Numerical Simulation of Mechanical Sensors using Hydrated IPMCs

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Abstract

The present study introduces the numerical simulation of mechanical sensors using IPMCs (ionic polymer-metal composites). IPMCs can be applied into both of actuators (from electricity to deformation) and mechanical sensors (from deformation to electricity), but the existing models of the actuators cannot be inversely applied to the mechanical sensors. The mechanical sensors generate very much smaller electric potential compared to the supplied electric potential of actuators with respect to the same displacement and structure. The non-invertible response of the mechanical sensors is numerically simulated, and the simulation considers hydration and transient behaviors. IPMCs have hydration effect that volume and mechanical stiffness are significantly changed with water uptake. In order to consider the volume swelling due to hydration, the total strains and pore pressure of IPMCs are respectively decomposed into stress-induced and hydration-induced parts. The hydration-induced strain is considered as eigen-strain, and the stress-induced strain and stress-induced pore pressure are employed into Biot poroelastic constitutive equations. The mechanical stiffness of a hydrated IPMC is expressed as empirical relations with water uptake. Furthermore, mechanical sensors using IPMCs show transient response with the relaxation and time lag of reaction force and electric potential. The transient response is modeled with a set of basic equations, e.g. layered Timoshenko beam model, Biot poroelastic model, Darcy-flow model, Poisson-Nernst-Plank model. The instantaneous peak of reaction force is estimated on undrained condition, the relaxation of reaction force is considered with pore pressure and its Poisson effect, and hydration-induced water migration is modeled with hydration potential. The hydration potential is modeled with an empirical chemical potential at free swelling equilibrium and is expressed as a function of water uptake. Next, discretization and numerical formulation with layered finite beam elements is introduced. Lastly, the transient responses of a Flemion-based mechanical sensor are numerically simulated with different deflections, and the distributions of stress, pore pressure, ion concentration and electric potential are obtained with time. Lastly, the numerical simulation is compared with the experiments of a reference.

Keywords : IPMC (Ionic Polymer-Metal Composite), mechanical sensor, numerical simulation, hydration

1. Introduction

IPMCs (ionic polymer-metal composites) have been known as intelligent materials having actuating and sensing functions, e.g. from electricity to deformation and from electricity to deformation. Owing to versatile characteristics such as low voltage, lightweight, easy access and flexibility, IPMCs have attracted attention as prospective applicants for artificial muscles, robotics, MEMS and so on (Jung et al., 2010). For example, Wang et al. (2009) introduced bio-inspired tactile sensors based on Flemion. Such applications are based on actuating and sensing functions with mechanical and electrical measures. In order to investigate the actuating and sensing functions, some papers (Nemat-Nasser and Zamani, 2006 and Jung et al., 2010) have attempted to simulate the actuating behaviors of IPMCs with more complicated models. However, the simulation of mechanical sensors using IPMCs has not been reported except for black box models. Numerical simulation is needed to understand and verify the complicated behaviors of the mechanical sensors, and its quantitative results are necessary for the design and control. Hence, the present study investigates a white box model with continuum mechanics, following macroscopic measurements.

The typical structure of IPMCs is illustrated in Fig. 1. It consists of an ionic conducting polymer and metal deposits. The ionic polymer is a membrane that play roles electrolyte in the transport of ions and solvent, and the metal deposits function as electrodes. In the membrane, ionomers are composed of hydrophobic backbone polymers and the covalently attached anions which are
hydrophilic. Through the backbone polymer, mobile cations and water molecules move with mechanical and elector-chemical forces (Nemat-Nasser and Wu, 2003).

The IPMCs have both of actuating and mechanical sensing functions, but the functions are not compatible with each other. Wang et al. (2009) investigated Flemion-based IPMCs with the generated voltage of mechanical sensing mode and the supplied voltage of actuating mode. The generated voltage of sensing mode is very much smaller than the supplied voltage of actuating mode with respect to the same displacement and structure. The non-invertible response of the mechanical sensors is shown in Fig. 2.

The very small electric potential of mechanical sensors using IPMCs is explained with the following mechanism. When porous IPMCs are mechanically deformed, the pressure of fluid in pores is changed by mechanical interaction between solid matrix and pore fluid. Subsequently, non-uniform pressure causes water and ions to be pushed out from shrunk side and to be sucked into the dilate side as shown in Fig. 1. The movement of the ions causes the instant unbalance of charge distribution, and the gradient of charge density results in electric potential. Therefore, electricity is generated corresponding to the magnitude of the mechanical stimulation (Wang et al., 2009).

Upon the above mechanism, the distributions of interesting parameters in the microscale thickness of IPMCs are very important, but the distributions are not easy to be obtained experimentally. Here, numerical simulation can be added to the quantitative estimation and design of mechanical sensors using IPMCs. Until now, the distributions of the interesting parameters in the mechanical sensing response have not been reported in spite of those in the actuating response. The detailed models of IPMC actuators (Nemat-Nasser and Zamani, 2006 and Jung et al., 2010) have been proposed, but the existing models of the actuators cannot be inversely applied to the mechanical sensors. Therefore, the present study proposes the simulation model of mechanical sensors using IPMCs and reports the simulation results with the distributions of interesting parameters in the thickness direction of IPMCs.

![Fig. 1 Typical structure (L) and ion transport (R) of IPMCs](image1)

![Fig. 2 Non-invertible black box of mechanical sensor and actuator using the same IPMC](image2)

We previously attempted the numerical simulation of mechanical sensors using conducting polymers such as PPy-PVDF-PPy composite (Yoo and Toi, 2013). Differently from the conducting polymers, the ionic conducting polymers of IPMCs, e.g. Flemion and Nafion, have significant hydration effects on volume and mechanical stiffness. Zhao et al. (2011) experimentally showed the volume swelling of Nafion with water absorption. Nemat-Nasser and Wu (2003) measured the volume and mechanical stiffness of Flemion and Nafion with respect to water uptake. The ionic conducting polymers have the significant changes of volume and
mechanical stiffness as water is absorbed due to hydration.

The above hydration effect is modeled in the present simulation. The total strains of ionic conducting polymers are decomposed as stress-induced and hydration-induced parts, and the hydration-induced strain is considered as eigen-strain. The hydration-induced strain covers the eigen-strain due to applied electric force in actuating mode. Nemat-Nasser and Zamani (2006) and Jung et al. (2010) expressed the actuating deformation of IPMCs with eigen-strain. They identified the parameter of ‘hydration’ as water uptake which is defined as the volume fraction of water to polymer, and the hydration parameter leads to eigen-strain in structural analysis. In the present study, the pressure of water in a hydrated IPMC is divided into stress-induced and hydration-induced parts. The stress-induced pressure accounts for the instantaneous increase and subsequent relaxation of pore pressure when mechanical forces are externally applied. The hydration-induced pressure accounts for the contribution of water migration due to hydration. Next, the stress-induced strain and stress-induced pressure are embedded into the constitutive equations of Biot poroelasticity (Biot, 1941). The coefficients of the constitutive equations, which mean mechanical stiffness, are expressed by empirical relations with water uptake from conventional mechanical tests.

The sensing response of IPMCs shows the relaxation and time lag of reaction force and electric potential. In the experiment of Wang et al. (2009), Flemion-based IPMCs show that reaction force is sharply increased and quickly relaxed but electrical potential is gradually increased and slowly decreased. The transient response is resultant from the interactions between polymer matrix, water, mobile ion and electric potential. Modeling the interaction terms, Yoo and Toi (2013) integrated basic equations, e.g. layered Timoshenko beam model, Biot poroelastic model, Darcy-flow model and Poisson-Nernst-Planck model, and they numerically expressed the relaxation and time lag of reaction force and electric potential.

The present study employs and modifies the above models with the followings. The layered Timoshenko beam model simplifies the deformation of the mechanical sensors to reduce the time cost of calculation (Yoo and Toi, 2013). The physical quantities over the thin thickness of IPMCs are dominant factors on the performance of the mechanical sensors, and the gradients of the interesting parameters over the thickness are very large compared with other directions. Biot poroelastic model, describing the behaviors of fluid-saturated elastic porous media, is one of pioneering theories in the poromechanics, and many researchers have still been working on Biot poroelastic model. Biot poroelastic model (Biot, 1941) considers the mechanical interaction between polymer matrix and water. Biot poroelastic model was introduced with undrained parameters (Rice and Cleary, 1976). The undrained parameters are also employed into the modeling the instantaneous peak of reaction force. In the bending of a poroelastic beam, axial stress is affected by the lateral distribution of pore pressure (Scherer et al., 2009). The Poisson effect is modeled with modified undrained Poisson’s ratio (Yoo and Toi, 2013). Darcy-flow model is well known as Darcy law describing the water migration of porous materials. In Darcy-flow model, water migration due to hydration is modeled by embedding hydration potential. The hydration potential is modeled with the empirical chemical potential in the free-swelling of IPMCs. Zhao et al. (2011) obtained the water activity of Nafion with respect to water uptake at free swelling equilibrium. The Poisson-Nernst-Planck model has been frequently employed to the analyses of ion transport and electric potential, e.g. fuel cell and bio-engineering. Nemat-Nasser and Zamani (2006) employed Poisson-Nernst-Planck equations to the simulation of IPMC actuators. The present study also employs Poisson-Nernst-Planck model to the mechanical sensors. With the existence of porous polymer, the total concentration of ion to bulk IPMCs is used, and the convention and diffusion terms are embedded into the Poisson-Nernst-Planck model. As a result, the distributions of mechanical stress, pore pressure, ion concentration and electric potential are estimated with time.

Next, the procedure of numerical simulation is introduced that the multi-fields of layered finite beam elements are discretized and the basic equations are numerically formulated. Lastly, the transient behaviors of a mechanical sensor using a Flemion are numerically simulated with different deflections, and the numerical simulation is compared with the experiment of Wang et al. (2009).

2. Constitutive equations

IPMCs consist of an ionic conducting polymer and metal deposits as shown in Fig. 1. The ionic conducting polymer is considered as a mixture of solid and fluid components. For example, the total volume of the mixture is decomposed into each volume of polymer matrix and water. On the conventional framework, the present study introduces two parameters, water uptake and porosity.

Water uptake is employed as a parameter to account for the hydration level of ionic conducting polymers. Dry ionic conducting polymers absorb a significant amount of water and their volume is spontaneously swollen. The phenomenon is known as hydration. Furthermore, mechanical stiffness is significantly changed with the water absorption. Nemat-Nasser and Wu (2003) identified the parameter of ‘hydration’ as water uptake, and they measured the volume and stiffness of ionic conducting polymers with water
uptake. The water uptake is actually estimated by the mass fraction of absorbed water to dry polymer. The present study would define the water uptake as

\[
W = \frac{m^f}{m^*} = \frac{m^{hydrated} - m^{dry}}{m^{dry}}
\]

(1)

where \( W \) is water uptake, \( m^f \) is the mass of absorbed water, \( m^* \) is the mass of polymer matrix, \( m^{hydrated} \) is the total mass of a hydrated ionic conducting polymer, and \( m^{dry} \) is the mass of the ionic conducting polymer on dry condition.

Porosity is a very common parameter in poromechanics, and is defined as the volume fraction of pores to bulk materials (Biot, 1976). If water is saturated in pores, porosity is equal to the volume fraction of fluid to bulk materials. The porosity is, referring to initial volume, introduced as

\[
\phi = \frac{V^f}{V^0}
\]

(2)

where \( \phi \) is porosity, \( V^f \) is the volume of fluid component, and \( V^0 \) is the total volume of a hydrated ionic polymer on initial condition (\( t = 0 \)).

Assuming that polymer matrix and water are incompressible (but total volume is changed with pore volume), the water uptake in Eq. (1) becomes \( W = V^f / V^0 \). Thus, the following relation between water uptake, porosity and volumetric strain is derived as

\[
e' = \varepsilon'_{kk} = \Delta \phi = \frac{\Delta W}{1 + w_0}
\]

(3)

where, \( e' \) is the total volumetric strain of bulk material, \( \varepsilon'_{kk} \) is the total strains of bulk material and \( W_0 \) is initial water uptake.

The volume of ionic conducting polymers is significantly changed with the humidity of surroundings. In case of actuating mode, the ionic conducting polymers are deformed when electricity is externally applied. On stress-free condition, the ionic conducting polymers can be deformed as electrochemical state is changed. In order to consider the electrochemically induced deformation, the present study decomposes the total strains of ionic conducting polymers into stress-induced strains and hydration-induced strains as follows.

\[
\varepsilon^i_{ij} = \varepsilon_{ij}^s + \frac{e^h}{3} \delta_{ij}
\]

(4)

where, \( \varepsilon^i_{ij} \) is stress-induced strains, \( \varepsilon^s_{ij} \) is hydration-induced strain and \( \delta_{ij} \) is Kronecker delta. The stress-induced strains are observed when mechanical forces are applied, and the hydration-induced strain is observed when water migrates by electrochemical forces. The hydration-induced strain is considered as eigen-strain in structural analysis. Nemat-Nasser and Zamani (2006) modeled the actuating deformation of IPMCs with eigen-strain. The eigen-strain due to electric force is also included in hydration-induced strain.

In the sensing mode of IPMCs, pore pressure is instantaneously changed and relaxed like reaction force (Yoo and Toi, 2013). On stress-free condition, the pore pressure of a hydrated ionic conducting polymer is initially different from the pressure of surroundings like osmosis. The present study decomposes the pore pressure as stress-induced and hydration-induced parts.

\[
p' = p^s + p^h
\]

(5)

where, \( p' \) is pore pressure, \( p^s \) is stress-induced pressure and \( p^h \) is hydration-induced pressure. The stress-induced pressure accounts for the transient part of pore pressure with relaxation when mechanical forces are externally applied. The hydration-induced pressure accounts for the contribution of water migration due to hydration. The hydration-induced pressure becomes total pore pressure at stress-free equilibrium including the actuation behaviors of IPMCs. Nemat-Nasser and Wu (2006) introduced the pressure of cluster assuming one spherical inclusion problem. Jung et al. (2010) modeled fluid pressure to lead to the volumetric strain of Flemion-based actuators. Those pressures in other actuator models are included in the hydration-induced pressure.
Yoo and Toi (2013) employed Biot poroelastic constitutive equations to consider the transient response of mechanical sensors using conducting polymers. Upon the decompositions of strain and pore pressure, the stress-induced strain and stress-induced pressure are embedded into the Biot poroelastic constitutive equations. The thickness of IPMCs is very thin compared to the length, so the bending deformation can be considered within elastic range. The constitutive equations of a hydrated ionic conducting polymer are proposed as

$$
\varepsilon_i^j = \frac{E}{3} \delta_{ij} + \frac{1+\nu}{E} (\sigma_i^j + b(p^i - p^h)\delta_{ij}) - \frac{3\nu}{E} (\sigma_{kk}^j + b(p^i - p^h))\delta_{ij}
$$

$$
= \varepsilon_i^j = \frac{1+\nu}{E}(\sigma_i^j + bp^s\delta_{ij}) - \frac{3\nu}{E} (\sigma_{kk}^j + bp^s)\delta_{ij}
$$

(6)

where, $\sigma_i^j$ is total stress, $E$ is elastic modulus, $\nu$ is Poisson’s ratio and $b$ is Biot coefficient. As mentioned before, the mechanical stiffness of ionic conducting polymers are changed as water is absorbed. Nemat-Nasser and Wu (2003) obtained the elastic modulus of Nafion and Flemion with respect to water uptake. They observed that elastic modulus exponentially decreases with water uptake. The coefficients in constitutive equations are conventionally obtained from static tensile experiments. On the conventional framework, the coefficients of a hydrated ionic conducting polymer are expressed with empirical relations with water uptake. For example, the elastic modulus and bulk modulus of a hydrated ionic conducting polymer can be expressed as exponential functions of water uptake.

$$
E = E_{dry} \exp(-Q w_0)
$$

(7)

$$
K = K_{dry} \exp(-R w_0)
$$

(8)

where $E_{dry}$ is elastic modulus on dry condition, $K$ is the bulk modulus of a hydrated ionic polymer, $K_{dry}$ is bulk modulus on dry condition, $Q$ and $R$ are empirical coefficients.

The Biot coefficient in Eq. (6) is introduced as (Scherer et al., 2009).

$$
b = 1 - \frac{K}{K_{solid}}
$$

(9)

where, $K_{solid}$ is the bulk modulus of solid matrix such that porosity is zero.

The sensing response of IPMCs shows the instantaneous peak of reaction force higher than static reaction force (Wang et al., 2009). The instantaneous peak is explained with the instant contribution of absorbed water to the total stiffness. If water in pores has not enough time to flow, then the water instantly resists deformation. The instantaneous behaviors are modeled with pore pressure on undrained condition as $\Delta(\phi_1) = 0$, where $\rho_1$ is the density of fluid. On the undrained condition, the relation between the stress-induced pressure and the undrained stress is introduced as (Rice, J.R. and Cleary, M.P., 1976)

$$
\Delta p^s = -B \frac{\Delta \sigma_{kk}}{3}
$$

(10)

$$
B = \frac{1}{K} - \frac{1}{K_{solid}} - \frac{1}{K_{fluid}} + \frac{1}{K}
$$

(11)

where, $B$ is Skempton coefficient, $K_{fluid}$ is the bulk modulus of fluid in the pores, and the others are mentioned previously.

On the undrained condition, the mechanical stiffness of IPMCs will be increased, so the undrained stiffness is additionally estimated. The undrained bulk modulus and Poisson’s ratio are introduced as (Rice, J.R. and Cleary, M.P., 1976)
\[ K^u = \frac{K}{1 - bB} \]  
\[ \nu^u = \frac{3\nu + B(1-2\nu)b}{3 - B(1-2\nu)b} \]

where, \( K^u \) is undrained bulk modulus and \( \nu^u \) is undrained Poisson’s ratio.

3. Basic equations

The solid stresses of IPMCs can be obtained by the quasi-static equilibrium equation as

\[ \frac{\partial \sigma_{ij}}{\partial x_j} = 0 \]

The deformation field of IPMCs can be idealized as a bending motion, thereby their constitutive equations are reduced (Nemat-Nasser and Wu, 2006). Yoo and Toi (2013) mentioned that pore pressure is non-linearly and significantly varied in micro-thickness of IPMCs and dominantly affects on the sensitivity of IPMCs. The present study employs layered approach into a beam model to obtain interesting parameters in thickness direction as shown in Fig. 3. The layered Timoshenko beam model (Toi, 2008) is introduced as follows.

\[ u_i = -x_2 \theta \]
\[ \varepsilon_{11} = \frac{\partial u_1}{\partial x_1} = -x_2 \frac{d \theta}{dx_1} \]
\[ \gamma_{21} = \frac{\partial u_2}{\partial x_1} + \frac{\partial u_1}{\partial x_2} = \frac{\partial u_2}{\partial x_1} - \theta \]

where, \( u_i \) is axial displacement, \( u_2 \) is lateral displacement, \( x_2 \) is distance from neutral axis, \( \theta \) is rotation of normal section, \( \varepsilon_{11} \) is axial strain, \( \gamma_{21} \) is transverse shear strain.

Scherer et al. (2009) mentioned Poisson effect that the uniaxial stress of a beam in transient behaviors is affected by the distribution of pore pressure. Yoo and Toi (2013) simply added the Poisson effect into axial stress of beam with modified undrained Poisson’s ratio. Employing the Poisson effect, the constitutive equations of Eq. (6) are reduced in layered Timoshenko beam model as follows.

Fig. 3 Layered Timoshenko beam element
\[
\sigma_{11} = E(\epsilon'_{11} - \frac{e^h}{3}) - bp' - 2v^b bp'
\]  
(18)

\[
\tau_{21} = \alpha G\gamma'_{21}
\]  
(19)

\[
v^b = \beta v^a
\]  
(20)

where, \( G \) is total shear modulus, \( \alpha \) is shear correction factor, \( v^b \) is beam undrained Poisson’s ratio, and \( \beta \) is the correction factor of Poisson effect.

Next, water transport in a hydrated ionic conducting polymer is introduced. From Biot poroelastic theory (Biot, 1941, and Rice and Cleary 1976), water transport in a porous material is modeled as

\[
\frac{\partial \phi}{\partial t} - \nabla \cdot f_w = \frac{\partial e'}{\partial t} - \nabla \cdot f_w = 0
\]  
(21)

where, \( f_w \) is the volume flux of water in pores.

Rice and Cleary (1976) introduced undrained volume change with undrained parameters from Biot poroelastic theory. The present study models the volumetric strain rate with stress-induced and hydration-induced pressures.

\[
\frac{\partial e'}{\partial t} = \frac{9(v^b - v)}{B^2 E(1 + v^b)} \left( B \frac{\sigma_{kk}^w}{3} + p' + p^b \right)
\]  
(22)

The volume flux in Eq. (21) can be described with Darcy law (Biot, 1941). The present study models water migration due to hydration with hydration potential, then the volume flux of water in a hydrated ionic conducting polymer becomes

\[
f_w = -\frac{\kappa_w}{\eta_w} \nabla (p' + p^b + H\Phi + z_{cation} c_{cation} F\psi)
\]  
(23)

where, \( \kappa_w \) is hydraulic permeability, \( \eta_w \) is dynamic viscosity, \( H \) is hydration coefficient, \( \Phi \) is hydration potential, \( z_{cation} \) is cation valence, \( c_{cation} \) is cation concentration, \( F \) is Faraday constant and \( \psi \) is electric potential.

Next, the modeling of hydration-induced strain is introduced with water migration due to hydration. Zhao et al. (2011) estimated the water activity of Nafion with respect to water uptake. From such experiments, the water activity of a hydrated ionic conducting polymer without any external force can be expressed as an empirical relation with water uptake. For example, the water activity in a certain range can be simply assumed as an exponential function with water uptake.

\[
a_w(w) = 1 - \exp(-Aw)
\]  
(24)

where, \( a_w \) is the water activity of a hydrated ionic conducting polymers without any external force and \( A \) is an empirical coefficient.

The hydration potential in Eq. (23) is modeled as an empirical chemical potential which is obtained from free swelling equilibrium without any external force. If the empirical chemical potential is expressed with the above water activity, then hydration potential is expressed as a function of water uptake.

\[
\Phi_w = \mu_{w0} + RT \ln a_w(w)
\]  
(25)

where, \( \mu_{w0} \) is the chemical potential of pure water, \( R \) is gas constant, and \( T \) is absolute temperature.

The hydration-induced pressure is resultant from electrolytically induced phenomena, but it is hydraulic pressure. On free swelling condition, the change of hydration potential accompanies the change of pore pressure with the balance of the mechanical interaction of fluid and solid. Therefore, the hydration coefficient in Eq. (23) is modeled as
\[ H(w) = \frac{\partial p^{h}}{\partial \Phi} \] (26)

Differentiating the hydration potential in Eq. (25) with water uptake leads to

\[ \frac{\partial \Phi}{\partial w} = \frac{RT}{a_{w}} \frac{da_{w}}{dw} = \frac{RTA \exp(-Aw)}{1 - \exp(-Aw)} \] (27)

From Eq. (3) and Eq. (22), the variation of hydration-induced pressure with water uptake is

\[ \frac{\partial p^{h}}{\partial w} = \frac{B^{2}E(1 + \nu^{b})}{9(\nu^{b} - \nu)(1 + w_{0})} \frac{1}{1 + w_{0}} \] (28)

From Eq. (27) and Eq. (28), the hydration coefficient in Eq. (23) is obtained as

\[ H(w) = \frac{B^{2}E(1 + \nu^{b})}{9(\nu^{b} - \nu)(1 + w_{0})} \frac{1 - \exp(-Aw)}{RTA \exp(-Aw)} \] (29)

From Eq. (23) and the related parameters, hydration-induced strain in the transient behaviors of IPMCs is obtained as

\[ \frac{\partial e^{h}}{\partial t} = -\frac{\kappa_{s}}{\eta_{w}} \nabla^{2} \left( p^{h} + HRT \ln a_{w} + \zeta_{\text{cation}} e_{\text{cation}} F \psi \right) \] (30)

Next, the transport phenomena of ions in IPMCs are analyzed with Poisson-Nernst-Plank model. The molar concentration of ions is obtained by Nernst-Planck equation, and electric potential is obtained by Poisson equation with the molar concentration of ions. In ionic conducting polymers of IPMCs, covalently attached anions are assumed as uniformly distributed and fixed, then mobile cations are only considered in the Poisson-Nernst-Plank model (Nemat-Nasser and Zamani, 2006). In the present study, the total concentration of mobile cation in ionic conducting polymers refers to the amount of ion to the total volume of bulk material, \( C_{i} = \phi_{i} c_{i} \), where \( c_{i} \) is molar concentration referring to the amount of ion to the volume of absorbed water. In the present study, the convection with water flow is added in order to express the generation of electric potential in sensing mode. In addition, the interaction between ions and polymer is added with the fraction of ions to the volume of polymer matrix, \( c_{i} V^{f} / V^{s} = c_{i} w_{0} \). The Poisson-Nernst-Plank equations are modified as follows.

\[ \frac{dC_{i}}{dt} = D_{i} \nabla \cdot (\nabla \Phi_{c_{i}}) + \frac{\zeta_{i} F}{RT} \nabla \cdot (\Phi_{c_{i}} \nabla \psi) + D_{i} \nabla^{2} (c_{i} w_{0}) - \nabla \cdot (c_{i} f_{w}) \] (31)

\[ \nabla^{2} \psi = \frac{z}{\kappa_{e}} (C_{i} - C_{i}^{0}) \] (32)

where, \( D_{i} \) is ion-water diffusivity, \( \zeta_{i} \) is valence, \( D_{\psi} \) is ion-polymer diffusivity, \( \kappa_{e} \) is electric permittivity, and \( C_{i}^{0} \) is concentration of immobile ions. As a result, water flow in pores results in the redistribution of ion concentration, then electric potential is generated. The aforementioned Poisson-Nernst-Plank equations, Biot poroelastic theory and layered Timoshenko beam model express the hydration and transient behaviors of mechanical sensors using IPMCs.

4. Numerical procedure

Finite element method is employed into the numerical analysis of mechanical sensors using IPMCs. The sensor is subdivided by layered Timoshenko beam elements. The beam elements are also discretized in the thickness direction on the fields of interesting parameters such as axial stress, pore pressure, ion concentration and electric potential as shown in Fig. 4. The spatial discretization is
firstly determined depending on the convergence in the distribution of pore pressure, because pore pressure near boundary can be rapidly changed. Next, time discretization is determined with numerical stability, e.g. Péclet number. Based on the discretization, basic equations are numerically formulated with the followings.

First, the instantaneous response of IPMCs, when mechanical force is applied, is obtained by considering the undrained condition. Using the undrained stiffness from Eq. (12) and (13), the incremental formulation of finite element stiffness equation is as follows.

\[
[S^u][\Delta u^u] = \{\Delta f^{ext} \}
\]  

(33)

\[
[S^u] = \int [B]^T[D^u][B]dV
\]  

(34)

\[
\{\Delta e^u\} = [B][\Delta u^u]
\]  

(35)

where \([S^u]\) is undrained stiffness matrix, \([\Delta u^u]\) is undrained nodal displacement increment vector, \([\Delta f^{ext}]\) is external force increment vector, \([B]\) is strain-displacement matrix, \([D^u]\) is undrained stress-strain relation matrix, \([\Delta e^u]\) is undrained strain increment vector. The coefficients in the undrained stress-strain relation matrix are determined by undrained parameters. Yoo and Toi (2013) introduced the components in the above stiffness matrix. The undrained behavior accompanies the instantaneous pore pressure on water in pores.

Second, the obtained increment if the undrained strains in Eq. (35) are used for the estimation of the instantaneous change of pore pressure with Eq. (10). The change of pore pressure on the undrained condition is added into stress-induced pressure. The stress-induced pressure of \(n\)-th layer, \(p_n^s\), is updated as follows.

\[
p_n^s = p_n^s - B \frac{\Delta \sigma^u_{ik}}{3} = p_n^s - BK_n^u \Delta \varepsilon_n^u = p_n^s - BK_n^u (1 - 2\nu_n^b) \Delta \varepsilon_{s,n}^u
\]  

(36)

Third, the above stress-induced pressure becomes non-uniformly distributed, so water in pores starts to flow. The amount of migrated water is estimated by the gradient of the non-uniform stress-induced pressure. At the same time, the stress-induced pressure is also relaxed as water is migrated. The phenomenon is modeled from Eq. (23), then the increment of stress-induced pressure is obtained with the followings.

\[
(f^{w1})_{n+1} = -\frac{\kappa_h}{\eta} \frac{p_{n+1}^s - p_n^s}{\Delta x_2}
\]  

\[
(f^{w1})_n = -\frac{\kappa_h}{\eta} \frac{p_n^s - p_{n-1}^s}{\Delta x_2}
\]  

(37)

\[
\Delta \varepsilon_n^s = \frac{\Delta t}{\Delta x_2} \left((f^{w1})_{n+1} - (f^{w1})_n\right) \Delta t
\]  

\[
\Delta p_n^s = \frac{B^2 E (1 + \nu_n^b)}{9(\nu_n^b - \nu)} \Delta \varepsilon_n^s
\]  

(38)

where, \(f^{w1}\) is volume flux with stress-induced pressure and \(x_2\) is local coordinate in the thickness of a beam element as shown in Fig. 3.

Fourth, hydration-induced strain is calculated with Eq. (30). The increment of the hydration-induced strain is as follows.

\[
(f^{w2})_{n+1} = \frac{\kappa_h}{\eta} \frac{p_{n+1}^h - p_n^h}{\Delta x_2}
\]  

\[
(f^{w2})_n = \frac{\kappa_h}{\eta} \frac{p_n^h - p_{n-1}^h}{\Delta x_2}
\]  

(39)

\[
(f^{w3})_{n+1} = \frac{\kappa_h}{\eta} \frac{H_{n+1}\Phi_n - H_n\Phi_n}{\Delta x_2}
\]  

\[
(f^{w3})_n = \frac{\kappa_h}{\eta} \frac{H_n\Phi_n - H_{n-1}\Phi_{n-1}}{\Delta x_2}
\]  

(40)
\[
(f^{w4})_{n+1} = \frac{K_b}{\eta} z_{\text{cation}} c_{\text{cation}} F E_{n+1} \\
(f^{w4})_n = \frac{K_b}{\eta} z_{\text{cation}} c_{\text{cation}} F E_n
\]

\[
\Delta e^h = -\frac{\Delta f}{\Delta x_2} \left\{ \left( (f^{w2})_{n+1} + (f^{w3})_{n+1} + (f^{w4})_{n+1} \right) - \left( (f^{w2})_n + (f^{w3})_n + (f^{w4})_n \right) \right\}
\]

(39)

where, \( f^{w2} \) is volume flux with hydration-induced pressure, \( f^{w3} \) is volume flux with hydration potential and \( f^{w4} \) is volume flux with electric potential.

Fifth, total pressure, porosity and water uptake are also updated from Eq. (3) and Eq. (21).

\[
\Delta p^h = \frac{B^2 E (1 + v^b)}{9(v^b - v)} \Delta e^h
\]

(40)

\[
p^n' = p^n_0 + \Delta p^n_0 + \Delta p^h
\]

(41)

\[
\phi^n = \phi^n_0 + \Delta \phi^n_0 = \phi^n_0 + \Delta e^n_0 + \Delta e^h
\]

(42)

\[
w^n = w^n_0 + \Delta w^n_0 = w^n_0 + (1 + w^n_0) \Delta \phi^n_0
\]

(43)

Using the stress-induced pressure in Eq. (38) and the hydration-induced strain in Eq. (39), the structural analysis of mechanical IPMC sensors is conducted with Biot poroelastic constitutive equation. The stress-induced pressure, beam Poisson effect and hydration-induced strain are implemented as external forces into finite element formulation. The finite element stiffness equation with initial strain method is as follows.

\[
[S] \{\Delta u\} = \{\Delta f^{\text{ext}}\} + \{\Delta f^{\text{x}}\} + \{\Delta f^h\} + \{\Delta f^{\text{beam}}\}
\]

(44)

\[
\{\Delta f^{\text{x}}\} = \int_V [L]^T \{b \Delta p' \delta_j\} dV
\]

(45)

\[
\{\Delta f^h\} = \int_V [L]^T [D] \left( \frac{\Delta e^h}{3} \delta_j \right) dV
\]

(46)

\[
\{\Delta f^{\text{beam}}\} = \int_V [L]^T \{2vn^b \Delta p' \delta_j\} dV
\]

(47)

where, \([S]\) is stiffness matrix, \(\{\Delta f^{\text{x}}\}\) is the force increment vector of stress-induced pressure, \(\{\Delta f^h\}\) is the force increment vector of hydration effect, \([D]\) is stress-strain relation matrix with water uptake. \(\{\Delta f^{\text{beam}}\}\) is the force increment vector of the Poisson effect only for beam models. Since the pore pressure acts on the lateral direction of uniaxial stress condition, the Poisson effect of the pore pressure is added. In order to match with the peak of reaction forces in experiments, the undrained Poisson's ratio is modified by the correction factor from the Eq. (20). Solving the Eq. (44), the mechanical states with the relaxation are numerically analyzed.

Lastly, the fields of mobile ion concentration and electric potential are obtained with Poisson-Nernst-Plank equations, Eq. (31) and Eq. (32). The field equations of mobile ion concentration and electric potential are obtained as follows.

\[
(f^{c1})_{n+1} = -D_{he} \frac{\phi^n_0 + \phi_{n+1}}{2} \frac{c_{n+1} - c_n}{\Delta x_2}
\]

\[
(f^{c1})_n = -D_{he} \frac{\phi^n_0 + \phi_{n-1}}{2} \frac{c_n - c_{n-1}}{\Delta x_2}
\]

\[
(f^{c2})_{n+1} = D_{he} \frac{zF}{RT} \frac{C_{n+1} + C_n}{2} E_{n+1}
\]

\[
(f^{c2})_n = D_{he} \frac{zF}{RT} \frac{C_n + C_{n-1}}{2} E_n
\]
\[
(f^{c^3})_{n+1} = -D_p \frac{C_{n+1}W_{n+1} - C_n W_n}{\Delta x_2} \quad (f^{c^3})_n = -D_p \frac{C_n W_n - C_{n-1}W_{n-1}}{\Delta x_2}
\]
\[
(f^w)_{n+1} = (f^{w^1})_{n+1} + (f^{w^2})_{n+1} + (f^{w^3})_{n+1} + (f^{w^4})_{n+1} \quad (f^w)_n = (f^{w^1})_n + (f^{w^2})_n + (f^{w^3})_n + (f^{w^4})_n
\]
\[
\Delta C = -\frac{\Delta t}{\Delta x_2} \left( (f^{c^3})_{n+1} - (f^{c^3})_n + (f^w)_{n+1} - (f^w)_n + \frac{c_{n+1} + c_n}{2} (f^w)_{n+1} - \frac{c_n + c_{n-1}}{2} (f^w)_n \right)
\]
\[
n = 2, 3, 4, \cdots, (N+1) \quad E_1 = 0 \quad E_n = E_{n-1} - \frac{F}{k_e} (C_n - C^n) \Delta x_2
\]
\[
n = 3, 5, 7, \cdots, (N+1) \quad \psi_1 = 0, \quad \psi_n = \psi_{n-2} - \frac{E_n + 4E_{n-1} + E_{n-2}}{3} \Delta x_2
\]

From the above equations, the electric potential is obtained as the output of the sensor model. Through the aforementioned procedure, the transient behaviors of mechanical sensors using IPMC are numerically analyzed with the fields of the stresses and strains of IPMCs, pore pressure, ion concentration and electric potential of each layer in Fig. 4.

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5. Simulation results

The numerical formulation in the previous chapter is adopted into the in-house code of finite element program, and the transient response of a mechanical sensor using Flemion in the experiment of Wang et al. (2009) is numerically simulated. The experiment measured electric potentials from the different deflections with time. The geometry of the mechanical sensor is illustrated in Fig. 1. Regarding the mechanical sensor, the physical parameters for the present simulation are determined as shown in Table 1. The material parameters can be determined by macroscale experiments. Flemion on dry condition is dense, so \( K_{\text{dry}} \) is approximated as \( K_{\text{solid}} \).

As shown in Fig. 4, the region of ionic conducting polymer is only discretized, and the outer surfaces of the polymer are assumed that water cannot be escaped. As the boundary condition of the structural analysis, the root of the mechanical sensors is fixed and the free tip is prescribed with different deflections as shown in Fig. 5. In details, 4, 5, 6 and 10mm deflections are prescribed for 1 sec, the deflections are kept for 9 sec, and return to initial positions for a second. As a results, reaction force, pore pressure, water uptake, ion concentration and electrical potential are obtained with respect to time and thickness direction as shown in Figs. 5-8.
In Fig 5, reaction forces are shown with different deflections. Responding to loads by the prescribed deflections, the reaction forces are instantaneously increased with small amounts of peaks. And then, the peaks are quickly relaxed and the reaction force becomes steady on constant deflections. The magnitudes of the peaks and steady reaction forces are almost proportional to the magnitudes of prescribed deflections. At 10 sec, the deflections are prescribed to initial position. The unloading behaviors apparently show the same peaks and relaxation with the loading behaviors, and return to zero reaction force.

From Fig 6, the numerical results are shown only for 4mm deflection case, because the time histories of interesting parameters are shown and the other 5, 6 and 10 mm deflection cases have the same characteristics in interesting parameters. The pore pressure is obtained as shown in Fig. 6. In the left graph, the stress-induced pressure is spontaneously increased with the deflection and reaction force for a second in Fig. 5, and subsequently the stress-induced pressure is relaxed to zero. As mentioned in previous chapters, the stress-induced pressure is induced on the undrained condition resulting in the transient reaction force. In the right side of Fig 6, the change of total pore pressure from initial pore pressure is shown with respect to time and thickness. At 1sec, the pore pressure peaks and then relaxed due to the contribution of the stress-induced pressure, but the pore pressure does not return to initial pore pressure even though deflection is sustained. The remained pore pressure means the change of hydration-induced pressure. After unloading, the hydration-induced pressure returns to initial pore pressure.

Water uptake is also shown in Fig. 7. The distribution of water uptake in thickness direction with time shows water migration in pores. During loading, the water is rapidly migrated due to the stress-induced pressure, and then slowly flows even when deflection is fixed. The moment of the pore pressure peak is delayed than the peak of pore pressure. That is, water tries to move due to the gradient of pore pressure. However, the driving force due to hydraulic pore pressure balances with the driving force due to hydration and electric potential resulting in the constant gradient of water uptake and pore pressure from 2sec to 10sec in Fig 7. On unloading, the water in pores rapidly is migrated and the volume flux is gradually decreased until uniform distribution. The transport of water solution in ionic conducting polymers also has an effect to the distribution of ion concentration that water flow delivers mobile ions.

<table>
<thead>
<tr>
<th>Table 1</th>
<th>Physical parameters for the present simulation</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Input parameters</strong></td>
<td></td>
</tr>
<tr>
<td>Initial water uptake</td>
<td>$w_0 = 0.3470$</td>
</tr>
<tr>
<td>Initial porosity</td>
<td>$\phi_0 = w_0 (1 + w_0) = 0.2576$</td>
</tr>
<tr>
<td>Elastic modulus on dry condition</td>
<td>$E_{dry} = 2461.2 , MPa$</td>
</tr>
<tr>
<td>Empirical coefficient of Elastic modulus</td>
<td>$Q = 7.2409$</td>
</tr>
<tr>
<td>Elastic modulus</td>
<td>$E = E_{dry} \exp(-Qw_0) = 199.5MPa$</td>
</tr>
<tr>
<td>Bulk modulus on dry condition</td>
<td>$K_{dry} = 10255 , MPa$</td>
</tr>
<tr>
<td>Empirical coefficient of Bulk modulus</td>
<td>$R = Q$</td>
</tr>
<tr>
<td>Bulk modulus</td>
<td>$K = K_{dry} \exp(-Rw_0) = 665MPa$</td>
</tr>
<tr>
<td>Bulk modulus of polymer matrix</td>
<td>$K^{poly} = K_{dry}$</td>
</tr>
<tr>
<td>Hydration coefficient</td>
<td>$A = Q$</td>
</tr>
<tr>
<td>Initial concentration of cation to water</td>
<td>$c_{i0} = 0.01M$</td>
</tr>
<tr>
<td>Initial total concentration of cation</td>
<td>$C_{i0} = \phi_0 c_{i0} = 0.002576M$</td>
</tr>
<tr>
<td>Valence of cation</td>
<td>$z_i = +1$</td>
</tr>
<tr>
<td>Hydraulic permeability</td>
<td>$k_s = 1.0 \times 10^{-14} , mm^2$</td>
</tr>
<tr>
<td>Dynamic viscosity</td>
<td>$\eta = 0.862 \times 10^{-7} , MPa \cdot s$</td>
</tr>
<tr>
<td>Ion-water diffusivity</td>
<td>$D_{iw} = 1.186 \times 10^{-15} , mm^2$</td>
</tr>
<tr>
<td>Ion-polymer diffusivity</td>
<td>$D_{ip} = 1.186 \times 10^{-15} , mm^2$</td>
</tr>
<tr>
<td>Electric permittivity</td>
<td>$k_e = 7.5 \times 10^{-6} , F/, mm$</td>
</tr>
<tr>
<td>Absolute temperature</td>
<td>$T = 293K$</td>
</tr>
<tr>
<td>Correction factor of shear stiffness</td>
<td>$\alpha = 5/6$</td>
</tr>
<tr>
<td>Correction factor of Poisson’s ratio</td>
<td>$\beta = 1.0$</td>
</tr>
<tr>
<td>Elastic modulus of gold</td>
<td>$E = 1946MPa$</td>
</tr>
<tr>
<td>Poisson’s ratio of gold</td>
<td>$\nu = 0.42$</td>
</tr>
</tbody>
</table>
The distribution of total concentration of cation is illustrated in Fig. 7. Following the water transport, the cations are redistributed, and spontaneously result in the redistribution of charge density. From well known Poisson equation in electrostatics, the redistribution of charge density determines electric potential. The distribution and time history of electric potentials are illustrated with thickness and time in Fig. 8. With the molar flux of cation, the electric potential is spontaneously changed, but the redistribution of cation is slower than the response of pore pressure. Related to the transports of water and ions, the electric potential shows relatively-smooth peaks and gradual relaxations with time. As the output of mechanical sensors using IPMCs, the time histories of electric potentials are obtained with different deflections. As shown in the right hand side in Fig. 8, the calculated electric potentials from the present simulation are well agreed with the experimental electric potential of Wang et al. (2009).

Therefore, the non-invertible black box between actuating and mechanical sensing modes is explained with interesting parameters from the numerical results, and the very small electric potential in mechanical sensing response is numerically expressed. Furthermore, the transient behavior, e.g. relaxation and time lag, are modeled and the time dependency of the mechanical sensing behaviors of IPMCs is shown. The hydration effect on the mechanical sensors is also estimated in the present study.

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![Prescribed deflection at beam tip](image1.png)

![Reaction force at fixed root](image2.png)

**Fig. 5** Prescribed tip deflection (L) and reaction force (R) with time

![Stress-induced pressure (L) and total pore pressure (R) over thickness with time in case of 4mm deflection](image3.png)

**Fig. 6** Stress-induced pressure (L) and total pore pressure (R) over thickness with time in case of 4mm deflection
Fig. 7 Water uptake (L) and ion concentration (R) over thickness with time in case of 4mm deflection

Fig. 8 Electrical potential over thickness (L) with time in case of 4mm deflection, and time history of voltage (R) with different deflections

Fig. 9 Relation between deflections in Fig.5 and electric potentials in Fig.8
The relations between deflections in Fig. 5 and electric potentials in Fig. 8 are shown in Fig. 9. The present study suggests a more complicated simulation rather than an empirical relation between deflection-input and voltage-output. The voltage-output of the mechanical sensors is continuously decreased as shown in Fig. 8. The peak of electric potential can employ as the output of the mechanical sensors, and the input-output relation can be employed for monotonic stimulation. However, the peak is induced from the transient response due to the migrations of water and ions. We conclude that black box models using only deflection and electric voltage are not reasonable unless the operation conditions are controlled like experiments. In order to obtained input-output relations for various conditions, many parameters and experiments are needed, since the mechanical sensors shows different behaviors depending on water uptake, velocities, concentration and so on. Therefore, the present study proposes the simulation of essential phenomena in the mechanism of the mechanical sensors.

6. Conclusion

The present study has attempted the numerical modeling of mechanical sensors using IPMCs and introduced its simulation procedure and results. The model of mechanical sensors using conducting polymers in the previous work (Yoo and Toi, 2013) was modified, and the hydration effects of IPMCs were newly considered with the followings.

First, the hydration effects on volume and stiffness has been considered. The present study decomposed the total strains of IPMCs as stress-induced strain and hydration-induced strain. The hydration-induced strain covered the electrochemically induced strains including actuating deformation, and the hydration-induced strain was handled as eigen-strain. Furthermore, pore pressure was decomposed into stress-induced pressure and hydration-induced pressure, and the stress-induced strain and stress-induced pressure were embedded into Biot constitutive equation. The volume swelling due to hydration was modeled with water migration due to electrochemically induced driving forces. The driving force was expressed with hydration potential employing an empirical chemical potential at free swelling equilibrium. The hydration potential was embedded into Darcy flow model with the modeling of hydration coefficient. As a result, the water migration due to hydration and electric potential was estimated and the hydration-induced strain was estimated with Biot poroelastic theory. The hydration-induced strain was added into the structural analysis of the mechanical sensing responses.

Second, the transient response with relaxation and time lag of reaction force and electric potential has been numerically simulated. The simulation of mechanical sensors using IPMCs was employed Timoshenko beam model, Biot poroelastic model and Poisson-Nernst-Plank model. The instantaneous pore pressure in the transient behaviors was estimated on the undrained condition, water migration due to the pore pressure was expressed and the ion transport following the water migration was modeled to induce electricity. In Timoshenko beam model, Poisson effect due to pore pressure was considered for the transient response of a porous beam. In Biot poroelastic model, hydration-induced strain and pressure were embedded, and undrained beam Poisson’s ratio was employed. In Poisson-Nernst-Plank equations, ion transport with water flow and interaction between ions and polymer were considered, and charge density to the total volume of bulk material was obtained. Next, the related models were numerically formulated, and they were employed into one-dimensional finite element program because the variations of physical quantities over microscale thickness are dominant.

Third, interesting parameters, which are significantly important in the mechanism of the mechanical sensors, were obtained with respect to time and thickness of IPMCs, and the non-invertible relation between deformation and electric potential in actuating and mechanical sensing modes were investigated. That is, the qualitative explanation for the very small output of electric potential was quantitatively estimated. From the simulation results, the distributions of solid stress, fluid pressure, water uptake, ion concentration and electric potential would be helpful for understanding the mechanism of IPMCs. The water transport in IPMCs is a key factor for design and control of mechanical IPMC sensors.

References


