Soft Matter

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Supplementary Material
From a distance: Shuttleworth revisited

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1 Surface Projection

To project fields defined over the domain Ω_0 onto the surface S_0, we introduce the surface projection tensor for a tangent surface with outward unit normal $\boldsymbol{N}$ as

$$\mathbb{P} = \boldsymbol{I} - \boldsymbol{N} \otimes \boldsymbol{N}, \tag{S.1}$$

where $\boldsymbol{I}$ is the second-order identity tensor[1]. A smooth vector field $\boldsymbol{v}$ (e.g. a displacement field) and a smooth second-order tensor field $\boldsymbol{T}$ (e.g. a stress field) are projected onto the surface as

$$\boldsymbol{v}_s = \mathbb{P}\boldsymbol{v} \quad \text{and} \quad \boldsymbol{T}_s = \mathbb{P}\boldsymbol{T}\mathbb{P}. \tag{S.2}$$

Furthermore, the surface gradient of a vector field follows from

$$\boldsymbol{\nabla}_s \boldsymbol{\phi} = \mathbb{P}\boldsymbol{\nabla}\boldsymbol{\phi} \quad \text{and} \quad \boldsymbol{\nabla}_s \boldsymbol{v} = (\boldsymbol{\nabla}\boldsymbol{v})\mathbb{P}. \tag{S.3}$$

As an example, the surface deformation gradient follows as

$$\boldsymbol{F}_s = \boldsymbol{I} + \mathbb{P}\nabla_s \boldsymbol{u}. \tag{S.4}$$

Note that within the manuscript, the projection tensor is dropped for simplicity as in previous studies[1].

2 Finite Kinematics

2.1 General setting

The non-linear Shuttleworth equations correspond to the expressions of the stress measures as a function of the surface energy in the current configuration W^{c}. In this setting, the second Piola-Kirchhoff stress follows as

$$\boldsymbol{S}_s = W^{\mathrm{c}} \frac{\partial J_s}{\partial \boldsymbol{E}_s} + J_s \frac{\partial W^{\mathrm{c}}}{\partial \boldsymbol{E}_s}. \tag{S.5}$$

Noting that $J_s = \det(\boldsymbol{F}_s) = \sqrt{\det(2\boldsymbol{E}_s + \boldsymbol{I})}$, and using the relationship

$$\frac{\partial \det \boldsymbol{M}}{\partial \boldsymbol{M}} = (\det \boldsymbol{M})\boldsymbol{M}^{-T} \tag{S.6}$$

for any invertible tensor $\boldsymbol{M}$, we obtain the non-linear Shuttleworth equation for the second Piola-Kirchhoff stress tensor

$$\boldsymbol{S}_s = W^{\mathrm{c}} J_s \left(\boldsymbol{F}_s^{\mathrm{T}} \boldsymbol{F}_s\right)^{-1} + J_s \frac{\partial W^{\mathrm{c}}}{\partial \boldsymbol{E}_s}. \tag{S.7}$$

Equivalently, the first Piola-Kirchhoff stress tensor reads

$$\boldsymbol{P}_s = W^{\mathrm{c}} J_s \boldsymbol{F}_s \left(\boldsymbol{F}_s^{\mathrm{T}} \boldsymbol{F}_s\right)^{-1} + J_s \boldsymbol{F}_s \frac{\partial W^{\mathrm{c}}}{\partial \boldsymbol{E}_s}, \tag{S.8}$$

and the Cauchy stress is given as

$$\boldsymbol{\sigma}_s = W^{\mathrm{c}} \mathbf{I} + \boldsymbol{F}_s \frac{\partial W^{\mathrm{c}}}{\partial \boldsymbol{E}_s} \boldsymbol{F}_s^{\mathrm{T}}. \tag{S.9}$$

2.2 Surface stress-strain relations

We can tailor stress measures to a specific constitutive material model by choosing, e.g., the St. Venant-Kirchhoff model

$$W^{\mathrm{R}}(\boldsymbol{E}_s) = \gamma J_s + \mu_s \operatorname{tr}(\boldsymbol{E}_s \boldsymbol{E}_s) + \frac{1}{2}\lambda_s (\operatorname{tr}(\boldsymbol{E}_s))^2. \tag{S.10}$$

In addition to relation (S.6), we note that the derivative of a tensor $\boldsymbol{M}$ with respect to that tensor is simply $\frac{\partial \operatorname{tr}(\boldsymbol{M})}{\partial \boldsymbol{M}} = \boldsymbol{I}$. Using the chain rule, the different stress measures under consideration then follow as

$$\begin{aligned} \boldsymbol{S}_s = \frac{\partial W^{\mathrm{R}}}{\partial \boldsymbol{E}_s} &= \boldsymbol{F}^{-1} \frac{\partial W^{\mathrm{R}}}{\partial \boldsymbol{F}_s} \\ &= \gamma J_s (\boldsymbol{F}_s^T \boldsymbol{F}_s)^{-1} + 2\mu_s \boldsymbol{E}_s + \lambda_s \operatorname{tr}(\boldsymbol{E}_s)\boldsymbol{I}, \end{aligned} \tag{S.11}$$

and the relations

$$\boldsymbol{P}_s = \boldsymbol{F}_s \boldsymbol{S}_s \text{ and } \boldsymbol{\sigma}_s = J_s^{-1} \boldsymbol{P}_s \boldsymbol{F}_s^T \tag{S.12}$$

lead to the main text equations (7) and (8).

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3 Linearization

For linearized surface relaxation, we express all quantities at play, J, $\boldsymbol{F}$ and $\boldsymbol{E}$ at first order in displacement gradient $\boldsymbol{\nabla u}$ or in strains $\boldsymbol{\varepsilon}$. The deformation gradient is readily defined at first order, $\boldsymbol{F} = \boldsymbol{I} + \boldsymbol{\nabla u}$, the Green-Lagrange strain tensor at first order simplifies to the linear strains $\boldsymbol{E} = \boldsymbol{\varepsilon} + \mathscr{O}(\boldsymbol{\varepsilon}^2)$, and the local area change simplifies to $J = [1 + \mathrm{tr}(\boldsymbol{\varepsilon})] + \mathscr{O}(\boldsymbol{\varepsilon}^2)$.

For finite surface relaxation, we expand all terms to linear order in $\boldsymbol{\nabla u}^0$, whereas terms involving $\boldsymbol{F}^*$ stay finite. The deformation gradient then simplifies to $\boldsymbol{F} = (\boldsymbol{I} + \boldsymbol{\nabla u}^0)\boldsymbol{F}^* + \mathscr{O}([\boldsymbol{\nabla u}^0]^2)$, the Green-Lagrange tensor to $\boldsymbol{E} = \boldsymbol{E}^* + \boldsymbol{F}^*\boldsymbol{\varepsilon}^0(\boldsymbol{F}^*)^T + \mathscr{O}([\boldsymbol{\nabla u}^0]^2)$, and the local area change to $J = \left[1 + \mathrm{tr}(\boldsymbol{\varepsilon}^0)\right] J^* + \mathscr{O}(\boldsymbol{\varepsilon}^2)$.

4 Finite surface relaxation in the absence of surface shear

In the absence of surface shear, the surface deformation gradient is diagonal

$$\mathbf{F}_\mathrm{s} = \begin{pmatrix} F_{\mathrm{s}\|} & 0 \\ 0 & F_{\mathrm{s}\perp} \end{pmatrix}, \tag{S.13}$$

with $F_\mathrm{si} = 1 + \varepsilon_\mathrm{si}$, where $i = \|$ or $i = \perp$ denote the two principal directions. Assuming the principal directions of the surface relaxation and of the imposed deformation coincide, we obtain the generic expression for the Cauchy principal stresses

$$\begin{aligned} \bar{\sigma}_\mathrm{si} =& \gamma + \bar{\sigma}^*_\mathrm{si} + 2\mu_\mathrm{s}\left((F^*_\mathrm{si})^4 + 2(F^*_\mathrm{si})^2 E^*_\mathrm{si}\right)\varepsilon^0_\mathrm{si} \\ & + \lambda_\mathrm{s}\left((F^*_\mathrm{si})^4 + 2(F^*_\mathrm{si})^2(E^*_\mathrm{si} + E^*_\mathrm{sj})\right)\varepsilon^0_\mathrm{si} \\ & + \lambda_\mathrm{s}(F^*_\mathrm{si}F^*_\mathrm{sj})^2\varepsilon^0_\mathrm{sj}, \end{aligned} \tag{S.14}$$

with $(i,j) = (\|,\perp)$ or $(i,j) = (\perp,\|)$ the principal directions, $\bar{\sigma}^*_\mathrm{si} = (2\mu_\mathrm{s} + \lambda_\mathrm{s})(F^*_\mathrm{si})^2 E^*_\mathrm{si} + \lambda_s (F^*_\mathrm{si})^2 E^*_\mathrm{sj}$ the surface relaxation stress contribution and $E_\mathrm{si} = \varepsilon_\mathrm{si}(2 + \varepsilon_\mathrm{si})/2$ the diagonal components of the Green-Lagrange strain tensor.

5 Relaxing cylinder

The total energy of the relaxing cylinder,

$$W_\mathrm{tot} = \min_{\lambda^*}\left(S_0\,\gamma + V_0\,W^\mathrm{R}(\lambda^*)\right), \tag{S.15}$$

comprises a surface contribution $S_0\,\gamma$ and a bulk contribution $V_0\,W^\mathrm{R}(\lambda^*)$, where S_0 and V_0 are the surface and volume of the relaxed cylinder, respectively. Here, $W^\mathrm{R}(\lambda^*) = \mu(2\lambda^{*2} + \lambda^{*-4} - 3)/2$ derives from the strain energy density of a Neo-Hookean incompressible material. The solutions to Eq. (S.15) are

$$\lambda^{*3}_\pm = \frac{\gamma/R \pm \sqrt{(\gamma/R)^2 + 4\mu^2}}{4\mu}, \tag{S.16}$$

from which we discard λ^*_- for physical reasons ($\lambda^* > 0$ when $L_{ec}/R \ll 1$), which leads to Eq. (19).

Notes and references

1 S. Krichen, L. Liu and P. Sharma, *JMPS*, 2019, **127**, 332–357.