# Tensorial implicit constitutive relations in mechanics of incompressible non-newtonian fluids

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# Constitutive relations

Governing equations (incompressible homogeneous material):

$$\operatorname{div} \mathbf{v} = 0$$

$$\rho \frac{\mathrm{d}\mathbf{v}}{\mathrm{d}t} = \operatorname{div} \mathbb{T} + \rho \mathbf{b}$$

$$\mathbb{T} = \mathbb{T}^{\top}$$

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Constitutive relations (Navier–Stokes), $\mathbb{D} =_{\mathrm{def}} \frac{1}{2}\left(\nabla \mathbf{v} + \nabla \mathbf{v}^\top\right)$:

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Different perspective, $\operatorname{Tr} \mathbb{D} = \operatorname{div} \mathbf{v} = 0$, $\mathbb{T}_\delta =_{\mathrm{def}} \mathbb{T} - \frac{1}{3} \operatorname{Tr}(\mathbb{T})\, \mathbb{I}$:

$$\mathbb{T}_\delta = 2\mu\mathbb{D}$$

# Constitutive relations for non-Newtonian fluids

Standard approach: Stress is an function of kinematical variables.

$$\mathbb{T}_\delta = \mathfrak{f}(\mathbb{D})$$

Example:

$$\mathbb{T}_\delta = 2\left(\mu_\infty + \frac{\mu_0 - \mu_\infty}{\left(1 + \alpha \left|\mathbb{D}\right|^2\right)^{\frac{n}{2}}}\right)\mathbb{D}$$

Pierre J. Carreau. Rheological equations from molecular network theories. *J. Rheol.*, 16(1):99–127, 1972

This approach dominates the standard phenomenological theory of constitutive relations.

C. Truesdell and W. Noll. The non-linear field theories of mechanics. In S. Flüge, editor, *Handbuch der Physik*, volume III/3. Springer, Berlin, 1965

# Constitutive relations for non-Newtonian fluids

Alternative approach: There is a relation between stress and kinematical variables.

$$\mathfrak{f}(\mathbb{T}_\delta, \mathbb{D}) = \mathbb{0}$$

Example:

$$\mathbb{T}_\delta = 2\left(\mu_\infty + (\mu_0 - \mu_\infty)\,\mathrm{e}^{-\frac{|\mathbb{T}_\delta|}{\tau_0}}\right)\mathbb{D}$$

Gilbert R. Seely. Non-newtonian viscosity of polybutadiene solutions. *AIChE J.*, 10(1):56–60, 1964

# Constitutive relations for non-Newtonian fluids

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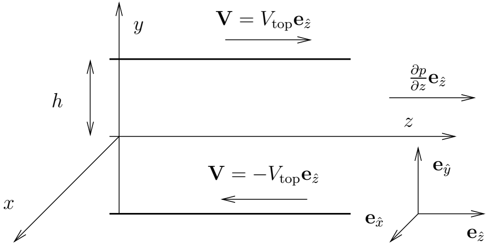
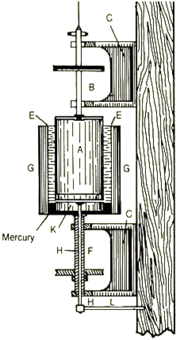
# Shear stress and shear rate



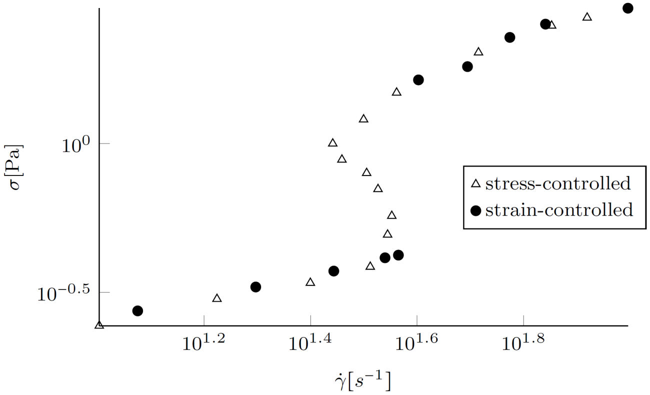




$$\mathbb{T} = \begin{bmatrix} \mathrm{T}_{\hat{x}\hat{x}} & 0 & 0 \\ 0 & \mathrm{T}_{\hat{y}\hat{y}} & \mathrm{T}_{\hat{y}\hat{z}} \\ 0 & \mathrm{T}_{\hat{z}\hat{y}} & \mathrm{T}_{\hat{z}\hat{z}} \end{bmatrix} \qquad \mathbb{D} = \frac{1}{2}\begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & \frac{\mathrm{d}v^{\hat{z}}}{\mathrm{d}y} \\ 0 & \frac{\mathrm{d}v^{\hat{z}}}{\mathrm{d}y} & 0 \end{bmatrix}$$

$$\sigma =_{\text{def}} \mathrm{T}_{\hat{y}\hat{z}} \quad \text{(shear stress)} \qquad \dot{\gamma} =_{\text{def}} \frac{\mathrm{d}v^{\hat{z}}}{\mathrm{d}y} \quad \text{(shear rate)}$$

# Example

$\mathbb{T} \approx \sigma$ (shear stress) $\quad \mathbb{D} \approx \dot{\gamma}$ (shear rate, strain rate)




Philippe Boltenhagen, Yuntao Hu, E. F. Matthys, and D. J. Pine. Observation of bulk phase separation and coexistence in a sheared micellar solution. *Phys. Rev. Lett.*, 79:2359–2362, Sep 1997

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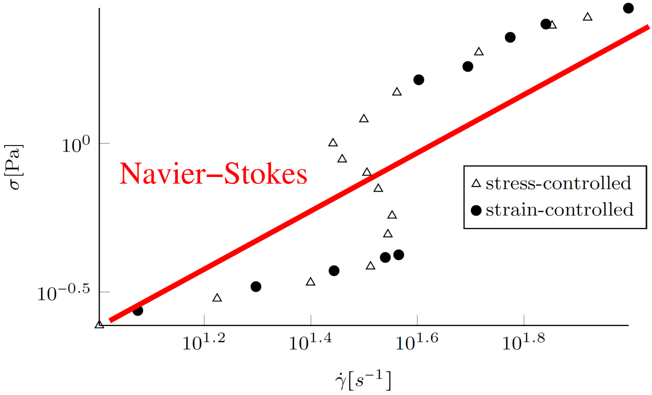


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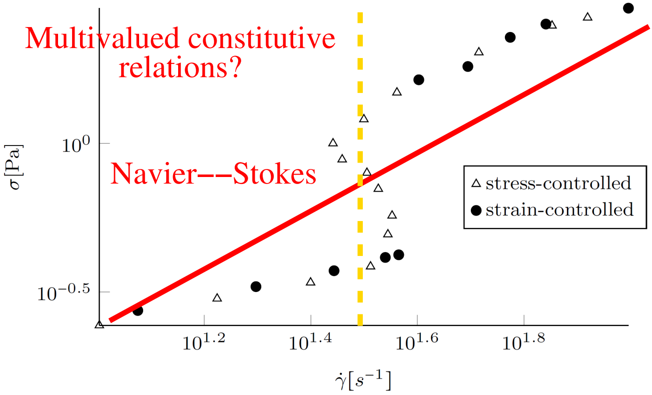


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# One-dimensional implicit type relations

One dimensional data:

$$\mathbb{T} \approx \sigma \text{ (shear stress)} \qquad \mathbb{D} \approx \dot{\gamma} \text{ (shear rate, strain rate)}$$

Standard approach (does not work):

$$\mathbb{T}_\delta = \mathfrak{f}(\mathbb{D})$$

Alternative approach:

$$\mathfrak{f}(\mathbb{T}_\delta, \mathbb{D}) = \mathbb{0} \qquad \text{or} \qquad \mathbb{D} = \mathfrak{f}(\mathbb{T}_\delta)$$

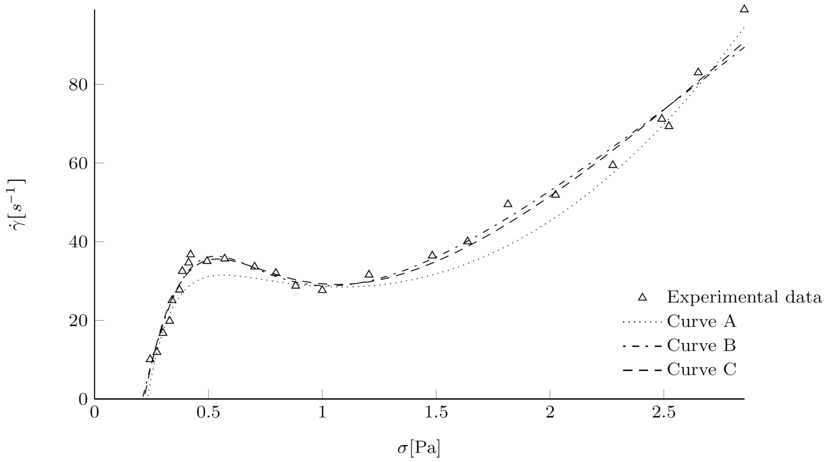
Curves:

$$\dot{\gamma} = \mathrm{e}^{-a\sigma}(a_1\sigma + b_1) + \left(1 - \mathrm{e}^{-b\sigma}\right)(a_2\sigma + b_2) \qquad \text{(A)}$$

$$\dot{\gamma} = \frac{p_1\sigma^3 + p_2\sigma^2 + p_3\sigma + p_4}{\sigma^2 + q_1\sigma + q_2} \qquad \text{(B)}$$

$$\dot{\gamma} = \left(\alpha\left(1 + \beta\sigma^2\right)^n + \gamma\right)\sigma \qquad \text{(C)}$$

# One dimensional implict type relations – curve fitting



# Reconstruction of the tensorial constitutive relation from one-dimensional data

Task:

$$f(\sigma, \dot{\gamma}) = 0 \mapsto \mathfrak{f}(\mathbb{T}_\delta, \mathbb{D}) = \mathbb{0}$$

Experimental data:

$$\mathbb{T} = \begin{bmatrix} \mathrm{T}_{\hat{x}\hat{x}} & 0 & 0 \\ 0 & \mathrm{T}_{\hat{y}\hat{y}} & \mathrm{T}_{\hat{y}\hat{z}} \\ 0 & \mathrm{T}_{\hat{z}\hat{y}} & \mathrm{T}_{\hat{z}\hat{z}} \end{bmatrix} \qquad \mathbb{D} = \frac{1}{2} \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & \frac{\mathrm{d}v^{\hat{z}}}{\mathrm{d}y} \\ 0 & \frac{\mathrm{d}v^{\hat{z}}}{\mathrm{d}y} & 0 \end{bmatrix}$$

$$\sigma =_{\mathrm{def}} \mathrm{T}_{\hat{y}\hat{z}} \quad \text{(shear stress)} \qquad \dot{\gamma} =_{\mathrm{def}} \frac{\mathrm{d}v^{\hat{z}}}{\mathrm{d}y} \quad \text{(shear rate)}$$

# Reconstruction of the tensorial constitutive relation from one-dimensional data – curve B

Task:

$$f(\sigma, \dot{\gamma}) = 0 \mapsto \mathfrak{f}(\mathbb{T}_\delta, \mathbb{D}) = \mathbb{0}$$

Fit of one dimensional experimental data:

$$\left(\sigma^2 + q_1\sigma + q_2\right)\dot{\gamma} = \left(p_1\sigma^2 + p_2\sigma + p_3\right)\sigma$$

Alternatives:

$$\left(\left|\mathbb{T}_\delta\right|^2 + q_1\left|\mathbb{T}_\delta\right| + q_2\right)\mathbb{D} = \left(p_1\left|\mathbb{T}_\delta\right|^2 + p_2\left|\mathbb{T}_\delta\right| + p_3\right)\mathbb{T}_\delta$$

$$\left(\mathbb{T}_\delta^2\mathbb{D} + \mathbb{D}\mathbb{T}_\delta^2\right)_\delta + \tilde{q}_1\left(\mathbb{T}_\delta\mathbb{D} + \mathbb{D}\mathbb{T}_\delta\right)_\delta + q_2\mathbb{D} = \left(p_4\left|\mathbb{T}_\delta\right|^2 + p_3\left|\mathbb{T}_\delta\right| + p_2\right)\mathbb{T}_\delta$$

$$\left(\mathbb{T}_\delta^2\mathbb{D} + \mathbb{D}\mathbb{T}_\delta^2\right)_\delta + q_1\left|\mathbb{T}_\delta\right|\mathbb{D} + q_2\mathbb{D} = \left(p_4\left|\mathbb{T}_\delta\right|^2 + p_2\right)\mathbb{T}_\delta + p_3\left(\mathbb{T}_\delta^2\right)_\delta$$

# Non-newtonian fluids and normal stress differences

(a) Weissenberg effect.

(b) Barus effect.

Normal stress differences:

$$N_1 =_{\text{def}} \mathrm{T}_{\hat{z}\hat{z}} - \mathrm{T}_{\hat{y}\hat{y}}$$
$$N_2 =_{\text{def}} \mathrm{T}_{\hat{y}\hat{y}} - \mathrm{T}_{\hat{x}\hat{x}}$$

# Non-newtonian fluids and normal stress differences

Navier–Stokes, $\mathbb{T} = -p\mathbb{I} + 2\mu\mathbb{D}$:

$$\begin{bmatrix} \mathrm{T}_{\hat{x}\hat{x}} & 0 & 0 \\ 0 & \mathrm{T}_{\hat{y}\hat{y}} & \mathrm{T}_{\hat{y}\hat{z}} \\ 0 & \mathrm{T}_{\hat{z}\hat{y}} & \mathrm{T}_{\hat{z}\hat{z}} \end{bmatrix} = -p \begin{bmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{bmatrix} + \mu \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & \frac{\mathrm{d}v^{\hat{z}}}{\mathrm{d}y} \\ 0 & \frac{\mathrm{d}v^{\hat{z}}}{\mathrm{d}y} & 0 \end{bmatrix}$$

A non-newtonian model, $\mathbb{T} = -p\mathbb{I} + 2\mu\mathbb{D} + 4\tilde{\mu}\mathbb{D}^2$:

$$\begin{bmatrix} \mathrm{T}_{\hat{x}\hat{x}} & 0 & 0 \\ 0 & \mathrm{T}_{\hat{y}\hat{y}} & \mathrm{T}_{\hat{y}\hat{z}} \\ 0 & \mathrm{T}_{\hat{z}\hat{y}} & \mathrm{T}_{\hat{z}\hat{z}} \end{bmatrix} = -p \begin{bmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{bmatrix} + \mu \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & \frac{\mathrm{d}v^{\hat{z}}}{\mathrm{d}y} \\ 0 & \frac{\mathrm{d}v^{\hat{z}}}{\mathrm{d}y} & 0 \end{bmatrix}$$

$$+ \tilde{\mu} \begin{bmatrix} 0 & 0 & 0 \\ 0 & \left(\frac{\mathrm{d}v^{\hat{z}}}{\mathrm{d}y}\right)^2 & 0 \\ 0 & 0 & \left(\frac{\mathrm{d}v^{\hat{z}}}{\mathrm{d}y}\right)^2 \end{bmatrix}$$

# Key question

What are the implications of the implicit constitutive relations with respect to the modelling of normal stress differences?

# General algebraic implicit constitutive relation – restrictions

Incompressible, homogeneous, isotropic fluid:

$$\alpha_1 \mathbb{T}_\delta + \alpha_2 \mathbb{D} + \alpha_3 \left(\mathbb{T}_\delta^2\right)_\delta + \alpha_4 \left(\mathbb{D}^2\right)_\delta + \alpha_5 (\mathbb{T}_\delta \mathbb{D} + \mathbb{D}\mathbb{T}_\delta)_\delta + \alpha_6 \left(\mathbb{T}_\delta^2 \mathbb{D} + \mathbb{D}\mathbb{T}_\delta^2\right)_\delta + \alpha_7 \left(\mathbb{T}_\delta \mathbb{D}^2 + \mathbb{D}^2 \mathbb{T}_\delta\right)_\delta + \alpha_8 \left(\mathbb{T}_\delta^2 \mathbb{D}^2 + \mathbb{D}^2 \mathbb{T}_\delta^2\right)_\delta = \mathbb{0}$$

Second law of thermodynamics:

$$\mathbb{T} : \mathbb{D} \geq 0$$

Dynamical admissibility in simple shear flow:

$$\mathbf{v} = \frac{V_{\mathrm{top}}}{h} \mathbf{e}_{\hat{z}}$$

## Simple implicit model $\alpha_1 \mathbb{T}_\delta + \alpha_2 \mathbb{D} + \alpha_3 \left(\mathbb{T}^2_\delta\right)_\delta = 0$

Cauchy stress tensor:

$$\mathbb{T} = \begin{bmatrix} \mathrm{C} & 0 & 0 \\ 0 & \mathrm{C} + \mathrm{A} & \mathrm{T} \\ 0 & \mathrm{T} & \mathrm{C} + \mathrm{B} \end{bmatrix}$$

Normal stress differences:

$$N_1 =_{\mathrm{def}} \mathrm{B} - \mathrm{A} \qquad N_2 =_{\mathrm{def}} \mathrm{A}$$

Dynamical admissibility:

$$(\mathrm{A} - \mathrm{B}) \left( \frac{\mathrm{A} + \mathrm{B}}{3} \alpha_3 + \alpha_1 \right) = 0$$

$$-\alpha_1 \frac{\mathrm{A} + \mathrm{B}}{3} - \alpha_3 \left( \frac{3 (\mathrm{A} - \mathrm{B})^2 - (\mathrm{A} + \mathrm{B})^2}{18} + \frac{2}{3} \mathrm{T}^2 \right) = 0$$

$$\alpha_1 \mathrm{T} + \frac{\alpha_2}{2} \frac{\mathrm{d} v^{\hat{z}}}{\mathrm{d} y} + \alpha_3 \mathrm{T} \frac{\mathrm{A} + \mathrm{B}}{9} = 0$$

# Simple implicit model $\alpha_1 \mathbb{T}_\delta + \alpha_2 \mathbb{D} + \alpha_3 \left(\mathbb{T}_\delta^2\right)_\delta = 0$

Simple model ($\alpha$ is a positive constant):

$$\alpha \mathbb{T}_\delta - \frac{1}{2} \frac{\left|\mathbb{T}_\delta^2\right| \sqrt{1 + \tilde{\gamma} \left|\mathbb{D}\right|^2}}{\left|\mathbb{D}\right|^2} \mathbb{D} + \left(\mathbb{T}_\delta^2\right)_\delta = 0$$

Features of the model:

- thermodynamically admissible
- dynamically admissible
- nonzero normal stress differences $(\mathrm{A} = \mathrm{B} \neq 0)$

Earlier, we have seen that implicit constitutive relations can:

- fit experimental data with $S$-shaped graphs of shear stress versus shear rate

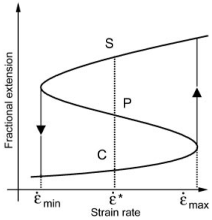



**Fig. 1.** Sketch of de Gennes' classic steady-state extension curve for polymers in extensional flow. De Gennes argued that polymers could exist in two physically realizable states (a stretched "S" and coiled "C" state) in a narrow range of flow strengths. The coiled and stretched polymer configurations correspond to free-energy minima $E/kT$ in a double-welled potential, separated by an energy barrier with a maximum at extension "P." If a coiled polymer is exposed to an adiabatic increase in the strain rate, there would exist a particular $\dot{\varepsilon} = \dot{\varepsilon}^*$ at which $E^C/kT = E^S/kT$, at which the polymer would spend equal amounts of time in the coiled and stretched states. However, given the limited residence and observation times for polymers in extensional flows, a hysteresis in extension would occur for most practical situations for $\dot{\varepsilon}$ between $\dot{\varepsilon}_{min}$ and $\dot{\varepsilon}_{max}$.

Using simple polymer kinetic theory, de Gennes arrived at an S-shaped curve for the steady-state polymer extension versus *De* (Fig. 1) and argued that in a narrow range of flow strengths near the coil-stretch transition, three molecular configurations were possible: a highly stretched "S" state, a compacted coiled "C" state, and a physically unstable "P" configuration.

The behavior of long-chain polymers near this phase transition has been a highly debated topic for several decades. Although the notion of conformation hysteresis was sup- neous mixture of two states with well-defined molar volumes. Saturated vapors can be subcooled into metastable states in the absence of liquid nucleation sites; similarly, saturated liquids may be superheated into the two-phase region specified by the dotted line. In both cases, the system is kinetically trapped in a local minimum of free energy. The presence of hysteresis is a signature of a first-order phase transition with an associated latent heat.

By analogy, polymers above a critical length $M_2$ can exist in two stable states of

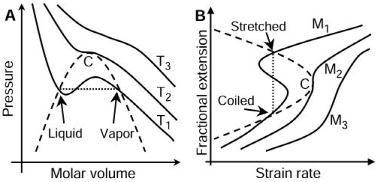



**Fig. 2.** (**A**) Classic first-order phase transition for vaporization or fusion of a pure substance. For temperatures $T$ less than a critical temperature $T_2$, vapor and liquid phases may coexist along an isotherm $T_1$ where the molar free energies of the liquid and vapor phases are equal. A cubic equation of state is merely an approximation to actual phase behavior (dashed line). (**B**) Coil-stretch phase transition for flexible polymers in extensional flows. For linear polymers with molecular weights $M_1$ greater than a critical molecular weight $M_2$, configuration hysteresis is possible in a range of *De*. Clearly, the steady-state extension will be a function of the deformation history of the polymer. The coil-stretch transition (given by the vertical dotted line) may be defined at the strain rate where the configurational free energies of the stretched and coiled states are equal for $M > M_2$.


Charles M. Schroeder, Hazen P. Babcock, Eric S. G. Shaqfeh, and Steven Chu. Observation of polymer conformation hysteresis in extensional flow. *Science*, 301(5639):1515–1519, 2003

# Example

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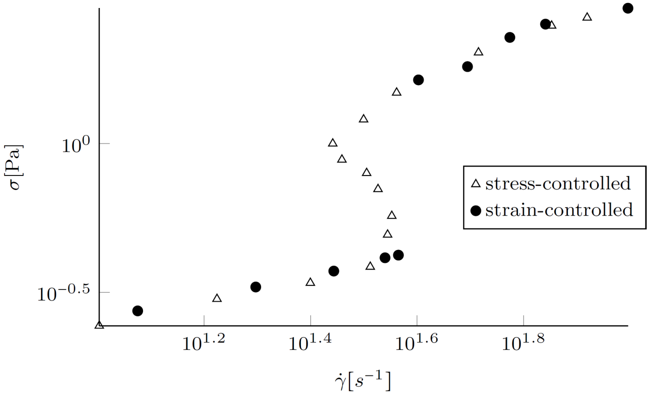


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would still not be present.) We also see that, contrary to the statement by Truesdell and Noll (ref. 11, p. 473), it is possible to distinguish some simple fluids from Reiner-Rivlin fluids in experiments in simple elongation, because we have here exhibited some simple fluids which do not always obey the usual rules for finite memory of the past, whereas the Reiner-Rivlin fluids respond only to the instantaneous kinematic field. Perhaps the most important aspect of the present investigation is that it serves as a cautionary tale. Despite the simplicity and deceptive innocence of the micro-

R. I. Tanner. Stresses in dilute-solutions of bead-nonlinear-spring macromolecules. 3. Friction coefficient varying with dumbbell extension. *Trans. Soc. Rheol.*, 19(4):557–582, 1975

# Conclusion

- Some experimental data that can not be interpreted using the standard models $\mathbb{T}_\delta = \mathfrak{f}(\mathbb{D})$.
- Implicit constitutive relations $\mathfrak{f}(\mathbb{T}_\delta, \mathbb{D}) = 0$ provide a tool how to develop constitutive models.
- Building a model using one-dimensional data is always a problem. (Rethinking of experimental procedures is necessary.)
- Construction of a specific constitutive relation with normal stress differences effect.
- Construction of a three dimensional fully implicit tensorial constitutive relations (thermodynamic background).

$$\begin{aligned}
\operatorname{div} \mathbf{v} &= 0 \\
\rho \frac{\mathrm{d}\mathbf{v}}{\mathrm{d}t} &= \operatorname{div} \mathbb{T} + \rho \mathbf{b} \\
\mathbb{T} &= \mathbb{T}^\top \\
\mathfrak{g}(\mathbb{T}_\delta, \mathbb{D}) &= \mathbb{0}
\end{aligned}$$