ACS PRF Grant No.: 57812-ND7
Project Title: *A Combined Theoretical Simulation and Experimental Study on Electrostriction in Polymer/Ferroelectric Particle Nanocomposites*
PI: Lei Zhu, Case Western Reserve University

1. Project Progress

In the first report year, we have focused on the reversible Maxwellian electroactuation of a dielectric elastomer, 3M Very High Bond (VHB), with prestrain. In the past, it has been reported that 3M VHB with prestrain can achieve over 100% actuation (*Science* **2010**, *330*, 1759). However, this is not true reversible actuation, because prior studies have overlooked an important viscoelastic property of polymeric elastomers, i.e., creep. When an electric field is applied to a dielectric elastomer, the Maxwellian force will be generated. Under this Maxwellian force, the dielectric elastomer will creep in addition to the reversible electroactuation. From quantitative analysis, the contribution of creep to the overall actuation is about 45%. Below, we will discuss our finding in the first report year.

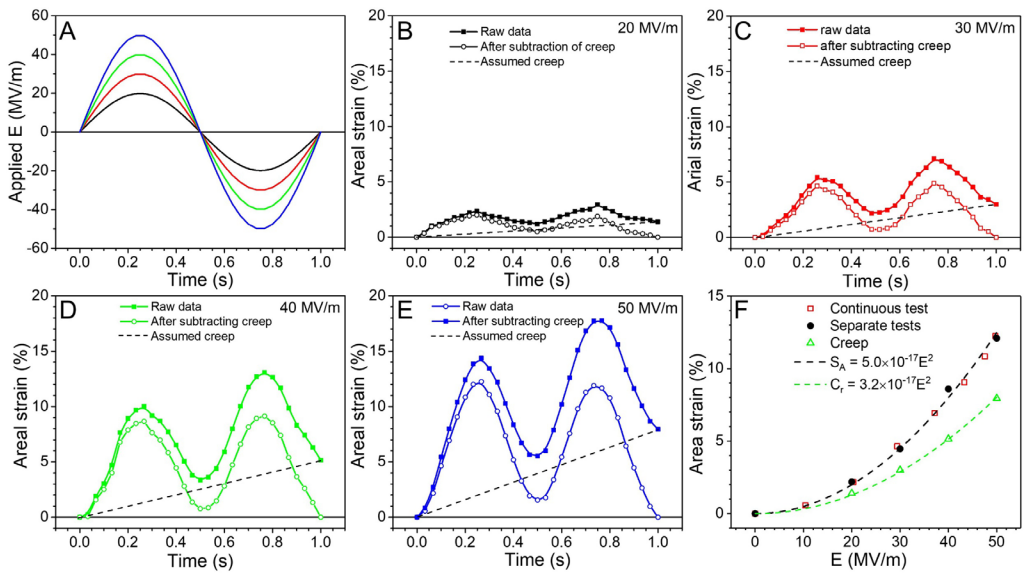

Fig. 1. (A) Profiles of the nominal applied AC fields at 1 Hz. Electroactuation areal strain profiles for the VHB-300% film under different nominal AC fields; (B) 20, (C) 30, (D), 40, and (E) 50 MV/m. The raw data are represented by solid symbols. The assumed creep is represented by the dashed lines. The open symbols represent areal strains after subtracting the creep. Lines connecting the symbols are used to guide the eyes. (F) Reversible areal strain as a function of applied electric field for VHB-300%. Data points from continuous [i.e., from the areal strain profile in (E)] and separate [i.e., from areal strains in (B-E)] tests are shown. The dashed lines are parabolic fitting curves.

In the past, the standard method to test Maxwellian electroactuation is to apply stepwise DC electric field with each step of 2 sec (*Smart Mater. Struct.* **2015**, *24*, 105025). However, this method overlooked creep of the dielectric elastomer. For example, the areal strain can reach 33% at 40 MV/m. To be able to eliminate the creep effect, an AC test method should be used (*J. Appl. Phys.* **2016**, *120*, 164502). Fig. 1 shows electroactuation of VHB 4905 under different AC electric fields at 1 Hz and a prestrain of 300% (Fig. 1A). The raw electroactuation data are shown as solid symbols in Figs. 1B-E. Obviously, there is significant creep at the end of the test at 1 sec. To subtract the creep, we choose to use a linear baseline. After subtraction of the irreversible creep, reversible Maxwellian electroactuation data are plotted as the open symbols. From these data, reversible electroactuation and creep values at different poling electric fields are plotted in Fig. 1F. Both reversible electroactuation and creep can be fitted into parabolic equations as a function of the applied electric field. Even under this AC test method, creep accounts for about 45% of the total actuation. After subtracting creep, the true reversible Maxwellian areal strain can only reach ca. 13% at 50 MV/m, much smaller than the value of >100% reported previously.

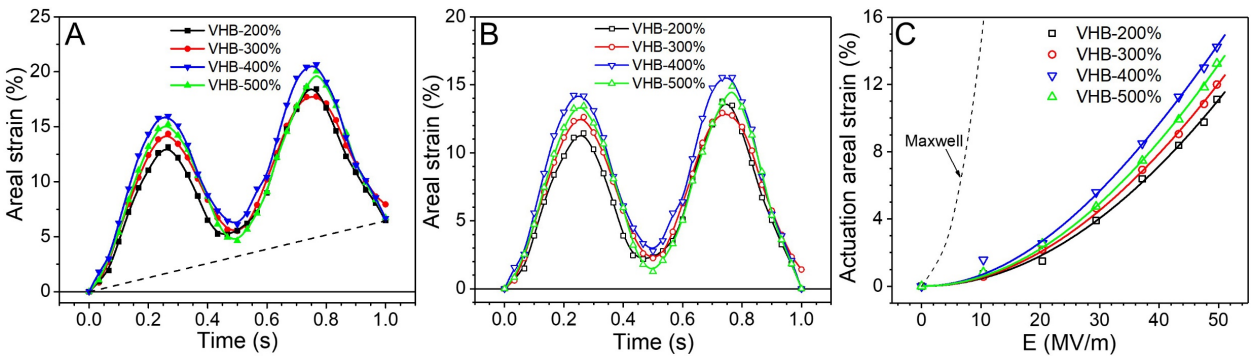

Fig. 2. Electroactuation under a nominal AC field of 50 MV/m at 1 Hz for VHB films with various prestrains: (A) raw data of areal strains (creep is represented by the dashed line) and (B) areal strains after subtraction of creep. (C) Areal strain as a function of electric field. Solid lines are Mooney-Rivlin fitting curves with two parameters, C_{10} and C_{01} (results see Table 1). Dashed line is predicted by the Maxwell equation using a static Young's modulus of 0.05 MPa.

After establishing the AC test method, electroactuation under different prestrains is studied. The raw data of electroactuation areal strains for various prestrained VHB films are shown in Fig. 2A. A master creep line (i.e., the dashed line) could be assumed. After subtraction of the contribution from creep, the reversible electroactuation is shown in Fig. 2B. From the first half peak in areal strain, the electric field dependence is plotted in Fig. 2C. Intriguingly, with increasing the areal prestrain up to 400%, the electroactuation areal strain gradually increased. For the 500% prestrain, the electroactuation decreased slightly. We consider that the strain-hardening at 500% prestrain prevented further enhancement in electroactuation, and the mechanical property of dielectric elastomers plays an important role in the electroactuation under the Maxwellian pressure.

In Fig. 2C, the Maxwell equation significantly over-estimates the electroactuation for dielectric elastomers. For fitting the experimentally measured areal actuation strains, we therefore use the two-constant hyperelastic Mooney-Rivlin (MR) model. This model is usually applied to initially non-prestrained films, for which the initial principal stretching ratio $\lambda = 1$. When the film is in a prestrained state (e.g., areal strain = 300%), its initial principal stretching ratio, $\lambda = 4\times4 = 16$. Therefore, the fitting of experimental data by the hyperelastic MR model needs to be modified in order to count for the non-zero elastic energy in the prestrained film. In the modified MR model, we can obtain both linear and nonlinear parts of the elastic response. With increasing the prestrain, the linear part of elastic contribution decreases before 400% prestrain, and then increases when the prestrain is 500%. Clearly, strain-hardening is observed at 500% prestrain. On the contrary, the nonlinear part of elastic contribution monotonically decreases with the prestrain, indicating that in highly prestrained VHB films the nonlinear effect became less important.

In summary, the reversible electroactuation strain is much smaller than the measured actuation values previously reported, which include creep because of the DC driving field used. We hoped that this study will be able to provide a viable standard for future electroactuation characterization of different dielectric elastomers. In addition, creep from viscoelasticity should be minimized for dielectric elastomers. For example, because of their extremely low T_g (<-100 °C), silicone-based elastomers can exhibit less significant viscoelasticity as in acrylic elastomers such as VHB. More study is needed in the future.

2. Program Impact

This project has initiated a new direction of the PI's research in the fields of electromechanical actuation, which may be viable for future grant applications to NSF and NIH. A visiting PhD student has been working on the project and trained for scientific research.

3. Publications

Liu, L.; Huang, Y.; Allahyarov, E.; Zhang, Z.; Lv, F.; Zhang, Y.; Zhu, L. Understanding reversible Maxwellian electroactuation in a 3M VHB dielectric elastomer with prestrain. *Polymer* **2018**, *144*, 150-158.