Experimental estimate of viscoelastic properties for ionic polymer-metal composites

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We propose an experimental method for estimating the general time-dependent elastic moduli of ionic polymer-metal composites (IPMCs). The materials exhibit fast and large bending motion even when a small voltage about 1 V is applied, and are expected to be used for polymer actuators. Experimental measurements for an IPMC beam of silver plated Nafion are given to demonstrate the usefulness of the proposed method. For the IPMC beam we also present a viscoelastic model, which describes the experimental results successfully.

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I. INTRODUCTION

Hydrated electrolytic polymer membranes sandwiched between thin metal layers, which are often called ionic polymer-metal composites (IPMCs), exhibit fast and large bending motion even when a small voltage about 1 V is applied [1–4]. Nafion, which consists of fluorocarbon chains and sulfonic acid groups, is the best known as such an electrolytic polymer [1,4]. Owing to their high compliance and low density as well as large bending motion and low energy consumption, IPMCs are expected to be used as new actuator materials. However, in spite of intensive investigations, their mechanical properties such as elastic moduli and generated force, which are essential for such applications, are not well understood at present, especially because they are time dependent and difficult to measure by standard methods.

In most cases IPMCs are modeled as linear elastic materials [2,3,5,6] and their static elastic moduli obtained experimentally [2,6]. In [7,8] a viscoelastic model was given for IPMCs and the model parameters, equivalently a dynamic elastic modulus, were experimentally estimated for a polymer transducer. However, the static or dynamic elastic moduli and the time-dependent generated force of IPMCs when some voltage is applied are not well understood.

In our previous study [9], we proposed a simple method to experimentally evaluate the nominal elastic moduli and generated force of IPMC beams in their process of self-bending. In the proposed method, the shapes of the beams and vertical force at their (hinged) ends are measured and used to calculate the nominal elastic moduli and generated force although their viscoelastic properties are neglected. Experimental results for an IPMC consisting of a Nafion sheet and two thin silver plates were also given to demonstrate the method. In particular, the nominal elastic modulus and generated force had large variations in the process of self-bending.

In this Brief Report, taking account of their viscoelasticity and extending the approach of [9], we propose an experimental method for estimating the general time-dependent elastic moduli of IPMC beams. Experimental measurements for an IPMC beam of silver plated Nafion are given to demonstrate the usefulness of the proposed method. For the IPMC beam we also present a viscoelastic model, which describes the experimental results successfully.

II. THEORY

The theoretical model is shown in Fig. 1. A beam with length ℓ is clamped at the left end, and it is hinged and subjected to the vertical force $f(t)$ at the right end. The right end is at the same level as the left one but free to move in the horizontal direction. It is also assumed to be inextensible and to deform according to the general law of viscoelastic materials (e.g., [10])

$$\sigma(t) = \int_0^t E(t-\tau)\dot{\epsilon}(\tau)d\tau, \tag{1}$$

where the overdot represents differentiation with respect to time t; $E(t)$ is the general time-dependent elastic modulus; and $\sigma(t)$ and $\epsilon(t)$ are the stress and strain at time t, respectively. The distance from the left end along the beam is denote by s. For simplicity we ignore the influence of the gravitational force and variation of the distance between both ends of the beam.

When its right end is free and a constant voltage V_0 is applied, the beam bends with some curvature in the negative direction due to the electric field across it thickness. The curvature, which we denote by $\alpha(t)$, depends on the value of V_0. So the bending moment generated in the electric field is given by

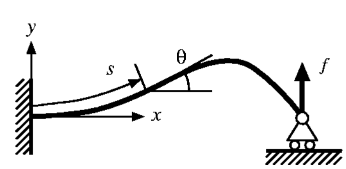

FIG. 1. Theoretical model.

$$M_0(t) = \int_0^t E(t-\tau) I \dot{\alpha}(\tau) d\tau, \qquad (2)$$

where I is the moment of inertia cross section of the beam. We also assume that the beam motion is so slow that we can neglect its inertial term.

Let M and θ be the bending moment and deflection angle of the beam, respectively. The equilibrium equation for the beam becomes

$$M'(t,s) + f(t)\cos\ \theta = 0, \qquad (3)$$

where the prime represents differentiation with respect to s. See, e.g., [11] for the derivation of Eq. (3). Here we ignored the influence of the gravitational force. The bending moment is given by

$$M(t) = \int_0^t E(t-\tau) I(\dot{\theta}'(\tau,s) + \dot{\alpha}(\tau)) d\tau. \qquad (4)$$

Substituting Eq. (4) into Eq. (3), we obtain an integro–partial-differential equation

$$\int_0^t E(t-\tau) I \dot{\theta}''(\tau,s) d\tau + f(t)\cos\ \theta = 0. \qquad (5)$$

The boundary condition is also given by

$$\theta(t,0) = 0, \quad \theta'(t,\ell) = -\alpha(t). \qquad (6)$$

Let $(x(t,s),y(t,s))$ be the position of the beam at time t and position s. Suppose that $\theta \approx 0$. Then $\theta \approx dy/dx$, $s \approx x$, and $\cos\ \theta \approx 1$, so that Eqs. (5) and (6) are, respectively, approximated by

$$\int_0^t E(t-\tau) I \dot{y}'''(\tau,x) d\tau + f(t) = 0 \qquad (7)$$

and

$$y(t,0) = y(t,\ell) = y'(t,0) = 0, \quad y''(t,\ell) = -\alpha(t), \qquad (8)$$

where the prime represents differentiation with respect to x. Performing the Laplace transformation in Eqs. (7) and (8), we obtain

$$p\hat{E}(p) I \hat{y}'''(p,x) + \hat{f}(p) = 0 \qquad (9)$$

and

$$\hat{y}(p,0) = \hat{y}(p,\ell) = \hat{y}'(p,0) = 0, \hat{y}''(p,\ell) = -\hat{\alpha}(p), \qquad (10)$$

where $\hat{E}(p)$, $\hat{y}(p,x)$, $\hat{f}(p)$, and $\hat{\alpha}(p)$ are the Laplace transforms of $E(t)$, $y(t,s)$, $f(t)$, and $\alpha(t)$, respectively, and are given by

$$\hat{E}(p) = \int_0^\infty E(t) e^{-pt} dt,$$

$$\hat{y}(p,x) = \int_0^\infty y(t,x) e^{-pt} dt,$$

$$\hat{f}(p) = \int_0^\infty f(t) e^{-pt} dt. \qquad (11)$$

Here we assumed that the beam is not initially deflected, i.e., $y(0,x)=0$. The solution of Eq. (9) with $\hat{y}(p,0)=\hat{y}(p,\ell)=\hat{y}'(p,0)=0$ is given by

$$\hat{y}(p,x) = \frac{\hat{f}(p) x^2 (\ell - x)}{6p\hat{E}(p) I}. \qquad (12)$$

By Eqs. (2) and (12) the Laplace transforms of $\alpha(t)$ and $M_0(t)$ become

$$\hat{\alpha}(p) = -\hat{y}''(p,\ell) = \frac{2\hat{f}(p)\ell}{3p\hat{E}(p) I} \qquad (13)$$

and

$$\hat{M}_0(p) = p\hat{E}(p) I \hat{\alpha}(p) = \frac{2}{3}\hat{f}(p)\ell, \qquad (14)$$

respectively. Here we assumed that $\alpha(0)=0$ and $M_0(0)=0$. From Eqs. (12)–(14) we obtain

$$y(t,x) = \frac{\alpha(t)}{4\ell} x^2 (\ell - x) \qquad (15)$$

and

$$M_0(t) = \frac{2}{3} f(t)\ell. \qquad (16)$$

Our approach to estimate the generated bending moment $M_0(t)$, curvature $\alpha(t)$, and time-dependent elastic modulus $E(t)$ in experiments is as follows. Let t_i, $i=1,2,\ldots$, be a sequence of times at the same interval such that $t_{i+1}-t_i=\Delta t$. We measure the load $f(t_i)$ and deflections of the beam $y_j(t_i)$ at some, say $N(>1)$, points $x=x_j$, $j=1,2,\ldots,N$, at $t=t_i$, $i=1,2,\ldots$. $M_0(t_i)$, $i=1,2,\ldots$, are obtained from Eq. (16) directly, and $\alpha(t_i)$, $i=1,2,\ldots$, are calculated by applying the least squares method to Eq. (15) with the data $f(t_i)$ and $y_j(t_i)$. The Laplace transformation is numerically performed to compute $\hat{f}(p)$ and $\hat{\alpha}(p)$, and then $\hat{E}(p)$ is obtained from Eq. (13). The general time-dependent elastic modulus $E(t)$ can be obtained, for example, by performing the inverse Laplace transformation for $\hat{E}(p)$ numerically.

III. EXPERIMENTS

Figure 2 is an illustration of our experimental setup. We prepared IPMC beams of silver plated Nafion through the method described in Sec. IV A of [9]. An IPMC beam was clamped horizontally near the left end, and its right end was placed slightly above the stand of balance so that the gap between it and the stand is negligibly small. The distance between the clamping point and the hinged end of the beam was 16 mm. When a voltage of 1 V is applied, the beam starts bending. The shape of the beam was tracked as a function of time with the image processing instrument CV-2000

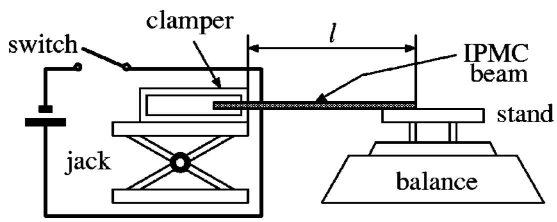

FIG. 2. Experimental setup.

(Keyence Corporation, Osaka, Japan). The positions (x_i, y_i), $i=1,\ldots,11$, of 11 representative points set on the beam including both the ends were automatically recorded every 122 ms. The vertical force f was also measured with a balance simultaneously at the same interval time of 122 ms. We did not observe plastic deformation of the beam for time periods shorter than 60 s.

Figure 3 shows a set of experimental results for a specimen. Here the Simpson rule was used as the numerical integrator to compute the Laplace transform in Fig. 3(d), in which numerical integration yields a smooth curve. Figures 3(a)–3(c) should be, respectively, compared with the corresponding results of Figs. 5, 8, and 9 in [9]. Qualitative agreement between both results is found although the experiment was performed for a different sample under different circumstances (e.g., temperature) from those in [9]. We note that mechanical properties of our material were very distinct among individual samples since Nafion does not have a perfect crystal structure. From Fig. 3(d) we see that the dependence of stress on instantaneous strain velocity is negligible since $\hat{E}(p)$ seems to tend to zero as $p \to \infty$. The theoretical prediction of Eq. (15) and the experimental measurement for the shape of the IPMC beam at $t=30.012$ s are shown in Fig. 4. A fine agreement between them is found. Note that we do not have to assume the value of $E(t)$ to obtain the theoretical result.

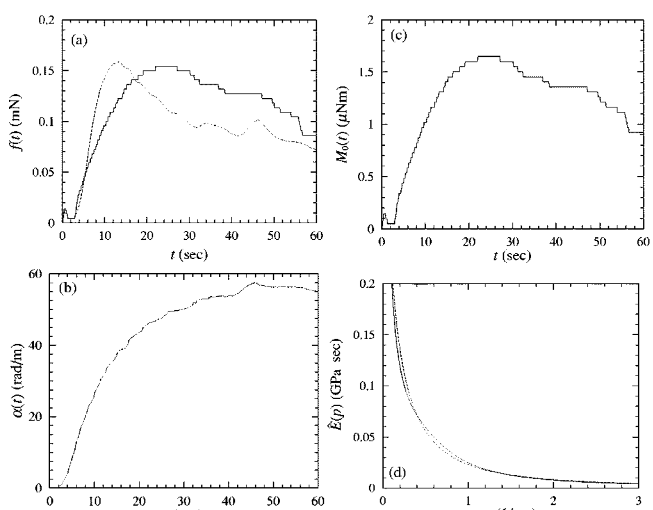

FIG. 3. Experimental results: (a) Vertical force at the (hinged) end of the beam; (b) equilibrium curvature; (c) generated bending moment; (d) Laplace transform of elastic modulus. The computation results of Eqs. (21) and (18) with the estimated parameter values are also drawn as broken curves in (a) and (d).

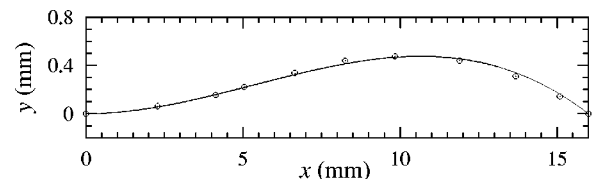

FIG. 4. Comparison between the theoretical prediction (solid curve) and experimental measurement (○) for the shape of the IPMC beam at $t=30.012$ s.

As stated at the end of Sec. II, by performing the inverse Laplace transformation, we can obtain the general time-dependent elastic modulus $E(t)$. However, precise inverse Laplace transformation is generally difficult and this is the case for the result of Fig. 3(d). Instead of doing so, we propose a viscoelastic model for the IPMC beam in the next section.

IV. VISCOELASTIC MODEL FOR THE IPMC BEAM

The Laplace transform of the elastic modulus in Fig. 3(d) is redrawn in Fig. 5 after it is multiplied by the variable p. The curve has a peak and it seems to tend to some constant as $p \to 0$ and $p \to \infty$. However, typical viscoelastic models like the Maxwell and Voigt materials [10] and the Golla-Hughes-McTavish (GHM) model used in [7,8] do not represent all the characteristics.

So we present a simple model having the properties

$$E(t) = E_0 + (E_1 t - E_2)e^{-\lambda t}, \tag{17}$$

where $E_0 > E_2 > 0$ and $E_1, \lambda > 0$. The Laplace transform of Eq. (17) becomes

$$\hat{E}(p) = \frac{E_0}{p} + \frac{E_1}{(p+\lambda)^2} - \frac{E_2}{p+\lambda}. \tag{18}$$

We easily see that

$$\lim_{p\to 0} \hat{E}(p) = \infty, \quad \lim_{p\to\infty} \hat{E}(p) = 0 \tag{19}$$

and

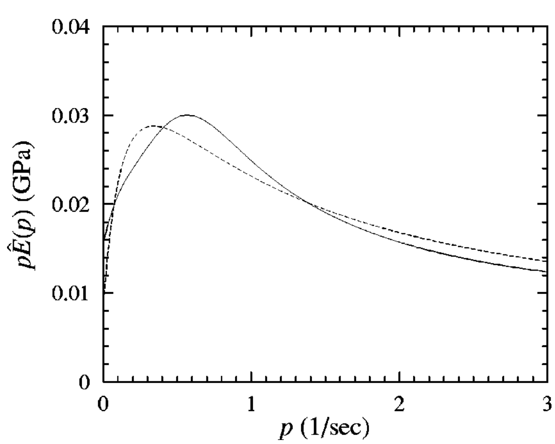

FIG. 5. Redrawing of the experimental result of Fig. 3(d). The computation result of Eq. (18) with the estimated parameter values is also drawn as a broken curve.

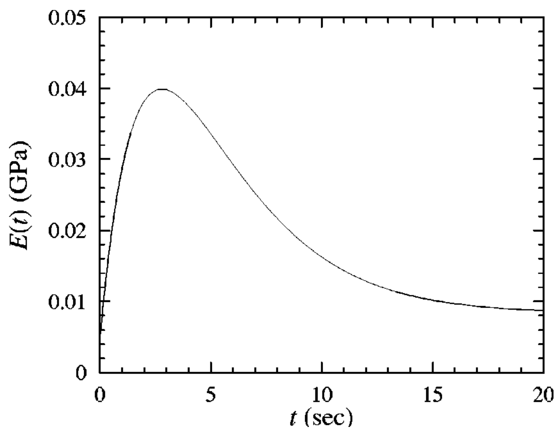

FIG. 6. Computed time-dependent elastic modulus (17) with the estimated parameter values.

$$\lim_{p\to 0} p\hat{E}(p) = E_0, \quad \lim_{p\to\infty} p\hat{E}(p) = E_0 - E_2. \tag{20}$$

Moreover, $p\hat{E}(p)$ attains its maximum $E_0+(E_1-E_2\lambda)^2/4E_1\lambda$ at $p=(E_1-E_2\lambda)\lambda/(E_1+E_2\lambda)$. Thus, the model (17) has key properties observed in Figs. 3(d) and 5.

Using the experimental data of Fig. 5 and applying the least squares technique to $p\hat{E}(p)$, we estimate

$$E_0 = 8.33 \times 10^{-3}(\text{GPa}), \quad E_1 = 3.35 \times 10^{-2}(\text{GPa}),$$

$$E_2 = 4.06 \times 10^{-3}(\text{GPa}), \quad \lambda = 0.373 \ (1/\text{s}).$$

Figure 6 shows the time-dependent elastic modulus $E(t)$ computed by Eq. (17) for the above parameter values. We observe that $E(t)$ has a peak at $t\approx 2.8$ s and gradually decreases as t increases after that. This can also explain why the beam deformed largely after the peak of the generated bending moment. From Eqs. (9), (15), and (17) we also obtain

$$\begin{aligned} f(t) &= \frac{3I}{2\ell}\left(\int_0^t E(t-\tau)\dot{\alpha}(\tau)d\tau\right) \\ &= \frac{3I}{2\ell}\left(\int_0^t [(E_1+E_2\lambda)-E_1\lambda(t-\tau)] \right. \\ &\quad \left. \times e^{-\lambda(t-\tau)}\alpha(\tau)d\tau + (E_0-E_2)\alpha(t)\right). \end{aligned} \tag{21}$$

The computation results of Eqs. (18) and (21) are also drawn in Figs. 3(a), 3(d), and 5. In Fig. 3(a) the data of Fig. 3(b) were used to compute Eq. (21). Especially in Figs. 3(d) and 5, we found good agreement between the computations and experimental measurements.

V. CONCLUDING REMARKS

In this Brief Report, taking into account their viscoelasticity, we extended the experimental method of [9] for estimating general time-dependent elastic moduli of IPMC beams. The proposed method was demonstrated in experiments of an IPMC beam of silver plated Nafion and a viscoelastic model was presented to describe the experimental results. We hope that our method and model will prove useful for understanding physical properties of IPMCs and other polymers.

One challenging issue is to construct a theory explaining the relationship between bending motions and applied voltages for IPMCs. It is necessary for manufacturing and controlling IPMC actuators. Some attempt was performed in [2,7,8] but that approach was based on a linear circuit model [2] although nonlinear behavior was observed in our preliminary experiments. Our research in this direction is in progress.

[1] K. Oguro, K. Asaka, and H. Takenaka, *Proceedings of the Fourth International Symposium on Micro Machine and Human Science* (Economic Affairs Bureau, Nagoya, 1993), p. 39.

[2] R. Kanno, S. Tadokoro, T. Takamori, M. Hattori, and K. Oguro, *Proceedings of the 1996 IEEE International Conference of Robotics and Automation* (IEEE, Piscataway, NJ, 1996), p. 219.

[3] M. Shahinpoor, Y. Bar-Cohen, J. O. Simpson, and J. Smith, Smart Mater. Struct. **7**, R15 (1998).

[4] S. Nemat-Nasser, J. Appl. Phys. **92**, 1 (2002).

[5] K. Kim and M. Shahinpoor, Proc. SPIE **4329**, 223 (2001).

[6] S. Nemat-Nasser and J. Y. Li, J. Appl. Phys. **87**, 3321 (2000).

[7] K. M. Newbury and D. J. Leo, J. Intell. Mater. Syst. Struct. **14**, 333 (2003).

[8] K. M. Newbury and D. J. Leo, J. Intell. Mater. Syst. Struct. **14**, 343 (2003).

[9] H. Tamagawa, K. Yagasaki, and F. Nogata, J. Appl. Phys. **92**, 7614 (2002).

[10] J. D. Ferry, *Viscoelastic Properties of Polymers* (Wiley, New York, 1960).

[11] B. D. Coleman and E. H. Dill, J. Acoust. Soc. Am. **91**, 2663 (1992).