Details on the numerical implementation of Stewart et al. (2022) in FEniCS

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In this supplementary document, details are provided for the numerical implementation of the theory in Stewart et al. (2022) in FEniCS. The notation and form of the equations here mirror the FEniCS code as closely as possible. If you use this in constructing your own code, please cite:

- E. M. Stewart, S. Narayan, and L. Anand. On modeling the infiltration of water in a PEG-DA hydrogel and the resulting swelling under unconstrained and mechanically-constrained conditions. Extreme Mechanics Letters, 54:101775, July 2022.

1 Unit system

FEniCS does not track units for physical quantities intrinsically, therefore we are free to select the most suitable system of units for the problem at hand. Here, we select the following base units

$$\text{Length: mm}, \quad \text{Time: s}, \quad \text{Mass: kg}, \quad \text{Moles: n-mol},$$

which implies derived units of

$$\text{Pressure: kPa}, \quad \text{Force: mN}, \quad \text{Energy: } \mu\text{J}.$$

The choice of millimeters follows naturally from the scale of the problems in this work, while the use of nano-moles is convenient in that 1 $\text{m}^3/\text{mol} = 1\ \text{mm}^3/\text{n-mol}$. The PEG-DA gel's material parameters and relevant physical constants are provided in Table 1 in the unit system of the FEniCS implementation.

Table 1: Physical constants and material parameters in the unit system of the numerical implementation.

Parameter	Value	Units
G	1×10^3	kPa
K	10×10^3	kPa
Ω	1.8×10^{-5}	$\text{mm}^3/\text{n-mol}$
D_0	2.0	mm^2/s
α	7.7	-
χ_0	0.52	-
β	1.9×10^{-4}	kPa^{-1}
ϑ	298	K
R	8.3145×10^{-3}	$\mu\text{J}/(\text{n-mol K})$

2 Degrees of freedom

We use a modified form of the mixed displacement-pressure $\{\mathbf{u}, \mathcal{P}\}$ finite element formulation, which is widely used for modeling nearly-incompressible materials. Specifically, we select the list of degrees of freedom

$$\big\{\mathbf{u},\, \mu,\, \mathcal{P}\big\}, \tag{2.1}$$

with $\mathbf{u}$ the vectorial displacement, μ the chemical potential, and $\mathcal{P}$ a pressure-like field. Guided by the form of the chemical potential, we select

$$\mathcal{P} \stackrel{\text{def}}{=} K \ln(J^e), \tag{2.2}$$

so that

$$\mu = R\vartheta\Big(\ln(1-\phi) + \phi + \chi\phi^2\Big) - \Omega\mathcal{P}\,. \tag{2.3}$$

2.1 Normalization of the degrees of freeedom

It is numerically advantageous for all degrees of freedom to take on values on the order of unity. For this reason, our numerical implementation actually uses a *normalized* list of degrees of freedom

$$\big\{\mathbf{u},\, \hat{\mu},\, \hat{\mathcal{P}}\big\}, \tag{2.4}$$

with normalization factors

$$\hat{\mu} = \frac{\mu}{R\vartheta}, \qquad \text{and} \qquad \hat{\mathcal{P}} = \frac{\mathcal{P}}{K}. \tag{2.5}$$

Note that since the base unit of length is millimeters, the displacement field $\mathbf{u}$ is already on the order of unity. We will use the normalized list of degrees of freedom $\{\mathbf{u},\, \hat{\mu},\, \hat{\mathcal{P}}\}$ in all discussion moving forward.

2.2 Basis functions for the degrees of freedom

Following a Taylor-Hood style of element, we interpolate the vectorial displacement degree of freedom $\mathbf{u}$ using quadratic Lagrangian basis functions, while we interpolate the normalized chemical potential $\hat{\mu}$ and the normalized pressure-like field $\hat{\mathcal{P}}$ using linear Lagrangian basis functions.

3 Governing equations

We solve weak forms of the governing equations of the theory in the reference configuration[1]. To derive weak forms of the governing equations, we introduce test functions for each degree of freedom as the list

$$\big\{\mathbf{u}_{\text{test}},\, \hat{\mu}_{\text{test}},\, \hat{\mathcal{P}}_{\text{test}}\big\}. \tag{3.1}$$

Multiplying a governing equation by one of these test fields and integrating over the entire domain yields the weak form of that equation[2]. Our implementation uses three weak forms, which we now describe in detail.

3.1 The force balance

The force balance can be expressed in the reference body as

$$\operatorname{Div}\mathbf{T}_{\text{R}} = \mathbf{0}, \tag{3.2}$$

with $\mathbf{T}_{\text{R}}$ the Piola stress. Using $\mathbf{T}_{\text{R}} = J\,\mathbf{T}\,\mathbf{F}^{-\top}$ and the Neo-Hookean Cauchy stress in (**??**), we find

$$\mathbf{T}_{\text{R}} = G(\mathbf{F} - \mathbf{F}^{-\top}) + J^s\,K\ln(J^e)\mathbf{F}^{-\top}. \tag{3.3}$$

Since $\phi = (J^s)^{-1}$ and $\hat{\mathcal{P}} = \ln(J^e)$, we calculate the Piola stress in the numerical implementation as

$$\mathbf{T}_{\text{R}} = G(\mathbf{F} - \mathbf{F}^{-\top}) + \phi^{-1}K\hat{\mathcal{P}}\mathbf{F}^{-\top}. \tag{3.4}$$

Multiplying through (3.2) by $\mathbf{u}_{\text{test}}$, integrating over the body, and neglecting surface tractions, we have

$$\int_B \mathbf{T}_{\text{R}} : \nabla\mathbf{u}_{\text{test}}\, dv_{\text{R}} = 0, \tag{3.5}$$

[1] Solution of the governing equations in the reference configuration is the default approach for FEniCS.
[2] Often, such weak forms are further simplified through integration by parts.

with the Piola stress $\mathbf{T}_{\mathrm{R}}$ given by (3.4). In practice, it is also useful to normalize the weak forms so that each term takes on values on the order of unity. To this end, in the FEniCS code we simply divide through the force balance weak form by the shear modulus G, viz.

$$\int_B \frac{1}{G}\mathbf{T}_{\mathrm{R}} : \nabla \mathbf{u}_{\mathrm{test}}\, dv_{\mathrm{R}} = 0, \tag{3.6}$$

3.2 The mass balance

The mass balance can be expressed in the reference body as

$$\frac{1}{\Omega}\dot{\overline{\left(\frac{1}{\phi}\right)}} + \mathrm{Div}\, \mathbf{j}_{\mathrm{R}} = 0, \tag{3.7}$$

where the referential mass flux $\mathbf{j}_{\mathrm{R}}$, following a Fick-type relation in the spatial configuration, is

$$\mathbf{j}_{\mathrm{R}} = -\mathbf{M}\nabla\mu, \tag{3.8}$$

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

$$\begin{aligned} \mathbf{M} &= \mathbf{C}^{-1}\left(\frac{Dc_{\mathrm{R}}}{R\vartheta}\right), \quad \text{where} \\ c_{\mathrm{R}} &= \frac{1}{\Omega}\left(\frac{1}{\phi} - 1\right), \quad \text{and} \\ D &= D_0\, \exp\left(-\alpha\frac{\phi}{1-\phi}\right) + \gamma. \end{aligned} \tag{3.9}$$

Multiplying through (3.7) by $\hat{\mu}_{\mathrm{test}}$, integrating over the body, and neglecting surface fluxes, we arrive at the weak form

$$\int_B \frac{1}{\Omega}\dot{\overline{\left(\frac{1}{\phi}\right)}}\,\hat{\mu}_{\mathrm{test}}\, dv_{\mathrm{R}} + \int_B \mathbf{M}\,\nabla\mu\cdot\nabla\hat{\mu}_{\mathrm{test}}\, dv_{\mathrm{R}} = 0. \tag{3.10}$$

We use a first-order finite difference approximation to write

$$\dot{\overline{\left(\frac{1}{\phi}\right)}} = \frac{\phi^{-1} - \phi_{\mathrm{old}}^{-1}}{\Delta t}. \tag{3.11}$$

Then, recalling that $\hat{\mu} = \mu/R\vartheta$ and rearranging constants to obtain terms of order unity, we have the final weak form which appears in the FEniCS code

$$\int_B \left(\phi^{-1} - \phi_{\mathrm{old}}^{-1}\right)\,\hat{\mu}_{\mathrm{test}}\, dv_{\mathrm{R}} + \int_B \Omega\,\Delta t\,\mathbf{M}\,(\nabla\hat{\mu}\,R\vartheta)\cdot\nabla\hat{\mu}_{\mathrm{test}}\, dv_{\mathrm{R}} = 0. \tag{3.12}$$

3.3 The chemical potential

We rearrange the constitutive relation for the chemical potential (2.3) to write

$$\mathcal{P} - \frac{1}{\Omega}\left[R\vartheta\Big(\ln(1-\phi) + \phi + \chi\phi^2\Big) - \mu\right] = 0. \tag{3.13}$$

The corresponding weak form of this equation can be obtained simply as

$$\int_B \left(\mathcal{P} - \frac{1}{\Omega}\left[R\vartheta\Big(\ln(1-\phi) + \phi + \chi\phi^2\Big) - \mu\right]\right)\,\mathcal{P}_{\mathrm{test}}\, dv_{\mathrm{R}} = 0, \tag{3.14}$$

or finally, in terms of the normalized degrees of freedom,

$$\int_B \left(\hat{\mathcal{P}} - \frac{R\vartheta}{K\Omega}\left[\ln(1-\phi) + \phi + \chi\phi^2 - \hat{\mu}\right]\right)\,\mathcal{P}_{\mathrm{test}}\, dv_{\mathrm{R}} = 0. \tag{3.15}$$

In the unit system of our numerical implementation we have $R\vartheta/(K\Omega) \approx 14$, and so the terms of this weak form are on the order of unity.

4 The solution procedure in the FEniCS code

In brief, our task in the FEniCS code is to use the known degrees of freedom

$$\left\{\mathbf{u},\, \hat{\mu},\, \hat{\mathcal{P}}\right\}, \tag{4.1}$$

as well as their known values from the previous step

$$\left\{\mathbf{u}_{\text{old}},\, \hat{\mu}_{\text{old}},\, \hat{\mathcal{P}}_{\text{old}}\right\}, \tag{4.2}$$

to calculate unknown *residuals* which correspond exactly to the weak form of the governing equations, viz.

$$\begin{aligned}
R_{\mathbf{u}} &= \int_B \frac{1}{G}\mathbf{T}_{\text{R}} : \nabla \mathbf{u}_{\text{test}}\, dv_{\text{R}}, \\
R_{\hat{\mu}} &= \int_B \left(\phi^{-1} - \phi_{\text{old}}^{-1}\right)\, \hat{\mu}_{\text{test}}\, dv_{\text{R}} + \int_B \Omega\, \Delta t\, \mathbf{M}\left(\nabla \hat{\mu}\, R\vartheta\right) \cdot \nabla \hat{\mu}_{\text{test}}\, dv_{\text{R}}, \\
R_{\hat{\mathcal{P}}} &= \int_B \left(\hat{\mathcal{P}} - \frac{R\vartheta}{K\Omega}\left[\ln(1-\phi) + \phi + \chi\phi^2 - \hat{\mu}\right]\right)\, \mathcal{P}_{\text{test}}\, dv_{\text{R}}.
\end{aligned} \tag{4.3}$$

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 $\{\mathbf{u},\, \hat{\mu},\, \hat{\mathcal{P}}\}$ for which the residuals are zero[3], and which therefore satisfy the governing equations as well as the initial and boundary conditions.

4.1 Algorithm for calculation of the residuals

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

1. Begin the step with known values of

$$\left\{\mathbf{u},\, \hat{\mu},\, \hat{\mathcal{P}}\right\} \quad \text{and} \quad \left\{\mathbf{u}_{\text{old}},\, \hat{\mu}_{\text{old}},\, \hat{\mathcal{P}}_{\text{old}}\right\}, \tag{4.4}$$

   where for the first step, $\{\mathbf{u}_{\text{old}},\, \hat{\mu}_{\text{old}},\, \hat{\mathcal{P}}_{\text{old}}\}$ are given by the initial conditions.

2. Calculate necessary kinematical quantities for the current step as

$$\begin{aligned}
&\mathbf{F} = \nabla \mathbf{u} + \mathbf{1}, \\
&J = \det \mathbf{F}, \\
&\mathbf{C}^{-1} = (\mathbf{F}^{\top}\mathbf{F})^{-1}
\end{aligned} \tag{4.5}$$

   and for the previous step as

$$\begin{aligned}
&\mathbf{F}_{\text{old}} = \nabla \mathbf{u}_{\text{old}} + \mathbf{1}, \\
&J_{\text{old}} = \det \mathbf{F}_{\text{old}}.
\end{aligned} \tag{4.6}$$

3. Use the pressure-like field $\hat{\mathcal{P}}$ to calculate J^e for the current and previous step as

$$J^e = \exp\left(\mathcal{P}\right), \quad \text{and} \quad J^e_{\text{old}} = \exp\left(\mathcal{P}_{\text{old}}\right). \tag{4.7}$$

4. Calculate the polymer volume fraction ϕ for the current and previous step as

$$\phi = \frac{J^e}{J}\phi_0, \quad \text{and} \quad \phi_{\text{old}} = \frac{J^e_{\text{old}}}{J_{\text{old}}}\phi_0, \tag{4.8}$$

   where ϕ_0 is the initial value of the polymer volume fraction.

[3] To within some small numerical tolerance.

5. Calculate the current Piola stress, Cauchy stress, and pressure p as

$$\begin{aligned}
&\mathbf{T}_{\mathrm{R}} = G(\mathbf{F} - \mathbf{F}^{-\top}) + \phi^{-1} K \hat{\mathcal{P}} \mathbf{F}^{-\top}, \\
&\mathbf{T} = J^{-1} \mathbf{T}_{\mathrm{R}} \mathbf{F}^{\top}, \\
&p = -\frac{1}{3} \operatorname{tr} \mathbf{T}.
\end{aligned} \tag{4.9}$$

6. Update the value of the Flory-Huggins parameter according to

$$\chi(p) = \chi_0 + \beta\, p. \tag{4.10}$$

7. Calculate the current mobility of the solvent as

$$\begin{aligned}
&\mathbf{M} = \mathbf{C}^{-1} \left(\frac{D c_{\mathrm{R}}}{R\vartheta} \right), \quad \text{where} \\
&c_{\mathrm{R}} = \frac{1}{\Omega} \left(\frac{1}{\phi} - 1 \right), \quad \text{and} \\
&D = D_0 \exp\left(-\alpha \frac{\phi}{1-\phi} \right) + \gamma.
\end{aligned} \tag{4.11}$$

8. Use the quantities from steps 1-7 to compute the residuals as

$$\begin{aligned}
R_{\mathbf{u}} &= \int_B \frac{1}{G} \mathbf{T}_{\mathrm{R}} : \nabla \mathbf{u}_{\text{test}} \, dv_{\mathrm{R}}, \\
R_{\hat{\mu}} &= \int_B \left(\phi^{-1} - \phi_{\text{old}}^{-1} \right) \hat{\mu}_{\text{test}} \, dv_{\mathrm{R}} + \int_B \Omega \, \Delta t \, \mathbf{M} \left(\nabla \hat{\mu} \, R\vartheta \right) \cdot \nabla \hat{\mu}_{\text{test}} \, dv_{\mathrm{R}}, \\
R_{\hat{\mathcal{P}}} &= \int_B \left(\hat{\mathcal{P}} - \frac{R\vartheta}{K\Omega} \left[\ln(1-\phi) + \phi + \chi \phi^2 - \hat{\mu} \right] \right) \mathcal{P}_{\text{test}} \, dv_{\mathrm{R}}.
\end{aligned} \tag{4.12}$$

9. FEniCS solves for the current values of $\{\mathbf{u},\, \hat{\mu},\, \hat{\mathcal{P}}\}$.

10. Set these current values as the "old" values for the next step in time,

$$\left\{ \mathbf{u},\, \hat{\mu},\, \hat{\mathcal{P}} \right\} \implies \left\{ \mathbf{u}_{\text{old}},\, \hat{\mu}_{\text{old}},\, \hat{\mathcal{P}}_{\text{old}} \right\}. \tag{4.13}$$

11. Repeat steps 1-10 for desired number of steps in time.

References

S. A. Chester, C. V. Di Leo, and L. Anand. A finite element implementation of a coupled diffusion-deformation theory for elastomeric gels. *International Journal of Solids and Structures*, 52:1–18, 2015.

E. M. Stewart, S. Narayan, and L. Anand. On modeling the infiltration of water in a PEG-DA hydrogel and the resulting swelling under unconstrained and mechanically-constrained conditions. *Extreme Mechanics Letters*, 54:101775, July 2022.