Electrostrictive Graft Elastomers and Applications

*National Research Council, NASA-Langley Research Center, Hampton, VA 23681, USA
**NASA-Langley Research Center, Hampton, VA 23681, USA
***Jet Propulsion Laboratory/CalTech, Pasadena, CA 91109, USA

Abstract

Efficient actuators that are lightweight, high performance, and compact are needed to support telerobotic requirements for future NASA missions. In this work, we present a new class of electromechanically active polymers that can potentially be used as actuators to meet many NASA needs. The materials are graft elastomers that offer high strain under an applied electric field. Due to its higher mechanical modulus, this elastomer also has a higher strain energy density as compared to previously reported electrostrictive polyurethane elastomers. The dielectric, mechanical and electromechanical properties of this new electrostrictive elastomer have been studied as a function of temperature and frequency. Combined with structural analysis using x-ray diffraction and differential scanning calorimetry on the new elastomer, structure-property interrelationship and mechanisms of the electric field induced strain in the graft elastomer have also been investigated. This electroactive polymer (EAP) has demonstrated high actuation strain and high mechanical energy density. The combination of these properties with its tailorable molecular composition and excellent processability makes it attractive for a variety of actuation tasks. The experimental results and applications will be presented.

Introduction

Materials that sustain mechanical displacement under controlled electrical excitation are needed as actuators for many applications. For aerospace applications, there is also a need for low mass, high performance, and ease of processability which are inherent characteristics of electroactive materials. Electroactive polymeric elastomers that show electromechanical activities, especially the large electric field induced strain, are being thought of potential candidates for such applications. Existing materials include polyurethane elastomers, [1-3] and silicon rubber [4,5]. Recently, we have demonstrated an electrostrictive response in graft elastomers. The elastomers offer large electric field induced strain and significant high mechanical modulus. Therefore, a high electromechanical output power, or high strain energy density is achieved. In addition to the high performance as a new class of electromechanically active polymeric materials, the electrostrictive graft elastomers also offer advantages such as excellent processability, and electrical and mechanical toughness.

Experiment

An electrostrictive graft elastomer consists of two components, a flexible backbone polymer and grafted crystalline groups. The schematics in Figures 1a and 1b show the structure and molecular morphology, respectively, of the graft elastomer. The graft crystalline phase provides the polarizable moieties and serves as cross-linking sites for the elastomer system.
The graft elastomer films were prepared by solution casting. Five grams of graft elastomer powder was added to N,N-dimethylformamide (as received from Aldrich) and was heated to 60 °C while stirring for 2 hours to make a 5 wt% solution. The solution was cooled to room temperature and cast on glass substrates and placed in a vacuum chamber. After drying overnight under vacuum, at room temperature, 20 micrometer thick films of the graft elastomer films were obtained. Gold electrodes were sputtered on opposing sides of the films using a plasma deposition device (Hummer III, Technics).

Dynamic mechanical and piezoelectric properties of the graft elastomer films were measured as a function of temperature and frequency using a Rheovibron DDV-II-C mechanical analyzer that has been modified to collect electric charge data as a function of applied stress. Dielectric data of the films was measured as a function of temperature and frequency using an HP4192A Impedance Analyzer.

The electric field induced strain was tested as a function of the applied electric field. The deflection of a bilayer bending actuator was measured and the strain was calculated using the relationship

\[ S_\perp = 2S_T = 2d(1/R) = 4dL/(L^2 + I^2) \]

where \( S_\perp \) is the longitudinal strain of the active elastomer layer, \( S_T \) is the strain of the active layer in the transversal direction, \( 1/R \) is the bending curvature of the actuator, \( d \) is the distance between the central layers of active and non-active films, and the \( L \) and \( I \) are the deflection of the tip of the actuator in in-surface and off-surface directions.[6,7]

The low temperature response of the bending actuator was tested in a Satec TCS1200 temperature controller with cryovac chamber (Satec Systems Inc.). Voltage was applied using a Trek 10/10A amplifier. Deflection was measured as a function of temperature (-55 °C to 25 °C) and voltage (up to 2.75 kV) using a Sony XC-55 progressive scan CCD and IMAQ 1407 PCI image acquisition board (National Instruments) controlled by LabView software (National Instruments).

The results of the x-ray diffraction (XRD) test of the films showed that the crystallinity is about 22%. Since the graft elastomer contains 30 wt% of the graft polymer, approximately, 70% of the graft polymer crystallized for the present processing condition. Differential scanning calorimetry (DSC) analysis showed that the melting temperature of the graft elastomer is about 160 °C with a melting range from 145 °C to 185 °C. DSC results also indicate that a glass transition occurs around 50 °C (from 40 °C to 60 °C). This transition should be the secondary glass transition, which is related to the cross linking grafted units.
RESULTS AND DISCUSSION

Figure 2 shows the relationship between the electric field induced strain, $S$, and the applied electric field at room temperature. The strain exhibits a quadratic dependence with the applied electric field. The electric field induced strain was observed to be as large as 3.9% at an applied electric field of 120MV/m.

![Graph showing the relationship between electric field and induced strain.](image)

Figure 2. Electric field dependence of the induced strain on the graft elastomers.

After subjecting the graft elastomer film to a high electric field, 130MV/m, the film was tested for piezoelectric activity. The result is shown in Figure 3. The piezoelectric strain coefficient, $d_{31}$, is only about 0.1 pC/N in the temperature range from 25 °C to around 100 °C at frequencies of 1 Hz, 10 Hz, and 100 Hz. This indicates that there is no significant remanent polarization in the graft elastomer after being treated under a high electric field.

![Graph showing piezoelectric activity.](image)

Figure 3. Piezoelectric activity of the graft elastomer after poling treatment.

In order to understand the mechanisms of the electrostrictive response of the graft elastomer, dynamic mechanical and dielectric properties of the elastomer were investigated as a function of frequency and temperature. The results of the dynamic mechanical analysis are shown in Figure 4. The temperature range for the test was from room temperature to 100 °C at frequencies of 0.1, 1, 10, 100 Hz. As can be seen, at room temperature, the mechanical modulus of the graft elastomer is in the range from 550 MPa to 700 MPa for the measured frequencies.
Figure 4. Temperature and frequency dependence of dynamic mechanical properties of the graft elastomer films.

The temperature and frequency dependence of the dielectric constant of the graft elastomer is shown in Figure 5. At room temperature, there is no significant disparity in the dielectric constant with frequency; however, as temperature increases, the dielectric constant varies for the range of frequencies measured.

Figure 5. Temperature and frequency dependence of dielectric constant of the graft elastomer.

In the polymeric elastomers exhibiting a large electric field induced strain, two intrinsic mechanisms are considered as primary contributors. They are electrostriction and the Maxwell stress effects.[7] The contributions from both mechanisms exhibit a quadratic dependence with an applied electric field. The electrostrictive effect is due to the reorientation of the polar phase in response to an applied electric field. The electrostrictive effect is due to the reorientation of the polar phase in response to an applied electric field while the Maxwell stress effects are attributed to the force generated by the accumulated charges on the opposing surface of films under an applied electric field. The strain response of an elastomer can be contributed by either one of them or both of them. The following equations give the relationships of the strain and the applied electric field through the two mechanisms:

\[ S_{\text{electrostriction}} = -Qe_0^2(K-1)^2E^2 \quad \text{and} \quad S_{\text{Maxwell}} = -se_0^2KE^2/2 \]  

(2)

where \( Q \) is the electrostrictive coefficient, \( K \) is the dielectric constant, \( s \) is the mechanical compliance, and \( E \) is the applied electric field. The total strain should be
\[ S = S_{\text{electrostriction}} + S_{\text{Maxwell}} = RE^2 \]  

where \( R \) is the electric field induced strain coefficient.

According to the experimental results, a comparison can be made relating the contributions of the different mechanisms to the strain response of polymeric elastomers, which is tabulated in Table 1. For silicon rubber, 100% of the strain response resulted from the Maxwell stress, or electrostatic, contribution.\[5\] For electrostrictive polyurethane, the majority of the strain response is due to electrostriction; however, the Maxwell stress contribution is still significant due to the low mechanical modulus of polyurethane.\[3\] For the newly developed electrostrictive graft elastomers, more than 95% of the strain response is contributed by electrostriction mechanism while the contribution from the Maxwell stress is less than 5%.

Table 1. Comparison of the contribution of mechanisms to the strain response

<table>
<thead>
<tr>
<th>Materials</th>
<th>Electrostriction</th>
<th>Maxwell Stress</th>
</tr>
</thead>
<tbody>
<tr>
<td>Silicon rubber</td>
<td>0</td>
<td>100</td>
</tr>
<tr>
<td>Polyurethane</td>
<td>65</td>
<td>35</td>
</tr>
<tr>
<td>Graft elastomer</td>
<td>95</td>
<td>5</td>
</tr>
</tbody>
</table>

Table 2 gives a comparison of some key properties of electrostrictive polyurethane and graft elastomers as electroactive polymeric materials. As can be seen,

Table 2. Comparison of electromechanical properties of electrostrictive elastomers

<table>
<thead>
<tr>
<th>Materials</th>
<th>Strain, ( S ) (%)</th>
<th>Modulus, ( Y ) (MPa)</th>
<th>Output power, ( P ) (MPa)</th>
<th>Energy density, ( E_{\text{strains}} ) (J/kg)</th>
<th>Dielectric Constant, ( K )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Graft elastomer</td>
<td>4</td>
<td>550</td>
<td>22</td>
<td>247</td>
<td>11</td>
</tr>
<tr>
<td>Polyurethane</td>
<td>11</td>
<td>17</td>
<td>2</td>
<td>87</td>
<td>6</td>
</tr>
</tbody>
</table>

the graft elastomers offers about 4% induced strain which is smaller than that of polyurethane. However, the graft elastomer exhibits a significantly higher mechanical modulus, which results in the high mechanical output power and high specific strain energy density. In addition to the good electromechanical properties, the electrostrictive graft elastomers also has a higher dielectric constant than that of the polyurethane elastomers. This is an advantage of the graft elastomer over the polyurethane in applications.

APPLICATIONS

Two types of actuators were fabricated using the electrostrictive graft elastomer: a unimorph actuator and a bimorph actuator. The unimorph is fabricated by adhering a layer of the graft elastomer film with electrodes and a layer of the graft elastomer film without electrode together. The adhesive used was a room temperature curable epoxy resin. Figures 7a and 7b show the unimorph in the unexcited state and excited state, respectively. The adhesive used was a room temperature curable epoxy resin. Figures 7a and 7b show the unimorph in the unexcited state and excited state, respectively. The unimorph actuator bends in one direction and its response frequency is two times of the frequency of the driving power supply, or \( f(\text{bending}) = 2f(\text{driving}) \).
Figure 7. The unimorph actuator (a) unexcited and (b) electrically excited state.

Figure 8 shows the bimorph actuator, which can bend in both directions when controlled by the power supply. The response frequency of the bimorph (two-direction bending actuator) is the same as the frequency of the power supply, \( f(\text{bending}) = f(\text{driving}) \).

Figure 8. The bimorph actuator in the state of unexcited (middle), one direction excited (left), and opposite direction excited.

In the low temperature test, it was observed that the actuators still function at \(-50^\circ\text{C}\).

CONCLUSIONS

A new class of electromechanically active polymer was developed using electrostrictive graft elastomers. These elastomers offer a unique combination of desirable promising properties including: light weight, large electric field induced strain, high performance, and excellent processability. Optimization of electromechanical properties of the graft elastomers can be realized by molecular design, composition adjustment, and processing to meet requirements of various applications.

REFERENCES