Linear Constitutive Model for Electromechanical Transduction in Ionic Polymer Materials
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ABSTRACT
A linear electromechanical model has been developed for a class of active materials fabricated from ionomeric polymers. A series of experiments are performed to assess the validity of an equivalent circuit model for ionic polymer transducers. The fundamental parameters of the model are the dielectric permittivity of the material, the viscoelastic modulus, and the effective strain coefficient of the transducer. In this work we present experimental results which highlight the key features of the linear electromechanical model. Our results demonstrate the importance of modeling the frequency-dependence of both the electric permittivity and the strain coefficient. Both of these material parameters exhibit strong frequency dependence below the maximum frequency range of the model tested (20 Hz).

INTRODUCTION
Ionic polymer materials fabricated from ion-exchange membranes are a relatively new class of active material. Like other types of materials, such as piezoelectric ceramics and polymers, ionic polymers exhibit sensing and actuation properties due to electromechanical coupling. The primary advantage of ionic polymers compared to piezoelectric materials is their ability to achieve strains on the order of 1-2% at electric fields on the order of kV/m. The primary disadvantages is that the material must remain hydrated to maximize electromechanical coupling. Seminal work in this field was performed by Oguro et al. (1992) and Sadeghipour et al. (1992). For a review of the advances made in the development of ionic polymer materials, see the recent reviews by Shahinpoor et al. (1998) and Nemat-Nasser and Thomas (2001).

Several models have been proposed for sensing and actuation using ionic polymer materials. Early work by Kanno et al. (1995) and Kanno et al. (1996) focused on the use of linear time invariant models of the electrical response and mechanical actuation properties. More recent models proposed by de Gennes et al. (2000), Nemat-Nasser and Li (2000), Asaka and Oguro (2000), Tadokoro et al. (2001), and Nemat-Nasser (2002) have modeled the physical characteristics of ionic motion within the polymer and related these processes to the electromechanical coupling. The various models account for different physical processes within the material. The primary difference between the models is that certain models propose a ‘hydraulic’ model that accounts for mechanical deformation, while others proposed an electrostatic model to account for electromechanical actuation. Although a number of the models are able to correlate experiment with theory, the exact physical mechanisms that produce electromechanical coupling is still a matter of debate.

The models developed by our group have taken a different approach to the analysis of electromechanical coupling in ionic polymer materials. Like the work by Kanno et al. (1995) and Kanno et al. (1996), we propose a linear model of electromechanical coupling based on the relationship between force, deflection, voltage, and current in the polymer. Our work has extended their results by developing a linear constitutive model that directly represents the electromechanical coupling in the material. Results published by Newbury and Leo (2002a) demonstrated that a two-port electromechanical model could properly account for both sensing and actuation in ionic polymer materials. This result was extended in Newbury (2002) and Newbury and Leo (2002c) to include geometric scaling in the model parameters. This extension enables the analysis of sensing and actuation as a function of fundamental material parameters and the geometry of bender transducers. The fundamental material parameters of the model are the electric permittivity, the viscoelastic modulus, and an effective strain coefficient that models electromechanical coupling. Any input-output relationship for both sensing and actuation can be determined from these three material parameters and the geometry of the transducer.

The purpose of this work is to present a rigorous experimental validation of this model within the linear operating regime of the material. This is not a straightforward task due to several factors. First, the material requires hydration to maximize the electromechanical coupling. Our experiments are performed in air, therefore it is of utmost importance to maintain a consistent level of hydration to produce repeatable experimental results. Second, the material exhibits noticeable memory which leads to non-repeatable measurements of force and deflection under (nominally) equivalent operating conditions. Finally, back relaxation occurs when subjected to step changes in
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applied field. At certain times this back relaxation produces steady-state force and deflection that is opposite to the initial motion, whereas at other times it produces a steady-state response that is consistent with the initial motion. This non-repeatability limits the ability to accurately predict the static response of the polymer.

In spite of these difficulties, our goal is to perform a set of experiments that validate the key aspects of the model developed by Newbury and Leo (2002b). These key aspects include (1) the simplifying assumption regarding the reflected impedance under blocked and free mechanical boundary conditions, (2) empirical methods for determining the three fundamental material parameters, and (3), the scaling in length and width as predicted by the model.

MODEL SUMMARY

A linear model of electromechanical transduction in ionic polymer benders has been derived by Newbury (2002) and Newbury and Leo (2002b). The frequency-dependent model takes the form

\[
\begin{bmatrix}
    v \\
    f
\end{bmatrix} = \frac{1}{s} \begin{bmatrix}
    \frac{t}{L_w \eta} & \frac{3}{4} \frac{t^2}{L_d} \frac{dY}{\eta^2} \\
    \frac{3}{4} \frac{t^2}{L_d} \frac{dY}{\eta^2} & \frac{Y_w t^3}{4L_d^2} + s^2 \frac{3L_{free} \rho_w \omega t}{L_d^3 \Gamma^4}
\end{bmatrix} \begin{bmatrix}
    i \\
    \dot{u}
\end{bmatrix}
\]

where \( v \) is the voltage, \( f \) is the force, \( i \) is the current, and \( \dot{u} \) is the velocity at the tip as shown in Figure 1. The geometry of the beam is defined by the width \( w \), the thickness \( t \), the free length \( L_{free} \), the total length \( L_t \), and the distance between the clamp and the driving end, \( L_d \). The material is defined by the density \( \rho_m \), the elastic modulus \( Y \), the electric permittivity \( \eta \), and the strain coupling coefficient \( d \). The parameter \( \Gamma \) is related to the solution of the cantilever beam equation for the first mode, and \( s \) is the Laplace variable. A detailed derivation of this model can be found in Newbury (2002) and Newbury and Leo (2002b).

To obtain quantitative information from the equivalent circuit model, one must first determine values for the material parameters \( d \), \( \eta \), and \( Y \). The fact that the underlying mechanisms responsible for ionic polymer transducer behavior have not yet been conclusively identified precludes an analytical approach to parameter estimation that is based only on well understood, fundamental physics and known (or easily measured) material properties. To avoid this issue, an experimental approach is taken in this work.

Because significant variations in the performance of different transducers with similar dimensions have been observed, all of the model parameter identification experiments were performed on the same ionic polymer transducer. Also, the validation experiments that are compared to simulated responses were performed using the same transducer used in the identification process.
IDENTIFICATION OF MODEL PARAMETERS

In general, the approach to parameter identification taken in this work was to consider the analytical expressions corresponding to several measurable input-output relationships and identify the expressions that are strongly affected by the term of interest but contain as few additional material parameters as possible. This approach allowed the material parameters to be isolated and then identified.

All the parameter identification experiments were performed on the same ionic polymer transducer. The transducer dimensions were: \( t = 0.2\text{mm} \), \( w = 5\text{mm} \), \( L_t = 33\text{mm} \), \( L_{free} = 25\text{mm} \), and the distance \( L_d \) between the electrode (the clamped end) and driving point was 20mm. A detailed discussion of the test setup and experimental procedure can be found in Newbury (2002).

One issue that hampered the parameter identification process as well as the model validation process was the relatively inconsistent behavior exhibited by the polymer transducers. Figure 2 shows results of identical blocked force with voltage input experiments that were performed only minutes apart, without disturbing any of the hardware used in the experiment. While the current responses overlay very well, there are significant differences in portions of the blocked force. In one experiment, the transducer exerted a steady state blocked force in the same direction as the initial force. In the other experiment, the opposite was true. With the polymer used in the identification process, the response with steady state force in the opposite direction from the initial force was seen more frequently. Inconsistent behavior was less of an issue for the experiments that were analyzed in the frequency domain. The greater consistency seen in frequency domain data may be due to the fact that data from a much greater period of time were averaged. Also, it is possible that the transducers behave more consistently above approximately 1Hz.

Mechanical Terms

Characterization of the mechanical terms in the model was performed with both frequency and time domain data. Measurements of the force response under a step change in the displacement were combined with measurements of the mechanical stiffness of the beam to obtain a measurement of the elastic modulus (see Figure 3). Initial work at characterizing the viscoelastic properties of the beam were complicated by poor measurement coherence above 20 Hz. The result of the mechanical characterization produced an elastic modulus of 0.4 GPa.

Electrical Terms

As derived in Newbury (2002) and Newbury and Leo (2002b), the electric terms in the transducer model are parameterized by the DC resistance

\[
R_{dc} = \frac{\rho_{dc} t}{L_t w}
\]
where $\rho_{dc}$ is the DC resistivity of the material, and the frequency-dependent electrical permittivity,

$$\eta(s) = \sum_{i=1}^{n} \frac{\varepsilon_i}{1 + s\varepsilon_i\rho_i}$$

(3)

where $\varepsilon_i$ and $\rho_i$ are the permittivities and resistivities of the individual branches of the electrical impedance model. Multiple branches are required to model the electrical impedance of an ionic polymer.

A weighted minimization of the time and frequency domain responses was used to obtain the electrical impedance model. The results are shown in Figure 4 and a summary of the electrical parameters is shown in Table 1. The inputs were a 1V voltage step, a 5mA current step, and a 0.18Vrms 0-50Hz random input. The experimental frequency response is based on 15 averages of a 2048 point FFT with a sampling rate of 128Hz.

Figure 4: Simulated versus experimental electrical response.

An important aspect of the electrical impedance is that it represents neither a purely capacitive nor a purely resistive response. Unlike other types of transducers (e.g. piezoelectric devices) which exhibit predominantly capacitive or resistive impedance, an accurate model of an ionic polymer transducer must properly account for the combination of capacitance and resistance which exists at low frequencies. Our results demonstrate that a multiple-branch linear model is able to account for this behavior over the frequency range tests (0-20 Hz).
Table 1: Resistivities and permittivities obtained from a weighted minimization.

<table>
<thead>
<tr>
<th>Permittivity (F/m)</th>
<th>$\epsilon_1$</th>
<th>$\epsilon_2$</th>
<th>$\epsilon_3$</th>
<th>$\epsilon_4$</th>
</tr>
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<tr>
<td>0.018</td>
<td>0.0084</td>
<td>0.0035</td>
<td>0.00040</td>
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</tbody>
</table>

<table>
<thead>
<tr>
<th>Resistivity (Ω·m)</th>
<th>$\rho_1$</th>
<th>$\rho_2$</th>
<th>$\rho_3$</th>
<th>$\rho_4$</th>
</tr>
</thead>
<tbody>
<tr>
<td>32.8</td>
<td>4.24</td>
<td>3.0</td>
<td>11.2</td>
<td></td>
</tr>
</tbody>
</table>

Electromechanical Coupling Term

In Newbury and Leo (2002b), the transformer turns ratio $N$, which represents the electromechanical coupling, was interpreted as the relationship between open-circuit voltage and applied force. For this interpretation to apply, the transducer’s DC resistance $R_{dc}$ (400Ω) must be large relative to the impedance $Z_p$, so that the charge generated by the electromechanical coupling is not ‘bled off’ through $R_{dc}$. Inspection of the electrical impedance plot in Figure 4 reveals that the parallel combination of $Z_p$ and $R_{dc}$ is at least an order of magnitude less than the value of $R_{dc}$ for approximately 0.1Hz and above. This order of magnitude ratio will not occur unless $R_{dc}$ is very large relative to $Z_p$. Therefore, the physical interpretation for $N$, along with the ensuing derivation in terms of dimensions and material parameters, is valid for 0.1Hz and above.

The identification of the strain coefficient $d$ is based on the expression for the blocked force exerted by the polymer transducer when excited by a voltage input. This experiment was chosen because it depends on only two material parameters, the modulus $Y$ and the strain coefficient $d$. Because the blocked force is independent of the other material parameters, the identification of the $d$ parameter can only be affected by inaccuracies in the identification of the modulus – it will be not be affected by errors in any of the other material parameters.

The blocked force expression, derived in Newbury and Leo (2002b) is

$$\left(\frac{f}{v}\right) = \frac{3dtwY}{4L_d}. \quad (4)$$

As with the terms responsible for the dynamics of the electrical impedance, a combination of time and frequency domain data was used in the identification process. Both the force produced by a 1V step input and the force produced by a 0-20Hz 0.61Vrms random input were considered. The frequency domain data is based on 10 averages of a 2048 point FFT with a sampling rate of 51.2Hz.

To help determine the nature of the function used to represent $d$, the blocked force step response, an example of which is shown in Figure 5a, was analyzed. One important response feature is that the initial force and the DC gain have opposite signs. By the following argument, this feature leads to the conclusion that $d$ is nonminimum phase.

![Figure 5: Blocked force with voltage input: a) step response, b) frequency response.](image-url)
as

\[ f = V_s(-A + Be^{-br})1(\tau) \quad \text{with} \quad B > A, \quad (5) \]

where \( A, B, \) and \( b \) are positive scalars, \( \tau \) represents time, and \( 1(\tau) \) is the unit step function. This expression will have a positive initial response of \( B - A \) and a DC value of \(-A\). Transforming equation 5 to the Laplace domain and solving for the transfer function \( f/v \) gives

\[ \frac{f}{v} = \frac{s(B - A) - Ab}{s + b}, \quad (6) \]

which has a pole at \(-b\) and a zero at \( \frac{Ab}{B - A} \). Since \( B > A \), the zero is in the right half plane, and the expression in equation 6 is nonminimum phase.

Further insight can be gained by also considering the frequency domain data for the blocked force, shown in Figure 5b. Above 2Hz, the phase starts to drop below 0 degrees. The phase of the single pole-zero combination in equation 6 will start at 180 degrees and end up at zero degrees, but it cannot produce a net phase change greater than 180 degrees. Therefore, additional terms will be necessary. Note that the additional phase lag cannot be accounted for by the frequency dependence of the modulus because the modulus adds phase lead to the blocked force transfer function.

The DC gain of the transfer function was first determined by examining the blocked force step response shown in Figure 5a and solving for the DC gain of \( d \) using equation 4. The MATLAB constrained minimization routine \textit{fmincon} was used to estimate the poles and zeros of a transfer function for \( d \). The constraints imposed were that one of the zeros be positive and that the remaining poles and zeros be negative. Equation 4 was used to predict the blocked force corresponding to \( d \). The prediction error for the step response was quantified using

\[ e_1 = \sqrt{\frac{1}{M} \sum_{i=1}^{M} (x_{i,\text{pred}} - x_{i,\text{meas}})^2}, \quad (7) \]

where the \( x_i \) are the force values at the \( M \) discrete points in time, and the subscripts ‘pred’ and ‘meas’ denote predicted and measured values. The function that was minimized is

\[ \Upsilon = w_1 e_1 + w_2 \sqrt{\frac{1}{P} \sum_{i=1}^{P} |F_{\text{pred}}(f_i) - F_{\text{meas}}(f_i)|^2}, \quad (8) \]

where \( F(f_i) \) denotes the blocked force per volt at frequency \( f_i \), and \( w_1 \) and \( w_2 \) are scalars that can be adjusted to bias the fit towards either the step response or the frequency domain measurement. A transfer function with three zeros and four poles gave a reasonable fit to both the step response and the frequency domain data. A comparison of the experimental and simulated responses is shown in Figure 6 with \( w_1=5 \) and \( w_2=1 \).

As with the identification process for the electrical parameters, it was not possible to obtain a ‘good’ fit to both the step response and the frequency domain data. If the fit was biased towards the frequency response, the peak value of the predicted step response was only 50% to 60% of the measured value. In the plot shown in Figure 6, the predicted peak of the step response is 72% of the measured value. This discrepancy is another indication that the material parameters may be dependent on excitation level.

Bode plots of the three key frequency dependent material parameters, \( \eta, d, \) and \( Y \) over the frequency range 0.01Hz to 20Hz are shown in Figure 7. A brief summary of the more noteworthy features of each follows. The electrical term \( \eta \) represents the relationship between charge density and the applied electric field. The highest charge densities will be achieved at very low frequencies, 0.1Hz and below. Above this range, the value of \( \eta \) starts to decrease (at less than one decade per decade), and the phase approaches -90 degrees, indicating that the resistive elements start to dominate the relationship between the charge density and the applied electric field. The strain coefficient \( d \), plotted in Figure 7b, represents the electromechanical coupling in terms of the strain induced at the transducer surface when an electric field is applied perpendicular to the transducer. At very low frequencies, below 0.1Hz, the magnitude of the coupling term decreases with decreasing frequency, indicating that the ionic polymer transducers considered in this work will make poor sensors and actuators for quasi-static applications. The coupling term magnitude also decreases with increasing frequency above approximately 5Hz, though the slope is more gentle than the sub 0.1Hz slope. This decrease in the strain coefficient will eventually limit the useful frequency range of ionic polymer transducers. Also, the term exhibits significant phase lag. The complex modulus shown in Figure 7c is
Figure 6: Simulated and experimental blocked force with voltage input: a) step response, b) frequency response.

Figure 7: Frequency domain plot of the key frequency dependent material parameters $\eta$, $d$, and $Y$

relatively flat across the 0.01-20Hz frequency range. The slight increase in magnitude along with a little phase lead represents the small amount of damping that was observed in the free deflection over voltage frequency response. The fact that the modulus plot is relatively flat indicates that viscoelasticity is not important in the frequency range considered.

**CONCLUSIONS**

The experiments summarized in this paper support the use of a linear model to analyze the electromechanical properties of ionic polymer transducers. An important conclusion from this work is the importance of modeling the frequency dependence of both the strain coefficient $d$ and electric permittivity $\eta$. Both parameters exhibited strong frequency dependence at frequencies below 20 Hz. This frequency dependence is important for accurate modeling of the time and frequency response of ionic polymer transducers.

**ACKNOWLEDGEMENTS**

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References


