Finite element simulations of the bending of the IPMC sheet

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ABSTRACT

Finite Element method is used to build and simulate an IPMC sheet. The physical bending of the realistic Nafion sheet due to the drift of counter-ions (e.g Na+) and dragged water in applied electric field are simulated. The effect of the charge imbalance caused by drift of counter-ions is related to physical bending of the IPMC sheet. Mechanical properties of nafion polymer and platinum coating have taken account separately in proposed model. All simulations are time dependent, thus transient model and need for few additional parameters such as damping coefficients are also explained. In addition to creating base model for simulating an IPMC muscle, electrochemical reactions are also introduced. Equations describing periodic adsorpion and desorpion of CO and OH in a platinum electrode in formaldehyde solution are coupled to the base model. This has allowed us to simulate self-oscillatory behavior of an IPMC sheet.

Keywords: Electroactive polymers, EAP, Finite element method, Electrochemical-mechanical analysis, Actuator, Coupled problem, Self-oscillating systems

1. INTRODUCTION

EAP-based electromechanical actuators are valuable for use in a number of applications starting with miniature robotics up to military and space. These actuators have light weight, noiseless motion, simple mechanical construction; large controlled displacement and good damage tolerance along with an ability to perform different movements like bending and contractions makes possible to use them as artificial muscles. In this letter we consider simulation of ionic polymer-metal composite (IPMC) materials with finite element method (FEM).

IPMC materials are highly porous polymer materials such as Nafion, Fleminon, Teflon, filled with some kind of ionic conductive liquid. There are water based IPMCs which operate in aquatic environment and conduction is caused by ions such as Na+, K+ dissociated in water. Ionic liquid based IPMCs do not need wet environment for operating. The sheet of the ionic polymer is coated with thin metal layer, usually platinum or gold. In applied electric field the freely movable cations inside the polymer migrate towards an electrode, causing expansion of the material at the one end of the sheet and contraction at the other end, which results in bending of the sheet.

For simulating actuation of an IPMC sheet we need to solve coupled problems due to the complex nature of IPMC actuation. It involves working in different domains such as mechanical, electrostatic and mass transfer. [REF] and [REF] have already simulated mass transfer and electrostatic effects. We have used similar approach for these problems. However, new approach is introduced for mechanical bending of IPMC strip. By coupling those domains together we get enough accurate finite element model for an IPMC muscle sheet. It allows us using the model as a base for solving more complex problems, thus we have also introduced a simulation of electrochemical reactions.

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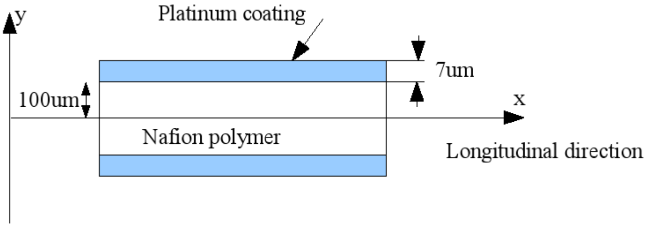

Figure 1. Illustration of domains and dimensions used in modeling.

1.1. Electrochemical oscillations

Spontaneous oscillations are common fenomenon in nature and it has been studied for many experiment, including electrochemical systems such as oxidation of organic materials and metals [REF]. Electrochemical systems exhibiting instabilities often behave like activator-inhibitor systems, where the potential of the electrode is an essential variable and takes on the role either of the activator or of the inhibitor. Under certain conditions the system can generate oscillations. We have conducted series of tests, where IPMC sheet have immersed into acidic formaldehyde, HCHO, solution and exposed to constant outer potential. However, measurements show current oscillations, which in turn result in oscillating bending of the IPMC sheet. Hence we have also introduced a model in this paper for describing such systems coupled with other physicals domains.

2. SIMULATION DETAILS

An IPMC sheet consists of backbone polymer and metal coating. We have used Nafion 117 coated with thin layer of platinum. Although it is written in the introduction that simulations in multiple physical domains are needed for getting bending model for an IPMC, most simulation is done in one mechanical domain - backbone polymer. Platinum coating is considered only in mechanics domain while calculating bending. So basically there are three mechanical domains as shown in Figure 1.

Most simulations are done for IPMC strip, 1.5cm long, 200um thick polymer coated with 7um thick platinum, in cantilever configuration - one end of the strip is not allowed to move. Gravitational forces are not considered in any of following simulations.

2.1. Migration of cations

Nernst-Planck equations describes diffusion, convection and in presence of electric field, also migration of particles. General form of the equation is

$$\frac{\partial C_i}{\partial t} + \nabla \cdot (-D_i \nabla C_i - z_i \mu_i F C_i \nabla \phi) = -\vec{u_i} \cdot \nabla C_i, \tag{1}$$

where subscript i denotes species and C is concentration, μ mobility of species, D is diffusion constant, T absolute temperature, R gas constant, F Faraday constant, $\vec{u}$ is velocity, z charge number and ϕ electric potential. This equations must be solved only for freely movable cations. As voltage is applied to the electrodes of an IPMC, all freely movable cations start migrating towards cathode causing current in circuit. As ions cannot move beyond boundary of Nafion, charges start to accumulate, resulting in increase of electric field of opposite direction to applied one and it could be described by Gauss' Law:

$$\nabla \vec{E} = \frac{F \cdot \rho}{\varepsilon}, \tag{2}$$

where ρ is charge density, ε is absolute dielectric constant and E is the strength of the electric field and can be expressed also as $\nabla \phi = -E$. Steady state of the cations forms when electric field created by distribution of cations cancels out applied electric field, i.e. electric field strength inside the polymer is approximately zero as shown in Figure 2. Charge distribution for cation concentration of $1200 \frac{mol}{m^3}$ is shown in Figure 3. It is interesting to notice that there are differences in charge distribution only in really thin boundary layers. There are no chare imbalances inside the polymer.

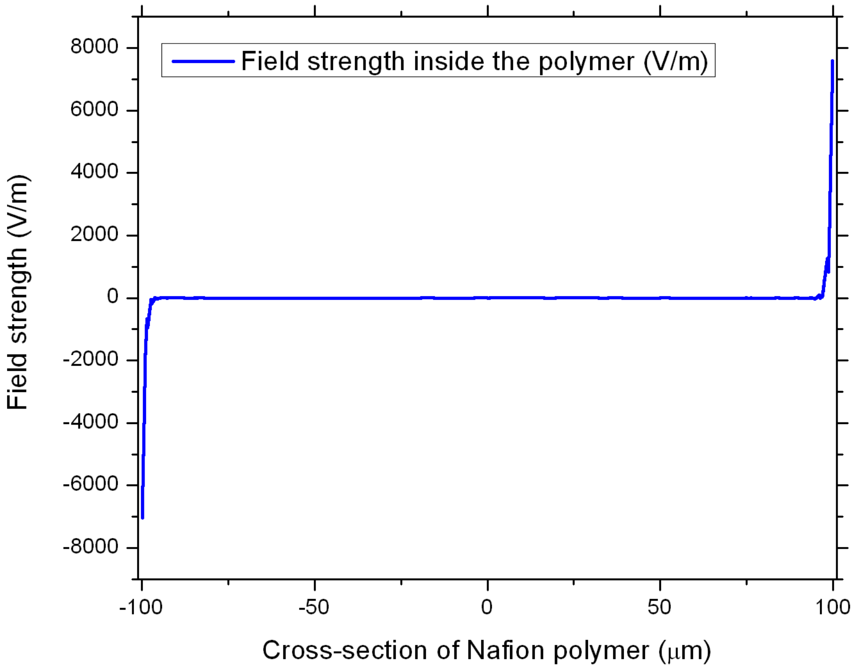

Figure 2. Electric field strength inside the IPMC in charge balance state. Notice that the field is zero inside the polymer, except really thin boundary layer.

2.2. Modeling actuation of an IPMC

Many authors have used Euler cantilever beam equation to model bending of an IPMC strip in cantilever configuration. [SOME ref's to authors?]. Though it describes position of the IPMC quite accurately for small displacements, it is not dynamical model - it does not show motion of the material in time. So using static Euler theory does not lead to really accurate time dependent model. Hence different approach is used for bending model: stiffness will be also considered when calculating. Importance of viscoelacity have been brought out also by some other authors like [REF].

2.2.1. Assumptions about bending mechanism

There are differences in charge distribution only in really thin boundary layers as we brought out before. General conclusion by many authors is that locally generated charge imbalance nearby platinum electrodes is main cause of bending of the IPMC [REFS]. Following example of Leo [REF], we have defined longitudinal force in each point of the IPMC as follows:

$$\vec{F} = (A \cdot \rho + sgn(\rho) \cdot B \cdot \rho^2) \cdot \hat{x}, \tag{3}$$

where ρ is charge density and A is a constant which could be found from different experiments. $sgn(\rho)$ is to preserve correct sign of excess charge. In our model the force depends on charge density and the shape of the force inside the polymer is also shown in Figure 3.

2.2.2. Mathematics behind the bending

Finite element method for solving physical equations is very powerful and allows us to get more precise results than by using analytical models. We used structural mechanics equations described in Comsol Multiphysics

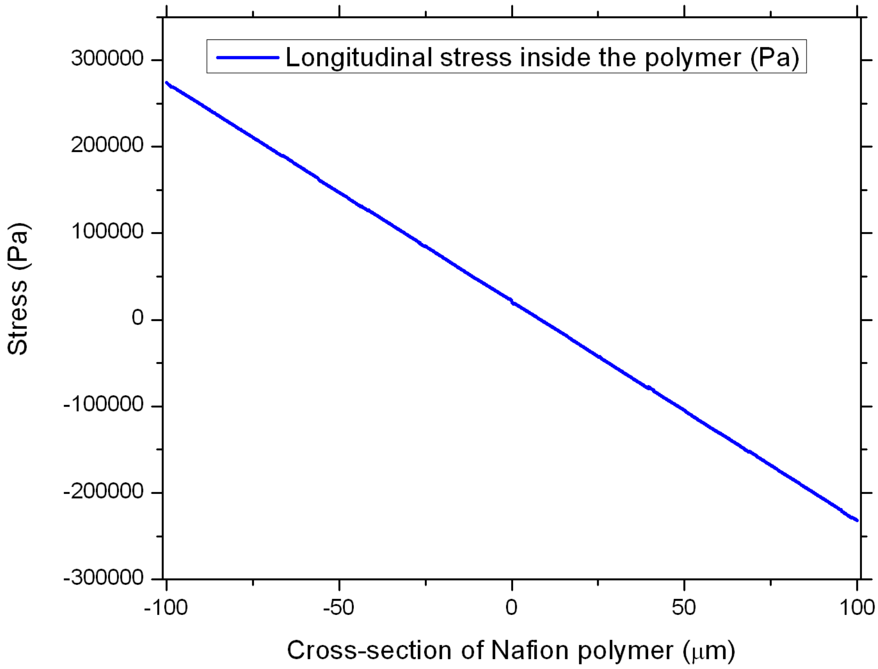

Figure 3. Cation distribution in charge balance state. Also longitudinal force according to modeling inside the polymer is shown. Notice that force grows faster near the boundaries of the polymer than cation distribution. The reason for that is quadratic term in force formula as shown in Equation 3. It is important to notice that the graph is somehow illustrative because real simulation maximum values for concentration and specially for stress are larger.

software package. Normal and shear strains are defined:

$$\varepsilon_i = \frac{\partial u_i}{\partial x_i}, \; \varepsilon_{ij} = \frac{1}{2}\left(\frac{\partial u_i}{\partial x_j} - \frac{\partial u_j}{\partial x_i}\right). \tag{4}$$

where u denotes displacement, x denotes a coordinate and indices i and j are from 1 to 3 and denote components correspondingly in x, y or z direction. As IPMC sheet in our simulations is allowed to move only in x-y plane, $\varepsilon_3 = 0$ and $\varepsilon_{23} = \varepsilon_{13} = 0$. Considering this we can write symmetric strain tensor in vector form and symmetric stress tensor in vector form as follows:

$$\varepsilon = \begin{bmatrix} \varepsilon_1 \\ \varepsilon_2 \\ 0 \\ 2\varepsilon_{12} \\ 0 \\ 0 \end{bmatrix}, \; \sigma = \begin{bmatrix} \sigma_1 \\ \sigma_2 \\ \sigma_3 \\ \tau_{12} \\ \tau_{23} \\ \tau_{13} \end{bmatrix}, \; \tau_{ij} = \tau_{ji}. \tag{5}$$

The stress-strain relationship is

$$\sigma = D\varepsilon. \tag{6}$$

D is 6 x 6 elasticity matrix and consists of components of Young's modulus and Poisson's ratio. The system is in equilibrium if the following equation is satisfied:

$$-\nabla \cdot \sigma = \vec{F}, \tag{7}$$

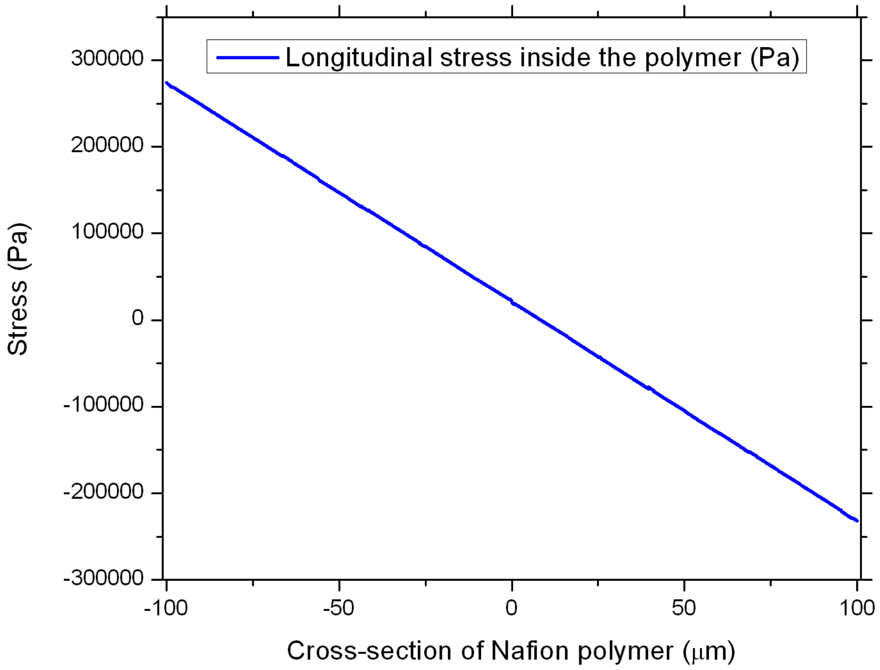

Figure 4. Longitudinal stress inside the polymer backbone in global coordinates.

which is basically Navier's equation for displacement. For instance, stress generated in the polymer is shown in Figure 4. The stress inside the platinum coating is very much bigger, thus it is not shown in the figure.

2.2.3. Transient analysis of bending

As our simulation is rather dynamic than static, we have to introduce equation for describing motion of the IPMC sheet. For that the Newton's Second law is used:

$$\rho\frac{\partial^2\vec{u}}{\partial t^2} - \nabla\cdot c\nabla\vec{u} = \vec{F}, \tag{8}$$

where the second term is static Navier's equation. Dynamic part is introduced by the first term of the equation. This equation describes a system without damping.

Motion with Rayleigh damping model for system with single degree of freedom can be described

$$m\frac{d^2u}{dt^2} + \xi\frac{du}{dt} + ku = f(t), \tag{9}$$

where ξ is damping parameter and can be expressed $\xi = \alpha m + \beta k$. Parameter m is mass and k is stiffness and α and β are corresponding damping coefficients. Comsol Multiphysics uses similar form for systems with multiple degrees of freedom:

$$\rho\frac{\partial^2\vec{u}}{\partial t^2} - \nabla\cdot\left[c\nabla\vec{u} + c\beta\nabla\frac{\partial\vec{u}}{\partial t}\right] + \alpha\rho\frac{\partial\vec{u}}{\partial t} = \vec{F}, \tag{10}$$

where c is constant used in Navier's equatin, in static analysis.

All values used to simulate previously described equations are shown in Table 1.

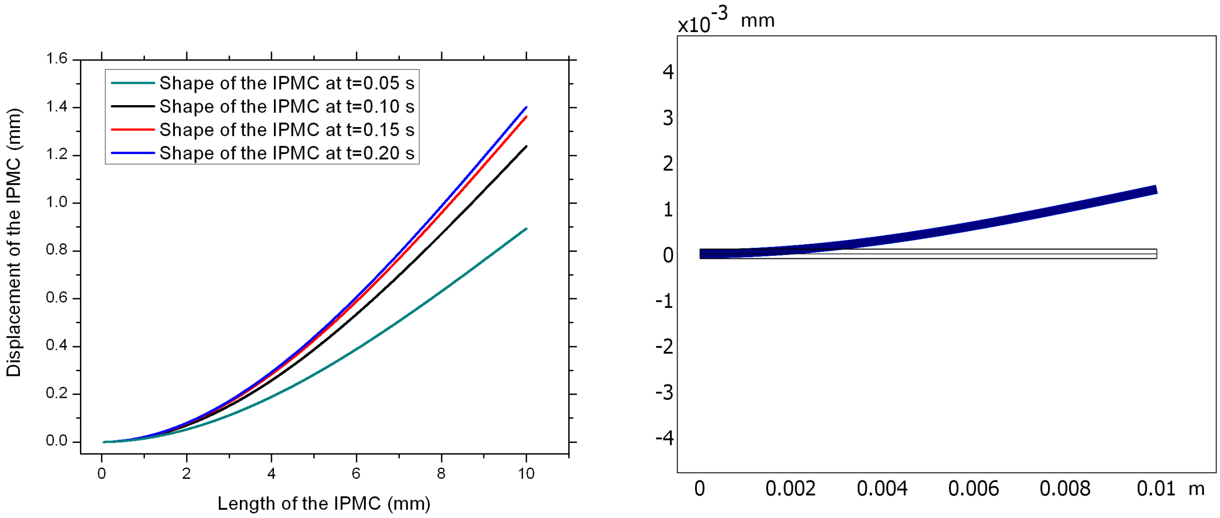

Figure 5. On the left: the shape of an IPMC muscle at different times (simulated with 2V, 1cm long strip). On the right: illustration of bending at t=0.2s

Variable	Value	Dimension	Comment
D_{cation}	$2 \cdot 10^{-9}$	$\frac{m^2}{s}$	Diffusion coefficient of cations, e.g Na+
ε	$3.8 \cdot 10^{-5}$	$\frac{F}{m}$	From capacitance measurment of an IPMC
μ	$8 \cdot 10^{-13}$	$\frac{mol \cdot s}{kg}$	From Nernst-Einsten relation $\mu = \frac{D}{R \cdot T}$ where $T = 293K$, $R = 8.31 \frac{J}{mol \cdot K}$
Y_N	$50 \cdot 10^6$	Pa	Young modulus of Nafion
Y_{Pt}	$169 \cdot 10^9$	Pa	Young modulus of platinum
ρ_N	2600	$\frac{kg}{m^3}$	Density of Nafion
ρ_{Pt}	21500	$\frac{kg}{m^3}$	Density of platinum
A	$5 \cdot 10^5$	$\frac{N \cdot m}{C}$	A constant in 3
B	$3 \cdot 10^4$	$\frac{N \cdot m^4}{C^2}$	A constant in 3
z	$+/-1$	-	Charge number for cations/anions

Table 1. Values used in simulations.

2.3. Bending simulation results

Although there are couple of parameters in Equation 3, which depend on experimental results and are not very uniquely specified for all IPMC sheets, the simulation results predict the bending of an IPMC sheet precisely enough to use in further modeling problems. There are illustration of bending and the a graph shown in Figure 5. As we are not modeling static problem, the time dependence of tip displacement of an IPMC muscle is shown in Figure 6.

By coupling previously described equations, we have developed a good base model. Now it is possible to extend it by adding more equations to get physically meaningful results for more complicated problems. Next subsection of the article gives good example, because electrochemical reactions which lead to self-oscillation of an IPMC muscle, are described and related to the model there.

2.4. Self-oscillations - coupling a problem of electrochemistry

We have conducted series of tests with IPMCs in formaldehyde (HCHO) solutions with applied constant electric field. Measurements show that current oscillations begin from potential of ca. 0.75V. Initial burst is caused by

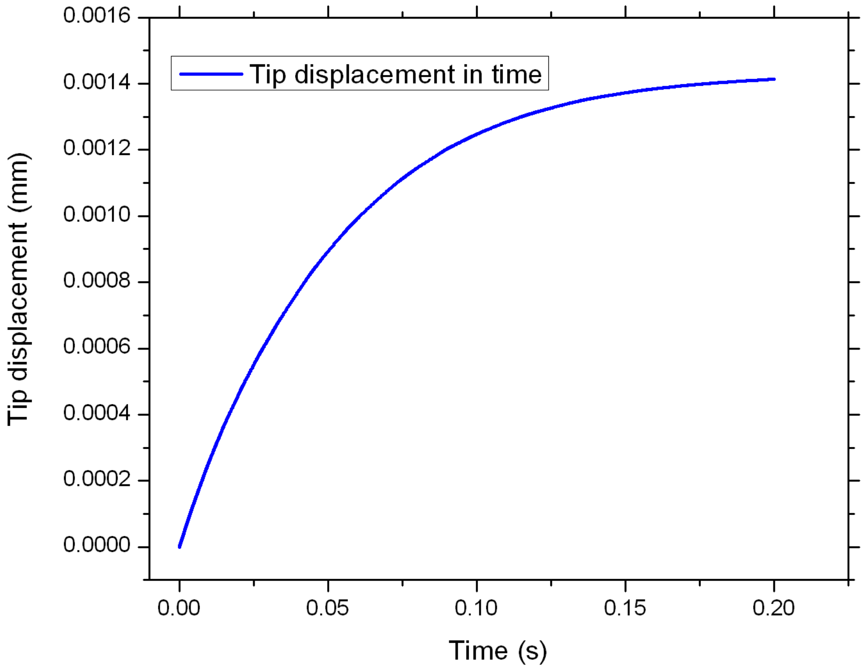

Figure 6. Tip displacement of an IPMC muscle in time.

the reaction

$$CO_{ads} + OH_{ads} \rightarrow CO_2 + H^+ + e^- + 2*, \tag{11}$$

where ads denotes species adsorbed to the surface of platinum and * denotes an active platinum site. The result of this reaction is freeing up 2 sites which causes CO to adsorb again. These reactions lead to oscillating potentials which in turn lead to self-rhythmic motion of an IPMC.

Chronopotentiometry scans show that before reaction 11, following reactions occur:

$$HCHO + * \rightarrow Pt - CHO_{ads} + H^+ + e^- \tag{12}$$

$$Pt - CHO \rightarrow Pt - CO_{ads} + H^+ + e^- \tag{13}$$

$$H_2O + * \leftrightarrow Pt - OH_{ads} + H^+ + e^- \tag{14}$$

HCHO is dissociated on the electrode surface at lower anodic potential. Higher anodic potential causes dehydrogenation of water which results in water oxidation with intermediate (Pt-OH) formation.

2.4.1. Simulating electrochemical reactions

It is suggested by [REF] that concentration of HCHO near the platinum sufrace is important for reaction 12 and it is one variable for altering frequency of oscillations. We introduce double layer near the surface of one electrode in our simulation model. Based on reactions 11 to 14 and conducted chronopotentiogram measurements we have

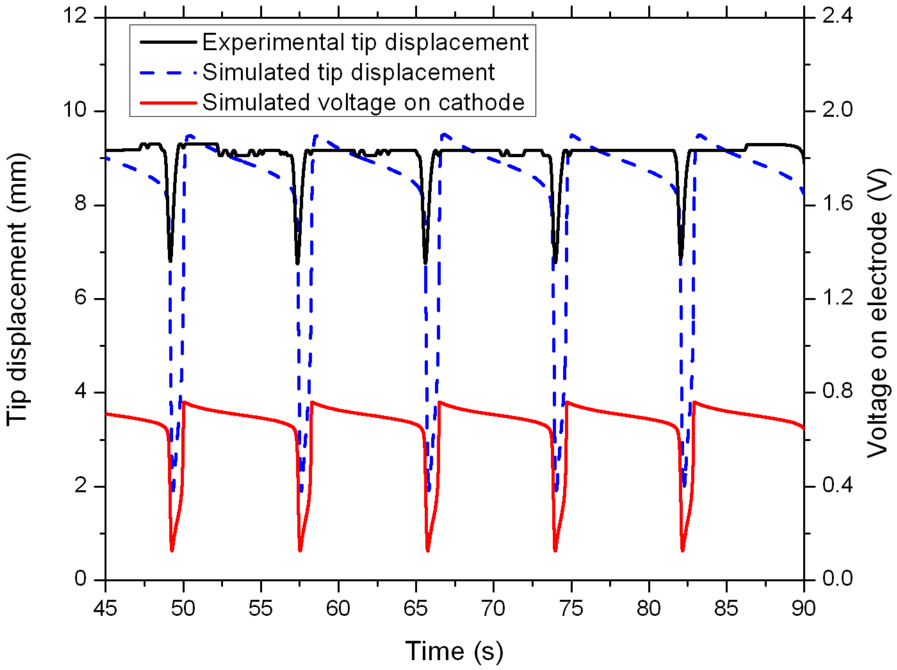

Figure 7. Simulation of voltage and tip displacement for 3M solution of HCHO + 3M H_2SO_4 and corresponding experimental data.

developed equations to describe adsorpion, desorpion, reaction of species and voltage fluctuation on cathode:

$$\dot{\theta}_{CO} = k_2(1-\theta_{CO}-\theta_{OH}) - k_4\theta_{CO}\theta_{OH} \tag{15}$$

$$\dot{\theta}_{OH} = k_3(1-\theta_{CO}-\theta_{OH}) - k_3\theta_{OH} - k_4\theta_{CO}\theta_{OH} \tag{16}$$

$$\dot{E} = \frac{1}{C_{dl}}\left[I_{th} + (I-I_{th})\frac{c}{c_{2M}}sgn(c-c_{2M}) - jS_{pt}(k_1(1-\theta_{OH}-\theta_{CO}) + k_4\theta_{OH}\theta_{CO})\right] \tag{17}$$

$$k_i = exp(s_i(E-E_i)) \tag{18}$$

Here θ is adsorpion coverage of corresponding species and is normalized to 1, E is potential on cathode, I_{th} threshold current and equals 10mA , I applied current, $c$concentration of HCHO near the electrode, c_{2M} equals $2000\frac{mol}{m^3}$ and s_i and E_i are correspondingly simulation parameters and reaction potentials as described in [D.K dissertation]. Double layer conductivity C_{dl} and maximum adsorpion coverage S_{pt} are taken equal to 1 in our simulations.

First version of this model was proposed in [Doyeon Kim disseration]. We have added second term to the equation 17, which brings in voltage dependance of concentration. As there is bigger concentration of HCHO near the electrode, there is also bigger direct current - direct oxidation of HCHO - which is described by second term in the equation. By coupling those equations with our base bending model, we get oscillating voltage and bending as shown in Figure 7. Similarly to other concentrations of HCHO in range of 1 to 3 $\frac{mol}{l}$ we get simulation results reflecting experimental values.

Though the effect of double layer concentration according to current simulations turned out to be really small, further models should consider it more significantly. It would probably help us to improve model dependance of applied voltage/current and would be physically more accurate.

3. CONCLUSIONS

We have developed finite element model for an IPMC sheet. Physical accuracy, flexibility and power of finite element method allows us to use this model as a base for coupling different equations corresponding to more complicated problems, such as was shown in the last subsection - electrochemical oscillations which occure on the surface of an electrode.

4. FUTURE WORK

Though the model we have developed has been enough accurate for current simulations, it could be developed further a lot. The effect of water dragged by ions is not explicitly taken account in current research, but to make 3 physically more meaningful, this effect should be written out more precisely for future simulations. The effect of surface resistance change due to bending of IPMC sheet is not considered in current model, but it could be taken account in future.

Also equations describing electrochemical oscillations need some revision, specially for making them more dependant of physical variables such as applied potential and concentration of formaldehyde in double layer, in wider range.

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