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Abstract. – We present experiments on the slow growth of a single crack in a fax paper sheet submitted to a constant force F. We find that statistically averaged crack growth curves can be described by only two parameters : the mean rupture time τ and a characteristic growth length ζ. We propose a model based on a thermally activated rupture process that takes into account the microstructure of cellulose fibers. The model is able to reproduce the shape of the growth curve, the dependence of ζ on F as well as the effect of temperature on the rupture time τ. We find that the length scale at which rupture occurs in this model is consistently close to the diameter of cellulose microfibrils.

Introduction. – A critical stress threshold is often defined to characterize material resistance to rupture. When the applied stress is smaller than the threshold, experiments show that rupture can still occur after a delay in time which decreases with the applied stress and with temperature [1–3]. A widespread approach has been to relate time-dependent fracture to the creep properties of the material [4,5]. In this general framework, the creep law is an empirical material dependent property, the most common dependence on the applied stress proposed in the literature being either a power law or an Arrhenius law. More recently, several authors have come back to a simpler situation where the material is elastic and subcritical rupture is thermally activated. The focus has been mainly on prediction of the rupture time (or lifetime) [6–8], but has also been extended to the slow growth of a single crack in brittle materials [9,10]. In this Letter, we present experimental results on the slow growth of a single crack in a sheet of paper which behaves as a quasi-brittle material. We show that the experimental results are compatible with a model of thermally activated crack growth in brittle materials when we take into account the microstructure of cellulose fibers in paper.

Experimental set-up. – Crack growth is obtained by loading in mode 1 at a constant force F a sheet of fax paper (Alrey) with an initial crack in the center (fig. 1a). The sample dimensions are : height $h = 21$cm, width $w = 24$cm, and thickness $e = 50\mu$m. The experimental set-up consists of a tensile machine driven by a motor (Micro Controle $UE42$) controlled electronically to move step by step (Micro Controle $ITL09$). The paper sheets are mounted on the tensile machine with both ends attached with glue tape and rolled twice over rigid bars clamped on jaws. The motor controls the displacement of one jaw (400 steps per

micrometer) while the other jaw is rigidly fixed to a force gage (Hydrotonics-TC). The tensile machine is placed in a box with a humidity level stabilized at 5%. In order to work on samples with the same initial crack shape and length L_i, we use calibrated razor blades mounted on a micrometric screw and we initiate a macroscopic crack precisely at the center of the sheet. The samples are loaded by increasing the distance between the jaws such that the resulting force F is perpendicular to the initial crack direction.
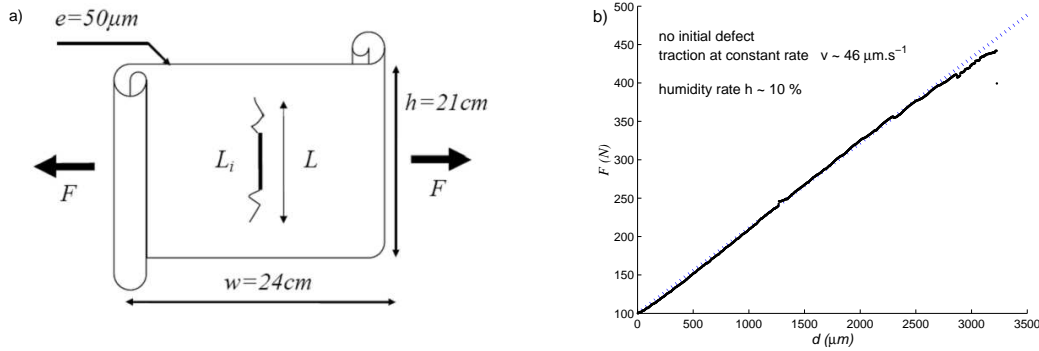

Fig. 1 – a) Sample geometry. b) Linear dependence between applied force and elongation until rupture.

A feedback loop allows us to adjust the displacement in order to keep the applied force F constant with a precision better than 0.5N and a response time less than 10ms. From the area A of a cross-section of the sheet, A being approximatively constant, we calculate the applied stress $\sigma_e = F/A$.

Physical properties of paper. – Sheets of fax paper break in a brittle manner. This is evidenced by the elastic stress-strain dependence which is quasi-linear until rupture (fig. 1b). Another sign that rupture is essentially brittle is given by the very good match between the two opposite lips of the fracture surfaces observed on post-mortem samples.

A sheet of paper is a complex network of cellulose fibers. Scanning electron microscopy on our samples shows fiber diameters between 4 and 50μm with an average of 18μm. Cellulose fibers are themselves a bundle of many microfibrils. Cellulose microfibrils have a cristalline structure (therefore, they are very brittle) and are consistently found to have a diameter $d = 2.5nm$ [11].

The mechanical properties of paper depend crucially on the humidity rate. To get reproducible results, the fax paper samples are kept at least one day at a low humidity level ($< 10\%$) and during the experiment ($\simeq 5\%$). At constant humidity level ($h \simeq 5\%$) and room temperature, the Young modulus of the fax paper sheets is typically $Y = 3.3\,10^9 \mathrm{N.m^{-2}}$.

Direct observation and image analysis. – We light the samples from the back. A high resolution and high speed digital camera (Photron Ultima 1024) collects the transmitted light and allows us to follow the crack growth. We observe that the global deformation of the paper sheet during a creep experiment is correlated in a rather reproducible way to the crack growth whatever the rupture time. We use this property to trigger the camera at fixed increment of elongation (one micron) rather than at fixed increment in time. This avoids saturation of the onboard memory card when the crack growth is slow and makes the acquisition rate faster when the crack grows faster and starts to have an effect on global deformation. We acquire 2 frames at 250fps at each trigger and obtain around one thousand images per experiment.

Image analysis is performed to extract the length of the crack projected on the main direction of propagation, i.e. perpendicular to the direction of the applied load (fig. 2). Although the crack actually follows a sinuous trajectory, its projected length L gives the main contribution to the stress intensity factor K which we compute as: $K = g(\pi L/2h)\sigma_e\sqrt{\pi L/2}$, where $g(x) = \sqrt{\tan x/x}$ takes into account the finite height h of the paper sheet [12].
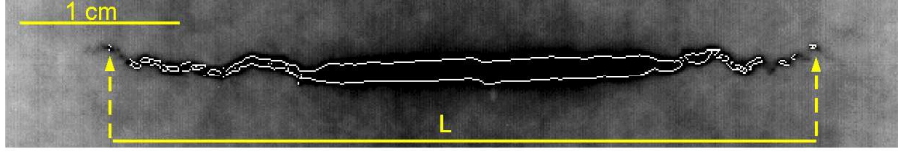

Fig. 2 – Extraction of the projected crack length L from the crack contour detected.

Single crack growth. – For a given initial crack length L_i, subcritical crack growth is obtained by applying a constant force F so that $K(L_i)$ is smaller than the critical rupture threshold of the material K_c above which fast crack propagation would occur. During an experiment, the crack length increases, and so does the stress intensity factor $K(L)$. This will cause the crack to accelerate until it reaches the critical length L_c for which $K(L_c) = K_c$.

On fig. 3a we show a typical growth curve obtained during a creep experiment with an applied force $F = 270$ N and an initial crack length $L_i = 1$cm. Since time to rupture τ is a statistical quantity, we prefer to plot time evolution as a function of the crack length. We observe that the crack growth is actually intermittent. Essentially, there are periods of rest during which the crack tip is pinned and does not move, and also moments when the crack suddenly opens and advances of a certain step size s. The crack advances by jumps until it reaches a critical length L_c where the paper sheet breaks suddenly. Measurements of L_c are used to estimate the critical stress intensity factor $K_c = 6 \pm 0.5\,\mathrm{MPa.m}^{1/2}$. Beyond L_c, the crack runs across the whole sample (about 18cm in this case) in less than one second, with a crack speed $v > 5\mathrm{m.s}^{-1}$. For the same experimental conditions (same stress, same initial crack length, same temperature and same humidity rate), we observe a strong dispersion in growth curves and in lifetime while the critical length seems to be rather well defined (see insert in fig. 3). In order to characterize both the average crack growth and the stepwise growth dynamics, a statistical analysis is required. In this Letter, we are going to focus our study on the average dynamics. The intermittent dynamics and in particular the step size statistics has already been described elsewhere [13].

Statistically averaged crack growth. – We have performed an extensive study of crack growth, varying the initial crack length from $L_i = 1$cm to $L_i = 4$cm and the applied force between $F = 140$N and $F = 280$N (corresponding to an initial stress intensity factor between $K_i = 2.7\mathrm{MPa.m}^{1/2}$ and $K_i = 4.2\mathrm{MPa.m}^{1/2}$) and repeating 5 to 20 experiments in the same conditions (stress, initial crack length, temperature and humidity rate). The resulting measured lifetime varied from a few seconds to a few days depending on the value of the applied stress or the temperature. In order to characterize the average growth dynamics, we examine for given experimental conditions the average time $\langle t \rangle(L)$ the crack takes to reach a length L.

Even though the lifetime distribution is large and the growth dynamics intermittent, the average growth offers a regular behavior. A typical mean growth, obtained by averaging ten experiments in the same conditions, is plotted in fig. 3b. This growth is qualitatively very
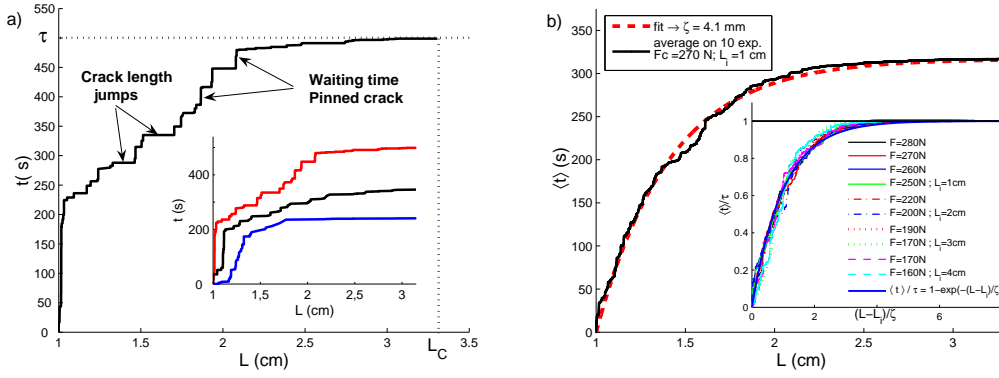
Fig. 3 – a) Typical stepwise growth curve for a creep experiment with an initial crack length $L_i = 1$cm submitted to a constant load $F = 270$N. The lifetime of the sample is $\tau = 500$s and the critical length $L_c = 3.3$cm. In insert, a strong dispersion is observed in crack growth profile and in lifetime for 3 creep experiments realized in the same conditions. b) Statistical average of growth curves for 10 creep experiments realized in the same conditions ($L_i = 1$cm, $F = 270$N). The dashed line corresponds to a fit using equation eq. (1) with a single free parameter $\zeta = 0.41$cm. Insert: rescaled average time $\langle t \rangle / \tau$ as function of rescaled crack length $(L - L_i)/\zeta$ for various initial crack lengths and applied stresses. The solid line corresponds to eq. (1).

close to an exponential evolution:

$$\langle t \rangle = \tau \left[1 - \exp\left(-\frac{L - L_i}{\zeta} \right) \right] \tag{1}$$

Indeed, we obtain a very good fit of the data in fig. 3b with eq. (1) setting the mean lifetime to the experimentally measured value and using ζ as a unique free parameter. Using the same procedure, we extract the characteristic growth length ζ for various experimental conditions. In the insert of fig. 3b, rescaling the crack length by ζ and the time by τ for many different experimental conditions, we show that the data collapse on the functional form given by eq. (1). Moreover, we have checked that the deviation from the predicted average behaviour is smaller when increasing the number of experiments.

We find that the experimental value of ζ is approximatively proportional to $1/F^2$ (fig. 4a). We also observe that the rupture time for all the experiments performed at room temperature is essentially a function of the initial stress intensity factor K_i (symbols with an error bar on fig. 5a. However, for a fixed value of applied force F and initial length L_i, varying temperature between 20°C and 120°C leads to variations of the rupture time up to four order of magnitude (symbols without error bar on fig. 5a).

Model of thermally activated crack growth. – In this part, we are going to describe a model that is able to reproduce several of the experimental features that we have just described.

First, we recall that the experimental crack growth curves have been obtained by performing a statistical average on many experiments in the same condition. The model will also be a statistical one. We are going to consider that in average the crack growth proceeds by breaking individual cellulose fibers at the crack tip. Still in average, the fibers who resist to the applied load will be considered parallel to each other and perpendicular to the crack direction. If λ is the typical diameter of a fiber and τ_L is the mean rupture time for the cellulose fiber at the
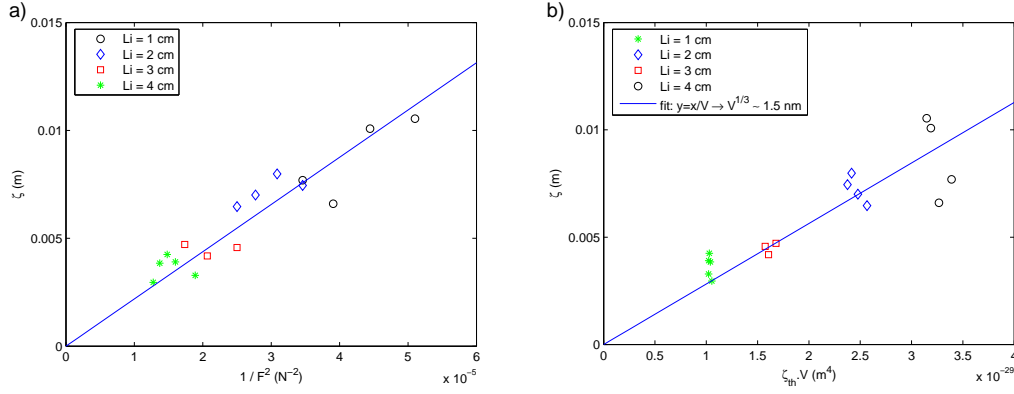
a)

b)

Fig. 4 – a) Experimental value of ζ extracted from the average growth profile as a function of $1/F^2$. b) ζ as a function of the prediction of the thermally activated rupture model. The line represents the best linear fit $y = x/V$. Its slope permits us to obtain a characteristic length scale for rupture: $V^{1/3} \sim 1.5\,\text{nm}$.

crack tip, the mean crack velocity is : $v = \lambda/\tau_L$. In that way, the increase in crack velocity as the crack length increases will correspond to a decrease of the rupture time τ_L.

Whatever is the rupture mechanism, we expect that the rupture time τ_L is going to depend on the applied load f on the fiber at the crack tip. Equivalently, we can refer to the applied stress $\sigma_L = f/S$ where $S \sim \lambda^2$ is the cross-sectional area of the fiber. We estimate σ_L assuming that the fiber acts as a cohesive zone of length λ and that the rest of the material behaves as an elastic continuum for a crack with a length $L + \lambda$. Following [14], we consider that the crack is at equilibrium if the elastic field presents no divergence of stress. This condition imposes that the stress level in the fiber is controlled by the size of the fiber and can be estimated as: $\sigma_L \propto \sigma_e \sqrt{L + \lambda}/\sqrt{\lambda} \sim \sigma_e \sqrt{L/\lambda}$. In our calculations, we will take $\sigma_L = K/\sqrt{2\pi\lambda}$ where K is
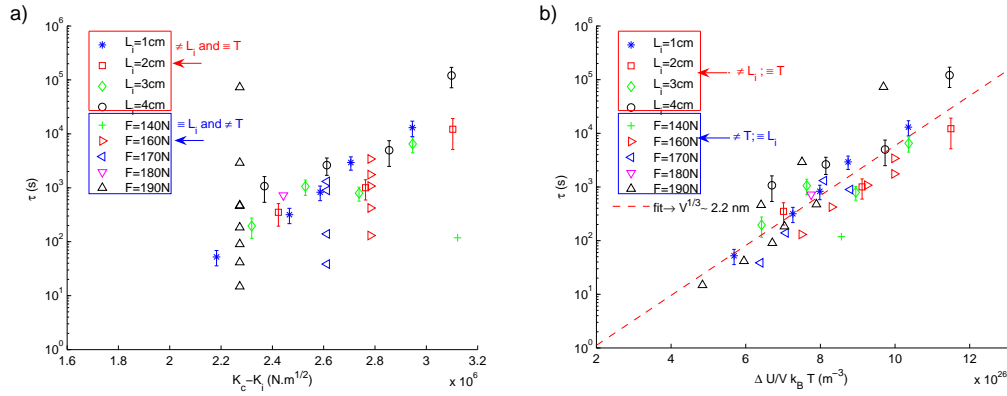
a)

b)

Fig. 5 – a) Logarithm of the mean rupture time τ as a function of $K_c - K_i$. Data points without error bars correspond to non-averaged measurements obtained by varying temperature with $L_i = 2$cm and various fixed values of F. b) Logarithm of lifetimes as a function of the dimensional factor $\Delta U/Vk_BT$ predicted by eq. (2) for different values of L_i, F and T. The best fit $\log \tau \propto \Delta U/k_BT$ (dashed line) slope gives an estimation of the characteristic length scale $V^{1/3} \sim 2.2$nm.

the stress intensity factor.

A common approach to describe the creep rupture of materials is to introduce a time-dependent creep compliance or a rate of rupture which is a power law of the applied stress. Instead of assuming a phenomenological law for the creep behavior, we are going to use a statistical approach that has been developed in the case of brittle materials to describe the rupture of fiber bundles. This approach is appropriate in the case of a cellulose fiber because it is actually a bundle of many identical brittle microfibrils. The idea of the rupture mechanism is that at equilibrium there are always statistical fluctuations of stress with a variance depending on the actual temperature of the material. Taking into account the probability that such stress fluctuations exceed the rupture threshold f_c of individual fibers in the bundle, it is possible to make some prediction on the rupture dynamics of a fiber bundle. In an homogenous fiber bundle where all the fibers have the same rupture thresholds, it was shown that the rupture time is [15,16]:

$$\tau \sim \exp\left[\frac{(f_c - f_0)^2}{2\alpha k_B T}\right]$$

where k_B is Boltzmann constant, T is temperature, α is the elastic stiffness of the fibers, f_0 is the applied force per fiber when the bundle is still intact, and f_c is the rupture threshold of the fibers. We can rewrite this expression in term of the Young modulus Y, the applied stress on the fiber σ_L and the rupture threshold σ_c. If $s = \pi d^2/4$ is the section area of a microfibril, we have $f_0 = \sigma_L s$ and $f_c = \sigma_c s$. Rupture in a microfibril is expected to occur in a small piece of the fiber with a length ℓ much smaller than the total fiber length. The relation between the stiffness α of this piece and the Young modulus is then: $\alpha = Ys/\ell$. Introducing the volume $V = s\ell$, we see that the rupture time can be written as :

$$\tau_L \sim \exp\left[\frac{(\sigma_c - \sigma_L)^2 V}{2Y k_B T}\right]$$

It must be understood that in this formula V represents the volume in which rupture of the microfibril actually occurs. Since the microfibril is broken whenever we cut a section through, it is physically reasonable to consider that the length ℓ of the piece involved in rupture will be comparable to the microfibril diameter d. Thus, we will expect to have $V \sim d^3$.

The expression obtained for τ_L gives an exponential dependence of the crack growth velocity. If we integrate this velocity following the procedure described in [10], we recover eq. (1) where the rupture time is :

$$\tau \sim \exp\left(\frac{\Delta U}{k_B T}\right), \qquad \text{with} \qquad \Delta U = \frac{(\sigma_c - \sigma_i)^2 V}{2Y} \tag{2}$$

and the characteristic growth length ζ:

$$\zeta = \frac{2Y k_B T}{V} \frac{L_i}{\sigma_i(\sigma_c - \sigma_i)}, \tag{3}$$

where σ_i is the value of σ_L when $L = L_i$.

Writing explicitly the dependence of stresses on F, we obtain from eq. (3) that $\zeta \propto 1/F^2$ in agreement with the experimental observations (fig. 4a). We have also $\zeta \propto \sqrt{L_i/(L_c - L_i)}$ but this dependence is difficult to verify experimentally because the ratio L_c/L_i does not change much. In fig. 4b, we see that there is a reasonable agreement between the experimental value of ζ and the theoretical one $\zeta V = 2Y k_B T L_i/[\sigma_i(\sigma_c - \sigma_i)]$. Using V as a free parameter, we find a characteristic scale $V^{1/3} = 1.5nm$ close to the microfibril diameter d.

The form of the energy barrier ΔU in the Arrhenius law (eq. (2)) is analogous to the one obtained for example in [9]. Note also that it is not simply a function of the external load σ_e as in [6–8]. In fig. 5b, we plot the rupture time as a function of $\Delta U/V k_B T = (\sigma_c - \sigma_i)^2/2Y k_B T$. We observe that the data points obtained by varying temperature are now collapsing with the one obtained at fixed temperature (the dispersion in this figure remains large because data as a function of temperature and constant F and L_i have been averaged less than those at constant temperature for technical reasons). From a fit of the data, we obtain independently a new estimate of V which gives a characteristic scale $V^{1/3} = 2.2nm$. Once again this estimate is close to the microfibril diameter d.

Conclusion. – We have studied the subcritical growth of a single crack in a sheet of paper during creep experiments. We have shown that statistically the crack growth curves can be well described with only two parameters: the rupture time τ and a characteristic growth length ζ. We have proposed a model which is able to reproduce the shape of the growth curves as well as the dependence of ζ with the applied load and the influence of temperature on the rupture time. The model takes advantage of the fact cellulose fibers are actually a bundle of many small brittle fibers with a diameter of nanometer scale. A previous prediction of the rupture time for thermally activated rupture of brittle fibers has been used to derive the crack velocity. We find that the experimental data are compatible with the idea that rupture occurs at a nanometric scale in the cellulose fibers. While the model we used work rather well, it remains to understand if the experimental results could be as well described by time-dependent fracture models based on the material rheological properties. Such a study would require for example a detailed investigation of the physical properties of individual fibers.

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