Some comments on the fracture of viscoelastic solids

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ABSTRACT

Crack propagation in viscoelastic solids like rubber is of great practical importance. Shrimali and Lopez–Pamies have proposed a new interesting approach for the crack propagation in viscoelastic solids. We give comments on the validity of the theory and point out some effects not included in the theory.

1. Introduction

Crack propagation in rubber-like materials has many applications, e.g. to tire wear [1,2], and has been studied for a long time [3–12]. In a recent series of papers Shrimali and Lopez–Pamies have presented an interesting new approach to crack propagation in viscoelastic solids [13–15]. The theory is based on the assumption that a slab of rubber fracture at a critical stretch that is independent of the stretch rate. Here we argue that the study is not as general as stated by the authors. In what follows we assume for simplicity linear viscoelasticity and neglect inertia effects.

2. Dependency of the strain at fracture on the strain rate

The theory of Shrimali and Lopez–Pamies is based on the assumption that fracture in "pure shear" (see Fig. 1) occurs at a critical stretch that is independent of the stretch rate. As support for this they cite several experimental studies where this is indeed observed [16,17]. However, the experimental studies are for a limited range of applied stretch rates, and it is easy to show that it cannot be true in general. To show this, consider a slab of elastomer (of undeformed height h_0) elongated at a rate $\dot{h}$. We assume that the length L of the slab is much bigger than the height h_0 and that the crack tip is not close (on the scale of h_0) to any end of the slab. For simplicity we assume linear viscoelasticity with the frequency-dependent modulus $E(\omega)$. The modulus in the low-frequency (rubbery) region $E(0) = E_0$ is much smaller than the modulus $E_1 = E(\infty)$ in the high frequency (glassy)

region. (Note that the rubbery region and the glassy region may be separated by 10 (or more) decades in frequency.) If the elongation strain rate $\dot{\epsilon} = \dot{h}/h_0$ (or rather strain rate frequency $\omega = \dot{\epsilon}/\epsilon$) is very low the rubber will effectively be in the rubbery state, and the onset of crack propagation will occur when the strain $\epsilon_0 = (2G_0/h_0 E_0)^{1/2}$ where $G_0 = \Delta\gamma$ (denoted by G_c in Ref. [13]) is the energy per unit surface area to breaks the bonds at the crack tip. However, if the strain rate is very high the fracture will occur almost instantaneously and rubber will effectively be in the glassy state everywhere both with respect to the deformations resulting from the elongation and from the movement of the crack tip. Hence, we can again apply the standard Griffith fracture criteria but now using the high frequency modulus so that the critical (fracture) strain $\epsilon_1 = (2G_0/h_0 E_1)^{1/2}$ which is smaller than the low strain rate fracture strain by a factor of $(E_0/E_1)^{1/2}$ which is typically in the range $0.1 - 0.01$ in practical cases. This conclusion is consistent with the temperature–frequency relation valid for (simple) amorphous rubber materials [18] where high frequencies are equivalent to low temperatures. At temperatures below the glass transition temperature the time scale of deformation is typically much shorter than that of the (Brownian motion) of the molecular chains of the rubber, and rubber compounds are hard and brittle like glass.

Even when the strain rate is very high, there will be energy dissipation by viscoelastic deformation (as discussed by one of the present authors in Ref. [12]) but this is irrelevant for the fracture problem since for very rapid elongation (or equivalently for an experiment performed
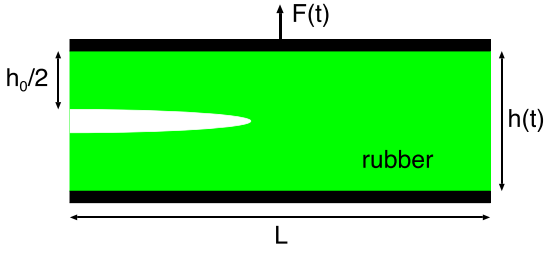
Fig. 1. Crack in a rubber sheet with clamped upper and lower edges. The height $h(t) = h_0 + u(t)$ of the slab increases with time (undeformed height is h_0). For an elastic solid (no viscoelasticity) the crack start to move when the elongation strain $\epsilon = u(t)/h_0$ reach the critical value $(2G_0/Eh_0)^{1/2}$ where $G_0 = \Delta\gamma$ is the energy per unit surface area to break the bonds at the crack tip.

at very low temperature) of a rubber strip the crack will fracture the rubber strip before the viscoelastic relaxation starts to occur.

We note that the energy release rate (or the effective energy of adhesion) $G(v)$ is not always increasing with the crack propagation speed v. Indeed for finite systems and high loading speed the effective energy of adhesion goes back to the value $G_0 = \Delta\gamma$ for purely elastic materials. But this is only true if the system is in the glassy state also during loading. If instead a sample is stretched without a crack to a very large final strain, and kept in the stretched state until equilibrium condition, and then a crack is inserted (e.g., using a razor blade) the crack will propagate so fast that the deformation frequency $\omega = \dot{\epsilon}/\epsilon$ is in the glassy region everywhere. In this case, in the initial fracture process only a fraction G_0/G of the stored elastic energy is used to break the bonds at the crack tip. After the crack has fully separated the system in two part a slow relaxation occurs where the stress and strain fields relax back to the equilibrium state. During this process the energy $G - G_0$ is dissipated (see [12,19]) and as a result $G/G_0 \to E_1/E_0$ as the crack speed $v \to \infty$. So only for the very fast loading case (where the solid is effectively in the glassy state everywhere also before crack propagation start) is it true that $G/G_0 \to 1$ as the loading rate increases.

We note that there is no reason for the critical tearing energy G_0 to be independent of the strain rate (and the temperature), but there is absolutely no reason that it should depend on the strain rate (and the temperature) in exactly the same way as the bulk viscoelastic modulus because it involves processes of a different nature, e.g., breaking of the polymer chains or cross-links, which are not involved in (or the origin of) the bulk viscoelasticity. In the papers by Shrimali and Lopez–Pamies it is assumed that the critical tearing energy G_0 is independent of the strain rate.

Experiments we have performed, showed that the critical stretch is independent of the stretch rate only for very special cases, mostly encountered in very tough viscoelastic materials and only in a finite range of stretch rates [20,21]. We have performed few tests with the pure shear geometry but many tests in single-edge notch geometry (not considered in Ref. [13]), where the critical stretch depends on notch length, and the stretch at break is markedly strain rate dependent, which may in part result from a strain rate dependency of the bond breaking in the process zone. See Ref. [20] and an earlier study on the fracture of very elastic polyurethanes [21].

3. Definition of the fracture energy

The fracture energy G (denoted as T_c in Ref. [13]) is usually measured under conditions where the strain far from the crack tip is in the relaxed state. For example, in some experiments the slab in Fig. 1 is first stretched without the presence of the crack and then kept in the stretched state until equilibrium is reached. Only after a crack is inserted using a razor blade. For this situation $G(v)$ is the energy to break the bonds at the crack tip plus the viscoelastic energy dissipation close to the crack tip due to its motion, and will only depend on the

crack tip speed v (and the temperature, see below). In this case $G(v)$ can be considered as a (useful) material parameter which can be tabulated for different rubber compounds as done by Gent [4] for three types of rubber [cis-polybutadiene (PB), styrene-butadiene copolymer (SB) and ethylene-propylenecopolymer(EPR)].

For a general situation where the strain field far from the crack is time dependent, as when the slab in Fig. 1 is elongated $h = h(t)$ in some arbitrary complex way, Shrimali and Lopez–Pamies *define* the fracture energy as $G = -dW/dA$ where dW/dA is the change in the total (stored and dissipated) deformation energy in the bulk with respect to added surface area A of the pre-existing crack. But using this definition G will depend on the (time-dependent) loading conditions and is no longer a material parameter but depends on the loading history. Shrimali and Lopez–Pamies only studied the onset of crack propagation where $v = 0$ but in the most general situation where the strain field far from the crack is time dependent, and the crack propagates with the speed v, the crack propagation need to be studied theoretically (or experimentally) for each such case separately using, e.g., numerical methods (see Sec. F). The material input parameters for such studies are the viscoelastic modulus and the energy to break the bonds per unit surface area $G_0 = \Delta\gamma$. (In general G_0 depends on complex processes occurring in the crack tip process zone, which depends on the crack tip speed and the temperature, i.e., $G_0 = G_0(v, T)$ is a function of the crack tip speed and the temperature.)

The theory proposed in Ref. [13] is stated to be valid only at the onset of crack propagation. This case is not of very big interest in practical applications since the (useful) toughness of rubber materials result from the fact that the crack propagation energy increases very strongly with increasing crack tip speed, e.g., by a factor of E_1/E_0 when the stress field far from the crack is in the relaxed state.

Consider the system in Fig. 1 exposed to time-dependent external forces. In Ref. [13] it was proposed that the crack will start to move at the time t when the condition

$$-\frac{dW^{\mathrm{Eq}}}{dA} = G_0 \tag{1}$$

is obeyed. Here W^{Eq} is the elastic energy stored in the system at equilibrium i.e. the elastic energy after keeping the system which prevail at time t for an infinite long time with fixed boundary conditions (as given by the boundary conditions at time t) on the part of the boundary that are not traction free. As support for this equation the authors use the experimental observation that the fracture for the system shown in Fig. 1 occurs at a fixed strain independent of the strain rate. However, we have argued that this may be true only for low enough strain rates, and will break down for strain rates where the deformation frequency $\omega = \dot{\epsilon}/\epsilon = \dot{u}/u$ (see Fig. 1) falls within the glassy response region. This result in a violation of (1) as can be easily seen by the following argument: Consider the system in Fig. 1 and assume first the case of extremely slow stretch rate. In this case the material behaves as an elastic body with low-frequency elastic modulus E_0 and the energy balance would then require

$$dW^{\mathrm{Eq}} = -G_0 dA \tag{2}$$

where $dW^{\mathrm{Eq}} = dU$ is the change of elastic energy $dU = E_0\epsilon_0^2 h_0 dA/2$ of the relaxed material. Next consider extremely high stretch rate, i.e. a step change of the remote displacement. In this case the material is in the glassy region and will behave elastically with modulus E_1 and the energy balance will require that the total change of the elastic energy must balance the fracture energy

$$dW^{\mathrm{Eq}} + dW^{\mathrm{NEq}} = -G_0 dA \tag{3}$$

Here the change in the total elastic energy equal $dU = dW^{\mathrm{Eq}} + dW^{\mathrm{NEq}} = E_1\epsilon_1^2 h_0 dA/2$. The conditions (2) and (3) give the Griffith fracture results for ϵ_0 ans ϵ_1 quoted in Sec. A. It is evident that Eq. (6) in Ref. [13] cannot hold true in all cases as the authors propose.
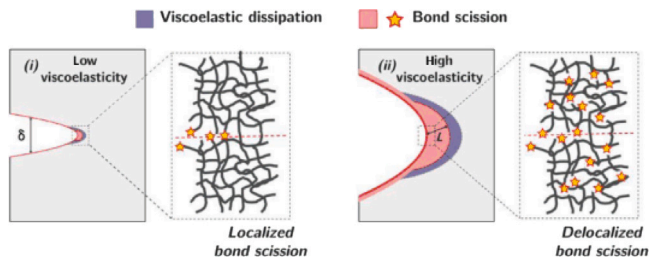
Fig. 2. Schematic coupling between viscoelasticity (blue domain) and strand breakage (red domain) at the crack tip. The enlarged region shows the occurrence of bond scission (yellow stars) in the elastomer network. Bond scission and viscoelastic dissipation are strongly coupled, with a joint increase in bond scission and viscoelastic dissipation between the low viscoelasticity (i) and large viscoelasticity regimes (ii). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)
Source: Adopted from Ref. [20].

4. Velocity dependency of the crack propagation energy $G(v)$

As we understand it the theory developed in Refs. [13–15] only address the onset of crack propagation. Therefore, the dependency of the fracture energy $G(v) = G_0[1 + f(v)]$ on the crack-tip speed cannot be deduced from the theory. When the crack is propagating at constant speed, one has to consider the influence of non-conservative work of internal stresses to write down the correct energy balance equation. This was done in Ref. [22] where Eqs. (6), (8), (9) and (10) give the change of the elastic energy, and of the non-conservative work of internal stresses, upon a small displacement of crack front both for the opening and closing crack, thus extending the Griffith criterion to viscoelastic cracks moving at a constant speed.

Here we note that in the classical experiments of Gent the $G(v)$ relation was measured by peeling rubber sheets apart [4]. By performing measurements at different peeling speeds and temperatures and using the temperature–velocity shifting procedure he could map out the full $G(v)$ (master) curve. Measurements of this type cannot be analyzed using the theory of Shrimali and Lopez–Pamies.

5. Crack tip process zone

Shrimali and Lopez–Pamies claim that the advantage of their approach compared to earlier theories is that they do not need to invoke a crack tip process zone. However, a crack tip process zone will occur in all real materials and it is important to study its influence on the crack propagation. If one assume a priory that no crack tip process zone occur one cannot claim that it will have no influence on the viscoelastic part of the crack propagation energy. Here, we note that for cohesive crack propagation the crack tip process zone can be very complex involving cavitation, formation of filaments (stringing) and a recent study has shown that breaking of chemical bonds may occur far away from the crack tip in a region overlapping with the region where viscoelastic energy dissipation occurs, see Fig. 2 and Ref. [20]. Thus bond scission, far from being restricted to a constant level near the crack plane, can be delocalized over hundreds of micrometers and increase by a factor of 100, depending on the temperature and stretch rate, and the energy dissipated by covalent bond scission accounts for a much larger fraction of the total fracture energy than was previously believed. The situation may be less severe for adhesive crack propagation (crack propagation at the interface between a flat rigid solid and an elastomer film adhering to the substrate) but even in these cases for weakly crosslinked elastomers (as in pressure sensitive adhesives) the crack tip process zone is very complex and spatially large [23].

The existence of a (complex) process zone in rubber-like materials can be (indirectly) seen through the roughness profiles of the surfaces formed by the crack propagation [24,25]. This is valid for all real materials, not only rubber. The morphology of fracture surfaces reflect the complex processes occurring on different length scales close to the crack tip, and depend on the microstructure of the material.

6. Numerical approaches to viscoelastic crack propagation

Shrimali and Lopez–Pamies performed a numerical study using the finite element method applied to an elastomer with non-linear rheology. To determine when the crack start to move they used the criteria (1) [see Eq. (6) in Ref. [13]] involving only $G_0 = \Delta\gamma$ and the equilibrium part of the stored elastic energy. In our opinion this approach may be valid only at the onset of crack propagation and for low enough stretch rate. An alternative more general approach, which can be used to obtain the full $G(v)$ curve, is to use a discretized solids where the "atoms" are connected by (realistic) non-linear springs or bonds, e.g using a non-linear Rouse-like model for the network chains [26]. Such atomistic models have been used to study the $G(v)$ relation for crack propagation in silicone [27] and recently also for adhesive cracks involving viscoelastic solids in contact with a rigid flat substrate [28]. An advantage with this approach is that it can also include inertia effects which are crucial in some cases (see Ref. [27]).

CRediT authorship contribution statement

B.N.J. Persson: Conceptualization, Formal analysis, Investigation. **G. Carbone:** Formal analysis. **C. Creton:** Formal analysis. **G. Heinrich:** Formal analysis. **T. Tada:** Formal analysis.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

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