

# Rheology of mucin films for molluscan adhesive locomotion

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## INTRODUCTION

Snails and slugs depend on the rheological properties of their pedal mucus for locomotion. These gastropods use a unique method to crawl called adhesive locomotion. Unlike an inchworm, or humans for that matter, no part of the animal is lifted from the ground to create differential friction. Rather, the mollusk exerts shear stresses on the thin layer of mucus holding it to the substrate. The pedal mucus exhibits an effective yield stress; under high applied stresses the network structure breaks enabling the foot to glide forward over a liquid-like layer, whereas in regions of low applied stress the network structure reforms into a solid layer connecting the foot to the substrate. Our current work explores the rheological properties required for adhesive locomotion, and is motivated by a robotic snail (RoboSnail2 developed by Hosoi and coworkers) that uses the same locomotive technique. Rheological measurements of natural mucin films shows that they are elasto-visco-plastic in nature with the characteristics of a physically crosslinked gel below the yield point and a strongly rate-dependent (or stress-dependent) apparent viscosity above a critical 'yield stress'. A simple model shows that any non-Newtonian fluid could be used for horizontal adhesive locomotion and a measure of efficiency is introduced to compare simulants. For inclined and inverted locomotion (e.g. wall climbing), a yield stress fluid is required. The rheological number of possible mucus simulants are contrasted with corresponding measurements of natural molluscan mucin films.

## BACKGROUND & EXPERIMENTAL METHODS

The rheological properties of invertebrate mucin films were first measured experimentally by Denny [1]. Natural pedal mucus films have a thickness of 10–20  $\mu\text{m}$  and are physically-crosslinked gels composed of 5–10% hydrated muco-polysaccharides in water [2]. The films have pronounced viscoelastic properties and a yield stress of 500 – 1500 Pa. More recently Taylor et al. [3] have studied the nonlinear rheology of pig gastric mucus films and mucin alginate gels using oscillatory stress sweeps. They note that these physically-crosslinked mucin gels exhibit a novel frequency-dependent stress-hardening response as the amplitude of the imposed deformation is increased. An analogous stress-hardening response in invertebrate mucin films would be important in controlling the efficiency of adhesive locomotion, however it has not been studied to date.

We use a controlled stress rheometer (TA Instruments AR1000) to impose large amplitude oscillatory stress (LAOS) sweeps of the form  $\sigma = \sigma_0 \sin(\omega t)$ . The native film depths (typically 10–20  $\mu\text{m}$ ) are below the measurement resolution of conventional rheometric fixtures. We utilize 8mm or 20mm diameter parallel plate fixtures with sample sizes of 100–200  $\mu\text{m}$  thickness that are formed by aggregating the native mucal secretions deposited naturally by the leopard slug (*Limax maximus*) as it crawls across the Peltier plate of the rheometer. Complementary measurements of mucin rheology as a function of film thickness are also being performed using a flexure-based microrheometer FMR described elsewhere [4].

The measured (and generally nonlinear) oscillatory deformation response can be represented in the general form of a complex compliance composed of a sequence of Fourier terms:

$$J^*(\omega; \sigma_0) = \frac{\gamma(t)}{\sigma_0} = \sum_i J_i'(\omega) \sin(\omega t) + J_i'' \cos(\omega t)$$

where  $J_i', J_i''$  are, respectively, the in-phase (elastic) and out of phase (viscous) compliances associated with each harmonic contribution. The leading order terms ( $i = 1$ ) correspond to the

conventional linear viscoelastic response functions and can be inverted to evaluate the more familiar viscoelastic storage modulus and loss modulus through the relationship  $J_i^*(\omega) G_i^*(\omega) = 1$ . As the material response becomes progressively more nonlinear the contribution of the higher harmonics become increasingly important and this transition can be systematically monitored using this Fourier-transform-based approach.

## LAOS RESULTS

Representative results from oscillatory stress sweeps are shown in Figure 1 & 2 below. For sufficiently small stress amplitudes that are below the yield stress ( $\sigma_0 \ll \sigma_y$ ) the mucin film is a viscoelastic solid with  $G_1' \sim 10G_1''$  and all other harmonic contributions negligible. As the stress amplitude is increased this ratio decreases and the material undergoes a rapid viscoplastic collapse at stresses  $\sigma_0 \geq 1000$  Pa.

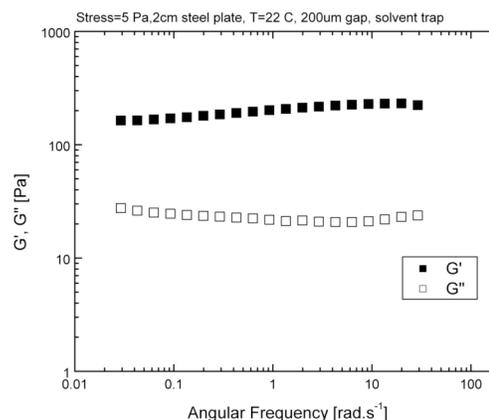


Figure 1. linear viscoelastic properties obtained from a frequency sweep of mucin slime from a leopard slug.

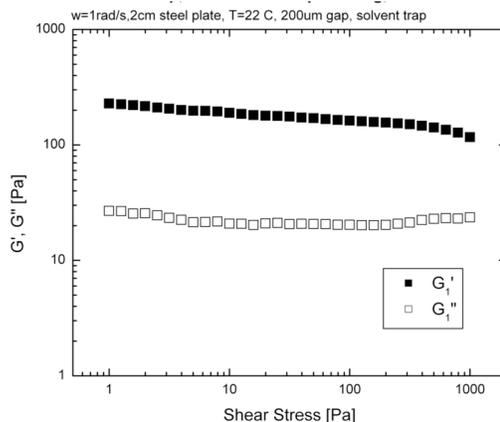
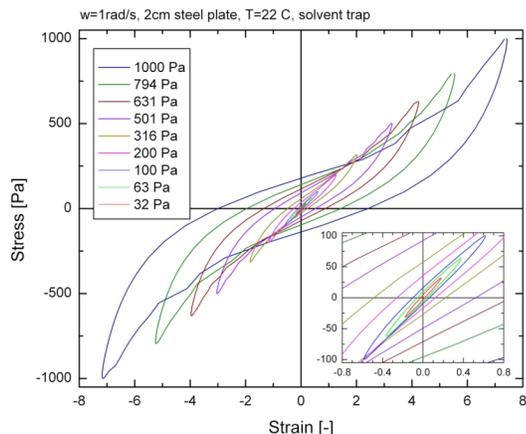


Figure 2. Linear viscoelastic moduli as a function of applied stress,  $\sigma_0$  at a frequency of 1 rad/s characteristic of adhesive locomotion.

The nonlinear characteristics of the material response can be represented more clearly in the form of Lissajous figures in which the stress and strain are plotted against each other as shown in Figure 3 (overleaf).

The linear and predominantly elastic response is evident from the increase in the instantaneous slope  $d\sigma/d\gamma$  at large stresses and strains. This local stress-hardening results from deformation of the chains in the physically-crosslinked mucin network. As the stress in the

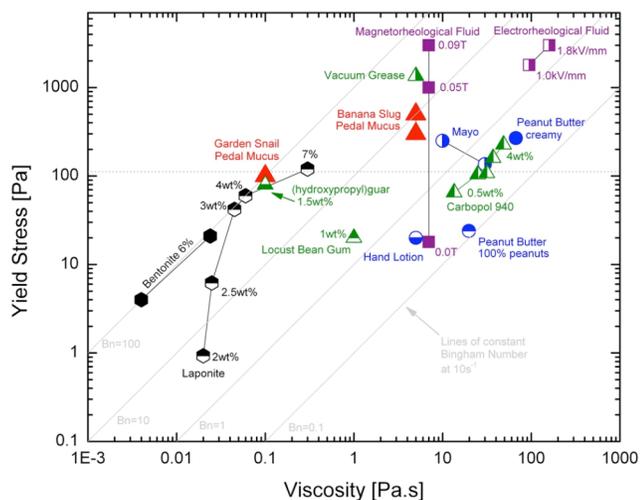
gel is increased towards 1 kPa the elastically-active chains are detached from the network and this results in increasing viscous dissipation. In the Lissajous figure this dissipation is related to the area under the curve during a single oscillatory stress cycle. By performing a series of similar stress sweeps as a function of different imposed frequencies it is possible to develop a complete 'rheological fingerprint' of the response of the mucin film. This fingerprint can be compactly represented in terms of a modified *Pipkin diagram* with contours of the viscous and elastic properties as a function of imposed stress and frequency. The yielding transition in such a rheologically-complex material is, in reality, a 'yield surface' that varies with the frequency of the imposed deformation.



**Figure 3.** Large amplitude oscillatory stress sweep for mucin slime film deposited by the Leopard slug (*Limax maximus*).

#### IMPACT OF RHEOLOGY ON ADHESIVE LOCOMOTION

These complex visco-elasto-plastic rheological properties of mucin films control the efficiency of locomotive adhesion, and also provide a reference point for the formulation of synthetic slime simulants that might be deployed by a robotic crawler. Two of the most important properties are the apparent yield stress of the material and its post-yield flow viscosity which controls the total viscous dissipation in the sheared film. Evidently a yield stress is required for stable inclined locomotion, and a line of minimum yield stress, can be drawn for a specified design platform. For the Robosnail project this critical value is O(100 Pa) and is shown in the figure below by a dotted line.



**Figure 4.** A two parameter design space for 'yield stress fluids' that might serve as mucin simulants. The native rheological properties of two species of gastropods are shown by the red triangles.

Furthermore, the post-yield viscosity should be minimized to reduce dissipation and increase the crawling speed for a given imposed actuator force. For a non-Newtonian fluid, the post-yield viscosity is not necessarily a constant, so viscosity values have been taken at a characteristic shear rate of  $\dot{\gamma} \sim 10s^{-1}$ . This is the order of magnitude of the shear rate for Robosnail with  $V \sim 1cm/s, \sim 1mm$ . These parameters are used to construct a two-dimensional design space which can rank the relative merits of different simulants as shown in Figure 4. A number of structured fluids have been surveyed, including biopolymer gels, synthetic polymer gels, particulate gels, emulsions, electrorheological fluids, and magnetorheological fluids. Lines of constant Bingham Number are plotted as a guide to the eye.

$$\text{Bingham Number } Bi = \frac{\sigma_{\text{yield}}}{\eta \dot{\gamma}} \sim \frac{\text{supporting force}}{\text{resisting force}}$$

Optimal propulsive efficiency for mechanical crawlers is obtained for high  $Bi \gg 1$  corresponding to large yield stresses and very little dissipation of mechanical energy through viscous effects. A simple two-dimensional diagram such as figure 4 does not permit us to capture the time-dependent viscoelastic structural recovery (i.e. "thixotropic elasticity") of natural glycoprotein gels and we are presently exploring appropriate three-dimensional representations of simulant properties. We determine the restructuring time via transient step strain-rate and creep/recovery tests with varying periods of recovery. For natural mucin gels, this restructuring time appears to be on the order of 1 second.

The insight obtained from complete rheological characterization of natural molluscan mucin films allows us to select optimal simulants that can be deposited by a robotic crawler that can successfully climb walls and travel with maximum efficiency. Having determined the rheological response of the materials, it is then possible to consider optimizing the shape of the imposed driving wave, as discussed by Hosoi and coworkers in accompanying work..

#### ACKNOWLEDGMENTS

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