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Sheared foam as a supercooled liquid?

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Abstract. – We conduct numerical simulations on a simple model of a two-dimensional steady-state sheared foam, and define a quantity Γ that measures stress fluctuations in the constant-area system. This quantity reduces to the temperature in an equilibrium system. We find that the relation between the viscosity and Γ is the same as that between viscosity and temperature in a very different system, namely a supercooled liquid. This is the first evidence of a common phenomenon linking these two systems.

A liquid foam consists of gas or liquid bubbles suspended in an immiscible liquid at a packing fraction that exceeds random close-packing. A quiescent foam has a nonzero static shear modulus, because the thermal energy is negligible compared to the energy barrier required to change the relative positions of bubbles. When a foam is sheared, however, it can be characterized by a well-defined viscosity. As the shear rate is lowered towards zero, the viscosity diverges. This behavior is reminiscent of a supercooled liquid, where the viscosity increases rapidly as the temperature is lowered towards the glass transition [1]. These two systems are completely different: a sheared foam is a driven, athermal system at steady state, while a supercooled liquid is a quiescent thermal system. However, in both cases, the systems are jamming (*i.e.* the systems are spontaneously restricting themselves to a small part of phase space). Other driven, athermal systems such as granular materials [2] also jam. We therefore ask: does the common phenomenon of jamming lead to any common behavior in driven, athermal systems and in quiescent, thermal systems?

Foam is a particularly simple athermal system that jams, because it can be driven homogeneously by steady shear flow. As foam is sheared, there are fluctuations caused by rearrangement events where bubbles change their relative positions [3–5]. In an *equilibrium* system, temperature is a measure of the size of fluctuations relative to how easy it is to create a fluctuation (a response function). This property of temperature is embodied in the linear response relation that connects fluctuations in the stress on the boundary normal to the y-direction, σ_{yy}, to the yy compression modulus [6, 7]

$$h\frac{\partial\langle\sigma_{yy}\rangle}{\partial h} = \frac{1}{\rho k_{\mathrm{B}}T}\left\langle(\sigma_{yy} - \langle\sigma_{yy}\rangle)^2\right\rangle, \tag{1}$$

where h is the fixed height of the system in the y-direction and ρ is its density. One way to compare the non-equilibrium sheared foam to a supercooled liquid would be to compare the functional form of the divergence of viscosity *vs.* temperature in the supercooled liquid to the form of viscosity *vs.* shear strain rate in the foam. However, strain rate and temperature are very different variables and cannot be compared directly.

We conduct simulations on a simple model of foam [8], and measure a quantity Γ that characterizes stress fluctuations due to rearrangement events in a two-dimensional, constant-area system that is steadily sheared in the x-direction with the shear gradient in the y-direction. In our non-equilibrium, steady-state system, we define Γ by analogy to eq. (1), so that it corresponds to the temperature in a non-driven, equilibrium system [9]:

$$\Gamma = \frac{\left\langle (\sigma_{yy} - \langle \sigma_{yy} \rangle)^2 \right\rangle_h}{\rho h \partial \langle \sigma_{yy} \rangle / \partial h} \,. \tag{2}$$

Note that Γ depends on shear rate and should not be interpreted as an effective temperature since there is no reason to believe that the equilibrium relation eq. (1) holds for our system. However, Γ does provide a measure of the size of fluctuations relative to how easy it is to create a fluctuation in our system, just as temperature does for an equilibrium system. In this sense, Γ provides a reasonable measure of the ability of the system to overcome energy barriers.

We measure Γ and the viscosity η as a function of shear rate. Our central result is that we find that the relation between viscosity and Γ is the *same* as the relation between viscosity and temperature in a supercooled liquid. This remarkable finding is the first quantitative evidence that a common mechanism may underlie jamming in a driven, athermal system and supercooled liquids.

Our simulations are carried out on a model introduced by Durian [8]. In two dimensions this model treats bubbles as circles interacting via two types of interactions. The first interaction is purely repulsive and originates physically in the energy cost to distort bubbles. If the distance between the centers of two bubbles is less than the sum of their radii, they will distort to avoid overlap. This gives rise to a harmonic repulsive force [10, 11]. The model therefore assumes that bubbles always remain circles, but includes the elastic energy of a compressed spring between two overlapping bubbles. The second interaction is a frictional force proportional to the velocity difference between neighboring bubbles. By modeling the dissipation in the thin liquid films and Plateau borders as a dynamic friction, we neglect lubrication effects that could be important in determining the stress/strain-rate relations for real foams [12]. However, this simple model already gives rise to remarkably complex behavior. Moreover, the regime in which the viscosity is high (the low shear rate regime) corresponds to the regime where elastic effects dominate viscous ones, so the specific dissipation mechanism may be unimportant there.

In the discussion that follows, we will scale all energies (such as Γ) by kr^2, where k is a characteristic spring constant and r is the average bubble radius. The strain rate $\dot{\gamma}$ is scaled by a characteristic time $\tau = b/k$, where b is the friction coefficient. Thus, $\dot{\gamma}$ is the Deborah number, or equivalently for this system, the capillary number.

Given the two types of forces on bubbles, we can solve the equations of motion [8] numerically and propagate the bubble positions forward in time, as in a molecular-dynamics simulation. Our simulations are conducted on polydisperse bubbles whose radii are drawn from a flat distribution of a specified width, and whose spring constants vary inversely with radius. We use square samples, with periodic boundary conditions in the x-direction and confining walls (separated by the fixed distance h) in the y-direction. The bubbles along the

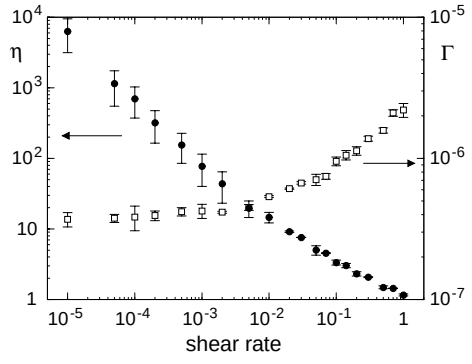

Fig. 1 – The quantity Γ (open squares, right axis) and the viscosity η (solid circles, left axis) as functions of the dimensionless shear rate $\dot{\gamma}$ (Deborah number). Neither one obeys simple power law behavior. Γ approaches a constant and η diverges in the limit $\dot{\gamma} \to 0$. All quantities are dimensionless as explained in the text.

confining walls are glued to them, and the system is sheared by moving one of the two walls at fixed velocity in the x-direction. We then measure the force per length in the y-direction on the wall (this is $-\sigma_{yy}$) and the force per length in the x-direction required to keep the wall moving at fixed velocity (this is σ_{xy}, the xy-component of the stress). These fluctuate in time as the system is sheared. The viscosity is defined by $\eta = \langle\sigma_{xy}\rangle/\dot{\gamma}$, where $\dot{\gamma}$ is the strain rate. In order to measure $\partial\langle\sigma_{yy}\rangle/\partial h$, we perturb the y-separation, h, of the confining walls and measure the average value of σ_{yy} during shear. We study fairly small systems, ranging in size from 63 to 621 bubbles. To obtain adequate statistics, averages (indicated by angle brackets) were taken over configurations as well as over time. Thus, each data point shown in figs. 1-3 corresponds to an average over 10000 time steps covering a total strain of at least 10, and at least 9 different initial configurations. We find that at sufficiently long times, the time average does approach the configurational average, but it proves more efficient to use a combination of configurational and time averaging.

In fig. 1, we show the viscosity, η, and Γ as functions of the strain rate, $\dot{\gamma}$. We have measured these quantities over 5 decades of strain rate for a 238-bubble system at a packing fraction of $\phi = 0.95$ (the close-packing fraction is roughly 0.84). Note that η, which diverges as $\dot{\gamma} \to 0$, is not a pure power law as a function of $\dot{\gamma}$; it requires two different power laws and an additive constant to fit the data over the range shown. In the infinite shear rate limit, η approaches a constant, η_∞, which can be calculated analytically by assuming perfectly laminar flow and integrating the viscous forces over the time that bubbles overlap.

Figure 1 also shows that Γ appears to level off at low strain rates. Although we cannot deduce the limiting behavior of Γ directly from the data, we know that it should approach a nonzero constant, Γ_0, in the limit $\dot{\gamma} \to 0$, by the following argument. As $\dot{\gamma}$ approaches zero, the system reduces to a simple network of springs, so the elastic constant $\partial\langle\sigma_{yy}\rangle/\partial h$ must approach a finite, nonzero constant. The stress fluctuations $\left\langle(\sigma_{yy} - \langle\sigma_{yy}\rangle)^2\right\rangle_h$ also approach a finite, nonzero constant, because as $\dot{\gamma} \to 0$ (*i.e.* the quasistatic limit), the system still explores its configuration space and the stress fluctuates, however slowly. (In the quasistatic limit the system is sheared infinitesimally and allowed to equilibrate before being sheared again.) *At* $\dot{\gamma} = 0$, of course, there are no stress fluctuations. In this sense, $\dot{\gamma} \to 0$ is a singular limit. Since both $\partial\langle\sigma_{yy}\rangle/\partial h$ and $\left\langle(\sigma_{yy} - \langle\sigma_{yy}\rangle)^2\right\rangle_h$ are nonzero and finite in the quasistatic limit, it follows that the ratio Γ defined in eq. (2) *must* approach a finite, nonzero constant, Γ_0. We

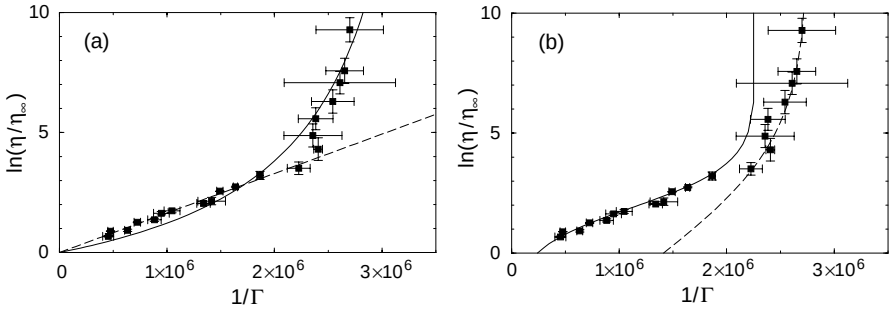

Fig. 2 – (a) An Arrhenius plot of $\ln\eta$ *vs.* $1/\Gamma$ from our simulations (solid squares) at $\phi = 0.95$. The dashed line is a fit to the Arrhenius form, $\eta/\eta_\infty = \exp[A_1/\Gamma]$, at high values of Γ. The solid line is a fit to the Vogel-Fulcher form, which is often used to fit data on supercooled liquids. The deviation from the dashed line indicates super-Arrhenius behavior. (b) Power law fits to the high-Γ (solid) and low-Γ (dashed) data. The solid curve corresponds to an exponent of 0.9 and the dashed curve to an exponent of 2.5. The latter exponent is consistent with mode-coupling theory, which is unsurprising given our limited dynamic range.

find that Γ_0 is comparable to the average elastic energy per bubble. However, our dynamic range is limited compared to that of experiments, so we cannot estimate the value of Γ_0 with much reliability.

Our aim is to determine whether jamming leads to common behavior in sheared foams and supercooled liquids. The most striking feature of jamming in a supercooled liquid is the stupendous rise in the viscosity with decreasing temperature. We have measured a quantity Γ that characterizes stress fluctuations in our driven system in the same way that temperature measures stress fluctuations in an equilibrium system. We therefore study the behavior of η as a function of Γ. This is shown in the solid symbols of fig. 2(a), where we have plotted $\ln\eta$ *vs.* $1/\Gamma$ [13]. A straight line on this plot would correspond to Arrhenius behavior. At high Γ, the behavior is indeed Arrhenius, as shown by the fit to the dashed line, which has the form $\eta/\eta_\infty = \exp[A_1/\Gamma]$, with $A_1 = 1.6 \times 10^{-6}$. The Arrhenius form implies that A_1 should characterize the height of energy barriers to bubble rearrangements in the foam. In our case, we can measure the height of these barriers by measuring the elastic energy per bubble as a function of strain as the system is sheared very slowly. As bubbles overlap (distort) the elastic energy rises, and when they rearrange the elastic energy drops. Thus, the distribution of energy rises measures the heights of barriers that the bubbles cross as the system is strained. The resulting distribution of barrier heights per bubble is shown in fig. 3. At low shear rates, the elastic energy rise distribution approaches a well-defined quasistatic limit [14]. The distribution is quite broad, with a power law region that is cut off at the high-barrier end. The average barrier height is $\langle\delta E\rangle = 5.3 \times 10^{-6}$, which is quite close to our measured value of A_1. Note that A_1 represents some unknown moment of the distribution and that the distribution is fairly broad, so it is not surprising that the two numbers are not exactly the same.

Clearly, the observed behavior is not Arrhenius over the entire range of Γ. Figure 2(a) shows that the behavior is super-Arrhenius at low Γ, in that the viscosity increases more rapidly than predicted by the Arrhenius law. The solid line is a fit to the Vogel-Fulcher form, $\eta/\eta_\infty = \exp[A/(\Gamma - \Gamma_0)]$, with $A = 9 \times 10^{-7}$ and $\Gamma_0 = 2.5 \times 10^{-7}$. *This is the same form that is often used to fit glass transition data* [15]. The ratio $A/\Gamma_0 \approx 3.6$ is a measure of fragility, and corresponds to an extremely fragile glass-forming system [13]. Note that the Vogel-Fulcher form does not fit particularly well at the high-Γ end; this is also typical of

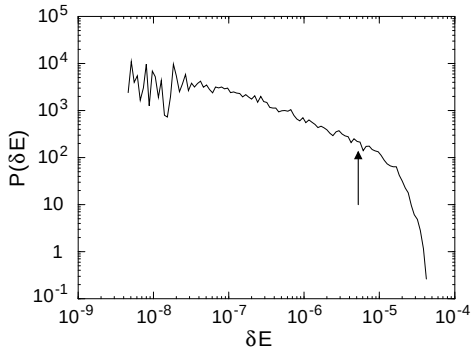

Fig. 3

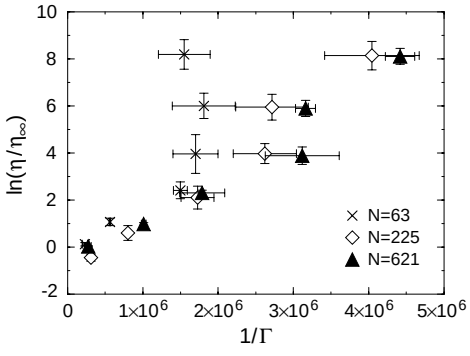

Fig. 4

Fig. 3 – The distribution of energy barriers (measured on a per-bubble basis) crossed by the system as it is strained in the low-shear-rate limit. The average of this distribution (marked by an arrow) provides an estimate of the characteristic energy barrier height that is independent of the Arrhenius parameter A_1.

Fig. 4 – Arrhenius plot of our data for different system sizes, where N is the total number of bubbles. Here, $\phi = 0.9$. The two larger systems yield nearly the same results, but there is a discernible trend towards more Arrhenius behavior with increasing size. Even in the infinite system size limit, however, the behavior must be super-Arrhenius, as discussed in the text.

supercooled liquids, where the Arrhenius form is often used to fit the high-temperature data, and the Vogel-Fulcher form is used to fit the lower-temperature data. Many other functional forms have also been used successfully for supercooled liquids [17]; these forms work equally well for our system.

Skeptics have asked whether our data can be fit by a power law, $\eta \propto 1/(\Gamma - \Gamma_c)^\alpha$. We find that a single power law does not fit the data well over the entire range. However, the high-Γ and low-Γ data can be fit reasonably well by two different power laws, as shown in fig. 2b. The high-Γ fit corresponds to an exponent of $\alpha = 0.9$ and diverges at $1/\Gamma_c = 2.3 \times 10^6$; this evidently underestimates the value of $1/\Gamma_c$. The low-Γ fit, which covers a viscosity range of nearly 3 decades, corresponds to an exponent of $\alpha = 2.5$ and diverges at $1/\Gamma_c = 2.8 \times 10^6$. The latter exponent is in good agreement with the prediction of mode-coupling theory [18]. Viscosity measurements near the colloidal glass transition, which cover a similar dynamic range, are consistent with a similar power law [19]. Data for supercooled liquids also can be fit by a comparable exponent over the same dynamic range [18], but this power law fit predicts a divergence well above the glass transition temperature so it is known to fail badly at higher viscosities.

It is not possible to provide a definite answer to whether our data should be interpreted in terms of a power law or super-Arrhenius form. The same difficulty plagues studies of the colloidal glass transition [19], which cover a similar dynamic range, as well as studies of supercooled liquids, which cover 10 more decades of dynamic range. However, given that a single power law does not fit our data well over the whole range, and that the Arrhenius form at high Γ agrees with an independent measurement of the barrier height, we believe that it is more natural to interpret our data as super-Arrhenius rather than power law.

Since our data are taken for a small system (238 bubbles), it is important to study the system size dependence of the observed trend. This is shown in fig. 4 for three different system sizes at a packing fraction of $\phi = 0.9$. The 63-bubble system is clearly too small, and shows a

very different dependence of η on Γ. There is also a difference between the results for 224 and 621 bubbles. It is small compared to the error bars but there is a systematic trend towards more Arrhenius behavior with increasing system size. Thus one might wonder if the behavior is Arrhenius for infinite systems. We argue that it must still be super-Arrhenius in that limit, because the viscosity diverges as $\dot{\gamma} \to 0$, but Γ approaches Γ_0, a nonzero constant. We note, however, that even if the behavior were Arrhenius, it would still be nontrivial and remarkable.

We find that the super-Arrhenius behavior depends only weakly on packing fraction. We have studied 3 different packing fractions: $\phi = 0.85$, just above close-packing, $\phi = 0.90$ and $\phi = 0.95$. At the lowest packing fraction, the behavior is nearly Arrhenius. Above $\phi = 0.85$, the behavior is fairly insensitive to ϕ, but Γ_0 increases somewhat with ϕ. This trend, as well as the much weaker dependence of viscosity on ϕ than on Γ, is consistent with experiments on supercooled liquids at constant density [20].

Finally, we have studied the effect of frustration on the super-Arrhenius behavior. A monodisperse system will crystallize under shear. We can therefore increase frustration by increasing the polydispersity. The results presented so far are for a flat distribution of width $w = 0.8$, so that the bubbles range from $1 - w = 0.2$ to $1 + w = 1.8$ times the average radius. We have also studied the case $w = 0.2$. We find that the curvature on an Arrhenius plot (with Γ on the abscissa instead of Γ/Γ_0) is greater for the less-frustrated system ($w = 0.2$); this reflects the increase of Γ_0 with decreasing w. This trend cannot be compared to experimental results for supercooled liquids, since there is no way to measure frustration for those systems. However, we note that a recent theoretical picture of the glass transition does predict the same trend [21].

In this paper, we have suggested a measure of fluctuations in an athermal system under steady-state shear, Γ defined in eq. (2), which reduces to $k_{\mathrm{B}}T$ in an equilibrium system. We have shown that the viscosity of a model foam obeys the same relation with Γ that the viscosity of a supercooled liquid obeys with temperature. While there is no *a priori* reason that fluctuations in a sheared foam should be described by an effective temperature, we note that this assumption appears implicitly in a recent approach to the rheology of soft glassy materials [22], which introduces a temperature variable into systems that are definitely athermal. The concept of a "granular temperature" has also been used extensively to describe driven granular flows [23]. Finally, there is numerical evidence that applying shear may be similar to raising the temperature in supercooled liquids. Molecular-dynamics simulations on a model supercooled liquid, namely a bidisperse repulsive Lennard-Jones system, study kinetic heterogeneities in the steady-state sheared supercooled liquid [24]. These simulations show that the kinetic heterogeneities in the sheared system are the same as those in an unsheared system at a higher temperature.

One reason that the concept of an effective temperature may be of some use to driven athermal systems is that the systems explore many different configurations. A different type of flow could explore a different set of bubble-packing configurations, but perhaps a limited amount of ergodicity is enough for statistical mechanics to be useful, within limits that must be determined. We note also that a steady-state driven system obeys a type of fluctuation-dissipation relation in the sense that the average flow gives rise to fluctuations, but the average flow and fluctuations must be determined self-consistently subject to the constraint that the total energy dissipated must be the same as the energy fed into the system. Clearly, a systematic study of the applicability of the concept of a temperature to driven systems is needed. We are currently investigating whether other definitions of an effective temperature yield results for our model that are consistent with the one proposed here. If there is indeed a common phenomenon of jamming underlying driven, athermal systems and supercooled liquids, however, it may not be necessary for the concept of temperature to be valid; it

is possible that *any* reasonable measure of fluctuations in the driven system will lead to qualitatively similar behavior.

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