Soft Matter

OPINION

Analogy between glass rheology and crystal plasticity: yielding at high strain rate

Yue Fan,[ab] Bilge Yildiz[a] and Sidney Yip[*ac]

An abrupt increase of the yield stress at sufficiently high strain rate, seen in glassy as well as crystalline structures, signifies a transition from classical thermal fluctuation to stress activated processes. For crystals this behavior has been recently explained using transition-state-theory with a stress-dependent activation barrier for dislocation glide. An equivalent approach, developed independently for amorphous solids, suggests the physical basis of the upturn behavior of the yield stress is more general. Insights into the interplay between thermal and stress activation processes can contribute to the current efforts toward identifying materials science frontiers at the mesoscale.

Up-turn behavior in dynamic yielding

Yielding is ubiquitous in all materials deformation when a sufficiently high stress or strain is imposed. Fig. 1 shows the behavior observed in two materials systems with very different microstructures, where one finds an upturn behavior in yielding at a characteristic rate of deformation. In a colloidal suspension of latex particles of size 146 nm under shear, the microstructure evolves over three stages of dynamic yielding prior to the onset of steady flow at a shear rate of $\sim 10^2$ s^{-1} (Fig. 1(a)).[1] An analogous response in the flow stress measured in a metal at a strain of 0.15 and 300 K by the Hopkinson pressure bar technique is given in Fig. 1(b).[2] An even more pronounced stress increase is seen at the critical strain rate of $\sim 10^4$ s^{-1}. Although the microstructure features of each material are quite distinct, and the deformation conditions are at different extremes, the similar upturn behavior suggests a dynamic yielding characteristic that is common to amorphous and crystalline matter. One may then ask whether this commonality extends also to the underlying rate-dependent processes and the formulation of yield stress. This is the question we address in the present Opinion article.

View from glass rheology

A nonlinear relation between yield stress and strain rate is of longstanding interest in the rheological behavior of dense suspensions, ranging from metallic to colloidal glasses. The temperature and strain rate dependence of shear yielding in amorphous systems can be explained by recalling the Eyring model,[3] concepts of shear transformation zone (STZ),[4] or models of soft glassy rheology (SGR) model.[5] Mode-coupling theory,[6] which has played a significant role in the dynamical analysis of the glass transition, also has been extended to nonlinear flow in dense suspensions in terms of shear thinning and dynamical yielding.[7] Along with these developments molecular dynamics (MD) simulations have provided valuable atomic-level details of how glassy model systems deform. Fig. 2 shows the flow curves of supercooled binary liquids (80/20 LJ system).[8,9] In Fig. 2(a), at low strain rate, the yield stress varies linearly as the logarithm of strain rate (solid lines), as described by the Eyring model. At high strain rate a more rapid increase develops, more like a power-law behavior (dashed lines), as given by the STZ and SGR models.[8] Fig. 2(b) shows a similar stress–strain rate behavior in a study that revealed fluidized shear bands at low strain rates, with increasing strain rate the bands grow thicker and disappear. Simulation studies also have focused on the interplay of loading, thermal activation, and mechanical noise, and the effects of temperature relative to athermal processes arising from avalanche dynamics.[10]

View from crystal plasticity

The experimental evidence of up-turn in yield response of metals is familiar in the community concerned with high impact and explosive deformation phenomena,[2] such variations have been observed in Fe, Ta, Cu, Al, and Zn.[11] There is general agreement that the essentially Arrhenius behavior at low strain rates is indicative of thermal activation. On the other hand, the underlying dislocation mechanism beyond the upturn, where a power-law behavior is indicated, is still a topic of debate. For discussions of empirical constitutive models, see Preston *et al.*,[12] and Remington *et al.*[13]

For glassy and crystalline systems yielding at the molecular level can be examined through MD simulations of stress *versus*

aDepartment of Nuclear Science and Engineering, Massachusetts Institute of Technology, 77 Massachusetts Avenue, Cambridge, MA, 02139, USA. E-mail: syip@mit.edu

*bMaterials Science and Technology Division, Oak Ridge National Laboratory, Oak Ridge, TN, 37831, USA*

cDepartment of Materials Science and Engineering, Massachusetts Institute of Technology, 77 Massachusetts Avenue, Cambridge, MA, 02139, USA
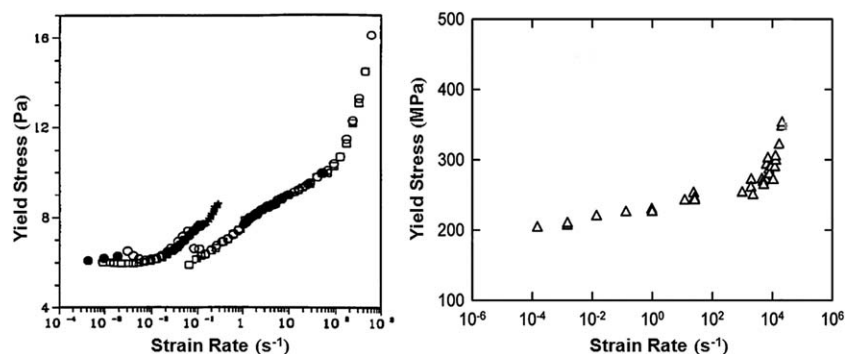
Fig. 1 (a) Experimental data on a colloidal suspension showing a significant increase in the shear stress beyond a certain shear rate (from ref. 1). (b) Measured flow stress in copper showing the up-turn behavior (from ref. 2).
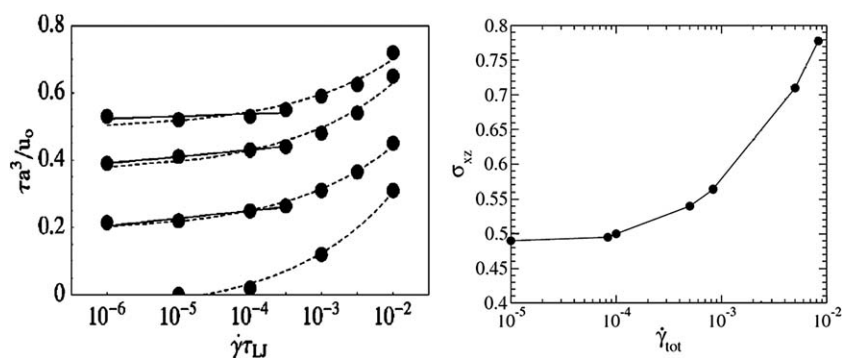
Fig. 2 (a) Variation of yield stress with strain rate in supercooled liquids (from ref. 8). Solid lines indicate a linear relation, while dashed lines are power-law fits, at reduced temperatures 0.01, 0.1, 0.2, 0.3 (top to bottom). (b) Variation of yield stress with imposed strain rate where analysis showed stress-induced shear bands at low strain rate and a broad transition to homogeneous shear expected at ~1.5 × 10^{-3} (from ref. 9).
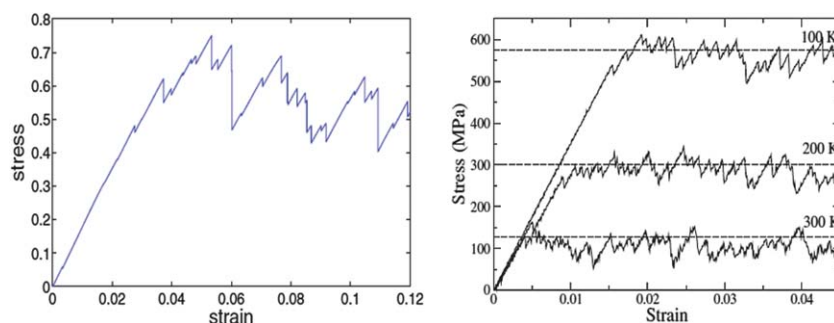

Fig. 3 (a) MD simulation results on a glassy system (from ref. 16). (b) MD simulation of stress–strain curves in single crystal Al containing a Lomer dislocation at several temperatures (from ref. 17).

strain. As seen in Fig. 3 after the initial elastic response, the shear stress undergoes small-scale fluctuations interrupted by a larger-magnitude relaxation event, reminiscent of stick-slip behavior. The intermittent discrete relaxation events in the glass (Fig. 3(a)) suggest activation (flip) of shear transformation,[14] or free volume zones,[15] while in the crystal (Fig. 3(b)) they can be interpreted as activation of dislocation displacement.[17]

Recently Fan *et al.* proposed a constitutive model for the gliding of a screw dislocation in a bcc metal to describe the onset of plastic flow in crystals.[18] A stress-dependent activation barrier, $E(\sigma)$, was introduced in the framework of transition-state theory as a quantity to be identified separately, including by direct determination by atomistic calculations. For strain-rate controlled environments, where the system's strain and stress are time-dependent, dislocation mobility is governed by a residence probability $P(t)$ that the dislocation does not glide to the next potential energy valley up to time t, with $P(t)$ given by the evolution equation

$$\frac{dP(t)}{dt} = -k^{\perp}(t)P(t) \tag{1}$$
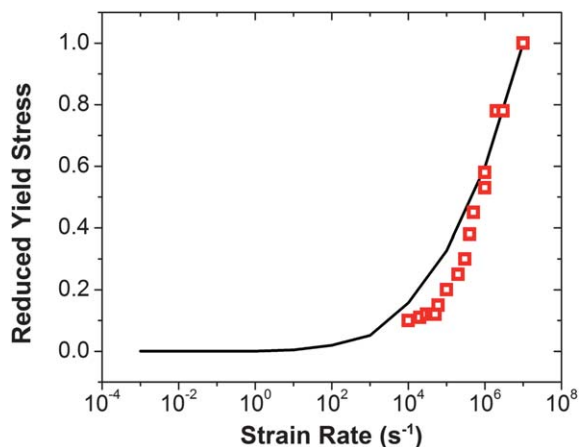
Fig. 4 Flow stress in Fe described by a stress-dependent activation model which couples thermal and stress activations (curve)[18] and experimental data.[2] Calculations and measurements are normalized at the strain rate 10^7 s^{-1}.

where $k^{\perp}(t)$ is a time-dependent transition rate. It is understood that $k^{\perp}(t)$ is also a stress-dependent quantity expressible through an activation barrier $E(\sigma)$,

$$k^{\perp}(\sigma) = k_0^{\perp} \exp\left[-\frac{E(\sigma)}{k_{\mathrm{B}}T}\right] \qquad (2)$$

along with the constitutive relation $\sigma = G\dot{\varepsilon}t$, where G is the shear modulus, and $\dot{\varepsilon}$ is the applied strain rate. The introduction of a stress-dependent activation barrier in eqn (2) naturally couples the stress to thermal activation. In this study the yield stress is calculated from $\bar{\sigma} = G\dot{\varepsilon}\bar{t}$, with $\bar{t}$ being the first moment of the escape probability distribution function $p(t)$, where $p(t) \equiv -\mathrm{d}P(t)/\mathrm{d}t$.

The only input to a prediction of the yield stress variation with strain rate is the quantity $E(\sigma)$, which can be determined by atomistic calculations using an appropriate inter-atomic potential for a given microstructure.[18] This approach therefore does not entail invoking any other dislocation mechanism or introducing bridging parameters. Moreover, the flow stress obtained is a coarse-grained quantity which can be compared directly with experiments, as shown in Fig. 4. One sees a smooth transition from the classical Arrhenius behavior of yielding, associated with thermally assisted relaxation, to a nonlinear behavior beyond the critical strain rate.

Implications of the analogy

We believe a key to physical understanding of the strain-rate effects on yield stress lies in the transition rate that describes the relevant plastic unit-process, namely, $k^{\perp}(\sigma)$, given by eqn (2), in the case of dislocation mobility. Independently, the notion of a strain-dependent activation energy has been put forth recently in analyzing the temperature effects on the rheology of amorphous solids,[10] as well as the elastoplasticity of glasses and dense colloidal suspensions.[16,19] Specifically the coupling between the thermally activated plastic units with applied strain rate was expressed through a conditional probability,[10]

$$P(\varepsilon; \varepsilon_0) = \exp\left[-\frac{1}{\dot{\varepsilon}}\int_{\varepsilon_0}^{\varepsilon} k^{\mathrm{flip}}(\varepsilon')\mathrm{d}\varepsilon'\right] \qquad (3)$$

$$k^{\mathrm{flip}}(\varepsilon) \propto \exp\left[-\frac{E(\varepsilon)}{k_{\mathrm{B}}T}\right] \qquad (4)$$

where $P(\varepsilon;\varepsilon_0)$ is the probability that the local zone does not flip at strain ε given that the initial strain is ε_0, and $k^{\mathrm{flip}}(\varepsilon)$ is the transition rate for a zone flip at local strain ε.

Comparison of eqn (1) and (2) with eqn (3) and (4) shows that essentially the same formulation is being considered independently in the two communities, both to probe the thermal and stress activation effects in yielding at finite strain rates. It would be interesting to compare further how $E(\sigma)$ in eqn (2) and $E(\varepsilon)$ in eqn (4) have been treated in their own context, crystal plasticity and glass rheology. Here one expects the microstructure differences between amorphous solids and crystals to play a major role, for example, in terms of local spatial and temporal organizations at the molecular and mesoscale levels. In the case of crystal plasticity we propose the up-turn behavior in Fig. 1(b) can be regarded as strain localization in that at the critical strain rate a localized strain does not have enough time to propagate through the system. The onset of a rising stress therefore signifies a kinetically trapped microstructure. Additionally analysis shows that the experimental data in Fig. 1(b) beyond the critical strain rate correspond to an "apparent" activation volume is reduced by three orders of magnitude.[2] It is this confinement that characterizes the rate sensitivity at high strain rate. A recent MD study of shear flow in supercooled liquids expressed the same notion of a critical strain rate governing the propagation of local strains.[20] It would be worthwhile to reconcile this interpretation with the stress-induced ordering discussed in ref. 9.

There is a surge of current interest in nonlinear rheological behavior of dense colloidal suspensions,[21–24] pointing to a broadening range of issues, for example, thermodynamic basis of STZ *versus* SGR,[22] yielding in model hard sphere glasses,[23] and residual stress relaxation.[24] It seems reasonable to regard the coupling between thermal activation and strain rate as a universal mechanism underpinning the microstructure sensitivity of dynamic yielding. Such studies can result in the construction of a $(T, \dot{\varepsilon})$ mechanism map, and the clarification of how to connect the microscale details to macroscale behavior. There are also emerging efforts in the materials community to identify the frontiers of mesoscale science,[25,26] which includes the mechanisms of plastic deformation[27] and flow.[21] Thus contributions from the soft matter community toward a broader appreciation of rate effects in dynamic yielding would be very timely.

Acknowledgements

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