Supporting Information

Silica Nanoparticles Treated by Cold Atmospheric-Pressure Plasmas Improve the Dielectric Performance of Organic-Inorganic Nanocomposites

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This Supporting Information file contains **Figures S1-S4** which show the system configuration and the sample dimension for the insulation performance test, **Figures S5** and **S6** which demonstrate the chemical structures and reactions of the fabricated materials, **Figure S7** which reveals the surface composition of the polymer nanocomposites, **Figure S8** which shows the results of the endurance test, and **Figures S9** and **S10** which explain the theory under the experimental observations. The detailed descriptions can be found in the main text of the article.

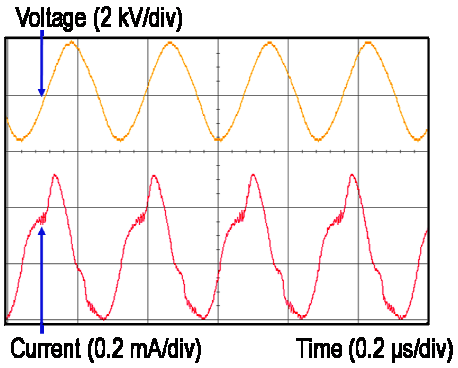

Figure S1. The voltage and current waveshapes of the plasma load. 4 kV peak-to-peak sinusoidal voltage is applied. The positive and negative peaks of the current waveform are 0.3 mA and -0.2 mA respectively.

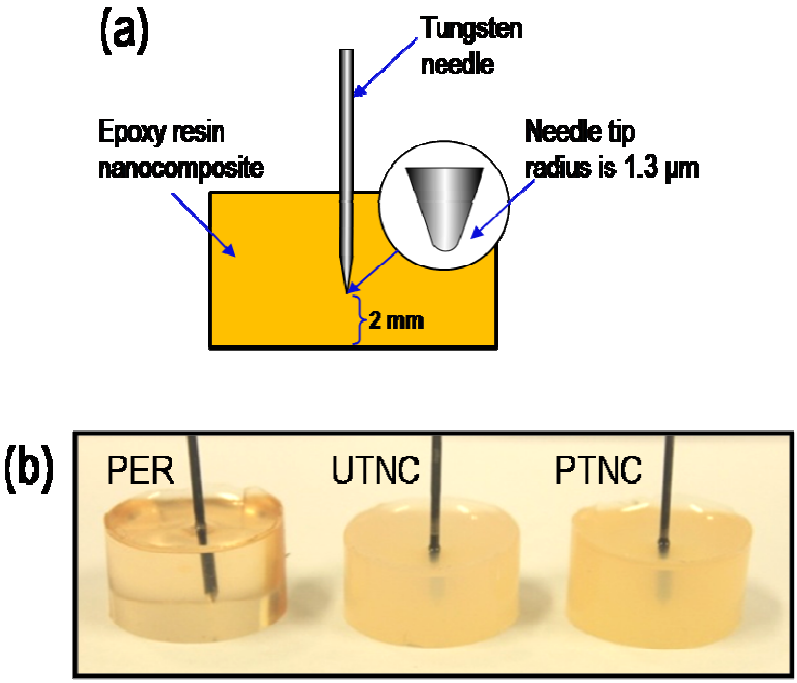

Figure S2. (a) The geometry and (b) images of molded samples for insulation tests. A tungsten needle is embedded into each sample for carrying out electrical tests.

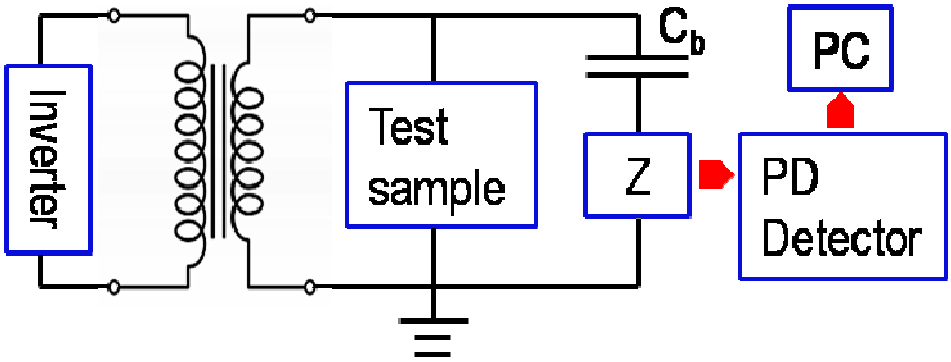

Figure S3. Partial discharge test circuit. The circuit is also applied for the ramp voltage test and the endurance test. Partial discharge activities can be simultaneously recorded and transmitted to computer.

Figure S4. Insulation-oil-filled test cell loaded with a sample. Unwanted surface discharge can be avoided, which insures that all detected discharges are created internally in the sample.

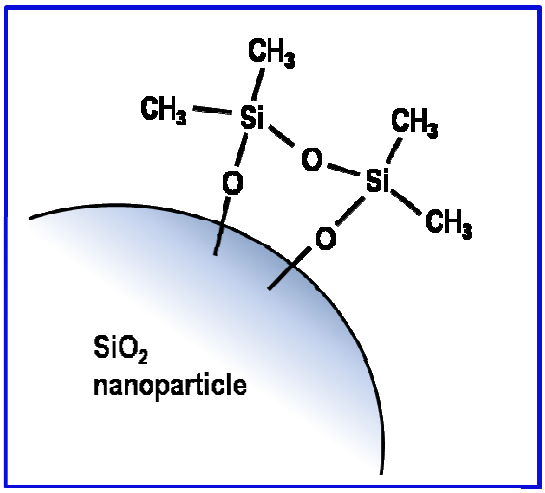

Figure S5. The surface structure of the as-received silane grafted SiO_2.

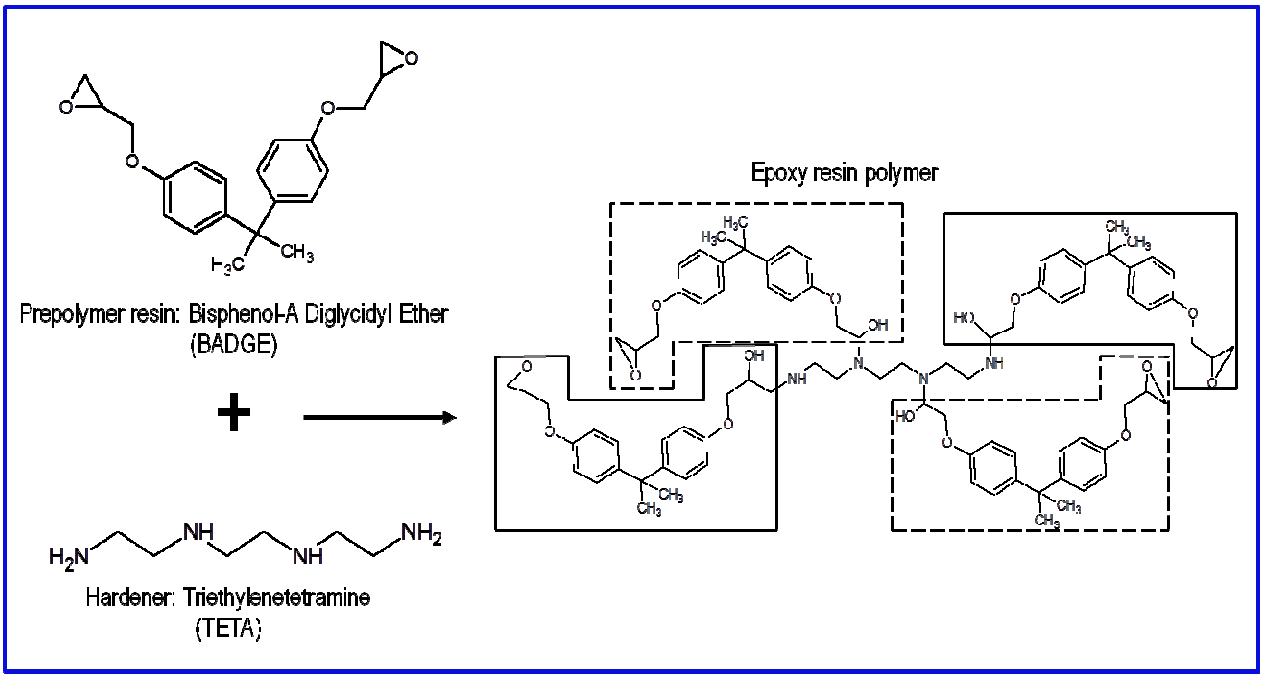

Figure S6. Reaction of the thermoset epoxy resin kit. The pre-polymer resin molecules are crosslinked through forming C-N bonds with the backbone Nitrogen atoms of the hardener molecules.

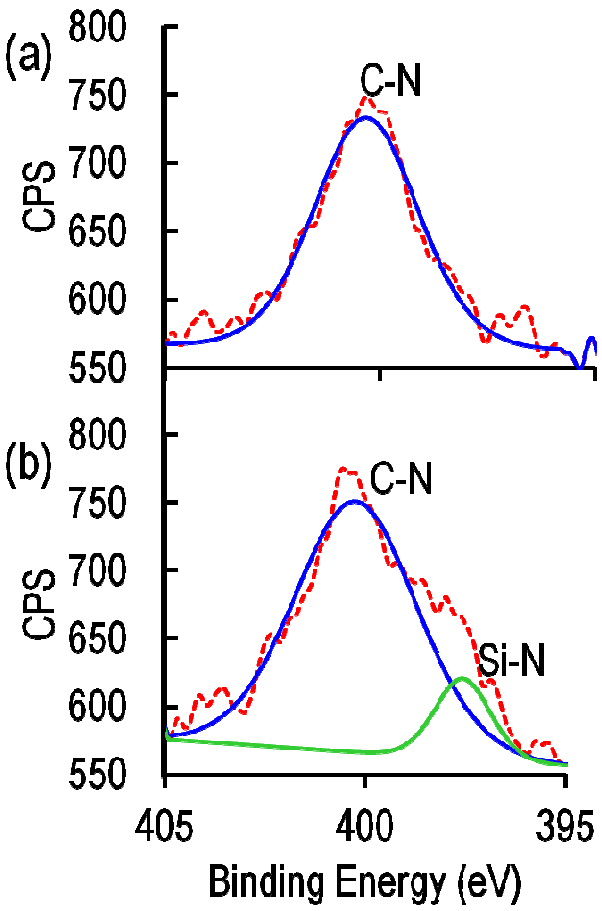

Figure S7. Surface composition of the polymer nanocomposites. Deconvoluted *N 1s* peaks (dotted line) of (a) the UTNC and (b) the PTNC samples. Only one peak, corresponding to the C-N bond (400.3 eV), can be found in the UTNC sample. However, the second peak, which corresponds to the Si-N bond (397.6 eV), is found in the PTNC sample. This result indicates the formation of Si-N bonds in the epoxy resin/plasma-treated SiO_2 nanocomposite.

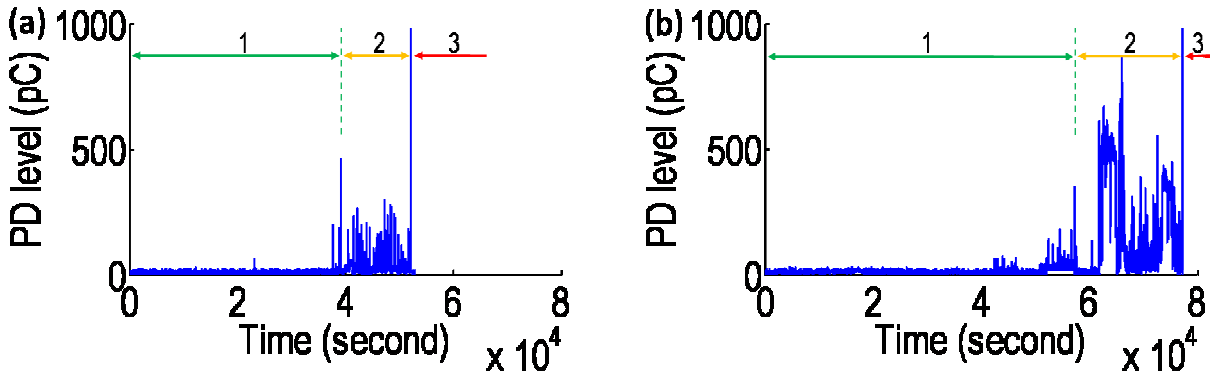

Figure S8. PD level spectra of 7.5 kV endurance test of (a) UTNC and (b) PTNC samples, the breakdown times are 14h 40min and 18h 50min, respectively. The endurance for the PTNC is 4.3 hours (in 300 Hz domain) longer than that of the UTNC. Regions **1**: the void formation region; **2**: the tree propagation region; **3**: the ultimate breakdown.

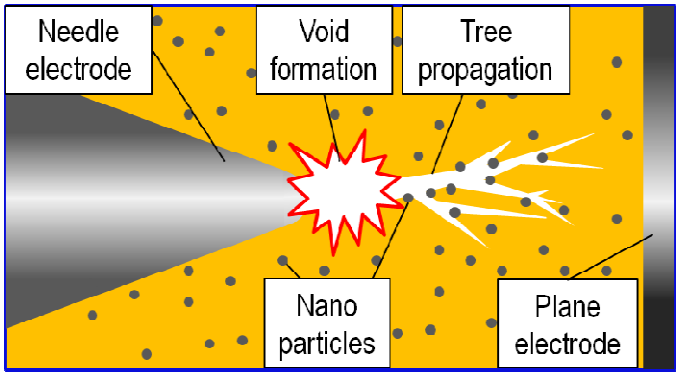

Figure S9. The electrical treeing process in the nanocomposite. Firstly the gaseous void is formed in the vicinity around the tip of the needle electrode. Partial discharges are then generated within the void,

leading to the "tree branches" propagating toward the plane electrode. The ultimate breakdown occurs once the tree reaches the plane electrode with a gaseous tunnel linking the two electrodes.

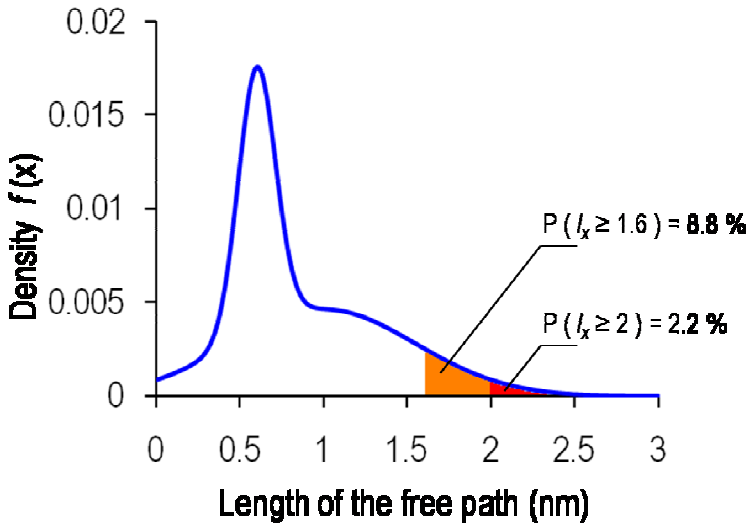

Figure S10. The distribution density of the mean free paths l_x of the electrons in the nanocomposite. The distribution can be fitted as a combination of two Gaussian distributions. The bond dissociation energy of the C-N and Si-N bonds is 3.1 and 3.7 eV, respectively. To break these bonds, the electrons need to move, without collisions, for at least 1.6 and 2 nm, respectively. According to the distribution of l_x, the probabilities of dissociation of the C-N and Si-N bonds are 8.8% and 2.2%, respectively.