Supplementary Information

Unstable crack propagation in Laponite gels: selection of a sinusoidal mode in an electric field

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Laponite gel of 6.29 wt% is prepared by adding 2.5 gm of Laponite clay to 40ml of deionized water and stirred in a magnetic stirrer for about 30s. The solution is initially turbid in nature but changes to a clear gel within a few minutes. This gel is used to perform the rheological experiments.

Storage and loss moduli of Laponite gel were measured using MCR102 SN81260812 for the frequency range 0.1 to 100 Hz. The results are shown in Figure 1. Further details will be published elsewhere. The storage modulus G' lies above the loss modulus G'' throughout the range, indicating predominantly solid-like behaviour. The apparent viscosity variation with frequency is given in Figure1c and it shows a monotonic decrease.

We attempt to reproduce the rheology results using a visco-elastic model with a minimum number of parameters. A 3-element Boltzmann model (Figure 1a) as discussed in¹ with a non-Newtonian character, attributed to the fluid component is found to fit the experimental data quite well. Simpler 2-element models like the Maxwell or Voigt models fail to give a realistic description of the system.

Figure 1a shows a schematic representation of the model. It consists of a Voigt element (i.e. a spring-dashpot pair in parallel) in series with another spring. It has been shown that introducing a fractional order time derivative (q) in the constitutive equation corresponding to the dashpot gives a realistic representation of a non-Newtonian fluid in the dissipative element^{2;3}. The parameter η represents the viscous element and E_0 , E_1 the elastic elements of the Boltzmann model.

Introducing the relations $a_1 = \frac{\eta}{E_0 + E_1}$, $m = \frac{E_0 E_1}{E_0 + E_1}$ and $b_1 = \frac{E_0 \eta}{E_0 + E_1}$, the equations for G' and G'' are obtained as follows:

$$G^*(\omega) = G'(\omega) + iG''(\omega)$$

where

$$G'(\omega) = \frac{m + (a_1m + b_1)\omega^q \cos(q\pi/2) + a_1b_1\omega^{2q}}{1 + 2a_1\omega^q \cos(q\pi/2) + a_1^2\omega^{2q}}$$
(1)

and

$$G''(\omega) = \frac{(b_1 - ma_1)\omega^q \sin(q\pi/2)}{1 + 2a_1\omega^q \cos(q\pi/2) + a_1^2\omega^{2q}}$$
(2)

G' and G'' calculated with best-fit parameters $E_0 = (10.045 \pm 0.126) \times 10^3$ Pa,

 $E_1 = (12.155 \pm 0.185) \times 10^3$ Pa, $\eta = (14.63 \pm 0.181) \times 10^3$ Pa.s and q = 0.8 are compared with experimental results in Figure 1b. The results remain invariant upto a change of the order of 0.001 in the value of q. The apparent viscosity is calculated from this model and the results are in close agreement with experimental values as shown in Figure 1c.

The shear wave velocity 4 of sound follows from the equation:

$$v = \sqrt{\left\{\frac{Re|G^*|}{\rho}\right\}} \tag{3}$$

It's variation with the angular frequency, as obtained from our theoretical modelling is shown in Figure 1d.

References

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Figure 1: The 3-element Boltzmann model for a visco-elastic medium is shown in (a). The storage and loss moduli for varying angular frequencies both calculated from model (a) (red and blue symbols) and experimental values (black and green symbols) are shown in (b). (The errors in the experimental data are less than the symbol sizes.) The apparent viscosity both calculated from model (a) (line) and experimental values (symbols) are shown in (c). The sound wave velocity from model is shown in (d).