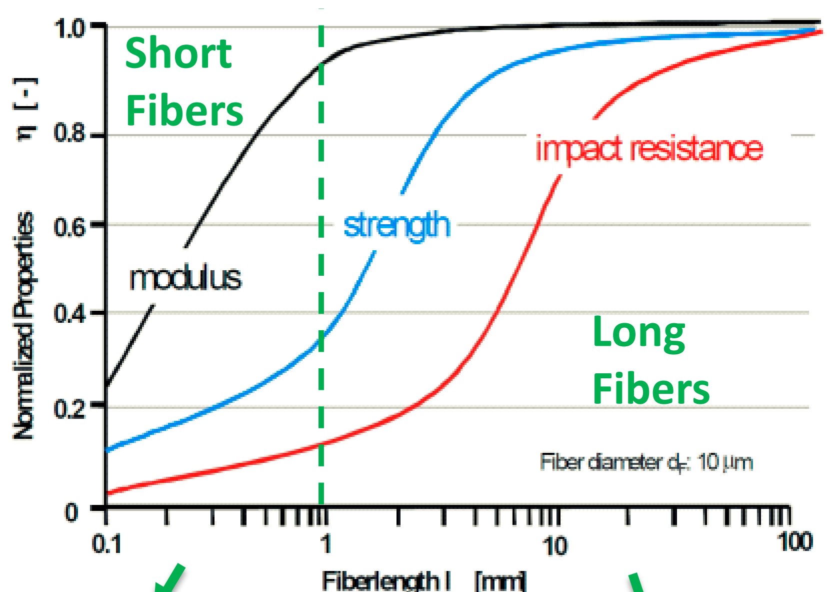
Role of Flow Type on the Evolution of Semi-Flexible Fiber Orientation

Flow Processes in Composite Materials 15th Conference

Hongyu Chen, Greg Lambert, Peter Wapperom*, and Donald G. Baird
Department of Chemical Engineering and the Department of Mathematics*, Virginia Polytechnic Institute and State University, Blacksburg, VA 24061-0201, USA and
Macromolecular Innovation Institute,

Short and Long Glass Fiber Composites



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$$f^{eff} = \frac{64\eta_m \dot{\gamma} a_r^{\,4}}{E_Y \pi}$$

$a_r < 100$

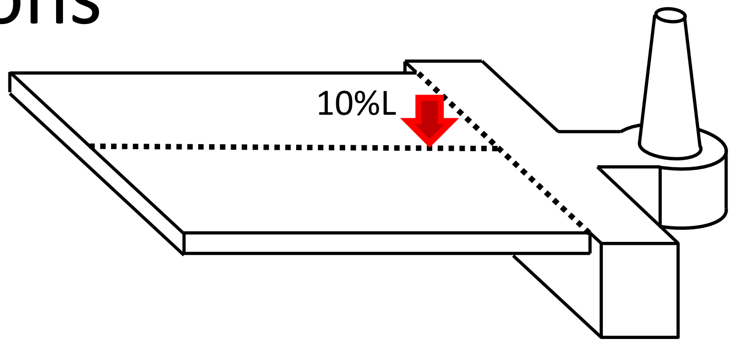
$a_r > 100$
(lengths > 1mm)

Plasticomp

Switzer and Klingenberg, 2003

Injection Molding Observations

50 wt% Carbon Fiber – Nylon 6,6



10% plaque length

Objectives

1. Develop a long-fiber orientation model for semi-flexible fibers and in which the strain reduction parameter depends on the flow type.
2. Initiate the development of a rheological test that incorporates both shear and extensional flow (non-lubricated squeeze flow, NLSF).
3. Verify that NLSF can be used to obtain orientation model parameters through fitting to the measured fiber orientation.
4. Test the model through the simulation of orientation in a basic processing flow, center-gated disc.

Background: Rheology

$$\sigma = -P\boldsymbol{I} + 2\eta_m\left[(1+2\varphi)\boldsymbol{D} + N_p \boldsymbol{D}:\boldsymbol{A}_4\right]$$ **Semi-Concentrated**

$$\sigma = -P\boldsymbol{I} + 2\eta_m \boldsymbol{D} + N_p \boldsymbol{D}:\boldsymbol{A}_4$$ **Dilute**

$$N_p = \left(\frac{2}{3}\right)\left(\frac{\varphi}{\ln(\pi/\varphi)}\right) a_r^2$$

$$N_p = \left(\frac{1}{2}\right)\left(\frac{a_r^2}{\ln a_r}\right)\varphi$$

$$N_p = \left(\frac{1}{3}\right)\left[C' + \ln(1/\varphi) - \ln(\ln(1/\varphi))\right]^{-1} a_r^2$$

Lipscomb Ii, G. G., Denn, M. M., Hur, D. U., & Boger, D. V. (1988). The flow of fiber suspensions in complex geometries. *Journal of Non-Newtonian Fluid Mechanics, 26*(3), 297-325. doi: http://dx.doi.org/10.1016/0377-0257(88)80023-5

Dinh, S. M., & Armstrong, R. C. (1984). A rheological equation of state for semiconcentrated fiber suspensions. *Journal of Rheology, 28*(3), 207-227. doi: 10.1122/1.549748

Shaqfeh, E. S. G., & Fredrickson, G. H. (1990). The hydrodynamic stress in a suspension of rods. *Physics of Fluids A: Fluid Dynamics (1989-1993), 2*(1), 7-24. doi: doi:http://dx.doi.org/10.1063/1.857683

Background: Orientation Models

$$\frac{DA}{Dt} = \alpha\Big(\big(\boldsymbol{W}\cdot\boldsymbol{A} - \boldsymbol{A}\cdot\boldsymbol{W}\big) + \xi\big(\boldsymbol{D}\cdot\boldsymbol{A} + \boldsymbol{A}\cdot\boldsymbol{D} - 2\boldsymbol{D}:\boldsymbol{A}_4\big) + 2C_I\dot{\gamma}\big(\boldsymbol{I} - 3\boldsymbol{A}\big)\Big)$$

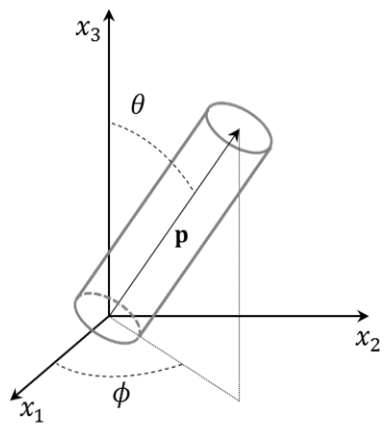
Matrix Deformation

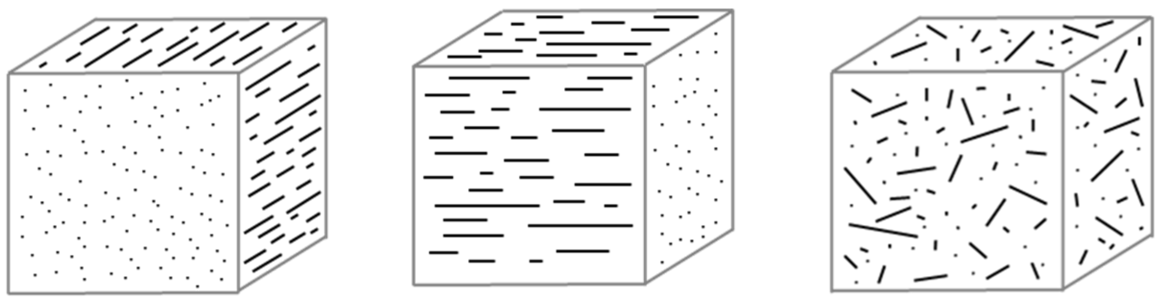
Isotropic Rotary Diffusion

Empirical Parameters	
α	Slows fiber motion
C_I	Fiber interactions

Folgar, F. and C.L. Tucker III, *Orientation behavior of fibers in concentrated suspensions.* Journal of Reinforced Plastics and Composites, 1984. 3(2): p. 98-119.
Huynh, H.M., *Improved Fiber Orientation Predictions for Injection-Molded Composites.* 2001, University of Illinois at Urbana-Champaign.

Background: Orientation Models





$$\mathbf{A} = \begin{pmatrix} 1 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix} \qquad \mathbf{A} = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 0 \end{pmatrix} \qquad \mathbf{A} = \begin{pmatrix} 1/3 & 0 & 0 \\ 0 & 1/3 & 0 \\ 0 & 0 & 1/3 \end{pmatrix}$$

$$\mathbf{A} = \int \mathbf{pp}\psi(\mathbf{p}, t)d\mathbf{p}$$

$$\mathbf{A_4} = \int \mathbf{pppp}\psi(\mathbf{p}, t)d\mathbf{p}$$

Advani, S.G. and C.L. Tucker III, *The Use of Tensors To Describe and Predict Fiber Orientation in Short Fiber Composites.* Journal of Rheology, 1987. **31**(8).

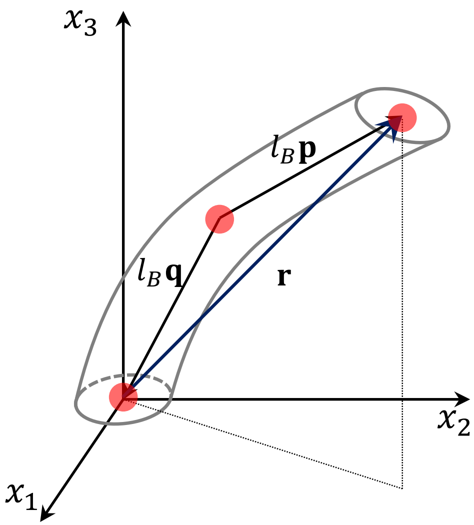
Semi-Flexible Fibers

$$\frac{D\mathbf{A}}{Dt} = \alpha\left[\mathbf{W}\cdot\mathbf{A} - \mathbf{A}\cdot\mathbf{W} + \xi\left(\mathbf{D}\cdot\mathbf{A} + \mathbf{A}\cdot\mathbf{D} - 2\mathbf{D}:\mathbf{A}_4\right) + 2C_I\dot{\gamma}\left(\mathbf{I} - 3\mathbf{A}\right) + \frac{l_B}{2}\left[\mathbf{Cm} + \mathbf{mC} - 2\left(\mathbf{m}\cdot\mathbf{C}\right)\mathbf{A}\right] + 2k\left(\mathbf{B} - \mathbf{A}\,tr\left(\mathbf{B}\right)\right)\right]$$

$$\frac{D\mathbf{B}}{Dt} = \alpha\left[\mathbf{W}\cdot\mathbf{B} - \mathbf{B}\cdot\mathbf{W} + \xi\left(\mathbf{D}\cdot\mathbf{B} + \mathbf{B}\cdot\mathbf{D} - \left(2\mathbf{D}:\mathbf{A}\right)\mathbf{B}\right) - 4C_I\dot{\gamma}\mathbf{B} + \frac{l_B}{2}\left[\mathbf{Cm} + \mathbf{mC} - 2\left(\mathbf{m}\cdot\mathbf{C}\right)\mathbf{B}\right] + 2k\left(\mathbf{A} - \mathbf{B}\,tr\left(\mathbf{B}\right)\right)\right]$$

$$\frac{D\mathbf{C}}{Dt} = \alpha\left[\nabla\mathbf{v}^t\cdot\mathbf{C} - \left(\mathbf{A}:\nabla\mathbf{v}^t\right)\mathbf{C} - 2C_I\dot{\gamma}\mathbf{C} + \frac{l_B}{2}\left[\mathbf{m} - \mathbf{C}\left(\mathbf{m}\cdot\mathbf{C}\right)\right] - k\mathbf{C}\left(1 - tr\left(\mathbf{B}\right)\right)\right]$$

Hydrodynamic | IRD | Bending From Flow | Bending Potential



$$\mathbf{m} = \sum_{i=1}^{3}\sum_{j=1}^{3}\sum_{k=1}^{3}\frac{\partial^2 v_i}{\partial x_j \partial x_k}A_{jk}\mathbf{e}_i$$

$$\mathbf{A} = \iint \mathbf{pp}\,\psi(\mathbf{p},\mathbf{q},t)\,d\mathbf{p}\,d\mathbf{q}$$

$$\mathbf{B} = \iint \mathbf{pq}\,\psi(\mathbf{p},\mathbf{q},t)\,d\mathbf{p}\,d\mathbf{q}$$

$$\mathbf{C} = \iint \mathbf{p}\,\psi(\mathbf{p},\mathbf{q},t)\,d\mathbf{p}\,d\mathbf{q}$$

$$\mathbf{A}_4 = \iint \mathbf{pppp}\,\psi(\mathbf{p},\mathbf{q},t)\,d\mathbf{p}\,d\mathbf{q}$$

$$\mathbf{r} = l_B\left(\mathbf{p} - \mathbf{q}\right)$$

$$\mathbf{R} = \frac{\langle\mathbf{rr}\rangle}{tr\left(\mathbf{rr}\right)} = \frac{\mathbf{A} - \mathbf{B}}{1 - tr\left(\mathbf{B}\right)}$$

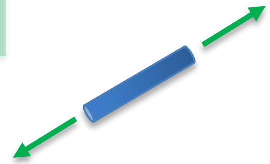
Strautins and Latz, 2007
Ortman et al., 2012

Coupling Orientation to Flow

Stress Equation for Rigid Fibers:

$$\boldsymbol{\sigma} = -P\mathbf{I} + 2\eta_m\mathbf{D} + 2\eta_m\phi(\,\mu_1\mathbf{D} + \mu_2\mathbf{D} : \mathbf{A_4})$$

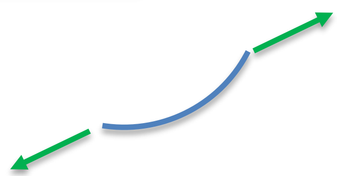
Matrix — Fibers

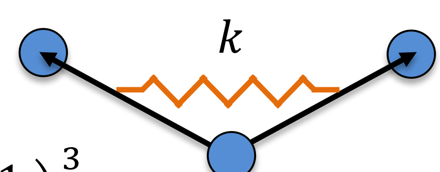


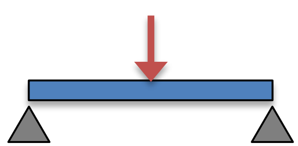
Proposed Stress Equation for Semi-Flexible Fibers:

$$\boldsymbol{\sigma} = -P\mathbf{I} + 2\eta_m\mathbf{D} + 2\eta_m\phi(\,\mu_1\mathbf{D} + \mu_2\mathbf{D} : \mathbf{R_4}) + \eta_m k\frac{3\phi a_r}{2}(\mathbf{B} - \mathbf{A}tr\mathbf{B})$$

Matrix — Fibers — Fiber Bending



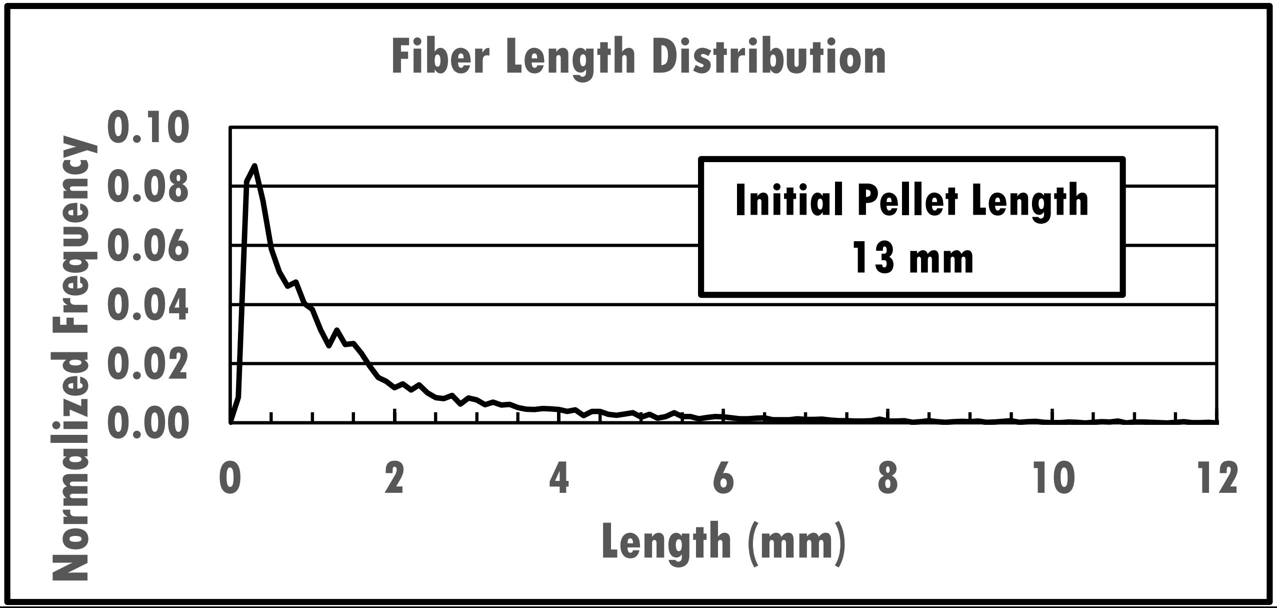




$$k = \frac{E_y}{8\eta_m}\left(\frac{1}{a_r}\right)^3$$

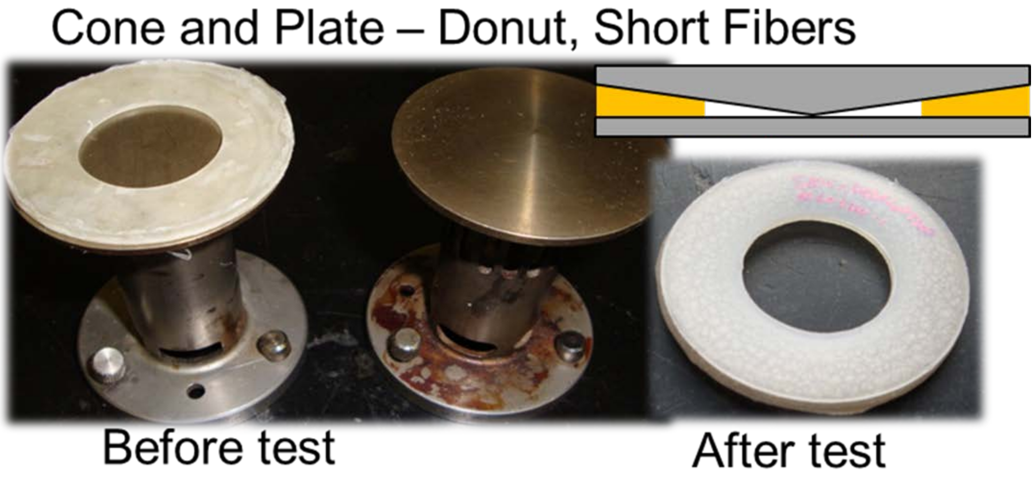
Lipscomb et al. 1988, Ortman et al. 2012

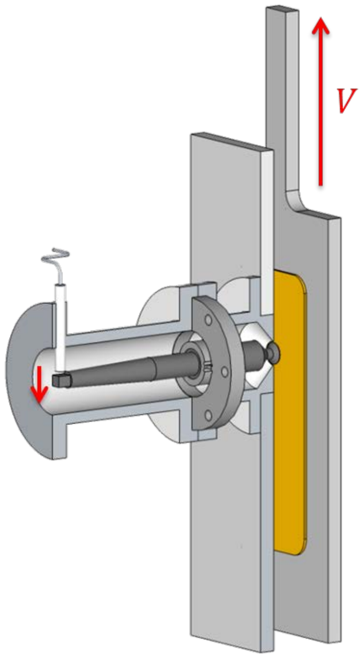
Shear vs PE



Matrix	Fiber Type	Weight % Fiber	Volume % Fiber	Number Average Length	Weight Average Length	Initial Orientation
SABIC Low Flow Polypropylene	Glass	10 %	3.6 %	1.55 mm	3.59 mm	“Planar Random”

Background: Rheology





Eberle, A.P.R., et al., *Using transient shear rheology to determine material parameters in fiber suspension theory.* J. Rheol., 2009. 53(3): p. 685-705.

Oakley, J.G. and A.J. Giacomin, *A sliding plate normal thrust rheometer for molten plastics.* Polym. Eng. Sci., 1994. 34(7): p. 580-4.

Ortman, K., et al., *Using startup of steady shear flow in a sliding plate rheometer to determine material parameters for the purpose of predicting long fiber orientation.* J. Rheol., 2012. 56(4): p. 955-981.

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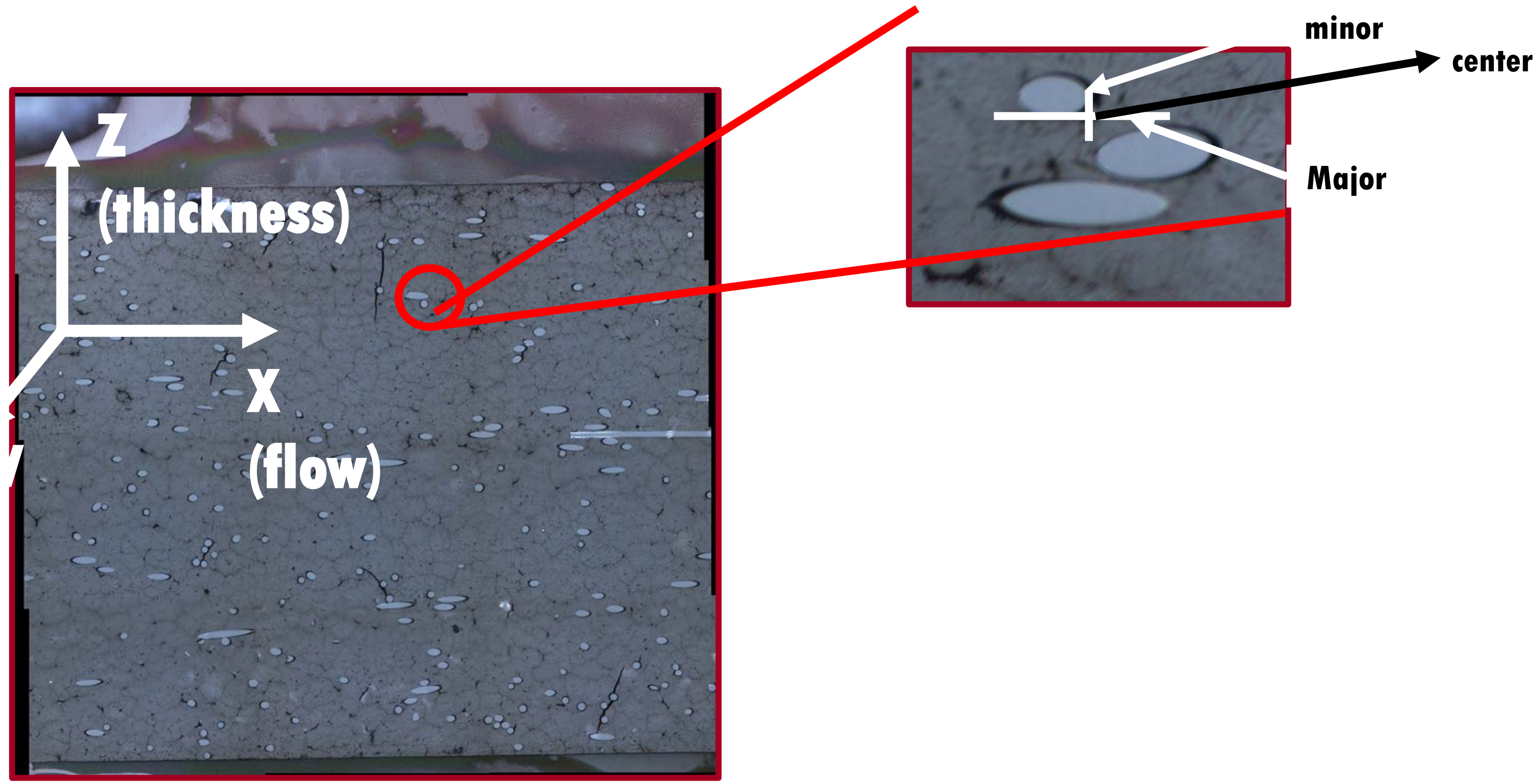
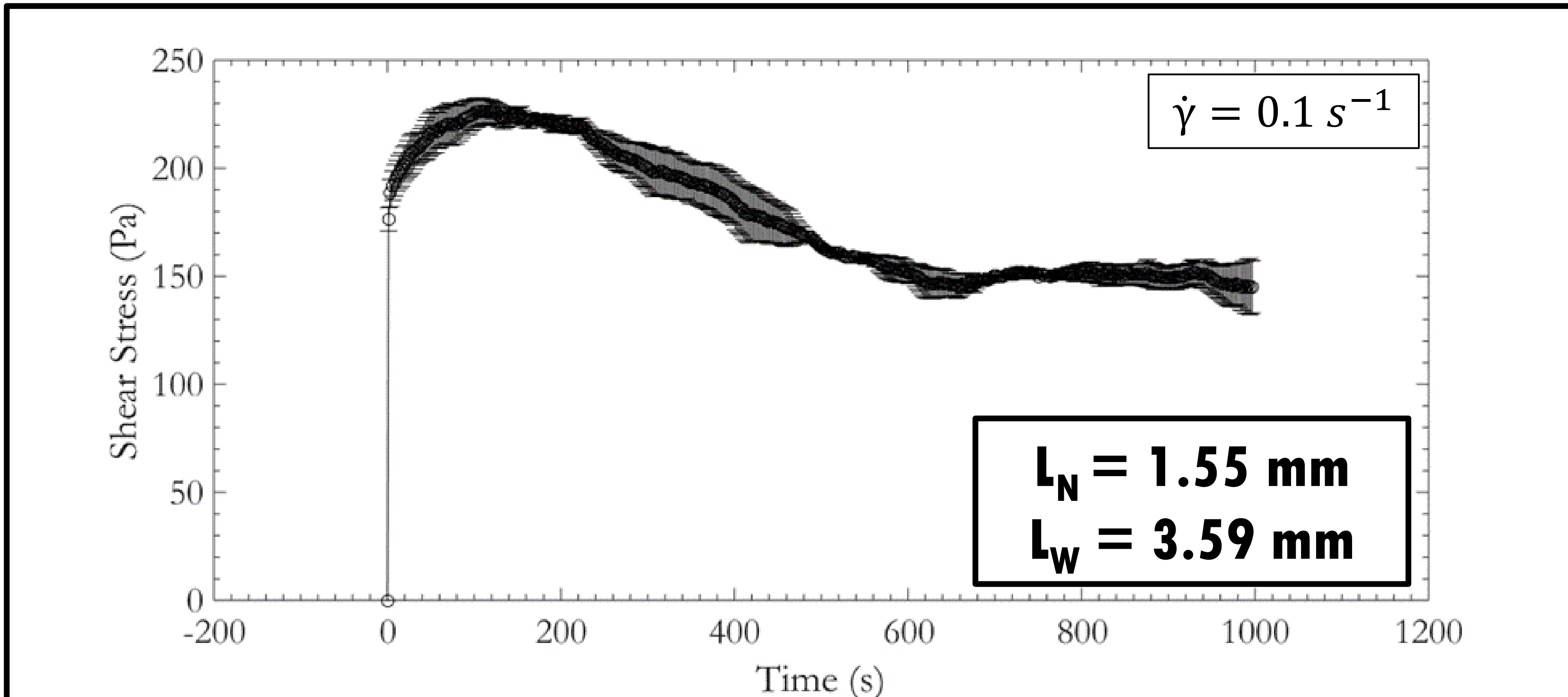
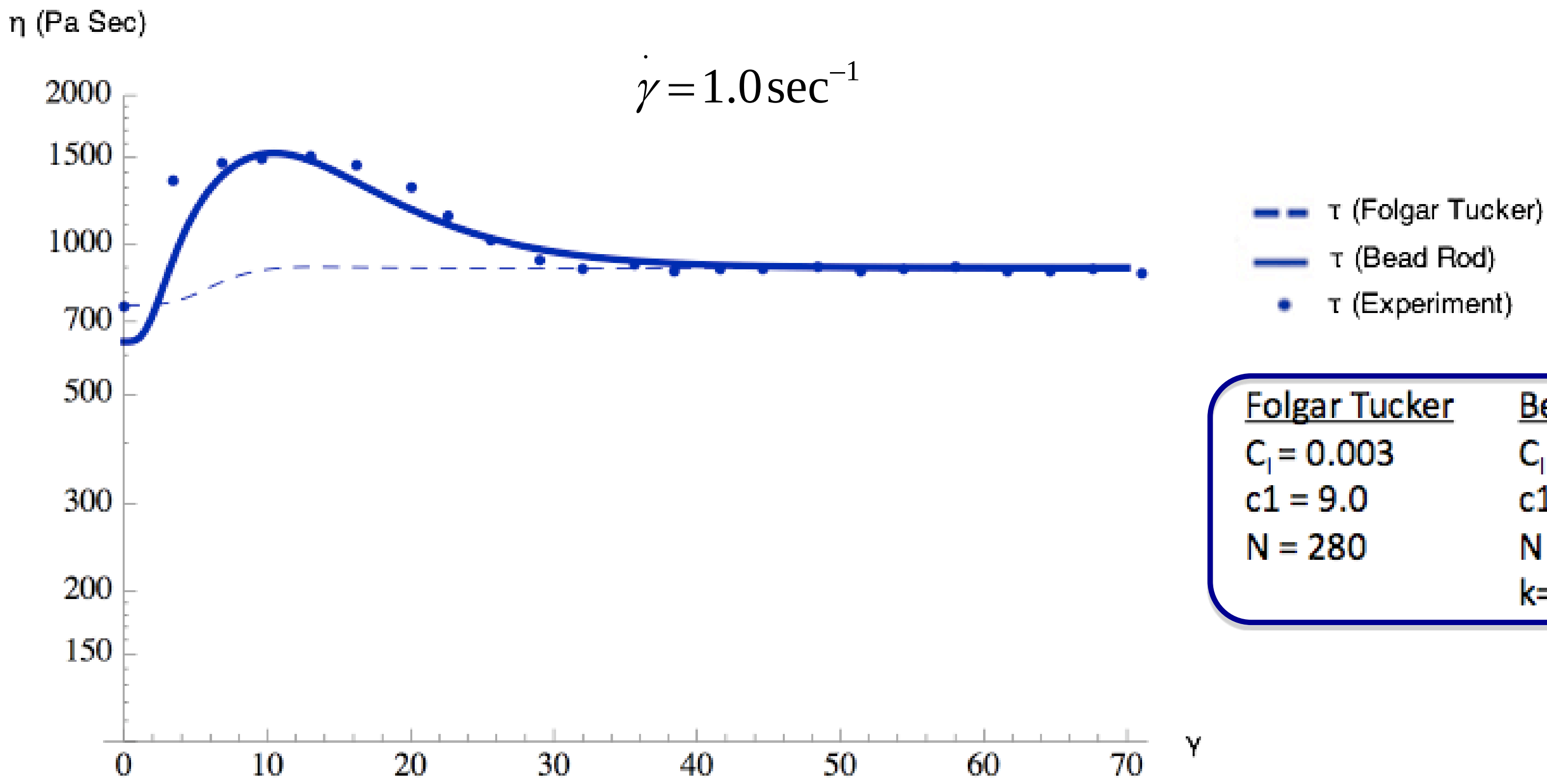
Background: Orientation Measurement(Leeds Method)



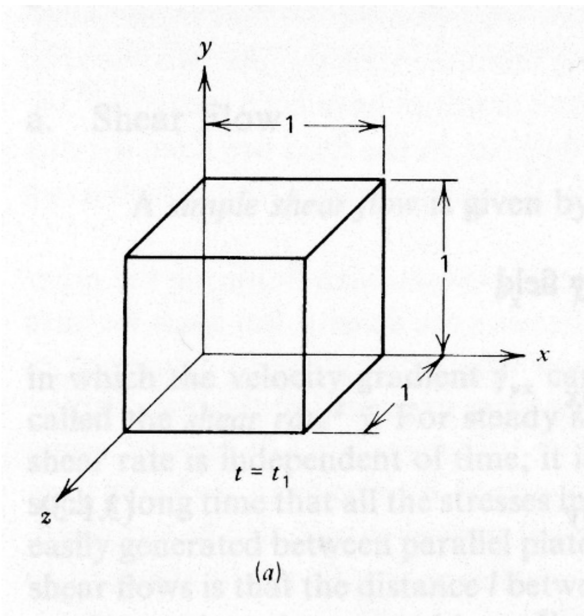
Shear vs PE: Shear Stress Growth

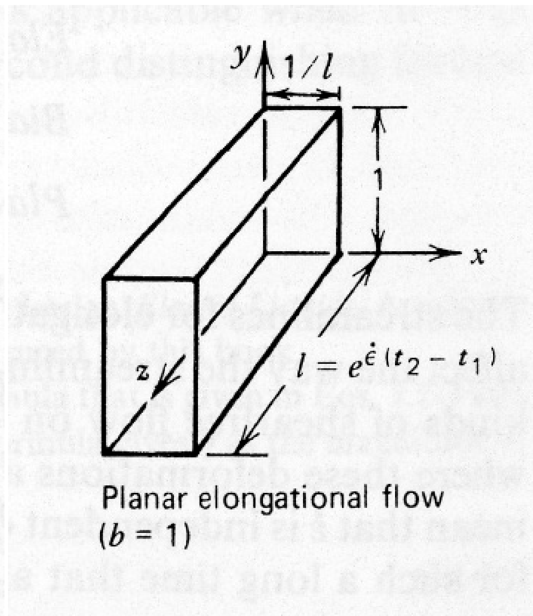




Folgar Tucker	Bead Rod
$C_I = 0.003$	$C_I = 0.001$
c1 = 9.0	c1 = 3.5
N = 280	N =830
	k= 0.15

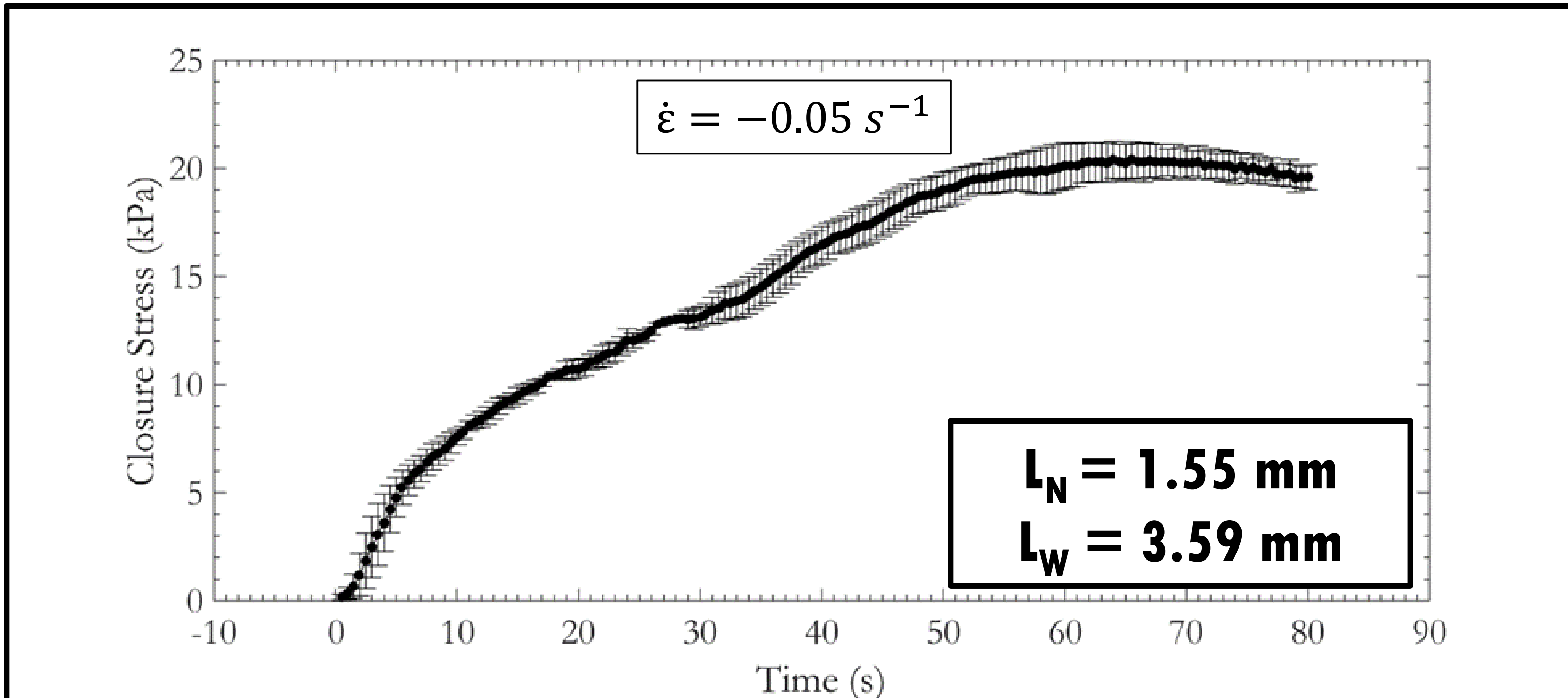
Shear vs PE



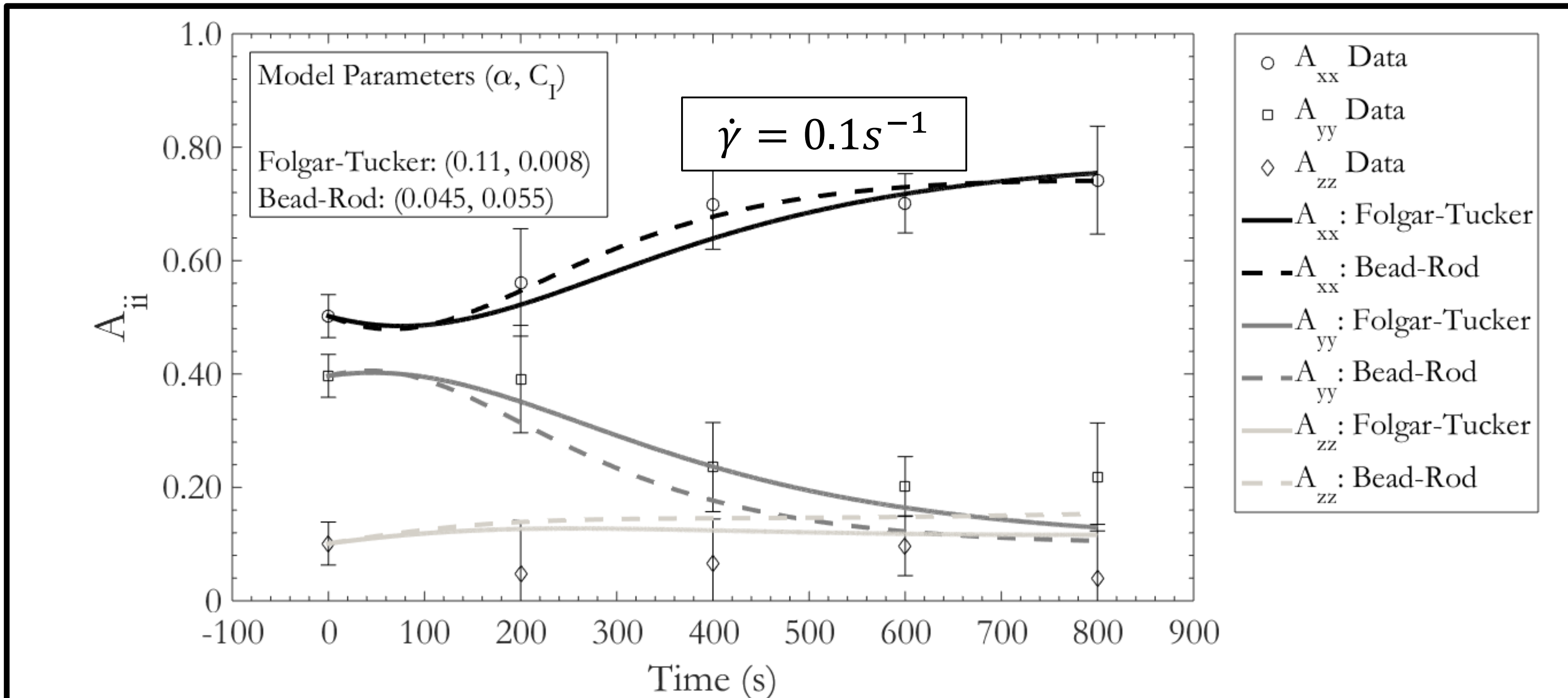


Bird, R.B., et al., *Dynamics of Polymeric Liquids*. 2 ed. Vol. 1. 1987, USA: John Wiley & Sons..

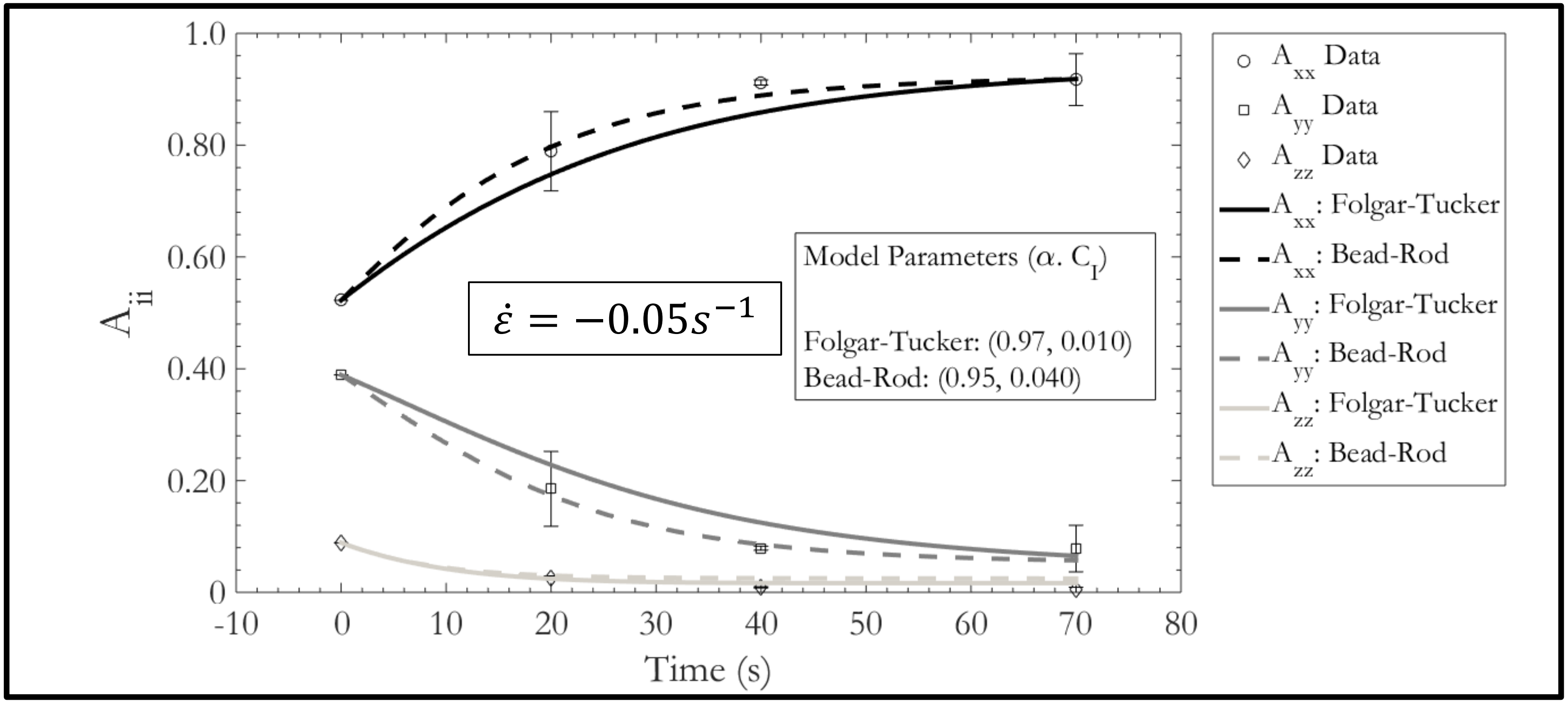
Shear vs PE: PE Stress Growth



Shear vs Planar Ext: Shear Orientation

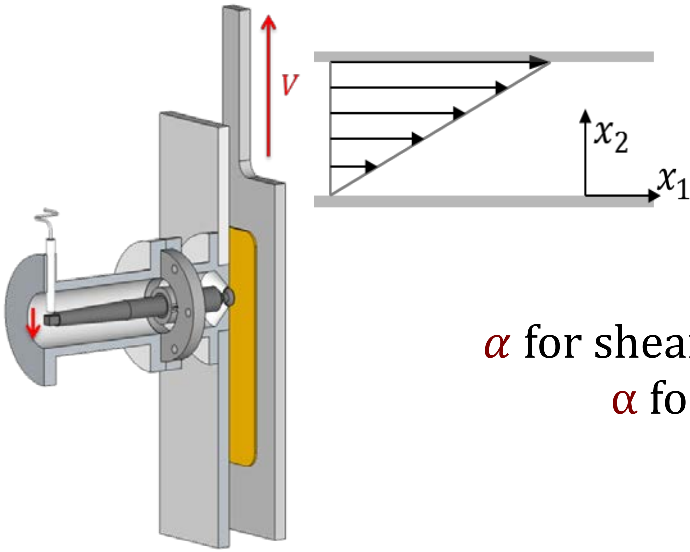


Shear vs Planar Ext: PE Orientation

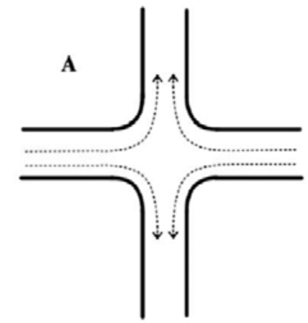


Flow-Type Dependent α ()

Sliding Plate (Shear)



Lubricated Squeeze(Extensional)



α for shear much smaller than
α for extensional

$\alpha_s < \alpha_e$

Models	α_s	α_e
SRF	0.25	1.0
RSC	0.2	1.0
ARD	0.2	1.0

Shear vs PE: Parameters

	Parameter	Shear	Extension
Rigid	α	0.11	0.97
	C_l	0.008	0.01
Flexible	α	0.045	0.95
	C_l	0.055	0.04

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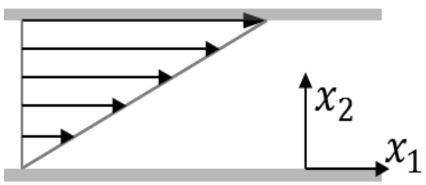
Flow-Type Parameter (Classifier)

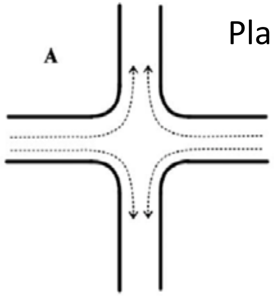
- The local flow-type of a complex flow can be identified by a dimensionless parameter β.
- β is evaluated from local velocity gradient.

Simple shear



$\beta = 0$

Planar extension



$\beta = 1$

- Values between 1 and 0 indicate a mixture of extension and shear.

when $0 \leq \beta \leq 1$: (shear, extensional, or the mix of both)

Variable Strain Reduction Factor: α_e (determined from extensional) α_s (determined from shear)

$$\alpha = \beta * \alpha_e + (1 - \beta) * \alpha_s$$

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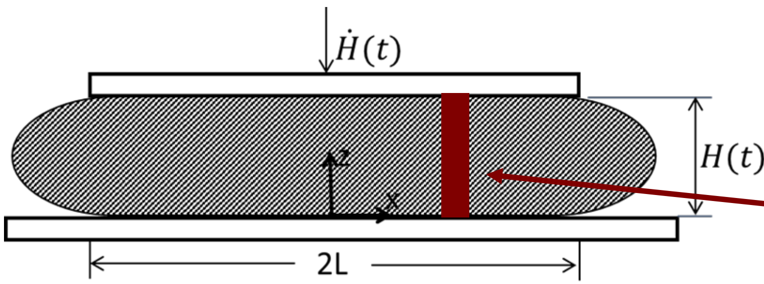
NLSF with Long-Fibers

Materials: **30 wt% glass fiber reinforced polypropylene**

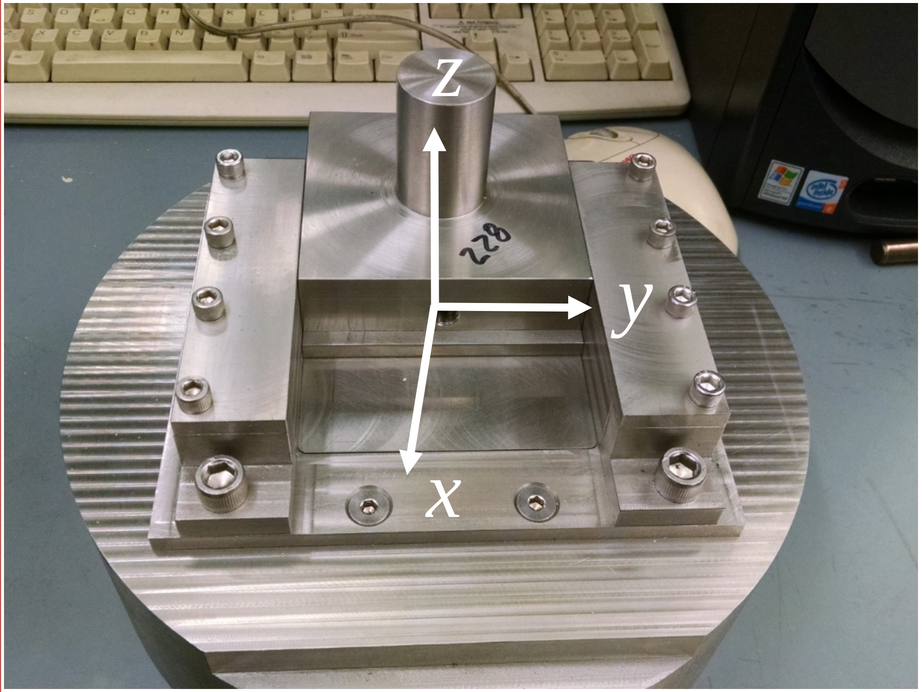


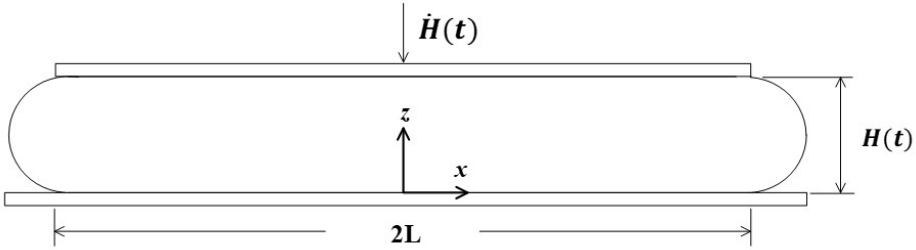
❖ Non lubricated squeeze flow (NLSF) with short fibers ($L_n = 0.8$ mm, $L_w = 2.5$ mm)

Through thickness orientation at $x = L/2$

Both are (comprise) complex flows

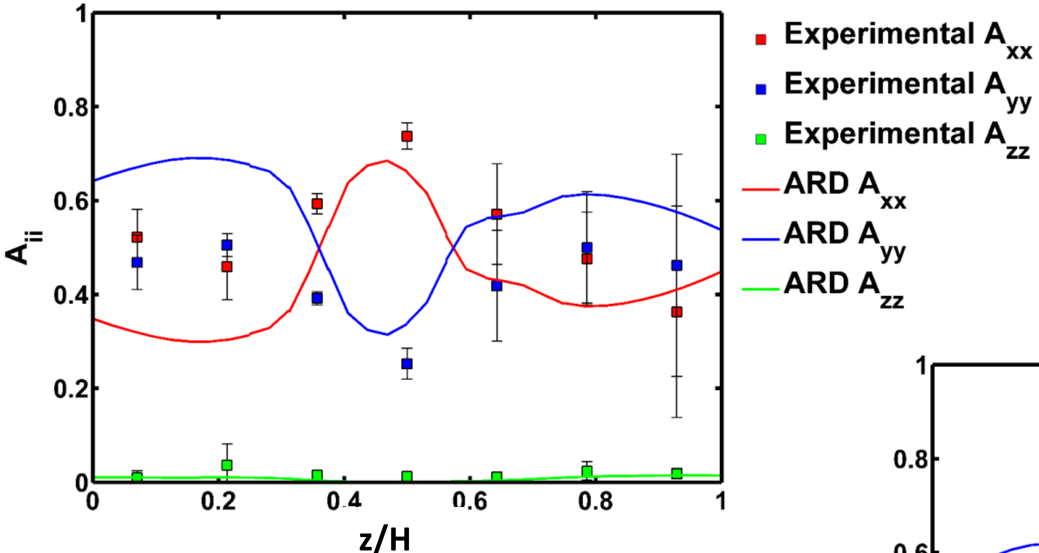
NLSF: Experimental





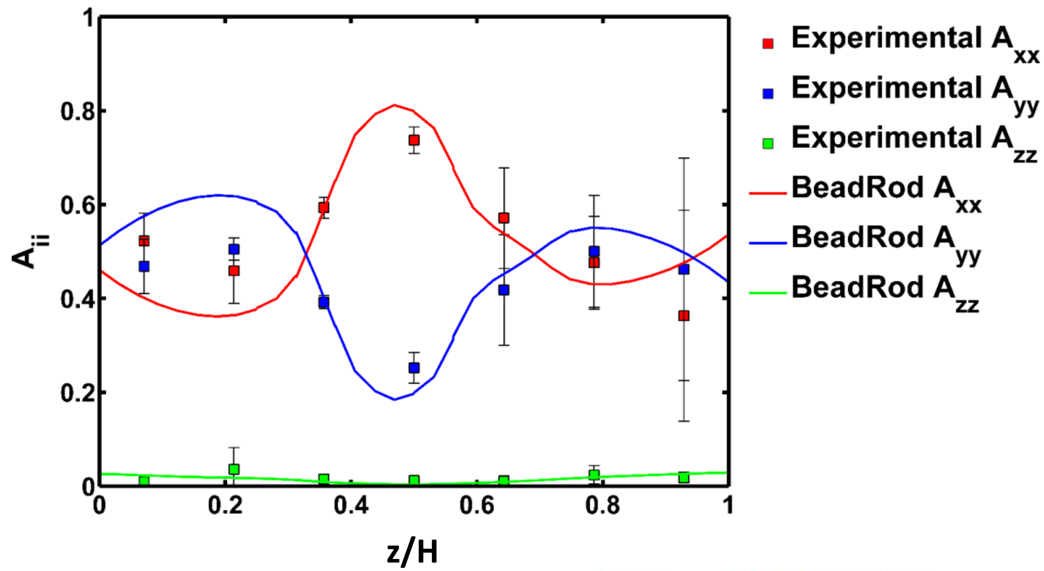
- Combination of shear and extension
- Second-order velocity gradients
- Closure stress easily measured

NLSF Simulations (LGF) Using Variable α



Rigid Rod
Orientation Model

Bead-Rod Semi-
Flexible Fiber Model



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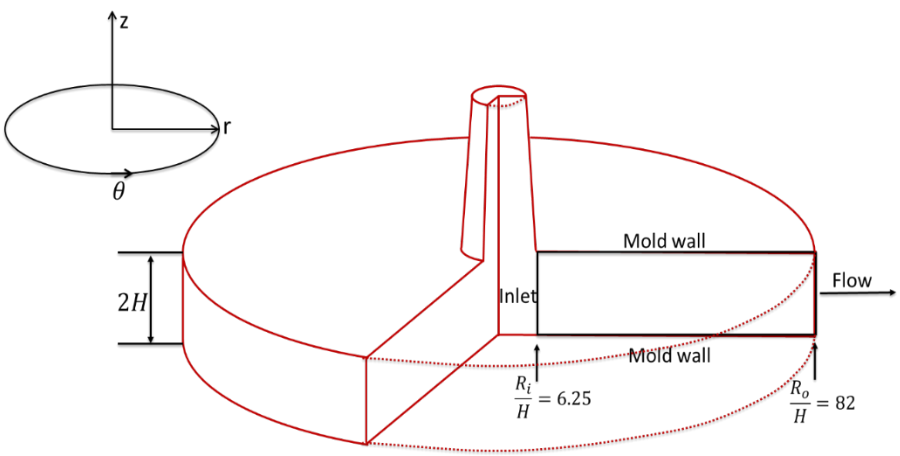
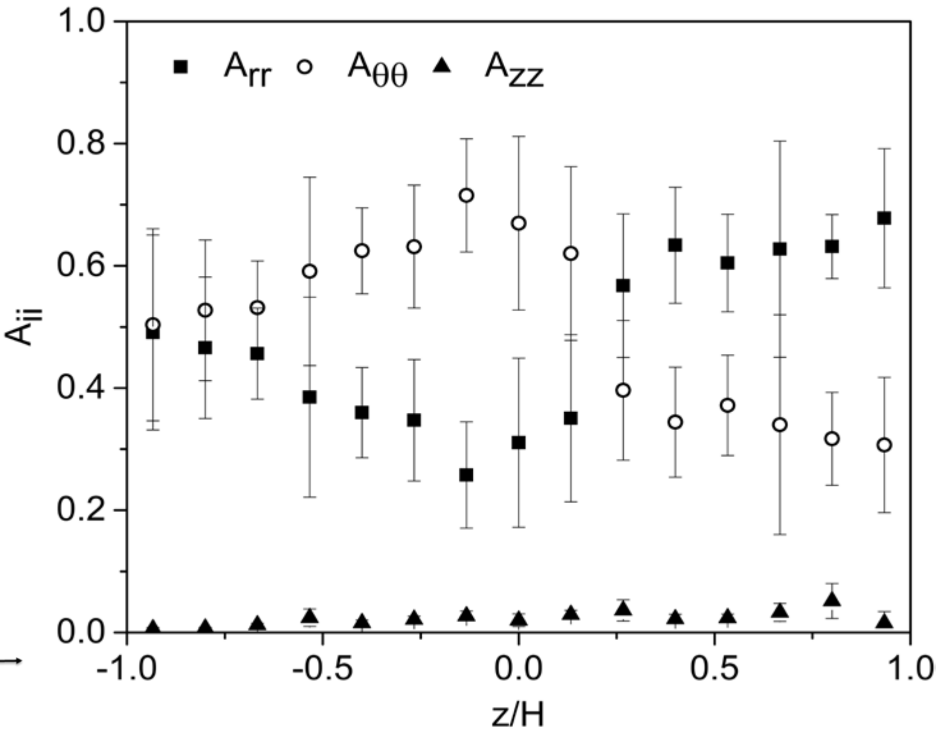
CGD Orientation at Inlet

CGD with 30 wt% long glass fibers in a polypropylene matrix

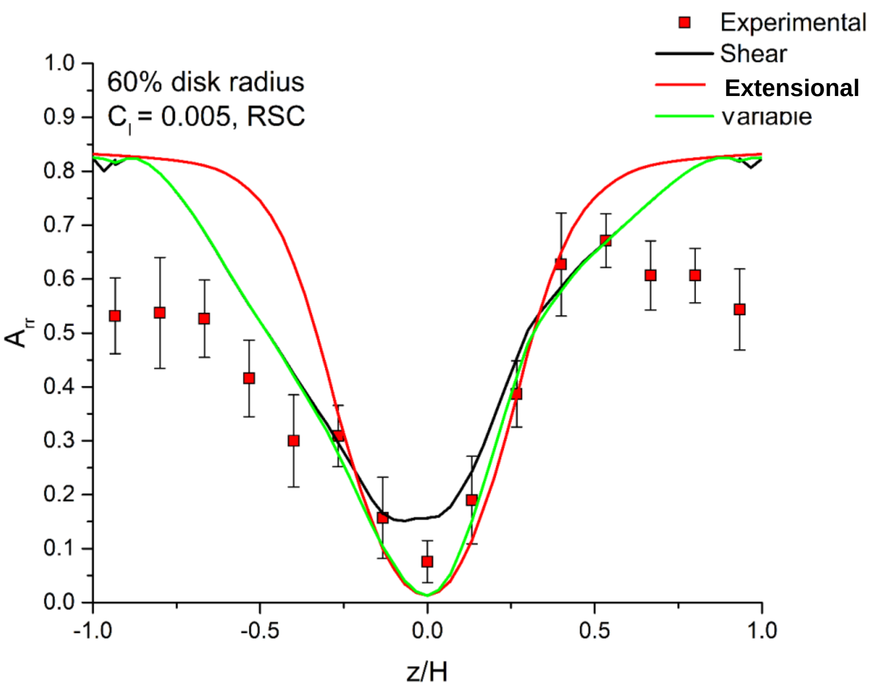
L_n (mm)	L_w (mm)
1.14±0.078	3.41±0.41



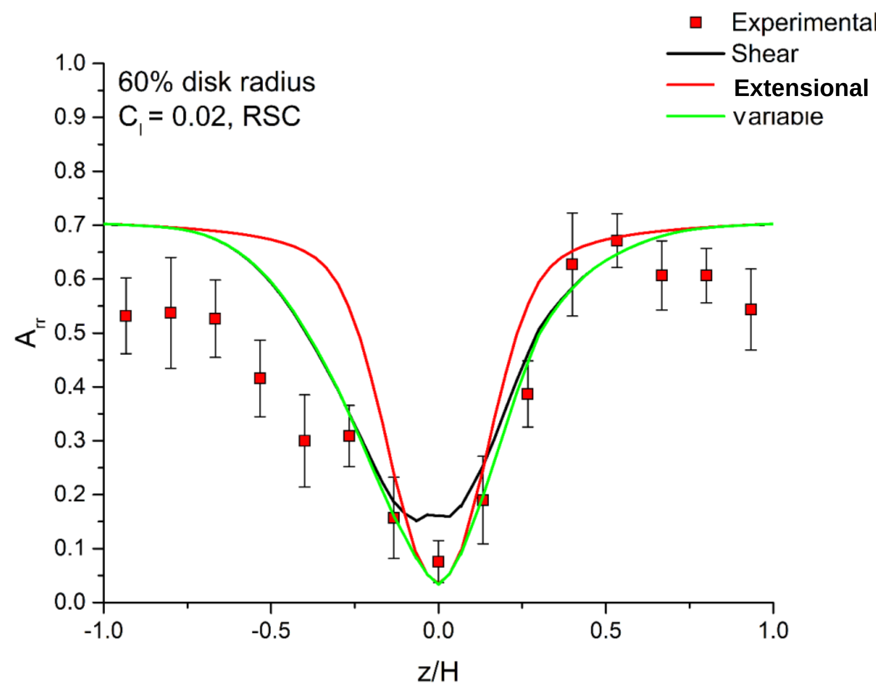


CGD at Hele-Shaw Region

RSC model



Bead-Rod Model



Conclusions

- ❖ α is dependent on the type of flow: Important for injection molding and compression molding
- ❖ Developed a variable strain-reduction factor based on the local flow conditions: shear, extension, and the mix of both.
- ❖ For the short fiber case the predicted orientation results using a variable α showed improved agreement of the profile shape with the experimental data in spite of the type of orientation model.
- ❖ For long fiber case, the bead-rod model with variable α did the best job predicting fiber orientations.

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Conclusions Continued

- Fiber orientation simulations for both non-lubricated squeeze flow and injection molded center-gated disk were conducted to verify this variable strain reduction factor method. The predicted orientation results showed improved qualitative agreement of the profile shape with the experimental data.

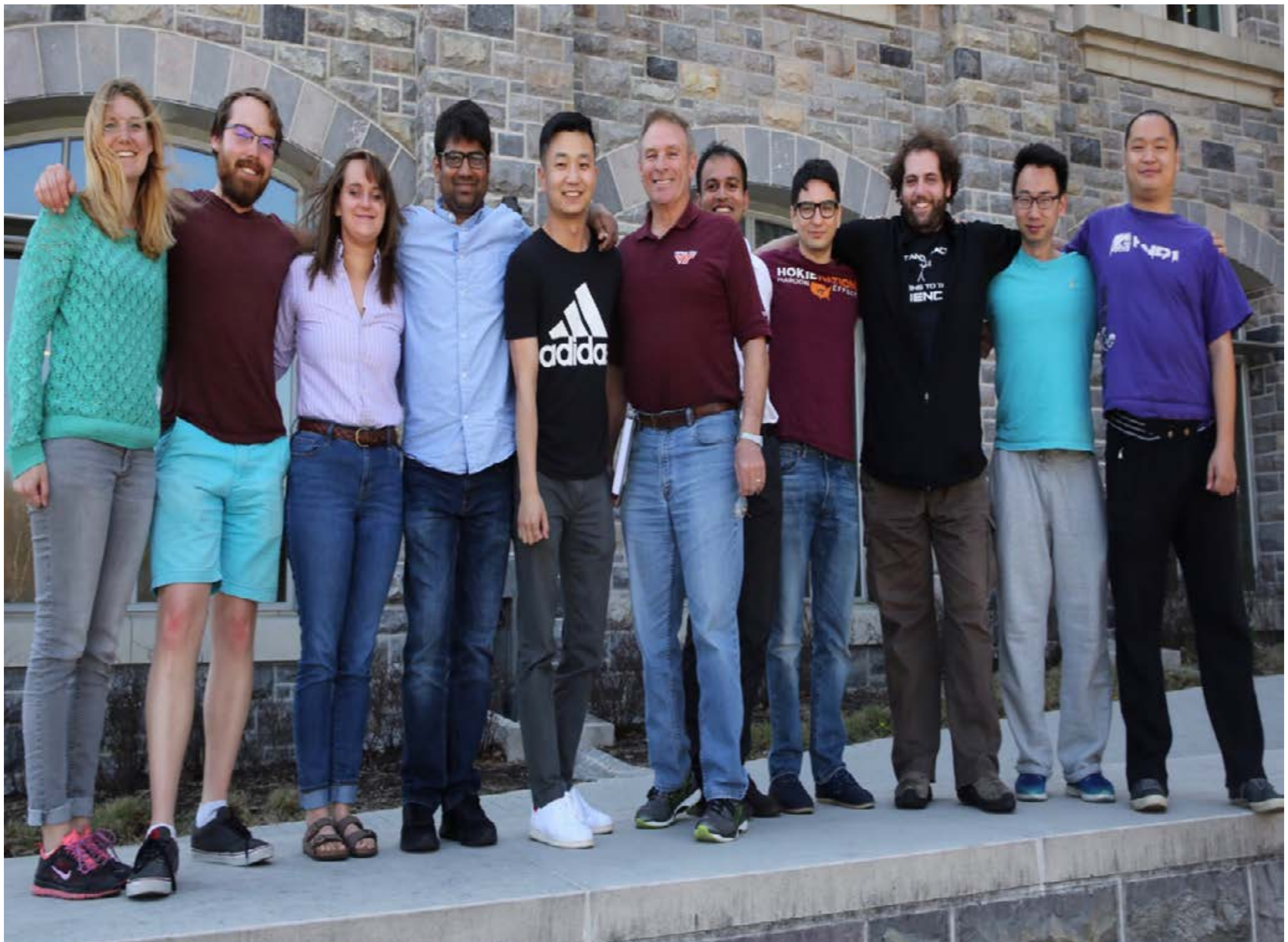
Future Efforts

- We need to confirm that NLSF can be used to efficiently obtain the parameters in the orientation and stress models.
- We need to develop a stress tensor for concentrated semi-flexible fiber suspensions.
- We need to develop a relation between orientation and fiber length and mechanical properties.

Acknowledgements

Macromolecules Innovation Institute
At the intersection of science, engineering, and society

Dr. Peter Wapperom



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Go Further

Long Fiber Orientation Behavior in Basic Flows of Fiber/ Polymer Melt Suspensions

Hongyu Chen, Greg Lambert, Peter Wapperom*, Kennedy Boyce, and Donald G. Baird, Department of Chemical Engineering, Macromolecules Innovation Institute, and the Department of Mathematics*, Virginia Polytechnic Institute and State University, Blacksburg, VA 24061-0201, USA

Overview

- Introduction & Background
  - Structure of semi-flexible fiber suspensions and origin to length
  - Stress and orientation tensors for rigid fiber suspensions
  - Fiber orientation model for semi-flexible fibers
  - Stress tensor for semi-flexible fiber suspensions
- Comparison of long fiber (semi-flexible) orientation evolution in shear and planar extension
- Modification of fiber orientation theory to incorporate flow type
- Investigation of non-lubricated squeeze flow (NLSF)
- Comparison of parameters obtained in shear and planar extension
- Prediction of fiber orientation in a basic molding flow
- Conclusions & Recommendations
- Acknowledgements

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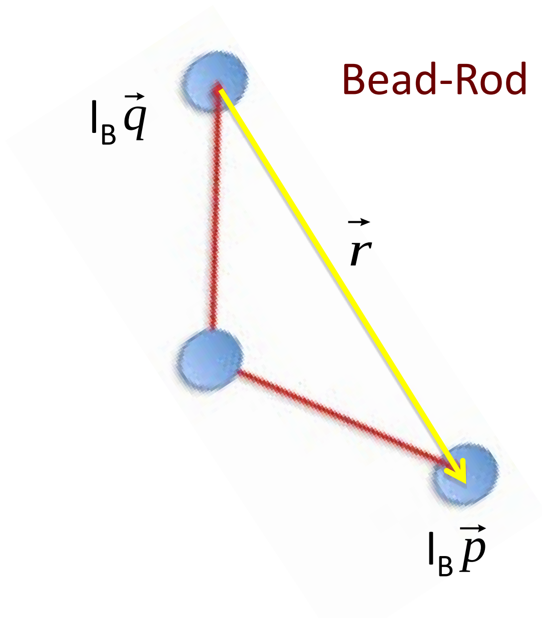


$$\underline{\underline{A}} = \int \vec{p}\vec{p}\,\psi(\vec{p},\vec{q},t)\,d\vec{p}\,d\vec{q}$$

$$\underline{\underline{B}} = \int \vec{p}\vec{q}\,\psi(\vec{p},\vec{q},t)\,d\vec{p}\,d\vec{q}$$

$$\vec{r} \equiv l_B\left(\vec{p}-\vec{q}\right)$$

What about constructing an end-to-end tensor to describe the orientation?

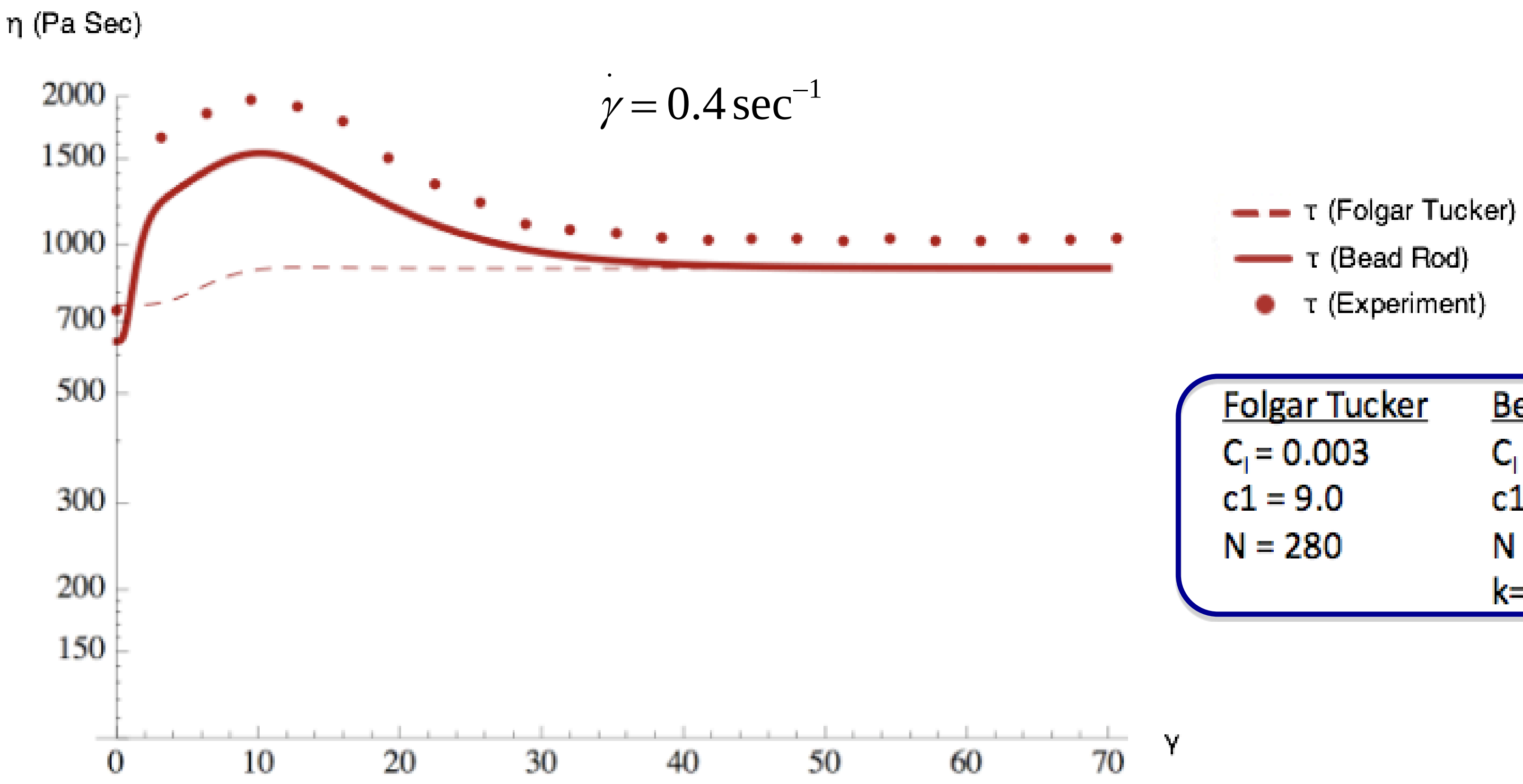
Libscomb Constitutive Model (quadratic closure):

$$\eta^{+} = \eta_s + c_1\varphi\eta_s + 2\varphi\eta_s N R_{12}^{2}$$

$$\underline{\underline{R}} = \frac{\underline{\underline{rr}}}{tr(\underline{\underline{rr}})}$$

η^{+} = (Newtonian Matrix + Fiber Concentration + Fiber Orientation)

Parameters to Fit = $\mathbf{c_1}$ and **N** ….. and (orientation model parameters $\mathbf{C_I}$ and/or **k**)



$$\dot{\gamma} = 0.4\,\text{sec}^{-1}$$

Folgar Tucker	Bead Rod
$C_I = 0.003$	$C_I = 0.001$
c1 = 9.0	c1 = 3.5
N = 280	N =830
	k= 0.15

Rheology: The Science of the Deformation and Flow of Matter

- How does the connection between flow behavior and properties evolve?
- The rheology of polymer composites provides a direct connection between processing conditions and properties generated.
- In the case of polymer composites, flow during processing controls fiber orientation and length.
- Physical properties are related to fiber orientation and fiber length.

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Background: Orientation Models

$$\frac{DA}{Dt} = \boldsymbol{W}\cdot\boldsymbol{A} - \boldsymbol{A}\cdot\boldsymbol{W} + \xi(\boldsymbol{D}\cdot\boldsymbol{A} + \boldsymbol{A}\cdot\boldsymbol{D} - 2[\boldsymbol{A}_4 + (1-\kappa)(\boldsymbol{L} - \boldsymbol{M}:\boldsymbol{A}_4)]:\boldsymbol{D}) + 2\kappa C_I D(\boldsymbol{I} - 3\boldsymbol{A})$$

$$\boldsymbol{L} = \sum_{i=1}^{3} \lambda_i \boldsymbol{e}_i\boldsymbol{e}_i\boldsymbol{e}_i\boldsymbol{e}_i \qquad \boldsymbol{M} = \sum_{i=1}^{3} \boldsymbol{e}_i\boldsymbol{e}_i\boldsymbol{e}_i\boldsymbol{e}_i$$

Wang, J., J.F. O'Gara, and C.L. Tucker III, *An objective model for slow orientation kinetics in concentrated fiber suspensions: Theory and rheological evidence.* J. Rheol., 2008. **52**(5): p. 1179-1200.

Shear vs NLSF: Experimental

Fiber Concentration	Number Average Length mm	Weight Average Length mm	Fiber Half Length l_B mm	Bending Potential Constant k s^{-1}
30 wt%	1.14	3.40	0.570	19.7
40 wt%	0.986	2.68	0.493	30.4
50 wt%	0.870	2.42	0.435	44.3

- Shear and NLSF
  o $\dot{\gamma} = 1\ s^{-1}$ and $\dot{\varepsilon} = -0.50\ s^{-1}$
- Initially oriented in y direction in xy plane

Objectives

1. Examine the assumption that the fiber orientation model parameters are independent of the flow field used to obtain them.
2. Develop a rheological test that incorporates both shear and extensional flow (non-lubricated squeeze flow), and verify that it can be used to obtain orientation model parameters through fitting to the measured fiber orientation.
3. Determine whether startup of shear or non-lubricated squeeze flow should be used for obtaining orientation model parameters in the future.

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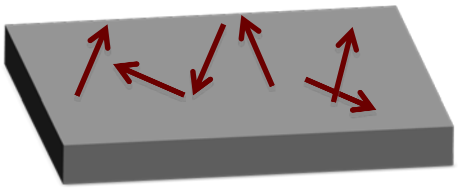
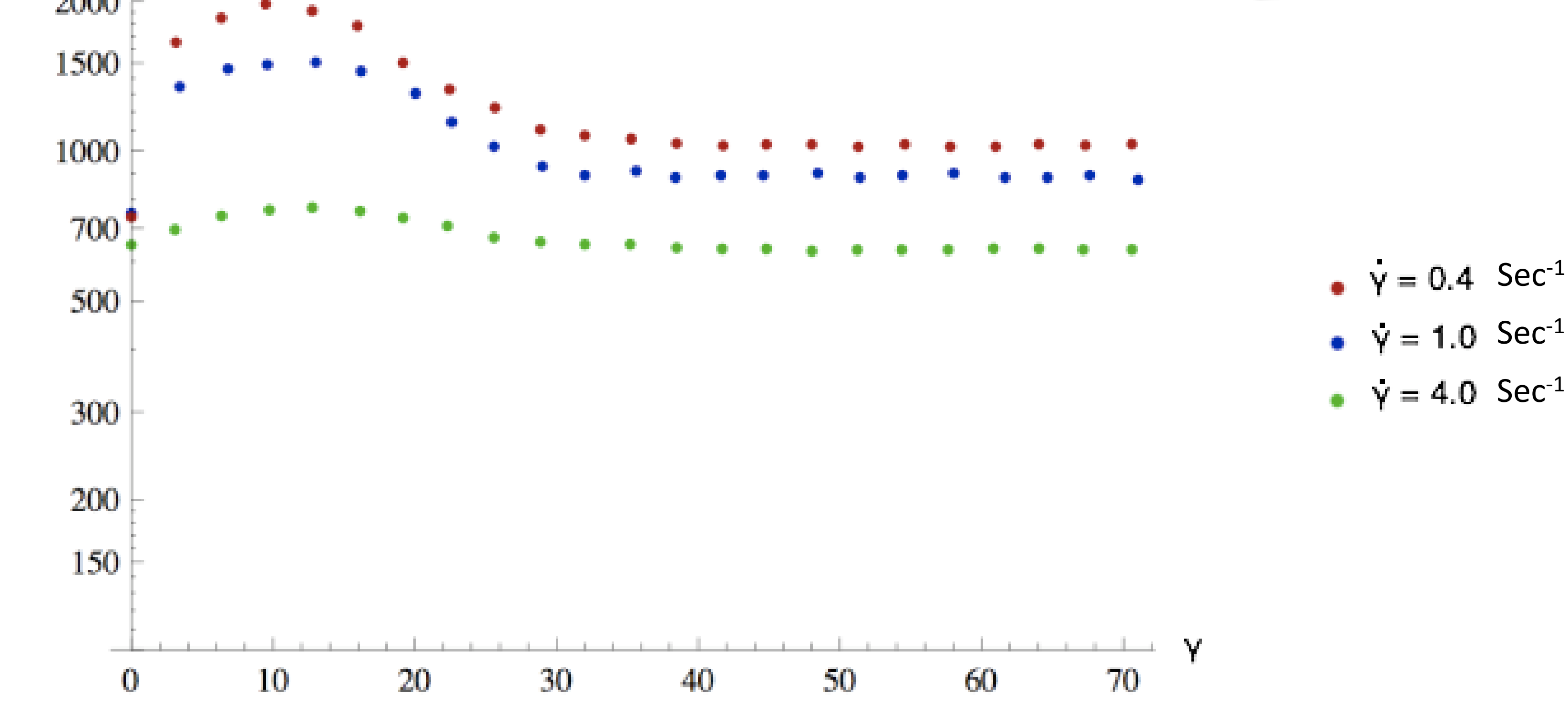
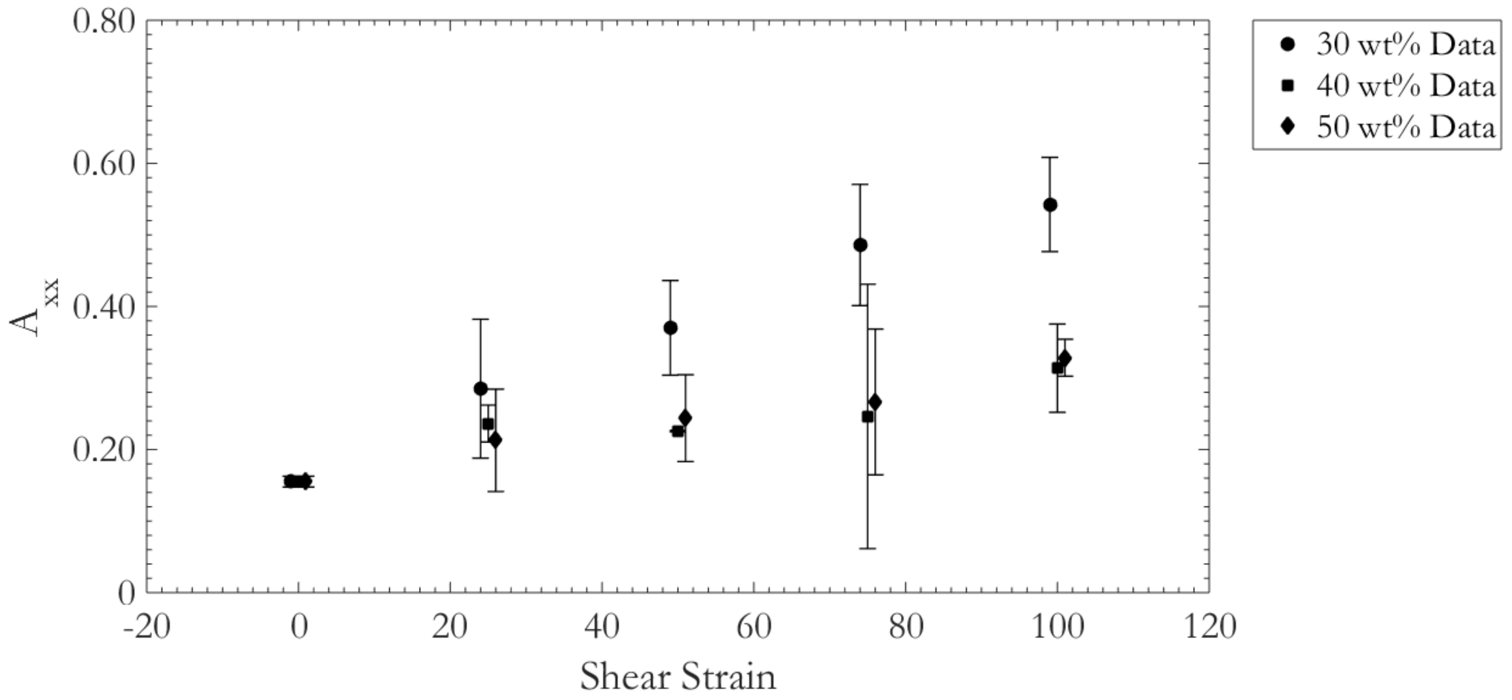
Startup rheology of 10% wt. Glass (~3.5 mm) filled Polypropylene initially Random

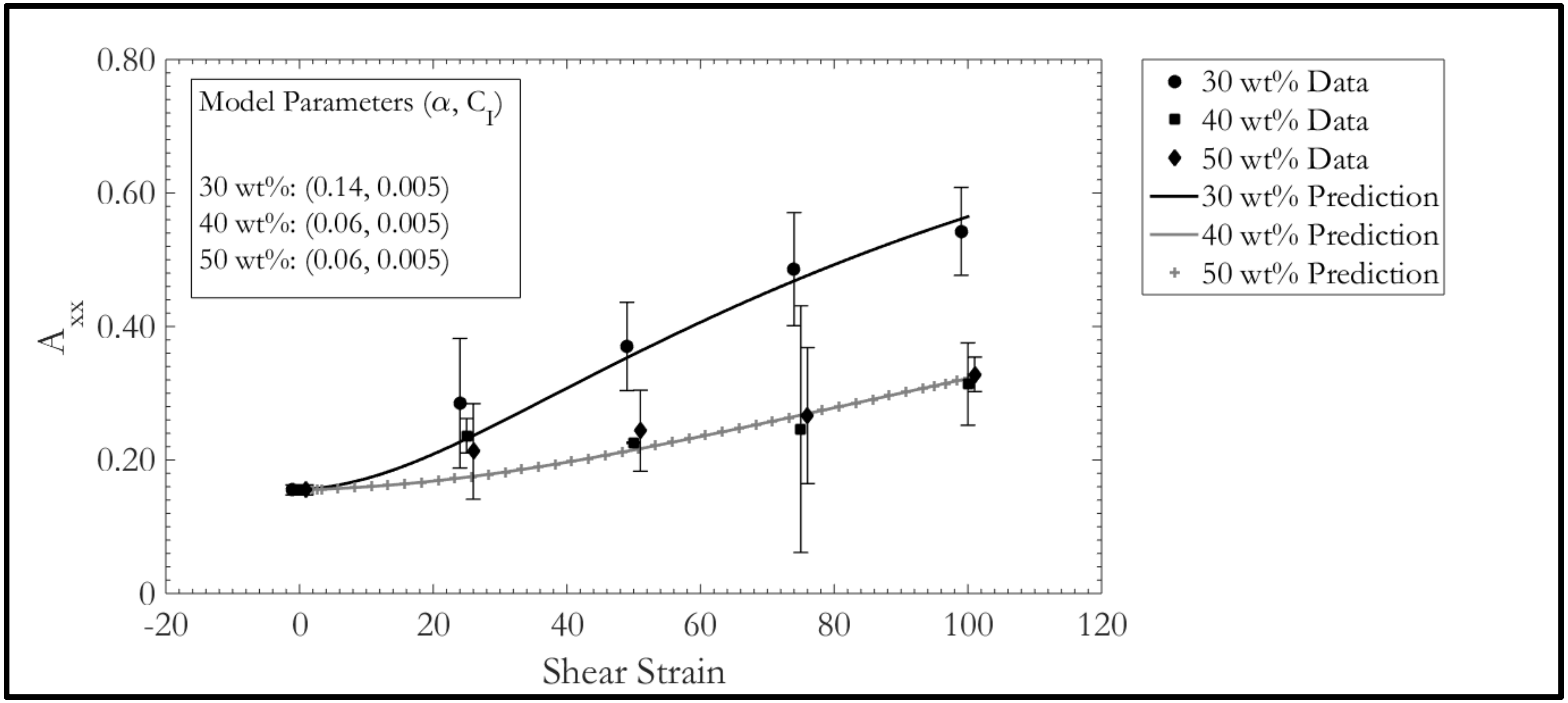




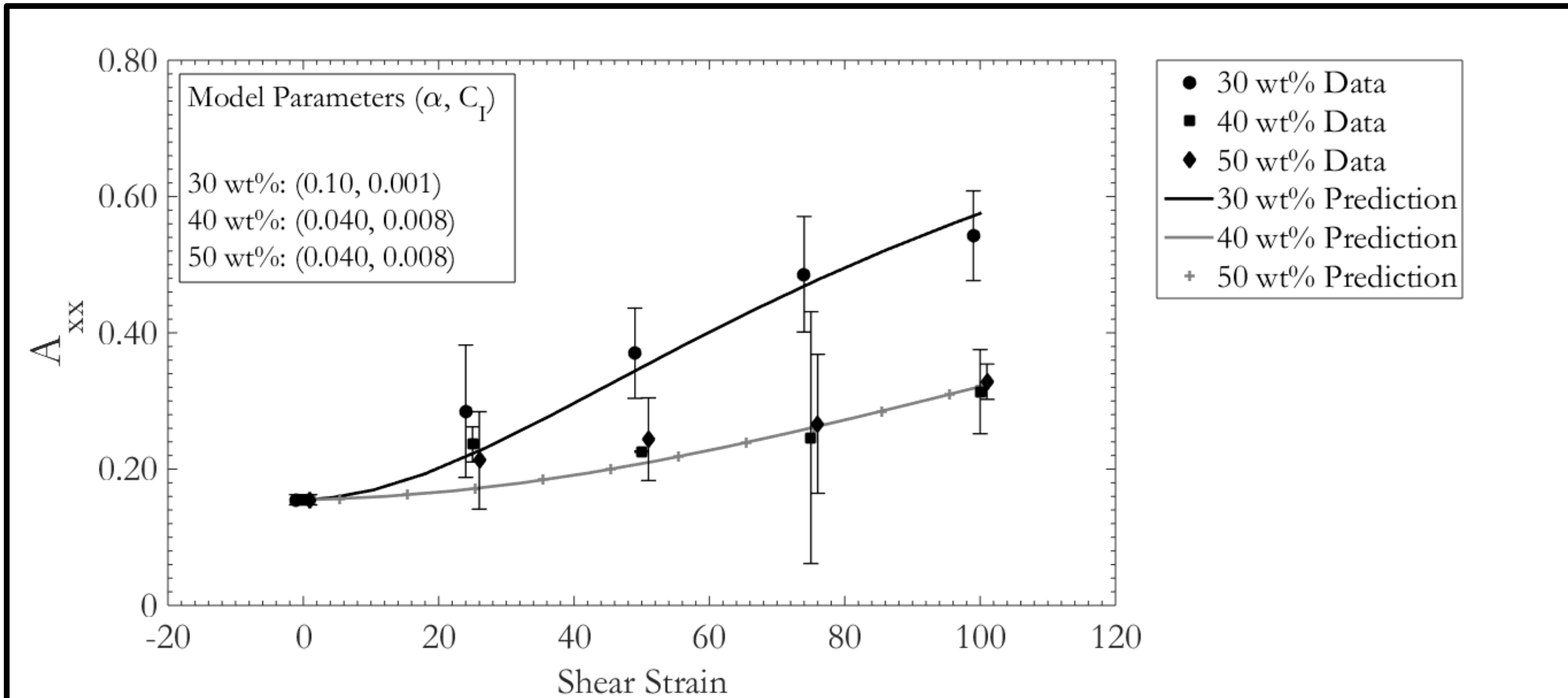
Shear vs NLSF: Shear Results



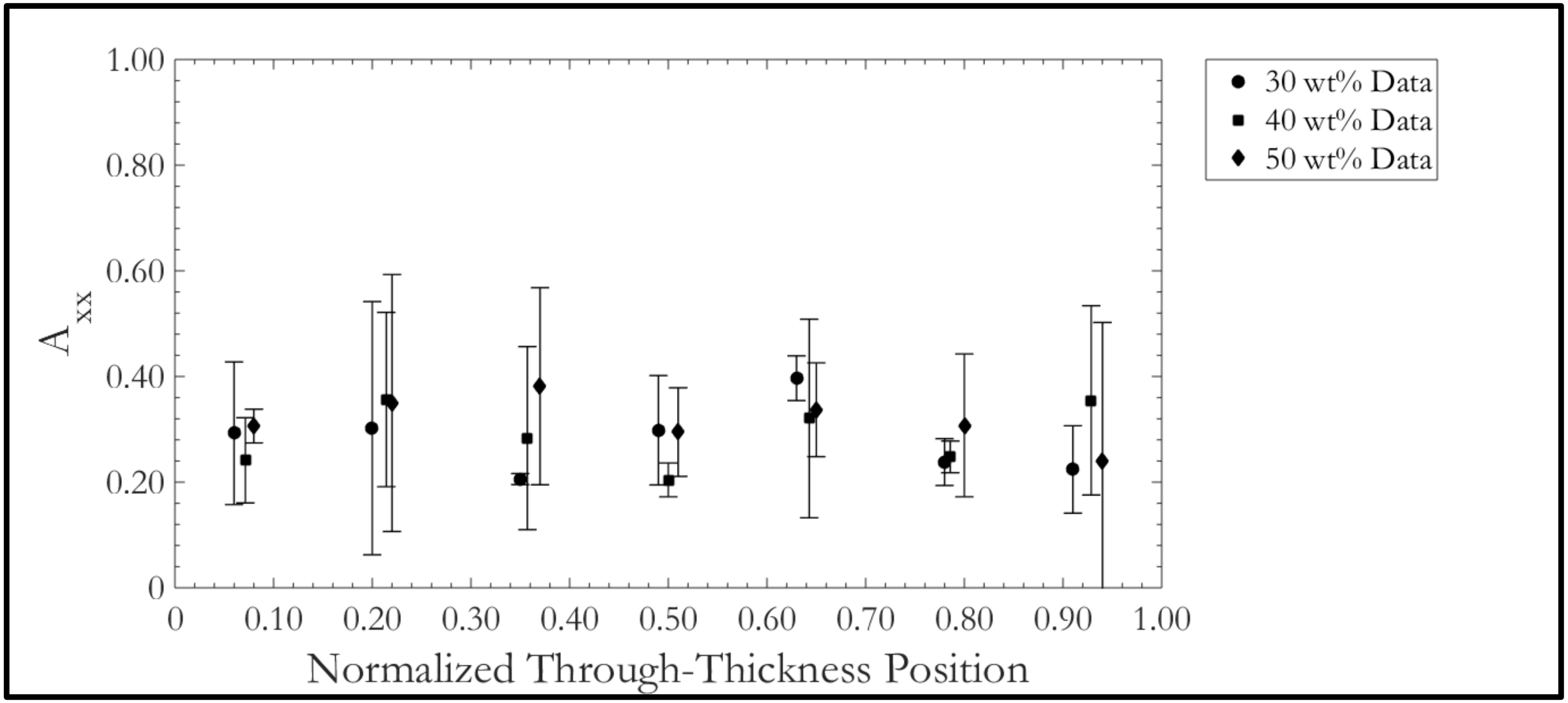
Shear vs NLSF: Shear Results



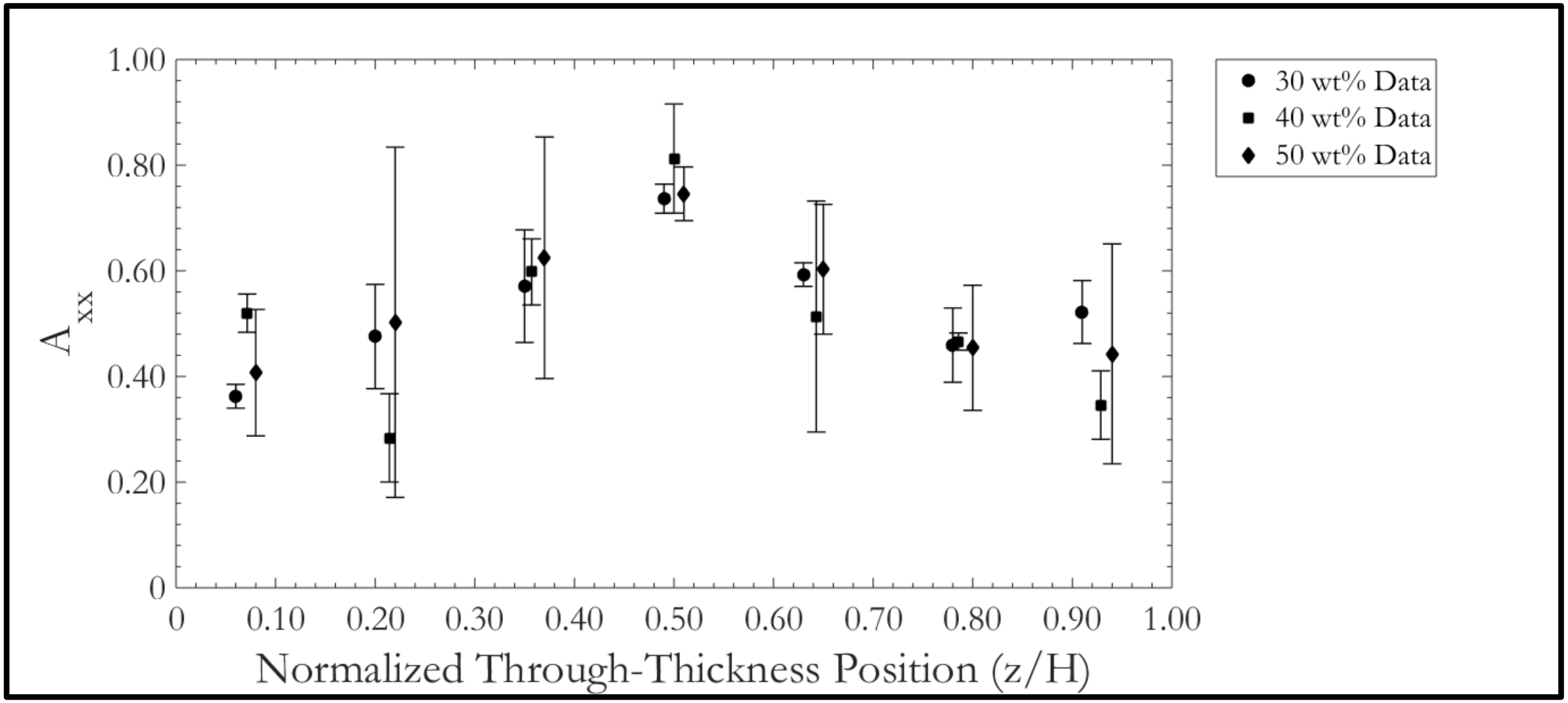
Shear vs NLSF: Shear Results



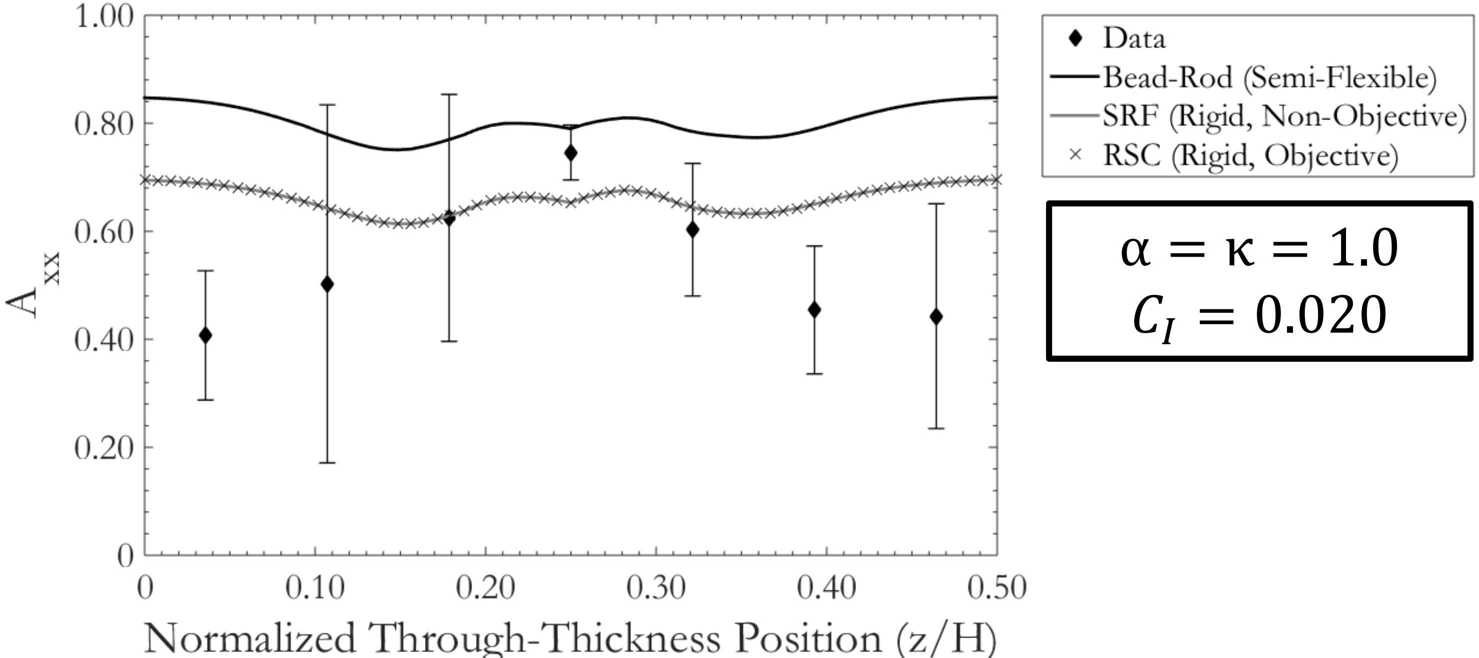
Shear vs NLSF: NLSF Initial Orientation



Shear vs NLSF: NLSF Orientation

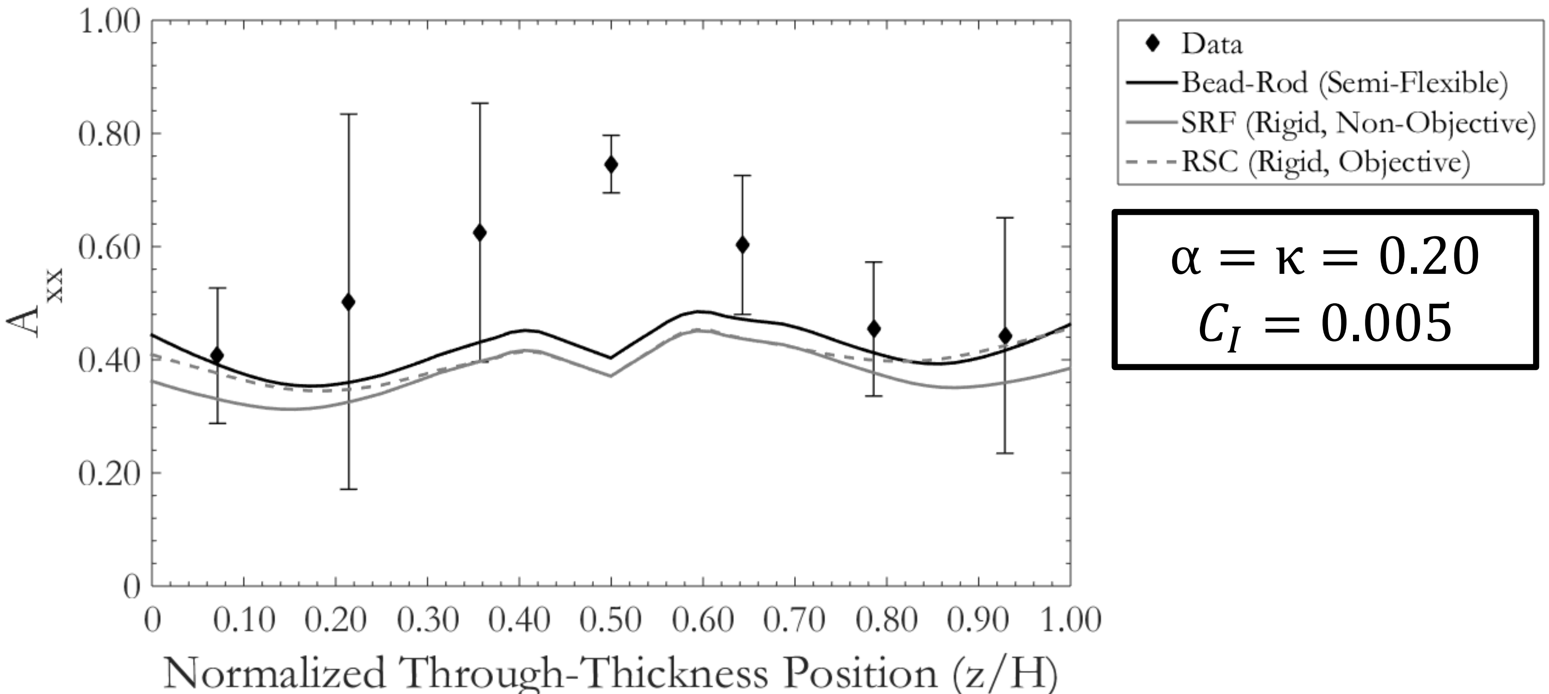


Shear vs NLSF: NLSF Fitting



$$\alpha = \kappa = 1.0$$
$$C_I = 0.020$$

Shear vs NLSF: NLSF



$$\alpha = \kappa = 0.20$$
$$C_I = 0.005$$

Shear vs NLSF: Conclusions

- Apparent interaction between concentration and initial orientation in shear
  - Orientation of 40 and 50 wt% material much slower than expected
  - Models do not have a mechanism that accounts for this
    - Just change the parameters for the same material
- Need to revise treatment of strain reduction
  - One constant value doesn't work in a mixed shear/extensional flow
  - Problems not rectified with objectivity
- Need more time points for the NLSF data
  - Establish transient behavior
  - Establish steady state

Background: Recap

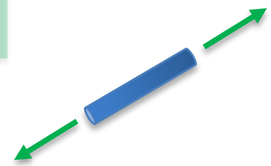
- **Need model parameters for part design**
  - **Really want to predict *mechanical properties***
  - **Common method → suboptimal parts**
- **Rheology might work**
  - **Independent of processing**
  - **Justified(?) in extrapolating to processing flows**
- **Limited success**
  - **CGD okay, EGP not so much**
- **Could extensional flow provide some insight?**

Coupling Orientation to Flow

Stress Equation for Rigid Fibers:

$$\boldsymbol{\sigma} = -P\mathbf{I} + 2\eta_m \mathbf{D} + 2\eta_m \phi(\mu_1 \mathbf{D} + \mu_2 \mathbf{D} : \mathbf{A_4})$$

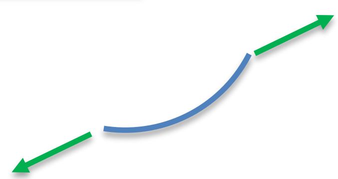
Matrix — Fibers

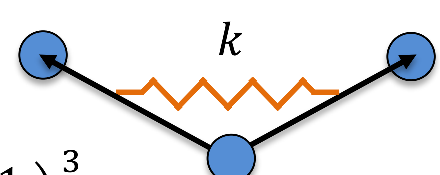


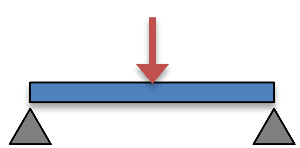
Proposed Stress Equation for Semi-Flexible Fibers:

$$\boldsymbol{\sigma} = -P\mathbf{I} + 2\eta_m \mathbf{D} + 2\eta_m \phi(\mu_1 \mathbf{D} + \mu_2 \mathbf{D} : \mathbf{R_4}) + \eta_m k \frac{3\phi a_r}{2}(\mathbf{B} - \mathbf{A} tr \mathbf{B})$$

Matrix — Fibers — Fiber Bending







$$k = \frac{E_y}{8\eta_m}\left(\frac{1}{a_r}\right)^3$$

Lipscomb et al. 1988, Ortman et al. 2012

Background: Orientation Models

$$\frac{D\boldsymbol{A}}{Dt} = \alpha\left((\boldsymbol{W}\cdot\boldsymbol{A} - \boldsymbol{A}\cdot\boldsymbol{W}) + (\boldsymbol{D}\cdot\boldsymbol{A} + \boldsymbol{A}\cdot\boldsymbol{D} - 2\boldsymbol{D}:\boldsymbol{A_4}) + 6C_I\dot{\gamma}\left(\frac{1}{3}\boldsymbol{I} - \boldsymbol{A}\right) + \frac{l_B}{2}(\boldsymbol{Cm} + \boldsymbol{mC} - 2(\boldsymbol{m}\cdot\boldsymbol{C})\boldsymbol{A}) - 2k(\boldsymbol{B} - \boldsymbol{A}tr(\boldsymbol{B}))\right)$$

$$\frac{D\boldsymbol{B}}{Dt} = \alpha\left((\boldsymbol{W}\cdot\boldsymbol{B} - \boldsymbol{B}\cdot\boldsymbol{W}) + (\boldsymbol{D}\cdot\boldsymbol{B} + \boldsymbol{B}\cdot\boldsymbol{D} - 2(\boldsymbol{D}:\boldsymbol{A})\boldsymbol{B}) - 4C_I\dot{\gamma}\boldsymbol{B} + \frac{l_B}{2}(\boldsymbol{Cm} + \boldsymbol{mC} - 2(\boldsymbol{m}\cdot\boldsymbol{C})\boldsymbol{B}) - 2k(\boldsymbol{A} - \boldsymbol{B}tr(\boldsymbol{B}))\right)$$

$$\frac{D\boldsymbol{C}}{Dt} = \alpha\left(\nabla\boldsymbol{v}^t\cdot\boldsymbol{C} - (\boldsymbol{A}:\nabla\boldsymbol{v}^t)\boldsymbol{C} - 2C_I\dot{\gamma}\boldsymbol{C} - \frac{l_B}{2}(\boldsymbol{m} - (\boldsymbol{m}\cdot\boldsymbol{C})\boldsymbol{C}) - k(1 - tr(\boldsymbol{B}))\boldsymbol{C}\right)$$

Matrix Deformation | Isotropic Rotary Diffusion | Flow-Coupled Bending | Bending Potential

$$\boldsymbol{m} = \sum_{i=1}^{3}\sum_{j=1}^{3}\sum_{k=1}^{3}\frac{\partial^2 v_i}{\partial x_j \partial x_k}A_{jk}\boldsymbol{\delta}_i$$

End-to-End Tensor

$$\boldsymbol{r} = l_B(\boldsymbol{p} - \boldsymbol{q})$$

$$\boldsymbol{R} = \frac{\langle\boldsymbol{rr}\rangle}{tr(\boldsymbol{rr})} = \frac{\boldsymbol{A} - \boldsymbol{B}}{1 - tr(\boldsymbol{B})}$$

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INTRODUCTION