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

Arora, S.; Shabbir, Aamir; Hassager, Ole; Ligoure, C.; Ramos, L.; Creton, Costantino; Rubinstein, Michael; Vlassopoulos, Dimitris; Olmsted, Peter; Colby, Ralph; van Ruymbeke, Evelyne

Questions JOR – Paper presented by C. Ligoure
Discussion of a paper by S. Arora, A Shabbir, O. Hassager, C. Ligoure, L. Ramos, entitled "Brittle fracture of polymer transient networks"

Costantino Creton:

The breakage or fracture can occur in both linear and non-linear extension regimes. Can you relate the fracture to the elastic energy at the point of fracture? This could be done by calculating the work required to break the material, and could be explained by the Hookean or non-linear forces during loading and unloading.

Answer: If we adopt a solid mechanical engineering representation (engineering stress versus $\lambda - 1/\lambda^2$, with λ the stretch ratio), we clearly see as well that the sample with a large amount of telechelic polymer (β=55%) breaks in the linear regime, whereas the sample with a low amount of polymer (β=5%) exhibits significant hardening before fracturing, as shown in the plot below. Unfortunately, because the samples are (viscoelastic) liquids, unloading experiments cannot be performed, preventing direct measurement of the dissipation during the fracture process.
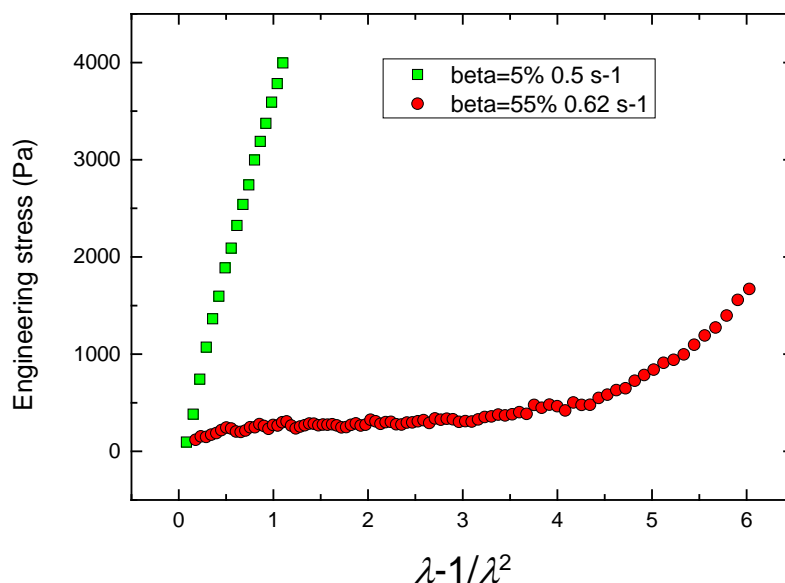
Michael Rubinstein:

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 (ref Nakaya-Yaegashi et al, Journal of Rheology **52**, 359,2008), as opposed to a simple network of telechelic chains. So we do not expect any kind of self-similarity due to the transition network connectivity.

Ole Hassager:

It is quite amazing to see that at 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?

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, non linear elasticity effects are crucial and lead to a non parabolic 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 or larger the sound speed, the system will undergo nonlinear behaviour. How to understand the fracture behaviour at such high speeds?
There were suggestions from the audience about the inertia effect in such cases.

Answer: We agree that, as shown in figure 9, the higher the crack velocity the higher is the non linear 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 non-linearity of the crack decrease. We believe this is due to the fact that non linear 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.

Ralph Colby:

Stress concentration is related to the radius of curvature of the crack tip. Probably the length delta is related in some way to that radius of curvature but since delta 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 non parabolic profile (as in figure 8a).

Evelyne van Ruymbeke:

I have a question related to Figure 5: In this figure, it is clear that the proportion of telechelic polymers strongly affects their strain hardening behaviour. Is this possible to relate the capacity of a sample to strain harden to the average molar mass of a strand between to telechelic polymers? (based on the idea that the smaller beta is, the larger is M_strand, and the more extensible is the sample).

Answer: We agree that when the amount of telechelic polymer increases, the average distance (or molecular weight) between to 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, see Ramos&Ligoure, Macromolecules **40**, 1248, 2007)