Notes on the numerical implementation of a coupled theory of the chemo-mechanics of PEG-DA hydrogels in FEniCS

Eric M. Stewart and Lallit Anand

Department of Mechanical Engineering

Massachusetts Institute of Technology

77 Massachusetts Avenue

Cambridge, MA 02139

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1 Governing partial differential equations and boundary conditions

pdes1

1. The governing partial differential equation for the mechanical problem in the absence of body forces and neglect of inertia is the force balance

$$\mathrm{Div}\,\mathbf{T}_{\mathrm{R}} = \mathbf{0} \qquad \forall\ \mathbf{X} \in \mathrm{B}\,, \tag{1.1}$$ mechpde1

where the Piola stress $\mathbf{T}_{\mathrm{R}} \in \mathrm{B}$ is given by

$$\mathbf{T}_{\mathrm{R}} = G(\mathbf{F} - \mathbf{F}^{-\top}) + (1 + \Omega c_{\mathrm{R}})\, K \ln\left(\frac{J}{1 + \Omega c_{\mathrm{R}}}\right) \mathbf{F}^{-\top}\,. \tag{1.2}$$ mechpde2

With $\partial \mathrm{B}_{\mathbf{u}}$ and $\partial \mathrm{B}_{\mathbf{t}_{\mathrm{R}}}$ denoting complementary subsurfaces of the boundary $\partial \mathrm{B}$ of the body B, we consider a pair of possible boundary conditions:

$$\begin{aligned} \mathbf{u} &= \bar{\mathbf{u}} \quad \text{on } \partial \mathrm{B}_{\mathbf{u}}, \\ \mathbf{T}_{\mathrm{R}}\mathbf{n}_{\mathrm{R}} &= \bar{\mathbf{t}}_{\mathrm{R}} \quad \text{on } \partial \mathrm{B}_{\mathbf{t}_{\mathrm{R}}}. \end{aligned} \tag{1.3}$$ mechpde2

In the boundary conditions above $\bar{\mathbf{u}}$ and $\bar{\mathbf{t}}_{\mathrm{R}}$ are prescribed displacement and tractions, both functions of $(\mathbf{X}, t)$.

2. The governing partial differential equation for mass balance is

$$\dot{c}_{\mathrm{R}} + \mathrm{Div}\,\mathbf{j}_{\mathrm{R}} = 0, \tag{1.4}$$ chempde1

where the referential mass flux $\mathbf{j}_{\mathrm{R}}$ is given by

$$\mathbf{j}_{\mathrm{R}} = -\mathbf{M}\nabla\mu, \tag{1.5}$$ chempde2

with chemical potential

$$\mu = R\vartheta\left(\ln\left(1 - \frac{1}{1 + \Omega c_{\mathrm{R}}}\right) + \frac{1}{1 + \Omega c_{\mathrm{R}}} + \frac{\chi}{(1 + \Omega c_{\mathrm{R}})^2}\right) - \Omega\, K\, \ln\left(\frac{J}{1 + \Omega c_{\mathrm{R}}}\right)\,, \tag{1.6}$$ chempot1a

and $\mathbf{M}$ the mobility tensor. Here, because of the constitutive assumptions made in Section 3 of Stewart et al. (2022), we have[1]

$$\begin{aligned} &\mathbf{M} = \mathbf{C}^{-1}\left(\frac{Dc_{\mathrm{R}}}{R\vartheta}\right), \quad \text{where} \\ &D = D_0\,\exp\left(-\alpha\frac{\phi}{1-\phi}\right)+\gamma, \quad \text{with} \quad \phi = \frac{1}{1+\Omega c_{\mathrm{R}}}. \end{aligned} \tag{1.7}$$

chempde3

With $\partial\mathrm{B}_\mu$ and $\partial\mathrm{B}_{\mathbf{j}_{\mathrm{R}}}$ denoting complementary subsurfaces of $\partial\mathrm{B}$, we consider a pair of possible boundary conditions:

$$\begin{aligned} \mu &= \bar{\mu} \quad \text{on } \partial\mathrm{B}_\mu, \\ \mathbf{j}_{\mathrm{R}}\cdot\mathbf{n}_{\mathrm{R}} &= \bar{j} \quad \text{on } \partial\mathrm{B}_{\mathbf{j}_{\mathrm{R}}}. \end{aligned} \tag{1.8}$$

chempde3

In the boundary conditions above $\bar{\mu}$ and $\bar{j}$ are *prescribed* chemical potential and normal species flux which are a function of $(\mathbf{X}, t)$.

Standard initial conditions involve the specifications

$$\mathbf{u}(\mathbf{X},0)=\mathbf{u}_0(\mathbf{X}), \qquad \mu(\mathbf{X},0)=\mu_0(\mathbf{X}), \qquad \text{and} \qquad c_{\mathrm{R}}(\mathbf{X},0)=c_{\mathrm{R}0}(\mathbf{X}), \qquad \text{on B}, \tag{1.9}$$

chempde4

with $\mathbf{u}_0$, μ_0 and $c_{\mathrm{R}0}$ prescribed functions of $\mathbf{X}\in\mathrm{B}$.

2 Weak forms of the governing partial differential equations

1. **Mechanical problem**:

   Consider a test field $\mathbf{u}_{\text{test}}(\mathbf{X})$ on B which satisfies $\mathbf{u}_{\text{test}}(\mathbf{X}) = \mathbf{0}$ on $\partial\mathrm{B}_{\mathbf{u}}$. If the balance equation $\mathrm{Div}\,\mathbf{T}_{\mathrm{R}} = \mathbf{0}$ on B and the traction condition $(\bar{\mathbf{t}}_{\mathrm{R}} - \mathbf{T}_{\mathrm{R}}\mathbf{n}_{\mathrm{R}}) = \mathbf{0}$ on $\partial\mathrm{B}_{\mathbf{t}_{\mathrm{R}}}$ are satisfied, then clearly

   $$\int_{\mathrm{B}} (\mathrm{Div}\,\mathbf{T}_{\mathrm{R}})\cdot\mathbf{u}_{\text{test}}\,dv_{\mathrm{R}} + \int_{\partial\mathrm{B}_{\mathbf{t}}} (\bar{\mathbf{t}}_{\mathrm{R}} - \mathbf{T}_{\mathrm{R}}\mathbf{n}_{\mathrm{R}})\cdot\mathbf{u}_{\text{test}}\,da_{\mathrm{R}} = 0\,, \tag{2.1}$$

   ntemp000

   or equivalently

   $$\int_{\mathrm{B}} (\mathrm{Div}\,\mathbf{T}_{\mathrm{R}})\cdot\mathbf{u}_{\text{test}}\,dv_{\mathrm{R}} - \int_{\partial\mathrm{B}_{\mathbf{t}}} \mathbf{T}_{\mathrm{R}}\mathbf{n}_{\mathrm{R}}\cdot\mathbf{u}_{\text{test}}\,da_{\mathrm{R}} + \int_{\partial\mathrm{B}_{\mathbf{t}}} \bar{\mathbf{t}}_{\mathrm{R}}\cdot\mathbf{u}_{\text{test}}\,da_{\mathrm{R}} = 0 \tag{2.2}$$

   ntemp00

   holds or all admissible test functions $\mathbf{u}_{\text{test}}$. Thus, using $\mathbf{u}_{\text{test}} = \mathbf{0}$ on $\partial\mathrm{B}_{\mathbf{u}}$ we may extend the first boundary integral in the equation above to the whole boundary $\partial\mathrm{B}$, and we may rewrite this equation as

   $$\int_{\partial\mathrm{B}_{\mathbf{t}}} \bar{\mathbf{t}}_{\mathrm{R}}\cdot\mathbf{u}_{\text{test}}\,da_{\mathrm{R}} = \int_{\partial\mathrm{B}} \mathbf{T}_{\mathrm{R}}\mathbf{n}_{\mathrm{R}}\cdot\mathbf{u}_{\text{test}}\,da_{\mathrm{R}} - \int_{\mathrm{B}} \mathrm{Div}\,\mathbf{T}_{\mathrm{R}}\cdot\mathbf{u}_{\text{test}}\,dv_{\mathrm{R}}\,. \tag{2.3}$$

   ntemp1

   Next, by the divergence theorem

   $$\int_{\partial\mathrm{B}} \mathbf{T}_{\mathrm{R}}\mathbf{n}_{\mathrm{R}}\cdot\mathbf{u}_{\text{test}}\,da_{\mathrm{R}} = \int_{\partial\mathrm{B}} (\mathbf{T}_{\mathrm{R}}^{\top}\mathbf{u}_{\text{test}})\cdot\mathbf{n}\,da_{\mathrm{R}} = \int_{\mathrm{B}} \mathrm{Div}\,\mathbf{T}_{\mathrm{R}}\cdot\mathbf{u}_{\text{test}}\,dv_{\mathrm{R}} + \int_{\mathrm{B}} \mathbf{T}_{\mathrm{R}}:\nabla\mathbf{u}_{\text{test}}\,dv_{\mathrm{R}}\,, \tag{2.4}$$

   ntemp1a

   use of which in (2.3) gives

   $$\int_{\partial\mathrm{B}_{\mathbf{t}}} \bar{\mathbf{t}}_{\mathrm{R}}\cdot\mathbf{u}_{\text{test}}\,da_{\mathrm{R}} = \int_{\mathrm{B}} \mathbf{T}_{\mathrm{R}}:\nabla\mathbf{u}_{\text{test}}\,dv_{\mathrm{R}}\,. \tag{2.5}$$

   ntemp3

[1] We introduce a small numerical factor $\gamma = 3\times10^{-4}$ which prevents zero-valued diffusivity as $\phi\to1$.

Hence, if the balance equation $\text{Div}\,\mathbf{T}_{\text{R}} = \mathbf{0}$ on B and the traction condition $(\bar{\mathbf{t}}_{\text{R}} - \mathbf{T}_{\text{R}}\mathbf{n}_{\text{R}}) = \mathbf{0}$ on $\partial\text{B}_{\mathbf{t}_{\text{R}}}$ are satisfied, then the weak form

$$\int_{\text{B}} \mathbf{T}_{\text{R}} : \nabla \mathbf{u}_{\text{test}}\, dv_{\text{R}} - \int_{\partial\text{B}_{\mathbf{t}}} \bar{\mathbf{t}}_{\text{R}} \cdot \mathbf{u}_{\text{test}}\, da_{\text{R}} = 0 \tag{2.6}$$ virpow1

holds for all admissible test fields $\mathbf{u}_{\text{test}}$ which vanish on $\partial\text{B}_{\mathbf{u}}$.

2. **Mass balance problem**:

Next, consider a scalar test field $\mu_{\text{test}}(\mathbf{X})$ on B which satisfies $\mu_{\text{test}}(\mathbf{X}) = \mathbf{0}$ on ∂B_{μ}. If the balance equation

$$\dot{c}_{\text{R}} + \text{Div}\,\mathbf{j}_{\text{R}} = 0 \qquad \text{on B},$$

and the boundary condition $\mathbf{j}_{\text{R}} \cdot \mathbf{n} = \bar{j}$ on $\partial\text{B}_{\mathbf{j}_{\text{R}}}$ are satisfied, then

$$\int_{\text{B}} \dot{c}_{\text{R}}\, \mu_{\text{test}}\, dv_{\text{R}} + \int_{\text{B}} (\text{Div}\mathbf{j}_{\text{R}})\, \mu_{\text{test}}\, dv_{\text{R}} + \int_{\partial\text{B}_{\mathbf{j}}} (\bar{j} - \mathbf{j}_{\text{R}} \cdot \mathbf{n})\ \mu_{\text{test}}\ da_{\text{R}} = 0\,, \tag{2.7}$$ nmtemp1aa

or equivalantly

$$\int_{\text{B}} \dot{c}_{\text{R}}\, \mu_{\text{test}}\, dv_{\text{R}} + \int_{\text{B}} (\text{Div}\mathbf{j}_{\text{R}})\, \mu_{\text{test}}\, dv_{\text{R}} - \int_{\partial\text{B}_{\mathbf{j}}} \mathbf{j}_{\text{R}} \cdot \mathbf{n}_{\text{R}}\, \mu_{\text{test}}\ da_{\text{R}} + \int_{\partial\text{B}_{\mathbf{j}}} \bar{j}\, \mu_{\text{test}}\ da_{\text{R}} = 0 \tag{2.8}$$ nmtemp1a

holds for all test fields μ_{test}. Thus, since $\mu_{\text{test}} = 0$ on ∂B_{μ} the first boundary integral in expression above may be extended to all of ∂B,

$$\int_{\text{B}} \dot{c}_{\text{R}}\, \mu_{\text{test}}\, dv_{\text{R}} + \int_{\text{B}} (\text{Div}\mathbf{j}_{\text{R}})\, \mu_{\text{test}}\, dv_{\text{R}} - \int_{\partial\text{B}} \mathbf{j}_{\text{R}} \cdot \mathbf{n}_{\text{R}}\, \mu_{\text{test}}\ da_{\text{R}} + \int_{\partial\text{B}_{\mathbf{j}}} \bar{j}\, \mu_{\text{test}}\ da_{\text{R}} = 0 \tag{2.9}$$ nmtemp1b

Next, by the divergence theorem

$$\int_{\partial\text{B}} \mathbf{j}_{\text{R}} \cdot \mathbf{n}_{\text{R}}\, \mu_{\text{test}}\, da_{\text{R}} = \int_{\partial\text{B}} (\mu_{\text{test}}\, \mathbf{j}_{\text{R}}) \cdot \mathbf{n}\, da_{\text{R}} = \int_{\text{B}} \mu_{\text{test}}\, \text{Div}\mathbf{j}_{\text{R}}\, dv_{\text{R}} + \int_{\text{B}} \mathbf{j}_{\text{R}} \cdot \nabla\mu_{\text{test}}\, dv_{\text{R}}\,. \tag{2.10}$$ nmtemp1a

Use of (2.10) in (2.9) gives the weak form

$$\int_{\text{B}} \dot{c}_{\text{R}}\, \mu_{\text{test}}\, dv_{\text{R}} - \int_{\text{B}} \mathbf{j}_{\text{R}} \cdot \nabla\mu_{\text{test}}\, dv_{\text{R}} + \int_{\partial\text{B}_{\mathbf{j}}} \bar{j}\, \mu_{\text{test}}\ da_{\text{R}} = 0\,. \tag{2.11}$$ nmtemp1bb

We use a first-order finite difference approximation to write

$$\dot{c}_{\text{R}} = \frac{c_{\text{R}} - c_{\text{R}\,\text{old}}}{\Delta t}, \tag{2.12}$$ nmtemp1c

so that the weak form (2.11) may be expressed as

$$\int_{\text{B}} \frac{c_{\text{R}} - c_{\text{R}\,\text{old}}}{\Delta t}\, \mu_{\text{test}}\, dv_{\text{R}} - \int_{\text{B}} \mathbf{j}_{\text{R}} \cdot \nabla\mu_{\text{test}}\, dv_{\text{R}} + \int_{\partial\text{B}_{\mathbf{j}}} \bar{j}\, \mu_{\text{test}}\ da_{\text{R}} = 0\,. \tag{2.13}$$ nmtemp1bbb

Summarizing, the weak forms of the governing equations for the coupled problem are

$$\boxed{\begin{aligned} \int_{\mathrm{B}} \mathbf{T}_{\mathrm{R}} : \nabla \mathbf{u}_{\mathrm{test}} \, dv_{\mathrm{R}} - \int_{\partial \mathrm{B}_{\mathbf{t}}} \bar{\mathbf{t}}_{\mathrm{R}} \cdot \mathbf{u}_{\mathrm{test}} \, da_{\mathrm{R}} = 0 \,, \\ \int_{\mathrm{B}} \frac{c_{\mathrm{R}} - c_{\mathrm{R}\,\mathrm{old}}}{\Delta t} \, \mu_{\mathrm{test}} \, dv_{\mathrm{R}} - \int_{\mathrm{B}} \mathbf{j}_{\mathrm{R}} \cdot \nabla \mu_{\mathrm{test}} \, dv_{\mathrm{R}} + \int_{\partial \mathrm{B}_{\mathbf{j}}} \bar{j} \, \mu_{\mathrm{test}} \, da_{\mathrm{R}} = 0 \,, \end{aligned}} \tag{2.14}$$

nweakforms

for all vector-valued test functions $\mathbf{u}_{\mathrm{test}}(\mathbf{X})$ which vanish on $\partial \mathrm{B}_{\mathbf{u}}$ and all scalar test functions $\mu_{\mathrm{test}}(\mathbf{X})$ which vanish on $\partial \mathrm{B}_{\mu}$.

Remarks:

1. As phrased above, the basic degrees of freedom in the boundary value problem are the displacement field and the chemical potential,

   $$\{\mathbf{u}, \mu\} \,.$$

   However, as is clear from $(2.14)_2$, we also need knowledge of the species concentration c_{R} to obtain a solution. Given $(\mathbf{u}, \mu)$ the concentration c_{R} may be calculated from the expression for the chemical potential

   $$\mu = R\vartheta \left(\ln \left(1 - \frac{1}{1 + \Omega c_{\mathrm{R}}} \right) + \frac{1}{1 + \Omega c_{\mathrm{R}}} + \frac{\chi}{(1 + \Omega c_{\mathrm{R}})^2} \right) - \Omega \, K \, \ln \left(\frac{J}{1 + \Omega c_{\mathrm{R}}} \right) \,, \tag{2.15}$$

   chempot1

   which, with $J = J(\mathbf{u})$ and μ known, may be written as an implicit equation for c_{R},

   $$\frac{\mu}{R\vartheta} - R\vartheta \left(\ln \left(1 - \frac{1}{1 + \Omega c_{\mathrm{R}}} \right) + \frac{1}{1 + \Omega c_{\mathrm{R}}} + \frac{\chi}{(1 + \Omega c_{\mathrm{R}})^2} \right) + \frac{\Omega \, K}{R\vartheta} \, \ln \left(\frac{J}{1 + \Omega c_{\mathrm{R}}} \right) = 0 \tag{2.16}$$

   chempot2

   On account of the inability of the FEniCS framework to solve nonlinear equations at the Gauss-point level, we consider solving equation (2.16) in a *weak sense* by introducing a test function c_{test},

   $$\int_{\mathrm{B}} \left[\frac{\mu}{R\vartheta} - R\vartheta \left(\ln \left(1 - \frac{1}{1 + \Omega c_{\mathrm{R}}} \right) + \frac{1}{1 + \Omega c_{\mathrm{R}}} + \frac{\chi}{(1 + \Omega c_{\mathrm{R}})^2} \right) + \frac{\Omega \, K}{R\vartheta} \, \ln \left(\frac{J}{1 + \Omega c_{\mathrm{R}}} \right) \right] \, c_{\mathrm{test}} = 0 \,. \tag{2.17}$$

   chempot3

2. Summarizing, the complete set of weak forms of the coupled problem are

   $$\boxed{\begin{aligned} \int_{\mathrm{B}} \mathbf{T}_{\mathrm{R}} : \nabla \mathbf{u}_{\mathrm{test}} \, dv_{\mathrm{R}} - \int_{\partial \mathrm{B}_{\mathbf{t}}} \bar{\mathbf{t}}_{\mathrm{R}} \cdot \mathbf{u}_{\mathrm{test}} \, da_{\mathrm{R}} = 0 \,, \\ \int_{\mathrm{B}} \frac{c_{\mathrm{R}} - c_{\mathrm{R}\,\mathrm{old}}}{\Delta t} \, \mu_{\mathrm{test}} \, dv_{\mathrm{R}} - \int_{\mathrm{B}} \mathbf{j}_{\mathrm{R}} \cdot \nabla \mu_{\mathrm{test}} \, dv_{\mathrm{R}} + \int_{\partial \mathrm{B}_{\mathbf{j}}} \bar{j} \, \mu_{\mathrm{test}} \, da_{\mathrm{R}} = 0 \,, \\ \int_{\mathrm{B}} \left[\frac{\mu}{R\vartheta} - \left(\ln \left(1 - \frac{1}{1 + \Omega c_{\mathrm{R}}} \right) + \frac{1}{1 + \Omega c_{\mathrm{R}}} + \frac{\chi}{(1 + \Omega c_{\mathrm{R}})^2} \right) + \frac{\Omega \, K}{R\vartheta} \, \ln \left(\frac{J}{1 + \Omega c_{\mathrm{R}}} \right) \right] \, c_{\mathrm{test}} \, dv_{\mathrm{R}} = 0 \,. \end{aligned}} \tag{2.18}$$

   totalweak1

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3 Normalization

Since we plan to solve all the governing equations related to multiple physical fields simultaneously, it is beneficial to rewrite the three residuals so that they have approximately the same magnitudes, and to use degrees of freedom which take on values on the order of unity. To this end:

1. The species concentration c_{R} mol/m^3 is normalized by the molar volume Ω [m^3/mol] as

$$\hat{c} \stackrel{\text{def}}{=} \Omega c_{\mathrm{R}} \,. \tag{3.1}$$ li6

2. The chemical potential μ [J/mol] is normalized by $R\vartheta$ [J/mol],

$$\hat{\mu} = \frac{\mu}{R\vartheta} \,. \tag{3.2}$$ li7

3. Recall the mass balance equation:

$$\dot{c}_{\mathrm{R}} + \mathrm{Div}\,\mathbf{j}_{\mathrm{R}} = 0.$$

   This may be written as,

$$\frac{1}{\Omega}\dot{\hat{c}} + \mathrm{Div}\,\mathbf{j}_{\mathrm{R}} = 0 \,.$$

4. Next, the species flux $\mathbf{j}_{\mathrm{R}}$ is given by the constitutive equation,

$$\mathbf{j}_{\mathrm{R}} = -\mathbf{M}\nabla\mu \qquad \text{with} \qquad \mathbf{M} = \mathbf{C}^{-1}\left(\frac{Dc_{\mathrm{R}}}{R\vartheta}\right) \,. \tag{3.3}$$ li10

   Using the normalized variables $\hat{\mu}$ and $\hat{c}$ we may rewrite it as follows:

$$\mathbf{j}_{\mathrm{R}} = -\mathbf{M}\nabla\hat{\mu} \times R\vartheta \qquad \text{with} \qquad \mathbf{M} = \mathbf{C}^{-1}\left(\frac{D\hat{c}}{\Omega R\vartheta}\right) \,, \tag{3.4}$$ li10a

   where D is given by

$$D = D_0 \exp\left(-\alpha\frac{\phi}{1-\phi}\right) + \gamma, \quad \text{with} \quad \phi = \frac{1}{1+\hat{c}} \,. \tag{3.5}$$ li10b

5. Finally we choose to normalize the stress with the shear modulus G:

$$\hat{\mathbf{T}}_{\mathrm{R}} \stackrel{\text{def}}{=} \frac{\mathbf{T}_{\mathrm{R}}}{G} = (\mathbf{F} - \mathbf{F}^{-\top}) + (1+\hat{c})\,\frac{K}{G}\,\ln\left(\frac{J}{1+\hat{c}}\right)\mathbf{F}^{-\top} \,. \tag{3.6}$$ li13

6. Summarizing the normalized governing equations may then be expressed as

$$\begin{aligned} \mathrm{Div}\,\hat{\mathbf{T}}_{\mathrm{R}} &= 0 \\ \dot{\hat{c}}\,\Omega^{-1} + \mathrm{Div}\,\mathbf{j}_{\mathrm{R}} &= 0 \,. \end{aligned} \tag{3.7}$$ li14

With the normalizations introduced above and the normalized parameters denoted by $\{\hat{\bullet}\}$, the weak forms of the coupled problem may be written as

$$\begin{aligned} &\int_{\mathrm{B}} \hat{\mathbf{T}}_{\mathrm{R}} : \nabla\mathbf{u}_{\mathrm{test}}\, dv_{\mathrm{R}} - \int_{\partial\mathrm{B}_{\mathbf{t}}} \bar{\hat{\mathbf{t}}}_{\mathrm{R}} \cdot \mathbf{u}_{\mathrm{test}}\, da_{\mathrm{R}} = 0 \,, \\ &\int_{\mathrm{B}} \frac{\hat{c} - \hat{c}_{\mathrm{old}}}{\Delta t}\,\hat{\mu}_{\mathrm{test}}\, dv_{\mathrm{R}} - \int_{\mathrm{B}} \Omega\,\mathbf{j}_{\mathrm{R}} \cdot \nabla\hat{\mu}_{\mathrm{test}}\, dv_{\mathrm{R}} + \int_{\partial\mathrm{B}_{\mathbf{j}}} \Omega\,\bar{j}\,\hat{\mu}_{\mathrm{test}}\; da_{\mathrm{R}} = 0 \,, \\ &\int_{\mathrm{B}} \left[\hat{\mu} - \left(\ln\left(1 - \frac{1}{1+\hat{c}}\right) + \frac{1}{1+\hat{c}} + \frac{\chi}{(1+\hat{c})^2}\right) + \frac{\Omega\,K}{R\vartheta}\,\ln\left(\frac{J}{1+\hat{c}}\right)\right]\,\hat{c}_{\mathrm{test}} = 0 \,. \end{aligned} \tag{3.8}$$ totalweak2

Remark. Note that in writing the second of the weak forms above we have multiplied through by Ω. This helps in the numerical implementation.

4 Solution procedure in the FEniCS code

In brief, in FEniCS, we use the following degrees of freedom

$$\big\{\mathbf{u},\, \hat{\mu},\, \hat{c}\big\}, \tag{4.1}$$

as well as their known values from the previous step

$$\big\{\mathbf{u}_{\text{old}},\, \hat{\mu}_{\text{old}},\, \hat{c}_{\text{old}}\big\}, \tag{4.2}$$

to calculate the *residuals* which correspond to the weak forms of the governing equations, viz.

$$\begin{aligned}
R_{\mathbf{u}} &= \int_{\text{B}} \hat{\mathbf{T}}_{\text{R}} : \nabla \mathbf{u}_{\text{test}}\, dv_{\text{R}} - \int_{\partial \text{B}_{\mathbf{t}}} \bar{\hat{\mathbf{t}}}_{\text{R}} \cdot \mathbf{u}_{\text{test}}\, da_{\text{R}}\,, \\
R_{\hat{\mu}} &= \int_{\text{B}} \left(\frac{\hat{c} - \hat{c}_{\text{old}}}{\Delta t} \right) \hat{\mu}_{\text{test}}\, dv_{\text{R}} - \int_{\text{B}} \Omega\, \mathbf{j}_{\text{R}} \cdot \nabla \hat{\mu}_{\text{test}}\, dv_{\text{R}} + \int_{\partial \text{B}_{\mathbf{j}}} \Omega\, \bar{j}\, \hat{\mu}_{\text{test}}\; da_{\text{R}}\,, \\
R_{\hat{c}} &= \int_{\text{B}} \left[\hat{\mu} - \left(\ln\left(1 - \frac{1}{1+\hat{c}}\right) + \frac{1}{1+\hat{c}} + \frac{\chi}{(1+\hat{c})^2} \right) + \frac{\Omega K}{R\vartheta}\, \ln\left(\frac{\hat{J}}{1+\hat{c}} \right) \right] \hat{c}_{\text{test}}\; da_{\text{R}}\,.
\end{aligned} \tag{4.3}$$

res1

FEniCS then automatically differentiates these residuals with respect to the separate degrees of freedom to obtain *tangents*. At each step in time, FEniCS uses the residuals and tangents in a global Newton-Raphson solver to determine current values of the degrees of freedom $\{\mathbf{u},\, \hat{\mu},\, \hat{c}\}$ for which the residuals are zero,[2] and which therefore satisfy the governing equations as well as the initial and boundary conditions (in a weak sense).

Remark. A mixed finite element method is utilized to solve for the displacement $\mathbf{u}$, chemical potential $\hat{\mu}$, and the concentration field $\hat{c}$, concurrently. To avoid numerical instabilities the basis functions for the displacement are taken as Lagrange quadratic, while those of the chemical potential are one-order lower, Lagrange linear. The field $\hat{c}$ is also interpolated as a linear Lagrange field.

[2] To within some small numerical tolerance.

4.1 Algorithm for calculation of the residuals

The algorithm we follow in our code to calculate the residuals in (4.3) is summarized below.

1. Begin the step with known values of
$$\left\{\mathbf{u}_{\text{old}},\, \hat{\mu}_{\text{old}},\, \hat{c}_{\text{old}}\right\}, \tag{4.4}$$
where for the first step these quantities are given by the initial conditions for $\mathbf{u}$, $\hat{\mu}$ and $\hat{c}_0$.

2. Calculate necessary kinematical quantities for the current step as,
$$\begin{aligned} &\mathbf{F} = \mathbf{1} + \nabla\,\mathbf{u}\,, \\ &J = \det\mathbf{F}, \\ &\mathbf{C}^{-1} = (\mathbf{F}^{\top}\mathbf{F})^{-1}\,. \end{aligned} \tag{4.5}$$

3. Calculate J^e,
$$J^e = \frac{J}{1+\hat{c}}\,. \tag{4.6}$$

4. Calculate the Piola stress and associated pressure
$$\hat{\mathbf{T}}_{\text{R}} = (\mathbf{F} - \mathbf{F}^{-\top}) + (1+\hat{c})\frac{K}{G}\ln(J^e)\,\mathbf{F}^{-\top}\,, \tag{4.7}$$
and
$$p = -\frac{1}{3}\,\text{tr}\left(G\,\hat{\mathbf{T}}_{\text{R}}\mathbf{F}^{\top}J^{-1}\right). \tag{4.8}$$

5. Calculate the species flux,
$$\mathbf{j}_{\text{R}} = -\mathbf{M}\nabla\hat{\mu} \times R\vartheta \qquad \text{with} \qquad \mathbf{M} = \mathbf{C}^{-1}\left(\frac{D\hat{c}}{\Omega R\vartheta}\right), \tag{4.9}$$
where D is given by
$$D = D_0\,\exp\left(-\alpha\frac{\phi}{1-\phi}\right) + \gamma, \quad \text{with} \quad \phi = \frac{1}{1+\hat{c}}\,. \tag{4.10}$$

6. Use the quantities from steps 1-5 to compute the residuals as
$$\begin{aligned} R_{\mathbf{u}} &= \int_{\text{B}} \hat{\mathbf{T}}_{\text{R}} : \nabla\mathbf{u}_{\text{test}}\, dv_{\text{R}} - \int_{\partial\text{B}_{\mathbf{t}}} \bar{\hat{\mathbf{t}}}_{\text{R}} \cdot \mathbf{u}_{\text{test}}\, da_{\text{R}}\,, \\ R_{\hat{\mu}} &= \int_{\text{B}} \left(\frac{\hat{c}-\hat{c}_{\text{old}}}{\Delta t}\right)\hat{\mu}_{\text{test}}\, dv_{\text{R}} - \int_{\text{B}} \Omega\,\mathbf{j}_{\text{R}} \cdot \nabla\hat{\mu}_{\text{test}}\, dv_{\text{R}} + \int_{\partial\text{B}_{\mathbf{j}}} \Omega\,\bar{j}\,\hat{\mu}_{\text{test}}\; da_{\text{R}}\,, \\ R_{\hat{c}} &= \int_{\text{B}} \left[\hat{\mu} - \left(\ln\left(1-\frac{1}{1+\hat{c}}\right) + \frac{1}{1+\hat{c}} + \frac{\chi(p)}{(1+\hat{c})^2}\right) + \frac{\Omega\,K}{R\vartheta}\,\ln\left(\frac{\hat{J}}{1+\hat{c}}\right)\right]\hat{c}_{\text{test}}\; da_{\text{R}}\,, \end{aligned} \tag{4.11}$$
res1b

including the pressure-dependence of the Flory-Huggins parameter,
$$\chi(p) = \chi_0 + \beta\,p. \tag{4.12}$$

7. FEniCS solves for the current values of $\{\mathbf{u},\, \hat{\mu},\, \hat{c}\}$.

8. Set these current values as the "old" values for the next step in time,
$$\left\{\mathbf{u},\, \hat{\mu},\, \hat{c}\right\} \implies \left\{\mathbf{u}_{\text{old}},\, \hat{\mu}_{\text{old}},\, \hat{c}_{\text{old}}\right\}. \tag{4.13}$$

9. Repeat steps 1-8 for desired number of steps in time.

5 Unit system and material parameters in SI (mm) units

Quantity	SI (mm)
Length	mm
Mass	tonne (10^3 kg)
Time	s
Force	N
Energy	mJ (10^{-3}J)
Mass Density	tonne/mm^3
Amount of substance	mol
c_{R}, species concentration	mol/mm^3
μ, Chemical potential	mJ/mol
G and K, Stress	MPa (N/mm^2)
Ω, molar volume	mm^3/mol
D species Diffusivity	mm^2/s
Gas Constant R	8.3145×10^3 mJ/(mol K)
Temperature	K

Quantity	Value SI (mm)
G Shear modulus	1 MPa
K, Bulk modulus	100 MPa
Ω, Molar volume of water	1.8×10^4 mm^3/mol
D_0, Solvent diffusivity when $\phi = 0$	2 mm^2/s
α, Yasuda form shape parameter	7.7
χ_0, Zero-pressure Flory-Huggins parameter	0.1
β, Flory-Huggins pressure-dependence parameter	1.9 $\times 10^{-1}$ MPa^{-1}
ϕ_0, Initial polymer volume fraction	0.999
Gas Constant R	8.3145×10^3 mJ/(mol K)
Temperature ϑ	298 K

References

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