

***In Situ* TEM Characterization of Nanostructured Dielectrics**

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Polymer nanocomposites are being considered as potential materials for high energy capacitor due to the possibility of obtaining high dielectric breakdown strength characteristic of the organic matrix and large dielectric permittivity from inorganic filler [1,2]. However in most case the increase in the content of inorganic ceramic filler in polymer nanocomposite inevitably causes a decrease in the dielectric breakdown strength due to the agglomeration of fillers, the existence of defects, and inorganic-organic interfacial effects. Many studies have attempted to improve the compatibility of organic and inorganic, however in the large majority the dielectric breakdown strength is poor and the dielectric loss is high. Additionally, as nanostructured dielectrics are a relatively new class of materials for this application, reports of investigating fundamental mechanisms of dielectric breakdown for the dielectric nanocomposite on the nanoscale have been limited.

Recently the dielectric performance of novel hairy polymer grafted nanoparticle assemblies has been compared to that of blended polymer nanocomposites; these two particular materials systems possessed well-dispersed morphology on the nanoscale and had low dielectric contrast. It was shown that “hairy” polymer grafted nanoparticle assemblies showed substantial improvement in reducing dielectric loss and maintaining charge/discharge efficiency [3].

Here we presented *in situ* TEM observation of these hairy polystyrene grafted SiO₂ nanoparticle assemblies (PS @ SiO₂ NPs, aHNP-PS-1) during pre-breakdown/breakdown process using a TEM MEMS device and STM-TEM holders [4]. Fig. 1 shows the deposition of thin PS @ SiO₂ NPs film, prepared by cryo-microtoming, across two electrodes on rectangular Si₃N₄ window in TEM MEMS device for *in situ* TEM experiment. HAADF-STEM images show the PS @ SiO₂ nanoparticles are well dispersed. Fig. 2 shows *in situ* TEM of a PS @ SiO₂ NPs film during a continuous voltage ramp within a STM-TEM holder, and significant microstructural morphology changes of the film at different electric fields were evident. Fig. 3 shows both morphology and electrical properties of PS @ SiO₂ NPs assemblies before and after breakdown, and it was found that a highly conductive, tree-like structure formed after breakdown. Several additional phenomena including nanoparticle motion and electrostriction were observed. Surprisingly, the measured breakdown strength of PS @ SiO₂ NPs film from *in situ* characterization can reach more than 900 V/μm when the assemblies on the nanoscale approach defect-free state [5].

References:

[1] Whittingham, M. S. MRS Bull. **vol.** 33 (2008), p. 411

[2] Barber, P.; Loye H.-C. Z. *et al* Materials **vol.** 2 (2009), p. 1697

[3] Grabowski, C. A.; Vaia, R. A. *et al* ACS Appl. Mater. Interfaces **vol.** 6 (2014) p. 21500.

[4] Hsiao, M.-S.; Drummy, L. F. *et al* In preparation

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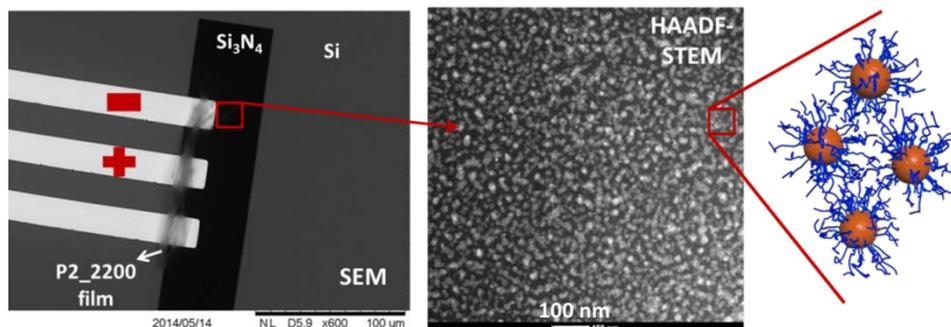


Fig. 1 SEM image of a cryo-microtoming PS @ SiO₂ NPs film depositing across two platinum electrodes on rectangular silicon nitrile window in the TEM MEMS holder. HAADF-STEM image shows silica nanoparticles disperse well in the polystyrene.

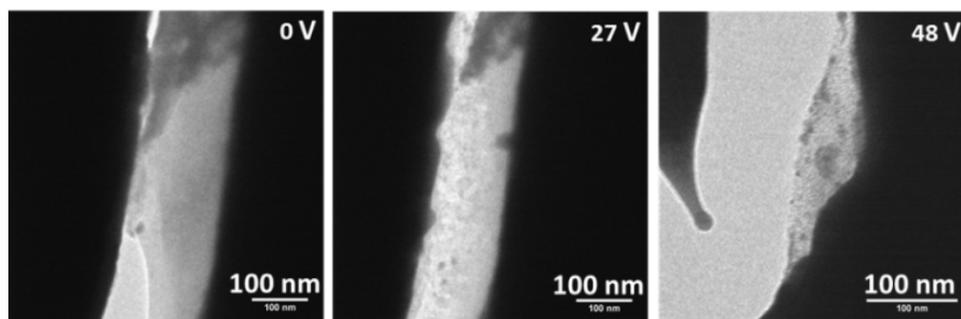


Fig. 2 *In situ* TEM observation of a PS @ SiO₂ NPs film at different electric field.

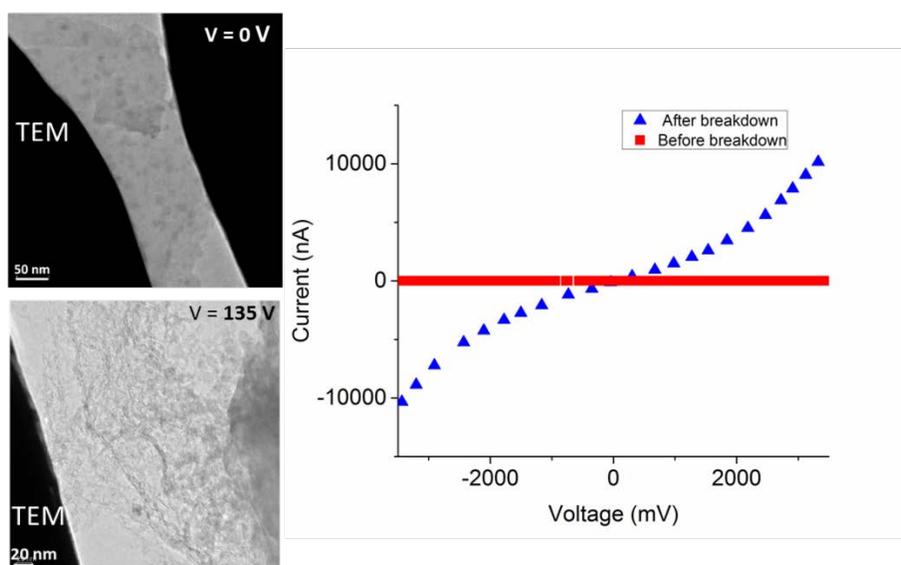


Fig. 3 Morphology and conducting behavior of a PS @ SiO₂ NPs film before and after breakdown, respectively.