

On structural models for ionic polymer metal composites

Alain Boldini^a, Lorenzo Bardella^b, and Maurizio Porfiri^{a,c}

^aDepartment of Mechanical and Aerospace Engineering, New York University Tandon School of Engineering, Six MetroTech Center, Brooklyn, NY 11201, USA

^bDepartment of Civil, Environmental, Architectural Engineering and Mathematics, University of Brescia, via Branze 43-45, 25123, Brescia, Italy

^cDepartment of Biomedical Engineering, New York University Tandon School of Engineering, Six MetroTech Center, Brooklyn, NY 11201, USA

ABSTRACT

Ionic polymer metal composites (IPMCs) are a class of soft electroactive polymers. IPMCs comprise a soft ionic polymer core, on which two stiff metal electrodes are plated. These active materials exhibit large bending upon the application of a small driving voltage across their electrodes, both in air and in aqueous environments. In a recent work, we presented compelling theoretical and numerical evidence suggesting that ionic polymer membranes exhibit complex deformation paths that are neglected by reduced-order structural models. While beam theories (including Euler-Bernoulli, Timoshenko, and most higher-order shear deformation models) would suggest vanishing through-the-thickness deformation, we discovered the onset of localized multiaxial deformation that reverberates into axial stretching. Building upon this effort, here we investigate the role of the electrodes on IPMCs deformation. We establish a novel structural theory for IPMCs, based on the Euler-Bernoulli kinematics enriched with a through-the-thickness deformation in the ionic polymer that is obtained from a Saint-Venant-like problem. We compare the results of our theory against finite element simulations modelling the IPMC as a two-dimensional continuum. The comparison indicates that our theory can predict the macroscopic displacement of the IPMC, along with localized deformation in the ionic polymer at the interface with the electrodes. This work paves the way to the development of more sophisticated structural theories for IPMCs and analogous active materials, affording an accurate description of deformations at a limited computational cost.

Keywords: beam theories, electrochemistry, non-uniform bending, reduced-order modeling, through-the-thickness deformation

1. INTRODUCTION

Within the range of electroactive polymers, ionic polymer metal composites (IPMCs) present unique characteristics that have attracted the attention of scientists and engineers for more than twenty years.^{1,2} The layout of these materials is that of a traditional sandwich structure,³ where a soft, ionic polymer core is plated by two stiffer metal skins that serve as electrodes.⁴ In the ionic polymer, anions are attached to the polymeric backbone, while cations can move throughout the solution that permeates the IPMC.^{1,2} When a voltage is applied across the electrodes, charge redistribution modulates osmotic pressure and Maxwell stress.⁵ Together, osmotic pressure and Maxwell stress define the eigenstress of the ionic polymer, which, through equilibrium with the mechanical stress, yields IPMC deformation. Dually, when the IPMC is mechanically deformed, the concentrations of cations and anions vary through the ionic polymer, thereby generating a voltage that can be sensed at the electrodes.^{1,2} The possibility of utilizing IPMCs underwater, along with their large compliance and low actuation voltage,^{1,2} make them viable actuators, sensors, and energy harvesters from soft robotics⁶ to biomedical engineering.⁷

Despite these advantages, practical, real-world applications of IPMCs are still very limited due to a number of technical hurdles, such as low blocking force and shape constraints,^{1,2} which hinder the use of IPMCs out of laboratory settings. A potential avenue to address these issues entails additive manufacturing of ionic polymers,

Further author information: (Send correspondence to M.P.)

M.P.: E-mail: mporfiri@nyu.edu, Telephone: 1 646 997 3681

recently proposed by the groups of Aureli, Leang, and Kim.^{8–10} This manufacturing technique allows engineers to tailor the physical and geometrical properties of ionic polymers, paving the way to novel, enticing free-form designs. To fully unleash the potential of this new manufacturing technique, one should rely on inverse design methodologies, grounded in low-cost, and yet accurate models of IPMC mechanics and electrochemistry.

While the literature on reduced-order circuit models of IPMC electrochemistry is well developed, effective tools to study IPMC mechanics remain relatively untapped, whereby most analyses are based on low-order structural theories.^{5,11,12} As first proposed in our recent work,¹³ the hypotheses underlying these theories are unlikely to be verified in IPMCs, where the variation of cations concentration and voltage along the thickness begets a complex form of the eigenstress. Therein, we suggested that IPMCs undergo multiaxial deformations that affect their bending, such that classical beam- and plate-like theories cannot predict IPMC actuation. More specifically, the longitudinal eigenstress acting on the IPMC is modified by the through-the-thickness strain, which is localized at the interface between the ionic polymer and electrodes, in correspondence to the electrochemical boundary layers. Strain localization is due to Maxwell stress, whereby the two charged electrodes, attracting each other as in a capacitor, elicit a contraction of the IPMC along its thickness. Under general working hypotheses, we established a finite element (FE) model of the nonlinear, fully coupled governing equations proposed by Cha and Porfiri⁵ by implementing a user-defined two-dimensional element in ABAQUSTM. However, FE simulations can hardly be utilized as tools for inverse design and optimization, as they are plagued by excessive computational burden.

Here, we seek to fill this gap in knowledge by developing a new structural theory that can provide accurate and affordable predictions of multiaxial deformations in slender IPMCs. Our work tackles general boundary conditions that may yield shear deformations and examines the presence of non-zero thickness electrodes - none of these aspects has never been considered in the technical literature of IPMC actuation. Building upon the classical Euler-Bernoulli beam theory,¹⁴ we propose an enriched kinematics that incorporates through-the-thickness strain localization in the vicinity of the ionic polymer-electrode interfaces. Strain localization is captured through an ad-hoc shape function, obtained from the analytical solution of a Saint-Venant-like problem for uniform bending through matched asymptotic expansions. Such shape function is modulated by an additional field to the longitudinal and transverse displacements of the midaxis within Euler-Bernoulli kinematics. The governing equations are derived from a total potential energy that encompasses the strain energy of ionic polymer and electrodes and the work done by the eigenstress. This yields a system of three coupled partial differential equations describing IPMC deformation as a function of the applied voltage across the electrodes. By comparing the predictions of this new structural theory, classical Euler-Bernoulli beam theory, and FE simulations, we finally draw general conclusions on the viability of the proposed approach and its added value with respect to beam models from literature.

This paper is organized as follows. In Section 2, we show the steps to derive the new structural theory. In Section 3, we summarize our FE framework, developed in our previous work.¹³ Section 4 illustrates the comparison between classical and enriched Euler-Bernoulli beam theories in a prototypical problem, assessing their accuracy against FE simulations. Finally, we summarize the main results of our work in Section 5, where we point at future research directions that can address the limitations of the present study.

2. DERIVATION OF THE NOVEL STRUCTURAL THEORY

Toward the definition of a physically-based reduced-order model for multiaxial deformations during IPMC actuation, we start by considering the continuum model proposed by Cha and Porfiri.⁵ Within this model, the ionic polymer is considered as a membrane with a fixed, negative, uniform charge distribution. The mechanics and electrochemistry of the ionic polymer are fully described by the displacement \mathbf{u} , the counterions' concentration C , and the electric potential ψ . These quantities are determined by conservation laws (balance of linear momentum, mass balance, and Gauss law) and by constitutive equations, derived in a thermodynamically-consistent framework through the definition of a suitable Helmholtz free-energy density¹⁵ for the ionic polymer. While electrodes enter the continuum model in Cha and Porfiri⁵ only to establish the boundary conditions, we assume that they behave as linear isotropic perfectly conductive materials that are impenetrable to ionic fluxes.

2.1 Exact solution for IPMC electrochemistry

Similar to our previous endeavor,¹³ we put forward several hypotheses to simplify the problem. Specifically, we consider a beam-like rectangular IPMC, with an ionic polymer cord of thickness $2h$, thin electrodes of thickness $e \ll h$, and length $l \gg h$. We assume that the IPMC undergoes small, plane-strain deformations in the $X - Y$ plane, where X runs from one end section through the IPMC length, and Y points through the thickness from the IPMC midaxis (see Fig. 1). Further, we hypothesize that the mechanics has no effect on the electrochemistry, such that the electrochemistry of the IPMC becomes completely independent of its mechanics.

Within this framework, we can solve electrochemistry once for all, independently of IPMC deformations, and then use its results to compute the eigenstress consisting of osmotic pressure and Maxwell stress. To this end, we consider a parallel-plate approximation of the electrochemistry,^{5,16} whereby we assume that variations of the electrochemical variables (counterions' concentration and voltage) along the axis X of the IPMC are negligible compared to their variations in the transversal direction Y . Under this hypothesis, we reduce ionic polymer electrochemistry to a one-dimensional Poisson-Nernst-Planck (PNP) system¹⁷ along the ionic polymer thickness.

The PNP system is a singularly-perturbed boundary value problem,¹⁸ which generates boundary layers of net charge and electric potential at the ionic polymer-electrode interfaces.¹⁷ This problem can be solved through the method of asymptotic expansions,^{18,19} which provides, as a function of the external voltage, the time (t) evolution of counterions' concentration $C(Y, t)$ and electric potential $\psi(Y, t)$. Further details about the solution of the PNP system in IPMCs through the method of asymptotic expansions can be found in Porfiri.¹⁹

2.2 Saint-Venant-like problem for uniform bending

As a fundamental step to incorporate through-the-thickness deformations within our structural model, we summarize herein the solution of the Saint-Venant-like problem for uniform bending studied in our previous work.¹³ Therein, we consider a simply supported IPMC, with zero-thickness electrodes, constrained on its midaxis. From the electrochemistry, we evaluate the in-plane eigenstress components $\sigma_{0_{XX}}(Y)$, $\sigma_{0_{YY}}(Y)$, and $\sigma_{0_{XY}}(Y)$ associated to osmotic pressure and Maxwell stress as

$$\sigma_{0_{XX}}(Y) = -\mathcal{RT}(C(Y) - C_0) - \frac{\epsilon}{2} \left(\frac{\partial \psi(Y)}{\partial Y} \right)^2, \quad (1a)$$

$$\sigma_{0_{YY}}(Y) = -\mathcal{RT}(C(Y) - C_0) + \frac{\epsilon}{2} \left(\frac{\partial \psi(Y)}{\partial Y} \right)^2, \quad (1b)$$

$$\sigma_{0_{XY}}(Y) = 0, \quad (1c)$$

where we have dropped the explicit dependence on time for ease of notation. Here, \mathcal{R} is the universal gas constant, \mathcal{T} indicates the absolute temperature, C_0 is the uniform concentration of fixed negative charges in the membrane, and ϵ is the dielectric constant of the ionomer.

Within this framework, we assume a linear longitudinal strain along the IPMC thickness

$$\varepsilon_{XX}(Y) = -kY + \varepsilon_0, \quad (2)$$

where k indicates the IPMC curvature and ε_0 the average longitudinal strain over the section. The equilibrium condition along the IPMC thickness, assuming negligible shear strain and stress, reads

$$\frac{\partial}{\partial Y} \sigma_{YY}(Y) = \frac{\partial}{\partial Y} [\lambda_L \varepsilon_{XX}(Y) + (\lambda_L + 2\mu_L) \varepsilon_{YY}(Y) + \sigma_{0_{YY}}(Y)] = 0, \quad (3)$$

where λ_L and μ_L are the Lamé parameters for the ionic polymer.²⁰ These parameters arise from the linearization of any isotropic constitutive model for small deformations.¹⁵

As IPMC is constrained on its midaxis and no external force is applied, the normal stress at the electrodes' surface should vanish. Combining these boundary conditions with Eq. (3), we obtain that σ_{YY} must be pointwise zero along Y , from which we can find the through-the-thickness strain

$$\varepsilon_{YY}(Y) = -\frac{1}{\lambda_L + 2\mu_L} [\lambda_L \varepsilon_{XX}(Y) + \sigma_{0_{YY}}(Y)]. \quad (4)$$

Contrary to the longitudinal strain in Eq. (2), the through-the-thickness strain directly depends on the eigenstress. Thus, the model predicts strain localization at the interfaces between the ionic polymer and electrodes, in correspondence of the electrochemical boundary layers. From this quantity, we can define the transverse displacement with respect to the IPMC midaxis as

$$\bar{f}(Y) = \int_0^Y \varepsilon_{YY}(\tilde{Y}) d\tilde{Y}. \quad (5)$$

Here and henceforth, we utilize this variable as a further shape function within our structural theory to account for through-the-thickness deformations.

The complete form of ε_{YY} is obtained by computing ε_0 and k in Eq. (2) by integrating the longitudinal stress along the Y axis to obtain the resultant longitudinal force and bending moment, which must vanish as no external force is applied on the IPMC. Such a derivation is provided in Boldini and Porfiri.¹³

2.3 Total potential energy

To obtain the governing equations for the structural model, we rely on a total potential energy per unit width, encompassing the strain energies of ionic polymer (U_{ion}) and electrodes (U_{el}), along with the work performed by the eigenstress \mathcal{W}_{eig} ,

$$U = U_{\text{mec}} + U_{\text{el}} + \mathcal{W}_{\text{eig}}. \quad (6)$$

The strain energy of the ionic polymer corresponds to the strain energy of a linear isotropic elastic material, with

$$U_{\text{mec}} = \int_{\mathcal{S}^i} W_{\text{mec}} dS = \frac{1}{2} \int_{\mathcal{S}^i} [(\lambda_L + 2\mu_L) [(\varepsilon_{XX}^i)^2 + (\varepsilon_{YY}^i)^2] + 2\lambda_L \varepsilon_{XX}^i \varepsilon_{YY}^i + \mu_L (\gamma_{XY}^i)^2] dS, \quad (7)$$

where \mathcal{S}^i is the ionic polymer domain in the $X - Y$ plane and $\gamma_{XY}^i = 2\varepsilon_{XY}^i$.

We assume that the electrodes are sufficiently slender that they can be modelled as Euler-Bernoulli beams, leading to

$$U_{\text{el}} = \frac{1}{2} \int_{\mathcal{S}^{\text{el}^+}} Y_{\text{el}}' (\varepsilon_{XX}^{\text{el}^+})^2 dS + \frac{1}{2} \int_{\mathcal{S}^{\text{el}^-}} Y_{\text{el}}' (\varepsilon_{XX}^{\text{el}^-})^2 dS, \quad (8)$$

where $\mathcal{S}^{\text{el}^\pm}$ are the domains in the $X - Y$ plane of anode and cathode, respectively, $\varepsilon_{XX}^{\text{el}^\pm}$ is the longitudinal strain of the anode and cathode, respectively, and $Y_{\text{el}}' = Y_{\text{el}} / (1 - \nu_{\text{el}}^2)$ is the longitudinal modulus of the electrodes resulting from the plane-strain constraint along with the condition $\sigma_{YY} = 0$ therein, where Y_{el} and ν_{el} are the electrodes' Young modulus and Poisson ratio, respectively.

Finally, we write the work performed by the eigenstress as

$$\mathcal{W}_{\text{eig}} = \int_{\mathcal{S}^i} (\sigma_{0XX} \varepsilon_{XX}^i + \sigma_{0YY} \varepsilon_{YY}^i) dS. \quad (9)$$

2.4 Kinematics of the proposed structural theory

The kinematics of our new structural theory is built on the classical Euler-Bernoulli beam theory,¹⁴ which assumes the following form for the ionic polymer displacement field in X ($u^i(X, Y)$) and Y ($w^i(X, Y)$):

$$u^i(X, Y) = u_0^i(X) - Y w_{0,X}^i(X), \quad (10a)$$

$$w^i(X, Y) = w_0^i(X). \quad (10b)$$

Here and henceforth, pedices after a comma indicate partial derivatives. Within this model, we cannot capture any shear or through-the-thickness strain, which are likely expected to influence IPMC actuation.¹³

To bridge this gap, we define an enriched Euler-Bernoulli beam theory, where we account for the through-the-thickness deformation of the ionic polymer. We hypothesize that such a deformation is not significantly affected by electrodes and shear deformations away from the constraints, unless we consider “severe” boundary

conditions.²¹ Thus, we include the function $\bar{f}(Y)$, encapsulating the transverse displacement of the ionic polymer with respect to the IPMC midaxis, within our structural model. We note that $\bar{f}(Y)$ may vary with time. To partially account for the effect of boundary conditions, we weigh the transverse displacement $\bar{f}(Y)$ from uniform bending by a function $p(X)$. Should we consider a shear-free condition with electrodes of negligible thickness, we would have $p(X) = 1$ throughout the IPMC span.

Therefore, the kinematics of our new structural theory for the ionic polymer reads

$$u^i(X, Y) = u_0^i(X) - Yw_{0,X}^i(X), \quad (11a)$$

$$w^i(X, Y) = w_0^i(X) + p(X)\bar{f}(Y). \quad (11b)$$

The solution of this structural theory requires the determination of three field variables along X : the midaxis longitudinal ($u_0^i(X)$) and transverse ($w_0^i(X)$) displacements, and the term modulating the ionomer through-the-thickness deformation along the IPMC axis ($p(X)$). This kinematics affords non-zero through-the-thickness and shear strain fields, which are expected to play an important role in macroscopic IPMC deformations.¹³

The deformation of the electrodes, which are modeled as Euler-Bernoulli beams, is determined by the displacement of the ionic polymer by imposing continuity of the displacements at the ionic polymer-electrode interfaces. Thus, we obtain the longitudinal strain of the electrodes as

$$\varepsilon_{XX}^{\text{el}\pm}(X, Y) = u_{0,X}^i(X) - Yw_{0,XX}^i(X) - (Y \mp h)p_{,XX}(X)\bar{f}(\pm h). \quad (12)$$

By substituting the expressions in Eqs. (11) and (12) into Eqs. (6), (7), (8), and (9), we derive the total potential energy for the enriched structural theory.

3. FINITE ELEMENT SIMULATIONS

To determine whether classical Euler-Bernoulli beam theory and its enriched version proposed herein can accurately describe IPMC deformations under actuation, we perform FE simulations of the fully coupled, nonlinear mechanics and electrochemistry of IPMCs, which we consider as our ground truth. Specifically, we utilize the plane-strain user defined element (UEL) implemented in the commercial FE software ABAQUSTM, as developed in our previous work.¹³

Our FE formulation relies on a standard Galerkin approximation²² of a weak formulation of the balance equations in Cha and Porfiri,⁵ without steric effects. The residuals of these equations are computed through a Gaussian integration scheme. We utilize a plane-strain bi-quadratic element with eight nodes, which implements a full integration scheme. To model the inert, isotropic electrodes, which were not considered in our original FE model,¹³ we employ the 8-noded bi-quadratic plane-strain built-in elements CPE8. Further details on the implementation of FE simulations can be found in Boldini and Porfiri.¹³

4. RESULTS AND DISCUSSION

We contrast the exact results of the proposed structural theory and of classical Euler-Bernoulli beam theory with FE simulations. Specifically, we consider a prototypical problem, shown in Fig. 1, with non-zero thickness electrodes and shear deformations, where the end cross-sections of the IPMC are constrained to move longitudinally, without any through-the-thickness contraction, while longitudinal rigid body motion is blocked at the center of the IPMC, such that

$$u_0^i\left(\frac{l}{2}\right) = 0, \quad (13a)$$

$$w_0^i(0) = 0, \quad (13b)$$

$$w_0^i(l) = 0, \quad (13c)$$

$$p(0) = 0, \quad (13d)$$

$$p(l) = 0. \quad (13e)$$

With these boundary conditions, we determine an analytical solution for the problem.

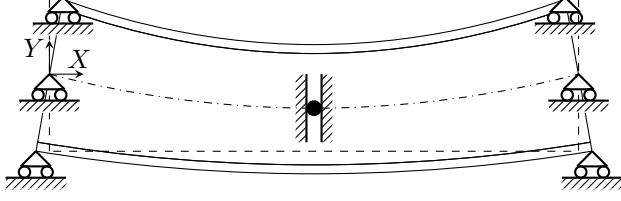


Figure 1. Schematics of the model problem. The frame X - Y sits on the left end section of the IPMC, with the X -axis aligned to the IPMC midaxis and the Y -axis along the thickness in the undeformed configuration. The end sections of the IPMC are constrained to only move longitudinally. The midpoint of the IPMC midaxis is constrained to only move transversally.

4.1 Analytical solution

From the total potential energy in which we have substituted the kinematics of our structural theory from Eqs. (11) and (12), we can find an analytical solution for the sample problem through minimization.²³ By imposing that the first variation with respect to the three unknown field variables $u_0^i(X)$, $w_0^i(X)$, and $p(X)$ is equal to zero, we obtain a system of partial differential Euler-Lagrange equations, along with natural boundary conditions complementing the essential boundary conditions in Eq. (13).

By solving the resulting system, we establish

$$u_0^i(X) = -\frac{1}{A_1} \left\{ \mathcal{I}_Y(\sigma_{0_{xx}}) \left(X - \frac{l}{2} \right) + A_2 \left[\mathcal{P}(X) - \mathcal{P} \left(\frac{l}{2} \right) \right] + A_3 \left[p_{,X} - p_{,X} \left(\frac{l}{2} \right) \right] \right\}, \quad (14a)$$

$$w_0^i(X) = \frac{1}{B_1} \left\{ \mathcal{I}_Y(\sigma_{0_{xx}} Y) \frac{X}{2} (X - l) - B_2 \left[\int_0^X \mathcal{P}(\tilde{X}) d\tilde{X} - \frac{X}{l} \int_0^l \mathcal{P}(\tilde{X}) d\tilde{X} \right] - B_3 p(X) \right\}. \quad (14b)$$

Here, we have defined $\mathcal{P}(X) = \int_0^X p(\tilde{X}) d\tilde{X}$ and the operator $\mathcal{I}_Y(\cdot) = \int_{-h}^h (\cdot) dY$. The constants in the solution are $A_1 = 2[(\lambda_L + 2\mu_L)h + Y'_{el}e]$, $A_2 = \lambda_L \mathcal{I}_Y(\bar{f}_{,Y})$, $A_3 = -\frac{1}{2} Y'_{el} e^2 \mathcal{I}_Y(\bar{f}_{,Y})$, $B_1 = \frac{2}{3} \left\{ (\lambda_L + 2\mu_L) + Y'_{el} \left[\left(1 + \frac{e}{h}\right)^3 - 1 \right] \right\}$, $B_2 = -\lambda_L \mathcal{I}_Y(\bar{f}_{,Y} Y)$, and $B_3 = \frac{1}{2} Y'_{el} e^2 \left(h + \frac{2}{3} e \right) [\bar{f}(h) + \bar{f}(-h)]$.

The field variable $p(X)$ is obtained as

$$p(X) = C_1 e^{\sigma(X-l)} \cos(\omega X) + C_2 e^{\sigma(X-l)} \sin(\omega X) + C_3 e^{-\sigma X} \cos(\omega X) + C_4 e^{-\sigma X} \sin(\omega X) + \bar{K}, \quad (15)$$

where σ and ω are the real and imaginary parts of the complex eigenvalues arising from the partial differential equation for $p(X)$, \bar{K} is a function of time that only depends on geometrical and material properties of ionic polymer and electrodes, along with the eigenstress in Eq. (1), while the constants C_1 , C_2 , C_3 , and C_4 are determined by appropriate essential and natural boundary conditions.

4.2 Comparison of structural theories predictions against FE simulations

We consider a step voltage of value $\bar{V} = 0.1285$ V, analyzing the steady-state response at 7.5 s. We utilize the same material parameters as in our previous work,¹³ selecting a Poisson ratio for the ionomer of 0.45. We consider a geometry with $h = 100 \mu\text{m}$ and $l = 20h$. For the electrodes, we consider a thickness of $2.3 \mu\text{m}$, Young modulus of 168×10^9 Pa, and a Poisson ratio of 0.39.^{5,24} Within the FE simulations, we use 36,000 elements, of which 4,000 for the electrodes. To resolve the boundary layers at the ionic polymer-electrode interfaces, the ionic polymer mesh is refined as it approaches the interfaces.

First of all, we consider the transverse displacement of the midaxis, see Fig. 2. The classical Euler-Bernoulli beam cannot even predict the sign of the IPMC curvature. By including the enrichment function, we instead demonstrate reasonably good agreement with FE simulations along the entire IPMC length. Specifically, both enriched Euler-Bernoulli beam theory and FE simulations anticipate back-relaxation of the IPMC – that is, bending toward the cathode after an initial deformation toward the anode.^{25,26} On the contrary, the classical Euler-Bernoulli beam theory predicts a qualitatively different result, where no back-relaxation occurs.

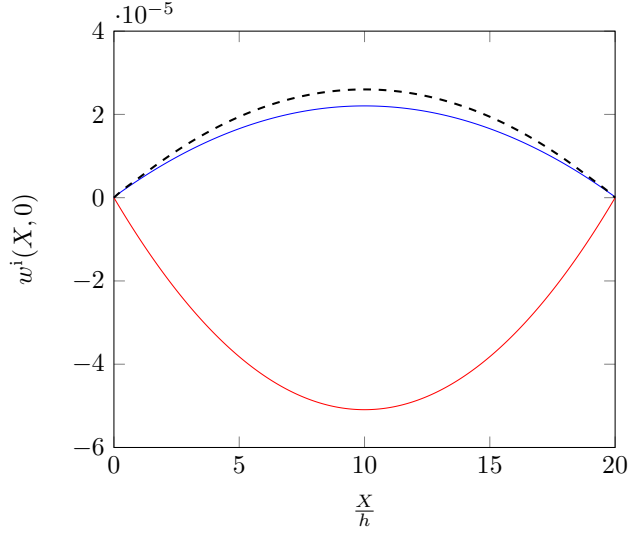


Figure 2. Transverse displacement of the midaxis of the IPMC. We compare the results from enriched Euler-Bernoulli beam theory (blue solid line), classical Euler-Bernoulli beam theory (red solid line), and FE simulations (black dashed line).

We further expand on the comparison by studying the longitudinal and through-the-thickness strain along the midspan section of the ionic polymer. Figure 3 shows the results for the steady-state longitudinal strain in the ionic polymer at the midspan section. As one would expect from the previous results, we register a qualitative difference between classical Euler-Bernoulli beam theory and FE simulations. While both display a linear longitudinal strain, the slope of these lines, which is associated to the curvature (see Eq. (2)), has a different sign. This discrepancy can be associated to the inability of classical Euler-Bernoulli beam theory to capture back-relaxation. More importantly, the results of the enriched structural theory are in close agreement with those of FE simulations, even if we acknowledge a small discrepancy approaching the anode.

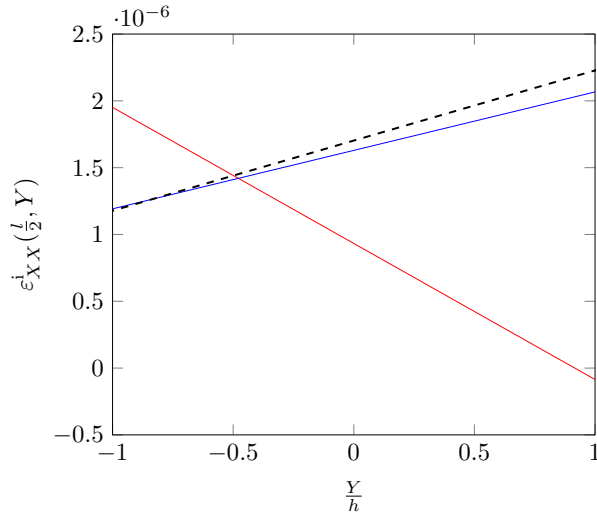


Figure 3. Longitudinal strain at the midspan section of the ionic polymer. We compare the results from enriched Euler-Bernoulli beam theory (blue solid line), classical Euler-Bernoulli beam theory (red solid line), and FE simulations (black dashed line).

Finally, in Fig. 4 we present the through-the-thickness strain at the midspan section of the ionic polymer. Similar to our previous results,¹³ FE simulations evidence through-the-thickness strain localization at the inter-

faces between ionic polymer and electrodes. We remark that these deformations are four orders of magnitude larger than longitudinal strain in Fig. 3. As expected, the classical Euler-Bernoulli beam theory cannot capture these deformations, whereby it assumes that IPMC cross-sections are rigid. The enriched Euler-Bernoulli beam theory, instead, accurately predicts the deformation along the IPMC thickness, in proximity of both electrodes. This result confirms our intuition that, far from the constraints, the through-the-thickness deformation is similar to the case of uniform bending.

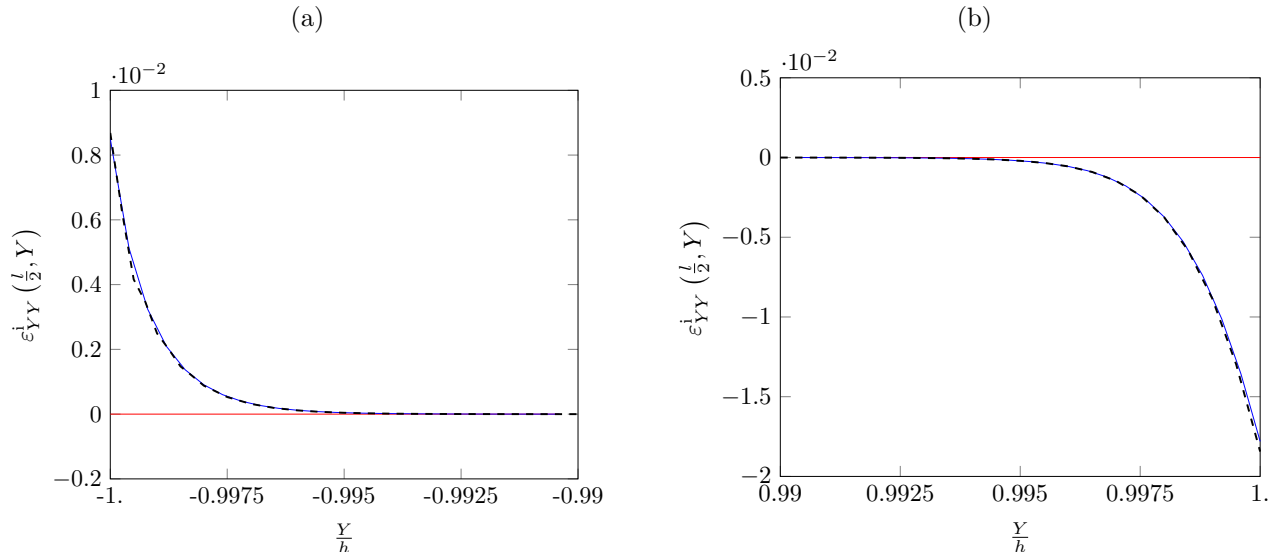


Figure 4. Through-the-thickness strain at the midspan section of the ionic polymer in the vicinity of the cathode (a) and anode (b). We compare the results from enriched Euler-Bernoulli beam theory (blue solid line), classical Euler-Bernoulli beam theory (red solid line), and FE simulations (black dashed line).

5. CONCLUSIONS

In this paper, we propose a new structural theory for ionic polymer metal composites (IPMCs), which can accurately describe multiaxial deformations of IPMCs under actuation at a limited computational cost. The theory builds upon our previous results on multiaxial deformations of IPMCs subject to uniform bending, toward including the effect of shear deformations and non-zero thickness electrodes. To this end, we enrich the classical Euler-Bernoulli beam theory with an additional term encompassing the transverse displacement of the ionic polymer observed during uniform bending, playing the role of a shape function, which is modulated by a new structural variable along the IPMC axis. The balance equations are obtained from the minimization of the total potential energy, accounting for the strain energies of ionic polymer and electrodes and the work performed by osmotic pressure and Maxwell stress that contribute to the eigenstress. By comparing classical and enriched Euler-Bernoulli beam theories against nonlinear finite element (FE) simulations of IPMC response to a step voltage, we demonstrate that the novel enriched theory accurately anticipates the results of FE simulations, whereas classical Euler-Bernoulli beam theory cannot even capture the main features of the macroscopic displacement field.

While promising, the application range of the proposed structural theory is partially limited by its underlying hypotheses. The enriched Euler-Bernoulli beam theory may not provide accurate results for more complex scenarios in terms of mechanical boundary conditions. Should one analyze more severe boundary conditions, typically associated with loading or constraints on a single skin, for example when testing blocking force in IPMCs, higher-order structural theories for cross-section warping and transverse deformability of the core^{21,27} have to be considered as the basis for an enriched structural theory. The assumption of inert electrodes could be relaxed by including the resistivity of IPMC electrodes, such that a different voltage would be applied across each cross-section.²⁸ More challenging is to consider larger applied voltages, which could hamper the validity

of the hypotheses of small strains and decoupling of the electrochemistry from the mechanics. Overcoming this constraint requires the adoption of a more accurate, nonlinear constitutive model for the ionic polymer and of iterative, staggered procedures to solve the fully coupled electrochemomechanical problem.⁵ Despite these limitations, this work is a first, critical step toward inverse design of IPMCs, which requires accurate and inexpensive tools for describing IPMC multiaxial deformations and stress concentrations.

ACKNOWLEDGMENTS

This research was supported by the National Science Foundation under Grant No. OISE-1545857.

REFERENCES

- [1] Shahinpoor, M., ed., [*Ionic Polymer Metal Composites (IPMCs): Smart Multi-Functional Materials and Artificial Muscles*], Smart Materials Series, Royal Society of Chemistry (2015).
- [2] Jo, C., Pugal, D., Oh, I.-K., Kim, K. J., and Asaka, K., “Recent advances in ionic polymer-metal composite actuators and their modeling and applications,” *Progress in Polymer Science* **38**(7), 1037–1066 (2013).
- [3] Allen, H. G., [*Analysis and design of structural sandwich panels*], Pergamon Press Ltd., Oxford (1969).
- [4] Oguro, K., “Preparation procedure - Ion-exchange polymer metal composites (IPMC) membranes,” (Retrieved on August 10, 2018).
- [5] Cha, Y. and Porfiri, M., “Mechanics and electrochemistry of ionic polymer metal composites,” *Journal of the Mechanics and Physics of Solids* **71**, 156–178 (2014).
- [6] Chen, Z., “A review on robotic fish enabled by ionic polymer-metal composite artificial muscles,” *Robotics and Biomimetics* **4**(24) (2017).
- [7] Carpi, F. and Smela, E., eds., [*Biomedical Applications of Electroactive Polymer Actuators*], Wiley (2009).
- [8] Carrico, J. D., Traeden, N. W., Aureli, M., and Leang, K. K., “Fused filament 3d printing of ionic polymer-metal composites (ipmcs),” *Smart Materials and Structures* **24**(12), 125021 (2015).
- [9] Carrico, J. D., Tyler, T., and Leang, K. K., “A comprehensive review of select smart polymeric and gel actuators for soft mechatronics and robotics applications: fundamentals, freeform fabrication, and motion control,” *International Journal of Smart and Nano Materials* **8**(4), 144–213 (2017).
- [10] Stalbaum, T., Trabia, S., Hwang, T., Olsen, Z., Nelson, S., Shen, Q., Lee, D.-C., Kim, K. J., Carrico, J., Leang, K. K., Palmre, V., Nam, J., Park, I., Tiwari, R., Kim, D., and Kim, S., “Guidelines for making ionic polymer-metal composite (IPMC) materials as artificial muscles by advanced manufacturing methods,” in [*Advances in Manufacturing and Processing of Materials and Structures*], Bar-Cohen, Y., ed., ch. 15, 379–395, CRC Press (2018).
- [11] Nemat-Nasser, S. and Li, J. Y., “Electromechanical response of ionic polymer-metal composites,” *Journal of Applied Physics* **87**(7) (2000).
- [12] Shahinpoor, M. and Kim, K. J., “Ionic polymer-metal composites: III. modeling and simulation as biomimetic sensors, actuators, transducers, and artificial muscles,” *Smart Materials and Structures* **13**(6) (2004).
- [13] Boldini, A. and Porfiri, M., “Multiaxial deformations of ionic polymer metal composites,” *International Journal of Engineering Science* **149**, 103227 (2020).
- [14] Bauchau, O. A. and Craig, J. I., [*Structural Analysis - With Applications to Aerospace Structures*], Springer (2009).
- [15] Gurtin, M. E., Fried, E., and Anand, L., [*The Mechanics and Thermodynamics of Continua*], Cambridge University Press (2013).
- [16] Pelesko, J. A. and Bernstein, D. H., [*Modeling MEMS and NEMS*], CRC Press (2002).
- [17] Bard, A. J. and Faulkner, L. R., [*Electrochemical Methods - Fundamentals and Applications*], John Wiley & Sons (2001).
- [18] Nayfeh, A. H., [*Introduction to Perturbation Techniques*], Wiley (2011).
- [19] Porfiri, M., “Charge dynamics in ionic polymer metal composites,” *Journal of Applied Physics* **104**(10) (2008).
- [20] Timoshenko, S. and Goodier, J., [*Theory of Elasticity*], McGrawHill, third ed. (2001).

- [21] Mattei, O. and Bardella, L., “A structural model for plane sandwich beams including transverse core deformability and arbitrary boundary conditions,” *European Journal of Mechanics - A/Solids* **58**, 172 – 186 (2016).
- [22] Belytschko, T., Liu, W. K., Moran, B., and Elkhodary, K. I., [*Nonlinear Finite Elements for Continua and Structures*], Wiley, second ed. (2014).
- [23] Lanczos, C., [*The variational principles of mechanics*], Courier Corporation (1949).
- [24] Cardarelli, F., [*Materials Handbook*], Springer, third ed. (2018).
- [25] Asaka, K., Oguro, K., Nishimura, Y., Mizuhata, M., and Takenaka, H., “Bending of polyelectrolyte membrane-platinum composites by electric stimuli I. Response characteristics to various waveforms,” *Polymer Journal* **27**, 436–440 (1995).
- [26] Porfiri, M., Leronni, A., and Bardella, L., “An alternative explanation of back-relaxation in ionic polymer metal composites,” *Extreme Mechanics Letters* **13**, 78–83 (2017).
- [27] Tonelli, D., Bardella, L., and Minelli, M., “A critical evaluation of mechanical models for sandwich beams,” *Journal of Sandwich Structures & Materials* **14**(6), 629–654 (2012).
- [28] Kim, H., Cha, Y., and Porfiri, M., “Voltage attenuation along the electrodes of ionic polymer metal composites,” *Journal of Intelligent Material Systems and Structures* **27**(17), 24262430 (2016).