

Constitutive modelling of Rubber-like materials

A thesis submitted
in partial fulfillment of the requirements
for the Degree of
Master of Technology

by
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Why do we need this?

Applications: Tires and tubes industry, Vibration dampers, Window seals in aircraft and spacecrafts and many more...

Saturated Rubber

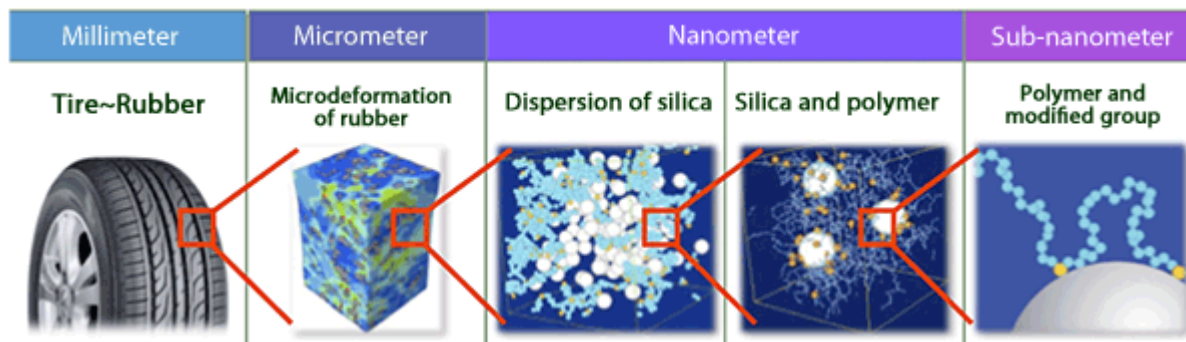
Polyacrylic rubber, Silicone rubber, EPM (ethylene propylene rubber), EPDM (ethylene propylene diene rubber)

Unsaturated Rubber

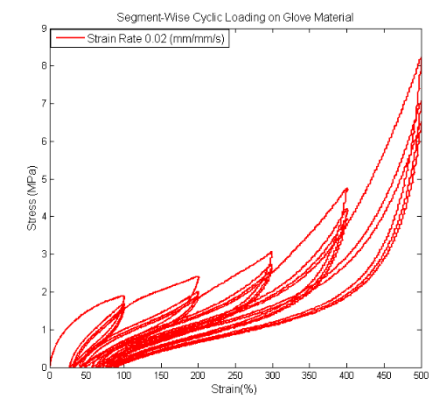
Natural isoprene (natural rubber), polybutadiene, styrene butadiene, nitrile rubber

Properties:

- Large deformation
- Enhanced properties by adding fillers.
- Long chain polymers
- Branching
- Crystallization induced by thermal/mechanical loading

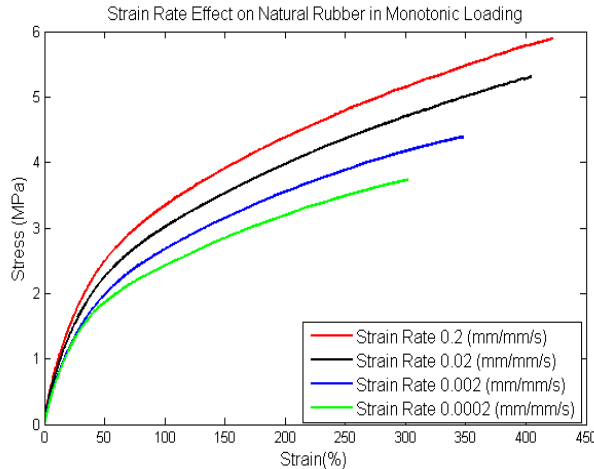


Multi-scale material pattern [Toyo tires]



Stress-Strain (Natural Rubber)

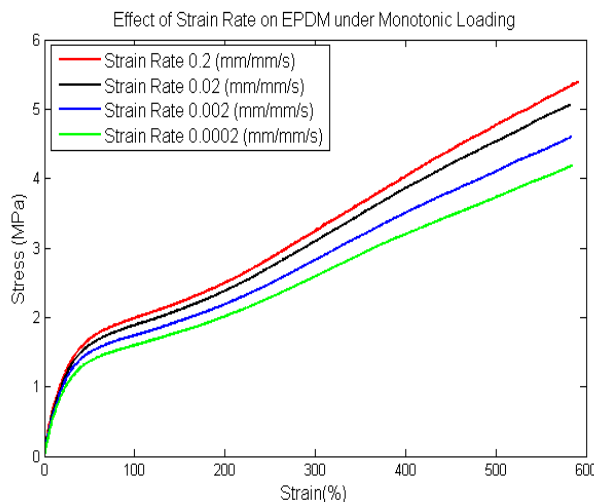
Experiment: Monotonic loading



Natural Rubber:

- 30% Recycled rubber
- 30% Calcium carbonate
- 5% Carbon black

- ❖ Non-linear behaviour
- ❖ Strain Rate dependence
- ❖ Large range of strain rate
- ❖ Similar rate-dependent behaviour irrespective of amount of fillers



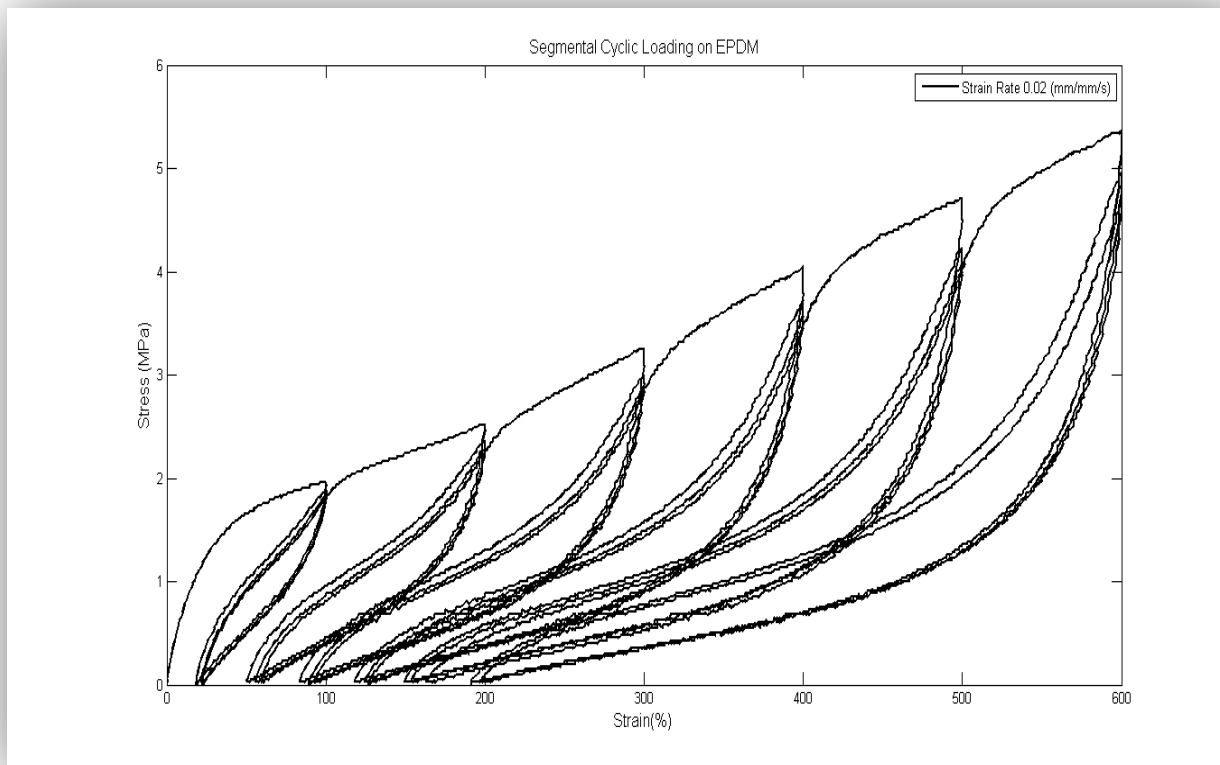
EPDM:

- 10% CaCO₃
- 5% Carbon Black

[Ref: Thesis by AS Khan (2015)]

Experiment: Segment-wise cyclic loading

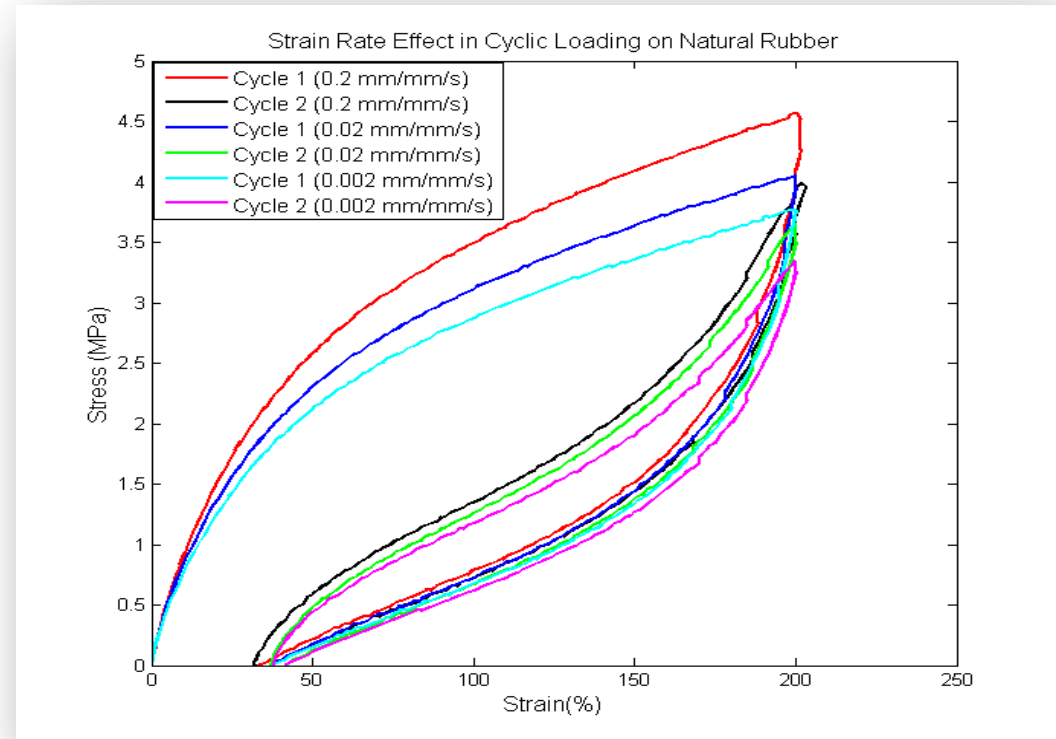
Mullin's Effect: Virgin material curve envelopes stress-softened material curve



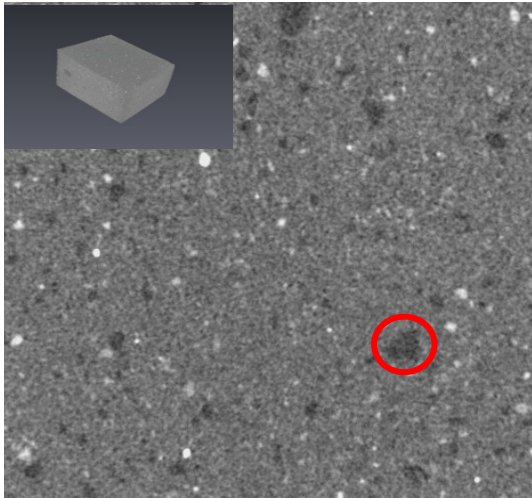
- ❖ 40% unrecovered strain set after a period of 2 months
- ❖ Hysteresis in stress softened material curve

What did we learn?

- ✓ Non-linearity
- ✓ Stress softening (Mullin's effect)
- ✓ Residual strain
- ✓ Rate dependence
- ✓ Hysteresis
- ✓ Large Deformation

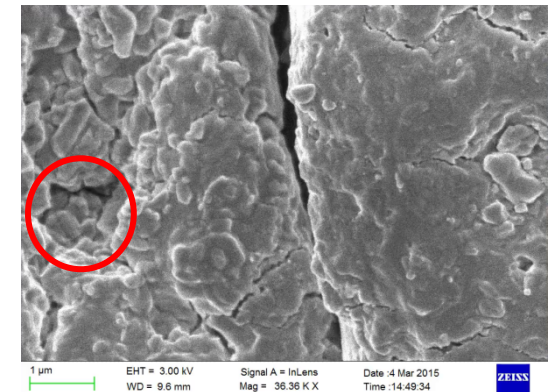
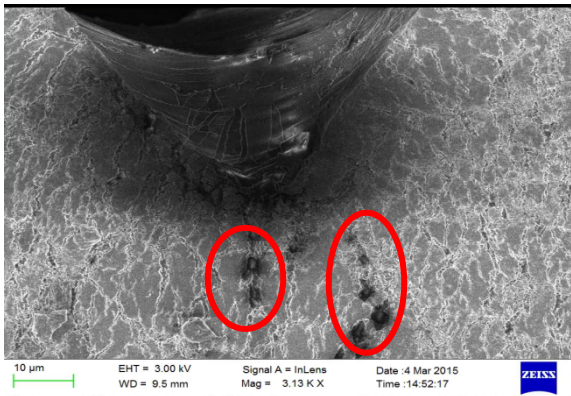


Microstructure: Void Growth



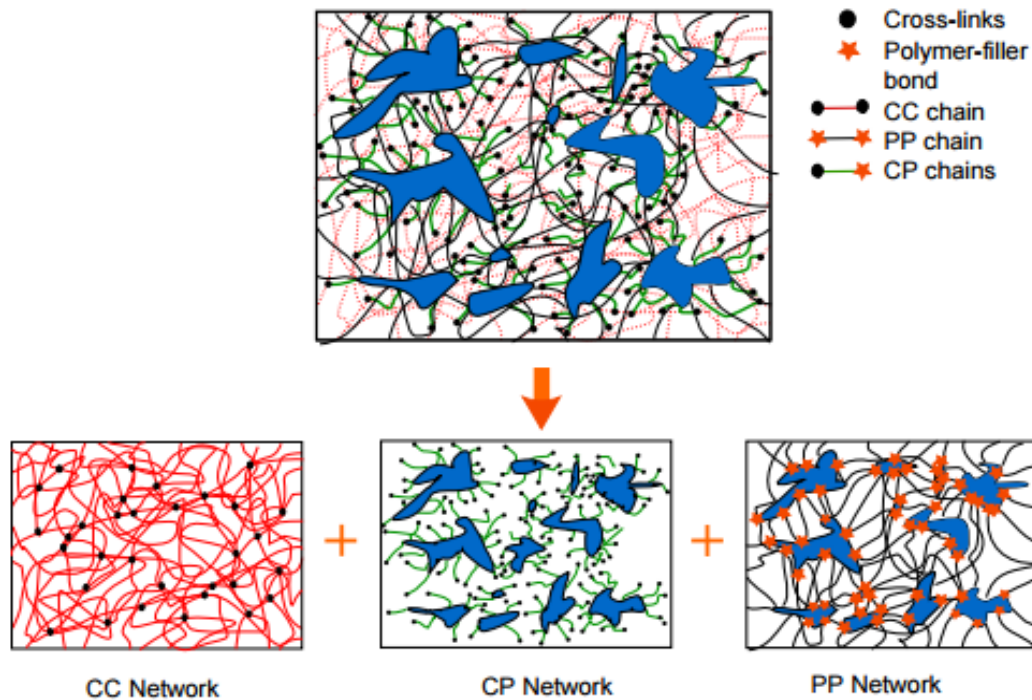
CT-Scan of Natural Rubber

- ❖ Post-loaded sample of Natural rubber shows growth in voids volume ratio
- ❖ Strain samples of EPDM shows void growth near a crack tip



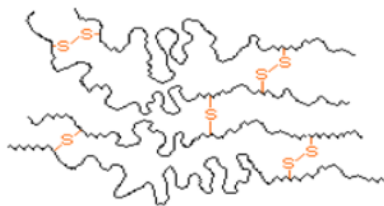
Scanning Electron Micrograph images of EPDM material near strained crack tip

Network Chains



Rubber Matrix decomposition in network chains [Itskov et al.]

- ❖ Random orientation in unloaded sample
- ❖ Orientation induced by loading
- ❖ New structures like fibrillar and lamella are formed
- ❖ Anisotropy induced via loading



Networks between polymeric chains and fillers

First step: Non-linearity

Free Energy Forms (Isothermal Case):

Helmholtz free energy

$$\Psi_H = \Psi_H(\epsilon)$$

$$\sigma = \frac{\partial \Psi_H}{\partial \epsilon}$$

← Legendre transform →

← Conjugate pairs →

Gibbs free energy

$$\Psi_G = \Psi_G(\sigma)$$

$$\epsilon = \frac{\partial \Psi_G}{\partial \sigma}$$

Similar expressions for other work conjugate pairs.

- Effort is employed majorly on formulating thermodynamically admissible free energy forms. Both statistical mechanics approaches (for instance, see Arruda and Boyce 1993) and phenomenological approaches (for instance, see Ogden 1972) are used to formulate these functions

Popular models by

Generalized Mooney-Rivlin solid , Generalized polynomial rubber elasticity potential, Ogden model, Arruda-Boyce 8 chain model, Ogden-Storakers hyperelastic foam and Blatz-Ko foam rubber etc.

[Ref: http://solidmechanics.org/Text/Chapter3_5/Chapter3_5.php]

Going beyond conservative systems

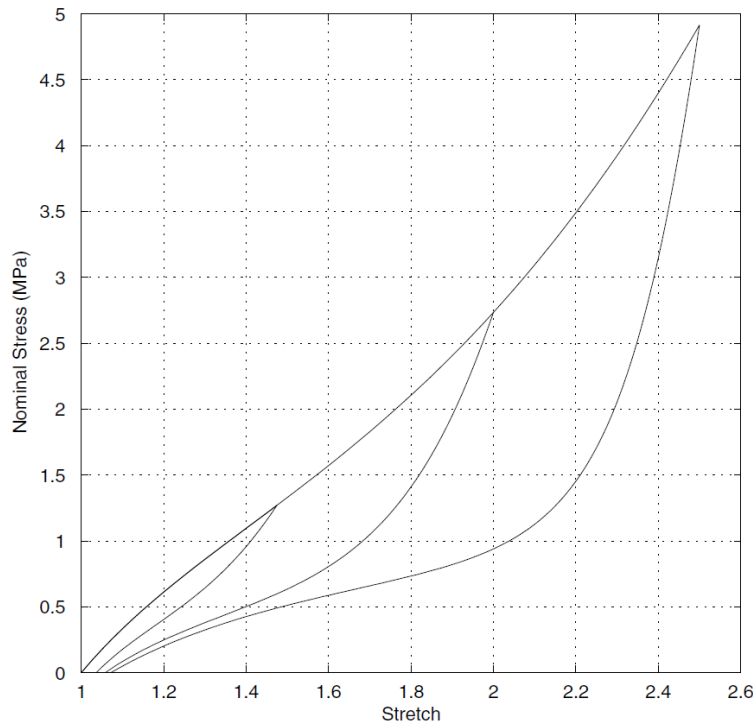
Pseudo-elasticity

$$\Psi(F, \eta_1, \eta_2) = \eta_1 \Psi_0(F) + (1 - \eta_2) N(F) + \Phi(\eta_1, \eta_2)$$

↓
Stress
softening

↓
Residual
strain

↓
Dissipation
potential



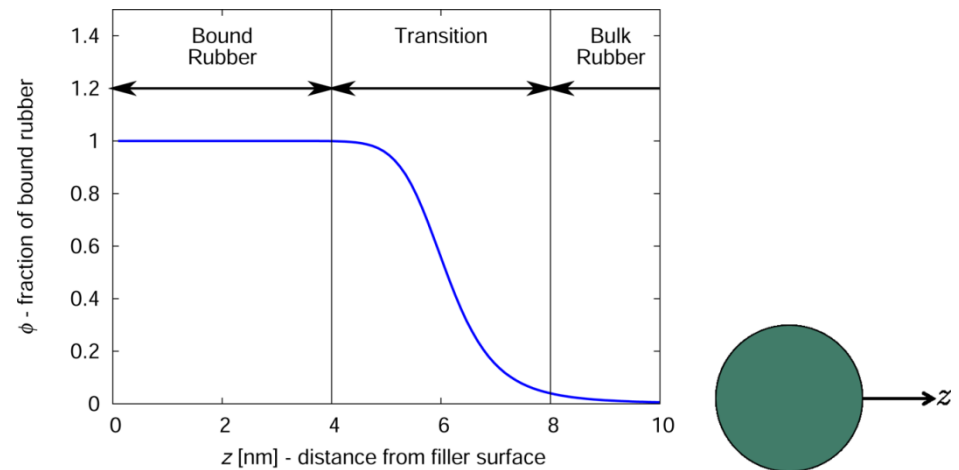
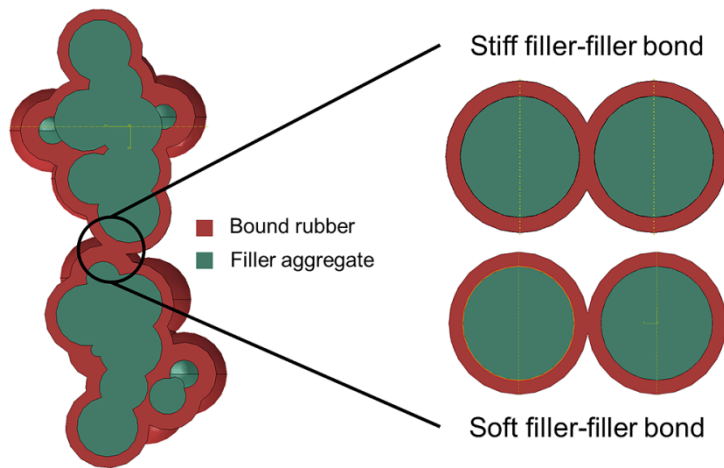
- Modified hyperelastic response to include damage parameters with initial value of unity
- Damage parameter asymptotically decreases to zero with increase in maximum deformation seen by the material
- This concept can be extended to include non virgin hysteresis too

[Ref: Dorfmann and Ogden (2003)]

Going beyond conservative systems

Hard and soft phases

- Rubber matrix contains 2 phases (Hard and Soft)
- Free energy is the sum of free energy of both phases.
- Hard Phase gets irreversibly converted to soft phase on loading.

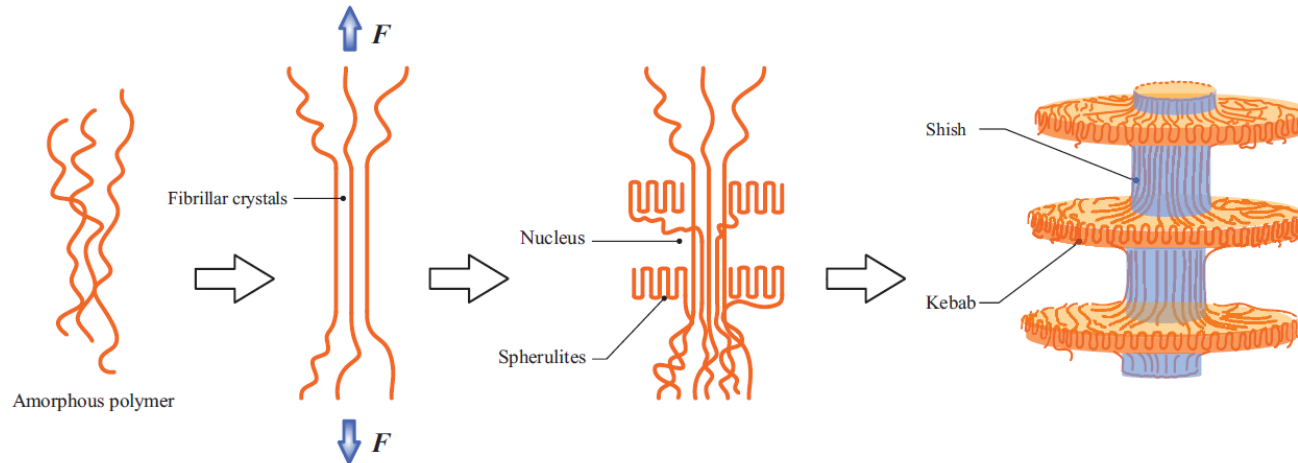


Representative work:

Beatty and Krishnaswamy (2000), Sodhani and Reese (2014)

Going beyond conservative systems

Strain induced crystallization



Formation of Shish-Kebab structure in tension [Ref: Itskov et. al. 2014]

❖ Total Strain energy = Sum of strain energy of different network (eg. Pure rubber (CC) network Polymer-Aggregate (PP) network Aggregate-fibrillar crystal (ST) network Aggregate-lamellar crystal (FL) network.

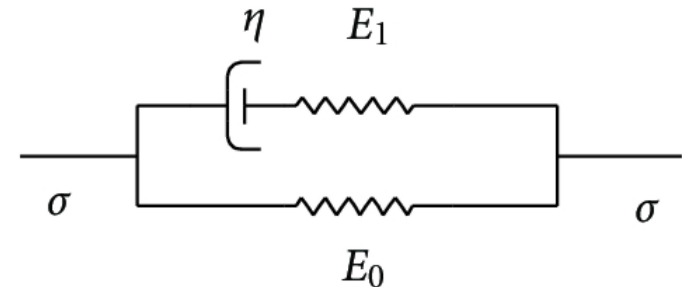
❖ The probability distribution of the networks in different directions differ for loading and unloading case.

Phenomenological models

Modelling by combining spring, damper and slider in different arrangements.

Viscoelastic material:

- Combination of spring and damper system
- Popular model include Kelvin-Voigt model and Maxwell model



Generalized Maxwell model

Damaged viscoelastic material:

- Generally the hyperelastic spring in a viscoelastic material is replaced by a damaged spring
- The damage increases with maximum deformation

Damaged viscoelastic plastic material:

- Networks of elasticity, viscosity and inelasticity are used in series/parallel/mixed arrangement
- Loading induces damage on few or all the networks

Modelling Techniques

✓ Hyperelastic models

- Free energy functions $\psi (F)$
- Mooney-Rivlin, Ogden and Arruda-Boyce model
- Good fit with quasi-static and monotonic experiments
- No viscous effect, hysteresis or residual strain

Pseudo-elasticity

Hard and Soft phases

Strain induced crystallization

Viscoelastic material

Damaged viscoelastic material

Damaged viscoelastic plastic material

Representative work:

Rivlin (1960), Arruda and Boyce (1993), Ogden (1972)

Modelling Techniques

✓ Hyperelastic models

✓ Pseudo-elasticity

- Free energy functions $\psi(\eta, F)$
- Damaged elasticity
- Stress softening and quasi-static hysteresis
- No viscous effect

☐ Hard and Soft phases

☐ Strain induced crystallization

☐ Viscoelastic material

☐ Damaged viscoelastic material

☐ Damaged viscoelastic plastic material

Representative work:

Ogden and Roxburgh (1999), Dorfmann and Ogden (2003)

Modelling Techniques

- ✓ Hyperelastic models
- ✓ Pseudo-elasticity
- ✓ Hard and Soft phases
 - Soft phase \equiv Rubbery phase
 - Hard Phase \equiv chain-filler bonds
 - Hard phase transforms to soft phase on loading
 - Mullin's effect but no viscous effect
- Strain induced crystallization
- Viscoelastic material
- Damaged viscoelastic material
- Damaged viscoelastic plastic material

Representative work:

Beatty and Krishnaswamy (2000), Sodhani and Reese (2014)

Modelling Techniques

- ✓ Hyperelastic models
- ✓ Pseudo-elasticity
- ✓ Hard and Soft phases
- ✓ Strain induced crystallization
 - Network models
 - Strain history dependent probability of networks
 - Total Strain energy = Sum of strain energy of network
 - Mullin's effect but no viscous effect
- Viscoelastic material
- Damaged viscoelastic material
- Damaged viscoelastic plastic material

Representative work:

Itskov et.al.(2014), Rault et.al.(2006), Flory (1962)

Modelling Techniques

- ✓ Hyperelastic models
- ✓ Pseudo-elasticity
- ✓ Hard and Soft phases
- ✓ Strain induced crystallization
- ✓ Viscoelastic material
 - Phenomenological model
 - Equilibrium and Non-equilibrium branches
 - Viscous effect and hysteresis due viscous dissipation
 - No stress softening and permanent strain set
- ❑ Damaged viscoelastic material
- ❑ Damaged viscoelastic plastic material

Representative work:

Reese and Govindjee (1998), Bergstrom and Boyce (1998)

Modelling Techniques

- ✓ Hyperelastic models
- ✓ Pseudo-elasticity
- ✓ Hard and Soft phases
- ✓ Strain induced crystallization
- ✓ Viscoelastic material
- ✓ Damaged viscoelastic material
 - Damaged elasticity
 - Damaged/undamaged viscous branch
 - Introduces stress-softening
 - No permanent strain set
- ❑ Damaged viscoelastic plastic material

Representative work:

Simo (1987), Kaliske and Rothert (1998)

Modelling Techniques

- ✓ Hyperelastic models
- ✓ Pseudo-elasticity
- ✓ Hard and Soft phases
- ✓ Strain induced crystallization
- ✓ Viscoelastic material
- ✓ Damaged viscoelastic material
- ✓ Damaged viscoelastic plastic material
 - Introduces permanent strain set
 - Cost: too complex and too many parameters

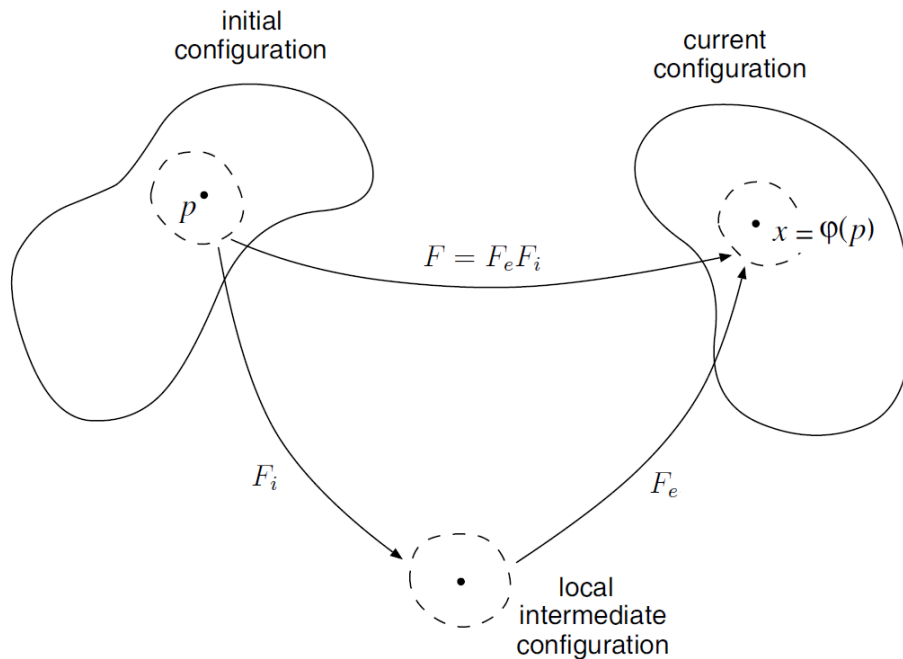
Representative work:

Meihe and Keck (2000), Lion (1996)

Damaged viscoelastic plastic model

Helmholtz energy:

$$\Psi = (1 - D) \Psi^0 (C_e) + \Psi^v (C_e, Q) + \Psi^I$$



Definitions

F : Deformation Gradient

$$F = F_e F_i$$

C : Right-Cauchy green tensor

$$C = F^T F$$

$$C_e = F_e^T F_e$$

D : Damage parameter

$$D \subset [0, 1]$$

Q : Internal State variable

Thermodynamic admissibility

Claussius-Duhem inequality (Isothermal version):

$$S^{PK2} : \dot{C}_e - 2\rho_0 \dot{\Psi} \geq 0$$

On substituting the energy form into the inequality, we get,

$$\begin{aligned} & \left[S^{PK2} - 2\rho_0 F_i^{-1} \frac{\partial ((1-D)\Psi^0 + \Psi^v)}{\partial C_e} F_i^{-T} \right] : \dot{C} \\ & - 2\rho_0 \frac{\partial ((1-D)\Psi^0 + \Psi^v)}{\partial C_e} : \left[\dot{F}_i^{-T} C F_i^{-1} + F_i^{-T} C \dot{F}_i^{-1} \right] \\ & + 2\rho_0 \left[\Psi^0 \dot{D} - \frac{\partial \Psi^v}{\partial Q} \dot{Q} - \dot{\Psi}^I \right] \geq 0 \end{aligned}$$

Thermodynamic admissibility

We can fix F_i , Q , D and vary C , we get stress as

$$S^{PK2} = 2\rho_0 F_i^{-1} \frac{\partial ((1 - D) \Psi^0 + \Psi^v)}{\partial C_e} F_i^{-T}$$

And dissipation as,

$$\begin{aligned} \frac{G}{2\rho_0} = & - \frac{\partial ((1 - D) \Psi^0 + \Psi^v)}{\partial C_e} \left\{ \dot{F}_i^{-1} C F_i^{-T} + F_i^{-1} C \dot{F}_i^{-T} \right\} \\ & + \Psi^0 \dot{D} - \frac{\partial \Psi^v}{\partial Q} \dot{Q} - \dot{\Psi}^I \geq 0 \end{aligned}$$

Thermodynamic admissibility

Dissipation can further be reduced to following form:

$$G = F_i S^{PK2} F_i^T : (L_i^T C_e + C_e L_i) \\ + 2\rho_0 \left[\Psi^0 \dot{D} - \frac{\partial \Psi^v}{\partial Q} \dot{Q} - \dot{\Psi}^I \right] \geq 0$$

Now assuming a strong form of dissipation,

$$F_i S^{PK2} F_i^T : (L_i^T C_e + C_e L_i) \geq 0$$

$$\Psi^0 \dot{D} \geq 0$$

$$\frac{\partial \Psi^v}{\partial Q} \dot{Q} + \dot{\Psi}^I \leq 0$$

Evolution Laws

Assuming linear evolutions (sufficient but not necessary !!!)
of inelasticity and damage,

$$\dot{D} = \dot{\beta} \Psi^0$$

$$L_i = C_e^{-1} \cdot \Upsilon \left(\dot{\beta} \right) : F_i \cdot S \cdot F_i^T$$

Where,

$$\Upsilon = \frac{v_1 \dot{\beta}}{3} I \otimes I + v_2 \dot{\beta} \left(I_S - \frac{1}{3} I \otimes I \right)$$

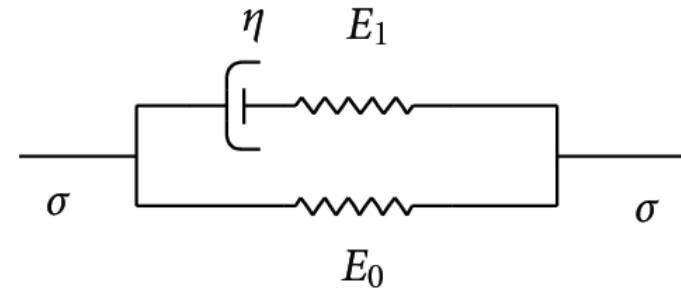
Yield surface: $\Phi = \sqrt{2\Psi^0} - \sqrt{2\Psi^0}_{max}$

Loading –unloading
condition: $\dot{\beta} > 0; \quad \Phi \leq 0; \quad \dot{\beta} \Phi = 0$

Viscous evolution

Motivated by **generalized Maxwell's model** we take evolution law of the form:

$$\dot{Q} = \gamma \dot{S}^{PK2} - \frac{1}{\zeta} Q$$



This can be achieved by taking the following energy forms,

$$\Psi^v = \int Q : dC_e - \frac{1}{2} Q : Q$$

$$\Psi^I = \zeta \int (\gamma \dot{S}^{PK2} - \dot{Q}) : dQ$$

So that the dissipation inequality reduces to

$$\zeta (\dot{Q} - \gamma \dot{S}^{PK2} + \frac{1}{\zeta} Q) \dot{Q} \geq 0$$

Hyperelastic form

Ogden type material

Free energy form:

$$\rho_0 \Psi^0 (\lambda_{e(1)}^*, \lambda_{e(2)}^*, \lambda_{e(3)}^*) = \sum_{p=1}^N \frac{\mu_p}{\alpha_p} \left(\lambda_{e(1)}^{*\alpha_p} + \lambda_{e(2)}^{*\alpha_p} + \lambda_{e(3)}^{*\alpha_p} - 3 \right) + \frac{1}{2} K \log (J_e^2)$$

Where the eigenvalues of elastic isochoric stretch tensor are related to the eigenvalues of elastic stretch tensor by the relation,

$$\lambda_{e(k)}^* = \frac{\lambda_{e(k)}}{J_e^{\frac{1}{3}}}$$

And the **Kirchhoff Stress** is given as

$$\tau_{(i)} = \lambda_{e(i)} \rho_0 \frac{\partial \Psi^0}{\partial \lambda_{e(i)}} = \sum_{p=1}^N \mu_p J_e^{-\alpha_p/3} \left[\lambda_{e(i)}^{\alpha_p} - \frac{1}{3} \left(\lambda_{e(1)}^{\alpha_p} + \lambda_{e(2)}^{\alpha_p} + \lambda_{e(3)}^{\alpha_p} \right) \right] + K \log J_e$$

Incompressibility constraint

$$\lambda_1 \lambda_2 \lambda_3 = 1$$

The Kirchhoff stress for this case can then be given by adding an extra hydrostatic stress as,

$$\tau_{(i)} = \lambda_{e(i)} \rho_0 \frac{\partial \Psi^0}{\partial \lambda_{e(i)}} - p$$

On eliminating this additional pressure we get,

$$\tau_{(1)} - \tau_{(3)} = \lambda_{e(1)} \rho_0 \frac{\partial \Psi^0}{\partial \lambda_{e(1)}}$$

$$\tau_{(2)} - \tau_{(3)} = \lambda_{e(2)} \rho_0 \frac{\partial \Psi^0}{\partial \lambda_{e(2)}}$$

1D reduction: Simple axial loading

Let us define (WLOG) 1 direction to be the loading direction. Then the stresses in 2-3 direction for a simple axial test will be given as,

$$\tau_2 = \tau_3 = 0$$

We can also take stretch to be symmetric in 2-3 direction, i.e. if we assume the stretch in 1 direction to be given as

$$\lambda_1 = \lambda$$

Then,

$$\lambda_2 = \lambda_3 = \frac{1}{\lambda^{\frac{1}{2}}}$$

Note: The implication of no transverse stress condition along with the assumed evolution equation is that there is no inelastic growth in 2-3 direction. Similar arguments can be made for evolution of Q

1D reduction: Evolution equations

Stretch assumes multiplicative decomposition given as,

$$\lambda = \lambda_e \lambda_i$$

The evolution equations can then be reduced to following 1-D form given as

$$\dot{\lambda}_i = c_1 \dot{\beta} S \frac{\lambda_i^3}{\lambda_e^2}$$

$$\dot{D} = c_2 \dot{\beta}$$

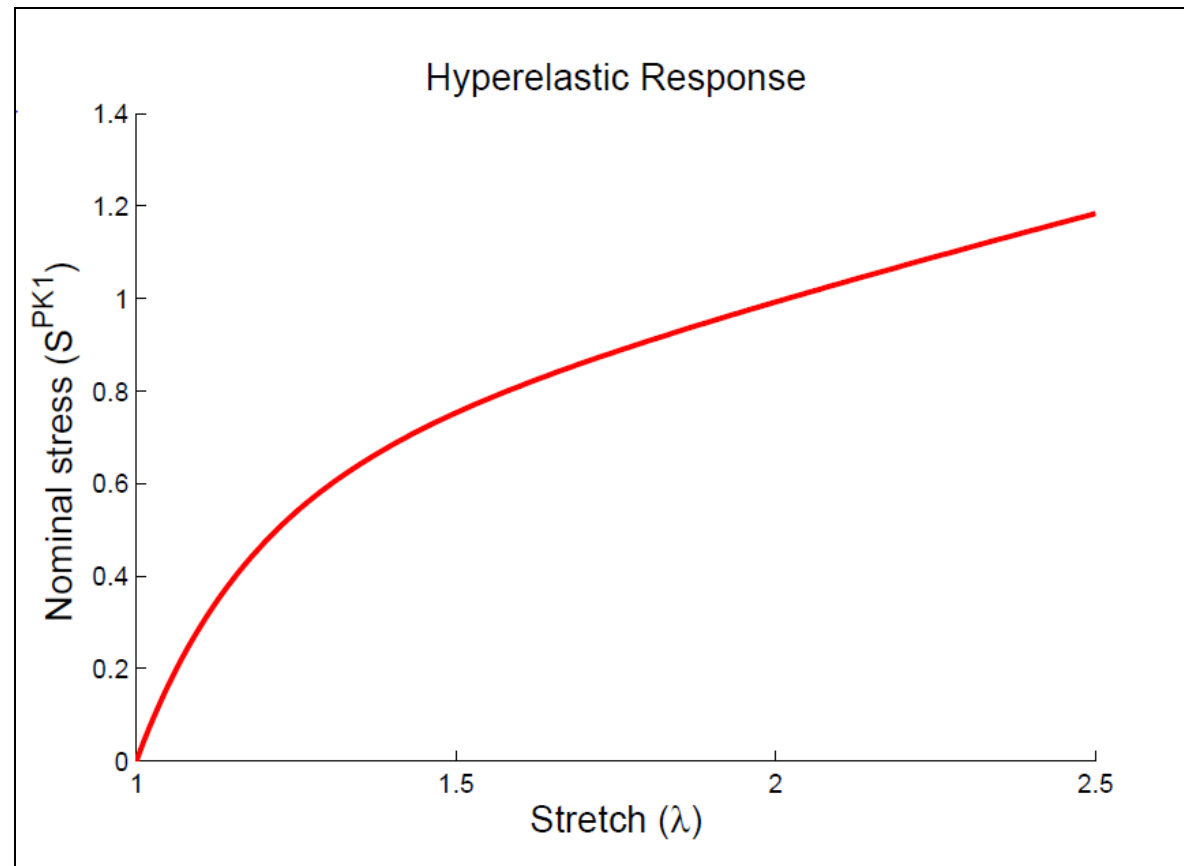
$$\dot{\beta} = \sqrt{2 \dot{\Psi}^0}_{max}$$

$$\dot{q} = \gamma \dot{S} - \frac{q}{\zeta}$$

Where S and q represents the (1,1) component of Second Piola-Kirchhoff stress and Q tensor respectively

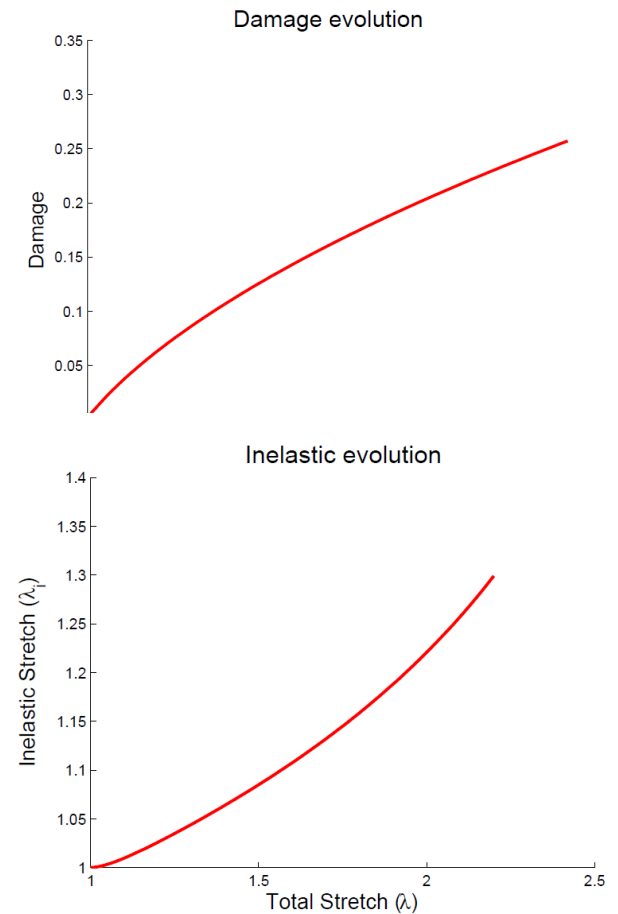
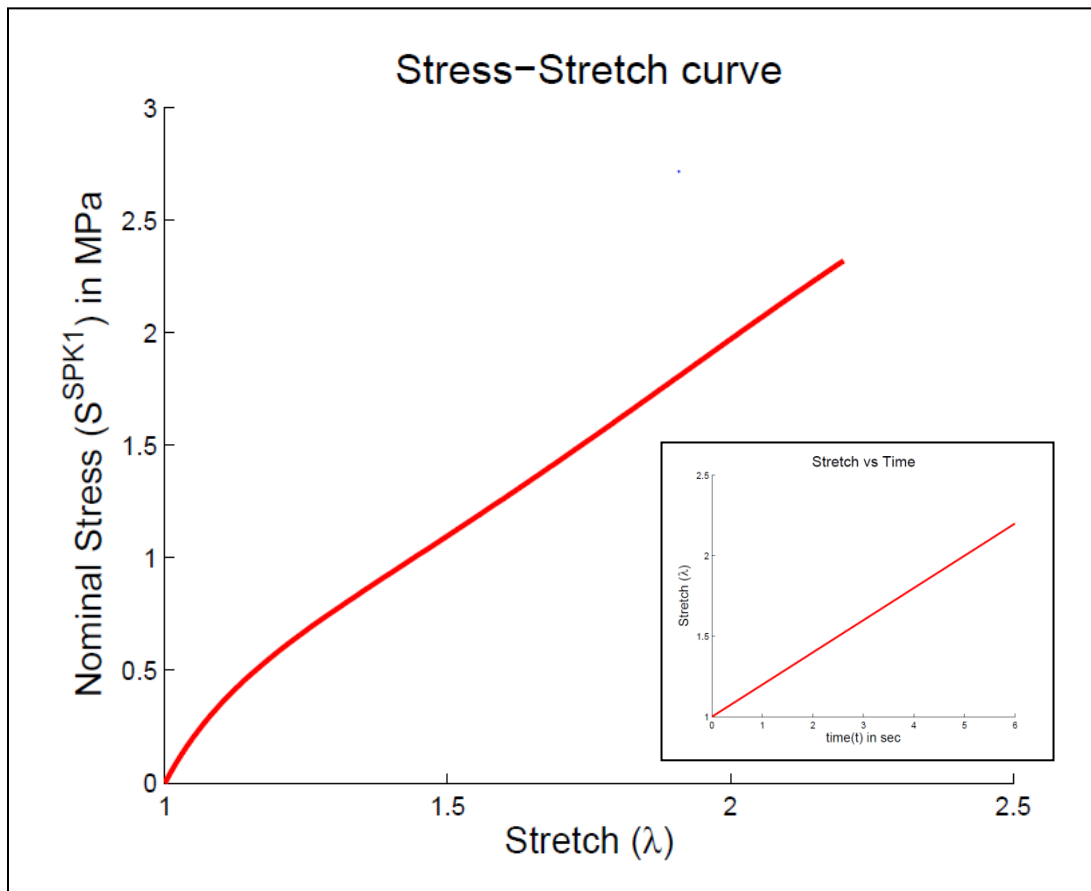
Qualitative behavior: Hyperelastic response

μ_1	α_1	μ_2	α_2	μ_3	α_3
-1.528380	-1.011467	0.222564	4.2047799	-1.13418E-3	-4.398598

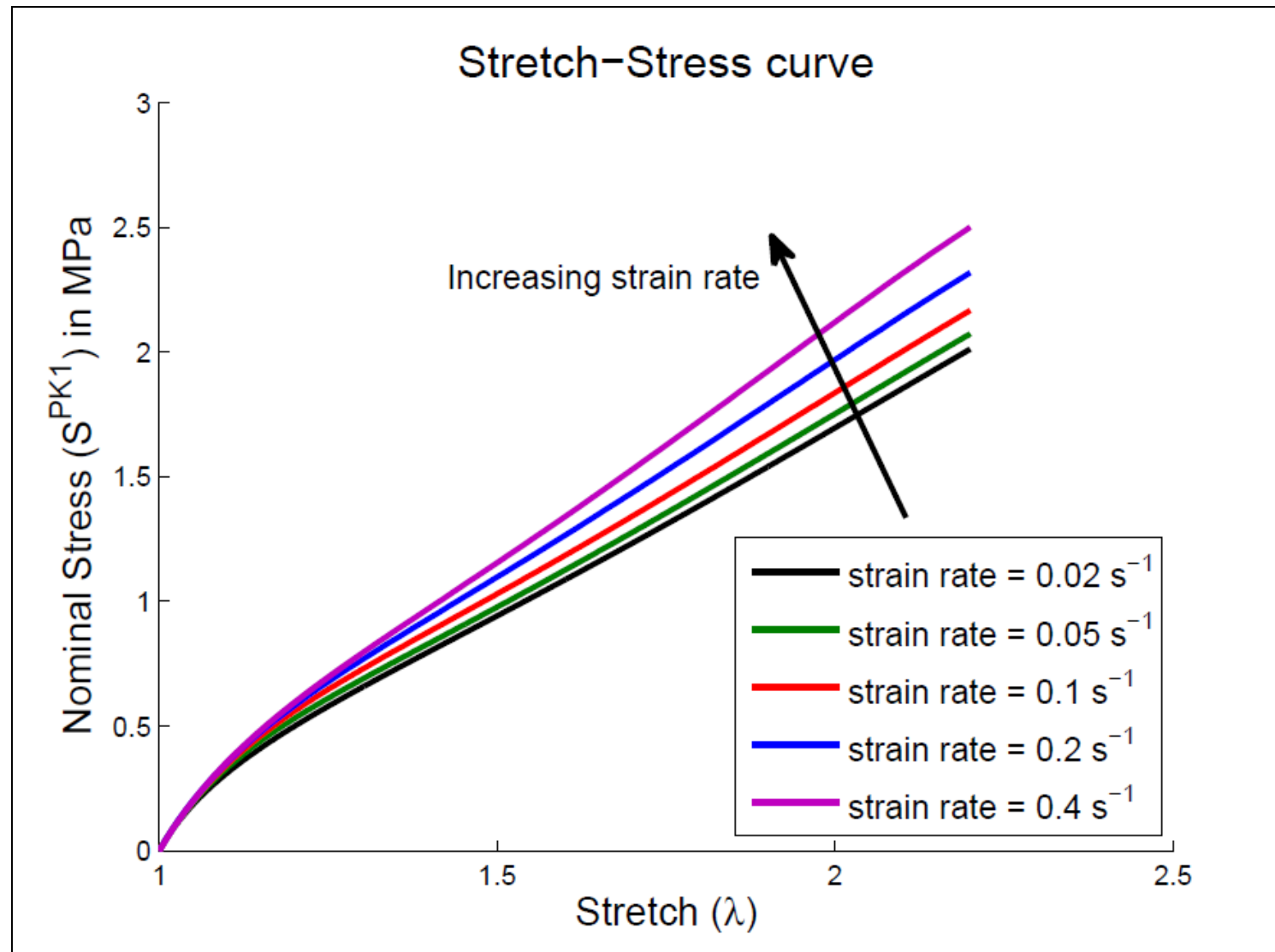


Qualitative behavior: Monotonic Loading

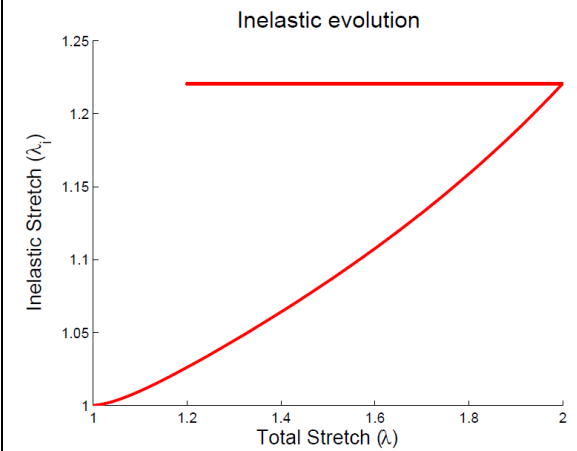
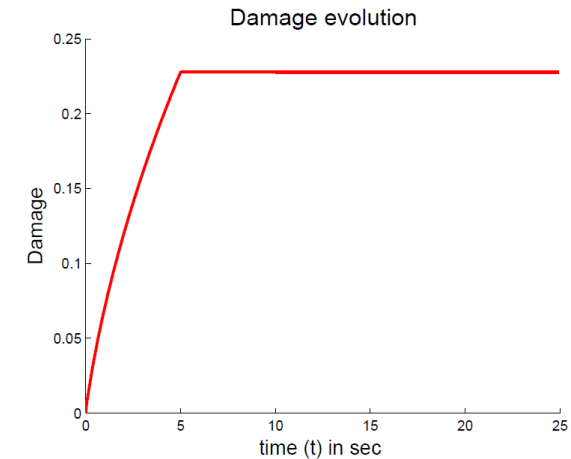
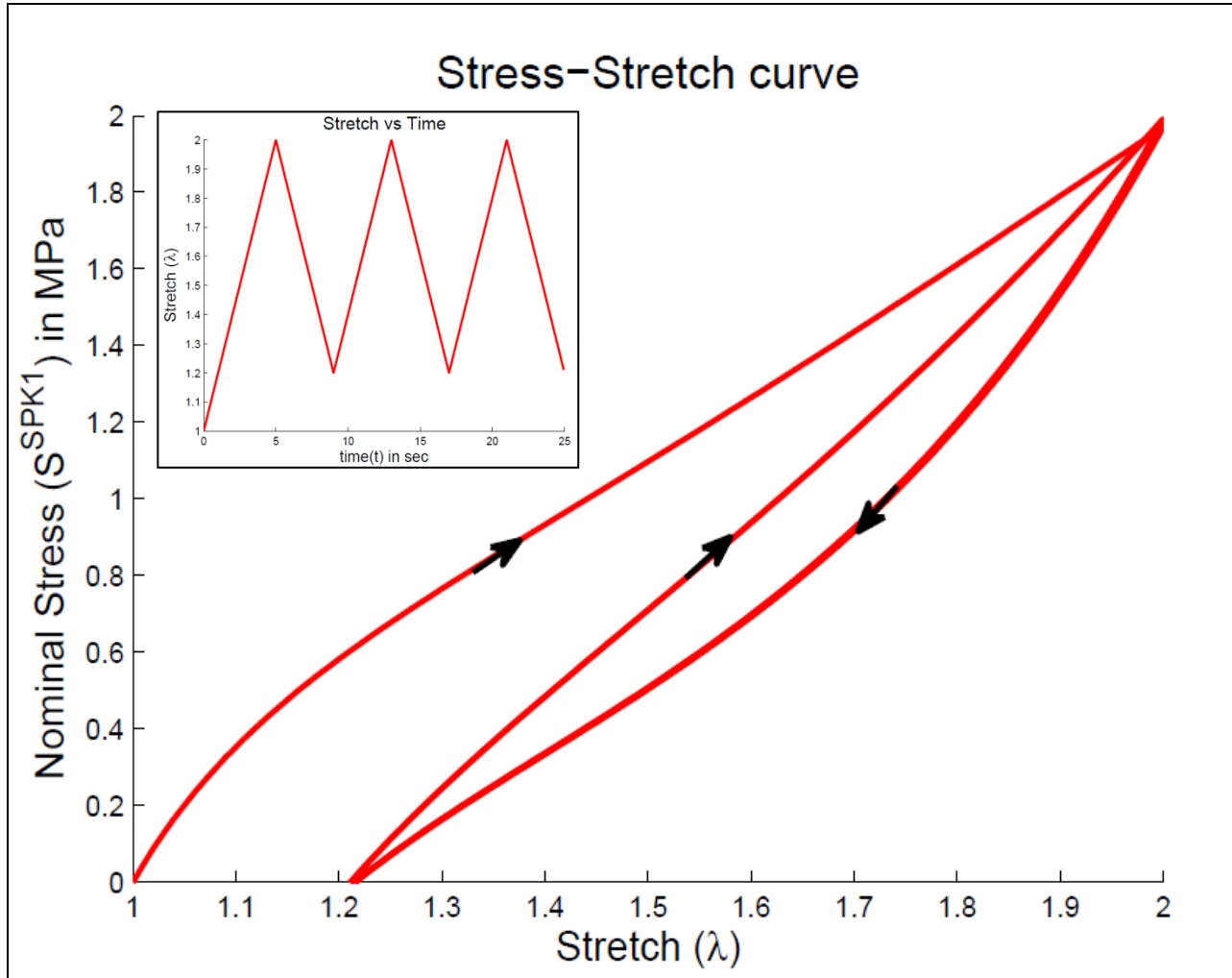
Inelasticity	Damage	Viscous	
c_1	c_2	γ	ζ
0.011404	0.127093	0.1	2



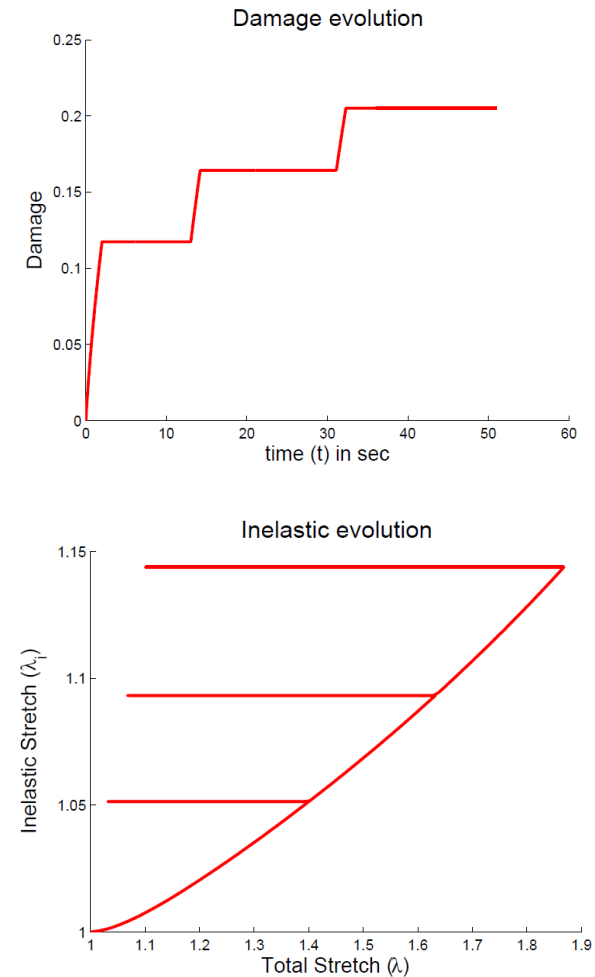
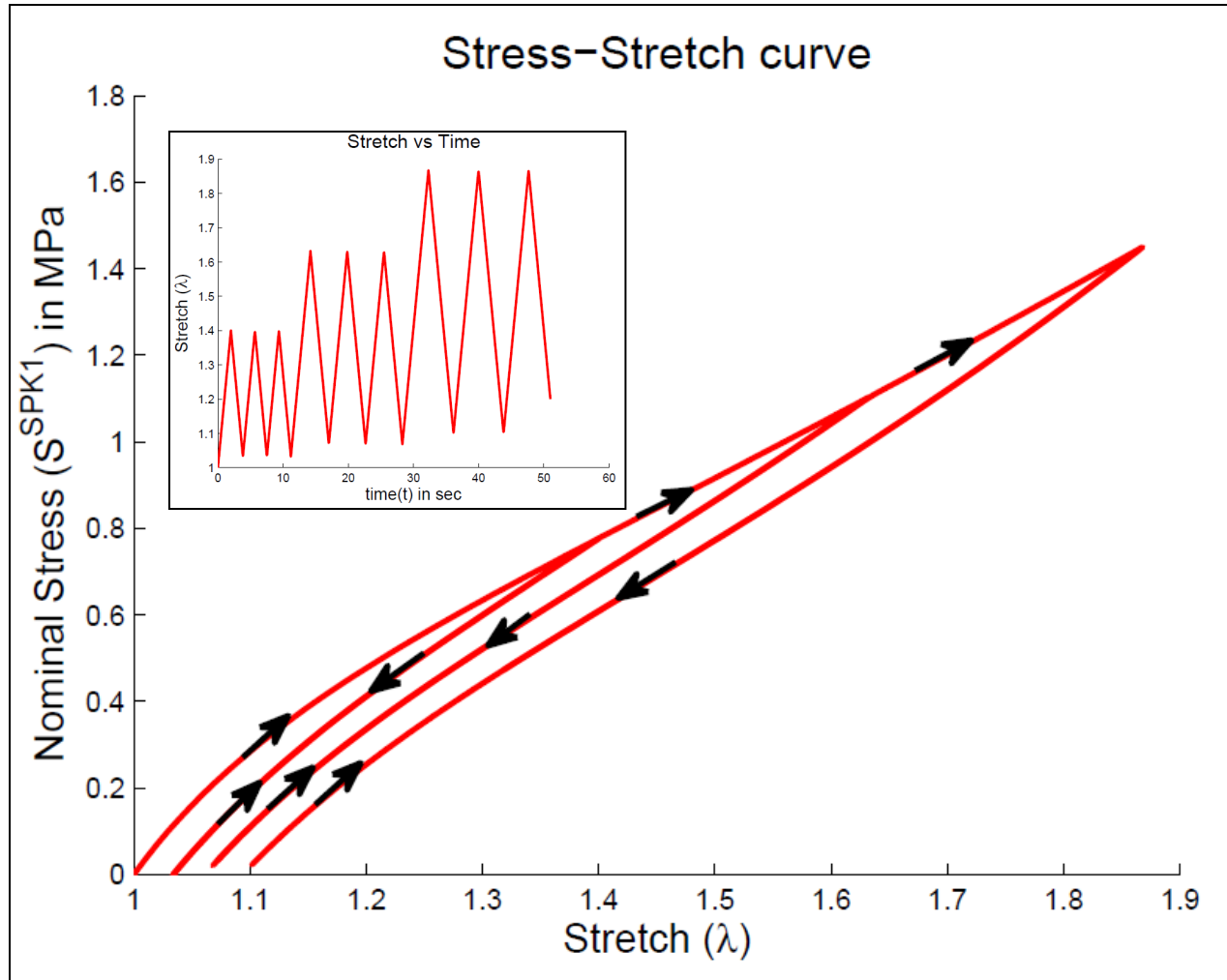
Qualitative behavior: Viscous Effect



Qualitative behavior: Cyclic Loading

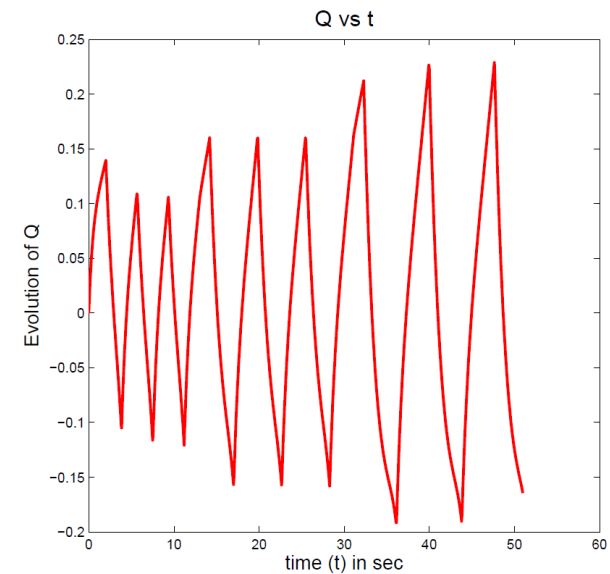
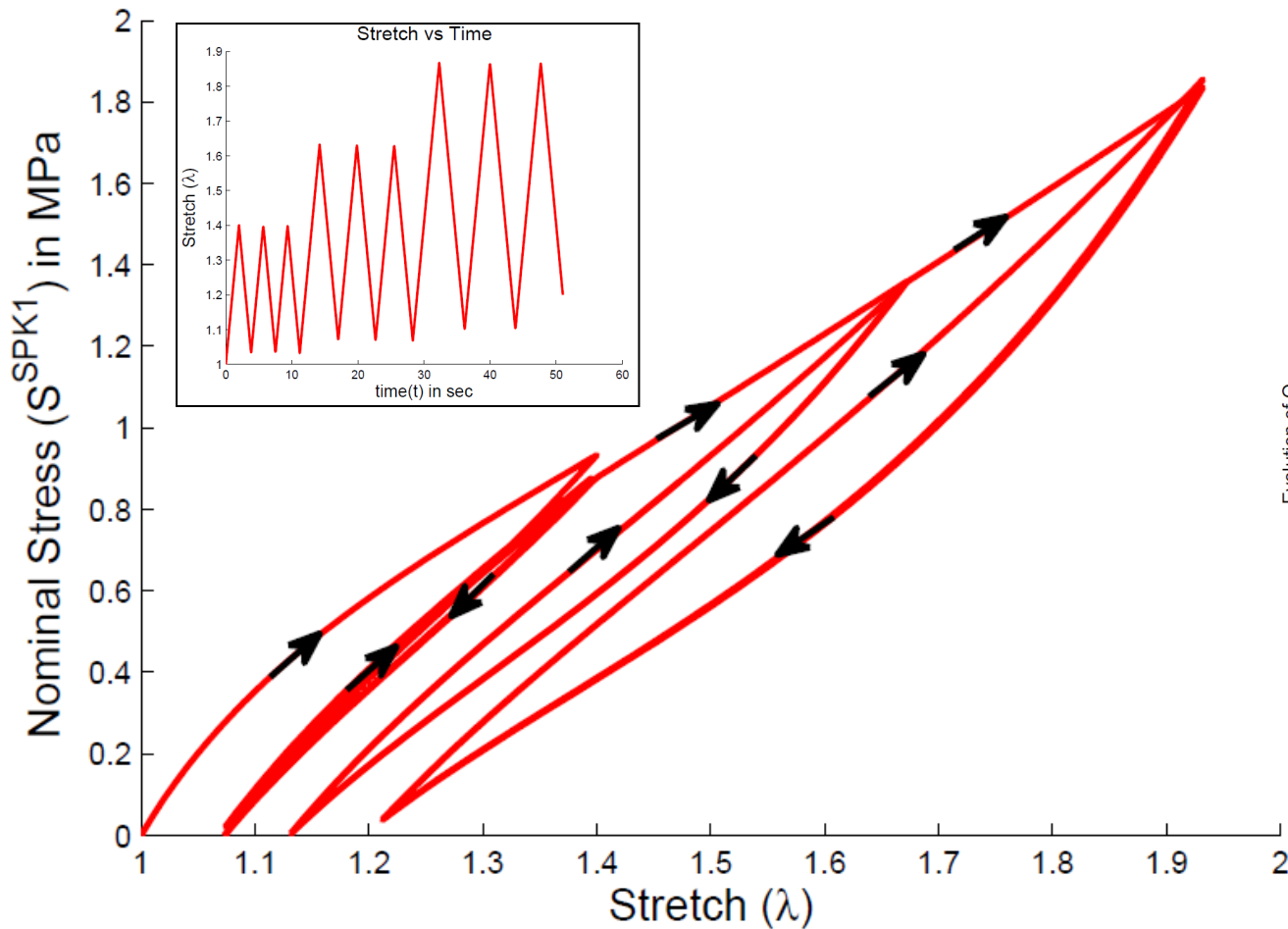


Qualitative behavior: without viscous branch



Qualitative behavior: with viscous branch

Stress–Stretch curve



All models are wrong but some models are useful!!!

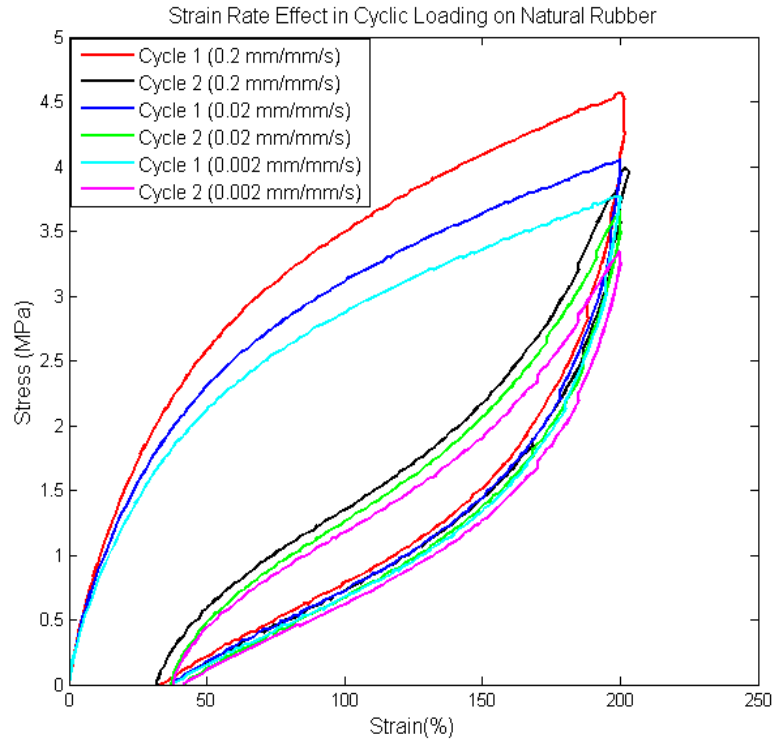
✓ Reviewed literature on “Rubber elasticity” with focus on constitutive modelling techniques like multiphase material modeling, strain induced crystallization and other phenomenological approaches like pseudo-elasticity and damage mechanics.

✓ Developed a damaged viscoelastic plastic model and verified its thermodynamic admissibility.

✓ Verified the qualitative behavior of this model in simple axial tensile loading case.

✓ The 1D axial case required only 4 parameters other than parameters for the hyperelastic form.

Does it solve everything?



Same dissipation in stress-softened hysteresis irrespective of strain rate.

Healing ??

Thank You

Objectivity of rate of deformation tensor

$$\chi = Q^T \dot{Q}$$

It can be shown that χ is antisymmetric.

$$\frac{D}{Dt} Q^T Q = \dot{Q}^T Q + Q^T \dot{Q} = \chi^T + \chi = 0$$

In the rotated frame, the velocity gradient L^* is given as,

$$L^* = Q(\dot{F} + \chi F)F^{-1}Q^T = Q(L + \chi)Q^T$$

$$Q(D + W + \chi)Q^T$$

From the antisymmetry of χ it follows that,

$$D^* = QDQ^T$$

$$W^* = Q(W + \chi)Q^T$$

Thus symmetric part of velocity gradient is objective.

Substituting energy form to CD inequality

$$\dot{\Psi} = (1 - D) \frac{\partial \Psi^0}{\partial C_e} : \dot{C}_e - \Psi^0 \dot{D} + \frac{\partial \Psi^v}{\partial C_e} : \dot{C}_e + \frac{\partial \Psi^v}{\partial Q} : \dot{Q} + \dot{\Psi}^I$$

$$C_e = F_i^{-T} C F_i^{-1}$$

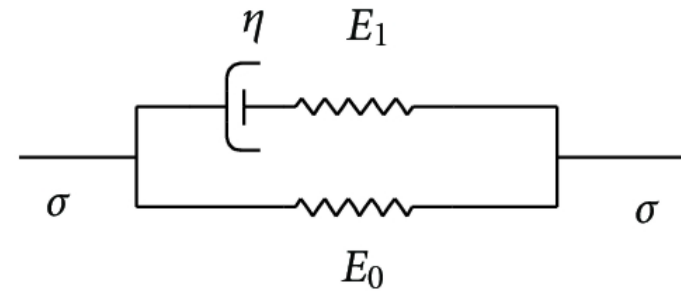
$$\dot{C}_e = \dot{F}_i^{-T} C F_i^{-1} + F_i^{-T} \dot{C} F_i^{-1} + F_i^{-T} C \dot{F}_i^{-1}$$

$$S^{PK2} : \dot{C} - 2\rho_0 \frac{\partial ((1 - D) \Psi^0 + \Psi^v)}{\partial C_e} :$$

$$\left[\dot{F}_i^{-T} C F_i^{-1} + F_i^{-T} \dot{C} F_i^{-1} + F_i^{-T} C \dot{F}_i^{-1} \right] + 2\rho_0 \left[\Psi^0 \dot{D} - \frac{\partial \Psi^v}{\partial Q} \dot{Q} - \dot{\Psi}^I \right] \geq 0$$

1D small strain linear viscous branch

$$\Psi = \underbrace{\frac{1}{2}E_0\varepsilon_0^2}_{E_0 \text{ Spring}} + \underbrace{\frac{1}{2}E_1\varepsilon_1^2}_{E_1 \text{ Spring}} + \underbrace{\int \eta \dot{\varepsilon}_2 d\varepsilon_2}_{\text{damper}}$$



$$\begin{aligned}\Psi &= \frac{1}{2}E_0\varepsilon_0^2 + \int \sigma_1 d(\varepsilon_0 - \varepsilon_2) + \int \eta \dot{\varepsilon}_2 d\varepsilon_2 \\ &= \frac{1}{2}E_0\varepsilon_0^2 + \int \sigma_1 d(\varepsilon_0 - \varepsilon_2) + \int \eta \left(\dot{\varepsilon}_0 - \frac{\dot{\sigma}_1}{E_1}\right) \dot{\varepsilon}_2 dt\end{aligned}$$

$$\begin{aligned}\dot{\Psi} &= E_0\varepsilon_0\dot{\varepsilon}_0 + \sigma_1(\dot{\varepsilon}_0 - \dot{\varepsilon}_2) + \eta\left(\dot{\varepsilon}_0 - \frac{\dot{\sigma}_1}{E_1}\right)\dot{\varepsilon}_2 \\ &= (E_0\varepsilon_0 + \sigma_1)\dot{\varepsilon}_0 + \eta\left(\dot{\varepsilon}_0 - \frac{\sigma_1}{\eta} - \frac{\dot{\sigma}_1}{E_1}\right)\dot{\varepsilon}_2\end{aligned}$$

$$\sigma = \underbrace{E_0\varepsilon_0}_{\sigma_{\text{equilibrium}}} + \underbrace{\sigma_1}_{\sigma_{\text{non-equilibrium}}}$$

$$\eta\left(\frac{\dot{\sigma}_{\text{equilibrium}}}{E_0} - \frac{\sigma_{\text{non-equilibrium}}}{\eta} - \frac{\dot{\sigma}_{\text{non-equilibrium}}}{E_1}\right)\dot{\varepsilon}_2 \geq 0$$

Rules of the game

Axioms:

Thermodynamic Determinism (CD inequality)

Memory

Equipresence

Local action

Objectivity

Admissibility

Simplifications:

Material Symmetry \rightarrow Isotropy

Internal State variables

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