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VISCOELASTIC RESPONSE OF ANISOTROPIC BIOLOGICAL MEMBRANES. PART II: CONSTITUTIVE MODELS

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According to: *Tib Journal Abbreviations (C) Mathematical Reviews*, the abbreviation TEOPM7 stands for TEORIJSKA I PRIMENJENA MEHANIKA.

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Viscoelastic response of anisotropic biological membranes. Part II: Constitutive models

Vlado A. Lubarda * **Robert J. Asaro** †

Dedicated to the memory of Dr. Jiddu Bezares, who was a great collaborator in our previous efforts of studying the mechanical response of biological membranes, before his untimely death.

Abstract

In Part I of this series [7] we described the structure of the biopolymer interlayers found in the shell of the mollusk *Haliotis rufescens* (the red abalone). There we described how the layers can be viewed as a viscoelastic composite reinforced by a network of chitin fibrils arranged in an often nearly unidirectional architecture. Mechanical testing documented the response to tensile testing of layers removed *via* demineralization. Herein in Part II we describe a general viscoelastic constitutive model for such layers that may be both transversely isotropic or orthotropic as would follow from the network of nearly aligned chitin fibrils described by Bezares *et al.* in Part I [7]. Part III of this series will be concerned with applying the models to more fully describing the response of these types of biological membranes to mechanical loading.

Key words: Abalone, Biological membranes, Orthotropy, Transverse isotropy, Viscoelasticity

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1 Introduction

Part I of this series presented a report detailing the structure of the biopolymer matrix of the nacreous shell of the mollusk *Halliotis rufescens* (the red abalone); this matrix, which is in the form of *interlayers* sandwiched between interdigitated layers of $CaCO_3$ tiles, is held to be responsible for the intriguing mechanical properties of the nacreous shell and being the mediator of growth of nacre as discussed, *e.g.*, in [1]-[3]. With specific respect to the organic matrix of *Haliotis rufescens*, we refer to the original work of Bezares *et al.* [4]-[7].

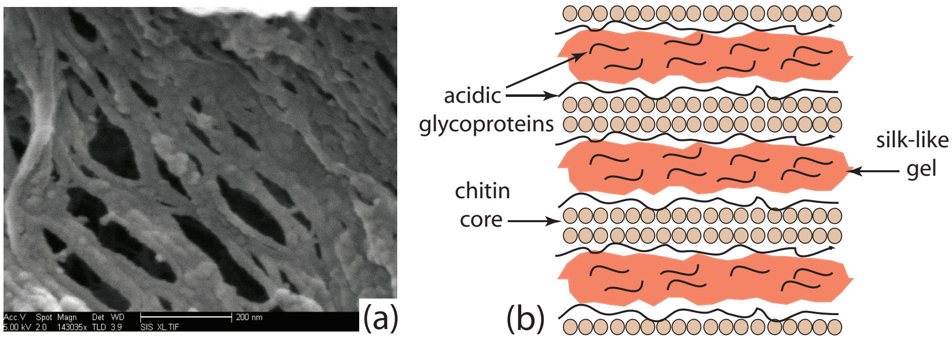

Figure 1: (a) An optical histochemical photograph of a section of biopolymer matrix extracted from *Haliotis rufescens via* enzymatic digestion that reveals the layout of chitin fibrils. (b) A schematic depiction of the layered structure of chitin fibrils and protein within the biopolymer *interlayers* of nacre.

Bezares *et al.* [5] extracted the biopolymer layers (*i.e.*, the interlayers) after demineralization with ethylene diamine tetraacetic acid (EDTA) and various forms of digestion to remove protein. Both types of procedures were carried out under ambient Ph conditions. They performed a series of tensile tests on the extracted layers and found them to be viscoelastic and to possess mechanical integrity over remarkably large length scales, which spanned literally hundreds of $CaCO_3$ tile diameters. The analysis performed by Bezares *et al.* [5] attributed the integrity of the biopolymer interlayers to the chitin core, earlier imaged *via* atomic force microscopy (AFM) by Bezares *et al.* [4], and modeled the chitin network as an organized mesh of chitin fibrils. Figure 1a shows an optical image of tissue extracted from *Haliotis rufescens*, again after demineralization with EDTA followed by progressively intense digestion to remove protein [7]. In this work the structure of chitin was documented in more detail. It was found that the fibrils were largely unidirectional and occasionally in the form of nearly parallel sets of fibrils with an angle in the range $10° \leq \phi \leq 90°$ (see Fig. 5). This is illustrated in the orthotropic model depicted in Fig. 5 that is developed in detail in Section 3. Figure 1a illustrates how most sections of the interlayers contained nearly unidirectional fibril bundles and yet in many areas the fibrils were oriented in the $\pm\phi$ manner as explained above. It is this type of material we aim to describe by an anisotropic viscoelastic constitutive model.

For generality, and application to a wider range of biological membranes, we develop two models that treat the membranes as either transversely isotropic or orthotropic. We believe the orthotropic model best fits the structural symmetry of the interlayers of nacre in *Haliolis rufescens*, but for other biological structures a transversely isotropic description may indeed be more appropriate; this is addressed at the end in the discussion and conclusions section. While elastic part of response was anisotropic, we assumed that viscous properties along and across chitin fibrils were the same, so that viscous part of response was isotropic.

2 Transversely isotropic membranes

Our membranes are modeled as viscoelastic materials of the general Kelvin–Voigt type, according to which the stress tensor is decomposed into its elastic and viscous parts as [8]-[10] as

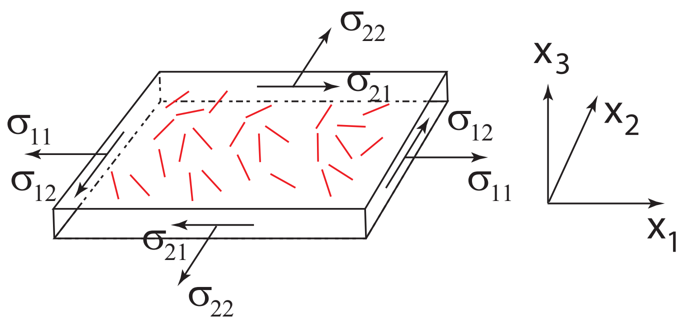

Figure 2: A thin viscoelastic biological membrane reinforced with isotropically distributed linear elastic fibers (shown in red).

$$\sigma_{ij} = \sigma^{\mathrm{e}}_{ij} + \sigma^{\mathrm{v}}_{ij}\,. \tag{2.1}$$

The materials are taken to be in the form of thin membranes that are reinforced with isotropically distributed arrays of linear elastic fibers; this is sketched in Fig. 2. The spherical part of the stress tensor is assumed to be independent of viscous processes, as illustrated *via* the models sketched in Fig. 3, so that

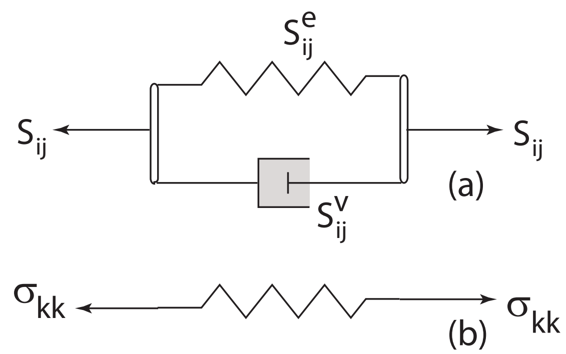

Figure 3: (a) Transversely isotropic Kelvin–Voigt viscoelastic model for the deviatoric part of stress S_{ij}. (b) Transversely isotropic elastic model for the spherical part of stress $\sigma_{kk}/3$.

$$\sigma_{kk} = \sigma^{\mathrm{e}}_{kk}\,, \quad \sigma^{\mathrm{v}}_{kk} = 0 \quad (\text{sum on}\, k)\,. \tag{2.2}$$

The deviatoric part of the stress tensor (S_{ij}), appearing in the decomposition $\sigma_{ij} = S_{ij} + (1/3)\sigma_{kk}\delta_{ij}$, where δ_{ij} is the Kronecker delta, is then partitioned as

$$S_{ij} = S^{\mathrm{e}}_{ij} + S^{\mathrm{v}}_{ij}\,, \quad S^{\mathrm{v}}_{ij} = \sigma^{\mathrm{v}}_{ij}\,. \tag{2.3}$$

Since σ^{v}_{ij} is deviatoric, it will be assumed that it is related to the deviatoric part of the strain rate tensor. Hence, by invoking Newton's linear relation, we have

$$\sigma^{\mathrm{v}}_{ij} = 2\eta\left(\dot\epsilon_{ij} - \frac{1}{3}\,\dot\epsilon_{kk}\delta_{ij}\right), \tag{2.4}$$

where η is the coefficient of viscosity. More involved viscous law could be adopted, such as one used in [11]. The viscous properties along and across chitin fibrils are assumed to be the same, so that viscous part of the response described by (2.4) is isotropic. Introducing the viscosity tensor with the components [8]

$$J_{ijkl} = 2\eta\left[\frac{1}{2}\left(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}\right) - \frac{1}{3}\,\delta_{ij}\delta_{kl}\right], \tag{2.5}$$

we can rewrite eq. (2.4) as

$$\sigma_{ij}^{\mathrm{v}} = J_{ijkl}\dot{\epsilon}_{kl}\,. \tag{2.6}$$

The elastic part of the stress tensor is related to strain *via* the well-known transversely isotropic constitutive relation [12, 13], which can be cast in the form

$$\begin{aligned}\sigma_{ij}^{\mathrm{e}} &= \lambda\epsilon_{kk}\delta_{ij} + 2\mu\epsilon_{ij} + \alpha(\epsilon_{33}\delta_{ij} + \epsilon_{kk}\delta_{i3}\delta_{j3}) \\ &\quad + 2(\mu_0 - \mu)(\delta_{i3}\epsilon_{j3} + \delta_{j3}\epsilon_{i3}) + \beta\delta_{i3}\delta_{j3}\epsilon_{33}\,,\end{aligned} \tag{2.7}$$

where λ, μ, μ_0, α, and β are five material parameters. To interpret eq. (2.7) we proceed as follows. The elastic shear modulus of the membrane within its plane of isotropy is μ *via* the response

$$\sigma_{12}^{\mathrm{e}} = \sigma_{21}^{\mathrm{e}} = 2\mu\epsilon_{12}\,.$$

The other Lamé constant is λ which, along with α, fully describes the isotropic in-plane stress response. Indeed, the in-plane normal stresses are given by

$$\sigma_{11}^{\mathrm{e}} = \lambda\epsilon_{kk} + 2\mu\epsilon_{11} + \alpha\epsilon_{33}\,, \quad \sigma_{22}^{\mathrm{e}} = \lambda\epsilon_{kk} + 2\mu\epsilon_{22} + \alpha\epsilon_{33}\,.$$

The out-of-plane shear modulus is μ_0, such that

$$\sigma_{13}^{\mathrm{e}} = \sigma_{31}^{\mathrm{e}} = 2\mu_0\epsilon_{13}\,, \quad \sigma_{23}^{\mathrm{e}} = \sigma_{32}^{\mathrm{e}} = 2\mu_0\epsilon_{23}\,.$$

The remaining material constant, β, completes the description of the out-of-plane response.

Expressed more generally, with respect to an otherwise arbitrary coordinate system in which n_i are components of the unit vector parallel to the axis of transverse isotropy, the constitutive expression of eq. (2.7) can be recast in the form

$$\begin{aligned}\sigma_{ij}^{\mathrm{e}} &= \lambda\epsilon_{kk}\delta_{ij} + 2\mu\epsilon_{ij} + \alpha(n_k n_l\epsilon_{kl}\delta_{ij} + n_i n_j\epsilon_{kk}) \\ &\quad + 2(\mu_0 - \mu)(n_i n_k\epsilon_{kj} + n_j n_k\epsilon_{ki}) + \beta n_i n_j n_k n_l\epsilon_{kl}\,.\end{aligned} \tag{2.8}$$

Upon summing the elastic and viscous parts of stress, given by eqs. (2.6) and (2.8), there follows

$$\sigma_{ij} = C_{ijkl}\epsilon_{kl} + J_{ijkl}\dot{\epsilon}_{kl}\,, \tag{2.9}$$

where C_{ijkl} are the components of the elastic moduli tensor associated with eq. (2.8). For a given strain history, eq. (2.9) specifies the corresponding stress history.

By inverting eq. (2.8), the strain tensor can be expressed as

$$\epsilon_{ij} = D_{ijkl}\sigma^{\mathrm{e}}_{kl}\,, \tag{2.10}$$

where D_{ijkl} are the components of the compliance tensor, the inverse of the elastic moduli tensor C_{ijkl}. Since $\sigma^{\mathrm{e}}_{ij} = \sigma_{ij} - \sigma^{\mathrm{v}}_{ij}$, eq. (2.10) becomes

$$\epsilon_{ij} = D_{ijkl}(\sigma_{kl} - \sigma^{\mathrm{v}}_{kl})\,. \tag{2.11}$$

In view of eq. (2.6) for σ^{v}_{kl}, eq. (2.11) can be written as

$$\epsilon_{ij} + D_{ijkl}J_{klmn}\dot{\epsilon}_{mn} = D_{ijkl}\sigma_{kl}\,. \tag{2.12}$$

For a given stress history, this is a set of differential equations for the corresponding strain history. Equation (2.12) also follows directly from eq. (2.9) by subjecting it to the trace product with the compliance tensor D_{mnij}.

2.1 Plane stress

If the membrane is loaded within its plane, plane stress conditions apply so that $\sigma_{i3} = 0$, for $i = 1, 2, 3$. For the shear stresses among these, this implies, from (2.1), (2.4) and (2.7), that

$$\sigma_{13} = 2\eta\dot{\epsilon}_{13} + 2\mu\epsilon_{13} = 0\,, \quad \sigma_{23} = 2\eta\dot{\epsilon}_{23} + 2\mu\epsilon_{23} = 0\,. \tag{2.13}$$

If initially $\epsilon_{13} = \epsilon_{23} = 0$, eqs. (2.13) require that, at all times t,

$$\epsilon_{13}(t) = \epsilon_{23}(t) = 0\,. \tag{2.14}$$

Moreover, the vanishing of the normal stress σ_{33} gives

$$\sigma_{33} = \sigma^{\mathrm{e}}_{33} + \sigma^{\mathrm{v}}_{33} = 0\,, \tag{2.15}$$

where, from (2.4) and (2.7),

$$\sigma^{\mathrm{v}}_{33} = \frac{4\eta}{3}\,\dot{\epsilon}_{33} - \frac{2\eta}{3}\,(\dot{\epsilon}_{11} + \dot{\epsilon}_{22})\,, \quad \sigma^{\mathrm{e}}_{33} = k\epsilon_{33} + (\lambda + \alpha)(\epsilon_{11} + \epsilon_{22})\,. \tag{2.16}$$

Here, the material parameter k is defined as

$$k = 3\lambda + 4\mu_0 - 2\mu + 4\alpha + \beta\,. \tag{2.17}$$

Thus, by substituting (2.16) into (2.15), we find that

$$\sigma_{33} = \frac{4\eta}{3}\,\dot{\epsilon}_{33} - \frac{2\eta}{3}\,(\dot{\epsilon}_{11} + \dot{\epsilon}_{22}) + k\epsilon_{33} + (\lambda + \alpha)(\epsilon_{11} + \epsilon_{22}) = 0\,. \tag{2.18}$$

The in-plane normal stresses, again from (2.1), (2.4) and (2.7), are

$$\sigma_{11} = 2\eta\Big(\dot\epsilon_{11} - \frac{1}{3}\,\dot\epsilon_{kk}\Big) + \lambda\epsilon_{kk} + 2\mu\epsilon_{11} + \alpha\epsilon_{33}\,, \tag{2.19}$$

$$\sigma_{22} = 2\eta\Big(\dot\epsilon_{22} - \frac{1}{3}\,\dot\epsilon_{kk}\Big) + \lambda\epsilon_{kk} + 2\mu\epsilon_{22} + \alpha\epsilon_{33}\,, \tag{2.20}$$

whereas the in-plane shear stress is given by

$$\sigma_{12} = 2\eta\dot\epsilon_{12} + 2\mu\epsilon_{12}\,. \tag{2.21}$$

The computation of the stress components may be organized as follows. If eq. (2.20) is subtracted from eq. (2.19), there follows

$$2\eta\,(\dot\epsilon_{11} - \dot\epsilon_{22}) + 2\mu\,(\epsilon_{11} - \epsilon_{22}) = \sigma_{11} - \sigma_{22}\,, \tag{2.22}$$

which is a differential equation for the strain difference $(\epsilon_{11} - \epsilon_{22})$. The objective is next to derive a differential equation for the strain sum $(\epsilon_{11}+\epsilon_{22})$. Toward that goal, in view of eq. (2.16), we can write $\sigma_{33}^{\mathrm{e}} = -\sigma_{33}^{\mathrm{v}}$, so that

$$\sigma_{kk} = \sigma_{kk}^{\mathrm{e}} = \sigma_{11}^{\mathrm{e}} + \sigma_{22}^{\mathrm{e}} + \sigma_{33}^{\mathrm{e}} = \sigma_{11}^{\mathrm{e}} + \sigma_{22}^{\mathrm{e}} - \sigma_{33}^{\mathrm{v}}\,. \tag{2.23}$$

Since $\sigma_{kk} = \sigma_{11} + \sigma_{22}$ in the case of plane stress, and since from (2.7) we have

$$\sigma_{11}^{\mathrm{e}} + \sigma_{22}^{\mathrm{e}} = 2(\lambda+\mu)(\epsilon_{11}+\epsilon_{22}) + 2(\lambda+\alpha)\epsilon_{33}\,, \tag{2.24}$$

while σ_{33}^{v} is specified by eq. (2.16), the substitution into eq. (2.23) yields

$$\sigma_{11}+\sigma_{22} = 2(\lambda+\mu)(\epsilon_{11}+\epsilon_{22}) + 2(\lambda+\alpha)\epsilon_{33} - \frac{4\eta}{3}\,\dot\epsilon_{33} + \frac{2\eta}{3}\,(\dot\epsilon_{11}+\dot\epsilon_{22})\,. \tag{2.25}$$

To eliminate the out-of-plane strain ϵ_{33}, we recall that $\sigma_{kk} = \sigma_{kk}^{\mathrm{e}}$, *i.e.*, in view of eq. (2.24) for $\sigma_{11}^{\mathrm{e}} + \sigma_{22}^{\mathrm{e}}$ and eq. (2.16) for σ_{33}^{e},

$$\sigma_{11}+\sigma_{22} = (3\lambda + 2\mu + \alpha)\,(\epsilon_{11}+\epsilon_{22}) + k\epsilon_{33}\,. \tag{2.26}$$

Thus,

$$\epsilon_{33} = \frac{1}{k}\,[(\sigma_{11}+\sigma_{22}) - (3\lambda+2\mu+\alpha)\,(\epsilon_{11}+\epsilon_{22})]\,. \tag{2.27}$$

Substitution of eq. (2.27) into eq. (2.25) yields the differential equation for the strain sum $(\epsilon_{11}+\epsilon_{22})$, which is

$$t_\star(\dot\epsilon_{11}+\dot\epsilon_{22}) + a\,(\epsilon_{11}+\epsilon_{22}) = \frac{1}{2}\,[b\,(\sigma_{11}+\sigma_{22}) + c\,t_\star(\dot\sigma_{11}+\dot\sigma_{22}]\,, \tag{2.28}$$

where $t_\star = \eta/\mu$ is a time like parameter, and

$$\begin{aligned} a &= 3\,\frac{(\lambda+\mu)k-(\lambda+\alpha)(3\lambda+2\mu+\alpha)}{\mu[k+2(3\lambda+2\mu+\alpha)]}\,,\\ b &= 3\,\frac{k-2(\lambda+\alpha)}{k+2(3\lambda+2\mu+\alpha)}\,,\\ c &= \frac{4\mu}{k+2(3\lambda+2\mu+\alpha)}\,. \end{aligned} \tag{2.29}$$

In summary, we have reduced the problem of finding the strain history, for the prescribed stress history, to solving a system of three decoupled 1^{st}- order ordinary differential equations (2.21), (2.22) and (2.28), *i.e.*,

$$t_\star\dot{\epsilon}_{12}+\epsilon_{12}=\frac{1}{2\mu}\,\sigma_{12}\,, \tag{2.30}$$

$$t_\star\left(\dot{\epsilon}_{11}-\dot{\epsilon}_{22}\right)+\left(\epsilon_{11}-\epsilon_{22}\right)=\frac{1}{2\mu}\left(\sigma_{11}-\sigma_{22}\right), \tag{2.31}$$

$$t_\star\left(\dot{\epsilon}_{11}+\dot{\epsilon}_{22}\right)+a\left(\epsilon_{11}+\epsilon_{22}\right)=\frac{1}{2\mu}\left[b\left(\sigma_{11}+\sigma_{22}\right)+c\,t_\star\left(\dot{\sigma}_{11}+\dot{\sigma}_{22}\right)\right]. \tag{2.32}$$

Each of these differential equations is of the form [14]

$$\dot{y}+\frac{r}{t_\star}\,y=f(t),\quad y(t_0)=y_0. \tag{2.33}$$

The initial value problem of eq. (2.33) has a closed form solution in terms of a hereditary integral given by

$$y(t)=\mathrm{e}^{-r(t-t_0)/t_\star}\left\{\int_{t_0}^{t}\mathrm{e}^{r(\tau-t_0)/t_\star}f(\tau)\mathrm{d}\,\tau+y(t_0)\right\}. \tag{2.34}$$

Applying this to eqs. (2.30)–(2.32), we obtain

$$\epsilon_{12}(t)=\mathrm{e}^{-(t-t_0)/t_\star}\Big\{\frac{1}{2\eta}\int_{t_0}^{t}\mathrm{e}^{(\tau-t_0)/t_\star}\sigma_{12}(\tau)\mathrm{d}\,\tau+\epsilon_{12}(t_0)\Big\}, \tag{2.35}$$

$$\begin{aligned} \epsilon_{11}(t)-\epsilon_{22}(t)=\mathrm{e}^{-(t-t_0)/t_\star}\Big\{&\frac{1}{2\eta}\int_{t_0}^{t}\mathrm{e}^{(\tau-t_0)/t_\star}[\sigma_{11}(\tau)-\sigma_{22}(\tau)]d\,\tau\\ &+\epsilon_{11}(t_0)-\epsilon_{22}(t_0)\Big\}, \end{aligned} \tag{2.36}$$

$$\begin{aligned}\epsilon_{11}(t)+\epsilon_{22}(t) = \mathrm{e}^{-a\,(t-t_0)/t_\star}\Big\{\frac{1}{2\eta}\int_{t_0}^{t}\mathrm{e}^{a\,(\tau-t_0)/t_\star}\Big(b\,[\sigma_{11}(\tau)+\sigma_{22}(\tau)] \\ + c\,t_\star[\dot\sigma_{11}(\tau)+\dot\sigma_{22}(\tau)]\Big)\mathrm{d}\,\tau+\epsilon_{11}(t_0)+\epsilon_{22}(t_0)\Big\}.\end{aligned} \tag{2.37}$$

Individual normal strain components may be calculated from

$$\begin{aligned}\epsilon_{11}(t) &= \frac{1}{2}\Big\{[\epsilon_{11}(t)+\epsilon_{22}(t)]+[\epsilon_{11}(t)-\epsilon_{22}(t)]\Big\},\\ \epsilon_{22}(t) &= \frac{1}{2}\Big\{[\epsilon_{11}(t)+\epsilon_{22}(t)]-[\epsilon_{11}(t)-\epsilon_{22}(t)]\Big\}.\end{aligned} \tag{2.38}$$

Having the sum $\epsilon_{11}(t)+\epsilon_{22}(t)$ determined, the out-of-plane normal strain, $\epsilon_{33}(t)$, follows *via* eq. (2.27).

2.2 Loading histories

The strain response corresponding to the loading history sketched in Fig. 4 will be considered first. The in-plane stresses are linearly increased from a

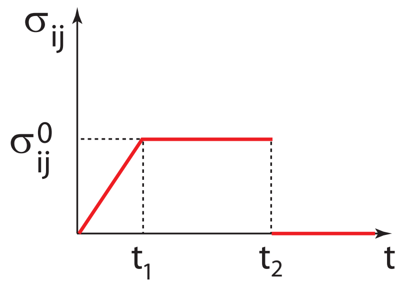

Figure 4: A load history involving a linear ramp up of stress from $t=0$ to $t=t_1$, followed by a constant load in the interval $t_1\le t\le t_2$. For $t>t_2$, the stress is reduced to zero.

zero initial value to the value σ^0_{ij} $(i=1,2,3)$, at time $t=t_1$. The stressing rate for each stress component, albeit constant, is generally different for each component and equal to σ^0_{ij}/t_1. In the subsequent time interval, $t_1\le t\le t_2^-$, the stress is held constant at σ^0_{ij} and thereafter set to zero. Thus

$$\sigma_{ij} = \begin{cases}\sigma^0_{ij}\,t/t_1\,, & 0\le t\le t_1\,,\\ \sigma^0_{ij}\,, & t_1\le t\le t_2^-\,,\\ 0\,, & t\ge t_2^+\,.\end{cases} \tag{2.39}$$

When this is substituted into eqs. (2.35)–(2.37), upon integration there follows in the time interval $0 \leq t \leq t_1$:

$$\epsilon_{12}(t) = \frac{\sigma_{12}^0}{2\mu}\,\frac{t_\star}{t_1}\left(\mathrm{e}^{-t/t_\star} + \frac{t}{t_\star} - 1\right), \tag{2.40}$$

$$\epsilon_{11}(t) - \epsilon_{22}(t) = \frac{\sigma_{11}^0 - \sigma_{22}^0}{2\mu}\,\frac{t_\star}{t_1}\left(\mathrm{e}^{-t/t_\star} + \frac{t}{t_\star} - 1\right), \tag{2.41}$$

$$\epsilon_{11}(t) + \epsilon_{22}(t) = \frac{\sigma_{11}^0 + \sigma_{22}^0}{2\mu}\,\frac{t_\star}{t_1}\frac{b}{a^2}\left[\left(1 - \frac{ac}{b}\right)\mathrm{e}^{-at/t_\star} + a\,\frac{t}{t_\star} + \frac{ac}{b} - 1\right]. \tag{2.42}$$

The initial conditions $\epsilon_{12}(0) = \epsilon_{11}(0) = \epsilon_{22}(0) = 0$ were used.

The above expressions specify the in-plane stress components for any time in the interval $0 \leq t \leq t_1$. The so calculated strain components are then used as initial conditions for the subsequent time interval $t_1 \leq t \leq t_2^-$, in which the stresses are held constant. Integration of eqs. (2.35)–(2.37) in this interval yields

$$\epsilon_{12}(t) = \frac{\sigma_{12}^0}{2\mu}\left[1 - e^{-(t-t_1)/t_\star}\right] + \epsilon_{12}(t_1)\,\mathrm{e}^{-(t-t_1)/t_\star}, \tag{2.43}$$

$$\epsilon_{11}(t) - \epsilon_{22}(t) = \frac{\sigma_{11}^0 - \sigma_{22}^0}{2\mu}\left[1 - \mathrm{e}^{-(t-t_1)/t_\star}\right] + \left[\epsilon_{11}(t_1) - \epsilon_{22}(t_1)\right]\mathrm{e}^{-(t-t_1)/t_\star}, \tag{2.44}$$

$$\epsilon_{11}(t) + \epsilon_{22}(t) = \frac{\sigma_{11}^0 + \sigma_{22}^0}{2\mu}\,\frac{b}{a}\left[1 - \mathrm{e}^{-a\,(t-t_1)/t_\star}\right] + \left[\epsilon_{11}(t_1) + \epsilon_{22}(t_1)\right]\mathrm{e}^{-a\,(t-t_1)/t_\star}. \tag{2.45}$$

The strain components $\epsilon_{12}(t_2^-)$, $\epsilon_{11}(t_2^-)$, and $\epsilon_{22}(t_2^-)$ are obtained from eqs. (2.43)–(2.45) by setting $t = t_2$.

2.3 Sudden loading or unloading

To begin we note that in a transversely isotropic Kelvin–Voigt viscoelastic constitutive model, a sudden change in stress, $[\sigma_{ij}]$ will result in a sudden change in strain $[\epsilon_{11}] = [\epsilon_{22}]$ and $[\epsilon_{33}]$ caused only be the spherical part of the stress change, *i.e.*, $[\sigma] = [\sigma_{kk}]/3$. This instantaneous response is purely elastic and from eq. (2.7) we can write for the spherical component of stress in x_1 or x_2 direction,

$$[\sigma] = 2(\lambda + \mu)[\epsilon_{11}] + (\lambda + \alpha)[\epsilon_{33}]\,, \tag{2.46}$$

and for the spherical component of stress in x_3 direction,

$$[\sigma] = 2(\lambda + \alpha)[\epsilon_{11}] + f[\epsilon_{33}]\,, \tag{2.47}$$

where

$$f = \lambda + 4\mu_0 - 2\mu + 2\alpha + \beta\,. \tag{2.48}$$

Solving eqs. (2.46) and (2.47) for $[\epsilon_{11}]$ and $[\epsilon_{33}]$, we obtain

$$[\epsilon_{11}] = [\epsilon_{22}] = \frac{\lambda + \alpha - f}{2[(\lambda+\alpha)^2 - (\lambda+\mu)f]}\,[\sigma]\,, \quad [\epsilon_{33}] = \frac{\alpha - \mu}{(\lambda+\alpha)^2 - (\lambda+\mu)f}\,[\sigma]\,. \tag{2.49}$$

Returning to the considered loading history eq. (2.39), the sudden removal of the stresses $\sigma^0_{11}, \sigma^0_{22}$ and σ^0_{12} gives rise to

$$[\sigma] = -\frac{1}{3}\,(\sigma^0_{11} + \sigma^0_{22}). \tag{2.50}$$

Therefore, from (2.49),

$$\epsilon_{11}(t_2^+) = \epsilon_{11}(t_2^-) - \frac{\lambda + \alpha - f}{6[(\lambda+\alpha)^2 - (\lambda+\mu)f]}\,(\sigma^0_{11} + \sigma^0_{22})\,, \tag{2.51}$$

$$\epsilon_{22}(t_2^+) = \epsilon_{22}(t_2^-) - \frac{\lambda + \alpha - f}{6[(\lambda+\alpha)^2 - (\lambda+\mu)f]}\,(\sigma^0_{11} + \sigma^0_{22})\,, \tag{2.52}$$

$$\epsilon_{33}(t_2^+) = \epsilon_{33}(t_2^-) - \frac{\alpha - \mu}{3[(\lambda+\alpha)^2 - (\lambda+\mu)f]}\,(\sigma^0_{11} + \sigma^0_{22})\,, \tag{2.53}$$

$$\epsilon_{12}(t_2^+) = \epsilon_{12}(t_2^-)\,. \tag{2.54}$$

Finally, for $t \geq t_2^+$ the stresses are zero and eqs. (2.35)–(2.37) directly yield

$$\epsilon_{12}(t) = \mathrm{e}^{-(t-t_2)/t_\star}\,\epsilon_{12}(t_2)\,, \tag{2.55}$$

$$\epsilon_{11}(t) - \epsilon_{22}(t) = \mathrm{e}^{-(t-t_2)/t_\star}\,[\epsilon_{11}(t_2^+) - \epsilon_{22}(t_2^+)]\,, \tag{2.56}$$

$$\epsilon_{11}(t) + \epsilon_{22}(t) = \mathrm{e}^{-a\,(t-t_2)/t_\star}\,[\epsilon_{11}(t_2^+) + \epsilon_{22}(t_2^+)]\,. \tag{2.57}$$

Note that the strain difference in eq. (2.56) is $\epsilon_{11}(t_2^+) - \epsilon_{22}(t_2^+) = \epsilon_{11}(t_2^-) - \epsilon_{22}(t_2^-)$, which follows via subtracting eq. (2.52) from eq. (2.51).

2.4 Prescribed strain histories

If the in-plane histories $\epsilon_{12}(t), \epsilon_{11}(t)$, and $\epsilon_{22}(t)$ are prescribed, still under conditions of plane stress, the governing equations for in-plane stress components are

$$\sigma_{12} = 2\mu\,(t_\star\dot{\epsilon}_{12} + \epsilon_{12})\,, \tag{2.58}$$

$$\sigma_{11} - \sigma_{22} = 2\mu\,[t_\star(\dot{\epsilon}_{11} - \dot{\epsilon}_{22}) + (\epsilon_{11} - \epsilon_{22})]\,, \tag{2.59}$$

$$t_\star(\dot{\sigma}_{11} + \dot{\sigma}_{22}) + \frac{b}{c}\,(\sigma_{11} + \sigma_{22}) = \frac{2\mu}{c}\,[t_\star(\dot{\epsilon}_{11} + \dot{\epsilon}_{22}) + a\,(\epsilon_{11} + \epsilon_{22})]\,. \tag{2.60}$$

The last equation is a differential equation for the sum in in-plane normal stresses, which has the solution

$$\begin{aligned}\sigma_{11}(t) + \sigma_{22}(t) = \mathrm{e}^{-b\,(t-t_0)/ct_\star}\Big\{\frac{2\mu}{c}\int_{t_0}^{t}\mathrm{e}^{b\,(\tau-t_0)/ct_\star}\Big(\frac{a}{t_\star}\,[\epsilon_{11}(\tau) + \epsilon_{22}(\tau)] \\ + \dot{\epsilon}_{11}(\tau) + \dot{\epsilon}_{11}(\tau)\Big)\mathrm{d}\,\tau + \sigma_{11}(t_0) + \sigma_{22}(t_0)\Big\}.\end{aligned} \tag{2.61}$$

In case of 3-D elastic isotropy, the moduli $\alpha = \beta = 0$ and $\mu_0 = \mu$, and the elastic part of the stress is related to strain as

$$\sigma^{\mathrm{e}}_{ij} = \lambda\epsilon_{kk}\delta_{ij} + 2\mu\epsilon_{ij}\,. \tag{2.62}$$

All the results of the previous section apply with the following specifications:

$$\begin{gathered}k = 3\lambda + 2\mu\,, \quad f = \lambda + 2\mu\,, \\ a = 1\,,\;\; b = \frac{\lambda + 2\mu}{3\lambda + 2\mu}\,,\;\; c = \frac{4\mu}{3(3\lambda + 2\mu)}\,.\end{gathered} \tag{2.63}$$

3 Orthotropic membranes

The mechanical response of a viscoelastic biological membrane reinforced by two sets of intersecting aligned fibers and subjected to biaxial in-plane loading is described *via* an orthotropic viscoelastic theory. The basic configuration is described in Fig. 5. The principal axes of orthotropy are along the unit vectors **a** and **b** that lie between the unit vectors **m** and **n** along the sets of fibers. With the angle ϕ as defined in Fig. 5, we note that $\mathbf{a} = (\mathbf{m} + \mathbf{n})/(2\cos\phi)$ and $\mathbf{b} = (\mathbf{m} - \mathbf{n})/(2\sin\phi)$.

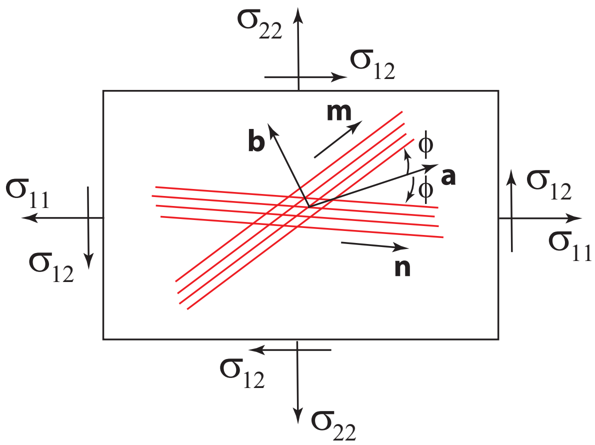

Figure 5: Membrane reinforced by two sets of parallel fibers oriented by the angles $\pm\phi$ with respect to an axis of orthotropic symmetry, $\mathbf{a}$. The fibers are aligned along the unit vectors $\mathbf{m}$ and $\mathbf{n}$.

The Cauchy stress tensor will again be decomposed into viscous and elastic parts as in the previous sections, consistent with a 3-dimensional generalization of the Kelvin model. The viscous part of the stress accounts for the time-dependent creep aspects of the membrane response; this is assumed to be purely deviatoric and related to the time rate of strain *via* eq. (2.4). The elastic part of the stress accounts for the anisotropic elastic properties of the fiber reinforced membrane matrix [15, 16]. From the representation theorems for orthotropic tensor functions [12, 13] this part of the stress can be expressed in terms of the strain tensor $\boldsymbol{\epsilon}$ and the director vectors $\mathbf{a}$ and $\mathbf{b}$ as

$$\begin{aligned}\sigma^{\mathrm{e}}_{ij} &= (\lambda\epsilon_{kk} + \alpha_1\epsilon_a + \alpha_2\epsilon_b)\,\delta_{ij} + 2\mu\epsilon_{ij} \\ &\quad + (\alpha_1\epsilon_{kk} + \beta_1\epsilon_a + \beta_3\epsilon_b)\,a_i a_j + (\alpha_2\epsilon_{kk} + \beta_3\epsilon_a + \beta_2\epsilon_b)\,b_i b_j \\ &\quad + 2\mu_1(a_i a_k\epsilon_{kj} + a_j a_k\epsilon_{ki}) + 2\mu_2(b_i b_k\epsilon_{kj} + b_j b_k\epsilon_{ki}).\end{aligned} \tag{3.1}$$

The longitudinal strains in the directions of the principal axes of orthotropy are $\epsilon_a = a_k a_l \epsilon_{kl}$ and $\epsilon_b = b_k b_l \epsilon_{kl}$. The nine elastic constants are $\lambda, \mu, \mu_1, \mu_2, \alpha_1, \alpha_2$ and $\beta_1, \beta_2, \beta_3$ [17].

If the coordinate axes are chosen to lie along the directions of the principal axes of orthotropy, then $a_i = \delta_{i1}$ and $b_i = \delta_{i2}$, so that $\epsilon_a = \epsilon_{11}$ and $\epsilon_b = \epsilon_{22}$

and eq. (3.1) becomes

$$\begin{aligned}\sigma^{\mathrm{e}}_{ij} &= (\lambda\epsilon_{kk} + \alpha_1\epsilon_{11} + \alpha_2\epsilon_{22})\,\delta_{ij} + 2\mu\epsilon_{ij} \\ &+ (\alpha_1\epsilon_{kk} + \beta_1\epsilon_{11} + \beta_3\epsilon_{22})\,\delta_{i1}\delta_{j1} + (\alpha_2\epsilon_{kk} + \beta_3\epsilon_{11} + \beta_2\epsilon_{22})\,\delta_{i2}\delta_{j2} \\ &+ 2\mu_1(\delta_{i1}\epsilon_{j1} + \delta_{j1}\epsilon_{i1}) + 2\mu_2(\delta_{i2}\epsilon_{j2} + \delta_{j2}\epsilon_{i2})\,.\end{aligned} \tag{3.2}$$

For the in-plane stresses we find that

$$\sigma_{11} = 2\eta\Big(\dot\epsilon_{11} - \frac{1}{3}\,\dot\epsilon_{kk}\Big) + (\lambda + \alpha_1)\epsilon_{kk} + (2\mu + 4\mu_1 + \alpha_1 + \beta_1)\epsilon_{11} + (\alpha_2 + \beta_3)\epsilon_{22}\,, \tag{3.3a}$$

$$\sigma_{22} = 2\eta\Big(\dot\epsilon_{22} - \frac{1}{3}\,\dot\epsilon_{kk}\Big) + (\lambda + \alpha_2)\epsilon_{kk} + (2\mu + 4\mu_2 + \alpha_2 + \beta_2)\epsilon_{22} + (\alpha_1 + \beta_3)\epsilon_{11}\,, \tag{3.3b}$$

$$\sigma_{12} = 2\eta\dot\epsilon_{12} + 2(\mu + \mu_1 + \mu_2)\epsilon_{12}\,. \tag{3.4}$$

3.1 Plane stress

For the plane stress the conditions $\sigma_{31} = \sigma_{32} = 0$ imply that $\epsilon_{31} = \epsilon_{32} = 0$, provided their initial values are set to zero. The vanishing of the normal stress component $\sigma_{33} = 0$ implies

$$2\eta\left(\dot\epsilon_{33} - \frac{1}{3}\,\dot\epsilon_{kk}\right) + \lambda\epsilon_{kk} + \alpha_1\epsilon_{11} + \alpha_2\epsilon_{22} + 2\mu\,\epsilon_{33} = 0. \tag{3.5}$$

If ϵ_{11} and ϵ_{22} were known, this would be a differential equation for ϵ_{33}. Its integration, however, can be circumvented by noting that

$$\sigma_{kk} = (3\lambda{+}2\mu{+}\alpha_1{+}\alpha_2)\epsilon_{kk}{+}(4\mu_1{+}3\alpha_1{+}\beta_1{+}\beta_3)\epsilon_{11}{+}(4\mu_2{+}3\alpha_2{+}\beta_2{+}\beta_3)\epsilon_{22}\,. \tag{3.6}$$

This leads to

$$\epsilon_{kk} = \frac{1}{k}\,(\sigma_{11} + \sigma_{22} - k_1\epsilon_{11} - k_2\epsilon_{22})\,, \tag{3.7}$$

where

$$k = 3\lambda{+}2\mu{+}\alpha_1{+}\alpha_2,\ k_1 = 4\mu_1{+}3\alpha_1{+}\beta_1{+}\beta_3,\ k_2 = 4\mu_2{+}3\alpha_2{+}\beta_2{+}\beta_3. \tag{3.8}$$

For given stresses, if ϵ_{11} and ϵ_{22} were determined, eq. (3.7) would specify the remaining normal strain component as

$$\epsilon_{33} = \frac{1}{k}\,[\sigma_{11} + \sigma_{22} - (k + k_1)\epsilon_{11} - (k + k_2)\epsilon_{22}]\,. \tag{3.9}$$

Moreover, when eq. (3.7) is substituted into eq. (3.3), we obtain the system of coupled differential equations

$$\begin{aligned} a_{11}\dot{\epsilon}_{11} + a_{12}\dot{\epsilon}_{22} + b_{11}\epsilon_{11} + b_{12}\epsilon_{22} &= f_1(t), \\ a_{21}\dot{\epsilon}_{11} + a_{22}\dot{\epsilon}_{22} + b_{21}\epsilon_{11} + b_{22}\epsilon_{22} &= f_2(t), \end{aligned} \tag{3.10}$$

with the coefficients

$$\begin{aligned} a_{11} &= \frac{2\eta}{3}\left(3 + \frac{k_1}{k}\right), \quad a_{22} = \frac{2\eta}{3}\left(3 + \frac{k_2}{k}\right), \\ a_{12} &= \frac{2\eta}{3}\,\frac{k_2}{k}\,, \quad a_{21} = \frac{2\eta}{3}\,\frac{k_1}{k}\,, \\ b_{11} &= 2\mu + 4\mu_1 + \alpha_1 + \beta_1 - (\lambda + \alpha_1)\,\frac{k_1}{k}\,, \\ b_{22} &= 2\mu + 4\mu_2 + \alpha_2 + \beta_2 - (\lambda + \alpha_2)\,\frac{k_2}{k}\,, \\ b_{12} &= \alpha_2 + \beta_3 - (\lambda + \alpha_2)\,\frac{k_2}{k}\,, \\ b_{21} &= \alpha_1 + \beta_3 - (\lambda + \alpha_2)\,\frac{k_1}{k}\,. \end{aligned} \tag{3.11}$$

In addition,

$$\begin{aligned} f_1(t) &= \sigma_{11} - \frac{\lambda + \alpha_1}{k}\,(\sigma_{11} + \sigma_{22}) + \frac{2\eta}{3k}\,(\dot{\sigma}_{11} + \dot{\sigma}_{22})\,, \\ f_2(t) &= \sigma_{22} - \frac{\lambda + \alpha_2}{k}\,(\sigma_{11} + \sigma_{22}) + \frac{2\eta}{3k}\,(\dot{\sigma}_{11} + \dot{\sigma}_{22})\,. \end{aligned} \tag{3.12}$$

Given stress histories for $\sigma_{11}(t)$ and $\sigma_{22}(t)$, eqs. (3.10) can be integrated to obtain the corresponding strain histories for $\epsilon_{11}(t)$ and $\epsilon_{22}(t)$. The numerical results will be presented in Part III, upon completion of the experimental determination of material parameters.

4 Discussion and conclusions

Two classes of linear viscoelastic models have been developed for thin biological membranes characterized by being reinforced by stiff fibers. The original impetus for this development was to provide an adequate model to simulate the mechanical response of the interlayers of the nacreous part of the shell of mollusks such as *Haliotis rufescens* (the red abalone). In Part I of this series it was described that the chitin fibril reinforcement within such interlayers are arranged in a predominantly unidirectional manner but with regions in which the fibrils are oriented at directions rotated by small angles, called ϕ above. Thus the interlayers are indeed anisotropic and may be reasonably described by the orthotropic model developed in Section 3. There are, however, many examples of biological membranes that are typically described as being transversely isotropic such as, for example, the cell wall of fungal cells, *e.g.* yeast cells. Figure 6 shows a schematic of the fungal cell wall as rendered from descriptions given by, *inter alia*, Herrera *et al.* [18, 19] or by [20, 21]. As in-

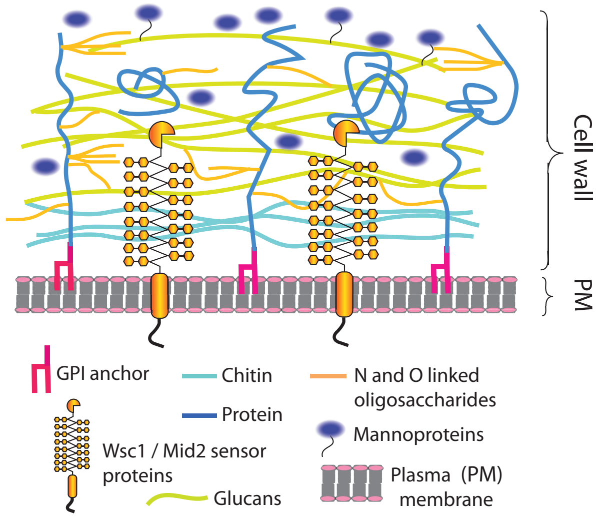

Figure 6: Representation of the structure of the fungal cell wall, depicting the mannan-glucan-chitin containing PM (plasma membrane) layers.

dicated, the fungal cell wall is characterized as being organized in layers that are, in turn, characterized by different macro-molecular constitution. The inner most layer is rich in chitin fibrils that are thought to be arranged in a transversely isotropic manner as described by the model developed in Section 2. Thus it is believed that this model may well be applicable to the structural layers in the fungal cell wall. Such applications are indeed underway.

In Part III of this series we indeed apply the models developed herein to two quite different cases that are the analysis of the interlayers of nacre in *Haliotis rufescens* and to the deformation of the fungal cell wall, or actually of the fungal cell itself. In the case of nacre in the inner shell of *Haliotis rufescens*, the goal will be to understand how its brick wall tile structure and the particular viscoelastic constitutive response of the interlayer matrix impart the well documented properties of nacre. In the case of the fungal cell wall, a key goal will be the understand the role of the individual viscoelastic layers in determining the deformation states within the cell wall and of the entire cell itself.

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Viskoelastično ponašanje anizotropnih bioloških membrana. Deo II: konstitutivni modeli

U prvom delu ove serije [7] opisana je struktura hitinskih vlakana u interlamelama sedefa *Haliotis rufescens* (crvena školjka). Opisano je kako se ova lamelarna struktura može modelirati kao viskoelastični kompozitni materijal ojačan hitinskim vlaknima koja su često približno jednosmerne arhitekture. Sprovedeno je mehaničko testiranje uzoraka dobijenih procesom demineralizacije. U ovom delu serije formiran je viskoelastični konstitutivni model za transverzno izotropnu ili ortotropnu lamelarne strukturu, saglasno analizi raspodele hitinskih vlakana iz dela I. Dio III serije biće posvećen primeni razvijenog konstitutivnog modela na analizu experimentalnih rezultata posmatrane vrste bioloških membrana pod dejstvom mehaničkog opterećenja.

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