Functional Grading of Rubber-Elastic Materials: From Chemistry to Mechanics

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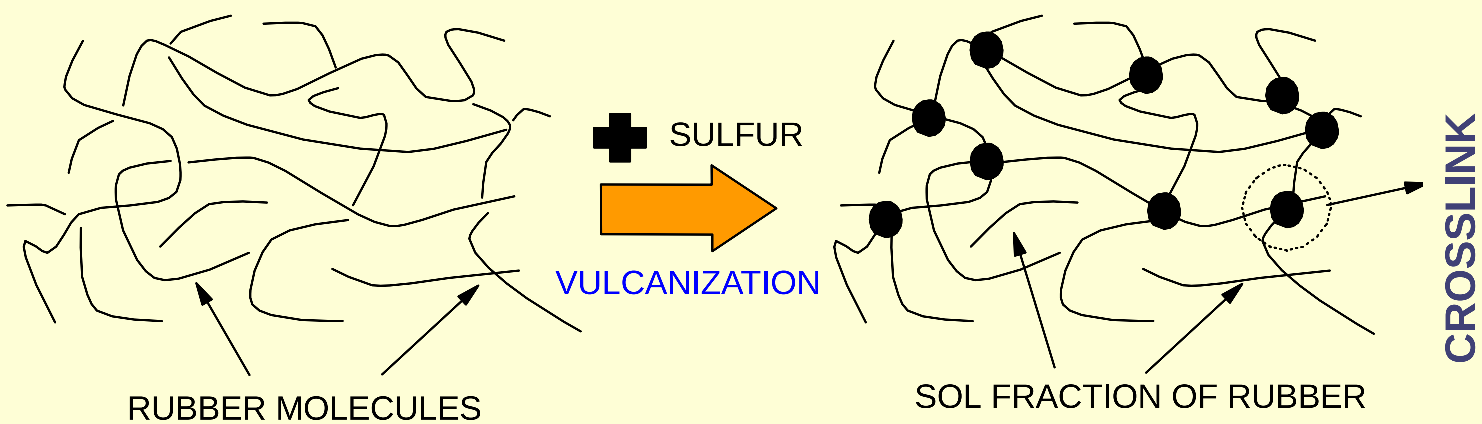
Outline of the Presentation

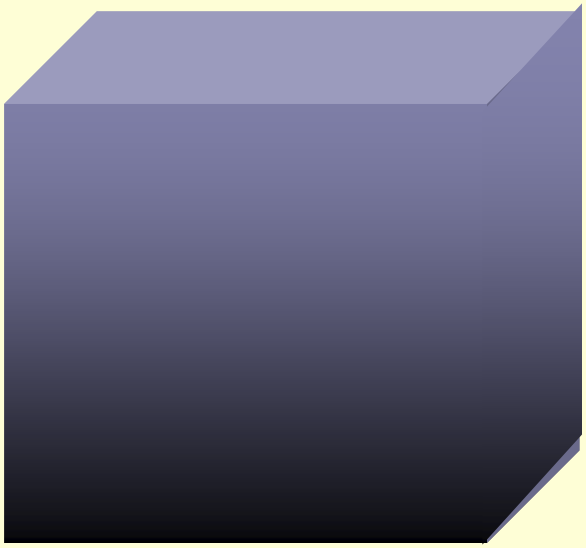
- Overview of rubbers and basic concepts
- Experimental results: potential of grading
- A theoretical study on the effects of
  - material non-homogeneity
  - temperature gradients
  - functional grading
- How to manufacture FGREMs?
  - modeling the structure development
- Conclusions

Overview of Rubber-like Materials

- Rubbers widely used in industry and daily life
- Applications: tires, bearings, bushings, sealants, etc.
- Relatively soft, light-weight, and flexible. Undergo large strains, energy dissipation, sub-ambient T_g.
- 3-D network structure of long, flexible polymeric chains:



Functional Grading Concept



Tailoring the material non-homogeneity

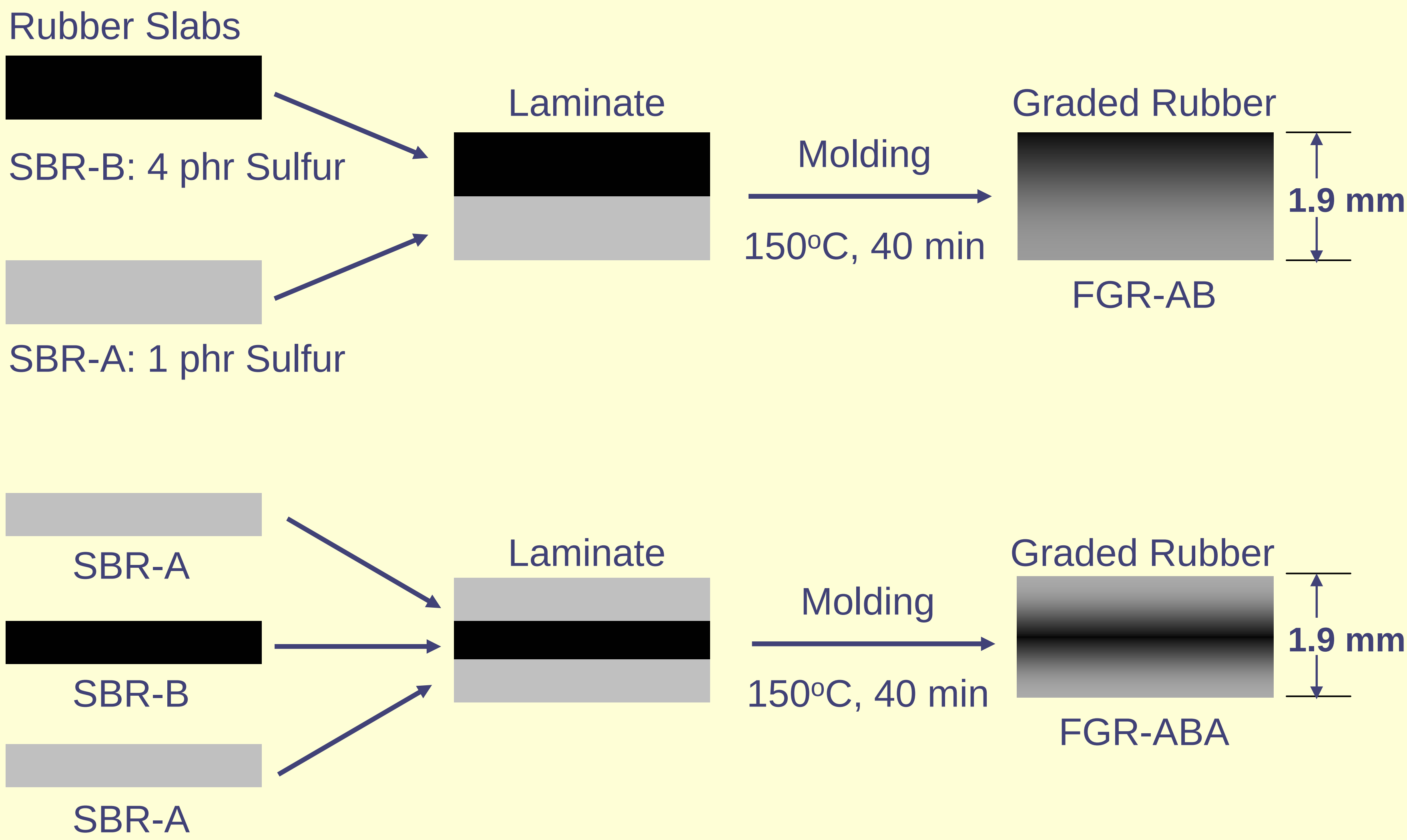
P(***X***) for

reducing stress-strain localizations

and temperature gradients

controlling the dynamic properties, surface properties, and the permeability to fluids

Graded Rubbers: Layering



Ikeda, Y. 2003. *J. Appl. Polym. Sci.*, **87**: 61-67.

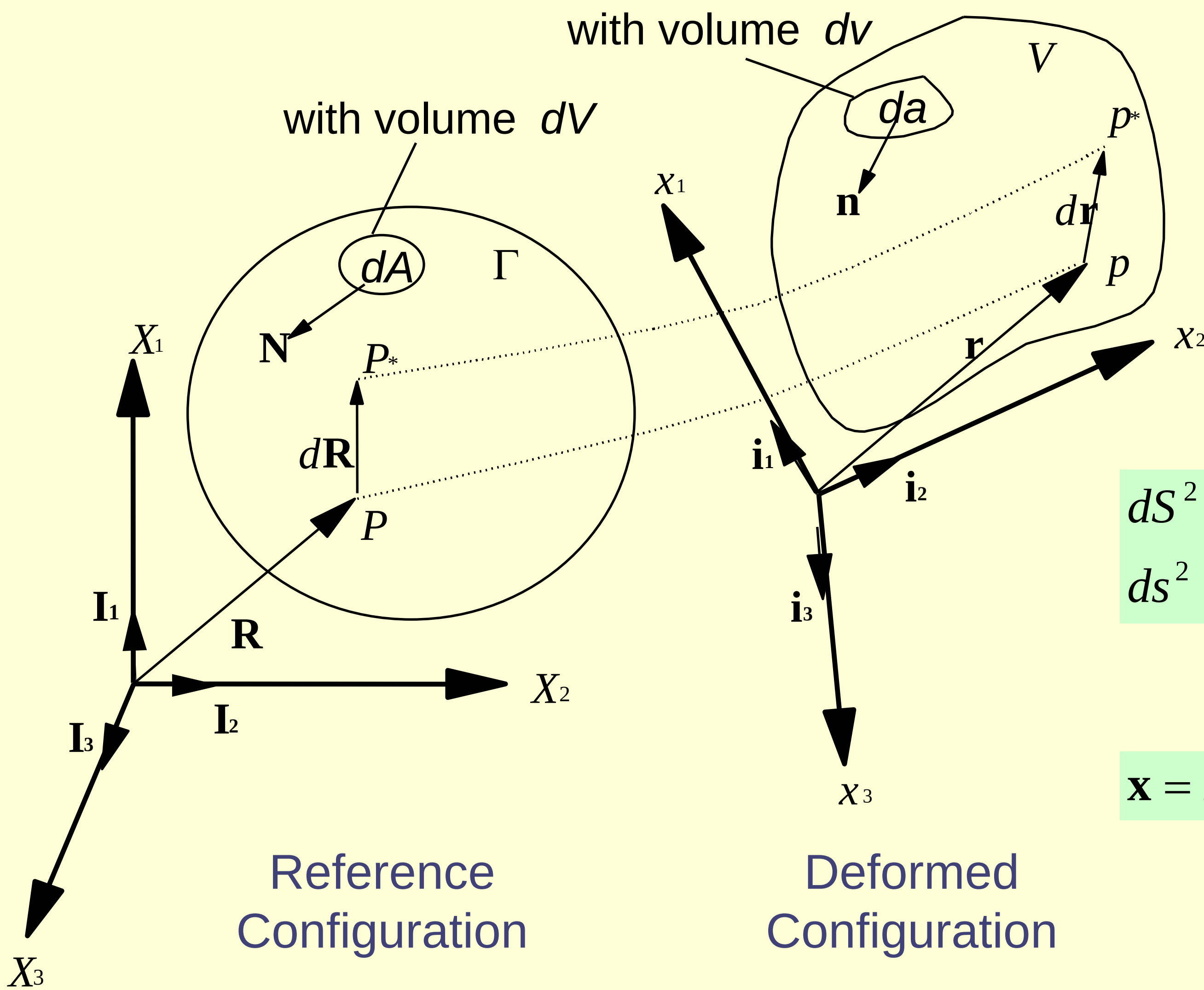
Milestones in Rubber Thermoelasticity

- Entropic origin for the stress (Anthony et al., 1942; Meyer and van der Wyk, 1946).
- Large, multi-axial, isothermal deformations (Treloar, 1944; Rivlin and Saunders,1951)
- Universal controllable deformations of isotropic elastic materials (Ericksen,1954,1955).
- Universal deformations within the context of finite thermo-elasticity (Petroski and Carlson, 1968,1970).
- Generalized thermoelastic models: Chadwick (1974), Chadwick and Creasy (1984), Ogden (1992).
- Many BVPs solved in the last decade (e.g., Horgan, Rajagopal, Saccomandi, Wineman). Non-homogeneity???

Consequences of Material Non-Homogeneity

- Material homogeneity is simply assumed in the rheological characterization of rubbers.
- What do standard tests really measure?
- Three fundamental issues to be addressed:
  - isothermal/non-isothermal behavior of non-homogeneous rubbers (forward problem)
  - tailoring the non-homogeneity
  - characterization of non-homogeneous rubbers (inverse problem)

Kinematics



$$dS^2 = d\mathbf{R}\cdot d\mathbf{R} = dX_K\, dX_K$$

$$ds^2 = d\mathbf{r}\cdot d\mathbf{r} = dx_k\, dx_k$$

Field Variables

$\mathbf{x} = \mathbf{x}(\mathbf{X},t)$ and $\theta = \theta(\mathbf{x},t)$

Deformation Measures

- The deformation gradient **F**

$$\mathbf{F} = \nabla \mathbf{x} \equiv \partial \mathbf{x} / \partial \mathbf{X}$$

- The left (right) Cauchy-Green deformation tensor **B** (**C**) is obtained via polar decomposition of **F**:

$$\mathbf{F} = \mathbf{RU} = \mathbf{VR} \Rightarrow \quad \mathbf{B} \equiv \mathbf{V}^2 = \mathbf{F}\mathbf{F}^{\mathrm{T}} \quad \text{and} \quad \mathbf{C} \equiv \mathbf{U}^2 = \mathbf{F}^{\mathrm{T}}\mathbf{F},$$

- Material isotropy requires that the response functions depend on the principal invariants of **B**, i.e., I_1, I_2, and I_3:

$$I_1 = \operatorname{tr}\mathbf{B}, \quad I_2 = \frac{1}{2}\left[(\operatorname{tr}\mathbf{B})^2 - \operatorname{tr}\mathbf{B}^2\right], \quad I_3 = \frac{1}{6}\left[(\operatorname{tr}\mathbf{B})^3 - 3\operatorname{tr}\mathbf{B}\operatorname{tr}\mathbf{B}^2 + 2\operatorname{tr}\mathbf{B}^3\right]$$

Finite Thermoelasticity

- Thermo-mechanical behavior of non-homogeneous, isotropic non-linearly elastic materials can be described by

$$\psi = \psi(I_1, I_2, I_3, \theta, \mathbf{X}), \quad \eta = -\partial\psi/\partial\theta, \quad \varepsilon = \psi + \theta\eta$$

- The Cauchy stress **T** and the heat flux vector **q** are given by

$$\mathbf{T} = \frac{2\rho_0(\mathbf{X})}{\sqrt{I_3}}\left[\left(I_2\frac{\partial\psi}{\partial I_2} + I_3\frac{\partial\psi}{\partial I_3}\right)\mathbf{I} + \frac{\partial\psi}{\partial I_1}\mathbf{B} - I_3\frac{\partial\psi}{\partial I_2}\mathbf{B}^{-1}\right]$$

$$\mathbf{q} = \mathbf{K}\nabla\theta = \left(\Gamma_0\mathbf{I} + \Gamma_1\mathbf{B} + \Gamma_{-1}\mathbf{B}^{-1}\right)\nabla\theta, \quad \Gamma_i = \Gamma_i(I_1, I_2, I_3, I_4, I_5, I_6, \theta, \mathbf{X})$$

where

$$i \equiv 0, 1, \text{or} -1, \quad I_4 = \nabla\theta\cdot\nabla\theta, \quad I_5 = \nabla\theta\cdot\mathbf{B}\nabla\theta, \quad I_6 = \nabla\theta\cdot\mathbf{B}^{-1}\nabla\theta$$

ψ, η , and ε : specific Helmholtz free energy, entropy, and internal energy

Γ_i : thermo-mechanical response functions

ρ_0 and θ : a reference density and temperature

Entropic Finite Thermoelasticity

- A constrained thermoelastic model:

$$\psi = \psi(I_1, I_2, I_3, \theta, \mathbf{X}) = \frac{\theta}{\rho_0(\mathbf{X})\theta_0} W(I_1, I_2, \mathbf{X}) - \int_{\theta_0}^{\theta} \left(\frac{\theta}{\bar{\theta}} - 1 \right) c(\mathbf{X}, \bar{\theta}) d\bar{\theta} - \frac{\pi}{\rho_0(\mathbf{X})} \left[\sqrt{I_3} - \frac{1}{\phi(\theta)} \right]$$

- The Cauchy stress **T** and the heat flux vector **q** are given by

$$\mathbf{T} = -p\mathbf{I} + 2\phi(\theta)\frac{\theta}{\theta_0}\left[\frac{\partial W}{\partial I_1}\mathbf{B} - \frac{1}{\phi^2(\theta)}\frac{\partial W}{\partial I_2}\mathbf{B}^{-1}\right]$$

$$\mathbf{q} = \left[K_0 - K_1 + K_2(\theta - \theta_0)\right]\mathbf{I} + K_1\mathbf{B}$$

- Clausius-Duhem Inequalities:

$$\sqrt{I_3}\,\mathbf{q}^T \nabla\theta \geq 0 \Rightarrow \Gamma_0 I_4 + \Gamma_1 I_5 + \Gamma_{-1} I_6 \geq 0$$

W : isothermal strain energy function; Constraint: $\rho/\rho_0 = \phi(\theta) = 1/\sqrt{I_3}$

Local Balance Equations

- Balance of Mass:

$$\frac{D\rho}{Dt} + \rho\nabla\cdot\mathbf{v} = 0 \quad \text{or} \quad \rho(\mathbf{x},t) = \frac{\rho_o(\mathbf{X})}{\det\mathbf{F}} = \frac{\rho_o(\mathbf{X})}{\sqrt{I_3}}$$

- Balance of Linear Momentum:

$$\rho\frac{D\mathbf{v}}{Dt} - \nabla\cdot\mathbf{T} - \rho\mathbf{b} = \mathbf{0}$$

- Balance of Thermal Energy:

$$\rho\frac{D\varepsilon}{Dt} - \mathbf{T}:\mathbf{d} - \nabla\cdot\mathbf{q} - \rho s = 0$$

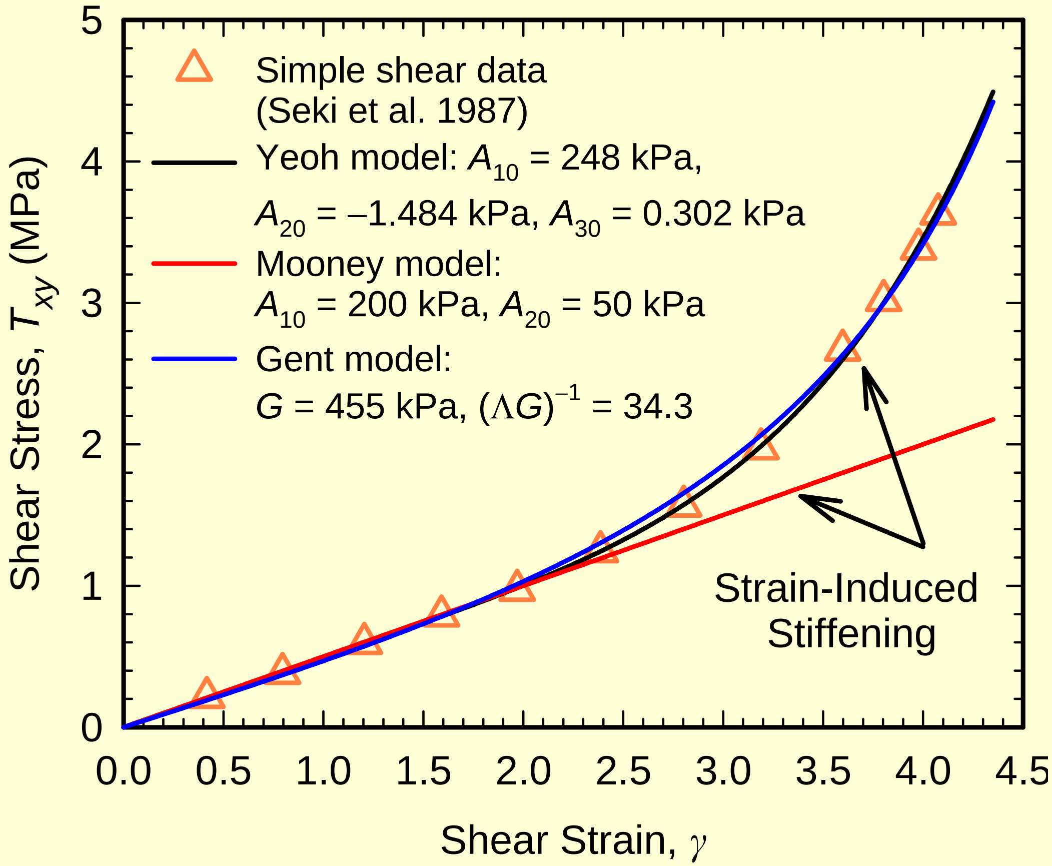
Isothermal Deformation of Homogeneous Rubbers

Neo-Hookean model: $W(I_1) = A_{10}(I_1 - 3)$

Mooney model: $W(I_1, I_2) = A_{10}(I_1 - 3) + A_{01}(I_2 - 3)$

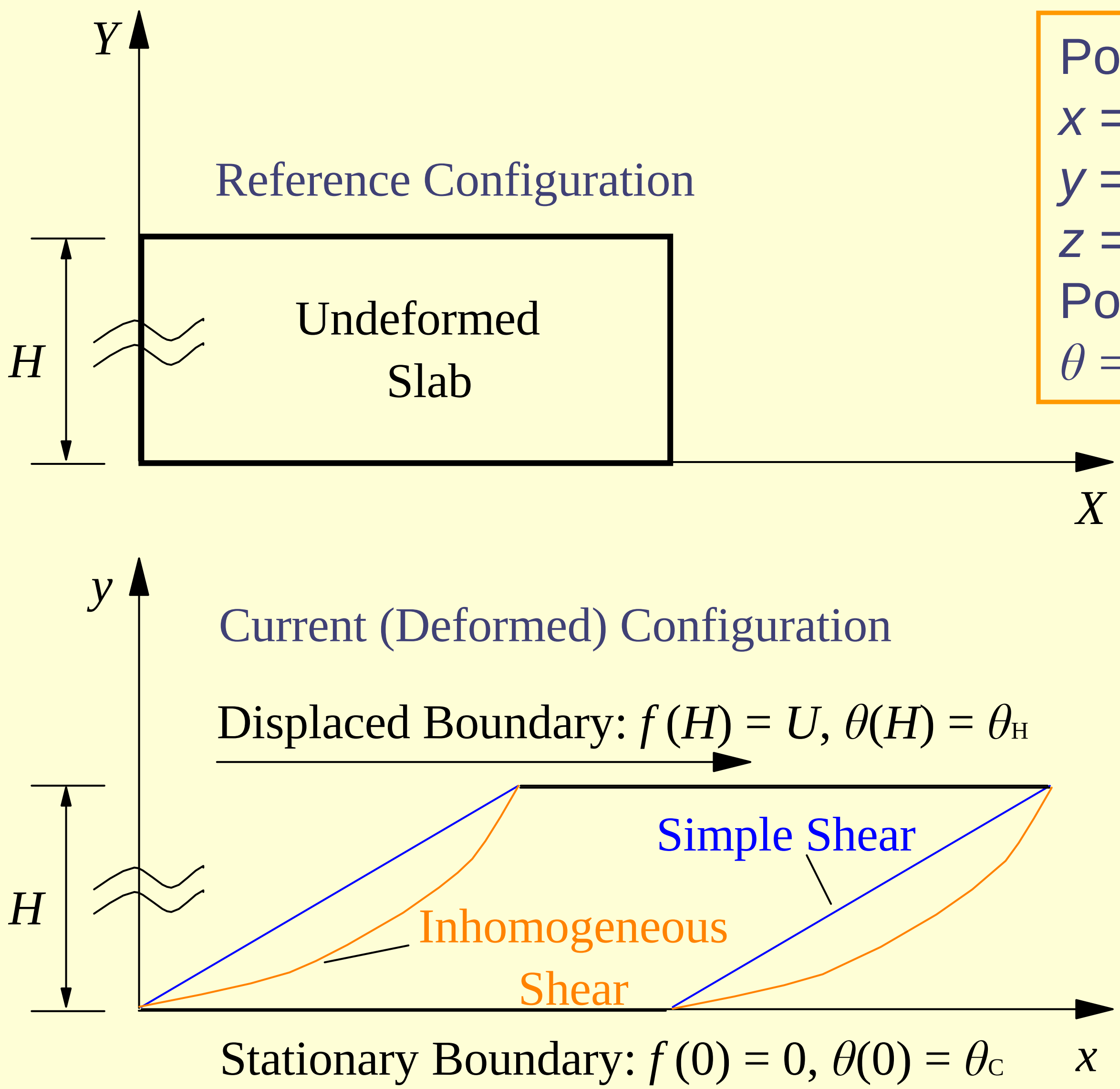
Yeoh model: $W(I_1) = A_{10}(I_1 - 3) + A_{20}(I_1 - 3)^2 + A_{30}(I_1 - 3)^3$

Gent model: $W(I_1) = -\mu J_m \ln[1 - (I_1 - 3)/J_m]/2$



Rectilinear Shearing of Rubber Slabs (I)

Postulated Deformation:
$x = X + f(Y)$
$y = Y$
$z = Z$
Postulated Temperature:
$\theta = \theta(y) = \theta(Y)$



Rectilinear Shear (II): Deformation Measures

- The deformation measures are implicitly given by

$$\mathbf{F} = \frac{\partial \mathbf{x}}{\partial \mathbf{X}} = \begin{bmatrix} 1 & f' & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{bmatrix}, \quad \mathbf{B} = \mathbf{F}\mathbf{F}^{\mathrm{T}} = \begin{bmatrix} 1+(f')^2 & f' & 0 \\ f' & 1 & 0 \\ 0 & 0 & 1 \end{bmatrix}$$

$$\mathrm{I}_1 = \mathrm{tr}\,\mathbf{B} = 3+(f')^2, \quad \mathrm{I}_2 = \frac{1}{2}\left[(\mathrm{tr}\,\mathbf{B})^2 - \mathrm{tr}\,\mathbf{B}^2\right] = 3+(f')^2, \quad \mathrm{I}_3 = \det\mathbf{B} = 1$$

- Due to det F = 1, incompressibility constraint is satisfied. The strain inhomogeneity is determined from:

$$\left\|\nabla\mathbf{F}\right\| := \sqrt{\frac{\partial F_{iJ}}{\partial X_M}\frac{\partial F_{iJ}}{\partial X_M}} = |f''| \qquad i \equiv x, y, z \quad \text{and} \quad J, M \equiv X, Y, Z$$

f and f' : shearing displacement and shear strain

$|f''|$: absolute value of the shear strain gradient

Rectilinear Shear (III): Field Equations

- The Cauchy's equations of motion can be recorded as

$$\nabla\cdot\mathbf{T}=\frac{\partial T_{ij}}{\partial x_j}=\frac{\partial T_{ij}}{\partial X_M}\frac{\partial X_M}{\partial x_j}=\mathbf{0}\quad i,j\equiv x,y,z\quad\text{and}\quad M\equiv X,Y,Z$$

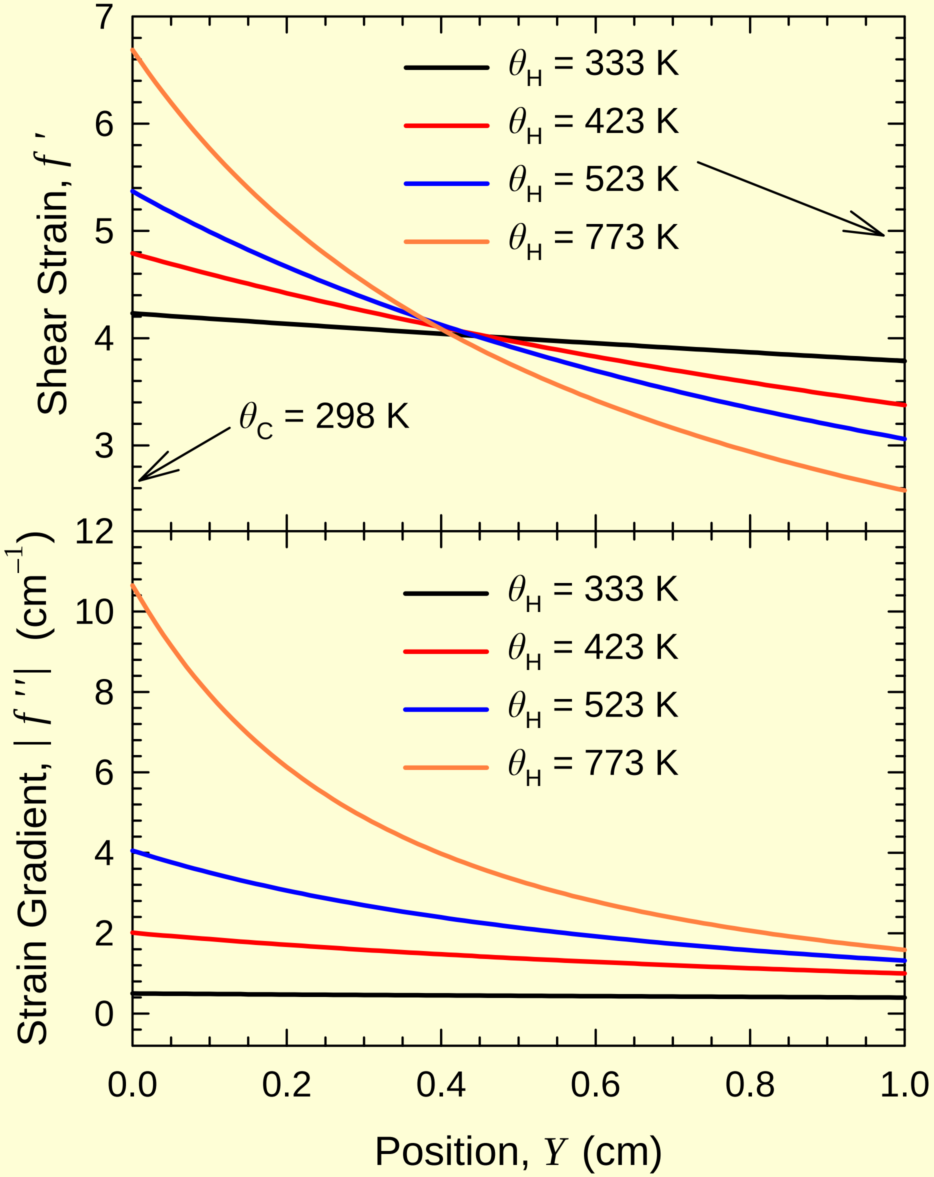
$$\frac{dT_{xy}}{dY}=\frac{d}{dY}\left[\frac{2\theta}{\theta_o}\left(\frac{\partial W}{\partial I_1}+\frac{\partial W}{\partial I_2}\right)f'\right]=0,\quad f(0)=0\quad\text{and}\quad f(H)=U\ \text{ or }\ T_{xy}(H)=S$$

- The local energy balance equation with Fourier's law:

$$\nabla\cdot\mathbf{q}=\frac{d}{dy}\left(k\frac{d\theta}{dy}\right)=k\theta''=0,\quad \theta(0)=\theta_C\quad\text{and}\quad\theta(H)=\theta_H$$

$$\theta=\theta(Y)=\theta_C+\frac{\theta_H-\theta_C}{H}Y$$

Rectilinear Shear (IV): Neo-Hookean Model



Non-isothermal Deformation of a Homogeneous Slab:

$$W(I_1) = A_{10}(I_1 - 3)$$

An increase in temperature gradient enhances the strain inhomogeneity.

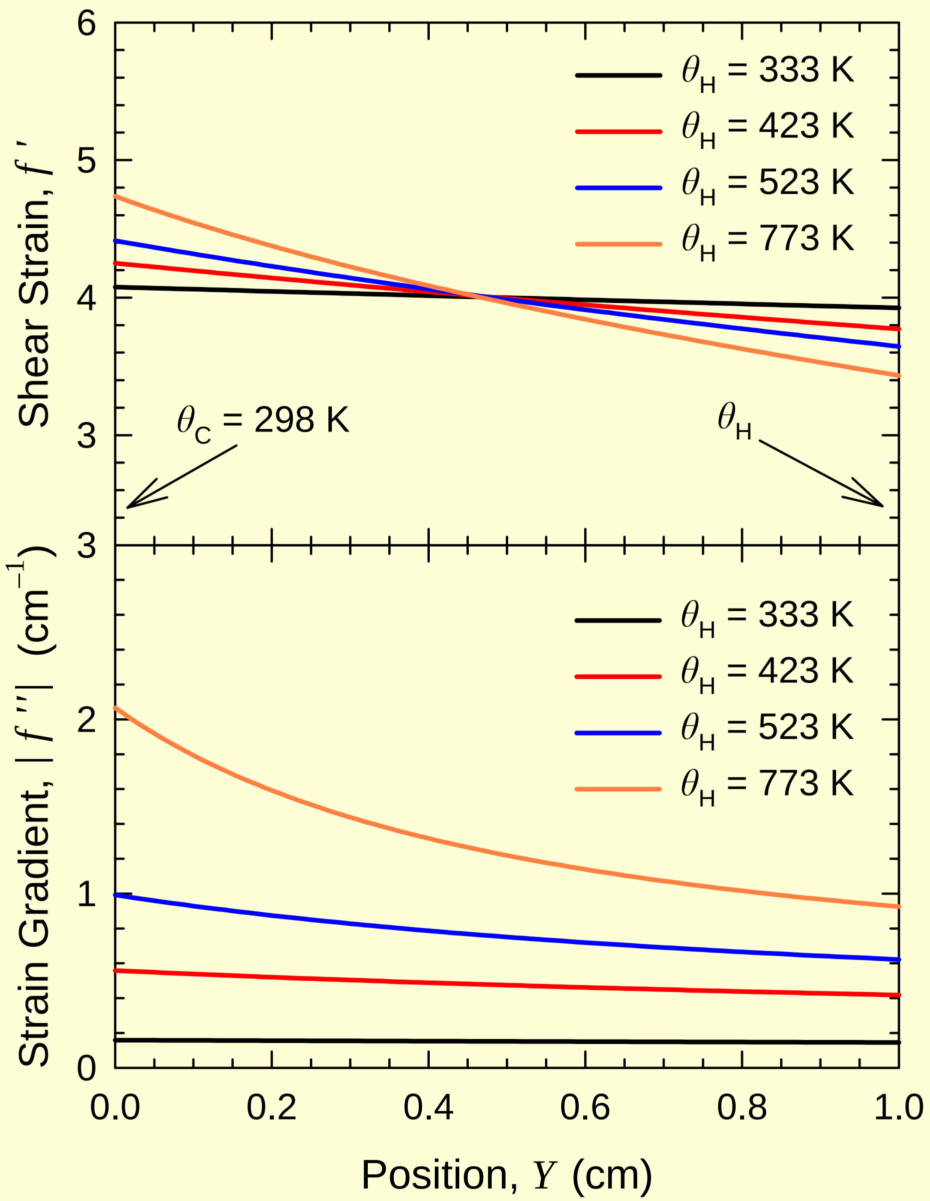
The strain inhomogeneity is more pronounced near the colder boundary.

H = 1 cm, U = 4 cm,

θ_0 = 296 K, θ_C = 298 K,

A_{10} = 250 kPa, θ_H varied

Rectilinear Shear (V): Yeoh Model



Non-isothermal Deformation of a Homogeneous Slab:

$$W(I_1) = A_{10}(I_1 - 3) + A_{20}(I_1 - 3)^2 + A_{30}(I_1 - 3)^3$$

The strain-induced stiffening homogenizes the strain field.

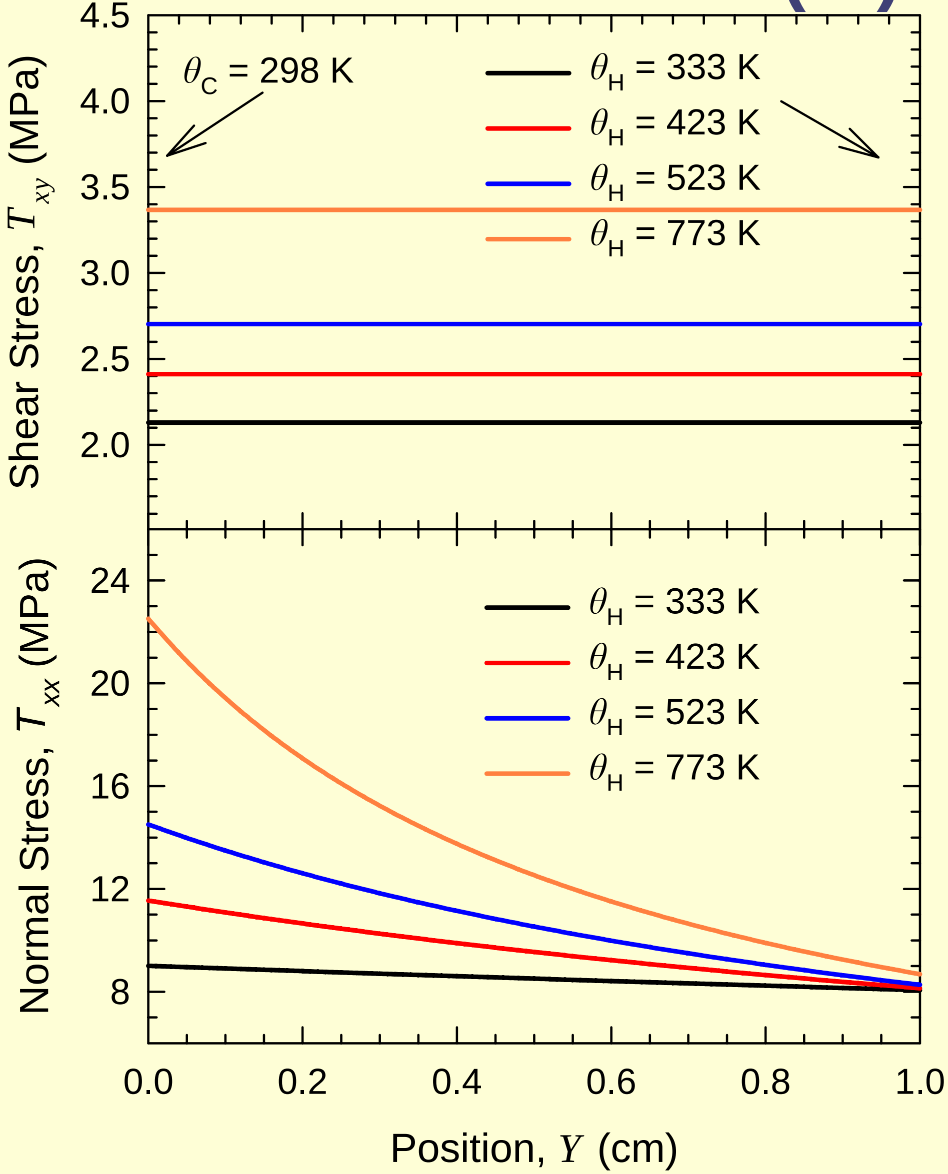
H = 1 cm, U = 4 cm,

θ_0 = 296 K, θ_C = 298 K,

A_{10} = 250 kPa, A_{10} = -1.484 kPa,

A_{30} = 0.302 kPa, θ_H varied

Rectilinear Shear (VI): Neo-Hookean Model



Non-isothermal Deformation of a Homogeneous Slab:

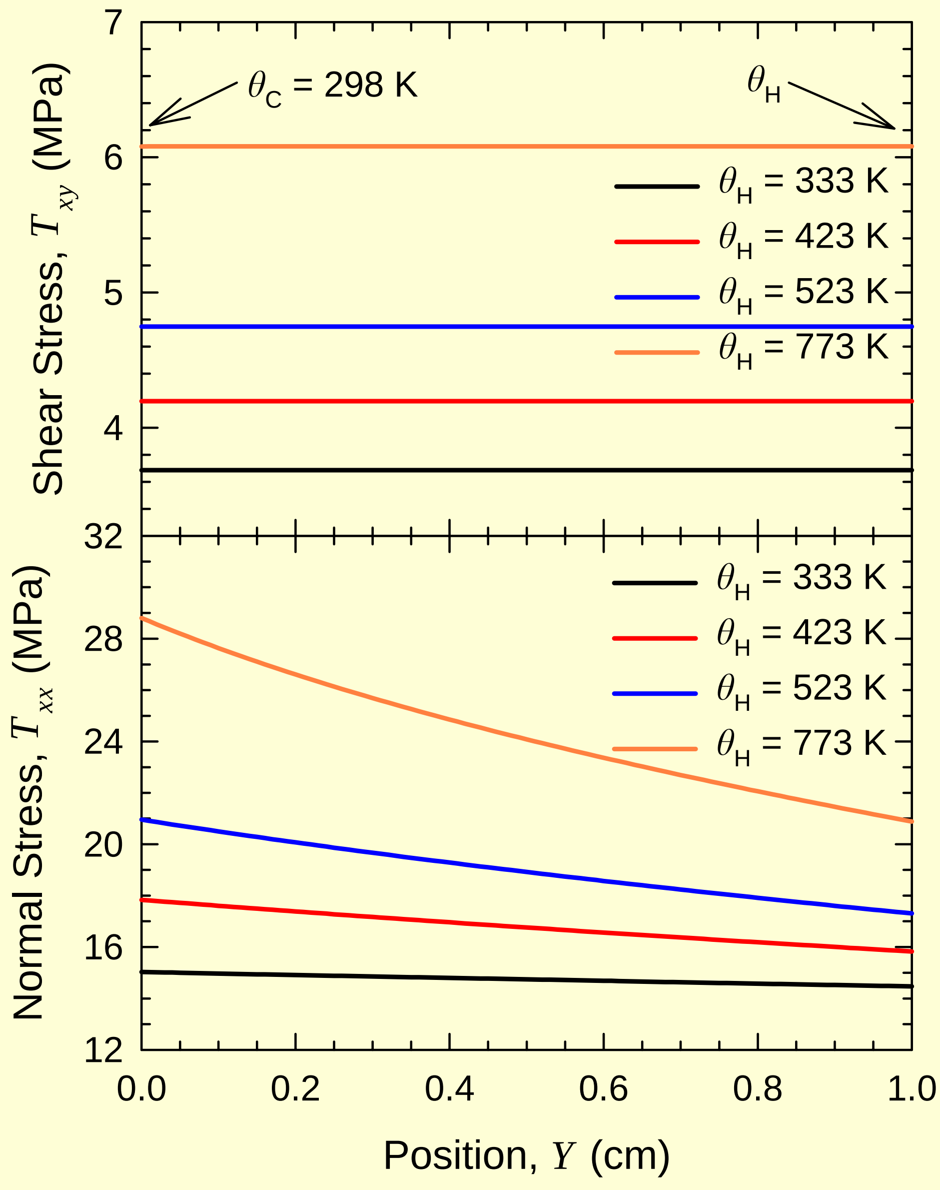
$$W(I_1) = A_{10}(I_1 - 3)$$

Stress increase due to temperature-induced stiffening

Colder boundary subjected to a higher normal stress!

H = 1 cm, U = 4 cm,
θ_0 = 296 K, θ_C = 298 K,
A_{10} = 250 kPa, θ_H varied

Rectilinear Shear (VII): Yeoh Model



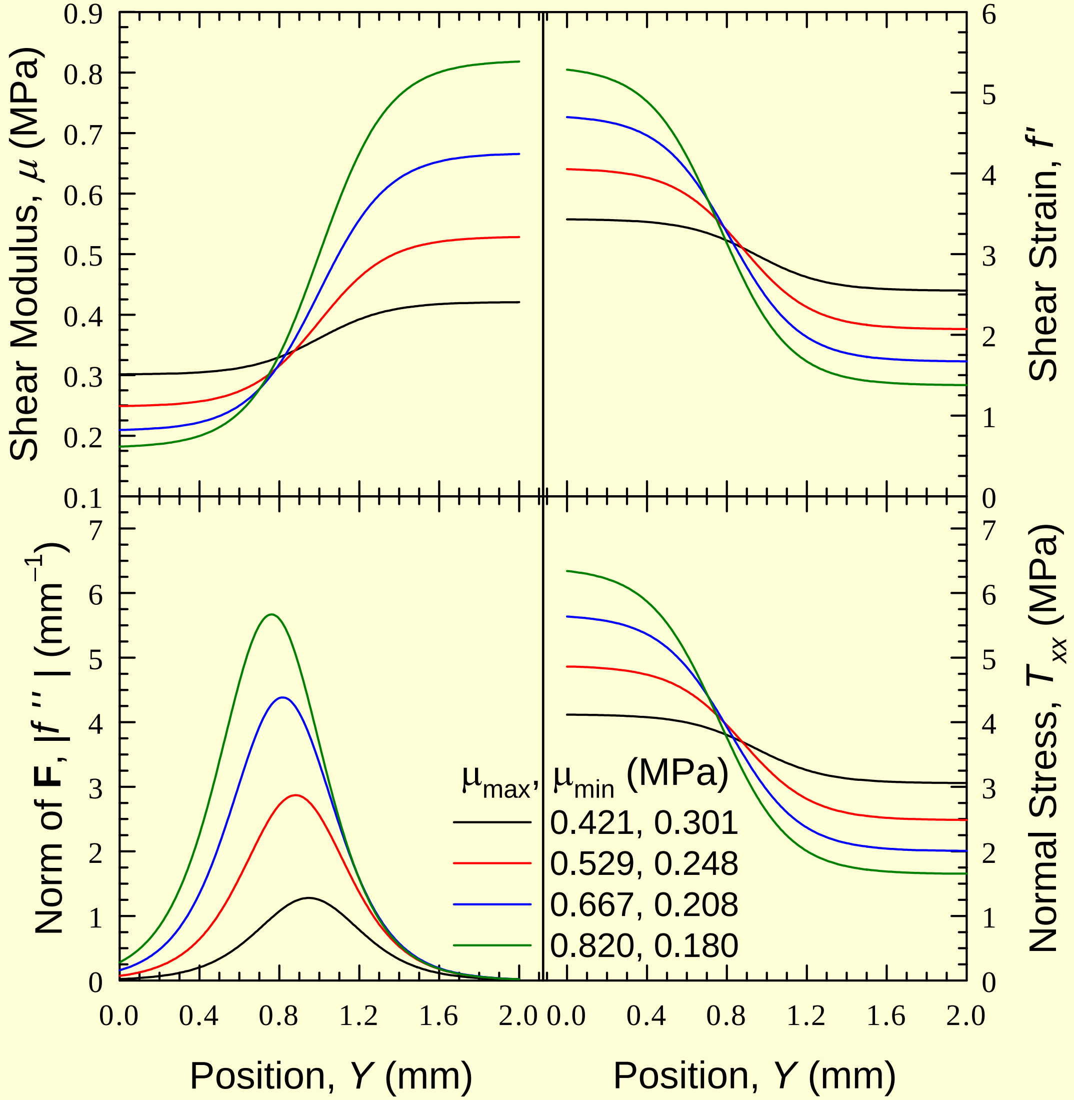
Non-isothermal Deformation of a Homogeneous Slab:

$$W(I_1) = A_{10}(I_1 - 3) + A_{20}(I_1 - 3)^2 + A_{30}(I_1 - 3)^3$$

The stresses are higher and less inhomogeneous in the Yeoh slab than in the Neo-Hookean slab.

H = 1 cm, U = 4 cm,
θ_0 = 296 K, θ_C = 298 K,
A_{10} = 250 kPa, A_{10} = -1.484 kPa,
A_{30} = 0.302 kPa, θ_H varied

Rectilinear Shear (VIII): AB Type Graded Rubbers



Isothermal Deformation of a Non-Homogeneous Slab:

FGR-AB

$$\mu = \mu(Y) = \mu_{\max} - \frac{\mu_{\max} - \mu_{\min}}{1 + 10^{\phi(Y - Y_{\inf})/H}}$$

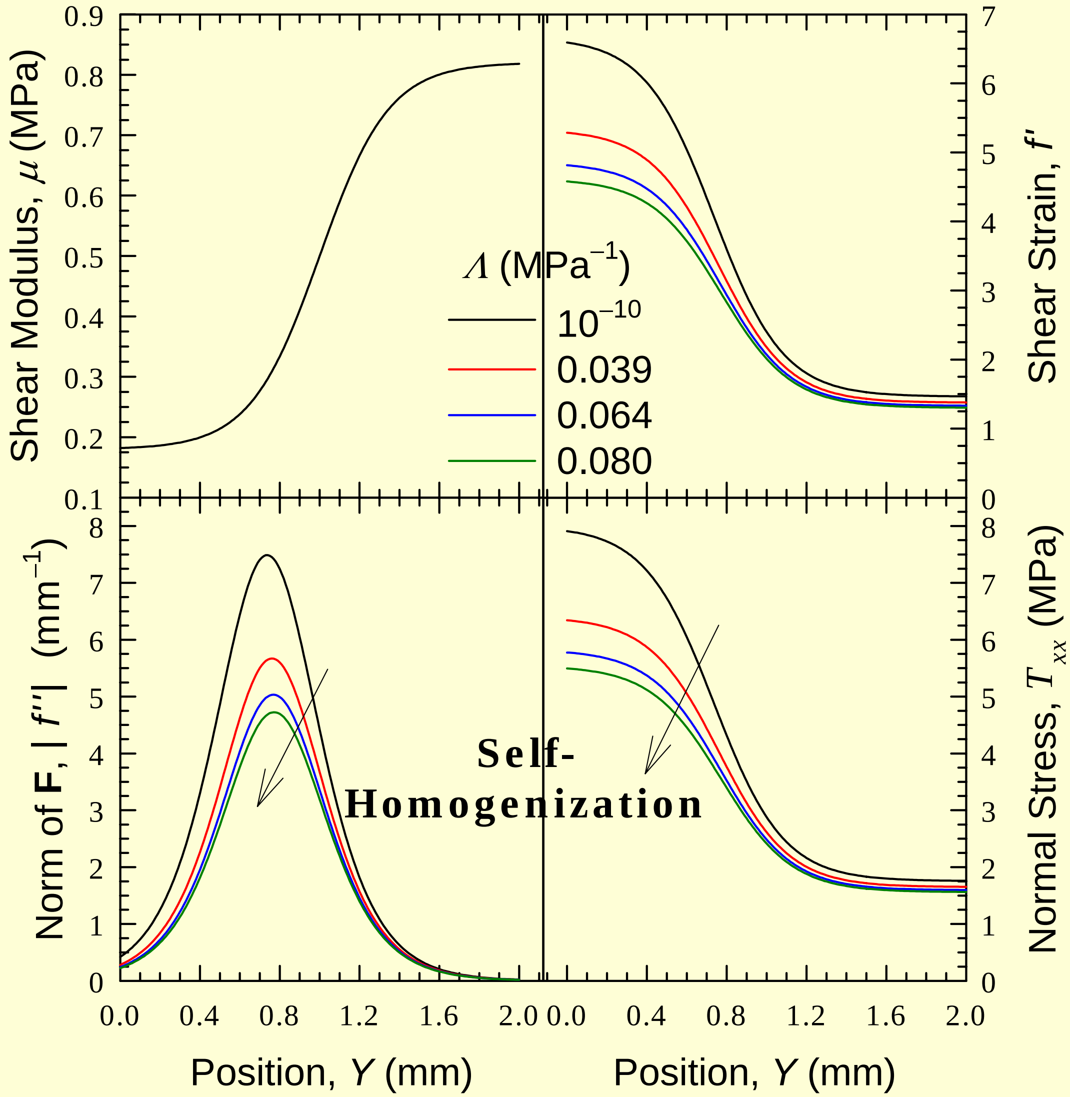
$$W(I_1, Y) = -\frac{\ln[1 - \Lambda\mu(Y)(I_1 - 3)]}{2\Lambda}$$

H = 2 mm, S = 1.2 MPa,

θ_0 = 296 K, ϕ = 5, $Y_{\inf}$ = 1 mm,

Λ = 0.039 MPa^{-1}

Rectilinear Shear (IX): AB Type Graded Rubbers



Isothermal Deformation
of a Non-Homogeneous Slab:

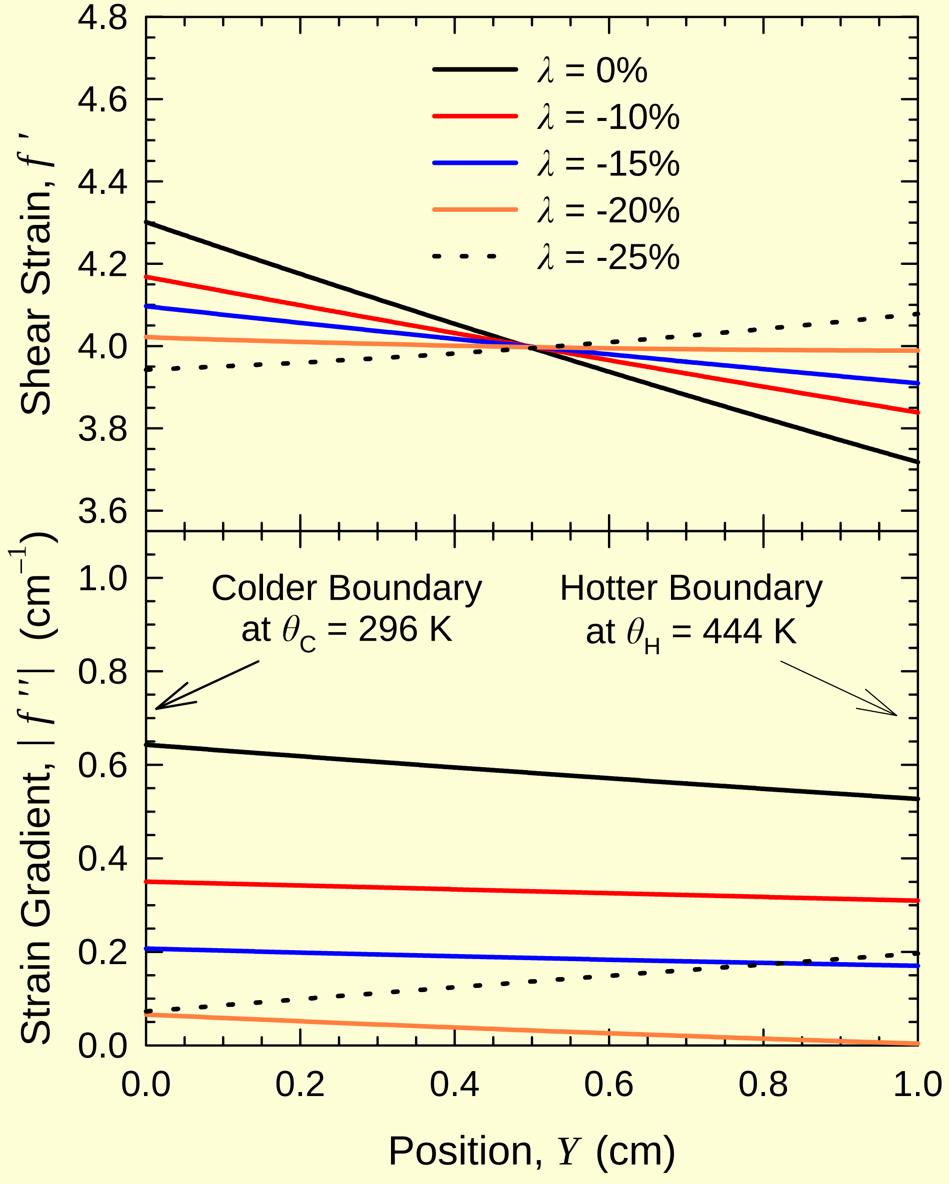
FGR-AB

$$\mu = \mu(Y) = \mu_{\max} - \frac{\mu_{\max} - \mu_{\min}}{1 + 10^{\phi(Y - Y_{\inf})/H}}$$

$$W(I_1, Y) = -\frac{\ln[1 - \Lambda\mu(Y)(I_1 - 3)]}{2\Lambda}$$

H = 2 mm, S = 1.2 MPa,
θ_0 = 296 K, ϕ = 5, $Y_{\inf}$ = 1 mm,
$\mu_{\max}$ = 0.82 MPa, $\mu_{\min}$ = 0.18 MPa

Rectilinear Shear (X): Functional Grading?



Non-isothermal Deformation of a Graded Slab:

$$\mu(Y) = \mu_B \left[1 + \frac{\lambda(1+\kappa)Y}{100(\kappa H + Y)} \right]$$

$$W(I_1, Y) = -\frac{\ln\left[1 - \Lambda\mu(Y)(I_1 - 3)\right]}{2\Lambda}$$

An optimum grading exists!

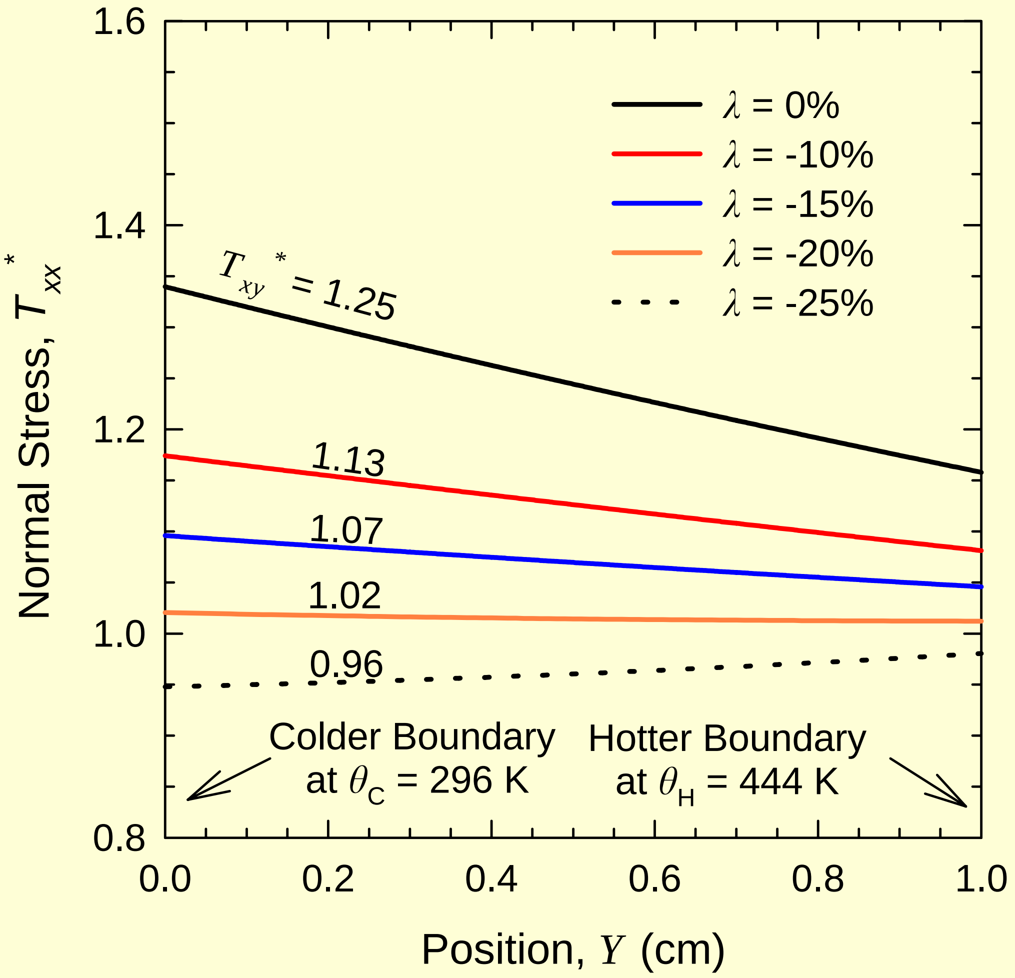
H = 1 cm, U = 4 cm,

$\theta_0 = \theta_C$ = 296 K, θ_H = 444 K,

κ = 5.0, μ_B = 455 kPa,

Λ = 0.0641 MPa^{-1}

Rectilinear Shear (XI): Functional Grading?



Non-isothermal Deformation of a Graded Slab:

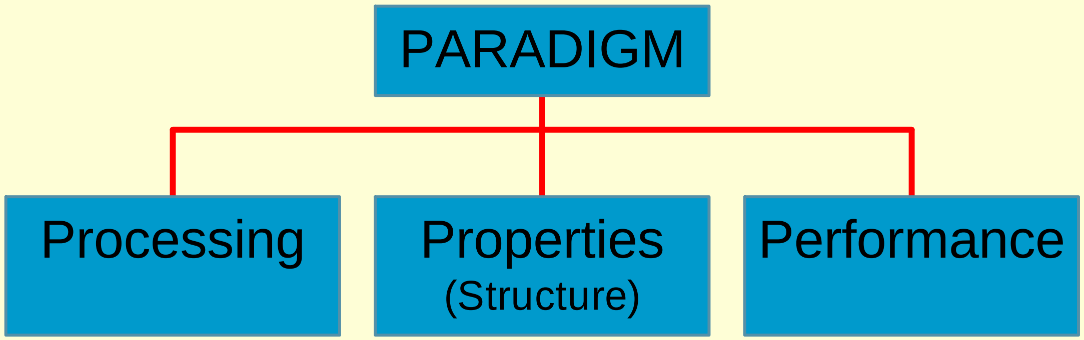
$$\mu(Y) = \mu_B\left[1 + \frac{\lambda(1+\kappa)Y}{100(\kappa H + Y)}\right]$$

$$W(I_1, Y) = -\frac{\ln[1 - \Lambda\mu(Y)(I_1 - 3)]}{2\Lambda}$$

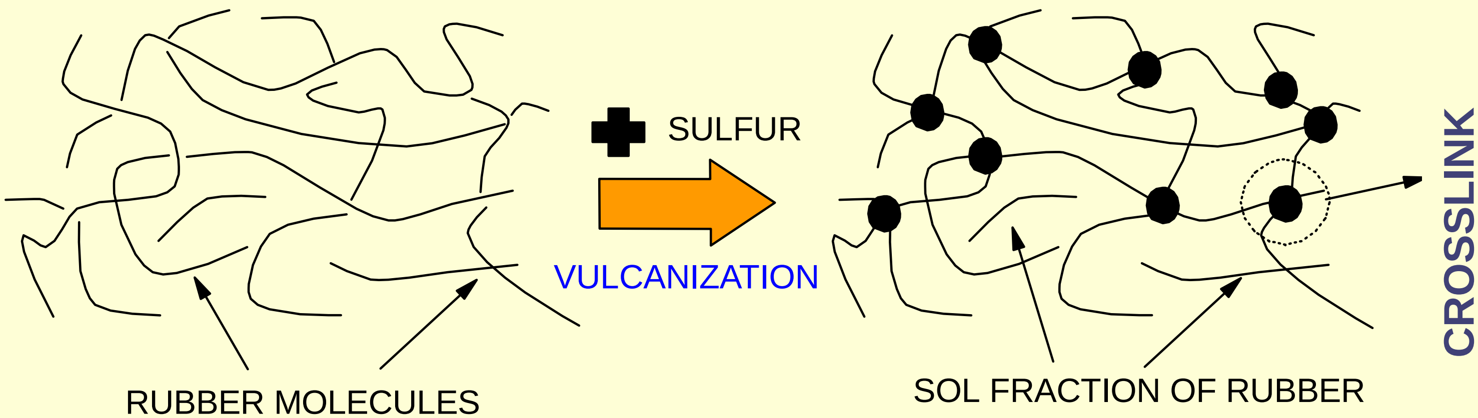
Grading leads to a reduction in stresses.

H = 1 cm, U = 4 cm,
$\theta_0 = \theta_C$ = 296 K, θ_H = 444 K,
κ = 5.0, μ_B = 455 kPa,
Λ = 0.0641 MPa^{-1}

A Perspective in Materials Engineering

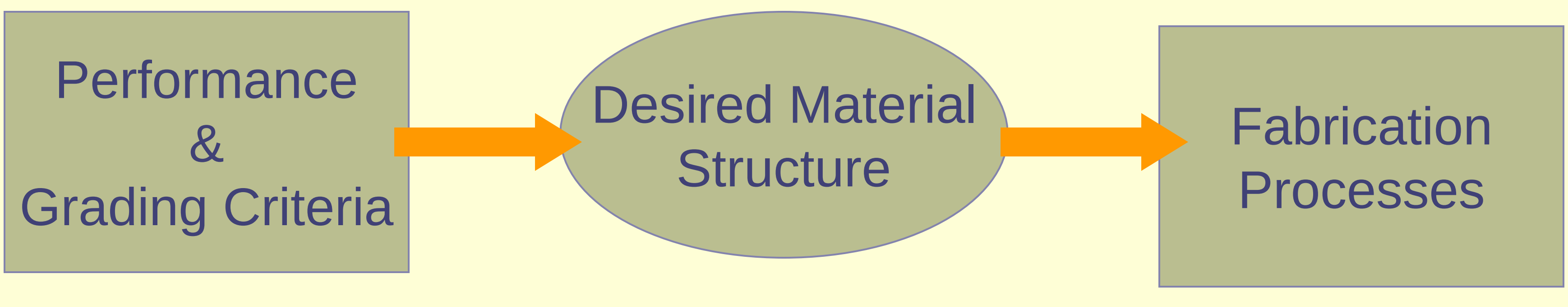


How to generate the spatial variation of the crosslink density $V(\boldsymbol{X})$?



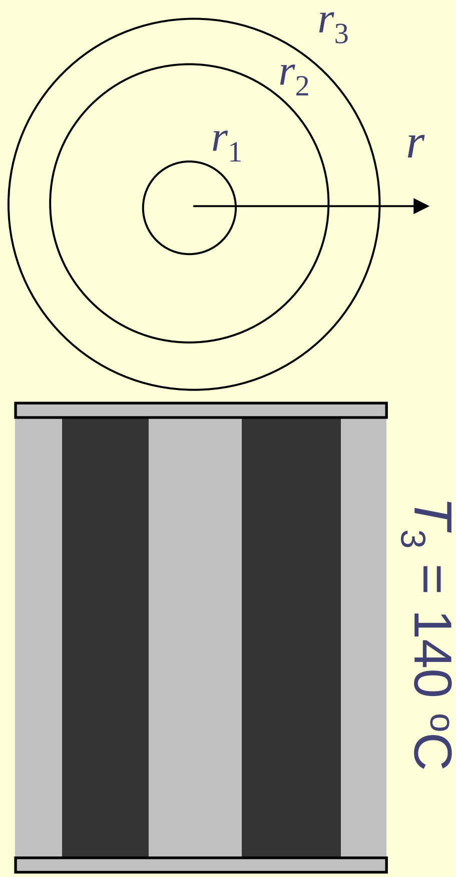
How to Produce FGREMs?

- Construction-based methods: layers with different contents of
  - curing agents (Ikeda, 2003)
  - secondary phase (fillers, recycled rubber powder)
  - rubber blends
- Transport-based methods:
  - Thermally-induced structure (crosslink density)



Molding of a Rubber Tube (I)

Geometry



Rubber: black, Gray: Metal mold

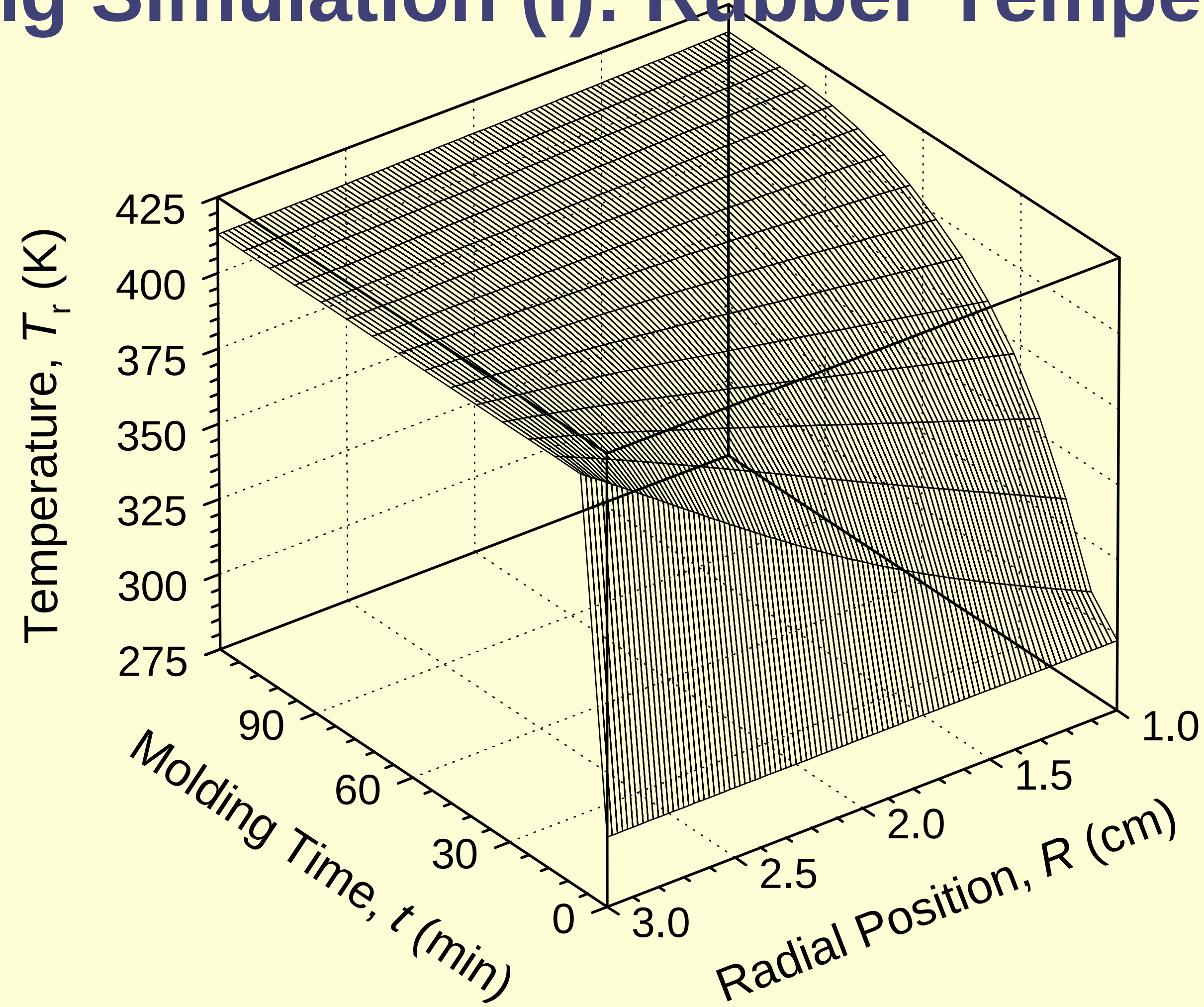
Rubber with curing agents is heated by the steam-jacketed metal mold.

The spatio-temporal development of temperature and crosslink density was simulated.

- Long tube, insulated ends, constant physical properties, except $\lambda_r = \lambda_0 - \lambda_1 (T_r - 273.15)$

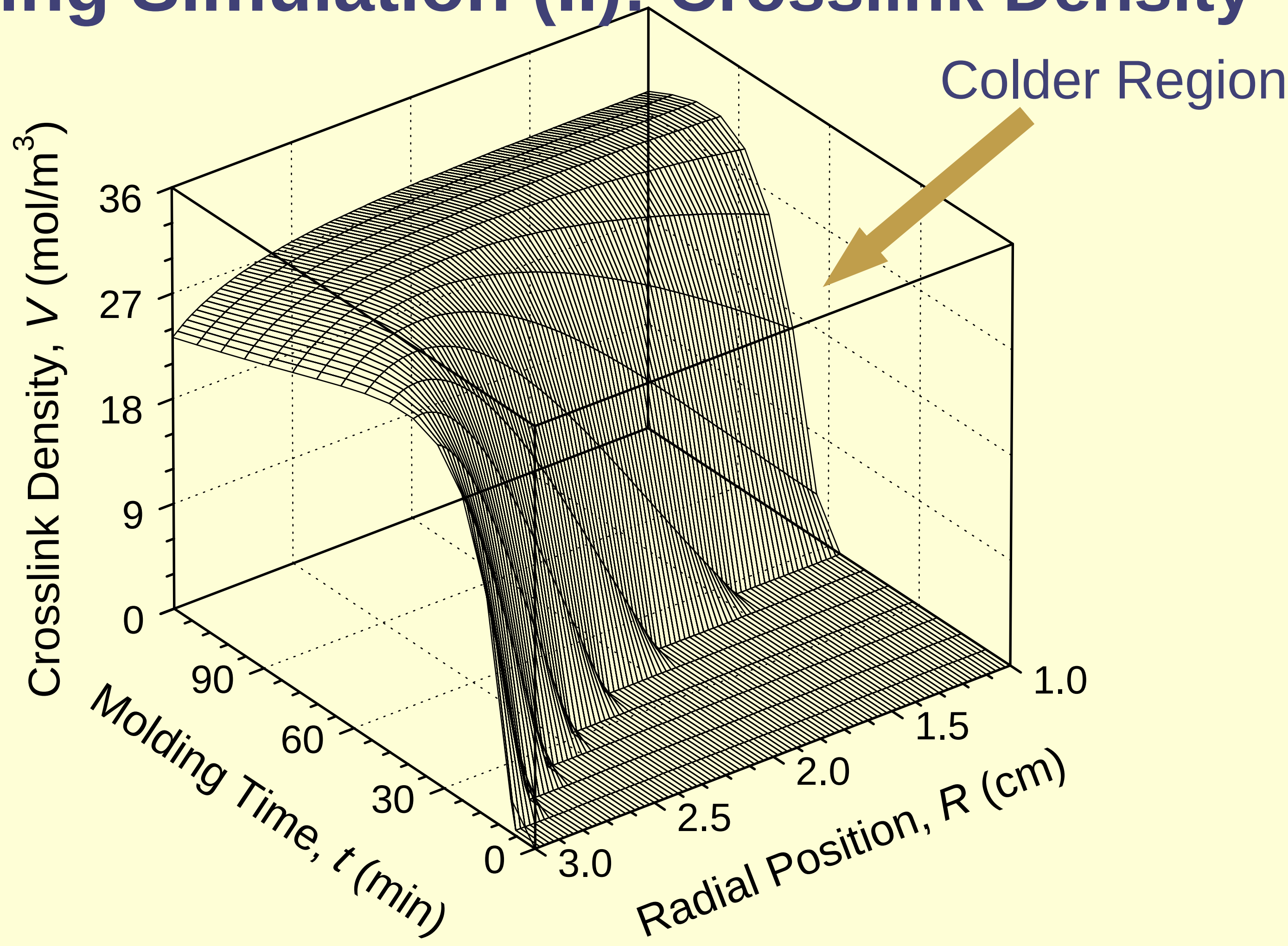
(E. Bilgili, AIChE Annu. Meet., 2003)

Molding Simulation (I): Rubber Temperature



The rubber temperature rises fast near the outer mold (about 413 K), which is heated by steam. It monotonically increases and evolves to a steady state.

Molding Simulation (II): Crosslink Density



The evolution of crosslink density *V* is non-monotonic due to reversion.
Transport-based grading of rubbers is possible via control of the process.

Conclusions

- Rubbers can be graded in a variety of ways.
- Functional grading can reduce stress-strain localizations.
- Sophisticated process models enable us to generate a desired structure.
- An attempt to connect processing-properties-performance aspects of rubbers has been made.

Publications

E. Bilgili, "Functional Grading of Rubber Tubes within the Context of a Molecularly Inspired Finite Thermoelastic Model," *Acta Mech.*, 169, 79-85 (2004).

E. Bilgili, "A Note on the Mechanical Characterization of Non-homogeneous and Graded Vulcanized Rubbers," *Polym. Polym. Compos.*, 12, 221-224 (2004).

E. Bilgili, "A Parametric Study of the Circumferential Shearing of Rubber Tubes: Beyond Isothermality and Material Homogeneity," *Kaut. Gummi Kunstst.*, 56, 671–676 (2003).

E. Bilgili, "Controlling the Stress–Strain Inhomogeneities in Axially Sheared and Radially Heated Hollow Rubber Tubes via Functional Grading," *Mech. Res. Commun.* 30, 257–266 (2003).

E. Bilgili, B. Bernstein, H. Arastoopour, "Effect of Material Non-homogeneity on the Inhomogeneous Shearing Deformation of a Gent Slab Subjected to a Temperature Gradient," *Int. J. Non-Linear Mech.* 38, 1351–1368 (2003).

E. Bilgili, "Computer Simulation as a Tool to Investigate the Shearing Deformation of Non-Homogeneous Elastomers," *J. Elastom. Plast.* 34, 239–264 (2002).

E. Bilgili, B. Bernstein, H. Arastoopour, "Influence of Material Non-Homogeneity on the Shearing Response of a Neo-Hookean Slab," *Rubber Chem. Technol.* 75, 347–363 (2002).

E. Bilgili, B. Bernstein, H. Arastoopour, "Inhomogeneous Shearing Deformation of a Rubber-like Slab within the Context of Finite Thermoelasticity with Entropic Origin for the Stress," *Int. J. Non-Linear Mech.* 36, 887–900 (2001).

Acknowledgments

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