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## Hysteresis in Bending Electrostriction of Organic-Inorganic Nanocomposite Electrodes

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Polymers have many attractive characteristics; they are lightweight, inexpensive, fracture tolerant, and flexible. Further, they can be configured into almost any conceivable shape and their properties can be tailored to suit a broad range of requirements. During the last ten years, new polymers have emerged that respond to electrical stimulation with a significant shape or size change, and this progress has added an important capability to these materials. This capability of the electroactive polymers (EAPs) has attracted the attention of engineers and scientists.<sup>[1]</sup>

Dielectric elastomer, one particular class of EAP, is known for their unique properties such as providing an excellent overall performance, combining large elongation. A key feature of dielectric elastomer technology is the use of compliant electrodes. If the electrode cannot be stretched in at least one planar direction of the film while its thickness contracts, actuation is reduced because the polymer is essentially incompressible. The selection of electrode material remains an ongoing research area.

Conductive grease works reasonably well but has drawbacks. In particular, it is difficult to get good adhesion with body and solid type electrode. One approach is to use sputter-deposited one such as ultrathin gold for compliant electrodes. Unfortunately, uniform gold electrodes easily crack and it is difficult to achieve strains greater than about 4% with this technique.<sup>[2]</sup> So, to improve adhesion with the body and to prevent a crack of electrode, we introduced conductive filler in waterborne polyurethane. In other words, we used a conductive nanocomposite for compliant electrodes.

Waterborne polyurethane (WPU) was synthesized by prepolymer mixing process. Diisocyanate for hard segments and polyol for soft segments were isophorone diisocyanate (IPDI) and polypropylene glycol (PPG,  $M_w=2000\text{g/mol}$ ), respectively. Chain extender was 1,3-butanediol (1,3-BD). Dimethylol butanoic acid (DMBA), as emulsifier and triethylamine (TEA) as counteragent were used. Dibutyltindilaurate (DBTDL), 2-butanone (MEK) were used without further purification. Solid content of synthesized WPU was 25 wt %.

Three types of conductive fillers such as carbon black (CB, particle diameter: 20 nm), vapor grown carbon fiber (VGCF, diameter: 0.1~0.3  $\mu\text{m}$ , length: 10~30  $\mu\text{m}$ ), and silver powder (Ag, particle diameter: 22 nm) were dispersed by stirring in WPU for 4 hours. We also used zirconia beads into the reactor to improve the dispersability of conductive filler in WPU matrix. And polyurethane (PU) film was used as dielectric elastomer.

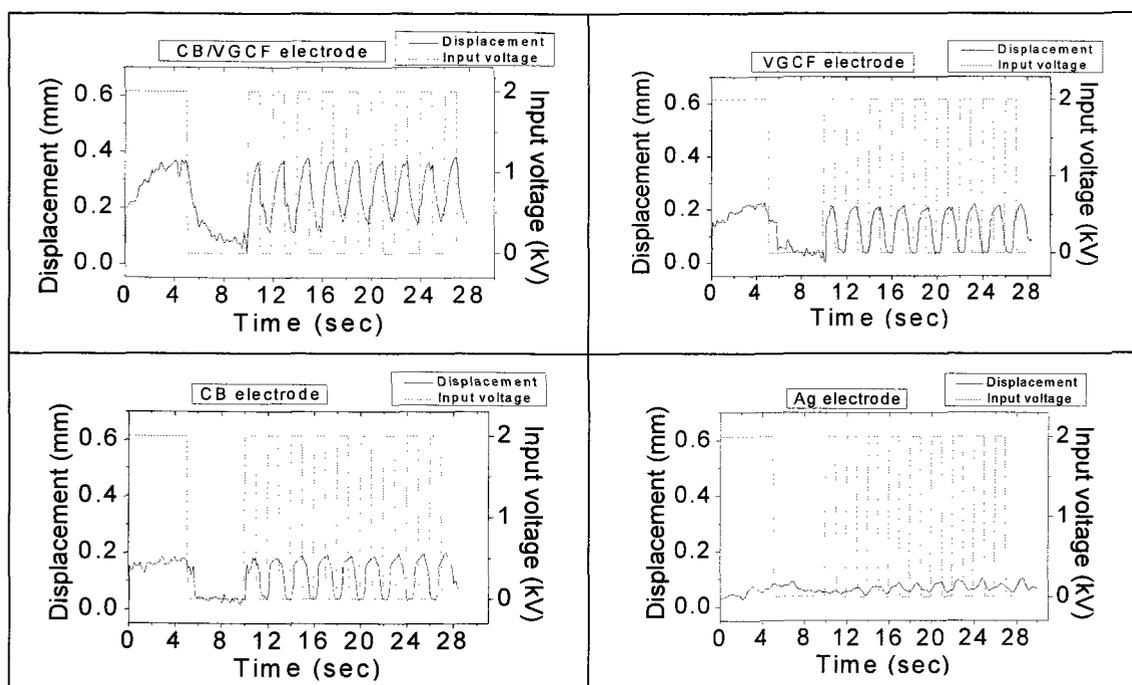
Electrode materials were coated on the both sides of PU film by spin coater. During the application of the electric field, the displacement of actuator was measured with OMRON Z4M-S40R laser sensor. The application of the electric field and the measurement of current were carried out with a SHV120-36K high voltage power supply.

In order to characterize the synthesized waterborne polyurethane, FT-IR analysis was performed at ambient temperature in the range between 4000 and 400 $\text{cm}^{-1}$ . Surface resistance of conductive nanocomposites was estimated by 4-point probe. All measurements were performed for ten replicates of sample and the average value was taken. And the tensile strength tests were carried out with a Universal Test machine according to the specifications of ASTM D-638-02, and the crosshead speed was set at 50mm/min.

The conductive nanocomposite containing VGCF exhibited the lowest threshold concentration at about 23 vol%, compared with 27 vol% for the WPU/ Ag nanocomposite and 46 vol% for the WPU/CB nanocomposite. VGCF with high aspect ratio possessed long conductive paths for electrons, leading to higher conductivity. And the blend of CB and VGCF (CB/VGCF) filler had a synergistic effect to electrical conductivity. The addition of a small amount of the VGCF (7 vol%) dramatically reduced the surface resistance of the conductive nanocomposite containing CB (20 vol%). The incorporation of VGCF into CB nanocomposite provides long conductive paths to link uncontacted CB particles and aggregates. And the CB particles and aggregates serve inter-fiber points of contact. This phenomenon is called as a synergy effect.<sup>[3]</sup>

CB/VGCF electrode had the largest displacement. It was caused by that the increase in relative dielectric constant enhanced the Maxwell stress effect.<sup>[4]</sup> On the other hand, Ag electrode with the lowest surface resistance had the smallest displacement. Because silver powder had heavier weight than CB and VGCF, it disturbed a vertical motion of diaphragm type actuator.

Fig.1 shows the hysteresis in bending deformation when the application of an electric field (8MV/m) was repeated. As shown in Fig. 1, the response speed for the second application of the electric field was much faster than that for the first application. In detail, it took 5 s for the displacement to reach 0.37mm at the first application (CB/VGCF electrode). On the other hand, it took only 0.8 s to reach the same displacement at the second application. Thus, the response speed at the second application was improved about six times than that at the first application.



**Figure 1** The hysteresis in bending deformation at the repeating application of an electric field (8MV/m).

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