Modeling and Simulation of Viscoelastic Flows in Polymer Processing with Integral Constitutive Equations

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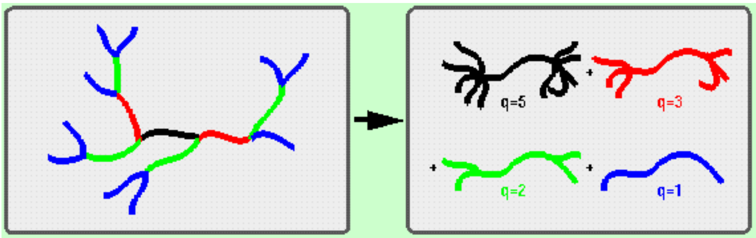
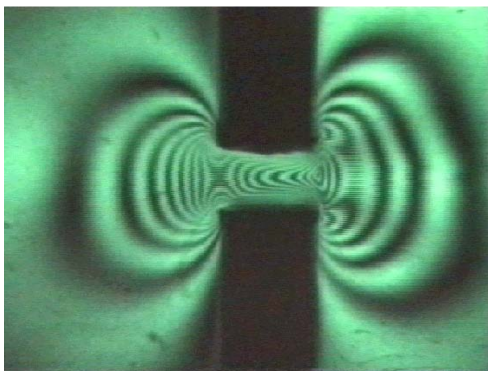
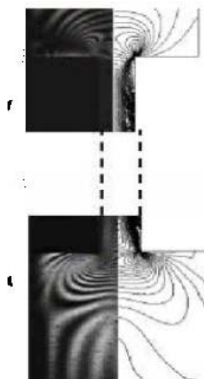
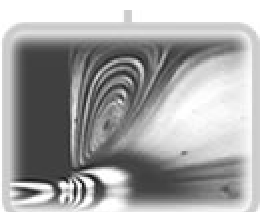
Molecular Structure

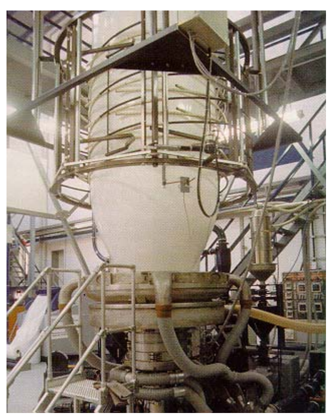


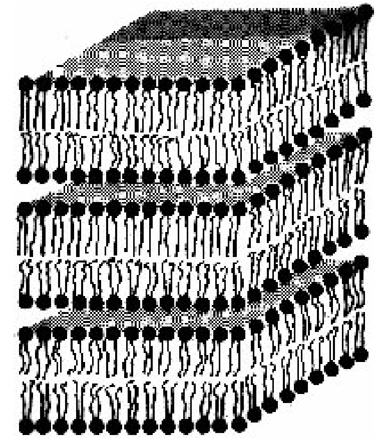
Processing











Product (Structure-Properties)

A. GOVERNING FLOW EQUATIONS (Conservation equations)

A1. Equation of conservation of mass

$$\nabla \cdot \overline{v} = 0$$

A2. Equation of conservation of momentum

$$\rho \overline{v} \cdot \nabla \overline{v} = -\nabla p + \nabla \cdot \overline{\overline{\tau}} + \rho \overline{g}$$

A3. Equation of conservation of energy

$$\rho C_p \overline{v} \cdot \nabla T = k \nabla^2 T + \overline{\overline{\tau}} : \nabla \overline{v}$$

****

B. RHEOLOGICAL MODEL (Constitutive equation)

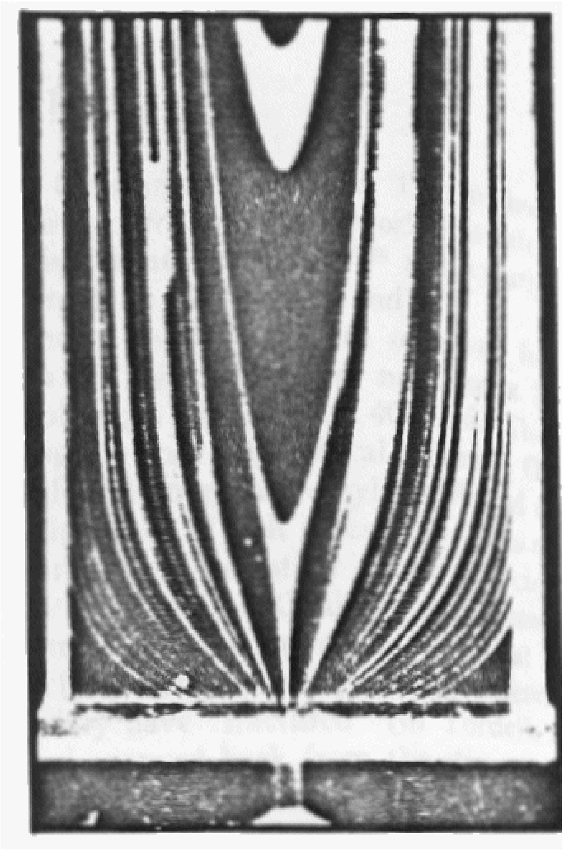
$$\overline{\overline{\tau}} = f(\overline{v}, \nabla \overline{v})$$

I Think It's STRESS

20:1 Axisymmetric Contraction

LDPE **HDPE**

τ_w = 0.1 MPa



Bagley and Birks, J. Appl. Phys., 1960

INTEGRAL CONSTITUTIVE EQUATIONS

$$\overline{\overline{\tau}}(t) = \int_{-\infty}^{t} \left[\mathrm{M}_1(\mathrm{t}-\mathrm{t}', \mathrm{I}_{\mathrm{C}^{-1}}, \mathrm{I}_{\mathrm{C}})\, \overline{\overline{C}}_t^{-1}(t') + \mathrm{M}_2(\mathrm{t}-\mathrm{t}', \mathrm{I}_{\mathrm{C}^{-1}}, \mathrm{I}_{\mathrm{C}})\, \overline{\overline{C}}_t(t') \right] dt'$$

where:

$M_1, M_2 = material - property \ \ functions$

$t = \ present \ time$

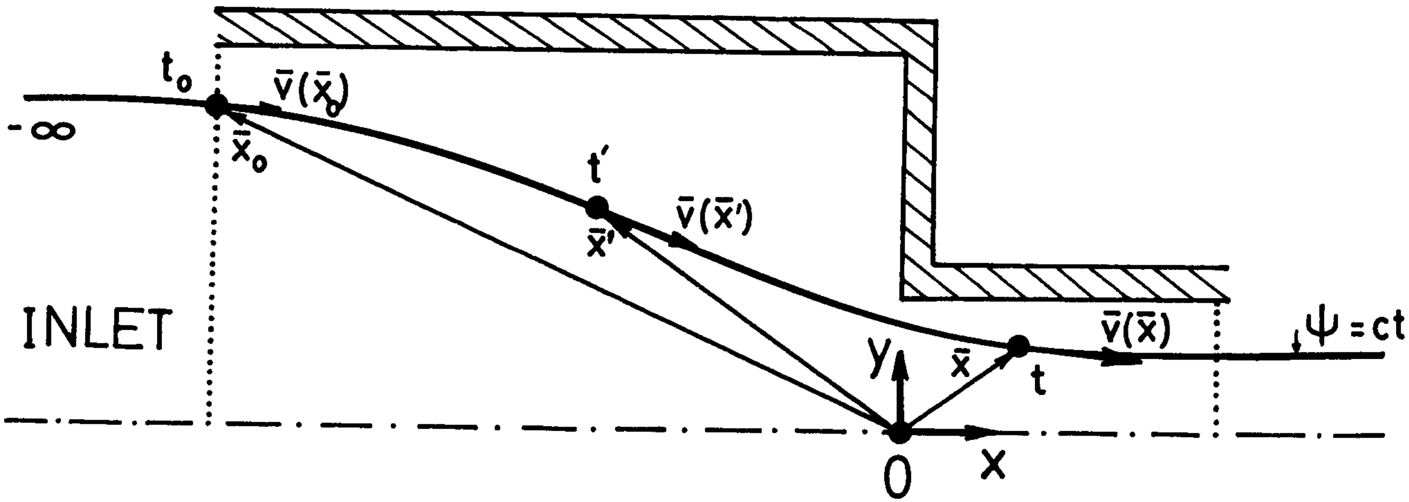
$t' = \ past \ time$

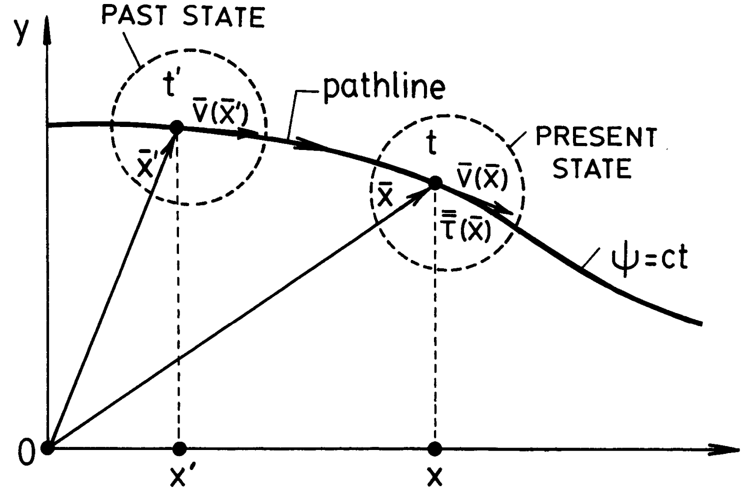
$I_{C^{-1}}, I_C = \ first \ invariants$

$\overline{\overline{C}}_t = Cauchy - Green \ tensor$

$\overline{\overline{C}}_t^{-1} = Finger \ strain \ \ tensor$

Particle Tracking





INTEGRAL CONSTITUTIVE EQUATIONS

K-BKZ EQUATION

- **Kaye (1962), Bernstein-Kearsley-Zapas (1963)**

$$\overline{\overline{\tau}}(t) = \int_{-\infty}^{t} \left[\frac{\partial U(t-t', I_{C^{-1}}, I_C)}{\partial I_{C^{-1}}} \overline{\overline{C}}_t^{-1}(t') - \frac{\partial U(t-t', I_{C^{-1}}, I_C)}{\partial I_C} \overline{\overline{C}}_t(t') \right] dt'$$

- **Factorized K-BKZ proposed by White and Tokita (1967)**

$$\overline{\overline{\tau}}(t) = \int_{-\infty}^{t} m(t-t') \left[\frac{\partial V(I_{C^{-1}}, I_C)}{\partial I_{C^{-1}}} \overline{\overline{C}}_t^{-1}(t') - \frac{\partial V(I_{C^{-1}}, I_C)}{\partial I_C} \overline{\overline{C}}_t(t') \right] dt'$$

where: $U = potential\ function$

$m(t-t') = time-dependent\ function$

$V = strain-dependent\ function$

$I_{C^{-1}}, I_C = first\ invariants$

$\overline{\overline{C}}_t = Cauchy-Green\ tensor$

$\overline{\overline{C}}_t^{-1} = Finger\ strain\ tensor$

The Faces behind the Names

Barry Bernstein, Elliot Kearsley and Louis Zapas at the Rochester Society of Rheology meeting in 1991.

From Rheology: An Historical Perspective, R.I. Tanner & K. Walters, Elsevier, 1998.

INTEGRAL CONSTITUTIVE EQUATIONS

RIVLIN-SAWYERS EQUATION

- **Rivlin and Sawyers (1971)**

$$\overline{\overline{\tau}}(t) = \int_{-\infty}^{t} \left[W_1(t-t', I_{C^{-1}}, I_C)\, \overline{\overline{C}}_t^{-1}(t') + W_2(t-t', I_{C^{-1}}, I_C)\, \overline{\overline{C}}_t(t') \right] dt'$$

- **Factorized R-S**

$$\overline{\overline{\tau}}(t) = \int_{-\infty}^{t} m(t-t') \left[H_1(I_{C^{-1}}, I_C)\, \overline{\overline{C}}_t^{-1}(t') + H_2(I_{C^{-1}}, I_C)\, \overline{\overline{C}}_t(t') \right] dt'$$

where:

$W = \text{not the potential function}$

$m(t-t') = \text{time-dependent function}$

$H = \text{strain-dependent function}$

$I_{C^{-1}}, I_C = \text{first invariants}$

$\overline{\overline{C}}_t = \text{Cauchy-Green tensor}$

$\overline{\overline{C}}_t^{-1} = \text{Finger strain tensor}$

INTEGRAL CONSTITUTIVE EQUATIONS

Special Cases of the Factorized

RIVLIN-SAWYERS EQUATION

- **Wagner Model (1980)**

$$M = \left[\sum_k \frac{a_k}{\lambda_k} \exp\left(-\frac{t-t'}{\lambda_k}\right) \right], \qquad H = \exp\left(-\beta \sqrt{\alpha I_{C^{-1}} + (1-\alpha) I_C - 3} \right)$$

- **Papanastasiou-Scriven-Macosko (PSM) Model (1983)**

$$M = \left[\sum_k \frac{a_k}{\lambda_k} \exp\left(-\frac{t-t'}{\lambda_k}\right) \right], \qquad H = \left[\frac{\alpha}{(\alpha - 3) + \beta I_{C^{-1}} + (1-\beta) I_C} \right]$$

where:

λ_k = *relaxation time*

a_k = *relaxation modulus*

α, β = *material constants*

$I_{C^{-1}}, I_C$ = *first invariants*

$\overline{\overline{C}}_t$ = *Cauchy – Green tensor*

$\overline{\overline{C}}_t^{-1}$ = *Finger strain tensor*

Relevant Literature

- **Kaye**, A. (1962) Non-Newtonian Flow in Incompressible Fluids, Part I: A General Rheological Equation of State; Part II: Some Problems in Steady Flow, Note No. 134, College of Aeronautics, Cranford, UK.
- **Bernstein**, B., **Kearsley**, E.A., and **Zapas**, L. (1963) A Study of Stress Relaxations with Finite Strain, *Trans. Soc. Rheol.*, **7**, 391-410.
- **Papanastasiou**, A.C., **Scriven**, L.E., and **Macosko**, C.W. (1983) An Integral Constitutive Equation for Mixed Flows: Viscoelastic Characterization, *J. Rheol.* **27**, 387-410.
- **Olley**, P. (2000) An adaptation of the separable KBKZ equation for comparable response in planar and axisymmetric flow, *J. Non-Newtonian Fluid Mech.*, **95**, 35-53.

Numerical Methods (2-D)

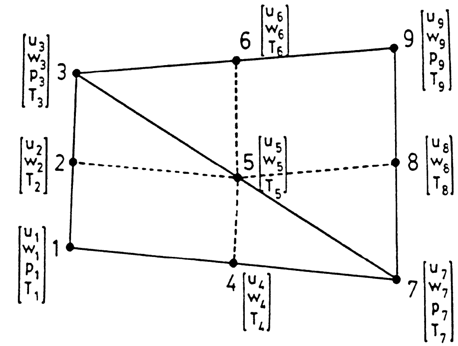
- ✦ **u-v-p formulation (primitive variables)**
- ✦ **u-v-p formulation (SI)**
- ✦ **mixed formulation (u-v-p-τ) (EVSS)**
- ✦ **enhanced mixed formulation (u-v-p-τ-g) (DEVSS-G)**
- ✦ **many others (EEME, SI, EVSS/SUPG, EVSS/SU, AVSS/SI, AVSS/SUPG, etc.)**

Numerical Methods (3-D)

- 3-D x-y-z-p-t formulation (primitive variables)
- Lagrangian Integral Method (LIM)
- H.K. Rasmussen, “Time-dependent finite-element method for the simulation of three-dimensional viscoelastic flow with integral models”, JNNFM, 84, 217-232 (1999).

Relative Difficulty of Pom-Pom vs. K-BKZ Models

(or differential vs. integral models)



2-D dof / element

K-BKZ	**Pom-pom**
8 u (serendipity)	**9 u (Lagrangian)**
8 v	**9 v**
4 p	**4 p**
8 T	**9 T**
20 (28)	22 (31)

planar **4 (τ_{xx}, τ_{yy}, τ_{xy}) = 12/mode x 8 = 96**

4 (g_{xx}, g_{yy}, g_{xy})= 12/mode x 8 = 96

46 (55) 214 (223)

axisymmetric **4 (τ_{rr}, τ_{zz}, τ_{rz}, $\tau_{\theta\theta}$) = 16/mode x 8 = 128**

4 (g_{rr}, g_{zz}, g_{rz}, $g_{\theta\theta}$)= 16/mode x 8 = 128

54 (63) 278 (287)

dof/elem/8 modes

Parameters for Fitting Data for the IUPAC-LDPE Melt

Pom-Pom

k	λ_k (s)	a_k (Pa)	q_k	$(\tau_b/\tau_s)_k$
1	0.0001	1.29×10^5	1	4.5
2	0.001	9.48×10^4	1	3.9
3	0.01	5.86×10^4	2	3.7
4	0.1	2.67×10^4	2	3.6
5	1.0	9.80×10^3	3	2.9
6	10.0	1.89×10^3	7	2.0
7	100.0	1.80×10^2	8	2.1
8	1000.0	1.00×10^0	9	1.3

Inkson, McLeish, Harlen, Groves, J. Rheol. (1999)

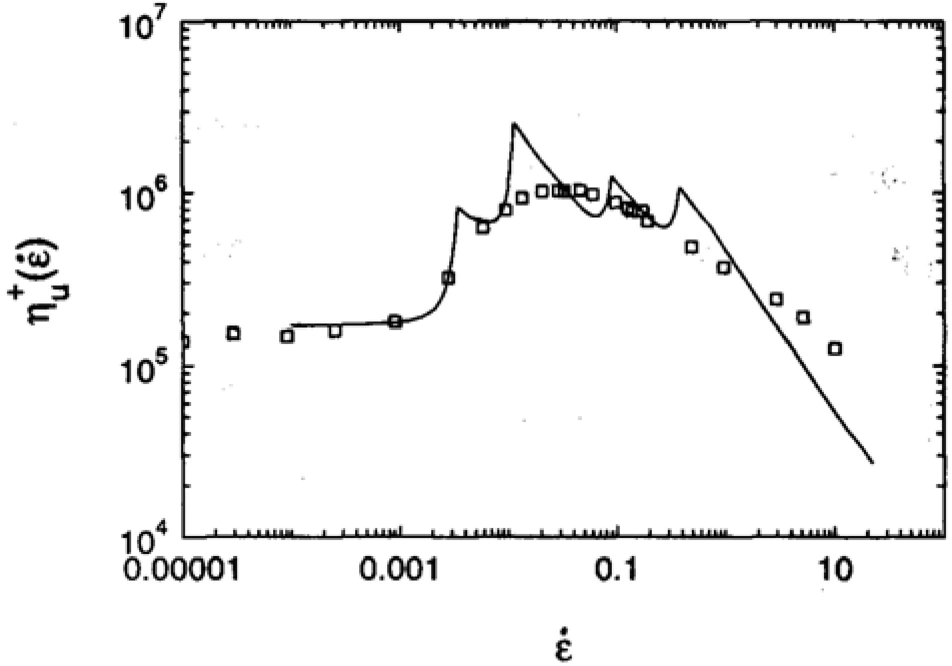
K-BKZ (PSM)

k	λ_k (s)	a_k (Pa)	α_k	β_k
1	0.0001	1.29×10^5	14.38	0.018
2	0.001	9.48×10^4	14.38	0.018
3	0.01	5.86×10^4	14.38	0.08
4	0.1	2.67×10^4	14.38	0.12
5	1.0	9.80×10^3	14.38	0.12
6	10.0	1.89×10^3	14.38	0.16
7	100.0	1.80×10^2	14.38	0.03
8	1000.0	1.00×10^0	14.38	0.002

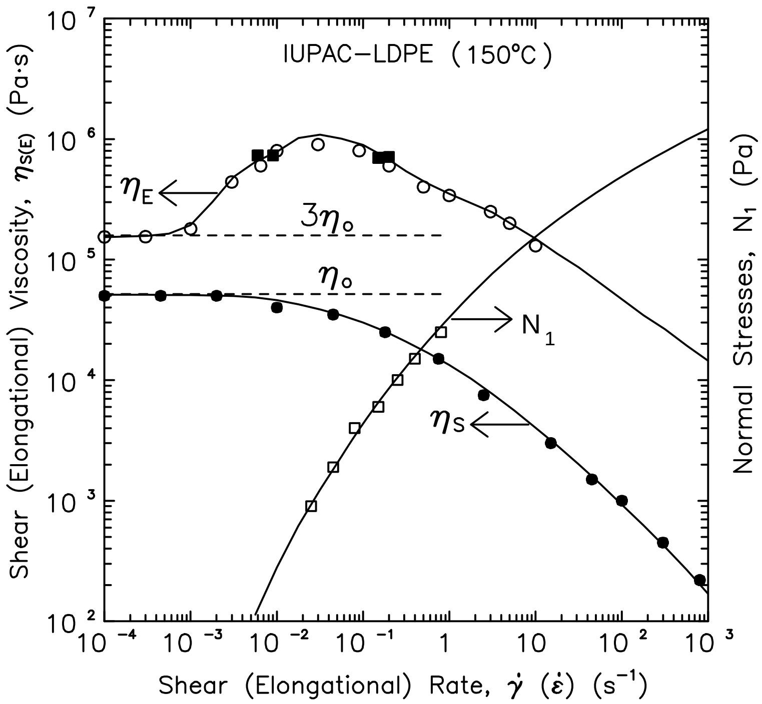
Barakos and Mitsoulis, J. Rheol. (1995)

Elongational Viscosity

Pom-Pom



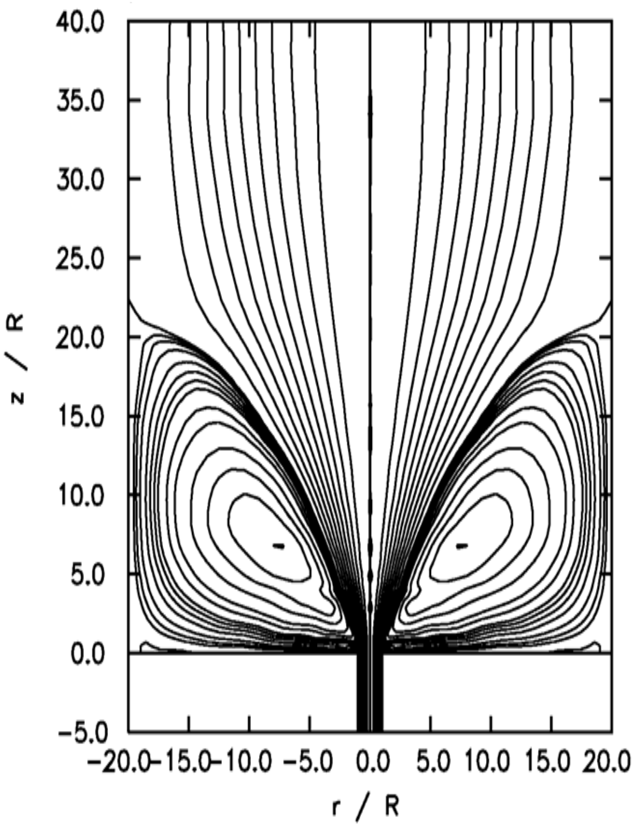
K-BKZ (PSM)



LDPE Melts

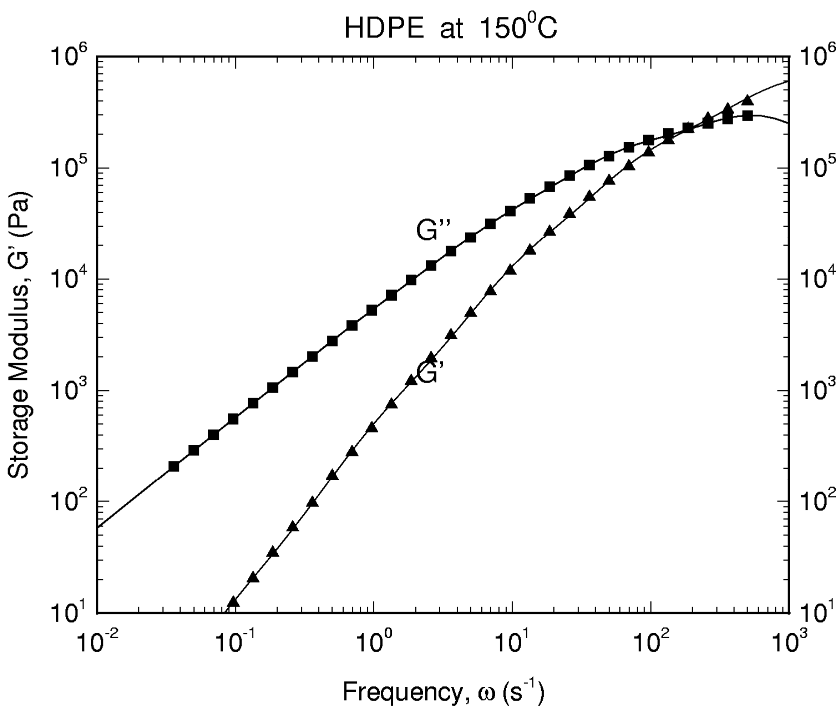
Simulations | Experiments

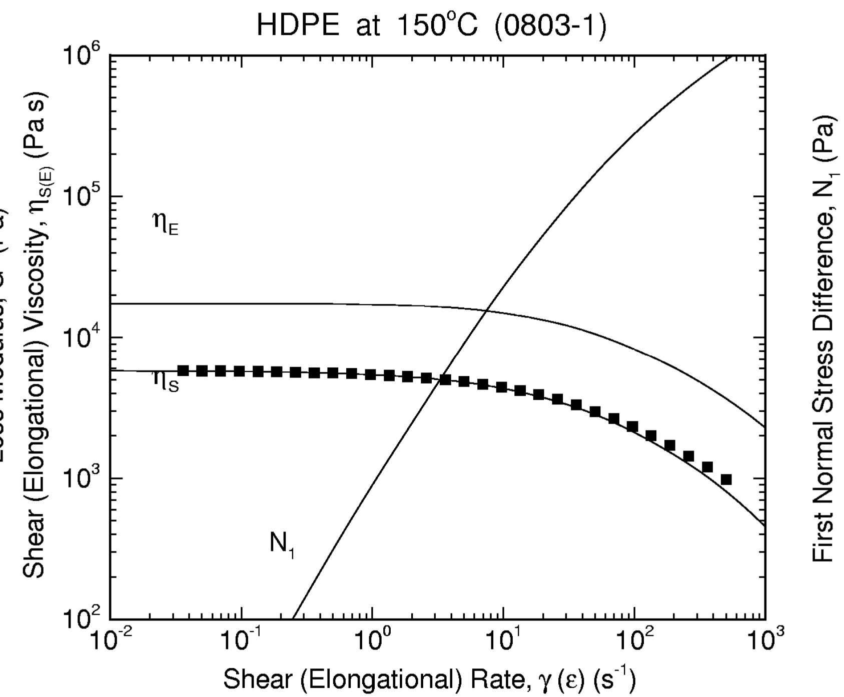
$\gamma_\alpha = 84\ s^{-1},\ \tau_w = 0.1\ MPa$



HDPE Melt (0803-1 at 150°C)

K-BKZ Model Original Fit





- A non-linear regression analysis is performed on the K-BKZ integral constitutive equation to determine the parameters in the model.

HDPE Melt (0803-1 at 150°C)
K-BKZ Model Parameters

Original Fit

k	λ_k (s)	a_k (Pa)	α_k	β_k
1	0.001723	5.46×10^5	4.0	0.9985
2	0.01299	1.73×10^5	4.0	0.9985
3	0.08644	2.15×10^4	4.0	0.9985
4	0.7991	7.69×10^2	4.0	0.9985
5	9.398	1.39×10^1	4.0	0.9985

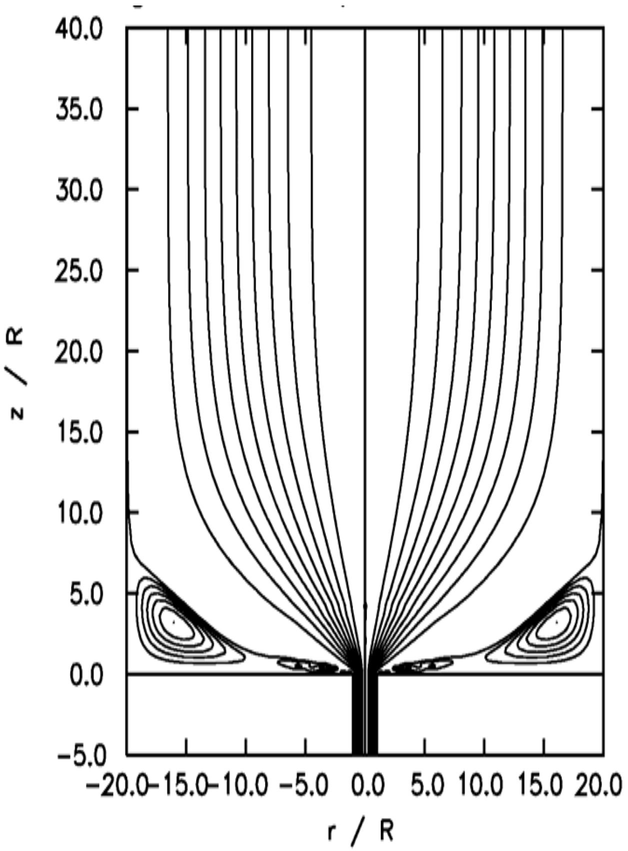
$\theta = 0$

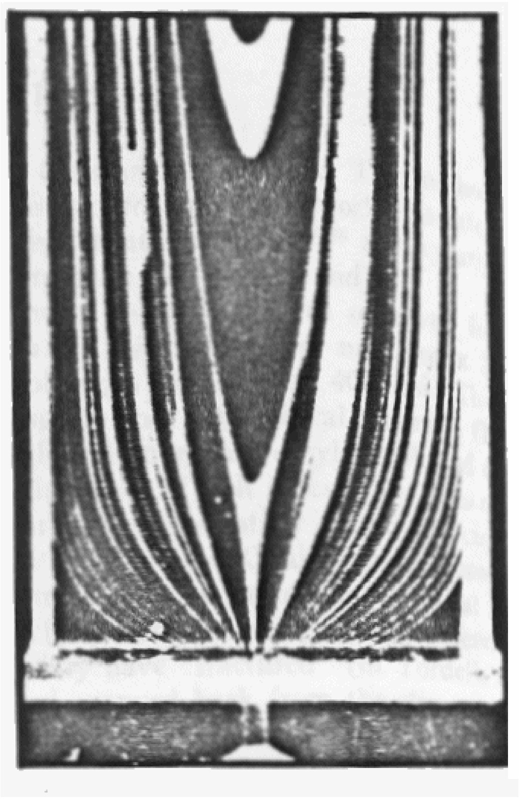
P. Wood-Adams, PhD Thesis, McGill University, 1999.

HDPE Melts

Simulations **Experiments**

γ_α = 248 s^{-1}, τ_w = 0.1 MPa





NON-ISOTHERMAL CONSTITUTIVE EQUATION

TIME-TEMPERATURE SHIFTING CONCEPT

$$\overline{\overline{\tau}}(t) = \frac{1}{1-\theta} \int_{-\infty}^{\xi(t)} \mathrm{M}\,\mathrm{H}\left[C_{t(\xi)}^{-1}(t'(\xi')) + \theta C_{t(\xi)}(t'(\xi')) \right] d\xi'$$

$$M = \left[\sum_k \frac{a_k}{\lambda_k} \exp\left(-\frac{\xi-\xi'}{\lambda_k}\right) \right], \qquad H = \left[\frac{\alpha}{(\alpha-3) + \beta I_{\mathbf{C}^{-1}} + (1-\beta) I_{\mathbf{C}}} \right]$$

where:

λ_k = *relaxation time*

a_k = *relaxation modulus*

α, β, θ = *material constants*

$I_{C^{-1}}, I_C$ = *first invariants*

$\overline{C_t}$ = *Cauchy – Green tensor*

$\overline{C_t}^{-1}$ = *Finger strain tensor*

Note: $\frac{N_2}{N_1} = \frac{\theta}{1-\theta}$

Non-Isothermal Analysis

- **Peclet Number:** $$Pe = \frac{\rho C_p \overline{V} R}{k}$$

- **Nahme Number:** $$Na = \frac{a\,\eta_o \overline{V}^2}{k}$$

where $$a(T) = \frac{\eta}{\eta_o} = \exp\left[-\frac{E_o}{R_g}\left(\frac{1}{T} - \frac{1}{T_o}\right)\right]$$

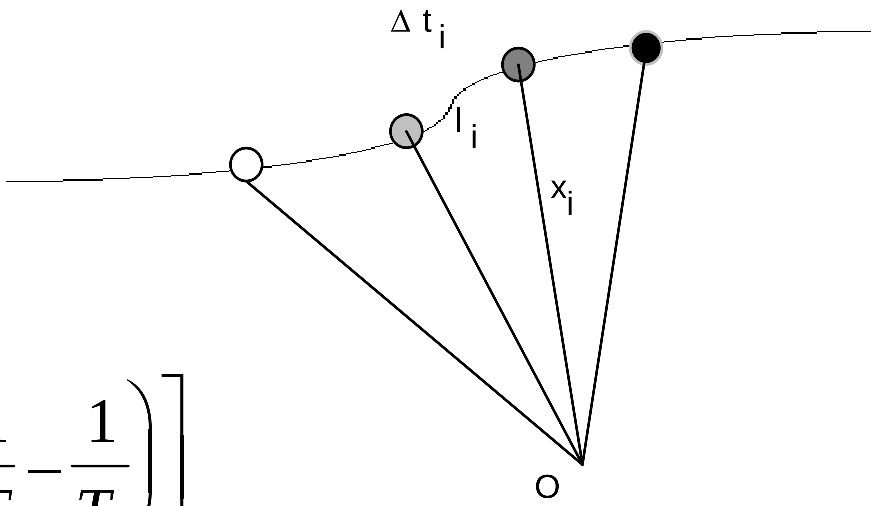
or $$a(T_o) = \frac{E_o}{R_g T_o^2}$$

Pseudo-Time Integral Method

$$d\xi = \frac{dt'}{a\left(T(t')\right)}$$

$$dt' = \frac{dl}{dV}$$

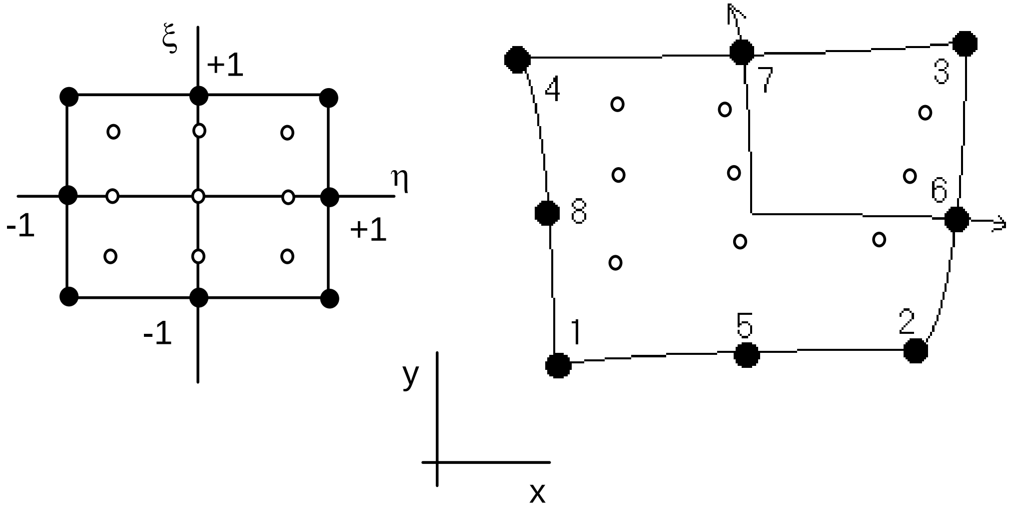
$$a(T) = \frac{\eta}{\eta_0} = \exp\left[-\frac{E_0}{R_0}\left(\frac{1}{T} - \frac{1}{T_0}\right)\right]$$



t': observer's time

ξ: particle's time

‘UPWIND’ FEM



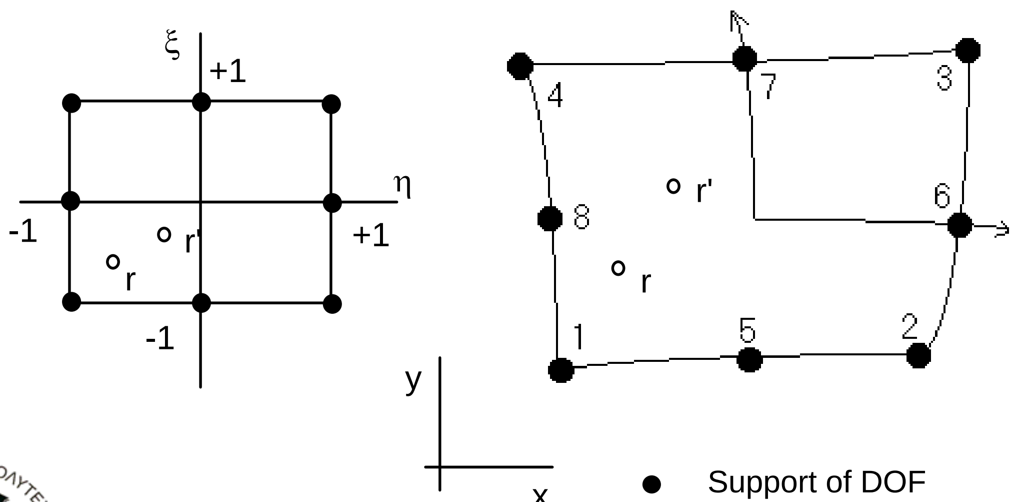
$$r' - r = a\,\frac{h}{2}$$

$$a = \coth\left(\frac{Pe}{2}\right)$$

$$Pe = \frac{|u|\,h}{k}$$

h=length of streamline

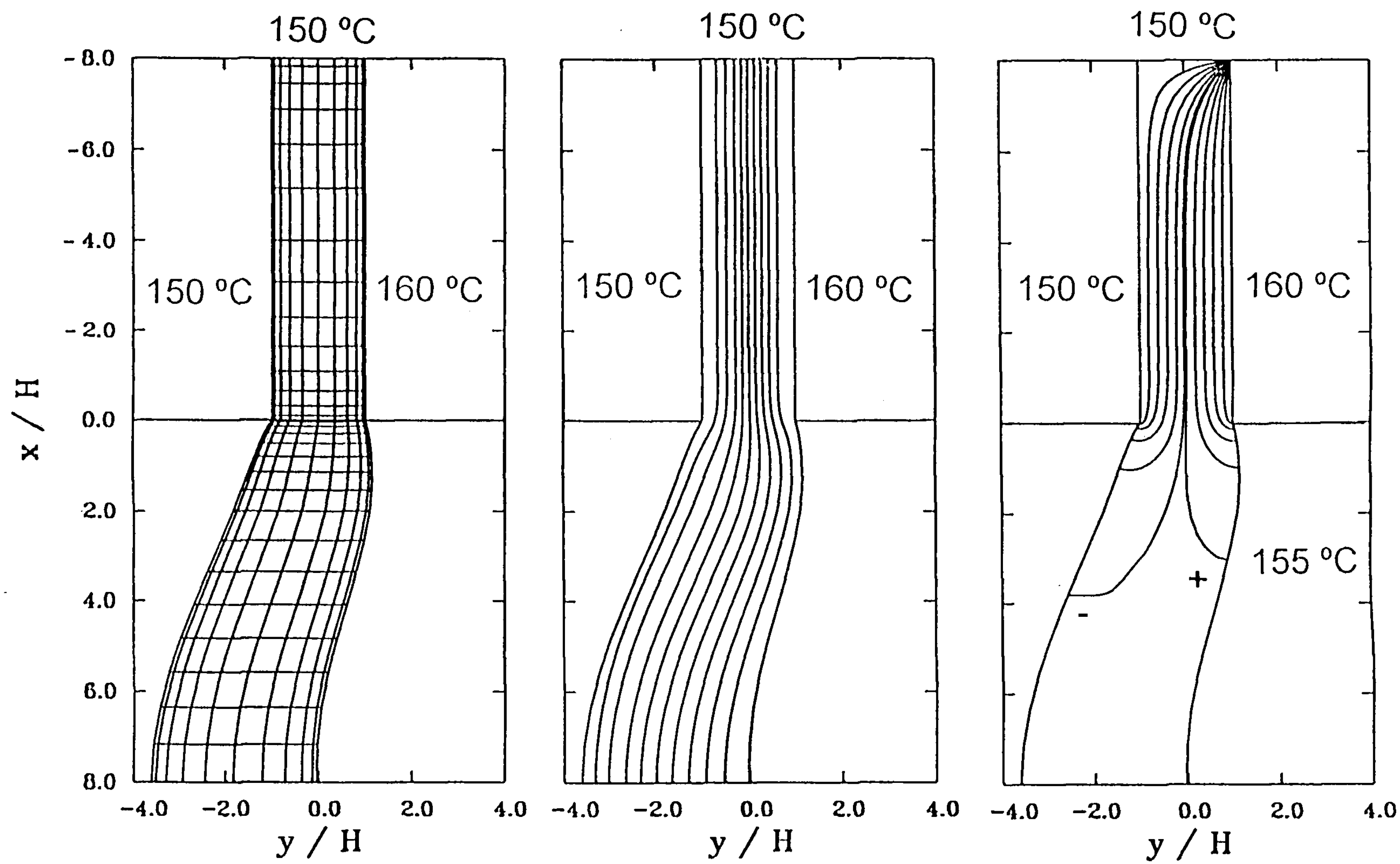
k=diffusion coefficient



● Support of DOF

○ Support of Integration Node

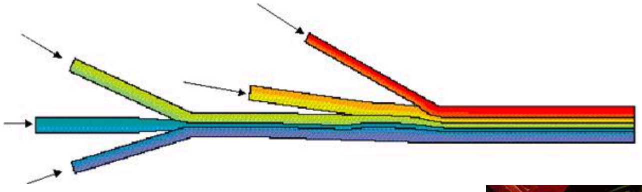
Extrudate Swell and Bending Phenomenon

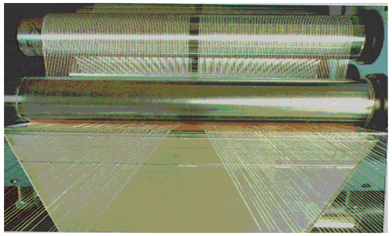


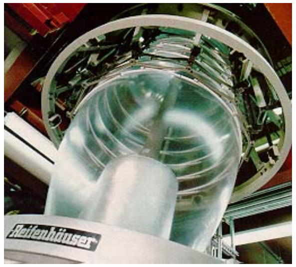
Polymer Processes

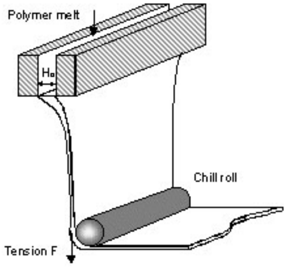
Polymer processes where the K-BKZ model has been used:

- Coextrusion
- Fiber Spinning
- Film Blowing
- Film Casting
- Blow Molding



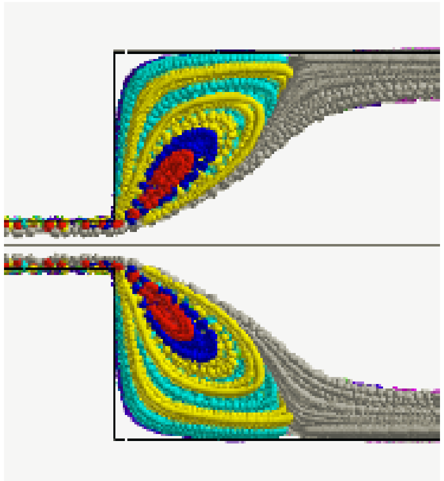


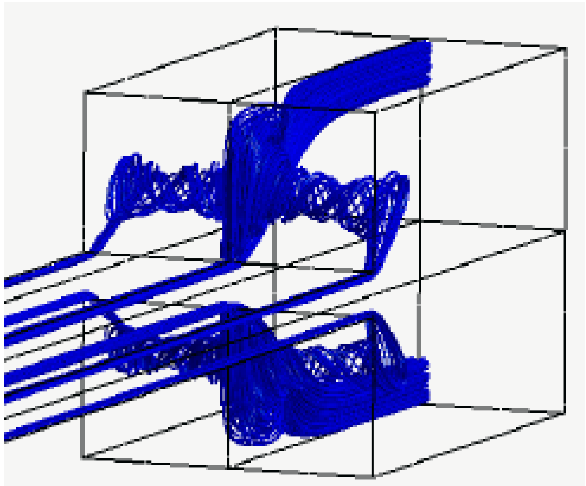


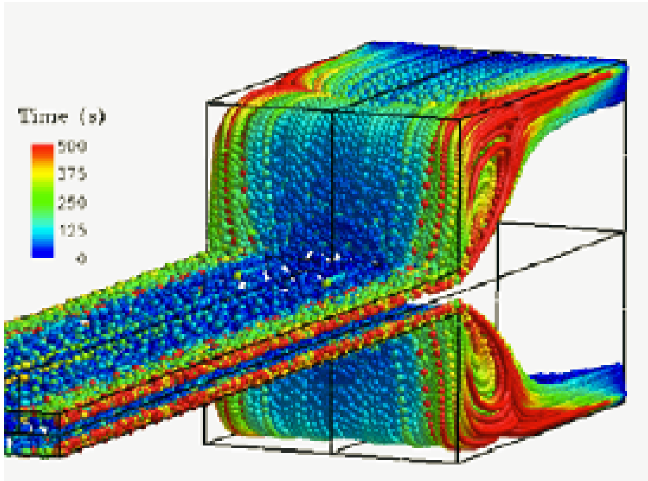


New Developments
3-D Simulations

Pom-Pom vs K-BKZ







From Sirakov (2006)

CONCLUSIONS

- Computational rheology has made good progress in the last 15 years due to rapid growth in computing power.
- From the modelling point of view, progress has been achieved mainly by using integral constitutive equations of the K-BKZ type, which are quite capable of fitting rheological data for polymer solutions and melts in simple flows.
- In the processing of non-Newtonian, complex materials, many unsolved problems still remain; however there are now available appropriate means of tackling them, so that the analysis and design of several processes becomes feasible.

FURTHER WORK

- The subject of viscoelasticity requires further study for more appropriate *damping functions* for fast deformations and better (faster) computational methods.
- Time-dependent flows
- Multiple-layer flows
- Three-Dimensional flows