.24
Cu-30 wt %	0.49	13.00	-	8.33	5.98
Cu/Cu_xO -3 wt %	0.35	4.96	4.86	33.33	8.24
Cu/Cu_xO -9 wt %	0.46	5.22	5.01	29.17	7.99
Cu/Cu_xO -15 wt %	0.57	5.96	5.46	29.17	8.42
Cu/Cu_xO -30 wt %	0.74	8.90	6.20	13.33	6.51