Discussion of paper by S. Arora, A. Shabbir, O. Hassager, C. Ligoure, L. Ramos, entitled 'Brittle fracture of polymer transient networks'

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Costantino Creton: The breakage or fracture can occur in either the linear or nonlinear extension regimes. Can you relate the onset of fracture to a critical value of stored elastic energy at the point of fracture? This could be done by first finding a proper material model fitting the uniaxial data at different strain rates with the same set of parameters and then simulate a loading curve up to the breaking point and then a fast unloading following the approach of [1]. The energy under the unloading curve is the stored elastic energy which may be a predictor of the onset of fracture.

Answer: If we adopt a solid mechanical engineering representation (engineering stress versus $\lambda - 1/\lambda^2$, with λ the stretch ratio), we clearly see that the sample with a large amount of telechelic polymer ($\beta = 55\%$) breaks in the linear regime, whereas the sample with a low amount of polymer ($\beta = 5\%$) exhibits significant hardening before fracturing, as shown in the plot (Fig. 1).

Unfortunately, because the samples are (viscoelastic) liquids, unloading experiments cannot be performed, preventing direct measurement of the dissipation during the fracture process.

Michael Rubinstein: From the Weissenberg numbers, can you relate the transition point to the transient network connectivity? (in relation to the plot of Wi-β%). Is there some kind of self-similarity?

Answer: Figure 3 shows that the transition from filament thinning to fracture is independent of the amount of polymer. In this specific double transition system, we have demonstrated that there is no percolation threshold for the network of telechelic polymers, due the underlying network of wormlike micelles [2] as opposed to a simple network of telechelic chains. So we do not expect any kind of self-similarity due to the transition of network connectivity

Comment from Ole Hassager: It is quite amazing to see that at $\mathrm{Wi} = 0.5$, there is already a liquid to solid transition.

Dimitris Vlassopoulos and Peter Olmsted: About the crack profiles: With the increase of the fraction of polymers, it appears that the fracture or cracking can take place even in the linear regime (see the evolution of the tip thickness). There seems no need to use the hyperelasticity concept to interpret the results. Is this a thermodynamic effect?

Answer: We agree that, when the amount of telechelic chains is large, hyperelasticty does not play any role and the crack profile is parabolic. However, we show in this paper that, when the amount of telechelic chains is low, nonlinear elasticity effects are crucial and lead to a nonparabolic crack profile.

Comment from Ole Hassager: Linear elastic fracture mechanics (LEFM) predicts a parabolic crack opening with a singular point at the crack tip. The observed departure from a parabolic crack profile near the tip represents the breakdown of LEFM. It is this departure that has been explained in terms of a nonlinear elastic process. The predicted displacement field near the crack tip is an expansion involving the LEFM term (depending on the square root of the distance from the tip) and a higher order term (that is linear in the distance).

Costantino Creton: When the fracture propagation speed is close to or larger than the sound speed, the system will undergo nonlinear behavior as demonstrated by the work of Jay Fibeberg and Eran Bouchbinder on polyacrylamide gels. How to understand the fracture behavior at such high speeds?

There were suggestions from the audience about the inertia effect in such cases.

Answer: We agree that, as shown in Fig. 9, the higher the crack velocity, the higher is the nonlinear character of the fracture process. However, it is interesting to notice that, as the telechelic polymer network is denser, both the crack propagation speed and the nonlinearity of the crack decrease. We believe this is due to the fact that nonlinear effects originate mainly from the wormlike micelle network, which may be impeded by the coupling to the network of telechelic chains. This is discussed in the last paragraph of the Discussion section of the paper.

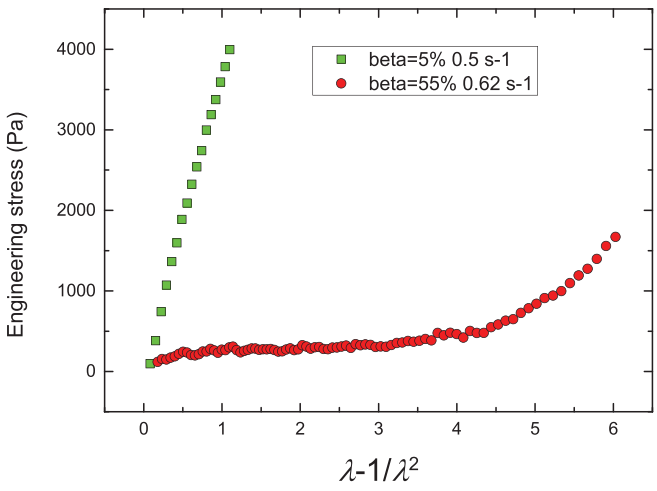

FIG. 1. Tensile engineering stress as a function of $\lambda - 1/\lambda^2$, where λ is the stretching ratio for samples with two polymer concentrations as indicated in the legend. Departure from a linear variation indicates a departure from the classical models of rubber elasticity.

Ralph Colby: Stress concentration is related to the radius of curvature of the crack tip. Probably the length δ is related in some way to that radius of curvature but since δ is quite large, maybe you can directly measure the radius of curvature of the crack tip?

Answer: Unfortunately, the spatial resolution is not sufficient to extract informative data on the radius of curvature at the crack tip, which is not measurable in the case of nonparabolic profile [as in Fig. 8(a)].

Evelyne van Ruymbeke: I have a question related to Fig. 5: In this figure, it is clear that the proportion of telechelic polymers strongly affects their strain hardening behavior. Is it possible to relate the capacity of a sample to strain harden to the average molar mass of a strand between two telechelic polymers? (based on the idea that a small β leads to large M_{strand}, and thus, more extensible sample).

Answer: We agree that when the amount of telechelic polymer increases, the average distance (or molecular weight) between two telechelic chains along a strand of wormlike micelle decreases, and thus the strain hardening decreases. We indeed could evaluate from the sample composition this average molecular weight as a function of the parameter β from the structural parameters as extracted from scattering techniques [3]).

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