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MODELING IPMC MATERIAL WITH DYNAMIC SURFACE CHARACTERISTICS

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ABSTRACT

This paper presents the Finite Element Analysis (FEA) of an ionic polymer-metal composite (IPMC) material. The IPMC materials are known to bend when electric field is applied on the electrodes. The material also produces potential difference on the electrodes when is bent. Several authors have used the FEA to describe that fenomenon and rather precise basic Finite Element (FE) models already exist. Therefore the current study is mainly focused on the modeling of the electrodes of IPMC. The first goal of this research is to model the electric currents in the electrodes. The basis of the electric current calculations is the Ramo-Shockley theorem, which has been used in the other areas of physics to describe the currents in a circuit due to a charge movement in a media. We have used the theorem to calculate the current density in the continuous electrodes of IPMC due to the ion migration in the backbone polymer. Along the current densities we are able to calculate voltage on the electrode at a given time moment. The model is demonstrated to give some physically reasonable results. However, the model is rather complex and as the solution times are quite large, some possible optimizations have been considered as well. The second goal of this study is to include the dynamic resistance and capacitance of the electrodes in our model. Lot of research has been done to develop a physically reasonable capacitor-resistor model of an IPMC and the results have been promising. Furthermore, some authors have managed to develop partial differential equations (PDE) to describe the model. We try to include some simplified versions of those equations into our physical model. As the FE model for IPMC is nonlinear and gets complicated very fast when additional equations are added, the final sections of this paper briefly considers some novel optimization ideas in regard to modeling IPMC with FE method.

1 INTRODUCTION

Electroactive polymer actuators have gained a lot of attention in many fields such as robotics and micro electronics. The advantages of EAP actuators are relatively simple mechanics and noiseless actuation. Additionally some EAPs, such as IPMCs [1], are able to function in aqueous environments. Those qualities make the materials possible to use as so called artificial muscles. In this paper we consider three dimensional time dependent simulations of IPMC type materials with the Finite Element Method.

One of the most important qualities of IPMC materials is

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Table 1. The simulation input values for the base model.

Variable	Value	Dimension	Comment
D_{cation}	$0.6 \cdot 10^{-11}$	$\frac{m^2}{s}$	Diffusion const.
ε	$2.5 \cdot 10^{-2}$	$\frac{F}{m}$	Dielectric const.
μ	$2.6 \cdot 10^{-15}$	$\frac{mol \cdot s}{kg}$	Mobility at $T = 293K$

relatively large amplitude bending in response to electrical stimulation. An ion exchange polymer membrane, such as Nafion, is covered with metal layers. The metal is typically platinum or gold. During the typical fabrication process the polymer membrane is saturated with certain solvent and ions. When voltage is applied to the metal electrodes, the ions start migrating due to the applied electric field. Migrating ions usually drag some solvent with them, causing expansion and contractions respectively near the surface layers. That in turn causes bending like actuation of IPMC sheet.

To predict the actuation, a good understanding of underlying processes must exist. Electrostatics, mass transfer and mechanical effects must be taken account to get a minimal functional base model which could predict actuation. Usually, a two dimensional time dependent model would be enough to get reasonable results. However, in this paper we consider three dimensional model of IPMC. This allows to take into account surface resistance changes for whole area of the metallic layer. Some authors [2–5] have already simulated mass transfer and electrostatic effects. We used similar approach in our model. Toi [6] has shown a Finite Element model including viscosity terms in transportation processes explicitly. The simulation is performed as time dependent and for three dimensions. However, the basis of the described model is a rectangular beam with 2 pairs of electrodes.

The current work extends an already published paper [7], where preliminary ideas of simulating electric current in the electrodes of IPMC and the voltage profile on the electrodes were proposed. The analytical surface resistance model is derived by Chen [8] for 2D domain. However, the proposed numeric FE simulations work both in 2D and 3D domain. In the following sections, a FE model for IPMC, where the electric processes inside the anode are coupled to the the processes which take place inside the polymer backbone of IPMC, is proposed. Also, some possible optimizations of the model are studied. The bending phenomenon of IPMC is not covered in this stage of the research, however, a simple FEA based deformation model of IPMC is demonstrated in our previous works [9].

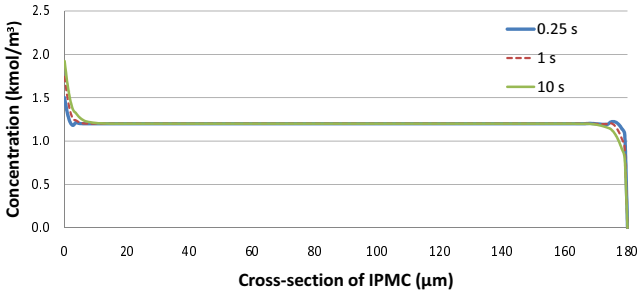

Figure 1. The cross section cation concentration in IPMC at $0.25s$, $1s$, and $10s$ after applying voltage to the polymer domain. the shape of the graphs is generally same for 2D and 3D simulations.

2 THE COUPLED ELECTRODE MODEL

We have used Nafion^{TM} 117, coated with thin layer of platinum in our experiments. Mass transfer and electrostatic simulations are done only for backbone polymer. Surface current calculations are carried out a electrode. All simulations are done for an IPMC strip of $180\mu m$ thick polymer coated with $5\mu m$ thick platinum. The following sections give an overview of the base model of IPMC and show how the electrode electric model is coupled to it.

2.1 The base model

The cation migration in the polymer backbone is described by the Nernst-Planck equation, which covers migration and diffusion part. The equation is:

$$\frac{\partial C}{\partial t} + \nabla \cdot (-D\nabla C - z\mu F C \nabla \phi) = -\vec{u} \cdot \nabla C, \qquad (1)$$

where C is concentration, μ mobility of species, D diffusion constant, T absolute temperature, R universal gas constant, F Faraday constant, $\vec{u}$ velocity, z charge number and ϕ electric potential. The equations is solved only for cations as anions are fixed in the polymer backbone. As voltage is applied to the platinum electrodes, all free cations start migrating towards cathode, causing current in the outer electric circuit. As ions cannot move beyond the boundary of the polymer, local charge intensity starts to increase near the surface of the platinum electrodes, resulting in increase of electric field in the opposite direction to the applied one. This effect could be described by by Poisson's Law:

$$\nabla \cdot \vec{E} = -\Delta\phi = \frac{F \cdot \rho}{\varepsilon}, \qquad (2)$$

where ρ is charge density, ε is absolute dielectric constant and E is the strength of the electric field and can be also expressed

as $\nabla\phi = -\vec{E}$. The formed steady state of the cations is shown in Figure 1. The values of simulation constants are shown in Table 1. So far we have described the base model, which is usable in both two dimensional and three dimensional modeling. Next we extend the model to include the current flow in the electrodes and also to estimate the currents and therefore the voltage distribution as well.

2.2 Electrode model

The model described in the previous section is sufficient for most cases and for two dimensional or three dimensional domains. However, solving the base model in three dimensional domain does not give us any extra information - it is basically the same as an extended two dimensional solution. At the same time there is a significant increase in the complexity in terms of meshing and increase of degrees of freedom of the system, which in turns leads to longer solving times. The usefulness of the third dimension becomes eminent when the model also takes into account the spatial and time dependent variation o the surface resistance of the electrodes. The electrode resistance is an important characteristic of an IPMC strip. Besides of varying for different IPMC sheets, it tends to depend on the curvature of the IPMC strip [10]. At the same time, the resistance is the parameter which could be rather easily altered. For instance, it is possible to make some areas of the muscle sheet less conductive. That is the case where three dimensional model would be applicable. However, even when the surface resistance is considered constant, there still exists a voltage drop along the surface and the proposed model helps to take this into account. The idea of the model is to couple the electrical model, which includes active resistances of the electrodes and the base model described in the previous section. Thus the cation transportation and continuum mechanics is coupled to the currents/voltage in the surface layers. Once having this model, the time dependent variation of the electrode resistance could be easily varied as well, however, this was not in the scope of the current paper.

An IPMC strip with an external power source forms a closed electric circuit. There are roughly two types of conduction mechanisms in the circuit: electron conduction in the outer part of the circuit and the ion migration inside the IPMC. Even though the ions move only inside the polymer backbone, there is a connection between current in outer circuit and displacement of the ions. The theorem which is more often used in plasma physics [11], is called Ramo-Shockley theorem. It connects movement of charged particles in a confined space to the currents in a connected electric circuit. The theory, for instance, has been used to create a model of ion channels [12]. The equation for the current in the external circuit due to the relocation of the ions is:

$$I = \frac{1}{V}\sum_i q_i \times W\vec{(}r_i)_i \vec{v}_i, \tag{3}$$

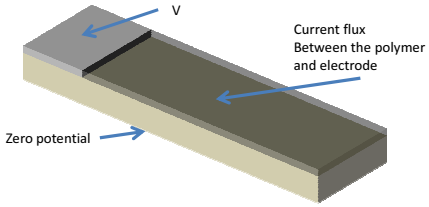

Figure 2. The model of the IPMC material. For both 2D and 3D case, it consists of polymer backbone domain, where Nernst-Placnk and Poisson equations are active and the electrode domain, where the electric current is modeled. The voltage is applied on the $1cm^2$ size of area in the one end of IPMC. Zero potential is applied to the bottom of the polymer domain and the electric current is modeled between the electrode and polymer domain.

where j is the index of a particle, q is the charge, and v is the velocity of a particle. W corresponds to a electric field which would exist without any charged particles present [12]. By using Eq. (3), we can calculate the current flowing in an electrode. This in turn is cause for the voltage drop in the surface. The electrode could be depicted as a series of resistors which are also connected to the polymer. Then the Eq. (3) could be written as

$$J = \frac{F}{d}\int_0^d \vec{f}\cdot\vec{dy}, \tag{4}$$

where d is the distance between electrodes and $\vec{f}$ is the ion flux inside the polymer $\left[\frac{mol}{s\ m^2}\right]$, F is Faraday constant. The J is the current density in the electrode. By knowing the J, the current and also voltage in the electrode could be calculated using numerical methods. Therefore the electrodes are described as a conductive media by using the following equation:

$$\sigma\nabla V = \vec{j}, \tag{5}$$

where V is voltage in the electrode, σ is electric conductivity and $\vec{j}$ is current density. Similar equation for the electrodes have been used also in so called gray box models [13]. The different way to express Eq. (5) is

$$\nabla V = RI, \tag{6}$$

where R is resistance per unit length and the equation is common form of the Ohm's law. The general boundary condtitions for the electrode are shown in Figure 2. The specific cases are discussed in the following sections.

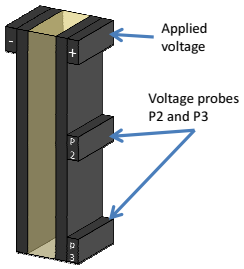

Figure 3. During the experiment, $1V$ was applied to the IPMC a... probes P2 and P3 were used to measure the voltage drop along the electrode.

3 EXPERIMENTAL SETUP

To validate the model, a series of experiments were conducted. The used material was Nafion 117 membrane, which was pretreated and plated with Pt. During the pretreatment, the membrane was immersed into 3% hydrogen preoxide (H_2O_2) to eliminate organic impurities and in $0.5M$ sulfuric acid (H_2SO_4) for removal of metallic impurities. Then Pt metal particles were deposited on the polymer membrane. The pretreated polymer membrane was immersed in a platinum complex solution $(Pt(NH_3)_4Cl_2 \cdot H_2O)$. In order to increase the surface conductivity, an additional platinum layer was deposited using a developing process. 5% aqueous solution of hydroxylamine hydrochloride $(H_2NOH \cdot HCl)$ and 20% solution of hydrazine $(NH_2NH_2 \cdot H_2O)$ were added to reduce the platinum. More details of the similar process could be found in [14].

During the experiment, different samples with various configurations were used. The overall experimental procedure was as follows: a sample was fixed in a certain configuration and three voltage probes were attached (see Figure 3). Then the voltage of $1V$ was applied to a one end of the sample for $10s$. The probes registred the voltages on the electrode and another probe also registred the overall current by measuring voltage drop on a 1Ω resistor. IODaq Personal Daq/56 device was used to measure and store the voltage values. The surface resistivity was measured with a four-point probe (Gardian, Model RSM-232).

Three different experimental configurations were used (see Figure 4). To validate the 2D model, the voltage measurements were done with a straight IPMC and with a significantly bent IPMC (a and b in Figure 4). The probes were attached to the middle of the cathode and to the end of the cathode. To validate the 3D model, a sample with nonuniform surface resistance was picked. This time, both probes were attached to the end of the cathode (c in Figure 4).

4 RESULTS AND DISCUSSION

The Comsol Multiphysics Finite Element software package was used to carry out the simulations. The choice of software al-

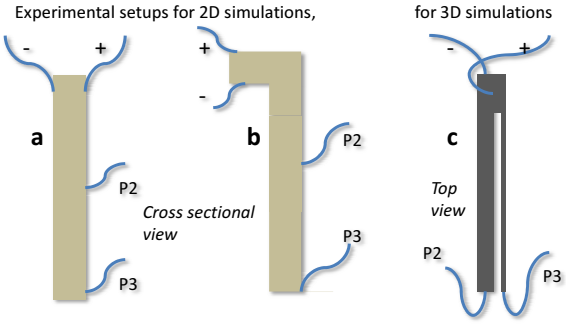

Figure 4. Different experimental setups were used to validate the model. The setups **a** and **b** were used to validate the 2D model - first for a straight IPMC and then for a considerably bent IPMC. The setup **c** was used to validate 3D model - probes were attached to the end of the forks.

lowed to use a predefined set of equations for different domains. The simulation of the metallic electrode coupled with the base model is rather complex, because the currents in the electrode cause the changes of voltage applied on the polymer, i.e this is a sort of feedback system. So building the model involved generally the following steps:

1. A domain for the base model was constructed and the equations (1) and (2) were applied.
2. One electrode domain was simulated. The electrode conductivity was obtained by measuring the surface resistance with 4 point probe. The simulation value of conductivity, however, was altered a bit as the surface is not uniform for the case of IPMC and exact thickness of Pt was unknown. The electrode thickness was assumed to be $5\mu m$. It is suggested though, that platinum layer is $1\mu m$, but as the model does not consider the platinum diffusion zone, the layer was chosen to be a bit thicker instead [15].
3. The voltage of the electrode boundaries which coincided with polymer, were set to be equal to the electric potential ϕ.

The tetrahedaral mesh was used for two dimensional domain. The mesh was refined near the boundaries and electrode. Meshing in a three dimensional domain was not as straightforward as it was for two dimensions. First of all, two dimensions of an IPMC sheet are relatively large (width and length are in range of centimeters) but the thickness is really small, much less than a millimeter. In addition, the model consists of two separate domains - a polymer backbone and a thin layer of metal coating. The tetrahedral meshing was used as this is the only type of mesh which supports projection couping (necessary to calculate Eq. (4)). The downside of the mesh is larger solution time. To optimize the calculations, two large dimensions of IPMC were reduced 100 times and this was taken into account for current and the electrode conductivity calculations.

Figure 5. The model to validate experimental setups **a** and **b** (see Figure 4). For the case **a**, conductivities $\sigma_1 = \sigma_2$. For the case **b**, $\sigma_2 > \sigma_1$.

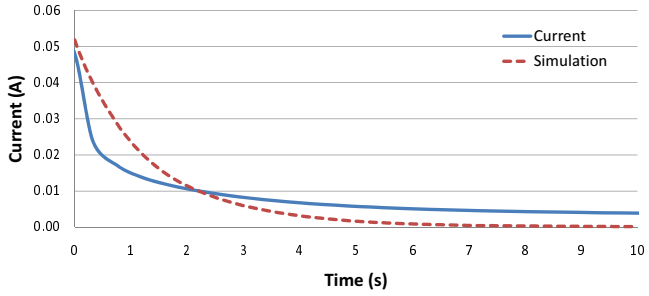

Figure 6. 2D model for experimental setup **a**. Simulated and measured total electric current.

4.1 2D and 3D simulations

To carry out the simulations, the boundary conditions between the backbone polymer and the electrode were described as follows:

1. For the electrode domain with Eq. (5), the boundary conditions was set as the current outflow, calculated using the Ramo-Shockley theorem Eq. (4).
2. For the backbone polymer domain, where the Gauss equation describes the electric potential, the electric field strength $E = V/d$ was set, where V is the voltage, calculated in the electrode domain and d is the polymer thickness with the value of $180\mu m$.
3. Positvive voltage of $1V$ was applied to the short section of the electrode (the very left section of electrode in figures 2 and 5).

Notice that those boundary conditions form sort of loop back system. The ionic current is caused by the voltage in the electrode. In turn, the voltage drop along the electrode is due to the ionic current.

To verify the model and the validity of the boundary conditions, the 2D simulations were carried out. The electrode of IPMC was described as two regions with conductivities σ_1 and σ_2 (see Figure 5). For the first set of the simulations, the condition

$$\sigma_1 = \sigma_2 = 29 \cdot 10^3 \frac{1}{\Omega m} \tag{7}$$

was set. This corresponds to the experimental setup **a** in Figure 4. The simulation results are shown in figures 6, 7, and 8. It could

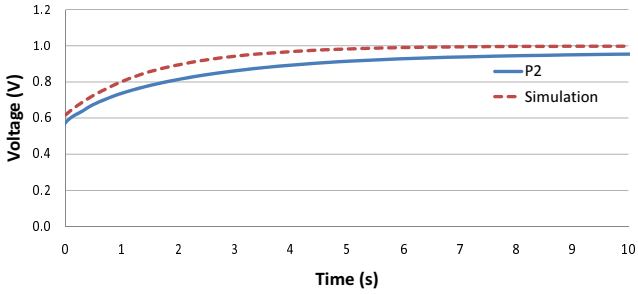

Figure 7. 2D model for experimental setup **a**. Simulated and measured voltage in the middle the electrode of IPMC.

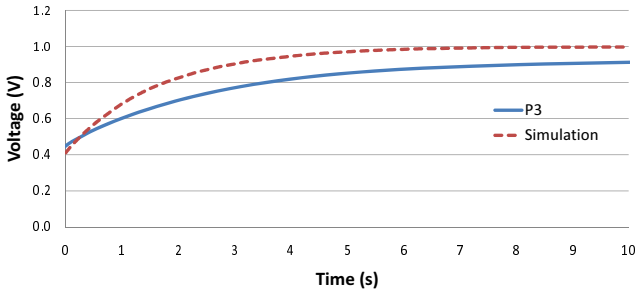

Figure 8. 2D model for experimental setup **a**. Simulated and measured voltage in the end the electrode of IPMC.

be seen that the simulation results correspond to the measured values sufficiently well. For the second set of simulation, the conductivity of σ_2 was decreased as this corresponds to a significantly bent IPMC. The conductivity value of the "bending" region was set to:

$$\sigma_2 = 14.5 \cdot 10^3 \frac{1}{\Omega m} \tag{8}$$

. The measurement results show that the conductivity of this region is roughtly two times smaller than the conductivity of the straight section of the electrode. It must be also noted, that even though the conductivity was measured with a four point probe, the simulation values were changed, because we do not have the exact knowledge of the thickness of the electrode. Furthermode, the Pt electrode is not a solid piece, but rather consists of small section of platinum [1]. Therefore the exact conductivity could not be measured, but was roughly estimated. However, comparing the simulated voltage drop on the electrodes to the measured one, we can see that the proposed model and the conductivity values are justified. The simulation results are shown in figures 9, 10, and 11.

The purpose of 3D simulations was to demonstrate the model viability when it comes to nonuniform surface resis-

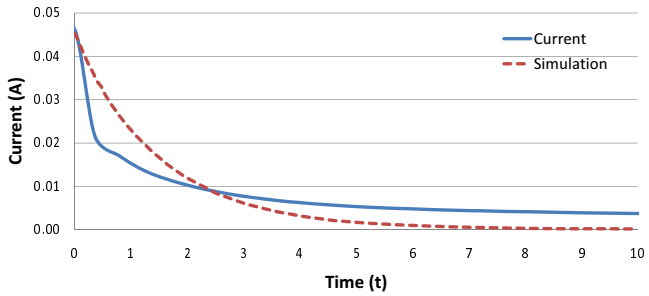

Figure 9. 2D model for experimental setup **b**. Simulated and measured total electric current.

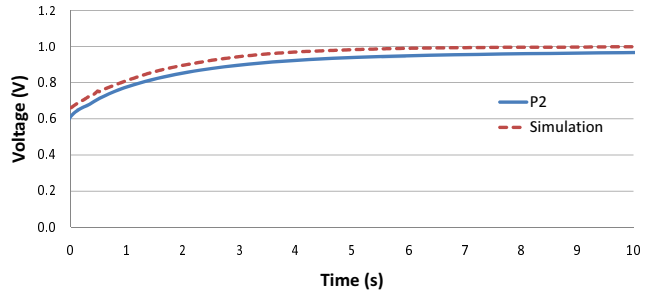

Figure 10. 2D model for experimental setup **b**. Simulated and measured voltage in the middle the electrode of IPMC.

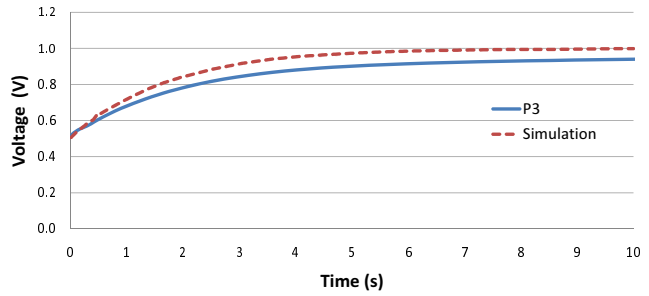

Figure 11. 2D model for experimental setup **a**. Simulated and measured voltage in the end the electrode of IPMC.

tance. A sample with variable surface characteristics was chosen and cut to the fork-like shape. The conductivity of both forsk is very different as also the conductivity of the base (see Figure 12). Again, some rough measurements were done and the estimated values based on the measurementes were used in the simulations:$\sigma_1 = 15 \cdot 10^3 \frac{1}{\Omega m}$, $\sigma_2 = 29 \cdot 10^3 \frac{1}{\Omega m}$, and $\sigma_3 = 10 \cdot 10^3 \frac{1}{\Omega m}$. The results of simulated and measured current is shown in Figure 13 and voltages in Figure 14. Notice that for the 3D simulations, the measured and simulated voltages are more off than for the 2D case. It is due to the fact that at this point

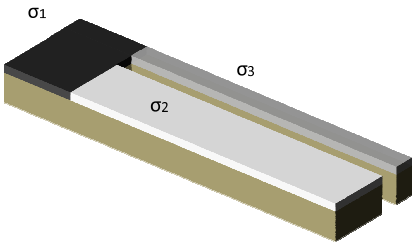

Figure 12. 3D model to validate experimental setup **c**. See Figure 4. Each area has different conductivity, so $\sigma_1 \neq \sigma_2 \neq \sigma_3$.

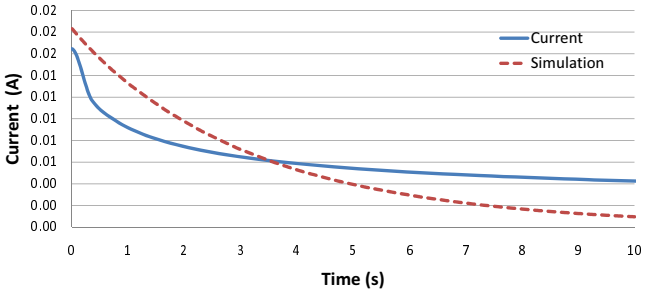

Figure 13. The measured and simulated current for setup **c**.

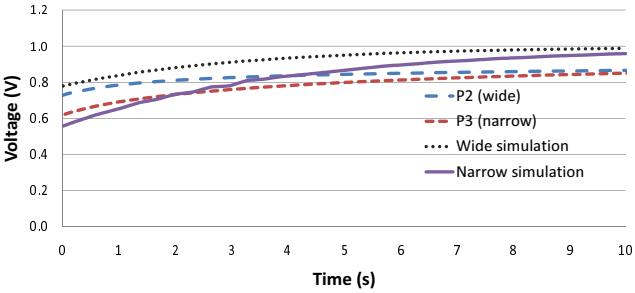

Figure 14. The measured and simulated voltages in the end of the forks for setup **c**.

of the research, the 3D model is still being developed and more optimizations are needed, i.e. the model does not converge for all simulation constant values, therefore varying of some critical constants such as diffusivity was necessary. However, the results do demonstrate, that the developed model is promising.

4.2 Optimization

As described before, the electrode model is very complicated due to the voltage-current dependencies between the polymer and electrode domains. The solution time is unreasonably large, especially for 3D simulations. Part of the work was to explore how to optimize the model to reduce the solution time and

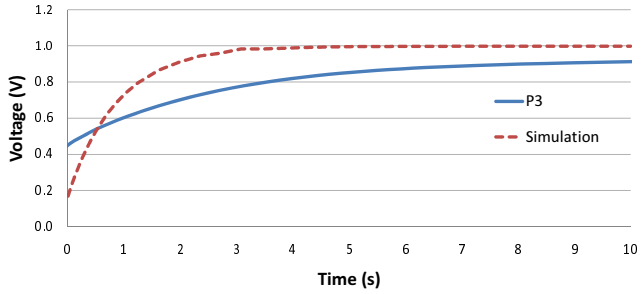

Figure 15. Optimized model for experimental setup **a**. Notice that the optimization has caused large discrepancy between the simulated values and the experimental data.

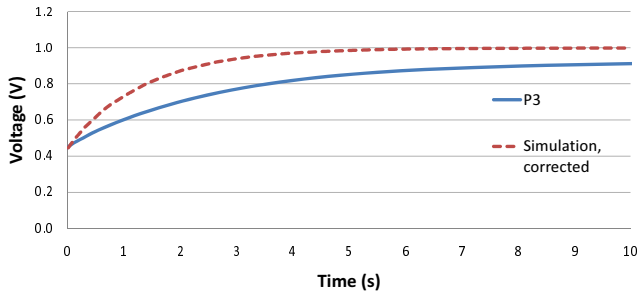

Figure 16. Optimized and enhanced model for experimental setup **a**. Changing the constant of diffusion reduced the gap between the simulated values and the experimental data.

improve the convergence for different constants. At this stage of the research, one possible simplification was studied. Instead of using boundary conditions $\phi = V/d$, where ϕ is potential inside the polymer and V is the potential calculated in the electrode, the condition $\phi = Vpos/d$ could be used. Here $Vpos$ is just a positive constant and equals to $1Volt$. The effect of the change is that the voltage drop, which depends on the ion flux, does not in turn influence the flux. The downside of this simplificatio is that the model underestimates the initial voltage drop and overestimates the voltage in time. This is demonstrated in Figure 15. On the other hand, the effect could be reduced when using different diffusion constant. Figure 16 shows the same simulation with slightly larger diffusion constant value. Despite of the fact, that simulated voltage value could be somewhat corrected, the calculated current value remains wrong.

Other possible optimizations include the technical enhancements of the simulations. Due to the current-voltage coupling, the weak solution form cannot be used with Comsol Multiphysics solvers. Also the mapped meshing is not available, when flux integration over a subdomain is used during the solution. But there are some areas, what could be studied. For instance, the meshing can be improved to reduce the solution time. One way is to use Comsol meshing engine to fine-tune the tetrahedral mesh. Also some alternatives could be consider. For instance open-source software Gmsh is very powerful tool to manipulate meshes [16]. A sample mesh for IPMC is shown in Figure 17. Furthemore, different Comsol solvers with fine tuned parameters can improve the convergence and solution time even more significantly.

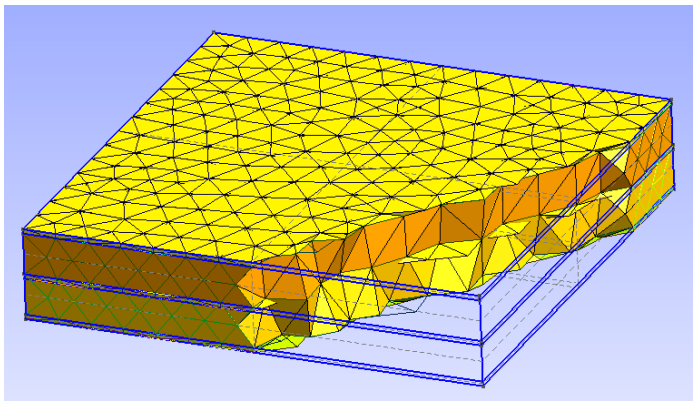

Figure 17. Example mesh generated with gmsh tool.

5 CONCLUSIONS

We have developed a base model to simulated the simple physical processes such as ion migration and electric field change in an IPMC. In this paper we have extended the base model to three dimensions, added surface electrodes and modeled the currents and voltage drop on them. The Ramo-Shockley theorem, which is mostly known in the other fields of physics, was used to couple the ionic current and the current in the electrodes. The non-uniform surface resistance was simulated and validated against the experimental results. Altough the developed model gives physically meaningful results, more optimization is needed to improve the model in terms of solution time and convergence. After the improvements, the model will be applicable for variety of IPMCs to estimate the current consumption and the voltage drop on the electrodes and therefore provide more precise deformation of IPMC.

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