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Finite Element Implementation of Mechanochemical Phenomena in Neutral Deformable Porous Media Under Finite Deformation

Biological soft tissues and cells may be subjected to mechanical as well as chemical (osmotic) loading under their natural physiological environment or various experimental conditions. The interaction of mechanical and chemical effects may be very significant under some of these conditions, yet the highly nonlinear nature of the set of governing equations describing these mechanisms poses a challenge for the modeling of such phenomena. This study formulated and implemented a finite element algorithm for analyzing mechanochemical events in neutral deformable porous media under finite deformation. The algorithm employed the framework of mixture theory to model the porous permeable solid matrix and interstitial fluid, where the fluid consists of a mixture of solvent and solute. A special emphasis was placed on solute-solid matrix interactions, such as solute exclusion from a fraction of the matrix pore space (solubility) and frictional momentum exchange that produces solute hindrance and pumping under certain dynamic loading conditions. The finite element formulation implemented full coupling of mechanical and chemical effects, providing a framework where material properties and response functions may depend on solid matrix strain as well as solute concentration. The implementation was validated using selected canonical problems for which analytical or alternative numerical solutions exist. This finite element code includes a number of unique features that enhance the modeling of mechanochemical phenomena in biological tissues. The code is available in the public domain, open source finite element program FEBio (http://mrl.sci.utah.edu/software). [DOI: 10.1115/1.4004810]

1 Introduction

Biological tissues and cells may be modeled as porous media consisting of a solid matrix and an interstitial fluid. This fluid is generally a water-based solution containing solutes of various molecular weights. In many applications the transport of solutes is of specific interest, as these solutes may be involved in a variety of biological processes. Solute transport in a free solution (in the absence of a solid matrix) is characterized by the frictional interactions between the solute and the solvent, as manifested in the classical mechanisms of diffusion and convection [1]. In the presence of a porous solid matrix, further interactions take place between the solute and the solid. These interactions give rise to additional diffusion and convection effects [2], as well as solute exclusion from some of the pore space as a result of steric volume and short-range electrostatic interactions [2-4]. When the porous medium is deformable, as in the case of cells and soft tissues, the interactions of the solute with the solid matrix may be significantly dependent on the state of strain [5-7]. Therefore, the ability to model solute transport in a deformable porous medium represents an important tool for biomedical engineers.

Solute transport in porous deformable media has been an intense subject of analysis in the biomechanics literature over the last two decades. A particular emphasis has been placed on the transport of ions in a charged, deformable solid matrix, as embod-

ied in the triphasic theory of Lai et al. [8], the quadriphasic theory of Huyghe and Janssen [9], and the multielectrolyte theory of Gu et al. [10]. These investigations have often focused on mechanoelectrochemical phenomena arising from the transport of charged species within a charged matrix, such as streaming and diffusion potentials and currents [11,12], Donnan osmotic swelling [8–10,13], and phenomena such as reverse osmosis [14]. Finite element implementations of charged porous media have been presented by several authors, which are applicable to infinitesimal deformations [15–20] and finite deformations [21]. Other investigators have used the analogy between thermal diffusion and solute transport to simulate a triphasic medium under infinitesimal deformation [22], or have constrained their finite element analyses to modeling the equilibrium response to Donnan osmotic swelling under finite deformation [23,24].

The neutral transport of solutes in porous deformable media has not been addressed as extensively as the transport of monovalent counterions in a charged matrix. Sengers et al. [25] formulated a finite element implementation of a biphasic (uncharged) medium, undergoing finite deformation, with solute transport and biosynthesis. Steck et al. [26] and Zhang and Szeri [27] used a commercial finite element code to combine mass (solute) transport with a poroelastic analysis using a two-stage solution procedure.

The objective of this study was to develop a finite element implementation of neutral solute transport in deformable porous media that incorporates a number of important phenomena at the interface of mechanics and physical chemistry: (1) Transport in deformable anisotropic media [28–31], including strain-induced alterations in permeability and diffusivity [6,7,32–34] and strain-

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induced anisotropy [35]. (2) Momentum exchange between solutes and the solid matrix, which is responsible for increased hindrance to transport [2,36] as well as enhanced convection under dynamic loading [37–40]. (3) Changes in tissue and cell volume due to alterations in their osmotic environment [5,41–43]. (4) Partial solute exclusion from pore spaces due to steric volume and short-range electrostatic effects [2–4], which may depend on solid matrix deformation [5,6] and solute concentration [44,45]. (5) Deviation of the physicochemical responses of solutions from ideal behavior with varying solute concentration [46,47] and solid matrix deformation.

All of these modeling features were implemented into FEBio, a free, open-source finite element code available to the general public (http://mrl.sci.utah.edu/software).

2 Finite Element Implementation

2.1 Mixture Framework. The governing equations are based on the framework of mixture theory [48,49]. Only a single solute is considered for notational simplicity, although extension to multiple solutes is straightforward. Various forms of the governing equations have been presented in the literature [27,38,41,50,51], though a presentation that incorporates all the desired features of this implementation has not been reported previously and is thus detailed here.

The fundamental assumptions adopted in this treatment are quasi-static conditions for linear momentum balance, intrinsic incompressibility of all constituents (invariant true densities), isothermal conditions, negligible volume fraction of solute relative to the solid and solvent, and negligible effects of solute and solvent viscosities (friction within constituents) relative to frictional interactions between constituents. These are standard assumptions for studies in biological tissues and cells. The incorporation of phenomena such as the action of external body forces or chemical reactions between constituents is relegated to future studies.

The various constituents of a mixture may be generically denoted by a superscript α or β . In the current implementation the three constituents of the mixture are the porous-permeable solid matrix ($\alpha = s$), the solvent ($\alpha = w$), and the solute ($\alpha = u$). The motion of the solid matrix is described by the displacement vector **u**, the pressure of the interstitial fluid (solvent + solute) is *p*, and the solute concentration (on a solution-volume basis) is *c*.

2.2 Balance of Momentum. The total (or mixture) stress may be described by the Cauchy stress tensor $\mathbf{T} = -p\mathbf{I} + \mathbf{T}^e$, where \mathbf{I} is the identity tensor and \mathbf{T}^e is the stress arising from the strain in the porous solid matrix. The solid matrix is compressible since the volume of pores changes as interstitial fluid enters or leaves the matrix. Under the conditions outlined above, the balance of linear momentum for the mixture reduces to

$$\operatorname{div} \mathbf{T} = -\operatorname{grad} p + \operatorname{div} \mathbf{T}^e = \mathbf{0}$$
 (2.1)

Similarly, the equations of balance of linear momentum for the solvent and solute are given by

$$-\rho^{w} \operatorname{grad} \tilde{\mu}^{w} + \mathbf{f}^{ws} \cdot (\mathbf{v}^{s} - \mathbf{v}^{w}) + \mathbf{f}^{wu} \cdot (\mathbf{v}^{u} - \mathbf{v}^{w}) = \mathbf{0} -\rho^{u} \operatorname{grad} \tilde{\mu}^{u} + \mathbf{f}^{us} \cdot (\mathbf{v}^{s} - \mathbf{v}^{u}) + \mathbf{f}^{uw} \cdot (\mathbf{v}^{w} - \mathbf{v}^{u}) = \mathbf{0}$$
(2.2)

where ρ^{α} is the apparent density (mass of α per volume of the mixture), $\tilde{\mu}^{\alpha}$ is the mechanochemical potential, and \mathbf{v}^{α} is the velocity of constituent α ; $\mathbf{f}^{\alpha\beta}$ is the diffusive drag tensor between constituents α and β representing momentum exchange via frictional interactions, which satisfies $\mathbf{f}^{\beta\alpha} = \mathbf{f}^{\alpha\beta}$. An important feature of (2.2) is the incorporation of a momentum exchange term between the solute and solid matrix, $\mathbf{f}^{us} \cdot (\mathbf{v}^s - \mathbf{v}^u)$, which is often neglected in other treatments but plays an important role for describing solid-solute interactions [38–40]. These momentum equations show that the driving force for the transport of solvent

or solute is the gradient in its mechanochemical potential, which is resisted by frictional interactions with other constituents.

The mechanochemical potential is the sum of the mechanical and chemical potentials. The chemical potential μ^{α} of α represents the rate at which the mixture free energy changes with increasing mass of α . The mechanical potential represents the rate at which the mixture free energy density changes with increasing volumetric strain of α . In a mixture of intrinsically incompressible constituents, where the volumetric strain is idealized to be zero, this potential is given by $(p - p_0)/\rho_T^{\alpha}$, where ρ_T^{α} is the invariant true density of α (mass of α per volume of α), and p_0 is some arbitrarily set reference pressure.

From classical physical chemistry, the general form of a constitutive relation for the chemical potential is $\mu^{\alpha} = \mu_0^{\alpha}(\theta)$ $+(R\theta/M^{\alpha}) \ln a^{\alpha}$ [46], where R is the universal gas constant, θ is the absolute temperature, M^{α} is the molecular weight (invariant), and a^{α} is the activity of constituent α (a nondimensional quantity that represents a measure of the effective concentration of α); $\mu_0^{\alpha}(\theta)$ is the chemical potential at some arbitrary reference state, at a given temperature. For solutes, physical chemistry treatments let $a^{\mu} = \gamma c/c_0$, where c_0 is the solute concentration in some standard reference state (an invariant, typically $c_0 = 1$ M), and γ is the nondimensional activity coefficient, which generally depends on the current state (e.g., concentration) but reduces to unity under the assumption of ideal physicochemical behavior [46]. Since this representation is strictly valid for free solutions only, whereas solutes may be partially excluded from some of the interstitial space of a porous solid matrix, Mauck et al. [38] extended this representation of the solute activity to let $a^{\mu} = \gamma c / \kappa c_0$, where the solubility κ represents the fraction of the pore space which is accessible to the solute ($0 < \kappa \le 1$). In this extended form it becomes clear that even under ideal behavior $(\gamma = 1)$, the solute activity may be affected by the solubility. Indeed, for neutral solutes, the solubility also represents the partition coefficient of the solute between the tissue and external bath [3,4].

When accounting for the fact that the solute volume fraction is negligible compared to the solvent volume fraction [46,52], the general expressions for the mechanochemical potentials $\tilde{\mu}^w$ and $\tilde{\mu}^u$ take the form

$$\tilde{\mu}^{w} = \mu_{0}^{w}(\theta) + \frac{1}{\rho_{T}^{w}}(p - p_{0} - R\theta\Phi c)$$

$$\tilde{\mu}^{u} = \mu_{0}^{u}(\theta) + \frac{R\theta}{\theta} \ln \frac{\gamma c}{\kappa c}$$
(2.3)

where Φ is the osmotic coefficient (a nondimensional function of solute concentration and solid strain) which deviates from unity under nonideal physicochemical behavior. Therefore, a complete description of the physicochemical state of solvent and solute requires constitutive relations for Φ and the effective solubility $\tilde{\kappa} = \kappa/\gamma$.

2.3 Balance of Mass. It is also necessary to satisfy the balance of mass for each of the constituents. In the absence of chemical reactions the statement of balance of mass for constituent α reduces to

$$\frac{\partial \rho^{\alpha}}{\partial t} + \operatorname{div}(\rho^{\alpha} \mathbf{v}^{\alpha}) = 0$$
(2.4)

The apparent density ρ^{α} may be related to the true density ρ_T^{α} via $\rho^{\alpha} = \varphi^{\alpha} \rho_T^{\alpha}$, where φ^{α} is the volume fraction of α in the mixture. Due to mixture saturation (no voids), the volume fractions add up to unity; since the solute volume fraction is considered negligible $(\varphi^u \ll \varphi^s, \varphi^w)$, it follows that $\varphi^{\alpha} \approx \varphi^s + \varphi^w = 1$. Since ρ_T^{α} of an incompressible constituent is invariant in space and time, these relations may be combined to produce the mixture balance of mass relation,

$$\operatorname{div}(\mathbf{v}^s + \mathbf{w}) = 0 \tag{2.5}$$

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where $\mathbf{w} = \varphi^w (\mathbf{v}^w - \mathbf{v}^s)$ is the volumetric flux of solvent relative to the solid. The balance of mass for the solute may also be written as

$$\frac{\partial(\varphi^{w}c)}{\partial t} + \operatorname{div}(\mathbf{j} + \varphi^{w}c\mathbf{v}^{s}) = 0$$
(2.6)

where $\mathbf{j} = \varphi^w c(\mathbf{v}^u - \mathbf{v}^s)$ is the molar flux of solute relative to the solid. This relation is obtained by recognizing that the solute apparent density is related to its concentration (moles per solution volume) via $\rho^u = (1 - \varphi^s) Mc \approx \varphi^w Mc$. The mass balance for the solid matrix requires that $\rho^s J$ should remain constant, where $J = \det \mathbf{F}$ and $\mathbf{F} = \mathbf{I} + \operatorname{Grad} \mathbf{u}$ is the deformation gradient of the solid matrix. Therefore, since $\rho^s = \varphi^s \rho_T^s$ and ρ_T^s is invariant, it follows that

$$\varphi^s = \frac{\varphi_r^s}{J} \tag{2.7}$$

where φ_r^s is the solid volume fraction in the reference state.

2.4 Flux Relations. Inverting the momentum balance equations in (2.3), it is now possible to relate the solvent and solute fluxes to the driving forces according to

$$\mathbf{w} = -\tilde{\mathbf{k}} \cdot \left(\rho_T^w \operatorname{grad} \tilde{\mu}^w + Mc \frac{\mathbf{d}}{d_0} \cdot \operatorname{grad} \tilde{\mu}^u \right)$$

$$\mathbf{j} = \mathbf{d} \cdot \left(-\frac{M}{R\theta} \varphi^w c \operatorname{grad} \tilde{\mu}^u + \frac{c}{d_0} \mathbf{w} \right)$$
 (2.8)

where **d** is the solute diffusivity tensor in the mixture (solid + solution) and d_0 is its (isotropic) diffusivity in free solution. $\tilde{\mathbf{k}}$ is the second-order hydraulic permeability tensor of the solution (solvent + solute) through the porous solid matrix, which depends explicitly on concentration according to

$$\tilde{\mathbf{k}} = \left[\mathbf{k}^{-1} + \frac{R\theta c}{\varphi^w d_0} \left(\mathbf{I} - \frac{\mathbf{d}}{d_0}\right)\right]^{-1}$$
(2.9)

where \mathbf{k} represents the hydraulic permeability tensor of the solvent through the solid matrix. The permeability and diffusivity tensors are related to the diffusive drag tensors appearing in (2.2) according to

$$\mathbf{k} = (\varphi^w)^2 (\mathbf{f}^{ws})^{-1}$$

$$\mathbf{d}_0 = R\theta\varphi^w c(\mathbf{f}^{uw})^{-1} \equiv d_0 \mathbf{I}$$

$$\mathbf{d} = R\theta\varphi^w c(\mathbf{f}^{us} + \mathbf{f}^{uw})^{-1}$$
(2.10)

though these explicit relationships are not needed here since **k**, **d**, and d_0 may be directly specified in a particular analysis. Since the axiom of entropy inequality requires that the tensors $\mathbf{f}^{\alpha\beta}$ be positive semidefinite (see Appendix of [35]), it follows that d_0 must be greater than or equal to the largest eigenvalue of **d**. Constitutive relations are needed for these transport properties, which relate them to the solid matrix strain [7,34,35,53–55] and solute concentration. Note that the relations in (2.8) represent generalizations of Darcy's law for fluid permeation through porous media, and Fick's law for solute diffusion in porous media or free solution.

2.5 Continuous Variables. In principle, the objective of the finite element analysis is to solve for the three unknowns u, p, and c using the partial differential equations that enforce mixture momentum balance in (2.1), mixture mass balance in (2.5), and solute mass balance in (2.6). The remaining solvent and solute momentum balances in (2.8) and solid mass balance in (2.7) have been reduced to algebraic equations that may be substituted into the three partial differential equations as needed. Solving these equations requires the application of suitable boundary conditions that are consistent with mass, momentum and energy balances

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across boundary surfaces or interfaces. When defining boundaries or interfaces on the solid matrix, whose outward unit normal is **n**, mass and momentum balance relations demonstrate that the mixture traction $\mathbf{t} = \mathbf{T} \cdot \mathbf{n}$ and normal flux components $w_n = \mathbf{w} \cdot \mathbf{n}$ and $j_n = \mathbf{j} \cdot \mathbf{n}$ must be continuous across the interface [52,56]. Therefore, \mathbf{t}, w_n , and j_n may be prescribed as boundary conditions.

Combining momentum and energy balances across an interface also demonstrates that $\tilde{\mu}^w$ and $\tilde{\mu}^u$ must be continuous [52,57], implying that these mechanochemical potentials may be prescribed as boundary conditions. However, because of the arbitrariness of the reference states μ_0^w , μ_0^u , p_0 , and c_0 , and the illconditioning of the logarithm function in the limit of small solute concentration, the mechanochemical potentials do not represent practical choices for primary variables in a finite element implementation. An examination of (2.3) also shows that continuity of these potentials across an interface does not imply continuity of the fluid pressure p or solute concentration c. Therefore, pressure and concentration are also unsuitable as nodal variables in a finite element analysis and they must be replaced by alternative choices. Based on the similar reasoning presented by Sun et al. [17], an examination of the expressions in (2.3) shows that continuity may be enforced by using

$$\tilde{p} = p - R\theta \,\Phi c \tag{2.11}$$

$$\tilde{c} = \frac{c}{\tilde{\kappa}}$$

where \tilde{p} is the effective fluid pressure and \tilde{c} is the effective solute concentration in the mixture. Note that \tilde{p} represents that part of the fluid pressure which does not result from osmotic effects (since the term $R\theta\Phi c$ may be viewed as the osmotic pressure contribution to p); and \tilde{c} is a straightforward measure of the solute activity, since $a^u = \tilde{c}/c_0$. Therefore these alternative variables have clear physical meanings.

Since the unknowns are now given by \mathbf{u}, \tilde{p} , and \tilde{c} , the governing partial differential equations may be rewritten in the form

$$-\operatorname{grad}(\tilde{p} + R\theta\Phi\tilde{\kappa}\tilde{c}) + \operatorname{div}\mathbf{T}^{\mathbf{e}} = 0$$

$$\operatorname{div}(\mathbf{v}^{s} + \mathbf{w}) = 0$$

$$\frac{\partial(\varphi^{w}\tilde{\kappa}\tilde{c})}{\partial t} + \operatorname{div}(\mathbf{j} + \varphi^{w}\tilde{\kappa}\tilde{c}\mathbf{v}^{s}) = 0$$
(2.12)

where

$$\mathbf{w} = -\tilde{\mathbf{k}} \cdot \left(\operatorname{grad} \tilde{p} + R\theta \frac{\tilde{\kappa}}{d_0} \mathbf{d} \operatorname{grad} \tilde{c} \right)$$

$$\mathbf{j} = \tilde{\kappa} \mathbf{d} \cdot \left(-\varphi^w \operatorname{grad} \tilde{c} + \frac{\tilde{c}}{d_0} \mathbf{w} \right)$$

$$\tilde{\mathbf{k}} = \left[\mathbf{k}^{-1} + \frac{R\theta}{\varphi^w} \frac{\tilde{\kappa}\tilde{c}}{d_0} \left(\mathbf{I} - \frac{\mathbf{d}}{d_0} \right) \right]^{-1}$$

$$\varphi^w = 1 - \frac{\varphi_I^*}{I}$$
(2.13)

Constitutive equations are needed to relate \mathbf{T}^{e} , \mathbf{k} , d, d_{0} , $\tilde{\kappa}$, and Φ to the solid matrix strain and effective solute concentration.

2.6 Principle of Virtual Work. The virtual work integral for this problem is given by

$$\delta W = -\int_{b} \delta \mathbf{v} \cdot \operatorname{div} \mathbf{T} dv -\int_{b} \delta \tilde{p} \operatorname{div}(\mathbf{v}^{s} + \mathbf{w}) dv -\int_{b} \delta \tilde{c} \left[\frac{\partial (\varphi^{w} \tilde{\kappa} \tilde{c})}{\partial t} + \operatorname{div}(\mathbf{j} + \varphi^{w} \tilde{\kappa} \tilde{c} \mathbf{v}^{s}) \right] dv$$
(2.14)

where $\delta \mathbf{v}$ is the virtual velocity of the solid, $\delta \tilde{p}$ is the virtual effective fluid pressure, and $\delta \tilde{c}$ is the virtual molar energy of the solute; *b* represents the mixture domain in the spatial frame and *dv* is an elemental mixture volume in *b*. In the last integral of δW , note that

$$\frac{\partial(\varphi^{w}\tilde{\kappa}\tilde{c})}{\partial t} + \operatorname{div}(\varphi^{w}\tilde{\kappa}\tilde{c}\mathbf{v}^{s}) = \frac{1}{J}\frac{D^{s}}{Dt}(J\varphi^{w}\tilde{\kappa}\tilde{c})$$
(2.15)

where $D^s f/Dt \equiv \partial f/\partial t + \mathbf{v}^s \cdot \operatorname{grad} f$ is the material time derivative of a scalar function f in the spatial frame, following the solid. Similarly, note that div $\mathbf{v}^s = J^{-1}(D^s J/Dt)$. Using the divergence theorem, the virtual work integral may be separated into internal and external contributions $\delta W = \delta W_{\text{int}} - \delta W_{\text{ext}}$, where

$$\delta W_{\text{int}} = \int_{b} \mathbf{T} : \delta \mathbf{D}^{s} dv + \int_{b} \left(\mathbf{w} \cdot \operatorname{grad} \delta \tilde{p} - \frac{\delta \tilde{p}}{J} \frac{D^{s} J}{Dt} \right) dv + \int_{b} \left[\mathbf{j} \cdot \operatorname{grad} \delta \tilde{c} - \frac{\delta \tilde{c}}{J} \frac{D^{s}}{Dt} (J \varphi^{w} \tilde{\kappa} \tilde{c}) \right] dv$$
(2.16)
$$\delta W_{\text{ext}} = \int_{\partial b} (\delta \mathbf{v} \cdot \mathbf{t} + \delta \tilde{p} w_{n} + \delta \tilde{c} j_{n}) da$$

with δW_{ext} being evaluated on the domains boundary surface ∂b . In the first expression $\delta \mathbf{D}^s = (\operatorname{grad} \delta \mathbf{v} + \operatorname{grad}^T \delta \mathbf{v})/2$ represents the virtual solid rate of deformation.

To solve this nonlinear system using an iterative Newton scheme, the virtual work must be linearized at trial solutions, along increments in \mathbf{u}, \tilde{p} , and \tilde{c} ,

$$\delta W + D\delta W[\Delta \mathbf{u}] + D\delta W[\Delta \tilde{p}] + D\delta W[\Delta \tilde{c}] \approx 0$$
(2.17)

where, for any function f(q), $Df[\Delta q]$ represents the directional derivative of f along Δq [58]. To operate the directional derivative on the integrand of δW_{int} , it is first necessary to convert the integrals from the spatial to the material domain [58]:

$$\delta W_{\text{int}} = \int_{B} \mathbf{S} : \delta \mathbf{E} dV + \int_{B} \left(\mathbf{W} \cdot \text{Grad} \, \delta \tilde{p} - \delta \tilde{p} \, \frac{\partial J}{\partial t} \right) dV + \int_{B} \left[\mathbf{J} \cdot \text{Grad} \, \delta \tilde{c} - \delta \tilde{c} \, \frac{\partial}{\partial t} (J \, \varphi^{w} \, \tilde{\kappa} \tilde{c}) \right] dV$$
(2.18)

where B represents the mixture domain in the material frame, dV is an elemental mixture volume in B, and

$$\mathbf{S} = J\mathbf{F}^{-1} \cdot \mathbf{T} \cdot \mathbf{F}^{-T}, \quad \delta \dot{\mathbf{E}} = \mathbf{F}^T \cdot \delta \mathbf{D}^s \cdot \mathbf{F} \mathbf{W} = J\mathbf{F}^{-1} \cdot \mathbf{w}, \qquad \mathbf{J} = J\mathbf{F}^{-1} \cdot \mathbf{j}$$
 (2.19)

These relations show that the second Piola-Kirchhoff stress tensor **S**, and material flux vectors **W** and **J**, are, respectively, related to **T**, **w**, and **j** by the Piola transformations for tensors and vectors [58,59]. Substituting (2.13) into (2.19) produces

$$\mathbf{W} = -\tilde{\mathbf{K}} \cdot \left(\operatorname{Grad} \tilde{p} + R\theta \frac{\tilde{\kappa}}{d_0} J^{-1} \mathbf{C} \cdot \mathbf{D} \cdot \operatorname{Grad} \tilde{c} \right)$$

$$\mathbf{J} = \tilde{\kappa} \mathbf{D} \cdot \left(-\varphi^{w} \operatorname{Grad} \tilde{c} + \frac{\tilde{c}}{d_0} J^{-1} \mathbf{C} \cdot \mathbf{W} \right)$$
(2.20)

where $\mathbf{C} = \mathbf{F}^T \cdot \mathbf{F}$ is the right Cauchy-Green deformation tensor; $\tilde{\mathbf{K}}$ and \mathbf{D} are the material representations of the permeability and diffusivity tensors, related to $\tilde{\mathbf{k}}$ and \mathbf{d} via the Piola transformation,

$$\tilde{\mathbf{K}} = J\mathbf{F}^{-1} \cdot \tilde{\mathbf{k}} \cdot \mathbf{F}^{-T}$$

$$\mathbf{D} = J\mathbf{F}^{-1} \cdot \mathbf{d} \cdot \mathbf{F}^{-T}$$
(2.21)

The linearization of δW_{int} is rather involved and a summary of the resulting lengthy expressions is provided in Appendix A. In consideration of the dearth of experimental data relating $\tilde{\kappa}$ and Φ to the complete state of solid matrix strain (such as C), this implementation assumes that the dependence of these functions on the strain is restricted to a dependence on the relative volume $J = (\det C)^{1/2}$. Furthermore, it is assumed that the free solution diffusivity d_0 is independent of the strain.

The linearization of δW_{ext} is described in Appendix B. Following the linearization procedure, the resulting expressions may be discretized by nodally interpolating **u**, \tilde{p} , and \tilde{c} over finite elements, producing a set of equations in matrix form, as described in Appendix C.

2.7 Implementation. The formulation presented in this study is implemented in the FEBio open-source finite element code (FEBio, http://mrl.sci.utah.edu/software) by introducing an

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additional module dedicated to solute transport in deformable porous media. The code is written in the C+ + object-oriented programming language, which employs classes to describe data structures and associated functions. Classes are implemented to describe material functions for \mathbf{T}^{e} , \mathbf{k} , \mathbf{d} (and d_{0}), $\tilde{\kappa}$, and Φ , which allow the formulation of any desired constitutive relation for these functions of **C** and \tilde{c} , along with corresponding derivatives of these functions with respect to **C** and \tilde{c} . The implementation accepts essential boundary conditions on \mathbf{u} , \tilde{p} , and \tilde{c} , or natural boundary conditions on \mathbf{t} , w_n , and j_n ; initial conditions may also be specified for \tilde{p} and \tilde{c} . Analysis results for pressure and concentration may be displayed either as \tilde{p} and \tilde{c} , or as p and c by inverting the relations of (2.11).

3 Validations

To validate the numerical solutions from the finite element implementation, analyses were performed for which alternative solutions were available, either from analytical solutions or alternative numerical schemes. In most cases these solutions were limited to the range of infinitesimal strains; therefore additional illustrations are also provided where large deformations were induced. An additional purpose of these validations was to illustrate fundamental phenomena encompassed in this implementation, including standard Fickian diffusion problems, osmotically induced solid matrix deformations, effects of solute partitioning $(\tilde{\kappa} < 1)$, and solute concentration enhancement under deformational loading. Though the code was formulated to allow strain and concentration-dependent constitutive relations for \mathbf{k} , \mathbf{d} , Φ , and $\tilde{\kappa}$, only the simplest constitutive models (constant properties) were adopted for the purpose of numerical validations, since the primary concern of this study did not revolve around the specific form of such constitutive relations.

3.1 Fickian Diffusion Consider a cylindrical disk of a porous material, with radius *a* and height *h*, placed in a large, well-stirred bath containing a solute with concentration c^* . A cylindrical coordinate system (r, z) is employed to describe the axisymmetric geometry. The bottom of the disk (z = 0) rests on a rigid impermeable substrate. Over time, the solute diffuses from the bath into the porous disk, across its lateral (r = a) and top (z = h) surfaces. If the deformation of the solid matrix is negligible (if the matrix is rigid, or if the osmotic pressure difference produced by the solute concentration is negligible in comparison to the stiffness of the solid matrix), standard Fickian diffusion [1] takes place and the resulting solute concentration in the porous material is given by the axisymmetric solution to the two-dimensional diffusion equation:

$$c(r, z, t) = c^* \left\{ 1 - \frac{4}{\pi} \sum_{m=0}^{\infty} \sum_{n=0}^{\infty} \frac{(-1)^n}{\gamma_m J_1(\gamma_m) \left(n + \frac{1}{2}\right)} J_0\left(\gamma_m \frac{r}{a}\right) \right.$$

$$\left. \cos\left[\left(n + \frac{1}{2}\right) \pi \frac{z}{h}\right] \exp\left(-d\lambda_{mn}^2 t\right) \right\}$$

$$(3.1)$$

where d is the solute diffusivity in the porous medium, J_0 and J_1 are Bessel functions of the first kind, of order 0 and 1, respectively,

$$\lambda_{mn}^2 = \left[\left(n + \frac{1}{2} \right) \frac{\pi}{h} \right]^2 + \left(\frac{\gamma_m}{a} \right)^2 \tag{3.2}$$

and γ_m is the *m*th root of $J_0(\gamma)$.

A 3D finite element model of a quarter disk is shown in Fig. 1, with a = 1 mm and h = 1 mm. Mesh biasing was employed to capture narrow boundary layers in solute concentration near the lateral and top surfaces; the mesh employed 20 increments along r, 20 along z, and 20 along the circumferential direction, for a total of 8000 elements. The solid matrix was described by a neo-Hookean elastic solid with a Young's modulus of 10 MPa and

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Fig. 1 Finite element mesh for 2D axisymmetric Fickian diffusion

Poisson's ratio of 0.3. The diffusivity tensor was taken to be isotropic, $\mathbf{d} = d\mathbf{I}$, with $d = 10^{-3} \text{ mm}^2/\text{s}$; the free diffusivity was also prescribed to be $d_0 = d$. The hydraulic permeability was given by $\mathbf{k} = k\mathbf{I}$, with $k = 1 \text{ mm}^4/\text{N} \cdot \text{s}$. It was also assumed that $\tilde{\kappa} = 1$, $\Phi = 1$, and $\varphi_r^s = 0.2$ An external bath concentration of $c^* = 1$ mM was prescribed on the lateral and top surfaces; similarly, the ambient fluid pressure in the bath was assumed to be $p^* = 0$. Therefore, in the finite element model, the boundary conditions were $\mathbf{t} = \mathbf{0}$ and, according to (2.11), $\tilde{p} = -R\theta c^*$ and $\tilde{c} = c^*$. The initial pressure and concentration in the disk were, respectively, set to $\tilde{p} = 0$ and $\tilde{c} = 0$. For these conditions, the osmotic pressure difference between the bath and the disk never exceeded $R\theta c^* = 2.5 \times 10^{-3}$ MPa (given that $R = 8.314 \times 10^{-6} \text{ mJ/nmol} \cdot \text{K}$ and assuming that $\theta = 298 \text{ K}$), a value negligible relative to the solid matrix modulus. Consequently, only variations in concentration were significant in this analysis, consistent with the premise of classical Fickian diffusion. Surface plots of the analytical and finite element solutions for the solute concentration in the (r, z) domain, at two representative time points, verified agreement between the two solutions (Fig. 2).

3.2 Osmotic Loading of a Spherical Gel. Osmotic loading (sometimes called osmotic shock) represents the process of subjecting a porous medium to a change in ambient solute concentration. In a prior study, Albro et al. [5] reported experimental measurements for osmotic loading of spherical alginate gels immersed in a solution containing dextran molecules. Results showed the gel initially shrunk, before slowly recovering to a steady-state volume smaller than its initial (reference) value in a solute-free bath. Under the assumption of infinitesimal deformations of a linear isotropic elastic solid matrix, they provided a set of coupled, nonlinear partial differential equations to solve for the radial deformation and solute concentration in the gel. These equations were solved numerically using a finite difference scheme. Their solution was employed here for comparison with the finite element implementation.

A finite element model of one-eighth of a sphere, of radius a = 0.6 mm, was used for this analysis [Fig. 3(*a*)]. A uniform mesh distribution, with 40 elements, was employed along the radial direction, to match the finite difference mesh; a total of 7680 elements comprised the entire finite element mesh. For both solutions schemes, the following material properties were used: A solid matrix with a Young's modulus of 6×10^{-3} MPa and Pois-



Fig. 2 2D axisymmetric Fickian diffusion, showing finite element results (symbol) and analytical solution (surface), at two representative time steps: (a) t = 4.64 s; (b) t = 72.08 s.

son's ratio of 0; $\varphi_r^s = 0.04$, $\tilde{\kappa} = 1$, $\Phi = 1$; the permeability and diffusivity were isotropic, with $k = 8 \times 10^{-2} \text{ mm}^4/\text{N} \cdot \text{s}$, $d = 2.75 \times 10^{-4} \text{ mm}^2/\text{s}$, and $d_0 = 3.9 \times 10^{-4} \text{ mm}^2/\text{s}$.

For the comparison with the finite difference solution, an external bath concentration of $c^* = 6 \times 10^{-3}$ mM was prescribed on the spherical surface, with the ambient pressure set to $p^* = 0$. Thus, $\tilde{c} = c^*$ and $\tilde{p} = -R\theta c^*$, and the initial osmotic pressure difference $(1.5 \times 10^{-5} \text{ MPa})$ remained small in comparison to the Young's modulus, as required for the infinitesimal strain solution of the finite difference analysis. Results for the radial displacement *u* [Fig. 3(*b*)] and solute concentration *c* [Fig. 3(*c*)], as a function of the radial position *r* and selected times *t*, verified agreement between the two solutions.

To further illustrate the significant influence of osmotic loading and solubility on solid matrix deformation, the finite element analysis was also repeated with $c^* = 6$ mM and $\tilde{\kappa} = 0.986$, and the solid matrix was modeled as neo-Hookean under finite deformation. The resulting initial osmotic pressure difference $(R\theta c^* = 1.5 \times 10^{-2} \text{ MPa})$ was significantly greater than the Young's modulus of the solid matrix. Results from the finite element analysis demonstrate that the gel volume decreased considerably upon the application of the osmotic load (Fig. 4), due to the exudation of interstitial fluid. However, as the average solute concentration in the gel increased monotonically with solute influx, the solvent flux reversed direction and the gel volume recovered until reaching a steady state. The steady-state volume was smaller than the initial volume V_0 , because the steady-state solute

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Fig. 3 Osmotic loading of a spherical gel, under infinitesimal strain conditions, comparing the finite element solution (symbol) to the finite difference solution (solid curve) at representative time points. (*a*) Finite element geometry and mesh. (b) Radial displacement of the solid matrix. (*c*) Solute concentration.

concentration in the gel, $\tilde{\kappa}c^*$, did not match the external bath concentration, thereby maintaining a small osmotic pressure difference of magnitude $R\theta(1-\tilde{\kappa})c^* = 2.1 \times 10^{-4}$ MPa on the gel.

3.3 Concentration Enhancement Under Step Compression. Just as osmotic loading may cause matrix deformation, deformational loading may produce solute transport. Mauck et al. [38] performed a finite difference analysis which



Fig. 4 Relative volume V/V_0 and average effective solute concentration \tilde{c}_{avg} during osmotic loading of a spherical gel, under finite strain conditions. V/V_0 initially decreases as a result of solvent outflux; as the effective solute concentration in the gel slowly increases by diffusion from the bath, solvent flows back into the gel and V/V_0 recovers to a steady-state value less than unity (since $\tilde{\kappa}$ <1).

demonstrated that, for certain combinations of material properties, dynamic deformational loading of a cylindrical disk of a porous tissue or gel may pump solute from the external bath into the disk to achieve concentrations in excess of the value in the bath. This theoretical prediction was verified in subsequent experimental studies that examined dextran and transferrin uptake in agarose and cartilage disks [39,60], further validated with a direct comparison of experiments and theory [40].

The numerical scheme for solving the coupled partial differential equations for this infinitesimal strain analysis [38,40] was employed here for comparison with the finite element implementation. A finite element mesh of a quarter disk (radius 2 mm, height 2.3 mm) was used, with 500 uniform increments along the radial direction (to match the axisymmetric finite difference analysis), one increment along the height, and 12 increments along the circumference, for a total of 6000 elements in the mesh. The solid matrix was ascribed a Young's modulus of 0.43 MPa and Poisson's ratio of 0; the isotropic hydraulic permeability was $k = 3.6 \times 10^{-3} \text{ mm}^4/\text{N} \cdot \text{s}$; the solute diffusivities were $d = 1.2 \times 10^{-6} \text{ mm}^2/\text{s}$ in the disk and $d_0 = 3.7 \times 10^{-5} \text{ mm}^2/\text{s}$ in free solution; the remaining properties were $\varphi_r^s = 0.098$, $\tilde{\kappa} = 1$, and $\Phi = 1$. The analysis assumed that the disk was initially equilibrated in a bath with solute concentration c^* . Since $\tilde{\kappa} = 1$ in this problem, the initial effective concentration in the disk was $\tilde{c} = c^*$; assuming an ambient pressure $p^* = 0$, the initial effective pressure in the disk was $\tilde{p} = -R\theta c^*$ (the initial osmotic pressure difference between the bath and disk was zero). The bath concentration was not altered in this analysis. A uniform axial engineering strain ε_0 was prescribed across the disk, to simulate compression with frictionless, impermeable, smooth, loading platens.

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For comparison with the infinitesimal strain analysis employing the finite difference scheme, a value of $\varepsilon_0 = -0.001$ was prescribed over a short ramp time of 0.01 s, then maintained constant until a steady state response was achieved. Since the resulting solid matrix deformation was nearly indistinguishable from the analytical solution for unconfined compression of a biphasic material [61], only the solute concentration results are presented here. The radial distribution in solute concentration is displayed for various time points in Fig. 5, showing agreement between the finite element and finite difference solutions. The solute concentration was observed to spike initially at the radial edge of the disk; over time, solute concentration rose throughout the disk, before slowly subsiding back to the bath concentration at steady state.

To illustrate the response under larger deformations, the analysis was repeated with $\varepsilon_0 = -0.2$, all other parameters remaining the same. The radial profile of the solute concentration remained qualitatively similar to the infinitesimal strain response (not shown); however the magnitude of solute concentration increased considerably under this larger prescribed strain, as illustrated in the temporal response of the concentration averaged over the entire disk (Fig. 6).

4 Discussion

This study reports the finite element implementation of mechanochemical phenomena in deformable porous media undergoing finite deformation. Though prior custom-written finite element implementations of related frameworks have been presented in the literature, several have focused on the mechano-electrochemical phenomena associated with the transport of monovalent counterions in a charged solid matrix, generally under infinitesimal strains [15–20] and, in a few cases, under finite deformation [21]. Finite element studies of neutral solute transport in deformable porous media have also been formulated [50], but have mostly relied on iterative solving of thermal and poroelastic transport in commercial codes [26,27]; however, Sengers et al. [25] provided a custom-written implementation, valid under finite deformation, which also modeled transport and biosynthesis.

A primary distinction with all these prior studies is that the current implementation incorporated the concept of solubility (or solute partitioning), embodied in $\tilde{\kappa}$, and solute-solid frictional interactions, embodied in the nonzero term \mathbf{f}^{us} [or alternatively, in the explicit distinction between the solute diffusivity tensor **d** in the mixture versus $\mathbf{d}_0 = d_0 \mathbf{I}$ in free solution, see (2.10)]. Solute partitioning [3,4] is a well-recognized phenomenon in biological tissues and gels, which is manifested by a smaller solute concentration in the porous medium relative to the surrounding bath, under steady-state conditions. This partitioning may occur both as



Fig. 5 Concentration enhancement under a step, infinitesimal, compression of a disk. The radial distribution of the solute concentration, normalized to the bath concentration, is displayed at selected time points. Finite element results (thick black curves) are compared to a finite difference solution (thinner gray curves).



Fig. 6 Concentration enhancement under a step, finite, compression of a disk. The temporal response of the average solute concentration in the disk (normalized to the bath concentration c^*), shows an initial rise before slowly subsiding back to the initial, ambient concentration.

a result of steric volume exclusion from a fraction of the pore space as well as long-range electrostatic interactions [2], therefore the solubility strictly equals the partition coefficient only in the absence of long-range electrostatic interactions (neutral solute in charged matrix, charged solute in neutral matrix, or a neutral/neutral combination). Experimental findings have demonstrated that this partitioning may vary with the state of solid matrix strain [5,6] as well as solute concentration [44,45]. Furthermore, partitioning may also explain partial volume recovery responses, in gels [5] and cells [43] subjected to osmotic loading, as a result of passive transport mechanisms rather than active regulatory processes as usually assumed in cells [62]. Partial volume recovery was illustrated in the problem of osmotic loading of a spherical gel (Fig. 4), which reproduced prior experimental findings [5]. Therefore, a finite element framework that incorporates a strain and concentration-dependent solubility $\tilde{\kappa}$ provides a valuable extension of prior implementations that can extend the range of modeling features.

It is also well known experimentally that solute diffusivity in a porous medium is generally slower than in free solution [2] (also see the cartilage-related literature review in the study of Mauck et al. [38]). This reduction in diffusivity has long been physically attributed to the additional transport hindrance imparted by the solid matrix. Mauck et al. [38] explicitly accounted for nonzero frictional interaction between solute and solvent and showed that a dynamically loaded solid matrix may pump solute into the porous medium as a result of this solute-solid momentum exchange. These predictions were subsequently confirmed experimentally in a series of studies that provided direct evidence for this mechanism [39,40,60]. Therefore, solute-solid interactions represent an important mechanism, especially when the solid may undergo dynamic deformation, that should be incorporated into a general finite element framework of solute transport in deformable porous media. The phenomenon of solute concentration enhancement was illustrated in the problem of step compression of a disk initially equilibrated in a solute bath (Figs. 5 and 6).

A consequence of modeling a variety of phenomena associated with solute transport in deformable porous media is that the number of material functions needed to model the solute behavior increases. Thus, the current implementation required the specification of the diffusivity **d** in the mixture and d_0 in free solution, the solubility $\tilde{\kappa}$, and the osmotic coefficient Φ . This stands in contrast with classical diffusion problems where it is generally required to only specify a single diffusion coefficient. Of course, this simplified case could be reproduced in the current framework by letting $\mathbf{d} = d_0 \mathbf{I}$, $\tilde{\kappa} = 1$, and $\Phi = 1$, under the assumption of negligible hindrance between solute and solid, no steric volume exclusion, and ideal physicochemical behavior. Nevertheless, since these material functions may generally depend on the solid matrix strain

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and solute concentration, the derivation of the tangent stiffness matrix in the finite element implementation becomes considerably more involved, as suggested by the lengthy expressions presented in the Appendix. In the C++ code employed in the FEBio implementation, vector and tensor classes were employed to facilitate the computation of these lengthy expressions by providing lines of code that closely reproduce the corresponding analytical expressions, rather than dealing directly with the Cartesian components of the vectors and tensors.

The governing equations (2.1)–(2.9) reduce to the framework of the biphasic theory for modeling solid-fluid mixtures [63], by simply letting the solute concentration reduce to zero. In the biphasic theory there is no distinction between the fluid pressure p and the corresponding effective fluid pressure \tilde{p} , as noted from (2.11). Therefore this biphasic-solute framework is fully compatible with the simpler biphasic framework that uses u and p as nodal variables, and FEBio seamlessly integrates models that contain both types of materials, as well as purely elastic or viscoelastic solids.

The availability of the FEBio source code, and binaries for a variety of platforms, in the public domain, also facilitates the dissemination of this implementation as well as the addition of newly formulated constitutive relations by the research community at large. Though the analysis of solute transport and mechanochemical phenomena in porous media is a classical science, modern derivations and investigations are uncovering interesting mechanisms and behaviors that enhance our understanding of cell and tissue physiology. Investigators who need an easier pathway to formulate novel constitutive formulations for the behavior of solutes in deformable porous media may find that the FEBio platform provides a broad and suitable foundation for this purpose. Future extensions of FEBio may include the incorporation of external body forces (e.g., to model solute sedimentation in centrifuges), chemical reactions with the solid and solvent (including binding and release kinetics), and active transport (e.g., transport by molecular motors).

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Appendix A: Linearization of Internal Virtual Work

The virtual work integral δW_{int} in (2.18) may be linearized term by term along increments in $\Delta \mathbf{u}$, $\Delta \tilde{p}$, and $\Delta \tilde{c}$ using the general form

$$D\left(\int_{B} F dV\right) \left[\Delta q\right] = \int_{B} DF[\Delta q] \, dV = \int_{b} f dv \tag{A1}$$

For notational simplicity, the integral sign is omitted and the linearization of each term is presented in the form $DF[\Delta q] dV = f dv$.

A.1 Linearization Along Δu

The linearization of the first term in δW_{int} along $\Delta \mathbf{u}$ yields

$$D(\mathbf{S} : \delta \dot{\mathbf{E}}) [\Delta \mathbf{u}] dV = [\delta \mathbf{D}^s : \mathcal{C} : \Delta \varepsilon + \mathbf{T} : (\operatorname{grad}^T \Delta \mathbf{u} \cdot \operatorname{grad} \delta \mathbf{v})] du$$
(A2)

where C is the spatial elasticity tensor of the mixture (tensor dyadic products \otimes , $\underline{\otimes}$, and $\overline{\otimes}$ are described in [64]),

$$\mathcal{C} = \mathcal{C}^{e} - \left(\tilde{p} + R\theta\Phi\tilde{\kappa}\tilde{c}\right)\left(\mathbf{I}\otimes\mathbf{I} - 2\mathbf{I}\underline{\overline{\otimes}}\mathbf{I}\right) - R\theta\tilde{c}J\frac{\partial(\Phi\tilde{\kappa})}{\partial J}\mathbf{I}\otimes\mathbf{I} \quad (A3)$$

and C^e is the spatial elasticity tensor of the solid matrix,

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$$\mathcal{C}^{e} = J^{-1}(\mathbf{F}\underline{\otimes}\mathbf{F}) \quad : \quad 2\frac{\partial \mathbf{S}^{e}}{\partial \mathbf{C}} \quad : \quad \left(\mathbf{F}^{T}\underline{\otimes}\mathbf{F}^{T}\right) \tag{A4}$$

The linearization of the second term is

$$D(\mathbf{W} \cdot \operatorname{Grad} \delta \tilde{p}) [\Delta u] dV = \operatorname{grad} \delta \tilde{p} \cdot \mathbf{w}'_{u} dv$$
(A5)

where

$$\mathbf{w}_{u}^{\prime} \equiv J^{-1}\mathbf{F} \cdot D\mathbf{W}[\Delta \mathbf{u}] = -\left(\tilde{\mathcal{K}} : \Delta \varepsilon\right) \cdot \left(\operatorname{grad} \tilde{p} + R\theta \frac{\tilde{\kappa}}{d_{0}} \mathbf{d} \cdot \operatorname{grad} \tilde{c}\right) \\ - \frac{R\theta}{d_{0}} \tilde{\mathbf{k}} \cdot \left[J^{2} \frac{\partial \left(J^{-1}\tilde{\kappa}\right)}{\partial J} \left(\operatorname{div} \Delta \mathbf{u}\right) \mathbf{I} + 2\tilde{\kappa}\Delta\varepsilon\right] \cdot \mathbf{d} \cdot \operatorname{grad} \tilde{c}$$

$$-\tilde{\kappa} \frac{R\theta}{d_{0}} \tilde{\mathbf{k}} \cdot \left(\mathcal{D} : \Delta\varepsilon\right) \cdot \operatorname{grad} \tilde{c}$$
(A6)

with

$$\widetilde{\mathcal{K}} = J^{-1}(\mathbf{F} \underline{\otimes} \mathbf{F}) : 2 \frac{\partial \mathbf{K}}{\partial \mathbf{C}} : (\mathbf{F}^T \underline{\otimes} \mathbf{F}^T)
\mathcal{D} = J^{-1}(\mathbf{F} \underline{\otimes} \mathbf{F}) : 2 \frac{\partial \mathbf{D}}{\partial \mathbf{C}} : (\mathbf{F}^T \underline{\otimes} \mathbf{F}^T)$$
(A7)

representing the spatial tangents, with respect to the strain, of the effective permeability and solute diffusivity, respectively. These fourth-order tensors exhibit minor symmetries but not major symmetry, as described in our recent study [35]. Since $\tilde{\mathbf{K}}$ is given by substituting (2.13)₃ into (2.21)₁, the evaluation of $\tilde{\mathcal{K}}$ is rather involved and it can be shown that

$$\tilde{\mathcal{K}} = 2(\tilde{\mathbf{k}} \otimes \mathbf{I} - 2\tilde{\mathbf{k}} \overline{\otimes} \mathbf{I}) - (\tilde{\mathbf{k}} \underline{\otimes} \tilde{\mathbf{k}}) : \mathcal{G}$$
(A8)

where

$$\mathcal{G} = 2(\mathbf{k}^{-1} \otimes \mathbf{I} - 2\mathbf{k}^{-1} \overline{\otimes} \mathbf{I}) - (\mathbf{k}^{-1} \underline{\otimes} \mathbf{k}^{-1}) : \mathcal{K} + \frac{R\theta\bar{c}}{d_0} J \frac{\partial}{\partial J} \left(\frac{\bar{\kappa}}{\phi^w}\right) \left(\mathbf{I} - \frac{\mathbf{d}}{d_0}\right) \otimes \mathbf{I} + \frac{R\theta\bar{c}}{d_0} \frac{\bar{\kappa}}{\phi^w} \left(\mathbf{I} \otimes \mathbf{I} - 2\mathbf{I} \underline{\otimes} \mathbf{I} - \frac{1}{d_0} \mathcal{D}\right)$$
(A9)

and

$$\mathcal{K} = J^{-1}(\mathbf{F}\underline{\otimes}\mathbf{F}) : 2\frac{\partial \mathbf{K}}{\partial \mathbf{C}} : (\mathbf{F}^T\underline{\otimes}\mathbf{F}^T)$$
(A10)

The next term in δW_{int} linearizes to

$$-D\left(\delta \tilde{p}\frac{\partial J}{\partial t}\right) [\Delta \mathbf{u}] dV = -\delta \tilde{p}\left(\left[(\operatorname{div} \Delta \mathbf{u}) \mathbf{I} - \operatorname{grad}^{T} \Delta \mathbf{u}\right]\right)$$

: grad $\mathbf{v}^{s} + \operatorname{div} \overline{\Delta \mathbf{u}} dv$ (A11)

where

$$\frac{1}{\Delta \mathbf{u}} \approx \frac{\Delta \mathbf{u}}{\Delta t}$$
 (A12)

and Δt represents the time increment relative to the previous time point. The next term is given by

$$D(\mathbf{J} \cdot \operatorname{Grad} \delta \tilde{c}) \left[\Delta \mathbf{u} \right] dV = \operatorname{grad} \delta \tilde{c} \cdot \mathbf{j}'_{u} dv \tag{A13}$$

where

$$\mathbf{j}'_{u} \equiv J^{-1}\mathbf{F} \cdot D\mathbf{J}[\Delta \mathbf{u}] = \left[J \frac{\partial \tilde{\kappa}}{\partial J} (\operatorname{div} \Delta \mathbf{u}) \, \mathbf{d} + \tilde{\kappa} \mathcal{D} : \Delta \varepsilon \right] \cdot \left(-\varphi^{w} \operatorname{grad} \tilde{c} + \frac{\tilde{c}}{d_{0}} \mathbf{w} \right) \\ + \tilde{\kappa} \mathbf{d} \cdot \left\{ -\varphi^{s} (\operatorname{div} \Delta \mathbf{u}) \operatorname{grad} \tilde{c} + \frac{\tilde{c}}{d_{0}} [2\Delta \varepsilon - (\operatorname{div} \Delta \mathbf{u}) \mathbf{I}] \cdot \mathbf{w} \right\} \\ + \tilde{\kappa} \frac{\tilde{c}}{d_{0}} \mathbf{d} \cdot \mathbf{w}'_{u}$$

(A14)

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The last term is

$$-D\left[\varphi \tilde{c} \frac{\partial (J \varphi^{w} \tilde{\kappa} \tilde{c})}{\partial t}\right] [\Delta u] dV = -\delta \tilde{c} \left\{ \tilde{c} \frac{\partial J}{\partial t} \frac{\partial^{2} (J \varphi^{w} \tilde{\kappa})}{\partial J^{2}} + \frac{\partial \tilde{c}}{\partial t} \frac{\partial}{\partial J} \left[J \varphi^{w} \left(\tilde{\kappa} + \frac{\partial \tilde{\kappa}}{\partial \tilde{c}} \tilde{c} \right) \right] \right\}$$

div $\Delta \mathbf{u} dv - \delta \tilde{c} \frac{\partial (J \varphi^{w} \tilde{\kappa})}{\partial J} \left\{ \left[(\operatorname{div} \Delta \mathbf{u}) \mathbf{I} - \operatorname{grad}^{T} \Delta \mathbf{u} \right] : \operatorname{grad} \mathbf{v}^{s} + \operatorname{div} \overline{\Delta \mathbf{u}} \right\} \tilde{c} dv$
(A15)

A.2 Linearization Along $\Delta \tilde{p}$

 $(0, \tau)$

The linearization of the various terms in δW_{int} along $\Delta \tilde{p}$ yields

$$D(\mathbf{S} : \delta \dot{\mathbf{E}}) [\Delta \tilde{p}] dV = -\Delta \tilde{p} \operatorname{div} \delta \mathbf{v} dv \qquad (A16)$$

$$D\left(\mathbf{W} \cdot \operatorname{Grad} \delta \tilde{p} - \delta \tilde{p} \frac{\partial J}{\partial t}\right) [\Delta \tilde{p}] dV = -\operatorname{grad} \delta \tilde{p} \cdot \tilde{\mathbf{k}} \cdot \operatorname{grad} \Delta \tilde{p} dv \quad (A17)$$
$$D\left[\mathbf{J} \cdot \operatorname{Grad} \delta \tilde{c} - \delta \tilde{c} \frac{\partial (J \varphi^{w} \tilde{\kappa} \tilde{c})}{\partial t}\right] [\Delta \tilde{p}] dV$$
$$= -\frac{\tilde{\kappa} \tilde{c}}{d_{0}} \operatorname{grad} \delta \tilde{c} \cdot \mathbf{d} \cdot \tilde{\mathbf{k}} \cdot \operatorname{grad} \Delta \tilde{p} dv \quad (A18)$$

A.3 Linearization Along $\Delta \tilde{c}$

The linearization of the first term in δW_{int} along $\Delta \tilde{c}$ yields

$$D(\mathbf{S} : \delta \dot{\mathbf{E}}) [\Delta \tilde{c}] dV = \Delta \tilde{c} \left[\mathbf{T}'_{c} : \delta \mathbf{D} - R\theta \frac{\partial (\Phi \tilde{\kappa} \tilde{c})}{\partial \tilde{c}} \operatorname{div} \delta \mathbf{v} \right] dv$$
(A19)

where

$$\mathbf{T}_{c}^{\prime} = J^{-1}\mathbf{F} \cdot \frac{\partial \mathbf{S}^{e}}{\partial \tilde{c}} \cdot \mathbf{F}^{T}$$
(A20)

represents the spatial tangent of the stress with respect to the effective concentration. The next term is

$$D(\mathbf{W} \cdot \operatorname{Grad} \delta \tilde{p}) \left[\Delta \tilde{c}\right] dV = \operatorname{grad} \delta \tilde{p} \cdot \mathbf{w}_{c}' dv \qquad (A21)$$

where

$$\mathbf{w}_{c}^{\prime} \equiv J^{-1}\mathbf{F} \cdot D\mathbf{W}[\Delta \tilde{c}]
= -\Delta \tilde{c} \tilde{\mathbf{k}}_{c}^{\prime} \cdot \left(\operatorname{grad} \tilde{p} + R\theta \frac{\tilde{\kappa}}{d_{0}} \mathbf{d} \cdot \operatorname{grad} \tilde{c} \right)
- R\theta \tilde{\mathbf{k}} \cdot \left\{ \Delta \tilde{c} \left[\frac{\partial}{\partial \tilde{c}} \left(\frac{\tilde{\kappa}}{d_{0}} \right) \mathbf{d} + \frac{\tilde{\kappa}}{d_{0}} \mathbf{d}_{c}^{\prime} \right] \cdot \operatorname{grad} \tilde{c} + \frac{\tilde{\kappa}}{d_{0}} \mathbf{d} \cdot \operatorname{grad} \Delta \tilde{c} \right\}$$
(A22)

and

$$\tilde{\mathbf{k}}_{c}' = J^{-1}\mathbf{F} \cdot \frac{\partial \mathbf{K}}{\partial \tilde{c}} \cdot \mathbf{F}^{T}$$
(A23)

is the spatial tangent of the effective hydraulic permeability with respect to the effective concentration. The next term reduces to

$$-D\left(\delta\tilde{p}\frac{\partial J}{\partial t}\right)\left[\Delta\tilde{c}\right]dV = 0 \tag{A24}$$

The following term is

$$D(J \operatorname{Grad} \delta \tilde{c}) [\Delta \tilde{c}] dV = \operatorname{grad} \delta \tilde{c} \cdot \mathbf{j}_c' dv \qquad (A25)$$

where

$$\mathbf{j}_{c}^{\prime} \equiv J^{-1}\mathbf{F} \cdot D\mathbf{j}_{0}[\Delta \tilde{c}]
= \Delta \tilde{c} \left(\frac{\partial \tilde{\kappa}}{\partial \tilde{c}} \mathbf{d} + \tilde{\kappa} \mathbf{d}_{c}^{\prime}\right) \cdot \left(-\varphi^{w} \operatorname{grad} \tilde{c} + \frac{\tilde{c}}{d_{0}} \mathbf{w}\right)$$

$$-\varphi^{w} \tilde{\kappa} \mathbf{d} \cdot \operatorname{grad} \Delta \tilde{c} + \tilde{\kappa} \frac{\tilde{c}}{d_{0}} \mathbf{d} \cdot \mathbf{w}_{c}^{\prime}$$
(A26)

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and

$$\mathbf{d}_{c}' = J^{-1}\mathbf{F} \cdot \frac{\partial \mathbf{D}}{\partial \tilde{c}} \cdot \mathbf{F}^{T}$$
(A27)

is the spatial tangent of the diffusivity with respect to the effective concentration. The last term is

 $-D\left[\frac{\partial (J\phi^{w}\bar{\kappa}\bar{c})}{\partial t}\delta\bar{c}\right] [\Delta\bar{c}] dV = -\delta\bar{c}\frac{1}{J}\frac{\partial (J\phi^{w})}{\partial t}\frac{\partial (\bar{\kappa}\bar{c})}{\partial\bar{c}}\Delta\bar{c}dv$ $-\delta\bar{c}\phi^{w}\left[\frac{\partial^{2}(\bar{\kappa}\bar{c})}{\partial\bar{c}^{2}}\Delta\bar{c}\frac{\partial\bar{c}}{\partial t} + \frac{\partial (\bar{\kappa}\bar{c})}{\partial\bar{c}}\overline{\Delta}\bar{c}\right]dv$ (A28) $-\delta\bar{c}\phi^{w}\frac{\partial I}{\partial\bar{c}}\partial\bar{c}(\bar{c}\frac{\partial\bar{\kappa}}{\partial I})\Delta\bar{c}dv$

where

$$\frac{1}{\Delta \tilde{c}} \approx \frac{\Delta \tilde{c}}{\Delta t}$$
 (A29)

Appendix B: Linearization of External Virtual Work

The linearization of δW_{ext} in (2.16) depends on whether natural boundary conditions are prescribed as area densities or total net values over an area. Thus, in the case when *tda* (net force), $w_n da$ (net volumetric flow rate), or $j_n da$ (net molar flow rate) are prescribed over the elemental area da, there is no variation in δW_{ext} and it follows that $D\delta W_{\text{ext}} = 0$. Alternatively, in the case when **t**, w_n , or j_n are prescribed, the linearization may be performed by evaluating the integral in the parametric space of the boundary surface ∂b , with parametric coordinates (η^1, η^2) . Accordingly, for a point $\mathbf{x}(\eta^1, \eta^2)$ on ∂b , surface tangents (covariant basis vectors) are given by

$$\mathbf{g}_{\alpha} = \frac{\partial \mathbf{x}}{\partial \eta^{\alpha}} \quad (\alpha = 1, 2)$$
 (B1)

and the outward unit normal is

$$\mathbf{n} = \frac{\mathbf{g}_1 \times \mathbf{g}_2}{|\mathbf{g}_1 \times \mathbf{g}_2|} \tag{B2}$$

The elemental area on ∂b is $da = |\mathbf{g}_1 \times \mathbf{g}_2| d\eta^1 d\eta^2$. Consequently, the external virtual work integral may be rewritten as

$$\delta W_{\text{ext}} = \int_{\partial b} (\delta \mathbf{v} \cdot \mathbf{t} + \delta \tilde{p} w_n + \delta \tilde{c} j_n) |\mathbf{g}_1 \times \mathbf{g}_2| \, d\eta^1 d\eta^2 \qquad (B3)$$

The directional derivative of δW_{ext} may then be applied directly to its integrand, since the parametric space is invariant [58].

If we restrict traction boundary conditions to the special case of normal tractions, then $\mathbf{t} = t_n \mathbf{n}$ where t_n is the prescribed normal traction component. Then it can be shown that the linearization of δW_{ext} along $\Delta \mathbf{u}$ produces

$$D(\delta W_{\text{ext}}) [\Delta \mathbf{u}] = \int_{\partial b} (t_n \delta \mathbf{v} + w_n \delta \tilde{p} \mathbf{n} + j_n \delta \tilde{c} \mathbf{n}) \left(\frac{\partial \Delta \mathbf{u}}{\partial \eta^1} \times \mathbf{g}_2 + \mathbf{g}_1 \times \frac{\partial \Delta \mathbf{u}}{\partial \eta^2} \right) d\eta^1 d\eta^2$$
(B4)

The linearizations along $\Delta \tilde{p}$ and $\Delta \tilde{c}$ reduce to zero, $D(\delta W_{\text{ext}}) [\Delta \tilde{p}] = 0$ and $D(\delta W_{\text{ext}}) [\Delta \tilde{c}] = 0$.

Appendix C: Discretization

To discretize the virtual work relations, let

$$\delta \mathbf{v} = \sum_{a=1}^{m} N_a \delta \mathbf{v}_a, \quad \Delta \mathbf{u} = \sum_{b=1}^{m} N_b \Delta \mathbf{u}_b$$

$$\delta \tilde{p} = \sum_{a=1}^{m} N_a \delta \tilde{p}_a, \quad \Delta \tilde{p} = \sum_{b=1}^{m} N_b \Delta \tilde{p}_b$$

$$\delta \tilde{c} = \sum_{a=1}^{m} N_a \delta \tilde{c}_a, \quad \Delta \tilde{c} = \sum_{b=1}^{m} N_b \Delta \tilde{c}_b$$

(C1)

where N_a represents the interpolation functions over an element, $\delta \mathbf{v}_a, \delta \tilde{p}_a, \delta \tilde{c}_a, \Delta \mathbf{u}_a, \Delta \tilde{p}_a$, and $\Delta \tilde{c}_a$, respectively, represent the nodal values of δv , $\delta \tilde{p}$, $\delta \tilde{c}$, Δu , $\Delta \tilde{p}$, and $\Delta \tilde{c}$; *m* is the number of nodes in an element.

The discretized form of δW_{int} in (2.16) may be written as

$$\delta W_{\text{int}} = \sum_{e=1}^{n_e} \sum_{k=1}^{n_{\text{int}}^{(e)}} W_k J_\eta \sum_{a=1}^{m} [\delta \mathbf{v}_a \quad \delta \tilde{p}_a \quad \delta \tilde{c}_a] \cdot \begin{bmatrix} \mathbf{r}_a^u \\ r_a^p \\ r_a^c \end{bmatrix}$$
(C2)

where n_e is the number of elements in b, $n_{int}^{(e)}$ is the number of integration points in the *e*th element, W_k is the quadrature weight associated with the *k*th integration point, and J_{η} is the Jacobian of the transformation from the current spatial configuration to the parametric space of the element. In the above expression,

$$\mathbf{r}_{a}^{\mu} = \mathbf{T} \cdot \operatorname{grad} N_{a}$$

$$r_{a}^{p} = \mathbf{w} \cdot \operatorname{grad} N_{a} - N_{a} \frac{1}{J} \frac{\partial J}{\partial t}$$

$$r_{a}^{c} = \mathbf{j} \cdot \operatorname{grad} N_{a} - N_{a} \frac{1}{J} \frac{\partial J}{\partial t} (J \varphi^{w} \tilde{\kappa} \tilde{c})$$
(C3)

and it is understood that J_{η} , \mathbf{r}_{a}^{μ} , r_{a}^{p} , and r_{a}^{c} are evaluated at the parametric coordinates of the *k*th integration point. Since the parametric space is invariant, time derivatives are evaluated in a material frame; for example, the time derivative $D^{s}J(x, t)/Dt$ appearing in (2.16) becomes $\partial J(\eta_{k}, t)/\partial t$ when evaluated at the parametric coordinates η_{k} of the *k*th integration point.

Similarly, the discretized form of $D\delta W_{int} = D\delta W_{int}[\Delta \mathbf{u}] + D\delta W_{int}[\Delta \tilde{\rho}] + D\delta W_{int}[\Delta \tilde{c}]$ may be written as

$$D\delta W_{\text{int}} = \sum_{e=1}^{n_e} \sum_{k=1}^{n_{\text{int}}^{(e)}} W_k J_\eta \sum_{a=1}^{m} \sum_{b=1}^{m} [\delta \mathbf{v}_a \ \delta \tilde{p}_a \ \delta \tilde{c}_a] \cdot \begin{bmatrix} \mathbf{K}_{ab}^{ua} \ \mathbf{k}_{ab}^{up} \ \mathbf{k}_{ab}^{uc} \\ \mathbf{k}_{ab}^{pa} \ \mathbf{k}_{ab}^{pp} \ \mathbf{k}_{ab}^{pc} \\ \mathbf{k}_{ab}^{cc} \ \mathbf{k}_{ab}^{pp} \ \mathbf{k}_{ab}^{cc} \end{bmatrix} \cdot \begin{bmatrix} \Delta \mathbf{u}_b \\ \Delta \tilde{p}_b \\ \Delta \tilde{c}_b \end{bmatrix}$$
(C4)

where the terms in the first column are the discretized form of the linearization along Δu :

$$\mathbf{K}_{ab}^{uu} = \operatorname{grad} N_a \cdot \mathcal{C} \cdot \operatorname{grad} N_b + \left(\operatorname{grad} N_a \cdot \mathbf{T} \cdot \operatorname{grad} N_b\right) \mathbf{I} \qquad (\mathbf{C5})$$

$$\mathbf{k}_{ab}^{pu} = \left(\mathbf{w}_{b}^{u}\right)^{T} \cdot \operatorname{grad} N_{a} + N_{a} \mathbf{q}_{b}^{pu}$$
(C6)

$$\mathbf{k}_{ab}^{cu} = \left(\mathbf{j}_{b}^{u}\right)^{T} \cdot \operatorname{grad} N_{a} + N_{a} \mathbf{q}_{b}^{cu}$$
(C7)

where

$$\mathbf{w}_{b}^{u} = -\left(\operatorname{grad}\tilde{p} + R\theta\frac{\tilde{\kappa}}{d_{0}}\mathbf{d}\cdot\operatorname{grad}\tilde{c}\right)\tilde{\mathcal{K}}\cdot\operatorname{grad}N_{b}$$
$$-\frac{R\theta}{d_{0}}\left[J^{2}\frac{\partial(J^{-1}\tilde{\kappa})}{\partial J}\left(\tilde{\mathbf{k}}\cdot\mathbf{d}\cdot\operatorname{grad}\tilde{c}\right)\otimes\operatorname{grad}N_{b} + 2\tilde{\kappa}(\operatorname{grad}N_{b}\cdot\mathbf{d}\cdot\operatorname{grad}\tilde{c})\tilde{\mathbf{k}}\right]$$
$$-\tilde{\kappa}\frac{R\theta}{d_{0}}\tilde{\mathbf{k}}(\operatorname{grad}\tilde{c}\cdot\mathcal{D}\cdot\operatorname{grad}N_{b})$$
(C8)

$$\begin{aligned} \mathbf{j}_{b}^{u} &= J_{\partial I}^{\underline{\delta} \overline{k}} \Big[\mathbf{d} \cdot \Big(-\varphi^{w} \operatorname{grad} \widetilde{c} + \frac{\widetilde{c}}{d_{0}} \mathbf{w} \Big) \Big] \otimes \operatorname{grad} N_{b} + \widetilde{\kappa} \Big\{ -\varphi^{s} (\mathbf{d} \cdot \operatorname{grad} \widetilde{c}) \otimes \operatorname{grad} N_{b} + \frac{\widetilde{c}}{d_{0}} [2(\operatorname{grad} N_{b} \cdot \mathbf{w}) \mathbf{d} - (\mathbf{d} \cdot \mathbf{w}) \otimes \operatorname{grad} N_{b}] \Big\} \\ &+ \widetilde{\kappa} \Big\{ -\varphi^{s} (\mathbf{d} \cdot \operatorname{grad} \widetilde{c}) \otimes \operatorname{grad} N_{b} + \frac{\widetilde{c}}{d_{0}} [2(\operatorname{grad} N_{b} \cdot \mathbf{w}) \mathbf{d} - (\mathbf{d} \cdot \mathbf{w}) \otimes \operatorname{grad} N_{b}] \Big\} \end{aligned}$$

$$\mathbf{q}_{b}^{pu} = -\left(\operatorname{div} \mathbf{v}^{s} + \frac{1}{\Delta t}\right)\operatorname{grad} N_{b} - \operatorname{grad}^{T} \mathbf{v}^{s} \cdot \operatorname{grad} N_{b} \qquad (C10)$$

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$$\mathbf{q}_{b}^{cu} = -\left\{\tilde{c}\frac{\partial J}{\partial t}\frac{\partial^{2}(J\varphi^{w}\tilde{\kappa})}{\partial J^{2}} + \frac{\partial \tilde{c}}{\partial t}\frac{\partial}{\partial J}\left[J\varphi^{w}\left(\tilde{\kappa} + \frac{\partial \tilde{\kappa}}{\partial \tilde{c}}\tilde{c}\right)\right]\right\}\operatorname{grad} N_{b} + \tilde{c}\frac{\partial (J\varphi^{w}\tilde{\kappa})}{\partial J}\mathbf{q}_{b}^{pu}$$
(C11)

The terms in the second column of the stiffness matrix in (C4) are the discretized form of the linearization along $\Delta \tilde{\rho}$:

$$\mathbf{k}_{ab}^{up} = -N_b \operatorname{grad} N_a, \tag{C12}$$

$$k_{ab}^{pp} = -\operatorname{grad} N_a \cdot \mathbf{k} \cdot \operatorname{grad} N_b \tag{C13}$$

$$k_{ab}^{cp} = -\frac{\tilde{\kappa}\tilde{c}}{d_0}\operatorname{grad} N_a \mathbf{d} \cdot \tilde{\mathbf{k}} \cdot \operatorname{grad} N_b \tag{C14}$$

The terms in the third column of the stiffness matrix in (C4) are the discretized form of the linearization along $\Delta \tilde{c}$:

$$\mathbf{k}_{ab}^{uc} = N_b \left(\mathbf{T}_c' \cdot \operatorname{grad} N_a - R\theta \frac{\partial (\Phi \tilde{\kappa} \tilde{c})}{\partial \tilde{c}} \operatorname{grad} N_a \right)$$
(C15)

$$k_{ab}^{pc} = \operatorname{grad} N_a \cdot \mathbf{w}_b^c \tag{C16}$$

$$k_{ab}^{cc} = \operatorname{grad} N_a \cdot \mathbf{j}_b^c + N_a q_b^c \tag{C17}$$

where

$$\mathbf{w}_{b}^{c} = -N_{b}\tilde{\mathbf{k}}_{c}^{\prime} \cdot \left(\operatorname{grad}\tilde{p} + R\theta \frac{\tilde{\kappa}}{d_{0}}\mathbf{d} \cdot \operatorname{grad}\tilde{c}\right) -R\theta\tilde{\mathbf{k}} \cdot \left\{N_{b}\left[\frac{\partial}{\partial\tilde{c}}\left(\frac{\tilde{\kappa}}{d_{0}}\right)\mathbf{d} + \frac{\tilde{\kappa}}{d_{0}}\mathbf{d}_{c}^{\prime}\right] \cdot \operatorname{grad}\tilde{c} + \frac{\tilde{\kappa}}{d_{0}}\mathbf{d} \cdot \operatorname{grad}N_{b}\right\}$$
(C18)

$$\mathbf{j}_{b}^{c} = N_{b} \left(\frac{\partial \tilde{\kappa}}{\partial \tilde{c}} \mathbf{d} + \tilde{\kappa} \mathbf{d}_{c}^{\prime} \right) \cdot \left(-\varphi^{w} \operatorname{grad} \tilde{c} + \frac{\tilde{c}}{d_{0}} \mathbf{w} \right) \\ + \tilde{\kappa} \mathbf{d} \cdot \left(-\varphi^{w} \operatorname{grad} N_{b} + \frac{\tilde{c}}{\tilde{c}} \mathbf{w}_{b}^{c} \right)$$
(C19)

$$q_b^c = -N_b \frac{1}{J} \frac{\partial (J\phi^w)}{\partial t} \frac{\partial (\tilde{\kappa}\tilde{c})}{\partial \tilde{c}} -N_b \phi^w \left[\frac{\partial^2 (\tilde{\kappa}\tilde{c})}{\partial \tilde{c}^2} \frac{\partial \tilde{c}}{\partial t} + \frac{1}{\Delta t} \frac{\partial (\tilde{\kappa}\tilde{c})}{\partial \tilde{c}} + \frac{\partial J}{\partial t} \frac{\partial}{\partial \tilde{c}} \left(\tilde{c} \frac{\partial \tilde{\kappa}}{\partial J} \right) \right]$$
(C20)

The discretization of δW_{ext} in (B3) has the form

$$\delta W_{\text{ext}} = \sum_{e=1}^{n_e} \sum_{k=1}^{n_{\text{int}}^{(e)}} W_k J_\eta \sum_{a=1}^{m} [\delta \mathbf{v}_a \quad \delta \tilde{p}_a \quad \delta \tilde{c}_a] \cdot \begin{bmatrix} N_a t_n \mathbf{n} \\ N_a w_n \\ N_a j_n \end{bmatrix}$$
(C21)

where $J_{\eta} = |\mathbf{g}_1 \times \mathbf{g}_2|$. The summation is performed over all surface elements on which these boundary conditions are prescribed. The discretization of $-D\delta W_{\text{ext}}$ has the form

$$-D\delta W_{\text{ext}} = \sum_{e=1}^{n_e} \sum_{k=1}^{n_{\text{int}}^{(e)}} W_k J_\eta \sum_{a=1}^{m} \sum_{b=1}^{m} [\delta \mathbf{v}_a \ \delta \tilde{p}_a \ \delta \tilde{c}_a] \cdot \begin{bmatrix} \mathbf{K}_{ab}^{uu} \ 0 \ 0 \\ \mathbf{k}_{ab}^{uu} \ 0 \ 0 \\ \mathbf{k}_{ab}^{cu} \ 0 \ 0 \end{bmatrix} \cdot \begin{bmatrix} \Delta \mathbf{u}_b \\ \Delta \tilde{p}_b \\ \Delta \tilde{c}_b \end{bmatrix}$$
(C22)

where

$$\begin{aligned} \mathbf{K}_{ab}^{uu} &= t_n N_a \mathcal{A} \Big\{ \frac{\partial N_b}{\partial \eta^1} \mathbf{g}_2 - \frac{\partial N_b}{\partial \eta^2} \mathbf{g}_1 \Big\} \\ \mathbf{k}_{ab}^{pu} &= -w_n N_a \mathcal{A} \Big\{ \frac{\partial N_b}{\partial \eta^1} \mathbf{g}_2 - \frac{\partial N_b}{\partial \eta^2} \mathbf{g}_1 \Big\} \cdot \mathbf{n} \\ \mathbf{k}_{ab}^{cu} &= -j_n N_a \mathcal{A} \Big\{ \frac{\partial N_b}{\partial \eta^1} \mathbf{g}_2 - \frac{\partial N_b}{\partial \eta^2} \mathbf{g}_1 \Big\} \cdot \mathbf{n} \end{aligned}$$
(C23)

In this expression, $\mathcal{A}\{v\}$ is the antisymmetric tensor whose dual vector is v (such that $\mathcal{A}\{v\} \cdot q = v \times q$ for any vector q).

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