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ANALYTICAL SOLUTIONS FOR A ONE-DIMENSIONAL CHEMO-MECHANICAL COUPLING PROBLEM**

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ABSTRACT Chemo-mechanical coupling exists in a lot of intelligent materials including hydrogels, biological tissues and other soft materials. These materials are able to respond to external stimulus, such as temperature, chemical concentration, and pH value. In this paper, a one-dimensional theoretical model for chemo-mechanical coupling is proposed for analyzing the uniaxial stress/strain state of coupling materials. Based on the chemo-mechanical coupled governing equation, the displacement function and concentration function are derived and the stress and chemical potential are obtained. It is shown that the present chemo-mechanical theory can characterize the chemo-mechanical coupling behavior of intelligent materials.

KEY WORDS intelligent materials, chemo-mechanical coupling, analytical solution, theoretical model, constitutive equations, hydrogel

I. INTRODUCTION

In modern industry and engineering practice, many materials employed have multi-field coupling behavior and are often known as smart material. Commonly used smart materials include shape memory alloys (SMA), mass-energy optical fibers, piezoelectric materials, pH-response hydrogels and their composites. Applications of these smart materials are widely found in the automotive industry, telephony, architecture, food and so on. Intelligent hydrogel is such a class of material that its properties can vary significantly with the changes of external environmental conditions, including physical and chemical stimuli. Physical stimuli are defined as physical environmental factors, including light, temperature, sound, electric and magnetic field, whereas chemical stimuli include pH value of the solution and ionic concentration.

It is noted that smart gels are generally swelling or shrinking in response to external chemical stimuli[1]. Such chemo-mechanical coupling phenomena also exist in other types of materials including clay[2–4], cement paste[5,6], geomaterials[7] and biological soft tissues[8–10] and hard tissues[11–13]. Therefore, it is important from the viewpoint of engineering application to evaluate the chemo-mechanical coupling behavior of these multifield materials. During the past few decades, researchers have developed some experimental methods, numerical simulations and theoretical models for analyzing chemo-mechanical coupling of these widely used materials. For example, Ballhause et al.[14] investigated the mechanical

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mechanism of the chemical stimulation, and he indicated that the osmotic pressure of polymer and solution would undergo a change in response to the variation of the external chemical condition. The concentration, the chemical potential, and the displacement can be analyzed by the coupled chemo-electro-mechanical model. Vallenton et al.[15] involved the material expansion caused by enzyme reaction in consideration of the coupled effects of fluid flow, diffusion and chemical reaction. Doi et al.[16] studied the colloid dynamic behavior under a static electric field. Thomas et al.[17] observed that the mechanical properties of the gel were affected by electric charge in the electrophoresis solution. The anion and cationic concentration and potential between the interior and exterior of the colloid can be calculated according to the ionic concentration in the surrounding solvent, when chemical and electric simulation sparks occurred at the same time. It is shown that, with the large expansion of colloid fibers, the ion concentration variation along the positive direction of the anion colloid leads to relatively large results. Recently, Lu et al.[18] designed a uniaxial tensile experiment to explore the critical strain of polymer-supported metal. Yashin et al.[19] found that self-consistent vibration of the hydrogel depends on its model size and external force. Loeffel et al.[20] dealt with the polymer surface properties of the TBC material, including the diffusion of oxygen of surface materials and redox characteristics.

For studies on numerical simulation in chemo-mechanical coupling problems, Li et al.[21] developed a Hermite-Cloud method to model chemo-mechanical deformation and ionic concentration of hygrogels subjected to chemical and electrical stimulation, and numerical results are in good agreement with experimental data. Kaasschieter et al.[22] applied the multiphasic theory and associated four-phase mixed finite element to model the mechanical behavior of cartilages. Hong et al.[23] presented a finite element method based on the Gibbs free energy and non-linear thermodynamics, and illustrated several examples including swelling-induced deformation, contact and bifurcation. Macrombe et al.[24] investigated the inhomogeneous swelling of pH-sensitive gels by implementing a finite element method. Furthermore, the theory has been embedded in the commercial software ABAQUS by writing a user-supplied subroutine. Yang et al.[25,26] formulated coupled constitutive equations for analyzing general thermo-electro-chemo-mechanical coupling behavior of hydrogels by introducing the thermo-electro-chemo-mechanical effects into the Gibbs free energy. Based on the Gibbs free energy functional, a coupled finite element procedure is developed to model the swelling, shrinking and redistribution of ions with the smart hydrogels in some chemo-mechanical environments.

For analytical solutions to chemo-mechanical problems, De et al.[27] derived steady- and transient-state chemo-mechanical coupled equations where the ion transportation is described by the Nernst-Planck flux equations, and the osmotic pressure of mechanical field was introduced by the extended Darcy's law. Li et al.[28,29] and Lai et al.[30] presented a MEC model for the glucose-stimulus hydrogel and pH-sensitive hydrogel considering the effects of enzyme catalysis. Giovanni et al.[31] derived a one-dimensional distributed model based on the Euler-Bernoulli beam theory and a parallel-plate approximation, and validated his theoretical findings through a series of experiments. Unlike the theoretical solutions mentioned above, this paper is focused on the theoretical stress/strain analysis of intelligent materials with the objective of deriving a series of closed-form solutions. The solution can then be used to obtain results of displacement, stress, concentration and chemical potential of smart gels. In particular, we proposed a uniaxial stress/strain theoretical model by considering mechanical equilibrium and mass conservation. Two numerical examples are considered to assess the effectiveness of the model and applicability of the corresponding solution.

II. GOVERNING EQUATIONS OF CHEMO-MECHANICAL COUPLING

Consider a chemo-mechanical body of volume Ω bounded by surface S. The governing equations are the equilibrium equations of stresses and diffusion equations of ions. The equilibrium equations are

$$\sigma_{ij,j} + f_i = \rho\ddot{u}_i \qquad (\text{in} \quad \Omega) \tag{1}$$

and boundary conditions

$$u_i = \bar{u}_i \quad (\text{on} \quad S_u), \qquad \sigma_{ij}n_j = \bar{t}_i \quad (\text{on} \quad S_t) \tag{2}$$

where σ_{ij} is stress tensor, f_i is the body force, u_i is displacement, $\bar{u}_i$ and $\bar{t}_i$ are the prescribed surface displacements and tractions on the surface S, n_j is the unit outward normal vector to the surface S, $S = S_u + S_t$.

The chemical field is described by the diffusion equation of ions

$$\dot{c} + \xi_{i,i} = 0 \qquad (\text{in} \quad \Omega) \tag{3}$$

and corresponding boundary conditions

$$c = \bar{c} \quad (\text{on} \quad S_c), \qquad \xi_i n_i = \bar{\xi}_n \quad (\text{on} \quad S_\xi) \tag{4}$$

where c are increments of concentrations. ξ_i is ionic flux, μ is chemical potential, and $\bar{\xi}_n$ is the specified ionic flux on the surface of the domain.

To consider the chemo-mechanical coupling, a modified form[26] of Eq.(3) is used:

$$\dot{\mu} + \tau \xi_{i,i} = 0 \tag{5}$$

where $\tau = R^* T / c_0$, R^* is the universal constant of ideal gas and T is the absolute temperature, c_0 is the reference concentration, and μ is the standard chemical potential.

The constitutive equations of chemo-mechanical coupling can be expressed by[26]

$$\sigma_{ij} = D_{ijkl} \varepsilon_{kl} + R_{ij} c, \qquad \mu = R_{kl} \varepsilon_{kl} + sc \tag{6}$$

where D_{ijkl} are elastic coefficients, R_{ij} are the mechanical-chemical coefficients, s is the chemical potential constant. It is noted that the coupled stress in the constitutive equation (6) contains two parts, one is elastic stress, the other is induced by the chemical effect due to ionic unbalance.

The gradient equations describing the relations between displacements u_i and strains ε_{ij} for elastic field, ionic flux ξ_i and concentration change c for chemical field, are given as follows:

$$\varepsilon_{ij} = \frac{1}{2}\left(u_{i,j} + u_{j,i}\right), \qquad \xi_i = -\Phi_{ij} c_{,j} \tag{7}$$

where Φ_{ij} denotes the diffusion coefficient, depending on the intrinsic features of the medium. It is noted that the theory described above is restricted to small deformation and small change in ion concentration.

III. SOLUTION FOR UNIAXIAL STRESS STATE

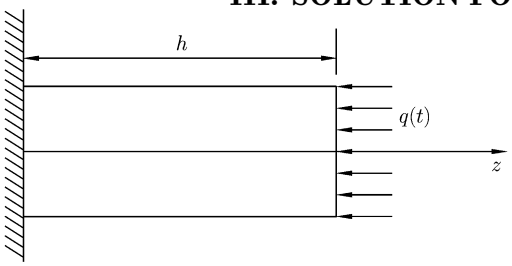

Fig. 1 One-dimensional model.

Let us consider a one-dimensional bar or a fiber subjected to coupled chemo-mechanical loads as shown in Fig.1. This model can be used to consider the one-dimensional coupling problem of chemistry and mechanics. The basic equations for chemo-mechanical coupling are

$$E\frac{\partial^2 w}{\partial z^2} + R\frac{\partial c}{\partial z} = \rho\frac{\partial^2 w}{\partial t^2} \tag{8a}$$

$$(1-2\nu) R\frac{\partial^2 w}{\partial z \partial t} + s\frac{\partial c}{\partial t} - \Phi\frac{R^* T}{c_0}\frac{\partial^2 c}{\partial z^2} = 0 \tag{8b}$$

where w is axial displacement, ν is the Poisson's ratio.

Consider a one-dimensional problem under time-dependent mechanical load $q = q_0 \mathrm{e}^{\alpha t}$. The displacement w and concentration c have the following forms:

$$w(z,t) = \bar{w}(z)\,\mathrm{e}^{\alpha t}, \qquad c(z,t) = \bar{c}(z)\,\mathrm{e}^{\alpha t} \tag{9}$$

For this case, substitution of Eq.(9) into Eqs.(8a) and (8b) yields

$$\begin{aligned} &\left(E\frac{\partial^2}{\partial z^2} - \rho\alpha^2\right)\bar{w}(z) + R\frac{\partial \bar{c}(z)}{\partial z} = 0 \\ &(1-2\nu) R\frac{\partial \bar{w}(z)}{\partial z} + \left(s - \frac{R^* T \Phi}{c_0 \alpha}\frac{\partial^2}{\partial z^2}\right)\bar{c}(z) = 0 \end{aligned} \tag{10}$$

Application of Cramer Rule to Eq.(10) leads to an equation for $\bar{w}(z)$ as

$$\left\{-\frac{E R^* T \Phi}{c_0 \alpha}\frac{\partial^4}{\partial z^4} + \left[Es + \frac{R^* T \Phi \rho \alpha}{c_0} - (1-2\nu) R^2\right]\frac{\partial^2}{\partial z^2} - \rho s \alpha^2\right\}\bar{w}(z) = 0 \tag{11}$$

Its characteristic equation is

$$-\frac{ER^*T\Phi}{c_0\alpha}r^4+\left[Es+\frac{R^*T\Phi\rho\alpha}{c_0}-(1-2\nu)R^2\right]r^2-\rho s\alpha^2=0 \tag{12}$$

Solution of Eq.(12) gives

$$r_{1,2}=\pm\eta=\pm\sqrt{\frac{\sqrt{k_2^2-4k_1k_3}-k_2}{2k_1}},\qquad r_{3,4}=\pm\mathrm{i}\beta=\pm\mathrm{i}\sqrt{\frac{\sqrt{k_2^2-4k_1k_3}+k_2}{2k_1}}$$

where $k_1=-ER^*T\Phi/(c_0\alpha)$, $k_2=Es+RT\Phi\rho\alpha/c_0-(1-2\nu)R^2$, $k_3=-\rho s\alpha^2$. Then the general form of displacement solution $w(z,t)$ can be given by

$$w(z,t)=\left[C_1\mathrm{e}^{\eta z}+C_2\mathrm{e}^{-\eta z}+C_3\cos(\beta z)+C_4\sin(\beta z)\right]\cdot\mathrm{e}^{\alpha t} \tag{13}$$

By substituting Eq.(13) into Eq.(10), we find the solution of concentration $c(z,t)$ as

$$c(z)=\left\{\left(\frac{\rho\alpha^2}{R\eta}-\frac{E\eta}{R}\right)\left(C_1\mathrm{e}^{\eta z}-C_2\mathrm{e}^{-\eta z}\right)+\left(\frac{\rho\alpha^2}{R\beta}+\frac{E\beta}{R}\right)\left[C_3\sin(\beta z)-C_4\cos(\beta z)\right]\right\}\cdot\mathrm{e}^{\alpha t} \tag{14}$$

Finally, by substituting Eqs.(13) and (14) into Eq.(6), the solutions of stress and chemical potential are obtained as

$$\sigma=\left\{\frac{\rho\alpha^2}{\eta}\left(C_1\mathrm{e}^{\eta z}-C_2\mathrm{e}^{-\eta z}\right)+\frac{\rho\alpha^2}{\beta}\left[C_3\cos(\beta z)-C_4\sin(\beta z)\right]\right\}\cdot\mathrm{e}^{\alpha t} \tag{15a}$$

$$\begin{aligned}\mu=&\left\{\frac{\rho\alpha^2s-Es\eta^2+R^2\eta^2}{R\eta}\left(C_1\mathrm{e}^{\eta z}-C_2\mathrm{e}^{-\eta z}\right)\right.\\&\left.+\frac{\rho\alpha^2s+Es\beta^2-R^2\beta^2}{R\beta}\left[C_3\sin(\beta z)-C_4\cos(\beta z)\right]\right\}\cdot\mathrm{e}^{\alpha t}\end{aligned} \tag{15b}$$

where the parameters C_1, C_2, C_3 and C_4 are integration constants.

The boundary conditions of the present problem are

$$\bar{w}\big|_{z=0}=0,\quad \bar{\mu}\big|_{z=0}=0,\quad \bar{\sigma}_z\big|_{z=h}=-q,\quad \bar{\xi}_n\big|_{z=0}=0 \tag{16}$$

Applying the boundary conditions to Eqs.(15a) and (15b), the constants can be determined as

$$\begin{aligned}&C_1=-C_2=-q_0\frac{\eta}{\rho\alpha^2}\Big/\left[\left(\mathrm{e}^{\eta h}+\mathrm{e}^{-\eta h}\right)-2\frac{\rho\alpha^2s-Es\eta^2+R^2\eta^2}{\rho\alpha^2s+Es\beta^2-R^2\beta^2}\sin(\beta h)\right],\qquad C_3=0\\&C_4=-2q_0\cdot\frac{\beta}{\rho\alpha^2}\cdot\frac{\rho\alpha^2s-Es\eta^2+R^2\eta^2}{\rho\alpha^2s+Es\beta^2-R^2\beta^2}\Big/\left[\left(\mathrm{e}^{\eta h}+\mathrm{e}^{-\eta h}\right)-2\frac{\rho\alpha^2s-Es\eta^2+R^2\eta^2}{\rho\alpha^2s+Es\beta^2-R^2\beta^2}\sin(\beta h)\right]\end{aligned} \tag{17}$$

Thus the exact solutions, i.e. displacement, concentration, stress and chemical potential, of the present model have been obtained.

IV. SOLUTIONS FOR UNIAXIAL STRAIN PROBLEM

It is assumed that the semi-infinite material is subjected to uniform pressure on the surface. This is a three-dimensional solid with infinite boundary. Because of the symmetry of the problem, it can be dealt with a uniaxial strain model. For simplicity, a coordinate (o, z) is used in the present analysis, as shown in Fig.2. The number of independent coefficients in Eq.(6) depends on the material symmetry. The constitutive equations for z axis can be given as follows:

$$\sigma_z=D\varepsilon_z+Rc,\qquad \mu=R\varepsilon_z+sc \tag{18}$$

The field equations for uniaxial strain state can be rewritten as

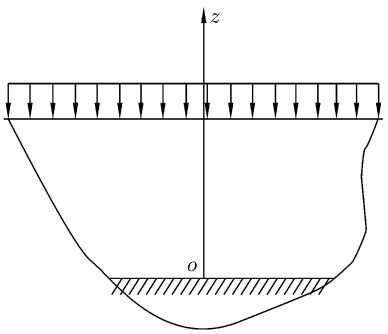

Fig. 2 Uniaxial strain state model.

$$\frac{\partial \sigma_z}{\partial z}=0, \qquad \frac{\partial \mu}{\partial z}-\frac{R^*T}{c_0}\frac{\partial c}{\partial z}=0 \tag{19}$$

and the gradient equations can be given by

$$\varepsilon_z=\frac{\partial w}{\partial z}, \qquad \xi=-\Phi\frac{\partial c}{\partial z} \tag{20}$$

where $D=E\left(1-\nu\right)/[\left(1+\nu\right)\left(1-2\nu\right)]$. Then the coupled equations can be given as

$$D\frac{\partial^2 w}{\partial z^2}+R\frac{\partial c}{\partial z}=\rho\frac{\partial^2 w}{\partial t^2}, \qquad R\frac{\partial^2 w}{\partial z\partial t}+s\frac{\partial c}{\partial t}-\Phi\frac{R^*T}{c_0}\frac{\partial^2 c}{\partial z^2}=0 \tag{21}$$

Consider a one-dimensional problem under time-dependent mechanical load $q=q_0\mathrm{e}^{\alpha t}$. It is assumed that displacement w and concentration c are in the following forms:

$$w\left(z,t\right)=\bar{w}\left(z\right)\mathrm{e}^{\alpha t}, \qquad c\left(z,t\right)=\bar{c}\left(z\right)\mathrm{e}^{\alpha t} \tag{22}$$

For this case, substituting Eq.(22) into Eq.(21) , we obtain

$$\left(D\frac{\partial^2}{\partial z^2}-\rho\alpha^2\right)\bar{w}\left(z\right)+R\frac{\partial}{\partial z}\bar{c}\left(z\right)=0, \qquad R\frac{\partial}{\partial z}\bar{w}\left(z\right)+\left(s-\frac{R^*T\Phi}{c_0\alpha}\frac{\partial^2}{\partial z^2}\right)\bar{c}\left(z\right)=0 \tag{23}$$

By introducing Cramer Rule into Eq.(23), the equation for $\bar{w}\left(z\right)$ is obtained

$$\left[-\frac{DR^*T\Phi}{c_0\alpha}\frac{\partial^4}{\partial z^4}+\left(Ds+\frac{\rho R^*T\Phi}{c_0\alpha}-R^2\right)\frac{\partial^2}{\partial z^2}-\rho\alpha^2 s\right]\bar{w}\left(z\right)=0 \tag{24}$$

Its characteristic equation is

$$-\frac{DR^*T\Phi}{c_0\alpha}r^4+\left(Ds+\frac{\rho R^*T\Phi}{c_0\alpha}-R^2\right)r^2-\rho\alpha^2 s=0 \tag{25}$$

Solutions of Eq.(25) give

$$r_{5,6}=\pm\eta'=\pm\sqrt{\frac{\sqrt{k_5^2-4k_4k_6}-k_5}{2k_4}}, \qquad r_{7,8}=\pm\mathrm{i}\beta'=\pm\mathrm{i}\sqrt{\frac{k_5+\sqrt{k_5^2-4k_4k_6}}{2k_4}}$$

where $k_4=-DR^*T\Phi/(c_0\alpha)$, $k_5=Ds+\rho R^*T\Phi/(c_0\alpha)-R^2$, $k_6=-\rho\alpha^2 s$. Then the general solution of displacement $w\left(z,t\right)$ can be given as

$$w\left(z\right)=\left[C_5\mathrm{e}^{\eta' z}+C_6\mathrm{e}^{-\eta' z}+C_7\cos(\beta' z)+C_8\sin(\beta' z)\right]\cdot\mathrm{e}^{\alpha t} \tag{26}$$

By substituting Eq.(26) into Eq.(23), we find the solution of concentration $c\left(z,t\right)$

$$c\left(z\right)=\left\{\left(-\frac{D\eta'}{R}-\frac{\rho\alpha^2}{R\eta'}\right)\left(C_5\mathrm{e}^{\eta' z}-C_6\mathrm{e}^{-\eta' z}\right)+\left(\frac{D\beta'}{R}+\frac{\rho\alpha^2}{R\beta'}\right)\left[C_7\sin(\beta' z)-C_8\cos(\beta' z)\right]\right\}\cdot\mathrm{e}^{\alpha t} \tag{27}$$

Finally, substituting Eq.(26) and Eq.(27) into Eq.(18), we can find the solution of stress and chemical potential

$$\begin{aligned}\sigma\left(z,t\right)&=\left\{\frac{\rho\alpha^2}{\eta'}\left(C_5\mathrm{e}^{\eta' z}-C_6\mathrm{e}^{-\eta' z}\right)+\frac{\rho\alpha^2}{\beta'}\left[C_7\sin(\beta' z)-C_8\cos(\beta' z)\right]\right\}\cdot\mathrm{e}^{\alpha t}\\ \mu\left(z,t\right)&=\left\{\frac{R^2\eta'^2+\rho\alpha^2 s-Ds\eta'^2}{R\eta'}\left(C_5\mathrm{e}^{\eta' z}-C_6\mathrm{e}^{-\eta' z}\right)\right.\\ &\quad\left.-\frac{Ds\beta'^2+R^2\beta'^2+\rho\alpha^2 s}{R\beta'}\left[C_7\sin(\beta' z)-C_8\cos(\beta' z)\right]\right\}\cdot\mathrm{e}^{\alpha t}\end{aligned} \tag{28}$$

where C_5, C_6, C_7 and C_8 are the integration constants, which can be determined by the boundary conditions.

Table 1. Material parameters used in the two numerical examples

Parameters	Value
Reference concentration c_0	15 mol/m^3
Universal ideal gas constant R^*	8.31 J/molK
Chemo-mechanical coupling coefficient R	1.75×10^5 Nm/mol
Young's modulus E	3.5×10^5 Pa
Poisson's ratio ν	0.45
Coefficients of the chemical potential s	1×10^6 Nm/mol
Diffusion coefficient Φ	4.5×10^{-10} m^2/s
Absolute temperature T	293 K

V. NUMERICAL EXAMPLES AND DISCUSSIONS

As application of the above solutions, we will give some numerical examples to demonstrate the coupled chemo-mechanical effect of polymer gel.

5.1. One-Dimensional Bar Subjected to An Axial Time-Dependent Load

A bar subjected to an axial time-dependent load is considered in this example, as shown in Fig.1. The length of the bar $h = 100$ mm and the constant load $q_0 = 400$ N/m^2 are used in the present calculation. The material parameters of the model are listed in Table 1. Numerical results of displacement, stress, concentration, and chemical potential are, respectively, illustrated in Figs.3∼6. It can be seen that the mechanical load can lead to a rapid change of the response of the intelligent hydrogel. Figure 3 gives the variation of the axial displacement with length of the bar at different times. It is shown that the compression deformation of the bar occurs under load $q = q_0 e^{\alpha t}$ and boundary condition (16), and therefore a compression stress is generated in the bar, as shown in Fig.4. It is more important that the deformation induces the variation of ionic concentration and chemical potential in the bar due to the chemo-mechanical coupling, as shown in Figs.5 and 6. As the external force is time-dependent, the response of the concentration and chemical potential varies with time. It is obvious that for a constant external force, the concentration and chemical potential will reach a balanced state.

5.2. Semi-Infinite Body Subjected to Time-Dependent Traction Load

A semi-infinite hydrogel subjected to time–dependent load in exponential form on the surface is considered in this example. The material parameters in this example are still given in Table 1. The general solutions of this problem are given by Eqs.(26)-(28). Figures 7 and 8 show that the displacement and stress in terms of the position z have the nonlinear parabolic-type evolution. It is noted that both displacement and stress reach their peak value at the surface boundary. As shown in Fig.9, with the passing of time, the changes of concentration are greater and greater. Figure 10 illustrates that the chemical potential presents a continuous sinusoidal variation at different times. These results are of value in studying the interaction between the displacements at any position and load applied to the boundary of a hydrogel.

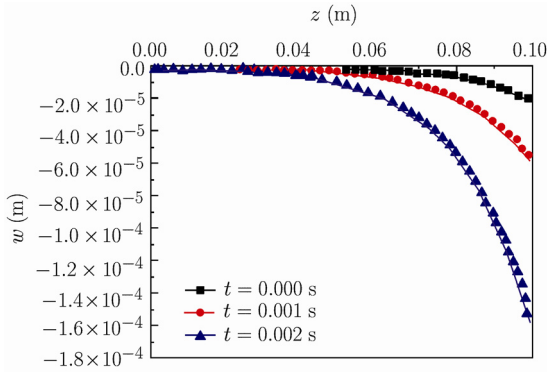

Fig. 3. Variations of the displacement versus length.

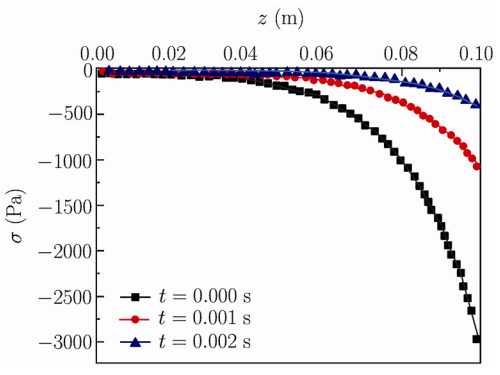

Fig. 4. Variations of the stress versus length.

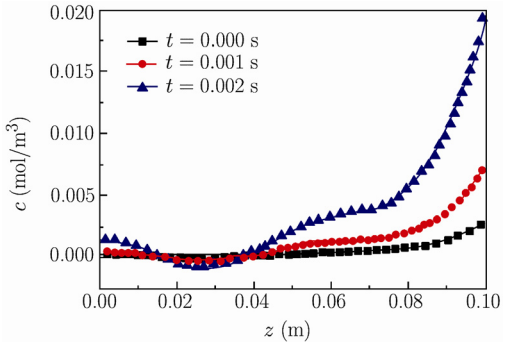

Fig. 5. Variations of the concentration versus length.

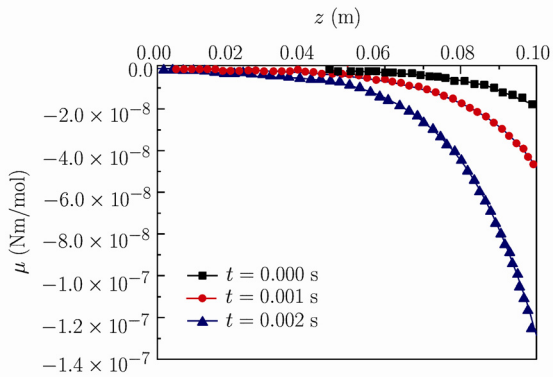

Fig. 6. Variations of the chemical potential versus length.

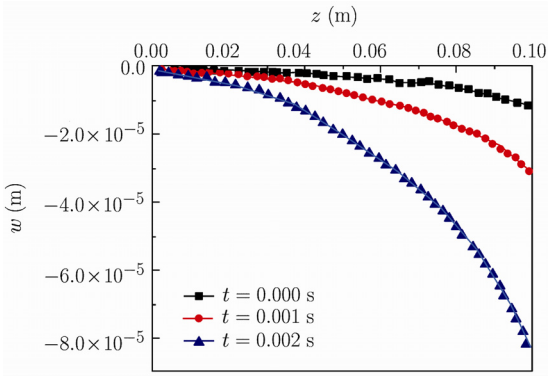

Fig. 7. Variation of displacement versus depth.

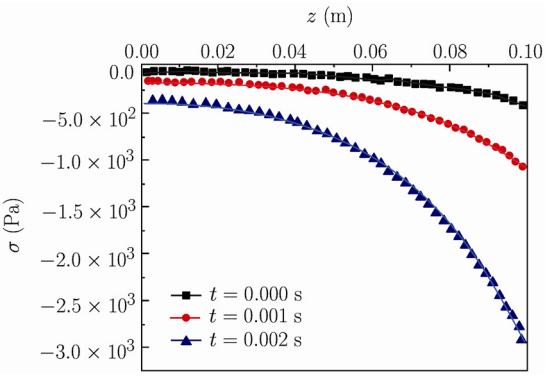

Fig. 8. Variation of stress versus depth.

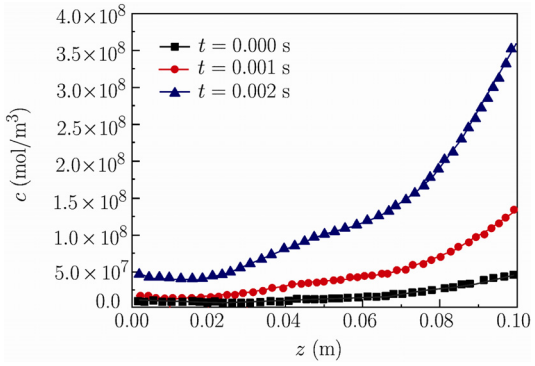

Fig. 9. Variation of concentration versus depth.

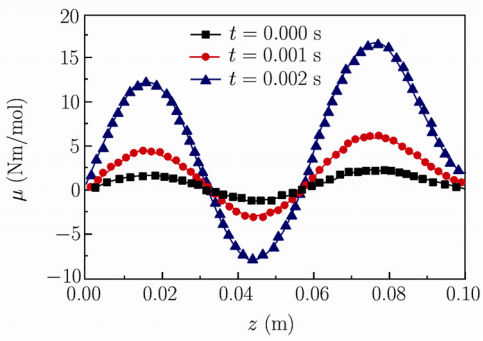

Fig. 10. Variation of chemical potential versus depth.

VI. CONCLUSIONS

This paper presents a theoretical model for analyzing chemical and mechanical interaction in intelligent materials. The analytical solutions of a one-dimensional chemo-mechanical coupling problem of intelligent hydrogel under the constant chemical stimulus and time-dependent mechanical load were deducted. These solutions can be used to analytically describe the chemo-mechanical coupling characterization of the materials. As typical applications, the numerical examples were used to illustrate the chemo-mechanical couplings of an intelligent hydrogel under steady and time-dependent loads.

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