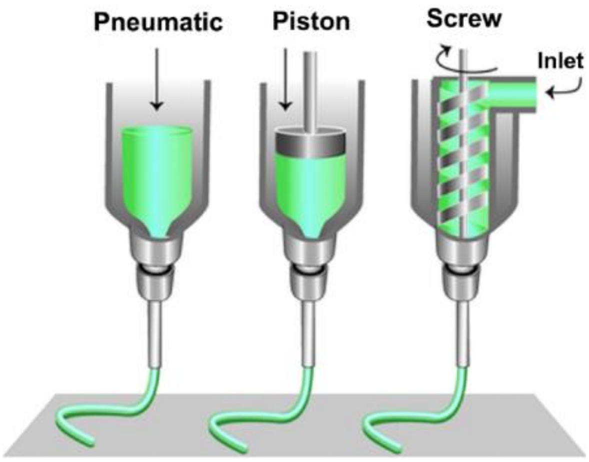
Numerical Simulation of Hydrogel-Based Bioprinting

Zelai Xu, James J. Feng

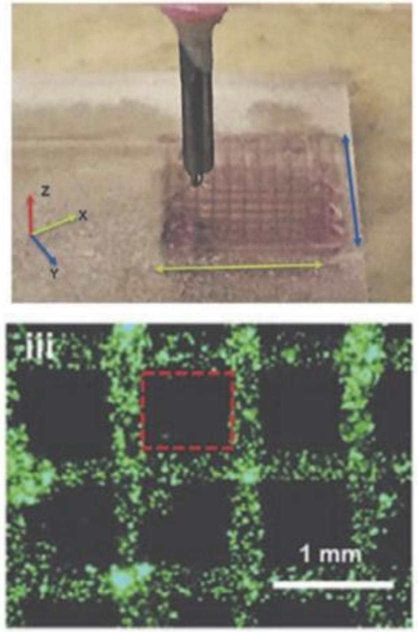
FREEFEM DAYS
17TH EDITION - PARIS

Motivation



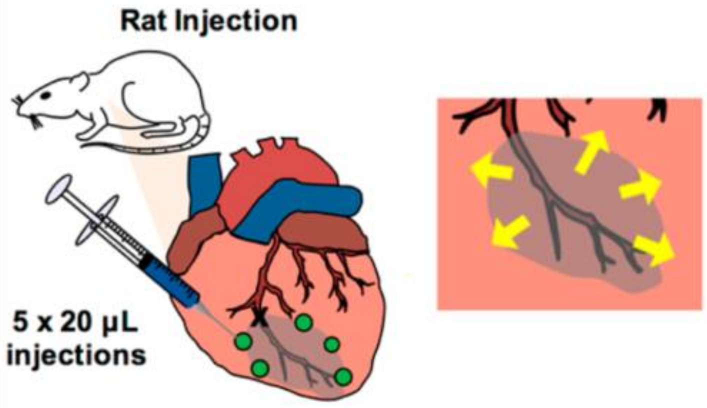
Nozzle-based 3-D printing system

Li *et. al.* (2020) Mater. Sci. Eng. R



Cell culture application

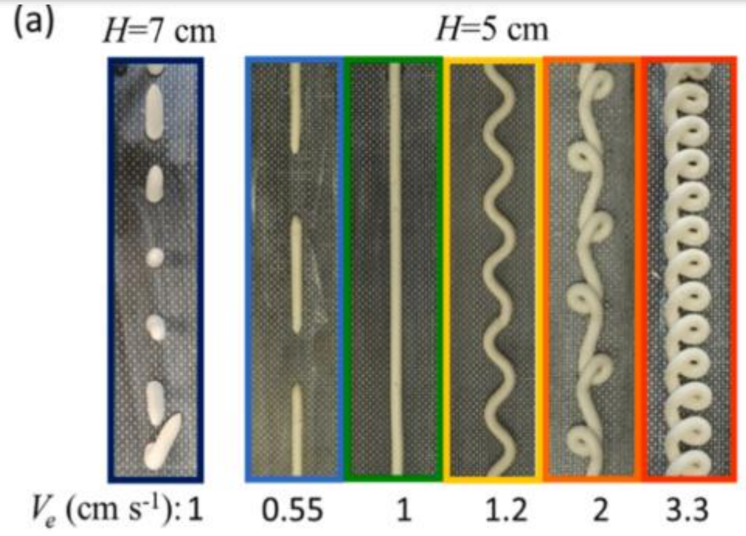
Zhu *et. al.* (2017) Adv. Funct. Mater.



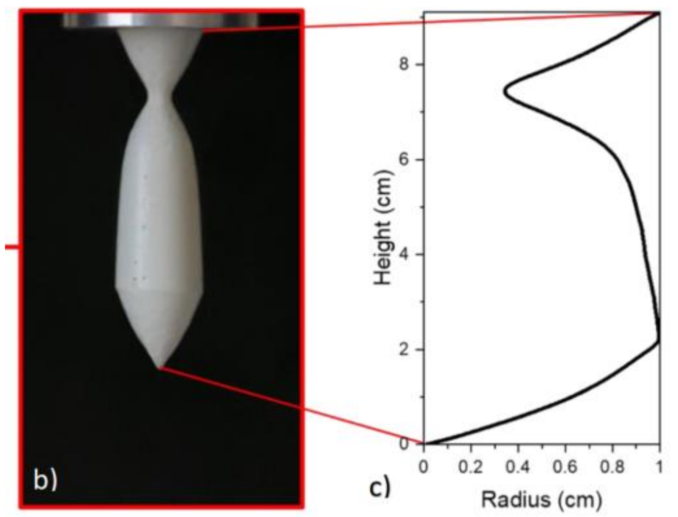
Tissue engineering

Chen *et. al.* (2017) ACS Biomater.

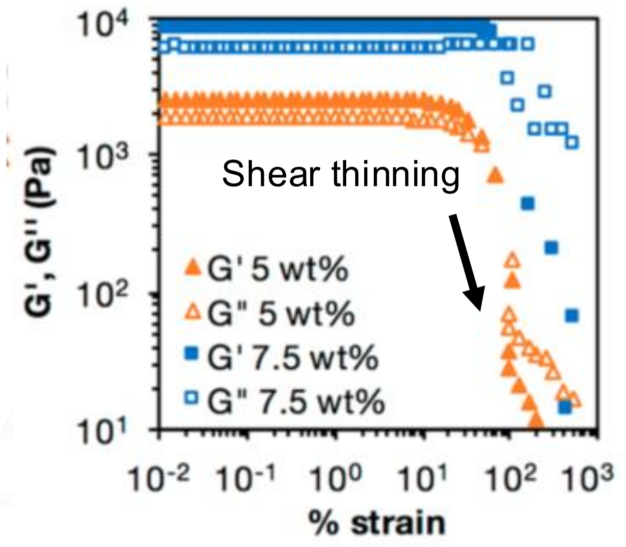
Features of our simulation



Printing outcomes depends on **inject rate** and **height**
Geffrault *et. al.* (2023) *Addit. Manuf*



Pinch-off dynamics
Geffrault *et. al.* (2021) *J. Rheol.*



Strong shear thinning property
Chen *et. al.* (2017) ACS Biomater.

Key experimental features

- **Yielding**
- **Viscoelastic behavior**
- **Solid and fluid transition**

Outline

1. A structural rheology model
2. Extension into a general tensorial form
3. Numerical results

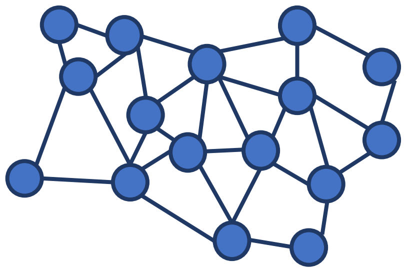
Preview of the simulation

Z. Xu & J. J. Feng. A structural model for yielding and degelation of colloidal gels. 2025, *J. Rheol.* 69, 1067–1085

Structure parameter

Structure parameter

Solid $\lambda \to 1$

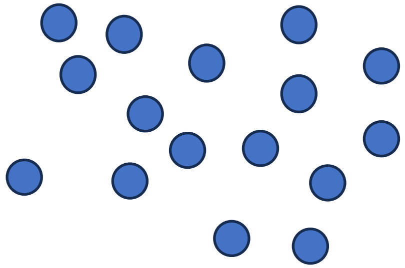


Resting – structure recovery

$$\frac{d\lambda}{dt} = k_1(1-\lambda) - k_2|\dot{\gamma}|\lambda$$

Deformation – structure breakage

Fluid $\lambda \to 0$



Strong elasticity

$$\sigma_e = \mu_s(\lambda)\gamma_e$$

$$\mu_s(\lambda) \to \mu_{s1}$$

$\mu_s(\lambda)$ -- elastic modulus
γ_e -- elastic strain

Strong viscosity

$$\sigma_v = \mu(\lambda)\dot{\gamma}$$

$$\mu(\lambda) \to \mu_1 + \mu_0$$

$\mu(\lambda)$ -- viscosity
$\dot{\gamma}$-- strain rate

Zero elasticity

$$\sigma_e = \mu_s(\lambda)\gamma_e$$

$$\mu_s(\lambda) \to 0$$

Weak viscosity

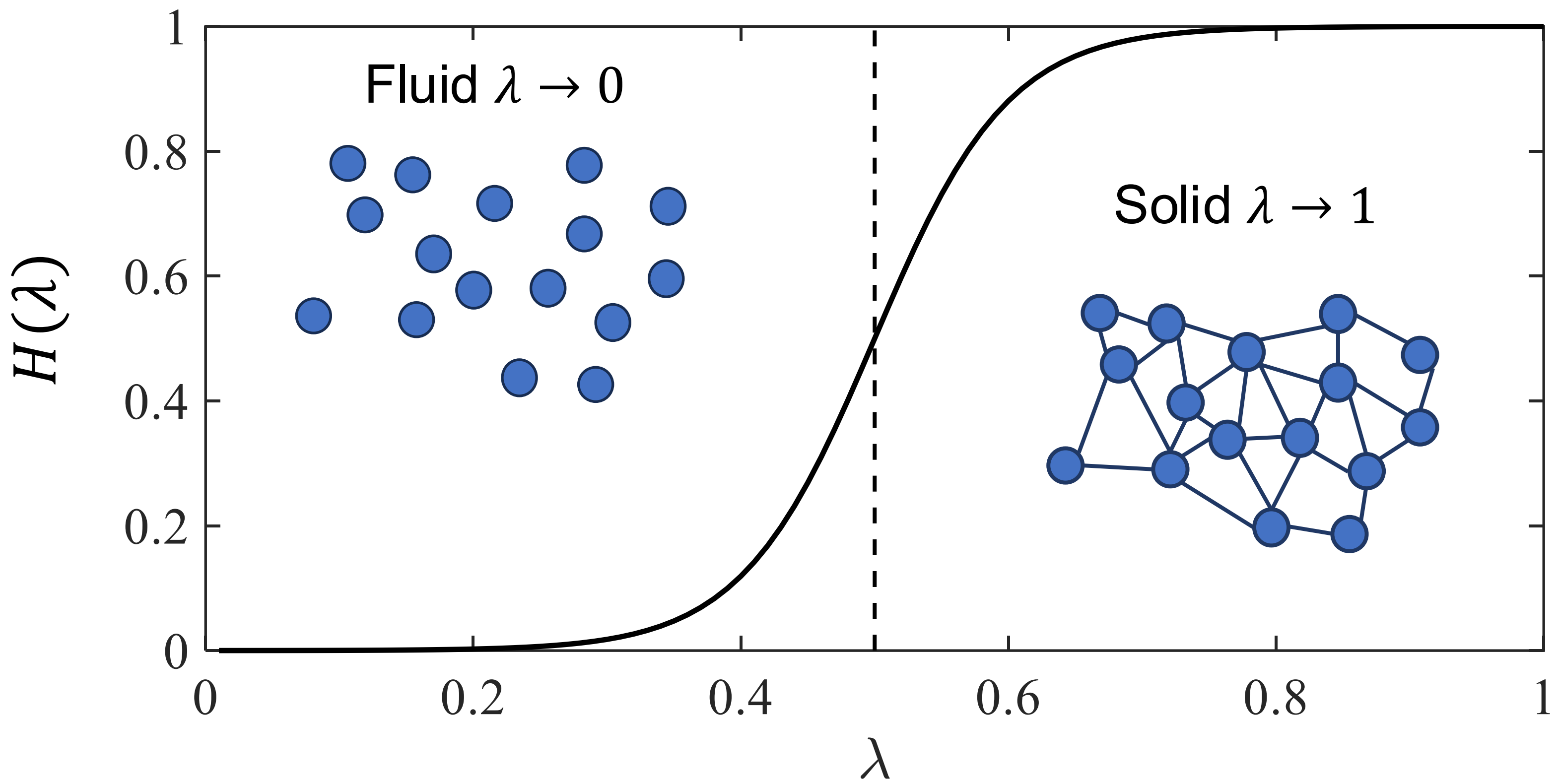
$$\sigma_v = \mu(\lambda)\dot{\gamma}$$

$$\mu(\lambda) \to \mu_0$$

Solid and Fluid transition

Stress consists of both elastic and viscous components:

$$\sigma = \mu_s(\lambda)\gamma_e + \mu(\lambda)\dot{\gamma}$$



The critical value for degelation:

$$\lambda_c = 0.5$$

When $\lambda > \lambda_c$, the elasticity and viscosity will be enhanced, *vice versa*.

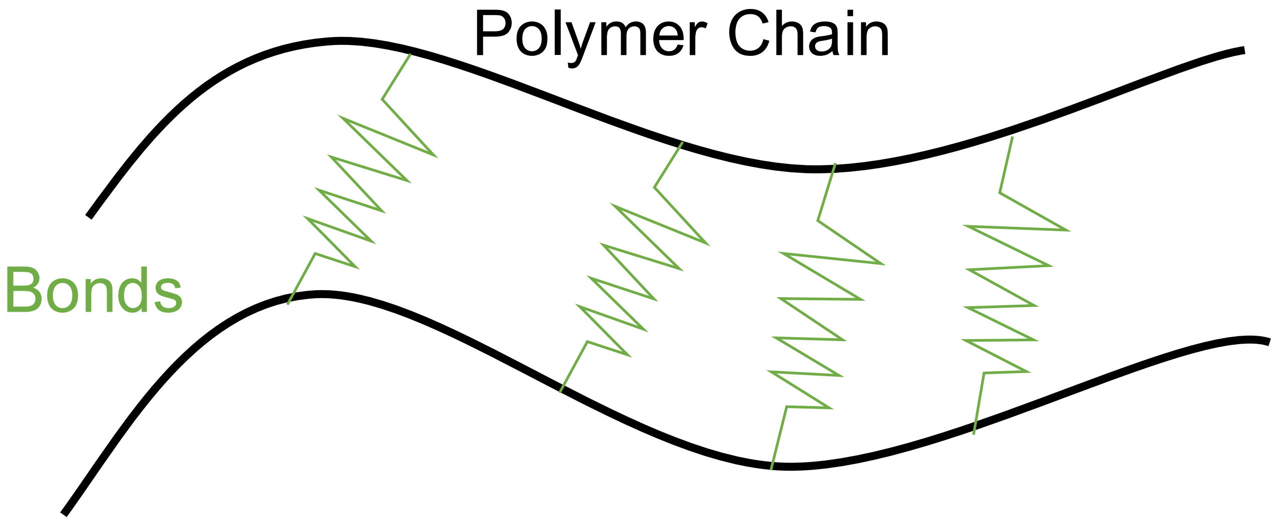
Elasticity $\mu_s(\lambda) = \lambda \cdot \mu_{s1} \cdot H(\lambda)$

Viscosity $\mu(\lambda) = \lambda \cdot \mu_1 \cdot H(\lambda) + \mu_0$

$$H(\lambda) = 0.5 + 0.5\tanh(\,(\lambda - \lambda_c)/\epsilon)$$

Innocenzi, P. *The Sol-to-Gel Transition*, 2019.

The evolution of elastic energy



Microscopic elastic energy

$$E_s = \sum_b \frac{1}{2} \mu_{s,b} \gamma_b^2$$

Macroscopic elastic energy

$$E_s = \frac{1}{2} \mu_s(\lambda) \gamma_e^2$$

$\mu_s(\lambda)$ -- averaged elastic modulus

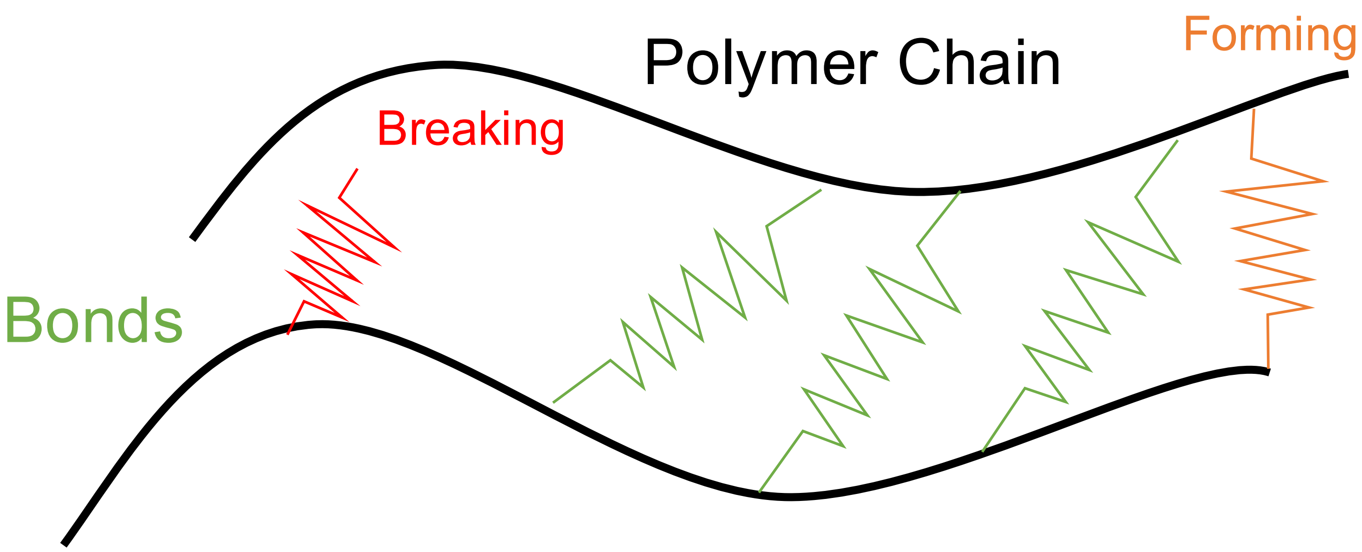
γ_e -- averaged elastic strain

Macroscopic elastic stress

$$\sigma_e = \mu_s(\lambda) \gamma_e$$

The evolution of elastic energy

Under shear rate $\dot{\gamma}$ after dt



Bond stretching **adds** elastic energy

Bond breaking **loses** elastic energy

Bond forming does **not change** elastic energy

Macroscopic elastic energy

$$E = \frac{1}{2}\mu_s(\lambda)\gamma_e^2$$

Work done by elastic stress:

$$dE_i = \mu_s(\lambda)\gamma_e \cdot \dot{\gamma}\, dt$$

Energy loss by breaking bonds:

$$dE_b = \frac{\partial E}{\partial \lambda}\dot{\lambda}_- dt$$

$$\frac{d\lambda}{dt} = k_1(1-\lambda) - \underbrace{\boxed{k_2|\dot{\gamma}|\lambda}}_{\dot{\lambda}_-}$$

The evolution of elastic energy

Macroscopic elastic energy

$$E = \frac{1}{2}\mu_s(\lambda)\gamma_e^2$$

Work done by elastic stress:

$$dE_i = \mu_s(\lambda)\gamma_e \cdot \dot{\gamma}\, dt$$

Energy loss by breaking bonds:

$$dE_b = \frac{\partial E}{\partial \lambda}\dot{\lambda}\_dt$$

Elastic energy evolves as

$$\dot{E} = \dot{E}_i - \dot{E}_b$$

$$\Downarrow$$

Governing equation for elastic strain

$$\dot{\gamma_e} = \dot{\gamma} - \frac{k_1}{2}\frac{{\mu_s}'}{\mu_s}(1-\lambda)\gamma_e$$

Elastic strain rate = Total strain rate - Plasticity due to new bond forming

Governing equations

The evolution of the structural parameter:

$$\dot{\lambda} = k_1(1-\lambda) - k_2|\dot{\gamma}|\lambda$$

Stress consists of both elastic and viscous components:

$$\sigma = \mu_s(\lambda)\gamma_e + \mu(\lambda)\dot{\gamma}$$

Solid elasticity $\mu_s(\lambda) = \lambda \cdot \mu_{s1} \cdot H(\lambda)$

Solid viscosity $\mu(\lambda) = \lambda \cdot \mu_1 \cdot H(\lambda) + \mu_0$

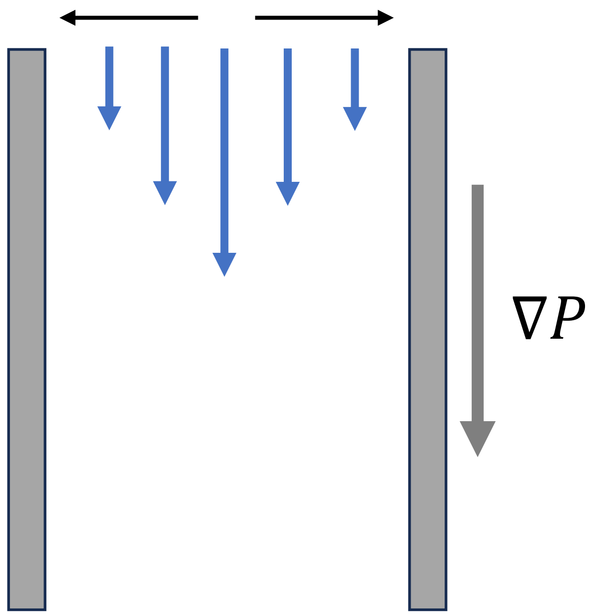
The evolution of elastic strain:

$$\dot{\gamma_e} = \dot{\gamma} - \frac{k_1}{2}\frac{G'}{G}\frac{1-\lambda}{\lambda}\gamma_e$$

1-D momentum equation:

$$\frac{d\sigma}{dy} - \frac{dp}{dx} = 0$$

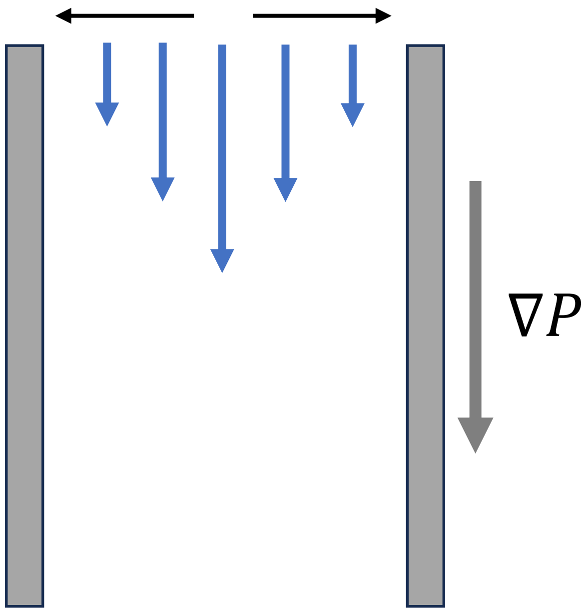
Shear stress increases from the **center to wall**



Pressure driven Poiseuille flow

Features of the model

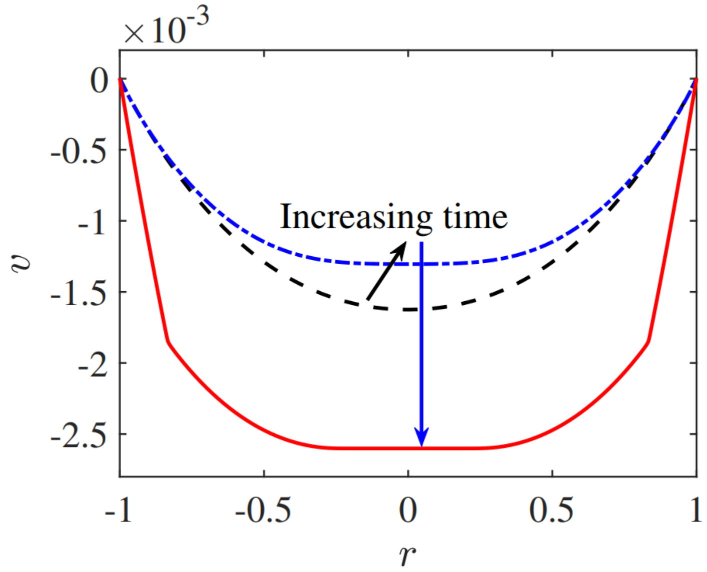
Shear stress increases
from the **center to wall**



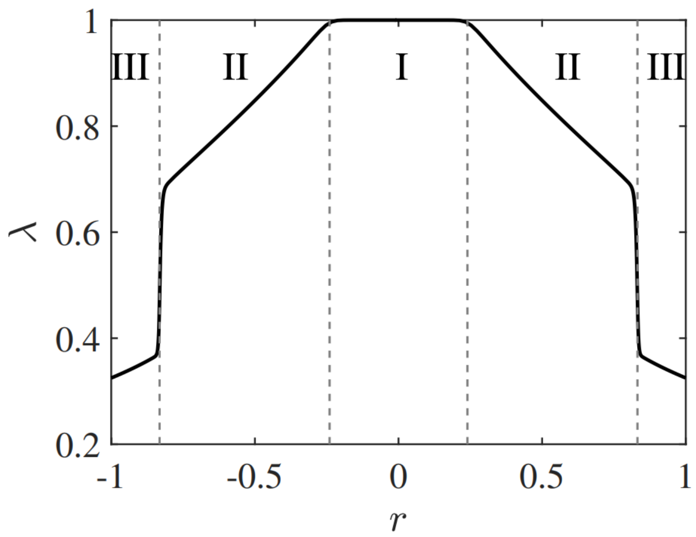
Pressure driven Poiseuille flow

Z. Xu & J. J. Feng. A structural model for yielding and degelation of colloidal gels. 2025, *J. Rheol.* 69, 1067–1085

Yielding and fluidization



Velocity profile



Structural parameter profile

(I) Solid region ➡ (II) Yielding region ➡ (III) Fluid region

$\lambda < 1$ $\qquad$ $\lambda < \lambda_c$

$\sigma_y = 2\mu_s/k_2$ $\qquad$ $\sigma_d = \dfrac{(\mu_{s0} + \mu_{s1}/4)}{k_2}$

Extension into a tensorial form

Momentum equation:

$$\nabla\boldsymbol{\sigma} - \nabla\cdot p = 0$$

Stress

$$\boldsymbol{\sigma} = \boldsymbol{\sigma}_s + \boldsymbol{\sigma}_f$$

Elastic stress $\boldsymbol{\sigma}_s = 2\mu_s\boldsymbol{e}$

Viscous stress $\boldsymbol{\sigma}_f = 2\mu\boldsymbol{D}$

$$\mu_s = \mu_{s0}\lambda H(\lambda - \lambda_c)$$

$$\mu = \mu_1\lambda H(\lambda - \lambda_c) + \mu_0$$

$$H(\lambda) = 0.5 + 0.5\tanh(\,(\lambda - \lambda_c)/\epsilon)$$

Elastic strain $\dfrac{\mathrm{d}\boldsymbol{e}}{\mathrm{dt}} = \boldsymbol{D} - \dfrac{k_1}{2}\dfrac{d\mu_s}{\mathrm{d}\lambda}\dfrac{1-\lambda}{\mu_s}\boldsymbol{e}$

Structure parameter $\dfrac{\mathrm{d}\lambda}{\mathrm{dt}} = k_1(C - \lambda) - k_2\big|\big|\boldsymbol{D}\big|\big|\lambda$

Interfacial tracking – diffuse interface

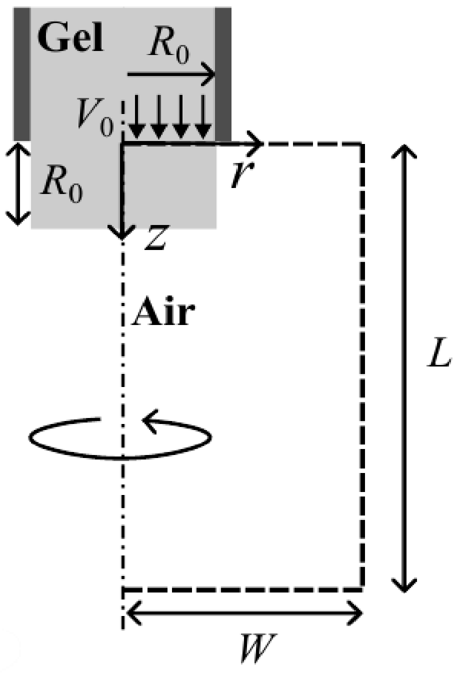
$$\frac{\partial \mathrm{C}}{\partial \mathrm{t}} + \nabla\cdot(vC) = \nabla\cdot\left(\delta\Delta C - C(1-C)\frac{\nabla \mathrm{C}}{|\nabla C|}\right)$$

Olsson & Kreiss (2005) J. Compt. Phys.

Velocity field – P2 elements; Pressure – P1 elements
Structural parameter λ – P1 element
Elastic strain – P1 element
Volume fraction C – P1 element

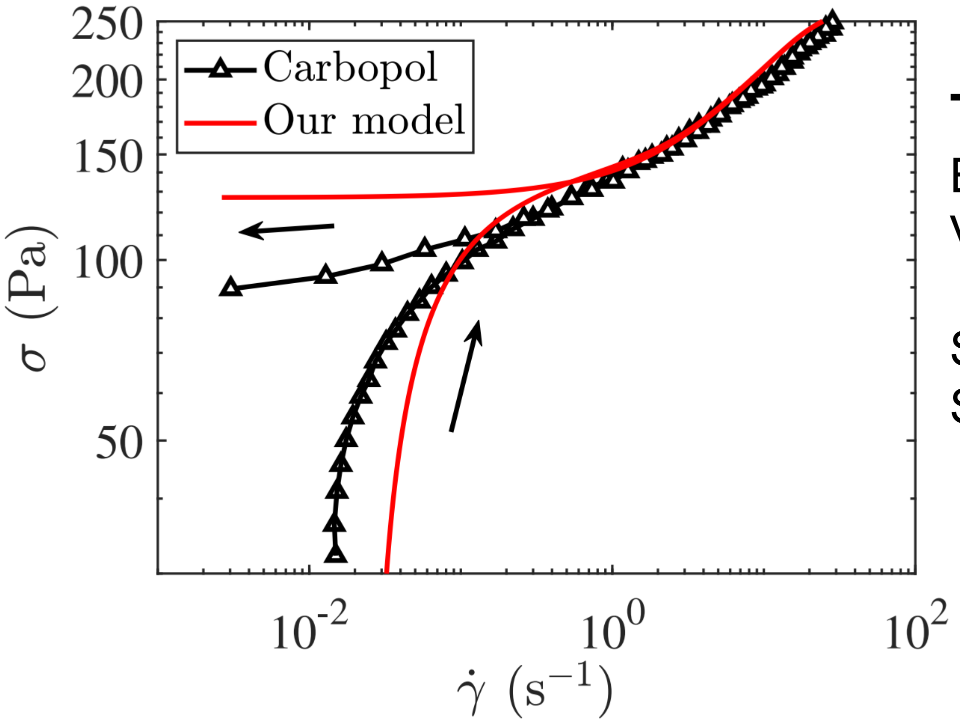
Solved by Freefem++

Numerical setup



Simulation domain

$R_0 = 5\,mm$
$L = 12R_0,\ \ W = 2R_0$

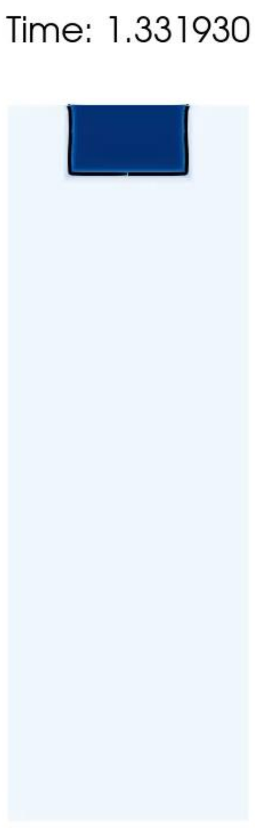


Flow curves of simple shear compare with Carbopol

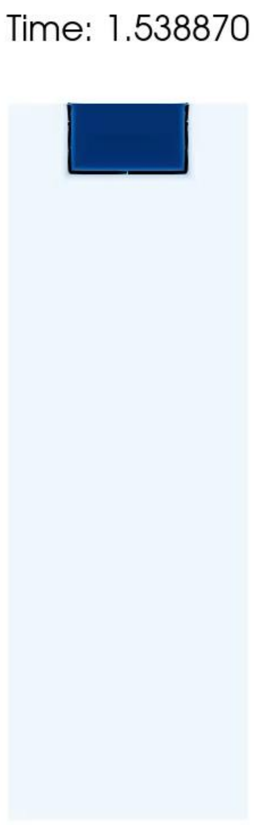
The parameters

Elasticity $\mu_{s1} = 200\ Pa$
Viscosity $\mu_1 = 15\ Pa\ s$ (before fluidization)
$\mu_0 = 5\ Pa\ s$ (after fluidization)
Structure healing $k_1 = 80\ s^{-1}$
Structure breakage $k_2 = 2.86$

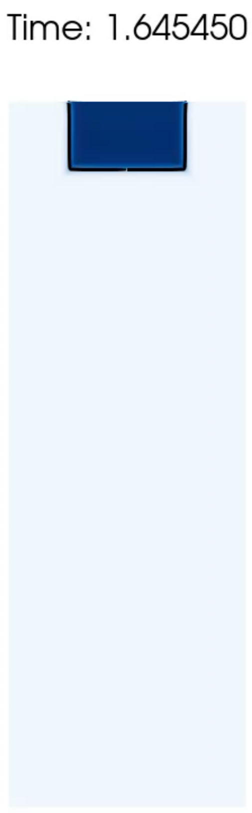
Gel printing with different elastic moduli



$\mu_s = 200\ Pa$

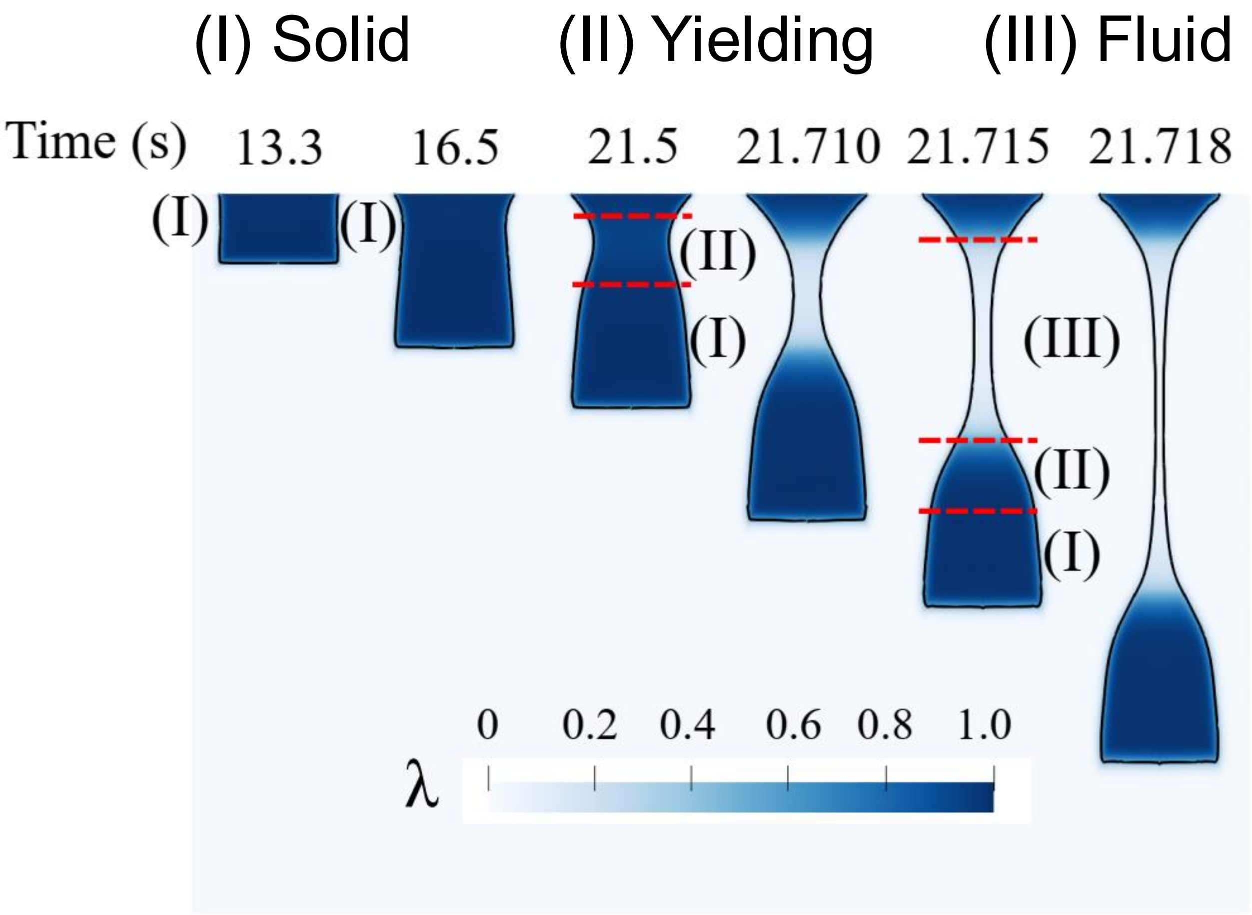


$\mu_s = 300\ Pa$



$\mu_s = 400\ Pa$

Dynamics of elongation

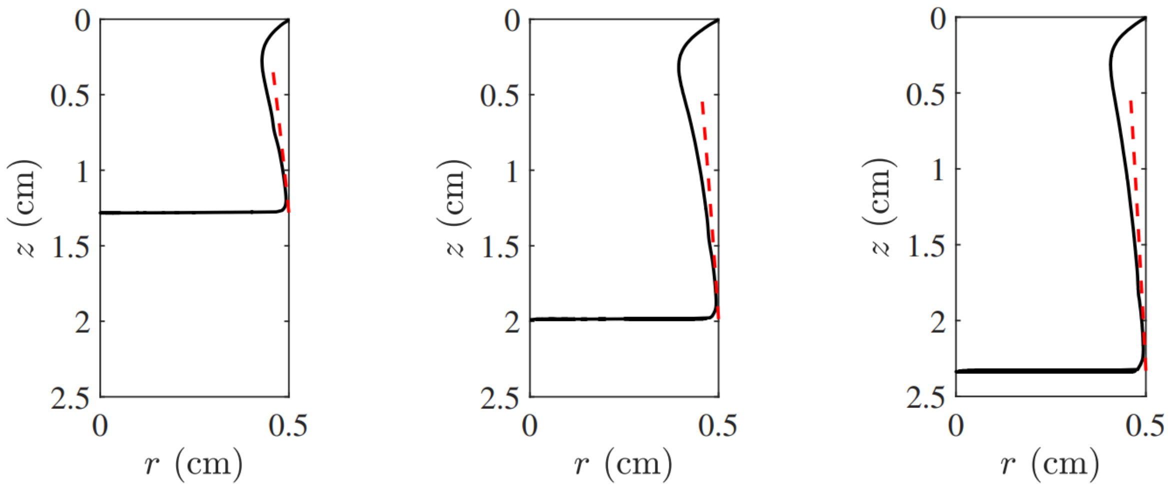


Time series of the snapshots of gel injection

Comparison with experiment

Geffrault *et. al.* (2021) *J. Rheol.*

Elastic deformation in non-yield region



$\mu_s = 200$ Pa $\mu_s = 300$ Pa $\mu_s = 400$ Pa

Elastic deformation of the solid regime

Red dashed line – theoretical elastic deformation
Black curve – numerical interface

Theoretical prediction – Pure elastic deformation

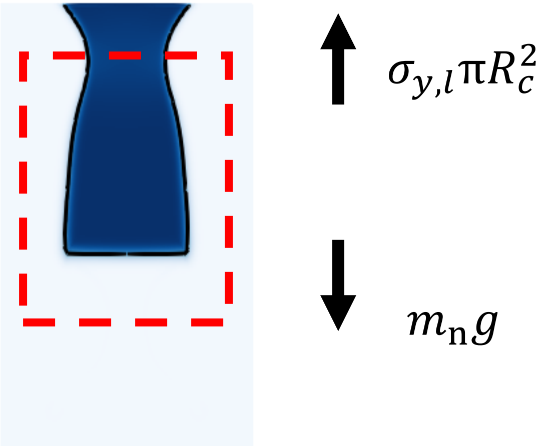
The balance between elastic stress and gravity

$$\frac{\mathrm{d}}{\mathrm{dz}}\left(\left(\sigma_{s,\mathrm{zz}} - p\right) \cdot \pi R(\mathrm{z})^2\right) + \rho g \cdot \pi R(\mathrm{z})^2 = 0$$

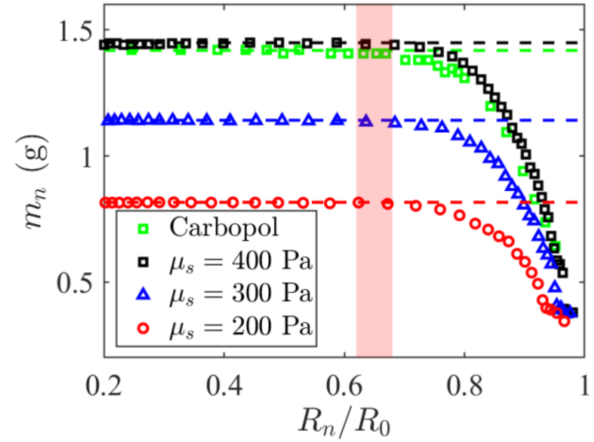
The deformation due to elastic deformation:

$$\frac{\mathrm{d}R(z)}{\mathrm{dz}} \approx \frac{\rho g R_0}{6u_s}$$

Elongational yield stress



The mass below the thinnest point



The mass VS neck radius

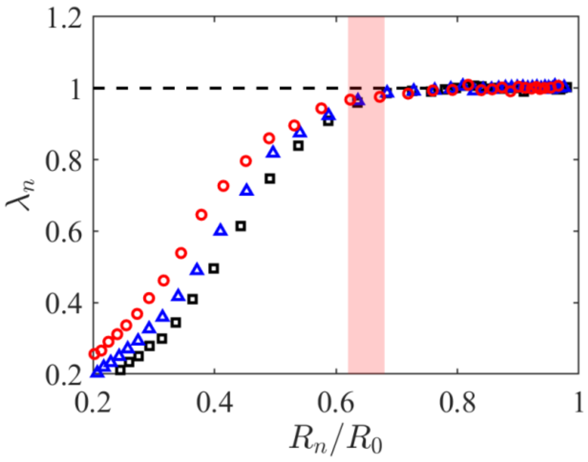
Theoretical yield stress of our model :

$$\sigma_{y,l} = 2\sqrt{3}\frac{\mu_s}{k_2}$$

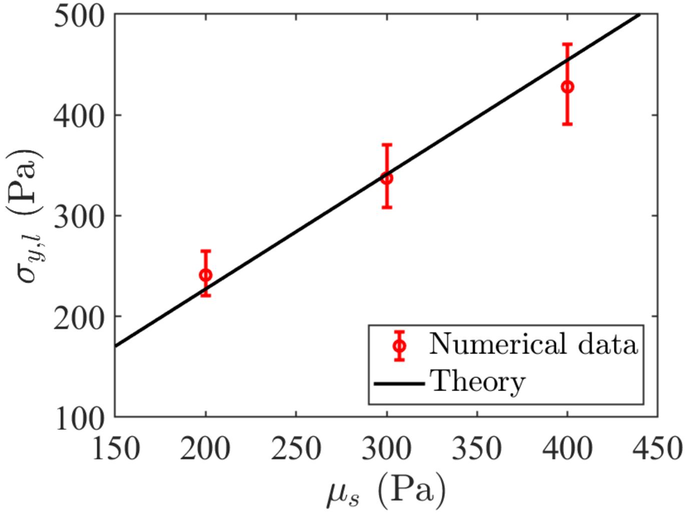
The experimental measured yield stress:

$$\sigma_{y,l} = \frac{m_c g}{\pi R_c}$$

Geffrault et al. (2023) J. Rheol.



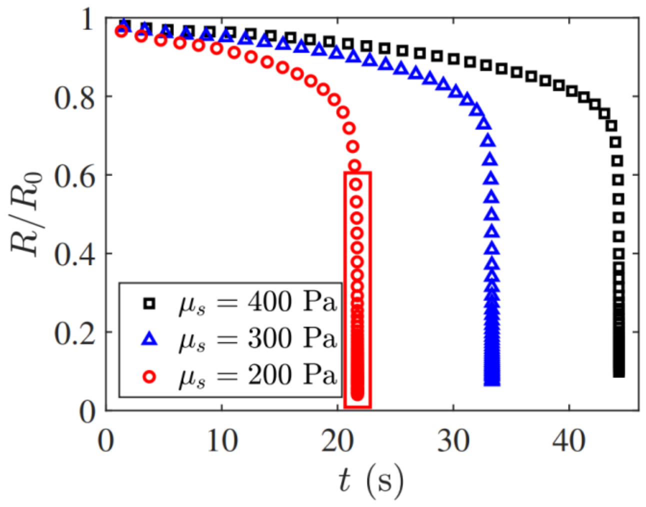
Local λ_n VS neck radius



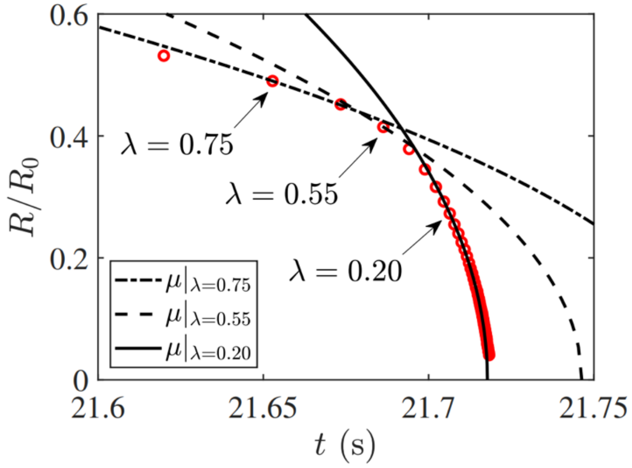
Comparison of the yield stress

Simulation helps determine $\boldsymbol{R_c}$

Viscous elongation



The evolution of neck thinning



Magnified view and theoretical prediction

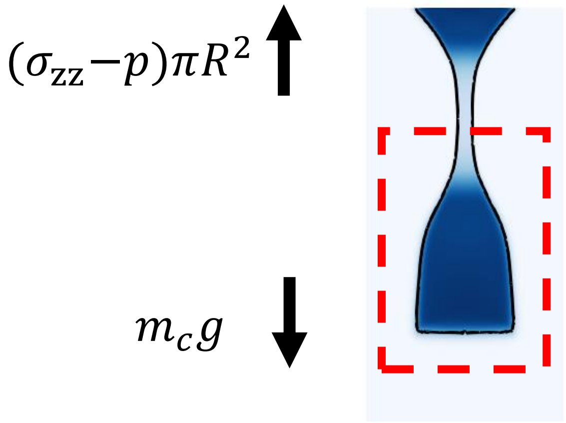
The balance between viscous stress and gravity:

Governing ODE:

$$6\pi\mu R\dot{R} = -m_c g$$

Theoretical solution:

$$R(t) = \left(R_s^2 + \frac{m_c g}{3\pi\mu(\lambda)}(t_s - t)\right)^{1/2}$$



R_s & t_s are the **initial condition** to specify the solution

Viscosity $\mu(\lambda)$ is determined by **structural parameter**

Conclusion

- The structural rheology model has captured features of yielding and fluidization;
- The 2-D simulations have revealed the dynamics after gel exiting the nozzle:
  1. Elastic deformation
  2. Yielding transition
  3. Viscous elongation

  The simulation results are validated by **experiments** and **theoretical solution**.