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Modeling and simulation of a chemically stimulated hydrogel bilayer bending actuator

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ABSTRACT

Stimuli-sensitive hydrogels are polymeric materials, which are able to reversibly swell in water in response to environmental changes. Relevant stimuli include variations of pH, temperature, concentration of specific ions etc. Stacked layers composed of multiple thin hydrogels - also referred to as hydrogel-layer composites - combine the distinct sensing properties of different hydrogels. This approach enables the development of sophisticated microfluidic devices such as bisensitive valves or fluid-sensitive deflectors. In order to numerically simulate the swelling of a polyelectrolyte hydrogel in response to an ion concentration change the multifield theory is adopted. The set of partial differential equations - including the description of the chemical, the electrical and the mechanical field - are solved using the Finite Element Method. Simulations are carried out on a two-dimensional domain in order to capture interactions between the different fields. In the present work, the ion transport is governed by diffusive and migrative fluxes. The distribution of ions in the gel and the solution bath result in an osmotic pressure difference, which is responsible for the mechanical deformation of the hydrogel-layer composite. The realized numerical investigation gives an insight into the evolution of the displacement field, the distribution of ions and the electric potential within the bulk material and the interface between gel and solution bath. The predicted behavior of the relevant field variables is in excellent agreement with results available in the literature.

Keywords: Multifield theory, hydrogel layer composites, modeling, simulation, Finite Element Method

1. INTRODUCTION

In the 1980s the wide field of microfluidics emerged. It is used for many applications as in micro-propulsion systems, lab on a chip technologies or DNA microarrays as well as in inkjet printheads. Conventional valves and actuators are fast in response, well-established and resilient. Nevertheless, it is complicated to use them in microfluidic devices, since they require an external energy supply (cables, hoses etc.), and are very limited when it comes to miniaturization. Intelligent materials, such as hydrogels, offer great opportunities to enhance present systems and make them smaller, simpler, cheaper and even responsive to different stimuli. As has been shown, hydrogel layer-systems are very promising for the development of microfluidic valves,¹⁻³ which for example act like autonomous venous valves.⁴ Also hydrogel layer-systems can be used for many sensoric applications⁵⁻⁸ as well as origami-like structures^{9,10} for use in microfluid devices for storage or computing purposes.

Polyelectrolyte hydrogels are three-dimensionally crosslinked polymers which are able to gain up to ten times of its original size by a reversible uptake of water. Since they are made from polyelectrolytes, they own a relatively large number of ionizable functional groups which are attached to the backbone polymer by strong covalent bonds. Within the void of the polymer network, water with mobile ions are present. The polymer network is a porous structure which allows the mobile ions to pass through and also to leave the hydrogel into the surrounding solution bath. Since the ion transport inside of the hydrogel is diffusion dominated,¹¹ the response time of hydrogels is size-dependent. This size dependence is the reason why hydrogel applications are mainly found on the microscale, since their response is too slow on the macroscale.

Despite of the promising applications of hydrogels and hydrogel-layer systems, only few were actually realized. This is mainly due to the lack of knowledge about their response behavior and material characteristics during the swelling / deswelling process. This contribution aims at filling a gap in modeling a hydrogel layer system in

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a classical bilayer configuration¹² as commonly used in bilayer thermostats. For the modeling of the hydrogel bilayer configuration, a coupled multifield theory has been developed.^{13,14} A set of partial differential equations describing the chemical, electrical and mechanical fields is solved on a two-dimensional domain using the Finite Element Method.

This contribution is structured in the following way: First, a brief introduction into the used coupled multifield theory is given in Section 2. In Section 3, the modeling of a simple hydrogel bilayer bending actuator is conducted and the obtained numerical results are discussed. For this, three different material combinations are investigated and compared. A short conclusion is drawn in Section 4.

2. FIELD FORMULATION

In the following, the used coupled chemo-electro-mechanical multifield theory is briefly described. For further information, the interested reader is referred to Refs.^{13–15}

2.1 Chemical Field

The transport of ions inside of the gel and the solution domain is governed by diffusion and migration. Under the assumption that neither chemical conversion nor convection occurs, the mass balance for the mobile ionic species can be formulated as

$$\dot{c}^\alpha = \left(D^\alpha c^\alpha_{,i} + D^\alpha \frac{F}{RT} z^\alpha c^\alpha \Psi_{,i} \right)_{,i} . \quad (1)$$

Here, the expressions $(\cdot)_{,i}$ and $\dot{(\cdot)}$ denote the derivatives $\partial(\cdot)/\partial x_i$ and $d(\cdot)/dt$, respectively. The index α indicates the affiliation to the mobile ion species α . Within this contribution, only two mobile species are considered that is one positive (+) and one negative (-) species. The variable c denotes the ion concentration, D the diffusion constant, $F = 96487 \text{ C/mol}$ the Faraday constant, $R = 8.3143 \text{ J/(mol}\cdot\text{K)}$ the universal gas constant, T the absolute temperature in Kelvin, z the valence of the respective ion and Ψ the scalar electric potential.

2.2 Reference Concentration

The reference concentration is determined by

$$\dot{c}^{\text{ref}\alpha} = \left[D^{\text{ref}\alpha} c^{\text{ref}\alpha}_{,i} \right]_{,i} , \quad (2)$$

where $D^{\text{ref}\alpha}$ denotes the Diffusion constant of the reference concentration of the mobile ion species α . In this contribution, only a diffusive-like behavior is modeled. For further information on the reference concentration, please refer to Sobczyk and Wallmersperger.¹⁶

2.3 Electrical Field

To describe the electrical field, it is possible to apply the Poisson equation for electrostatics

$$\Psi_{,ii} = -\frac{\rho}{\varepsilon^0 \varepsilon^r} , \quad (3)$$

with the volume charge density

$$\rho = F \sum_{\gamma} (z^\gamma c^\gamma) , \quad (4)$$

where $\varepsilon^0 = 8.8542 \cdot 10^{-12} \text{ As/(Vm)}$ denotes the vacuum permittivity and ε^r the relative permittivity of the material. The index γ indicates summation index over the different mobile and fixed ion species. These can be mobile cations (+) and anions (-), as well as fixed anions (fc-) or cations (fc+). The operator $(\cdot)_{,ii}$ denotes the Laplace operator.

Figure 1. Geometry of the investigated hydrogel bilayer system.

2.4 Mechanical Field

In order to obtain a field solution for the mechanics, the linear momentum equation in the form

$$\sigma_{ij,j} = E_{ijkl}(\varepsilon_{kl}^{\text{tot}} - \varepsilon_{kl}^{\pi}) \quad (5)$$

is utilized. Here, E_{ijkl} denotes the tensor of elasticity, $\varepsilon_{kl}^{\text{tot}}$ the tensor of total strain and ε_{kl}^{π} the strain contribution resulting from isotropic swelling. The swelling strain is obtained by

$$\varepsilon_{kl}^{\pi} = \Gamma \delta_{kl} \Delta \pi^{\text{osm}}. \quad (6)$$

Here, Γ denotes a swelling coefficient and δ_{kl} the Kronecker Delta. The osmotic pressure $\Delta \pi^{\text{osm}}$ can be approximated using

$$\Delta \pi^{\text{osm}} = RT \sum_{\alpha} (\Delta c^{\alpha} - \Delta c^{\alpha 0}) \quad \text{with} \quad \begin{cases} \Delta c^{\alpha} = c^{\alpha} - c^{\alpha \text{ref}} \\ \Delta c^{\alpha 0} = c^{\alpha} - c^{\alpha \text{ref}0} \end{cases}. \quad (7)$$

3. NUMERICAL INVESTIGATION

In the following, the field equations are numerically solved by using the Finite Element Method. A bilayer configuration is chosen to demonstrate the capabilities of the applied method. Different kinds of hydrogel combinations are investigated in order to obtain a qualitative understanding of the processes in the respective configurations.

A simple bilayer with a size of $L_G \times (B_{G1} + B_{G2})$ as depicted in Figure 1 is investigated. The geometric dimensions of the gels are chosen to be $B_{G1} = B_{G2} = 0.3 \text{ mm}$ and $L_G = 5 \text{ mm}$. The solution bath is modeled as a thin layer around the bilayer, having a thickness of $L_S = B_S = 2 \cdot 10^{-4} \text{ mm}$. Only a thin film of the solution bath is modeled in order to minimize the time of diffusive transport in the solution bath, but still have the possibility to prescribe the concentration in the solution bath as Dirichlet Boundary condition. This approach is applicable as long as the presence of the gel system does not influence the ion concentration in the solution bath. The bilayer is in a clamped configuration on the left side, while the other side can move freely. The bilayer consists of two different gels, which are stacked. The bottom gel layer is an anionic gel, the other gel layer is either anionic (testcase 1), cationic (testcase 2) or neutral (testcase 3). Initially, the bilayer is located in a solution bath with a mobile ion concentration of $c^+ = c^- = 1 \text{ mM}$. At $t = 0$, the solution bath is exchanged and the concentration of mobile ions is reduced to $c^+ = c^- = 0.5 \text{ mM}$. No external voltage is applied to the system.

The diffusion constants inside of the solution bath are chosen to be equal to the ones of Na^+ and Cl^- ions in water as described by Yuan-Hui et al.¹⁷ for sea water. Inside of the gel layers, the diffusion is slower due to the existence of the polymer network. Here, a value as used by Acartürk et al.¹⁸ is assumed. The relative permittivity ε_r inside of the gels as well as in the solution bath is chosen to be equal to the one of water at room temperature, as measured by Catenaccio et al.¹⁹ The swelling coefficient in gel 2 and gel 1 are chosen to be

equal. The neutral gel is assumed to exhibit no swelling behavior and therefore the swelling coefficient is set to zero. The Young's modulus of the gel layers is set to the value of Poly(N-isopropylacrylamide) as measured by Matzelle et al.²⁰ using Scanning Force Microscopy. The hydrogels are assumed to be almost incompressible and therefore a Poisson's ration close to 0.5 is used for the gel layers. Inside of gel 1, the concentration of fixed anions is set to $c^{\text{fc-}} = 1 \text{ mM}$, which is a value as used by Wallmersperger et al.¹³ The concentration of fixed charges inside of gel 2 is chosen to be dependent on the respective testcase, but to be in the same order of magnitude as in gel layer 1. All used material parameters are listed in Table 1. The material properties for the different testcases are listed in Table 2. To solve the initial boundary value problem suitable initial and boundary conditions have to be prescribed.

Table 1. Used material parameters.

material parameter	solution bath	gel 1	gel 2
Young's modulus E (kPa)	-	10	10
Poisson ration ν (-)	-	0.499	0.499
expansion coefficient Γ (1/Pa)	-	$1 \cdot 10^{-4}$	see Table 2
diffusion coefficient D^+ (m^2/s)	$1.33 \cdot 10^{-9}$	$5 \cdot 10^{-10}$	$5 \cdot 10^{-10}$
diffusion coefficient D^- (m^2/s)	$2.03 \cdot 10^{-9}$	$5 \cdot 10^{-10}$	$5 \cdot 10^{-10}$
relative permittivity ε_r (-)	80	80	80
fixed anion concentration $c^{\text{fc-}}$ (mM)	0	0.5	see Table 2
fixed cation concentration $c^{\text{fc+}}$ (mM)	0	0	see Table 2

Table 2. Used material parameters for gel 2 in the different testcases.

parameter	testcase 1 (anionic-anionic)	testcase 2 (anionic-cationic)	testcase 3 (anionic-neutral)
fixed anion concentration $c^{\text{fc-}}$ (mM)	1	0	0
fixed cation concentration $c^{\text{fc+}}$ (mM)	0	1	0
expansion coefficient Γ (1/Pa)	$1 \cdot 10^{-4}$	$1 \cdot 10^{-4}$	0

Initial Conditions In initial state, the bilayer is undeformed. In order to obtain correct initial conditions for the chemo-electrical field, a foregoing simulation is made. This simulation is performed in order to find the correct values for the mobile ion concentration inside of the interface layers. For this foregoing simulation, initial conditions are prescribed according to the Donnan equilibrium with a concentration of positive and negative ions in the solution bath of $c^+ = c^- = 1 \text{ mM}$ and no applied electric potential.

Boundary Conditions On the left side of the system, a no flux condition $J_i^+ n_i = J_i^- n_i = 0$ is prescribed since the container of the system is assumed to be closed. Here, n_i is the outward normal vector. To account for the chemical stimulation by exchange of the solution bath, the ion concentrations at the other outer boundaries of the thin solution bath film are set to a fixed value of mobile ion concentrations $c^+ = c^- = 0.5 \text{ mM}$ at $t \geq 0$. This is a good assumption if the whole solution bath is large and the overall ion concentration inside of the solution bath is not altered due to the presence of the hydrogel layer system. The electric potential is prescribed as a Dirichlet boundary condition of $\Psi = 0 \text{ V}$ on the top, right and bottom side of the solution bath. On the left side, a vanishing electric field $\mathcal{E}_i n_i = 0$ is prescribed. The momentum balance is only solved inside of the gel layers and not in the solution bath. Therefore, the boundary conditions for the mechanical field are prescribed on the outer boundaries of the bilayer. The fluid structure interaction between the gel bilayer and the solution bath is neglected, therefore a stress free boundary between gel and solution bath is prescribed $\sigma_{ij} n_i = 0$. The left side of the gel is in a clamped configuration. It is assumed, that the displacement in x -direction is blocked, that is $u_x(x=0) = 0$, and that the center of the gel layer is fixed in the y -direction, that is $u_y(x=0, y=0) = 0$.

Analytical results First, the results of the chemo-electrical field are discussed. Due to the reduction of the mobile ion concentration in the solution bath at $t = 0$, the ion concentration in the gel layers is also reduced. The concentration of mobile ions in the different gels can be analytically calculated by the equilibrium condition of equal chemical potentials in the gel μ^{gel} and the solution bath μ^{sol}

$$\mu_{\text{gel}} = \mu_{\text{sol}} \quad (8)$$

and the electroneutrality condition

$$\sum_{\gamma} (z^{\gamma} c^{\gamma}) = 0. \quad (9)$$

The chemical potential μ^{α} of ion species α can be calculated by

$$\mu^{\alpha} = \left[\frac{\partial U(S, V, n^{\alpha})}{\partial n^{\alpha}} \right]_{S, V, n^{\alpha}} = \mu_0^{\alpha} + RT \ln \frac{c^{\alpha}}{c_0}, \quad (10)$$

for ideal solutions. Under the assumption of only one fixed ionic species per domain this leads to

$$c_{\text{gel}}^{+} = -\frac{z^{\text{fc}}}{2z^{+}} \pm \sqrt{\left(\frac{z^{\text{fc}}}{2z^{+}} \right)^2 + (c_{\text{sol}}^{+})^2} \quad (11a)$$

$$c_{\text{gel}}^{-} = -\frac{z^{\text{fc}}}{2z^{-}} \pm \sqrt{\left(\frac{z^{\text{fc}}}{2z^{-}} \right)^2 + (c_{\text{sol}}^{-})^2}. \quad (11b)$$

Here, U denotes the internal energy of the system, S the entropy, V the volume, c_0 a normalization concentration, μ_0^{α} the chemical reference potential and n^{α} the amount of ion species α . The calculated analytical solution for the investigated testcases are listed in Table 3 for the new equilibrium.

Figure 2. Distribution of the mobile and fixed ions in the hydrogel bilayer system along y at $x = \frac{1}{2}L_G$ in equilibrium state.

Numerical results The field equations (1), (2), (3) and (5) with the aforementioned initial and boundary conditions are numerically solved using the Finite Element Method. In Figure 2, the numerical results for the distribution of mobile and fixed ions in the equilibrium state is depicted for the different testcases along the y -coordinate at $x = \frac{1}{2}L_G$. Outside of the interface layers between different domains (solution bath, gel 1 and gel 2), the analytical solution and the numerical results are identical for both the ion concentrations and the

electrical potential. The described analytical model is not capable of predicting the distribution of the respective field variables in regions where an electric potential gradient is present, in this case inside of the boundary layer between adjacent domains.

Figure 3. Investigation at the interface layer between gel 1 and gel 2: Distribution of a) mobile cations, b) mobile anions c) reference concentrations, d) electric potential, e) volume charge density and f) osmotic pressure difference along y at $x = \frac{1}{2}L_G$ in equilibrium state.

In Figures 3a and 3b, the numerical solution for the distribution of mobile anions and cations at the boundary layer between the gel layers is given for the different testcases at equilibrium, plotted along the y -coordinate at $x = \frac{1}{2}L_G$. The mobile ion concentration distribution between gel 1 and gel 2 is continuous and significantly

Table 3. Analytical solution for the mobile ion concentration and the electric potential inside the gels in equilibrium state for the different testcases.

field variable and domain	testcase 1 (anionic-anionic)	testcase 2 (anionic-cationic)	testcase 3 (anionic-neutral)
c^+ (mM) in gel 1	0.8090	0.8090	0.8090
c^- (mM) in gel 1	0.3090	0.3090	0.3090
c^+ (mM) in gel 2	1.2071	0.2071	0.5000
c^- (mM) in gel 2	0.2071	1.2071	0.5000
Ψ (V) in gel 1	-0.0124	-0.0124	-0.0124
Ψ (V) in gel 2	-0.0226	0.0226	0

affected by the gradient on the electric potential due to migrative fluxes. The distribution of the electric potential is shown in Figure 3d. It is obvious that the gradient in the electric potential is much steeper if an anionic gel is combined with a cationic gel (testcase 2) than if the anionic gel is combined with an anionic gel (testcase 1). This increased gradient leads in fact to a higher migrative flux in the interface layer between the gels and therefore has an impact on the shape of the respective mobile ion distribution. The different shape of the ion distribution in the interface layer also leads to a different shape in the osmotic pressure difference $\Delta\pi^{\text{osm}}$: For the testcase 1 and 3, the osmotic pressure difference monotonically increases or decreases, while in testcase 2 it first decreases and then increases towards the cationic gel 2, see Figure 3f. Please note, that in the evaluation of the osmotic pressure difference in the equilibrium state, the reference concentrations $c^{\text{ref}+}$ and $c^{\text{ref}-}$ are equal and constant as depicted in Figure 3c.

For the deflection of the investigated bilayer, the distribution of osmotic pressure inside of the interface layer does not have any impact. Therefore, the deflection of the anionic-anionic (testcase 1) and the anionic-cationic (testcase 2) bilayer configuration is equal. For the chemical stimulation, the osmotic pressure difference depends on the concentration of fixed charges, but not on their valence. The resulting deflection of the bilayer tip in the different configurations is given in Table 4. It should be mentioned that the bilayer bends downwards for testcase 1 and 2 and upwards for testcase 3. Due to the swelling of both gels of the bilayer in testcase 1 and 2, the deflection is smaller compared to testcase 3, but the tip displacement in x -direction is larger.

Table 4. Tip displacement of the hydrogel bilayer u_y^{tip} and u_x^{tip} in x - and y -direction in equilibrium.

	testcase 1 (anionic-anionic)	testcase 2 (anionic-cationic)	testcase 3 (anionic-neutral)
u_x^{tip} (mm)	0.1429	0.1429	0.0344
u_y^{tip} (mm)	-0.6520	-0.6520	0.3027

4. CONCLUSION

In this contribution, the chemo-electro-mechanical behavior of a hydrogel bilayer is analyzed. For this, a coupled multifield theory is adopted and solved on a two-dimensional domain using the Finite Element Method. Three different material combinations are investigated, namely an anionic-anionic, an anionic-cationic and an anionic-neutral hydrogel bilayer. The obtained solution for the chemo-electrical behavior is in excellent agreement with analytical models. Furthermore, the model gives an insight into the distribution of the relevant field variables in both - the domains and the interface layer - between adjacent domains. The model predicts a reduction of the osmotic pressure difference in the interface layer between the two adjacent gels of the anionic-cationic bilayer, which was not observed in the other hydrogel combinations. For macroscale applications like the investigated hydrogel bilayer, this effect does not show any impact on the deflection of the bending actuator. Furthermore it could be shown, that the deflection is equal for the anionic-anionic and the anionic-cationic bilayer and significantly higher for the anionic-neutral configuration. For the fabrication of hydrogel bending actuators for chemical stimulation, we therefore advise to use an ionic hydrogel paired with a soft passive layer.

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