Finite element modeling of hydrogel chemo-mechanics in Abaqus/Standard through user element (UEL) subroutine interface

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This chapter contains the details of finite element implementation of the neutral hydrogel model as a user-defined element subroutine (UEL) in Abaqus/Standard. The coupled chemo-mechanical theory, constitutive model, and finite element implementation are similar to those proposed by Chester and co-workers in a series of papers (Chester & Anand, 2010, 2011; Chester et al., 2015).

In the current implementation, I chose the PK-2 stress-based total Lagrangian framework, and the hydrogel is considered to be in a pre-swollen configuration. The current version of the subroutine supports linear triangular and bilinear quadrilateral plane-strain elements and linear tetrahedral and trilinear hexahedral three-dimensional elements with the constitutive choice of Neo-Hookean or Arruda-Boyce type potential for entropic stretching of elastomeric network with binary Flory-Huggins mixture potential for polymer-solvent mixing. Hydrogel has been modeled as a quasi-incompressible elastomer using a penalty function formulation. For the bilinear quadrilateral and trilinear hexahedral elements, the F-bar method (Chester et al., 2015; Neto et al., 1996) has been implemented to alleviate the volumetric locking issue.

Acknowledgments

1 Summary of the coupled theory and constitutive model

Hydrogels are solvent-filled crosslinked elastomeric networks of uncharged polymer. The constitutive behaviors of hydrogels are governed by the entropic stretching of the polymer network and the chemical mixing of polymer chains and the solvent. As-prepared hydrogels have a certain %wt of polymer and a certain %wt of solvent based on the chemical recipes. Depending on the properties of hydrogels, they can imbibe more solvent to obtain a chemomechanical equilibrium. In the current work, I will write the necessary finite element formulations and procedures for implementing a transient chemo-mechanical theory and constitutive model for hydrogels.

1.1 Finite strain kinematics

Let $\Omega_0(\mathbf{X}) \in \mathbb{R}^3$ be an arbitrary volume in the reference state of hydrogel where material coordinate $\mathbf{X} \in \Omega_0 \cup \partial\Omega_0$, where $\partial\Omega_0$ is the boundary of Ω_0. For a quasi-incompressible hydrogel with initial polymer volume fraction, ϕ_0^p, undergoing swelling or deswelling and elastic deformation, $\mathbf{F}$, represents the total deformation gradient. I prescribe the kinematics of deformation as multiplicative decomposition of the elastic and swelling deformation gradient and constitutive law for swelling volume change as,

$$
\begin{aligned}
\mathbf{F} &= (\phi_0^p)^{1/3}\mathbf{F}^e\mathbf{F}^s, \\
J^e &= \det(\mathbf{F}^e), \\
J^s = \det(\mathbf{F}^s) = \frac{1}{\phi_0^p}\left(C^p\mathcal{V}^p + C^w\mathcal{V}^w\right) &\quad \Rightarrow \mathbf{F}^s = \left(\frac{C^p\mathcal{V}^p + C^w\mathcal{V}^w}{\phi_0^p}\right)^{1/3}\mathbb{1}, \\
J = \det(\mathbf{F}) = \phi_0^p J^e J^s &= J^e\left(C^p\mathcal{V}^p + C^w\mathcal{V}^w\right).
\end{aligned}
\tag{1.1}
$$

Here, C^w and $\mathcal{V}^w$ are the nominal (referential) molar concentration and molar volume of the solvent, respectively. $\mathbf{F}^e$ and $\mathbf{F}^s$ are the deformation gradients related to the elastic and swelling deformation, respectively, and J^e and J^s are the volume change associated with them, respectively. The swelling volume change of hydrogel is entirely attributed to the solvent uptake governed by the diffusion process. However, the polymer chains and solvent are often considered incompressible or quasi-incompressible, thus, the elastic volume change of hydrogel is often modeled as incompressible or quasi-incompressible. In this work, I will consider the hydrogels to be quasi-incompressible with a large bulk modulus compared to the shear modulus, or in other words, the Poisson ratio is close to 0.5.

Remark 1. Ideally, $\mathbf{F} = \lambda_0^{-1}\mathbf{F}^e\mathbf{F}^s$ where λ_0 is the initial stretch from the dry state of the hydrogel to the hydrated state of the gel, however, because of the quasi-incompressible behavior, I assumed $\lambda_0 \approx (\phi_0^p)^{-1/3}$. Additionally, for non-reactive gels in which the polymer mass stays the same throughput swelling, I can show $J^s = 1/\phi^p$.

1.2 Strong form of the governing equations, boundary, and initial conditions in material (reference) configuration

In the reference configuration, Γ_g and Γ_T, and Γ_h and Γ_{I^w} be two distinct pairs of subsurfaces of the boundary $\partial\Omega_0$. The subsurfaces can be defined as follows,

$$\begin{aligned}
\partial\Omega_0 &= \Gamma_g \cup \Gamma_T \quad \text{and} \quad \Gamma_g \cap \Gamma_T = \varnothing, \\
&= \Gamma_h \cup \Gamma_{I^w} \quad \text{and} \quad \Gamma_h \cap \Gamma_{I^w} = \varnothing.
\end{aligned} \tag{1.2}$$

For a time interval $t \in [0, T]$, the governing partial differential equation for stress equilibrium as well as the boundary conditions in terms of the displacement vector, $\mathbf{u}$, on Γ_g and the Piola-Kirchhoff traction, $\mathbf{T}$, on Γ_T are given by,

$$\begin{aligned}
\mathrm{Div}(\mathbf{FS}) + \rho_R \mathbf{B} &= 0 \quad && \text{in } \Omega_0 \times [0, T], \\
\mathbf{u} &= \mathbf{g} \quad && \text{on } \Gamma_g \times [0, T], \\
\mathbf{P} \cdot \mathbf{N} = (\mathbf{FS}) \cdot \mathbf{N} &= \mathbf{T} \quad && \text{on } \Gamma_T \times [0, T].
\end{aligned} \tag{1.3}$$

Here, $\mathbf{P}$ and $\mathbf{S}$ are the first and the second Piola-Kirchhoff stress, respectively, and $\mathbf{B}$ is the body force per unit of mass in the hydrated reference configuration. The constitutive relation for the second Piola-Kirchhoff stress is given by (1.9). I expressed the governing equation for linear momentum balance in terms of the second Piola-Kirchhoff stress to take advantage of its symmetric nature during numerical implementation. For a time interval $t \in [0, T]$, the mass balance equation for the solvent, and the initial condition μ_0^w in Ω_0 and the boundary conditions in terms of the solvent chemical potential, μ^w, on Γ_h and solvent flux, $\mathbf{J}^w$, on Γ_{I^w} are given by,

$$\begin{aligned}
\dot{C}^w &= -\mathrm{Div}\,\mathbf{J}^w \quad && \text{in } \Omega_0 \times [0, T] \\
\mu^w(\mathbf{X}, t = 0) &= \mu_0^w \quad && \text{in } \Omega_0, \\
\mu^w &= h \quad && \text{on } \Gamma_h \times [0, T], \\
-\mathbf{J}^w \cdot \mathbf{N} &= I^w \quad && \text{on } \Gamma_{I^w} \times [0, T].
\end{aligned} \tag{1.4}$$

Here, $\mathbf{J}^w$ is the molar flux of the solvent, and the constitutive relation for $\mathbf{J}^w$ is given by (1.13).

1.3 Constitutive relations

In this work, I assumed the Langevin chain-based 8-chain Arruda-Boyce network describes the entropic elasticity of the polymer chain network. The chemical mixing of polymer chains and the solvent is described using a binary Flory-Huggins potential. Thus, the total Helmholtz

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free energy density, Ψ, for a pre-swollen hydrogel can be written as follows,

$$\Psi = \mu_w^0 C^w + \phi_0^p N R\theta \left[\lambda_{\mathrm{L}}^2 \left(\frac{\lambda_c \beta_c}{\lambda_{\mathrm{L}}} + \ln \frac{\beta_c}{\sinh \beta_c} \right) - \left(\frac{\lambda_{\mathrm{L}}}{3} \beta_0 \right) \ln J \right] + \frac{\phi_0^p \kappa}{2} J^s (\ln J^e)^2$$

$$+ \frac{\phi_0^p R\theta}{\mathcal{V}^w} \frac{1 - \phi^p}{\phi^p} \left[\ln \left(1 - \phi^p \right) + \chi \phi^p \right], \tag{1.5}$$

where, $\beta_c = \mathcal{L}^{-1} \left(\frac{\lambda_c}{\lambda_{\mathrm{L}}} \right)$, $\beta_0 = \mathcal{L}^{-1} \left(\frac{\lambda^0}{\lambda_{\mathrm{L}}} \right)$, and $\lambda_c = \sqrt{\frac{I_1}{3}} = (J^s)^{1/3} \sqrt{\frac{I_1^e}{3}}$.

Here, μ_w^0 is the chemical potential for the pure solvent, and θ is the temperature under consideration. N is the molar density, λ_{L} is the locking stretch, and ϕ^p is the volume fraction of polymer chains. $\mathcal{L}^{-1}(x)$ represents the inverse of Langevin function, $\mathcal{L}(x) = \coth(x) - 1/x$. The Flory-Huggins interaction parameter, χ, describes the energetic contribution of polymer solvent mixing which could depend on temperature and polymer concentration, etc. A higher value of the Flory-Huggins interaction parameter indicates unfavorable mixing, and thus, a smaller degree of swelling or deswelling, depending on the current or reference state of the hydrogel.

The Cauchy stress, $\boldsymbol{\sigma}$, is given by,

$$\boldsymbol{\sigma} = 2J^{-1} \left[\mathbf{F}^e \left(\frac{\partial \Psi}{\partial \mathbf{C}^e} \right) \mathbf{F}^{e\top} \right],$$

$$= J^{-1} \left[G_0 \left(\frac{\lambda_{\mathrm{L}}}{3\lambda_c} \beta_c \right) \mathbf{b} - G_0 \left(\frac{\lambda_{\mathrm{L}}}{3} \beta_0 \right) (\phi_0^p)^{2/3} \mathbb{1} + \phi_0^p J^s \kappa (\ln J^e) \mathbb{1} \right], \tag{1.6}$$

where, $G_0 = N R\theta (\phi_0^p)^{1/3}$.

The mean pressure, p, is given by,

$$p = -\frac{1}{3} J^e \operatorname{tr}(\boldsymbol{\sigma}),$$

$$= \frac{-1}{3\phi_0^p J^s} \left[G_0 \left(\frac{\lambda_{\mathrm{L}}}{3\lambda_c} \beta_c \right) I_1 - G_0 (\phi_0^p)^{2/3} \lambda_{\mathrm{L}} \right] - \kappa (\ln J^e). \tag{1.7}$$

The first Piola-Kirchhoff stress, $\mathbf{P}$, can be calculated using the pull-back operation on the Cauchy stress, $\boldsymbol{\sigma}$, as,

$$\mathbf{P} = J \boldsymbol{\sigma} \mathbf{F}^{-\top} = \frac{\partial \Psi}{\partial \mathbf{F}^e} = G_0 \left(\frac{\lambda_{\mathrm{L}}}{3\lambda_c} \beta_c \right) \mathbf{F} - \left[(\phi_0^p)^{2/3} G_0 \left(\frac{\lambda_{\mathrm{L}}}{3} \right) - \phi_0^p J^s \kappa (\ln J^e) \right] \mathbf{F}^{-\top}. \tag{1.8}$$

Similarly, the second Piola-Kirchhoff stress is given by,

$$\mathbf{S} = J \mathbf{F}^{-1} \boldsymbol{\sigma} \mathbf{F}^{-\top} = \mathbf{F}^{-1} \mathbf{P} = G_0 \left(\frac{\lambda_{\mathrm{L}}}{3\lambda_c} \beta_c \right) \mathbb{1} - \left[(\phi_0^p)^{2/3} G_0 \left(\frac{\lambda_{\mathrm{L}}}{3} \right) - \phi_0^p J^s \kappa (\ln J^e) \right] \mathbf{C}^{-1}, \tag{1.9}$$

where, $\mathbf{C} = \mathbf{F}^\top \mathbf{F}$ is known as the right Cauchy-Green deformation tensor.

The chemical potential for the solvent, μ^w, is,

$$\mu^w = \frac{\partial \Psi}{\partial C^w} + p\mathcal{V}^w,$$

$$= \mu_w^0 + \frac{R\theta}{J^s}\left[1 + \frac{1}{\phi^p}\ln(1-\phi^p) + \chi\phi^p\right] - \kappa\mathcal{V}^w(\ln J^e) + \frac{\kappa\mathcal{V}^w}{2}(\ln J^e)^2, \qquad (1.10)$$

$$= \mu_w^0 + R\theta\left[\phi^p + \ln(1-\phi^p) + \chi(\phi^p)^2\right] - \kappa\mathcal{V}^w(\ln J^e) + \frac{\kappa\mathcal{V}^w}{2}(\ln J^e)^2.$$

Chemical potential for the solvent, μ^w, can be expressed in terms of concentrations using the definition of polymer volume fraction as

$$\phi^p = \frac{C^p\mathcal{V}^p}{C^p\mathcal{V}^p + C^w\mathcal{V}^w}. \qquad (1.11)$$

Hence, it can be written as,

$$\mu_w^0 + R\theta\left[\phi^p + \ln(1-\phi^p) + \chi(\phi^p)^2\right] - \kappa\mathcal{V}^w(\ln J^e) + \frac{\kappa\mathcal{V}^w}{2}(\ln J^e)^2 - \mu^w = 0, \qquad (1.12)$$

$$\Rightarrow \mathcal{G}\left(\mathbf{C}, \mu^w, \phi^p(C^w)\right) = 0.$$

For solvent transport, I assumed a Fickian-type diffusion flux relation for the solvent, $\mathbf{J}^w$, which is given as,

$$\mathbf{J}^w = -\frac{D^w C^w}{R\theta}\mathbf{C}^{-1}\operatorname{Grad}\mu^w, \qquad (1.13)$$

where, D^w is the diffusion coefficient of the solvent, and $\mathbf{C} = \mathbf{F}^\top\mathbf{F}$ is the right Cauchy-Green deformation tensor. To simplify the calculations, I define the isotropic mobility tensor, $\mathbf{M}^w$, as,

$$\mathbf{M}^w = \frac{D^w C^w}{R\theta}\mathbf{C}^{-1} \qquad (1.14)$$

Remark 2. The Langevin statistics-based 8-chain Arruda-Boyce network model has a stretch-dependent shear modulus. In contrast, the Gaussian statistics-based N-chain network model has a constant shear modulus. In the limit of infinite (numerically large) locking stretch, $\lambda_{\mathrm{L}} \to \infty$, I get $\zeta \approx 1$, and thus, $G_0\zeta = G_0$. Thus, the expressions for the free energy density function and the stress from the Langevin statistics-based 8-chain Arruda-Boyce model reduce to the N-chain Gaussian network (Neo-Hookean) model. The free energy for a pre-swollen Neo-Hookean elastomer is given by,

$$\Psi^{\mathrm{mech}} = \frac{G_0}{2}\left[(\phi_0^p J^s)^{2/3}I_1^e - 3 - 2(\phi_0^p)^{2/3}\ln(J)\right] + \frac{\kappa\phi_0^p}{2}J^s(\ln J^e)^2 \qquad (1.15)$$

The Cauchy stress, $\boldsymbol{\sigma}$, and the first Piola-Kirchhoff stress, $\mathbf{P}$, and the second Piola-Kirchhoff

stress, $\mathbf{S}$, are given by,

$$\boldsymbol{\sigma} = J^{-1} \left[G_0 \mathbf{b} - \left(G_0 (\phi_0^p)^{2/3} - \kappa \phi_0^p J^s (\ln J^e) \right) \mathbb{1} \right],$$
$$\mathbf{P} = G_0 \mathbf{F} - \left[(\phi_0^p)^{2/3} G_0 - \kappa \phi_0^p J^s (\ln J^e) \right] \mathbf{F}^{-\top}, \qquad (1.16)$$
$$\mathbf{S} = G_0 \mathbb{1} - \left[(\phi_0^p)^{2/3} G_0 - \kappa \phi_0^p J^s (\ln J^e) \right] \mathbf{C}^{-1}.$$

2 Mixed finite element (u-μ) formulation

The first step towards finite element formulation is to develop the corresponding weak forms of the governing equation. Typically, the variational method or weighted residual method is used to derive the weak form. In this work, I will use a weighted residual approach with a standard Galerkin approach to develop the finite element formulation. In Abaqus/Standard, the user element subroutine (UEL) requires defining the element stiffness (tangent) matrix, `AMATRX`, and the element residual vector, `RHS`. Abaqus performs the global assembly and iterative solution procedures.

2.1 Weighted residual based weak formulation of the governing equations

Let us assume $\mathbf{W} \in \mathscr{W}$ be a vector test (or weight) function which satisfies $W_i = 0$ on Γ_g. I can write the weak form of the momentum balance equation as

$$\int_{\Omega_0} \mathrm{Div}(\mathbf{FS}) \cdot \mathbf{W} \, dV + \int_{\Omega_0} \rho_\mathrm{R} \mathbf{B} \cdot \mathbf{W} \, dV = 0,$$
$$\Rightarrow \int_{\Omega_0} \mathbf{FS} : \mathrm{Grad}(\mathbf{W}) \, dV - \int_{\Omega_0} \rho_\mathrm{R} \mathbf{B} \cdot \mathbf{W} \, dV - \int_{\Gamma_T} \mathbf{T} \cdot \mathbf{W} \, dS = 0. \qquad (2.1)$$

Applying the divergence theorem on $(2.1)_1$ results in the final weak form of stress equilibrium $(2.1)_2$. In the context of the principle of virtual work, the first term in $(2.1)_2$ is called internal work, and the second and third terms are collectively known as external work.

Let us assume $\Theta \in \mathscr{V}$ be a scalar weight function that satisfies $\Theta = 0$ on Γ_h, then I can write the weak form of mass balance equation for the solvent as,

$$\int_{\Omega_0} \dot{C}^w \, \Theta \, dV + \int_{\Omega_0} \mathrm{Div} \, \mathbf{J}^w \, \Theta \, dV = 0,$$
$$\Rightarrow \int_{\Omega_0} \dot{C}^w \, \Theta \, dV - \int_{\Omega_0} \mathbf{J}^w \cdot \mathrm{Grad}(\Theta) \, dV - \int_{\Gamma_{I^w}} I^w \, \Theta \, dS = 0. \qquad (2.2)$$

Similar to the weak form of stress equilibrium, application of the divergence theorem on $(2.2)_1$ results in the final weak form of mass balance $(2.2)_2$.

2.2 Discretization and Buvnov-Galerkin finite element approximation

The computational domain and its boundary are discretized using finite elements, *i.e.*, $\Omega_0 = \cup\, \Omega_0^e$ and $\partial\Omega_0 = \cup\, \partial\Omega_0^e$. Using this approximation (Zienkiewicz et al., 2014, Chapter 2) and based on the final weak forms for stress equilibrium $(2.1)_2$ and mass balance $(2.2)_2$, I can write the weak forms for system of governing equations for each element as,

$$\mathcal{W}_{\mathbf{u}}^e(\mathbf{u}, \mu^w) = -\int_{\Omega_0^e} \mathbf{FS} : \text{Grad}(\mathbf{W})\ dV + \int_{\Omega_0^e} \rho_{\mathrm{R}}\mathbf{B}^e \cdot \mathbf{W}\ dV + \int_{\Gamma_T^e} \mathbf{T}^e \cdot \mathbf{W}\ dS = 0,$$

$$\mathcal{W}_{\mu}^e(\mathbf{u}, \mu^w) = \int_{\Omega_0^e} \dot{C}^w\ \Theta\ dV - \int_{\Omega_0^e} \mathbf{J}^w \cdot \text{Grad}(\Theta)\ dV - \int_{\Gamma_{Iw}^e} I^{we}\ \Theta\ dS = 0. \tag{2.3}$$

Remark 3. In most diffusion-based transport problems, the concentration of the relevant species is typically taken as the nodal variable in finite element implementation. In those cases, the solution mixture is assumed to be ideal and because of the simplified constitutive choice for the ideal mixture, it can be easily shown that the transport is driven by the gradient of concentration. Thus, it becomes convenient to use concentration as the degree of freedom (or nodal variable). However, for hydrogels, the polymer-solvent solution is non-deal, and the gradient of the chemical potential of the solvent drives the transport until it reaches an equilibrium with the external solution. It is difficult to obtain an explicit expression for flux in terms of concentration. Thus, it is more convenient and physically meaningful to use chemical potential as the nodal variable instead of concentration.

Let N^a denote the interpolation functions in terms of local coordinates, and X_i^a is the i-th component coordinate of node a within the element. Then the coordinate of any point within an element, $\mathbf{X}$, can be approximated as,

$$X_i = \sum_{a=1}^{\text{nnode}} N^a X_i^a \quad \Rightarrow \mathbf{X} = \mathbf{N_u} \mathbf{X}_e, \tag{2.4}$$

where, $\mathbf{N_u}$ is the matrix form of shape functions and $\mathbf{X}$ is the vector form of nodal coordinates within the element. For a three-dimensional computational domain, I can write the matrix form of $\mathbf{N_u}$ and the vector form of $\mathbf{x}_e$, as,

$$\mathbf{N_u} = \begin{bmatrix} N_1 & 0 & 0 & N_2 & 0 & 0 & \cdots & \cdots & N_{\text{nnode}} & 0 & 0 \\ 0 & N_1 & 0 & 0 & N_2 & 0 & \cdots & \cdots & 0 & N_{\text{nnode}} & 0 \\ 0 & 0 & N_1 & 0 & 0 & N_2 & \cdots & \cdots & 0 & 0 & N_{\text{nnode}} \end{bmatrix}_{\text{ndim}\times\text{nnode}*\text{ndim}},$$

$$\mathbf{X}_e = \begin{bmatrix} X_1 & Y_1 & Z_1 & X_2 & Y_2 & Z_2 & \cdots & \cdots & X_{\text{nnode}} & Y_{\text{nnode}} & Z_{\text{nnode}} \end{bmatrix}_{\text{nnode}*\text{ndim}\times 1}^{\top}. \tag{2.5}$$

By inspecting the structure of $[\mathbf{N_u}]$, I can see there exist repeating blocks of sub-matrices corresponding to each node within the element. Let define those sub-matrices as $\mathbf{N_u^a}$, then

for a three dimensional case, I can write $\mathbf{N_u^a}$ and X_a^e as,

$$\mathbf{N_u^a} = \begin{bmatrix} N_i & 0 & 0 \\ 0 & N_i & 0 \\ 0 & 0 & N_i \end{bmatrix}_{\text{ndim} \times \text{ndim}} , \quad \mathbf{X}^a = \begin{bmatrix} X_i \\ Y_i \\ Z_i \end{bmatrix}_{\text{ndim} \times 1} , \tag{2.6}$$

where, N_i is the interpolation function corresponds to the node i within the element. These matrix-vector forms of shape functions and nodal coordinates can be easily reduced to two-dimensional cases, as needed.

The trial solutions for the nodal variables, displacement, $\mathbf{u}$, and chemical potential, μ^w, are approximated using the interpolation function matrix, $\mathbf{N_u}$, and interpolation function vector, $\mathbf{N}_\mu$, as,

$$\mathbf{u}(\mathbf{X}) = \mathbf{N_u}\mathbf{u}^e = \sum_{a=1}^{\text{nnode}} \mathbf{N_u^a}(\mathbf{X})\mathbf{u}^a \quad \Rightarrow \quad u_i(\mathbf{X}) = \sum_{a=1}^{\text{nnode}} N^a(\mathbf{X})u_i^a,$$

$$\text{and} \quad \mu^w(\mathbf{X}) = \mathbf{N}_\mu \mu^{we} = \sum_{a=1}^{\text{nnode}} N^a \mu^{wa}. \tag{2.7}$$

where, $\mathbf{u}^e$ and μ^{we} are the vectors of nodal displacements and chemical potentials for an element, respectively. Since chemical potential, μ^w, is a scalar field, the corresponding shape function vector, $\mathbf{N}_\mu$, is given by,

$$\mathbf{N}_\mu = \begin{bmatrix} N_\mu^1 & N_\mu^2 & N_\mu^3 & \cdots & \cdots & N_\mu^{\text{nnode}} \end{bmatrix}. \tag{2.8}$$

Employing a standard Galerkin approximation, $i.e.$, approximating the test functions, $\mathbf{W}$ and Θ, using the same interpolation functions as the nodal variables, I can write,

$$\mathbf{W}(\mathbf{X}) = \mathbf{N_u}\mathbf{W}^e \quad \Rightarrow \quad W_i(\mathbf{X}) = \sum_{a=1}^{\text{nnode}} N^a(\mathbf{X})W_i^a,$$

$$\Theta(\mathbf{X}) = \mathbf{N}_\mu \mathbf{\Theta}^e = \sum_{a=1}^{\text{nnode}} N^a(\mathbf{X})\Theta^a. \tag{2.9}$$

The deformation gradient, $\mathbf{F}$, thus can be calculated as,

$$\mathbf{F} = \frac{\partial \mathbf{x}}{\partial \mathbf{X}} = \mathbb{1} + \frac{\partial \mathbf{u}}{\partial \mathbf{X}} = \mathbb{1} + [\bar{\mathbf{u}}_e]\left[\frac{\partial \mathbf{N_u}}{\partial \mathbf{X}}\right] \quad \Rightarrow \quad F_{iJ} = \delta_{ij} + \sum_{a=1}^{\text{nnode}} \frac{\partial N^a}{\partial X_J}u_i^a. \tag{2.10}$$

where u_i^a denotes i-th nodal displacement component of node a, and N^a is the shape function corresponding to the same node. $\left[\frac{\partial \mathbf{N_u}}{\partial \mathbf{X}}\right]$ is a matrix which has a dimension of $\left[\frac{\partial \mathbf{N_u}}{\partial \mathbf{X}}\right]_{\text{nnode} \times \text{ndim}}$, and $[\bar{\mathbf{u}}_e]$ has a dimension of $[\bar{\mathbf{u}}_e]_{\text{ndim} \times \text{nnode}}$. Once the deformation gradient is obtained, additional deformation tensors and strain tensors can be calculated based on that.

With the Galerkin discretization and Euler implicit time integration scheme, the system of

element residuals can be written as,

$$\mathbf{R_u^e}(\mathbf{u}^e, \mu^{we}) = -\int_{\Omega_0^e} \mathbf{FS} : \text{Grad}(\mathbf{N_u}) \, dV + \int_{\Omega_0^e} \rho_R \mathbf{N_u^\top} \mathbf{B}^e \, dV + \int_{\Gamma_T^e} \mathbf{N_u^\top} \mathbf{T}^e \, dS = 0,$$

$$\mathbf{R_\mu^e}(\mathbf{u}^e, \mu^{we}) = -\int_{\Omega_0^e} \mathbf{N_\mu^\top} \left(\frac{C^w - C_t^w}{\Delta t} \right) \, dV + \int_{\Omega_0^e} \mathbf{J}^w \cdot \text{Grad}(\mathbf{N_\mu}) \, dV + \int_{\Gamma_{Iw}^e} \mathbf{N_\mu^\top} I^{we} \, dS = 0.$$

(2.11)

Here, $\dot{C}^w = \frac{C^w - C_t^w}{\Delta t}$ represents the discretized time derivative with C^w being the referential concentration of the solvent at time $t + \Delta t$ and C_t^w being the referential concentration of the solvent at time t.

The element residual vectors, $\mathbf{R}_\mathbf{u}^e$ and $\mathbf{R}_\mu^e$, correspond to all of the degrees of freedom (DOFs) in an element. In the Abaqus/ Standard user element subroutine (UEL), it is required to have the degrees of freedom correspond to each node together, thus, it would be convenient to write the residual vector for each node as follows,

$$\mathbf{R_u^a}(\mathbf{u}^e, \mu^{we}) = -\int_{\Omega_0^e} \mathbf{FS} : \text{Grad}(\mathbf{N_u^a}) \, dV + \int_{\Omega_0^e} \rho_R \mathbf{N_u^{a\top}} \mathbf{B}^e \, dV + \int_{\Gamma_T^e} \mathbf{N_u^{a\top}} \mathbf{T}^e \, dS = 0,$$

$$R_\mu^a(\mathbf{u}^e, \mu^{we}) = -\int_{\Omega_0^e} N_\mu^{a\top} \left(\frac{C^w - C_t^w}{\Delta t} \right) \, dV + \int_{\Omega_0^e} \mathbf{J}^w \cdot \text{Grad}(N_\mu^a) \, dV + \int_{\Gamma_{Iw}^e} N_\mu^{a\top} I^{we} \, dS = 0.$$

(2.12)

2.3   Consistent linearization and Newton-Raphson method

As mentioned in the previous section, the second Piola-Kirchhoff stress, $\mathbf{S}$, the time derivative of the referential molar concentration of the solvent, $\dot{C}^w$, and referential molar flux of the solvent, $\mathbf{J}^w$ in the system of residuals are nonlinear functions of the displacement vector, $\mathbf{u}$, and the chemical potential of the solvent, μ^w. Thus, I need to linearize the system of nodal residuals along the independent variables (degrees of freedom) to employ Newton-Raphson iteration. Assuming, at any time $t = t_{n+1}$, the initial guess for the independent nodal variables are $\mathbf{u}^b$ and μ^{wb}. Then the system of nodal residuals can be perturbed by $\Delta \mathbf{u}^b$ and $\Delta \mu^{wb}$, and using first-order Taylor series expansion, I have,

$$\mathbf{R_u^a} \left(\mathbf{u}^b + \Delta \mathbf{u}^b, \mu^{wb} + \Delta \mu^{wb} \right) = \mathbf{R_u^a}(\mathbf{u}, \mu^w) + \frac{\partial \mathbf{R_u^a}(\mathbf{u}, \mu^w)}{\partial \mathbf{u}^b} \Delta \mathbf{u}^b + \frac{\partial \mathbf{R_u^a}(\mathbf{u}, \mu^w)}{\partial \mu^{wb}} \Delta \mu^{wb},$$

$$R_\mu^a \left(\mathbf{u}^b + \Delta \mathbf{u}^b, \mu^{wb} + \Delta \mu^{wb} \right) = R_\mu^a(\mathbf{u}, \mu^w) + \frac{\partial R_\mu^a(\mathbf{u}, \mu^w)}{\partial \mathbf{u}^b} \Delta \mathbf{u}^b + \frac{\partial R_\mu^a(\mathbf{u}, \mu^w)}{\partial \mu^{wb}} \Delta \mu^{wb}.$$

(2.13)

To obtain the solution of the nonlinear system of residuals, the perturbed residuals need to be zero, thus, I have,

$$-\frac{\partial \mathbf{R_u^a}}{\partial \mathbf{u}^b} \Delta \mathbf{u}^b - \frac{\partial \mathbf{R_u^a}}{\partial \mu^{wb}} \Delta \mu^{wb} = \mathbf{R_u^a},$$

$$-\frac{\partial R_\mu^a}{\partial \mathbf{u}^b} \Delta \mathbf{u}^b - \frac{\partial R_\mu^a}{\partial \mu^{wb}} \Delta \mu^{wb} = R_\mu^a.$$

(2.14)

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In matrix form, it can be written as,

$$
\begin{bmatrix} -\frac{\partial \mathbf{R}_{\mathbf{u}}^a}{\partial \mathbf{u}^b} & -\frac{\partial \mathbf{R}_{\mathbf{u}}^a}{\partial \mu^{wb}} \\ -\frac{\partial R_\mu^a}{\partial \mathbf{u}^b} & -\frac{\partial R_\mu^a}{\partial \mu^{wb}} \end{bmatrix} \begin{Bmatrix} \Delta \mathbf{u}^b \\ \Delta \mu^{wb} \end{Bmatrix} = \begin{Bmatrix} \mathbf{R}_{\mathbf{u}}^a \\ R_\mu^a \end{Bmatrix}. \tag{2.15}
$$

In finite element literature, the matrix containing the derivatives of the system of residuals and the right-hand side vector containing the nodal residual is known as the stiffness (or tangent) matrix, $\mathbf{k}^{ab}$, and the element residual vector, $\mathbf{R}^a$, respectively.

$$
\mathbf{k}^{ab} = \begin{bmatrix} \mathbf{k}_{\mathbf{uu}}^{ab} & \mathbf{k}_{\mathbf{u}\mu}^{ab} \\ \mathbf{k}_{\mu\mathbf{u}}^{ab} & k_{\mu\mu}^{ab} \end{bmatrix} = \begin{bmatrix} -\frac{\partial \mathbf{R}_{\mathbf{u}}^a}{\partial \mathbf{u}^b} & -\frac{\partial \mathbf{R}_{\mathbf{u}}^a}{\partial \mu^{wb}} \\ -\frac{\partial R_\mu^a}{\partial \mathbf{u}^b} & -\frac{\partial R_\mu^a}{\partial \mu^{wb}} \end{bmatrix}, \quad \text{and} \quad \mathbf{R}^a = \begin{Bmatrix} \mathbf{R}_{\mathbf{u}}^a \\ R_\mu^a \end{Bmatrix}. \tag{2.16}
$$

The element tangent matrix, $\mathbf{k}^e$ and element residual vector, $\mathbf{R}^e$ can be obtained by assembling the nodal tangent matrix, $\mathbf{k}^{ab}$, and nodal residual vector, $\mathbf{R}^a$, as follows

$$
\mathbf{k}^e = \overset{\text{nnode}}{\underset{a,b=1}{\mathbf{A}}} \mathbf{k}^{ab}, \qquad \text{and} \qquad \mathbf{R}^e = \overset{\text{nnode}}{\underset{a=1}{\mathbf{A}}} \mathbf{R}^a. \tag{2.17}
$$

The element tangent matrix, $\mathbf{k}^e$, and element residual vector, $\mathbf{R}^e$, for a three-dimensional chemo-mechanical element with M nodes are given by,

$$
\mathbf{k}^e = \begin{bmatrix}
k_{u_1 u_1}^{11} & k_{u_1 u_2}^{11} & k_{u_1 u_3}^{11} & k_{u_1 \mu}^{11} & \cdots & k_{u_1 u_1}^{1M} & k_{u_1 u_2}^{1M} & k_{u_1 u_3}^{1M} & k_{u_1 \mu}^{1M} \\
k_{u_2 u_1}^{11} & k_{u_2 u_2}^{11} & k_{u_2 u_3}^{11} & k_{u_2 \mu}^{11} & \cdots & k_{u_2 u_1}^{1M} & k_{u_2 u_2}^{1M} & k_{u_2 u_3}^{1M} & k_{u_2 \mu}^{1M} \\
k_{u_3 u_1}^{11} & k_{u_3 u_2}^{11} & k_{u_3 u_3}^{11} & k_{u_3 \mu}^{11} & \cdots & k_{u_3 u_1}^{1M} & k_{u_3 u_2}^{1M} & k_{u_3 u_3}^{1M} & k_{u_3 \mu}^{1M} \\
k_{\mu u_1}^{11} & k_{\mu u_2}^{11} & k_{\mu u_3}^{11} & k_{\mu \mu}^{11} & \cdots & k_{\mu u_1}^{1M} & k_{\mu u_2}^{1M} & k_{\mu u_3}^{1M} & k_{\mu \mu}^{1M} \\
\vdots & \vdots & & \vdots & \ddots & \vdots & & \vdots & \vdots \\
\vdots & \vdots & & \vdots & \ddots & \vdots & & \vdots & \vdots \\
k_{u_1 u_1}^{M1} & k_{u_1 u_2}^{M1} & k_{u_1 u_3}^{M1} & k_{u_1 \mu}^{M1} & \cdots & k_{u_1 u_1}^{MM} & k_{u_1 u_2}^{MM} & k_{u_1 u_3}^{MM} & k_{u_1 \mu}^{MM} \\
k_{u_2 u_1}^{M1} & k_{u_2 u_2}^{M1} & k_{u_2 u_3}^{M1} & k_{u_2 \mu}^{M1} & \cdots & k_{u_2 u_1}^{MM} & k_{u_2 u_2}^{MM} & k_{u_2 u_3}^{MM} & k_{u_2 \mu}^{MM} \\
k_{u_3 u_1}^{M1} & k_{u_3 u_2}^{M1} & k_{u_3 u_3}^{M1} & k_{u_3 \mu}^{M1} & \cdots & k_{u_3 u_1}^{MM} & k_{u_3 u_2}^{MM} & k_{u_3 u_3}^{MM} & k_{u_3 \mu}^{MM} \\
k_{\mu u_1}^{M1} & k_{\mu u_2}^{M1} & k_{\mu u_3}^{M1} & k_{\mu \mu}^{M1} & \cdots & k_{\mu u_1}^{MM} & k_{\mu u_2}^{MM} & k_{\mu u_3}^{MM} & k_{\mu \mu}^{MM}
\end{bmatrix}_{\text{ndofel} \times \text{ndofel}}
, \mathbf{R}^e = \begin{bmatrix}
R_{u_1}^1 \\ R_{u_2}^1 \\ R_{u_3}^1 \\ R_\mu^1 \\ \vdots \\ R_{u_1}^M \\ R_{u_2}^M \\ R_{u_3}^M \\ R_\mu^M
\end{bmatrix}_{\text{ndofel} \times 1}
\tag{2.18}
$$

The global stiffness (tangent) matrix, $[\mathbf{K}]$, and the global residual vector, $\{\mathbf{R}\}$, can be obtained by assembling the element-level quantities. Assembly of the global stiffness (tangent) matrix and global residual vector, and application of kinematic (or essential) boundary conditions are done by Abaqus once the element stiffness (tangent) matrix and element residual vectors are defined in the user element subroutine (UEL).

$$
\mathbf{K} = \overset{\text{nel}}{\underset{e=1}{\mathbf{A}}} \mathbf{k}^e, \qquad \text{and} \qquad \mathbf{R} = \overset{\text{nel}}{\underset{e=1}{\mathbf{A}}} \mathbf{R}^e. \tag{2.19}
$$

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In matrix form, the global stiffness (tangent) matrix and residual vector appear as,

$$[\mathbf{K}]\{\Delta \mathbf{d}\} = \{\mathbf{R}\}, \tag{2.20}$$

Knowing the initial guess for $\mathbf{d}$, this system of linear equations can be solved for nodal variables, $\Delta \mathbf{d}$. Consequently, the initial guess is updated for any $k+1$ iteration as below, and the solution procedure is iteratively repeated until $\Delta \mathbf{d}$ satisfies the tolerance.

$$\mathbf{d}_{k+1} = \mathbf{d}_k + \Delta \mathbf{d} \tag{2.21}$$

This is known as the Newton-Raphson procedure for solving a nonlinear system of equations.

2.4 Expressions for element stiffness (tangent) matrix and residual vector

Based on the coupled theory and constitutive model, I assumed the following functional forms,

$$\mathbf{S} \equiv \mathbf{S}\left(\mathbf{C}, C^w\right), \quad \mathcal{G}\left(\mathbf{C}, \mu^w, C^w\right) = 0, \quad \mathbf{J}^w \equiv \mathbf{J}^w\left(\mathbf{C}, C^w, \mathrm{Grad}(\mu^w)\right) \tag{2.22}$$

2.4.1 Mechanical tangent matrix, $\mathbf{k}^e_{\mathbf{uu}}$

In index notation, the residual for the linear momentum balance equation is given by,

$$R^a_{\mathbf{u}}(u_i, \mu^w) = -\int_{\Omega_0^e} \frac{\partial N^a}{\partial X_I} F_{iJ} S_{JI} \; dV + \int_{\Omega_0^e} \rho_{\mathrm{R}} N^a B_i^e \; dV + \int_{\Gamma_T^e} N^a T_i^e \; dS = 0. \tag{2.23}$$

The mechanical tangent, $k^{ab}_{u_i u_k}$, is given by,

$$
\begin{aligned}
k^{ab}_{u_i u_k} = -\frac{\partial R^a_{u_i}}{\partial u^b_k} &= \int_{\Omega^e_0} \frac{\partial N^a}{\partial X_I} \left(\frac{\partial F_{iJ}}{\partial u^b_k} S_{JI} + F_{iJ} \frac{\partial S_{JI}}{\partial u^b_k} \right) \ dV, \\
&= \int_{\Omega^e_0} \frac{\partial N^a}{\partial X_I} \left[\frac{\partial F_{iJ}}{\partial u^b_k} S_{JI} + F_{iJ} \left(\frac{\partial S_{JI}}{\partial C_{KL}} + \frac{\partial S_{JI}}{\partial C^w} \frac{\partial C^w}{\partial C_{kL}} \right) \frac{\partial C_{KL}}{\partial F_{mN}} \frac{\partial F_{mN}}{\partial u^b_k} \right] \ dV, \\
&= \int_{\Omega^e_0} \frac{\partial N^a}{\partial X_I} \left[\frac{\partial F_{iJ}}{\partial u^b_k} S_{JI} + 2F_{iJ} \left(\frac{\partial S_{JI}}{\partial C_{KL}} + \frac{\partial S_{JI}}{\partial C^w} \frac{\partial C^w}{\partial C_{kL}} \right) F_{mL} \delta_{KN} \frac{\partial F_{mN}}{\partial u^b_k} \right] \ dV, \\
&= \int_{\Omega^e_0} \frac{\partial N^a}{\partial X_I} \left(\frac{\partial F_{iJ}}{\partial u^b_k} S_{JI} + F_{iJ} \mathbb{C}_{IJKL} F_{mL} \frac{\partial F_{mK}}{\partial u^b_k} \right) \ dV, \\
&= \int_{\Omega^e_0} \frac{\partial N^a}{\partial X_I} \left(\frac{\partial N^b}{\partial X_J} \delta_{ik} S_{JI} + F_{iJ} \mathbb{C}_{IJKL} F_{mL} \frac{\partial N^b}{\partial X_K} \delta_{mk} \right) \ dV, \\
&= \int_{\Omega^e_0} \frac{\partial N^a}{\partial X_I} \left(\frac{\partial N^b}{\partial X_J} \delta_{ik} S_{JI} + F_{iJ} \mathbb{C}_{IJKL} F_{kL} \frac{\partial N^b}{\partial X_K} \right) \ dV, \\
&= \int_{\Omega^e_0} \frac{\partial N^a}{\partial X_I} S_{JI} \delta_{ik} \frac{\partial N^b}{\partial X_J} \ dV + \int_{\Omega^e_0} \frac{\partial N^a}{\partial X_I} \left(F_{iJ} \mathbb{C}_{IJKL} F_{kL} \right) \frac{\partial N^b}{\partial X_K} \ dV, \\
&= \int_{\Omega^e_0} \frac{\partial N^a}{\partial X_J} S_{JL} \delta_{ik} \frac{\partial N^b}{\partial X_L} \ dV + \int_{\Omega^e_0} \frac{\partial N^a}{\partial X_J} \left(F_{iI} \mathbb{C}_{IJKL} F_{kK} \right) \frac{\partial N^b}{\partial X_L} \ dV.
\end{aligned}
$$

$$(2.24)$$

Here,

$$
\mathbb{C}_{IJKL} = 2 \left(\frac{\partial S_{IJ}}{\partial C_{KL}} + \frac{\partial S_{IJ}}{\partial C^w} \frac{\partial C^w}{\partial C_{KL}} \right) \tag{2.25}
$$

is defined as the material tangent tensor (or second elasticity tensor). Owing to the symmetric nature of the second Piola-Kirchhoff stress, $\mathbf{S} = \mathbf{S}^\top \Rightarrow S_{IJ} = S_{JI}$, and the right Cauchy-Green deformation tensor, $\mathbf{C} = \mathbf{C}^\top \Rightarrow C_{KL} = C_{LK}$, the material tangent tensor, $\mathbb{C}$, possesses minor symmetry, *i.e.*, $\mathbb{C}_{IJKL} = \mathbb{C}_{JILK}$.

Additionally, I used the properties of summation indices (or dummy indices) and the following intermediate results to obtain the final expression for $k^{ab}_{u_i u_k}$.

$$
\frac{\partial C_{KL}}{\partial F_{mN}} = F_{mL} \delta_{KN} + F_{mK} \delta_{LN} = 2 F_{mL} \delta_{KN} \quad \text{(since } C_{KL} = C_{LK}\text{)},
$$

$$
\text{and,} \quad F_{iJ} = \delta_{ij} + \sum_{b=1}^{\text{nnode}} \frac{\partial N^b}{\partial X_J} u^b_i \quad \Rightarrow \quad \frac{\partial F_{iJ}}{\partial u^b_k} = \frac{\partial N^b}{\partial X_J} \delta_{ik}.
$$

$$(2.26)$$

Thus, the matrix form of the mechanical element tangent matrix is given by,

$$[\mathbf{k}_{\mathbf{uu}}^e]_{\text{nnode}*\text{ndim}\times\text{nnode}*\text{ndim}} = \int_{\Omega_0^e} \left[\mathbf{G}_{\mathbf{u}}^\top \mathbf{\Sigma}_{\mathbf{S}} \mathbf{G}_{\mathbf{u}} + (\mathbf{B}_{\mathbf{u}}\mathbf{\Sigma}_{\mathbf{F}})^\top \mathbf{D}_{\mathbb{C}}(\mathbf{B}_{\mathbf{u}}\mathbf{\Sigma}_{\mathbf{F}}) \right] \, dV \qquad (2.27)$$

where, $\mathbf{D}_{\mathbb{C}}$ is the Voigt matrix form of the referential mechanical tangent $\mathbb{C}_{IJKL}$. The form of the non-symmetric gradient matrix, $\mathbf{G}_{\mathbf{u}}$, the symmetric gradient matrix, $\mathbf{B}_{\mathbf{u}}$, the sparse second Piola-Kirchhoff stress matrix, $\mathbf{\Sigma}_{\mathbf{S}}$, the sparse deformation gradient matrix, $\mathbf{\Sigma}_{\mathbf{F}}$, are given in the next section.

2.4.2 Mechano-chemical tangent matrix, $\mathbf{k}_{\mathbf{u}\mu}^e$

The components of mechano-chemical element sub-matrix, $k_{u_i\mu}^{ab}$, can be written as,

$$
\begin{aligned}
k_{u_i\mu}^{ab} = -\frac{\partial R_{u_i}^a}{\partial \mu^{wb}} &= \int_{\Omega_0^e} \frac{\partial N^a}{\partial X_I} \left(F_{iJ} \frac{\partial S_{JI}}{\partial C^w} \frac{\partial C^w}{\partial \mu^w} \right) \frac{\partial \mu^w}{\partial \mu^{wb}} \, dV, \\
&= \int_{\Omega_0^e} \frac{\partial N^a}{\partial X_I} \left(F_{iJ} \frac{\partial S_{JI}}{\partial C^w} \frac{\partial C^w}{\partial \mu^w} \right) N^b \, dV,
\end{aligned}
\qquad (2.28)
$$

Let define,

$$\mathbb{S}_{iI} = F_{iJ} \frac{\partial S_{JI}}{\partial C^w} \frac{\partial C^w}{\partial \mu^w}. \qquad (2.29)$$

Thus, I can write,

$$k_{u_i\mu}^{ab} = \int_{\Omega_0^e} \frac{\partial N^a}{\partial X_I} \mathbb{S}_{iI} N^b \, dV. \qquad (2.30)$$

In matrix form, it can be expressed as,

$$\left[\mathbf{k}_{\mathbf{u}\mu}^e \right]_{\text{nnode}*\text{ndim}\times\text{nnode}} = \int_{\Omega_0^e} \mathbf{G}_{\mathbf{u}}^\top \mathbf{d}_{\mathbf{u}\mu} \mathbf{N}_\mu \, dV \qquad (2.31)$$

where $\mathbf{d}_{\mathbf{u}\mu}$ is the vector form of following second-order tangent

$$\mathbb{S}_{iI} = F_{iJ} \frac{\partial S_{JI}}{\partial C^w} \frac{\partial C^w}{\partial \mu^w} \quad \xrightarrow{\text{reshape to vector}} \quad \mathbf{d}_{\mathbf{u}\mu}, \qquad (2.32)$$

2.4.3 Chemo-mechanical tangent matrix, $\mathbf{k}_{\mu\mathbf{u}}^e$

The residual for the mass balance equation of solvent can be written as,

$$R_\mu^a = -\int_{\Omega_0^e} N^a \left(\frac{C^w - C_t^w}{\Delta t} \right) \, dV + \int_{\Omega_0^e} \frac{\partial N^a}{\partial X_I} J_I^w \, dV + \int_{\Gamma_{Iw}^e} N^a I^{we} \, dS. \qquad (2.33)$$

Thus, the components of chemo-mechanical tangent matrix, $\mathbf{k}_{\mu u_k}^{ab}$, can be written as,

$$\mathbf{k}_{\mu u_k}^{ab} = -\frac{\partial R_\mu^a}{\partial u_k^b} = \int\limits_{\Omega_0^e} \left[N^a \left(\frac{1}{\Delta t}\frac{\partial C^w}{\partial F_{mL}} \right) - \frac{\partial N^a}{\partial X_I} \left(\frac{\partial J_I^w}{\partial F_{mL}} + \frac{\partial J_I^w}{\partial C^w}\frac{\partial C^w}{\partial F_{mL}} \right) \right] \frac{\partial F_{mL}}{\partial u_k^b} \; dV,$$

$$= \int\limits_{\Omega_0^e} \left[N^a \left(\frac{1}{\Delta t}\frac{\partial C^w}{\partial F_{mL}} \right) - \frac{\partial N^a}{\partial X_I} \left(\frac{\partial J_I^w}{\partial F_{mL}} + \frac{\partial J_I^w}{\partial C^w}\frac{\partial C^w}{\partial F_{mL}} \right) \right] \frac{\partial}{\partial u_k^b} \left(\delta_{mL} + \sum_{b=1}^{\text{nnode}} \frac{\partial N^b}{\partial X_J}u_m^b \right) \; dV,$$

$$= \int\limits_{\Omega_0^e} \left[N^a \left(\frac{1}{\Delta t}\frac{\partial C^w}{\partial F_{mL}} \right) - \frac{\partial N^a}{\partial X_I} \left(\frac{\partial J_I^w}{\partial F_{mL}} + \frac{\partial J_I^w}{\partial C^w}\frac{\partial C^w}{\partial F_{mL}} \right) \right] \frac{\partial N^b}{\partial X_L}\delta_{mk} \; dV,$$

$$= \int\limits_{\Omega_0^e} N^a \left(\frac{1}{\Delta t}\frac{\partial C^w}{\partial F_{mL}} \right) \frac{\partial N^b}{\partial X_L}\delta_{mk} \; dV - \int\limits_{\Omega_0^e} \frac{\partial N^a}{\partial X_I} \left(\frac{\partial J_I^w}{\partial F_{mL}} + \frac{\partial J_I^w}{\partial C^w}\frac{\partial C^w}{\partial F_{mL}} \right) \frac{\partial N^b}{\partial X_L}\delta_{mk} \; dV,$$

$$= \int\limits_{\Omega_0^e} N^a \left(\frac{1}{\Delta t}\frac{\partial C^w}{\partial F_{kL}} \right) \frac{\partial N^b}{\partial X_L} \; dV - \int\limits_{\Omega_0^e} \frac{\partial N^a}{\partial X_I} \left(\frac{\partial J_I^w}{\partial F_{kL}} + \frac{\partial J_I^w}{\partial C^w}\frac{\partial C^w}{\partial F_{kL}} \right) \frac{\partial N^b}{\partial X_L} \; dV,$$

$$= \int\limits_{\Omega_0^e} N^a \left(\frac{1}{\Delta t}\frac{\partial C^w}{\partial F_{kL}} \right) \frac{\partial N^b}{\partial X_L} \; dV - \int\limits_{\Omega_0^e} \frac{\partial N^a}{\partial X_I} \mathbb{J}_{IkL} \frac{\partial N^b}{\partial X_L} \; dV.$$

$$(2.34)$$

In matrix form, I can write,

$$\left[\mathbf{k}_{\mu\mathbf{u}}^e \right]_{\text{nnode}\times\text{nnode}*\text{ndim}} = \int\limits_{\Omega_0^e} \left(\mathbf{N}_\mu^\top \frac{\partial C^w}{\partial \mathbf{F}}\mathbf{G_u} - \mathbf{B}_\mu^\top \mathbf{D}_{\mu\mathbf{u}}\mathbf{G_u} \right) \; dV \qquad (2.35)$$

where, $\frac{\partial C^w}{\partial \mathbf{F}}$ is reshaped into a vector of dimension $\left[\frac{\partial C^w}{\partial \mathbf{F}} \right]_{1\times\text{ndim}^2}$, and $\mathbf{D}_{\mu\mathbf{u}}$ is the matrix form of following third-order tangent,

$$(\mathbb{J})_{IkL} = \frac{\partial J_I^w}{\partial F_{kL}} + \frac{\partial J_I^w}{\partial C^w}\frac{\partial C^w}{\partial F_{kL}} \qquad \xrightarrow{\text{reshape to matrix}} \qquad \mathbf{D}_{\mu\mathbf{u}}. \qquad (2.36)$$

2.4.4 Chemical tangent matrix, $k_{\mu\mu}^e$

Finally, the chemical tangent matrix, $k_{\mu\mu}^{ab}$, is given by,

$$k_{\mu\mu}^{ab} = -\frac{\partial R_\mu^a}{\partial \mu^{wb}},$$

$$= \int\limits_{\Omega_0^e} N^a \left(\frac{1}{\Delta t}\frac{\partial C^w}{\partial \mu^w} \right) \frac{\partial \mu^w}{\partial \mu^{wb}} \; dV - \int\limits_{\Omega_0^e} \frac{\partial N^a}{\partial X_I}\frac{\partial J_I^w}{\partial C^w}\frac{\partial C^w}{\partial \mu^w}\frac{\partial \mu^w}{\partial \mu^{wb}} \; dV - \int\limits_{\Omega_0^e} \frac{\partial N^a}{\partial X_I}\frac{\partial J_I^w}{\partial (\text{Grad }\mu^w)}N^b \; dV,$$

$$= \int\limits_{\Omega_0^e} N^a \left(\frac{1}{\Delta t}\frac{\partial C^w}{\partial \mu^w} \right) N^b \; dV - \int\limits_{\Omega_0^e} \frac{\partial N^a}{\partial X_I} \left(\frac{\partial J_I^w}{\partial C^w}\frac{\partial C^w}{\partial \mu^w} \right) N^b \; dV - \int\limits_{\Omega_0^e} \frac{\partial N^a}{\partial X_I} \left(\frac{\partial J_I^w}{\partial (\text{Grad }\mu^w)_J} \right) \frac{\partial N^b}{\partial X_J} \; dV,$$

$$(2.37)$$

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Let define,

$$(\mathbf{m}_{\mu\mu})_I = \frac{\partial J_I^w}{\partial C^w} \frac{\partial C^w}{\partial \mu^w}.$$ (2.38)

Additionally, by recognizing $\frac{\partial J_I^w}{\partial (\mathrm{Grad}\,\mu^w)_J} = -M_{IJ}^w$ is the mobility tensor, the matrix form of the chemical tangent can be written as,

$$\left[\mathbf{k}_{\mu\mu}^e\right]_{\mathrm{nnode}\times\mathrm{nnode}} = \int_{\Omega_0^e} \left(\mathbf{N}_\mu^\top \frac{\partial C^w}{\partial \mu^w} \mathbf{N}_\mu - \mathbf{B}_\mu^\top \mathbf{m}_{\mu\mu} \mathbf{N}_\mu + \mathbf{B}_\mu^\top \mathbf{M}^w \mathbf{B}_\mu \right)\, dV,$$ (2.39)

2.4.5 Matrix form of element residual vectors

The members of the element residual vector, $\mathbf{R}^e$, are given by,

$$[\mathbf{R}_\mathbf{u}^e]_{\mathrm{nnode}*\mathrm{ndim}\times 1} = -\int_{\Omega_0^e} (\mathbf{B_u}\Sigma_\mathbf{F})^\top \mathbf{S}\, dV + \int_{\Omega_0^e} \rho_\mathrm{R}\mathbf{N}_\mathbf{u}^\top \mathbf{B}^e\, dV + \int_{\Gamma_T^e} \mathbf{N}_\mathbf{u}^\top \mathbf{T}^e\, dS,$$

$$\text{and} \quad \left[\mathbf{R}_\mu^e\right]_{\mathrm{nnode}\times 1} = -\int_{\Omega_0^e} \mathbf{N}_\mu^\top \left(\frac{C^w - C_t^w}{\Delta t}\right)\, dV + \int_{\Omega_0^e} \mathbf{B}_\mu^\top \mathbf{J}^w\, dV + \int_{\Gamma_{Iw}^e} \mathbf{N}_\mu^\top I^{we}\, dS.$$ (2.40)

2.5 Summary of the element tangent matrix and residual vector components

Using the index notations, the components of the element residual vector can be expressed as follows,

$$R_\mathbf{u}^a = -\int_{\Omega_0^e} \frac{\partial N^a}{\partial X_I} F_{iJ} S_{JI}\, dV + \int_{\Omega_0^e} \rho_\mathrm{R} N^a B_i^e\, dV + \int_{\Gamma_T^e} N^a T_i^e\, dS = 0,$$

$$R_\mu^a = -\int_{\Omega_0^e} N^a \left(\frac{C^w - C_t^w}{\Delta t}\right)\, dV + \int_{\Omega_0^e} \frac{\partial N^a}{\partial X_I} J_I^w\, dV + \int_{\Gamma_{Iw}^e} N^a I^{we}\, dS.$$ (2.41)

Similarly, using index notation, the components of the element tangent matrix can be

expressed as below,

$$k_{u_i u_k}^{ab} = \int\limits_{\Omega_0^e} \frac{\partial N^a}{\partial X_J} S_{JL} \delta_{ik} \frac{\partial N^b}{\partial X_L} \, dV + \int\limits_{\Omega_0^e} \frac{\partial N^a}{\partial X_J} F_{iI} \left[2 \left(\frac{\partial S_{JI}}{\partial C_{KL}} + \frac{\partial S_{JI}}{\partial C^w} \frac{\partial C^w}{\partial C_{kL}} \right) \right] F_{kK} \frac{\partial N^b}{\partial X_L} \, dV,$$

$$k_{u_i \mu}^{ab} = \int\limits_{\Omega_0^e} \frac{\partial N^a}{\partial X_I} \left(F_{iJ} \frac{\partial S_{JI}}{\partial C^w} \frac{\partial C^w}{\partial \mu^w} \right) N^b \, dV,$$

$$k_{\mu u_k}^{ab} = \int\limits_{\Omega_0^e} N^a \left(\frac{1}{\Delta t} \frac{\partial C^w}{\partial F_{kL}} \right) \frac{\partial N^b}{\partial X_L} \, dV - \int\limits_{\Omega_0^e} \frac{\partial N^a}{\partial X_I} \left(\frac{\partial J_I^w}{\partial F_{kL}} + \frac{\partial J_I^w}{\partial C^w} \frac{\partial C^w}{\partial F_{kL}} \right) \frac{\partial N^b}{\partial X_L} \, dV,$$

$$k_{\mu \mu}^{ab} = \int\limits_{\Omega_0^e} N^a \left(\frac{1}{\Delta t} \frac{\partial C^w}{\partial \mu^w} \right) N^b \, dV - \int\limits_{\Omega_0^e} \frac{\partial N^a}{\partial X_I} \left(\frac{\partial J_I^w}{\partial C^w} \frac{\partial C^w}{\partial \mu^w} \right) N^b \, dV - \int\limits_{\Omega_0^e} \frac{\partial N^a}{\partial X_I} \left(\frac{\partial J_I^w}{\partial (\mathrm{Grad}\, \mu^w)_J} \right) \frac{\partial N^b}{\partial X_J} \, dV.$$

$$(2.42)$$

The matrix forms of element residual vector components are given by,

$$[\mathbf{R_u^e}]_{\mathrm{nnode*ndim} \times 1} = - \int\limits_{\Omega_0^e} (\mathbf{B_u \Sigma_F})^\top \mathbf{S} \, dV + \int\limits_{\Omega_0^e} \rho_{\mathrm{R}} \mathbf{N_u^\top B}^e \, dV + \int\limits_{\Gamma_T^e} \mathbf{N_u^\top T}^e \, dS,$$

$$\text{and} \quad \left[\mathbf{R_\mu^e}\right]_{\mathrm{nnode} \times 1} = - \int\limits_{\Omega_0^e} \mathbf{N_\mu^\top} \left(\frac{C^w - C_t^w}{\Delta t} \right) \, dV + \int\limits_{\Omega_0^e} \mathbf{B_\mu^\top J}^w \, dV + \int\limits_{\Gamma_{Iw}^e} \mathbf{N_\mu^\top} I^{we} \, dS.$$

$$(2.43)$$

The matrix forms of the different element tangent sub-matrices are given by,

$$[\mathbf{k_{uu}^e}]_{\mathrm{nnode*ndim} \times \mathrm{nnode*ndim}} = -\frac{\partial \mathbf{R_u^e}}{\partial \mathbf{u}^e} = \int\limits_{\Omega_0^e} \left[\mathbf{G_u^\top \Sigma_S G_u} + (\mathbf{B_u \Sigma_F})^\top \mathbf{D_\mathbb{C}} (\mathbf{B_u \Sigma_F}) \right] \, dV,$$

$$\left[\mathbf{k_{u\mu}^e}\right]_{\mathrm{nnode*ndim} \times \mathrm{nnode}} = -\frac{\partial \mathbf{R_u^e}}{\partial \mu^{we}} = \int\limits_{\Omega_0^e} \mathbf{G_u^\top d_{u\mu} N_\mu} \, dV,$$

$$\left[\mathbf{k_{\mu u}^e}\right]_{\mathrm{nnode} \times \mathrm{nnode*ndim}} = -\frac{\partial \mathbf{R_\mu^e}}{\partial \mathbf{u}^e} = \int\limits_{\Omega_0^e} \left(\mathbf{N_\mu^\top} \left(\frac{1}{\Delta t} \frac{\partial C^w}{\partial \mathbf{F}} \right) \mathbf{G_u} - \mathbf{B_\mu^\top D_{\mu u} G_u} \right) \, dV,$$

$$\left[\mathbf{k_{\mu \mu}^e}\right]_{\mathrm{nnode} \times \mathrm{nnode}} = -\frac{\partial \mathbf{R_\mu^e}}{\partial \mu^{we}} = \int\limits_{\Omega_0^e} \left(\mathbf{N_\mu^\top} \left(\frac{1}{\Delta t} \frac{\partial C^w}{\partial \mu^w} \right) \mathbf{N_\mu} - \mathbf{B_\mu^\top m_{\mu \mu} N_\mu} + \mathbf{B_\mu^\top M}^w \mathbf{B_\mu} \right) \, dV,$$

$$(2.44)$$

Here, the non-symmetric gradient matrix, $[\mathbf{G_u}]$, in the geometric stiffness term appears as,

$$\mathbf{G_u} = \left[\mathbf{G_u^1} \quad \mathbf{G_u^2} \quad \mathbf{G_u^3} \quad \cdots \quad \cdots \quad \mathbf{G_u^{\mathrm{nnode}}} \right]_{\mathrm{ndim}^2 \times \mathrm{nnode*ndim}}, \tag{2.45}$$

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where the sub-matrix, $[\mathbf{G}_\mathbf{u}^a]$, for two-dimensional and three-dimensional cases are given by,

$$\mathbf{G}_\mathbf{u}^a = \begin{bmatrix} N_{,1}^a & 0 \\ 0 & N_{,1}^a \\ N_{,2}^a & 0 \\ 0 & N_{,2}^a \end{bmatrix}_{\mathrm{ndim}^2 \times \mathrm{ndim}} , \qquad \mathbf{G}_\mathbf{u}^a = \begin{bmatrix} N_{,1}^a & 0 & 0 \\ 0 & N_{,1}^a & 0 \\ 0 & 0 & N_{,1}^a \\ N_{,2}^a & 0 & 0 \\ 0 & N_{,2}^a & 0 \\ 0 & 0 & N_{,2}^a \\ N_{,3}^a & 0 & 0 \\ 0 & N_{,3}^a & 0 \\ 0 & 0 & N_{,3}^a \end{bmatrix}_{\mathrm{ndim}^2 \times \mathrm{ndim}} . \qquad (2.46)$$

$[\Sigma_\mathbf{S}]$ is the stress matrix. For a two-dimensional and a three-dimensional case, it is given by,

$$\Sigma_\mathbf{S} = \begin{bmatrix} \mathbf{S}_{11} & 0 & \mathbf{S}_{12} & 0 \\ 0 & \mathbf{S}_{11} & 0 & \mathbf{S}_{12} \\ \mathbf{S}_{12} & 0 & \mathbf{S}_{22} & 0 \\ 0 & \mathbf{S}_{12} & 0 & \mathbf{S}_{22} \end{bmatrix} , \quad \Sigma_\mathbf{S} = \begin{bmatrix} \mathbf{S}_{11} & 0 & 0 & \mathbf{S}_{12} & 0 & 0 & \mathbf{S}_{13} & 0 & 0 \\ 0 & \mathbf{S}_{11} & 0 & 0 & \mathbf{S}_{12} & 0 & 0 & \mathbf{S}_{13} & 0 \\ 0 & 0 & \mathbf{S}_{11} & 0 & 0 & \mathbf{S}_{12} & 0 & 0 & \mathbf{S}_{13} \\ \mathbf{S}_{12} & 0 & 0 & \mathbf{S}_{22} & 0 & 0 & \mathbf{S}_{23} & 0 & 0 \\ 0 & \mathbf{S}_{12} & 0 & 0 & \mathbf{S}_{22} & 0 & 0 & \mathbf{S}_{23} & 0 \\ 0 & 0 & \mathbf{S}_{12} & 0 & 0 & \mathbf{S}_{22} & 0 & 0 & \mathbf{S}_{23} \\ \mathbf{S}_{13} & 0 & 0 & \mathbf{S}_{23} & 0 & 0 & \mathbf{S}_{33} & 0 & 0 \\ 0 & \mathbf{S}_{13} & 0 & 0 & \mathbf{S}_{23} & 0 & 0 & \mathbf{S}_{33} & 0 \\ 0 & 0 & \mathbf{S}_{13} & 0 & 0 & \mathbf{S}_{23} & 0 & 0 & \mathbf{S}_{33} \end{bmatrix} . $$
$$(2.47)$$

$[\Sigma_\mathbf{S}]$ has a dimension of $[\Sigma_\mathbf{S}]_{\mathrm{ndim}^2 \times \mathrm{ndim}^2}$. It is also possible to represent $\mathbf{G}_\mathbf{u}^a$ and consequently $\mathbf{G}_\mathbf{u}$ and $\Sigma_\mathbf{S}$ matrices in alternative matrix forms which will essentially give the same result (de Borst et al., 2012; Reddy, 2015).

$[\Sigma_\mathbf{F}]_{\mathrm{nnode*ndim} \times \mathrm{nnode*ndim}}$ is a square banded diagonal matrix of dimension $\mathrm{nnode} * \mathrm{ndim} \times \mathrm{nnode} * \mathrm{ndim}$, and for a three-dimensional case, it appears as,

$$\Sigma_\mathbf{F} = \begin{bmatrix} F_{11} & F_{12} & F_{13} & 0 & 0 & 0 & \cdots & 0 & 0 & 0 \\ F_{21} & F_{22} & F_{23} & 0 & 0 & 0 & \cdots & 0 & 0 & 0 \\ F_{31} & F_{32} & F_{33} & 0 & 0 & 0 & \cdots & 0 & 0 & 0 \\ & & & \cdots & & & & & & \\ & & & \cdots & & & & & & \\ & & & \cdots & & & & & & \\ & & & \cdots & & & & & & \\ & & & \cdots & & & & & & \\ & & & \cdots & & & & & & \\ 0 & 0 & 0 & 0 & 0 & 0 & \cdots & F_{11} & F_{12} & F_{13} \\ 0 & 0 & 0 & 0 & 0 & 0 & \cdots & F_{21} & F_{22} & F_{23} \\ 0 & 0 & 0 & 0 & 0 & 0 & \cdots & F_{31} & F_{32} & F_{33} \end{bmatrix}_{\mathrm{nnode*ndim} \times \mathrm{nnode*ndim}} \qquad (2.48)$$

For a two-dimensional plane strain case, I can eliminate the rows and columns related to the

third dimension and reduce the size of $\Sigma_{\mathbf{F}}$.

2.6 PK-II stress-based total Lagrangian F-bar formulation

Since I considered the hydrogel quasi-incompressible, volumetric locking could happen under certain loading conditions. Similar to the hyperelastic materials, I employed the F-bar approach (Chester et al., 2015; Neto et al., 1996) to mitigate the volumetric locking in 4-noded bilinear quadrilateral element and 8-noded trilinear hexahedral element. I should note that, for a coupled chemo-mechanical model, the modified deformation gradient, $\overline{\mathbf{F}}$ will cause changes to $\mathbf{k}_{\mathbf{u}\mu}$ and $\mathbf{k}_{\mu\mathbf{u}}$ besides $\mathbf{k}_{\mathbf{uu}}$. However, as of now, these changes have been ignored, and only the mechanical part of the element tangent matrix, $\mathbf{k}_{\mathbf{uu}}$, is modified.

2.6.1 Three-dimensional case

$$\overline{\mathbf{F}} = \mathbf{F}^{\mathrm{dev}}\mathbf{F}_0^{\mathrm{vol}} = \left(\frac{\det \mathbf{F}_0}{\det \mathbf{F}}\right)^{1/3}\mathbf{F}. \tag{2.49}$$

For three-dimensional cases, using the definition of the second Piola-Kirchhoff stress, $\mathbf{S}$, the matrix form of element residual vector, $\mathbf{R}_{\mathbf{u}}^e(\mathbf{u}^e)$, is given by,

$$\mathbf{R}_{\mathbf{u}}^e(\mathbf{u}^e) = -\int_{\Omega_0^e} \left(\frac{\det \mathbf{F}_0}{\det \mathbf{F}}\right)^{-2/3}(\mathbf{B}_{\mathbf{u}}\Sigma_{\mathbf{F}})^{\top}\mathbf{S}\ dV + \int_{\Omega_0^e} \rho_{\mathrm{R}}\mathbf{N}_{\mathbf{u}}^{\top}\mathbf{B}^e\ dV + \int_{\Gamma_T^e} \mathbf{N}_{\mathbf{u}}^{\top}\mathbf{T}^e\ dS \tag{2.50}$$

The matrix form of the element tangent matrix is given by,

$$\begin{aligned}
[\mathbf{k}_{\mathbf{uu}}^e] &= \int_{\Omega_0^e} \left(\frac{\det \mathbf{F}_0}{\det \mathbf{F}}\right)^{-1/3}\left[\mathbf{G}_{\mathbf{u}}^{\top}\Sigma_{\mathbf{S}}\mathbf{G}_{\mathbf{u}} + (\mathbf{B}_{\mathbf{u}}\Sigma_{\mathbf{F}})^{\top}\mathbf{D}_{\mathbb{C}}(\mathbf{B}_{\mathbf{u}}\Sigma_{\mathbf{F}})\right]\ dV \\
&+ \int_{\Omega_0^e} \left[\left(\frac{\det \mathbf{F}_0}{\det \mathbf{F}}\right)^{-2/3}\left(\mathbf{G}_{\mathbf{u}}^{\top}\mathbf{Q}_{\mathrm{R}}^0\mathbf{G}_{\mathbf{u}}^0 - \mathbf{G}_{\mathbf{u}}^{\top}\mathbf{Q}_{\mathrm{R}}\mathbf{G}_{\mathbf{u}}\right)\right]\ dV
\end{aligned} \tag{2.51}$$

where, $\mathbf{Q}_{\mathrm{R}}^0$ and $\mathbf{Q}_{\mathrm{R}}$ are the matrix form of the following fourth-order tensors

$$\begin{aligned}
(\mathbb{Q}_{\mathrm{R}}^0)_{iJkL} &= \frac{1}{3}\overline{F}_{iI}\left[2\left(\frac{\partial \overline{S}_{IJ}}{\partial \overline{C}_{MN}} + \frac{\partial \overline{S}_{IJ}}{\partial C^w}\frac{\partial C^w}{\partial \overline{C}_{kL}}\right)\right]\overline{C}_{MN}F_{0,kL}^{-\top} - \frac{1}{3}\overline{P}_{iJ}F_{0,kL}^{-\top}, \\
(\mathbb{Q}_{\mathrm{R}})_{iJkL} &= \frac{1}{3}\overline{F}_{iI}\left[2\left(\frac{\partial \overline{S}_{IJ}}{\partial \overline{C}_{MN}} + \frac{\partial \overline{S}_{IJ}}{\partial C^w}\frac{\partial C^w}{\partial \overline{C}_{kL}}\right)\right]\overline{C}_{MN}F_{kL}^{-\top} - \frac{1}{3}\overline{P}_{iJ}F_{kL}^{-\top}.
\end{aligned} \tag{2.52}$$

2.6.2 Plane strain case

For two-dimensional plane strain cases, the modified deformation gradient, $\overline{\mathbf{F}}$, needs to be computed as,

$$\overline{\mathbf{F}} = \left[\begin{array}{c|c} [\overline{\mathbf{F}}_{\text{pe}}] & \begin{matrix} 0 \\ 0 \end{matrix} \\ \hline 0 \quad 0 & 1 \end{array} \right], \tag{2.53}$$

where, $[\overline{\mathbf{F}}_{\text{pe}}]_{2\times 2}$ is defined as the following

$$\overline{\mathbf{F}}_{\text{pe}} = \mathbf{F}^{\text{dev}}\mathbf{F}_0^{\text{vol}} = \left(\frac{\det \mathbf{F}_0}{\det \mathbf{F}} \right)^{1/2} \mathbf{F}_{\text{pe}}, \quad \text{where,} \quad \mathbf{F}_{\text{pe}} = \begin{bmatrix} F_{11} & F_{12} \\ F_{21} & F_{22} \end{bmatrix}. \tag{2.54}$$

Using the definition of the second Piola-Kirchhoff stress, for plane-strain case, the matrix form of element residual vector, $\mathbf{R}_{\mathbf{u}}^e(\mathbf{u}^e)$, can be written as,

$$\mathbf{R}_{\mathbf{u}}^e(\mathbf{u}^e) = - \int_{\Omega_0^e} \left(\frac{\det \mathbf{F}_0}{\det \mathbf{F}} \right)^{-1/2} (\mathbf{B}_{\mathbf{u}}\Sigma_{\mathbf{F}})^{\top}\mathbf{S} \; dV + \int_{\Omega_0^e} \rho_R \mathbf{N}_{\mathbf{u}}^{\top}\mathbf{B}^e \; dV + \int_{\Gamma_T^e} \mathbf{N}_{\mathbf{u}}^{\top}\mathbf{T}^e \; dS \tag{2.55}$$

The matrix form of the element tangent matrix is given by,

$$[\mathbf{k}_{\mathbf{uu}}^e] = \int_{\Omega_0^e} \left[\mathbf{G}_{\mathbf{u}}^{\top}\Sigma_{\mathbf{S}}\mathbf{G}_{\mathbf{u}} + (\mathbf{B}_{\mathbf{u}}\Sigma_{\mathbf{F}})^{\top}\mathbf{D}_{\mathbb{C}}(\mathbf{B}_{\mathbf{u}}\Sigma_{\mathbf{F}}) \right] \; dV$$

$$+ \int_{\Omega_0^e} \left[\left(\frac{\det \mathbf{F}_0}{\det \mathbf{F}} \right)^{-1/2} \mathbf{G}_{\mathbf{u}}^{\top} \left(\mathbf{Q}_R^0 \mathbf{G}_{\mathbf{u}}^0 - \mathbf{Q}_R \mathbf{G}_{\mathbf{u}} \right) \right] \; dV \tag{2.56}$$

where, $\mathbf{Q}_R^0$ and $\mathbf{Q}_R$ are the matrix form of the following fourth-order tensors

$$(\mathbb{Q}_R^0)_{iJkL} = \frac{1}{2}\overline{F}_{iI} \left[2 \left(\frac{\partial \overline{S}_{IJ}}{\partial \overline{C}_{MN}} + \frac{\partial \overline{S}_{IJ}}{\partial C^w}\frac{\partial C^w}{\partial \overline{C}_{kL}} \right) \right] \overline{C}_{MN} F_{0,kL}^{-\top},$$

$$(\mathbb{Q}_R)_{iJkL} = \frac{1}{2}\overline{F}_{iI} \left[2 \left(\frac{\partial \overline{S}_{IJ}}{\partial \overline{C}_{MN}} + \frac{\partial \overline{S}_{IJ}}{\partial C^w}\frac{\partial C^w}{\partial \overline{C}_{kL}} \right) \right] \overline{C}_{MN} F_{kL}^{-\top}. \tag{2.57}$$

2.7 Some remarks on the tangents and matrix operators

1. Contribution of deformation-dependent body force, traction, and flux was ignored in the stiffness (tangent) matrix.

2. $\mathbf{N}_{\mathbf{u}}$ is the shape function matrix of dimension $[\mathbf{N}_{\mathbf{u}}]_{\text{ndim}\times\text{nnode}*\text{ndim}}$.

3. $\mathbf{B}_{\mathbf{u}} = \text{sym}(\text{Grad}(\mathbf{N}_{\mathbf{u}}))$ is the symmetric gradient matrix of shape functions (also called strain-displacement matrix) of dimension $[\mathbf{B}_{\mathbf{u}}]_{\text{nstress}\times\text{nnode}*\text{ndim}}$.

4. $\mathbf{G}_{\mathbf{u}} = \text{Grad}(\mathbf{N}_{\mathbf{u}})$ is referred to as the non-symmetric gradient matrix which has a

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dimension of $[\mathbf{G_u}]_{\text{ndim}^2 \times \text{nnode}*\text{ndim}}$.

5. $\Sigma_{\mathbf{S}}$ and $\Sigma_{\mathbf{F}}$ have dimensions of $[\Sigma_{\mathbf{S}}]_{\text{ndim}^2 \times \text{ndim}^2}$ and $[\Sigma_{\mathbf{F}}]_{\text{nnode}*\text{ndim} \times \text{nnode}*\text{ndim}}$, respectively.

6. $\mathbf{N}_\mu$ is a vector of dimension of $[\mathbf{N}_\mu]_{1 \times \text{nnode}}$.

7. $\mathbf{B}_\mu = \text{Grad}(\mathbf{N}_\mu)$ is a matrix of dimension $[\mathbf{B}_\mu]_{\text{ndim} \times \text{nnode}}$.

8. $[\mathbf{D}_{\mathbb{C}}]_{\text{nstress} \times \text{nstress}}$ in the mechanical element stiffness (tangent) matrix, $\mathbf{k}^e_{\mathbf{uu}}$, is the Voigt stiffness matrix mapped from the material tangent, $\mathbb{C}$, defined as,

$$\mathbb{C}_{IJKL} = 2 \left(\frac{\partial S_{IJ}}{\partial C_{KL}} + \frac{\partial S_{JI}}{\partial C^w} \frac{\partial C^w}{\partial C_{KL}} \right)$$

9. $\frac{\partial \dot{C}^w}{\partial \mathbf{F}}$ is a vector of dimension $\left[\frac{\partial \dot{C}^w}{\partial \mathbf{F}} \right]_{1 \times \text{ndim}^2}$.

10. $\mathbf{d}_{\mathbf{u}\mu}$ is a vector of dimension $[\mathbf{d}_{\mathbf{u}\mu}]_{\text{ndim}^2 \times 1}$ obtained from the second-order mechano-chemical tangent tensor, $\mathbb{S}$, defined as,

$$\mathbb{S}_{iI} = F_{iJ} \frac{\partial S_{IJ}}{\partial C^w} \frac{\partial C^w}{\partial \mu^w}.$$

11. $\mathbf{D}_{\mu\mathbf{u}}$ is a matrix of dimension $[\mathbf{D}_{\mu\mathbf{u}}]_{\text{ndim} \times \text{ndim}^2}$ obtained from the third-order chemo-mechanical tangent tensor, $\mathbb{J}$ defined as,

$$(\mathbb{J})_{IkL} = \frac{\partial J^w_I}{\partial F_{kL}} + \frac{\partial J^w_I}{\partial C^w} \frac{\partial C^w}{\partial F_{kL}}$$

12. $\mathbf{m}_{\mu\mu}$ is a vector of dimension $[\mathbf{m}_{\mu\mu}]_{\text{ndim} \times 1}$ defined as,

$$(\mathbf{m}_{\mu\mu})_I = \frac{\partial J^w_I}{\partial C^w} \frac{\partial C^w}{\partial \mu^w}. \tag{2.58}$$

3 Constitutive model-specific tangent moduli (derivatives)

Calculating the stiffness (tangent) matrix for the chemo-mechanical element $(\mathbf{u} - \mu)$ requires evaluating a few derivative terms that depend on the material-specific constitutive choices. In this section, I will derive those terms based on the constitutive model chosen for hydrogel. However, before that, I need to calculate the internal variable, ϕ^p, as it appears in the constitutive relations using a local Newton-Raphson iteration.

3.1 Local iteration to calculate polymer volume fraction

At every time step, the solution of the nodal variables, *i.e.*, displacement vector, $\mathbf{u}$, and solvent chemical potential, μ^w, are passed to the user element subroutine in Abaqus. Based on the nodal solutions, the deformation gradient, chemical potential, and gradient of chemical potential are calculated at each integration point in the element subroutine using the

interpolation functions and their derivatives. Consequently, these variables are passed to the material point calculation subroutine, `umatHydrogel` (see next section). In the material point subroutine, necessary tangents and constitutive expressions are evaluated using the formulations from the previous section and passed back to the element subroutine to form the components of the tangent matrix and the residual vector to perform the calculation in the next time step.

To calculate the material response, firstly, the polymer volume fraction is calculated. With chemical potential being known at the current time, the following nonlinear constitutive expression for chemical potential is solved using the Newton-Raphson method.

$$\mathcal{G}(\phi^p) = \mu_w^0 - \mu^w + R\theta \left[\phi^p + \ln(1 - \phi^p) + \chi(\phi^p)^2\right] - \kappa\mathcal{V}^w\left[\ln\left(\frac{J\phi^p}{\phi_0^p}\right)\right] + \frac{\kappa\mathcal{V}^w}{2}\left[\ln\left(\frac{J\phi^p}{\phi_0^p}\right)\right]^2 = 0.$$

(3.1)

The derivative used in the Newton-Raphson method is given by,

$$\frac{d\mathcal{G}(\phi^p)}{d\phi^p} = R\theta\left[1 - \frac{1}{1 - \phi^p} + 2\chi\phi^p\right] - \frac{\kappa\mathcal{V}^w}{\phi^p} + \frac{\kappa\mathcal{V}^w}{\phi^p}\ln\left(\frac{J\phi^p}{\phi_0^p}\right).$$

(3.2)

I should note that, since J^e is unknown in the expression for chemical potential, it has been substituted with the known parameter (J, ϕ_0^p) and unknown variable of interest, ϕ^p. Once the polymer volume fraction is calculated, referential concentrations for the polymer and the solvent are calculated. These values are used in calculating different tangents with the exception being the tangent, $\frac{\partial \dot{C}^w}{\partial \mu^w}$. By perturbing the chemical potential in the above equation, polymer volume fractions are calculated for two different conditions and consequently used in calculating the tangent.

3.2 Calculation of $\dfrac{\partial C^w}{\partial \mu^w}$

The constitutive relation for the solvent chemical potential is given by,

$$\mu^w = \mu_w^0 + R\theta\left[\phi^p + \ln(1 - \phi^p) + \chi(\phi^p)^2\right] - \kappa\mathcal{V}^w(\ln J^e) + \frac{\kappa\mathcal{V}^w}{2}(\ln J^e)^2,$$

$$\Rightarrow \mu_w^0 + R\theta\left[\phi^p + \ln(1 - \phi^p) + \chi(\phi^p)^2\right] - \kappa\mathcal{V}^w\left[\ln\left(\frac{J\phi^p}{\phi_0^p}\right)\right] + \frac{\kappa\mathcal{V}^w}{2}\left[\ln\left(\frac{J\phi^p}{\phi_0^p}\right)\right]^2 - \mu^w = 0,$$

$$\Rightarrow \mathcal{G}(\mathbf{C}(\mathbf{F}), \mu^w, \phi^p(C^w)) = 0.$$

(3.3)

Thus, the partial derivative of the implicit constitutive function, $\mathcal{G}$, with respect to μ^w can be written as,

$$\frac{d\mathcal{G}}{d\mu^w} = \left.\frac{\partial \mathcal{G}}{\partial \mathbf{F}}\right|_{\mu^w,\phi^p} \frac{\partial \mathbf{F}}{\partial \mu^w} + \left.\frac{\partial \mathcal{G}}{\partial \mu^w}\right|_{\mathbf{F},\phi^p} \frac{\partial \mu^w}{\partial \mu^w} + \left.\frac{\partial \mathcal{G}}{\partial \phi^p}\right|_{\mathbf{F},\mu^w} \frac{\partial \phi^p}{\partial C^w} \frac{\partial C^w}{\partial \mu^w},$$

$$\Rightarrow \left.\frac{\partial \mathcal{G}}{\partial \mu^w}\right|_{\mathbf{F},\phi^p} + \left.\frac{\partial \mathcal{G}}{\partial \phi^p}\right|_{\mathbf{F},\mu^w} \frac{\partial \phi^p}{\partial C^w} \frac{\partial C^w}{\partial \mu^w} = 0, \qquad \left(\text{since, } \frac{\partial \mu^w}{\partial \mu^w} = 1 \text{ and } \frac{\partial \mathbf{F}}{\partial \mu^w} = 0\right), \quad (3.4)$$

$$\Rightarrow \frac{\partial C^w}{\partial \mu^w} = -\frac{\frac{\partial \mathcal{G}}{\partial \mu^w}}{\frac{\partial \mathcal{G}}{\partial \phi^p}\frac{\partial \phi^p}{\partial C^w}} = \frac{\partial C^w}{\partial \phi^p}\left(\frac{\partial \mathcal{G}}{\partial \phi^p}\right)^{-1} \qquad \left(\text{since, } \frac{\partial \mathcal{G}}{\partial \mu^w} = -1\right)$$

Polymer volume fraction and its derivative with respect to the referential concentration of the solvent are given by,

$$\phi^p = \frac{C^p \mathcal{V}^p}{C^p \mathcal{V}^p + C^w \mathcal{V}^w} \qquad \Rightarrow \qquad \frac{\partial C^w}{\partial \phi^p} = -\frac{C^p \mathcal{V}^p}{\mathcal{V}^w (\phi^p)^2} = -\frac{\phi_0^p}{\mathcal{V}^w (\phi^p)^2}. \qquad (3.5)$$

In writing the second expression for μ^w, I used $J = \phi_0^p J^e J^s$ and $J^s = 1/\phi^p$. Thus, I have,

$$\frac{\partial \mathcal{G}}{\partial \phi^p} = R\theta\left[1 - \frac{1}{1-\phi^p} + 2\chi\phi^p\right] - \frac{\kappa \mathcal{V}^w}{\phi^p} + \frac{2\kappa \mathcal{V}^w}{\phi^p}\ln\left(\frac{J\phi^p}{\phi_0^p}\right). \qquad (3.6)$$

By substituting the expressions for $\frac{\partial C^w}{\partial \phi^p}$ and $\left(\frac{\partial \mathcal{G}}{\partial \phi^p}\right)^{-1}$, I can obtain the expression for $\frac{\partial C^w}{\partial \mu^w}$ during implementation.

3.3   Calculation of $\frac{\partial C^w}{\partial \mathbf{F}}$

We have,

$$\mathcal{G} \equiv \mu_w^0 + R\theta\left[\phi^p + \ln(1-\phi^p) + \chi(\phi^p)^2\right] - \kappa \mathcal{V}^w\left[\ln\left(\frac{J\phi^p}{\phi_0^p}\right)\right] + \frac{\kappa \mathcal{V}^w}{2}\left[\ln\left(\frac{J\phi^p}{\phi_0^p}\right)\right]^2 - \mu^w = 0$$

$$(3.7)$$

Following the same procedure as before, the partial derivative of the implicit constitutive function, $\mathcal{G}$, with respect to $\mathbf{F}$ can be written as,

$$\frac{\partial \mathcal{G}}{\partial \mathbf{F}}\frac{\partial \mathbf{F}}{\partial \mathbf{F}} + \frac{\partial \mathcal{G}}{\partial \mu^w}\frac{\partial \mu^w}{\partial \mathbf{F}} + \frac{\partial \mathcal{G}}{\partial \phi^p}\frac{\partial \phi^p}{\partial C^w}\frac{\partial C^w}{\partial \mathbf{F}} = 0,$$

$$\Rightarrow \frac{\partial C^w}{\partial \mathbf{F}} = -\frac{\frac{\partial \mathcal{G}}{\partial \mathbf{F}}}{\frac{\partial \mathcal{G}}{\partial \phi^p}\frac{\partial \phi^p}{\partial C^w}} = -\left(\frac{\partial \mathcal{G}}{\partial \mathbf{F}}\right)\left(\frac{\partial C^w}{\partial \phi^p}\right)\left(\frac{\partial \mathcal{G}}{\partial \phi^p}\right)^{-1}$$

$$(3.8)$$

Now,

$$\frac{\partial \mathcal{G}}{\partial \mathbf{F}} = -\kappa \mathcal{V}^w \mathbf{F}^{-\top} + \kappa \mathcal{V}^w \ln\left(\frac{J\phi^p}{\phi_0^p}\right) \mathbf{F}^{-\top},$$

$$= \kappa \mathcal{V}^w \left[\ln(J^e) - 1\right] \mathbf{F}^{-\top},$$

$$\Rightarrow \left(\frac{\partial \mathcal{G}}{\partial \mathbf{F}}\right)_{kL} = \kappa \mathcal{V}^w \left[\ln(J^e) - 1\right] F_{kL}^{-\top},$$

$$= \kappa \mathcal{V}^w \left[\ln(J^e) - 1\right] F_{Lk}^{-1}.$$

(3.9)

By substituting the expressions for $\frac{\partial \mathcal{G}}{\partial \mathbf{F}}$, $\frac{\partial C^w}{\partial \phi^p}$, and $\left(\frac{\partial \mathcal{G}}{\partial \phi^p}\right)^{-1}$, I can calculate $\frac{\partial C^w}{\partial \mathbf{F}}$ during implementation.

$$\frac{\partial C^w}{\partial \mathbf{F}} = \left(\frac{\phi_0^p}{\mathcal{V}^w (\phi^p)^2}\right) \left(\frac{\partial \mathcal{G}}{\partial \phi^p}\right)^{-1} \kappa \mathcal{V}^w \left[\ln(J^e) - 1\right] F_{Lk}^{-1}$$

(3.10)

3.4 Calculation of $\dfrac{\partial C^w}{\partial \mathbf{C}}$

$$\left(\frac{\partial C^w}{\partial \mathbf{C}}\right)_{KL} = \frac{\partial C^w}{\partial F_{mN}} \frac{\partial F_{mN}}{\partial C_{KL}}$$

$$= \left(\frac{\partial C^w}{\partial \phi^p}\right) \left(\frac{\partial \mathcal{G}}{\partial \phi^p}\right)^{-1} \left(\kappa \mathcal{V}^w \left[\ln(J^e) - 1\right] F_{Nm}^{-1}\right) \left(\frac{\partial C_{KL}}{\partial F_{mN}}\right)^{-1},$$

$$= \frac{1}{2} \left(\frac{\partial C^w}{\partial \phi^p}\right) \left(\frac{\partial \mathcal{G}}{\partial \phi^p}\right)^{-1} \kappa \mathcal{V}^w \left[\ln(J^e) - 1\right] F_{Nm}^{-1} F_{Lm}^{-1} \delta_{KN},$$

$$= \frac{1}{2} \left(\frac{\partial C^w}{\partial \phi^p}\right) \left(\frac{\partial \mathcal{G}}{\partial \phi^p}\right)^{-1} \kappa \mathcal{V}^w \left[\ln(J^e) - 1\right] C_{KL}^{-1}.$$

(3.11)

3.5 Calculation of $\dfrac{\partial \mathbf{S}}{\partial C^w}$

The second Piola-Kirchhoff stress, $\mathbf{S}$, for a Arruda-Boyce type elastomeric gel is given by,

$$\mathbf{S} = G_0 \left(\frac{\lambda_{\mathrm{L}}}{3\lambda_c} \beta_c\right) \mathbb{1} - \left[(\phi_0^p)^{2/3} G_0 \left(\frac{\lambda_{\mathrm{L}}}{3}\right) - \phi_0^p J^s \kappa (\ln J^e)\right] \mathbf{C}^{-1},$$

(3.12)

On the other hand, the second Piola-Kirchhoff stress, $\mathbf{S}$, for a Neo-Hookean type elastomeric gel is given by,

$$\mathbf{S} = G_0 \mathbb{1} - \left[(\phi_0^p)^{2/3} G_0 - \phi_0^p J^s \kappa (\ln J^e)\right] \mathbf{C}^{-1}, \quad \text{where,} \quad J^s = \frac{1}{\phi_0^p}(C^P \mathcal{V}^p + C^w \mathcal{V}^w)$$

(3.13)

For both type of elastomers, $\frac{\partial \mathbf{S}}{\partial C^w}$ is the same. By substituting $J^e = \frac{J}{\phi_0^p J^s}$, I can compute $\frac{\partial \mathbf{S}}{\partial C^w}$ as,

$$\frac{\partial \mathbf{S}}{\partial C^w} = \kappa \mathcal{V}^w [\ln(J^e) - 1] \mathbf{C}^{-1},$$
$$\Rightarrow \left(\frac{\partial \mathbf{S}}{\partial C^w}\right)_{IJ} = \kappa \mathcal{V}^w [\ln(J^e) - 1] C_{IJ}^{-1}. \tag{3.14}$$

3.6 Calculation of $\mathbb{C}$

Let's recall the definition of the material tangent for the quasi-incompressible hydrogel,

$$\mathbb{C}_{IJKL} = 2 \left(\frac{\partial S_{IJ}}{\partial C_{KL}} + \frac{\partial S_{IJ}}{\partial C^w} \frac{\partial C^w}{\partial C_{KL}} \right) \qquad \Rightarrow \qquad \mathbb{C} = \mathbb{C}^{\text{mech}} + \mathbb{C}^{\text{chem}}. \tag{3.15}$$

To calculate the so-called material tangent, I will make use of the following tensor derivative identities,

$$\frac{\partial \mathbf{F}}{\partial \mathbf{F}} = \mathbb{I} \quad \Rightarrow (\mathbb{I})_{ijkl} = \delta_{ik}\delta_{jl},$$
$$\frac{\partial \ln J}{\partial \mathbf{F}} = \frac{1}{J} \det(\mathbf{F}) \mathbf{F}^{-\top} = \mathbf{F}^{-\top}, \quad \Rightarrow \frac{\partial \ln J}{\partial F_{kL}} = F_{Lk}^{-1},$$
$$\left(\frac{\partial \mathbf{F}^{-1}}{\partial \mathbf{F}}\right)_{IjkL} = -F_{Ik}^{-1} F_{Lj}^{-1}, \quad \left(\frac{\partial \mathbf{F}^{-\top}}{\partial \mathbf{F}}\right)_{iJkL} = -F_{Li}^{-1} F_{Jk}^{-1}, \tag{3.16}$$
$$\left(\frac{\partial \mathbf{C}^{-1}}{\partial \mathbf{C}}\right)_{IJKL} = -\frac{1}{2} \left(C_{IK}^{-1} C_{JL}^{-1} + C_{JK}^{-1} C_{IL}^{-1}\right) = \mathbb{I}_{\mathbf{C}^{-1}}, \qquad [\text{since, } \mathbf{C} = \mathbf{C}^{\top}]$$

Recalling the second Piola-Kirchhoff stress, $\mathbf{S}$, for hydrogels with Arruda-Boyce type elastomer network is given by,

$$\mathbf{S} = G_0 \left(\frac{\lambda_{\text{L}}}{3\lambda_c} \beta_c\right) \mathbb{1} - \left[(\phi_0^p)^{2/3} G_0 \left(\frac{\lambda_{\text{L}}}{3}\right) - \phi_0^p J^s \kappa (\ln J^e)\right] \mathbf{C}^{-1}, \tag{3.17}$$

Thus, the material tangent, $\mathbb{C}$, for hydrogel with an Arruda-Boyce type network can be

calculated as,

$$
\begin{aligned}
\mathbb{C}^{\text{mech}} &= 2\frac{\partial \mathbf{S}}{\partial \mathbf{C}} \\
&= \frac{2G_0\lambda_{\mathrm{L}}}{3}\left(\frac{1}{\lambda_c}\frac{\partial \beta_c}{\partial \mathbf{C}} + \beta_c\frac{\partial \lambda_c^{-1}}{\partial \mathbf{C}}\right)\mathbb{1} + 2\phi_0^p J^s\kappa\left(\frac{\partial \ln J^e}{\partial \mathbf{C}}\right)\mathbf{C}^{-1} - 2\left[\frac{G_0\lambda_{\mathrm{L}}}{3} - \phi_0^p J^s\kappa(\ln J^e)\right]\frac{\partial \mathbf{C}^{-1}}{\partial \mathbf{C}}, \\
&= \frac{G_0}{9\lambda_c^2}\left(\frac{\partial \beta_c}{\partial\left(\frac{\lambda_c}{\lambda_{\mathrm{L}}}\right)} - \frac{\lambda_{\mathrm{L}}}{\lambda_c}\beta_c\right)\mathbb{1}\otimes\mathbb{1} + \phi_0^p J^s\kappa\mathbf{C}^{-1}\otimes\mathbf{C}^{-1} - 2\left[\frac{G_0\lambda_{\mathrm{L}}}{3} - \phi_0^p J^s\kappa(\ln J^e)\right]\mathbb{I}_{\mathbf{C}^{-1}},
\end{aligned}
$$

$$
\text{where, } \frac{\partial \ln J^e}{\partial \mathbf{C}} = \frac{\partial}{\partial \mathbf{C}}\left[\ln\left(\frac{J\phi^p}{\phi_0^p}\right)\right] = \frac{\partial \ln J}{\partial \mathbf{C}}
$$

$$(3.18)$$

For a hydrogel with a Neo-Hookean type elastomeric network, the second Piola-Kirchhoff stress, $\mathbf{S}$, is given by,

$$
\mathbf{S} = G_0\mathbb{1} - \left[(\phi_0^p)^{2/3}G_0 - \phi_0^p J^s\kappa(\ln J^e)\right]\mathbf{C}^{-1} \tag{3.19}
$$

Thus, following the calculation from the previous section, the material tangent, $\mathbb{C}$, for Neo-Hookean type elastomeric gel is given by,

$$
\begin{aligned}
\mathbb{C}^{\text{mech}} &= 2\frac{\partial \mathbf{S}}{\partial \mathbf{C}} = \phi_0^p J^s\kappa\mathbf{C}^{-1}\otimes\mathbf{C}^{-1} - 2\left[(\phi_0^p)^{2/3}G_0 - \phi_0^p J^s\kappa\ln J\right]\mathbb{I}_{\mathbf{C}^{-1}}, \\
\Rightarrow \mathbb{C}^{\text{mech}}_{IJKL} &= \kappa\phi_0^p J^s C_{IJ}^{-1}C_{KL}^{-1} + \left((\phi_0^p)^{2/3}G_0 - \phi_0^p J^s\kappa\ln J^e\right)\left(C_{IK}^{-1}C_{JL}^{-1} + C_{JK}^{-1}C_{IL}^{-1}\right).
\end{aligned}
$$

$$(3.20)$$

Now the chemical part of the material tangent is given by,

$$
\begin{aligned}
\mathbb{C}^{\text{chem}}_{IJKL} &= 2\frac{\partial S_{IJ}}{\partial C^w}\frac{\partial C^w}{\partial C_{KL}}, \\
&= 2\left(\kappa\mathcal{V}^w[\ln(J^e) - 1]C_{IJ}^{-1}\right)\left[\frac{1}{2}\left(\frac{\partial C^w}{\partial\phi^p}\right)\left(\frac{\partial\mathcal{G}}{\partial\phi^p}\right)^{-1}\kappa\mathcal{V}^w[\ln(J^e) - 1]C_{KL}^{-1}\right], \\
&= \left(\frac{\partial C^w}{\partial\phi^p}\right)\left(\frac{\partial\mathcal{G}}{\partial\phi^p}\right)^{-1}(\kappa\mathcal{V}^w[\ln(J^e) - 1])^2 C_{IJ}^{-1}C_{KL}^{-1}, \\
&= \left(\frac{\partial C^w}{\partial\phi^p}\right)\left(\frac{\partial\mathcal{G}}{\partial\phi^p}\right)^{-1}(\kappa\mathcal{V}^w[\ln(J^e) - 1])^2 \mathbf{C}^{-1}\otimes\mathbf{C}^{-1}.
\end{aligned}
$$

$$(3.21)$$

Thus the material tangent, $\mathbb{C}$, is given by,

$$
\begin{aligned}
\mathbb{C}_{IJKL} &= \kappa\phi_0^p J^s C_{IJ}^{-1}C_{KL}^{-1} + \left((\phi_0^p)^{2/3}G_0 - \phi_0^p J^s\kappa\ln J^e\right)\left(C_{IK}^{-1}C_{JL}^{-1} + C_{JK}^{-1}C_{IL}^{-1}\right) \\
&\quad + \left(\frac{\partial C^w}{\partial\phi^p}\right)\left(\frac{\partial\mathcal{G}}{\partial\phi^p}\right)^{-1}(\kappa\mathcal{V}^w[\ln(J^e) - 1])^2 C_{IJ}^{-1}C_{KL}^{-1}
\end{aligned}
$$

$$(3.22)$$

3.7 Calculation of $\mathbb{J}$

Recalling $\mathbb{J}$ is defined as,

$$(\mathbb{J})_{IkL} = \frac{\partial J_I^w}{\partial F_{kL}} + \frac{\partial J_I^w}{\partial C^w}\frac{\partial C^w}{\partial F_{kL}}. \tag{3.23}$$

The molar flux for the solvent diffusion is given by,

$$\mathbf{J}^w = -\mathbf{M}^w \operatorname{Grad}\mu^w \quad\Rightarrow\quad J_I^w = -M_{IJ}(\operatorname{Grad}\mu^w)_J,$$

$$\text{where, } \mathbf{M}^w = \frac{D^w C^w}{R\theta}\mathbf{C}^{-1} \quad\Rightarrow\quad M_{IJ} = \frac{D^w C^w}{R\theta}C_{IJ}^{-1}. \tag{3.24}$$

Now,

$$\frac{\partial \mathbf{J}^w}{\partial \mathbf{F}} = -\frac{\partial \mathbf{C}^{-1}}{\partial \mathbf{F}}\left(\frac{D^w C^w}{R\theta}\operatorname{Grad}\mu^w\right) - \frac{D^w C^w}{R\theta}\mathbf{C}^{-1}\frac{\operatorname{Grad}\mu^w}{\partial \mathbf{F}}. \tag{3.25}$$

Let me recall the following kinematic definitions and identities to aid the calculation

$$\mathbf{C} = \mathbf{F}^\top\mathbf{F} \quad\Rightarrow\quad \mathbf{C}^{-1} = \mathbf{F}^{-1}\mathbf{F}^{-\top}, \quad\Rightarrow C_{IJ}^{-1} = F_{Im}^{-1}F_{mJ}^{-\top} = F_{Im}^{-1}F_{Jm}^{-1},$$

$$\left(\frac{\partial \mathbf{F}^{-1}}{\partial \mathbf{F}}\right)_{IjkL} = -F_{Ik}^{-1}F_{Lj}^{-1}, \quad \left(\frac{\partial \mathbf{F}^{-\top}}{\partial \mathbf{F}}\right)_{iJkL} = -F_{Li}^{-1}F_{Jk}^{-1}. \tag{3.26}$$

Hence, the derivative in the first term of (3.25) can be written as,

$$\begin{aligned}\frac{\partial \mathbf{C}^{-1}}{\partial \mathbf{F}} = \frac{\partial C_{IJ}^{-1}}{\partial F_{kL}} &= \frac{\partial}{\partial F_{kL}}\left(F_{Im}^{-1}F_{Jm}^{-1}\right) = \frac{\partial F_{Im}^{-1}}{\partial F_{kL}}F_{Jm}^{-1} + \frac{\partial F_{Jm}^{-1}}{\partial F_{kL}}F_{Im}^{-1}\\ &= -F_{Ik}^{-1}F_{Lm}^{-1}F_{Jm}^{-1} - F_{Jk}^{-1}F_{Lm}^{-1}F_{Im}^{-1},\\ &= -F_{Ik}^{-1}C_{LJ}^{-1} - F_{Jk}^{-1}C_{LI}^{-1}. \end{aligned} \tag{3.27}$$

which results in

$$\frac{\partial \mathbf{C}^{-1}}{\partial \mathbf{F}}\left(\frac{D^w C^w}{R\theta}\operatorname{Grad}\mu^w\right) = -\frac{D^w C^w}{R\theta}\left[F_{Ik}^{-1}C_{LJ}^{-1} + F_{Jk}^{-1}C_{LI}^{-1}\right](\operatorname{Grad}\mu^w)_J. \tag{3.28}$$

Now, the derivative in the second term of (3.25) can be written as,

$$\frac{\partial}{\partial \mathbf{F}}\left(\operatorname{Grad}\mu^w\right) = \frac{\partial}{\partial F_{kL}}\left(\frac{\partial \mu^w}{\partial X_J}\right) = \frac{\partial}{\partial X_J}\left(\frac{\partial \mu^w}{\partial F_{kL}}\right) = \frac{\partial}{\partial X_J}\left(\frac{\partial \mu^w}{\frac{\partial x_k}{\partial X_L}}\right) = \delta_{LJ}F_{Mk}^{-1}(\operatorname{Grad}\mu^w)_M, \tag{3.29}$$

which leads to,

$$\frac{D^w C^w}{R\theta} \mathbf{C}^{-1} \frac{\operatorname{Grad} \mu^w}{\partial \mathbf{F}} = \frac{D^w C^w}{R\theta} C_{JI}^{-1} \delta_{LJ} F_{Mk}^{-1} (\operatorname{Grad} \mu^w)_M,$$
$$= \frac{D^w C^w}{R\theta} C_{LI}^{-1} F_{Mk}^{-1} (\operatorname{Grad} \mu^w)_M, \tag{3.30}$$
$$= \frac{D^w C^w}{R\theta} C_{LI}^{-1} F_{Jk}^{-1} (\operatorname{Grad} \mu^w)_J.$$

$$\frac{\partial J_I^w}{\partial C^w} \frac{\partial C^w}{\partial F_{kL}} = \left[-\frac{D^w}{R\theta} C_{IJ}^{-1} (\operatorname{Grad} \mu^w)_J \right] \left[\left(\frac{\phi_0^p}{\mathcal{V}^w (\phi^p)^2} \right) \left(\frac{\partial \mathcal{G}}{\partial \phi^p} \right)^{-1} \left(\kappa \mathcal{V}^w \left[\ln(J^e) - 1 \right] F_{Lk}^{-1} \right) \right],$$
$$= -\left(\frac{\kappa \mathcal{V}^w \phi_0^p}{\mathcal{V}^w (\phi^p)^2} \right) \left(\frac{\partial \mathcal{G}}{\partial \phi^p} \right)^{-1} \frac{D^w}{R\theta} \left[\ln(J^e) - 1 \right] F_{Lk}^{-1} C_{IJ}^{-1} (\operatorname{Grad} \mu^w)_J \tag{3.31}$$

Thus, by adding all the terms together, I have,

$$\mathbb{J}_{IkL} = \frac{D^w C^w}{R\theta} F_{Ik}^{-1} C_{LJ}^{-1} (\operatorname{Grad} \mu^w)_J - \left(\frac{\kappa \mathcal{V}^w \phi_0^p}{\mathcal{V}^w (\phi^p)^2} \right) \left(\frac{\partial \mathcal{G}}{\partial \phi^p} \right)^{-1} \frac{D^w}{R\theta} \left[\ln(J^e) - 1 \right] F_{Lk}^{-1} C_{IJ}^{-1} (\operatorname{Grad} \mu^w)_J. \tag{3.32}$$

3.8 Calculation of $\dfrac{\partial \mathbf{J}^w}{\partial C^w} \dfrac{\partial C^w}{\partial \mu^w}$

Molar flux for the solvent, $\mathbf{J}^w$, is given by,

$$\mathbf{J}^w = -\mathbf{C}^{-1} \frac{D^w C^w}{R\theta} \operatorname{Grad} \mu^w \tag{3.33}$$

Thus, I have,

$$\frac{\partial \mathbf{J}^w}{\partial C^w} = -\frac{D^w}{R\theta} \mathbf{C}^{-1} \operatorname{Grad} \mu^w,$$
$$\Rightarrow \frac{\partial \mathbf{J}^w}{\partial \mu^w} = -\frac{D^w}{R\theta} \mathbf{C}^{-1} \operatorname{Grad} \mu^w \left(\frac{\partial C^w}{\partial \mu^w} \right), \tag{3.34}$$
$$\Rightarrow \left(\frac{\partial \mathbf{J}^w}{\partial \mu^w} \right)_I = \left[-\frac{D^w}{R\theta} C_{IJ}^{-1} (\operatorname{Grad} \mu^w)_J \right] \left(\frac{\partial C^w}{\partial \mu^w} \right)$$

By substituting the derivative, $\dfrac{\partial C^w}{\partial \mu^w}$, in the above expression, $\dfrac{\partial \mathbf{J}^w}{\partial \mu^w}$ can be calculated.

4 Implementation of u-μ element in Abaqus/Standard

4.1 Algorithmic procedure and a brief description of the source code

To keep the algorithm presentation concise, I eliminated some common procedural statements and matrix indices that were used in the previous chapters. Additionally, the algorithm presented in Procedure 1 does not include the additional calculation for F-bar implementation. The standard and F-bar finite element formulation with the appropriate constitutive model has been implemented in the Fortran source codes `uel_hydrogel.for` and `uel_hydrogel_Fbar.for`, respectively.

Procedure 1: Total Lagrangian finite element implementation of the hydrogel as user element subroutine (UEL) in Abaqus/ Standard

---

Input : PROPS, COORDS, JELEM, JTYPE, nnode, NDOFEL, TIME, DTIME, U, DU, V, A, PREDEF, JDLTYP, NDLOAD, MDLOAD, DDLMAG, ALMAG

Output : AMATRX, RHS, PNEWDT, ENERGY, SVARS

1   Get nInt $\leftarrow$ PROPS and ndim, nStress $\leftarrow$ JTYPE

2   Initialize: $\{\mathbf{N}_\mathbf{u}^a, \mathbf{B}_\mathbf{u}^a, \mathbf{G}_\mathbf{u}^a, \mathbf{N}, \mathbf{B}, \mathbf{G}_\mathbf{u}, \Sigma_\mathbf{F}, \Sigma_\mathbf{S}, \mathbf{k}_{\mathbf{uu}}^e, \mathbf{k}_{\mathbf{u}\mu}^e, \mathbf{k}_{\mu\mathbf{u}}^e, \mathbf{k}_{\mu\mu}^e, \mathbf{R}_\mathbf{u}^e, \mathbf{R}_\mu^e\} = 0$

3   Split nodal displacements and chemical potential from U: $\mathbf{u}^e, \mu^{we}$

4   Reshape nodal coordinates and displacement vector in the matrix form: $\overline{\mathbf{x}}_e, [\overline{\mathbf{u}}_e]$

5   Get $w_\text{int}, \boldsymbol{\xi}_\text{int} \leftarrow$ SUBROUTINE gaussQuadrtr(ndim, nnode)

6   **for** $k = 1$ **to** nInt **do**

7   Get $\phi^p(t=0) = \phi_0^p$ or $\phi(t_n) \leftarrow$ SVARS

8   Get $\mathbf{N}, \frac{\partial \mathbf{N}_\mathbf{u}}{\partial \boldsymbol{\xi}} \leftarrow$ SUBROUTINE interpFunc(ndim, nnode, $\boldsymbol{\xi}_\text{int}(k)$)

9   Calculate: $\frac{\partial \mathbf{X}}{\partial \boldsymbol{\xi}} = \overline{\mathbf{x}}_e \frac{\partial \mathbf{N}_\mathbf{u}}{\partial \boldsymbol{\xi}}$ // map to the reference configuration

10   Calculate: $\frac{\partial \mathbf{N}_\mathbf{u}}{\partial \mathbf{X}} = \frac{\partial \mathbf{N}_\mathbf{u}}{\partial \boldsymbol{\xi}} \left(\frac{\partial \mathbf{X}}{\partial \boldsymbol{\xi}}\right)^{-1}$ and J_ξ

11   Calculate deformation gradient: $\mathbf{F} = \mathbb{1} + [\overline{\mathbf{u}}_e] \frac{\partial \mathbf{N}_\mathbf{u}}{\partial \mathbf{X}}$

12   Calculate chemical potential and its gradient: $\mu^w = \mathbf{N}_\mu \mu^{we}, \text{Grad}(\mu^w) = \mathbf{B}_\mu \mu^{we}$

13   Get $\mathbf{S}, \dot{C}^w, \mathbf{J}^w, \mathbf{D}_\mathbb{C}, \mathbf{d}_{\mathbf{u}\mu}, \frac{\partial \dot{C}^w}{\partial \mathbf{F}}, \mathbf{D}_{\mu\mathbf{u}}, \frac{\partial \dot{C}^w}{\partial \mu^w}, \mathbf{m}_{\mu\mu}, \mathbf{M}^w, \phi^p(t_{n+1}) \leftarrow$ SUBROUTINE umatHydrogel(PROPS, $\mathbf{F}, \mu, \text{Grad}(\mu^w), \phi^p(t_n), \Delta t$)   // UMAT returns coupled material tangents and force vectors

14   Store $\phi^p(t_{n+1}) \rightarrow$ SVARS.

15   Form $\mathbf{N}_\mathbf{u}^a, \mathbf{B}_\mathbf{u}^a, \mathbf{G}_\mathbf{u}^a, \mathbf{N}_\mathbf{u}, \mathbf{B}_\mathbf{u}, \mathbf{G}_\mathbf{u}, \Sigma_\mathbf{F}, \Sigma_\mathbf{S}$ matrices   // this section is similar to total Lagrangian hyperelasticity formulation

16   Calculate the element tangent sub-matrices: $\mathbf{k}_{\mathbf{uu}}^e, \mathbf{k}_{\mathbf{u}\mu}^e, \mathbf{k}_{\mu\mathbf{u}}^e$, and $\mathbf{k}_{\mu\mu}^e$ // use the formulation developed in the previous section

17   Calculate the residual sub-vectors, $\mathbf{R}_\mathbf{u}^e$ and $\mathbf{R}_\mu^e$ // use the formulation developed in the previous section

18   **end**

                                  // end of integration point loop

19   Get AMATRX, RHS $\leftarrow$ SUBROUTINE assembleElement $\left(\mathbf{k}_{\mathbf{uu}}^e, \mathbf{k}_{\mathbf{u}\mu}^e, \mathbf{k}_{\mu\mathbf{u}}^e, \mathbf{k}_{\mu\mu}^e, \mathbf{R}_\mathbf{u}^e, \mathbf{R}_\mu^e\right)$

4.2 Element technology and properties

Current Abaqus UEL implementation includes a variety of three-dimensional and two-dimensional (plane strain) elements for hydrogel. It includes 4-node tetrahedral and 8-node hexahedral elements in three dimensions and 3-node triangular and 4-node quadrilateral elements in two dimensions (plane strain). Available element types and their integration schemes are listed in Table 1. I strongly recommend that users choose the 8-node trilinear hexahedral and 4-node bilinear quadrilateral elements with the full integration scheme whenever possible. To circumvent volumetric locking, the mechanical part of the element tangent matrices for these elements have been modified with F-bar element formulation. Triangular and tetrahedral elements should only be used with the full integration scheme. It is up to the users to ensure they correctly specify the integration points. This element formulation can be easily extended to include axisymmetric-type elements as well; however, extending it to plane stress cases is not straightforward. The latter requires a local Newton-Raphson procedure to determine the remaining component of the deformation gradient, F_{33}, using $\sigma_{33} = 0$.

Currently, it is programmed to store the Cauchy stress tensor, Euler-Almansi strain tensor, and polymer volume fraction at each integration point as element output (`UVARM`). If the user would like to have other quantities as output, they will have to program it within the `umat_hydrogel` subroutine.

Type of element	UEL element tag	Integration points (`nInt`)
4-node tetrahedral	U1	1 (full)
8-node hexahedral	U2	8 (full) and 1 (reduced)
3-node triangular (plane-strain)	U3	1 (full)
4-node quadrilateral (plane-strain)	U4	4 (full) and 1 (reduced)

Table 1: Types of small displacement mechanical elements in `uel_hydrogel.for`.

A list of physical constants, ambient conditions (temperature), and material properties to specify by the user are given in Table 2.

Remark 4. For mixed elements, using a similar order interpolation function (either linear or quadratic) for all the fields violates the Ladyzhenskaya–Babuška–Brezzi (LBB) stability criterion and may result in unwanted convergence issues because of spurious oscillation in the solution (Bouklas et al., 2015). Thus, it is preferred to have a higher-order interpolation (quadratic) for the displacement field and a lower-order (linear) interpolation for the chemical potential field, which satisfies the LBB criteria. Implementation of such an element will require the user to specify the degrees of freedom correctly in the Abaqus input file and then alter the current implementation to split the degrees of freedom accordingly. Finally, it will require the user to program a separate element tangent matrix and residual vector assembly subroutine as well. The element formulation and material procedure will remain the same.

Property Name	Symbol	Variable in UEL
Universal gas constant	R	Rgas
Absolute temperature	θ	theta
Initial polymer volume fraction	ϕ_0^p	phi0
Density of the gel	ρ	rho
Shear modulus at reference state	G	Gshear
Bulk modulus at reference state	κ	Kappa
Locking stretch	λ_L	lam_L
Molar volume of polymer	$\mathcal{V}^p$	Vp
Chemical potential of pure solvent	μ_w^0	mu0
Molar volume of the solvent	$\mathcal{V}^w$	Vw
Flory-Huggins interaction parameter	χ	chi
Diffusion coefficient of the solvent	D^w	Dw
No. of integration points		nInt
Elastomeric material ID		matID
No. of post-processed variables		nPostVars

Table 2: List of properties used in element and material definition.

4.3 Modification of Abaqus input file

Development of this UEL has been inspired by the UEL source code and example available from Chester et al., 2015. Interested users are requested to follow the detailed step-by-step tutorial on setting up a coupled finite element model for the hydrogel, which is available in the supplementary information of this article. This article also provides several examples for interested users. For even simpler mechanical models, users can also see Datta, 2024a, 2024b in which multiple examples are included in the repository.

Bibliographic references

Bouklas, N., Landis, C. M., & Huang, R. (2015). A nonlinear, transient finite element method for coupled solvent diffusion and large deformation of hydrogels. *Journal of the mechanics and physics of solids*, *79*, 21–43. https://doi.org/10.1016/j.jmps.2015.03.004

Chester, S. A., & Anand, L. (2010). A coupled theory of fluid permeation and large deformations for elastomeric materials. *Journal of the mechanics and physics of solids*, *58*(11), 1879–1906. https://doi.org/10.1016/j.jmps.2010.07.020

Chester, S. A., & Anand, L. (2011). A thermo-mechanically coupled theory for fluid permeation in elastomeric materials: Application to thermally responsive gels. *Journal of the mechanics and physics of solids*, *59*(10), 1978–2006. https://doi.org/10.1016/j.jmps.2011.07.005

Chester, S. A., Di Leo, C. V., & Anand, L. (2015). A finite element implementation of a coupled diffusion-deformation theory for elastomeric gels. *International journal of solids and structures*, *52*, 1–18. https://doi.org/10.1016/j.ijsolstr.2014.08.015

Datta, B. (2024a). *An Abaqus user element (UEL) implementation of linear elastostatics.* Zenodo. https://doi.org/10.5281/zenodo.11075088

Datta, B. (2024b). *A nonlinear user element (UEL) implementation for hyperelastic materials in Abaqus/Standard.* Zenodo. https://doi.org/10.5281/zenodo.11078801

de Borst, R., Crisfield, M. A., Remmers, J. J. C., & Verhoosel, C. V. (2012). *Non-linear Finite Element Analysis of Solids and Structures* (2nd ed.). John Wiley & Sons, Inc.

Neto, E. A. d. S., Perić, D., Dutko, M., & Owen, D. (1996). Design of simple low order finite elements for large strain analysis of nearly incompressible solids. *International journal of solids and structures*, *33*(20-22), 3277–3296. https://doi.org/10.1016/0020-7683(95)00259-6

Reddy, J. N. (2015). *An Introduction to Nonlinear Finite Element Analysis* (1st ed.). Oxford University Press. Retrieved December 7, 2023, from https://academic.oup.com/book/32507

Zienkiewicz, O. C., Taylor, R. L., & Fox, D. (2014). *The Finite Element Method for Solid and Structural Mechanics* (7th ed.). Butterworth-Heinemann.