Implementation of Abaqus user element (UEL) subroutine of electro-chemo-mechanical model for polyelectrolyte hydrogels

Bibekananda Datta

Mechanical Engineering, Johns Hopkins University, Baltimore, MD 21211, USA

This chapter contains the details of finite element implementation of the polyelectrolyte hydrogel model as a user-defined element subroutine (UEL) in Abaqus/Standard (Dassault Systèmes, 2024). The coupled electro-chemo-mechanical theory, constitutive model, and finite element implementation have similarities with Narayan and Anand, 2022; Zhang et al., 2020. However, compared to the aforementioned articles, in the current approach, I omitted the governing equations for the electric field and enforced electroneutrality as a local constraint, making the current implementation numerically efficient.

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 model encompassing free energy components from Neo-Hookean potential for entropic stretching of elastomeric network, binary Flory-Huggins mixture potential for polymer-solvent mixing, and dilute ionic solution. Hydrogel has been modeled as a quasi-incompressible elastomer using a penalty function formulation. For fully-integrated 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.

1 Summary of the coupled theory and constitutive model

1.1 Finite strain kinematics

Following the kinematics prescribed for neutral hydrogel in the previous chapter, for pre-swollen polyelectrolyte hydrogel with an initial polymer volume fraction of ϕ_0^p, I can write

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

where, $\mathbf{F}^e$ and $\mathbf{F}^s$ are the elastic and swelling deformation gradients, respectively. The referential concentration for the polymer, C^p, and for the solvent, C^w are defined as the moles per unit hydrated reference volume. Finally, $\mathcal{V}^p$ and $\mathcal{V}^w$, represent the molar volume of the polymer and solvent, respectively. Since the concentration of the counterions and coions present in polyelectrolyte hydrogel is considered to be dilute, I assumed their contribution to the swelling to be negligible and did not include it in the constitutive relation for J^s. Similar to the neutral hydrogel, I can define polymer volume fraction, ϕ^p, as follows,

$$
\phi^p = \frac{C^p\mathcal{V}^p}{C^p\mathcal{V}^p + C^w\mathcal{V}^w}. \tag{1.2}
$$

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

Because of the additional ionic species within the hydrogel, I will have to assume another additional pair of subsurface to describe the additional field equations and corresponding initial and boundary conditions for the dilute species. Thus, in the reference configuration, Γ_g and Γ_T, Γ_h and Γ_{I^w}, and Γ_{H^β} and Γ_{I^β} be three distinct pairs of subsurfaces of the boundary $\partial\Omega_0$. The subsurfaces can be defined as follows,

$$
\begin{aligned}
\partial\Omega_0 &= \Gamma_g \cup \Gamma_T \quad &&\text{and} \quad \Gamma_g \cap \Gamma_T &&= \varnothing, \\
&= \Gamma_h \cup \Gamma_{I^w} \quad &&\text{and} \quad \Gamma_h \cap \Gamma_{I^w} &&= \varnothing, \\
&= \Gamma_{H^\beta} \cup \Gamma_{I^\beta} \quad &&\text{and} \quad \Gamma_{H^\beta} \cap \Gamma_{I^\beta} &&= \varnothing.
\end{aligned}
\tag{1.3}
$$

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,

$$
\begin{aligned}
\mathrm{Div}(\mathbf{FS}) + \rho_R\mathbf{B} = 0 \qquad &\text{in } \Omega_0, \\
\mathbf{u} = \mathbf{g} \qquad &\text{on } \Gamma_g, \\
\mathbf{P}\cdot\mathbf{N} = (\mathbf{FS})\cdot\mathbf{N} = \mathbf{T} \qquad &\text{on } \Gamma_T.
\end{aligned}
\tag{1.4}
$$

Here, $\mathbf{P}$ and $\mathbf{S}$ are the first and the second Piola-Kirchhoff stress tensors, respectively, and $\mathbf{B}$ is the body force per unit of mass in the reference configuration. 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,

$$
\begin{aligned}
\dot{C}^w &= -\operatorname{Div} \mathbf{J}^w && \text{in } \Omega_0, \\
\mu^w(\mathbf{X}, t=0) &= \mu_0^w && \text{in } \Omega_0, \\
\mu^w &= h && \text{on } \Gamma_h, \\
-\mathbf{J}^w \cdot \mathbf{N} &= I^w && \text{on } \Gamma_{I^w}.
\end{aligned}
\tag{1.5}
$$

For the same time interval, $t \in [0, T]$, the diffusion equations for any dilute ionic species, β, and initial and boundary conditions associated with the governing equations are given by,

$$
\begin{aligned}
\dot{C}^\beta &= -\operatorname{Div} \mathbf{J}^\beta + \widehat{C}^\beta && \text{in } \Omega_0, \\
\omega^\beta(\mathbf{X}, t=0) &= \omega_0^\beta && \text{in } \Omega_0, \\
\omega^\beta &= H^\beta && \text{on } \Gamma_{H^\beta}, \\
-\mathbf{J}^\beta \cdot \mathbf{N} &= I^\beta && \text{on } \Gamma_{I^\beta}.
\end{aligned}
\tag{1.6}
$$

Here, $\hat{C}^\beta$ is known as the source or sink term, often relating to chemical production or absorption of the species because of chemical reactions. In this case, I will consider the ionic species to be non-reactive and ignore this term, hence, $\hat{C}^\beta = 0$.

1.3 Electroneutrality condition

The mass flow of ions in and out of PE gel results in the accumulation of charges which is referred to as the referential charge density, Q, and can be written as the sum of all charges as,

$$
Q = F\left(z^p C^p + \sum_\beta z^\beta C^\beta\right). \tag{1.7}
$$

where F is Faraday's constant, z^p and z^β are the charge numbers of polymer chains and ions, β, respectively. The charge density has a unit of charge per unit volume. The non-uniform distribution of ions within the gel and external solution bath gives rise to an electric potential, ψ, within the gel which is known as the Donnan potential. For bulk polyelectrolyte gel, when the length scale is larger than the Debye length scale, the electroneutrality condition states that the net charge in a media has to be zero because of Coulombic interactions among the oppositely charged ionic species which is written as,

$$
z^p C^p + \sum_\beta z^\beta C^\beta = 0. \tag{1.8}
$$

In what follows, I will enforce the electroneutrality condition to compute the electric potential locally within an element during the numerical implementation.

Remark 1. Since I assumed the PE gel to be electroneutral in the proposed theory, the surrounding external solvent also needs to be electroneutral. Hence, I should ensure the electroneutrality condition is maintained when the boundary conditions for the ion transport are prescribed. This also tells us that, I need at least two ionic species within the gel and in the external bath to model the coupled behavior accurately.

1.4 Constitutive relations

The Helmholtz free energy density function, Ψ, for a polyelectrolyte hydrogel with multi-species diffusion, can be described using the following components in an additive manner,

$$\begin{aligned}\Psi = \mu_w^0 C^w + \sum_\beta \omega_\beta^0 C^\beta + \phi_0^p N R\theta \left[\lambda_{\mathrm{L}}^2 \left(\frac{\lambda_c \beta_c}{\lambda_{\mathrm{L}}} + \ln \frac{\beta_c}{\sinh \beta_c}\right) - \left(\frac{\lambda_{\mathrm{L}}}{3}\beta_0\right) \ln J\right] \\ + \frac{\phi_0^p \kappa}{2} J^s (\ln J^e)^2 + \phi_0^p \left(\frac{R\theta}{\mathcal{V}^w}\frac{1-\phi^p}{\phi^p}\left[\ln\left(1-\phi^p\right) + \chi\phi^p\right]\right) + R\theta \sum_\beta C^\beta \left[\ln\left(\frac{C^\beta}{C^w}\right) - 1\right], \\ \text{where, } \beta_c = \mathcal{L}^{-1}\left(\frac{\lambda_c}{\lambda_{\mathrm{L}}}\right), \quad \beta_0 = \mathcal{L}^{-1}\left(\frac{\lambda^0}{\lambda_{\mathrm{L}}}\right), \quad \text{and } \lambda_c = \frac{1}{\sqrt{3}}(J^s)^{1/3}\sqrt{I_1^e}.\end{aligned} \tag{1.9}$$

Compared to the previous free energy description of the neutral gel, an additional term is added here to account for the dilute mixture (entropic) of multiple species, β. In case the behavior of the elastomeric network is described by Gaussian chain statistics, the free energy for the pre-swollen gel can be written as,

$$\begin{aligned}\Psi = \mu_w^0 C^w + \sum_\beta \omega_\beta^0 C^\beta + \frac{G_0}{2}\left[(\phi_0^p J^s)^{2/3} I_1^e - 3 - 2(\phi_0^p)^{2/3}\ln(J)\right] + \frac{\phi_0^p \kappa}{2} J^s (\ln J^e)^2 \\ + \phi_0^p \left(\frac{R\theta}{\mathcal{V}^w}\frac{1-\phi^p}{\phi^p}\left[\ln\left(1-\phi^p\right) + \chi\phi^p\right]\right) + R\theta \sum_\beta C^\beta \left[\ln\left(\frac{C^\beta}{C^w}\right) - 1\right].\end{aligned} \tag{1.10}$$

As the additional species do not interact with the polymer network, the stress expression remains the same as in neutral gel. For Arruda-Boyce type elastomeric network, the Cauchy stress, $\boldsymbol{\sigma}$, is given by,

$$\begin{aligned}\boldsymbol{\sigma} &= 2J^{-1}\left[\mathbf{F}^e \left(\frac{\partial \Psi}{\partial \mathbf{C}^e}\right)\mathbf{F}^{e\top}\right], \\ &= J^{-1}\left[G_0\left(\frac{\lambda_{\mathrm{L}}}{3\lambda_c}\beta_c\right)\mathbf{b} - G_0\left(\frac{\lambda_{\mathrm{L}}}{3}\beta_0\right)(\phi_0^p)^{2/3}\mathbb{1} + \phi_0^p J^s \kappa (\ln J^e)\mathbb{1}\right], \\ &\text{where, } G_0 = NR\theta(\phi_0^p)^{1/3}.\end{aligned} \tag{1.11}$$

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

$$\begin{aligned}
\mathbf{P} = J\boldsymbol{\sigma}\mathbf{F}^{-\top} = \frac{\partial \Psi}{\partial \mathbf{F}^e} &= G_0 \left(\frac{\lambda_{\mathrm{L}}}{3\lambda_c} \beta_c \right) \mathbf{F} - \left[(\phi_0^p)^{2/3} G_0 \left(\frac{\lambda_{\mathrm{L}}}{3} \right) - \phi_0^p J^s \kappa (\ln J^e) \right] \mathbf{F}^{-\top}, \\
\mathbf{S} = J\mathbf{F}^{-1}\boldsymbol{\sigma}\mathbf{F}^{-\top} = \mathbf{F}^{-1}\mathbf{P} &= G_0 \left(\frac{\lambda_{\mathrm{L}}}{3\lambda_c} \beta_c \right) \mathbb{1} - \left[(\phi_0^p)^{2/3} G_0 \left(\frac{\lambda_{\mathrm{L}}}{3} \right) - \phi_0^p J^s \kappa (\ln J^e) \right] \mathbf{C}^{-1}.
\end{aligned} \tag{1.12}$$

where, $\mathbf{C} = \mathbf{F}^\top \mathbf{F}$ is known as the right Cauchy-Green deformation tensor. The mean pressure, p, is given by,

$$\begin{aligned}
p &= -\frac{1}{3} J^e \operatorname{tr}(\boldsymbol{\sigma}), \\
&= \frac{-1}{3\phi_0^p J^s} \left[G_0 \left(\frac{\lambda_{\mathrm{L}}}{3\lambda_c} \beta_c \right) I_1 - 3G_0 \left(\frac{\lambda_{\mathrm{L}}}{3} \beta_0 \right) (\phi_0^p)^{2/3} \right] - \kappa (\ln J^e).
\end{aligned} \tag{1.13}$$

On the other hand, for the Neo-Hookean type elastomeric network, the Cauchy stress, $\boldsymbol{\sigma}$, the first Piola-Kirchhoff stress, $\mathbf{P}$, and the second Piola-Kirchhoff stress, $\mathbf{S}$, are given by,

$$\begin{aligned}
\boldsymbol{\sigma} &= J^{-1} \left[G_0 \mathbf{b} - G_0 (\phi_0^p)^{2/3} \mathbb{1} + \phi_0^p J^s \kappa (\ln J^e) \mathbb{1} \right], \\
\mathbf{P} &= G_0 \mathbf{F} - \left[(\phi_0^p)^{2/3} G_0 - \phi_0^p J^s \kappa (\ln J^e) \right] \mathbf{F}^{-\top}, \\
\mathbf{S} &= G_0 \mathbb{1} - \left[(\phi_0^p)^{2/3} G_0 - \phi_0^p J^s \kappa (\ln J^e) \right] \mathbf{C}^{-1}.
\end{aligned} \tag{1.14}$$

whereas the mean pressure, p, is given by,

$$p = \frac{-1}{3\phi_0^p J^s} \left(G_0 I_1 - 3G_0 (\phi_0^p)^{2/3} \right) - \kappa (\ln J^e). \tag{1.15}$$

Because of the dilute mixture formed between the solvent and the additional ionic species, the chemical potential for the solvent, μ^w, will have an additional osmotic pressure term as follows,

$$\begin{aligned}
\mu^w &= \frac{\partial \Psi}{\partial C^w} + p\mathcal{V}^\beta \\
&= \mu_w^0 + R\theta \left[\phi^p + \ln(1 - \phi^p) + \chi (\phi^p)^2 \right] - \kappa \mathcal{V}^w (\ln J^e) + \frac{\kappa \mathcal{V}^w}{2} (\ln J^e)^2 - R\theta \sum_\beta \frac{C^\beta}{C^w}.
\end{aligned} \tag{1.16}$$

where,

$$\phi^p = \frac{C^p \mathcal{V}^p}{C^p \mathcal{V}^p + C^w \mathcal{V}^w}. \tag{1.17}$$

Let define,

$$\mathcal{P} = \frac{\kappa}{2} (\ln J^e)^2 - \kappa \ln J^e = \frac{\kappa}{2} \left[\ln \left(\frac{J\phi^p}{\phi_0^p} \right) \right]^2 - \kappa \left[\ln \left(\frac{J\phi^p}{\phi_0^p} \right) \right]. \tag{1.18}$$

I used $J^e = \frac{J\phi^p}{\phi^p_0}$ in writing this expression.

By substituting $\mathcal{P}$ and rearranging the constitutive equation for the chemical potential of the solvent, I can write,

$$\mu^0_w + R\theta\left[\phi^p + \ln(1-\phi^p) + \chi(\phi^p)^2\right] - R\theta\sum_\beta \frac{C^\beta}{C^w} + \mathcal{P}\mathcal{V}^w - \mu^w = 0. \tag{1.19}$$

Similarly, the electrochemical potential for dilute ionic species, ω^β, is given by,

$$\begin{aligned}\omega^\beta &= \frac{\partial\Psi}{\partial C^\beta} + p\mathcal{V}^\beta,\\ &= \omega^0_\beta + R\theta\ln\left(\frac{C^\beta}{C^w}\right) + F\psi z^\beta + p\mathcal{V}^\beta\end{aligned} \tag{1.20}$$

By recognizing $\mathcal{P}$ and rearranging the equation, it can be written as,

$$\omega^0_\beta + R\theta\ln\left(\frac{C^\beta}{C^w}\right) + F\psi z^\beta + p\mathcal{V}^\beta - \omega^\beta = 0. \tag{1.21}$$

By substituting the expression for C^β from the above equation into the electroneutrality condition, I have,

$$\sum_\beta C^\beta z^\beta + C^p z^p = 0, \qquad \Rightarrow C^w\sum_\beta z^\beta \exp\left(\frac{\omega^\beta - \omega^0_\beta - F\psi z^\beta - p\mathcal{V}^\beta}{R\theta}\right) + C^p z^p = 0. \tag{1.22}$$

The diffusion-based transport relation for the solvent is given by,

$$\mathbf{J}^w = -\mathbf{M}^w\,\mathrm{Grad}\,\mu^w, \qquad \text{where,}\quad \mathbf{M}^w = \frac{D^w C^w}{R\theta}\mathbf{C}^{-1} \tag{1.23}$$

Similar to the case for the solvent transport, I assumed a diffusion-based referential molar flux relation for the dilute species as,

$$\mathbf{J}^\beta = -\mathbf{M}^\beta\,\mathrm{Grad}\,\omega^\beta, \qquad \text{where,}\quad \mathbf{M}^\beta = \frac{D^\beta C^\beta}{R\theta}\mathbf{C}^{-1}. \tag{1.24}$$

where, D^β is the diffusion coefficient of the species, β, and $\mathbf{M}^\beta$ is referred to as the mobility tensor of the same species.

Remark 2. The gradient of electrochemical potential can be evaluated as,

$$\mathrm{Grad}\,\omega^\beta = \frac{R\theta}{C^\beta}\mathrm{Grad}(C^\beta) + Fz^\beta\,\mathrm{Grad}(\psi) + \mathcal{V}^\beta\,\mathrm{Grad}(p). \tag{1.25}$$

Thus, the constitutive relation for the referential molar flux, $\mathbf{J}^\beta$, can be written as,

$$\mathbf{J}^\beta = -\mathbf{C}^{-1} D^\beta \left[\mathrm{Grad}(C^\beta) + \frac{F C^\beta z^\beta}{R\theta} \mathrm{Grad}(\psi) + \frac{C^\beta \mathcal{V}^\beta}{R\theta} \mathrm{Grad}(p) \right] \tag{1.26}$$

In absence of deformation, *i.e.*, $\mathbf{F} = \mathbb{1} \Rightarrow \mathbf{C}^{-1} = \mathbb{1} \Rightarrow p = 0$, the above flux relation is reduced to,

$$\mathbf{J}^\beta = -D^\beta \left[\mathrm{Grad}(C^\beta) + \frac{F C^\beta z^\beta}{R\theta} \mathrm{Grad}(\psi) \right] \tag{1.27}$$

This is known as the Nernst-Planck equation for ionic species in a polyelectrolyte fluid medium in the absence of advection.

2 Finite element formulation

Based on the prescribed constitutive model for the pre-swollen polyelectrolyte hydrogel, it is convenient to select $\mathbf{u}$, μ^w, and ω^β as the nodal variables for the finite element framework which leaves C^w, C^β, and ψ as the internal variables.

Remark 3. For an electroneutral polyelectrolyte gel, at least two ions of opposite types of charge are required. In developing the finite element formulation, I will just consider one ionic species. However, the formulation can be readily extended for multiple ionic species. This extension will require defining additional tangent matrix and residual vectors and performing local iteration for additional internal variables.

2.1 Weighted residual based weak formulation

Let $\mathbf{u} \in \mathscr{U}$ be the trial solution which satisfies $u_i = g_i$, and $\mathbf{w} \in \mathscr{W}$ be a vector test (or weight) function which satisfies $w_i = 0$ on Γ_g, I can write the weak form of momentum balance equation as

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

Application of the divergence theorem results in the final weak form of stress equilibrium. In the context of the principle of virtual work, the first term in the weak form is called internal work and the second and third terms are collectively known as external work.

Now, assuming Θ represents a test function that vanishes on Γ_h, I can write the weak form of mass balance equation for the solvent as,

$$
\begin{aligned}
&\int_{\Omega_0} \dot{C}^w \; \Theta \; dV + \int_{\Omega_0} \operatorname{Div} \mathbf{J}^w \; \Theta \; dV = 0, \\
\Rightarrow &\int_{\Omega_0} \dot{C}^w \; \Theta \; dV - \int_{\Omega_0} \mathbf{J}^w \cdot \operatorname{Grad}(\Theta) \; dV - \int_{\Gamma_{I^w}} I^w \; \Theta \; dS = 0.
\end{aligned}
\tag{2.2}
$$

Similar to the weak form of stress equilibrium, applying the divergence theorem results in the final weak form of mass balance.

Following the same procedure as before, for any arbitrary test function η which vanishes on Γ_{H^β}, the generic weak form of the governing equation for the mass balance of the ionic species at the element level is given by,

$$
\begin{aligned}
&\int_{\Omega_0} \dot{C}^{\beta_1} \; \eta_1 \; dV - \int_{\Omega_0} \mathbf{J}^{\beta_1} \cdot \operatorname{Grad}(\eta_1) \; dV - \int_{\Gamma_{I^{\beta_1}}} I^{\beta_1} \; \eta_1 \; dS = 0, \\
&\int_{\Omega_0} \dot{C}^{\beta_2} \; \eta_2 \; dV - \int_{\Omega_0} \mathbf{J}^{\beta_2} \cdot \operatorname{Grad}(\eta_2) \; dV - \int_{\Gamma_{I^{\beta_2}}} I^{\beta_2} \; \eta_2 \; dS = 0
\end{aligned}
\tag{2.3}
$$

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, mass balance for the solvent, mass balance for the ionic species, and electroneutrality condition, I can write the weak forms for a system of governing equations for each element as,

$$
\begin{aligned}
\mathcal{W}_e^{\mathbf{u}} &= -\int_{\Omega_0^e} \mathbf{FS} : \operatorname{Grad}(\mathbf{W}) \; dV + \int_{\Omega_0^e} \rho_{\mathrm{R}} \mathbf{B} \cdot \mathbf{W} \; dV + \int_{\Gamma_T^e} \mathbf{T} \cdot \mathbf{W} \; dS = 0, \\
\mathcal{W}_e^{\mu} &= \int_{\Omega_0} \dot{C}^w \; \Theta \; dV - \int_{\Omega_0} \mathbf{J}^w \cdot \operatorname{Grad}(\Theta) \; dV - \int_{\Gamma_{I^w}} I^{we} \; \Theta \; dS = 0, \\
\mathcal{W}_e^{\omega_1} &= \int_{\Omega_0^e} \dot{C}^{\beta_1} \; \eta_1 \; dV - \int_{\Omega_0^e} \mathbf{J}^{\beta_1} \cdot \operatorname{Grad}(\eta_1) \; dV - \int_{\Gamma_{I^{\beta_1}}^e} I^{\beta_1 e} \; \eta_1 \; dS = 0, \\
\mathcal{W}_e^{\omega_2} &= \int_{\Omega_0^e} \dot{C}^{\beta_2} \; \eta_2 \; dV - \int_{\Omega_0^e} \mathbf{J}^{\beta_2} \cdot \operatorname{Grad}(\eta_2) \; dV - \int_{\Gamma_{I^{\beta_2}}^e} I^{\beta_2 e} \; \eta_2 \; dS = 0.
\end{aligned}
\tag{2.4}
$$

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,

$$
X_i = \sum_{a=1}^{\mathrm{n_{en}}} N_a X_a^i \quad \Rightarrow \mathbf{X} = \mathbf{N_u} \mathbf{X}^e, \tag{2.5}
$$

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,

$$\mathbf{N_u} = \begin{bmatrix} N_1 & 0 & 0 & N_2 & 0 & 0 & \cdots & \cdots & N_{\mathrm{n_{en}}} & 0 & 0 \\ 0 & N_1 & 0 & 0 & N_2 & 0 & \cdots & \cdots & 0 & N_{\mathrm{n_{en}}} & 0 \\ 0 & 0 & N_1 & 0 & 0 & N_2 & \cdots & \cdots & 0 & 0 & N_{\mathrm{n_{en}}} \end{bmatrix}_{\mathrm{n_{dim}}\times \mathrm{n_{en}} * \mathrm{n_{dim}}}, \tag{2.6}$$
$$\mathbf{X}^e = \begin{bmatrix} X_1 & Y_1 & Z_1 & X_2 & Y_2 & Z_2 & \cdots & \cdots & X_{\mathrm{n_{en}}} & Y_{\mathrm{n_{en}}} & Z_{\mathrm{n_{en}}} \end{bmatrix}^\top_{\mathrm{n_{en}} * \mathrm{n_{dim}} \times 1}.$$

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 $\mathbf{N}^a_{\mathbf{u}}$, then for a three dimensional case, I can write $\mathbf{N}^{\mathbf{u}}_a$ and X^e_a as,

$$\mathbf{N}^{\mathbf{u}}_a = \begin{bmatrix} N_i & 0 & 0 \\ 0 & N_i & 0 \\ 0 & 0 & N_i \end{bmatrix}_{\mathrm{n_{dim}} \times \mathrm{n_{dim}}}, \quad \mathbf{X}_a = \begin{bmatrix} X_i \\ Y_i \\ Z_i \end{bmatrix}_{\mathrm{n_{dim}} \times 1}, \tag{2.7}$$

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.

I now employ the standard Galerkin procedure to approximate the degrees of freedom, $\mathbf{u}_e$, μ^w_e, $\omega^{\beta_1}_e$, and $\omega^{\beta_2}_e$, and their corresponding weight functions within each element as follows,

$$\begin{aligned} u^i_e(\mathbf{X}) &= \sum_a N^{\mathbf{u}}_a(\mathbf{X})\ u^i_a, \quad \text{and} \quad W^i(\mathbf{X}) = \sum_a N^{\mathbf{u}}_a(\mathbf{X})\ W^i_a, \\ \mu^w_e(\mathbf{X}) &= \sum_a N^{\mu}_a(\mathbf{X})\ \mu^w_a, \quad \text{and} \quad \Theta(\mathbf{X}) = \sum_a N^{\mu}_a(\mathbf{X})\ \Theta_a, \\ \omega^{\beta_2}_e(\mathbf{X}) &= \sum_a N^{\omega}_a(\mathbf{X})\ \omega^{\beta_1}_a, \quad \text{and} \quad \eta_1(\mathbf{X}) = \sum_a N^{\omega}_a(\mathbf{X})\ \eta_{1a}, \\ \omega^{\beta_2}_e(\mathbf{X}) &= \sum_a N^{\omega}_a(\mathbf{X})\ \omega^{\beta_2}_a, \quad \text{and} \quad \eta_2(\mathbf{X}) = \sum_a N^{\omega}_a(\mathbf{X})\ \eta_{2a}. \end{aligned} \tag{2.8}$$

Here, $N^{\mathbf{u}}_a$, N^{μ}_a, and N^{ω}_a are the interpolation functions correspond to the nodal values of $\mathbf{u}_a$, μ^w_a, and $\omega^{\beta_i}_a$, respectively. In the standard Galerkin weighted residual approach, the same interpolation functions are used for any nodal variable and its weight function pair. However, it does not restrict using different interpolation functions for other nodal fields and their weight functions in a multi-field finite element framework. Here, N_a is the interpolation function corresponding to each element node. $\mathbf{u}^e$, μ^{we}, ${\omega^{\beta_1}}^e$, and ${\omega^{\beta_2}}^e$ are the vectors containing nodal variables for the element. Unlike displacement field, solvent chemical potential, μ^w, and ionic species electrochemical potential, ω^{β_1} and ω^{β_2} are scalar fields, the corresponding shape function matrix, $\mathbf{N_u}$, and vectors, $\mathbf{N}_\mu$ and $\mathbf{N}_\omega$. are given by, Here, $\mathbf{N_u}$, $\mathbf{N}_\mu$, and $\mathbf{N}_\omega$ are the interpolation function matrix for the element nodal displacement vector, $\mathbf{u}$, nodal solvent chemical potential, μ^w, and nodal electrochemical potential for ions, ω^{β_1} and ω^{β_2}, respectively.

These matrices are represented as follows,

$$
\begin{aligned}
\mathbf{N}_{\mathbf{u}} &= \begin{bmatrix} N_1^{\mathbf{u}} & 0 & 0 & N_2^{\mathbf{u}} & 0 & 0 & \cdots & \cdots & N_{\mathrm{n_{en}}}^{\mathbf{u}} & 0 & 0 \\ 0 & N_1^{\mathbf{u}} & 0 & 0 & N_2^{\mathbf{u}} & 0 & \cdots & \cdots & 0 & N_{\mathrm{n_{en}}}^{\mathbf{u}} & 0 \\ 0 & 0 & N_1^{\mathbf{u}} & 0 & 0 & N_2^{\mathbf{u}} & \cdots & \cdots & 0 & 0 & N_{\mathrm{n_{en}}}^{\mathbf{u}} \end{bmatrix}, \\
\mathbf{N}_{\mu} &= \begin{bmatrix} N_1^{\mu} & N_2^{\mu} & N_3^{\mu} & \cdots & \cdots & N_{\mathrm{n_{en}}}^{\mu} \end{bmatrix}, \\
\text{and} \quad \mathbf{N}_{\omega} &= \begin{bmatrix} N_1^{\omega} & N_2^{\omega} & N_3^{\omega} & \cdots & \cdots & N_{\mathrm{n_{en}}}^{\omega} \end{bmatrix}.
\end{aligned} \tag{2.9}
$$

Using these matrix and vector forms of shape function, the Galerkin approximation for nodal variables can be written as,

$$
\mathbf{u}_e = \mathbf{N}_{\mathbf{u}}\mathbf{u}, \qquad \mu_e^w = \mathbf{N}_{\mu}\mu^w, \qquad \omega_e^{\beta_1} = \mathbf{N}_{\omega}\omega^{\beta_1}, \quad \text{and } \omega_e^{\beta_2} = \mathbf{N}_{\omega}\omega^{\beta_2}. \tag{2.10}
$$

Here, $\mathbf{N}_{\mathbf{u}}$, $\mathbf{N}_{\mu}$, and $\mathbf{N}_{\omega}$ are the interpolation function matrix for the element nodal displacement vector, $\mathbf{u}$, nodal solvent chemical potential, μ^w, and nodal electrochemical potential for ions, ω^{β_1}, and ω^{β_2} respectively. The deformation gradient, $\mathbf{F}$, thus can be calculated as,

$$
\mathbf{F} = \frac{\partial \mathbf{x}}{\partial \mathbf{X}} = \mathbb{1} + \frac{\partial \mathbf{u}}{\partial \mathbf{X}} = \mathbb{1} + [\overline{\mathbf{u}}_e]\left[\frac{\partial \mathbf{N}_{\mathbf{u}}}{\partial \mathbf{X}}\right] \quad \Rightarrow F_{iJ} = \delta_{ij} + \sum_{a=1}^{\mathrm{n_{en}}} \frac{\partial N_a}{\partial X_J} u_a^i. \tag{2.11}
$$

where u_a^i denotes i-th nodal displacement component of node a, and N_a is the shape function corresponding to the same node. $\left[\frac{\partial \mathbf{N}_{\mathbf{u}}}{\partial \mathbf{X}}\right]$ 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.

Thus, for arbitrary test functions, $\mathbf{W}$, Θ, η_1, and η_2, the system of residuals for each element can be written as,

$$
\begin{aligned}
\mathbf{R}_e^{\mathbf{u}} &= -\int_{\Omega_0^e} \mathbf{F}\mathbf{S} : \mathrm{Grad}(\mathbf{N}_{\mathbf{u}})\ dV + \int_{\Omega_0^e} \rho_{\mathrm{R}} \mathbf{N}_{\mathbf{u}}^{\top}\mathbf{B}\ dV + \int_{\Gamma_T^e} \mathbf{N}_{\mathbf{u}}^{\top}\mathbf{T}\ dS, \\
\mathbf{R}_e^{\mu} &= -\int_{\Omega_0^e} \mathbf{N}_{\mu}^{\top}\dot{C}^w\ dV + \int_{\Omega_0^e} \mathbf{J}^w \cdot \mathrm{Grad}(\mathbf{N}_{\mu})\ dV + \int_{\Gamma_{I^w}^e} \mathbf{N}_{\mu}^{\top} I^w\ dS, \\
\mathbf{R}_e^{\omega_1} &= -\int_{\Omega_0^e} \mathbf{N}_{\omega}^{\top}\dot{C}^{\beta_1}\ dV + \int_{\Omega_0^e} \mathbf{J}^{\beta_1} \cdot \mathrm{Grad}(\mathbf{N}_{\omega_1})\ dV + \int_{\Gamma_{I^\beta}^e} \mathbf{N}_{\omega}^{\top} I^{\beta_1}\ dS, \\
\mathbf{R}_e^{\omega_2} &= -\int_{\Omega_0^e} \mathbf{N}_{\omega}^{\top}\dot{C}^{\beta_2}\ dV + \int_{\Omega_0^e} \mathbf{J}^{\beta} \cdot \mathrm{Grad}(\mathbf{N}_{\omega_1})\ dV + \int_{\Gamma_{I^\beta}^e} \mathbf{N}_{\omega}^{\top} I^{\beta_2}\ dS
\end{aligned} \tag{2.12}
$$

I now use first-order discretization on the time derivative of referential concentrations at current time $t + \Delta t$

$$
\dot{C}^w = \frac{C^w - C_t^w}{\Delta t}, \quad \dot{C}^{\beta_1} = \frac{C^{\beta_1} - C_t^{\beta_1}}{\Delta t}, \quad \text{and} \quad \dot{C}^{\beta_2} = \frac{C^{\beta_2} - C_t^{\beta_2}}{\Delta t}, \tag{2.13}
$$

where, C^w, C^{β_1}, C^{β_2} are the referential concentration of the corresponding species at time $t + \Delta t$, and C_t^w, $C_t^{\beta_1}$, $C_t^{\beta_2}$ are the referential concentration of the corresponding species at previous time t. By using the spatial and the temporal discretizations (2.8)-(2.13), I now write the element level residuals as follows,

$$
\begin{aligned}
R_a^{u_i} &= -\int\limits_{\Omega_0^e} \frac{\partial N_a^{\mathbf{u}}}{\partial X_I} F_{iJ} S_{JI}\ dV + \int\limits_{\Omega_0^e} \rho_{\mathrm{R}} N_a^{\mathbf{u}} B_i\ dV + \int\limits_{\Gamma_{\mathbf{T}}^e} N_a^{\mathbf{u}} T_i\ dS, \\
R_a^{\mu} &= -\int\limits_{\Omega_0^e} N_a^{\mu} \left(\frac{C^w - C_t^w}{\Delta t}\right)\ dV + \int\limits_{\Omega_0^e} \frac{\partial N_a^{\mu}}{\partial X_I} J_I^w\ dV + \int\limits_{\Gamma_{I^w}^e} N_a^{\mu} I^w\ dS, \\
R_a^{\omega_1} &= -\int\limits_{\Omega_0^e} N_a^{\omega} \left(\frac{C^{\beta_1} - C_t^{\beta_1}}{\Delta t}\right)\ dV + \int\limits_{\Omega_0^e} \frac{\partial N_a^{\omega}}{\partial X_I} J_I^{\beta_1}\ dV + \int\limits_{\Gamma_{I^{\beta_1}}^e} N_a^{\omega} I^{\beta_1}\ dS, \\
R_a^{\omega_2} &= -\int\limits_{\Omega_0^e} N_a^{\omega} \left(\frac{C^{\beta_2} - C_t^{\beta_2}}{\Delta t}\right)\ dV + \int\limits_{\Omega_0^e} \frac{\partial N_a^{\omega}}{\partial X_I} J_I^{\beta_2}\ dV + \int\limits_{\Gamma_{I^{\beta_2}}^e} N_a^{\omega} I^{\beta_2}\ dS.
\end{aligned}
\tag{2.14}
$$

The element residual vector can be constructed using its components as follows,

$$
\mathbf{R}_e = \{\mathbf{R}_a^{\mathbf{u}},\ R_a^{\mu},\ R_a^{\omega_1},\ R_a^{\omega_2}\}_{\mathrm{nen}\times 1}^{\top}. \tag{2.15}
$$

2.3 Consistent linearization and Newton-Raphson method

Constitutive relations for the second Piola-Kirchhoff stress, $\mathbf{S}$, referential molar flux of the solvent, $\mathbf{J}^w$ and the ionic species $\mathbf{J}^{\beta}$, as well as the internal variables are nonlinear functions degrees of freedom, *i.e.*, displacement vector, $\mathbf{u}$, chemical potential of the solvent, μ^w, and electrochemical potential for the ionic species, ω^{β_1} and ω^{β_2}.

$$
\begin{aligned}
\mathbf{S} &= \hat{\mathbf{S}}\left(\mathbf{C}; C^w, C^{\beta_1}, C^{\beta_2}\right), \\
\mu^w &= \hat{\mu}^w\left(\mathbf{C}; C^w, C^{\beta_1}, C^{\beta_2}\right), \\
\omega^{\beta_1} &= \hat{\omega}^{\beta_1}\left(\mathbf{C}; C^w, C^{\beta_1}, C^{\beta_2}, \psi\right), \\
\omega^{\beta_2} &= \hat{\omega}^{\beta_2}\left(\mathbf{C}; C^w, C^{\beta_1}, C^{\beta_2}, \psi\right) \\
\mathbf{J}^w &= \hat{\mathbf{J}}^w\left(\mathbf{C}, \mathrm{Grad}\,\mu^w, \mathrm{Grad}\,\omega^{\beta_1}, \mathrm{Grad}\,\omega^{\beta_1}; C^w, C^{\beta_1}, C^{\beta_2}\right), \\
\mathbf{J}^{\beta_1} &= \hat{\mathbf{J}}^{\beta_1}\left(\mathbf{C}, \mathrm{Grad}\,\mu^w, \mathrm{Grad}\,\omega^{\beta_1}, \mathrm{Grad}\,\omega^{\beta_2}; C^w, C^{\beta_1}, C^{\beta_2}\right), \\
\mathbf{J}^{\beta_2} &= \hat{\mathbf{J}}^{\beta_2}\left(\mathbf{C}, \mathrm{Grad}\,\mu^w, \mathrm{Grad}\,\omega^{\beta_1}, \mathrm{Grad}\,\omega^{\beta_2}; C^w, C^{\beta_1}, C^{\beta_2}\right).
\end{aligned}
\tag{2.16}
$$

Within each element, the constitutive expressions for solvent chemical potential, ion electrochemical potentials, and electroneutrality condition form a system of nonlinear equations as follows,

$$\begin{aligned}
\mathcal{G}_1\left(C^w, C^{\beta_1}, C^{\beta_2}, \psi\right) &\equiv \mu^w - \mu_e^w = 0,\\
\mathcal{G}_2\left(C^w, C^{\beta_1}, C^{\beta_2}, \psi\right) &\equiv \omega^{\beta_1} - \omega_e^{\beta_1} = 0,\\
\mathcal{G}_3\left(C^w, C^{\beta_1}, C^{\beta_2}, \psi\right) &\equiv \omega^{\beta_2} - \omega_e^{\beta_2} = 0,\\
\mathcal{G}_4\left(C^w, C^{\beta_1}, C^{\beta_2}, \psi\right) &\equiv z^p C^p + \sum_{\beta} z^{\beta} C^{\beta} = 0,
\end{aligned} \tag{2.17}$$

where μ^w, ω^{β_1}, and ω^{β_2} are the nonlinear constitutive expressions obtained using specific material model and constitutive definitions, and μ_e^w, $\omega_e^{\beta_1}$, and $\omega_e^{\beta_2}$ are the approximation of nodal solution within the element at $t + \Delta t$. This system of nonlinear equations can be expressed in a compact form as follows,

$$\mathcal{G}\left(\mathbf{F}, \mu^w, \omega^{\beta_1}, \omega^{\beta_2}; C^w, C^{\beta_1}, C^{\beta_2}, \psi\right) = \begin{Bmatrix} \mathcal{G}_1 \\ \mathcal{G}_2 \\ \mathcal{G}_3 \\ \mathcal{G}_4 \end{Bmatrix} = \begin{Bmatrix} 0 \\ 0 \\ 0 \\ 0 \end{Bmatrix}. \tag{2.18}$$

I need to linearize the system of element residuals along the independent variables (degrees of freedom) to employ Newton-Raphson iteration. Assuming, at any time $t + \Delta t$, the initial guess for the independent nodal variables are $\mathbf{u}_b$, μ_b^w, $\omega_b^{\beta_1}$, and $\omega_b^{\beta_2}$, following matrix form of a system of linear equations can be obtained by using Newton-Raphson procedure,

$$\begin{bmatrix}
\frac{\partial \mathbf{R}_a^{\mathbf{u}}}{\partial \mathbf{u}_b} & \frac{\partial \mathbf{R}_a^{\mathbf{u}}}{\partial \mu_b^w} & \frac{\partial \mathbf{R}_a^{\mathbf{u}}}{\partial \omega_b^{\beta 1}} & \frac{\partial \mathbf{R}_a^{\mathbf{u}}}{\partial \omega_b^{\beta 2}} \\
\frac{\partial R_a^{\mu}}{\partial \mathbf{u}_b} & \frac{\partial R_a^{\mu}}{\partial \mu_b^w} & \frac{\partial R_a^{\mu}}{\partial \omega_b^{\beta 1}} & \frac{\partial R_a^{\mu}}{\partial \omega_b^{\beta 2}} \\
\frac{\partial R_a^{\omega 1}}{\partial \mathbf{u}_b} & \frac{\partial R_a^{\omega 1}}{\partial \mu_b^w} & \frac{\partial R_a^{\omega 1}}{\partial \omega_b^{\beta 1}} & \frac{\partial R_a^{\omega 1}}{\partial \omega_b^{\beta 2}} \\
\frac{\partial R_a^{\omega 2}}{\partial \mathbf{u}_b} & \frac{\partial R_a^{\omega 2}}{\partial \mu_b^w} & \frac{\partial R_a^{\omega 2}}{\partial \omega_b^{\beta 1}} & \frac{\partial R_a^{\omega 2}}{\partial \omega_b^{\beta 2}}
\end{bmatrix}
\begin{Bmatrix} \Delta \mathbf{u}_b \\ \Delta \mu_b^w \\ \Delta \omega_b^{\beta_1} \\ \Delta \omega_b^{\beta_2} \end{Bmatrix}
= \begin{Bmatrix} \mathbf{R}_a^{\mathbf{u}} \\ R_a^{\mu} \\ R_a^{\omega_1} \\ R_a^{\omega_2} \end{Bmatrix}. \tag{2.19}$$

By assuming nonlinear constitutive relations for the PE gel (2.16), the element residual vector (2.14) can be linearized at the current time step, $t + \Delta t$, to obtain the element tangent stiffness matrix, $\mathbf{K}_e$, as below,

$$\mathbf{K}_e = -\frac{d\mathbf{R}_e}{d\mathbf{d}_e} = -\begin{bmatrix}
\frac{\partial \mathbf{R}_a^{\mathbf{u}}}{\partial \mathbf{u}_b} & \frac{\partial \mathbf{R}_a^{\mathbf{u}}}{\partial \mu_b^w} & \frac{\partial \mathbf{R}_a^{\mathbf{u}}}{\partial \omega_b^{\beta 1}} & \frac{\partial \mathbf{R}_a^{\mathbf{u}}}{\partial \omega_b^{\beta 2}} \\
\frac{\partial R_a^{\mu}}{\partial \mathbf{u}_b} & \frac{\partial R_a^{\mu}}{\partial \mu_b^w} & \frac{\partial R_a^{\mu}}{\partial \omega_b^{\beta 1}} & \frac{\partial R_a^{\mu}}{\partial \omega_b^{\beta 2}} \\
\frac{\partial R_a^{\omega 1}}{\partial \mathbf{u}_b} & \frac{\partial R_a^{\omega 1}}{\partial \mu_b^w} & \frac{\partial R_a^{\omega 1}}{\partial \omega_b^{\beta 1}} & \frac{\partial R_a^{\omega 1}}{\partial \omega_b^{\beta 2}} \\
\frac{\partial R_a^{\omega 2}}{\partial \mathbf{u}_b} & \frac{\partial R_a^{\omega 2}}{\partial \mu_b^w} & \frac{\partial R_a^{\omega 2}}{\partial \omega_b^{\beta 1}} & \frac{\partial R_a^{\omega 2}}{\partial \omega_b^{\beta 2}}
\end{bmatrix}. \tag{2.20}$$

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 condition is done by Abaqus once the element stiffness (tangent) matrix and element residual vectors are defined in the user element subroutine (UEL).

$$\mathbf{K} = \mathop{\mathsf{A}}_{e=1}^{\mathrm{nel}} \mathbf{k}^e, \qquad \text{and} \qquad \mathbf{R} = \mathop{\mathsf{A}}_{e=1}^{\mathrm{nel}} \mathbf{R}^e. \tag{2.21}$$

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

$$[\mathbf{K}]\{\Delta\mathbf{d}\} = \{\mathbf{R}\}, \tag{2.22}$$

In Newton-Raphson procedure, with an initial guess for $\mathbf{d}$, this system of linear equations can be solved for the 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.

$$\mathbf{d}_{k+1} = \mathbf{d}_k + \Delta\mathbf{d} \tag{2.23}$$

$$
\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} & k_{11}^{u_1\omega_1} & k_{11}^{u_1\omega_2} & \cdots & \cdots & \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_{1\mathrm{n_{en}}}^{u_1\omega_1} & k_{1\mathrm{n_{en}}}^{u_1\omega_2} \\
k_{11}^{u_2u_1} & k_{11}^{u_2u_2} & k_{11}^{u_2u_3} & k_{11}^{u_2\mu} & k_{11}^{u_2\omega_1} & k_{11}^{u_2\omega_2} & \cdots & \cdots & \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_{1\mathrm{n_{en}}}^{u_2\omega_1} & k_{1\mathrm{n_{en}}}^{u_2\omega_2} \\
k_{11}^{u_3u_1} & k_{11}^{u_3u_2} & k_{11}^{u_3u_3} & k_{11}^{u_3\mu} & k_{11}^{u_3\omega_1} & k_{11}^{u_3\omega_2} & \cdots & \cdots & \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_{1\mathrm{n_{en}}}^{u_3\omega_1} & k_{1\mathrm{n_{en}}}^{u_3\omega_1} \\
k_{11}^{\mu u_1} & k_{11}^{\mu u_2} & k_{11}^{\mu u_3} & k_{11}^{\mu\mu} & k_{11}^{\mu\omega_1} & k_{11}^{\mu\omega_2} & \cdots & \cdots & \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} & k_{1\mathrm{n_{en}}}^{\mu\omega_1} & k_{1\mathrm{n_{en}}}^{\mu\omega_2} \\
k_{11}^{\omega_1u_1} & k_{11}^{\omega_1u_2} & k_{11}^{\omega_1u_3} & k_{11}^{\omega_1\mu} & k_{11}^{\omega_1\omega_1} & k_{11}^{\omega_1\omega_1} & \cdots & \cdots & \cdots & k_{1\mathrm{n_{en}}}^{\omega_1u_1} & k_{1\mathrm{n_{en}}}^{\omega_1u_2} & k_{1\mathrm{n_{en}}}^{\omega_1u_3} & k_{1\mathrm{n_{en}}}^{\omega_1\mu} & k_{1\mathrm{n_{en}}}^{\omega_1\omega_1} & k_{1\mathrm{n_{en}}}^{\omega_1\omega_2} \\
k_{11}^{\omega_2u_1} & k_{11}^{\omega_2u_2} & k_{11}^{\omega_2u_3} & k_{11}^{\omega_2\mu} & k_{11}^{\omega_2\omega_1} & k_{11}^{\omega_2\omega_2} & \cdots & \cdots & \cdots & k_{1\mathrm{n_{en}}}^{\omega_2u_1} & k_{1\mathrm{n_{en}}}^{\omega_2u_2} & k_{1\mathrm{n_{en}}}^{\omega_2u_3} & k_{1\mathrm{n_{en}}}^{\omega_2\mu} & k_{1\mathrm{n_{en}}}^{\omega_2\omega_1} & k_{1\mathrm{n_{en}}}^{\omega_2\omega_2} \\
\vdots & & \vdots & & \vdots & & \ddots & & & & \vdots & & \vdots & & \vdots \\
\vdots & & \vdots & & \vdots & & & \ddots & & & \vdots & & \vdots & & \vdots \\
\vdots & & \vdots & & \vdots & & & & \ddots & & \vdots & & \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} & k_{\mathrm{n_{en}}1}^{u_1\omega_1} & k_{\mathrm{n_{en}}1}^{u_1\omega_2} & \cdots & \cdots & \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}n_{en}}}^{u_1\omega_1} & k_{\mathrm{n_{en}n_{en}}}^{u_1\omega_2} \\
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} & k_{\mathrm{n_{en}}1}^{u_2\omega_1} & k_{\mathrm{n_{en}}1}^{u_2\omega_2} & \cdots & \cdots & \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}n_{en}}}^{u_2\omega_1} & k_{\mathrm{n_{en}n_{en}}}^{u_2\omega_2} \\
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} & k_{\mathrm{n_{en}}1}^{u_3\omega_1} & k_{\mathrm{n_{en}}1}^{u_3\omega_2} & \cdots & \cdots & \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}n_{en}}}^{u_3\omega_1} & k_{\mathrm{n_{en}n_{en}}}^{u_3\omega_2} \\
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} & k_{\mathrm{n_{en}}1}^{\mu\omega_1} & k_{\mathrm{n_{en}}1}^{\mu\omega_2} & \cdots & \cdots & \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} & k_{\mathrm{n_{en}n_{en}}}^{\mu\omega_1} & k_{\mathrm{n_{en}n_{en}}}^{\mu\omega_2} \\
k_{\mathrm{n_{en}}1}^{\omega_1u_1} & k_{\mathrm{n_{en}}1}^{\omega_1u_2} & k_{\mathrm{n_{en}}1}^{\omega_1u_3} & k_{\mathrm{n_{en}}1}^{\omega_1\mu} & k_{\mathrm{n_{en}}1}^{\omega_1\omega_1} & k_{\mathrm{n_{en}}1}^{\omega_1\omega_2} & \cdots & \cdots & \cdots & k_{\mathrm{n_{en}n_{en}}}^{\omega_1u_1} & k_{\mathrm{n_{en}n_{en}}}^{\omega_1u_2} & k_{\mathrm{n_{en}n_{en}}}^{\omega_1u_3} & k_{\mathrm{n_{en}n_{en}}}^{\omega_1\mu} & k_{\mathrm{n_{en}n_{en}}}^{\omega_1\omega_1} & k_{\mathrm{n_{en}n_{en}}}^{\omega_1\omega_2} \\
k_{\mathrm{n_{en}}1}^{\omega_2u_1} & k_{\mathrm{n_{en}}1}^{\omega_2u_2} & k_{\mathrm{n_{en}}1}^{\omega_2u_3} & k_{\mathrm{n_{en}}1}^{\omega_2\mu} & k_{\mathrm{n_{en}}1}^{\omega_2\omega_1} & k_{\mathrm{n_{en}}1}^{\omega_2\omega_2} & \cdots & \cdots & \cdots & k_{\mathrm{n_{en}n_{en}}}^{\omega_2u_1} & k_{\mathrm{n_{en}n_{en}}}^{\omega_2u_2} & k_{\mathrm{n_{en}n_{en}}}^{\omega_2u_3} & k_{\mathrm{n_{en}n_{en}}}^{\omega_2\mu} & k_{\mathrm{n_{en}n_{en}}}^{\omega_2\omega_1} & k_{\mathrm{n_{en}n_{en}}}^{\omega_2\omega_2}
\end{bmatrix},
$$

$$
\mathbf{R}^e = \left\{ R_1^{u_1} \quad R_1^{u_2} \quad R_1^{u_3} \quad R_1^{\mu} \quad R_1^{\omega_1} \quad R_1^{\omega_2} \quad \cdots \quad R_{\mathrm{n_{en}}}^{u_1} \quad R_{\mathrm{n_{en}}}^{u_2} \quad R_{\mathrm{n_{en}}}^{u_3} \quad R_{\mathrm{n_{en}}}^{\mu} \quad R_{\mathrm{n_{en}}}^{\omega_1} \quad R_{\mathrm{n_{en}}}^{\omega_2} \right\}^{\top}
$$

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

2.4.1 Tangent matrix, $\mathbf{k}_e^{\mathbf{uu}}$

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

$$R_a^{\mathbf{u}}(u_a^i, \mu^w) = -\int\limits_{\Omega_0^e} \frac{\partial N_a^{\mathbf{u}}}{\partial X_I} F_{iJ} S_{JI}\ dV + \int\limits_{\Omega_0^e} \rho_{\mathrm{R}} N_a^{\mathbf{u}} B_i\ dV + \int\limits_{\Gamma_T^e} N_a^{\mathbf{u}} T_i\ dS = 0, \tag{2.24}$$

where, $\mathbf{S} \equiv \mathbf{S}(\mathbf{C}, C^w)$ and $\mathcal{G}(\mathbf{C}, \mu^w, \omega^{\beta_1}, \omega^{\beta_2}; C^w, C^{\beta_1}, C^{\beta_2}, \psi) = \mathbf{0}$. Thus, 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\limits_{\Omega_0^e} \frac{\partial N_a^{\mathbf{u}}}{\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\limits_{\Omega_0^e} \frac{\partial N_a^{\mathbf{u}}}{\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\limits_{\Omega_0^e} \frac{\partial N_a^{\mathbf{u}}}{\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\limits_{\Omega_0^e} \frac{\partial N_a^{\mathbf{u}}}{\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\limits_{\Omega_0^e} \frac{\partial N_a^{\mathbf{u}}}{\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^{\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{2.25}$$

Here,

$$\mathbb{C}_{IJKL} = 2 \left(\frac{\partial S_{IJ}}{\partial C_{KL}} + \frac{\partial S_{IJ}}{\partial C^w} \frac{\partial C^w}{\partial C_{KL}} \right) \tag{2.26}$$

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 $k_{ab}^{u_i u_k}$.

$$\begin{aligned} \frac{\partial C_{KL}}{\partial F_{mN}} &= F_{mL}\delta_{KN} + F_{mK}\delta_{LN} = 2F_{mL}\delta_{KN} \quad (\text{since } C_{KL} = C_{LK}), \\ \text{and,} \quad F_{iJ} &= \delta_{ij} + \sum_{b=1}^{\mathrm{n_{en}}} \frac{\partial N_b^{\mathbf{u}}}{\partial X_J} u_b^i \qquad \Rightarrow \qquad \frac{\partial F_{iJ}}{\partial u_b^k} = \frac{\partial N_b^{\mathbf{u}}}{\partial X_J}\delta_{ik}. \end{aligned} \tag{2.27}$$

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

$$[\mathbf{k}_e^{\mathbf{uu}}]_{\mathrm{n_{en}}*\mathrm{n_{dim}}\times\mathrm{n_{en}}*\mathrm{n_{dim}}} = \int\limits_{\Omega_0^e} \Big[\mathbf{G}_{\mathbf{u}}^\top \Sigma_{\mathbf{S}} \mathbf{G}_{\mathbf{u}} + (\mathbf{B}_{\mathbf{u}}\mathbf{\Sigma}_{\mathbf{F}})^\top \mathbf{D}_{\mathbb{C}} (\mathbf{B}_{\mathbf{u}}\Sigma_{\mathbf{F}})\Big] \; dV \tag{2.28}$$

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 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,

$$\begin{aligned} k_{ab}^{u_i\mu} &= -\frac{\partial R_a^{u_i}}{\partial \mu_b^w}, \\ &= \int\limits_{\Omega_0^e} \frac{\partial N_a^{\mathbf{u}}}{\partial X_I} \left(F_{iJ} \frac{\partial S_{JI}}{\partial C^w} \frac{\partial C^w}{\partial \mu^w} \right) \frac{\partial \mu^w}{\partial \mu_b^w} \; dV, \\ &= \int\limits_{\Omega_0^e} \frac{\partial N_a^{\mathbf{u}}}{\partial X_I} \left(F_{iJ} \frac{\partial S_{JI}}{\partial C^w} \frac{\partial C^w}{\partial \mu^w} \right) N_b^\mu \; dV, \end{aligned} \tag{2.29}$$

Let define,

$$\mathbb{S}_{iI}^{\mu} = F_{iJ} \frac{\partial S_{JI}}{\partial C^w} \frac{\partial C^w}{\partial \mu^w}. \tag{2.30}$$

Thus, I can write,

$$k_{ab}^{u_i\mu} = \int\limits_{\Omega_0^e} \frac{\partial N_a^{\mathbf{u}}}{\partial X_I} \mathbb{S}_{iI}^{\mathbf{u}\mu} N_b^\mu \; dV. \tag{2.31}$$

In matrix form, it can be expressed as,

$$\left[\mathbf{k}_\mu^e\right]_{\mathrm{n_{en}}*\mathrm{n_{dim}}\times\mathrm{n_{en}}} = \int\limits_{\Omega_0^e} \mathbf{G}_{\mathbf{u}}^\top \mathbf{a}_{\mathbf{u}\mu} \mathbf{N}_\mu \; dV \tag{2.32}$$

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

$$\mathbb{S}^{\mu}_{iI} = F_{iJ}\frac{\partial S_{JI}}{\partial C^w}\frac{\partial C^w}{\partial \mu^w} \xrightarrow{\text{reshape to vector}} \mathbf{a}_{\mathbf{u}\mu}, \tag{2.33}$$

2.4.3 Tangent matrix, $\mathbf{k}^{\mathbf{u}\omega_1}_e$ *and* $\mathbf{k}^{\mathbf{u}\omega_2}_e$

The components of mechano-chemical element sub-matrix, $k^{u_i\omega_1}_{ab}$, can be written as,

$$\begin{aligned} k^{u_i\omega_1}_{ab} &= -\frac{\partial R^{u_i}_a}{\partial \omega^{\beta_1}_b}, \\ &= \int\limits_{\Omega^e_0} \frac{\partial N^{\mathbf{u}}_a}{\partial X_I}\left(F_{iJ}\frac{\partial S_{JI}}{\partial C^w}\frac{\partial C^w}{\partial \omega^{\beta_1}}\right)\frac{\partial \omega^{\beta}}{\partial \omega^{\beta_1}_b}\; dV, \\ &= \int\limits_{\Omega^e_0} \frac{\partial N^{\mathbf{u}}_a}{\partial X_I}\left(F_{iJ}\frac{\partial S_{JI}}{\partial C^w}\frac{\partial C^w}{\partial \omega^{\beta_1}}\right)N^{\omega}_b\; dV, \end{aligned} \tag{2.34}$$

Let define,

$$\mathbb{S}^{\omega_1}_{iI} = F_{iJ}\frac{\partial S_{JI}}{\partial C^w}\frac{\partial C^w}{\partial \omega^{\beta_1}}. \tag{2.35}$$

Thus, I can write,

$$k^{u_i\omega_1}_{ab} = \int\limits_{\Omega^e_0} \frac{\partial N^{\mathbf{u}}_a}{\partial X_I}\mathbb{S}^{\mathbf{u}\omega_1}_{iI}N^{\omega}_b\; dV. \tag{2.36}$$

In matrix form, it can be expressed as,

$$[\mathbf{k}^{\mathbf{u}\omega_1}_e]_{\mathrm{n_{en}}*\mathrm{n_{dim}}\times\mathrm{n_{en}}} = \int\limits_{\Omega^e_0} \mathbf{G}^{\top}_{\mathbf{u}}\mathbf{a}_{\mathbf{u}\omega_1}\mathbf{N}_{\omega}\; dV, \tag{2.37}$$

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

$$\mathbb{S}^{\beta_1}_{iI} = F_{iJ}\frac{\partial S_{JI}}{\partial C^w}\frac{\partial C^w}{\partial \omega^{\beta_1}} \xrightarrow{\text{reshape to vector}} \mathbf{a}_{\mathbf{u}\omega_1}, \tag{2.38}$$

The index form, $k^{u_i\omega_2}_{ab}$, and matrix form, $\mathbf{k}^{\mathbf{u}\omega_2}_e$, are as follows,

$$\begin{aligned} k^{u_i\omega_2}_{ab} &= \int\limits_{\Omega^e_0} \frac{\partial N^{\mathbf{u}}_a}{\partial X_I}\left(F_{iJ}\frac{\partial S_{JI}}{\partial C^w}\frac{\partial C^w}{\partial \omega^{\beta_1}}\right)N^{\omega}_b\; dV, \\ \Rightarrow [\mathbf{k}^{\mathbf{u}\omega_2}_e]_{\mathrm{n_{en}}*\mathrm{n_{dim}}\times\mathrm{n_{en}}} &= \int\limits_{\Omega^e_0} \mathbf{G}^{\top}_{\mathbf{u}}\mathbf{a}_{\mathbf{u}\omega_2}\mathbf{N}_{\omega}\; dV \end{aligned} \tag{2.39}$$

Here,

$$\mathbb{S}^{\beta_2}_{iI} = F_{iJ}\frac{\partial S_{JI}}{\partial C^w}\frac{\partial C^w}{\partial \omega^{\beta_2}} \xrightarrow{\text{reshape to vector}} \mathbf{a}_{\mathbf{u}\omega_2} \tag{2.40}$$

2.4.4 Tangent matrix, $\mathbf{k}_e^{\mu\mathbf{u}}$

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

$$R_a^\mu = -\int\limits_{\Omega_0^e} N_a^\mu \left(\frac{C^w - C_t^w}{\Delta t}\right)\, dV + \int\limits_{\Omega_0^e} \frac{\partial N_a^\mu}{\partial X_I} J_I^w\, dV + \int\limits_{\Gamma_{I^w}^e} N_a^\mu I^w\, dS \tag{2.41}$$

where, the following constitutive relations are valid,

$$\begin{aligned} \mathcal{G}(\mathbf{C}, \mu^w, \omega^{\beta_1}, \omega^{\beta_2}; C^w, C^{\beta_1}, C^{\beta_2}, \psi) = \mathbf{0}, \\ \mathbf{J}^w = \hat{\mathbf{J}}^w \left(\mathbf{C}, \operatorname{Grad}\mu^w, \operatorname{Grad}\omega^{\beta_1}, \operatorname{Grad}\omega^{\beta_2}; C^w, C^{\beta_1}, C^{\beta_2}\right). \end{aligned} \tag{2.42}$$

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

$$\begin{aligned} k_{ab}^{\mu u_k} &= -\frac{\partial R_a^\mu}{\partial u_b^k}, \\ &= \int\limits_{\Omega_0^e} \left[N_a^\mu \left(\frac{1}{\Delta t}\frac{\partial C^w}{\partial F_{mL}}\right) - \frac{\partial N_a^\mu}{\partial X_I}\left(\frac{\partial J_I^w}{\partial F_{mL}} + \frac{\partial J_I^w}{\partial C^w}\frac{\partial C^w}{\partial F_{mL}}\right)\right]\frac{\partial F_{mL}}{\partial u_b^k}\, dV, \\ &= \int\limits_{\Omega_0^e} \left[N_a^\mu \left(\frac{1}{\Delta t}\frac{\partial C^w}{\partial F_{mL}}\right) - \frac{\partial N_a^\mu}{\partial X_I}\left(\frac{\partial J_I^w}{\partial F_{mL}} + \frac{\partial J_I^w}{\partial C^w}\frac{\partial C^w}{\partial F_{mL}}\right)\right]\frac{\partial}{\partial u_b^k}\left(\delta_{mL} + \sum_{b=1}^{\mathrm{n_{en}}}\frac{\partial N_b^{\mathbf{u}}}{\partial X_J}u_b^m\right) dV, \\ &= \int\limits_{\Omega_0^e} \left[N_a^\mu \left(\frac{1}{\Delta t}\frac{\partial C^w}{\partial F_{mL}}\right) - \frac{\partial N_a^\mu}{\partial X_I}\left(\frac{\partial J_I^w}{\partial F_{mL}} + \frac{\partial J_I^w}{\partial C^w}\frac{\partial C^w}{\partial F_{mL}}\right)\right]\frac{\partial N_b^{\mathbf{u}}}{\partial X_L}\delta_{mk}\, dV, \\ &= \int\limits_{\Omega_0^e} N_a^\mu \left(\frac{1}{\Delta t}\frac{\partial C^w}{\partial F_{mL}}\right)\frac{\partial N_b^{\mathbf{u}}}{\partial X_L}\delta_{mk}\, dV - \int\limits_{\Omega_0^e} \frac{\partial N_a^\mu}{\partial X_I}\left(\frac{\partial J_I^w}{\partial F_{kL}} + \frac{\partial J_I^w}{\partial C^w}\frac{\partial C^w}{\partial F_{kL}}\right)\frac{\partial N_b^{\mathbf{u}}}{\partial X_L}\delta_{mk}\, dV, \\ &= \int\limits_{\Omega_0^e} N_a^\mu \left(\frac{1}{\Delta t}\frac{\partial C^w}{\partial F_{kL}}\right)\frac{\partial N_b^{\mathbf{u}}}{\partial X_L}\, dV - \int\limits_{\Omega_0^e} \frac{\partial N_a^\mu}{\partial X_I}\mathbb{J}_{IkL}^w\frac{\partial N_b^{\mathbf{u}}}{\partial X_L}\, dV. \end{aligned} \tag{2.43}$$

In matrix form, I can write,

$$[\mathbf{k}_e^{\mu\mathbf{u}}]_{\mathrm{n_{en}}\times\mathrm{n_{en}}*\mathrm{n_{dim}}} = \int\limits_{\Omega_0^e} \left(\mathbf{N}_\mu^\top \left(\frac{1}{\Delta t}\mathbf{c}_{\mu\mathbf{u}}\right)\mathbf{G}_\mathbf{u} - \mathbf{B}_\mu^\top \mathbf{D}_{\mu\mathbf{u}}\mathbf{G}_\mathbf{u}\right)\, dV \tag{2.44}$$

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

$$(\mathbb{J}^w)_{IkL} = \frac{\partial J_I^w}{\partial F_{kL}} + \frac{\partial J_I^w}{\partial C^w}\frac{\partial C^w}{\partial F_{kL}} \quad \xrightarrow{\text{reshape to matrix}} \quad \mathbf{D}_{\mu\mathbf{u}}. \tag{2.45}$$

2.4.5 Tangent matrix, $k_e^{\mu\mu}$

The tangent matrix, $k_{ab}^{\mu\mu}$, is given by,

$$
\begin{aligned}
k_{ab}^{\mu\mu} &= -\frac{\partial R_a^\mu}{\partial \mu_b^w}, \\
&= \int\limits_{\Omega_0^e} N_a^\mu \left(\frac{1}{\Delta t} \frac{\partial C^w}{\partial \mu^w} \right) \frac{\partial \mu^w}{\partial \mu_b^w} \, dV - \int\limits_{\Omega_0^e} \frac{\partial N_a^\mu}{\partial X_I} \frac{\partial J_I^w}{\partial C^w} \frac{\partial C^w}{\partial \mu^w} \frac{\partial \mu^w}{\partial \mu_b^w} \, dV \\
&\quad - \int\limits_{\Omega_0^e} \frac{\partial N_a^\mu}{\partial X_I} \frac{\partial J_I^w}{\partial (\operatorname{Grad} \mu^w)_J} \frac{\partial (\operatorname{Grad} \mu^w)_J}{\partial \mu^w} \, dV, \\
&= \int\limits_{\Omega_0^e} N_a^\mu \left(\frac{1}{\Delta t} \frac{\partial C^w}{\partial \mu^w} \right) N_b^\mu \, dV - \int\limits_{\Omega_0^e} \frac{\partial N_a^\mu}{\partial X_I} \left(\frac{\partial J_I^w}{\partial C^w} \frac{\partial C^w}{\partial \mu^w} \right) N_b^\mu \, dV \\
&\quad - \int\limits_{\Omega_0^e} \frac{\partial N_a^\mu}{\partial X_I} \left(\frac{\partial J_I^w}{\partial (\operatorname{Grad} \mu^w)_J} \right) \frac{\partial N_b^\mu}{\partial X_J} \, dV,
\end{aligned}
\tag{2.46}
$$

Let define,

$$
(\mathbf{m}_{\mu\mu})_I = \frac{\partial J_I^w}{\partial C^w} \frac{\partial C^w}{\partial \mu^w}. \tag{2.47}
$$

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

$$
[\mathbf{k}_e^{\mu\mu}]_{\mathrm{n_{en}} \times \mathrm{n_{en}}} = \int\limits_{\Omega_0^e} \left(\mathbf{N}_\mu^\top \left(\frac{1}{\Delta t} \frac{\partial C^w}{\partial \mu^w} \right) \mathbf{N}_\mu - \mathbf{B}_\mu^\top \mathbf{m}_{\mu\mu} \mathbf{N}_\mu + \mathbf{B}_\mu^\top \mathbf{M}^w \mathbf{B}_\mu \right) \, dV, \tag{2.48}
$$

2.4.6 Tangent matrix, $k_e^{\mu\omega_1}$ and $k_e^{\mu\omega_2}$

The tangent matrix, $k_{ab}^{\mu\omega_1}$, is given by,

$$
\begin{aligned}
k_{ab}^{\mu\omega_1} &= -\frac{\partial R_a^\mu}{\partial {\omega^{\beta_1}}^b}, \\
&= \int\limits_{\Omega_0^e} N_a^\mu \left(\frac{1}{\Delta t} \frac{\partial C^w}{\partial \omega^{\beta_1}} \right) \frac{\partial \omega^{\beta_1}}{\partial {\omega^{\beta_1}}^b} \, dV - \int\limits_{\Omega_0^e} \frac{\partial N_a^\mu}{\partial X_I} \frac{\partial J_I^w}{\partial C^w} \frac{\partial C^w}{\partial \omega^{\beta_1}} \frac{\partial \omega^{\beta_1}}{\partial {\omega^{\beta_1}}^b} \, dV \\
&\quad - \int\limits_{\Omega_0^e} \frac{\partial N_a^\mu}{\partial X_I} \frac{\partial J_I^w}{\partial (\operatorname{Grad} \omega^{\beta_1})_J} \frac{\partial (\operatorname{Grad} \omega^{\beta_1})_J}{\partial \omega^{\beta_1}} \, dV, \\
&= \int\limits_{\Omega_0^e} N_a^\mu \left(\frac{1}{\Delta t} \frac{\partial C^w}{\partial \omega^{\beta_1}} \right) N_b^\omega \, dV - \int\limits_{\Omega_0^e} \frac{\partial N_a^\mu}{\partial X_I} \left(\frac{\partial J_I^w}{\partial C^w} \frac{\partial C^w}{\partial \omega^{\beta_1}} \right) N_b^\omega \, dV \\
&\quad - \int\limits_{\Omega_0^e} \frac{\partial N_a^\mu}{\partial X_I} \left(\frac{\partial J_I^w}{\partial (\operatorname{Grad} \omega^{\beta_1})_J} \right) \frac{\partial N_b^\omega}{\partial X_J} \, dV
\end{aligned}
\tag{2.49}
$$

Let define,

$$(\mathbf{m}_{\mu\omega_1})_I = \frac{\partial J_I^w}{\partial C^w}\frac{\partial C^w}{\partial \omega^{\beta_1}}, \quad \text{and} \quad (\mathbf{M}^{w\beta_1})_{IJ} = -\frac{\partial J_I^w}{\partial (\operatorname{Grad}\omega^{\beta_1})_J} \tag{2.50}$$

Thus, the matrix form of the tangent can be written as,

$$[\mathbf{k}_e^{\mu\omega_1}]_{\mathrm{n_{en}}\times\mathrm{n_{en}}} = \int_{\Omega_0^e} \left(\mathbf{N}_\mu^\top \left(\frac{1}{\Delta t}\frac{\partial C^w}{\partial \omega^{\beta_1}}\right)\mathbf{N}_\omega - \mathbf{B}_\mu^\top \mathbf{m}_{\mu\omega_1}\mathbf{N}_\omega + \mathbf{B}_\mu^\top \mathbf{M}^{w\beta_1}\mathbf{B}_\omega\right)\, dV \tag{2.51}$$

The index form and matrix form of $\mathbf{k}_e^{\mu\omega_2}$ is as follows,

$$k_{ab}^{\mu\omega_2} = \int_{\Omega_0^e} N_a^\mu \left(\frac{1}{\Delta t}\frac{\partial C^w}{\partial \omega^{\beta_2}}\right) N_b^\omega \; dV - \int_{\Omega_0^e} \frac{\partial N_a^\mu}{\partial X_I}\left(\frac{\partial J_I^w}{\partial C^w}\frac{\partial C^w}{\partial \omega^{\beta_2}}\right) N_b^\omega \; dV, - \int_{\Omega_0^e} \frac{\partial N_a^\mu}{\partial X_I}\left(\frac{\partial J_I^w}{\partial (\operatorname{Grad}\omega^{\beta_2})_J}\right)\frac{\partial N_b^\omega}{\partial X_J}\; dV$$

$$\Rightarrow [\mathbf{k}_e^{\mu\omega_2}]_{\mathrm{n_{en}}\times\mathrm{n_{en}}} = \int_{\Omega_0^e} \left(\mathbf{N}_\mu^\top \left(\frac{1}{\Delta t}\frac{\partial C^w}{\partial \omega^{\beta_2}}\right)\mathbf{N}_\omega - \mathbf{B}_\mu^\top \mathbf{m}_{\mu\omega_2}\mathbf{N}_\omega + \mathbf{B}_\mu^\top \mathbf{M}^{w\beta_2}\mathbf{B}_\omega\right)\, dV, \tag{2.52}$$

where,

$$(\mathbf{m}_{\mu\omega_2})_I = \frac{\partial J_I^w}{\partial C^w}\frac{\partial C^w}{\partial \omega^{\beta_2}}, \quad \text{and} \quad (\mathbf{M}^{w\beta_2})_{IJ} = -\frac{\partial J_I^w}{\partial (\operatorname{Grad}\omega^{\beta_2})_J}. \tag{2.53}$$

2.4.7 Tangent matrix, $\mathbf{k}_e^{\omega_1\mathbf{u}}$ *and* $\mathbf{k}_e^{\omega_2\mathbf{u}}$

In index notation, the residual for the mass balance equation of the ionic species can be written as,

$$\begin{aligned} R_a^{\omega_1} &= -\int_{\Omega_0^e} N_a^\omega \left(\frac{C^{\beta_1} - C_t^{\beta_1}}{\Delta t}\right)\; dV + \int_{\Omega_0^e} \frac{\partial N_a^\omega}{\partial X_I} J_I^{\beta_1}\; dV + \int_{\Gamma_{I^{\beta_1}}^e} N_a^\omega I^{\beta_1}\; dS, \\ R_a^{\omega_2} &= -\int_{\Omega_0^e} N_a^\omega \left(\frac{C^{\beta_2} - C_t^{\beta_2}}{\Delta t}\right)\; dV + \int_{\Omega_0^e} \frac{\partial N_a^\omega}{\partial X_I} J_I^{\beta_2}\; dV + \int_{\Gamma_{I^{\beta_2}}^e} N_a^\omega I^{\beta_2}\; dS, \end{aligned} \tag{2.54}$$

where following constitutive relations are held

$$\begin{aligned} \mathcal{G}(\mathbf{C}, \mu^w, \omega^{\beta_1}, \omega^{\beta_2}, C^w, C^{\beta_1}, C^{\beta_2}, \psi) &= \mathbf{0}, \\ \mathbf{J}^{\beta_1} = \hat{\mathbf{J}}^{\beta_1}\left(\mathbf{C}, \operatorname{Grad}\mu^w, \operatorname{Grad}\omega^{\beta_1}, \operatorname{Grad}\omega^{\beta_2}; C^w, C^{\beta_1}, C^{\beta_2}\right)&, \\ \mathbf{J}^{\beta_2} = \hat{\mathbf{J}}^{\beta_2}\left(\mathbf{C}, \operatorname{Grad}\mu^w, \operatorname{Grad}\omega^{\beta_1}, \operatorname{Grad}\omega^{\beta_2}; C^w, C^{\beta_1}, C^{\beta_2}\right)&. \end{aligned} \tag{2.55}$$

Thus, the components of tangent matrix, $\mathbf{k}_e^{\omega_1 \mathbf{u}}$ can be computed as,

$$
\begin{aligned}
k_{ab}^{\omega_1 u_k} &= -\frac{\partial R_a^{\omega_1}}{\partial u_b^k}, \\
&= \int\limits_{\Omega_0^e} \left[N_a^{\omega} \left(\frac{1}{\Delta t} \frac{\partial C^{\beta_1}}{\partial F_{mL}} \right) - \frac{\partial N_a^{\omega}}{\partial X_I} \left(\frac{\partial J_I^{w}}{\partial F_{mL}} + \frac{\partial J_I^{\beta_1}}{\partial C^{\beta_1}} \frac{\partial C^{\beta_1}}{\partial F_{mL}} \right) \right] \frac{\partial F_{mL}}{\partial u_b^k} \, dV, \\
&= \int\limits_{\Omega_0^e} \left[N_a^{\omega} \left(\frac{1}{\Delta t} \frac{\partial C^{\beta_1}}{\partial F_{mL}} \right) - \frac{\partial N_a^{\omega}}{\partial X_I} \left(\frac{\partial J_I^{\beta_1}}{\partial F_{mL}} + \frac{\partial J_I^{\beta_1}}{\partial C^{\beta_1}} \frac{\partial C^{\beta_1}}{\partial F_{mL}} \right) \right] \frac{\partial}{\partial u_b^k} \left(\delta_{mL} + \sum_{b=1}^{\mathrm{n_{en}}} \frac{\partial N_b^{\mathbf{u}}}{\partial X_J} u_b^m \right) \, dV, \\
&= \int\limits_{\Omega_0^e} \left[N_a^{\omega} \left(\frac{1}{\Delta t} \frac{\partial C^{\beta_1}}{\partial F_{mL}} \right) - \frac{\partial N_a^{\omega}}{\partial X_I} \left(\frac{\partial J_I^{\beta_1}}{\partial F_{mL}} + \frac{\partial J_I^{\beta_1}}{\partial C^{\beta_1}} \frac{\partial C^{\beta_1}}{\partial F_{mL}} \right) \right] \frac{\partial N_b^{\mathbf{u}}}{\partial X_L} \delta_{mk} \, dV, \\
&= \int\limits_{\Omega_0^e} N_a^{\omega} \left(\frac{1}{\Delta t} \frac{\partial C^{\beta_1}}{\partial F_{mL}} \right) \frac{\partial N_b^{\mathbf{u}}}{\partial X_L} \delta_{mk} \, dV - \int\limits_{\Omega_0^e} \frac{\partial N_a^{\omega}}{\partial X_I} \left(\frac{\partial J_I^{\beta_1}}{\partial F_{kL}} + \frac{\partial J_I^{\beta_1}}{\partial C^{\beta_1}} \frac{\partial C^{\beta_1}}{\partial F_{kL}} \right) \frac{\partial N_b^{\mathbf{u}}}{\partial X_L} \delta_{mk} \, dV, \\
&= \int\limits_{\Omega_0^e} N_a^{\omega} \left(\frac{1}{\Delta t} \frac{\partial C^{\beta_1}}{\partial F_{kL}} \right) \frac{\partial N_b^{\mathbf{u}}}{\partial X_L} \, dV - \int\limits_{\Omega_0^e} \frac{\partial N_a^{\omega}}{\partial X_I} \mathbb{J}_{IkL}^{\beta_1} \frac{\partial N_b^{\mathbf{u}}}{\partial X_L} \, dV.
\end{aligned}
\tag{2.56}
$$

Thus, in matrix form, I can write,

$$
[\mathbf{k}_e^{\omega_1 \mathbf{u}}]_{\mathrm{n_{en}} \times \mathrm{n_{en}} * \mathrm{n_{dim}}} = \int\limits_{\Omega_0^e} \left(\mathbf{N}_{\omega}^{\top} \left(\frac{1}{\Delta t} \mathbf{c}_{\omega_1 \mathbf{u}} \right) \mathbf{G}_{\mathbf{u}} - \mathbf{B}_{\omega}^{\top} \mathbf{D}_{\omega_1 \mathbf{u}} \mathbf{G}_{\mathbf{u}} \right) \, dV \tag{2.57}
$$

where, $\frac{\partial C^{\beta_1}}{\partial \mathbf{F}}$ is reshaped into a vector of dimension $[\mathbf{c}_{\omega_1 \mathbf{u}}]_{1 \times \mathrm{n_{dim}^2}}$, and $[\mathbf{D}_{\omega_1 \mathbf{u}}]_{\mathrm{n_{dim}} \times \mathrm{n_{dim}^2}}$ is the matrix form of following third-order tangent,

$$
(\mathbb{J}^{\beta_1})_{IkL} = \frac{\partial J_I^{\beta_1}}{\partial F_{kL}} + \frac{\partial J_I^{\beta_1}}{\partial C^{\beta_1}} \frac{\partial C^{\beta_1}}{\partial F_{kL}} \quad \xrightarrow{\text{reshape to matrix}} \quad \mathbf{D}_{\omega_1 \mathbf{u}}. \tag{2.58}
$$

The index form, $k_{ab}^{\omega_2 u_k}$, and the corresponding matrix form, $\mathbf{k}_e^{\omega_1 \mathbf{u}}$, are given as,

$$
\begin{aligned}
k_{ab}^{\omega_2 u_k} &= \int\limits_{\Omega_0^e} N_a^{\omega} \left(\frac{1}{\Delta t} \frac{\partial C^{\beta_2}}{\partial F_{kL}} \right) \frac{\partial N_b^{\mathbf{u}}}{\partial X_L} \, dV - \int\limits_{\Omega_0^e} \frac{\partial N_a^{\omega}}{\partial X_I} \mathbb{J}_{IkL}^{\beta_2} \frac{\partial N_b^{\mathbf{u}}}{\partial X_L} \, dV, \\
[\mathbf{k}_e^{\omega_2 \mathbf{u}}]_{\mathrm{n_{en}} \times \mathrm{n_{en}} * \mathrm{n_{dim}}} &= \int\limits_{\Omega_0^e} \left(\mathbf{N}_{\omega}^{\top} \left(\frac{1}{\Delta t} \mathbf{c}_{\omega_2 \mathbf{u}} \right) \mathbf{G}_{\mathbf{u}} - \mathbf{B}_{\omega}^{\top} \mathbf{D}_{\omega_2 \mathbf{u}} \mathbf{G}_{\mathbf{u}} \right) \, dV,
\end{aligned}
\tag{2.59}
$$

where,

$$
\begin{aligned}
\frac{\partial C^{\beta_2}}{\partial \mathbf{F}} \quad &\xrightarrow{\text{reshape to vector}} \quad [\mathbf{c}_{\omega_2 \mathbf{u}}]_{1 \times \mathrm{n_{dim}^2}}, \\
(\mathbb{J}^{\beta_2})_{IkL} = \frac{\partial J_I^{\beta_2}}{\partial F_{kL}} + \frac{\partial J_I^{\beta_2}}{\partial C^{\beta_2}} \frac{\partial C^{\beta_2}}{\partial F_{kL}} \quad &\xrightarrow{\text{reshape to matrix}} \quad [\mathbf{D}_{\omega_2 \mathbf{u}}]_{\mathrm{n_{dim}} \times \mathrm{n_{dim}^2}}.
\end{aligned}
\tag{2.60}
$$

2.4.8 Tangent matrix, $k_e^{\omega_1\mu}$ and $k_e^{\omega_2\mu}$

Thus, the components of tangent matrix, $\mathbf{k}_e^{\omega_1\mu}$, can be computed as,

$$\begin{aligned} k_{ab}^{\omega_1\mu} &= -\frac{\partial R_a^{\omega_1}}{\partial \mu_b^w}, \\ &= \int\limits_{\Omega_0^e} N_a^\omega \left(\frac{1}{\Delta t}\frac{\partial C^{\beta_1}}{\partial \mu^w}\right)\frac{\partial \mu^w}{\partial \mu_b^w}\ dV - \int\limits_{\Omega_0^e} \frac{\partial N_a^\omega}{\partial X_I}\frac{\partial J_I^{\beta_1}}{\partial C^{\beta_1}}\frac{\partial C^{\beta_1}}{\partial \mu^w}\frac{\partial \mu^w}{\partial \mu_b^w}\ dV, \\ &= \int\limits_{\Omega_0^e} N_a^\omega \left(\frac{1}{\Delta t}\frac{\partial C^{\beta_1}}{\partial \mu^w}\right) N_b^\mu\ dV - \int\limits_{\Omega_0^e} \frac{\partial N_a^\omega}{\partial X_I}\left(\frac{\partial J_I^{\beta_1}}{\partial C^{\beta_1}}\frac{\partial C^{\beta_1}}{\partial \mu^w}\right) N_b^\mu\ dV \end{aligned} \tag{2.61}$$

Let define,

$$(\mathbf{m}_{\omega_1\mu})_I = \frac{\partial J_I^{\beta_1}}{\partial C^{\beta_1}}\frac{\partial C^{\beta_1}}{\partial \mu^w}. \tag{2.62}$$

Thus, the matrix form of the tangent can be written as,

$$[\mathbf{k}_e^{\omega_1\mu}]_{\mathrm{n_{en}}\times\mathrm{n_{en}}} = \int\limits_{\Omega_0^e} \left(\mathbf{N}_\omega^\top \left(\frac{1}{\Delta t}\frac{\partial C^{\beta_1}}{\partial \mu^w}\right)\mathbf{N}_\mu - \mathbf{B}_\omega^\top \mathbf{m}_{\omega_1\mu}\mathbf{N}_\mu\right)\ dV. \tag{2.63}$$

Similarly, the index form of $k_{ab}^{\omega_2\mu}$ and corresponding matrix form $\mathbf{k}_e^{\omega_2\mu}$ can be written as,

$$\begin{aligned} k_{ab}^{\omega_2\mu} &= \int\limits_{\Omega_0^e} N_a^\omega \left(\frac{1}{\Delta t}\frac{\partial C^{\beta_2}}{\partial \mu^w}\right) N_b^\mu\ dV - \int\limits_{\Omega_0^e} \frac{\partial N_a^\omega}{\partial X_I}\left(\frac{\partial J_I^{\beta_2}}{\partial C^{\beta_2}}\frac{\partial C^{\beta_2}}{\partial \mu^w}\right) N_b^\mu\ dV, \\ [\mathbf{k}_e^{\omega_1\mu}]_{\mathrm{n_{en}}\times\mathrm{n_{en}}} &= \int\limits_{\Omega_0^e} \left(\mathbf{N}_\omega^\top \left(\frac{1}{\Delta t}\frac{\partial C^{\beta_2}}{\partial \mu^w}\right)\mathbf{N}_\mu - \mathbf{B}_\omega^\top \mathbf{m}_{\omega_2\mu}\mathbf{N}_\mu\right)\ dV. \end{aligned} \tag{2.64}$$

where,

$$(\mathbf{m}_{\omega_2\mu})_I = \frac{\partial J_I^{\beta_2}}{\partial C^{\beta_2}}\frac{\partial C^{\beta_2}}{\partial \mu^w}. \tag{2.65}$$

2.4.9 Tangent matrix, $k_e^{\omega_1\omega_1}$ and $k_e^{\omega_2\omega_2}$

Thus, the components of tangent matrix, $\mathbf{k}_e^{\omega_1\omega_1}$ can be computed as,

$$
\begin{aligned}
k_{ab}^{\omega_1\omega_1} &= -\frac{\partial R_a^{\omega_1}}{\partial \omega_b^{\beta_1}}, \\
&= \int\limits_{\Omega_0^e} N_a^\omega \left(\frac{1}{\Delta t} \frac{\partial C^{\beta_1}}{\partial \omega^{\beta_1}} \right) \frac{\partial \omega^{\beta_1}}{\partial \omega_b^{\beta_1}} \ dV - \int\limits_{\Omega_0^e} \frac{\partial N_a^\omega}{\partial X_I} \frac{\partial J_I^{\beta_1}}{\partial C^{\beta_1}} \frac{\partial C^{\beta_1}}{\partial \omega^{\beta_1}} \frac{\partial \omega^{\beta_1}}{\partial \omega_b^{\beta_1}} \ dV \\
&\quad - \int\limits_{\Omega_0^e} \frac{\partial N_a^\omega}{\partial X_I} \frac{\partial J_I^{\beta_1}}{\partial (\operatorname{Grad} \omega^{\beta_1})} \frac{\partial (\operatorname{Grad} \omega^{\beta_1})_J}{\partial \omega^{\beta_1}} \ dV, \\
&= \int\limits_{\Omega_0^e} N_a^\omega \left(\frac{1}{\Delta t} \frac{\partial C^{\beta_1}}{\partial \omega^{\beta_1}} \right) N_b^\omega \ dV - \int\limits_{\Omega_0^e} \frac{\partial N_a^\omega}{\partial X_I} \left(\frac{\partial J_I^{\beta_1}}{\partial C^{\beta_1}} \frac{\partial C^{\beta_1}}{\partial \omega^{\beta_1}} \right) N_b^\omega \ dV \\
&\quad - \int\limits_{\Omega_0^e} \frac{\partial N_a^\omega}{\partial X_I} \left(\frac{\partial J_I^{\beta_1}}{\partial (\operatorname{Grad} \omega^{\beta_1})_J} \right) \frac{\partial N_b^\omega}{\partial X_J} \ dV,
\end{aligned}
\tag{2.66}
$$

Let define,

$$
(\mathbf{m}_{\omega_1\omega_1})_I = \frac{\partial J_I^{\beta_1}}{\partial C^{\beta_1}} \frac{\partial C^{\beta_1}}{\partial \omega^{\beta_1}}, \quad \text{and} \quad (\mathbf{M}^{\beta_1\beta_1})_{IJ} = -\frac{\partial J_I^{\beta_1\beta_1}}{\partial (\operatorname{Grad} \omega^{\beta_1})_J}
\tag{2.67}
$$

Additionally, by recognizing $\frac{\partial J_I^{\beta_1}}{\partial (\operatorname{Grad} \omega^{\beta_1})_J} = -M_{IJ}^{\beta_1}$ is the mobility tensor, the matrix form of the tangent can be written as,

$$
[\mathbf{k}_e^{\omega_1\omega_1}]_{\mathrm{n_{en}} \times \mathrm{n_{en}}} = \int\limits_{\Omega_0^e} \left(\mathbf{N}_\omega^\top \left(\frac{1}{\Delta t} \frac{\partial C^{\beta_1}}{\partial \omega^{\beta_1}} \right) \mathbf{N}_\omega - \mathbf{B}_\omega^\top \mathbf{m}_{\omega_1\omega_1} \mathbf{N}_\omega + \mathbf{B}_\omega^\top \mathbf{M}^{\beta_1} \mathbf{B}_\omega \right) \ dV,
\tag{2.68}
$$

Similarly, $k_{ab}^{\omega_2\omega_2}$ can be written as,

$$
\begin{aligned}
k_{ab}^{\omega_2\omega_2} &= -\frac{\partial R_a^{\omega_2}}{\partial \omega_b^{\beta_2}}, \\
&= \int\limits_{\Omega_0^e} N_a^\omega \left(\frac{1}{\Delta t} \frac{\partial C^{\beta_2}}{\partial \omega^{\beta_2}} \right) N_b^\omega \ dV - \int\limits_{\Omega_0^e} \frac{\partial N_a^\omega}{\partial X_I} \left(\frac{\partial J_I^{\beta_2}}{\partial C^{\beta_2}} \frac{\partial C^{\beta_2}}{\partial \omega^{\beta_2}} \right) N_b^\omega \ dV \\
&\quad - \int\limits_{\Omega_0^e} \frac{\partial N_a^\omega}{\partial X_I} \left(\frac{\partial J_I^{\beta_2}}{\partial (\operatorname{Grad} \omega^{\beta_2})_J} \right) \frac{\partial N_b^\omega}{\partial X_J} \ dV.
\end{aligned}
\tag{2.69}
$$

The corresponding matrix form, $\mathbf{k}_e^{\omega_2\omega_2}$, is given by,

$$
[\mathbf{k}_e^{\omega_2\omega_2}]_{\mathrm{n_{en}} \times \mathrm{n_{en}}} = \int\limits_{\Omega_0^e} \left(\mathbf{N}_\omega^\top \left(\frac{1}{\Delta t} \frac{\partial C^{\beta_2}}{\partial \omega^{\beta_2}} \right) \mathbf{N}_\omega - \mathbf{B}_\omega^\top \mathbf{m}_{\omega_2\omega_2} \mathbf{N}_\omega + \mathbf{B}_\omega^\top \mathbf{M}^{\beta_2\beta_2} \mathbf{B}_\omega \right) \ dV,
\tag{2.70}
$$

where,

$$(\mathbf{m}_{\omega_2\omega_2})_I = \frac{\partial J_I^{\beta_2}}{\partial C^{\beta_2}} \frac{\partial C^{\beta_2}}{\partial \omega^{\beta_2}}, \quad \text{and} \quad (\mathbf{M}^{\beta_2\beta_2})_{IJ} = -\frac{\partial J_I^{\beta_2}}{\partial (\operatorname{Grad} \omega^{\beta_2})_J} \tag{2.71}$$

2.4.10 Tangent matrix, $k_e^{\omega_1\omega_2}$ and $k_e^{\omega_2\omega_1}$

Following the same procedure, $k_e^{\omega_1\omega_2}$ can be written in index notation as follows,

$$\begin{aligned} k_{ab}^{\omega_1\omega_2} &= -\frac{\partial R_a^{\omega_2}}{\partial \omega_b^{\beta_2}}, \\ &= \int_{\Omega_0^e} N_a^\omega \left(\frac{1}{\Delta t}\frac{\partial C^{\beta_1}}{\partial \omega^{\beta_2}}\right) N_b^\omega \; dV - \int_{\Omega_0^e} \frac{\partial N_a^\omega}{\partial X_I}\left(\frac{\partial J_I^{\beta_2}}{\partial \omega^{\beta_2}}\right) N_b^\omega \; dV \\ &\quad - \int_{\Omega_0^e} \frac{\partial N_a^\omega}{\partial X_I}\left(\frac{\partial J_I^{\beta_2}}{\partial (\operatorname{Grad} \omega^{\beta_2})_J}\right)\frac{\partial N_b^\omega}{\partial X_J} \; dV \end{aligned} \tag{2.72}$$

Accordingly, the matrix form, $\mathbf{K}_e^{\omega_1\omega_2}$, is given as,

$$[\mathbf{K}_e^{\omega_1\omega_2}]_{\mathrm{n_{en}}\times\mathrm{n_{en}}} = \int_{\Omega_0^e} \left(\mathbf{N}_\omega^\top \left(\frac{1}{\Delta t}\frac{\partial C^{\beta_1}}{\partial \omega^{\beta_2}}\right)\mathbf{N}_\omega - \mathbf{B}_\omega^\top \mathbf{m}_{\omega_1\omega_2}\mathbf{N}_\omega + \mathbf{B}_\omega^\top \mathbf{M}^{\beta_1\beta_2}\mathbf{B}_\omega\right) dV, \tag{2.73}$$

where,

$$(\mathbf{m}_{\omega_1\omega_2})_I = \frac{\partial J_I^{\beta_1}}{\partial C^{\beta_2}} \frac{\partial C^{\beta_2}}{\partial \omega^{\beta_2}} \quad \text{and} \quad (\mathbf{M}^{\beta_1\beta_2})_{IJ} = -\frac{\partial J_I^{\beta_1}}{\partial (\operatorname{Grad} \omega^{\beta_2})_J} \tag{2.74}$$

Finally, using index notation $k_e^{\omega_2\omega_1}$ can be expressed as,

$$\begin{aligned} k_{ab}^{\omega_2\omega_1} &= -\frac{\partial R_a^{\omega_2}}{\partial \omega_b^{\beta_1}}, \\ &= \int_{\Omega_0^e} N_a^\omega \left(\frac{1}{\Delta t}\frac{\partial C^{\beta_2}}{\partial \omega^{\beta_1}}\right) N_b^\omega \; dV - \int_{\Omega_0^e} \frac{\partial N_a^\omega}{\partial X_I}\left(\frac{\partial J_I^{\beta_2}}{\partial \omega^{\beta_1}}\right) N_b^\omega \; dV - \int_{\Omega_0^e} \frac{\partial N_a^\omega}{\partial X_I}\left(\frac{\partial J_I^{\beta_2}}{\partial (\operatorname{Grad} \omega^{\beta_1})_J}\right)\frac{\partial N_b^\omega}{\partial X_J} \; dV. \end{aligned} \tag{2.75}$$

The corresponding matrix form, $\mathbf{K}_e^{\omega_2\omega_1}$, is given by,

$$[\mathbf{K}_e^{\omega_2\omega_1}]_{\mathrm{n_{en}}\times\mathrm{n_{en}}} = \int_{\Omega_0^e} \left(\mathbf{N}_\omega^\top \left(\frac{1}{\Delta t}\frac{\partial C^{\beta_2}}{\partial \omega^{\beta_1}}\right)\mathbf{N}_\omega - \mathbf{B}_\omega^\top \mathbf{m}_{\omega_2\omega_1}\mathbf{N}_\omega + \mathbf{B}_\omega^\top \mathbf{M}^{\beta_2\beta_1}\mathbf{B}_\omega\right) dV, \tag{2.76}$$

where, where,

$$(\mathbf{m}_{\omega_2\omega_1})_I = \frac{\partial J_I^{\beta_2}}{\partial C^{\beta_1}} \frac{\partial C^{\beta_1}}{\partial \omega^{\beta_1}} \quad \text{and} \quad (\mathbf{M}^{\beta_2\beta_1})_{IJ} = -\frac{\partial J_I^{\beta_2}}{\partial (\operatorname{Grad} \omega^{\beta_1})_J} \tag{2.77}$$

2.4.11 *Matrix form of element residual vectors*

$$
\begin{aligned}
[\mathbf{R}^{\mathbf{u}}_e]_{\mathrm{n_{en}*n_{dim}}\times 1} &= -\int\limits_{\Omega_0^e} (\mathbf{B_u}\Sigma_{\mathbf{F}})^\top \mathbf{S}\ dV + \int\limits_{\Omega_0^e} \rho_{\mathrm{R}} \mathbf{N}_{\mathbf{u}}^\top \mathbf{B}\ dV + \int\limits_{\Gamma^e_{\mathbf{T}}} \mathbf{N}_{\mathbf{u}}^\top \mathbf{T}\ dS,\\
[\mathbf{R}^{\mu}_e]_{\mathrm{n_{en}}\times 1} &= -\int\limits_{\Omega_0^e} \mathbf{N}_{\mu}^\top \left(\frac{C^w - C^w_t}{\Delta t}\right)\ dV + \int\limits_{\Omega_0^e} \mathbf{B}_{\mu}^\top \mathbf{J}^w\ dV + \int\limits_{\Gamma^e_{I^w}} \mathbf{N}_{\mu}^\top I^w\ dS,\\
[\mathbf{R}^{\omega_1}_e]_{\mathrm{n_{en}}\times 1} &= -\int\limits_{\Omega_0^e} \mathbf{N}_{\omega}^\top \left(\frac{C^{\beta_1} - C^{\beta_1}_t}{\Delta t}\right)\ dV + \int\limits_{\Omega_0^e} \mathbf{B}_{\omega}^\top \mathbf{J}^{\beta_1}\ dV + \int\limits_{\Gamma^e_{I^{\beta_1}}} \mathbf{N}_{\omega}^\top I^{\beta_1}\ dS,\\
[\mathbf{R}^{\omega_2}_e]_{\mathrm{n_{en}}\times 1} &= -\int\limits_{\Omega_0^e} \mathbf{N}_{\omega}^\top \left(\frac{C^{\beta_2} - C^{\beta_2}_t}{\Delta t}\right)\ dV + \int\limits_{\Omega_0^e} \mathbf{B}_{\omega}^\top \mathbf{J}^{\beta_2}\ dV + \int\limits_{\Gamma^e_{I^{\beta_2}}} \mathbf{N}_{\omega}^\top I^{\beta_2}\ dS.
\end{aligned} \tag{2.78}
$$

2.5 Summary of the element tangent matrix and residual vector components

$$
\begin{aligned}
R^{u_i}_a &= -\int\limits_{\Omega_0^e} \frac{\partial N^{\mathbf{u}}_a}{\partial X_I} F_{iJ} S_{JI}\ dV + \int\limits_{\Omega_0^e} \rho_{\mathrm{R}} N^{\mathbf{u}}_a B_i\ dV + \int\limits_{\Gamma^e_{\mathbf{T}}} N^{\mathbf{u}}_a T_i\ dS,\\
R^{\mu}_a &= -\int\limits_{\Omega_0^e} N^{\mu}_a \left(\frac{C^w - C^w_t}{\Delta t}\right)\ dV + \int\limits_{\Omega_0^e} \frac{\partial N^{\mu}_a}{\partial X_I} J^w_I\ dV + \int\limits_{\Gamma^e_{I^w}} N^{\mu}_a I^w\ dS,\\
R^{\omega_1}_a &= -\int\limits_{\Omega_0^e} N^{\omega}_a \left(\frac{C^{\beta_1} - C^{\beta_1}_t}{\Delta t}\right)\ dV + \int\limits_{\Omega_0^e} \frac{\partial N^{\omega}_a}{\partial X_I} J^{\beta_1}_I\ dV + \int\limits_{\Gamma^e_{I^{\beta_1}}} N^{\omega}_a I^{\beta_1}\ dS,\\
R^{\omega_2}_a &= -\int\limits_{\Omega_0^e} N^{\omega}_a \left(\frac{C^{\beta_2} - C^{\beta_2}_t}{\Delta t}\right)\ dV + \int\limits_{\Omega_0^e} \frac{\partial N^{\omega}_a}{\partial X_I} J^{\beta_2}_I\ dV + \int\limits_{\Gamma^e_{I^{\beta_2}}} N^{\omega}_a I^{\beta_2}\ dS.
\end{aligned} \tag{2.79}
$$

In index notation, the element tangent matrix can be expressed as follows,

$$
\begin{aligned}
k^{u_i u_k}_{ab} &= -\frac{\partial R^{u_i}_a}{\partial u^k_b} &&= \int\limits_{\Omega_0^e} \frac{\partial N^{\mathbf{u}}_a}{\partial X_J} S_{JL} \delta_{ik} \frac{\partial N^{\vec{u}}_b}{\partial X_L}\ dV + \int\limits_{\Omega_0^e} \frac{\partial N^{\mathbf{u}}_a}{\partial X_J} F_{iI} \left(2\frac{dS_{IJ}}{dC_{KL}}\right) F_{kK} \frac{\partial N^{\vec{u}}_b}{\partial X_L}\ dV,\\
k^{u_i \mu}_{ab} &= -\frac{\partial R^{u_i}_a}{\partial \mu^w_b} &&= \int\limits_{\Omega_0^e} \frac{\partial N^{\mathbf{u}}_a}{\partial X_I} \left(F_{iJ}\frac{dS_{IJ}}{d\mu^w}\right) N^{\mu}_b\ dV,\\
k^{u_i \omega_1}_{ab} &= -\frac{\partial R^{u_i}_a}{\partial \omega^{\beta_1}_b} &&= \int\limits_{\Omega_0^e} \frac{\partial N^{\mathbf{u}}_a}{\partial X_I} \left(F_{iJ}\frac{dS_{IJ}}{d\omega^{\beta_1}}\right) N^{\omega}_b\ dV,\\
k^{u_i \omega_2}_{ab} &= -\frac{\partial R^{u_i}_a}{\partial \omega^{\beta_2}_b} &&= \int\limits_{\Omega_0^e} \frac{\partial N^{\mathbf{u}}_a}{\partial X_I} \left(F_{iJ}\frac{dS_{IJ}}{d\omega^{\beta_2}}\right) N^{\omega}_b\ dV,\\
k^{\mu u_k}_{ab} &= -\frac{\partial R^{\mu}_a}{\partial u^k_b} &&= \int\limits_{\Omega_0^e} N^{\mu}_a \left(\frac{1}{\Delta t}\frac{\partial C^w}{\partial F_{kL}}\right) \frac{\partial N^{\vec{u}}_b}{\partial X_L}\ dV - \int\limits_{\Omega_0^e} \frac{\partial N^{\mu}_a}{\partial X_I} \left(\frac{dJ^w_I}{dF_{kL}}\right) \frac{\partial N^{\vec{u}}_b}{\partial X_L}\ dV,
\end{aligned}
$$

$$
\begin{aligned}
k_{ab}^{\mu\mu} &= -\frac{\partial R_a^\mu}{\partial \mu_b^w} &&= \int\limits_{\Omega_0^e} N_a^\mu \left(\frac{1}{\Delta t}\frac{\partial C^w}{\partial \mu^w}\right) N_b^\mu \, dV - \int\limits_{\Omega_0^e} \frac{\partial N_a^\mu}{\partial X_I} \left(\frac{\partial J_I^w}{\partial \mu^w}\right) N_b^\mu \, dV \\
& && \quad - \int\limits_{\Omega_0^e} \frac{\partial N_a^\mu}{\partial X_I} \left(\frac{\partial J_I^w}{\partial (\operatorname{Grad}\mu^w)_J}\right) \frac{\partial N_b^\mu}{\partial X_J} \, dV, \\
k_{ab}^{\mu\omega_1} &= -\frac{\partial R_a^\mu}{\partial \omega_b^{\beta_1}} &&= \int\limits_{\Omega_0^e} N_a^\mu \left(\frac{1}{\Delta t}\frac{\partial C^w}{\partial \omega^{\beta_1}}\right) N_b^\omega \, dV - \int\limits_{\Omega_0^e} \frac{\partial N_a^\mu}{\partial X_I} \left(\frac{\partial J_I^w}{\partial \omega^{\beta_1}}\right) N_b^\omega \, dV, \\
& && \quad - \int\limits_{\Omega_0^e} \frac{\partial N_a^\mu}{\partial X_I} \left(\frac{\partial J_I^w}{\partial (\operatorname{Grad}\omega^{\beta_1})_J}\right) \frac{\partial N_b^\omega}{\partial X_J} \, dV, \\
k_{ab}^{\mu\omega_2} &= -\frac{\partial R_a^\mu}{\partial \omega_b^{\beta_2}} &&= \int\limits_{\Omega_0^e} N_a^\mu \left(\frac{1}{\Delta t}\frac{\partial C^w}{\partial \omega^{\beta_2}}\right) N_b^\omega \, dV - \int\limits_{\Omega_0^e} \frac{\partial N_a^\mu}{\partial X_I} \left(\frac{\partial J_I^w}{\partial \omega^{\beta_2}}\right) N_b^\omega \, dV, \\
& && \quad - \int\limits_{\Omega_0^e} \frac{\partial N_a^\mu}{\partial X_I} \left(\frac{\partial J_I^w}{\partial (\operatorname{Grad}\omega^{\beta_2})_J}\right) \frac{\partial N_b^\omega}{\partial X_J} \, dV, \\
k_{ab}^{\omega_1 u_k} &= -\frac{\partial R_a^{\omega_1}}{\partial u_b^k} &&= \int\limits_{\Omega_0^e} N_a^\omega \left(\frac{1}{\Delta t}\frac{\partial C^{\beta_1}}{\partial F_{kL}}\right) \frac{\partial N_b^{\vec{u}}}{\partial X_L} \, dV - \int\limits_{\Omega_0^e} \frac{\partial N_a^\omega}{\partial X_I} \left(\frac{d J_I^{\beta_1}}{d F_{kL}}\right) \frac{\partial N_b^{\vec{u}}}{\partial X_L} \, dV, \\
k_{ab}^{\omega_1 \mu} &= -\frac{\partial R_a^{\omega_1}}{\partial \mu_b^w} &&= \int\limits_{\Omega_0^e} N_a^\omega \left(\frac{1}{\Delta t}\frac{\partial C^{\beta_1}}{\partial \mu^w}\right) N_b^\mu \, dV - \int\limits_{\Omega_0^e} \frac{\partial N_a^\omega}{\partial X_I} \left(\frac{\partial J_I^{\beta_1}}{\partial \mu^w}\right) N_b^\mu \, dV, \qquad (2.80) \\
k_{ab}^{\omega_1\omega_1} &= -\frac{\partial R_a^{\omega_1}}{\partial \omega_b^{\beta_1}} &&= \int\limits_{\Omega_0^e} N_a^\omega \left(\frac{1}{\Delta t}\frac{\partial C^{\beta_1}}{\partial \omega^{\beta_1}}\right) N_b^\omega \, dV - \int\limits_{\Omega_0^e} \frac{\partial N_a^\omega}{\partial X_I} \left(\frac{\partial J_I^{\beta_1}}{\partial \omega^{\beta_1}}\right) N_b^\omega \, dV \\
& && \quad - \int\limits_{\Omega_0^e} \frac{\partial N_a^\omega}{\partial X_I} \left(\frac{\partial J_I^{\beta_1}}{\partial (\operatorname{Grad}\omega^{\beta_1})_J}\right) \frac{\partial N_b^\omega}{\partial X_J} \, dV, \\
k_{ab}^{\omega_1\omega_2} &= -\frac{\partial R_a^{\omega_1}}{\partial \omega_b^{\beta_2}} &&= \int\limits_{\Omega_0^e} N_a^\omega \left(\frac{1}{\Delta t}\frac{\partial C^{\beta_1}}{\partial \omega^{\beta_2}}\right) N_b^\omega \, dV - \int\limits_{\Omega_0^e} \frac{\partial N_a^\omega}{\partial X_I} \left(\frac{\partial J_I^{\beta_1}}{\partial \omega^{\beta_2}}\right) N_b^\omega \, dV \\
& && \quad - \int\limits_{\Omega_0^e} \frac{\partial N_a^\omega}{\partial X_I} \left(\frac{\partial J_I^{\beta_1}}{\partial (\operatorname{Grad}\omega^{\beta_2})_J}\right) \frac{\partial N_b^\omega}{\partial X_J} \, dV, \\
k_{ab}^{\omega_2 u_k} &= -\frac{\partial R_a^{\omega_2}}{\partial u_b^k} &&= \int\limits_{\Omega_0^e} N_a^\omega \left(\frac{1}{\Delta t}\frac{\partial C^{\beta_2}}{\partial F_{kL}}\right) \frac{\partial N_b^{\vec{u}}}{\partial X_L} \, dV - \int\limits_{\Omega_0^e} \frac{\partial N_a^\omega}{\partial X_I} \left(\frac{d J_I^{\beta_2}}{d F_{kL}}\right) \frac{\partial N_b^{\vec{u}}}{\partial X_L} \, dV, \\
k_{ab}^{\omega_2 \mu} &= -\frac{\partial R_a^{\omega_2}}{\partial \mu_b^w} &&= \int\limits_{\Omega_0^e} N_a^\omega \left(\frac{1}{\Delta t}\frac{\partial C^{\beta_2}}{\partial \mu^w}\right) N_b^\mu \, dV - \int\limits_{\Omega_0^e} \frac{\partial N_a^\omega}{\partial X_I} \left(\frac{\partial J_I^{\beta_2}}{\partial \mu^w}\right) N_b^\mu \, dV, \\
k_{ab}^{\omega_2\omega_1} &= -\frac{\partial R_a^{\omega_2}}{\partial \omega_b^{\beta_1}} &&= \int\limits_{\Omega_0^e} N_a^\omega \left(\frac{1}{\Delta t}\frac{\partial C^{\beta_2}}{\partial \omega^{\beta_1}}\right) N_b^\omega \, dV - \int\limits_{\Omega_0^e} \frac{\partial N_a^\omega}{\partial X_I} \left(\frac{\partial J_I^{\beta_2}}{\partial \omega^{\beta_1}}\right) N_b^\omega \, dV
\end{aligned}
$$

$$
\begin{aligned}
& \qquad - \int\limits_{\Omega_0^e} \frac{\partial N_a^\omega}{\partial X_I} \left(\frac{\partial J_I^{\beta_2}}{\partial (\operatorname{Grad} \omega^{\beta_1})_J} \right) \frac{\partial N_b^\omega}{\partial X_J} \; dV, \\
k_{ab}^{\omega_2 \omega_2} &= - \frac{\partial R_a^{\omega_2}}{\partial \omega_b^{\beta_2}} = \int\limits_{\Omega_0^e} N_a^\omega \left(\frac{1}{\Delta t} \frac{\partial C^{\beta_2}}{\partial \omega^{\beta_2}} \right) N_b^\omega \; dV - \int\limits_{\Omega_0^e} \frac{\partial N_a^\omega}{\partial X_I} \left(\frac{\partial J_I^{\beta_2}}{\partial \omega^{\beta_2}} \right) N_b^\omega \; dV \\
& \qquad - \int\limits_{\Omega_0^e} \frac{\partial N_a^\omega}{\partial X_I} \left(\frac{\partial J_I^{\beta_2}}{\partial (\operatorname{Grad} \omega^{\beta_2})_J} \right) \frac{\partial N_b^\omega}{\partial X_J} \; dV.
\end{aligned}
$$

In the matrix form, the residual sub-vectors are expressed as,

$$
\begin{aligned}
[\mathbf{R}_e^{\mathbf{u}}]_{\mathrm{n_{en}} * \mathrm{n_{dim}} \times 1} &= - \int\limits_{\Omega_0^e} (\mathbf{B_u} \Sigma_{\mathbf{F}})^\top \mathbf{S} \; dV + \int\limits_{\Omega_0^e} \rho_{\mathrm{R}} \mathbf{N}_{\mathbf{u}}^\top \mathbf{B} \; dV + \int\limits_{\Gamma_{\mathbf{T}}^e} \mathbf{N}_{\mathbf{u}}^\top \mathbf{T} \; dS, \\
[\mathbf{R}_e^{\mu}]_{\mathrm{n_{en}} \times 1} &= - \int\limits_{\Omega_0^e} \mathbf{N}_\mu^\top \left(\frac{C^w - C_t^w}{\Delta t} \right) \; dV + \int\limits_{\Omega_0^e} \mathbf{B}_\mu^\top \mathbf{J}^w \; dV + \int\limits_{\Gamma_{I^w}^e} \mathbf{N}_\mu^\top I^w \; dS, \\
[\mathbf{R}_e^{\omega_1}]_{\mathrm{n_{en}} \times 1} &= - \int\limits_{\Omega_0^e} \mathbf{N}_\omega^\top \left(\frac{C^{\beta_1} - C_t^{\beta_1}}{\Delta t} \right) \; dV + \int\limits_{\Omega_0^e} \mathbf{B}_\omega^\top \mathbf{J}^{\beta_1} \; dV + \int\limits_{\Gamma_{I^{\beta_1}}^e} \mathbf{N}_\omega^\top I^{\beta_1} \; dS, \\
[\mathbf{R}_e^{\omega_2}]_{\mathrm{n_{en}} \times 1} &= - \int\limits_{\Omega_0^e} \mathbf{N}_\omega^\top \left(\frac{C^{\beta_2} - C_t^{\beta_2}}{\Delta t} \right) \; dV + \int\limits_{\Omega_0^e} \mathbf{B}_\omega^\top \mathbf{J}^{\beta_2} \; dV + \int\limits_{\Gamma_{I^{\beta_2}}^e} \mathbf{N}_\omega^\top I^{\beta_2} \; dS.
\end{aligned}
\tag{2.81}
$$

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

$$
\begin{aligned}
[\mathbf{K}_e^{\mathbf{uu}}]_{\mathrm{n_{en}} * \mathrm{n_{dim}} \times \mathrm{n_{en}} * \mathrm{n_{dim}}} &= \int\limits_{\Omega_0^e} \left(\mathbf{G}_{\mathbf{u}}^\top \Sigma_{\mathbf{S}} \mathbf{G}_{\mathbf{u}} + (\mathbf{B_u} \Sigma_{\mathbf{F}})^\top \mathbf{D}_{\mathbb{C}} (\mathbf{B_u} \Sigma_{\mathbf{F}}) \right) dV, \\
[\mathbf{K}_e^{\mathbf{u}\mu}]_{\mathrm{n_{en}} * \mathrm{n_{dim}} \times \mathrm{n_{en}}} &= \int\limits_{\Omega_0^e} \mathbf{G}_{\mathbf{u}}^\top \mathbf{a}_{\mathbf{u}\mu} \mathbf{N}_\mu \; dV, \\
[\mathbf{K}_e^{\mathbf{u}\omega_1}]_{\mathrm{n_{en}} * \mathrm{n_{dim}} \times \mathrm{n_{en}}} &= \int\limits_{\Omega_0^e} \mathbf{G}_{\mathbf{u}}^\top \mathbf{a}_{\mathbf{u}\omega_1} \mathbf{N}_\omega \; dV, \\
[\mathbf{K}_e^{\mathbf{u}\omega_2}]_{\mathrm{n_{en}} * \mathrm{n_{dim}} \times \mathrm{n_{en}}} &= \int\limits_{\Omega_0^e} \mathbf{G}_{\mathbf{u}}^\top \mathbf{a}_{\mathbf{u}\omega_2} \mathbf{N}_\omega \; dV, \\
[\mathbf{K}_e^{\mu\mathbf{u}}]_{\mathrm{n_{en}} \times \mathrm{n_{en}} * \mathrm{n_{dim}}} &= \int\limits_{\Omega_0^e} \left(\mathbf{N}_\mu^\top \left(\frac{1}{\Delta t} \mathbf{c}_{\mu\mathbf{u}} \right) \mathbf{G}_{\mathbf{u}} - \mathbf{B}_\mu^\top \mathbf{D}_{\mu\mathbf{u}} \mathbf{G}_{\mathbf{u}} \right) dV, \\
[\mathbf{K}_e^{\mu\mu}]_{\mathrm{n_{en}} \times \mathrm{n_{en}}} &= \int\limits_{\Omega_0^e} \left(\mathbf{N}_\mu^\top \left(\frac{1}{\Delta t} \frac{\partial C^w}{\partial \mu^w} \right) \mathbf{N}_\mu - \mathbf{B}_\mu^\top \mathbf{m}_{\mu\mu} \mathbf{N}_\mu + \mathbf{B}_\mu^\top \mathbf{M}^w \mathbf{B}_\mu \right) dV, \\
[\mathbf{K}_e^{\mu\omega_1}]_{\mathrm{n_{en}} \times \mathrm{n_{en}}} &= \int\limits_{\Omega_0^e} \left(\mathbf{N}_\mu^\top \left(\frac{1}{\Delta t} \frac{\partial C^w}{\partial \omega^{\beta_1}} \right) \mathbf{N}_\omega - \mathbf{B}_\mu^\top \mathbf{m}_{\mu\omega_1} \mathbf{N}_\omega + \mathbf{B}_\mu^\top \mathbf{M}^{w\beta_1} \mathbf{B}_\omega \right) dV,
\end{aligned}
$$

$$
\begin{aligned}
[\mathbf{K}_e^{\mu\omega_2}]_{\mathrm{n_{en}}\times\mathrm{n_{en}}} &= \int\limits_{\Omega_0^e} \left(\mathbf{N}_\mu^\top \left(\frac{1}{\Delta t}\frac{\partial C^w}{\partial \omega^{\beta_2}}\right)\mathbf{N}_\omega - \mathbf{B}_\mu^\top \mathbf{m}_{\mu\omega_2}\mathbf{N}_\omega + \mathbf{B}_\mu^\top \mathbf{M}^{w\beta_2}\mathbf{B}_\omega\right) dV,\\
[\mathbf{K}_e^{\omega_1\mathbf{u}}]_{\mathrm{n_{en}}\times\mathrm{n_{en}}*\mathrm{n_{dim}}} &= \int\limits_{\Omega_0^e} \left(\mathbf{N}_\omega^\top \left(\frac{1}{\Delta t}\mathbf{c}_{\omega_1\mathbf{u}}\right)\mathbf{G}_\mathbf{u} - \mathbf{B}_\omega^\top \mathbf{D}_{\omega_1\mathbf{u}}\mathbf{G}_\mathbf{u}\right) dV, \qquad (2.82)\\
[\mathbf{K}_e^{\omega_1\mu}]_{\mathrm{n_{en}}\times\mathrm{n_{en}}} &= \int\limits_{\Omega_0^e} \left(\mathbf{N}_\omega^\top \left(\frac{1}{\Delta t}\frac{\partial C^{\beta_1}}{\partial \mu^w}\right)\mathbf{N}_\mu - \mathbf{B}_\omega^\top \mathbf{m}_{\omega_1\mu}\mathbf{N}_\mu\right) dV,\\
[\mathbf{K}_e^{\omega_1\omega_1}]_{\mathrm{n_{en}}\times\mathrm{n_{en}}} &= \int\limits_{\Omega_0^e} \left(\mathbf{N}_\omega^\top \left(\frac{1}{\Delta t}\frac{\partial C^{\beta_1}}{\partial \omega_1}\right)\mathbf{N}_\omega - \mathbf{B}_\omega^\top \mathbf{m}_{\omega_1\omega_1}\mathbf{N}_\omega + \mathbf{B}_\omega^\top \mathbf{M}^{\beta_1\beta_1}\mathbf{B}_\omega\right) dV,\\
[\mathbf{K}_e^{\omega_1\omega_2}]_{\mathrm{n_{en}}\times\mathrm{n_{en}}} &= \int\limits_{\Omega_0^e} \left(\mathbf{N}_\omega^\top \left(\frac{1}{\Delta t}\frac{\partial C^{\beta_1}}{\partial \omega_2}\right)\mathbf{N}_\omega - \mathbf{B}_\omega^\top \mathbf{m}_{\omega_1\omega_2}\mathbf{N}_\omega + \mathbf{B}_\omega^\top \mathbf{M}^{\beta_1\beta_2}\mathbf{B}_\omega\right) dV,\\
[\mathbf{K}_e^{\omega_2\mathbf{u}}]_{\mathrm{n_{en}}\times\mathrm{n_{en}}*\mathrm{n_{dim}}} &= \int\limits_{\Omega_0^e} \left(\mathbf{N}_\omega^\top \left(\frac{1}{\Delta t}\mathbf{c}_{\omega_2\mathbf{u}}\right)\mathbf{G}_\mathbf{u} - \mathbf{B}_\omega^\top \mathbf{D}_{\omega_2\mathbf{u}}\mathbf{G}_\mathbf{u}\right) dV,\\
[\mathbf{K}_e^{\omega_2\mu}]_{\mathrm{n_{en}}\times\mathrm{n_{en}}} &= \int\limits_{\Omega_0^e} \left(\mathbf{N}_\omega^\top \left(\frac{1}{\Delta t}\frac{\partial C^{\beta_2}}{\partial \mu^w}\right)\mathbf{N}_\mu - \mathbf{B}_\omega^\top \mathbf{m}_{\omega_2\mu}\mathbf{N}_\mu\right) dV,\\
[\mathbf{K}_e^{\omega_2\omega_1}]_{\mathrm{n_{en}}\times\mathrm{n_{en}}} &= \int\limits_{\Omega_0^e} \left(\mathbf{N}_\omega^\top \left(\frac{1}{\Delta t}\frac{\partial C^{\beta_2}}{\partial \omega^{\beta_1}}\right)\mathbf{N}_\omega - \mathbf{B}_\omega^\top \mathbf{m}_{\omega_2\omega_1}\mathbf{N}_\omega + \mathbf{B}_\omega^\top \mathbf{M}^{\beta_2\beta_1}\mathbf{B}_\omega\right) dV,\\
[\mathbf{K}_e^{\omega_2\omega_2}]_{\mathrm{n_{en}}\times\mathrm{n_{en}}} &= \int\limits_{\Omega_0^e} \left(\mathbf{N}_\omega^\top \left(\frac{1}{\Delta t}\frac{\partial C^{\beta_2}}{\partial \omega^{\beta_2}}\right)\mathbf{N}_\omega - \mathbf{B}_\omega^\top \mathbf{m}_{\omega_2\omega_2}\mathbf{N}_\omega + \mathbf{B}_\omega^\top \mathbf{M}^{\beta_2\beta_2}\mathbf{B}_\omega\right) dV.
\end{aligned}
$$

$[\mathbf{G}_\mathbf{u}]$ in the geometric stiffness term is the non-symmetric gradient matrix of the shape functions, which appears as,

$$
\mathbf{G}_\mathbf{u} = \begin{bmatrix}\mathbf{G}_\mathbf{u}^1 & \mathbf{G}_\mathbf{u}^2 & \mathbf{G}_\mathbf{u}^3 & \cdots & \cdots & \mathbf{G}_\mathbf{u}^{\mathrm{n_{en}}}\end{bmatrix}_{\mathrm{n_{dim}^2}\times\mathrm{n_{en}}*\mathrm{n_{dim}}}, \qquad (2.83)
$$

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

$$
\mathbf{G}_\mathbf{u}^a = \begin{bmatrix} N_{a,1} & 0\\ 0 & N_{a,1}\\ N_{a,2} & 0\\ 0 & N_{a,2}\end{bmatrix}_{\mathrm{n_{dim}^2}\times\mathrm{n_{dim}}}, \qquad
\mathbf{G}_\mathbf{u}^a = \begin{bmatrix} N_{a,1} & 0 & 0\\ 0 & N_{a,1} & 0\\ 0 & 0 & N_{a,1}\\ N_{a,2} & 0 & 0\\ 0 & N_{a,2} & 0\\ 0 & 0 & N_{a,2}\\ N_{a,3} & 0 & 0\\ 0 & N_{a,3} & 0\\ 0 & 0 & N_{a,3}\end{bmatrix}_{\mathrm{n_{dim}^2}\times\mathrm{n_{dim}}}. \qquad (2.84)
$$

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

$$\Sigma_{\mathbf{S}} = \begin{bmatrix} \mathbf{S}_{11} & 0 & \mathbf{S}_{12} & 0 \\ 0 & \mathbf{S}_{11} & 0 & \mathbf{S}_{12} \\ \mathbf{S}_{12} & 0 & \mathbf{S}_{22} & 0 \\ 0 & \mathbf{S}_{12} & 0 & \mathbf{S}_{22} \end{bmatrix}, \quad \Sigma_{\mathbf{S}} = \begin{bmatrix} \mathbf{S}_{11} & 0 & 0 & \mathbf{S}_{12} & 0 & 0 & \mathbf{S}_{13} & 0 & 0 \\ 0 & \mathbf{S}_{11} & 0 & 0 & \mathbf{S}_{12} & 0 & 0 & \mathbf{S}_{13} & 0 \\ 0 & 0 & \mathbf{S}_{11} & 0 & 0 & \mathbf{S}_{12} & 0 & 0 & \mathbf{S}_{13} \\ \mathbf{S}_{12} & 0 & 0 & \mathbf{S}_{22} & 0 & 0 & \mathbf{S}_{23} & 0 & 0 \\ 0 & \mathbf{S}_{12} & 0 & 0 & \mathbf{S}_{22} & 0 & 0 & \mathbf{S}_{23} & 0 \\ 0 & 0 & \mathbf{S}_{12} & 0 & 0 & \mathbf{S}_{22} & 0 & 0 & \mathbf{S}_{23} \\ \mathbf{S}_{13} & 0 & 0 & \mathbf{S}_{23} & 0 & 0 & \mathbf{S}_{33} & 0 & 0 \\ 0 & \mathbf{S}_{13} & 0 & 0 & \mathbf{S}_{23} & 0 & 0 & \mathbf{S}_{33} & 0 \\ 0 & 0 & \mathbf{S}_{13} & 0 & 0 & \mathbf{S}_{23} & 0 & 0 & \mathbf{S}_{33} \end{bmatrix}. \tag{2.85}$$

$[\Sigma_{\mathbf{S}}]$ has a dimension of $[\Sigma_{\mathbf{S}}]_{\mathrm{n}^2_{\mathrm{dim}} \times \mathrm{n}^2_{\mathrm{dim}}}$. 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,

$$\Sigma_{\mathbf{F}} = \begin{bmatrix} F_{11} & F_{12} & F_{13} & 0 & 0 & 0 & \cdots & 0 & 0 & 0 \\ F_{21} & F_{22} & F_{23} & 0 & 0 & 0 & \cdots & 0 & 0 & 0 \\ F_{31} & F_{32} & F_{33} & 0 & 0 & 0 & \cdots & 0 & 0 & 0 \\ & & & & \cdots & & & & & \\ & & & & \cdots & & & & & \\ & & & & \cdots & & & & & \\ & & & & \cdots & & & & & \\ & & & & \cdots & & & & & \\ & & & & \cdots & & & & & \\ 0 & 0 & 0 & 0 & 0 & 0 & \cdots & F_{11} & F_{12} & F_{13} \\ 0 & 0 & 0 & 0 & 0 & 0 & \cdots & F_{21} & F_{22} & F_{23} \\ 0 & 0 & 0 & 0 & 0 & 0 & \cdots & F_{31} & F_{32} & F_{33} \end{bmatrix}_{\mathrm{n_{en}} * \mathrm{n_{dim}} \times \mathrm{n_{en}} * \mathrm{n_{dim}}}. \tag{2.86}$$

For a two-dimensional case, I can eliminate the rows and columns related to the third dimension and reduce the size of $\Sigma_{\mathbf{F}}$.

2.6 Some remarks on the matrix operators and tangents

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 $[\mathbf{N}_{\mathbf{u}}]_{\mathrm{n_{dim}} \times \mathrm{n_{en}} * \mathrm{n_{dim}}}$.
3. $\mathbf{B}_{\mathbf{u}} = \mathrm{sym}(\mathrm{Grad}(\mathbf{N}_{\mathbf{u}}))$ is the symmetric gradient matrix of the shape functions (also called the strain-displacement matrix) of dimension $[\mathbf{B}_{\mathbf{u}}]_{\mathrm{nstress} \times \mathrm{n_{en}} * \mathrm{n_{dim}}}$.
4. $\mathbf{G}_{\mathbf{u}} = \mathrm{Grad}(\mathbf{N}_{\mathbf{u}})$ is referred to as the non-symmetric gradient matrix of the shape function which has a dimension of $[\mathbf{G}_{\mathbf{u}}]_{\mathrm{n}^2_{\mathrm{dim}} \times \mathrm{n_{en}} * \mathrm{n_{dim}}}$.

5. $\Sigma_{\mathbf{S}}$ and $\Sigma_{\mathbf{F}}$ have dimensions of $[\Sigma_{\mathbf{S}}]_{\mathrm{n}_{\mathrm{dim}}^2 \times \mathrm{n}_{\mathrm{dim}}^2}$ and $[\Sigma_{\mathbf{F}}]_{\mathrm{n_{en}} * \mathrm{n_{dim}} \times \mathrm{n_{en}} * \mathrm{n_{dim}}}$, respectively.

6. $\mathbf{N}_\mu$ and $\mathbf{N}_\omega$ are vectors of dimension of $[\bullet]_{1\times \mathrm{n_{en}}}$.

7. $\mathbf{B}_\mu = \mathrm{Grad}(\mathbf{N}_\mu) = \frac{\partial \mathbf{N}_\mu}{\partial \mathbf{X}}$ and $\mathbf{B}_\omega = \mathrm{Grad}(\mathbf{N}_\omega) = \frac{\partial \mathbf{N}_\omega}{\partial \mathbf{X}}$ are matrices of dimension $[\bullet]_{\mathrm{n_{dim}}\times \mathrm{n_{en}}}$.

8. $[\mathbf{D}_{\mathbb{C}}]_{\mathrm{nstress}\times\mathrm{nstress}}$ 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,

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

9. $\mathbf{c}_{\mu\mathbf{u}}$, $\mathbf{c}_{\omega_1\mathbf{u}}$, and $\mathbf{c}_{\omega_2\mathbf{u}}$ are vectors of dimension $[\bullet]_{1\times \mathrm{n}_{\mathrm{dim}}^2}$.

10. $\mathbf{a}_{\mathbf{u}\mu}$, $\mathbf{a}_{\mathbf{u}\omega_1}$, and $\mathbf{a}_{\mathbf{u}\omega_2}$ are vectors of dimension $[\bullet]_{\mathrm{n}_{\mathrm{dim}}^2\times 1}$ obtained from the second-order tangent tensors, $\mathbb{S}^\mu$, $\mathbb{S}^{\beta_1}$, and $\mathbb{S}^{\beta_2}$, respectively, which are defined as,

$$\mathbb{S}^\mu_{iI} = F_{iJ}\frac{\partial S_{JI}}{\partial C^w}\frac{\partial C^w}{\partial \mu^w}, \quad \mathbb{S}^{\beta_1}_{iI} = F_{iJ}\frac{\partial S_{JI}}{\partial C^w}\frac{\partial C^w}{\partial \omega^{\beta_1}}, \quad \text{and} \quad \mathbb{S}^{\beta_2}_{iI} = F_{iJ}\frac{\partial S_{JI}}{\partial C^w}\frac{\partial C^w}{\partial \omega^{\beta_2}}$$

11. $\mathbf{D}_{\mu\mathbf{u}}$, $\mathbf{D}_{\omega_1\mathbf{u}}$, and $\mathbf{D}_{\omega_2\mathbf{u}}$ are matrices of dimension $[\bullet]_{\mathrm{n_{dim}}\times \mathrm{n}_{\mathrm{dim}}^2}$ obtained from the third-order chemo-mechanical tangent tensor, $\mathbb{J}$, defined as,

$$\begin{aligned}
(\mathbb{J}^w)_{IkL} &= \frac{\partial J^w_I}{\partial F_{kL}} + \frac{\partial J^w_I}{\partial C^w}\frac{\partial C^w}{\partial F_{kL}},\\
(\mathbb{J}^{\beta_1})_{IkL} &= \frac{\partial J^{\beta_1}_I}{\partial F_{kL}} + \frac{\partial J^{\beta_1}_I}{\partial C^{\beta_1}}\frac{\partial C^{\beta_1}}{\partial F_{kL}},\\
(\mathbb{J}^{\beta_2})_{IkL} &= \frac{\partial J^{\beta_2}_I}{\partial F_{kL}} + \frac{\partial J^{\beta_2}_I}{\partial C^{\beta_2}}\frac{\partial C^{\beta_2}}{\partial F_{kL}}.
\end{aligned}$$

12. $\mathbf{m}_{\mu\mu}$, $\mathbf{m}_{\mu\omega_1}$, $\mathbf{m}_{\omega_1\mu}$, $\mathbf{m}_{\omega_1\omega_1}$, $\mathbf{m}_{\mu\omega_2}$, $\mathbf{m}_{\omega_2\mu}$, $\mathbf{m}_{\omega_1\omega_2}$, $\mathbf{m}_{\omega_2\omega_1}$, and $\mathbf{m}_{\omega_2\omega_2}$ are vectors of dimension $[\bullet]_{\mathrm{n_{dim}}\times 1}$ defined as,

$$\begin{aligned}
(\mathbf{m}_{\mu\mu})_I &= \frac{\partial J^w_I}{\partial C^w}\frac{\partial C^w}{\partial \mu^w}, & (\mathbf{m}_{\mu\omega_1})_I &= \frac{\partial J^w_I}{\partial C^w}\frac{\partial C^w}{\partial \omega^{\beta_1}}, & (\mathbf{m}_{\omega_1\mu})_I &= \frac{\partial J^{\beta_1}_I}{\partial C^{\beta_1}}\frac{\partial C^{\beta_1}}{\partial \mu^w},\\
(\mathbf{m}_{\omega_1\omega_1})_I &= \frac{\partial J^{\beta_1}_I}{\partial C^{\beta_1}}\frac{\partial C^{\beta_1}}{\partial \omega^{\beta_1}}, & (\mathbf{m}_{\mu\omega_2})_I &= \frac{\partial J^w_I}{\partial C^w}\frac{\partial C^w}{\partial \omega^{\beta_2}}, & (\mathbf{m}_{\omega_2\mu})_I &= \frac{\partial J^{\beta_2}_I}{\partial C^{\beta_2}}\frac{\partial C^{\beta_2}}{\partial \mu^w},\\
(\mathbf{m}_{\omega_1\omega_2})_I &= \frac{\partial J^{\beta_1}_I}{\partial C^{\beta_2}}\frac{\partial C^{\beta_2}}{\partial \omega^{\beta_2}}, & (\mathbf{m}_{\omega_2\omega_1})_I &= \frac{\partial J^{\beta_2}_I}{\partial C^{\beta_1}}\frac{\partial C^{\beta_1}}{\partial \omega^{\beta_1}}, & (\mathbf{m}_{\omega_2\omega_2})_I &= \frac{\partial J^{\beta_2}_I}{\partial C^{\beta_2}}\frac{\partial C^{\beta_2}}{\partial \omega^{\beta_2}},
\end{aligned} \tag{2.87}$$

13. The time derivative of the referential concentration of the solvent, $\dot{C}^w$, and concentration

of the ionic species, C^{β_1} and C^{β_2}, are calculated using the following formula

$$\dot{C}^w = \frac{C^w - C^w_{t+\Delta t}}{\Delta t}, \quad \dot{C}^{\beta_1} = \frac{C^{\beta_1} - C^{\beta_1}_{t+\Delta t}}{\Delta t}, \quad \text{and} \quad \dot{C}^{\beta_2} = \frac{C^{\beta_2} - C^{\beta_2}_{t+\Delta t}}{\Delta t}. \tag{2.88}$$

3 Constitutive model-specific tangent moduli

Calculating the stiffness (tangent) matrix for the chemo-mechanical element ($\mathbf{u}$-μ-ω) 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 polyelectrolyte hydrogel. To reduce the computation burden and simplify the implementation procedure, I will neglect some small terms in the constitutive model.

3.1 Local iteration to solve internal variables

Let me recall the constitutive relations for the chemical potential of the solvent, the electrochemical potential of the ionic species, and the electroneutrality condition,

$$\begin{aligned}
\mathcal{G}_1 &\equiv \mu^0_w + R\theta \left[\phi^p + \ln(1-\phi^p) + \chi(\phi^p)^2\right] + \mathcal{P}\mathcal{V}^w - R\theta \sum_\beta \frac{C^\beta}{C^w} - \mu^w = 0,\\
\mathcal{G}_2 &\equiv \omega^0_{\beta_1} + R\theta \ln\left(\frac{C^{\beta_1}}{C^w}\right) + F\psi z^{\beta_1} + p\mathcal{V}^{\beta_1} - \omega^{\beta_1} = 0,\\
\mathcal{G}_3 &\equiv \omega^0_{\beta_2} + R\theta \ln\left(\frac{C^{\beta_2}}{C^w}\right) + F\psi z^{\beta_2} + p\mathcal{V}^{\beta_2} - \omega^{\beta_2} = 0,\\
\mathcal{G}_4 &\equiv C^w \sum_\beta z^\beta \exp\left(\frac{\omega^\beta - F\psi z^\beta - p\mathcal{V}^\beta - \omega^0_\beta}{R\theta}\right) + C^p z^p = 0,
\end{aligned} \tag{3.1}$$

where,

$$\mathcal{P} = \frac{\kappa}{2}(\ln J^e)^2 - \kappa \ln J^e = \frac{\kappa}{2}\left[\ln\left(\frac{J\phi^p}{\phi^p_0}\right)\right]^2 - \kappa\left[\ln\left(\frac{J\phi^p}{\phi^p_0}\right)\right]. \tag{3.2}$$

The mean pressure, p, for Neo-Hookean type elastomeric gel is given by,

$$\begin{aligned}
p &= \frac{-1}{3\phi^p_0 J^s}\left(G_0 I_1 - 3G_0(\phi^p_0)^{2/3}\right) - \kappa(\ln J^e),\\
&= \frac{G_0\phi^p}{3\phi^p_0}\left(3(\phi^p_0)^{2/3} - I_1\right) - \kappa\left[\ln\left(\frac{J\phi^p}{\phi^p_0}\right)\right].
\end{aligned} \tag{3.3}$$

On the other hand, for Arruda-Boyce type elastomeric gel, the mean pressure, p, is given by,

$$p = \frac{G_0\phi^p}{3\phi^p_0}\left[3\left(\frac{\lambda_{\mathrm{L}}}{3}\beta_0\right)(\phi^p_0)^{2/3} - \left(\frac{\lambda_{\mathrm{L}}}{3\lambda_c}\beta_c\right) I_1\right] - \kappa\left[\ln\left(\frac{J\phi^p}{\phi^p_0}\right)\right]. \tag{3.4}$$

The nonlinear constitutive functions, $\mathcal{G}_1$, $\mathcal{G}_2$, $\mathcal{G}_3$ and $\mathcal{G}_4$ form a coupled system of equations.

In a finite element framework, knowing the nodal degrees of freedom $(\mathbf{u}, \mu^w, \omega^{\beta_1}, \omega^{\beta_2})$, a local Newton iteration is performed to solve for the set of internal variables $(C^w, C^{\beta_1}, C^{\beta_2}, \psi)$. For a set of initial guesses for the unknown variables, the solution is iteratively updated until a user-specified convergence criterion is achieved. To solve for the unknown internal variables, a local Newton-Raphson procedure is employed within the element,

$$
\underbrace{-\begin{bmatrix} \frac{\partial \mathcal{G}_1}{\partial C^w} & \frac{\partial \mathcal{G}_1}{\partial C^{\beta_1}} & \frac{\partial \mathcal{G}_1}{\partial C^{\beta_2}} & \frac{\partial \mathcal{G}_1}{\partial \psi} \\ \frac{\partial \mathcal{G}_2}{\partial C^w} & \frac{\partial \mathcal{G}_2}{\partial C^{\beta_1}} & \frac{\partial \mathcal{G}_2}{\partial C^{\beta_2}} & \frac{\partial \mathcal{G}_2}{\partial \psi} \\ \frac{\partial \mathcal{G}_3}{\partial C^w} & \frac{\partial \mathcal{G}_3}{\partial C^{\beta_1}} & \frac{\partial \mathcal{G}_3}{\partial C^{\beta_2}} & \frac{\partial \mathcal{G}_3}{\partial \psi} \\ \frac{\partial \mathcal{G}_4}{\partial C^w} & \frac{\partial \mathcal{G}_4}{\partial C^{\beta_1}} & \frac{\partial \mathcal{G}_4}{\partial C^{\beta_2}} & \frac{\partial \mathcal{G}_4}{\partial \psi} \end{bmatrix}}_{\frac{d\mathcal{G}}{d\mathbf{f}_e}} \underbrace{\begin{Bmatrix} \Delta C^w \\ \Delta C^{\beta_1} \\ \Delta C^{\beta_2} \\ \Delta \psi \end{Bmatrix}}_{\Delta \mathbf{f}_e} = \underbrace{\begin{Bmatrix} \mathcal{G}_1 \\ \mathcal{G}_2 \\ \mathcal{G}_3 \\ \mathcal{G}_4 \end{Bmatrix}}_{\mathcal{G}} \tag{3.5}
$$

where the unknown internal variables are solved iteratively as follows until convergence is achieved,

$$
\mathbf{f}_e^{(k+1)} = \mathbf{f}_e^{(k)} + \Delta \mathbf{f}_e^{(k+1)}, \quad \text{where,} \;\; \Delta \mathbf{f}_e^{(k+1)} = -\left(\frac{d\mathcal{G}}{d\mathbf{f}_e}\right)^{-1} \mathcal{G}^{(k)}, \tag{3.6}
$$

Following are the required tangents for the local Newton-Raphson iteration for the internal variables,

$$
\begin{aligned}
\frac{\partial \mathcal{G}_1}{\partial C^w} &= -\frac{\mathcal{V}^w(\phi^p)^2}{\phi_0^p}\left[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)\right]-R\theta\sum_\beta C^\beta,\\
\frac{\partial \mathcal{G}_1}{\partial C^{\beta_1}} &= \frac{\partial \mathcal{G}_1}{\partial C^{\beta_2}} = -\frac{R\theta}{C^w},\\
\frac{\partial \mathcal{G}_1}{\partial \psi} &= 0,\\
\frac{\partial \mathcal{G}_2}{\partial C^w} &= -\frac{R\theta}{C^w}+\frac{(\phi^p)^2\mathcal{V}^w}{\phi_0^p}\left[\frac{G_0}{3\phi_0^p}\left(I_1-3(\phi_0^p)^{2/3}\right)+\frac{\kappa}{\phi^p}\right]\mathcal{V}^{\beta_1},\\
\frac{\partial \mathcal{G}_2}{\partial C^{\beta_1}} &= \frac{R\theta}{C^{\beta_1}},\\
\frac{\partial \mathcal{G}_2}{\partial C^{\beta_2}} &= 0,\\
\frac{\partial \mathcal{G}_2}{\partial \psi} &= Fz^{\beta_1}\\
\frac{\partial \mathcal{G}_3}{\partial C^w} &= -\frac{R\theta}{C^w}+\frac{(\phi^p)^2\mathcal{V}^w}{\phi_0^p}\left[\frac{G_0}{3\phi_0^p}\left(I_1-3(\phi_0^p)^{2/3}\right)+\frac{\kappa}{\phi^p}\right]\mathcal{V}^{\beta_2},\\
\frac{\partial \mathcal{G}_3}{\partial C^{\beta_1}} &= 0\\
\frac{\partial \mathcal{G}_3}{\partial C^{\beta_2}} &= \frac{R\theta}{C^{\beta_2}},\\
\frac{\partial \mathcal{G}_3}{\partial \psi} &= Fz^{\beta_2}\\
\frac{\partial \mathcal{G}_4}{\partial C^w} &= \sum_\beta z^\beta \exp\left(\frac{\omega^\beta-p\mathcal{V}^\beta-F\psi z^\beta-\omega_\beta^0}{R\theta}\right)\left(1-\frac{(\phi^p)^2C^w\mathcal{V}^w}{R\theta\phi_0^p}\left[\frac{G_0}{3\phi_0^p}\left(I_1-3(\phi_0^p)^{2/3}\right)+\frac{\kappa}{\phi^p}\right]\mathcal{V}^\beta\right),\\
\frac{\partial \mathcal{G}_4}{\partial C^{\beta_1}} &= \frac{\partial \mathcal{G}_4}{\partial C^{\beta_2}} = 0,\\
\frac{\partial \mathcal{G}_4}{\partial \psi} &= -\frac{FC^w}{R\theta}\sum_\beta\left(z^\beta\right)^2\exp\left(\frac{\omega^\beta-p\mathcal{V}^\beta-F\psi z^\beta-\omega_\beta^0}{R\theta}\right).
\end{aligned}
\tag{3.7}
$$

Remark 4. The second term in $\frac{\partial \mathcal{G}_2}{\partial C^w}$ and $\frac{\partial \mathcal{G}_3}{\partial C^w}$ are small enough that they can be ignored during implementation (the term $p\mathcal{V}^\beta$ is often ignored in literature for the same reason). In this case, it is equivalent to not considering $p \equiv p(C^w)$.

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

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

$$\begin{aligned}\mu_w^0 + R\theta\left[\phi^p + \ln(1-\phi^p) + \chi(\phi^p)^2\right] - \kappa\mathcal{V}^w\left[\ln\left(\frac{J\phi^p}{\phi_0^p}\right)\right] \\ + \frac{\kappa\mathcal{V}^w}{2}\left[\ln\left(\frac{J\phi^p}{\phi_0^p}\right)\right]^2 - R\theta\sum_\beta \frac{C^\beta}{C^w} - \mu^w = 0, \\ \Rightarrow \mathcal{G}_1\left(\mathbf{F}, \mu^w; C^w, C^{\beta_1}, C^{\beta_2}\right) = 0.\end{aligned} \tag{3.8}$$

Thus, the partial derivative of the implicit constitutive function, $\mathcal{G}_1$, with respect to μ^w can be written as,

$$\begin{aligned}&\frac{d\mathcal{G}_1}{d\mu^w} = \left.\frac{\partial\mathcal{G}_1}{\partial\mathbf{F}}\right|_{\mu^w,\phi^p,C^\beta}\frac{\partial\mathbf{F}}{\partial\mu^w} + \left.\frac{\partial\mathcal{G}_1}{\partial\mu^w}\right|_{\mathbf{F},\phi^p,C^\beta}\frac{\partial\mu^w}{\partial\mu^w} + \left.\frac{\partial\mathcal{G}_1}{\partial\phi^p}\right|_{\mathbf{F},\mu^w,C^\beta}\frac{\partial\phi^p}{\partial C^w}\frac{\partial C^w}{\partial\mu^w} + \left.\frac{\partial\mathcal{G}_1}{\partial C^\beta}\right|_{\mathbf{F},\mu^w,\phi^p}\frac{\partial C^\beta}{\partial\mu^w}, \\ &\Rightarrow \frac{\partial\mathcal{G}_1}{\partial\mu^w} + \left.\frac{\partial\mathcal{G}_1}{\partial\phi^p}\right|_{\mathbf{F},\mu^w,C^\beta}\frac{\partial\phi^p}{\partial C^w}\frac{\partial C^w}{\partial\mu^w} = 0 \qquad \left(\text{since, } \frac{\partial\mu^w}{\partial\mu^w} = 1 \text{ and } \frac{\partial\mathbf{F}}{\partial\mu^w} = \frac{\partial C^\beta}{\partial\mu^w} = 0\right), \\ &\qquad\Rightarrow \frac{\partial C^w}{\partial\mu^w} = -\frac{\frac{\partial\mathcal{G}_1}{\partial\mu^w}}{\frac{\partial\mathcal{G}_1}{\partial\phi^p}\frac{\partial\phi^p}{\partial C^w}} = \frac{\partial C^w}{\partial\phi^p}\left(\frac{\partial\mathcal{G}_1}{\partial\phi^p}\right)^{-1} \qquad \left(\text{since, } \frac{\partial\mathcal{G}_1}{\partial\mu^w} = -1\right)\end{aligned} \tag{3.9}$$

Polymer volume fraction and its derivative with respect to the referential concentration of the solvent is

$$\phi^p = \frac{C^p\mathcal{V}^p}{C^p\mathcal{V}^p + C^w\mathcal{V}^w} \qquad \Rightarrow \qquad \frac{\partial C^w}{\partial\phi^p} = -\frac{C^p\mathcal{V}^p}{\mathcal{V}^w(\phi^p)^2} = -\frac{\phi_0^p}{\mathcal{V}^w(\phi^p)^2}. \tag{3.10}$$

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,

$$\frac{\partial\mathcal{G}_1}{\partial\phi^p} = R\theta\left(1 - \frac{1}{1-\phi^p} + 2\chi\phi^p\right) - \frac{\kappa\mathcal{V}^w}{\phi^p} + \frac{2\kappa\mathcal{V}^w}{\phi^p}\ln\left(\frac{J\phi^p}{\phi_0^p}\right) \tag{3.11}$$

By substituting the expressions for $\frac{\partial C^w}{\partial\phi^p}$ and $\left(\frac{\partial\mathcal{G}_1}{\partial\phi^p}\right)^{-1}$, I can obtain the expression for $\frac{\partial C^w}{\partial\mu^w}$ during implementation.

3.3 Calculation of $\dfrac{\partial C^w}{\partial \omega^{\beta_1}}$

As discussed before, the simplified form of the constitutive relation for ω^{β_1} is given by,

$$\omega_{\beta_1}^0 + R\theta\ln\left(\frac{C^{\beta_1}}{C^w}\right) + F\psi z^{\beta_1} - \omega^{\beta_1} = 0 \qquad \Rightarrow \mathcal{G}_2\left(\omega^{\beta_q}; C^w, C^{\beta_1}, \psi\right) = 0. \tag{3.12}$$

By taking partial derivative of $\mathcal{G}_2$, I have,

$$\frac{d\mathcal{G}_2}{d\omega^{\beta_1}} = \frac{\partial \mathcal{G}_2}{\partial \omega^{\beta_1}}\bigg|_{C^w, C^{\beta_1}, \psi} \frac{\partial \omega^{\beta_1}}{\partial \omega^{\beta_1}} + \frac{\partial \mathcal{G}_2}{\partial C^w}\bigg|_{\omega^{\beta_1}, C^{\beta_1}, \psi} \frac{\partial C^w}{\partial \omega^{\beta_1}} + \frac{\partial \mathcal{G}_2}{\partial C^{\beta_1}}\bigg|_{\omega^{\beta_1}, C^w, \psi} \frac{\partial C^{\beta_1}}{\partial \omega^{\beta_1}} + \frac{\partial \mathcal{G}_2}{\partial \psi}\bigg|_{\omega^{\beta_1}, C^w, C^{\beta_1}} \frac{\partial \psi}{\partial \omega^{\beta_1}},$$

$$\Rightarrow \frac{\partial \mathcal{G}_2}{\partial \omega^{\beta_1}} + \frac{\partial \mathcal{G}_2}{\partial C^w}\frac{\partial C^w}{\partial \omega^{\beta_1}} = 0,$$

$$\Rightarrow \frac{\partial C^w}{\partial \omega^{\beta_1}} = -\frac{\frac{\partial \mathcal{G}_2}{\partial \omega^{\beta_1}}}{\frac{\partial \mathcal{G}_2}{\partial C^w}} = -\frac{\partial \mathcal{G}_2}{\partial \omega^{\beta_1}}\left(\frac{\partial \mathcal{G}_2}{\partial C^w}\right)^{-1},$$

$$\Rightarrow \frac{\partial C^w}{\partial \omega^{\beta_1}} = -\frac{C^w}{R\theta}. \tag{3.13}$$

This can be generalized for $\frac{\partial C^w}{\partial \omega^{\beta_k}} = -\frac{C^w}{R\theta}$.

3.4 Calculation of $\frac{\partial C^{\beta_1}}{\partial \mu^w}$

Following the same procedure as before for $\mathcal{G}_1$, I can write,

$$\frac{\partial C^{\beta_1}}{\partial \mu^w} = -\frac{\partial \mathcal{G}_1}{\partial \mu^w}\left(\frac{\partial \mathcal{G}_1}{\partial C^{\beta_1}}\right)^{-1} = -\frac{C^w}{R\theta}. \tag{3.14}$$

This can be generalized for $\frac{\partial C^{\beta_k}}{\partial \mu^w} = -\frac{C^w}{R\theta}$.

3.5 Calculation of $\frac{\partial C^{\beta_1}}{\partial \omega^{\beta_1}}$

By applying the similar procedure on $\mathcal{G}_2$, I can write,

$$\frac{\partial C^{\beta_1}}{\partial \omega^{\beta_1}} = -\frac{\partial \mathcal{G}_2}{\partial \omega^{\beta_1}}\left(\frac{\partial \mathcal{G}_2}{\partial C^{\beta_1}}\right)^{-1} = \frac{C^{\beta_1}}{R\theta}. \tag{3.15}$$

This can be generalized for $\frac{\partial C^{\beta_k}}{\partial \omega^{\beta_k}} = \frac{C^{\beta_1}}{R\theta}$.

3.6 Calculation of $\frac{\partial C^{\beta_1}}{\partial \omega^{\beta_2}}$

$$\frac{\partial C^{\beta_1}}{\partial \omega^{\beta_2}} = 0. \tag{3.16}$$

This can be generalized for $\frac{\partial C^{\beta_i}}{\partial \omega^{\beta_k}} = 0$.

3.7 Calculation of $\dfrac{\partial C^w}{\partial \mathbf{F}}$, $\dfrac{\partial C^{\beta 1}}{\partial \mathbf{F}}$, and $\dfrac{\partial C^{\beta 2}}{\partial \mathbf{F}}$

Let me recall the constitutive relations for the solvent chemical potential and ionic species electrochemical potential,

$$
\begin{aligned}
\mathcal{G}_1 &\equiv \mu_w^0 + R\theta\left[\phi^p + \ln(1-\phi^p) + \chi(\phi^p)^2\right] + \mathcal{P}\mathcal{V}^w - R\theta \sum_\beta \frac{C^\beta}{C^w} - \mu^w = 0, \\
\mathcal{G}_2 &\equiv \omega_{\beta_1}^0 + R\theta \ln\left(\frac{C^{\beta_1}}{C^w}\right) + F\psi z^{\beta_1} + p\mathcal{V}^{\beta_1} - \omega^{\beta_1} = 0, \\
\mathcal{G}_3 &\equiv \omega_{\beta_2}^0 + R\theta \ln\left(\frac{C^{\beta_2}}{C^w}\right) + F\psi z^{\beta_2} + p\mathcal{V}^{\beta_2} - \omega^{\beta_2} = 0.
\end{aligned}
\tag{3.17}
$$

where,

$$
\begin{aligned}
\mathcal{P} &= \frac{\kappa}{2}\left[\ln\left(\frac{J\phi^p}{\phi_0^p}\right)\right]^2 - \kappa\left[\ln\left(\frac{J\phi^p}{\phi_0^p}\right)\right], \\
p &= \frac{G_0\phi^p}{3\phi_0^p}\left(3(\phi_0^p)^{2/3} - I_1\right) - \kappa\left[\ln\left(\frac{J\phi^p}{\phi_0^p}\right)\right].
\end{aligned}
\tag{3.18}
$$

In functional form, these can be expressed as,

$$
\begin{aligned}
\mathcal{G}_1\left(\mathbf{F}, \mu^w; C^w, C^{\beta_1}, C^{\beta_2}\right) &= 0, \\
\mathcal{G}_2\left(\mathbf{F}, \omega^{\beta_1}; C^w, C^{\beta_1}, \psi\right) &= 0, \\
\mathcal{G}_3\left(\mathbf{F}, \omega^{\beta_2}; C^w, C^{\beta_2}, \psi\right) &= 0,
\end{aligned}
\tag{3.19}
$$

Internal variables, C^w, C^{β_1}, and C^{β_2} are related to the deformation gradient, $\mathbf{F}$ through this set of nonlinear implicit constitutive relations. For the above set of nonlinear implicit functions, I can write,

$$
\begin{aligned}
\frac{\partial \mathcal{G}_1}{\partial \mathbf{F}}\frac{\partial \mathbf{F}}{\partial \mathbf{F}} + \frac{\partial \mathcal{G}_1}{\cancel{\partial \mu^w}}\frac{\cancelto{0}{\partial \mu^w}}{\partial \mathbf{F}} + \frac{\partial \mathcal{G}_1}{\partial C^w}\frac{\partial C^w}{\partial \mathbf{F}} + \frac{\partial \mathcal{G}_1}{\partial C^{\beta_1}}\frac{\partial C^{\beta_1}}{\partial \mathbf{F}} + \frac{\partial \mathcal{G}_1}{\partial C^{\beta_2}}\frac{\partial C^{\beta_2}}{\partial \mathbf{F}} &= \frac{d\mathcal{G}_1}{d\mathbf{F}} = 0, \\
\frac{\partial \mathcal{G}_2}{\partial \mathbf{F}}\frac{\partial \mathbf{F}}{\partial \mathbf{F}} + \frac{\partial \mathcal{G}_2}{\cancel{\partial \mu^w}}\frac{\cancelto{0}{\partial \mu^w}}{\partial \mathbf{F}} + \frac{\partial \mathcal{G}_2}{\partial C^w}\frac{\partial C^w}{\partial \mathbf{F}} + \frac{\partial \mathcal{G}_2}{\partial C^{\beta_1}}\frac{\partial C^{\beta_1}}{\partial \mathbf{F}} + \frac{\partial \mathcal{G}_2}{\cancel{\partial \psi}}\frac{\cancelto{0}{\partial \psi}}{\partial \mathbf{F}} &= \frac{d\mathcal{G}_2}{d\mathbf{F}} = 0, \\
\frac{\partial \mathcal{G}_3}{\partial \mathbf{F}}\frac{\partial \mathbf{F}}{\partial \mathbf{F}} + \frac{\partial \mathcal{G}_3}{\cancel{\partial \mu^w}}\frac{\cancelto{0}{\partial \mu^w}}{\partial \mathbf{F}} + \frac{\partial \mathcal{G}_3}{\partial C^w}\frac{\partial C^w}{\partial \mathbf{F}} + \frac{\partial \mathcal{G}_3}{\partial C^{\beta_2}}\frac{\partial C^{\beta_2}}{\partial \mathbf{F}} + \frac{\partial \mathcal{G}_3}{\cancel{\partial \psi}}\frac{\cancelto{0}{\partial \psi}}{\partial \mathbf{F}} &= \frac{d\mathcal{G}_3}{d\mathbf{F}} = 0.
\end{aligned}
\tag{3.20}
$$

The above system of linear equations can be expressed in matrix form as follows and solved for $\frac{\partial C^w}{\partial \mathbf{F}}$, $\frac{\partial C^{\beta_1}}{\partial \mathbf{F}}$, and $\frac{\partial C^{\beta_2}}{\partial \mathbf{F}}$ as follows,

$$\begin{Bmatrix} \frac{\partial C^w}{\partial \mathbf{F}} \\ \frac{\partial C^{\beta_1}}{\partial \mathbf{F}} \\ \frac{\partial C^{\beta_2}}{\partial \mathbf{F}} \end{Bmatrix} = - \begin{bmatrix} \frac{\partial \mathcal{G}_1}{\partial C^w} & \frac{\partial \mathcal{G}_1}{\partial C^{\beta_1}} & \frac{\partial \mathcal{G}_1}{\partial C^{\beta_2}} \\ \frac{\partial \mathcal{G}_2}{\partial C^w} & \frac{\partial \mathcal{G}_2}{\partial C^{\beta_1}} & \frac{\partial \mathcal{G}_2}{\partial C^{\beta_2}} \\ \frac{\partial \mathcal{G}_3}{\partial C^w} & \frac{\partial \mathcal{G}_3}{\partial C^{\beta_1}} & \frac{\partial \mathcal{G}_3}{\partial C^{\beta_2}} \end{bmatrix}^{-1} \begin{Bmatrix} \frac{\partial \mathcal{G}_1}{\partial \mathbf{F}} \\ \frac{\partial \mathcal{G}_2}{\partial \mathbf{F}} \\ \frac{\partial \mathcal{G}_3}{\partial \mathbf{F}}. \end{Bmatrix} \tag{3.21}$$

The derivatives within the coefficient matrix were calculated previously and listed below,

$$\begin{aligned}
\frac{\partial \mathcal{G}_1}{\partial C^w} &= -\frac{\mathcal{V}^w(\phi^p)^2}{\phi_0^p}\left[R\theta\left(1 - \frac{1}{1-\phi^p} + 2\chi\phi^p\right) - \frac{\kappa\mathcal{V}^w}{\phi^p} + \frac{\kappa\mathcal{V}^w}{\phi^p}\ln\left(\frac{J\phi^p}{\phi_0^p}\right)\right] - R\theta\sum_\beta C^\beta, \\
\frac{\partial \mathcal{G}_1}{\partial C^{\beta_1}} &= \frac{\partial \mathcal{G}_1}{\partial C^{\beta_2}} = -\frac{R\theta}{C^w}, \\
\frac{\partial \mathcal{G}_2}{\partial C^w} &= -\frac{R\theta}{C^w} + \frac{(\phi^p)^2\mathcal{V}^w}{\phi_0^p}\left[\frac{G_0}{3\phi_0^p}\left(I_1 - 3(\phi_0^p)^{2/3}\right) + \frac{\kappa}{\phi^p}\right]\mathcal{V}^{\beta_1}, \\
\frac{\partial \mathcal{G}_3}{\partial C^w} &= -\frac{R\theta}{C^w} + \frac{(\phi^p)^2\mathcal{V}^w}{\phi_0^p}\left[\frac{G_0}{3\phi_0^p}\left(I_1 - 3(\phi_0^p)^{2/3}\right) + \frac{\kappa}{\phi^p}\right]\mathcal{V}^{\beta_2}, \\
\frac{\partial \mathcal{G}_2}{\partial C^{\beta_1}} &= \frac{R\theta}{C^{\beta_1}}, \\
\frac{\partial \mathcal{G}_2}{\partial C^{\beta_2}} &= 0, \\
\frac{\partial \mathcal{G}_3}{\partial C^{\beta_1}} &= \frac{R\theta}{C^{\beta_2}}, \\
\frac{\partial \mathcal{G}_3}{\partial C^{\beta_2}} &= 0.
\end{aligned} \tag{3.22}$$

Accordingly, the residuals are given as,

$$\begin{aligned}
\frac{\partial \mathcal{G}_1}{\partial \mathbf{F}} &= -\kappa\mathcal{V}^w\mathbf{F}^{-\top} + \kappa\mathcal{V}^w\ln\left(\frac{J\phi^p}{\phi_0^p}\right)\mathbf{F}^{-\top} = \kappa\mathcal{V}^w\left[\ln(J^e) - 1\right]\mathbf{F}^{-\top}, \\
\frac{\partial \mathcal{G}_2}{\partial \mathbf{F}} &= -\left(\frac{2G_0\phi^p}{3\phi_0^p}\mathbf{F} + \kappa\mathcal{V}^w\mathbf{F}^{-\top}\right)\mathcal{V}^{\beta_1}, \\
\frac{\partial \mathcal{G}_3}{\partial \mathbf{F}} &= -\left(\frac{2G_0\phi^p}{3\phi_0^p}\mathbf{F} + \kappa\mathcal{V}^w\mathbf{F}^{-\top}\right)\mathcal{V}^{\beta_2}.
\end{aligned} \tag{3.23}$$

An alternative approach

By ignoring the coupling between the system of implicit nonlinear equations, we can calculate the derivatives as follows,

$$
\begin{aligned}
\frac{\partial C^w}{\partial \mathbf{F}} &= -\left(\frac{\partial \mathcal{G}_1}{\partial C^w}\right)^{-1} \frac{\partial \mathcal{G}_1}{\partial \mathbf{F}}, \\
\frac{\partial C^{\beta_1}}{\partial \mathbf{F}} &= -\left(\frac{\partial \mathcal{G}_2}{\partial C^{\beta_1}}\right)^{-1} \frac{\partial \mathcal{G}_1}{\partial \mathbf{F}}, \\
\frac{\partial C^{\beta_2}}{\partial \mathbf{F}} &= -\left(\frac{\partial \mathcal{G}_3}{\partial C^{\beta_2}}\right)^{-1} \frac{\partial \mathcal{G}_1}{\partial \mathbf{F}}.
\end{aligned}
\tag{3.24}
$$

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

Once $\frac{\partial C^w}{\partial \mathbf{F}}$ is calculated, I can perform the following procedure,

$$
\begin{aligned}
\left(\frac{\partial C^w}{\partial \mathbf{C}}\right)_{KL} &= \frac{\partial C^w}{\partial F_{mN}} \frac{\partial F_{mN}}{\partial C_{KL}} \\
&= \frac{\partial C^w}{\partial F_{mN}} \left(\frac{\partial C_{KL}}{\partial F_{mN}}\right)^{-1}, \\
&= \frac{1}{2} \frac{\partial C^w}{\partial F_{mN}} F^{-1}_{Lm} \delta_{KN}, \\
&= \frac{1}{2} \frac{\partial C^w}{\partial F_{mK}} F^{-1}_{Lm}.
\end{aligned}
\tag{3.25}
$$

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

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

$$
\mathbf{S} = G_0 \left(\frac{\lambda_{\mathrm{L}}}{3\lambda_c} \beta_c\right) \mathbb{1} - \left[(\phi_0^p)^{2/3} G_0 \left(\frac{\lambda_{\mathrm{L}}}{3}\right) - \phi_0^p J^s \kappa (\ln J^e)\right] \mathbf{C}^{-1}, \tag{3.26}
$$

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

$$
\mathbf{S} = G_0 \mathbb{1} - \left[(\phi_0^p)^{2/3} G_0 - \phi_0^p J^s \kappa (\ln J^e)\right] \mathbf{C}^{-1}, \quad \text{where,} \quad J^s = \frac{1}{\phi_0^p} (C^P \mathcal{V}^p + C^w \mathcal{V}^w) \tag{3.27}
$$

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

$$\begin{aligned} \frac{\partial \mathbf{S}}{\partial C^w} &= \kappa \mathcal{V}^w [\ln(J^e) - 1]\mathbf{C}^{-1}, \\ \Rightarrow \left(\frac{\partial \mathbf{S}}{\partial C^w}\right)_{IJ} &= \kappa \mathcal{V}^w [\ln(J^e) - 1] C^{-1}_{IJ}. \end{aligned} \tag{3.28}$$

3.10 Calculation of $\mathbb{C}$

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

$$\mathbb{C}_{IJKL} = 2\left(\frac{\partial S_{IJ}}{\partial C_{KL}} + \frac{\partial S_{IJ}}{\partial C^w}\frac{\partial C^w}{\partial C_{KL}}\right) \qquad \Rightarrow \qquad \mathbb{C} = \mathbb{C}^{\text{mech}} + \mathbb{C}^{\text{chem}}. \tag{3.29}$$

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

$$\begin{aligned} \frac{\partial \mathbf{F}}{\partial \mathbf{F}} &= \mathbb{I} \quad \Rightarrow (\mathbb{I})_{ijkl} = \delta_{ik}\delta_{jl}, \\ \frac{\partial \ln J}{\partial \mathbf{F}} &= \frac{1}{J}\det(\mathbf{F})\mathbf{F}^{-\top} = \mathbf{F}^{-\top}, \quad \Rightarrow \frac{\partial \ln J}{\partial F_{kL}} = F^{-1}_{Lk}, \\ \left(\frac{\partial \mathbf{F}^{-1}}{\partial \mathbf{F}}\right)_{IjkL} &= -F^{-1}_{Ik}F^{-1}_{Lj}, \quad \left(\frac{\partial \mathbf{F}^{-\top}}{\partial \mathbf{F}}\right)_{iJkL} = -F^{-1}_{Li}F^{-1}_{Jk}, \\ \left(\frac{\partial \mathbf{C}^{-1}}{\partial \mathbf{C}}\right)_{IJKL} &= -\frac{1}{2}\left(C^{-1}_{IK}C^{-1}_{JL} + C^{-1}_{JK}C^{-1}_{IL}\right) = \mathbb{I}_{\mathbf{C}^{-1}}, \qquad [\text{since, } \mathbf{C} = \mathbf{C}^\top] \end{aligned} \tag{3.30}$$

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

$$\mathbf{S} = G_0\left(\frac{\lambda_{\mathrm{L}}}{3\lambda_c}\beta_c\right)\mathbb{1} - \left[(\phi_0^p)^{2/3}G_0\left(\frac{\lambda_{\mathrm{L}}}{3}\right) - \phi_0^p J^s \kappa(\ln J^e)\right]\mathbf{C}^{-1}, \tag{3.31}$$

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

$$
\begin{aligned}
\mathbb{C}^{\text{mech}} &= 2\frac{\partial \mathbf{S}}{\partial \mathbf{C}} \\
&= \frac{2G_0\lambda_{\mathrm{L}}}{3}\left(\frac{1}{\lambda_c}\frac{\partial \beta_c}{\partial \mathbf{C}} + \beta_c\frac{\partial \lambda_c^{-1}}{\partial \mathbf{C}}\right)\mathbb{1} + 2\phi_0^p J^s \kappa\left(\frac{\partial \ln J^e}{\partial \mathbf{C}}\right)\mathbf{C}^{-1} - 2\left[\frac{G_0\lambda_{\mathrm{L}}}{3} - \phi_0^p J^s\kappa(\ln J^e)\right]\frac{\partial \mathbf{C}^{-1}}{\partial \mathbf{C}}, \\
&= \frac{G_0}{9\lambda_c^2}\left(\frac{\partial \beta_c}{\partial\left(\frac{\lambda_c}{\lambda_{\mathrm{L}}}\right)} - \frac{\lambda_{\mathrm{L}}}{\lambda_c}\beta_c\right)\mathbb{1}\otimes\mathbb{1} + \phi_0^p J^s\kappa\mathbf{C}^{-1}\otimes\mathbf{C}^{-1} - 2\left[\frac{G_0\lambda_{\mathrm{L}}}{3} - \phi_0^p J^s\kappa(\ln J^e)\right]\mathbb{I}_{\mathbf{C}^{-1}}, \\
&\text{where, } \frac{\partial \ln J^e}{\partial \mathbf{C}} = \frac{\partial}{\partial \mathbf{C}}\left[\ln\left(\frac{J\phi^p}{\phi_0^p}\right)\right] = \frac{\partial \ln J}{\partial \mathbf{C}}
\end{aligned}
\tag{3.32}
$$

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

$$
\mathbf{S} = G_0\mathbb{1} - \left[(\phi_0^p)^{2/3}G_0 - \phi_0^p J^s\kappa(\ln J^e)\right]\mathbf{C}^{-1} \tag{3.33}
$$

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

$$
\begin{aligned}
\mathbb{C}^{\text{mech}} &= 2\frac{\partial \mathbf{S}}{\partial \mathbf{C}} = \phi_0^p J^s\kappa\mathbf{C}^{-1}\otimes\mathbf{C}^{-1} - 2\left[(\phi_0^p)^{2/3}G_0 - \phi_0^p J^s\kappa\ln J\right]\mathbb{I}_{\mathbf{C}^{-1}}, \\
\Rightarrow \mathbb{C}^{\text{mech}}_{IJKL} &= \kappa\phi_0^p J^s C^{-1}_{IJ}C^{-1}_{KL} + \left((\phi_0^p)^{2/3}G_0 - \phi_0^p J^s\kappa\ln J^e\right)\left(C^{-1}_{IK}C^{-1}_{JL} + C^{-1}_{JK}C^{-1}_{IL}\right).
\end{aligned}
\tag{3.34}
$$

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

$$
\begin{aligned}
\mathbb{C}^{\text{chem}}_{IJKL} &= 2\frac{\partial S_{IJ}}{\partial C^w}\frac{\partial C^w}{\partial C_{KL}}, \\
&= 2\left(\kappa\mathcal{V}^w[\ln(J^e)-1]C^{-1}_{IJ}\right)\left[\frac{1}{2}\frac{\partial C^w}{\partial F_{mK}}F^{-1}_{Lm}\right], \\
&= \kappa\mathcal{V}^w[\ln(J^e)-1]C^{-1}_{IJ}F^{-1}_{Lm}\frac{\partial C^w}{\partial F_{mK}}
\end{aligned}
\tag{3.35}
$$

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

$$
\begin{aligned}
\mathbb{C}_{IJKL} = \kappa\phi_0^p J^s C^{-1}_{IJ}C^{-1}_{KL} &+ \left((\phi_0^p)^{2/3}G_0 - \phi_0^p J^s\kappa\ln J^e\right)\left(C^{-1}_{IK}C^{-1}_{JL} + C^{-1}_{JK}C^{-1}_{IL}\right) \\
&+ \kappa\mathcal{V}^w[\ln(J^e)-1]C^{-1}_{IJ}F^{-1}_{Lm}\frac{\partial C^w}{\partial F_{mK}}
\end{aligned}
\tag{3.36}
$$

3.11 Calculation of $\mathbb{J}^w$

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

$$
(\mathbb{J}^w)_{IkL} = \frac{\partial J^w_I}{\partial F_{kL}} + \frac{\partial J^w_I}{\partial C^w}\frac{\partial C^w}{\partial F_{kL}} \tag{3.37}
$$

The molar flux for the solvent diffusion is given by,

$$\begin{aligned}
\mathbf{J}^w &= -\mathbf{M}^w \,\mathrm{Grad}\,\mu^w \qquad \Rightarrow \quad J^w_I = -M_{IJ}(\mathrm{Grad}\,\mu^w)_J, \\
\text{where, } \mathbf{M}^w &= \frac{D^w C^w}{R\theta}\mathbf{C}^{-1} \qquad \Rightarrow \quad M_{IJ} = \frac{D^w C^w}{R\theta} C^{-1}_{IJ}.
\end{aligned} \tag{3.38}$$

Now,

$$\frac{\partial \mathbf{J}^w}{\partial \mathbf{F}} = -\frac{\partial \mathbf{C}^{-1}}{\partial \mathbf{F}}\left(\frac{D^w C^w}{R\theta}\,\mathrm{Grad}\,\mu^w\right) - \mathbf{C}^{-1}\frac{D^w C^w}{R\theta}\frac{\partial}{\partial \mathbf{F}}\left(\mathrm{Grad}\,\mu^w\right). \tag{3.39}$$

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

$$\begin{aligned}
&\mathbf{C} = \mathbf{F}^\top \mathbf{F} \quad \Rightarrow \mathbf{C}^{-1} = \mathbf{F}^{-1}\mathbf{F}^{-\top}, \quad \Rightarrow C^{-1}_{IJ} = F^{-1}_{Im}F^{-\top}_{mJ} = F^{-1}_{Im}F^{-1}_{Jm}, \\
&\left(\frac{\partial \mathbf{F}^{-1}}{\partial \mathbf{F}}\right)_{IjkL} = -F^{-1}_{Ik}F^{-1}_{Lj}, \quad \left(\frac{\partial \mathbf{F}^{-\top}}{\partial \mathbf{F}}\right)_{iJkL} = -F^{-1}_{Li}F^{-1}_{Jk}.
\end{aligned} \tag{3.40}$$

Hence,

$$\begin{aligned}
\frac{\partial \mathbf{C}^{-1}}{\partial \mathbf{F}} = \frac{\partial C^{-1}_{IJ}}{\partial F_{kL}} = \frac{\partial}{\partial F_{kL}}\left(F^{-1}_{Im}F^{-1}_{Jm}\right) &= \frac{\partial F^{-1}_{Im}}{\partial F_{kL}}F^{-1}_{Jm} + \frac{\partial F^{-1}_{Jm}}{\partial F_{kL}}F^{-1}_{Im} \\
&= -F^{-1}_{Ik}F^{-1}_{Lm}F^{-1}_{Jm} - F^{-1}_{Jk}F^{-1}_{Lm}F^{-1}_{Im}, \\
&= -F^{-1}_{Ik}C^{-1}_{LJ} - F^{-1}_{Jk}C^{-1}_{LI}.
\end{aligned} \tag{3.41}$$

Thus, I have,

$$\left(\frac{\partial \mathbf{J}^w}{\partial \mathbf{F}}\right)_{IkL} = \frac{D^w C^w}{R\theta}\left[F^{-1}_{Ik}C^{-1}_{LJ} + F^{-1}_{Jk}C^{-1}_{LI}\right](\mathrm{Grad}\,\mu^w)_J. \tag{3.42}$$

The second term will result in,

$$\frac{\partial}{\partial \mathbf{F}}\left(\mathrm{Grad}\,\mu^w\right) = \frac{\partial}{\partial F_{kL}}\left(\frac{\partial \mu^w}{\partial X_J}\right) = \frac{\partial}{\partial X_J}\left(\frac{\partial \mu^w}{\partial F_{kL}}\right) = \frac{\partial}{\partial X_J}\left(\frac{\partial \mu^w}{\frac{\partial x_k}{\partial X_L}}\right) = \delta_{LJ}F^{-1}_{Mk}(\mathrm{Grad}\,\mu^w)_M, \tag{3.43}$$

which leads to,

$$\begin{aligned}
\frac{D^w C^w}{R\theta}\mathbf{C}^{-1}\frac{\mathrm{Grad}\,\mu^w}{\partial \mathbf{F}} &= \frac{D^w C^w}{R\theta}C^{-1}_{JI}\delta_{LJ}F^{-1}_{Mk}(\mathrm{Grad}\,\mu^w)_M, \\
&= \frac{D^w C^w}{R\theta}C^{-1}_{LI}F^{-1}_{Mk}(\mathrm{Grad}\,\mu^w)_M, \\
&= \frac{D^w C^w}{R\theta}C^{-1}_{LI}F^{-1}_{Jk}(\mathrm{Grad}\,\mu^w)_J.
\end{aligned} \tag{3.44}$$

Now, the second term of $\mathbb{J}^w$ can be written as,

$$\frac{\partial J^w_I}{\partial C^w}\frac{\partial C^w}{\partial F_{kL}} = \left[-\frac{D^w}{R\theta}C^{-1}_{IJ}(\mathrm{Grad}\,\mu^w)_J\right]\left(\frac{\partial C^w}{\partial F_{kL}}\right). \tag{3.45}$$

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

$$\mathbb{J}^w_{IkL} = \frac{D^w C^w}{R\theta} F^{-1}_{Ik} C^{-1}_{LJ} (\mathrm{Grad}\,\mu^w)_J - \left[\frac{D^w}{R\theta} C^{-1}_{IJ} (\mathrm{Grad}\,\mu^w)_J\right] \left(\frac{\partial C^w}{\partial F_{kL}}\right). \tag{3.46}$$

3.12 Calculation of $\mathbb{J}^\beta$

Following a similar procedure as the previous step, I can obtain,

$$\mathbb{J}^\beta_{IkL} = \frac{D^\beta C^\beta}{R\theta} \left[F^{-1}_{Ik} C^{-1}_{LJ}\right] (\mathrm{Grad}\,\omega^\beta)_J - \left[\frac{D^\beta}{R\theta} C^{-1}_{IJ} (\mathrm{Grad}\,\omega^\beta)_J\right] \left(\frac{\partial C^\beta}{\partial F_{kL}}\right) \tag{3.47}$$

3.13 Calculation of $\frac{\partial \mathbf{J}^w}{\partial C^w} \frac{\partial C^w}{\partial \mu^w}$

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

$$\mathbf{J}^w = -\mathbf{C}^{-1} \frac{D^w C^w}{R\theta}\,\mathrm{Grad}\,\mu^w \tag{3.48}$$

Thus, I have,

$$\begin{aligned} \frac{\partial \mathbf{J}^w}{\partial C^w} &= -\frac{D^w}{R\theta} \mathbf{C}^{-1}\,\mathrm{Grad}\,\mu^w, \\ \Rightarrow \frac{\partial \mathbf{J}^w}{\partial C^w} \frac{\partial C^w}{\partial \mu^w} &= -\frac{D^w}{R\theta} \mathbf{C}^{-1}\,\mathrm{Grad}\,\mu^w \left(\frac{\partial C^w}{\partial \mu^w}\right), \\ \Rightarrow \left(\frac{\partial \mathbf{J}^w}{\partial C^w}\right)_I \frac{\partial C^w}{\partial \mu^w} &= \left[-\frac{D^w}{R\theta} C^{-1}_{IJ} (\mathrm{Grad}\,\mu^w)_J\right] \left(\frac{\partial C^w}{\partial \mu^w}\right) \end{aligned} \tag{3.49}$$

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

3.14 Calculation of $\frac{\partial \mathbf{J}^w}{\partial C^w} \frac{\partial C^w}{\partial \omega^\beta{}_i}$

Similarly, I can calculate the following where $\frac{\partial C^w}{\partial \omega^{\beta_i}}$ was previously calculated.

$$\left(\frac{\partial \mathbf{J}^w}{\partial C^w}\right)_I \frac{\partial C^w}{\partial \omega^{\beta_i}} = \left[-\frac{D^w}{R\theta} C^{-1}_{IJ} (\mathrm{Grad}\,\mu^w)_J\right] \left(\frac{\partial C^w}{\partial \omega^{\beta_i}}\right) \tag{3.50}$$

3.15 Calculation of $\frac{\partial \mathbf{J}^\beta}{\partial C^\beta{}_i} \frac{\partial C^\beta{}_i}{\partial \mu^w}$

Similarly, I can calculate the following,

$$\left(\frac{\partial \mathbf{J}^{\beta_i}}{\partial C^{\beta_i}}\right)_I \frac{\partial C^{\beta_i}}{\partial \mu^w} = \left[-\frac{D^{\beta_i}}{R\theta} C^{-1}_{IJ} (\mathrm{Grad}\,\omega^{\beta_i})_J\right] \left(\frac{\partial C^{\beta_i}}{\partial \mu^w}\right) \tag{3.51}$$

where, $\frac{\partial C^{\beta_i}}{\partial \mu^w}$ is calculated previously is substituted.

3.16 Calculation of $\frac{\partial \mathbf{J}^{\beta_i}}{\partial C^{\beta_i}} \frac{\partial C^{\beta_i}}{\partial \omega^{\beta_i}}$

Similarly, I can calculate the following,

$$\left(\frac{\partial \mathbf{J}^{\beta_i}}{\partial C^{\beta_i}}\right)_I \frac{\partial C^{\beta_i}}{\partial \omega^{\beta_i}} = \left[-\frac{D^{\beta_i}}{R\theta} C^{-1}_{IJ} (\mathrm{Grad}\, \omega^{\beta_i})_J\right] \left(\frac{\partial C^{\beta_i}}{\partial \omega^{\beta_i}}\right) \tag{3.52}$$

where, $\frac{\partial C^{\beta_i}}{\partial \omega^{\beta_i}}$ is substituted from previously performed calculation.

3.17 Calculation of $\frac{\partial \mathbf{J}^{\beta_i}}{\partial C^{\beta_j}} \frac{\partial C^{\beta_j}}{\partial \omega^{\beta_j}}$

$$\frac{\partial \mathbf{J}^{\beta_i}}{\partial C^{\beta_j}} \frac{\partial C^{\beta_j}}{\partial \omega^{\beta_j}} = 0. \tag{3.53}$$

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