TRANSITION TO NON-NEWTONIAN DYNAMICS IN DROPS OF POLYMER SOLUTIONS

VIRGILE THIEVENAZ, SREERAM RAJESH, NATHAN VANI AND ALBAN SAURET

a: PMMH, CNRS, ESPCI Paris, Université PSL, Sorbonne Université, Université Paris Cité, F-75005, Paris, France **b**: Department of Mechanical Engineering, University of California, Santa Barbara, California 93106, USA Contact: virgile.thievenaz@espci.fr — website: vthievenaz.fr

THE COIL-STRETCH TRANSITION

At rest, a polymer chain tends to take the shape of a coil since it maximizes its entropy. However, in a flow with a strong velocity gradient the free energy minimum corresponds to the unwinding and stretching the chain. For a critical value of the local rate of strain occurs the coil-stretch transition.



ABSTRACT

Many natural or industrial flows involve the breakup of liquids into droplets, and many of these liquids contain polymers or suspended grains. At low scales and high strain, the flow reveals the composition of the liquid, since the components may deform, organize, separate, etc, and thereby change the rheology. We first study the pinch-off of drops of polymer solutions; we show that the transition between the well-known Newtonian and viscoelastic regimes follows a universal dynamic, linked to the coil-stretch transition. Then, we add spherical particles (diameter 20-250µm) to the solution. Their presence increases the local strain rate felt by the polymer and so enhances the coil-stretch transition. This experiment enables the measure of the amplification of strain in the fluid phase of a granular suspension; we perform these measurements for monodisperse and bidisperse suspensions.

THINNING AND DROP DETACHMENT

Depending on the state of the chains, the thinning of a polymer solution may follow two distinct dynamics. In the coiled state, polymer chains act like rigid beads and increase the fluid viscosity although the flow remains Newtonian. In the stretched state, their elasticity becomes relevant and the flow becomes viscoelastic.



De Gennes, **Coil-stretch transition of dilute flexible polymers under ultrahigh velocity gradient**, J. Chem. Phys. 60 (1974)

READ ABOUT IT

1- Thiévenaz and Sauret, Pinch-off of viscoelastic particulate suspension, Physical Review Fluids 6 (2021)

2- Rajesh, Thiévenaz and Sauret, Transition to the viscoelastic regime in thinning of polymer suspension, Soft Matter 18 (2022)

3- Thiévenaz, Vani and Sauret, Caging and fluid deformations in dense bidisperse suspensions, in revision, ArXiV 2311.09112 (2023)

PINCH-OFF OF A DROP OF POLYMER SOLUTION

Drop of a mixture of water (75%) and glycerol (25%) in which polyethylene oxide (M_{w} = 300 kg/mol) has been dissolved at weight fraction 1%. The nozzle is 2.75mm-wide.



The same solution as on the left, adding 140 μ m-big polystyrene beads at volume fraction 40%.





UNIVERSAL TRANSITION TO THE VISCOELASTIC REGIME

We follow the thinning dynamics h(t) for solutions of polyethylene oxide in water-glycerol mixtures. We vary the molar mass (300 or 4000 kg/mol), the

STRAIN RATE AMPLIFICATION BY DISPERSED PARTICLES^{1,3}

We add polystyrene beads (diameter 20-250µm) to the polymer solution. In the Newtonian regime, adding solid particles slows down the thinning; the suspension behaves exactly like an equivalent fluid of identitcal viscosity. In the viscoelastic regime, particles have no effect on the relaxation time, likely because the polymer chains are much smaller, even stretched. Adding particles does not change the universal transition, only the value of $\hat{\varepsilon}_{\rm c}$.¹





Because the coil-stretch transition is driven by the flow at the polymer chain scale, its microscopic threshold $\dot{\varepsilon}_{c,0}$ should not depend on the particle volume fraction, and can be measured when there is no particles. Therefore, we can use the polymer as a **strain** probe to measure the deformations of the fluid in a dense suspension; it also works in bidisperse suspensions (particles of two sizes). The caging of small particles, if they are small enough, by the large ones, then appears to be important. 3

 $\dot{arepsilon}_{
m c}/\dot{arepsilon}_{
m c,0}$

 $\dot{arepsilon}_{
m c}/\dot{arepsilon}_{
m c,0}$

We show that the strain rate dynamics at the transition is universal. The critical strain rate at the transition $\dot{\varepsilon}_c$ also defines the transition time scale, which differs significantly from the relaxation time λ .





