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# A theoretical model and finite element formulation for coupled thermo-electro-chemo-mechanical media

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## 1. Introduction

# With the rapid development of material sciences and technologies, many new multifunction materials have been created and applied to industrial engineering, including materials that exhibit coupled multi-field behavior and interaction among fields. For example, conducting polymers have been widely used as artificial muscles and biosensors (Baughman, 1996; Garard et al., 2002; MacDiarmid, 2002; Yoseph, 2000), as the conducting polymers can accomplish the transformation of electrical, chemical and mechanical energy and demonstrate response to external environmental variables including temperature, pH and electrical and mechanical loadings. Such versatile polymers, which seem to be increasingly manufactured, include hydrogel and various advanced polymers (Otero et al., 1995; Doi et al., 1992; Santa et al., 1997). In general, these multi-field materials consist of a solid network and

#### ABSTRACT

Many synthetic and natural media which are often described as multifunctional smart materials demonstrate thermo-electro-chemo-mechanical coupling behavior and are sensitive to external environmental stimuli. This paper presents a set of basic equations, a variational principle and a finite element procedure for investigating the coupled behavior of thermo-electro-chemo-elastic media. Emphasis here is placed on introducing chemical effects into the coupled equation system. Using the governing equations of thermal conduction, electric flow, ionic diffusion and momentum balance, a variational principle is deduced for a linearly coupled system by means of the extended Gibb's free energy function. The variational principle is then used to derive a fully coupled multi-field finite element formulation for simulating the coupled thermo-electro-chemo-elastic behavior of biological tissues. Numerical examples are considered to illustrate the coupled phenomena of the materials and to verify the proposed variational theory and numerical procedure.

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interval fluid, and deform in volume and shape. They can be applied to practical engineering as biosensors, artificial skins of robots, artificial muscles, and actuators of adaptive structures (Garard et al., 2002; Yoseph, 2000).

By comparison, natural materials such as biological tissues, clays and shales exhibit strong swelling and contractive behavior under chemical, electrical and mechanical stimuli. For example, articular cartilage is a porous medium bathed in an electrolyte and its electro-chemomechanical coupling behavior cannot be ignored. This cartilage consists of hydrated proteoglycans and collagen fibers which form fibrillar structures that trap their own water. To sustain external loads, including mechanical and electrochemical loads, cartilage modifies its internal configuration by means of water and ion exchanges (Simom et al., 1998; Garikipatia et al., 2004; Loret and Simões 2005). The performance of saturated porous media has also attracted the attention of many researchers and scientists over the past decades (de Boer, 2000; 2003; Ehler, 2002). Early studies in this direction focused on the interactions between the solid and fluid of the saturated porous media. A so-called poroelastic theory was

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developed (Biot, 1955; Bowen, 1982) and used for deriving various numerical algorithms. The concept of equivalent stress and the equations of mass balance and momentum balance form the primary framework of the poroelastic theory.

Using poroelastic theory (Bowen, 1982), a triphasic mixture model of porous media was proposed to consider the electric and diffusion effect induced by ions in the fluid (Lai et al., 1991). In the triphasic theoretical model, the porous medium was assumed to be composed of solid, fluid and ions. Modified mass balance and momentum balance equations, in addition to the ionic diffusion equation, were introduced. These equations were then used to describe the deformation and stress of biological soft tissues like cartilage and to derive corresponding finite element (FE) formulations (Snijders et al., 1995). Later, a quadriphasic model was presented, to investigate quasi-static finite deformation of swelling of incompressible porous media, where the ions in the fluid are decompounded as anions and cations (Huyghe and Janssen, 1997). In this model, balance laws are derived for each phase and for the mixture as a whole. The guadriphasic model, considering electricosmosis and streaming current effects, can be applied to the analysis of intervertebral disk tissue (Frijns et al., 1997). More recently, a thermo-electro-chemo-mechanical formulation based on the quadriphasic mixture model has been developed for quasi-static finite deformation of swelling incompressible porous media (Huyghe and Janssen 1999). It is noted that both triphasic and quadriphasic models belong to the category of mixture theory based on the poroelastic framework of porous media. However, these two models provide balance equations of the mixture only, and an explicit form of constitutive law does not appear in the related literature. Therefore, the solution of these theoretical models largely depends on the form of constitutive law used.

In addition to the mixture methods discussed above, other types of multi-field approaches have been presented in recent years (Moyne and Murad, 2002; Loret et al., 2002; De Sudipto and Aluru, 2004; Wallmersperger et al., 2004; Gajo and Loret, 2003; Yang et al., 2004, 2005; Zohdi, 2004) to reveal the electro-chemo-mechanical coupling behavior of porous media and to try to explain interactions among the fields. On the basis of these theories, some numerical methods have been developed to solve the coupled multi-field differential equations, including direct iteration procedures (De Sudipto and Aluru, 2004; Wallmersperger et al., 2004) and the FE method (Gajo and Loret, 2003; Yang et al., 2004, 2005; Zohdi, 2004).

In this paper, a theoretical model and the corresponding FE formulation for thermo-electro-chemo-mechanical coupled problems are presented, developed by redefining linearly coupled constitutive relations and extending the traditional Gibb's free energy to include chemical effects. In contrast to previous work, the theoretical model proposed is based on a newly introduced linear constitutive chemical law instead of the balance laws which have been used in the triphasic and quadriphasic mixture models (Lai et al., 1991; Snijders et al., 1995; Huyghe and Janssen, 1997; Frijns et al., 1997; Huyghe and Janssen, 1999). As existing chemical governing equations are not suitable

for FE analysis, we start by deriving a modified form of basic equation for the chemical field (Section 2). By extending the traditional Gibb's free energy to include chemical field, we obtain linear forms of coupled constitutive laws and a variational principle including chemical effect (Sections 3 and 4). As a special case of the coupled system, coupling between chemical and mechanical fields is discussed in detail, and the determination of some coupled property parameters is also demonstrated (Section 5). Finally, numerical examples are performed to assess the coupled chemo-mechanical behavior for medium (Section 6).

#### 2. Governing equations of fields

Consider a thermo-electro-chemo-mechanical body of volume  $\Omega$  bounded by surface *S*. The governing equations, including heat conduction equation, Maxwell equation of electrostatics, equilibrium equations of stresses, and diffusion equations of ions, are as follows:

(1) Equilibrium equations of stresses:

$$\sigma_{ij,j} + f_i = 0 \quad (\text{in } \Omega) \tag{1}$$

(2) Boundary conditions:

$$u_i = \bar{u}_i \quad (\text{on } S_u) \tag{2a}$$

$$\sigma_{ij}n_j = t_i \quad (\text{on } S_t) \tag{2b}$$

(3) Maxwell's equation of electrostatics:

$$D_{i,i} = q_b \quad (\text{in } \Omega) \tag{3}$$

(4) Electric boundary conditions:

$$\phi = \bar{\phi} \quad (\text{on } S_{\phi}) \tag{4a}$$

$$D_i n_i = -\bar{q}_s \quad (\text{on } S_D) \tag{4b}$$

(5) Heat conduction equation:

$$h_{i,i} = -T_0 \dot{\eta} \quad (\text{in } \Omega) \tag{5}$$

where

$$h_{i,i} = \frac{\partial h_1}{\partial x_1} + \frac{\partial h_2}{\partial x_2} + \frac{\partial h_3}{\partial x_3}$$

(6) Thermal boundary conditions:

$$T = T \quad (\text{on } S_T) \tag{6a}$$

$$h_i n_i = h_n \quad (\text{on } S_h) \tag{6b}$$

In these equations,  $\sigma_{ij}$ ,  $D_i$ ,  $\phi$ ,  $h_i$  and T are, respectively, stress tensor, electric displacement vector, electric potential, heat flux vector and temperature,  $f_i$  and  $q_b$  are the mechanical body force and body electric charge density,  $S_u$ ,  $S_t$ ,  $S_\phi$ ,  $S_D$ ,  $S_T$  and  $S_h$  denote, respectively, the corresponding surface of displacement, traction, electric potential, electric displacement, temperature and heat flux.  $\bar{u}_i$  and  $\bar{t}_i$  are the prescribed surface displacements and tractions,  $\bar{\phi}$  and  $\bar{q}_s$  are the prescribed electric potential and surface electric charge,  $\bar{T}$  and  $\bar{h}_n$  are the prescribed temperature change and heat flux on the surface *S*,  $n_i$  is the unit outward normal vector on the surface *S*,  $\eta$  is the entropy density,  $T_0$  is reference temperature, and  $S = S_u + S_t = S_\phi + S_D = S_T + S_h$ .

E

(7) Basic equations of chemical field:

Fick's law shows that the mass flux  $\xi^{\pm}$  is proportional to the gradient of the ionic concentrations  $c_{j}^{\pm}$  by Levine Ira (2002):

$$\xi_i^{\pm} = -\varphi_{ij}^{\pm} c_j^{\pm} \tag{7}$$

where '+' and '-' denote anion and cation, respectively. The proportional coefficients  $\varphi_{ij}^{\pm}$  denote the diffusion coefficients of anions and cations and depend on the intrinsic features of the medium. For isotropic medium,  $\varphi_{ij}^{\pm} = \varphi^{\pm} \delta_{ij}$ .  $c^{\pm}$  are increments of concentrations for the anion and cation. Therefore,  $c^{\pm}$  can be related to the current concentrations. The convection-diffusion equations of the ions can thus be written as

$$\frac{\partial c^{\pm}}{\partial t} + \left(c^{\pm} \nu_{i}\right)_{,i} - \left(\frac{\varphi_{ij}^{\pm} c^{\pm}}{RT^{*}} \mu_{,j}^{\pm}\right)_{,i} = 0$$
(8)

where **v** is the convective velocity of the ions, *R* is the universal gas constant and  $T^*$  is the absolute temperature. The first term in Eq. (8) represents the change rate of the concentrations with respect to the time. The second term stands for the convection effect that describes the macroscopic motion of the ions. The third term is the diffusion of the ions. The electric potential produced by ion is very small comparing with one by the applied electric field and therefore is ignored.

For the motion of ions in fluid, the primary mechanism is due to the ionic diffusion. By ignoring the convection effect in Eq. (8), which means the macroscopic motion of the ions is not been considered, we have

$$\frac{\partial c^{\pm}}{\partial t} - \left(\frac{\varphi_{ij}^{\pm}c^{\pm}}{RT^{*}}\mu_{j}^{\pm}\right)_{,i} = 0$$
(9)

where  $\mu^{\pm}$  is chemical potential. In classical physical chemistry, the chemical potential and concentration of the ions have the following relations:

$$\mu^{\pm} = \mu_0^{\pm} + RT^* \ln \bar{c}^{\pm} \tag{10}$$

where  $\mu_0^\pm$  is a reference potential of anion and cation in the standard state.

Substituting Eq. (10) into Eq. (9) leads to

$$\frac{\partial c^{\pm}}{\partial t} - \left(\varphi_{ij}^{\pm}c_{j}^{\pm}\right)_{,i} = 0 \tag{11}$$

It is noted that Eqs. (8) and (11) agree with ones given by Moyne and Murad (2002) for the motion of ions. For an isotropic medium, we have

$$\frac{\partial c^{\pm}}{\partial t} + \xi_{i,i}^{\pm} = 0 \quad (\text{in } \Omega)$$
(12)

The corresponding natural boundary condition is

$$\xi_i^{\pm} n_i = \xi_n^{\pm} \quad (\text{on } S) \tag{13}$$

where  $\xi_n^{\pm}$  is the ionic flux on the surface of the domain.

Eq. (12) and natural boundary condition (13) are the governing equations of ionic diffusion. For the multi-field coupling case, we must modify Eq. (12) to consider the coupling effect. For this purpose, taking differentiation with respect to time t leads to

$$\frac{\partial \mu^{\pm}}{\partial t} = \frac{RT^*}{c_0^{\pm}} \frac{\partial c^{\pm}}{\partial t}$$
(14)

Using Eqs. (12) and (14), we obtain the diffusion equations of ions in the following form:

$$\frac{\partial \mu^{\pm}}{\partial t} + \frac{RT^{*}}{c_{0}^{\pm}} \xi_{i,i}^{\pm} = 0$$
(15)

Relations between displacements  $u_i$  and strains  $\varepsilon_{ij}$  for elastic field, electric potential  $\phi$  and electric fields  $E_i$  for electrostatics, temperature change T and heat flux  $h_i$  for heat conduction, ionic flux  $\xi_i^{\pm}$  and concentration change  $c^{\pm}$  for chemical field are as follows:

$$s_{ij} = \frac{1}{2}(u_{i,j} + u_{j,i}) \tag{16}$$

$$E_i = -\phi_{,i} \tag{17}$$

$$h_i = -k_{ij}T_{,j} \tag{18}$$

$$\xi_i^{\pm} = -\varphi_{ij}^{\pm} c_j^{\pm} \tag{19}$$

where  $k_{ij}$  are heat conduction coefficients. It is noted that the above mentioned theory is restricted to small deformation.

#### 3. Free energy and constitutive laws

In this section, the Gibb's free energy function in Qin (2001) and Mason (1950) is extended to include chemical effect and is used to derive the linear constitutive law for thermo-electro-chemo-mechanical systems. For a system including thermal, electrical, chemical and mechanical interaction, the extended Gibb's free energy per volume can be written by adding chemical energy in the following form:

$$g = U - E_i D_i - \eta T + \sum_{\alpha = +, -} \mu^{\alpha} c^{\alpha}$$
<sup>(20)</sup>

where g is extended Gibb's free energy density, U denotes the internal energy density, the second and third terms stand for the energy contributions of electric and temperature fields, respectively, and the last term is the chemical energy. The Gibb's free energy containing the first three terms in Eq. (20) has discussed elsewhere (Qin, 2001; Mason, 1950). The last term should be added to the Gibb's free energy when the chemical effect is considered (Levine Ira, 2002). An exact differential of Gibb's free energy function (20) with respect to its independent variables ( $\varepsilon$ , *E*, *T* and *c*) leads to

$$dg = \sigma_{ij} d\varepsilon_{ij} - D_m dE_m - \eta dT + \sum_{\alpha = +, -} \mu^{\alpha} dc^{\alpha}$$
(21)

Thus, we obtain

$$\sigma_{ij} = \frac{\partial g}{\partial \varepsilon_{ij}}, \quad D_i = -\frac{\partial g}{\partial E_i}, \quad \eta = -\frac{\partial g}{\partial T}, \quad \mu^{\pm} = \frac{\partial g}{\partial c^{\pm}}$$
(22)

When the function g is expanded with respect to T,  $\varepsilon_{ij}$ ,  $E_m$  and  $c^{\pm}$  within the scope of linear interactions, we have

$$g = \frac{1}{2} \left( \sigma_{ij} \varepsilon_{ij} - D_i E_i - \eta T + \sum_{\alpha = +, -} \mu^{\alpha} c^{\alpha} \right)$$
(23)

The following constants can then be defined:

$$\begin{split} C_{ijkl} &= \left[\frac{\partial^2 g}{\partial \varepsilon_{ij} \partial \varepsilon_{kl}}\right], \quad \kappa_{nm} = -\left[\frac{\partial^2 g}{\partial E_n \partial E_m}\right], \\ R_{ij}^{\pm} &= -\left[\frac{\partial^2 g}{\partial \varepsilon_{ij} \partial c^{\pm}}\right], \quad \nu^{\pm} = -\left[\frac{\partial^2 g}{\partial T \partial c^{\pm}}\right], \\ \alpha &= \frac{\rho C_{\nu}}{T_0} = -\left[\frac{\partial^2 g}{\partial T^2}\right], \quad e_{mij} = -\left[\frac{\partial^2 g}{\partial \varepsilon_{ij} \partial E_m}\right], \\ w_m^{\pm} &= -\left[\frac{\partial^2 g}{\partial E_m \partial c^{\pm}}\right], \quad \lambda_{ij} = -\left[\frac{\partial^2 g}{\partial \varepsilon_{ij} \partial T}\right], \\ \chi_m &= -\left[\frac{\partial^2 g}{\partial T \partial E_m}\right], \quad s^{\pm} = \left[\frac{\partial^2 g}{\partial c^{\pm} \partial c^{\pm}}\right] \end{split}$$
(24)

where  $c_{ijkl}$  are the elastic coefficients,  $\kappa_{nm}$  the dielectric constants,  $\rho$  the density,  $C_{\nu}$  the specific heat per unit mass,  $e_{mij}$  the piezoelectric coefficients,  $\lambda_{ij}$  the thermal-stress coefficients, and  $\chi_m$  the pyroelectric coefficients. The newly introduced constants  $R_{ij}^{\pm}$ ,  $\nu^{\pm}$ ,  $w_m^{\pm}$ , and  $s^{\pm}$  are, respectively, the mechanical-chemical coefficients measured at a constant temperature and electric field, the thermo-chemical coefficients measured at constant strain and electric field, the electrical-chemical coefficients measured at a constant strain and temperature, and the chemical potential constant measured at a constant strain and temperature and electric field for anion and cation.

When the function g is differentiated according to Eq. (21), and the above constants are used, we find

$$\sigma_{ij} = C_{ijkl} \varepsilon_{ij} - \lambda_{ij} T - e_{ijn} E_n - \sum_{\alpha = +, -} R_{ij}^{\alpha} c^{\alpha}$$
(25a)

$$\eta = \lambda_{ij}\varepsilon_{ij} + \alpha T + \chi_n E_n + \sum_{\alpha=+,-} \nu^{\alpha} c^{\alpha}$$
(25b)

$$D_m = e_{klm} \varepsilon_{kl} + \chi_m T + \kappa_{mn} E_n + \sum_{\alpha = +, -} w_m^{\alpha} c^{\alpha}$$
(25c)

$$\mu^{\pm} = -R_{kl}^{\pm}\varepsilon_{kl} - \nu^{\pm}T - w_n^{\pm}E_n + s^{\pm}c^{\pm}$$
(25d)

A set of these equations is the constitutive relation in the coupled system.

It is noted that the constitutive equations (25a)–(25d) are extensions of known thermo-electro-mechanical coupling (Qin, 2001; Mason, 1950) to include chemical field. In classical physical chemistry, the relation between chemical potential and ionic concentrations is expressed by a logarithm function [see Eq. (10)], but here we assume a linear relationship between the potential and the concentration changes, which means that Eqs. (25a)–(25d) apply for a small change of ionic concentrations only. This assumption allows us to develop the corresponding numerical model for FE formulation in a simple way. Actually, the classical logarithm relation is applied to determine the present linear coefficients, as shown in the following section.

Finally, using the material parameters defined in Eqs. (25a)-(25d) we can rewrite the Gibb's free energy function equation (23) as

$$g = \frac{1}{2} C_{ijkl} \varepsilon_{kl} \varepsilon_{ij} - \lambda_{ij} T \varepsilon_{ij} - e_{ijn} E_n \varepsilon_{ij} - \sum_{\alpha = +, -} R^{\alpha}_{ij} c^{\alpha} \varepsilon_{ij}$$
$$- \frac{1}{2} \alpha T^2 - \chi_n E_n T - \sum_{\alpha = +, -} \nu^{\alpha} c^{\alpha} T - \frac{1}{2} \kappa_{mn} E_n E_m$$
$$- \sum_{\alpha = +, -} w^{\alpha}_m c^{\alpha} E_m + \frac{1}{2} \sum_{\alpha = +, -} s^{\alpha} c^{\alpha} c^{\alpha}$$
(26)

This energy function is used as a basis for developing FE formulations in the following sections.

#### 4. Variational principle

Variational functional plays a central role in the formulation of the fundamental governing equations in finite element method (FEM). For the boundary value problem described in Section 2 and the linear constitutive equations (25a)–(25d), the variational functional used for deriving FE formulation of thermo-electro-chemo-mechanical coupling system can be constructed in the form

$$\Pi = \int_{\Omega} \left( \dot{B} + F + J \right) d\Omega - \int_{\Omega} \left( f_i \dot{u}_i - q_b \dot{\phi} \right) d\Omega$$
$$- \int_{S} \left( \bar{t}_i \dot{u}_i + \bar{q}_s \dot{\phi} - \frac{T}{T_0} \bar{h}_n - \tau \sum_{\alpha = +, -} \bar{\xi}_n^{\alpha} c^{\alpha} \right) dS$$
(27)

where  $\dot{B} = \frac{\partial B}{\partial t}$ ,  $\tau = \frac{RT^*}{c_0^+}$ . The vanishing variation of functional (27) leads to

$$\delta\Pi = \int_{\Omega} \delta\dot{B} d\Omega + \delta \int_{\Omega} (F+J) d\Omega - \int_{\Omega} \left( f_i \delta\dot{u}_i - q_b \delta\dot{\phi} \right) d\Omega$$
$$- \int_{S} \left( \bar{t}_i \delta\dot{u}_i + \bar{q}_s \delta\dot{\phi} - \frac{\bar{h}_n}{T_0} \delta T - \tau \sum_{\alpha=+,-} \bar{\xi}_n^{\alpha} \delta c^{\alpha} \right) dS = 0$$
(28)

where *B* is the generalized Biot's free energy (Mason, 1950) of the coupled four-field system. The function *B* should satisfy the following conditions:

$$\frac{\partial B}{\partial \varepsilon_{ij}} = \sigma_{ij}, \quad \frac{\partial B}{\partial T} = \eta, \quad \frac{\partial B}{\partial E_i} = D_i, \quad \frac{\partial B}{\partial c^{\pm}} = \mu^{\pm}$$
(29)

It should be noted that the minus sign in Eq. (22) disappears here. Thus the function *B* has following differential form:

$$\delta \dot{B} = \delta \dot{g} + 2D_n \delta \dot{E}_n + 2\dot{\eta} \delta T \tag{30}$$

The function J in Eq. (27) is the dissipation energy caused by ionic diffusion and F is the heat dissipation. They are defined by

$$J = -\frac{1}{2}\tau \sum_{\alpha=+,-} \xi_i^{\alpha} c_{,i}^{\alpha}$$
(31a)

$$F = -\frac{1}{2T_0}h_iT_{,i} \tag{31b}$$

Substituting Eqs. (30) and (31) into Eq. (28) yields

$$\begin{split} \delta\Pi &= \int_{\Omega} \left( \sigma_{ij} \delta \dot{\varepsilon}_{ij} + \dot{\eta} \delta T + D_n \delta \dot{E}_n + \sum_{\alpha = +, -} \dot{\mu}^{\alpha} \delta c^{\alpha} \right) d\Omega \\ &- \frac{1}{2} \delta \int_{\Omega} \left( \frac{1}{T_0} h_i T_{,i} + \tau \sum_{\alpha = +, -} \xi_i^{\alpha} c_{,i}^{\alpha} \right) d\Omega \\ &- \int_{\Omega} \left( f_i \delta \dot{u}_i - q_b \delta \dot{\phi} \right) d\Omega \\ &- \int_{S} \left( \bar{t}_i \delta \dot{u}_i + \bar{q}_s \delta \dot{\phi} - \frac{\bar{h}_n}{T_0} \delta T - \tau \sum_{\alpha = +, -} \bar{\xi}_n^{\alpha} \delta c^{\alpha} \right) dS = 0 \end{split}$$
(32)

Integrating by parts, Eq. (32) can be further written as

$$\begin{split} \delta\Pi &= \int_{\Omega} \left[ -(\sigma_{ijj} + f_i) \delta \dot{u}_i + \left(\frac{h_{i,i}}{T_0} + \dot{\eta}\right) \delta T + (D_{i,i} - q_b) \delta \dot{\phi} \\ &+ \sum_{\alpha = +, -} (\tau \xi^{\alpha}_{i,i} + \dot{\mu}^{\alpha}) \delta c^{\alpha} \right] d\Omega + \int_{S} \left[ (\sigma_{ij} n_j - \bar{t}_i) \delta \dot{u}_i \\ &+ (D_i n_i + \bar{q}_s) \delta \dot{\phi} + \left(\frac{\bar{h}_n}{T_0} - \frac{h_i n_i}{T_0}\right) \delta T \\ &+ \sum_{\alpha = +, -} \tau (\bar{\xi}^{\alpha}_n - \xi^{\alpha}_i n_i) \delta c^{\alpha} \right] dS = 0 \end{split}$$
(33)

Due to the arbitrariness of  $\delta u_i$ ,  $\delta \phi$ ,  $\delta T$  and  $\delta c^{\pm}$ , the variational equation (33) leads to following governing equations and natural boundary conditions:

$$\begin{aligned} \sigma_{ij,i} + f_i &= 0 \quad \sigma_{ij}n_j = \bar{t}_i \\ D_{i,i} &= q_b \quad D_i n_i = -\bar{q}_s \\ h_{i,i} &= -T_0 \dot{\eta} \quad h_i n_i = \bar{h}_n \\ \xi^{\pm}_{i,i} + \frac{1}{\tau} \dot{\mu}^{\pm} &= 0 \quad \xi^{\pm}_i n_i = \bar{\xi}^{\pm}_n \end{aligned}$$

Obviously, they are the Euler equations of functional (27) and represent the governing equations (1), (3), (5) and (15) of the mechanical, thermal, electrical and chemical fields, respectively, as well as the corresponding natural boundary conditions, respectively.

It is interesting to note that the governing equations of the chemical field have the same forms as those of heat conduction problem. In other words, there is an analogous relation between ionic diffusion and heat conduction. Table 1 lists the analogous relations between chemical problems and heat conduction problems.

 Table 1

 Analogies of ionic diffusion and heat conduction.

Heat conduction	lonic diffusion
Temperature T	Concentration $c$
Heat flux $h_i = -\kappa_{ij}T_j$	Mass flux $\xi_i = -\varphi_{ij}c_j$
Entropy change rate $\dot{\eta}$	Chemical potential change rate $\mu$
Energy form $\eta T$	Energy form $\mu c$
Governing equation	Governing equation
$h_{i,i} + \frac{1}{T_0}\dot{\eta} = 0$	$\xi_{i,i} + \frac{\xi_0}{R^2} \cdot \mu = 0$
Boundary condition $h_i n_i = \bar{h}_n$	Boundary condition $\xi_i n_i = \overline{\xi}_n$

### 5. Chemo-mechanical formulation

For some media, the electric and thermal effects are weak with respect to chemical and mechanical effects and they can be ignored. Thus the governing equations of chemo-mechanical couplings can be expressed as

$$\varphi^{\pm}c_{,ii}^{\pm} - \frac{1}{\tau}\dot{\mu}^{\pm} = 0 \quad \sigma_{ij,j} + f_i = 0$$
(34)

where the inertial effect is neglected. The natural boundary conditions are

$$\varphi^{\pm}c_{,i}^{\pm}n_{i} = -\bar{\zeta}_{n}^{\pm} \quad \sigma_{ij}n_{j} - \bar{t}_{i} = 0$$
(35)

As in previous sections, the chemical governing equations modified by coupling coefficient are used. The generalized variational principle for the chemo-mechanical coupling is then given by

$$\delta \int_{\Omega} (\dot{g} + J) d\Omega - \int_{\Omega} f_i \delta \dot{u}_i \, d\Omega - \int_{S_t} \bar{t}_i \delta \dot{u}_i \, dS$$
$$- \sum_{\alpha = +, -} \int_{S_\xi} \tau \bar{\xi}_n^{\alpha} \delta c^{\alpha} \, dS = 0$$
(36)

where g is Gibb's free energy for chemo-mechanical coupling problem, J is chemical dissipation energy with the forms

$$\delta g = \sigma_{ij} \delta \varepsilon_{ij} + \mu \delta c \tag{37a}$$

$$J = -\frac{1}{2}\tau \sum_{\alpha = +, -} \xi_{i}^{\alpha} c_{,i}^{\alpha} = \frac{1}{2}\tau \sum_{\alpha = +, -} \varphi^{\alpha} c_{,i}^{\alpha} c_{,i}^{\alpha}$$
(37b)

Substituting Eqs. (37a) and (37b) into Eq. (36), we have

$$\begin{split} \delta\Pi &= \int_{\Omega} \left( \sigma_{ij} \delta \dot{\varepsilon}_{ij} + \dot{\mu} \delta c + \tau \sum_{\alpha = +, -} \varphi^{\alpha} c_{,i}^{\alpha} c_{,i}^{\alpha} \right) d\Omega - \int_{\Omega} f_{i} \delta \dot{u}_{i} d\Omega \\ &- \int_{s} \bar{t}_{i} \delta \dot{u}_{i} dS + \sum_{\alpha = +, -} \int_{s} \tau \bar{\xi}_{n}^{\alpha} \delta c^{\alpha} dS = \mathbf{0} \\ &= \int_{\Omega} \left( C_{ijkl} \varepsilon_{ij} \delta \dot{\varepsilon}_{kl} - \sum_{\alpha = +, -} R_{ij}^{\alpha} c^{\alpha} \delta \dot{\varepsilon}_{kl} - \sum_{\alpha = +, -} R_{kl}^{\alpha} \dot{\varepsilon}_{kl} \delta c^{\alpha} \\ &+ \sum_{\alpha = +, -} s^{\alpha} \dot{c}^{\alpha} \delta c^{\alpha} + \tau \sum_{\alpha = +, -} \varphi^{\alpha} c_{,i}^{\alpha} c_{,i}^{\alpha} \right) d\Omega - \int_{\Omega} f_{i} \delta \dot{u}_{i} d\Omega \\ &- \int_{s} \bar{t}_{i} \delta \dot{u}_{i} dS + \sum_{\alpha = +, -} \int_{s} \tau \bar{\xi}_{n}^{\alpha} \delta c^{\alpha} dS = \mathbf{0} \end{split}$$
(38)

After some algebra, the equation becomes

$$\delta \Pi = \int_{\Omega} (\sigma_{ij} + f_i) \delta \dot{u}_i d\Omega + \int_{S} (\sigma_{ij} n_j - \bar{t}_i) \delta \dot{+} u_i dS$$
  
+ 
$$\sum_{\alpha = +, -} \int_{\Omega} (-\tau \varphi^{\alpha} c^{\alpha}_{,ii} + \dot{\mu}) \delta c^{\alpha} d\Omega$$
  
+ 
$$\sum_{\alpha = +, -} \int_{S} \tau (\varphi^{\alpha} c^{\alpha}_{,i} n_i + \xi^{\alpha}_n) \delta c^{\alpha} dS = 0$$
(39)

Eq. (39) is equivalent to Eqs. (34) and (35).

Eq. (38) can be written in compact matrix form for the purpose of FE formulation:

$$\delta \Pi = \int_{\Omega} \left( \delta \dot{u} \mathbf{B}^{\mathrm{T}} \mathbf{C} \mathbf{B} u - \delta \dot{u} \mathbf{B}^{\mathrm{T}} \mathbf{R} \mathbf{N} c - \delta c \mathbf{N}^{\mathrm{T}} \mathbf{R} \mathbf{B} \frac{\partial u}{\partial t} + \delta c \mathbf{N}^{\mathrm{T}} \mathbf{s} \mathbf{N} \frac{\partial c}{\partial t} \right. \\ \left. + \tau \delta c \mathbf{B}_{c}^{\mathrm{T}} \varphi \mathbf{B}_{c} c \right) d\Omega - \int_{\Omega} \delta \dot{u} \mathbf{N}^{\mathrm{T}} \mathbf{f} \, d\Omega - \int_{\mathrm{s}} \delta \dot{u} \mathbf{N}^{\mathrm{T}} \mathbf{\bar{t}} \, dS \\ \left. + \tau \int_{\mathrm{s}} \delta c \mathbf{N}^{\mathrm{T}} \bar{\boldsymbol{\xi}} \, dS = \mathbf{0}$$
(40a)

$$\delta \Pi = \delta \dot{u} \left( \int_{\Omega} \left( \mathbf{B}^{\mathrm{T}} \mathbf{C} \mathbf{B} u - \mathbf{B}^{\mathrm{T}} \mathbf{R} \mathbf{N} c \right) d\Omega - \int_{\Omega} \mathbf{N}^{\mathrm{T}} \mathbf{f} d\Omega - \int_{\mathrm{s}} \mathbf{N}^{\mathrm{T}} \bar{\mathbf{t}} dS \right) + \delta c \left( \int_{\Omega} -\mathbf{N}^{\mathrm{T}} \mathbf{R} \mathbf{B} \dot{u} + \mathbf{N}^{\mathrm{T}} \mathbf{s} \mathbf{N} \dot{c} + \tau \mathbf{B}_{c}^{\mathrm{T}} \varphi \mathbf{B}_{c} c + \tau \int_{\mathrm{s}} \mathbf{N}^{\mathrm{T}} \bar{\boldsymbol{\xi}} dS \right)$$
(40b)

This leads to following FE equations:

r

$$\begin{bmatrix} \mathbf{0} & \mathbf{0} \\ \mathbf{K}_{cm}^{T} & \mathbf{M}_{c} \end{bmatrix} \left\{ \begin{array}{c} \dot{u} \\ \dot{c} \end{array} \right\} + \begin{bmatrix} \mathbf{K}_{mm} & \mathbf{K}_{mc} \\ \mathbf{0} & \mathbf{K}_{cc} \end{bmatrix} \left\{ \begin{array}{c} u \\ c \end{array} \right\} = \left\{ \begin{array}{c} \mathbf{F}_{m} \\ \mathbf{F}_{c} \end{array} \right\}$$
(41)

in which the coefficient matrices have following forms:

$$\mathbf{M}_{c} = \int_{\Omega} \mathbf{N}^{T} \mathbf{S} \mathbf{N} \, d\Omega \tag{42}$$
$$\mathbf{K}_{mm} = \int_{\Omega^{e}} \mathbf{B}^{T} \mathbf{C} \mathbf{B} \, d\Omega, \mathbf{K}_{mc} = \mathbf{K}_{cm}^{T} = -\int_{\Omega^{e}} \mathbf{B}^{T} \mathbf{R} \widetilde{\mathbf{N}} \, d\Omega,$$
$$\mathbf{K}_{cc} = \int_{\Omega} \tau \mathbf{B}_{c}^{T} \varphi \mathbf{B}_{c} \, d\Omega \tag{43}$$

The equivalent nodal force vectors are

$$\mathbf{F}_{m}^{e} = \int_{\Omega^{e}} \mathbf{N}^{T} \mathbf{f} \, d\Omega + \int_{S_{t}^{e}} \mathbf{N}^{T} \bar{\mathbf{t}}^{e} \, dS$$
$$\mathbf{F}_{c}^{e} = -\int_{S} \tau \mathbf{N}^{T} \boldsymbol{\xi}_{n}^{e} \, dS$$
(44)

For material parameters of chemo-mechanical media, the mechanical stiffness matrix can be easily obtained through Young's modulus and Poisson's ratio, and the diffusion coefficient can also be measured by a physicochemical method (Levine Ira, 2002). Here, the new parameters to be determined are the proportional coefficients and the coupling coefficient of the chemical and mechanical fields, which can be estimated by a theoretical method.

Let us first consider the linearly proportional coefficients of chemical potential and concentration. In this study the linear relation  $\mu^{\pm} = s^{\pm}c^{\pm}$  is assumed to be satisfied. Based on the theory of physical chemistry, the dimensions of  $c^{\pm}$  and  $\mu^{\pm}$  are, respectively, mol m<sup>-3</sup> and J m<sup>-3</sup>, thus  $s^{\pm}$  should have the dimension of J mol<sup>-1</sup>.

We use the same notation to rewrite Eq. (10) as

$$\mu^{\pm} = \frac{RT^{*}}{\overline{V}^{\pm}} \ln \bar{c}^{\pm} = \frac{RT^{*}}{\overline{V}^{\pm}} \ln \left( c_{0}^{\pm} + c^{\pm} \right)$$
(45)

Then the differentiation of Eq. (45) is given by

$$d\mu^{\pm} = s^{\pm} dc^{\pm} = \frac{RT^{*}}{\overline{V}^{\pm}} \frac{dc^{\pm}}{c_{0}^{\pm} + c^{\pm}}$$
(46)

Considering an infinitesimal change in concentration, we have

$$s^{\pm} = \frac{RT^{*}}{\overline{V}^{\pm}} \lim_{c^{\pm} \to 0} \frac{1}{c_{0}^{\pm} + c^{\pm}} = \frac{RT^{*}}{\overline{V}^{\pm}c_{0}^{\pm}}$$
(47)

If the temperature change is very small, i.e.  $T^{\dagger} = T_0 + T \approx T_0$ , Eq. (47) can be rewritten as

$$s^{\pm} = \frac{RT_0}{\overline{V}^{\pm}c_0^{\pm}} \tag{48}$$

Since the dimensions of the parameters R,  $T_0$  and  $\overline{V}^{\pm}$  are, respectively, J mol<sup>-1</sup> K<sup>-1</sup>, K, and m<sup>3</sup> mol<sup>-1</sup>, the dimension of  $s^{\pm}$  should be J mol<sup>-1</sup>, which is consistent with the dimension in the present linear relation. The value of  $s^{\pm}$  depends on the properties of the material.

Concerning the coupled coefficient of chemical and mechanical effects, we can use the analogous relation of chemical and heat conduction to predict its value. Considering an isotropic material and supposing that concentration changes induce only swelling and contraction, and no shear deformation of the medium is produced, the constitutive laws for chemo-mechanical coupling can be written as

$$\begin{aligned} \sigma_{ij} &= C_{ijkl} \varepsilon_{ij} - \sum_{\alpha = +, -} R^{\alpha}_{ij} c^{\alpha} \\ &= \frac{\nu E}{(1+\nu)(1-2\nu)} \varepsilon_{kk} \delta_{ij} + \frac{E}{1+\nu} \varepsilon_{ij} - \sum_{\alpha = +, -} R^{\alpha}_{0} \delta_{ij} c^{\alpha} \end{aligned}$$
(49a)

$$\mu^{\pm} = \frac{R_{kl}^{\pm}}{V^{\pm}} \varepsilon_{kl} + s^{\pm} c^{\pm} = \frac{R_{0}^{\pm}}{V^{\pm}} \delta_{kl} \varepsilon_{kl} + s^{\pm} c^{\pm}$$
(49b)

In order to estimate the coupled coefficients  $R_0^+$ , setting i = jin the above equations and denoting  $\varepsilon_{ii} = \varepsilon_{11} + \varepsilon_{22} + \varepsilon_{33}$  as the volume strain, the volume stress is given by

$$\sigma_{ii} = \frac{E}{1 - 2\nu} \varepsilon_{ii} - 3R_0^+ c^+ - 3R_0^- c^-$$
(50)

It is assumed that the swelling and contraction of the material induced by concentration change starts from the free stress state, e.g.  $\sigma_{ii}$  = 0. Thus we can obtain the volume expansion coefficient

$$K^{+} = \left(\frac{\varepsilon_{ii}}{c^{+}}\right)_{\sigma=0} = \frac{3(1-2\nu)}{E}R_{0}^{+}$$
(51a)

$$K^{-} = \left(\frac{\varepsilon_{ij}}{c^{-}}\right)_{\sigma=0} = \frac{3(1-2\nu)}{E}R_{0}^{-}$$
(51b)

Then the coupled coefficients are

$$R_0^{\pm} = \frac{E}{3(1-2\nu)} K^{\pm}$$
(52)

Note that the dimension of *E* is N m<sup>-2</sup> and dimension of  $K^{\pm}$  is m<sup>3</sup> mol<sup>-1</sup>, the dimension of  $R_0^{\pm}$  is, thus, N m mol<sup>-1</sup>.

#### 6. Numerical examples

Since the main purpose of this paper is to outline the basic principles of the proposed multi-field approach including a chemical effect, the assessment is limited to the swelling of a rectangular plate subjected to chemical load and a rectangular strip subjected to a chemical stimulus on its longer side. The two examples illustrate the coupling between chemical and mechanical fields. The material parameters and values used in present calculations are listed in Table 2.

Table 2
Material parameters used in numerical examples.

Young's modulus E (Pa)	Poisson's ratio v	Diffusion coefficient $\phi^+$ (m <sup>2</sup> /s)	Diffusion coefficient $\phi^-$ (m <sup>2</sup> /s)	Coefficients of the chemical potential $s^+$ , $s^-$ (N m mol <sup>-1</sup> )	Chemo-mechanical coupling coefficient $R_0^+$ , $R_0^-$ (N m mol <sup>-1</sup> )
$3.5\times10^{5}$	0.45	$\textbf{4.8}\times \textbf{10}^{-10}$	$\textbf{7.8}\times 10^{-10}$	$1.0\times10^3$	$1.75\times10^4$



**Fig. 1.** Mechanical boundary conditions and the input concentrations of a square plate.

**Example 1** (*A square plate subjected to chemical load*). Consider the swelling of a square plate of sides 0.01 m subjected to chemical load. The body force is assumed to vanish. The displacements vanish on the lower boundary and the other sides are traction free. The ionic concentration is applied on the upper boundary, which varies from 0.001 mol/l to 0.01 mol/l, and the concentration is zero on the lateral sides as shown in Fig. 1. As a result, the concentrations of the ions vary linearly in x direction and display symmetry with respect to the *y*-direction. The same distributions for the anions and cations are assumed here.

The deformation of the plate obtained from the proposed formulation is shown in Fig. 2. It is apparent that a volume swelling of the plate occurs under the chemical stimuli, while the degree of swelling varies from point to point. The symmetry of the deformation in x-direction can be found because of the symmetric model and load. Due to the linear distribution of ionic concentration, the maximum swelling occurs on the upper boundary and no swelling occurs on the lower boundary. It is shown that chemical swelling of the medium is very similar to heat expansion where volume expansion produces under thermal load. The variations of maximum displacements versus the ionic concentrations are shown in Fig. 3. It is noted that the maximum displacements  $u_{xmax}$  and  $u_{ymax}$ both increase linearly with an increase of the ionic concentration. The magnitudes of  $u_{xmax}$  and  $u_{ymax}$  are very different due to the difference of ionic distribution in the two directions.

# **Example 2** (*A* rectangular strip subjected to a chemical stimulus on its longer side).

For a rectangle strip of sides  $0.01 \text{ m} \times 0.004 \text{ m}$ , the displacement is fixed in the *y*-direction at the lower side. To avoid rigid body motion, the displacement of the middle point of the lower side is constrained in *x*-direction. A chemical stimulus is applied to the strip on one of its longer sides. This boundary value problem highlights the fact that the swelling deformation of the medium can be affected by the boundary condition and chemical load.

The ionic diffusion results in a linear distribution of ions in the strip. The calculated deformation of the strip is shown in Fig. 4. In contrast with Example 1, a bending deformation of the strip appears due to the non-symmetric swelling which leads to different expansions on the two opposite sides of the strip. It is concluded that the defor-



Fig. 2. Schematic plot of the swelling of a square plate under chemical stimulation: undeformed (left) and deformed state (right).



Fig. 3. Variations of maximum displacement via ionic concentrations.



**Fig. 4.** Schematic plot of bending deformation of a strip under chemical load: undeformed state (left) and deformed state (right).

mation model of the whole sample can be controlled by applying proper constraints on the boundaries. A similar phenomenon for a gel fiber has been demonstrated in Wallmersperger et al. (2004). This example illustrates the capability of the present theory to modeling of the deformation of coupling medium under chemical stimulus.

# 7. Conclusions

A finite element formulation is derived based on the governing equations of coupled thermal, electrical, chemical and mechanical fields proposed in this paper. Using the proposed four-field equations, a variational principle for deriving the FE formulation can be easily constructed. The coupling between the chemical field and the other fields is enforced through out-of-diagonal coefficients. Thus the resulting FE procedure is fully coupled in terms of the four fields. Two numerical examples are considered to illustrate the application of the FEM and to verify the proposed theory.

A linearly constitutive relation is obtained using the concept of extended Gibb's free energy. The materials parameters for the chemo-mechanical coupling problem are discussed from the point of the view of theoretical estimations and their dimensions. It is shown that material parameters in the present linear model are related with the physical constants in classical physical chemistry.

It should be pointed out that modification of the basic equation governing the chemical field is necessary for the treatment of coupled behavior. The present investigation modifies the ionic diffusion equations, allowing the chemical field to be coupled with the other fields.

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