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Loïc Vanel, Sergio Ciliberto, Pierre-Philippe Cortet, Stéphane Santucci. Time-dependent rupture and slow crack growth: elastic and viscoplastic dynamics. *Journal of Physics D: Applied Physics*, 2009, 42 (21), pp.214007. 10.1088/0022-3727/42/21/214007 . hal-01729166

HAL Id: hal-01729166

<https://univ-lyon1.hal.science/hal-01729166v1>

Submitted on 12 Mar 2018

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REVIEW ARTICLE

Time-dependent rupture and slow crack growth: Elastic and viscoplastic dynamics

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Abstract. We review some theoretical and experimental works describing the slow, thermally activated, growth of a crack in a solid material under stress. Theoretical approaches fall into two main classes: creep crack growth models and elastic trap models. On one hand, creep crack growth models describe the viscoplastic flow of matter until some characteristic rupture strain is reached. This first category of models applies especially to the case of polymer rupture. On the other hand, elastic trap models assume that a rupture energy barrier is overcome by elastic stress fluctuations. While this second category of models is more restricted since it applies only to materials with brittle rupture features, it offers a framework that can be interestingly and importantly extended to the case of heterogeneous materials. Models will be discussed in the light of recent experimental works.

PACS numbers: 62.20.Mk, 46.50.+a, 46.35+z

1. Introduction

Understanding strength of solids has always been an important concern in order to be able to design properly structures of various kind (buildings, vehicles, ships, airplanes, etc.). Experimentally, the strength of solids is usually found much lower than would predict a theoretical estimate assuming rupture of covalent bonds. Griffith described how the presence of flaws in a material and the corresponding stress inhomogeneities resulting from it could explain a significant decrease in strength [1]. It is also well-known that strength of solids depends on the volume of the specimen, the strength becoming smaller in larger samples. This property has been mainly described as a statistical effect in a distribution of flaws using the weakest link concept, originally introduced by Pierce [2] and developed into a theory by Weibull [3]. This theory, mathematically connected to the problem of extreme-value statistics [4], has helped to understand how strength of wires depend on their diameter or their length, and explains why, nowadays, very small objects such as nanotubes or nanowires have often a very high strength. However, another well-known observation, that can not be described by Weibull's statistical theory, is the time-dependence of strength of solids, namely the fact that materials will break at smaller loads when the duration over which the load is applied becomes longer [5]. It has been recognized since at least the early 1940s' that the decrease in strength with time was due to rupture processes that are thermally activated [6]. Several models involving a thermally-activated mechanism have been proposed to explain the experimental observations. Nowadays, there are still many unanswered questions on the proper way to describe slow rupture, alternatively called subcritical rupture or time-dependent rupture.

The present paper intends to review historical and also more recent works on time-dependent rupture with a focus on slow crack growth dynamics in the second half of the paper. Section 2 will first discuss various laws for the breaking time of a material under a constant applied load. We will see that many different kinds of materials can be described according to a simple phenomenological law. We will discuss the limits of this phenomenological law, in particular the scale at which rupture is supposed to occur and the precise dependence with the applied stress. We will also review various theoretical models that have been proposed in order to predict the experimental observations on the breaking time. Section 3 will discuss several models that can be used to predict slow crack growth dynamics under a constant load. We will concentrate on two models describing the case of brittle materials, and compare their predictions with recent experimental results. We will also discuss briefly the case of viscoplastic materials for which predictions are more difficult to make and often rely on very phenomenological descriptions of the material mechanical properties.

2. Breaking time of materials under a constant load

2.1. Early experiments and the BBZ model

Time-dependent rupture, subcritical rupture, or more simply put, slow rupture has been very early described as a consequence of creep flow. Maybe the first historical evidence for such a possible connection between creep flow and slow rupture was made around 1830 by Vicat [5], a bridge construction engineer who studied the creep of iron wires under different loads. Vicat discovered that, even though corrosion of the wire by the atmosphere was prevented with the use of drying oil, wires that were submitted to the largest loads would eventually break after some time, which meant in Vicat's experiments about three years!

A century after Vicat, Busse [6], a tire engineer, studied the slow rupture properties of cotton yarns, i. e. bundles of cotton fibers. He found that the breaking time τ decreases exponentially with the applied stress σ and he inferred from his observations on various yarn lengths that this behaviour was due to viscoplastic flow of individual fibers, but neither slippage of the fibers between each other nor chemical degradation. Busse proposed that the inverse of the breaking time should be proportional to the viscous strain-rate predicted by Eyring's reaction-rate theory of viscous flow [7]:

$$\tau^{-1} \sim \tau_0^{-1} \exp\left(\frac{-E_0}{k_B T}\right) \sinh\left(\frac{\sigma V}{k_B T}\right) \quad (1)$$

where k_B is Boltzmann constant, T the thermodynamic temperature, E_0 a characteristic energy barrier and V an activation volume. In Eyring's theory, the activation volume is half the average volume occupied by a molecule. The \sinh term in equation (1) comes from the fact molecules have a thermally activated probability proportional to $\exp(\sigma V/k_B T)$ to jump a unit distance in the direction of the flow but also a small probability proportional to $\exp(-\sigma V/k_B T)$ to jump in the opposite direction. When the applied stress σ is large enough so that $\sigma V \gg k_B T$, the probability to move backward is very small. In such a case, the rupture process is irreversible and the rupture time takes a simpler form:

$$\tau = \tau_0 \exp\left(\frac{E_0 - \sigma V}{k_B T}\right) \quad (2)$$

Busse found equation (2) to be in qualitative agreement with his experimental observations. Shortly afterwards, Tobolsky and Eyring [8] calculated the rupture time of a fiber bundle submitted to a constant force, taking into account the progressive increase of the load on the remaining fibers. They found that, in the limit where $\sigma V \gg k_B T$, it added only logarithmic corrections to equation (2). Along the same line, Coleman described how knowledge of the statistical distribution of breaking times under a constant load could be used to predict breaking times for other load histories (linear increase in time or sinusoidal) [9]. In particular, the stain-rate dependence observed for the strength of some materials (polymers, rocks) can be understood as a consequence of the thermally activated behaviour predicted by equation (2) [10, 11].

Bueche, who was specifically interested in thermally activated rupture of polymer chains, observed for glassy polymers, i.e. at temperatures below the glass transition, an exponential dependence of the rupture time with applied force [12]. He derived a model where rupture occurs in a polymer chain when bonds are pulled apart up to a critical distance δ that is expected to be a fraction of an interatomic distance. He also assumed that applying a constant force \mathcal{F} to a bond decrease the rupture energy barrier E to be overcome by thermal activation such that $E = E_0 - \mathcal{F}\delta$, where E_0 represents now the bond energy. Finally, he suggested that the characteristic frequency τ_0^{-1} should be of the order of the bond vibration frequency. Finally, Bueche's prediction can be put exactly in the same form as equation (2) writing the activation volume: $V = A_0\delta$, where A_0 is the typical cross-sectional area per monomer through the rupture plane.

Zhurkov's kinetic theory of rupture [13] contains the same ideas than Bueche's theory. Zhurkov has verified experimentally for a wide range of temperatures that the scaling law equation (2) works reasonably well for many different materials (polycrystalline metals, alloys, non-metallic crystals, polymers). He compared the experimental value of the energy barrier E_0 with the sublimation energy and found they were close. His numerous observations came in support of the idea that time-dependent rupture results from thermally activated bond cut. In the case of polymers, Zhurkov found more direct evidence of bond rupture by measuring the rate of radical formation using EPR (Electron Paramagnetic Resonance) and showed it had the same qualitative and quantitative dependence on applied stress than the inverse of the rupture time.

Even though Busse, Bueche and Zhurkov have different physical arguments to justify their models (viscous flow, bond rupture), they all have in common that they predict the same dependence of the time-to-rupture on the applied stress. For convenience, we shall refer to equation (2) as the BBZ model.

2.2. Problems with the BBZ model

2.2.1. The scale of rupture. Although the simplicity of the BBZ model and the theories proposed to justify it are rather appealing, some difficulties arise when looking at the scale at which the rupture process occurs according to the models. In Table 1, we have reported or estimated the energy scale and the activation volume extracted from several experiments published in the literature. While it seems true that E_0 is of the order of magnitude of an atomic bond energy, we see right away that the length scale associated to the activation volume is consistently of the order of a few nanometres. This means that the thermally-activated rupture process described by the BBZ model should involve several hundred atomic bonds simultaneously. If one tries to estimate the critical distance δ in Bueche's model with a typical value of the cross-sectional area per monomer $A_0 = 70\text{\AA}^2$, one still finds $\delta \sim 1\text{nm}$ which can clearly not be interpreted as a critical atomic bond distance for rupture. Bueche was aware of this problem and tried, unsuccessfully, to correct his model by taking into account several possible effects, such as the progressive increase of load on polymer chains due to viscous relaxation of

	$T(^{\circ}\text{K})$	$V^{1/3}$ (nm)	E_0 (eV)
Cotton yarns [6]	411	1.2	1.79
Nylon yarn [9]	296	2.56	0.9
PEMA [12]	375	0.95	\emptyset
Polystyrene [12]	393	0.98	\emptyset
Polystyrene [16]	293	1.4	1.56
PMMA [13]	291	1.1	2.74
Zn - polycrist [13]	291 - 523	1.03	1.31
Al - polycrist [13]	291 - 573	1.65	2.29
Granite [10]	~ 300	2.4	1.9

Table 1. Characteristic energy E_0 and volume V estimated for various experimental data found in the literature using the BBZ model. E_0 is computed assuming $\tau_0 = 10^{-13}s$ as discussed in [13].

the load on other chains [14].

2.2.2. The limit of long rupture times. Another serious limitation of the BBZ model are the deviations from the linear dependence of $\log(\tau)$ with applied stress σ observed in many cases. For instance, both Bueche and Zhurkov have observed that at very low stresses or high temperatures, the rupture time is much longer than expected by simply extrapolating the behaviour at high stress or low temperatures. Note also that in the case of rubbery polymers, i.e. polymers at temperatures above the glass transition, the deviations can be especially large [15]. Bueche argued that, in the long rupture time regime, glassy polymers tend to behave like rubbery polymers for which he had made specific predictions based on a non-standard model of rubbery elasticity, but the agreement he found with experimental data remained qualitatively and quantitatively unsatisfying.

Zhurkov suggested that the deviations might arise from the reversibility of the rupture process at small applied stress [13], with the probability for an atomic bond to heal becoming equal to the probability to break at zero stress. Such a reversible process is actually naturally included in the non-approximated version of the BBZ model given by equation (1) which predicts, in the limit of small applied stresses, $\tau \propto \sigma^{-1}$. Following the idea that rupture is a reversible process, Kausch *et al* [17] have studied a fiber bundle model where the rate of bond rupture and bond healing is given by Eyring reaction-rate theory. Their model takes into account the progressive rupture of bonds, the corresponding increase of the load shared equally by the remaining bonds and assumes that the bonds break when a critical load is reached. The model is able to reproduce the divergence of rupture time at small stresses. However, in order to fit experimental data, Kausch *et al* chose, without any physical justifications, a time scale τ_0 , an energy scale E_0 and an activation volume V that were different for bond rupture and for bond healing [17]. In practice, the activation volume for bond healing was found to be 10 to

30 times smaller than the activation volume for bond rupture and the elementary rate of bound rupture 20 to 2000 times the elementary rate of bound healing. Note that, as seen in the previous paragraph, the order of magnitude of the activation volume for bond rupture was still about $(1 - 2 \text{ nm})^3$.

2.3. Elastic energy barrier models

A completely different explanation for the deviations from the BBZ model observed experimentally could be that the linear decrease of the energy barrier with the applied stress σ , assumed in Eyring reaction-rate theory, is simply not the correct functional dependence, even at large stresses. Several alternative scaling relations between the breaking time and the applied stress have been proposed in the case of purely elastic materials.

2.3.1. Taylor's model. In order to explain early experiments on time-dependent rupture in glass by Baker *et al* [18], Glathart and Preston have suggested the following empirical fit of their data [19]:

$$\tau = \tau_0 \exp\left(\frac{\sigma_0}{\sigma}\right) \quad (3)$$

In order to justify this empirical scaling, Taylor introduced shortly afterwards a model where a perfectly elastic material breaks when it reaches a critical elongation. Although the physical justifications behind Taylor's model are quite unclear, his core assumption is that the volume of material involved in the calculation of the energy barrier decreases proportionally to the inverse of the applied stress [20]. In the specific case of glass, Baker *et al* have shown that in addition to temperature, the atmosphere conditions (humidity) had a strong influence on the rupture dynamics (this behaviour has been studied extensively later by Wiederhorn [21], see also [22] for a recent review). This rupture regime, called stress-corrosion, comes from the fact the energy barrier for bond rupture in silicate glass involves a chemical reaction with water molecules and thus depends on the specific amount of humidity in the environment. Such effects involving a chemical reaction and the detailed change in the energy barrier related to it are out of the scope of the present review.

2.3.2. Global energy balance (GEB) models. More recently, Pomeau [23, 24] has determined the breaking time of materials under a constant load by assuming it comes from the nucleation of a critical crack with a size determined according to Griffith's rupture criterium [1]. Indeed, Griffith idea of energy balance between the energy required to create a crack surface and the total mechanical energy (elastic + potential energy of the external load) predicts that a solid stretched at constant load is in a metastable state. The energy barrier to overcome is then the one corresponding to the nucleation of a critical crack. This approach assumes that the material response is elastic up to the rupture point, a case which is usually referred to as brittle rupture. For a $2d$ geometry,

i. e. a plane of small thickness e , the nucleation of a critical Griffith crack across the plane will take a time:

$$\tau = \tau_0 \exp\left(\frac{\sigma_0^2}{\sigma^2}\right) \quad (4)$$

where $\sigma_0^2 \sim \gamma^2 Y e / k_B T$, Y is the Young modulus and γ is the energy needed to create a unit surface of the crack. For a $3d$ geometry, where the nucleated crack can be thought in first approximation as a spherical cavity, the breaking time is predicted to be:

$$\tau = \tau_0 \exp\left(\frac{\sigma_0^4}{\sigma^4}\right) \quad (5)$$

where $\sigma_0^4 \sim \gamma^3 Y^2 / k_B T$. Note that if one considers in $3d$ the nucleation of a crack corresponding to a straight needle cut of atomic diameter, one could expect to get a scaling with applied stress corresponding to the case of the $2d$ geometry where the thickness of the crack e is of the order of magnitude of an atomic size. Some authors have shown that the breaking time keep the same dependence on applied stress if one includes the effect of curviness of the crack [25]. The case where a crack pre-exists in the material changes slightly the energy barrier and has been expressed both for the $2d$ and $3d$ geometry [26, 27].

In the last 15 years, several experimental works have tried to verify Pomeau's prediction. Experiments in 2d rods of NBD stearic acids [28] seem to agree with the 2d prediction of equation (4) while experiments in wood composites [29], fiber glass composites [30] or sintered glass beads [31] agree better with the 3d prediction of equation (5). Some authors even generalized Pomeau's prediction to fractal dimensions arguing it corresponds to the case of gels [32]. However, note that from the experimental values of σ_0 , Y and T , obtained for instance in [30], one would find using a fit with equation (5) a value $\gamma \sim \mu\text{J.m}^{-2}$ [27]. This is an abnormally low value for the fracture energy suggesting that the energy barrier in equation (5) has been strongly overestimated.

One of the main fallout of Pomeau's model [23] and derivatives is that they neglect the possibility that rupture results from the accumulation of small irreversible rupture events in the material that would require a much lower energy barrier. Indeed, according to Pomeau's initial idea, a crack is supposed to reversibly explore due to thermal noise several sizes until it reaches the critical one. This might be relevant in the case of very large loads for which the critical crack size, and thus the energy barrier, is very small, but it will not in general describe properly the practical case of materials that can have rather large preexisting flaws that will not be able to close back in a reversible manner. Golubovic *et al* [33] have suggested that irreversibility of the rupture process arises as soon as the distance between the two crack lips is larger than an atomic size. Following this idea, they defined a critical crack size as the crack for which the maximum opening is larger than an atomic size. Their prediction yields a change in the stress dependence of the energy barrier such that, in a $2d$ geometry, the rupture time takes the form corresponding to equation (3) while in $3d$, it takes the form of equation (4).

Note that this approach neglects the time it takes afterwards for the crack to grow until it reaches the Griffith length. In order to predict what would happen in that case, the energy barrier corresponding to the progressive growth of a crack has been expressed by Santucci *et al* [26] assuming that, due to irreversibility, the Griffith potential could be progressively climbed in elementary steps δ , possibly of atomic size, up to the Griffith critical crack size. For a $2d$ geometry, this will give an energy barrier per unit thickness:

$$E = (G_c - G)\delta \quad (6)$$

where $G_c = 2\gamma$ is the critical elastic energy release rate corresponding to Griffith rupture criteria and G the elastic release rate corresponding to the actual crack length[‡], assumed here smaller than the Griffith critical size ($G < G_c$).

Another way to go around the problem of oversized barriers in Pomeau's model is to take into account the effect of disorder (or heterogeneity) in the material properties. For instance, in order to explain their experimental observations, Guarino *et al* [29] introduced the idea that the disordered nature of the samples could result in an effective temperature at least an order of magnitude higher than the thermodynamic temperature T . This idea has been mainly discussed in the context of local elastic barrier models (see paragraph 2.3.3). In the context of Global Energy Balance Models, Nattermann *et al* have shown how disorder could decrease the energy barrier and thus the critical crack size for rupture [34]. However, relying also on global energy balance models, Kierfeld *et al* [35, 36] have recently shown that, for a single crack, disorder might instead have the effect to slow down rupture and even cause the complete arrest of the crack (see also [37], for a similar prediction in the context of local elastic barrier models).

2.3.3. Local elastic barrier (LEB) models. In order to explain slow rupture at high temperature of sapphire whiskers, Brenner [38] took into account a preexisting crack and proposed that the energy barrier to overcome should correspond to the elastic energy increment needed to reach in a small volume near the crack tip a local rupture threshold. Brenner predicted that the rate of rupture p at the crack tip should have the following form:

$$p \propto \exp\left(-\frac{(\sigma_c - \sigma_m)^2 V}{2Yk_B T}\right) \quad (7)$$

where σ_c is the local value of the rupture stress and σ_m the mean stress in volume V . In order to understand Brenner's formula, let us consider for an elastic material the temporal stress fluctuations σ_f that occur around the statistical mean value σ_m in a given fixed volume V . Statistical physics predicts that these fluctuations follow a normal distribution:

$$p(\sigma_f) = \sqrt{\frac{V}{2\pi Yk_B T}} \exp\left[-\frac{(\sigma_f - \sigma_m)^2 V}{2Yk_B T}\right] \quad (8)$$

[‡] The elastic energy release rate is defined by the relation $G = -\partial U_m / \partial A$, with U_m the sum of the elastic energy and the potential energy of the external loading, and A the fracture surface area.

The probability to break, i.e. to have a fluctuation larger than the breaking threshold, is then: $P(\sigma_c - \sigma_m) = \int_{\sigma_c}^{\infty} p(\sigma_f) d\sigma_f = \frac{1}{2} \operatorname{erfc}[(\sigma_c - \sigma_m) \sqrt{V/2Yk_B T}]$. If the energy barrier is large compared to the energy of thermal noise, i. e. if $(\sigma_c - \sigma_m)^2 V \gg 2Yk_B T$, then it is possible to show that the volume V will break after a time:

$$\tau = \tau_0 \sqrt{\frac{2\pi V}{Yk_B T}} (\sigma_c - \sigma_m) \exp \left[\frac{(\sigma_c - \sigma_m)^2 V}{2Yk_B T} \right] \quad (9)$$

Equation (9) indeed shows that the rate of rupture ($\propto \tau^{-1}$) is dominated by the same exponential term as the one proposed by Brenner. Thanks to the local nature of the rupture process, the local energy barrier models have been very useful to describe thermally activated rupture in elastic disordered materials [37, 39, 40, 41, 42, 43, 44].

3. Slow crack growth under a constant load

The predictions of time to rupture presented in section 2 very often ignore the fact that rupture is usually a gradual process, involving many elementary rupture events. Two main scenarios of progressive rupture are often referred to: in the first one, the material break after the accumulation of many diffuse rupture events; in the second one, there is a main defect, a main crack, that will grow slowly until reaching a critical size. In this second part of the paper, we will be discussing mainly the slow growth of a crack. As we have seen previously, there is some uncertainty regarding the precise dependence of the energy barrier with the applied stress. Looking at the detailed dynamics of slow crack growth under constant stress can, in principle, help to discriminate between the various predictions. Indeed, under a constant load, the stress intensity factor will increase as the crack grows and the crack will accelerate. But, the acceleration of the crack will depend on the precise functional form of the energy barrier so that the crack length evolution with time will have different shapes for various models of the energy barrier. In addition, one might wonder if the experimental values of the activation volume, observed to be larger than expected for a model of bound rupture at the atomic scale, might be due to the failure of properly describing the progressive damage processes leading to rupture. In the following, we review some predictions made by models of brittle rupture where the material is assumed to be elastic up to the rupture point. Afterwards, we discuss the case of viscoplastic rupture for which predictions are more difficult to make and often rely on very phenomenological descriptions of the material mechanical properties.

3.1. Brittle materials

3.1.1. Slow crack growth in the GEB model: lattice trapping. Brittle materials are assumed to behave elastically up to the rupture point. For elastic materials, the Griffith energy balance approach predicts that, under a constant load σ , there exists a critical crack length ℓ_g corresponding to an unstable equilibrium point that verifies exactly the condition: $G = \Gamma$ where G is the elastic energy release rate and $\Gamma = 2\gamma$ the energy cost per unit area to create two fracture interfaces. Note that G is here an increasing

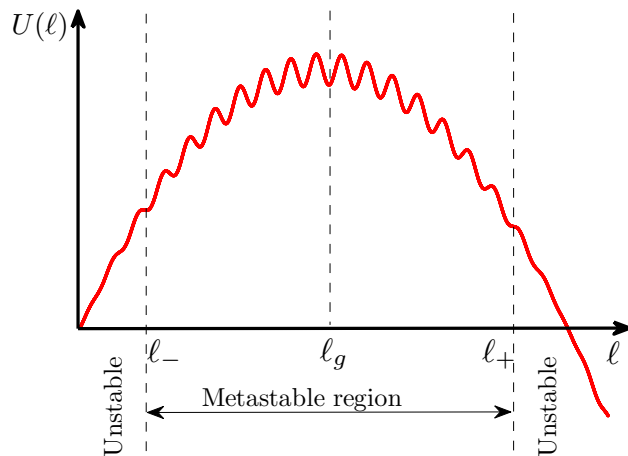


Figure 1. Thomson potential energy as a function of crack length ℓ and for a constant applied stress σ . For $\ell_- < \ell < \ell_+$, the crack can be trapped in metastable states, while for $\ell < \ell_-$ and $\ell > \ell_+$ it is always unstable. The Griffith length ℓ_g corresponds to a case where statistically the crack has the same probability to move forward or backward.

function of the crack length ℓ . If rupture is a reversible process and the crack length is such that $\ell < \ell_g$ (equivalently $G < \Gamma$), the crack will be unstable and close back entirely. On the other hand, if the crack length ℓ is such that $\ell > \ell_g$ (equivalently $G > \Gamma$), the crack will also be unstable and grow indefinitely. In order to explain the existence of stable cracks under a tensile load at zero temperature and their slow growth at finite temperature, Thomson *et al* have developed a lattice theory where the crack is trapped due to the discreteness of matter at the atomic scale δ [49]. The model assumes that this discreteness introduces a frozen spatial modulation of the fracture energy, with a minimum value $\Gamma_- < 2\gamma$ and a maximum value $\Gamma_+ > 2\gamma$. When $\Gamma_- < G < \Gamma_+$, the crack will be in a metastable state due to what Thomson *et al* have called a lattice trapping effect. At constant load, there will exist a length $\ell_- < \ell_g$ and a length $\ell_+ > \ell_g$ between which the crack can move only by thermally activated jumps over energy barriers (see figure 1). As shown below, the resulting motion will be either a slow growth or a slow healing of the crack depending on the value of the crack length. For $\ell > \ell_+$ (resp. $\ell < \ell_-$), the trapping barrier becomes zero and the crack can grow (resp. heal) quickly. That the trapping model explains the existence of a lower threshold for a crack to be able to start growing and also an upper threshold above which the crack becomes mechanically unstable has made this model realistic and thus attractive [50, 51, 52, 53, 54, 55, 56, 57]. Linearizing the trapping energy barrier, in the same spirit as done originally by Hsieh *et al* [50], we can write the energy barrier to move a discrete step δ_s forward E^+ and the energy barrier to move a discrete step δ_s backward E^- :

$$E^+ = (\Gamma_+ - G)\delta_s \quad \text{and} \quad E^- = (G - \Gamma^-)\delta_s \quad (10)$$

Note that the expression for E^+ is very similar to equation (6), except that the threshold Γ_+ is now larger than the Griffith one 2γ . Also, in equation (10), the energy E^+ and

E^- have been written per unit width of the crack front. If the crack front were to move at once, the energy barriers E^+ and E^- would have to be multiplied by the width of the crack front to get the actual energy barrier of the rupture process. Realizing that this was not very realistic since it would lead to very large energy barriers when the crack front is of macroscopic size, Hsieh *et al* [50] had originally suggested that the crack could advance by forming a small kink at the atomic scale and that afterwards the kink would propagate along the width of the crack front. With a kink mechanism, the energy barrier of the process can be roughly obtained by multiplying the energy barriers in equation (10) by a scale δ_k of the order of a few atomic sizes. The kink propagation mechanism itself has been described in detail by Marder [56]. Assuming that the healing of the crack is possible, the crack velocity can be written:

$$v \equiv \frac{d\ell}{dt} = v_0 \exp\left[-\frac{E^+\delta_k}{k_B T}\right] - v_0 \exp\left[-\frac{E^-\delta_k}{k_B T}\right] \quad (11)$$

which can be rearranged so that:

$$v = 2v_0 \exp\left[-\frac{\Delta\Gamma\delta_s\delta_k}{k_B T}\right] \sinh\left[\frac{(G - \Gamma_0)\delta_s\delta_k}{k_B T}\right] \quad (12)$$

where:

$$\Delta\Gamma = \frac{\Gamma_+ - \Gamma_-}{2} \quad \text{and} \quad \Gamma_0 = \frac{\Gamma_+ + \Gamma_-}{2} \quad (13)$$

The quantity Γ_0 represents the average value of the fracture energy, and when there is no lattice trapping ($\Gamma_+ = \Gamma_-$), the usual Griffith condition gives $G = \Gamma_0$. In equation (12), one notices that the crack velocity is zero when $G = \Gamma_0$, hence when the crack verifies Griffith condition for the mean value of the fracture energy. It is important to realize that while Γ_0 gives a threshold for crack propagation, it does not give a threshold for spontaneous healing which will occur only when $G < \Gamma_- (< \Gamma_0)$. Spontaneous fracture will on the other hand occur only when $G > \Gamma_+ (> \Gamma_0)$. Note that in order to observe spontaneous healing in the lattice trapping model, one will have to decrease G below Γ_0 by the same amount we would need to increase G above Γ_0 to observe spontaneous crack growth. If a thermally activated rupture process is allowed, slow healing of the crack will occur when $\Gamma_- < G < \Gamma_0$ and slow crack growth when $\Gamma_0 < G < \Gamma_+$.

In practice, slow crack growth will be observed in a reasonable amount of time if $(G - \Gamma_0)\delta_s\delta_k \gg k_B T$. In that case, eq. (12) reduces to:

$$v = v_0 \exp\left[\frac{(G - \Gamma_+)\delta_s\delta_k}{k_B T}\right] \quad (14)$$

One can apply equation (14) to determine the velocity of a crack of length ℓ centred in a plane sheet under a constant tensile stress σ perpendicular to the crack direction. In that case, $G = K^2/E = \pi\sigma^2\ell/2Y$, where $K = \sigma\sqrt{\pi\ell/2}$ is the mode I stress intensity factor. The upper limit of the fracture energy Γ_+ can be used to define a critical crack size $\ell_c \equiv \ell_+$ and a critical stress intensity factor K_c through the relation: $\Gamma_+ = K_c^2/E = \pi\sigma^2\ell_+/2Y$. Then, one can write the velocity:

$$v = v_0 \exp\left[-\frac{(K_c^2 - K^2)\delta_s\delta_k}{Yk_B T}\right] = v_0 \exp\left[-\frac{\pi\sigma^2(\ell_c - \ell)\delta_s\delta_k}{2Yk_B T}\right] \quad (15)$$

Note that this velocity increases exponentially with the crack length. Equation (15) is a differential equation that can be solved to find $\ell(t)$ with the initial condition that at $t = 0$, $\ell = \ell_i$:

$$t = \tau_G \left[1 - \exp \left(-\frac{\ell - \ell_i}{\zeta_G} \right) \right] \quad (16)$$

where we have introduced the characteristic length scale ζ_G :

$$\zeta_G = \frac{2Yk_B T}{\pi\sigma^2\delta_s\delta_k} \quad (17)$$

and the characteristic time τ_G :

$$\tau_G = \frac{\zeta_G}{v_0} \exp \left[\frac{U_G}{k_B T} \right] \quad (18)$$

with:

$$U_G = \frac{(K_c^2 - K_i^2)\delta_s\delta_k}{Yk_B T} = \frac{\pi\sigma^2(\ell_c - \ell_i)\delta_s\delta_k}{2Yk_B T} \quad (19)$$

Derivating again the solution with respect to time, one can write:

$$v = \frac{\zeta_G}{\tau_G - t} \quad (20)$$

It is clear from the last formula that τ_G will represent the rupture time of the sample while, as can be seen from equation (16), ζ_G sets a typical growth length before the crack length starts to diverge. We will compare these predictions with experimental results in paragraph 3.1.3.

3.1.2. Slow crack growth in the LEB model. In paragraph 2.3.3, we have introduced the LEB model which corresponds to a description where an intrinsic rupture threshold exists at a local scale. We insist that this rupture threshold does not come from a Griffith rupture criteria or from any global energy balance reasoning. In the LEB model, the stress at the crack tip is assumed to have a finite value and if this value is smaller than a rupture threshold σ_c , the crack will be effectively trapped. Contrary to what has been discussed previously, there is no need at this point to introduce a discretization at the atomic scale in order to create a trap. There are many possible reasons why, in an elastic material, one can expect to have a finite value of the stress at the crack tip. It will occur for instance if the crack tip has a well-defined radius of curvature. Alternatively, there might exist some cut-off length, related to the microstructure of the material, below which the stress can not diverge. Finally, in the spirit of Barenblatt, there might exist a cohesive zone of small fixed size that will prevent divergence of the stress [58] (the same calculation was also derived in a different context by Dugdale [59]). In any case, we will introduce a characteristic size λ so that the stress at the crack tip can be written:

$$\sigma_m = \frac{K}{\sqrt{\lambda}} \quad \text{where} \quad K = \sigma \sqrt{\frac{\pi}{2}} \ell \quad (21)$$

Now, the scale d at which thermally activated rupture occurs has nothing to do with the scale λ that acts as a cut-off for the stress divergence. Remember for instance that, in

the BBZ model, the rupture scale was about a nanometre (see table 1), and we expect a similar order of magnitude in the LEB model. The time τ to rupture a volume $V = d^3$ at the end of each crack tip is known from eq. (9). The crack velocity can be defined as:

$$v \equiv \frac{2d}{\tau} = v_0 \sqrt{\frac{2Yk_B T}{\pi(\sigma_c - \sigma_m)^2 V}} \exp \left[-\frac{(\sigma_c - \sigma_m)^2 V}{2Yk_B T} \right] \quad (22)$$

where we set $v_0 = d/\tau_0$. This differential equation is not as easy to solve as equation (15). However it is possible to solve an approximate version of this equation valid as long as the critical crack size ℓ_c is not too large compared to the initial crack size ℓ_i [26, 46]. With the initial condition that at $t = 0$, $\ell = \ell_i$ and defining $\sigma_i = \sigma_m(\ell_i)$, the solution $\ell(t)$ of equation (22) can be expressed as:

$$t = \tau_L \left[1 - \exp \left(-\frac{\ell - \ell_i}{\zeta_L} \right) \right] \quad (23)$$

where we have introduced the characteristic length scale ζ_L :

$$\zeta_L = \frac{2Yk_B T \ell_i}{\sigma_i(\sigma_c - \sigma_i)V} = \frac{4\pi Yk_B T \ell_i}{K_i(K_c - K_i)} \frac{\lambda}{V} \quad (24)$$

and the characteristic time τ_L :

$$\tau_L = \frac{\zeta_L}{v_0} \sqrt{\frac{\pi U_L}{k_B T}} \exp \left[\frac{U_L}{k_B T} \right] \quad (25)$$

with:

$$U_L = \frac{(\sigma_c - \sigma_i)^2 V}{2Y} = \frac{(K_c - K_i)^2 V}{2Y\lambda} = \frac{\pi\sigma^2(\sqrt{\ell_c} - \sqrt{\ell_i})^2 V}{4Y\lambda} \quad (26)$$

The prediction of the crack growth dynamics given by equation (23) is qualitatively the same than the one obtained with the GEB model. However, the characteristic length ζ_L , rupture time τ_L and energy barrier U_L have a different dependence on the physical parameters. In the next paragraph, we will compare both the LEB and GEB model predictions with experimental observations. The detailed statistics of crack jumps that are observed in these experiments and that can be successfully understood in the framework of the LEB model [60] will not be discussed here (for a review, see [46]).

3.1.3. An experimental test of the GEB and LEB models. We will now compare the predictions of both the GEB and LEB models with the experiments of Santucci *et al* [45, 46] on slow crack growth in a sheet of paper under a constant applied load. In addition to measuring the rupture time, Santucci *et al* have measured the crack length as a function of time and after averaging over several experiments in the same conditions, they have determined the characteristic growth length ζ using a fit with equation (16) or equivalently, equation (23) [45, 46]. In figure 2 (a) and 2 (b), we compare the experimental values of ζ with the predicted values $\zeta_G \cdot (\delta_s \delta_k)$ and $\zeta_L \cdot (V/\lambda)$ that can be computed using the experimental values of the applied stress σ , the initial crack length ℓ_i , the critical crack length ℓ_c (or critical stress intensity factor K_c), the temperature and the measured tensile modulus of paper $E = 3.3 \cdot 10^9$ GPa. We note in both cases that

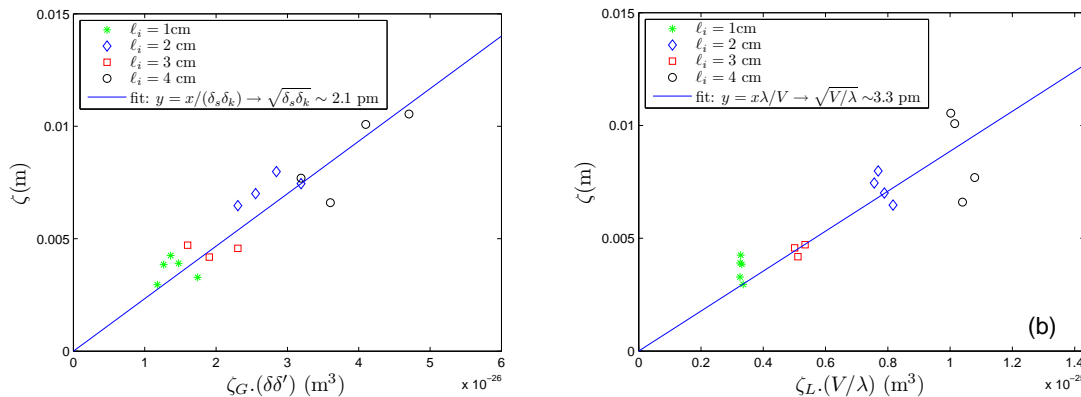


Figure 2. Experimental characteristic growth length extracted from average crack growth curves in a sheet of paper [45] compared with the prediction of (a) the GEB model and (b) the LEB model. Note that several values of initial crack length ℓ_i have been used. A linear fit allows to extract very similar values for the combined scales $\delta_s \delta_k$ and V/λ .

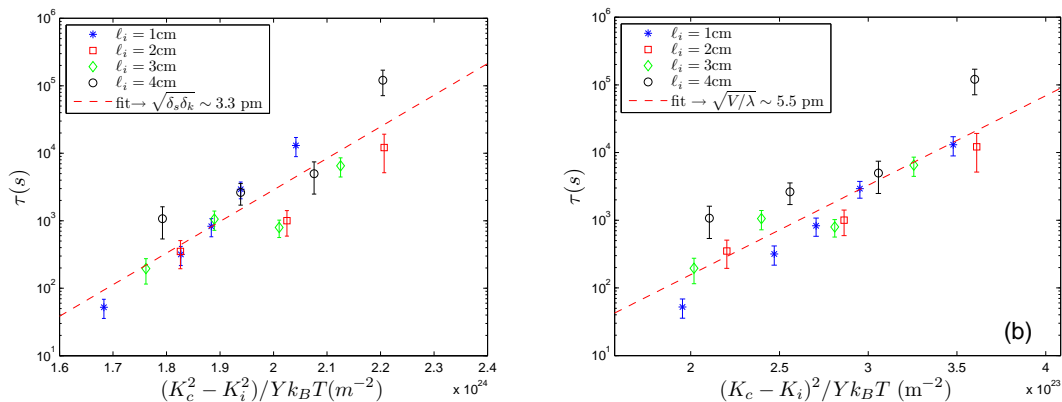


Figure 3. Experimental breaking time extracted from average crack growth curves in a sheet of paper [45] compared with the prediction of (a) the GEB model and (b) the LEB model. Note that several values of initial crack length ℓ_i have been used. A linear fit allows to extract very similar values for the combined scales $\delta_s \delta_k$ and V/λ .

there is good correlation between the experimental values and the models. From a linear fit, we can obtain the following values for the combined scales of rupture: $\sqrt{\delta_s \delta_k} = 2.1$ pm and $\sqrt{V/\lambda} = 3.3$ pm. In figure 3 (a) and 3 (b), we compare the logarithm of the rupture time with the energy barriers corresponding to both models, and we also find that they describe the experiments equally well. A fit enables us to extract independently another set of values for the combined scales of rupture: $\sqrt{\delta_s \delta_k} = 3.3$ pm and $\sqrt{V/\lambda} = 5.5$ pm. Although the combined scales determined from the rupture time τ and the ones from the characteristic length ζ are not exactly the same, they are close enough to consider that both the GEB and LEB models give a consistent picture of slow crack growth. Overall, there is apparently not much difference between the two models.

Let us now discuss in more details the scales involved in both models. In the GEB

model, δ_s is supposed to correspond to an interatomic distance while δ_k is supposed to be several interatomic distances. However, choosing $\delta_s = \delta_k$, we see from the comparison of this model with the experimental data that the typical scale is $\delta_s \sim 3 \cdot 10^{-12}$ m, which is clearly non-physical. If we force a more physical value for $\delta_s \sim \text{\AA}$, then the result is even worse since it yields $\delta_k \sim 10^{-14}$ m. In the LEB model, although the ratio V/λ is of the same order of magnitude than $\delta_s \delta_k$, the physical interpretation is quite different. The scale λ is a cut-off length at which the divergence of stress is suppressed. Santucci *et al* initially assumed that a natural cut-off length was the scale of the paper fibers, i.e. about $20 \mu\text{m}$ in diameter, which leads here to a value $V^{1/3} = 7 \pm 1 \text{\AA}$. This value is much more reasonable physically than the values obtained for δ_s or δ_k in the GEB model. Also, it is quite remarkable that we have now an activation volume that turns out to be not so different from the one usually found with the simpler BBZ model (see Table 1). Furthermore, it is important to realize that the results obtained with the LEB model would still give physical estimates of rupture scales if we had chosen a very different scale for the cut-off λ . For instance, it is well-known that paper fibers are made of cellulose which contain fibrils of diameter $D = 2.5$ nm [47] and Young modulus $Y = 138$ GPa [48]. These fibrils are nanofibres with a crystalline structure for which the LEB model is especially appropriate since they will break in a brittle manner. If we compute the stress at the scale of the nanofibres choosing $\lambda = 2.5$ nm and use the elastic modulus of the nanofibres, one finds now $V^{1/3} = 1.4 \text{\AA}$. Although it is questionable that the nanofibres is a physically correct cut-off scale for the stress divergence, the value obtained for the volume is now very close to an interatomic distance and thus is very consistent with the idea that thermally activated rupture occurs at the level of single atomic bonds.

3.2. Viscoplastic materials

3.2.1. Introduction. Many models of viscoplastic slow crack growth have been developed with the specific case of polymers in mind [54, 61, 62, 63, 64, 65, 66, 67]. However, most of these models correspond to the case of a linear viscoelastic material, and they have been shown to predict the correct behaviour mostly for elastomeric polymers or polymers in a rubbery state, i.e. at temperatures above the glass transition [63, 67]. Furthermore, breaking time laws such as the one given by the BBZ model discussed in section 2, work better in the case of polymers in a glassy state, i. e. below the glass transition temperature [12]. This is probably because viscous flow in glassy polymers typically follows an Eyring law, like in most solids, while the viscous flow of polymers in a rubbery state resembles more viscous flow in a fluid. Recent, direct evidences of fluid-like viscous crack propagation have been observed in gels for which the crack velocity can be either proportional to the elastic energy release rate [68] or have a power law dependence [69]. In our brief review of slow crack growth in viscoplastic materials under a constant load, we will mostly focus on the case of glassy polymers. Modelling the mechanical behaviour of polymers in a glassy state, and particularly the appearance of a yield stress and of strain hardening at large strains, has been recently

a very active topic [70, 71, 72, 73, 74, 75]. On the other hand, experimental works have mostly focused on understanding the fracture energy needed to propagate a crack, including the influence of strain-rate [76, 77]. Overall, there is relatively much less work regarding the slow crack dynamics in glassy polymers, both from an experimental and a theoretical point of view.

3.2.2. Slow crack growth in a polycarbonate film. We describe here some recent experimental results obtained on slow crack growth in polycarbonate films (for more details, see [78, 79]). A centred crack of initial length ℓ_i is loaded in mode I at constant velocity until reaching a stress σ that is kept constant during the slow growth of the crack until complete rupture of the sample. During the loading phase of the film, a macroscopic flame-shaped plastic zone appears at each tip of the crack and grows with the applied stress (cf. figure 4 where are defined ℓ , the crack length and, ℓ_{pz} the plastic zone length from tip to tip). This zone was previously observed by Donald and Kramer [80]. The formation of the plastic zone corresponds to a necking instability which brings the film thickness from $125\mu\text{m}$ to about $75 \pm 5\mu\text{m}$. Our polycarbonate films are isotropic and have the same mechanical properties than annealed polycarbonate. Consequently, the thickness of the plastic zone can be considered constant and uniform except in two small neighbourhoods close to the crack tip and close to the plastic zone tip [80].

Due to the influence of rate-effect on the polymer mechanical properties, the stress level in the plastic zone is expected to be dependent on the growth dynamics and must be understood as a dynamical stress. In order to estimate the stress in the plastic zone, we can use the Dugdale-Barenblatt cohesive zone model [58, 59] which predicts the relation between ℓ_{pz} and ℓ at equilibrium. Assuming that the stress in the plastic zone is uniformly equal to the plastic yield stress σ_y of the material, Dugdale predicts:

$$\sigma_y = \frac{\pi}{2} \frac{\sigma}{\arccos\left(\frac{\ell}{\ell_{pz}}\right)}. \quad (27)$$

The experimental values of ℓ , ℓ_{pz} and σ can be used to estimate σ_y . Here, we are using engineering stresses but could have equivalently used true stresses since, as mentioned above, the thickness of the plastic zone can be considered constant almost everywhere. Note that equation (27) does not require the plastic zone to be small compared to the crack size and is obtained from the condition that the stress intensity factor is zero at the tip of the plastic zone. It is also important to realize that the values of σ_y obtained in that way are very consistent with the values of the yield stress that can be determined directly from a tensile test in a polycarbonate strip without any cracks [79, 81].

Typical growth curves of the fracture and plastic zone are shown in figure 5. We can see right away that the prediction of slow crack growth for brittle materials, equation (16) or (23), can not describe the shape of the crack growth curve observed here, showing first a slowing down of the growth until the length ℓ_x and an acceleration afterwards. Furthermore, the shape of the accelerated part of the crack motion is incompatible with the prediction obtained for brittle crack growth [81].

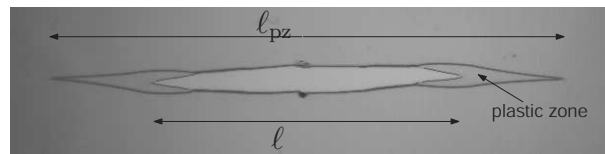


Figure 4. Image of a crack in a polycarbonate film with its macroscopic plastic zone at each tip. ℓ is the crack length and ℓ_{pz} is the plastic zone length from tip to tip.

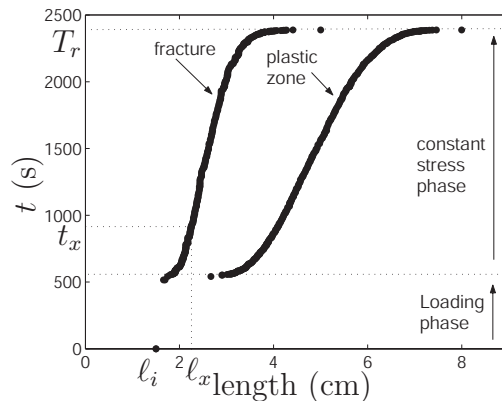


Figure 5. Time as a function of both the crack and plastic zone lengths for an imposed stress experiment ($\ell_i = 1.5\text{cm}$, $F = 900\text{N}$). We indicate the position of the inflexion point t_x , ℓ_x of the crack growth curve.

We have seen in section 2 that the Eyring model for creep flow had been proposed as a possible interpretation of the observed stress dependence of breaking times for many different materials. In addition, several authors have observed that plastic zones formed during a necking instability can grow in creep conditions and that their growth velocity also follow an Eyring law [79, 82]. Thus, we could expect that the crack growth dynamics is controlled, at least partly, by the creep dynamics of the plastic zone. In figure 6(a), we plot the instantaneous crack growth velocity as a function of the stress σ_y for eight experiments performed with various experimental conditions. Although there is not a direct correlation between crack velocity and the plastic stress σ_y , it is important to realize that the behaviour observed at the beginning of the experiment, when the crack is slowing down, give a correlation that is not very far from an Eyring law with a slope quantitatively close to the one observed for the creep velocity of the necking instability [79]. We discovered that introducing a correction to σ_y linear with the crack length ℓ allows us to collapse the experimental data on a straight line (cf. figure 3.2(b)). This correction can be written:

$$\sigma_y^{\text{corr}} = \sigma_y + \kappa(\ell - \ell_x) \quad (28)$$

where it is found that $\kappa = (3.4 \pm 0.6) 10^8 \text{N.m}^{-3}$. This rescaling means that the crack growth velocity follows:

$$v = v_0 \exp(\alpha \sigma_y^{\text{corr}}) \quad (29)$$

where $\alpha = 6.8 10^{-7} \text{m}^2.\text{N}^{-1}$. Interestingly, there are indications that $\kappa \ell_x = \sigma_s - \sigma$, where

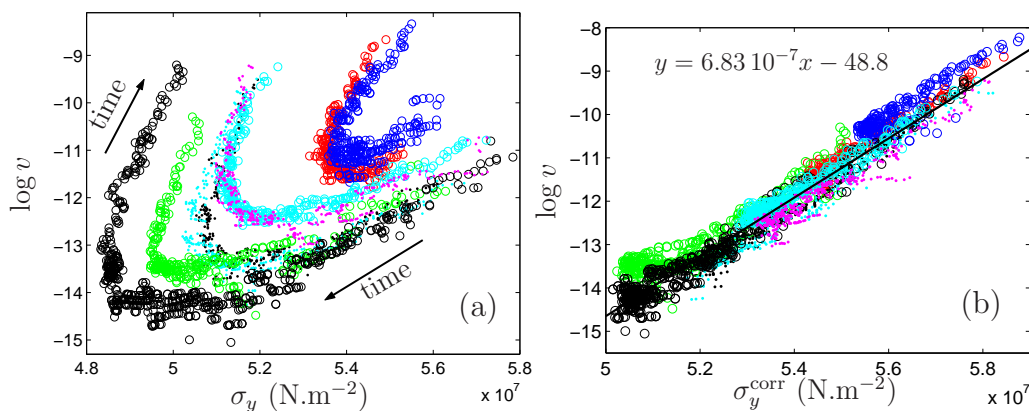


Figure 6. Natural logarithm of the instantaneous crack growth velocity as a function of (a) the Dugdale-Barenblatt stress, (b) the corrected Dugdale-Barenblatt stress σ_y^{corr} according to equation (28) for eight experiments performed with various experimental conditions ($\ell_i = 1.5, 2, 3\text{cm}$ and $2.9 < \sigma < 3.8 \cdot 10^7 \text{N.m}^{-2}$). In figure (b), the black line is the result of a linear data fit.

σ_s represents the breaking stress of the polycarbonate film when there is no crack [79]. In the following, we emphasize two important features of the phenomenological law, equation (29), obtained for the instantaneous crack velocity.

First, one can interpret the exponential dependence of the velocity as a function of σ_y^{corr} in a rather simple way if one compares this scaling with the Eyring creep law of polycarbonate relating its strain rate $\dot{\epsilon}$ to the applied stress σ [8]:

$$\dot{\epsilon} = \dot{\epsilon}_1 \exp(\sigma V/k_B T) \quad (30)$$

Creep experiments performed at room temperature on our own polycarbonate samples give $V/k_B T = 7.67 \cdot 10^{-7} \text{m}^2 \cdot \text{N}^{-1}$ [79]. The fact this prefactor $V/k_B T$ is of the same magnitude than α in the exponential law for the crack velocity (cf. equation (29)) reinforces the idea that the creep of the plastic zone plays an important role in the mechanisms of crack growth. If one identifies now $\alpha \simeq V/k_B T$, we find $V^{1/3} = 1.4 \text{nm}$. This value is close to the one normally obtained for the breaking time of polymers when the BBZ model is used (see table 1).

Second, it is striking that the linear correction in ℓ that was added to define σ_y^{corr} introduces an exponential dependence of the instantaneous velocity on the crack length ℓ . Such a dependence was already predicted in the case of brittle slow crack growth, both in the GEB and LEB models. The characteristic growth length scale $\zeta_{\text{VP}} = (\alpha \kappa)^{-1} \simeq 4.3 \text{mm}$ is also of the same order of magnitude than the one obtained in the case of slow crack growth in a sheet of paper (see paragraph 3.1.3). However, it is not clear yet whether or not ζ_{VP} depends on the applied stress σ in the same way as in the GEB and LEB models.

Thus, equation (29) means that the crack growth velocity in a glassy polymer such as polycarbonate can be expressed roughly as the product of a creep velocity, that takes into account the rate dependence of the plastic stress, times a brittle-like velocity, that depends exponentially on the crack length. As a consequence of this important property,

we can expect that slow crack growth curves in any solid materials, even viscoplastic ones, will have the same shape than for a brittle material as long as the influence of rate effects on the plastic stress remains small, such that it can be considered constant during the whole crack growth.

3.2.3. Possible origin of the brittle-like term in viscoplastic materials. Here, we want to emphasize that it is not obvious to understand the physical reason behind the observed dependence of the crack velocity on crack length in polycarbonate films. The size of the plastic zone that Dugdale predicted was obtained by noting that the total stress intensity factor at the tip of the plastic zone must vanish. Hence, as it was already discussed by Rice [54], the elastic energy release rate must also be zero. All the variations of elastic energy during crack growth have to go into the plastic zone so that nothing can be used to help the crack grow. This argument has led Chudnovsky *et al* [67] to postulate the existence of an out of equilibrium situation where the total stress intensity factor is not exactly zero. However, allowing for a time relaxation of the fracture energy needed to propagate a crack, Chudnovsky finds a crack growth law that has exactly the same form than the one obtained in linear viscoelastic models [62].

Another approach to solve the problem could be to recognize that, once the plastic zone has been formed by necking, it can behave again elastically (it is well-known that the plastic plateau ends after the necking instability occurred and is followed by an hardening elastic response [84]). Then, it is very likely that there will be a stress intensification at the crack tip. A rough estimate of the stress intensification for an ideally straight crack and assuming that the plastic zone behaves with the same elastic response as the rest of the film, would predict that the stress is larger than twice the applied stress σ at distances from the crack tip smaller than about 0.16ℓ . Since we have a ratio $\ell_{pz}/\ell \sim 2$ and thus $\sigma_y/\sigma \sim 1.5$ [79], we can conclude right away that most of the plastic zone is submitted to a Dugdale plastic stress while stress intensification remains confined in a smaller region close to the crack tip. Now, if one applies the LEB model using the local stress at the crack tip, the rate of rupture will increase with crack length and we can expect to recover the observed dependence on crack length. Thus, we reach again the conclusion that it is easier to understand the experimental observations by reasoning at a local scale rather than by reasoning using global energy balance models.

4. Conclusion

A large corpus of experiments shows that the breaking time of solid materials submitted to a constant load is proportional to an Arrhenius factor with an energy barrier that decreases with the applied stress. An exception is, for instance, the case of polymers in a rubbery or elastomeric state. The Arrhenius behaviour is a clear sign that thermally activated processes cause rupture and this picture is reinforced by several experiments showing how breaking time depends on temperature. We have reviewed several models that are consistent with experiments and differ mainly with the precise functional

dependence of the energy barrier on the applied stress. The BBZ model, that can be viewed as a first order approach to describe experimental data, leads to experimental values of the maximum energy barrier close to the theoretical value for the rupture of atomic bonds. On the other hand, it predicts a scale of rupture of the order of a few nanometres which is a little bit too large if bond rupture is to be the correct mechanism. We have also discussed various GEB models, based on global energy balance as inspired by Griffith's original approach, and have shown they often tend to overestimate the size of the energy barrier, and thus lead to unphysical predictions. Finally, we have introduced the LEB model which considers that the rupture rate depends only on the local state of stress in the material and how far this stress is from an intrinsic rupture threshold. A major fault of many of the models presented here is that they neglect the progressive accumulation of damage before complete macroscopic rupture of a sample.

In the second part of this review, we have focused on the growth of a single crack under a constant load, which is a very simple case where the slow dynamics of rupture can be followed in time. We have first discussed the case of brittle materials that have a simple linear elastic response up to rupture. In the framework of the GEB model, lattice trapping effects can help understanding the existence of a range of crack length for which thermally activated crack growth exist. In the framework of the LEB model, the crack is naturally in subcritical condition as long as the local stress at the crack tip is smaller than the rupture threshold. Both the GEB and LEB model predict that the crack velocity should increase exponentially with crack length. Both models also introduce two characteristic length scales in the problem. However, comparisons with recent experiments on paper sheets, show that the GEB model fails to predict physically realistic values for both length scales that are found to be much too small. This is in our view another proof that the GEB model overestimates the energy barrier. On the other hand, the LEB model leads to much more physical values for the length scales. One of them seems to correspond well to a characteristic cut-off scale of the stress divergence at the crack tip while the other one is coherent with a rupture process at the atomic scale. Finally, we have discussed the case of viscoplastic materials by focusing on recent experimental results obtained by us. We have shown that in a glassy polymer such as polycarbonate the crack velocity can be obtained, roughly, as the product of a creep velocity, taking into account strain-rate effects on the plastic stress, times a velocity with the same functional dependence on crack length than the crack velocity in brittle materials. We argue that the LEB model is the best framework to understand the remarkable appearance of a brittle-like term for the instantaneous crack velocity in a glassy polymer.

Acknowledgments

L. Vanel acknowledge the support of grant ANR-05-JCJC-0121-01.

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