Supplementary materials to: Hyperelastic models for hydration of cellular tissue

R.G.M. van der Sman
Agrotechnology and Food Sciences Group,
Wageningen University & Research, the Netherlands

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1 Homogeneous swelling of hydrogels described with scaling laws

The swelling of hydrogels is described via thermodynamics using a free energy functional as in the Flory-Rehner theory. The free energy functional has two independent contributions: 1) the mixing free energy, and 2) the strain energy function. The strain energy function accounts for the contribution of the elastic deformation. It is described in terms of the stretching of the polymers λ_i in the three principal directions. In the relaxed state $\lambda_i = 1$, and is taken as the reference state. During tension polymers are stretched $\lambda_i > 1$, while during compression $\lambda_i < 1$. In compression the polymer will behave more like a collapsed random coil. For isotropic swelling it holds all three principal stretches are equal: $\lambda_i = \lambda$. In the relaxed state the polymer volume fraction has the value ϕ_{ref}. Under the assumption of incompressibility of polymer and solvent (water), we have the following relation between stretching and polymer volume fraction:

$$\lambda_1\lambda_2\lambda_3 = \lambda^3 = \frac{\phi_{ref}}{\phi} \tag{1}$$

Below, we will define $\tilde{\phi} = \phi/\phi_{ref}$.

Often, it suffices to use the Neo-Hookean model to describe the elastic deformation of hydrogels. The strain energy function is the following:

$$W(\lambda) = \tfrac{1}{2}G\textstyle\sum_i(\lambda_i^2 - 1) \tag{2}$$

The mechanical stress is derived from the strain energy function as follows:

$$\sigma_{ii} = \frac{1}{\lambda_j\lambda_k}\frac{\partial W}{\partial \lambda_i} - \Pi_{mix} = \frac{G}{\lambda} - \Pi_{mix} = G\tilde{\phi}^{\frac{1}{3}} - \Pi_{mix} \tag{3}$$

Π_{mix} is the osmotic pressure, derived from the mixing free energy. The mixing free energy is traditionally described by Flory-Huggins theory. At equilibrium the mechanical stress is zero everywhere in the gel. At equilibrium the polymer volume fraction is ϕ_0, which will be different from ϕ_{ref}.

Recently, we have investigated an alternative formulation with the Cloizeaux scaling law [1], which states that the osmotic pressure scales as $\Pi_{mix} \sim \phi^{\beta}$, with β an universal scaling exponent [2]. For Gaussian coils in a theta-solvent $\beta = 9/4$, which applies also to biopolymer gels. We have shown that the scaling law can be formulated in terms of $\tilde{\phi}$ using the c*-theorem of deGennes [3], and the relation $\phi_0 = 2/3\phi_{ref}$ [4], which holds universally for biopolymer gels. The c*-theorem related the elastic modulus G to ϕ_{ref}: $G \sim \phi_{ref}^{\beta}$, with β the same scaling exponent as in the Cloizeaux scaling law.

$$\Pi_{mix} = \alpha G \tilde{\phi}^{\beta} \tag{4}$$

The value of α is determined by the relation $\phi_0 = 2/3\phi_{ref}$, which holds at equilibrium defined by:

$$\sigma_{ii} = G\tilde{\phi}^{\frac{1}{3}} - \alpha G \tilde{\phi}^{\beta} \tag{5}$$

The above expression of the osmotic pressure has is more compact formulation than the Flory-Huggins theory, and is formulated in terms of $\tilde{\phi}$. Hence, it is better fitting with the framework of large deformation of elastic hydrogels - possibly allowing for analytical solutions of problems with simple geometry.

2 Inhomogeneous deformation of elastic shells

It is known that non-swellable elastic shells with a pressurized internal cavity undergo inhomogeneous deformation with respect to their reference state. For spherical and cylindrical shells the mechanical equilibrium under a prescribed internal pressure has an analytical solution. It is instructive to show how this analytical solution is obtained, as it provides the framework for mechanical equilibrium for swellable, elastic shells - which we have taken as a model system for the cell wall.

2.1 Spherical shells

It is custom to describe first the kinetics of the large deformation. The elastic shell is thought of to consists of infinitesimal thin control volumes. In the deformed state, the control volume are characterized by their radius r and thickness dr. The deformation is described with respect to a reference state, which is often taken as the relaxed state where all polymers are unstretched, i.e. $\lambda_i = 1$. In the reference state the control volume has a radius R and thickness dR.

For spherical geometries the principal stresses are the radial stress, σ_{rr} and the two hoop stresses, which are equal due to symmetry $\sigma_{\theta\theta} = \sigma_{\phi\phi}$. The stretches in these principal directions are $\lambda_\theta = r/R$ and $\lambda_r = dr/dR$. The elastic material is assumed to be incompressible, meaning that the volume of the control volume is invariant under deformation: $dV = 4\pi r^2 dr = 4\pi R^2 dR$. In terms of the stretches this reads:

$$\lambda_\theta^2 \lambda_r = 1 \tag{6}$$

This shows that the stretches are *not* independent. It is custom to define $\lambda_\theta = \lambda$, and consequently, $\lambda_r = 1/\lambda^2$.

The condition for mechanical equilibrium in spherical coordinates is [5]:

$$\frac{d\sigma_{rr}}{dr} + 2\frac{\sigma_{rr} - \sigma_{\theta\theta}}{r} = 0 \tag{7}$$

The expressions for the stresses are derived from the strain energy function. For the Neo-Hookean model it has the following form: $W = \frac{1}{2}G(\lambda_r^2 + \lambda_\theta^2 + \lambda_\phi^2 - 3)$. G is the elastic modulus. The difference between the hoop and radius stress is:

$$\sigma_{\theta\theta} - \sigma_{rr} = G(\lambda_\theta \frac{\partial W}{\partial \lambda_\theta} - \lambda_r \frac{\partial W}{\partial \lambda_r}) = G(\lambda^2 - \lambda^{-4}) \tag{8}$$

Consequently:

$$d\sigma_{rr} = 2G(\lambda^2 - \lambda^{-4})\frac{dr}{r} \tag{9}$$

The last term dr/r can be expressed in λ using the incompressibility condition [5, 6]. Recall that $\lambda = r/R$, and the total derivative gives:

$$d\lambda = \frac{dr}{R} - \frac{r}{R^2}dR \tag{10}$$

using $R = r/\lambda$, and

$$dr = \frac{R^2}{r^2}dR = \frac{1}{\lambda^2}dR \tag{11}$$

This renders:

$$\frac{dr}{r} = \frac{d\lambda}{\lambda^4 - \lambda} \tag{12}$$

Now, we can write:

$$d\sigma_{rr} = 2G\frac{\lambda^2 - \lambda^{-4}}{\lambda^4 - \lambda}d\lambda \tag{13}$$

For an arbitrary strain energy function, the following relation holds [7]:

$$d\sigma_{rr} = \frac{\lambda \partial_\lambda W}{\lambda^3 - 1}d\lambda \tag{14}$$

The above equation can be integrated over the shell, using the boundary values. At the inner surface the radius stress equals the internal pressure $\sigma_A = p_{int}$, and its stretch equals $\Lambda_a = a/A$, where a is the deformed inner radius, and A is the inner radius in the reference state. At the outer surface the radial stress equals the external pressure $\sigma_B = p_{ext}$, and the stretch is $\Lambda_b = b/B$. Integration over the shell renders [8]:

$$\frac{2(p_{int} - p_{ext}}{G} = \frac{1 + 4\Lambda_a^3}{\Lambda_a^4} - \frac{1 + 4\Lambda_b^3}{\Lambda_b^4} \tag{15}$$

The stretch of the outer surface can be expressed in Λ_a via the incompressibility of the total shell. From the reference state the radii A and B are known, and consequently: $B^3 - A^3 = b^3 - a^3 = \Lambda_b^3 B^3 - \Lambda_a^3 A^3$. Hence, one can compute the relation between the pressure difference $\Delta p = p_{int} - p_{ext}$ and Λ_a.

The above expression is known as the thick shell theory [7]. For cells often the thin shell approximation applies [9].

In absence of surface tension, $\sigma_A = -p_{int}$, and $\sigma_B = -p_{ext}$. Hence, to inflate the sphere, $p_{int} > p_{ext}$. It is assumed that $\sigma_{rr} \ll \sigma_{\theta\theta}$, and thus $\sigma_{\theta\theta} \approx G(\lambda^2 - \lambda^{-4})$. Mechanical equilibrium over a cross section of the thin shell with thickness h and radius r renders:

$$(p_{int} - p_{ext})\pi r^2 = 2\pi r h \sigma_{\theta\theta} \tag{16}$$

using $r = R\lambda$, and $h = H\lambda^2$ (with H the thickness of a relaxed shell) [7]:

$$(p_{int} - p_{ext}) = \frac{2H}{R} G \frac{(\lambda^2 - \lambda^{-4})}{\lambda^3} = \frac{2H}{R} G(\lambda^{-1} - \lambda^{-7}) \tag{17}$$

The thick and thin shell theories are compared in the left pane of figure 1.

2.2 Cylindrical shells

For shells with cylindrical geometry the principal directions for the stress are the radial, circumferential, and axial direction. We assume very long cylinders with negligible axial stretch: $\lambda_z = 1$. Hence, the incompressibilty condition becomes: $\lambda_\theta \lambda_r = 1$, with $\lambda_\theta = \lambda = r/R$, and $\lambda_r = 1/\lambda = dr/dR$.

Mechanical equilibrium in cylindrical coordinates:

$$\frac{d\sigma_{rr}}{dr} + \frac{\sigma_{rr} - \sigma_{\theta\theta}}{r} = 0 \tag{18}$$

For the Neo-Hookean model the difference between hoop and radial becomes:

$$\sigma_{\theta\theta} - \sigma_{rr} = G(\lambda^2 - \lambda^{-2}) \tag{19}$$

The last term dr/r can be expressed in λ via the incompressibility condition:

$$\frac{dr}{r} = \frac{d\lambda}{\lambda^3 - \lambda} \tag{20}$$

Hence, we can write:

$$d\sigma_{rr} = G\frac{\lambda^2 - \lambda^{-2}}{\lambda^3 - \lambda}d\lambda \tag{21}$$

Integration over the shell renders [10]:

$$\frac{p_{int} - p_{ext}}{G} = \left(\frac{1}{2\Lambda_a^2} - \frac{1}{2\Lambda_b^2} - \ln(\Lambda_a/\Lambda_b)\right) \tag{22}$$

In the thin membranes approximation it is assumed that $\sigma_{rr} \ll \sigma_{\theta\theta}$. Mechanical equilibrium over the axial cross section of the cylinder, with inner radius r and thickness h, dictates:

$$(p_{int} - p_{ext})2Lr = 2Lh\sigma_{\theta\theta} \tag{23}$$

using the incompressibility condition:

$$(p_{int} - p_{ext}) = \frac{H}{R}G(1 - \lambda^{-4}) \tag{24}$$

The thick and thin shell theories are compared in the right pane of figure 1.

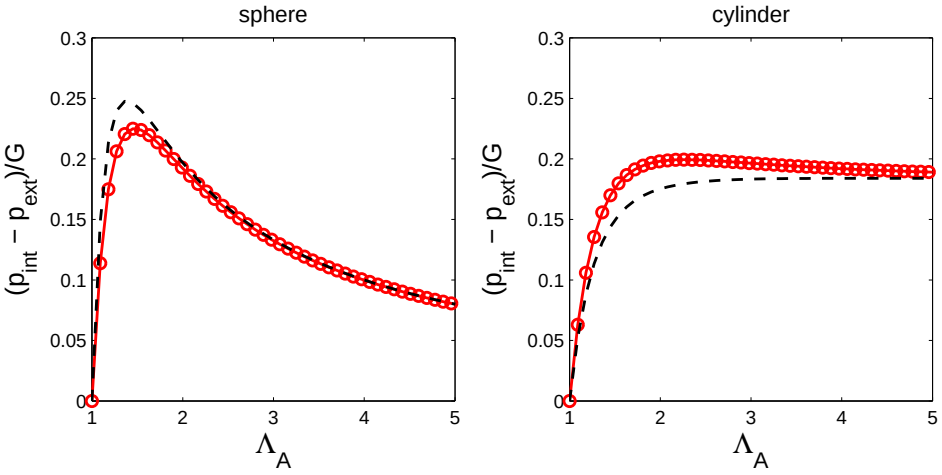

Figure 1: Pressure stretch relations for elastic shells of spherical and cylindrical shape. the solid line with circles indicate the solution for thick shells with $H/R = 0.2$, and the dashed lines indicate the solution for thin shells. We observe mainly deviations for moderate deformations, $\Lambda \approx 2$.

3 Cylindrical cell swelling for different fiber angles

We have performed a parameter study on the swelling of cylindrical cells, having microfibers in the cell wall having different angles with the main axis.

The values of the fiber angles we have taken are 5, 22, 33, and 44 degrees. (Angles with 0 degrees angle are oriented along the circumferential direction). For larger degrees the numerical solution is instable. We think this is caused by our assumption of a homogeneous axial deformation Λ_z, which is incompatible with microfiber angles larger than 45 degrees. Simulations are performed with $k_1/G = 1$ and $k_2/G = 2.9$ and $B = 1.05R_0$ - using Eq.(32) in the main text as the strain energy function. Results are shown in figures 2-3. Similar to the presentation of results in the main text, we show a) the stretching in axial and circumferential direction (Λ_i/Λ_0) as function of cell volume V/V_0, b) the pressure/volume curve $\Delta p/G$ vs. V/V_0, and c) the relative water content of the cell wall $RWC = V_{wall}/V_{tot}$ as function of cell volume.

For small angles (5 degrees) the circumferential stretching is very limited, but there is a large axial stretching. The pressure/volume curve does not show a limiting cell volume. The relative water content in the cell wall is decreasing with increasing cell volume, but reaches an asymptotic value. This kind of behaviour is very similar to the growth of hyphen of fungi, which grow via stretching in the axial direction. Indeed, this kind of fiber orientation has been observed for hyphen in their growth stage. During maturation glucan fibers are synthesized, which are oriented in axial direction - thereby changing the average fiber orientation towards 45 degrees.

At larger angles we observe effects of strain hardening in the pressure/volume curves. The limiting cell volume is quite similar for angles $\psi_0 \geq \pi/10$, but with increasing angles a higher pressure is required to reach the limiting cell volume. Surprisingly, the RWC is showing a minimum. We view it out of scope of our paper to investigate this point into depth.

As can be expected, the stretching in the axial and circumferential direction depends highly on the fiber orientation. For small angles, $\psi_0 < \pi/8$ there is little stretching in circumferential direction, while for large angles, $\psi_0 \approx \pi/4$, there is little stretching in axial direction. For intermediate angles there is a mixed behaviour, stretching occurs in both directions without little signs of strain hardening.

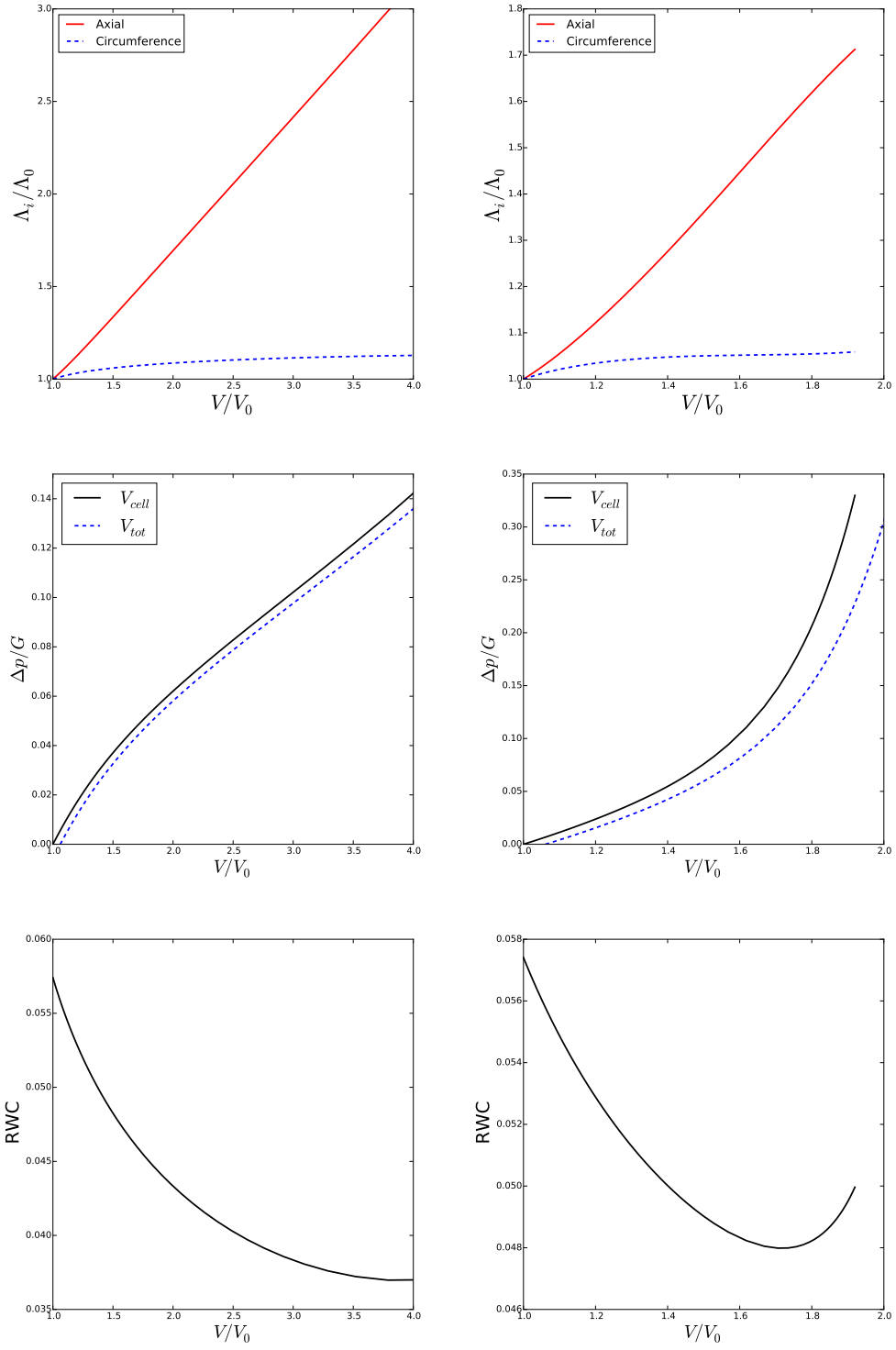

Figure 2: Swelling of cylindrical cells with fiber angles 5 (left) and 22 (right) degrees

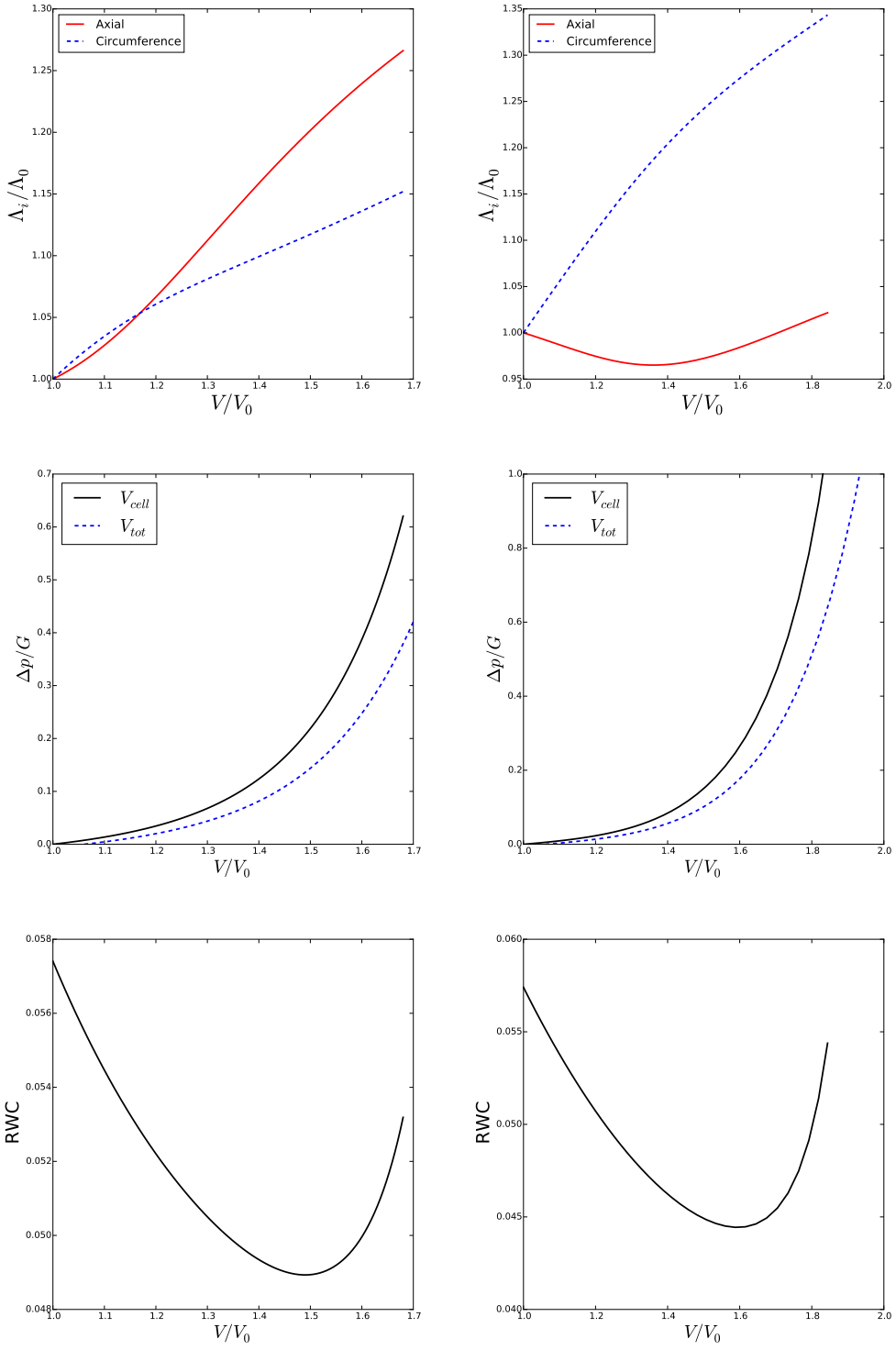

Figure 3: Swelling of cylindrical cells with fiber angles 33 (left) and 44 (right) degrees

4 Numerical solution procedure

The cell wall has volume $V_{wall} = B^3 - A^3$. It will be divided in N control volumes, with radius r, and thickness dr. The volume is then $dV = 4\pi r^2 dr$, with polymer volume fraction ϕ. In the relaxed state the radius is R, the thickness dR, and the polymer volume fraction ϕ_{ref}, with volume $dV_{ref} = 4\pi R^2 dR$. Amount of polymer is conserved during swelling:

$$\phi_{ref} dV_{ref} = \phi dV \tag{25}$$

or rather with introducing the stretch factors $\lambda_\theta = r/R$, and $\lambda_r = dr/dR$:

$$1 = \tilde{\phi}\lambda_\theta^2 \lambda_r \tag{26}$$

At swelling the tension is:

$$\begin{aligned} \sigma_{\theta\theta} &= G\tilde{\phi}\lambda_\theta^2 - \Pi_{mix} \\ \sigma_{rr} &= G\tilde{\phi}\lambda_r^2 - \Pi_{mix} \end{aligned} \tag{27}$$

with $\Pi_{mix} = \alpha G \tilde{\phi}^\beta$. α is determined by the fact that free isotropic swelling occurs at $\tilde{\phi} = 2/3$. Isotropic stretching at free swelling conditions imposes $\lambda = \lambda_\theta = \lambda_r$. Hence, $\lambda = \tilde{\phi}^{-\frac{1}{3}}$. Substitution of that shows that equilibrium is obtained at free swelling in pure water. The stretching is $\Lambda_0 = (2/3)^{-\frac{1}{3}}$. The control volume has a radius of $\Lambda_0 R$, and thickness $\Lambda_0 dR$.

4.1 Algoritm for swelling spherical shell

Initialisation: set $\lambda = \Lambda_b$, $r = b = \Lambda_b B$, and $\sigma = \sigma_b = 0$.
Iterate for all control volumes, starting at the outside
  Solve ϕ via σ_{rr}, cf. Eq.(27).
  Compute thickness of control volume $dr/dR = \lambda_r = 1/(\tilde{\phi}\lambda^2)$
  Compute $\lambda(r - dr) = (r - dr)/(R - dR)$.
  Compute $d\sigma/dr$
  Compute $\sigma(r - dr) = \sigma(r) + dr \times d\sigma/dr$
  Advance to next control volume
$\Delta p = \sigma_b - \sigma_a$

4.2 Algoritm for swelling cylindrical shell

Initialisation: set $\lambda = \Lambda_b$, $r = b = \Lambda_b B$, and $\sigma = \sigma_b = 0$.
Assume a certain Λ_z
Iterate until convergence
  Iterate for all control volumes, starting at the outside
    Solve ϕ via σ_{rr}
    Compute thickness of control volume $dr/dR = \lambda_r = 1/(\tilde{\phi}\lambda\Lambda_z)$

Compute $\lambda(r - dr) = (r - dr)/(R - dR)$.
Compute $d\sigma/dr$
Compute $\sigma(r - dr) = \sigma(r) + dr \times d\sigma/dr$
Advance to next control volume
$\Delta p = \sigma_b - \sigma_a$
Compute Λ_z from force balance in axial direction via iteration

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