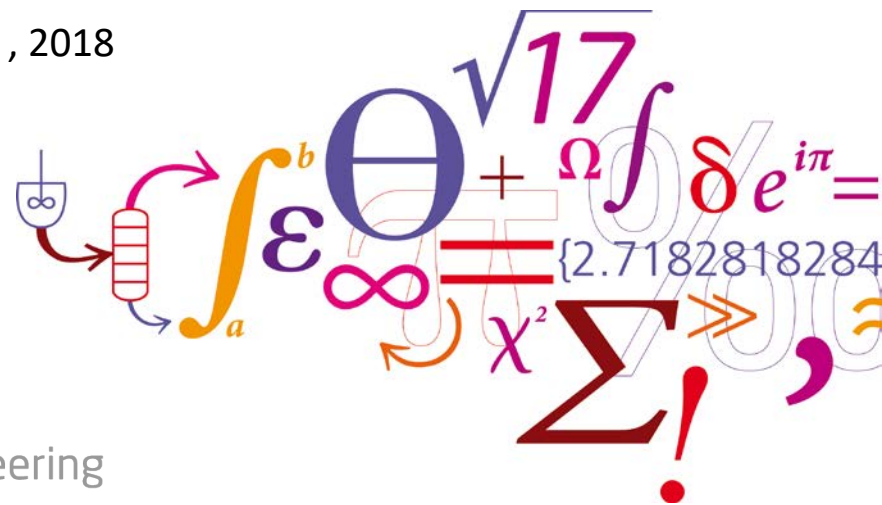


Lecture 7 and 8: Extensional Rheology

Ole Hassager and Qian Huang

Nanjing, August , 2018



A Chinese Nobel laureate:

Experimental science is the foundation of all natural science.

National Museum of China, Beijing



Do you know this feeling?



DTU Chemical Engineering

Department of Chemical and Biochemical Engineering

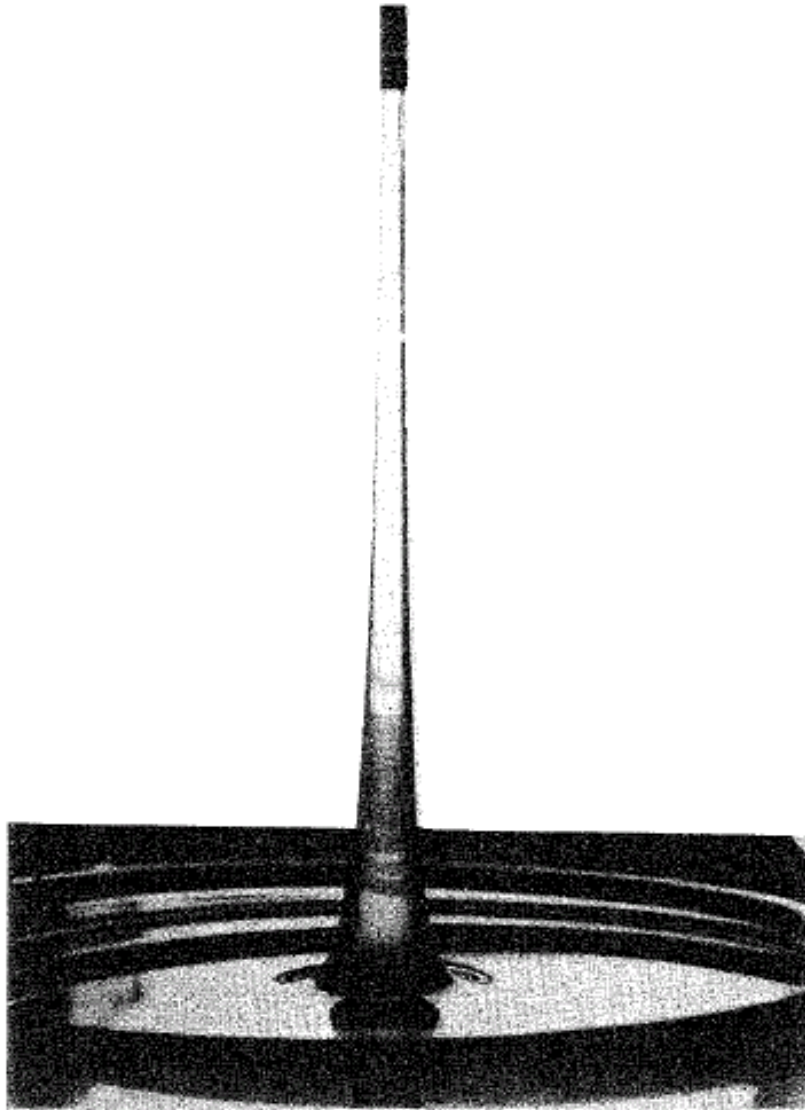


FIGURE 2.5-3. Fluid column in tubeless siphon experiment with a high molecular weight hydrocarbon polymer, AM-1 in JP-8 aviation fuel. [Reproduced from S. T. J. Peng and R. F. Landel, *J. Appl. Phys.* **47**, 4255 (1976)].

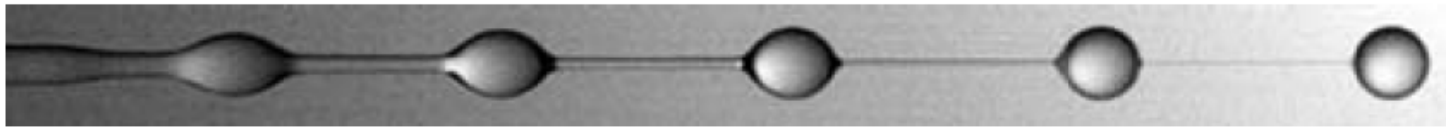


FIGURE 2. High-speed video-image of a jet of dilute (0.01 wt%) aqueous polyacrylamide solution (surface tension $\gamma = 62 \text{ mN m}^{-1}$) undergoing capillary thinning. The sharp-edged jet orifice is at the left of the image (radius $R_0 = 0.30 \text{ mm}$) and the free jet velocity is 30 cm s^{-1} . The polymeric contribution to the viscosity is $\eta_p = 0.0119 \text{ Pa s}$, and the polymer time scale is found to be $\lambda = 0.012 \text{ s}$. This corresponds to a Deborah number of $De = 18.2$.

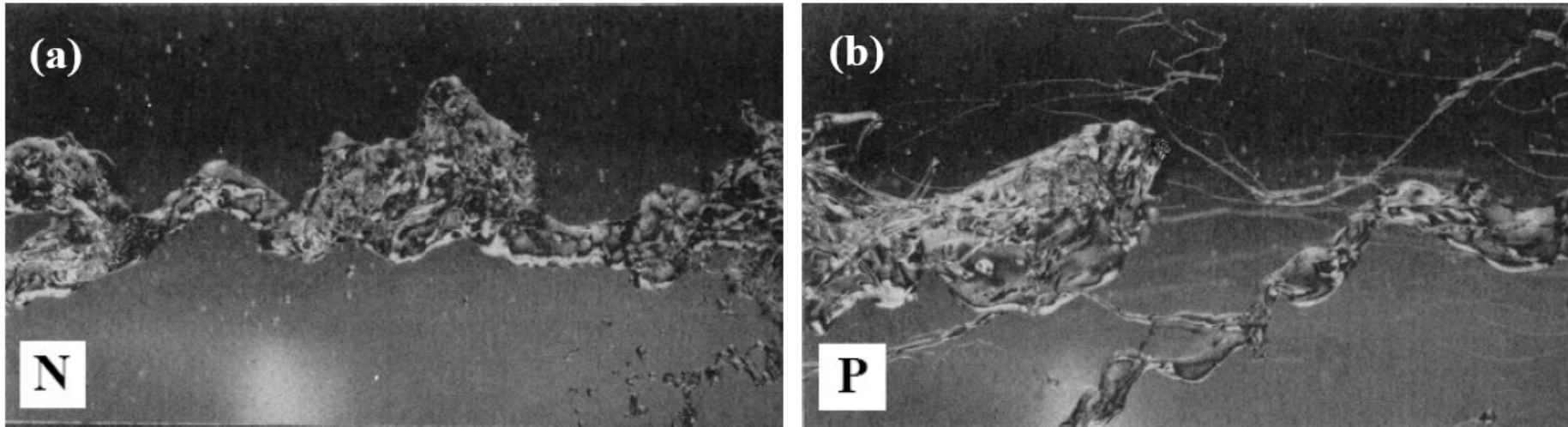
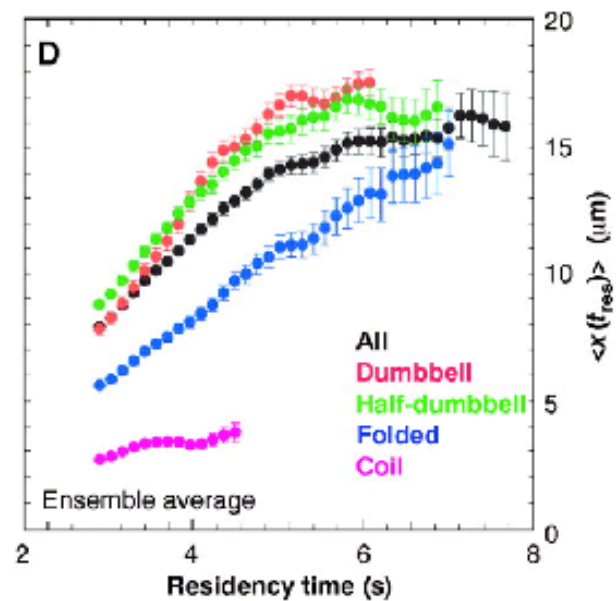
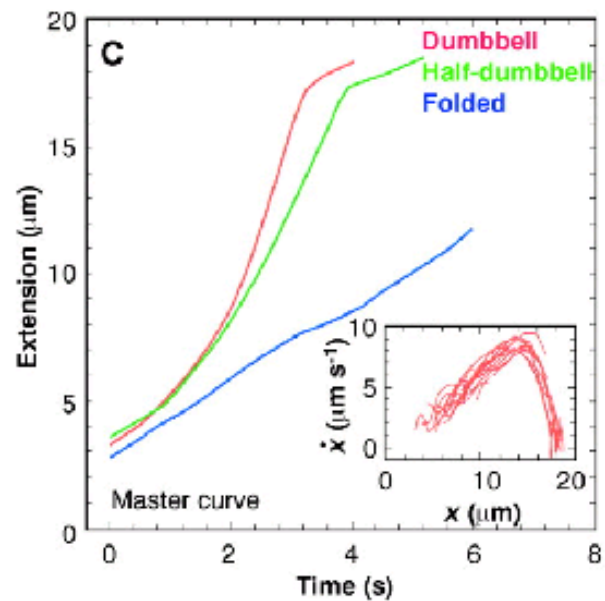
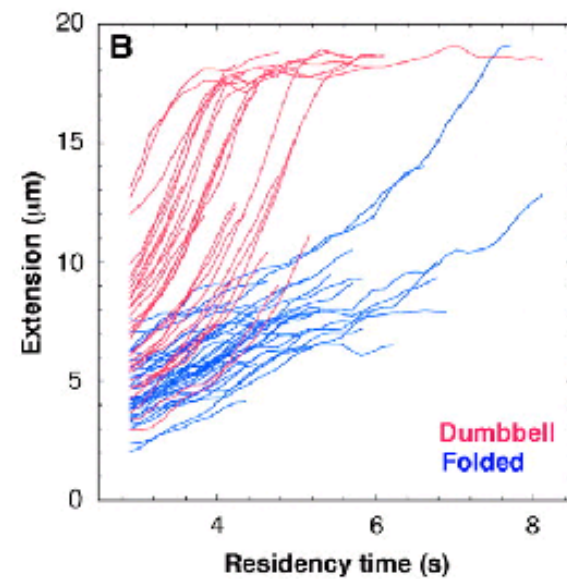
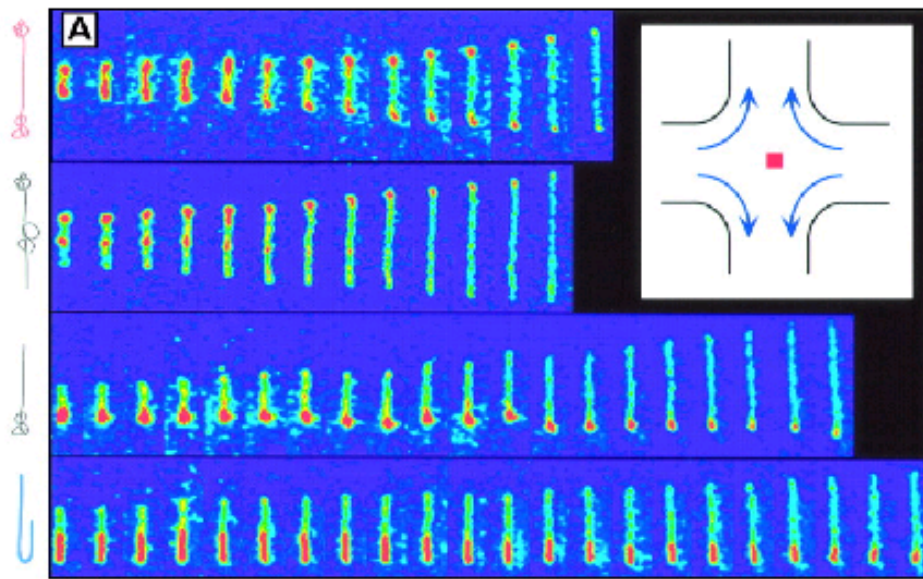
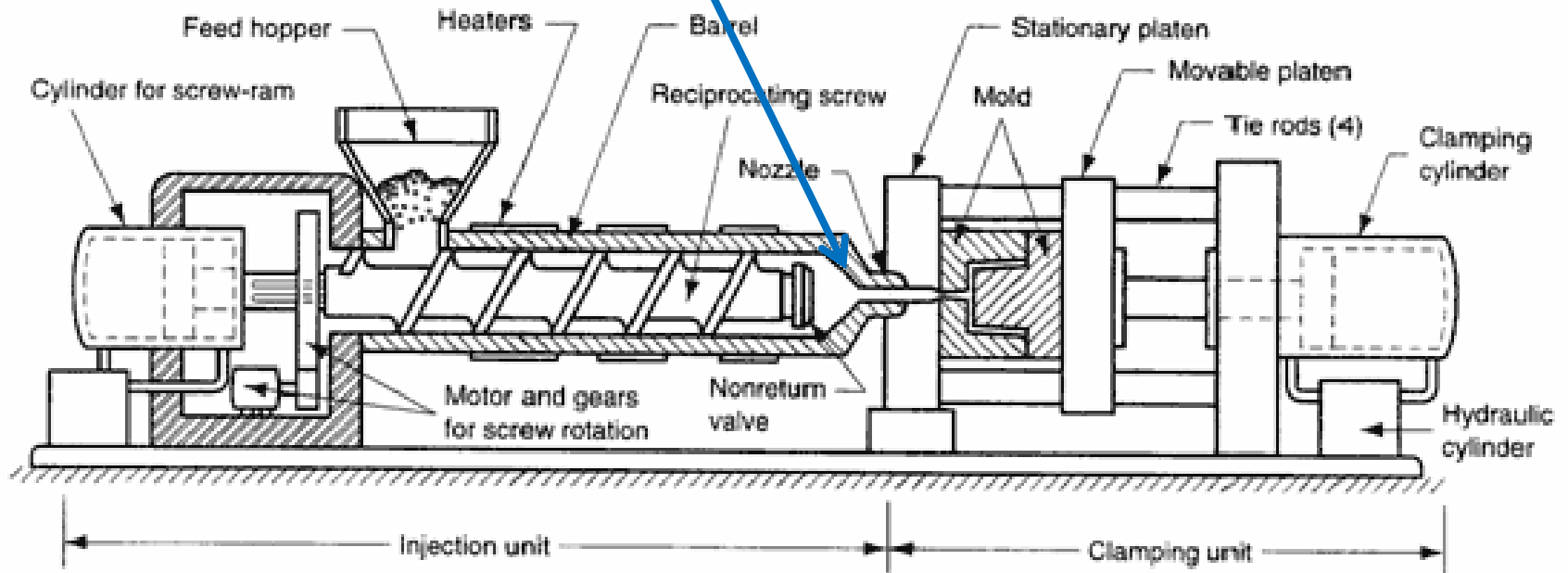


Fig. 8 (a) A turbulent jet of pure water, N for Newtonian, forming droplets. (b) A turbulent jet of water with the addition of only 200 parts per million of the macromolecule polyethylene-oxide forming filaments, from Ref. [80].



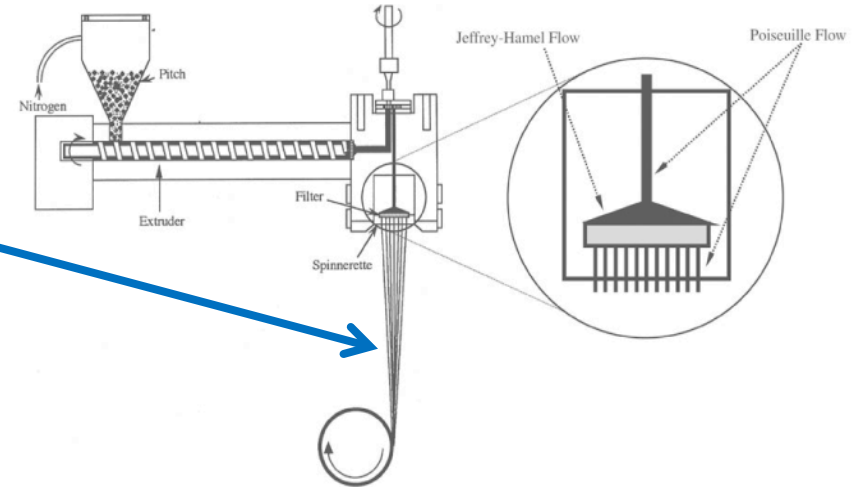
Why study extensional rheology?

Injection Moulding:

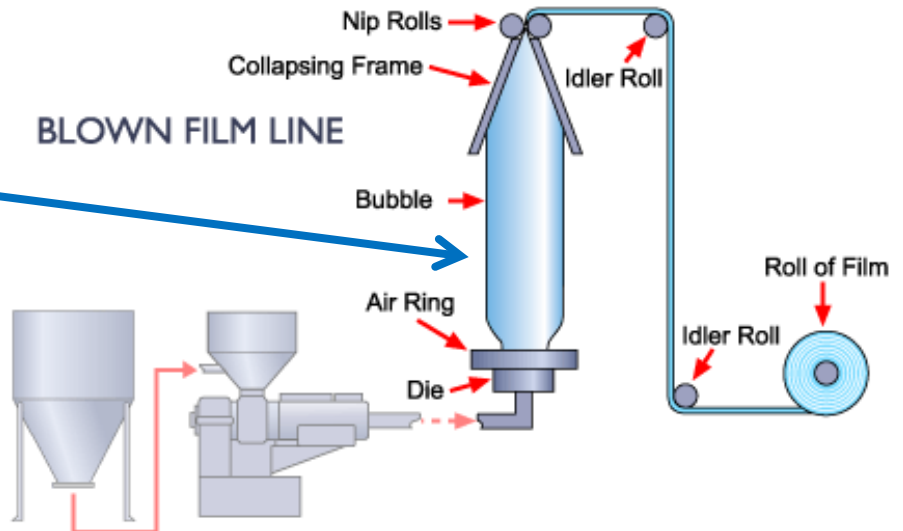


Other Polymer processing operations with Extension:

Fibre spinning:
(draw down ratio)



Film blowing:



"The World's Strongest Fibre™"



Dyneema® is an UHMwPE (Ultra High Molecular weight Polyethylene) fibre developed by DSM in the Netherlands

High Strength: On a weight for weight basis, Dyneema® is 15 times stronger than steel wire

Water resistant: Dyneema® is hydrophobic and does not absorb water.

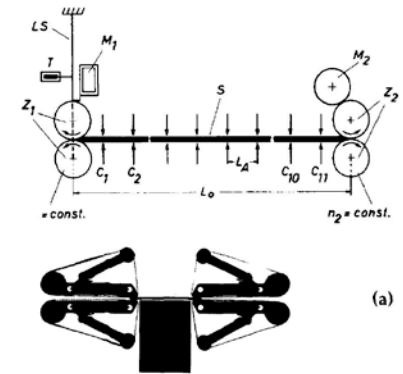
Chemical resistance: Dyneema® is chemically inert.

Historical: Extensional Rheometer Designs

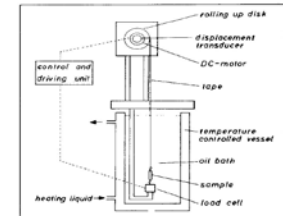
Meissner (1969), (1971), rotary clamp

$$\epsilon_{max} \approx 7$$

Meissner and Hostettler (1994), conveyor belt



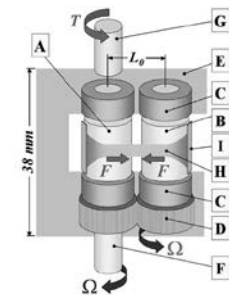
Münstedt and Laun (1979)



Sentmanat (2004): SER

$$\epsilon_{max} \approx 3.5$$

TA: EVF



$$\epsilon_N = \log(L/L_0)$$

All materials:

$$\text{Mass: } \nabla \cdot v = 0$$

$$\text{Force: } \rho \left(\frac{\partial}{\partial t} + v \cdot \nabla \right) v = -\nabla p + \nabla \cdot \sigma$$

Newtonian liquids:

$$\text{True Stress: } \sigma(t) = \mu \dot{\gamma}(t)$$

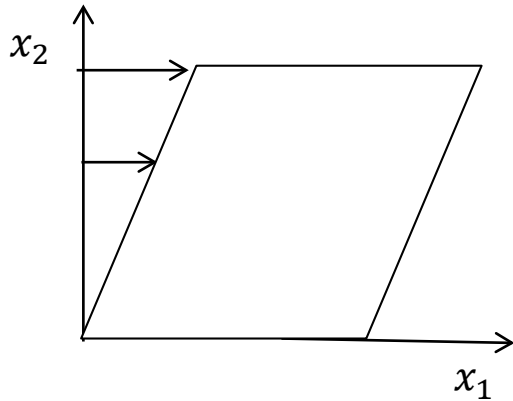
$$\text{Viscosity: } \mu$$

$$\text{Rate-of-deformation tensor: } \dot{\gamma} = [\nabla v + (\nabla v)^\dagger]$$

Polymeric liquids:

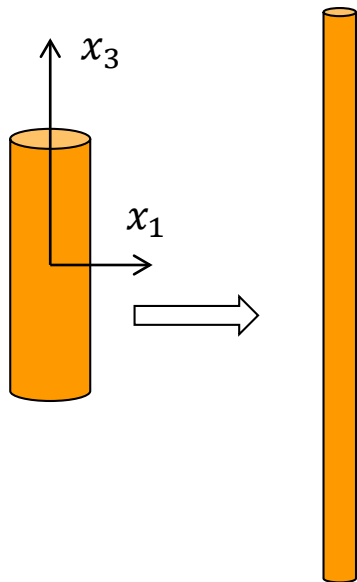
$$\text{True Stress: } \sigma(t) = \sigma\{v(t'), t' \in [-\infty; t]\}$$

Two basic frequently studied classes of flow:



Shear Flow:

Shear rate κ [1/s]



Uniaxial Extensional flow:
 $v_1 = -\dot{\epsilon}x_1/2$ $\sigma_{33} - \sigma_{11} = \eta_E(\dot{\epsilon})\dot{\epsilon}$
 $v_2 = -\dot{\epsilon}x_2/2$ $\dot{\epsilon} \geq 0$
 $v_3 = \dot{\epsilon}x_3$

Hencky strain rate $\dot{\epsilon}$ [1/s]

Problem: In general no connection between rheometric functions in shear and extension

Except in small deformations:

$$\sigma_{ij}(t) = \int_{t'=-\infty}^t G(t-t') \dot{\gamma}_{ij}(t') dt'$$

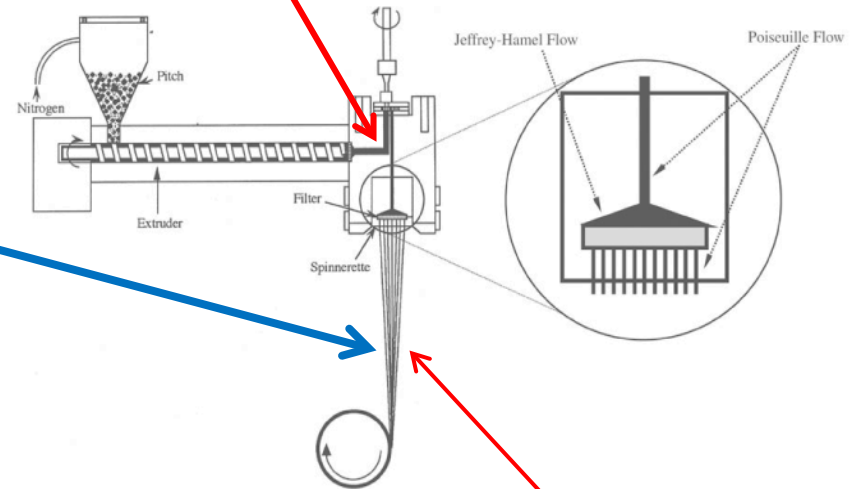
$G(t-t')$: Linear Viscoelastic Relaxation modulus

But polymer processing operations does not
involve small deformations!

Mainly shear flow

Fibre spinning:
(draw down ratio)

Find pressure, velocity, polymer conformation, crystallinity...



Mainly extensional flow

Rheometry: Determine rheometric functions $\eta_E(\dot{\epsilon})...$

If we can control flow locally and measure stress locally the flow field needs not be homogeneous globally.

Homogeneous Extensional Flow

For incompressible liquids, the local Hencky strain of a filament may be calculated from the diameter:

Hencky Strain:

$$\epsilon = \ln \lambda = \ln \frac{L(t)}{L_0}$$

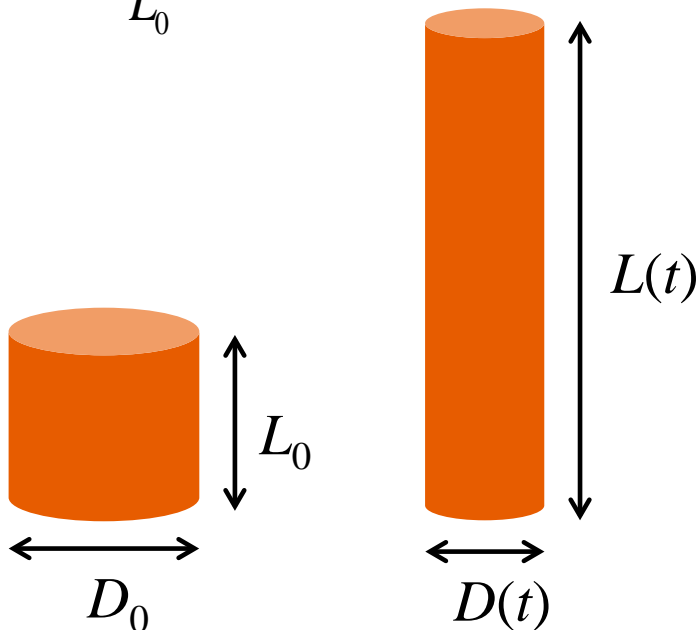
$$\epsilon = -2 \ln \frac{D(t)}{D_0}$$

Hencky Strain Rate:

$$\dot{\epsilon} = \frac{d\epsilon}{dt}$$

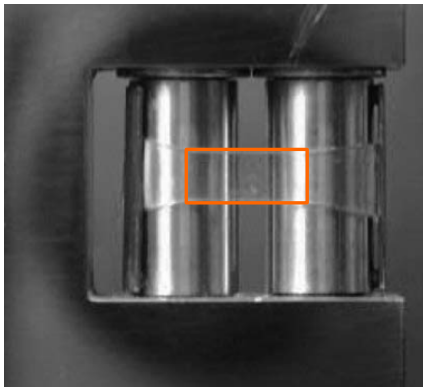
Stretch Ratio:

$$\lambda = \frac{L(t)}{L_0}$$



Commercially available extensional rheometers do not provide globally homogeneous flow :

**FSR locally homogeneous
in mid-plane**

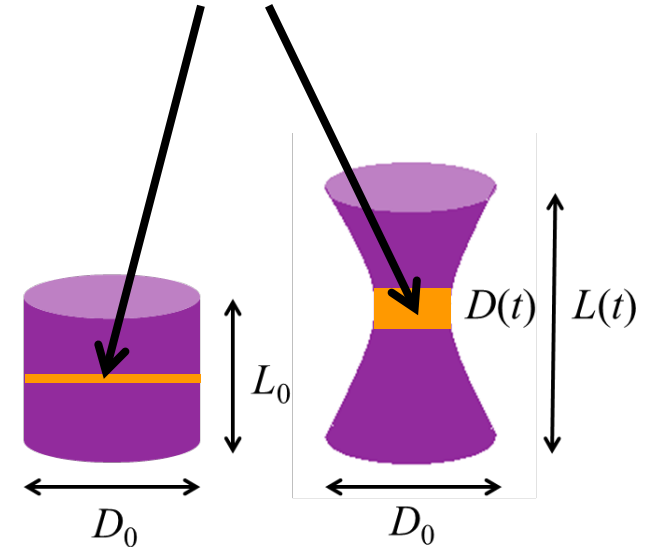


Sentmanat Extensional Rheometer (SER)

Extensional Viscosity Fixture (EVF)



Filament Stretching Rheometer (FSR)

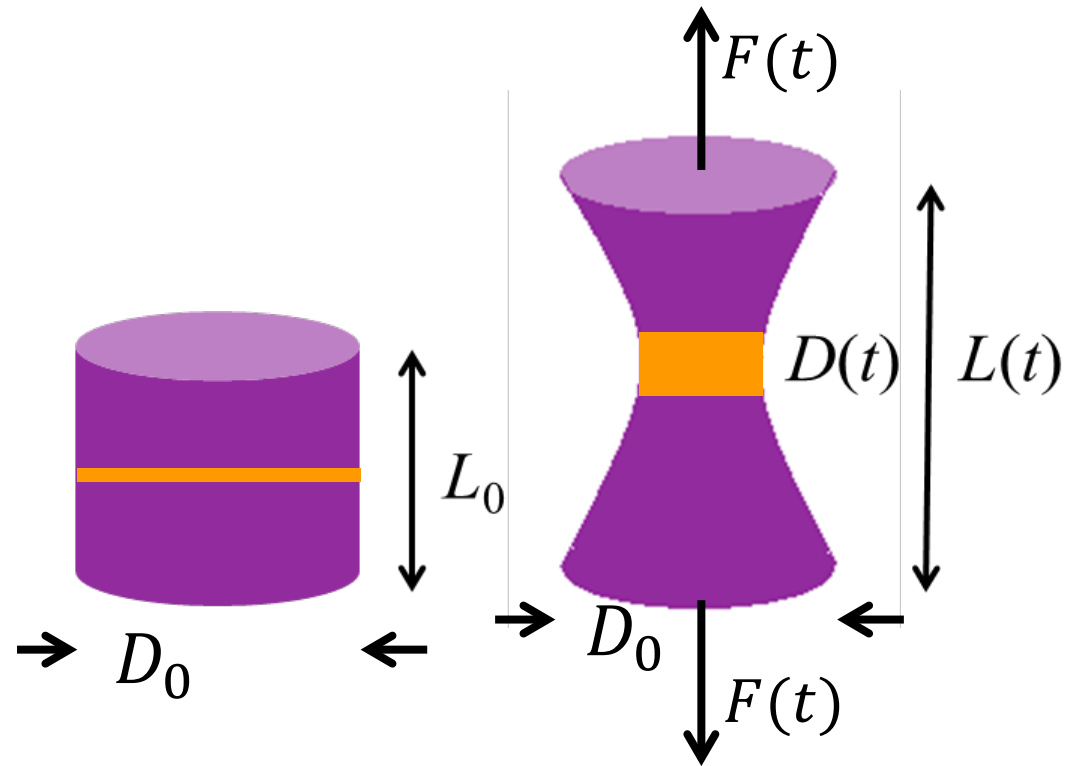


$$\varepsilon = -2 \ln \frac{D(t)}{D_0}$$

$$\varepsilon_z = \ln \frac{L(t)}{L_0}$$

Determination of stress:

Filament Stretching Rheometer (FSR)

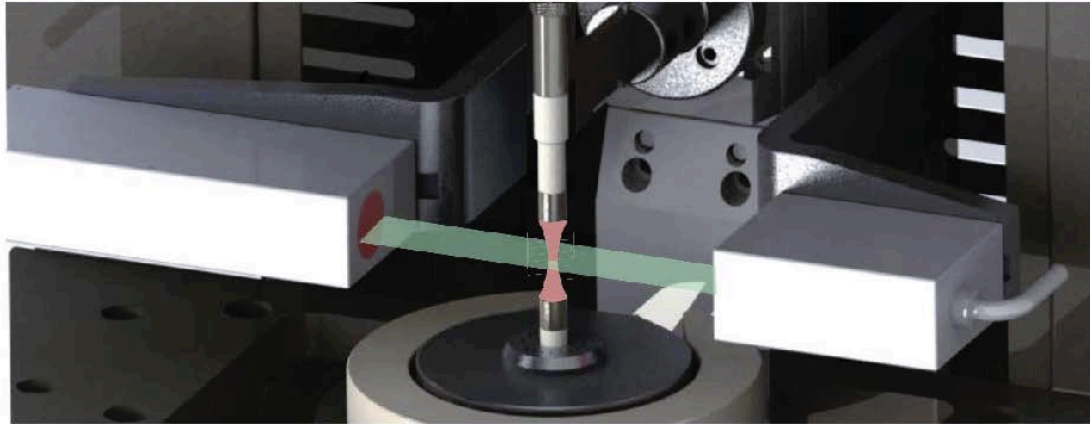


Engineering stress at t: $\sigma^E = F(t)/(\pi(\frac{D_0}{2})^2)$

(True) stress in yellow material at t: $\sigma = F(t)/(\pi(\frac{D(t)}{2})^2)$

Versatile Accurate Deformation Extensional Rheometer (VADER) provides:

- Local diameter measurement by laser micrometer
- Fast feedback loop for controlled deformation

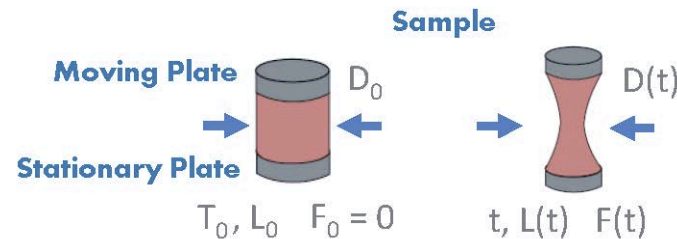


Uniaxial Hencky Strain

$$\epsilon = 2 \ln(D_0/D(t))$$

True Stress

$$\sigma = \frac{\text{Force}}{\text{Area}} = \frac{4 F(t)}{\pi D(t)^2}$$

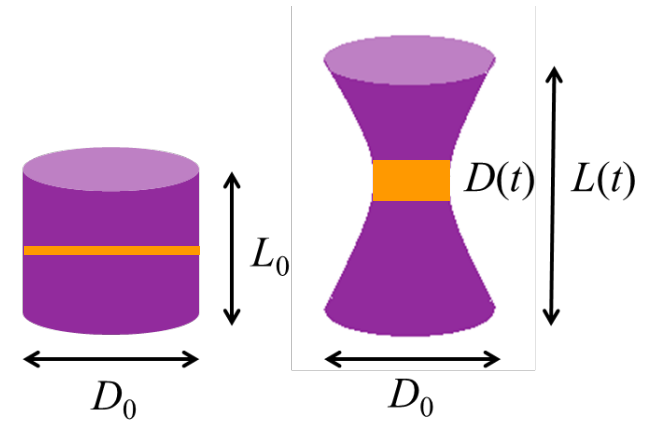
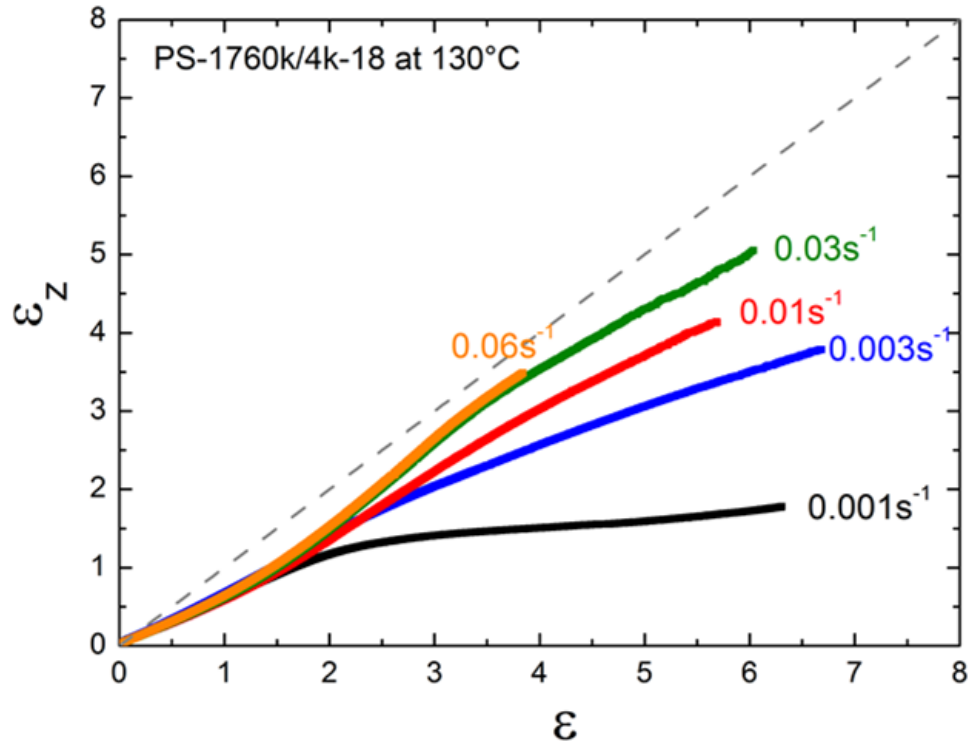


Force measured at bottom plate. (correction for gravity may be needed)

Available protocols:

- Constant rate (start-up)
- True Stress Relaxation ($\dot{\epsilon} = 0$)
- Constant stress (creep)
- Small Amplitude Oscillation

Filament Stretching



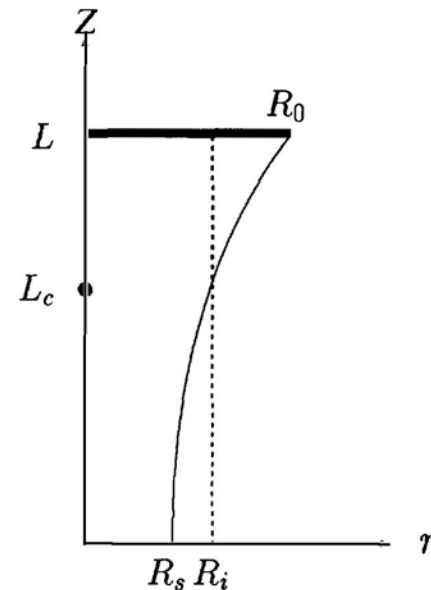
$$\epsilon = -2 \ln \frac{D(t)}{D_0}$$

$$\epsilon_z = \ln \frac{L(t)}{L_0}$$

What happens at constant stretch rate no feed-back control?

$$\sigma(t) = \int_{-\infty}^t M(t - t') \phi(I_1, I_2) B(t, t') dt'$$

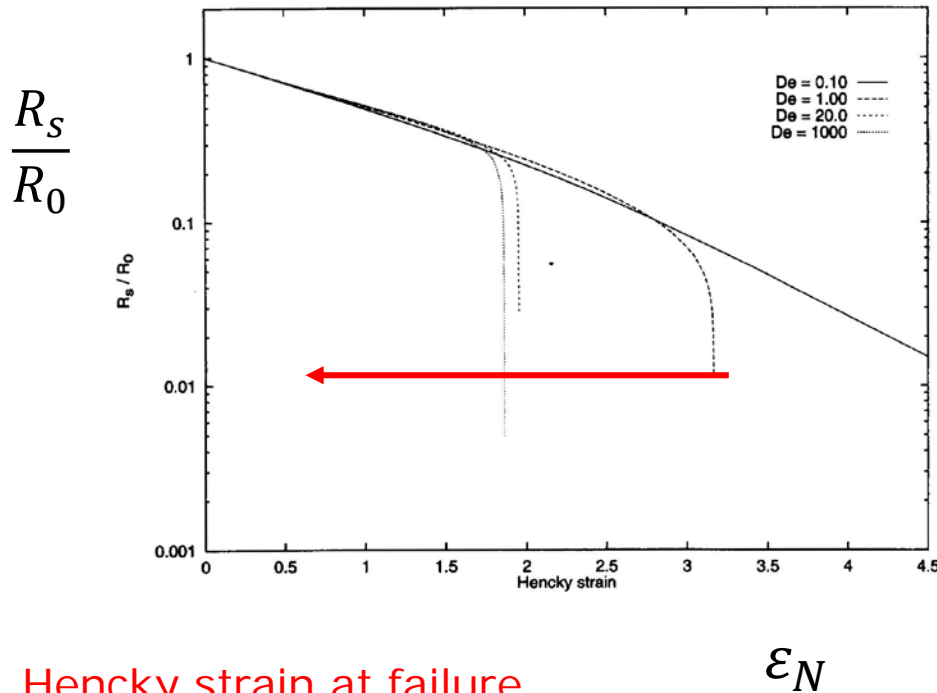
$$\phi(I_1, I_2) = \frac{\alpha}{(\alpha - 3) + \beta i_1 + (1 - \beta) I_2}$$



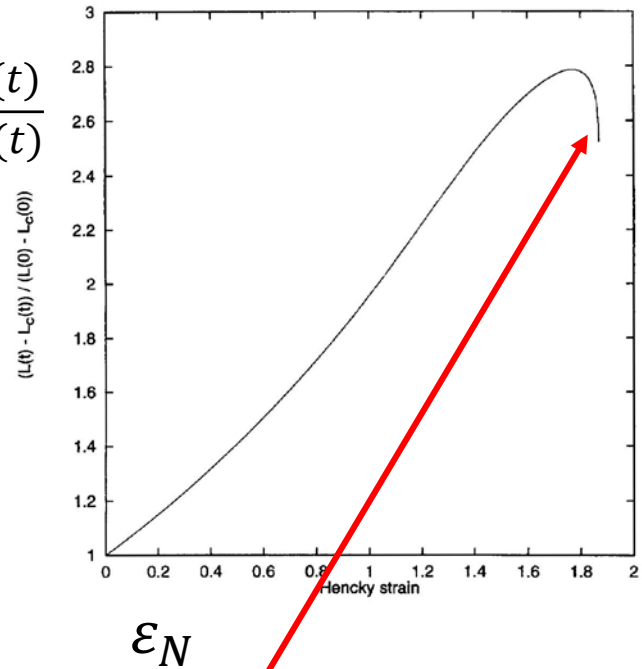
Hassager, Kolte and Renardy

J. Non-Newtonian Fluid Mech., 76, 137-151 (1998)

What happens at constant stretch rate no feed-back control?



$$\frac{L(t) - L_c(t)}{L(0) - L_c(0)}$$



Hencky strain at failure decreases with increasing rate

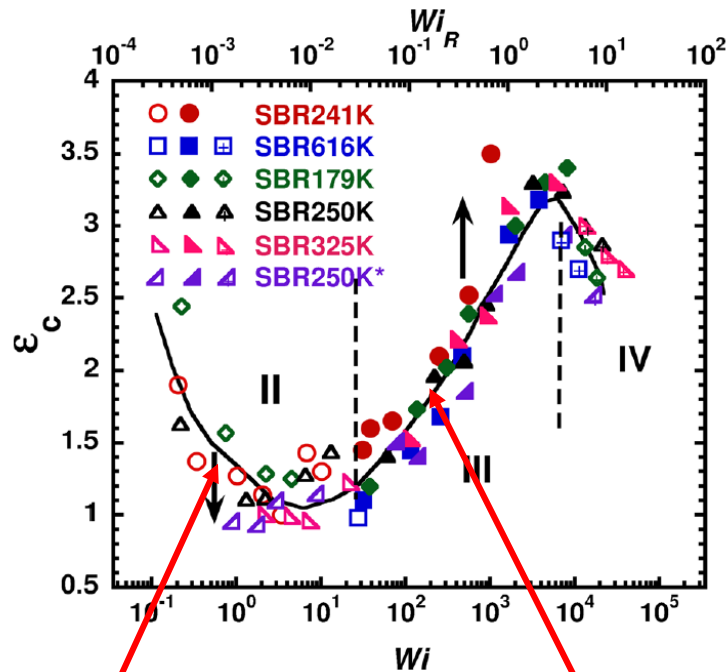
Material near plates recoils

Hassager, Kolte and Renardy

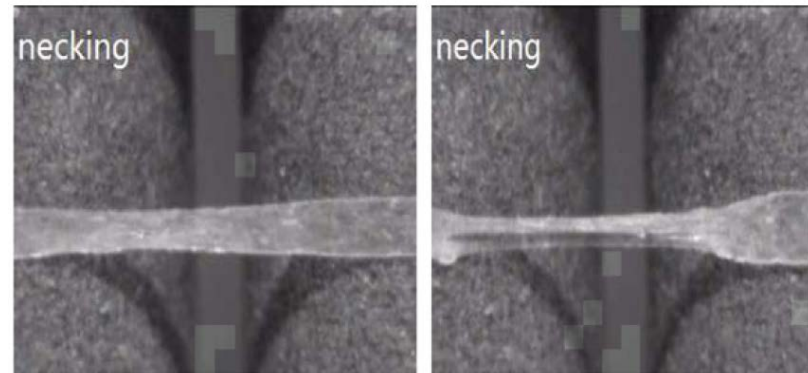
J. Non-Newtonian Fluid Mech., 76, 137-151 (1998)

No feedback loop: Viscoelastic Instability

Both FSR and SER:



Zhu et al., *J. Rheol.* (2013) 57: 223



Zhu et al., *J. Rheol.* (2013) 57: 223

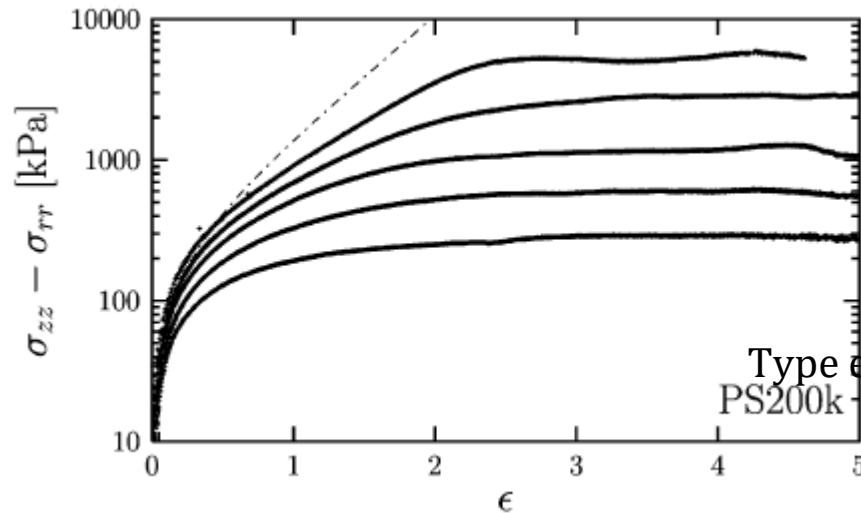
Simulations by e.g.
Hassager, Kolte Renardy

Molecular
stretching.

See also Malkin et al., *Prog. Poly. Sci.*
(2014) 39: 959-978

With feed-back control: Steady-state Extensional Flow

PS melts and solutions



Bach et al., Macromolecules (2003) 34: 5174-6179

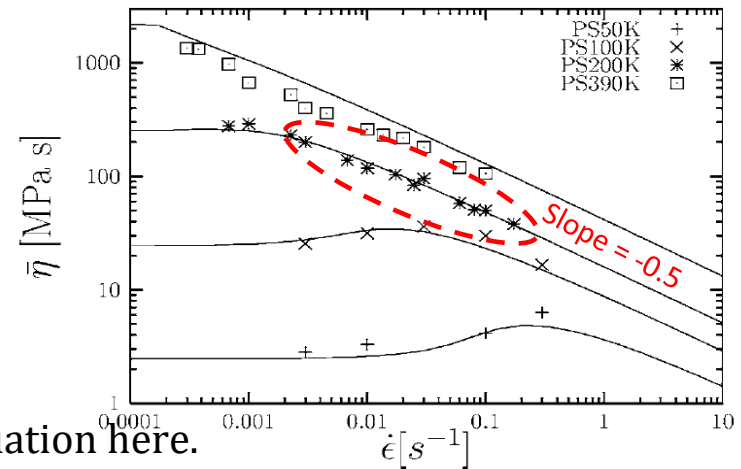
Rolie-Poly and Doi-Edwards without stretch:

$$\eta_E \sim \dot{\epsilon}^{-1}$$

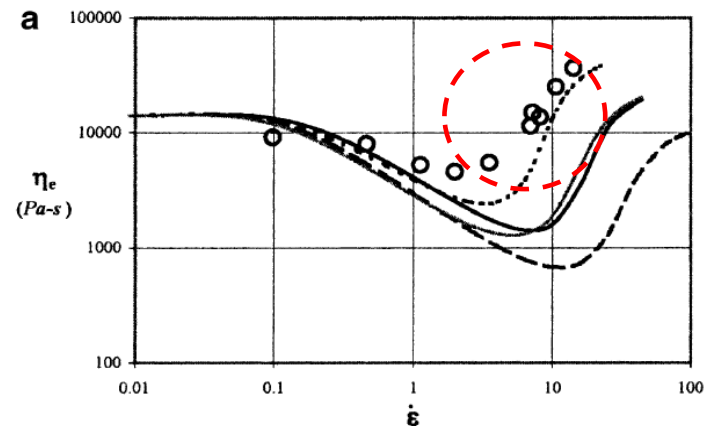
UCM and Gaussian dumbbells:

$$\eta_E \rightarrow \infty \text{ for } \dot{\epsilon} \rightarrow \infty$$

Wiest model: $\eta_E \sim \dot{\epsilon}^{-1/2}$ for $\dot{\epsilon} \rightarrow \infty$



Nielsen et al., J. Rheol. (2006) 50: 453-476



Bhattacharjee et al., Macromolecules (2002) 35: 10131-10148

- Elongational viscosity of model linear polymers
- Elongational viscosity at processing (PE-A and PE-B)
- Elongational viscosity of model branched polymers
- Stress maximum in branched polymers (creep)
- Stress maximum in branched polymers (SAXS)
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- True stress relaxation (SANS)
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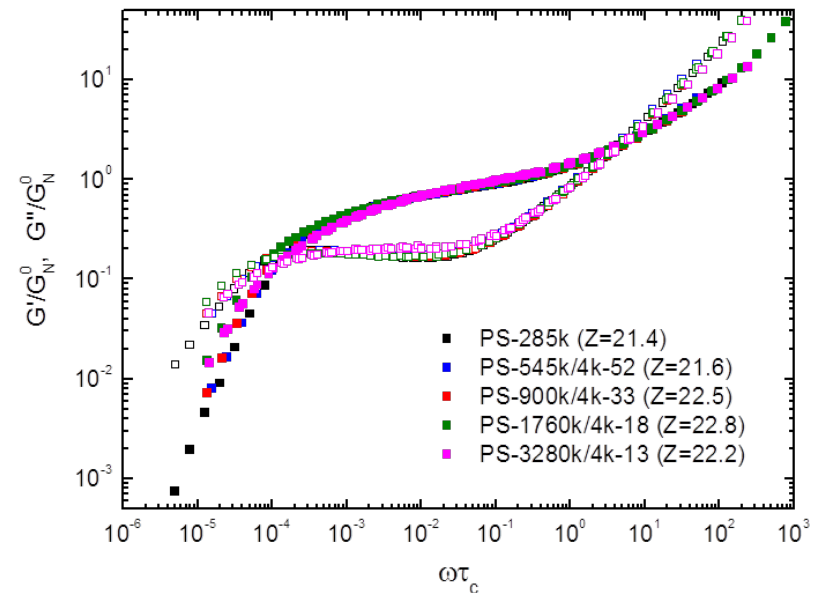
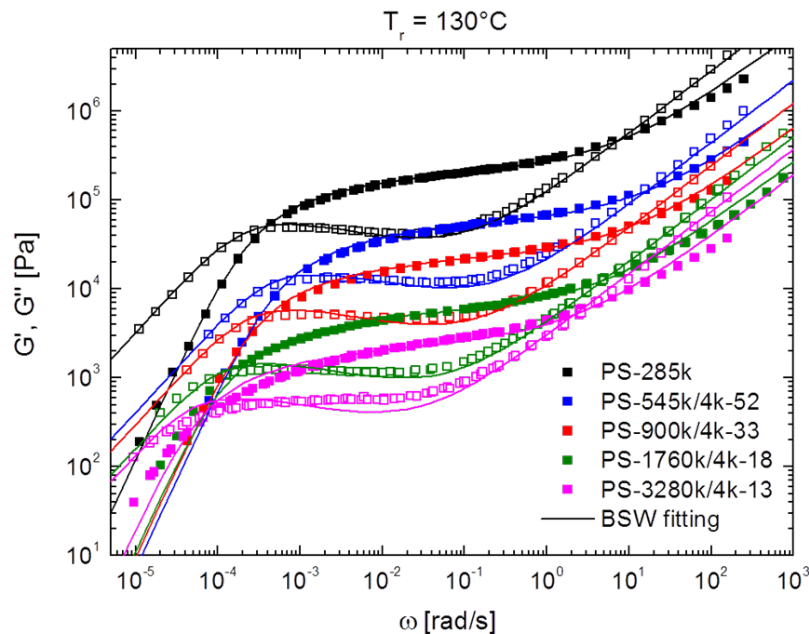
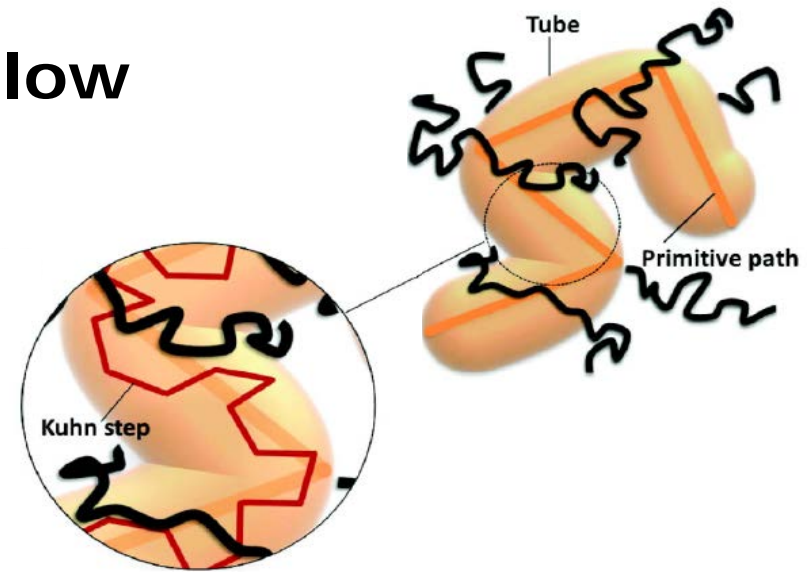
Steady-state Extensional Flow

Tube model parameters:

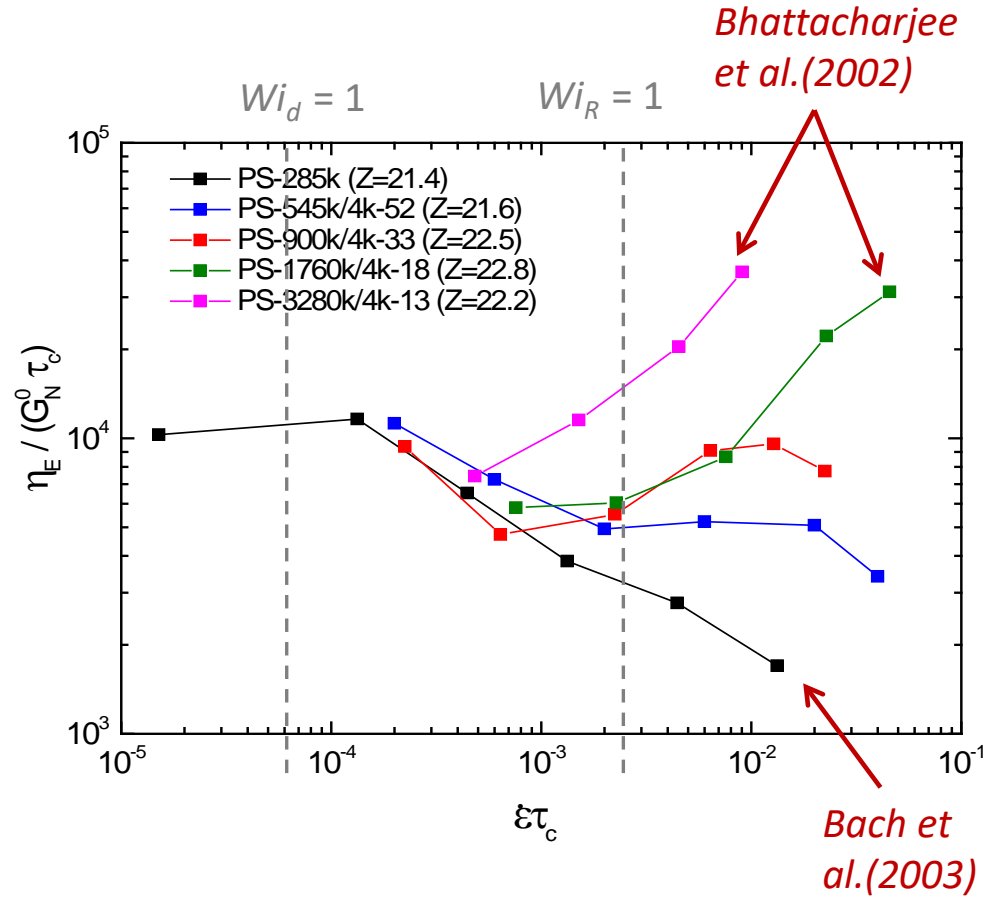
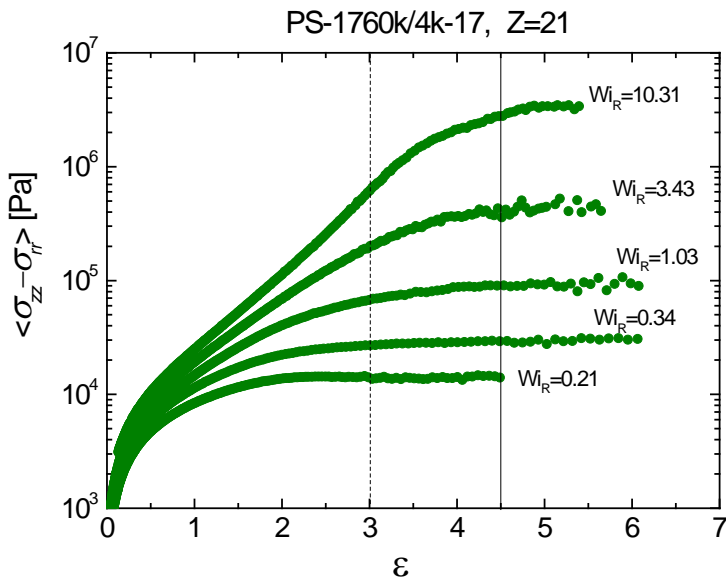
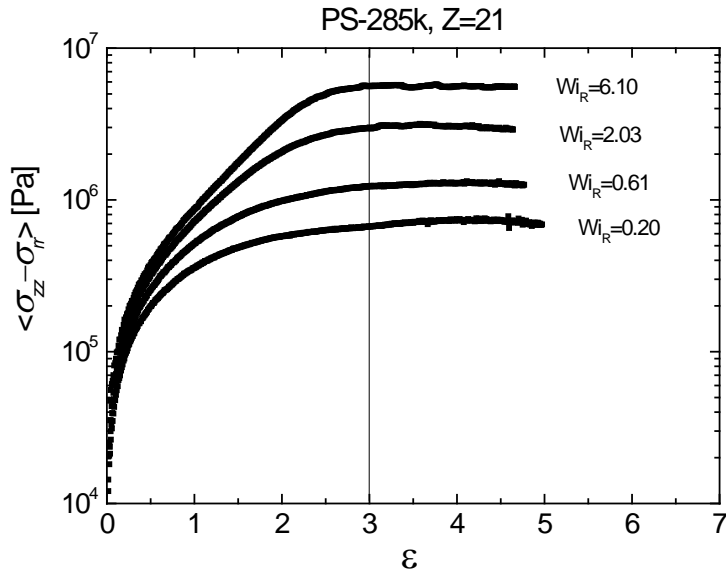
Number of entanglements per chain: Z

Number of Kuhn steps per entanglement: N_e

Entanglement Relaxation time: τ_e



Steady-state Extensional Flow

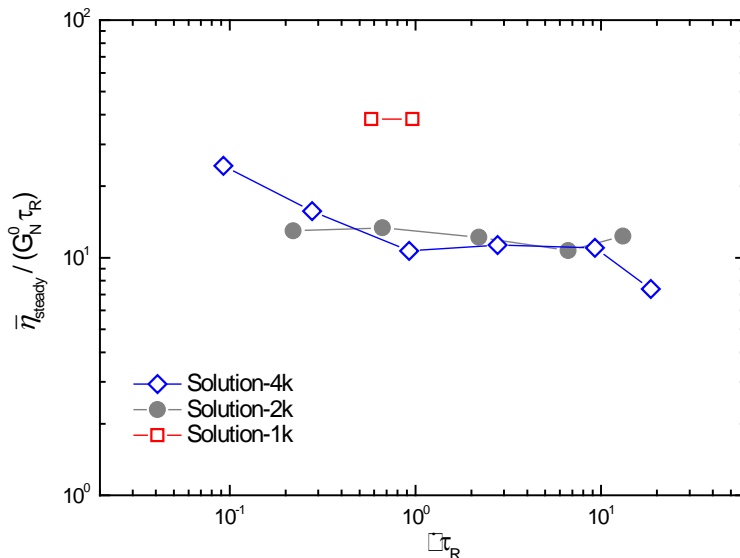
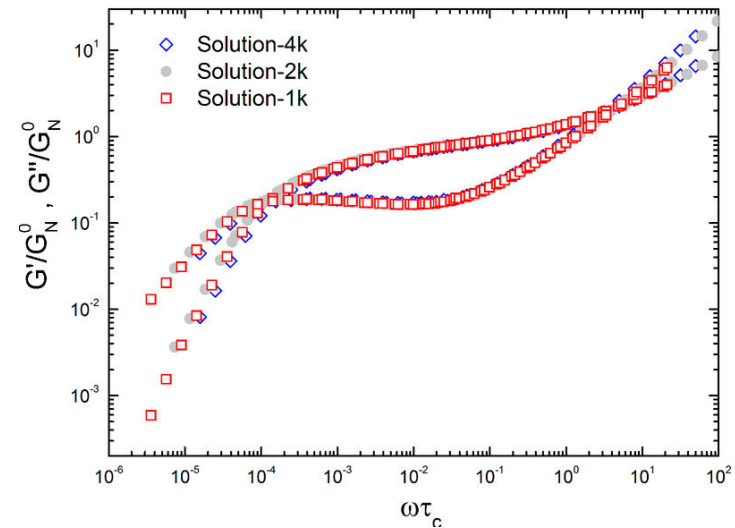
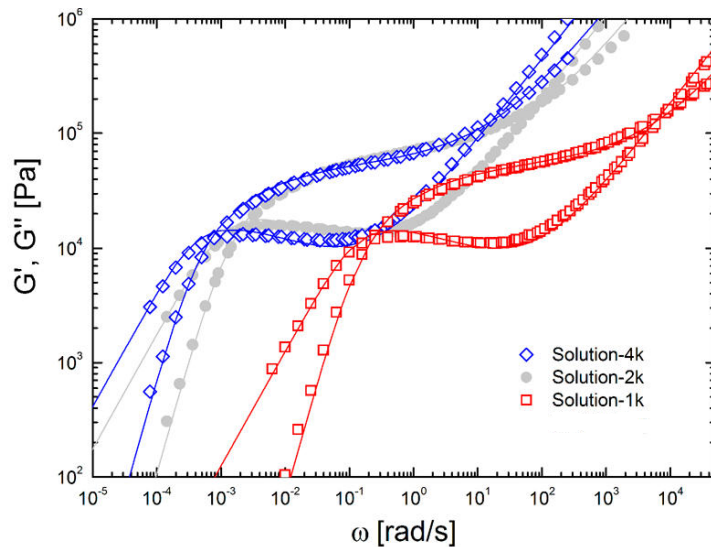


Number of entanglements per chain: Z

Entanglement Relaxation time: τ_e

Number of Kuhn steps per entanglement: N_e

Steady-state Extensional Flow



Number of entanglements per chain: Z

Entanglement Relaxation time: τ_e

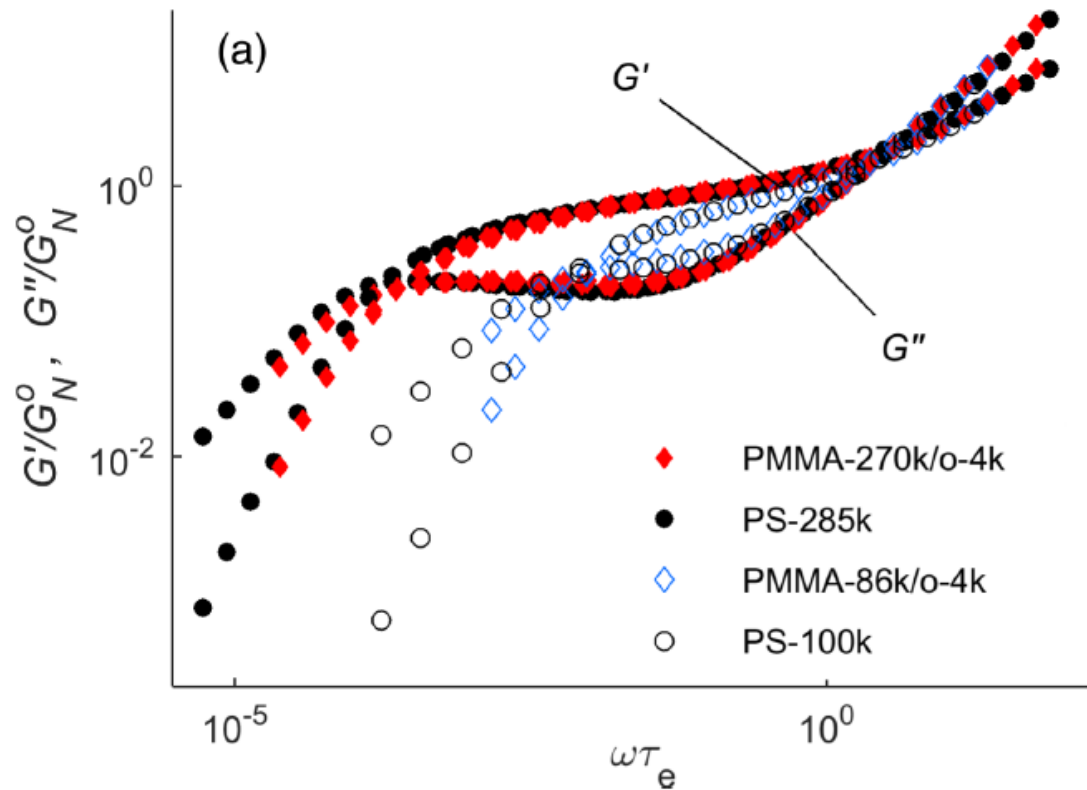
Number of Kuhn steps per entanglement: N_e

Ianniruberto et al. (2012) & Yaoita et al. (2012):

Reduction of monomeric friction

Steady-state Extensional Flow

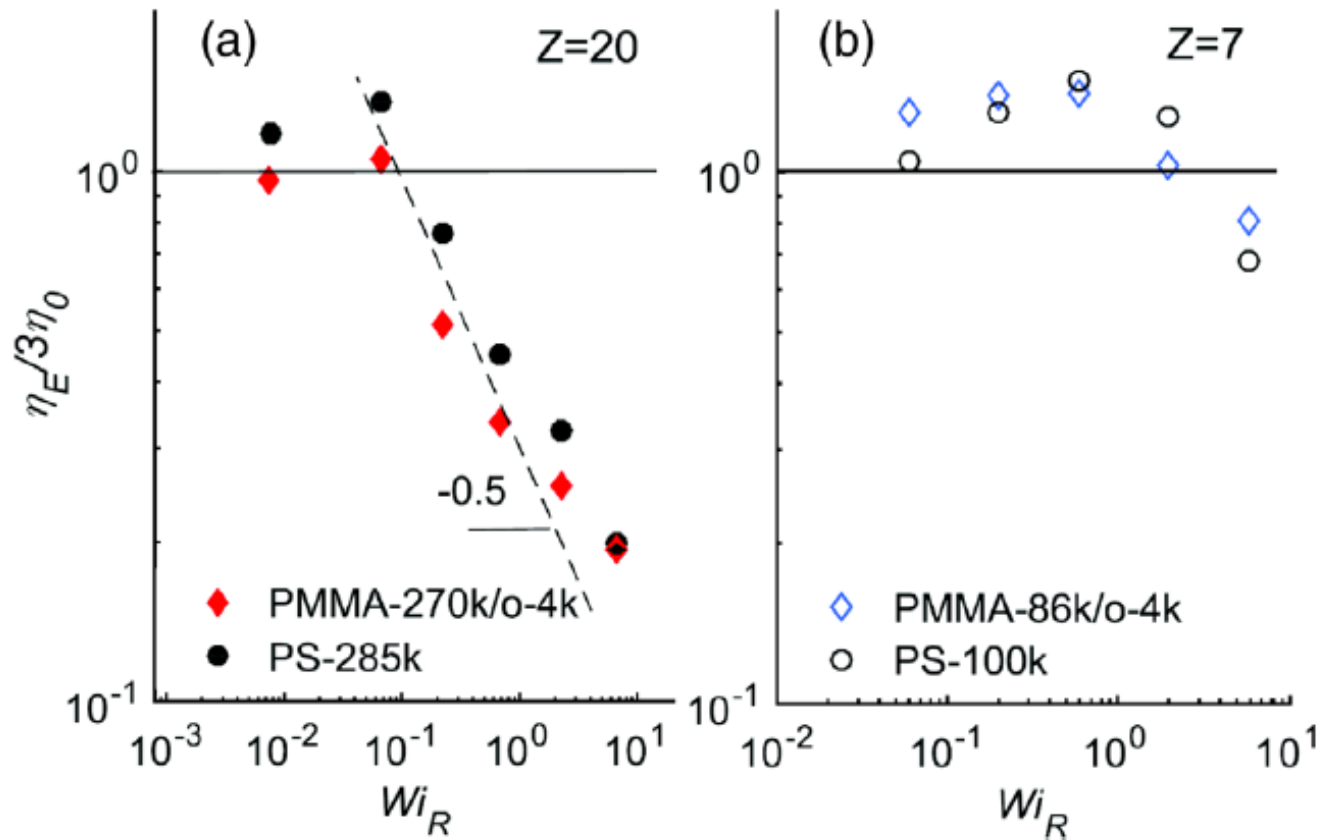
PS melts & PMMA solutions



Z
 N_e
 τ_e

Steady-state Extensional Flow

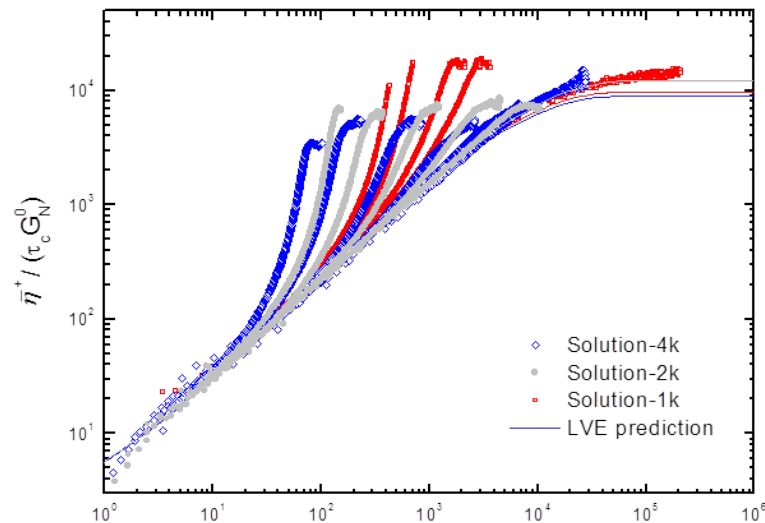
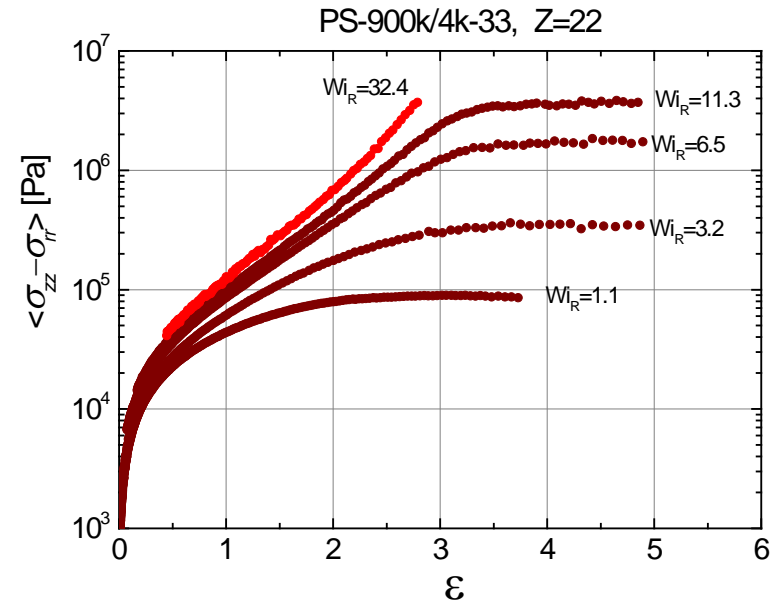
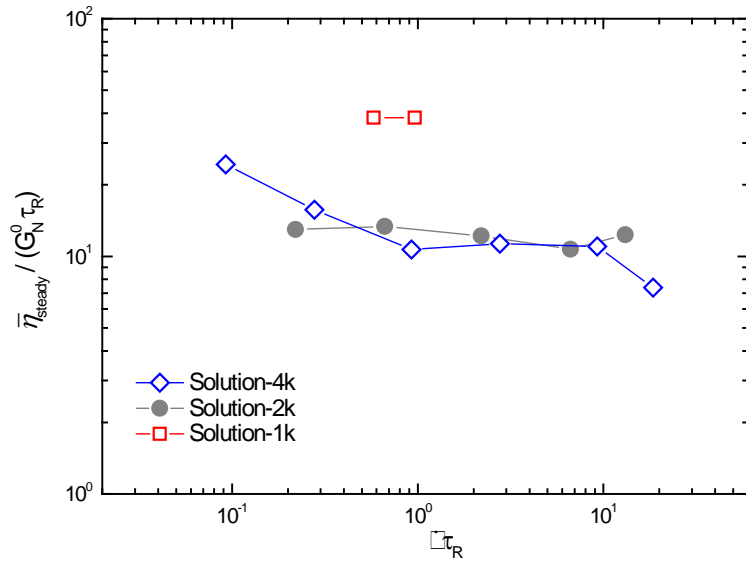
PS melts & PMMA solutions



Z
 N_e
 τ_e

Similar
monomeric
friction

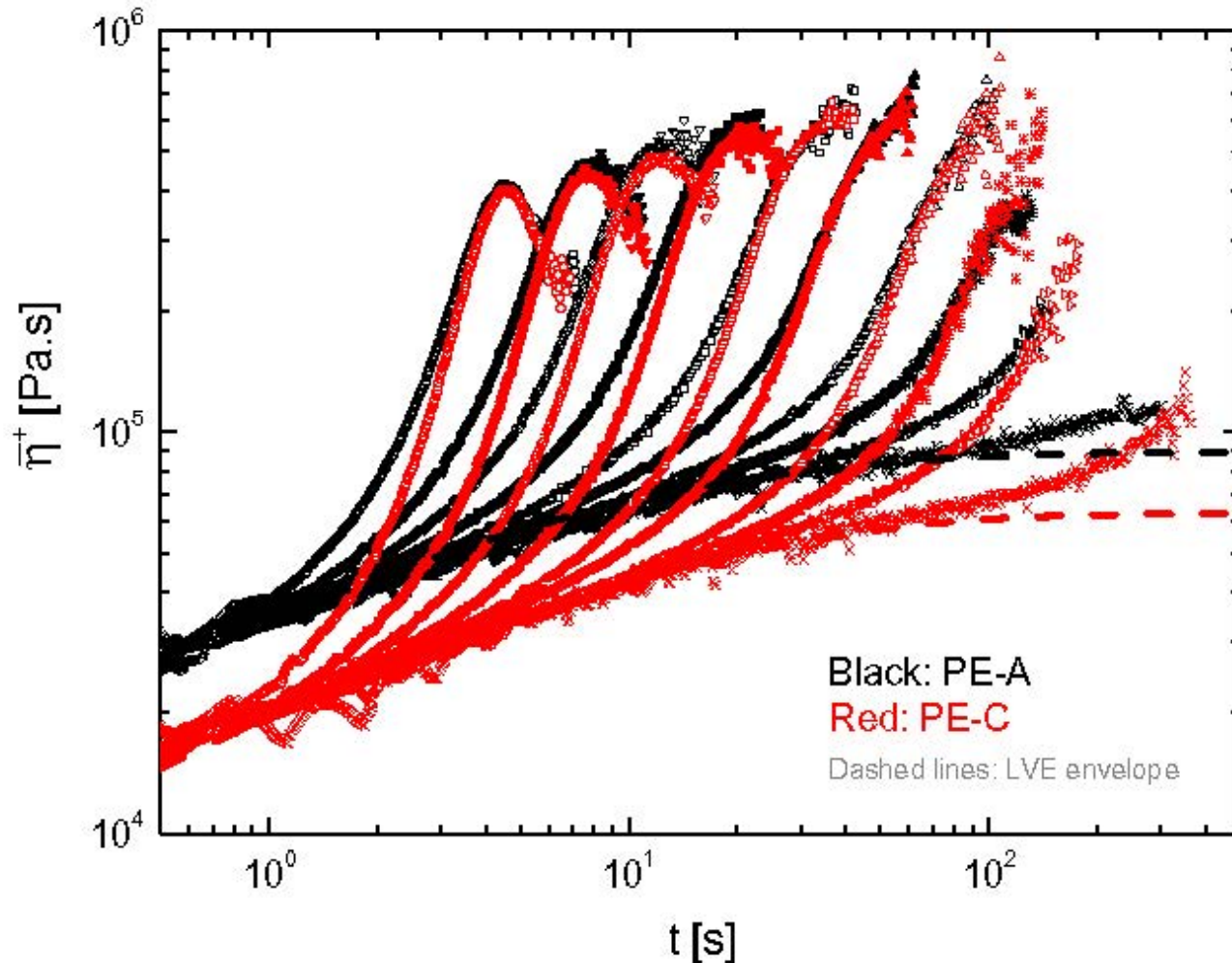
Steady-state \rightarrow Fracture



Fracture occurs on a fast time scale to make direct visual observations impossible

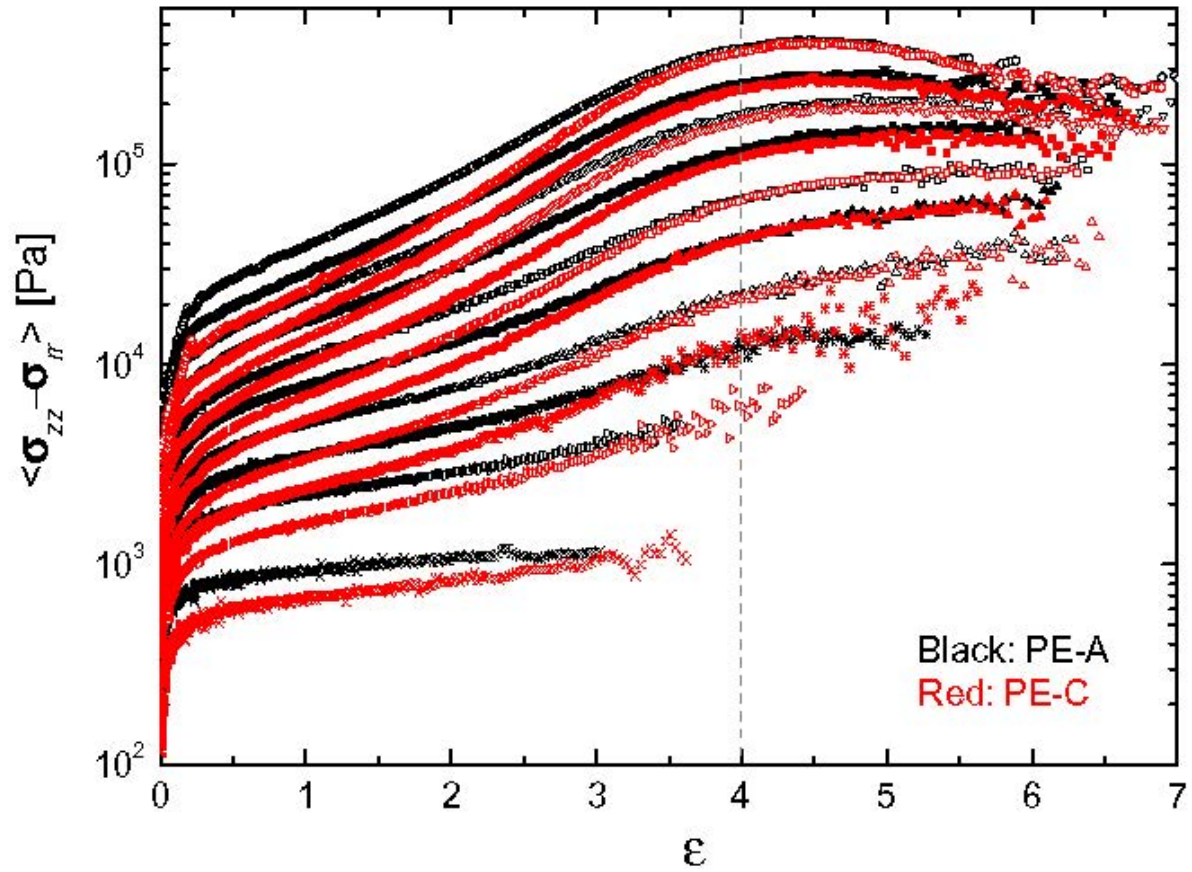
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- Challenges of Stress Relaxation
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Comparison of two commercial polyethylene samples (PE-A and PE-C) both at 150C.



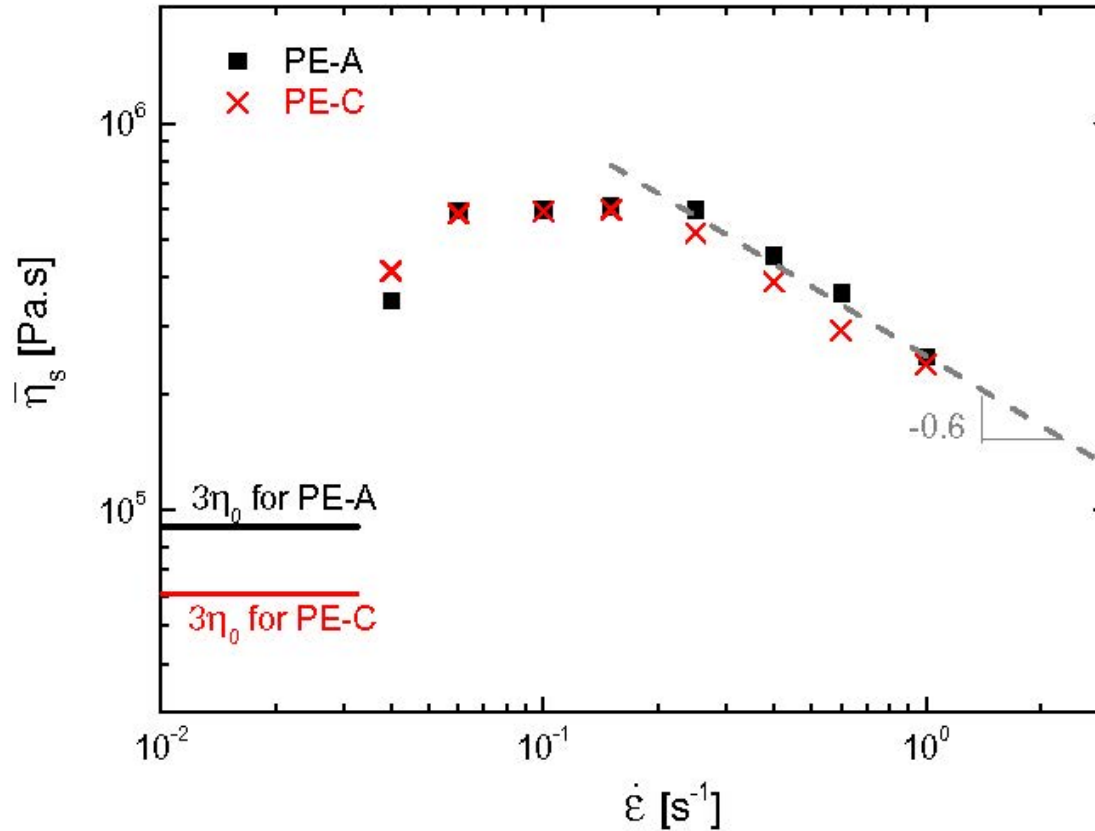
Extensional stress growth coefficient as function of time. Strain rate (from left to right): 1.0, 0.6, 0.4, 0.25, 0.15, 0.1, 0.06, 0.04, 0.025, and 0.01 (1/s).

Comparison of two commercial polyethylene samples (PE-A and PE-C) both at 150C.



Extensional stress as function of strain. Strain rate (top to bottom): 1.0, 0.6, 0.4, 0.25, 0.15, 0.1, 0.06, 0.04, 0.025, and 0.01 (1/s).

Comparison of two commercial polyethylene samples (PE-A and PE-C) both at 150C.



Steady extensional viscosity (η_E) as function of strain rate $\dot{\epsilon}$.

- Elongational viscosity of model linear polymers
- Elongational viscosity at processing (PE-A and PE-B)
- Elongational viscosity of model branched polymers
- Stress maximum in branched polymers (creep)
- Stress maximum in branched polymers (SAXS)
- Challenges of Stress Relaxation
- True stress relaxation (SANS)
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Comparison of model branched polymers:



Rouse time: $\tau_R \approx 85\text{s}$ at 130°C

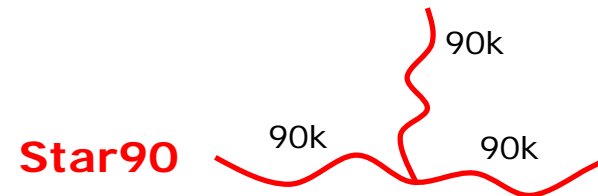
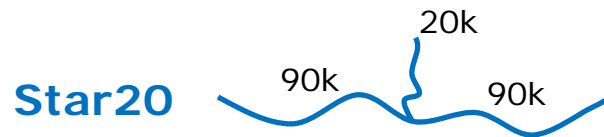
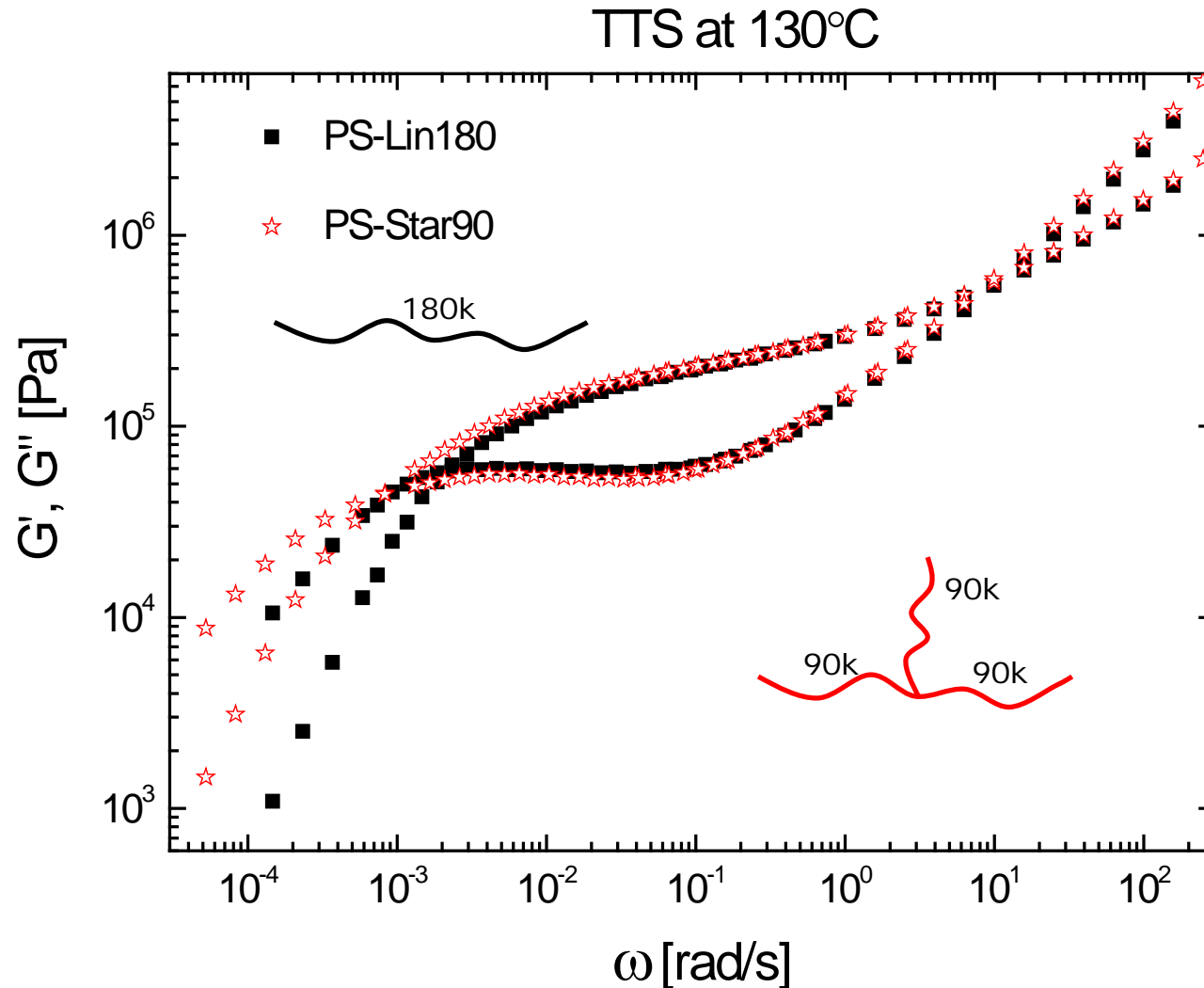


Table 1: The properties of the polystyrenes

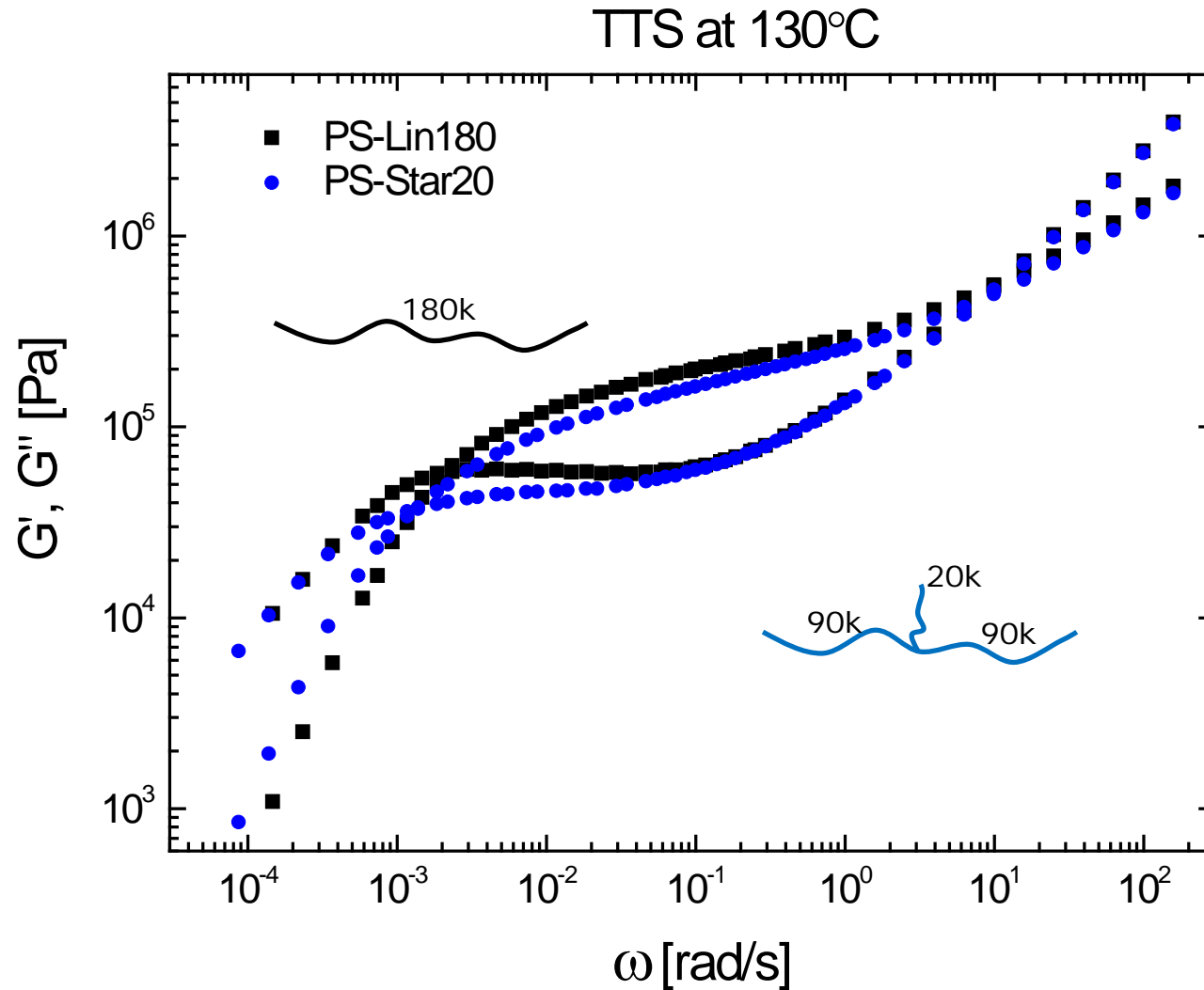
Sample name	Molar mass of long arm		Molar mass of short arm		Molar mass of polymer	
	M_w [g/mol]	\mathcal{D}	M_w [g/mol]	\mathcal{D}	M_w [g/mol]	\mathcal{D}
Lin180	–	–	–	–	187000	1.02
Star20	92400	1.03	20500	1.05	208300	1.03
Star90	92400	1.03	–	–	289100	1.03

Agostini et al., *European Polymer Journal* 49, 2769-2784(2013)

Linear Viscoelasticity

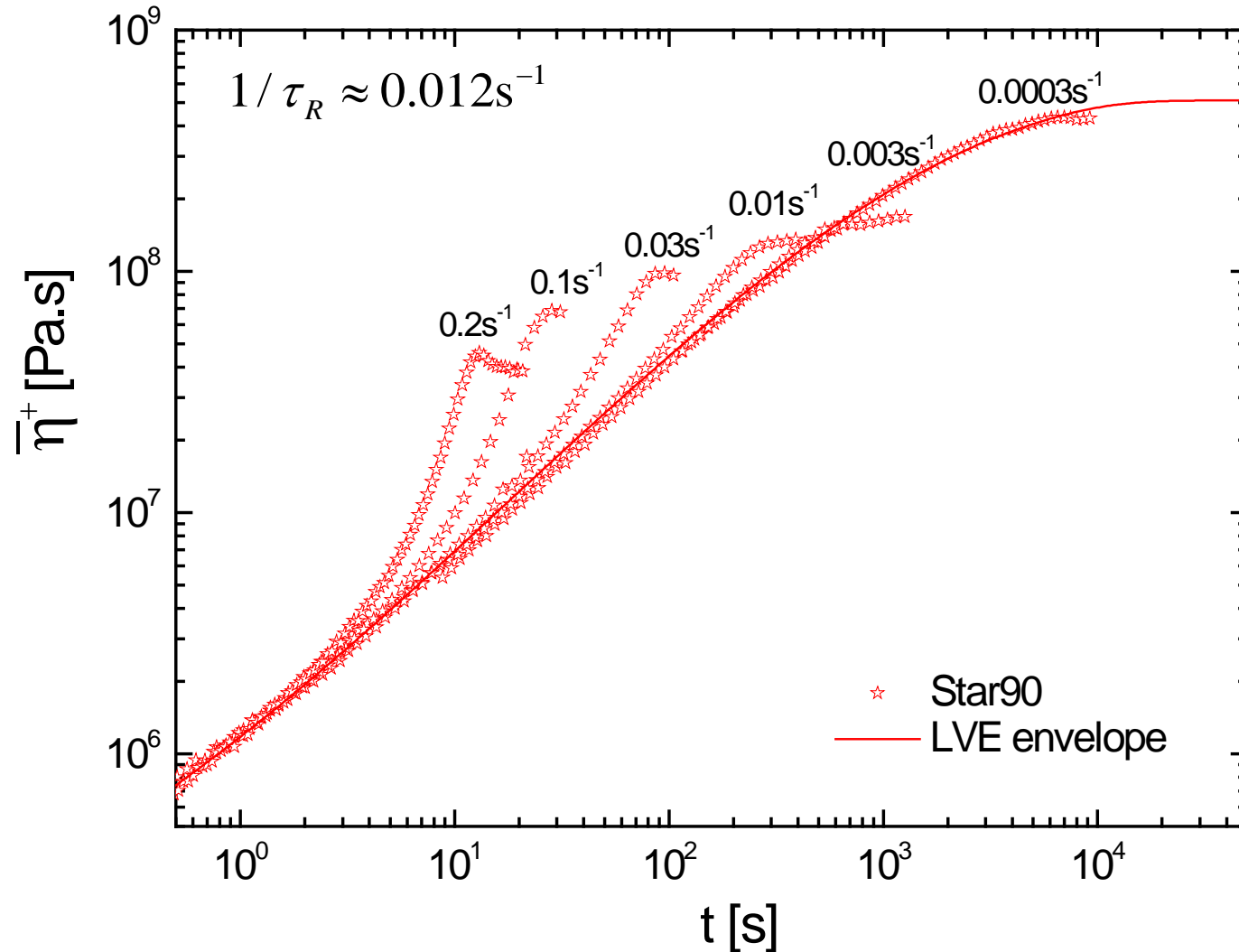


Linear Viscoelasticity

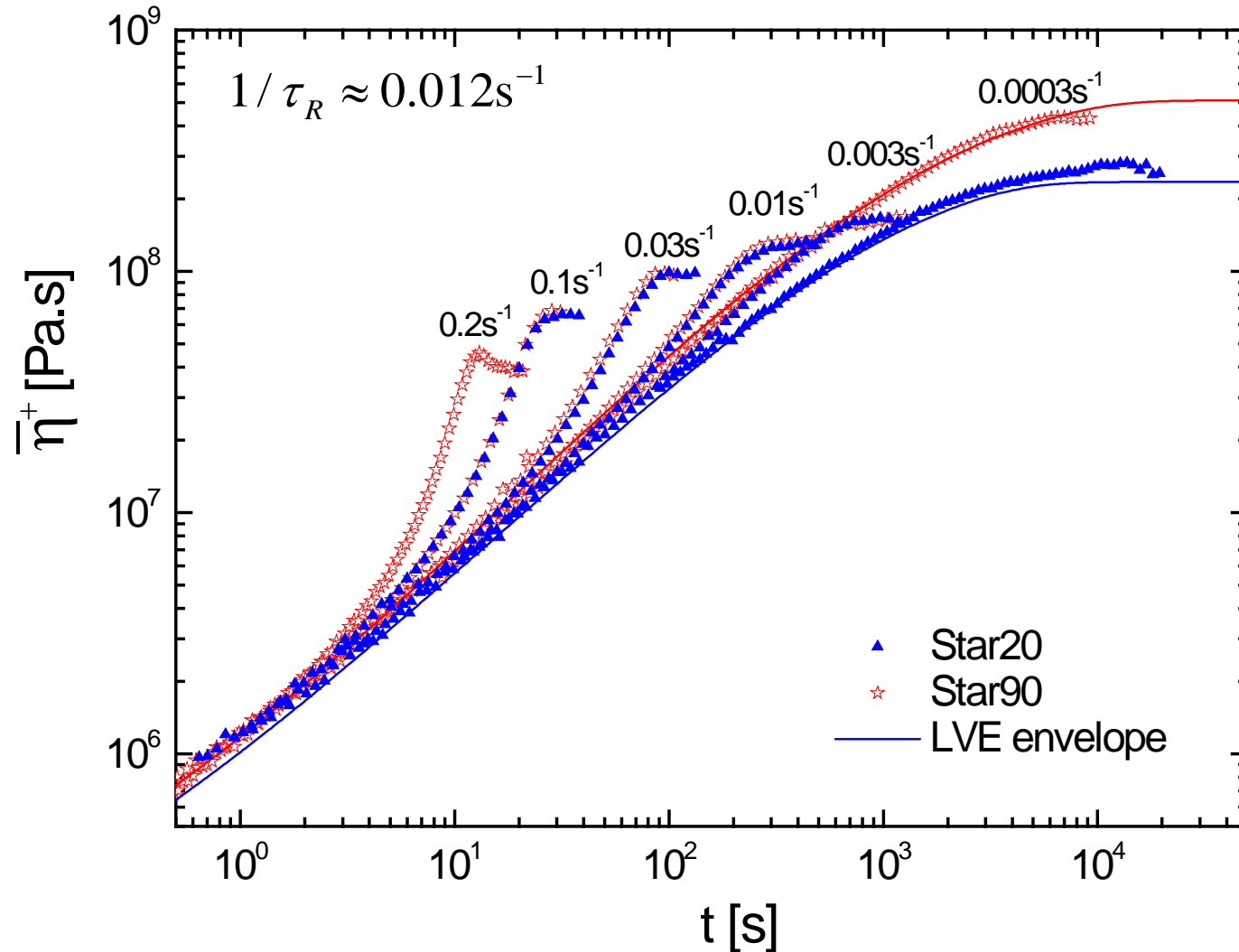


Extensional Flow

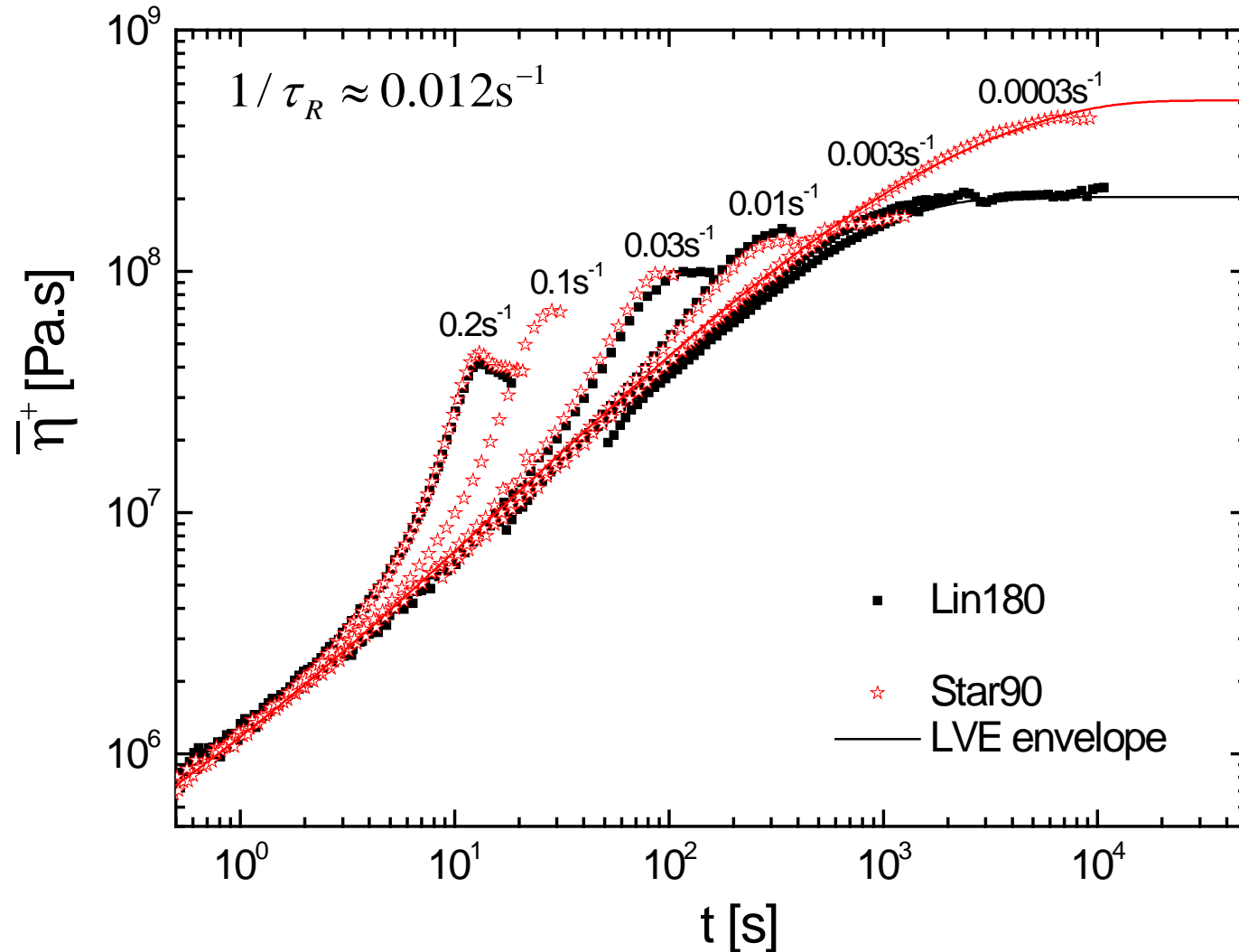
$$\bar{\eta}^+ = \langle \sigma_{zz} - \sigma_{rr} \rangle / \dot{\epsilon}$$



Extensional Flow

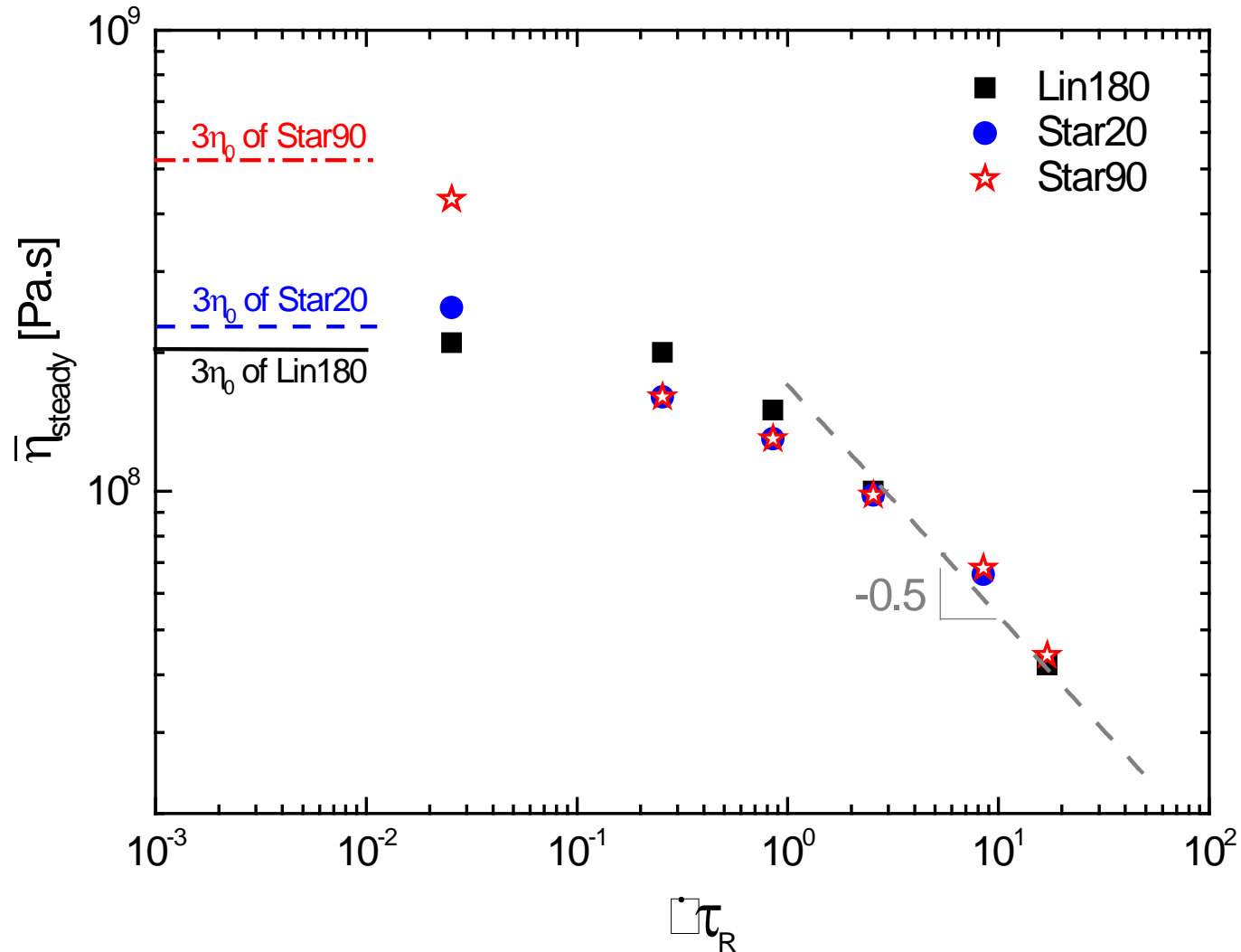


Extensional Flow



Steady state

Entangled star PS behave like linear PS in the **steady state** of fast elongational flow



- Elongational viscosity of model linear polymers
- Elongational viscosity at processing (PE-A and PE-B)
- Elongational viscosity of model branched polymers
- Stress maximum in branched polymers (creep)
- Stress maximum in branched polymers (SAXS)
- Challenges of Stress Relaxation
- True stress relaxation (SANS)
- Transient free surface viscoelastic flow (FEM)

Stress growth coefficient for BASF Lupolen 1840D and 3020D observed with first feedback loop on plate motion:

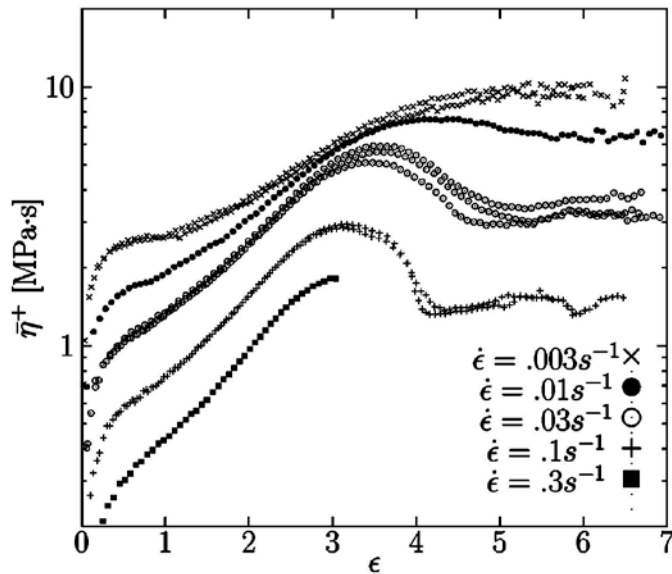


FIG. 1. The uncorrected transient elongation viscosities $\bar{\eta}^+$ of Lupolen 3020D measured at 130 °C, using Eq. (3), shown as a function of the Hencky strain, ϵ . $\bar{\eta}^+$ are measured at five different elongational rates $\dot{\epsilon}$ as shown in the figure.

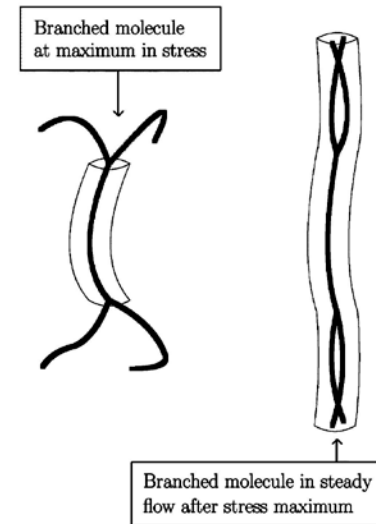


FIG. 6. Interpretation of reduction in stress in terms of Pom-Pom picture. At the maximum in stress, the arms contribute to the tension in the backbone. At steady state, the molecule becomes effectively a linear polymer without arms.

Viscosity overshoot in the start-up of uniaxial elongation of low density polyethylene melts, by H.K. Rasmussen, Nielsen, Bach and Hassager, J. Rheol. (2005)

Creep Measurements Confirm Steady Flow after Stress Maximum in Extension of Branched Polymer Melts

Nicolas J. Alvarez, José Manuel Román Marín, Qian Huang, Michael Locht Michelsen, and Ole Hassager*



FIG. 1. Polyethylene sample before (bottom, diameter 9 mm) and after (top) stretching. The midplane deformation corresponds to a Hencky strain of 6.7. The Hencky strain in the filament midplane is measured on-line by a laser micrometer during the experiment.

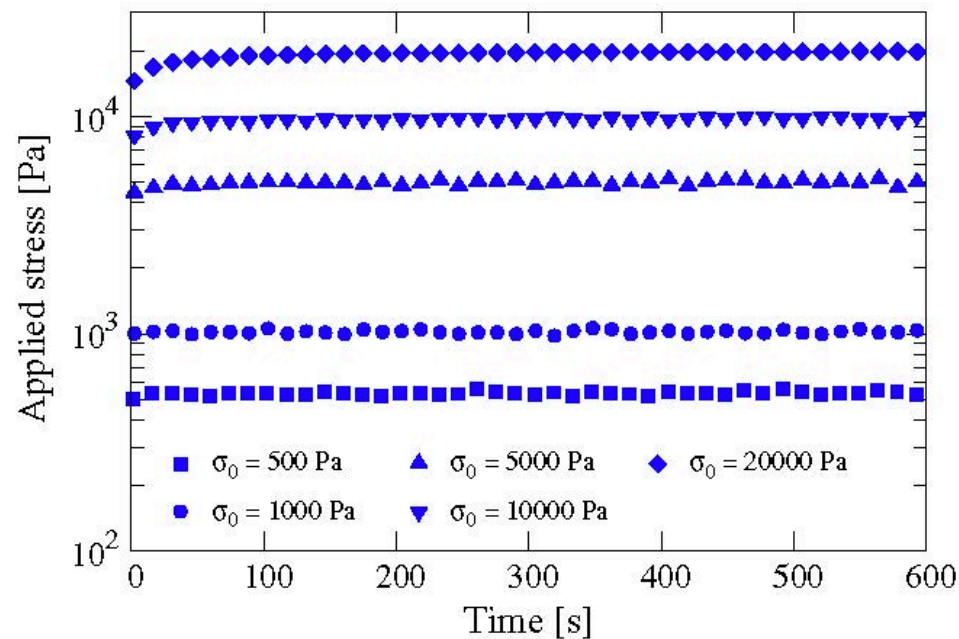
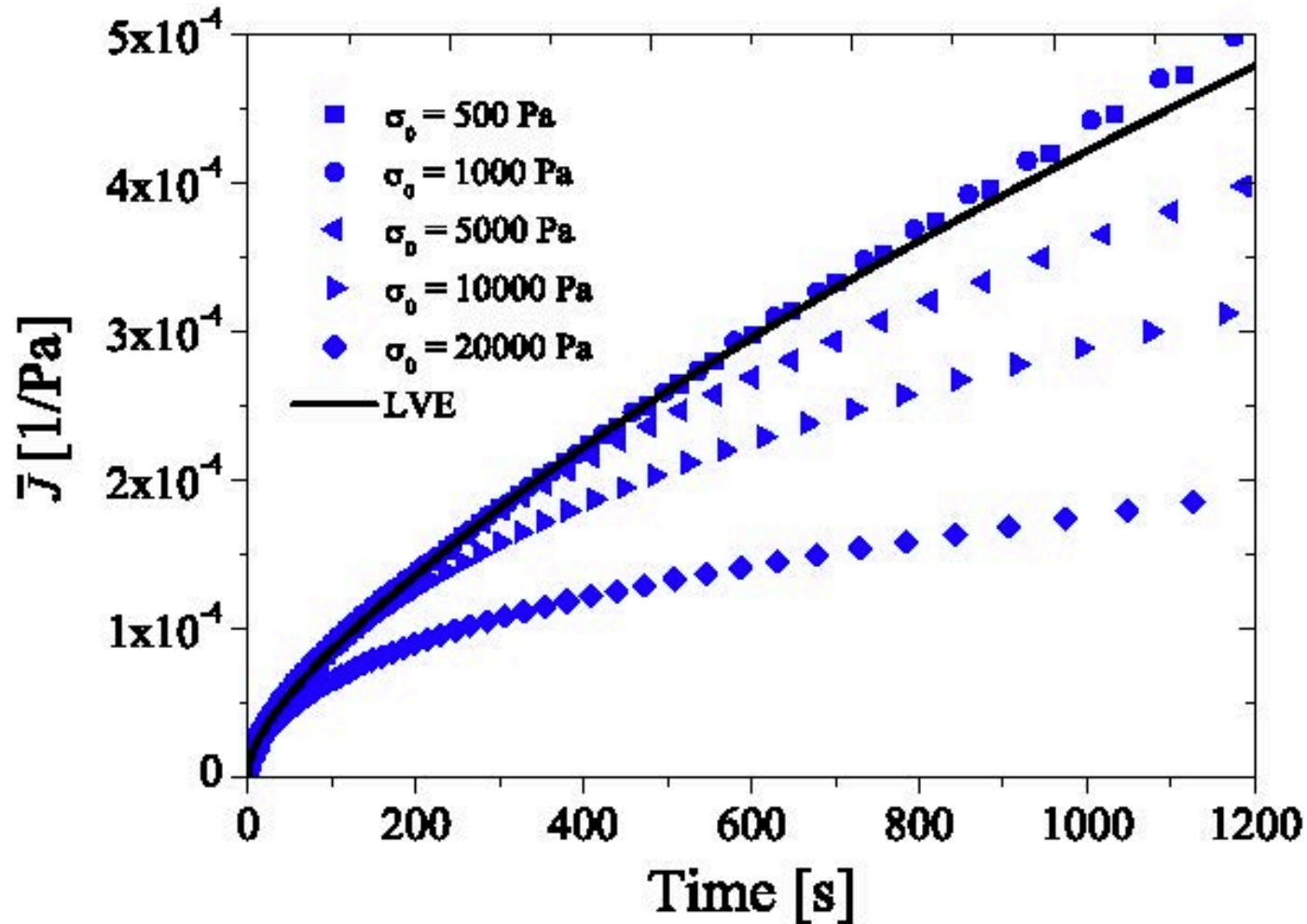


FIG. 2 (color online). Stress in the filament midplane measured as a function of time for five predefined stress levels.

Creep compliance: $J(t) = \epsilon(t)/\sigma$



Compliance: Comparison shear vs. extension:

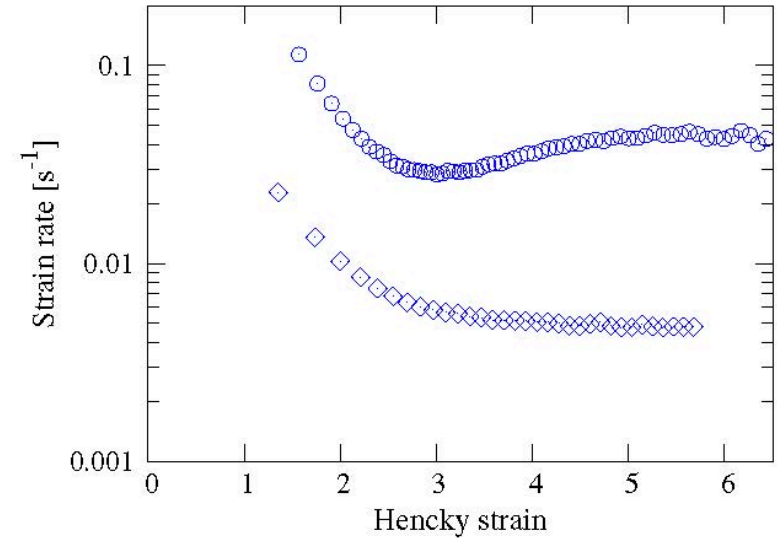
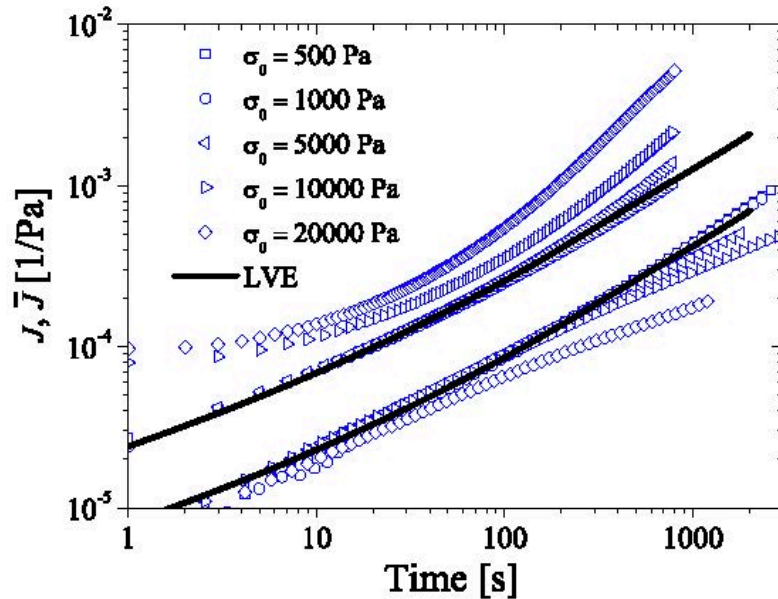


FIG. 3 (color online). Top: Extensional compliance \bar{J} versus time at five applied stresses. Bottom: Comparison of shear compliance (top curves) and extensional compliance versus time for five applied stresses (log-log scales). The solid lines in both graphs correspond to the predicted LVE compliance from Eq. (3).

FIG. 4 (color online). Hencky strain rate $\dot{\epsilon}$ as a function of Hencky strain ϵ for constant applied σ_0 equal to 40 kPa (diamonds, bottom) and 150 kPa (circles, top). For σ_0 larger than approximately 80 kPa, the strain rate goes through a minimum before reaching a steady state value.

Comparison constant stress (creep) vs. constant strain rate:

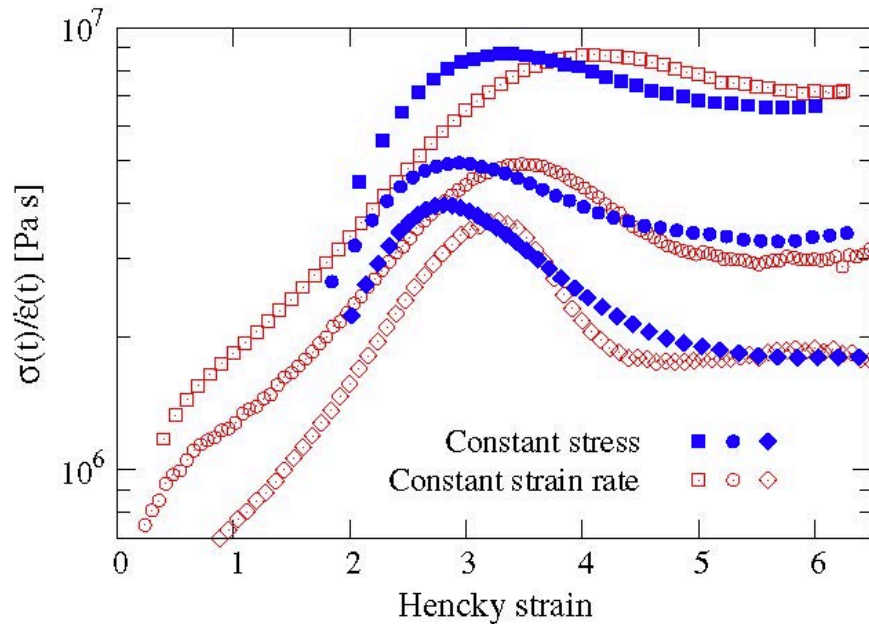


FIG. 5 (color online). $\sigma/\dot{\epsilon}$ as a function of Hencky strain ϵ for constant stress (closed symbols) and constant strain rate (open symbols) experiments. The constant stress experiments correspond to (filled squares) 80, (filled circles) 100, and (filled small diamonds) 200 kPa. The constant strain-rate experiments correspond to (squares) 0.01, (circles) 0.03, and (big diamonds) 0.1 s⁻¹.

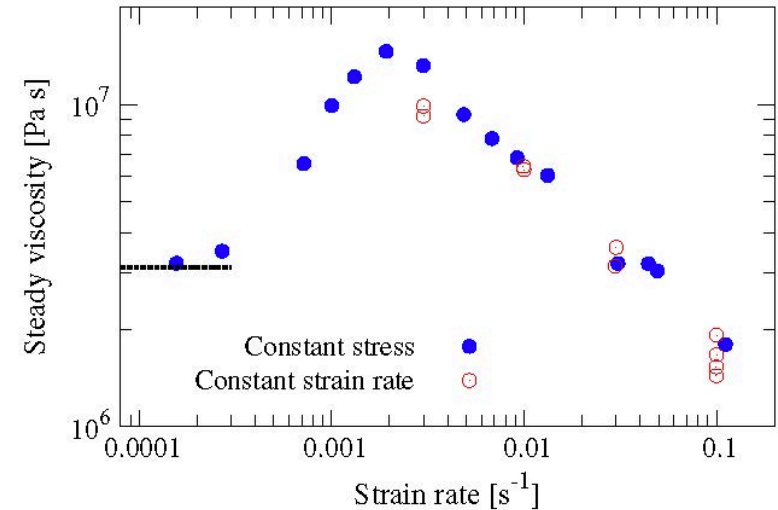


FIG. 6 (color online). Steady viscosity as a function of Hencky strain rate $\dot{\epsilon}$ for constant stress (closed symbols) and constant strain-rate (open symbols) experiments [4]. The dashed line represents the zero shear rate viscosity determined from the LVE.

Comparison constant stress (creep) vs. constant strain rate:

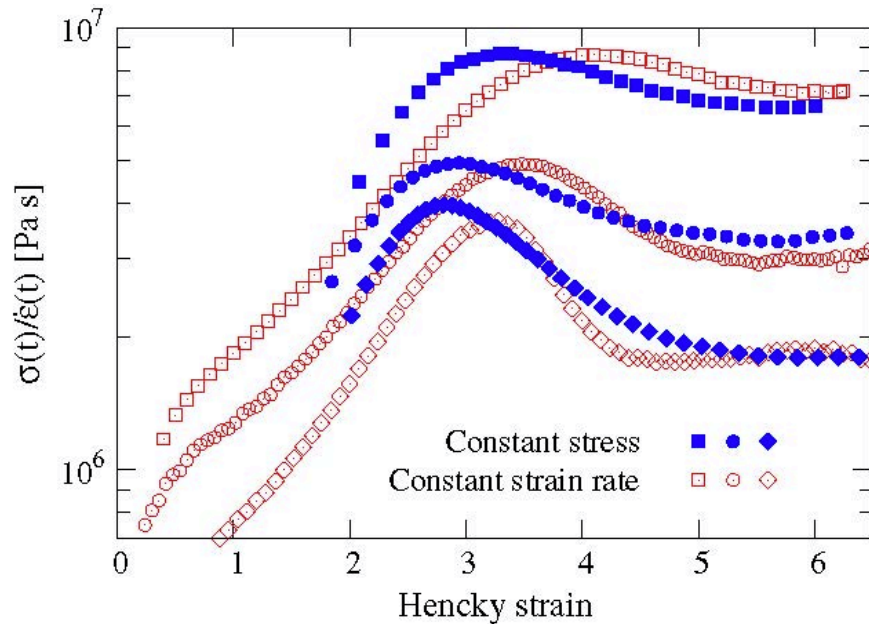


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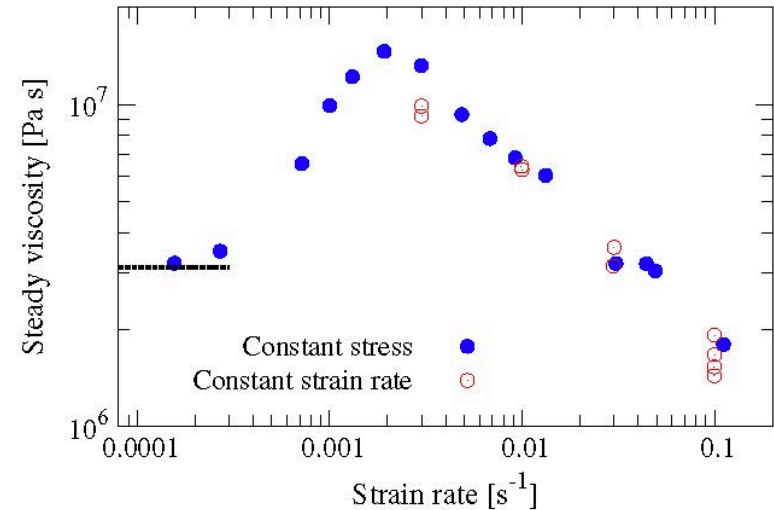


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Alvarez et al., Physical Review Letters, (2013).

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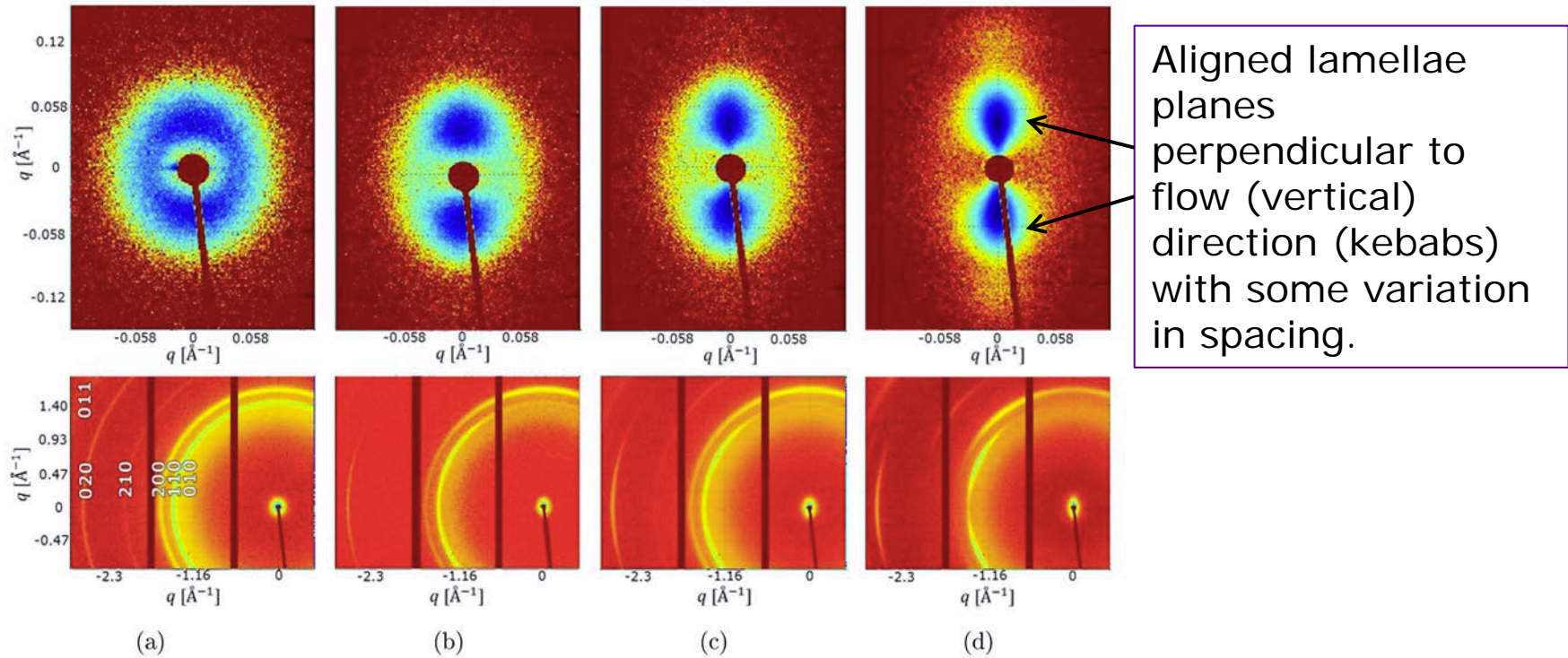
VADER 1000

Knob for lifting oven

Oven surrounding sample can be lifted up for rapid quenching of sample

www.rheofilament.com

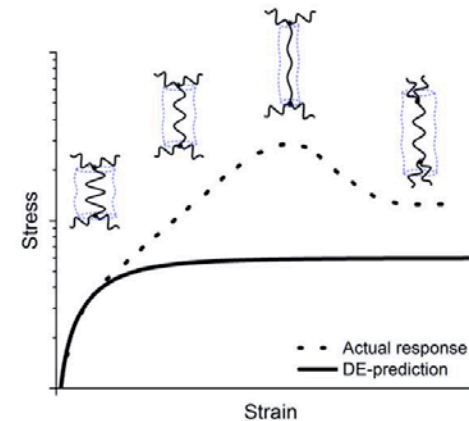
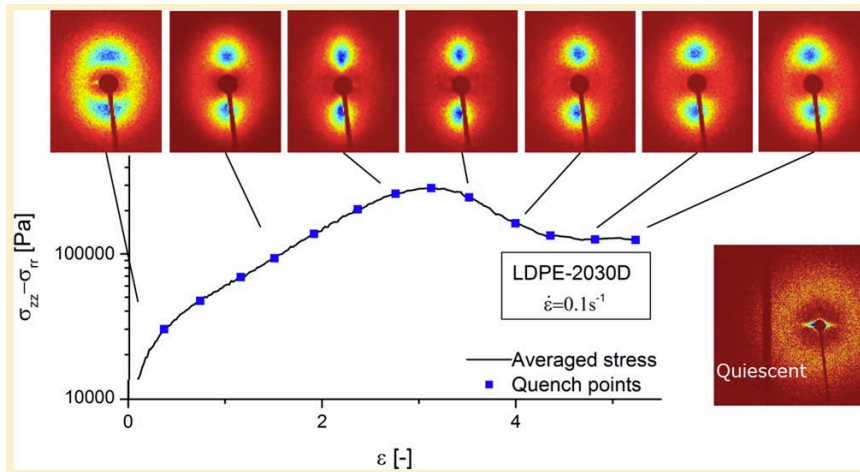
Ex-Situ X-ray Measurements on quenched filaments:



SAXS (top) and WAXD (bottom) patterns of LDPE filaments quenched at various stress (a) 30, (b) 126, (c) 262, and (d) 518 kPa. All filaments have been elongated at $T = 130\text{C}$, and flow direction is vertical.

Wingstrand et al., *Macromolecules* (2017)

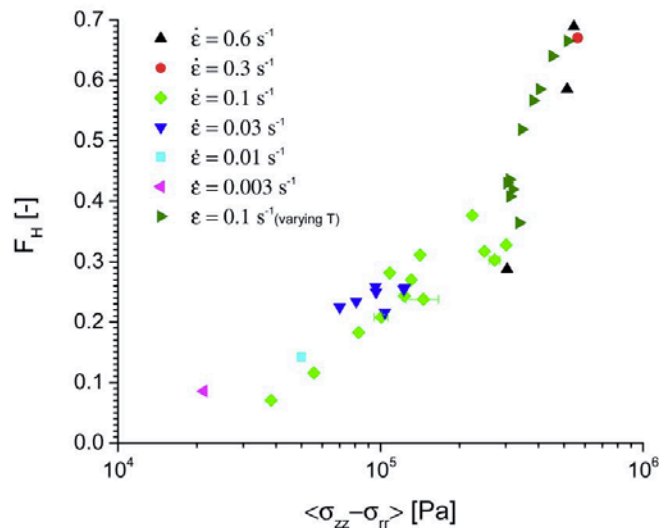
Visual appearance of SAXS patterns suggest correlation with stress (before and after maximum).



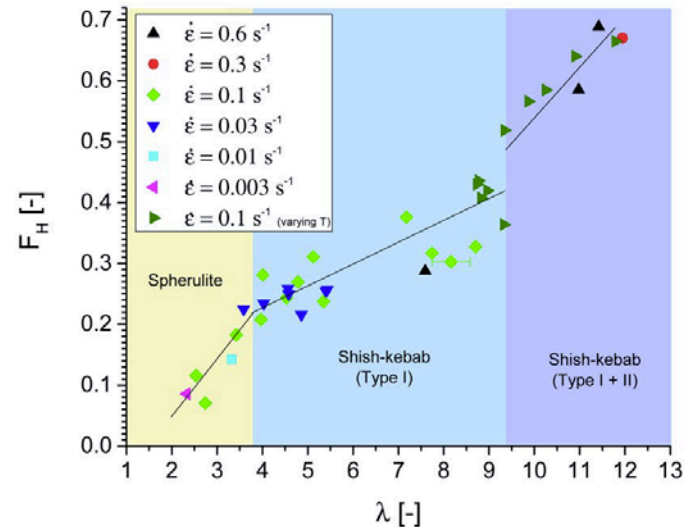
The X-ray wavelength was 1.54 Å for both WAXD and SAXS while the distance to the detector was ~118 mm for the WAXD measurements and either ~1020 or ~2240 mm for the SAXS measurements.

Wingstrand et al., Macromolecules (2017)

Herman's orientation factor for SAXS patterns confirm unique relation between stress before quench and crystalline orientation.



Herman's orientation factor vs stress at quench. All samples have been stretched at $T = 130\text{C}$ except (green) which have been stretched at varying T from 110 to 130C.



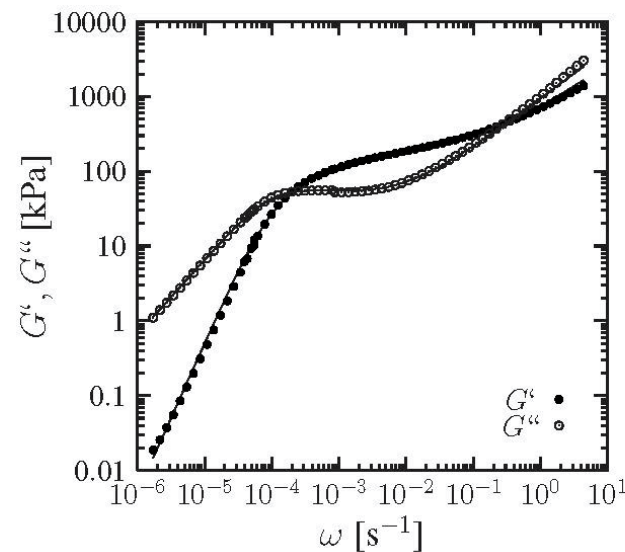
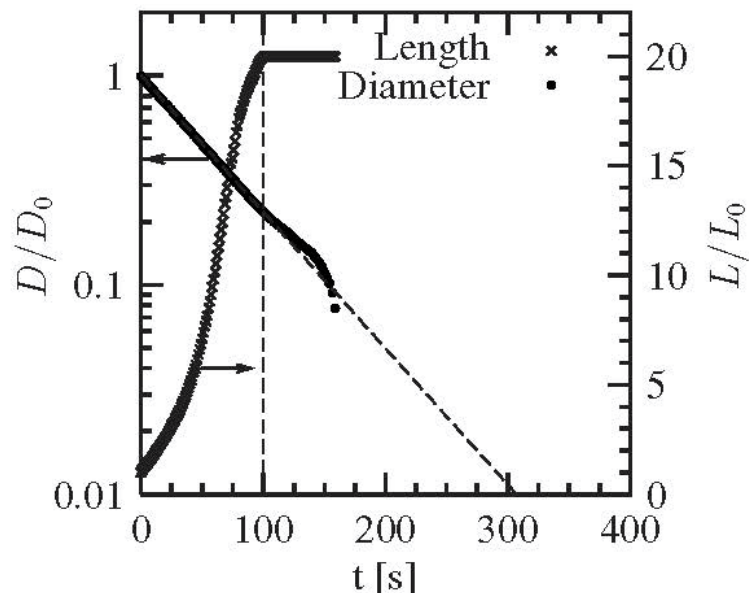
Herman's orientation factor vs average stretch of the backbone for the two highest Maxwell models. Vertical lines indicate regions of different morphologies.

Wingstrand et al., Macromolecules (2017)

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- **Challenges of Stress Relaxation**
- True stress relaxation (SANS)
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The incorrect stress relaxation experiment: $\dot{\epsilon}_N = 0$

No feed-back control

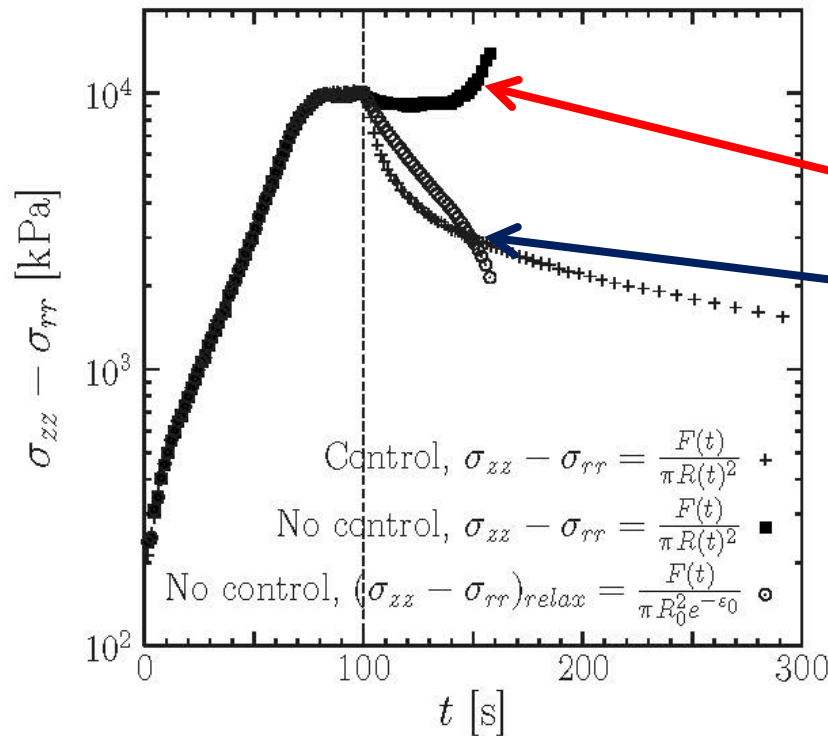


Evolution of filament diameter, $D(t)$, and plate separation, $L(t)$, for an elongational rate of 0.03 s^{-1} at 120C , stretched for 100 s and then relaxed. $D_0=3.40 \text{ mm}$ and $L_0=8.82 \text{ mm}$.

Sample: NMMD polystyrene
Mw = 145 kg/mol, 120C .

Failure by necking after about 60 s relaxation.

The incorrect stress relaxation experiment: $\dot{\epsilon}_N = 0$



How do you report stress in uncontrolled stress relaxation experiments?

- True stress?
- Engineering stress?

Stress relaxation after $\epsilon = 3$

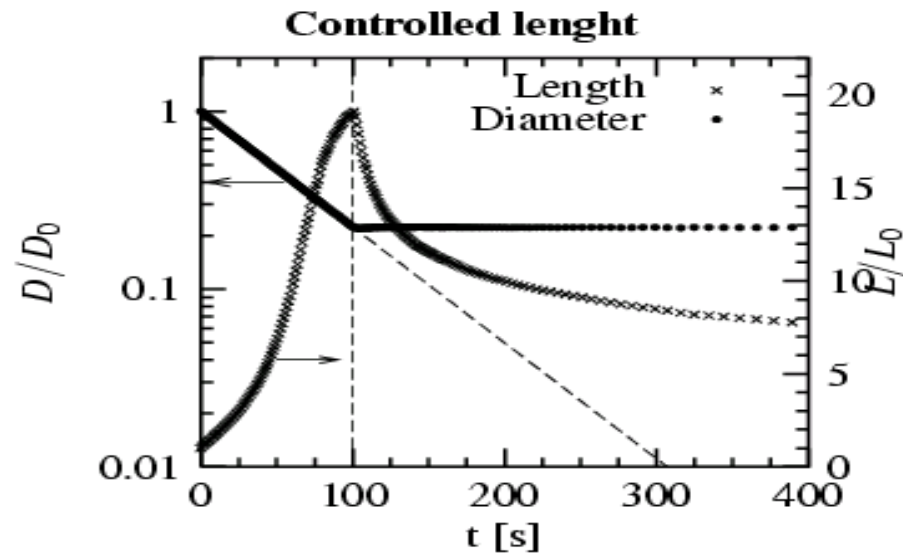
Total time 14400 s = 4 h

Note: The true stress actually increases with time in (some) uncontrolled stress relaxation experiments!

J. Rheol. 524, 885-899 July/August 2008
0148-6055

- Elongational viscosity of model linear polymers
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The correct stress relaxation experiment: $\dot{\epsilon} = 0$



Evolution of plate separation $L(t)$ and mid-filament diameter $D(t)$.

145 kg/mol polystyrene at 120 C.

Elongation rate 0.03 s^{-1} for $0 < t < 100 \text{ s}$.

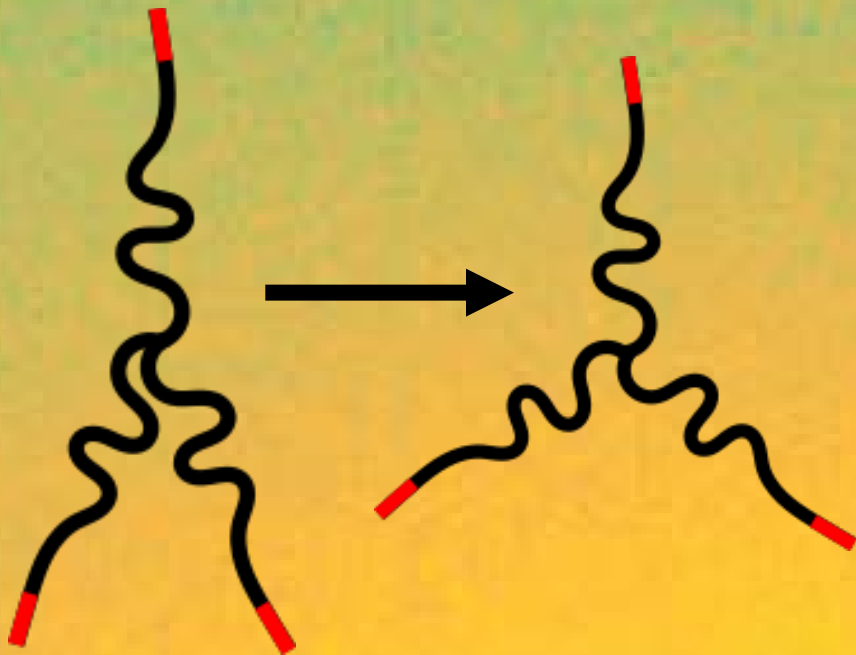
For $t > 100 \text{ s}$ the plates are either held fixed (left) or controlled to keep mid-filament diameter constant (right)

$D_0 = 3.40 \text{ mm}$, $L_0 = 8.82 \text{ mm}$



145 kg/mol polystyrene at 120 C. Elongation rate 0.03 s^{-1} for 100 s.

Samples quenched after [0, 7, 47, 126, 350, 3000, 7000] s of relaxation



Star polymer relaxation after fast extensional flow using neutrons

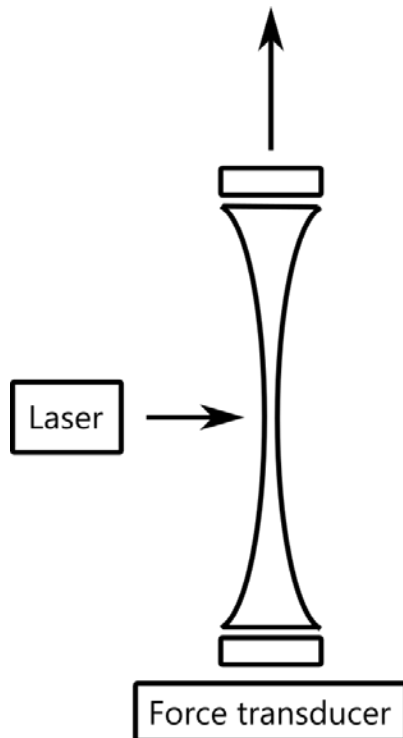
Anine Borger, PhD student, Niels Bohr Institute.

AERC 2018, Sorrento, April 18th

K. Mortensen, Q. Huang, O. Hassager, J. Kirkensgaard, K. Almdal, C. Garvey



Filament stretch rheometer



Hencky strain

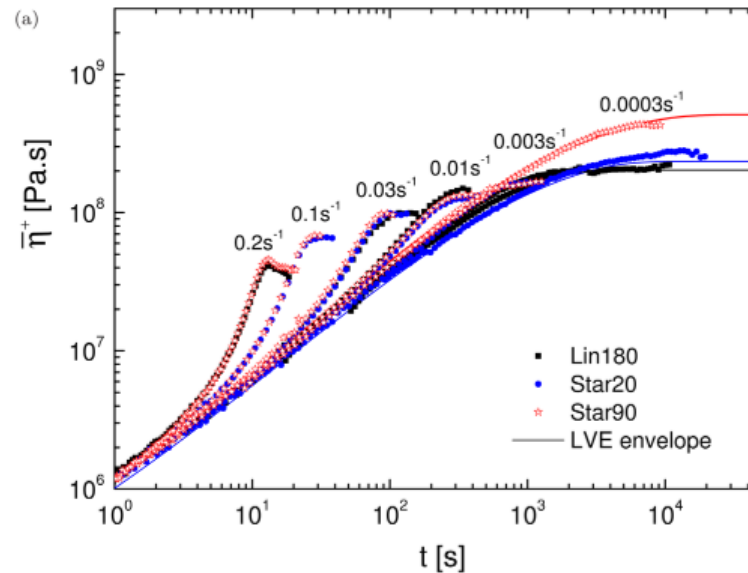
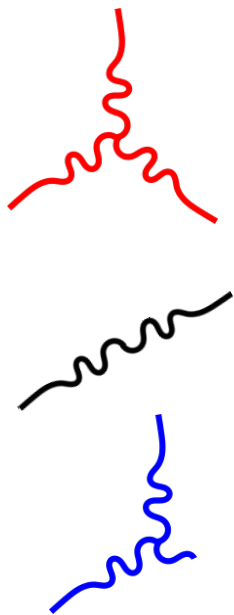
$$\varepsilon(t) = -2 \ln \left(\frac{R(t)}{R_0} \right)$$

Control scheme:

- Stretch at constant Hencky strain rate
- Stress relaxation at constant Hencky strain

A. Bach et al, J. Rheol., 47(2), 429-441 (2003)

Branched polymers behave like linear in steady state of fast extensional flow

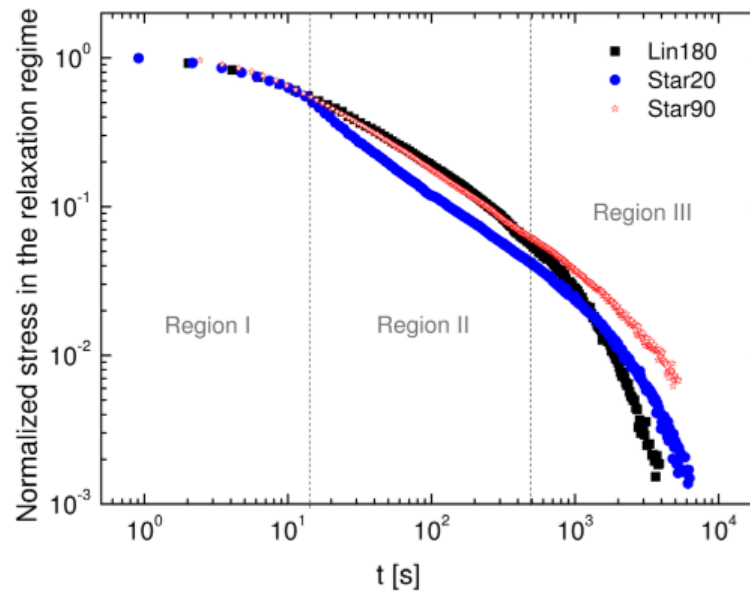
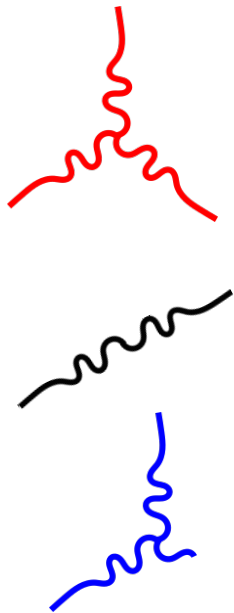


$$\eta^{\pm} = \langle \sigma_{zz} - \sigma_{rr} \rangle / \dot{\epsilon}$$

$$\dot{\epsilon} > 1/\tau_r \approx 0.01 s^{-1}$$

Q. Huang et al. *Macromolecules* 2016, 49, 6694-6699

The relaxation of star and linear polymers differ at long time scales

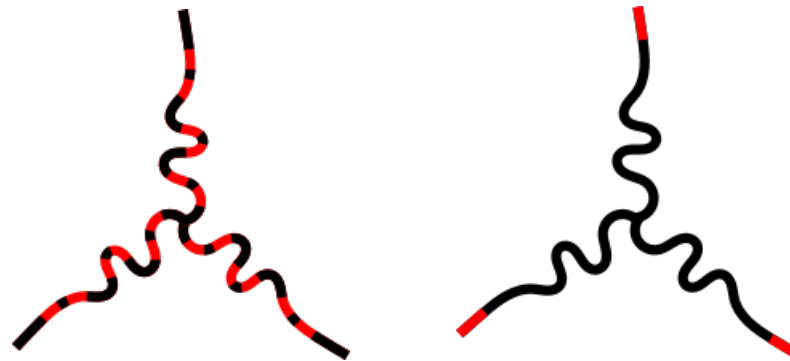


I : < 15 s
 II: $15\text{-}500$ s
 III: > 500 s

$$\dot{\epsilon} = 0.03 s^{-1}$$

Q. Huang et al. Macromolecules 2016, 49, 6694-6699

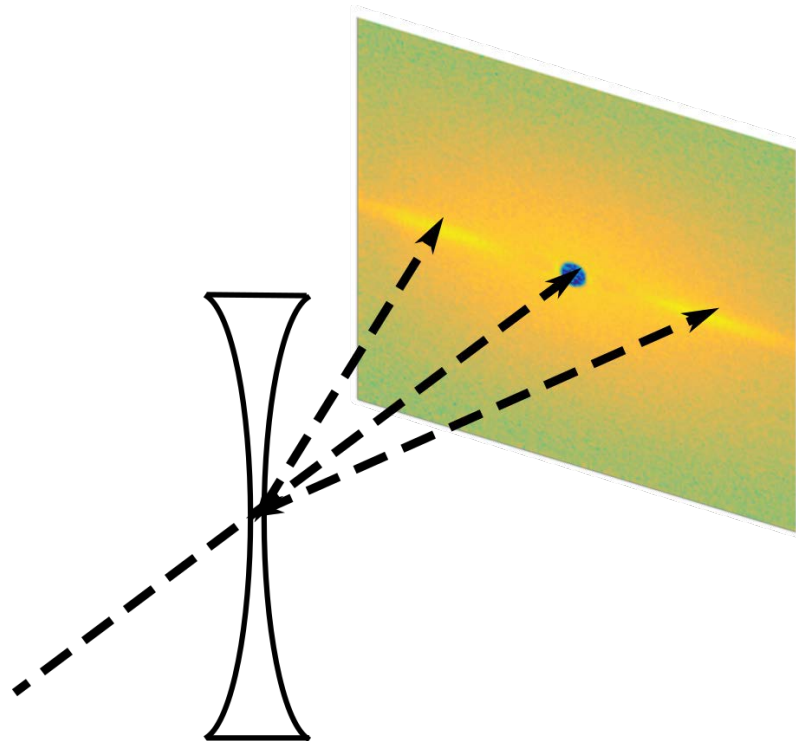
Two model sample series for neutron scattering



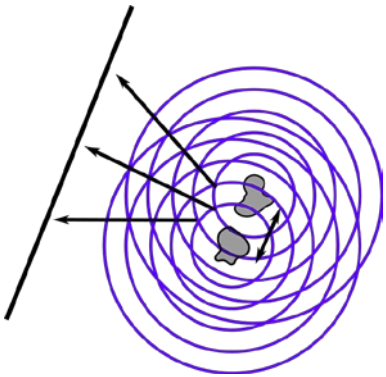
I : < 15 s
 II: 15-500 s
 III: >500 s

	Random deuterated	End-deuterated
Arm length	100 k	
W_{iR}	27	
Relaxation times (s)	0, 10, 100, 1000, fully	0, 5, 200, 4000, fully

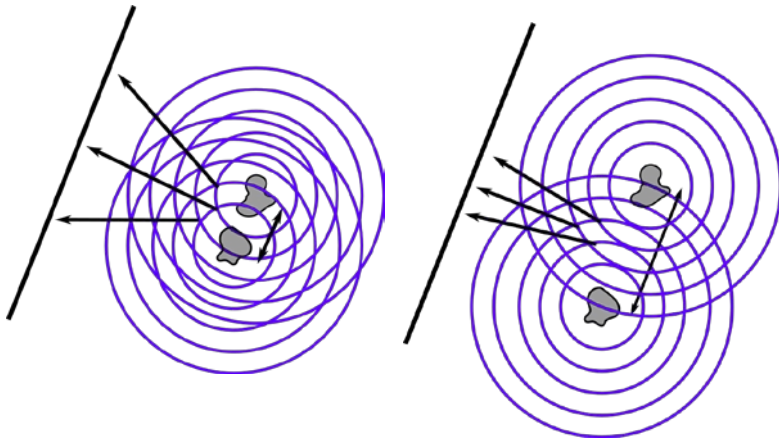
Neutron scattering data: Setup



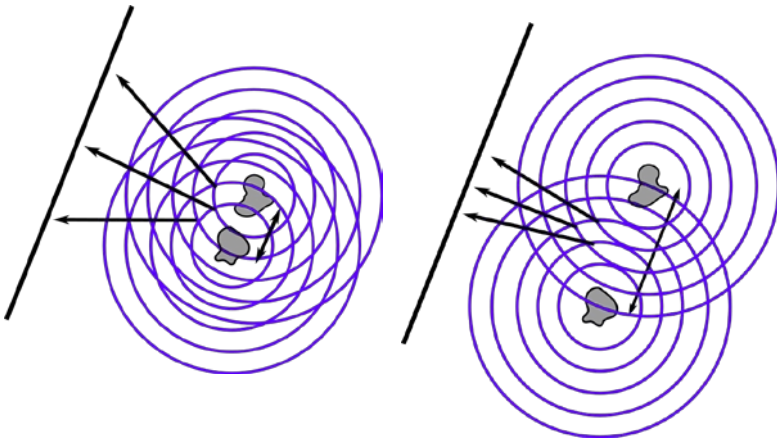
Neutron scattering: Correlations and sizes



Neutron scattering: Correlations and sizes

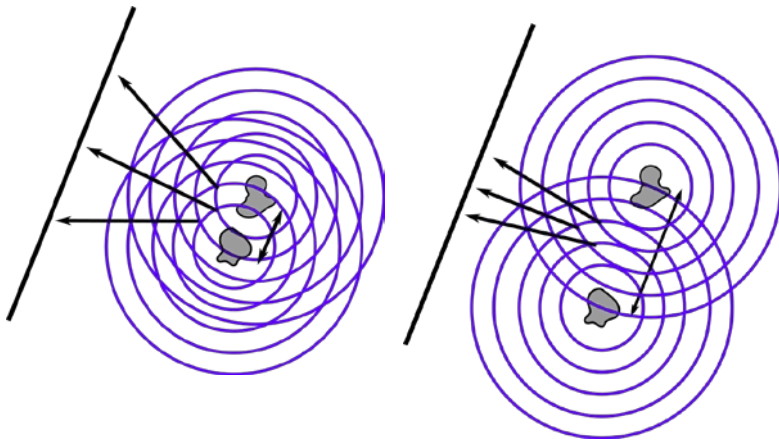


Neutron scattering: Correlations and sizes



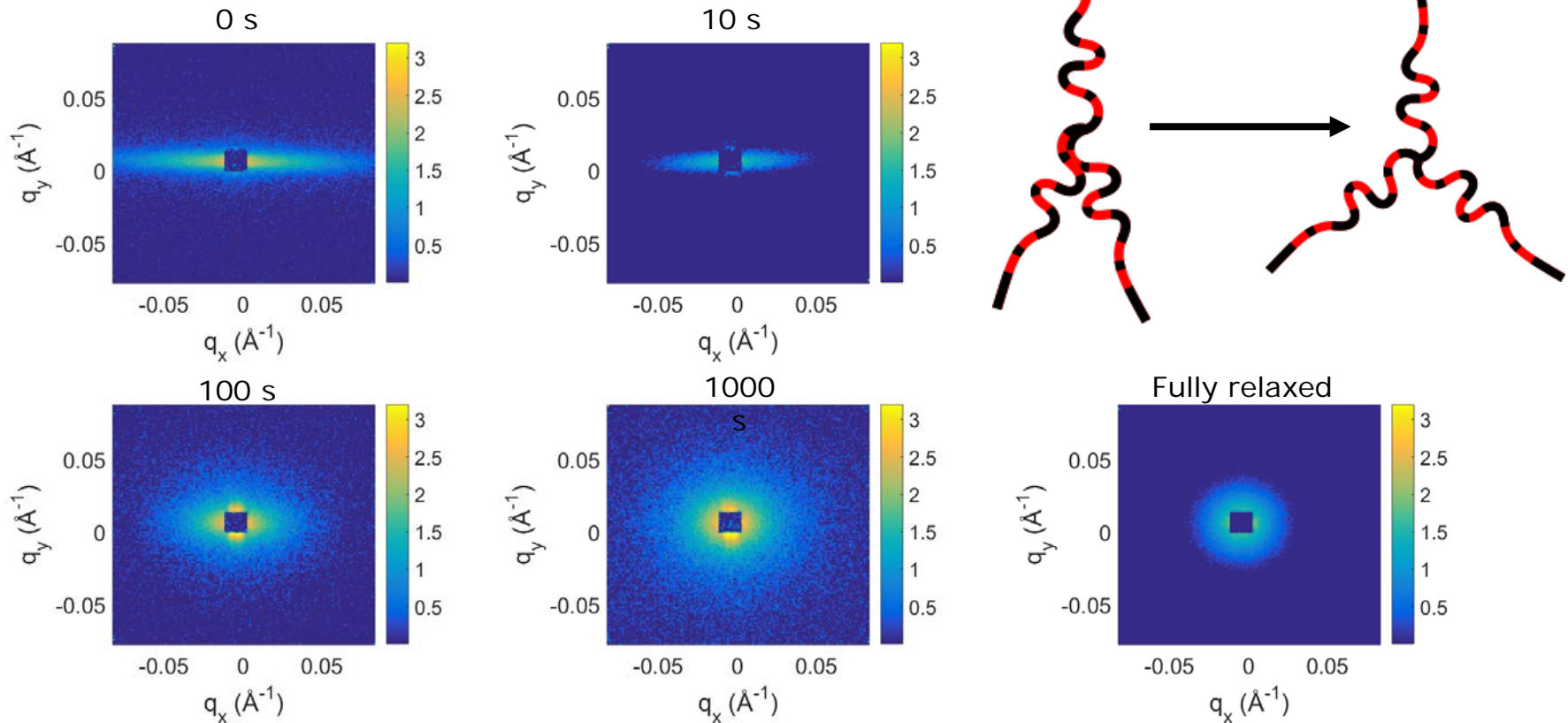
- Characteristic distances in the sample give peaks in intensity
- Scattering maps reciprocal space

Neutron scattering: Correlations and sizes



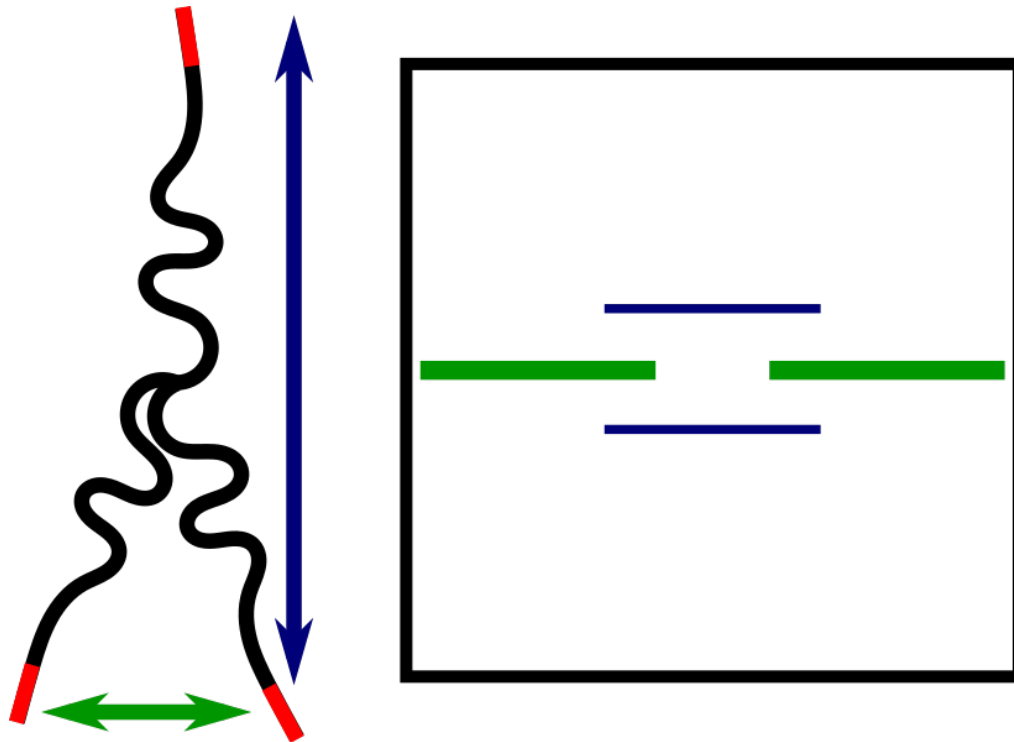
- Characteristic distances in the sample give peaks in intensity $q = \frac{4\pi \sin \theta}{\lambda}$
- Scattering maps reciprocal space
- Scattering vector

Random-deuterated stars

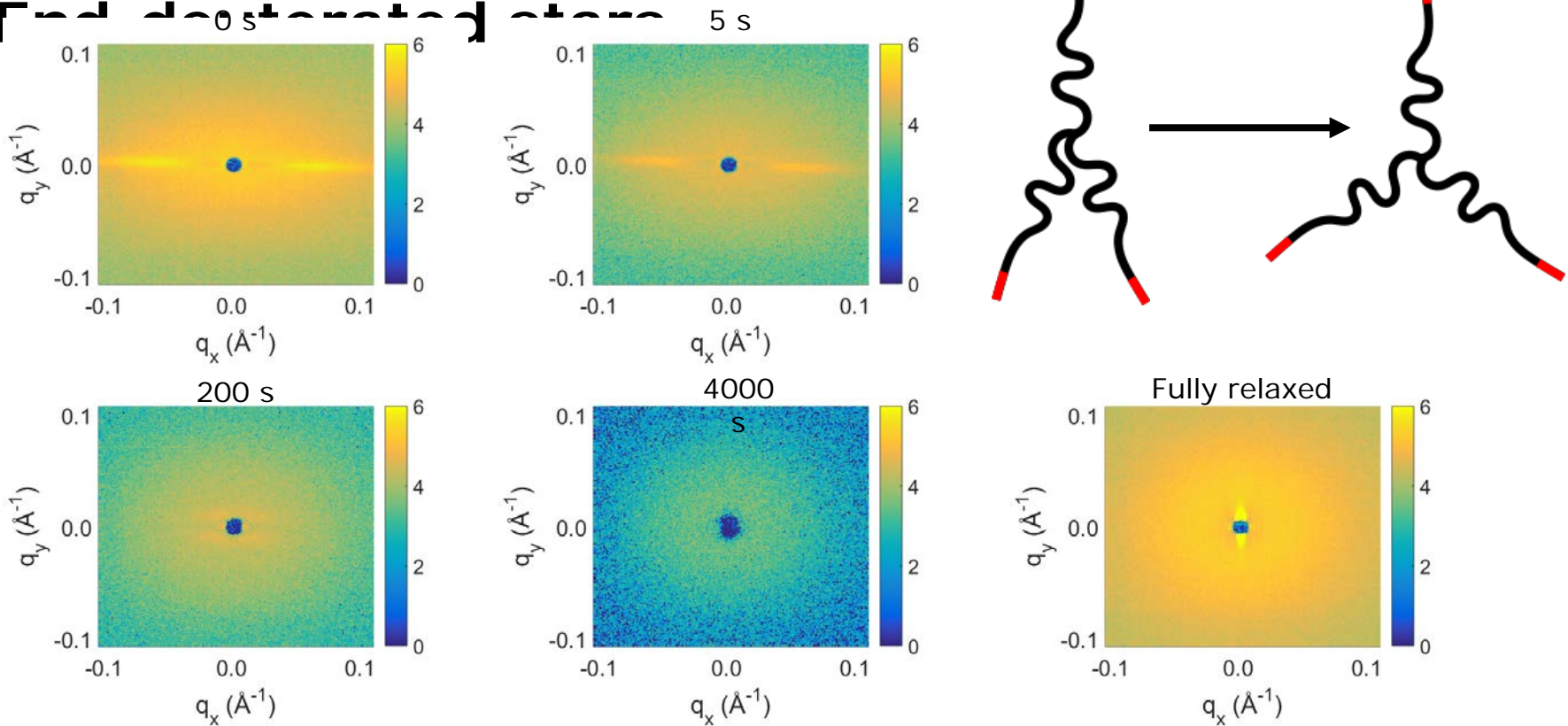


PSI:
SANS-I

End-deuterated stars: Expectation



End deuterated styrene



0s and fully relaxed: K. Mortensen et al., accepted for publication(2018)

ANSTO: QUOKKA

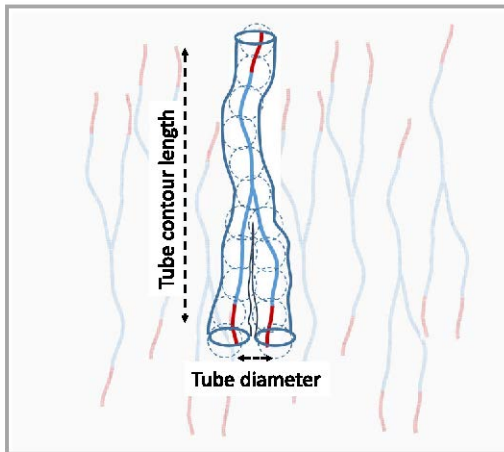
Polystyrene: $a = 85 \text{ \AA}$

Total length: $2 N_{arm} b = 4960 \text{ \AA}$

Length of oriented tubes: $2 Z_{arm} a = 1040 \text{ \AA}$

Measured axial distance: 1300 \AA

Measured transverse distance: 100 \AA



Conclusions:

Oriented tube elements with small stretching.

Arms more or less inside one tube section.

- Elongational viscosity at processing (PE-A and PE-B)
- Molecular extensional rheology (model polymers)
- Stress maximum in branched polymers (creep)
- Stress maximum in branched polymers (SAXS)
- Challenges of Stress Relaxation
- True stress relaxation (SANS)
- Transient free surface viscoelastic flow (FEM)

All materials:

$$\text{Mass: } \nabla \cdot v = 0$$

$$\text{Force: } \rho \left(\frac{\partial}{\partial t} + v \cdot \nabla \right) v = -\nabla p + \nabla \cdot \sigma$$

Newtonian liquids:

$$\text{True Stress: } \sigma(t) = \mu \dot{\gamma}(t)$$

$$\text{Viscosity: } \mu$$

$$\text{Rate-of-deformation tensor: } \dot{\gamma} = [\nabla v + (\nabla v)^\dagger]$$

Polymeric liquids:

$$\text{True Stress: } \sigma(t) = \sigma\{v(t'), t' \in [-\infty; t]\}$$

Simulations of transient viscoelastic flow:
Lagrangian finite element method.

All materials:

$$\text{Mass: } \nabla \cdot v = 0$$

$$\text{Force: } \rho \left(\frac{\partial}{\partial t} + v \cdot \nabla \right) v = -\nabla p + \nabla \cdot \sigma$$

Polymeric liquids:

$$\text{True Stress: } \sigma(t) = \sigma\{v(t'), t' \in [-\infty; t]\}$$

Yu, Marin, Rasmussen and Hassager,
J. Non-Newtonian Fluid Mech. 165 (2010)

Polyisoprene



Doi-Edwards constitutive equation in K-BKZ integral form

$$\boldsymbol{\sigma}(t) = \int_{-\infty}^t \lambda^2(t, t') M(t - t') (\phi_1 \mathbf{B} - \phi_2 \mathbf{B}^{-1}) dt'$$

$$\phi_1 = \frac{f_1}{U} \quad \phi_2 = \frac{f_2}{U}$$

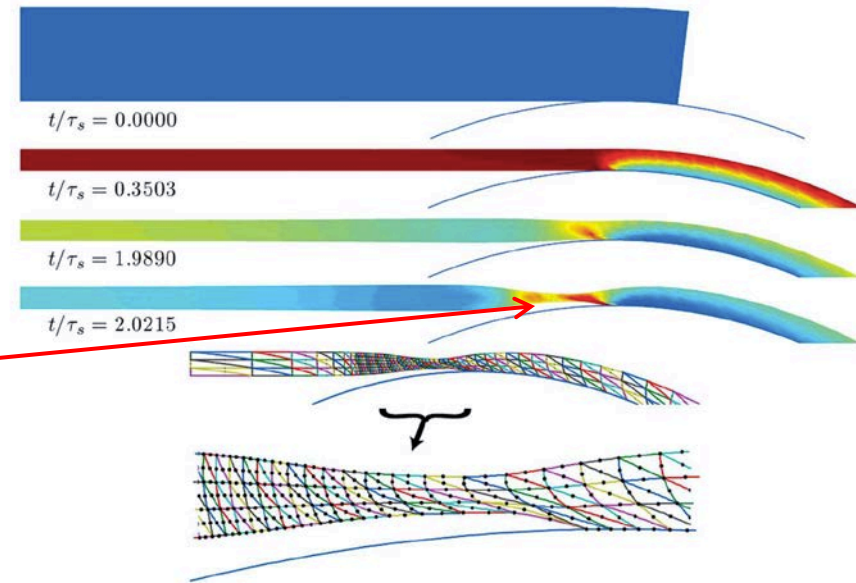
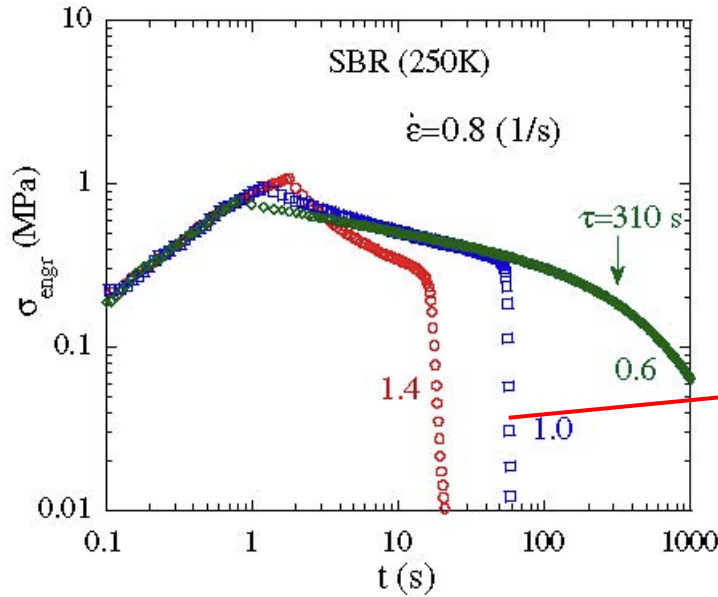
$$\lambda^2(t, t') = \left[1 + (\sqrt{U} - 1) e^{-\frac{(t-t')}{\tau_R}} \right]^2 \quad \tau_R = 0.67s$$

The effect of independent alignment(IA) approximation

	With IA	Without IA
f_1	$\frac{5}{7}$	$\frac{15}{16} \left(1 - \frac{3}{2} \frac{I_2^{3/2}}{I_1} + \frac{I_2}{I_1^2} \left(\frac{9}{8} - \log I_1 + \frac{1}{2} \log I_2 \right) - \frac{4a_1}{I_1^2} \right)$
f_2	$\frac{5}{7} \frac{1}{\sqrt{I_2 + 13/4}}$	$\frac{15}{16} \left(\frac{9}{8} \frac{I_2^{1/2}}{I_1^2} + \frac{1}{I_1} \left(-\frac{5}{8} + \log I_1 - \frac{1}{2} \log I_2 \right) - \frac{4a_2}{I_2^2} \right)$
U	$\frac{1}{7} (I_1 + 2\sqrt{I_2 + 13/4} - 1)$	$\frac{1}{4} I_1 + \frac{1}{4} \frac{I_2}{I_1} \left(-\frac{1}{8} + \frac{3}{4} \frac{I_2^{1/2}}{I_1} + \log I_1 - \frac{1}{2} \log I_2 \right) + \frac{a_1}{I_1} + \frac{a_2}{I_2} + a_3$

Relaxation with $\dot{\epsilon}_N = 0$ constant nominal Hencky strain may result in highly inhomogeneous flow and necking.

Necking close to cylinder:

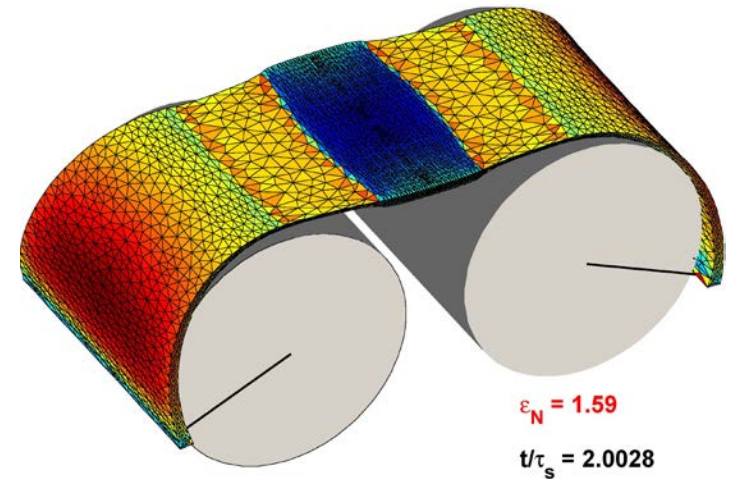
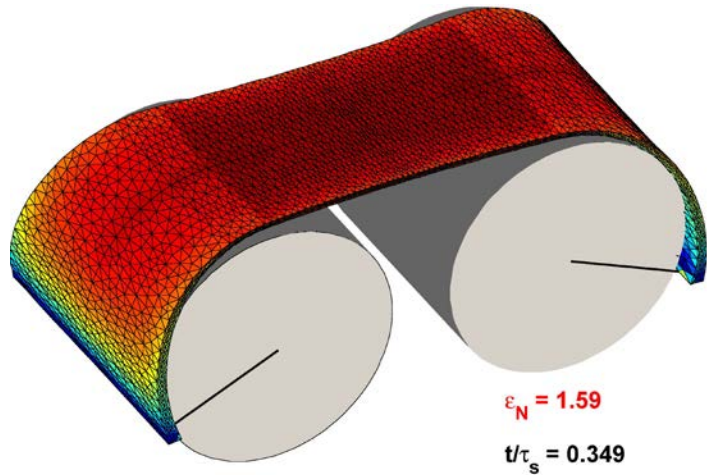
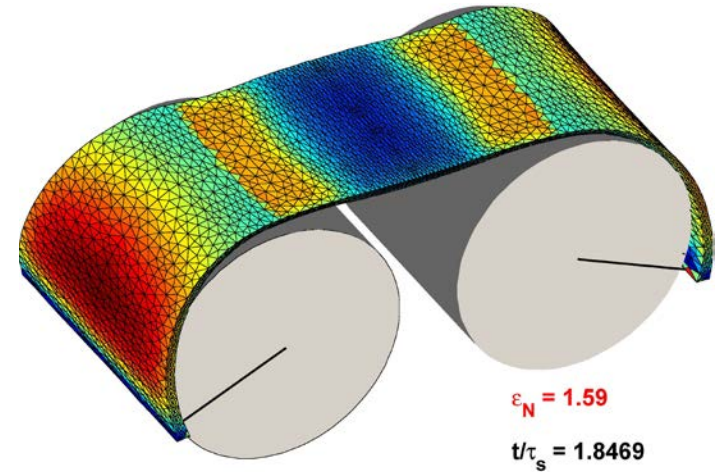
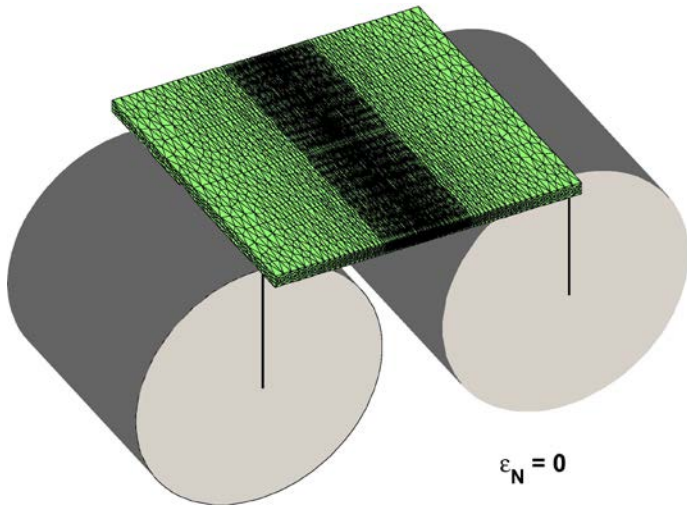


Wang, Boukani, Wang and Wang, PRL 99, 237801 (2007)

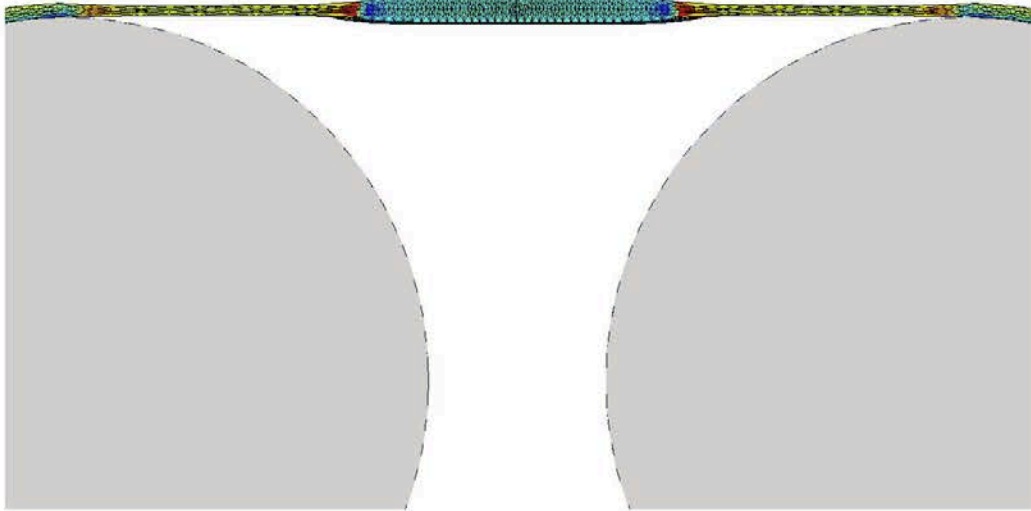
SER fixture coupled to an Anton Paar MCR 301 rotational rheometer.

2D planar simulation by Lyhne, Rasmussen and Hassager PRL 102, 138301 (2009)

3D simulation of inhomogeneous "stress reaxation" by Yu, Marin, Rasmussen and Hassager, J. Non-Newtonian Fluid Mech. (2010).



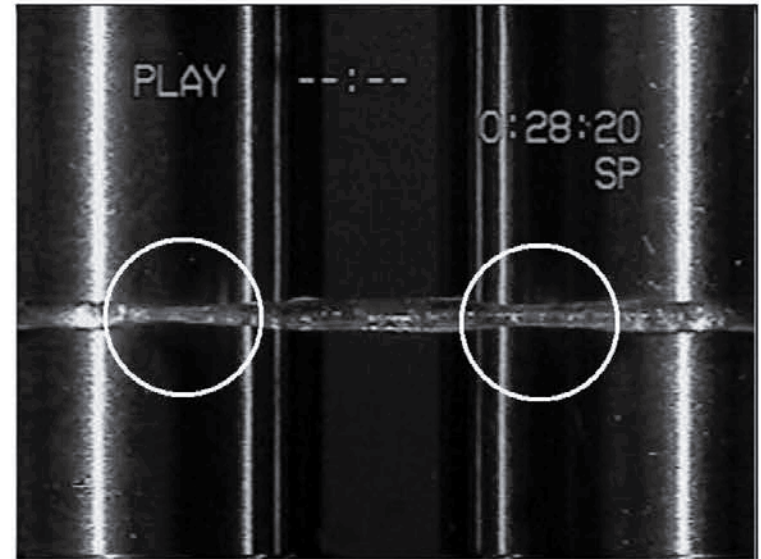
3D simulation of inhomogeneous "stress reaxation" by Yu, Marin, Rasmussen and Hassager, *J. Non-Newtonian Fluid Mech.* (2010).



Marin *et al.* simulations (2009)

3D Lagrangian Finite Element

$$H/W = 20$$

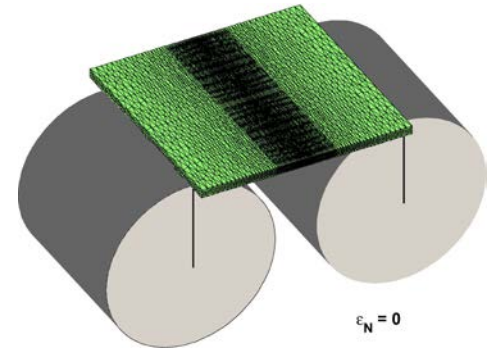
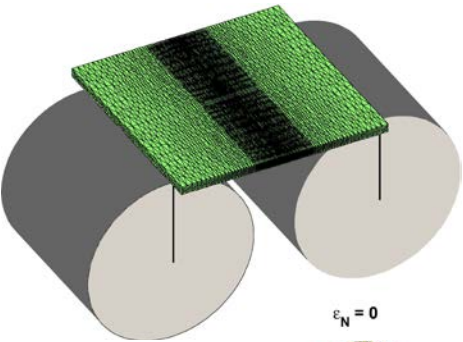


Wang *et al.* experiments

J. Rheol. (2008)

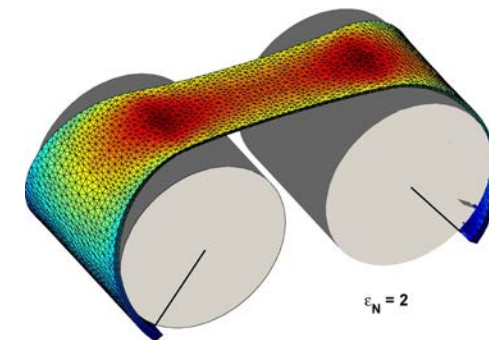
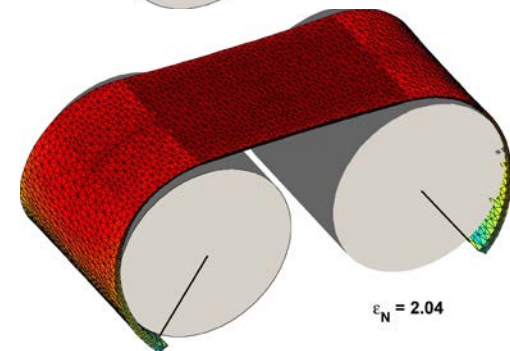
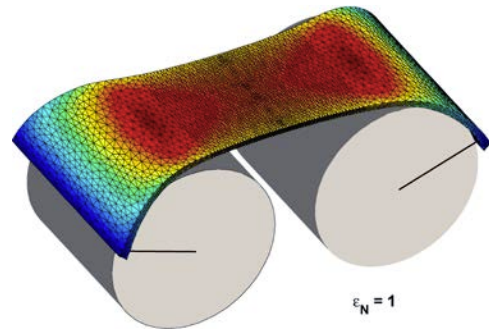
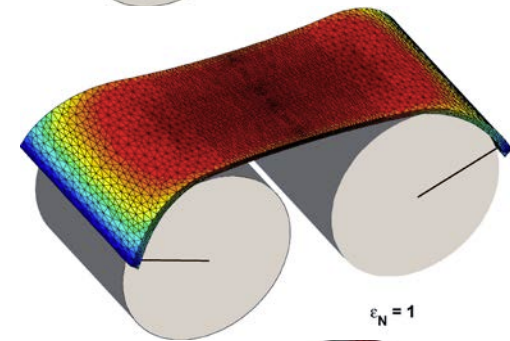
Yu, Marin, Rasmussen

Influence of non-linear material behavior on inhomogeneous deformation in dual-drum wind-up apparatus.



Left: neo-Hookean rubber material

Right: Doi-Edwards (independent alignment) tube model.



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The Danish Council for Independent Research

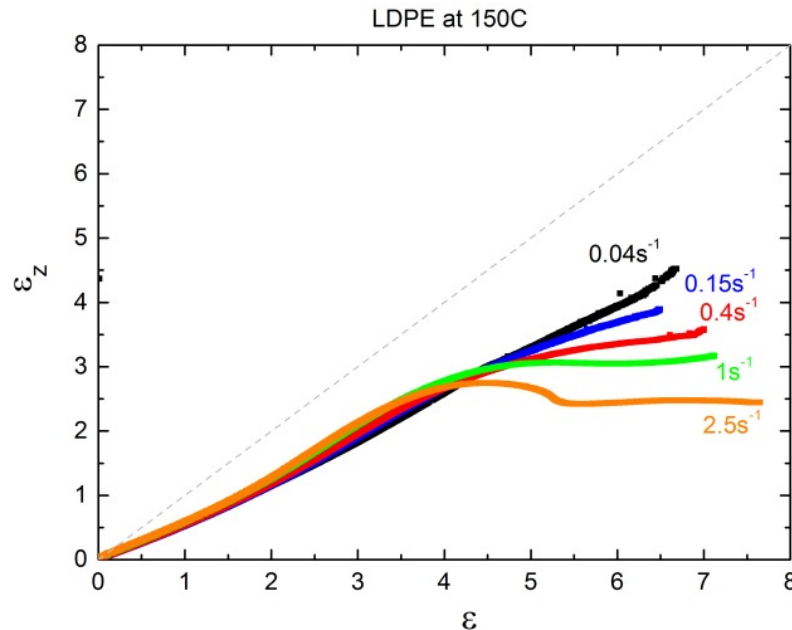
The Aage and Johanne Louis-Hansen Foundation

The European Science Foundation

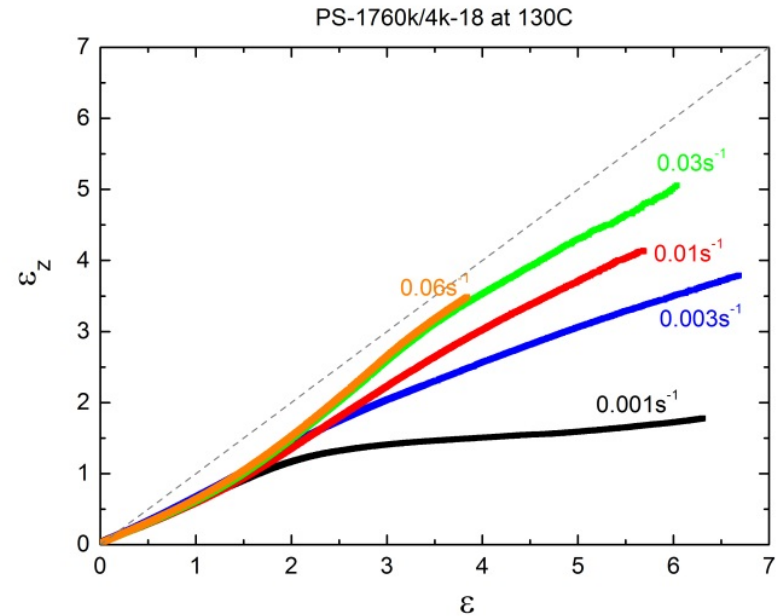
The Otto Mønsted Foundation

Nominal Hencky strain: $\varepsilon_z = \ln(L(t)/L_0)$

True Hencky strain: $\epsilon = 2 \ln(D_0/D(t))$



Nominal Hencky strain as function of true Hencky strain for constant rate experiment with LDPE.



Nominal Hencky strain as function of true Hencky strain for constant rate experiment with polystyrene solution.

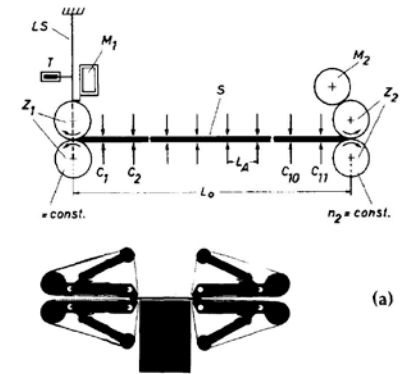
(Similar difference between Engineering stress vs. true stress.)

Historical: Extensional Rheometer Designs

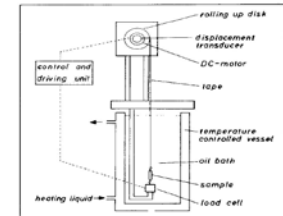
Meissner (1969), (1971), rotary clamp

$$\epsilon_{max} \approx 7$$

Meissner and Hostettler (1994), conveyor belt



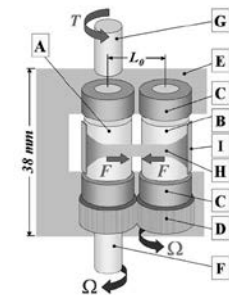
Münstedt and Laun (1979)



Sentmanat (2004): SER

$$\epsilon_{max} \approx 3.5$$

TA: EVF



$$\epsilon_N = \log(L/L_0)$$