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A 3-D stress-strain relation for glassy polymers

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Summary.

In constitutive equations for finite elastic-plastic deformation of polymers the elastic part of the deformation is usually formulated by assuming an isotropic relation between the Jaumann rate of the Cauchy-stress tensor and the rate of strain tensor. The Jaumann-stress rate however is known to display aphysical behaviour in the elastic region. In this paper we will derive a "compressible-Leonov model" which reduces to the Jaumann-stress rate in the limit of complete plastic deformation, but is also capable of giving a correct description of the elastic region.

In polymer rheology it is now recognized that constitutive equations are most naturally formulated using evolution equations for state variables^{1,2} (the variables that determine the Helmholtz-free energy (A) of the system). Jongschaap² derived a formalism which describes how external rate variables (the velocity gradient \underline{L}) and external forces (the Cauchy-stress tensor $\underline{\sigma}$) are coupled to the rate of change of the state variables and the internal thermodynamic forces (derivatives of the free energy with respect to the state variables). An important concept in this formalism is that of macroscopic time reversal where one determines to what extend the rate of change of the state variables and the stress tensor $\underline{\sigma}$ are effected by a reversal of the macroscopic external velocity gradient \underline{L} . Here it should be noted that any function $f(\underline{L})$ may be decomposed in an even part f^+ and an odd part f^- according to:

$$f(\underline{L}) = f^+(\underline{L}) + f^-(\underline{L}) = \frac{1}{2}(f(\underline{L}) + f(-\underline{L})) + \frac{1}{2}(f(\underline{L}) - f(-\underline{L})) \quad (\text{thus } \underline{L} = \underline{L} \text{ per definition})$$

From the notion that the state variables are even per definition and due to the second law of thermodynamics the dissipation function² is also even, the formalism describes how a constitutive equation can be decomposed as follows:

$$\begin{pmatrix} \underline{\sigma} \\ \underline{\dot{S}} \end{pmatrix} = \begin{pmatrix} \underline{\eta} & -\underline{A}^T \\ \underline{\Lambda} & \underline{\beta} \end{pmatrix} : \begin{pmatrix} \underline{L} \\ -\underline{M} \end{pmatrix}, \quad \underline{M} = \rho \left(\frac{\partial A}{\partial \underline{S}} \right) \quad (1)$$

Where it is assumed that the state variable determining the free energy is a second order tensor \underline{S} with an associated thermodynamic force \underline{M} (ρ is the density). The fourth-order tensor $\underline{\eta}$, $\underline{\Lambda}$ and $\underline{\beta}$ are even with respect to \underline{L} .

One important result is that the reversible (elastic) part of the stress tensor, $\underline{\sigma}^*$, is coupled to the reversible

(odd) part of the rate of change of the state variable, \underline{S} :

$$\underline{\sigma}^{ev} = \underline{\sigma}^* = \underline{\Lambda}^T : \underline{M} = \underline{M} : \underline{\Lambda}$$

We will now use this formalism to derive a constitutive relation that gives a correct description of the elastic as well as the plastic behaviour of polymers assuming that the volume response remains pure elastic. We will use polycarbonate as a model system to compare some of the existing models.

In the case of isotropic elastic behaviour one can assume that the state variable determining the free energy is the left-Cauchy-Green-deformation tensor \underline{B} , $\underline{B} = \underline{F} \cdot \underline{F}^T$ (\underline{F} is the deformation gradient). Because of isotropy we then have:

$$A = A(I_B, II_B, III_B) \quad I_B = \text{tr}(\underline{B}), \quad II_B = \frac{1}{2}(I_B^2 - \text{tr}(\underline{B} \cdot \underline{B})), \quad III_B = \det(\underline{B})$$

The thermodynamic force therefore equals:

$$\underline{M} = \rho \left(\frac{\partial A}{\partial \underline{B}} \right) = \rho \left(\frac{\partial A}{\partial I_B} \frac{\partial I_B}{\partial \underline{B}} + \frac{\partial A}{\partial II_B} \frac{\partial II_B}{\partial \underline{B}} + \frac{\partial A}{\partial III_B} \frac{\partial III_B}{\partial \underline{B}} \right) = \rho (\alpha_1 \underline{I} + \alpha_2 (\text{tr}(\underline{B}) \underline{I} - \underline{B}) + \alpha_3 \det(\underline{B}) \underline{B}^{-1})$$

Here, α_i are the derivatives of the free energy with respect to the invariants of \underline{B} .

From kinematics we know that the upper convected or Truesdell derivative of \underline{B} equals $\underline{\dot{B}}$ and this leads us to the evolution equation for \underline{B} :

$$\underline{\dot{B}} = \underline{L} \cdot \underline{B} + \underline{B} \cdot \underline{L}^T = \underline{\Delta} : \underline{L} \quad ; \quad \Lambda_{ijkm} = \delta_{ik} B_{mj} + B_{im} \delta_{jk} \quad (2)$$

Since we have no dissipation \underline{n} and \underline{B} are \underline{Q} and the constitutive equation for elastic behaviour follows directly from (1) and (2):

$$\begin{cases} \underline{\sigma} = \underline{M} : \underline{\Lambda} = 2\rho (\alpha_3 \det(\underline{B}) \underline{I} + (\alpha_1 + \alpha_2 \text{tr}(\underline{B})) \underline{B} - \alpha_2 \underline{B}^2) \\ \underline{\nabla} \\ \underline{B} = 0 \end{cases}$$

This is the well-known expression for Green-elastic behaviour.

Because we want to develop an expression for elastic-plastic behaviour where the volume response is pure elastic, we now consider the case of pure elastic behaviour where the volume deformation is independent of the change of shape. Because of the independent volume deformation we assume that the free energy is determined by two state variables: the relative volume deformation J ($J = \det(\underline{E})$) (for convenience we will use J_I instead of J) and the left-Cauchy-Green deformation tensor at constant volume \underline{B}^* , $\underline{B}^* = J^{-2/3} \underline{B}$. In the case of isotropic behaviour we then have³:

$$A = A(J, I_{B^*}, II_{B^*}, \dots) \quad (III_{B^*} = \det(\underline{B}) = 1)$$

There are now two conjugated thermodynamic forces, one related to the volume deformation (\underline{N}) and one related to the shape deformation (\underline{M}):

$$\underline{N} = \rho \left(\frac{\partial A}{\partial J_I} \right) = \frac{1}{3} \rho \left(\frac{\partial A}{\partial J} \right) \underline{I} \quad \text{and} \quad \underline{M} = \rho \left(\frac{\partial A}{\partial \underline{B}^*} \right) = \rho (\alpha_1^* \underline{I} + \alpha_2^* (\text{tr}(\underline{B}^*) \underline{I} - \underline{B}^{*T}))$$

From kinematics we have:

$$\dot{J}\underline{I} = J \text{tr}(\underline{D})\underline{I} = J \underline{II} : \underline{L} \quad \text{and} \quad \underline{\dot{B}}^* = \underline{L}^d \cdot \underline{B}^* + \underline{B}^* \cdot \underline{L}^d = \underline{\Lambda}^* : \underline{L} \quad \left(\underline{\Lambda}^* = \underline{\Lambda} - \frac{2}{3} \underline{B}\underline{I} \right)$$

The constitutive equation therefore becomes:

$$\begin{cases} \underline{\sigma} = \underline{N} : \underline{J}\underline{I}\underline{I} + \underline{M} : \underline{\Lambda}^* = \rho_0 \left(\frac{\partial A}{\partial J} \right) \underline{I} + \rho \left(2\alpha_1^* \underline{B}^d + 2\alpha_2^* \text{tr}(\underline{B}^*) \underline{B}^d + 2\alpha_2^* (\underline{B}^*)^d \right) = \underline{\sigma}^h + \underline{\sigma}^d \\ \underline{\dot{B}}^* = \underline{L}^d \cdot \underline{B}^* + \underline{B}^* \cdot \underline{L}^d \\ \underline{J}\underline{I} = J \text{tr}(\underline{D})\underline{I} \end{cases} \quad (3)$$

As might be expected the hydrostatic stress is determined solely by the volume deformation, whereas the deviatoric stress is governed by \underline{B}^* .

The evolution equation for \underline{B}^* may also be written as:

$$\overset{\circ}{\underline{B}}^* = \underline{D}^d \cdot \underline{B}^* - \underline{B}^* \cdot \underline{D}^d \quad \text{where } \overset{\circ}{\underline{B}}^* \text{ is the Jaumann rate of } \underline{B}^* : \overset{\circ}{\underline{B}}^* = \underline{\dot{B}}^* - \underline{W} \cdot \underline{B}^* - \underline{B}^* \cdot \underline{W}^T$$

In case of plastic deformation we now assume that the accumulation of elastic strain (at constant volume) is reduced because of the existence of a (deviatoric) plastic strain rate \underline{D}_p . The evolution equation for \underline{B}^* therefore changes to (note that the plastic spin equals 0):

$$\overset{\circ}{\underline{B}}^* = (\underline{D}^d - \underline{D}_p) \cdot \underline{B}^* - \underline{B}^* \cdot (\underline{D}^d - \underline{D}_p) \quad (4)$$

For polymers in general and polycarbonate special it is known⁴ that the plastic-strain rate \underline{D}_p is well described by an Eyring-flow process:

$$\underline{\sigma}^d = 2\eta (II_{\sigma^d}) \underline{D}_p ; \quad \text{where} \quad \eta(II_{\sigma^d}) = A\tau_0 \frac{\sqrt{\frac{II_{\sigma^d}}{2\tau_0^2}}}{\sinh\left(\sqrt{\frac{II_{\sigma^d}}{2\tau_0^2}}\right)}$$

For polycarbonate we have $A = 8.6 \cdot 10^{20} \text{ s}$ and $\tau_0 = 0.856 \text{ MPa}^4$.

A constitutive equation for elastic-plastic behaviour with an elastic-volume response is now obtained by combining the equation for elastic behaviour (3) with the new evolution equation for \underline{B}^* (4). We will assume that $\alpha_2^* = 0$ (physical linear elastic behaviour) and identifying $2\rho\alpha_1^*$ with the shear modulus G and $\rho_0(\partial A/\partial J)$ with $K(J-1)$ (where K is the bulkmodulus) we have (at small volume deformations, $J \approx 1$):

$$\begin{aligned} \underline{\sigma} &= K(J-1)\underline{I} + G\underline{B}_e^* \\ \overset{\circ}{\underline{B}}_e^* &= (\underline{D}^d - \underline{D}_p) \cdot \underline{B}_e^* + \underline{B}_e^* \cdot (\underline{D}^d - \underline{D}_p) \\ \underline{J}\underline{I} &= \text{tr}(\underline{D})\underline{I} \\ \underline{\sigma}^d &= 2\eta(II_{\sigma^d})\underline{D}_p \end{aligned}$$

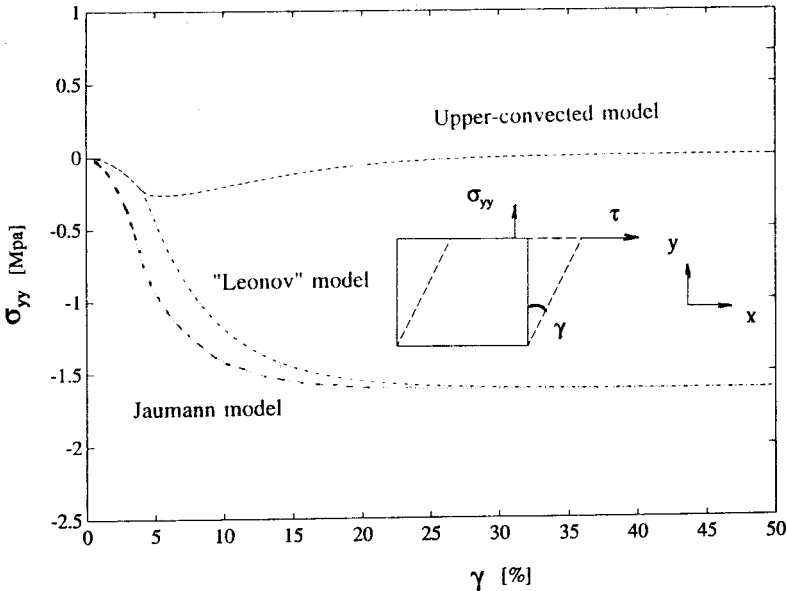
We can now compare our model with the widely used Jaumann-stress-rate equation⁵:

$$\frac{\eta(II_{\sigma^d})}{G} \overset{\circ}{\underline{\sigma}} + \underline{\sigma}^d = 2\eta(II_{\sigma^d})\underline{D} \quad \text{together with} \quad \text{tr}(\underline{\sigma}) = 3K \text{tr}(\underline{D})$$

and the upper-convected equivalent of this:

$$\frac{\eta(I_T)}{G} \frac{\nabla}{I} + \underline{T}^d = 2\eta(I_T)\underline{D} ; \underline{\sigma}^d = \underline{T}^d \text{ together with } \text{tr}(\underline{\sigma}) = 3K\text{tr}(\underline{D})$$

As an example we will calculate the normal-stress response of the three models for a plane-stress-shear deformation (plane-stress condition in the z-direction). The Young's modulus of polycarbonate $E = 2400 \text{ MPa}$ and the Poisson constant $\nu = 0.33$.



Calculations show that the three models give virtually the same shear-stress response. The normal stresses however differ, as can be seen from the figure.

It is known⁶ that the upper-convected model gives a good description of the elastic behaviour, but apparently fails to describe the (experimentally observed) normal stress during plastic deformation. The Jaumann model on the other hand gives a reasonable description of the normal stress, but is not capable of capturing the correct elastic behaviour⁶. Our "compressible Leonov model" provides a natural link between these two extremes.

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