A finite element model and Abaqus user element (UEL) implementation of hydrogel chemo-mechanics

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This report contains the details of the 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, we 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 first-order elements for three-dimensional cases, two-dimensional axisymmetric, and plane strain cases. For the constitutive model, we chose Neo-Hookean potential with Flory-Huggins mixing potential to describe the behavior of the hydrogel. 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.

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1 Summary of the coupled theory and constitutive model

Considering a pre-swollen (as-prepared) hydrogel with polymer volume fraction, ϕ_0^p, to be the reference state, the material coordinate, $\mathbf{X} \in \Omega_0$, can be mapped to the spatial coordinate, $\mathbf{x} \in \Omega$, via one-to-one mapping. Based on this, the deformation gradient, $\mathbf{F}$, is given by,

$$\mathbf{F} = (\phi_0^p)^{1/3}\mathbf{F}^e\mathbf{F}^s. \tag{1}$$

Accordingly, the volume change associated with the total deformation is given by,

$$J = \det(\mathbf{F}) = \phi_0^p J^e J^s, \tag{2}$$

where $J^e = \det(\mathbf{F}^e)$ and $J^s = \det(\mathbf{F}^s)$ are the elastic and swelling volume changes from the dry state of the hydrogel.

Volume change of non-ionic hydrogel is completely attributed to the imbibition of solvent; thus, we can prescribe the following constitutive relation,

$$J^s = \frac{1}{\phi_0^p}\left(C^w\mathcal{V}^w + C^p\mathcal{V}^p\right), \tag{3}$$

where C^w and C^p are referential molar concentration, measured as the moles per the reference volume, of the solvent and polymer, respectively. $\mathcal{V}^w$ and $\mathcal{V}^p$ are the molar volumes of the solvent and polymer, respectively.

In the reference configuration, Ω_0, $\Gamma_{\mathbf{g}}$ and $\Gamma_{\mathbf{T}}$, and Γ_h and Γ_{I^w} be two distinct pairs of non-overlapping subsurfaces of the boundary $\partial\Omega_0$. The subsurfaces can be defined as follows,

$$\begin{aligned} \partial\Omega_0 &= \Gamma_{\mathbf{g}} \cup \Gamma_{\mathbf{T}} \quad \text{and} \quad \Gamma_{\mathbf{g}} \cap \Gamma_{\mathbf{T}} = \varnothing, \\ &= \Gamma_h \cup \Gamma_{I^w} \quad \text{and} \quad \Gamma_h \cap \Gamma_{I^w} = \varnothing. \end{aligned} \tag{4}$$

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 $\Gamma_{\mathbf{g}}$ and the Piola-Kirchhoff traction, $\mathbf{T}$, on $\Gamma_{\mathbf{T}}$ are given by,

$$\begin{aligned} \mathrm{Div}(\mathbf{FS}) + \rho_R\mathbf{B} &= 0 && \text{in } \Omega_0, \\ \mathbf{u} &= \mathbf{g} && \text{on } \Gamma_{\mathbf{g}}, \\ \mathbf{P}\cdot\mathbf{N} = (\mathbf{FS})\cdot\mathbf{N} &= \mathbf{T} && \text{on } \Gamma_{\mathbf{T}}. \end{aligned} \tag{5}$$

Here, $\mathbf{P}$ and $\mathbf{S}$ are the first and second Piola-Kirchhoff stress, respectively, ρ_{R} is the referential mass density, and $\mathbf{B}$ is the body force per unit of mass in the hydrated reference configuration. We 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 the same time interval, 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,

$$\begin{aligned} \dot{C}^w &= -\operatorname{Div}\mathbf{J}^w && \text{in } \Omega_0, \\ \mu^w(\mathbf{X}, t=0) &= \mu_0^w && \text{in } \Omega_0, \\ \mu^w &= h && \text{on } \Gamma_h, \\ -\mathbf{J}^w \cdot \mathbf{N} &= I^w && \text{on } \Gamma_{I^w}, \end{aligned} \tag{6}$$

where $\mathbf{J}^w$ is the molar flux of the solvent, measured as moles per unit area in the reference configuration, per unit time.

1.1 Constitutive model for non-ionic hydrogel

Helmholtz free energy density, Ψ, of a polymeric hydrogel encompasses an additive split of free energy density of pure solvent, Ψ^w, free energy density from mechanical deformation, Ψ^{mech}, and the mixture of polymer and solvent, $\Psi^{\text{mix,pol}}$,

$$\Psi = \Psi^w + \Psi^{\text{mech}} + \Psi^{\text{mix,pol}}. \tag{7}$$

Assuming the polymer network in the hydrogel behaves as N-chain Gaussian network and their mixing is described by binary Flory-Huggins mixture theory, we can then write,

$$\begin{aligned} \Psi = \mu^{0,w} C^w + \frac{G}{2}\left[I_1 - 3 - 2(\phi_0^p)^{2/3}\ln(J)\right] + \frac{\phi_0^p \kappa}{2} J^s (\ln J^e)^2 \\ + \frac{\phi_0^p R\theta}{\mathcal{V}^w}\left[\left(\frac{1}{\phi^p} - 1\right)\ln(1-\phi^p) + \chi(1-\phi^p)\right]. \end{aligned} \tag{8}$$

where G is the shear modulus, κ is the bulk modulus, and χ is the Flory-Huggins interaction parameter characterizing the enthalpic contribution in a binary polymer-solvent mixture.

Based on the theory proposed by Chester and Anand, 2010, the thermodynamic restrictions on the state variables are given by,

$$\begin{aligned} \boldsymbol{\sigma} &= J^{-1}\left[\mathbf{F}^e\left(2\frac{\partial \Psi}{\partial \mathbf{C}^e}\right)\mathbf{F}^{e\top}\right], \\ \mu^w &= \frac{\partial \Psi}{\partial C^w} + p\mathcal{V}^w, \end{aligned} \tag{9}$$

where

$$p = \frac{-1}{3} J^e \operatorname{tr}(\boldsymbol{\sigma}) \tag{10}$$

is known as the mean pressure. We can use these restrictions to calculate the constitutive response of the hydrogel for a specific material model.

Using the constitutive definition, the Cauchy stress, $\boldsymbol{\sigma}$, can be calculated as,

$$\boldsymbol{\sigma} = J^{-1}\left[G\mathbf{b} - \left(G(\phi_0^p)^{2/3} - \kappa\phi_0^p J^s(\ln J^e)\right)\mathbb{1}\right], \tag{11}$$

and the mean pressure, p, is given by,

$$p = \frac{-1}{3\phi_0^p J^s}\left(GI_1 - 3G(\phi_0^p)^{2/3}\right) - \kappa(\ln J^e). \tag{12}$$

By using pull-back operation on the Cauchy stress, $\boldsymbol{\sigma}$, the first Piola-Kirchhoff stress, $\mathbf{P}$, and the second Piola-Kirchhoff stress, $\mathbf{S}$, can be obtained as

$$\begin{aligned} \mathbf{P} &= G\mathbf{F} - \left[G(\phi_0^p)^{2/3} - \kappa\phi_0^p J^s(\ln J^e)\right]\mathbf{F}^{-\top}, \\ \mathbf{S} &= G\mathbb{1} - \left[G(\phi_0^p)^{2/3} - \kappa\phi_0^p J^s(\ln J^e)\right]\mathbf{C}^{-1}. \end{aligned} \tag{13}$$

The chemical potential of the solvent, μ^w, is given by

$$\mu^w = \mu^{0,w} + R\theta\left[\phi^p + \ln(1-\phi^p) + \chi(\phi^p)^2\right] + \mathcal{P}\mathcal{V}^w, \tag{14}$$

where

$$\mathcal{P} = \frac{\kappa}{2}(\ln J^e)^2 - \kappa(\ln J^e) \tag{15}$$

acts as a pressure-like Lagrange multiplier for quasi-incompressibility.

To describe diffusion within the gel, we now prescribe the following kinetic law for the diffusion of the solvent,

$$\mathbf{J}^w = -\mathbf{M}^w\,\mathrm{Grad}\,\mu^w, \qquad \text{where} \quad \mathbf{M}^w = \frac{D^w C^w}{R\theta}\mathbf{C}^{-1}. \tag{16}$$

Here, D^w is known as the diffusion coefficient of the solvent in the hydrogel, and $\mathbf{M}^w$ is known as the mobility tensor in the reference configuration and adopted from Chester and Anand, 2010.

2 Mixed finite element (u-μ) formulation

The first step towards finite element formulation is to develop the corresponding weak forms of the governing equations. Typically, the variational method or weighted residual method is used to derive the weak form. In this work, we 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. We can write the weak form of the momentum balance equation as

$$\begin{aligned} &\int_{\Omega_0} \text{Div}(\mathbf{FS}) \cdot \mathbf{W} \; dV + \int_{\Omega_0} \rho_{\text{R}} \mathbf{B} \cdot \mathbf{W} \; dV = 0, \\ \Rightarrow &\int_{\Omega_0} \mathbf{FS} : \text{Grad}(\mathbf{W}) \; dV - \int_{\Omega_0} \rho_{\text{R}} \mathbf{B} \cdot \mathbf{W} \; dV - \int_{\Gamma_T} \mathbf{T} \cdot \mathbf{W} \; dS = 0. \end{aligned} \tag{17}$$

Applying the divergence theorem on $(17)_1$ results in the final weak form of stress equilibrium $(17)_2$. In the context of the principle of virtual work, the first term in $(17)_2$ 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 we can write the weak form of the mass balance equation for the solvent as,

$$\begin{aligned} &\int_{\Omega_0} \dot{C}^w \; \Theta \; dV + \int_{\Omega_0} \text{Div}\, \mathbf{J}^w \; \Theta \; dV = 0, \\ \Rightarrow &\int_{\Omega_0} \dot{C}^w \; \Theta \; dV - \int_{\Omega_0} \mathbf{J}^w \cdot \text{Grad}(\Theta) \; dV - \int_{\Gamma_{Iw}} I^w \; \Theta \; dS = 0. \end{aligned} \tag{18}$$

Similar to the weak form of stress equilibrium, application of the divergence theorem on $(18)_1$ results in the final weak form of mass balance $(18)_2$.

2.2 Discretization and Bubnov-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 $(17)_2$ and mass balance $(18)_2$, we can write the weak forms for the system of governing equations for each element as,

$$\begin{aligned} \mathcal{W}_e^{\mathbf{u}}(\mathbf{u}, \mu^w) &= -\int_{\Omega_0^e} \mathbf{FS} : \text{Grad}(\mathbf{W}) \; dV + \int_{\Omega_0^e} \rho_{\text{R}} \mathbf{B}_e \cdot \mathbf{W} \; dV + \int_{\Gamma_T^e} \mathbf{T}_e \cdot \mathbf{W} \; dS = 0, \\ \mathcal{W}_e^{\mu}(\mathbf{u}, \mu^w) &= \int_{\Omega_0^e} \dot{C}^w \; \Theta \; dV - \int_{\Omega_0^e} \mathbf{J}^w \cdot \text{Grad}(\Theta) \; dV - \int_{\Gamma_{Iw}^e} I_e^w \; \Theta \; dS = 0. \end{aligned} \tag{19}$$

Remark 1. 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_a^i be 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,

$$X_i = \sum_{a=1}^{\mathrm{n_{en}}} N_a^{\mathbf{u}} X_a^i \quad \Rightarrow \mathbf{X} = \mathbf{N_u} \mathbf{X}_e, \tag{20}$$

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, we can write the matrix form of $\mathbf{N_u}$ and the vector form of $\mathbf{x}_e$, as,

$$\begin{aligned} \mathbf{N_u} &= \begin{bmatrix} N_1^{\mathbf{u}} & 0 & 0 & N_2^{\mathbf{u}} & 0 & 0 & \cdots & \cdots & N_{\mathrm{n_{en}}}^{\mathbf{u}} & 0 & 0 \\ 0 & N_1^{\mathbf{u}} & 0 & 0 & N_2^{\mathbf{u}} & 0 & \cdots & \cdots & 0 & N_{\mathrm{n_{en}}}^{\mathbf{u}} & 0 \\ 0 & 0 & N_1^{\mathbf{u}} & 0 & 0 & N_2^{\mathbf{u}} & \cdots & \cdots & 0 & 0 & N_{\mathrm{n_{en}}}^{\mathbf{u}} \end{bmatrix}_{\mathrm{n_{dim}} \times \mathrm{n_{en}} * \mathrm{n_{dim}}}, \\ \mathbf{X}_e &= \begin{bmatrix} X_1 & Y_1 & Z_1 & X_2 & Y_2 & Z_2 & \cdots & \cdots & X_{\mathrm{n_{en}}} & Y_{\mathrm{n_{en}}} & Z_{\mathrm{n_{en}}} \end{bmatrix}^{\top}_{\mathrm{n_{en}} * \mathrm{n_{dim}} \times 1}. \end{aligned} \tag{21}$$

By inspecting the structure of $[\mathbf{N_u}]$, we can see that there exist repeating blocks of sub-matrices corresponding to each node within the element. Let define those sub-matrices as $\mathbf{N}_a^{\mathbf{u}}$, then for a three dimensional case, we can write $\mathbf{N}_a^{\mathbf{u}}$ and X_a^e as,

$$\mathbf{N}_a^{\mathbf{u}} = \begin{bmatrix} N_a^{\mathbf{u}} & 0 & 0 \\ 0 & N_a^{\mathbf{u}} & 0 \\ 0 & 0 & N_a^{\mathbf{u}} \end{bmatrix}_{\mathrm{n_{dim}} \times \mathrm{n_{dim}}}, \quad \mathbf{X}^a = \begin{bmatrix} X_i \\ Y_i \\ Z_i \end{bmatrix}_{\mathrm{n_{dim}} \times 1}, \tag{22}$$

where, $N_a^{\mathbf{u}}$ is the interpolation function for displacement corresponding to the node a 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,

$$\begin{aligned} \mathbf{u}(\mathbf{X}) = \mathbf{N_u} \mathbf{u}_e = \sum_{a=1}^{\mathrm{n_{en}}} \mathbf{N}_a^{\mathbf{u}}(\mathbf{X}) \mathbf{u}_a \quad &\Rightarrow \quad u^i(\mathbf{X}) = \sum_{a=1}^{\mathrm{n_{en}}} N_a^{\mathbf{u}}(\mathbf{X}) u_a^i, \\ \text{and} \quad \mu^w(\mathbf{X}) = \mathbf{N}_\mu \mu_e^w = \sum_{a=1}^{\mathrm{n_{en}}} N_a^\mu \mu_a^w. & \end{aligned} \tag{23}$$

where $\mathbf{u}_e$ and μ_e^w 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,

$$\mathbf{N}_\mu = \begin{bmatrix} N_1^\mu & N_2^\mu & N_3^\mu & \cdots & \cdots & N_{\mathrm{n_{en}}}^\mu \end{bmatrix}. \tag{24}$$

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

$$\begin{aligned} \mathbf{W}(\mathbf{X}) = \mathbf{N}_{\mathbf{u}}\mathbf{W}^e \quad \Rightarrow \quad W_i(\mathbf{X}) = \sum_{a=1}^{\mathrm{n_{en}}} N_a^{\mathbf{u}}(\mathbf{X}) W_a^i, \\ \Theta(\mathbf{X}) = \mathbf{N}_{\mu}\boldsymbol{\Theta}^e = \sum_{a=1}^{\mathrm{n_{en}}} N_a^{\mu}(\mathbf{X})\Theta_a. \end{aligned} \tag{25}$$

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

$$\mathbf{F} = \frac{\partial \mathbf{x}}{\partial \mathbf{X}} = \mathbb{1} + \frac{\partial \mathbf{u}}{\partial \mathbf{X}} = \mathbb{1} + [\overline{\mathbf{u}}_e]\left[\frac{\partial \mathbf{N}_{\mathbf{u}}}{\partial \mathbf{X}}\right] \quad \Rightarrow F_{iJ} = \delta_{ij} + \sum_{a=1}^{\mathrm{n_{en}}} \frac{\partial N_a^{\mathbf{u}}}{\partial X_J} u_a^i. \tag{26}$$

where u_a^i denotes i-th nodal displacement component of node a, and $N_a^{\mathbf{u}}$ is the shape function corresponding to the same node. $\left[\frac{\partial \mathbf{N}_{\mathbf{u}}}{\partial \mathbf{X}}\right]$ is a matrix which has a dimension of $\left[\frac{\partial \mathbf{N}_{\mathbf{u}}}{\partial \mathbf{X}}\right]_{\mathrm{n_{en}} \times \mathrm{n_{dim}}}$, and $[\overline{\mathbf{u}}_e]$ has a dimension of $[\overline{\mathbf{u}}_e]_{\mathrm{n_{dim}} \times \mathrm{n_{en}}}$. 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,

$$\begin{aligned} \mathbf{r}_e^{\mathbf{u}} &= -\int_{\Omega_0^e} \mathbf{FS} : \mathrm{Grad}(\mathbf{N}_{\mathbf{u}})\, dV + \int_{\Omega_0^e} \rho_{\mathrm{R}} \mathbf{N}_{\mathbf{u}}^{\top} \mathbf{B}_e\, dV + \int_{\Gamma_T^e} \mathbf{N}_{\mathbf{u}}^{\top} \mathbf{T}_e\, dS = 0, \\ \mathbf{r}_e^{\mu} &= -\int_{\Omega_0^e} \mathbf{N}_{\mu}^{\top} \left(\frac{C^w - C_t^w}{\Delta t}\right) dV + \int_{\Omega_0^e} \mathbf{J}^w \cdot \mathrm{Grad}(\mathbf{N}_{\mu})\, dV + \int_{\Gamma_{Iw}^e} \mathbf{N}_{\mu}^{\top} I_e^w\, dS = 0. \end{aligned} \tag{27}$$

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, $\mathbf{r}_e^{\mathbf{u}}$ and $\mathbf{r}_e^{\mu}$, 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,

$$\begin{aligned} \mathbf{r}_a^{\mathbf{u}} &= -\int_{\Omega_0^e} \mathbf{FS} : \mathrm{Grad}(\mathbf{N}_a^{\mathbf{u}})\, dV + \int_{\Omega_0^e} \rho_{\mathrm{R}} \mathbf{N}_a^{\mathbf{u}\top} \mathbf{B}_e\, dV + \int_{\Gamma_T^e} \mathbf{N}_a^{\mathbf{u}\top} \mathbf{T}_e\, dS = 0, \\ r_a^{\mu} &= -\int_{\Omega_0^e} N_a^{\mu\top} \left(\frac{C^w - C_t^w}{\Delta t}\right) dV + \int_{\Omega_0^e} \mathbf{J}^w \cdot \mathrm{Grad}(N_a^{\mu})\, dV + \int_{\Gamma_{Iw}^e} N_a^{\mu\top} I_e^w\, dS = 0. \end{aligned} \tag{28}$$

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, we 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_b^w. Then the system of nodal residuals can be perturbed by $\Delta\mathbf{u}_b$ and $\Delta\mu_b^w$, and using first-order Taylor series expansion, we have,

$$\begin{aligned}
\mathbf{r}_a^{\mathbf{u}}\left(\mathbf{u}_b + \Delta\mathbf{u}_b, \mu_b^w + \Delta\mu_b^w\right) &= \mathbf{r}_a^{\mathbf{u}}(\mathbf{u}, \mu^w) + \frac{\partial \mathbf{r}_a^{\mathbf{u}}(\mathbf{u}, \mu^w)}{\partial \mathbf{u}_b}\Delta\mathbf{u}_b + \frac{\partial \mathbf{r}_a^{\mathbf{u}}(\mathbf{u}, \mu^w)}{\partial \mu_b^w}\Delta\mu_b^w, \\
r_a^{\mu}\left(\mathbf{u}_b + \Delta\mathbf{u}_b, \mu_b^w + \Delta\mu_b^w\right) &= r_a^{\mu}(\mathbf{u}, \mu^w) + \frac{\partial r_a^{\mu}(\mathbf{u}, \mu^w)}{\partial \mathbf{u}_b}\Delta\mathbf{u}_b + \frac{\partial r_a^{\mu}(\mathbf{u}, \mu^w)}{\partial \mu_b^w}\Delta\mu_b^w.
\end{aligned} \tag{29}$$

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

$$\begin{aligned}
-\frac{\partial \mathbf{r}_a^{\mathbf{u}}}{\partial \mathbf{u}_b}\Delta\mathbf{u}_b - \frac{\partial \mathbf{r}_a^{\mathbf{u}}}{\partial \mu_b^w}\Delta\mu_b^w &= \mathbf{r}_a^{\mathbf{u}}, \\
-\frac{\partial r_a^{\mu}}{\partial \mathbf{u}_b}\Delta\mathbf{u}_b - \frac{\partial r_a^{\mu}}{\partial \mu_b^w}\Delta\mu_b^w &= r_a^{\mu}.
\end{aligned} \tag{30}$$

In matrix form, it can be written as,

$$\begin{bmatrix} -\frac{\partial \mathbf{r}_a^{\mathbf{u}}}{\partial \mathbf{u}_b} & -\frac{\partial \mathbf{r}_a^{\mathbf{u}}}{\partial \mu_b^w} \\ -\frac{\partial r_a^{\mu}}{\partial \mathbf{u}_b} & -\frac{\partial r_a^{\mu}}{\partial \mu_b^w} \end{bmatrix} \begin{Bmatrix} \Delta\mathbf{u}_b \\ \Delta\mu_b^w \end{Bmatrix} = \begin{Bmatrix} \mathbf{r}_a^{\mathbf{u}} \\ r_a^{\mu} \end{Bmatrix}. \tag{31}$$

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.

$$\mathbf{k}_{ab} = \begin{bmatrix} \mathbf{k}_{ab}^{\mathbf{uu}} & \mathbf{k}_{ab}^{\mathbf{u}\mu} \\ \mathbf{k}_{ab}^{\mu\mathbf{u}} & k_{ab}^{\mu\mu} \end{bmatrix} = \begin{bmatrix} -\frac{\partial \mathbf{r}_a^{\mathbf{u}}}{\partial \mathbf{u}_b} & -\frac{\partial \mathbf{r}_a^{\mathbf{u}}}{\partial \mu_b^w} \\ -\frac{\partial r_a^{\mu}}{\partial \mathbf{u}_b} & -\frac{\partial r_a^{\mu}}{\partial \mu_b^w} \end{bmatrix}, \quad \text{and} \quad \mathbf{r}_a = \begin{Bmatrix} \mathbf{r}_a^{\mathbf{u}} \\ r_a^{\mu} \end{Bmatrix}. \tag{32}$$

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

$$\mathbf{K}_e = \bigA_{a,b=1}^{\mathrm{n_{en}}} \mathbf{k}_{ab}, \qquad \text{and} \qquad \mathbf{r}_e = \bigA_{a=1}^{\mathrm{n_{en}}} \mathbf{r}_a. \tag{33}$$

The element tangent matrix, $\mathbf{K}_e$, and element residual vector, $\mathbf{r}_e$, for a three-dimensional

chemo-mechanical element with n nodes are given by,

$$\mathbf{K}_e = \begin{bmatrix} k_{11}^{u_1u_1} & k_{11}^{u_1u_2} & k_{11}^{u_1u_3} & k_{11}^{u_1\mu} & \cdots & k_{1\mathrm{n_{en}}}^{u_1u_1} & k_{1\mathrm{n_{en}}}^{u_1u_2} & k_{1\mathrm{n_{en}}}^{u_1u_3} & k_{1\mathrm{n_{en}}}^{u_1\mu} \\ k_{11}^{u_2u_1} & k_{11}^{u_2u_2} & k_{11}^{u_2u_3} & k_{11}^{u_2\mu} & \cdots & k_{1\mathrm{n_{en}}}^{u_2u_1} & k_{1\mathrm{n_{en}}}^{u_2u_2} & k_{1\mathrm{n_{en}}}^{u_2u_3} & k_{1\mathrm{n_{en}}}^{u_2\mu} \\ k_{11}^{u_3u_1} & k_{11}^{u_3u_2} & k_{11}^{u_3u_3} & k_{11}^{u_3\mu} & \cdots & k_{1\mathrm{n_{en}}}^{u_3u_1} & k_{1\mathrm{n_{en}}}^{u_3u_2} & k_{1\mathrm{n_{en}}}^{u_3u_3} & k_{1\mathrm{n_{en}}}^{u_3\mu} \\ k_{11}^{\mu u_1} & k_{11}^{\mu u_2} & k_{11}^{\mu u_3} & k_{11}^{\mu\mu} & \cdots & k_{1\mathrm{n_{en}}}^{\mu u_1} & k_{1\mathrm{n_{en}}}^{\mu u_2} & k_{1\mathrm{n_{en}}}^{\mu u_3} & k_{1\mathrm{n_{en}}}^{\mu\mu} \\ \vdots & & \vdots & & \ddots & & \vdots & & \vdots \\ \vdots & & \vdots & & \ddots & & \vdots & & \vdots \\ k_{\mathrm{n_{en}}1}^{u_1u_1} & k_{\mathrm{n_{en}}1}^{u_1u_2} & k_{\mathrm{n_{en}}1}^{u_1u_3} & k_{\mathrm{n_{en}}1}^{u_1\mu} & \cdots & k_{\mathrm{n_{en}n_{en}}}^{u_1u_1} & k_{\mathrm{n_{en}n_{en}}}^{u_1u_2} & k_{\mathrm{n_{en}n_{en}}}^{u_1u_3} & k_{\mathrm{n_{en}n_{en}}}^{u_1\mu} \\ k_{\mathrm{n_{en}}1}^{u_2u_1} & k_{\mathrm{n_{en}}1}^{u_2u_2} & k_{\mathrm{n_{en}}1}^{u_2u_3} & k_{\mathrm{n_{en}}1}^{u_2\mu} & \cdots & k_{\mathrm{n_{en}n_{en}}}^{u_2u_1} & k_{\mathrm{n_{en}n_{en}}}^{u_2u_2} & k_{\mathrm{n_{en}n_{en}}}^{u_2u_3} & k_{\mathrm{n_{en}n_{en}}}^{u_2\mu} \\ k_{\mathrm{n_{en}}1}^{u_3u_1} & k_{\mathrm{n_{en}}1}^{u_3u_2} & k_{\mathrm{n_{en}}1}^{u_3u_3} & k_{\mathrm{n_{en}}1}^{u_3\mu} & \cdots & k_{\mathrm{n_{en}n_{en}}}^{u_3u_1} & k_{\mathrm{n_{en}n_{en}}}^{u_3u_2} & k_{\mathrm{n_{en}n_{en}}}^{u_3u_3} & k_{\mathrm{n_{en}n_{en}}}^{u_3\mu} \\ k_{\mathrm{n_{en}}1}^{\mu u_1} & k_{\mathrm{n_{en}}1}^{\mu u_2} & k_{\mathrm{n_{en}}1}^{\mu u_3} & k_{\mathrm{n_{en}}1}^{\mu\mu} & \cdots & k_{\mathrm{n_{en}n_{en}}}^{\mu u_1} & k_{\mathrm{n_{en}n_{en}}}^{\mu u_2} & k_{\mathrm{n_{en}n_{en}}}^{\mu u_3} & k_{\mathrm{n_{en}n_{en}}}^{\mu\mu} \end{bmatrix}_{\mathrm{n_{dofel}}\times\mathrm{n_{dofel}}}, \tag{34}$$

$$\mathbf{r}_e = \left[r_1^{u_1}, r_1^{u_2}, r_1^{u_3}, r_1^{\mu},\ \cdots\ \cdots,\ r_{\mathrm{n_{en}}}^{u_1}, r_{\mathrm{n_{en}}}^{u_2}, r_{\mathrm{n_{en}}}^{u_3}, r_{\mathrm{n_{en}}}^{\mu}\right]^{\top}_{\mathrm{n_{dofel}}\times 1},$$

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).

$$\mathbf{K} = \bigwedge_{e=1}^{\mathrm{n_{el}}} \mathbf{K}_e, \qquad \text{and} \qquad \mathbf{r} = \bigwedge_{e=1}^{\mathrm{n_{el}}} \mathbf{r}_e. \tag{35}$$

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

$$[\mathbf{K}]\{\Delta\mathbf{d}\} = \{\mathbf{r}\}, \tag{36}$$

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.

$$\mathbf{d}_{k+1} = \mathbf{d}_k + \Delta\mathbf{d} \tag{37}$$

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

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

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

$$\mathbf{S} \equiv \mathbf{S}\left(\mathbf{C}, C^w\right), \quad \mathbf{J}^w \equiv \mathbf{J}^w\left(\mathbf{C}, C^w, \mathrm{Grad}(\mu^w)\right), \quad \mathcal{G}\left(\mathbf{C}, \mu^w, C^w\right) = 0. \tag{38}$$

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,

$$r_a^{\mathbf{u}}(u_i,\mu^w) = -\int\limits_{\Omega_0^e} \frac{\partial N_a^{\mathbf{u}}}{\partial X_I} F_{iJ} S_{JI}\; dV + \int\limits_{\Omega_0^e} \rho_{\mathrm{R}} N_a^{\mathbf{u}} B_i^e\; dV + \int\limits_{\Gamma_T^e} N_a^{\mathbf{u}} T_e^i\; dS = 0. \tag{39}$$

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_k^b} &= \int\limits_{\Omega_0^e} \frac{\partial N_a^{\mathbf{u}}}{\partial X_I} \left(\frac{\partial F_{iJ}}{\partial u_k^b} S_{JI} + F_{iJ} \frac{\partial S_{JI}}{\partial u_k^b} \right)\; dV, \\
&= \int\limits_{\Omega_0^e} \frac{\partial N_a^{\mathbf{u}}}{\partial X_I} \left[\frac{\partial F_{iJ}}{\partial u_k^b} S_{JI} + F_{iJ} \left(\frac{\partial S_{JI}}{\partial C_{KL}} + \frac{\partial S_{JI}}{\partial C^w} \frac{dC^w}{dC_{kL}} \right) \frac{\partial C_{KL}}{\partial F_{mN}} \frac{\partial F_{mN}}{\partial u_k^b} \right]\; dV, \\
&= \int\limits_{\Omega_0^e} \frac{\partial N_a^{\mathbf{u}}}{\partial X_I} \left[\frac{\partial F_{iJ}}{\partial u_k^b} S_{JI} + 2F_{iJ} \left(\frac{\partial S_{JI}}{\partial C_{KL}} + \frac{\partial S_{JI}}{\partial C^w} \frac{dC^w}{dC_{kL}} \right) F_{mL} \delta_{KN} \frac{\partial F_{mN}}{\partial u_k^b} \right]\; dV, \\
&= \int\limits_{\Omega_0^e} \frac{\partial N_a^{\mathbf{u}}}{\partial X_I} \left(\frac{\partial F_{iJ}}{\partial u_k^b} S_{JI} + F_{iJ} \mathbb{C}_{IJKL} F_{mL} \frac{\partial F_{mK}}{\partial u_k^b} \right)\; dV, \\
&= \int\limits_{\Omega_0^e} \frac{\partial N_a^{\mathbf{u}}}{\partial X_I} \left(\frac{\partial N_b^{\mathbf{u}}}{\partial X_J} \delta_{ik} S_{JI} + F_{iJ} \mathbb{C}_{IJKL} F_{mL} \frac{\partial N_b^{\mathbf{u}}}{\partial X_K} \delta_{mk} \right)\; dV, \\
&= \int\limits_{\Omega_0^e} \frac{\partial N_a^{\mathbf{u}}}{\partial X_I} \left(\frac{\partial N_b^{\mathbf{u}}}{\partial X_J} \delta_{ik} S_{JI} + F_{iJ} \mathbb{C}_{IJKL} F_{kL} \frac{\partial N_b^{\mathbf{u}}}{\partial X_K} \right)\; dV, \\
&= \int\limits_{\Omega_0^e} \frac{\partial N_a^{\mathbf{u}}}{\partial X_I} S_{JI} \delta_{ik} \frac{\partial N_b^{\mathbf{u}}}{\partial X_J}\; dV + \int\limits_{\Omega_0^e} \frac{\partial N_a^{\mathbf{u}}}{\partial X_I} \left(F_{iJ} \mathbb{C}_{IJKL} F_{kL} \right) \frac{\partial N_b^{\mathbf{u}}}{\partial X_K}\; dV, \\
&= \int\limits_{\Omega_0^e} \frac{\partial N_a^{\mathbf{u}}}{\partial X_J} S_{JL} \delta_{ik} \frac{\partial N_b^{\mathbf{u}}}{\partial X_L}\; dV + \int\limits_{\Omega_0^e} \frac{\partial N_a^{\mathbf{u}}}{\partial X_J} \left(F_{iI} \mathbb{C}_{IJKL} F_{kK} \right) \frac{\partial N_b^{\mathbf{u}}}{\partial X_L}\; dV.
\end{aligned} \tag{40}$$

Here,

$$\mathbb{C}_{IJKL} = 2\frac{dS_{IJ}}{dC_{KL}} = 2\left(\frac{\partial S_{IJ}}{\partial C_{KL}} + \frac{\partial S_{IJ}}{\partial C^w} \frac{dC^w}{dC_{KL}} \right) \tag{41}$$

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, we used the properties of summation indices (or dummy indices) and the

following intermediate results to obtain the final expression for $k_{ab}^{u_i u_k}$.

$$\begin{aligned}\frac{\partial C_{KL}}{\partial F_{mN}} &= F_{mL}\delta_{KN} + F_{mK}\delta_{LN} = 2F_{mL}\delta_{KN} \quad (\text{since } C_{KL} = C_{LK}),\\ \text{and,} \quad F_{iJ} &= \delta_{ij} + \sum_{b=1}^{\mathrm{n_{en}}} \frac{\partial N_b^{\mathbf{u}}}{\partial X_J} u_i^b \qquad \Rightarrow \quad \frac{\partial F_{iJ}}{\partial u_k^b} = \frac{\partial N_b^{\mathbf{u}}}{\partial X_J}\delta_{ik}.\end{aligned} \tag{42}$$

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

$$[\mathbf{k}_e^{\mathbf{uu}}]_{\mathrm{n_{en}}*\mathrm{n_{dim}}\times\mathrm{n_{en}}*\mathrm{n_{dim}}} = \int_{\Omega_0^e} \left[\mathbf{G}_{\mathbf{u}}^\top \Sigma_{\mathbf{S}} \mathbf{G}_{\mathbf{u}} + (\mathbf{B}_{\mathbf{u}}\boldsymbol{\Sigma}_{\mathbf{F}}{}^\top)^\top \mathbf{D}_{\mathbb{C}} (\mathbf{B}_{\mathbf{u}}\Sigma_{\mathbf{F}}{}^\top)\right]\, dV \tag{43}$$

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, $\Sigma_{\mathbf{S}}$, the sparse deformation gradient matrix, $\Sigma_{\mathbf{F}}$, are given in the next section.

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

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

$$\begin{aligned}k_{ab}^{u_i\mu} &= -\frac{\partial r_{u_i}^a}{\partial \mu_b^w} = \int_{\Omega_0^e} \frac{\partial N_a^{\mathbf{u}}}{\partial X_I} \left(F_{iJ}\frac{\partial S_{JI}}{dC^w}\frac{dC^w}{d\mu^w}\right)\frac{\partial \mu^w}{\partial \mu_b^w}\, dV,\\ &= \int_{\Omega_0^e} \frac{\partial N_a^{\mathbf{u}}}{\partial X_I} \left(F_{iJ}\frac{\partial S_{JI}}{dC^w}\frac{dC^w}{d\mu^w}\right) N_b^\mu\, dV,\end{aligned} \tag{44}$$

Let define,

$$\mathbb{S}_{iI} = F_{iJ}\frac{\partial S_{JI}}{dC^w}\frac{dC^w}{d\mu^w}. \tag{45}$$

Thus, we can write,

$$k_{ab}^{u_i\mu} = \int_{\Omega_0^e} \frac{\partial N_a^{\mathbf{u}}}{\partial X_I}\mathbb{S}_{iI} N_b^\mu\, dV. \tag{46}$$

In matrix form, it can be expressed as,

$$[\mathbf{k}_e^{\mathbf{u}\mu}]_{\mathrm{n_{en}}*\mathrm{n_{dim}}\times\mathrm{n_{en}}} = \int_{\Omega_0^e} \mathbf{G}_{\mathbf{u}}^\top \mathbf{d}_{\mathbf{u}\mu}\mathbf{N}_\mu\, dV \tag{47}$$

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

$$\mathbb{S}_{iI} = F_{iJ}\frac{\partial S_{JI}}{\partial C^w}\frac{dC^w}{d\mu^w} \quad \xrightarrow{\text{reshape to vector}} \quad \mathbf{d}_{\mathbf{u}\mu}, \tag{48}$$

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

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

$$r_a^\mu = -\int_{\Omega_0^e} N_a^\mu \left(\frac{C^w - C_t^w}{\Delta t}\right)\, dV + \int_{\Omega_0^e} \frac{\partial N_a^\mu}{\partial X_I} J_I^w \, dV + \int_{\Gamma_{Iw}^e} N_a^\mu I_e^w \, dS. \tag{49}$$

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

$$\begin{aligned}
\mathbf{k}_{ab}^{\mu u_k} = -\frac{\partial r_a^\mu}{\partial u_k^b} &= \int_{\Omega_0^e} \left[N_a^\mu \left(\frac{1}{\Delta t}\frac{dC^w}{dF_{mL}}\right) - \frac{\partial N_a^\mu}{\partial X_I}\left(\frac{\partial J_I^w}{\partial F_{mL}} + \frac{\partial J_I^w}{\partial C^w}\frac{dC^w}{dF_{mL}}\right)\right]\frac{\partial F_{mL}}{\partial u_k^b}\, dV, \\
&= \int_{\Omega_0^e} \left[N_a^\mu \left(\frac{1}{\Delta t}\frac{dC^w}{dF_{mL}}\right) - \frac{\partial N_a^\mu}{\partial X_I}\left(\frac{\partial J_I^w}{\partial F_{mL}} + \frac{\partial J_I^w}{\partial C^w}\frac{dC^w}{dF_{mL}}\right)\right]\frac{\partial}{\partial u_k^b}\left(\delta_{mL} + \sum_{b=1}^{\mathrm{n_{en}}}\frac{\partial N_b^{\mathbf{u}}}{\partial X_J}u_m^b\right)\, dV, \\
&= \int_{\Omega_0^e} \left[N_a^\mu \left(\frac{1}{\Delta t}\frac{dC^w}{dF_{mL}}\right) - \frac{\partial N_a^\mu}{\partial X_I}\left(\frac{\partial J_I^w}{\partial F_{mL}} + \frac{\partial J_I^w}{\partial C^w}\frac{dC^w}{dF_{mL}}\right)\right]\frac{\partial N_b^{\mathbf{u}}}{\partial X_L}\delta_{mk}\, dV, \\
&= \int_{\Omega_0^e} N_a^\mu \left(\frac{1}{\Delta t}\frac{dC^w}{dF_{mL}}\right)\frac{\partial N_b^{\mathbf{u}}}{\partial X_L}\delta_{mk}\, dV - \int_{\Omega_0^e}\frac{\partial N_a^\mu}{\partial X_I}\left(\frac{\partial J_I^w}{\partial F_{mL}} + \frac{\partial J_I^w}{\partial C^w}\frac{dC^w}{dF_{mL}}\right)\frac{\partial N_b^{\mathbf{u}}}{\partial X_L}\delta_{mk}\, dV, \\
&= \int_{\Omega_0^e} N_a^\mu \left(\frac{1}{\Delta t}\frac{dC^w}{dF_{kL}}\right)\frac{\partial N_b^{\mathbf{u}}}{\partial X_L}\, dV - \int_{\Omega_0^e}\frac{\partial N_a^\mu}{\partial X_I}\left(\frac{\partial J_I^w}{\partial F_{kL}} + \frac{\partial J_I^w}{\partial C^w}\frac{dC^w}{dF_{kL}}\right)\frac{\partial N_b^{\mathbf{u}}}{\partial X_L}\, dV, \\
&= \int_{\Omega_0^e} N_a^\mu \left(\frac{1}{\Delta t}\frac{dC^w}{dF_{kL}}\right)\frac{\partial N_b^{\mathbf{u}}}{\partial X_L}\, dV - \int_{\Omega_0^e}\frac{\partial N_a^\mu}{\partial X_I}\mathbb{J}_{IkL}\frac{\partial N_b^{\mathbf{u}}}{\partial X_L}\, dV.
\end{aligned} \tag{50}$$

In matrix form, we can write,

$$[\mathbf{k}_e^{\mu\mathbf{u}}]_{\mathrm{n_{en}}\times\mathrm{n_{en}}*\mathrm{n_{dim}}} = \int_{\Omega_0^e}\left(\mathbf{N}_\mu^\top \frac{\partial C^w}{\partial \mathbf{F}}\mathbf{G}_{\mathbf{u}} - \mathbf{B}_\mu^\top \mathbf{D}_{\mu\mathbf{u}}\mathbf{G}_{\mathbf{u}}\right)\, dV \tag{51}$$

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

$$(\mathbb{J})_{IkL} = \frac{dJ_I^w}{dF_{kL}} = \frac{\partial J_I^w}{\partial F_{kL}} + \frac{\partial J_I^w}{\partial C^w}\frac{dC^w}{dF_{kL}} \quad \xrightarrow{\text{reshape to matrix}} \quad \mathbf{D}_{\mu\mathbf{u}}. \tag{52}$$

2.4.4 Chemical tangent matrix, $k_e^{\mu\mu}$

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

$$\begin{aligned}
k_{ab}^{\mu\mu} &= -\frac{\partial r_a^\mu}{\partial \mu_b^w}, \\
&= \int_{\Omega_0^e} N_a^\mu \left(\frac{1}{\Delta t}\frac{dC^w}{d\mu^w}\right)\frac{\partial \mu^w}{\partial \mu_b^w}\, dV - \int_{\Omega_0^e} \frac{\partial N_a^\mu}{\partial X_I}\frac{\partial J_I^w}{\partial C^w}\frac{dC^w}{d\mu^w}\frac{\partial \mu^w}{\partial \mu_b^w}\, dV - \int_{\Omega_0^e} \frac{\partial N_a^\mu}{\partial X_I}\frac{dJ_I^w}{d(\mathrm{Grad}\,\mu^w)} N_b^\mu\, dV, \\
&= \int_{\Omega_0^e} N_a^\mu \left(\frac{1}{\Delta t}\frac{dC^w}{d\mu^w}\right) N_b^\mu\, dV - \int_{\Omega_0^e} \frac{\partial N_a^\mu}{\partial X_I}\left(\frac{\partial J_I^w}{\partial C^w}\frac{dC^w}{d\mu^w}\right) N_b^\mu\, dV - \int_{\Omega_0^e} \frac{\partial N_a^\mu}{\partial X_I}\left(\frac{dJ_I^w}{d(\mathrm{Grad}\,\mu^w)_J}\right)\frac{\partial N_b^\mu}{\partial X_J}\, dV,
\end{aligned} \tag{53}$$

Let define,

$$(\mathbf{m}_{\mu\mu})_I = \frac{dJ_I^w}{dC^w} = \frac{\partial J_I^w}{\partial C^w}\frac{dC^w}{d\mu^w}. \tag{54}$$

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,

$$[\mathbf{k}_e^{\mu\mu}]_{\mathrm{n_{en}}\times\mathrm{n_{en}}} = \int_{\Omega_0^e} \left(\mathbf{N}_\mu^\top \frac{dC^w}{d\mu^w}\mathbf{N}_\mu - \mathbf{B}_\mu^\top \mathbf{m}_{\mu\mu}\mathbf{N}_\mu + \mathbf{B}_\mu^\top \mathbf{M}^w \mathbf{B}_\mu\right)\, dV, \tag{55}$$

2.4.5 Matrix form of element residual vectors

The members of the element residual vector, $\mathbf{r}_e$, are given by,

$$\begin{aligned}
[\mathbf{r}_e^{\mathbf{u}}]_{\mathrm{n_{en}}*\mathrm{n_{dim}}\times 1} &= -\int_{\Omega_0^e} (\mathbf{B}_{\mathbf{u}}\Sigma_{\mathbf{F}}{}^\top)^\top \mathbf{S}\, dV + \int_{\Omega_0^e} \rho_{\mathrm{R}} \mathbf{N}_{\mathbf{u}}^\top \mathbf{B}_e\, dV + \int_{\Gamma_T^e} \mathbf{N}_{\mathbf{u}}^\top \mathbf{T}_e\, dS, \\
\text{and} \quad [\mathbf{r}_e^\mu]_{\mathrm{n_{en}}\times 1} &= -\int_{\Omega_0^e} \mathbf{N}_\mu^\top \left(\frac{C^w - C_t^w}{\Delta t}\right)\, dV + \int_{\Omega_0^e} \mathbf{B}_\mu^\top \mathbf{J}^w\, dV + \int_{\Gamma_{I^w}^e} \mathbf{N}_\mu^\top I_e^w\, dS.
\end{aligned} \tag{56}$$

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,

$$\begin{aligned}
r_a^{\mathbf{u}} &= -\int_{\Omega_0^e} \frac{\partial N_a^{\mathbf{u}}}{\partial X_I} F_{iJ} S_{JI}\, dV + \int_{\Omega_0^e} \rho_{\mathrm{R}} N_a^{\mathbf{u}} B_e^i\, dV + \int_{\Gamma_T^e} N_a^{\mathbf{u}} T_e^i\, dS = 0, \\
r_a^\mu &= -\int_{\Omega_0^e} N_a^\mu \left(\frac{C^w - C_t^w}{\Delta t}\right)\, dV + \int_{\Omega_0^e} \frac{\partial N_a^\mu}{\partial X_I} J_I^w\, dV + \int_{\Gamma_{I^w}^e} N_a^\mu I_e^w\, dS.
\end{aligned} \tag{57}$$

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

$$
\begin{aligned}
k_{ab}^{u_i u_k} &= -\frac{\partial r_a^{u_i}}{\partial u_b^k} &&= \int_{\Omega_0^e} \frac{\partial N_a^{\mathbf{u}}}{\partial X_J} S_{JL} \delta_{ik} \frac{\partial N_b^{\mathbf{u}}}{\partial X_L}\, dV + \int_{\Omega_0^e} \frac{\partial N_a^{\mathbf{u}}}{\partial X_J} F_{iI} \left(2 \frac{dS_{IJ}}{dC_{KL}} \right) F_{kK} \frac{\partial N_b^{\mathbf{u}}}{\partial X_L}\, dV, \\
k_{ab}^{u_i \mu} &= -\frac{\partial r_a^{u_i}}{\partial \mu_b^w} &&= \int_{\Omega_0^e} \frac{\partial N_a^{\mathbf{u}}}{\partial X_I} \left(F_{iJ} \frac{dS_{IJ}}{d\mu^w} \right) N_b^{\mu}\, dV, \\
k_{ab}^{\mu u_k} &= -\frac{\partial r_a^{\mu}}{\partial u_b^k} &&= \int_{\Omega_0^e} N_a^{\mu} \left(\frac{1}{\Delta t} \frac{dC^w}{dF_{kL}} \right) \frac{\partial N_b^{\mathbf{u}}}{\partial X_L}\, dV - \int_{\Omega_0^e} \frac{\partial N_a^{\mu}}{\partial X_I} \left(\frac{dJ_I^w}{dF_{kL}} \right) \frac{\partial N_b^{\mathbf{u}}}{\partial X_L}\, dV, \\
k_{ab}^{\mu \mu} &= -\frac{\partial r_a^{\mu}}{\partial \mu_b^w} &&= \int_{\Omega_0^e} N_a^{\mu} \left(\frac{1}{\Delta t} \frac{dC^w}{d\mu^w} \right) N_b^{\mu}\, dV - \int_{\Omega_0^e} \frac{\partial N_a^{\mu}}{\partial X_I} \left(\frac{dJ_I^w}{d\mu^w} \right) N_b^{\mu}\, dV \\
& & &\quad - \int_{\Omega_0^e} \frac{\partial N_a^{\mu}}{\partial X_I} \left(\frac{dJ_I^w}{d(\mathrm{Grad}\, \mu^w)_J} \right) \frac{\partial N_b^{\mu}}{\partial X_J}\, dV.
\end{aligned}
\tag{58}
$$

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

$$
\begin{aligned}
[\mathbf{r}_e^{\mathbf{u}}]_{\mathrm{n_{en}} * \mathrm{n_{dim}} \times 1} &= -\int_{\Omega_0^e} (\mathbf{B_u} \Sigma_{\mathbf{F}}{}^{\top})^{\top} \mathbf{S}\, dV + \int_{\Omega_0^e} \rho_{\mathrm{R}} \mathbf{N}_{\mathbf{u}}^{\top} \mathbf{B}_e\, dV + \int_{\Gamma_T^e} \mathbf{N}_{\mathbf{u}}^{\top} \mathbf{T}_e\, dS, \\
\text{and} \quad [\mathbf{r}_e^{\mu}]_{\mathrm{n_{en}} \times 1} &= -\int_{\Omega_0^e} \mathbf{N}_{\mu}^{\top} \left(\frac{C^w - C_t^w}{\Delta t} \right)\, dV + \int_{\Omega_0^e} \mathbf{B}_{\mu}^{\top} \mathbf{J}^w\, dV + \int_{\Gamma_{Iw}^e} \mathbf{N}_{\mu}^{\top} I_e^w\, dS.
\end{aligned}
\tag{59}
$$

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

$$
\begin{aligned}
[\mathbf{k}_e^{\mathbf{uu}}]_{\mathrm{n_{en}} * \mathrm{n_{dim}} \times \mathrm{n_{en}} * \mathrm{n_{dim}}} &= -\frac{\partial \mathbf{r}_e^{\mathbf{u}}}{\partial \mathbf{u}_e} &&= \int_{\Omega_0^e} \left[\mathbf{G}_{\mathbf{u}}^{\top} \Sigma_{\mathbf{S}} \mathbf{G}_{\mathbf{u}} + (\mathbf{B_u} \mathbf{\Sigma_F}{}^{\top})^{\top} \mathbf{D}_{\mathbb{C}} (\mathbf{B_u} \Sigma_{\mathbf{F}}{}^{\top}) \right]\, dV, \\
[\mathbf{k}_e^{\mathbf{u}\mu}]_{\mathrm{n_{en}} * \mathrm{n_{dim}} \times \mathrm{n_{en}}} &= -\frac{\partial \mathbf{r}_e^{\mathbf{u}}}{\partial \mu_e^w} &&= \int_{\Omega_0^e} \mathbf{G}_{\mathbf{u}}^{\top} \mathbf{d}_{\mathbf{u}\mu} \mathbf{N}_{\mu}\, dV, \\
[\mathbf{k}_e^{\mu\mathbf{u}}]_{\mathrm{n_{en}} \times \mathrm{n_{en}} * \mathrm{n_{dim}}} &= -\frac{\partial \mathbf{r}_e^{\mu}}{\partial \mathbf{u}_e} &&= \int_{\Omega_0^e} \left(\mathbf{N}_{\mu}^{\top} \left(\frac{1}{\Delta t} \frac{\partial C^w}{\partial \mathbf{F}} \right) \mathbf{G}_{\mathbf{u}} - \mathbf{B}_{\mu}^{\top} \mathbf{D}_{\mu\mathbf{u}} \mathbf{G}_{\mathbf{u}} \right)\, dV, \\
[\mathbf{k}_e^{\mu\mu}]_{\mathrm{n_{en}} \times \mathrm{n_{en}}} &= -\frac{\partial \mathbf{r}_e^{\mu}}{\partial \mu_e^w} &&= \int_{\Omega_0^e} \left(\mathbf{N}_{\mu}^{\top} \left(\frac{1}{\Delta t} \frac{dC^w}{d\mu^w} \right) \mathbf{N}_{\mu} - \mathbf{B}_{\mu}^{\top} \mathbf{m}_{\mu\mu} \mathbf{N}_{\mu} + \mathbf{B}_{\mu}^{\top} \mathbf{M}^w \mathbf{B}_{\mu} \right)\, dV.
\end{aligned}
\tag{60}
$$

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

$$
\mathbf{G_u} = \left[\mathbf{G}_{\mathbf{u}}^1 \quad \mathbf{G}_{\mathbf{u}}^2 \quad \mathbf{G}_{\mathbf{u}}^3 \quad \cdots \quad \cdots \quad \mathbf{G}_{\mathbf{u}}^{\mathrm{n_{en}}} \right]_{\mathrm{n_{dim}}{}^2 \times \mathrm{n_{en}} * \mathrm{n_{dim}}},
\tag{61}
$$

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

$$\mathbf{G}_a^{\mathbf{u}} = \begin{bmatrix} N_{a,1} & 0 \\ 0 & N_{a,1} \\ N_{a,2} & 0 \\ 0 & N_{a,2} \end{bmatrix}_{\mathrm{n_{dim}}^2 \times \mathrm{n_{dim}}}, \qquad \mathbf{G}_a^{\mathbf{u}} = \begin{bmatrix} N_{a,1} & 0 & 0 \\ 0 & N_{a,1} & 0 \\ 0 & 0 & N_{a,1} \\ N_{a,2} & 0 & 0 \\ 0 & N_{a,2} & 0 \\ 0 & 0 & N_{a,2} \\ N_{a,3} & 0 & 0 \\ 0 & N_{a,3} & 0 \\ 0 & 0 & N_{a,3} \end{bmatrix}_{\mathrm{n_{dim}}^2 \times \mathrm{n_{dim}}}. \tag{62}$$

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

$$\Sigma_{\mathbf{S}} = \begin{bmatrix} \mathbf{S}_{11} & 0 & \mathbf{S}_{12} & 0 \\ 0 & \mathbf{S}_{11} & 0 & \mathbf{S}_{12} \\ \mathbf{S}_{12} & 0 & \mathbf{S}_{22} & 0 \\ 0 & \mathbf{S}_{12} & 0 & \mathbf{S}_{22} \end{bmatrix}, \quad \Sigma_{\mathbf{S}} = \begin{bmatrix} \mathbf{S}_{11} & 0 & 0 & \mathbf{S}_{12} & 0 & 0 & \mathbf{S}_{13} & 0 & 0 \\ 0 & \mathbf{S}_{11} & 0 & 0 & \mathbf{S}_{12} & 0 & 0 & \mathbf{S}_{13} & 0 \\ 0 & 0 & \mathbf{S}_{11} & 0 & 0 & \mathbf{S}_{12} & 0 & 0 & \mathbf{S}_{13} \\ \mathbf{S}_{12} & 0 & 0 & \mathbf{S}_{22} & 0 & 0 & \mathbf{S}_{23} & 0 & 0 \\ 0 & \mathbf{S}_{12} & 0 & 0 & \mathbf{S}_{22} & 0 & 0 & \mathbf{S}_{23} & 0 \\ 0 & 0 & \mathbf{S}_{12} & 0 & 0 & \mathbf{S}_{22} & 0 & 0 & \mathbf{S}_{23} \\ \mathbf{S}_{13} & 0 & 0 & \mathbf{S}_{23} & 0 & 0 & \mathbf{S}_{33} & 0 & 0 \\ 0 & \mathbf{S}_{13} & 0 & 0 & \mathbf{S}_{23} & 0 & 0 & \mathbf{S}_{33} & 0 \\ 0 & 0 & \mathbf{S}_{13} & 0 & 0 & \mathbf{S}_{23} & 0 & 0 & \mathbf{S}_{33} \end{bmatrix}. \tag{63}$$

$[\Sigma_{\mathbf{S}}]$ has a dimension of $[\Sigma_{\mathbf{S}}]_{\mathrm{n_{dim}}^2 \times \mathrm{n_{dim}}^2}$. It is also possible to represent $\mathbf{G}_a^{\mathbf{u}}$ 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).

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

$$\Sigma_{\mathbf{F}} = \begin{bmatrix} F_{11} & F_{12} & F_{13} & 0 & 0 & 0 & \cdots & 0 & 0 & 0 \\ F_{21} & F_{22} & F_{23} & 0 & 0 & 0 & \cdots & 0 & 0 & 0 \\ F_{31} & F_{32} & F_{33} & 0 & 0 & 0 & \cdots & 0 & 0 & 0 \\ & & & & \cdots & & & & & \\ & & & & \cdots & & & & & \\ & & & & \cdots & & & & & \\ & & & & \cdots & & & & & \\ & & & & \cdots & & & & & \\ & & & & \cdots & & & & & \\ 0 & 0 & 0 & 0 & 0 & 0 & \cdots & F_{11} & F_{12} & F_{13} \\ 0 & 0 & 0 & 0 & 0 & 0 & \cdots & F_{21} & F_{22} & F_{23} \\ 0 & 0 & 0 & 0 & 0 & 0 & \cdots & F_{31} & F_{32} & F_{33} \end{bmatrix}_{\mathrm{n_{en}}*\mathrm{n_{dim}} \times \mathrm{n_{en}}*\mathrm{n_{dim}}} \tag{64}$$

For a two-dimensional plane strain case, we 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 we considered the hydrogel quasi-incompressible, volumetric locking could happen under certain loading conditions. Similar to the hyperelastic materials, we employed the F-bar approach (Chester et al., 2015; Neto et al., 1996) to mitigate the volumetric locking in 4-noded bilinear quadrilateral elements and 8-noded trilinear hexahedral elements. We 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

$$\overline{\mathbf{F}} = \mathbf{F}^{\text{dev}}\mathbf{F}_0^{\text{vol}} = \left(\frac{\det \mathbf{F}_0}{\det \mathbf{F}}\right)^{1/3} \mathbf{F}. \tag{65}$$

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

$$\mathbf{r}_e^{\mathbf{u}}(\mathbf{u}_e) = -\int_{\Omega_0^e} \left(\frac{\det \mathbf{F}_0}{\det \mathbf{F}}\right)^{-2/3} (\mathbf{B}_{\mathbf{u}}\Sigma_{\mathbf{F}}{}^{\top})^{\top}\mathbf{S}\; dV + \int_{\Omega_0^e} \rho_{\text{R}}\mathbf{N}_{\mathbf{u}}^{\top}\mathbf{B}^e\; dV + \int_{\Gamma_T^e} \mathbf{N}_{\mathbf{u}}^{\top}\mathbf{T}^e\; dS \tag{66}$$

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

$$\begin{aligned}[\mathbf{k}_{\mathbf{uu}}^e] = &\int_{\Omega_0^e} \left(\frac{\det \mathbf{F}_0}{\det \mathbf{F}}\right)^{-1/3} \left[\mathbf{G}_{\mathbf{u}}^{\top}\Sigma_{\mathbf{S}}\mathbf{G}_{\mathbf{u}} + (\mathbf{B}_{\mathbf{u}}\boldsymbol{\Sigma}_{\mathbf{F}}{}^{\top})^{\top}\mathbf{D}_{\mathbb{C}}(\mathbf{B}_{\mathbf{u}}\Sigma_{\mathbf{F}}{}^{\top})\right]\; dV \\ &+ \int_{\Omega_0^e} \left[\left(\frac{\det \mathbf{F}_0}{\det \mathbf{F}}\right)^{-2/3} \left(\mathbf{G}_{\mathbf{u}}^{\top}\mathbf{Q}_{\text{R}}^0\mathbf{G}_{\mathbf{u}}^0 - \mathbf{G}_{\mathbf{u}}^{\top}\mathbf{Q}_{\text{R}}\mathbf{G}_{\mathbf{u}}\right)\right]\; dV\end{aligned} \tag{67}$$

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

$$\begin{aligned}(\mathbb{Q}_{\text{R}}^0)_{iJkL} &= \frac{1}{3}\overline{F}_{iI}\left[2\left(\frac{\partial \overline{S}_{IJ}}{\partial \overline{C}_{MN}} + \frac{\partial \overline{S}_{IJ}}{\partial C^w}\frac{\partial C^w}{\partial \overline{C}_{kL}}\right)\right]\overline{C}_{MN}F_{0,kL}^{-\top} - \frac{1}{3}\overline{P}_{iJ}F_{0,kL}^{-\top}, \\ (\mathbb{Q}_{\text{R}})_{iJkL} &= \frac{1}{3}\overline{F}_{iI}\left[2\left(\frac{\partial \overline{S}_{IJ}}{\partial \overline{C}_{MN}} + \frac{\partial \overline{S}_{IJ}}{\partial C^w}\frac{\partial C^w}{\partial \overline{C}_{kL}}\right)\right]\overline{C}_{MN}F_{kL}^{-\top} - \frac{1}{3}\overline{P}_{iJ}F_{kL}^{-\top}.\end{aligned} \tag{68}$$

2.6.2 Plane strain case

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

$$\overline{\mathbf{F}} = \left[\begin{array}{cc|c} & & 0 \\ & [\overline{\mathbf{F}}_{\text{pe}}] & 0 \\ \hline 0 & 0 & 1 \end{array}\right], \tag{69}$$

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

$$\overline{\mathbf{F}}_{\text{pe}} = \mathbf{F}^{\text{dev}}\mathbf{F}_0^{\text{vol}} = \left(\frac{\det \mathbf{F}_0}{\det \mathbf{F}}\right)^{1/2} \mathbf{F}_{\text{pe}}, \quad \text{where, } \mathbf{F}_{\text{pe}} = \begin{bmatrix} F_{11} & F_{12} \\ F_{21} & F_{22} \end{bmatrix}. \tag{70}$$

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

$$\mathbf{r}_e^{\mathbf{u}}(\mathbf{u}_e) = -\int_{\Omega_0^e} \left(\frac{\det \mathbf{F}_0}{\det \mathbf{F}}\right)^{-1/2} (\mathbf{B}_{\mathbf{u}}\Sigma_{\mathbf{F}}{}^{\top})^{\top}\mathbf{S}\ dV + \int_{\Omega_0^e} \rho_{\text{R}}\mathbf{N}_{\mathbf{u}}^{\top}\mathbf{B}^e\ dV + \int_{\Gamma_T^e} \mathbf{N}_{\mathbf{u}}^{\top}\mathbf{T}^e\ dS \tag{71}$$

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

$$\begin{aligned}[\mathbf{k}_{\mathbf{uu}}^e] = &\int_{\Omega_0^e} \left[\mathbf{G}_{\mathbf{u}}^{\top}\Sigma_{\mathbf{S}}\mathbf{G}_{\mathbf{u}} + (\mathbf{B}_{\mathbf{u}}\boldsymbol{\Sigma}_{\mathbf{F}}{}^{\top})^{\top}\mathbf{D}_{\mathbb{C}}(\mathbf{B}_{\mathbf{u}}\Sigma_{\mathbf{F}}{}^{\top})\right]\ dV \\ &+ \int_{\Omega_0^e} \left[\left(\frac{\det \mathbf{F}_0}{\det \mathbf{F}}\right)^{-1/2} \mathbf{G}_{\mathbf{u}}^{\top}\left(\mathbf{Q}_{\text{R}}^0\mathbf{G}_{\mathbf{u}}^0 - \mathbf{Q}_{\text{R}}\mathbf{G}_{\mathbf{u}}\right)\right]\ dV\end{aligned} \tag{72}$$

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

$$\begin{aligned}(\mathbb{Q}_{\text{R}}^0)_{iJkL} &= \frac{1}{2}\overline{F}_{iI}\left[2\left(\frac{\partial \overline{S}_{IJ}}{\partial \overline{C}_{MN}} + \frac{\partial \overline{S}_{IJ}}{\partial C^w}\frac{\partial C^w}{\partial \overline{C}_{kL}}\right)\right]\overline{C}_{MN}F_{0,kL}^{-\top}, \\ (\mathbb{Q}_{\text{R}})_{iJkL} &= \frac{1}{2}\overline{F}_{iI}\left[2\left(\frac{\partial \overline{S}_{IJ}}{\partial \overline{C}_{MN}} + \frac{\partial \overline{S}_{IJ}}{\partial C^w}\frac{\partial C^w}{\partial \overline{C}_{kL}}\right)\right]\overline{C}_{MN}F_{kL}^{-\top}.\end{aligned} \tag{73}$$

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 $[\mathbf{N}_{\mathbf{u}}]_{\text{n}_{\text{dim}}\times \text{n}_{\text{en}}*\text{n}_{\text{dim}}}$.
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 $[\mathbf{B}_{\mathbf{u}}]_{\text{nstress}\times \text{n}_{\text{en}}*\text{n}_{\text{dim}}}$.
4. $\mathbf{G}_{\mathbf{u}} = \text{Grad}(\mathbf{N}_{\mathbf{u}})$ is referred to as the non-symmetric gradient matrix which has a dimension of $[\mathbf{G}_{\mathbf{u}}]_{\text{n}_{\text{dim}}^2\times \text{n}_{\text{en}}*\text{n}_{\text{dim}}}$.
5. $\Sigma_{\mathbf{S}}$ and $\Sigma_{\mathbf{F}}$ have dimensions of $[\Sigma_{\mathbf{S}}]_{\text{n}_{\text{dim}}^2\times \text{n}_{\text{dim}}^2}$ and $[\Sigma_{\mathbf{F}}]_{\text{n}_{\text{en}}*\text{n}_{\text{dim}}\times \text{n}_{\text{en}}*\text{n}_{\text{dim}}}$, respectively.
6. $\mathbf{N}_{\mu}$ is a vector of dimension of $[\mathbf{N}_{\mu}]_{1\times \text{n}_{\text{en}}}$.
7. $\mathbf{B}_{\mu} = \text{Grad}(\mathbf{N}_{\mu})$ is a matrix of dimension $[\mathbf{B}_{\mu}]_{\text{n}_{\text{dim}}\times \text{n}_{\text{en}}}$.

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,

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

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

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

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

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

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

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

$$(\mathbf{m}_{\mu\mu})_I = \frac{\partial J_I^w}{\partial C^w}\frac{dC^w}{d\mu^w}. \tag{74}$$

3 Constitutive update procedure

Constitutive relations are evaluated at each integration point of the element to compute the element residual vector and element tangent stiffness matrix. Element residuals require evaluation of the constitutive relations, and element tangent matrices require material tangent moduli.

3.1 Local Newton solver for internal variable

The first step in constitutive evaluation is to solve for the internal variable, C^w. We employ a local Newton-Raphson solver to calculate ϕ^p from the constitutive equation of solvent chemical potential, μ^w, and then calculate C^w.

$$(\phi^p)^{(j+1)} = (\phi^p)^{(j)} + \Delta(\phi^p)^{(j+1)}, \quad \text{where, } \Delta(\phi^p)^{(j+1)} = -\left(\frac{d\mathcal{G}}{d\phi^p}\right)^{-1}\mathcal{G}^{(j)}, \tag{75}$$

where (j) and $(j+1)$ are the consecutive iteration counters for the Newton-Raphson procedure, and the local residual, $\mathcal{G}$, is given by,

$$\mathcal{G}(\phi^p) = \mu^{0,w} + R\theta\left[\phi^p + \ln(1-\phi^p) + \chi(\phi^p)^2\right] + \mathcal{P}\mathcal{V}^w - \mu^w = 0. \tag{76}$$

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

$$\frac{d\mathcal{G}}{d\phi^p} = R\theta\left[1 - \frac{1}{1-\phi^p} + 2\chi\phi^p\right] + \left(\frac{\partial\mathcal{P}}{\partial\phi^p}\right)\mathcal{V}^w, \tag{77}$$

where,

$$\frac{\partial\mathcal{P}}{\partial\phi^p} = \frac{\kappa}{\phi^p}\left(\ln J^e - 1\right). \tag{78}$$

The solvent concentration, C^w, is then calculated as,

$$C^w = \frac{1}{\mathcal{V}^w}\left(\frac{\phi_0^p}{\phi^p} - \phi_0^p\right). \tag{79}$$

3.2 Calculation of $\frac{dC^w}{d\mu^w}$

By applying the implicit function theorem, the partial derivative of the implicit constitutive function, $\mathcal{G}(\mathbf{C}(\mathbf{F}), \mu^w, \phi^p(C^w)) = 0$, with respect to μ^w can be written as,

$$\frac{dC^w}{d\mu^w} = \frac{\partial C^w}{\partial\phi^p}\left(\frac{\partial\mathcal{G}}{\partial\phi^p}\right)^{-1}. \qquad \left(\text{since, } \frac{\partial\mathcal{G}}{\partial\mu^w} = -1\right) \tag{80}$$

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

$$\phi^p = \frac{C^p\mathcal{V}^p}{C^p\mathcal{V}^p + C^w\mathcal{V}^w} \qquad \Rightarrow \quad \frac{\partial C^w}{\partial\phi^p} = -\frac{C^p\mathcal{V}^p}{\mathcal{V}^w(\phi^p)^2} = -\frac{\phi_0^p}{\mathcal{V}^w(\phi^p)^2}. \tag{81}$$

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

3.3 Calculation of $\frac{dC^w}{d\mathbf{F}}$

By using the implicit function theorem on $\mathcal{G}(\mathbf{C}(\mathbf{F}), \mu^w, \phi^p(C^w)) = 0$, we have,

$$\frac{dC^w}{d\mathbf{F}} = -\left(\frac{\partial C^w}{\partial\phi^p}\right)\left(\frac{\partial\mathcal{G}}{\partial\phi^p}\right)^{-1}\left(\frac{\partial\mathcal{G}}{\partial\mathbf{F}}\right). \tag{82}$$

Now,

$$\begin{aligned}
\frac{\partial\mathcal{G}}{\partial\mathbf{F}} &= -\kappa\mathcal{V}^w\mathbf{F}^{-\top} + \kappa\mathcal{V}^w\ln\left(\frac{J\phi^p}{\phi_0^p}\right)\mathbf{F}^{-\top}, \\
&= \kappa\mathcal{V}^w\left[\ln(J^e) - 1\right]\mathbf{F}^{-\top}, \\
\Rightarrow \left(\frac{\partial\mathcal{G}}{\partial\mathbf{F}}\right)_{kL} &= \kappa\mathcal{V}^w\left[\ln(J^e) - 1\right]F_{kL}^{-\top}, \\
&= \kappa\mathcal{V}^w\left[\ln(J^e) - 1\right]F_{Lk}^{-1}.
\end{aligned} \tag{83}$$

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

$$\frac{dC^w}{d\mathbf{F}} = -\left(\frac{\partial C^w}{\partial \phi^p}\right)\left(\frac{\partial \mathcal{G}}{\partial \phi^p}\right)^{-1} \kappa \mathcal{V}^w \left[\ln(J^e) - 1\right] F^{-1}_{Lk}. \tag{84}$$

3.4 Calculation of $\frac{dC^w}{d\mathbf{C}}$

$$\begin{aligned}\left(\frac{dC^w}{d\mathbf{C}}\right)_{KL} &= -\left(\frac{\partial C^w}{\partial \phi^p}\right)\left(\frac{\partial \mathcal{G}}{\partial \phi^p}\right)^{-1}\left(\frac{\partial \mathcal{G}}{\partial \mathbf{C}}\right)_{kL} \\ &= -\left(\frac{\partial C^w}{\partial \phi^p}\right)\left(\frac{\partial \mathcal{G}}{\partial \phi^p}\right)^{-1} \kappa \mathcal{V}^w \left[\ln(J^e) - 1\right] \frac{C^{-1}_{KL}}{2}\end{aligned} \tag{85}$$

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

$$\begin{aligned}\frac{\partial \mathbf{S}}{\partial C^w} &= \kappa \mathcal{V}^w (\ln J^e - 1)\mathbf{C}^{-1}, \\ \Rightarrow \left(\frac{\partial \mathbf{S}}{\partial C^w}\right)_{IJ} &= \kappa \mathcal{V}^w (\ln J^e - 1) C^{-1}_{IJ}.\end{aligned} \tag{86}$$

3.6 Calculation of $\mathbb{C}$

Let us define the material tangent, $\mathbb{C}$, for the quasi-incompressible hydrogel as

$$\mathbb{C}_{IJKL} = 2\left(\frac{\partial S_{IJ}}{\partial C_{KL}} + \frac{\partial S_{IJ}}{\partial C^w}\frac{dC^w}{dC_{KL}}\right) \qquad \Rightarrow \quad \mathbb{C} = \mathbb{C}^{\text{mech}} + \mathbb{C}^{\text{chem}}. \tag{87}$$

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

$$\begin{aligned}\frac{\partial \mathbf{F}}{\partial \mathbf{F}} &= \mathbb{I} \quad \Rightarrow (\mathbb{I})_{iJkL} = \delta_{ik}\delta_{jl}, \\ \frac{\partial \ln J}{\partial \mathbf{F}} &= \frac{1}{J}\det(\mathbf{F})\mathbf{F}^{-\top} = \mathbf{F}^{-\top}, \quad \Rightarrow \frac{\partial \ln J}{\partial F_{kL}} = F^{-1}_{Lk}, \\ \left(\frac{\partial \mathbf{F}^{-1}}{\partial \mathbf{F}}\right)_{IjkL} &= -F^{-1}_{Ik}F^{-1}_{Lj}, \quad \left(\frac{\partial \mathbf{F}^{-\top}}{\partial \mathbf{F}}\right)_{JikL} = -F^{-1}_{Jk}F^{-1}_{Li}, \\ \left(\frac{\partial \mathbf{C}^{-1}}{\partial \mathbf{C}}\right)_{IJKL} &= -\frac{1}{2}\left(C^{-1}_{IK}C^{-1}_{JL} + C^{-1}_{JK}C^{-1}_{IL}\right) = \mathbb{I}_{\mathbf{C}^{-1}}. \qquad [\text{since, } \mathbf{C} = \mathbf{C}^{\top}]\end{aligned} \tag{88}$$

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

$$\mathbf{S} = G\mathbb{1} - \left[G(\phi_0^p)^{2/3} - \kappa\phi_0^p J^s(\ln J^e)\right]\mathbf{C}^{-1}. \tag{89}$$

Thus, the mechanical part of the material tangent, $\mathbb{C}^{\text{mech}}$, is given by,

$$\begin{aligned} \mathbb{C}^{\text{mech}}_{IJKL} &= \frac{\partial S_{IJ}}{C_{KL}} = \kappa\phi_0^p J^s C^{-1}_{IJ}C^{-1}_{KL} + \left((\phi_0^p)^{2/3}G - \phi_0^p J^s \kappa \ln J^e\right)\left(C^{-1}_{IK}C^{-1}_{JL} + C^{-1}_{JK}C^{-1}_{IL}\right), \\ \Rightarrow \mathbb{C}^{\text{mech}} &= \phi_0^p J^s \kappa \mathbf{C}^{-1}\otimes\mathbf{C}^{-1} - 2\left[(\phi_0^p)^{2/3}G - \phi_0^p J^s\kappa\ln J\right]\mathbb{I}_{\mathbf{C}^{-1}}. \end{aligned} \tag{90}$$

Similarly, the chemical part of the material tangent, $\mathbb{C}^{\text{chem}}$ is given by,

$$\begin{aligned} \mathbb{C}^{\text{chem}}_{IJKL} &= 2\frac{\partial S_{IJ}}{\partial C^w}\frac{dC^w}{dC_{KL}}, \\ &= 2\left(\kappa\mathcal{V}^w(\ln J^e - 1)C^{-1}_{IJ}\right)\left[\frac{1}{2}\left(\frac{\partial C^w}{\partial\phi^p}\right)\left(\frac{\partial\mathcal{G}}{\partial\phi^p}\right)^{-1}\kappa\mathcal{V}^w\left[\ln(J^e)-1\right]C^{-1}_{KL}\right], \\ &= \left(\frac{\partial C^w}{\partial\phi^p}\right)\left(\frac{\partial\mathcal{G}}{\partial\phi^p}\right)^{-1}\left(\kappa\mathcal{V}^w(\ln J^e - 1)\right)^2 C^{-1}_{IJ}C^{-1}_{KL}, \\ &= \left(\frac{\partial C^w}{\partial\phi^p}\right)\left(\frac{\partial\mathcal{G}}{\partial\phi^p}\right)^{-1}\left(\kappa\mathcal{V}^w(\ln J^e - 1)\right)^2 \mathbf{C}^{-1}\otimes\mathbf{C}^{-1}. \end{aligned} \tag{91}$$

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

$$\begin{aligned} \mathbb{C}_{IJKL} = \kappa\phi_0^p J^s C^{-1}_{IJ}C^{-1}_{KL} &+ \left((\phi_0^p)^{2/3}G - \phi_0^p J^s\kappa\ln J^e\right)\left(C^{-1}_{IK}C^{-1}_{JL} + C^{-1}_{JK}C^{-1}_{IL}\right) \\ &+ \left(\frac{\partial C^w}{\partial\phi^p}\right)\left(\frac{\partial\mathcal{G}}{\partial\phi^p}\right)^{-1}\left(\kappa\mathcal{V}^w(\ln J^e - 1)\right)^2 C^{-1}_{IJ}C^{-1}_{KL} \end{aligned} \tag{92}$$

3.7 Calculation of $\mathbb{J}$

Let us define $\mathbb{J}$ as,

$$(\mathbb{J})_{IkL} = \frac{\partial J^w_I}{\partial F_{kL}} + \frac{\partial J^w_I}{\partial C^w}\frac{dC^w}{dF_{kL}}. \tag{93}$$

The molar flux for the solvent diffusion is given by,

$$\begin{aligned} &\mathbf{J}^w = -\mathbf{M}^w\,\text{Grad}\,\mu^w \qquad \Rightarrow \quad J^w_I = -M_{IJ}(\text{Grad}\,\mu^w)_J, \\ &\text{where } \mathbf{M}^w = \frac{D^wC^w}{R\theta}\mathbf{C}^{-1} \qquad \Rightarrow \quad M_{IJ} = \frac{D^wC^w}{R\theta}C^{-1}_{IJ}. \end{aligned} \tag{94}$$

Now,

$$\frac{\partial\mathbf{J}^w}{\partial\mathbf{F}} = -\frac{\partial\mathbf{C}^{-1}}{\partial\mathbf{F}}\left(\frac{D^wC^w}{R\theta}\,\text{Grad}\,\mu^w\right) - \frac{D^wC^w}{R\theta}\mathbf{C}^{-1}\frac{\text{Grad}\,\mu^w}{\partial\mathbf{F}}. \tag{95}$$

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

$$\begin{aligned}\mathbf{C}=\mathbf{F}^{\top}\mathbf{F} \quad &\Rightarrow \mathbf{C}^{-1}=\mathbf{F}^{-1}\mathbf{F}^{-\top}, \quad \Rightarrow C^{-1}_{IJ}=F^{-1}_{Im}F^{-\top}_{mJ}=F^{-1}_{Im}F^{-1}_{Jm},\\ \left(\frac{\partial \mathbf{F}^{-1}}{\partial \mathbf{F}}\right)_{IjkL}&=-F^{-1}_{Ik}F^{-1}_{Lj}, \quad \left(\frac{\partial \mathbf{F}^{-\top}}{\partial \mathbf{F}}\right)_{JikL}=-F^{-1}_{Jk}F^{-1}_{Li}.\end{aligned} \tag{96}$$

Hence, the derivative in the first term of (95) can be written as,

$$\begin{aligned}\frac{\partial \mathbf{C}^{-1}}{\partial \mathbf{F}}=\frac{\partial C^{-1}_{IJ}}{\partial F_{kL}}=\frac{\partial}{\partial F_{kL}}\left(F^{-1}_{Im}F^{-1}_{Jm}\right)&=\frac{\partial F^{-1}_{Im}}{\partial F_{kL}}F^{-1}_{Jm}+\frac{\partial F^{-1}_{Jm}}{\partial F_{kL}}F^{-1}_{Im}\\ &=-F^{-1}_{Ik}F^{-1}_{Lm}F^{-1}_{Jm}-F^{-1}_{Jk}F^{-1}_{Lm}F^{-1}_{Im},\\ &=-F^{-1}_{Ik}C^{-1}_{LJ}-F^{-1}_{Jk}C^{-1}_{LI},\end{aligned} \tag{97}$$

which results in

$$\frac{\partial \mathbf{C}^{-1}}{\partial \mathbf{F}}\left(\frac{D^wC^w}{R\theta}\operatorname{Grad}\mu^w\right)=-\frac{D^wC^w}{R\theta}\left[F^{-1}_{Ik}C^{-1}_{LJ}+F^{-1}_{Jk}C^{-1}_{LI}\right](\operatorname{Grad}\mu^w)_J. \tag{98}$$

Now, the derivative in the second term of (95) can be written as,

$$\frac{\partial}{\partial \mathbf{F}}(\operatorname{Grad}\mu^w)=\frac{\partial}{\partial F_{kL}}\left(\frac{\partial \mu^w}{\partial X_J}\right)=\frac{\partial}{\partial X_J}\left(\frac{\partial \mu^w}{\partial F_{kL}}\right)=\frac{\partial}{\partial X_J}\left(\frac{\partial \mu^w}{\frac{\partial x_k}{\partial X_L}}\right)=\delta_{LJ}F^{-1}_{Mk}(\operatorname{Grad}\mu^w)_M, \tag{99}$$

which leads to,

$$\begin{aligned}\frac{D^wC^w}{R\theta}\mathbf{C}^{-1}\frac{\operatorname{Grad}\mu^w}{\partial \mathbf{F}}&=\frac{D^wC^w}{R\theta}C^{-1}_{JI}\delta_{LJ}F^{-1}_{Mk}(\operatorname{Grad}\mu^w)_M,\\ &=\frac{D^wC^w}{R\theta}C^{-1}_{LI}F^{-1}_{Mk}(\operatorname{Grad}\mu^w)_M,\\ &=\frac{D^wC^w}{R\theta}C^{-1}_{LI}F^{-1}_{Jk}(\operatorname{Grad}\mu^w)_J.\end{aligned} \tag{100}$$

Now,

$$\begin{aligned}\frac{\partial J^w_I}{\partial C^w}\frac{dC^w}{dF_{kL}}&=\left[-\frac{D^w}{R\theta}C^{-1}_{IJ}(\operatorname{Grad}\mu^w)_J\right]\left[\left(\frac{\phi^p_0}{\mathcal{V}^w(\phi^p)^2}\right)\left(\frac{\partial \mathcal{G}}{\partial \phi^p}\right)^{-1}\left(\kappa\mathcal{V}^w\left[\ln(J^e)-1\right]F^{-1}_{Lk}\right)\right],\\ &=-\left(\frac{\kappa\mathcal{V}^w\phi^p_0}{\mathcal{V}^w(\phi^p)^2}\right)\left(\frac{\partial \mathcal{G}}{\partial \phi^p}\right)^{-1}\frac{D^w}{R\theta}\left[\ln(J^e)-1\right]F^{-1}_{Lk}C^{-1}_{IJ}(\operatorname{Grad}\mu^w)_J\end{aligned} \tag{101}$$

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

$$\mathbb{J}_{IkL} = \frac{D^w C^w}{R\theta} F^{-1}_{Ik} C^{-1}_{LJ} (\text{Grad}\,\mu^w)_J - \left(\frac{\kappa \mathcal{V}^w \phi^p_0}{\mathcal{V}^w (\phi^p)^2}\right) \left(\frac{\partial \mathcal{G}}{\partial \phi^p}\right)^{-1} \frac{D^w}{R\theta} \left[\ln(J^e) - 1\right] F^{-1}_{Lk} C^{-1}_{IJ} (\text{Grad}\,\mu^w)_J. \tag{102}$$

3.8 Calculation of $\frac{\partial \mathbf{J}^w}{\partial C^w} \frac{dC^w}{d\mu^w}$

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

$$\mathbf{J}^w = -\mathbf{C}^{-1} \frac{D^w C^w}{R\theta} \text{Grad}\,\mu^w \tag{103}$$

Thus, we have,

$$\begin{aligned} \frac{\partial \mathbf{J}^w}{\partial C^w} &= -\frac{D^w}{R\theta} \mathbf{C}^{-1} \text{Grad}\,\mu^w, \\ \Rightarrow \frac{\partial \mathbf{J}^w}{\partial \mu^w} &= -\frac{D^w}{R\theta} \mathbf{C}^{-1} \text{Grad}\,\mu^w \left(\frac{dC^w}{d\mu^w}\right), \\ \Rightarrow \left(\frac{\partial \mathbf{J}^w}{\partial \mu^w}\right)_I &= \left[-\frac{D^w}{R\theta} C^{-1}_{IJ} (\text{Grad}\,\mu^w)_J\right] \left(\frac{dC^w}{d\mu^w}\right) \end{aligned} \tag{104}$$

By substituting the derivative, $\frac{dC^w}{d\mu^w}$, in the above expression, $\frac{\partial \mathbf{J}^w}{\partial \mu^w}$ 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, we 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 within `uel_hydrogel.for`/

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`

Get `nInt` $\leftarrow$ `PROPS` and $\mathtt{n_{dim}}, \mathtt{nStress} \leftarrow$ `JTYPE`

Initialize: $\{\mathbf{N}_a^{\mathbf{u}}, \mathbf{B}_a^{\mathbf{u}}, \mathbf{G}_a^{\mathbf{u}}, \mathbf{N}, \mathbf{B}, \mathbf{G}_{\mathbf{u}}, \Sigma_{\mathbf{F}}, \Sigma_{\mathbf{S}}, \mathbf{k}_e^{\mathbf{uu}}, \mathbf{k}_e^{\mathbf{u}\mu}, \mathbf{k}_e^{\mu\mathbf{u}}, \mathbf{k}_e^{\mu\mu}, \mathbf{r}_e^{\mathbf{u}}, \mathbf{r}_e^{\mu}\} = 0$

Split nodal displacements and chemical potential from `U`: $\mathbf{u}_e, \mu_e^w$

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

Get $w_{\text{int}}, \boldsymbol{\xi}_{\text{int}} \leftarrow$ SUBROUTINE `gaussQuadrtr`$(\mathtt{n_{dim}}, \mathtt{n_{en}})$

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

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

  Get $\mathbf{N}$, $\frac{\partial \mathbf{N_u}}{\partial \boldsymbol{\xi}} \leftarrow$ SUBROUTINE `interpFunc`$(\mathtt{n_{dim}}, \mathtt{n_{en}}, \boldsymbol{\xi}_{\text{int}}(k))$

  Calculate: $\frac{\partial \mathbf{X}}{\partial \boldsymbol{\xi}} = \overline{\mathbf{x}}_e \frac{\partial \mathbf{N_u}}{\partial \boldsymbol{\xi}}$ `// map to the reference configuration`

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

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

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

  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 `neohookean`$_{\mathtt{f}}$`lory`$(\mathtt{PROPS}, \mathbf{F}, \mu, \text{Grad}(\mu^w), \phi^p(t_n), \Delta t)$ `// UMAT returns coupled material tangents and force vectors`

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

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

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

  Calculate the residual sub-vectors, $\mathbf{r}_e^{\mathbf{u}}$ and $\mathbf{r}_e^{\mu}$ `// use the formulation developed in the previous section`

end `// end of integration point loop`

Get `AMATRX, RHS` $\leftarrow$ SUBROUTINE `assembleElement` $(\mathbf{k}_e^{\mathbf{uu}}, \mathbf{k}_e^{\mathbf{u}\mu}, \mathbf{k}_e^{\mu\mathbf{u}}, \mathbf{k}_e^{\mu\mu}, \mathbf{r}_e^{\mathbf{u}}, \mathbf{r}_e^{\mu})$

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. We 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 `neohookean_flory` 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 (axisymmetry)	`U3`	1 (full)
4-node quadrilateral (axisymmetry)	`U4`	4 (full) and 1 (reduced)
3-node triangular (plane-strain)	`U5`	1 (full)
4-node quadrilateral (plane-strain)	`U6`	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.

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.

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