High Electromechanical Performance of Electroelastomers Based on Interpenetrating Polymer Networks

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ABSTRACT

The electromechanical performance of interpenetrating polymer networks (IPN) in which one elastomer network is under high tension balanced by compression of the second network, were investigated. Uniaxial stress relaxation analysis confirmed significant decrease in viscoelasticity in comparison with 3M VHB films, the primary component network in the IPN films. In dynamic mechanical analysis, the IPN composite showed a higher mechanical efficiency, suggesting delayed relaxation of the acrylic chains in the presence of IPN formation. This improvement was found to be dependant on the contents of poly(TMPTMA). Actuation performance without mechanical prestrain showed that these IPN electroelastomers had demonstrated high elastic strain energy density (3.5 MJ/m\textsuperscript{3}) and a high electromechanical coupling factor (93.7\%). These enhanced electromechanical performances indicate that IPN electroelastomer should be suitable for diverse applications.

Keywords : Electroelastomer, Interpenetrating Polymer Networks, Electromechanical Performance, Actuator

1. INTRODUCTION

Novel high-performance actuator materials with a muscle-like functionality have long been desired for biomimetic applications such as mini- and micro-robotics and active prosthetics. In the last decade or so, new category of polymer artificial muscles has emerged that exhibit considerable deformation in response to external electrical stimulation.\textsuperscript{1,5} These responsive materials reversibly contract and expand in area which is in strong resemblance to biological muscle. Diverse candidate materials under investigation have been broadly divided into two groups: ionic (including carbon nanotube, conductive polymers, and ionic polymer-metal composites) and electronic (including electroelastomers, also known as dielectric elastomers, and ferroelectric (co)polymers).\textsuperscript{6,7} Among them, electroelastomers based on acrylic copolymer elastomers (e.g., 3M VHB4910) have been particularly attractive because reversible electromechanical strain (380\% in area), elastic energy density (3.4 MJ/m\textsuperscript{3} calculated from acrylic film itself) and electromechanical coupling efficiency (60-90\%) are all extraordinarily high.\textsuperscript{8} Even though these stupendous performances surpass that of natural muscle in many respects, challenges for commercial application still remain due to other properties electroelastomers possess. The first is that electroelastomers commonly require large uniaxial or biaxial prestrain, as much as 25-fold in area to achieve the before mentioned performance. The large prestrain significantly enhances performance by increasing the dielectric breakdown strength particularly with acrylic elastomers.\textsuperscript{9} Common applicable configurations such as bow, bowtie, rigid-frame, diaphragm and rolled actuator, ought to support the required high prestrain despite significantly more space and weight than that of film themselves.\textsuperscript{10,11} Therefore, overall performance of the packaged actuators is substantially lower such as reduced net elastic energy density, limited actuator lifetime due to mechanical instability, and time dependant stress relaxation.\textsuperscript{8} Another challenges of using electroelastomer as artificial muscles is improving electromechanical response such as speed of response and efficiency. The electromechanical response is limited by how fast the electronic circuitry charges and discharges the electrodes on electroelastomers, as well as how fast the film expand and relax in response.\textsuperscript{12} Since the electronic driving circuitry is sufficiently fast, the apparent fundamental limitation on actuator speed is the inherent viscoelastic loss. The viscoelastic
response of electroelastomers can be monitored with respect to time and frequencies. Most of which should be surmounted via development of new materials or improvement in material properties. We have, for instance, recently demonstrated that IPN as new electroelastomers can exhibit high actuation performance without mechanical prestrain.\textsuperscript{13,14}

In our approach, VHB acrylic elastomers were used as the initial elastomeric network to take advantage of the high dielectric breakdown field obtained in highly prestrained acrylic elastomer film. Crosslinkable liquid monomers were swollen into highly-prestrained acrylic network and cured to form the second elastomeric network. When the interpenetrating elastomeric network films were allowed to relax at zero external stress, the acrylic network would contract, compressing the additive network. In the resulting free-standing films, the two networks were in balance, one in high tension and the other in high compression. A schematic explanation of this approach is shown in Figure 1.

To evaluate the overall performance of the IPN as a new category of electroelastomers, the IPN films formed from VHB4905 and 4910 containing 6.59% and 5.16% by weight of poly(TMPTMA) were analyzed. The free standing films of IPN-VHB4905 and IPN-VHB4910 showed 275% and 244% preserved prestrain compared to the pristine acrylic films.

![Fig. 1. Schematic illumination of the fabrication process of IPN electroelastomer.](https://www.spiedigitallibrary.org/conference-proceedings-of-spie)

2. EXPERIMENTAL

To prepare the IPN films, acrylic copolymer elastomers (3M VHB4905 and VHB4910) were stretched to 5 times their original length and width, or 400% biaxial strain. Trimethylolpropane trimethacrylate (TMPTMA), purchased from Sigma Aldrich, was used as the thermally curable additive. Each TMPTMA molecule has three methacrylate functional groups and weighs 338 Dalton. When fully polymerized, TMPTMA forms a highly crosslinked network. Bezoyl
peroxide, also from Sigma Aldrich, was co-dissolved with TMPTMA in ethyl acetate and then sprayed onto the 400% biaxially prestrained VHB films. The films were then placed in a vacuum oven and heated up to 85 °C to induce free-radical polymerization of the trimethacrylate.

Tensile tests were carried out to characterize mechanical properties such as elongation, and tensile stress at break, using a universal testing machine (Instron 5565). Samples were punched out using a dog-shaped punch mold of which size followed ASTM 638 and 882. The tensile test was performed with 50 mm/min test speed for 5 samples per each electroelastomer type. Dynamic mechanical analysis (DMA) was carried out using the Pyris Diamond DMA (PerkinElmer) with a Seiko Instruments Inc, analysis program. Frequency sweep experiments in shear mode were conducted in the range from 0.1 to 100 Hz at room temperature. In uniaxial relaxation tests, samples were stretched to a predetermined strain using a hand-operating setup. The exerted force was measured as a function of time with a force transducer (HBM, type S2) which was connected to a PC running LabVIEW. The original sample geometry was set to a ratio between length and width of 8:1, in order to ensure uniaxial stress conditions.

Diaphragm actuators were fabricated from free-standing composite films. Conductive carbon grease was smeared onto both sides of the film. Thin strips of aluminum foil were used as electrical contacts. Electrically-induced strain was monitored using a CCD camera to capture the changes in diameter, height, and curvatures. Strain in area was computed from geometric relations found between the diameter and height of the bulge.

3. RESULTS AND DISCUSSION.

3.1 Mechanical Properties of the IPN films

Several mechanical properties of the electroelastomer materials influence the actuator performance. For instance low Young’s modulus is critically important in achieving high electromechanical strain and energy density. The response speed and electromechanical conversion efficiency can be limited by viscoelastic losses. Experimental studies of these mechanical properties of IPN films in comparison with pristine and prestrained acrylic elastomers are as follows:

3.1.1 Young’s Modulus

The stiffness of materials is conventionally tested by uniaxial stress-strain measurements. Such loading condition does not reflect the biaxial stress-strain characteristics of the IPN films. Figure 2 shows a simple setup to measure the biaxial deformation of clamped electroelastomer films under hydraulic load. Water was added into the tube to provide a hydraulic pressure to biaxially stress the acrylic film.\(^{15}\) Deflection of the central point in the acrylic film was measured by CCD camera imaging. The value was used to calculate the biaxial strain of the acrylic film by the hydraulic pressure. At small stretch ratios, the Young’s modulus \(Y\), of a clamped circular plate under a uniform applied pressure \(P\) is given by:\(^{16}\)

\[
Y = \frac{3Pr^4}{16h^4} \left( \frac{h}{t} + \frac{0.042}{1 - \nu^2} \left( \frac{h}{t} \right)^3 \right) 
\]

where \(t\), \(h\), \(r\), and \(\nu\) are film thickness, deflection at the film center, and radius of plate and Poisson’s ratio, respectively. By this method, the Young’s moduli of non-prestrained VHBs and free-standing VHB-poly(TMPTMA) are determined at 1.03 stretch ratio. Mechanical properties such as biaxial Young’s modulus, tensile elongation, and tensile stress at break are summarized in Table 1.

<table>
<thead>
<tr>
<th>Electroelastomer</th>
<th>Biaxial Young’s Modulus (MPa)</th>
<th>Tensile Elongation (%)</th>
<th>Tensile Stress at Break (MPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>VHB4905-poly(TMPTMA)</td>
<td>3.94</td>
<td>541.8</td>
<td>6.87</td>
</tr>
<tr>
<td>VHB4910-poly(TMPTMA)</td>
<td>4.15</td>
<td>672</td>
<td>5.7</td>
</tr>
</tbody>
</table>
Fig. 2. Photos of electroelastomers mounted onto lower end of a vertical acrylic tube that has a 25.4 mm inner diameter and a 1.22 m tube length. The geometric change (height and diameter) on hydraulic pressure is calculated for biaxial strain

\[
\lambda = 1 + \left( \frac{\left( r^2 + h^2 \right) / h \cdot \arcsin \left( \frac{2rh}{r^2 + h^2} \right) - 2r}{2r} \right)
\]

3.1.2 Stress Relaxation

Acrylic elastomers showed relatively high viscoelastic loss that contributes to their reduced energy efficiency and response speed.\(^8\) To evaluate the effect of IPN formation on the time dependent mechanical behavior, samples were stretched to a predetermined strain and measured exerted force as function of time.

As Figure 3 shows, the relaxation function (nominal stress at each time divided by infinite time stress) of a VHB film decreases continually for thousands of seconds. On the other hands, the rates of decrease in the IPN film subdues rapidly within 1 s. At the same time, the magnitude of the initial fall in relaxation function of the IPN film is much smaller than that of the VHB film (Figure 3 inset). These phenomena indicate that IPN film shows less viscous behavior. The significantly increased elasticity suggests that the IPN film has greater response speed than the VHB film.

Fig. 3. Comparison of the average relaxation functions \(s_x(t)\) of VHB4905 (X), VHB4910 (open square), VHB4905-poly(TMPTMA) (star), and VHB4910-poly(TMPTMA) (open circle). The inset shows initial relaxation functions from 0 to 10 s.
3.1.3 Mechanical Efficient
As mentioned before, the overall efficiency of an actuator is generally proportional to the mechanical efficiency ($\eta_m$) and is affected by viscoelastic property, loss tangent of the materials, as

$$\eta_m = \frac{1}{1 + \pi \tan \delta} \quad (2)$$

The loss $\tan \delta$ is a measure of the energy lost, expressed in terms of the recoverable energy, and represents mechanical damping or internal friction in viscoelastic system.\(^{17}\) A high $\tan \delta$ value designates that material has a high nonelastic strain component, while a low value shows more elasticity.\(^{18}\) Figure 4 and 5 show the influence of IPN formation on $\tan \delta$ and mechanical efficiency with different frequencies. For VHB film, the $\tan \delta$ increases monotonically with increase frequency and then exhibits a peak at transition frequencies ($\omega_t$), 20 Hz. However, IPN electroelastomer clearly demonstrates a shift in the transition frequency towards the lower frequency side, less frequency dependence, and much lower values of $\tan \delta$. These phenomena also confirm the elastic behavior and the restrained relaxation in the presence of poly(TMPTMA).\(^{19}\) The observed reduction in $\tan \delta$ values is attributed to increased mechanical efficiency (also shown in figure 5).

Similar behavior was reported in prestrained VHB films.\(^{20}\) The applied biaxial prestrain basically reorients the entangled polymer chain in the plane of the film leading to the decrease in viscoelastic loss, and an increase in mechanical efficiency. Polymer realignments are also expected in the IPN films as a result of the locking-in prestrain of the acrylic network. It is interesting to note that the mechanical efficiency of the IPN films is further improved even compared with acrylic films under high prestrain. The measured maximum mechanical efficiencies was 56 % for VHB-poly(TMPTMA), which is about 50% higher than the best reported efficiency, 38% with a 300% biaxially prestrained acrylic film.\(^{20}\) Therefore, the formation of IPN configuration is more effective in withholding the acrylic chain realignment and restricting the viscoelastic relaxation of the polymer chains.

![Fig. 4. Frequency dependence of $\tan \delta$ of IPN electroelastomers](image-url)
3.2 Actuation Performance

Performance metrics of actuator materials include several important parameters such as effective compressive stress (also known as Maxwell stress, \( p \)), elastic strain energy density \( u_e \) and electromechanical coupling factor \( K^2 \). To characterize the performance metrics of the IPN electroelastomers, diaphragm configuration shown in Figure 6(a) is used to measure the strain in area corresponding to recorded images provided in Figure 6(b).\(^{13,14}\)

The effective compressive stress, electrostatic pressure acting perpendicular to the film surface, is determined from

\[
p = \varepsilon \varepsilon_0 E^2
\]

Where, \( \varepsilon \) and \( \varepsilon_0 \) are, respectively, the relative dielectric constant of material and the permittivity of the free space.\(^{5,7,12}\)

\( E \) is the applied electric field (volt per meter). For electrical induced strains greater than 20\%, the elastic strain energy density in the actuator material itself, \( u_e \) (the amount of work generated in one actuation cycle per unit volume of material), is given by

\[
u_e = -\frac{1}{2} \rho \ln(1 + s_z)
\]

where, \( s_z \) is defined as \( t_{\text{act}} / t_0 - 1 \), the film thicknesses before \( t_0 \) and after \( t_{\text{act}} \), and determined from measured area strains by assuming incompressible deformation (shown in Figure 6(b)).\(^{5,7,12}\) Another actuation performance metric is the electromechanical coupling factor, which is defined as the ratio of energy converted into mechanical work to applied electrical energy.\(^{12}\) Assuming no losses from capacitance change for the given strain, electromechanical coupling factor is given by

\[
K^2 = -2s_z - s_z^2
\]

Measured values of maximum thickness strain \( s_z \), maximum electric field conditions \( E \) and materials property \( \varepsilon \) are listed in Table 2 for convenient comparison to the elastic strain energy density and electromechanical coupling factor discussed elsewhere. As indicated by the values, IPN electroelastomers, although they were not under external high prestrain, illustrate the good overall performance in terms of electromechanical thickness strain (75\%), \( u_e \) (the 3.5 MJ/m\(^3\) maximum is 400 times that of our muscle)\(^6\) and \( K^2 \) (93.7\%), comparable to those of VHB4910 films under 300\% biaxial prestrain. Further more, freestanding IPN electroelastomers should improve the overall performance of packaged actuators, such as the net elastic energy density and reliable actuation due to mineralize mechanical supporting structure.
Fig. 6. (a) Diaphragm actuator structure based on IPN composite film. The films are under a constant, small tension maintained in the diaphragm chamber. (b) a series of digital images showing the actuation. The geometric change (height and diameter) in response to applied electric field is calculated for electrically induced area strain. 

\[ \text{Strain}_{\text{Area}} = \left( h^2 + r^2 \right) - \left( h_0^2 + r_0^2 \right) = \left( h_0^2 + r_0^2 \right) \]

Table 2. Actuation performance metric of IPN elastomer and other electroelastomers

<table>
<thead>
<tr>
<th></th>
<th>Prestrain (x,y) (%)</th>
<th>ε</th>
<th>-sₓ (%)</th>
<th>E  (MV/m)</th>
<th>P  (MPa)</th>
<th>uₓ  (MJ/m³)</th>
<th>K² (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>VHB4905-poly(TMPTMA)</td>
<td>(0,0)</td>
<td>2.43⁺</td>
<td>59.36</td>
<td>265.4</td>
<td>1.51</td>
<td>0.68</td>
<td>83.5</td>
</tr>
<tr>
<td>VHB4910-poly(TMPTMA)</td>
<td>(0,0)</td>
<td>3.27⁺</td>
<td>74.97</td>
<td>418.05</td>
<td>5.06</td>
<td>3.5</td>
<td>93.7</td>
</tr>
<tr>
<td>VHB4910 acrylic</td>
<td>(300,300)³</td>
<td>4.8¹</td>
<td>61⁵</td>
<td>412⁵</td>
<td>7.2⁵</td>
<td>3.4⁵</td>
<td>85⁷</td>
</tr>
<tr>
<td></td>
<td>(15,15)³</td>
<td>4.8¹</td>
<td>29⁵</td>
<td>160⁵</td>
<td>0.6⁵</td>
<td>0.091⁵</td>
<td>43.6⁷</td>
</tr>
<tr>
<td>HS3 silicone</td>
<td>(68,68)³</td>
<td>2.8¹</td>
<td>48⁵</td>
<td>110⁵</td>
<td>0.3⁵</td>
<td>0.098⁵</td>
<td>73⁷</td>
</tr>
<tr>
<td>CF19-2186 silicone</td>
<td>(45,45)³</td>
<td>2.8¹</td>
<td>39⁵</td>
<td>350⁵</td>
<td>3.0⁵</td>
<td>0.75⁵</td>
<td>62.8⁷</td>
</tr>
</tbody>
</table>

⁺ ε was measured at 10 Hz
* K² are calculated values from reported -sₓ using equation (5)

The dependence of K² and η on the amount of poly(TMPTMA) additive in the IPN films is shown in Figure 7. The values of η increase up to 70 % and then remains invariant with increasing additive. On the other hand, K² initially
increases with the amount of poly(TMPTMA), but after reaching 93.7%, it turns to sharply diminish. This rapid reduction in $K^2$ is due to decreasing strain: the IPN films become increasingly stiffer and thus require much higher energy to obtain large strain. These results indicate that there exists a compromised IPN composition for high energy conversion efficiency and high coupling efficiency.

Fig. 7. The dependence of mechanical efficiency ($\eta$) at 100 Hz and electromechanical coupling factor ($K^2$) on poly(TMPTMA) concentration for VHB4910. Note that values of zero weight percent correspond to 300% biaxially prestrained VHB4910.

4. CONCLUSION

The benefits of the prestrain are reproduced in IPN films consisting of two internal tension-compression balanced elastomer networks. The IPN films showed less viscous behavior than the corresponding acrylic elastomers. Dynamic mechanical analysis showed that the IPN films additionally exhibit reduced viscoelastic loss and consequently increased mechanical energy efficiency and response speed. The actuation metrics, such as elastic strain energy density and electromechanical coupling factor from actuation performance with no prestrain, also demonstrated high overall performance. After comprehensive comparisons with prestrained VHB films, it can be certain that it is possible to use such ‘prestrain-less’ electroelastomers to fabricate novel actuators without (or with minimal) supporting structure/mass and to delivery greater actuator performance.

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REFERENCE


