Slow crack growth in polycarbonate films

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Abstract. – We study experimentally the slow growth of a single crack in polycarbonate films submitted to uniaxial and constant imposed stress. The specificity of fracture in polycarbonate films is the appearance of flame-shaped macroscopic process zones at the tips of the crack. Supported by an experimental study of the mechanical properties of polycarbonate films, an analysis of the stress dependence of the mean ratio between the process zone and crack lengths, during the crack growth, shows a quantitative agreement with the Dugdale-Barenblatt model of the plastic process zone. We find that the fracture growth curves obey strong scaling properties that lead to a well-defined growth master curve.

Introduction. – Solids with a single crack usually break at a critical rupture stress. However, experiments [1] show that a given solid submitted to a subcritical stress breaks after a certain amount of time. Therefore, understanding the mechanisms of subcritical macroscopic fracture growth in solids has become an important goal of fracture physics in order to improve the resistance of structures to failure. Recent experimental works [2,3] have shown that sub-critical crack growth in paper can be successfully described by a thermal activation model for elastic brittle media. In this letter, we present an experimental study of slow growth of a single crack in a polycarbonate film which is a highly non-brittle material. We observe that a large flame-shaped area, the process zone, forms ahead of each crack tip. We find that the dependence of the process zone length with the applied stress, during the crack growth, is in reasonable quantitative agreement with the Dugdale-Barenblatt model. In that respect, polycarbonate appears to be a good model material to understand the mechanisms of crack growth in non-elastic media. We show that the crack growth curve obeys remarkable scaling properties that are not theoretically understood yet.

The experimental setup and the experiment. – The experiment consists in loading $125\,\mu m$ thick isotropic polycarbonate films (height $21\,cm$, length $24\,cm$) with uniaxial and constant imposed stress σ. The polycarbonate films used are made of Bayer Makrofol® and present the properties of bulk material. An initial crack of length ℓ_i is initiated at the center of each polycarbonate sample using calibrated blades of different lengths (from $0.5\,cm$ to $3\,cm$). Then, a constant force F_c is applied to the film perpendicularly to the crack direction, so that we get
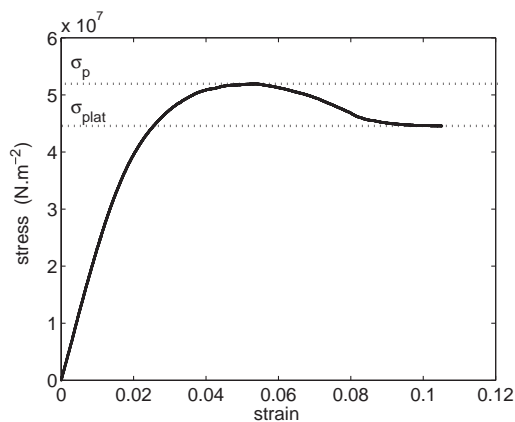
Fig. 1 – Stress as a function of the strain for a $125\,\mu$m thick polycarbonate film loaded with a $46.25\,\mu$m s^{-1} strain rate.

a mode 1 crack opening type. For more details about the setup see [2,3]. A high-resolution and high-speed camera (Photron Ultima 1024) is used to follow the crack growth.

We follow the growth of the single linear fracture and its process zones, under constant applied stress σ, till the total rupture of the sample. The applied stress σ is chosen such that crack growth is slow, *i.e.*, smaller than the critical one, σ_c, above which a fast crack propagation occurs.

Mechanical properties of polycarbonate films. – In order to characterize the material in which the crack will grow, we performed some preliminary experiments on polycarbonate films, without crack, submitted to uniaxial deformation at a constant deformation rate ($46.25\,\mu$m s^{-1}). A typical experimental stress-strain plot is presented in fig. 1. The polymer films show the classical behavior of a plastic material with a quasi-elastic behavior for small strains followed by a bell profile and a plateau. The different characteristic values observed on this graph are in good agreement with the ones measured by Lu and Ravi-Chandar in bulk polycarbonate [4]. We measured the experimental values of the maximum reachable stress, $\sigma_p = 5.2\,10^7\,$N m^{-2}, the plastic plateau stress $\sigma_{plat} = 4.45\,10^7\,$N m^{-2} and the Young modulus for small strains $Y = 194\,10^7\,$N m^{-2}.

The flame-shaped process zone ahead of the crack tip. – In each experiment, during the loading phase of the film, a macroscopic flame-shaped process zone appears at each tip of the
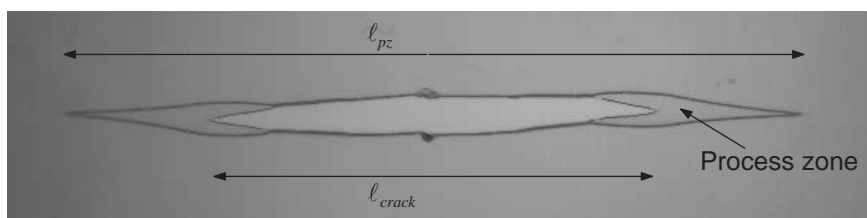

Fig. 2 – Image of a crack in a polycarbonate film with its macroscopic process zone at each tip; ℓ_{crack} is the crack length and ℓ_{pz} is the process zone length from tip to tip.
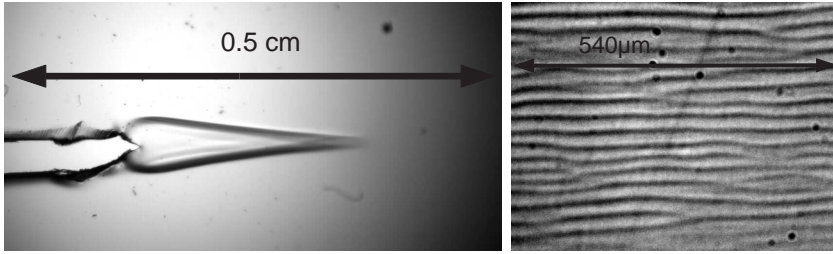
Fig. 3 – On the left: process zone at the tip of a growing crack in a polycarbonate film; on the right: microscopic image showing striations quasi-parallel to the fracture direction in the process zone.

crack and grows with the applied stress (cf. figs. 2 and 3). This zone was previously noticed by Donald and Kramer [5]. In the late loading stage, the fracture may also grow a little. Consequently, the real experimental initial condition, obtained when the constant stress σ is reached, is not exactly ℓ_i. During the imposed stress stage, the process zone and the fracture are both growing till the final breakdown of the sample in a way that the fracture never catches up the process zone tip.

Inside the process zone, the film is subjected to a thinning which brings its thickness from $125\,\mu$m to about $70\,\mu$m (measured on *post mortem* samples). It is worth noticing that on microscopic images (cf. fig. 3) one can see in the process zone the presence of striations quasi-parallel to the fracture front with a wavelength of about $22\,\mu$m. These striations seem to be thickness oscillations of the film. It is still an open question whether this process zone, once it has been formed, continues to behave as a visco-plastic zone or as an elastic zone with an effective macroscopic Young modulus different from the one of the rest of the film. Understanding the mechanical nature of the process zone is the key to reach a model of fracture growth in polycarbonate.

Dependence of the process zone length on the fracture length. – The experimental relation between the process zone length (defined in fig. 2), ℓ_{pz}, and the crack length, ℓ_{crack}, during the crack growth process, is plotted in fig. 4a). It seems that this relation is not statistical because all curves for identical experimental conditions are almost identical. The experimental stress ranges for different ℓ_i do not overlap, so that it is not possible to plot the curves for the same ℓ_i and very different σ. However, it is expected that the ℓ_{pz} against ℓ_{crack} curve depends on the applied stress only.

It is important to notice that during an experiment the fracture length grows on time scales much larger than the time scale of relaxation of the process zone shape. This has been checked by suddenly forcing the fracture length to grow using a blade during an imposed stress experiment. This action induces a quasi-instantaneous growth of the process zone to its new equilibrium state. So, it is appropriate to think that during the fracture experiments, the process zone is in a quasi-equilibrium state for each given crack length (below the critical one) and applied load. Therefore, we can analyze these data using an equilibrium model for the process zone.

The relation between ℓ_{pz} and ℓ_{crack} has first been theoretically described by the Dugdale-Barenblatt model [6,7]. Dugdale considers the process zone as an isotropic plastic material in which the stress is uniformly equal to the plastic yield stress σ_y. Assuming the non-divergence of the stress at the tip of the process zone, he concludes to a zero stress intensity factor (SIF) [8] at the tip of the process zone: $K_{tot} = K_{el}(\sigma; \ell_{pz}) + \tilde{K}(\sigma_y; \ell, \ell_{pz}) = 0$, where K_{el} is the traditional SIF at the tip of a fracture of length ℓ_{pz} in an elastic film submitted to σ at its border and $\tilde{K}$ the SIF for a film fractured on a length ℓ_{pz} and submitted only to σ_y on the fracture lips
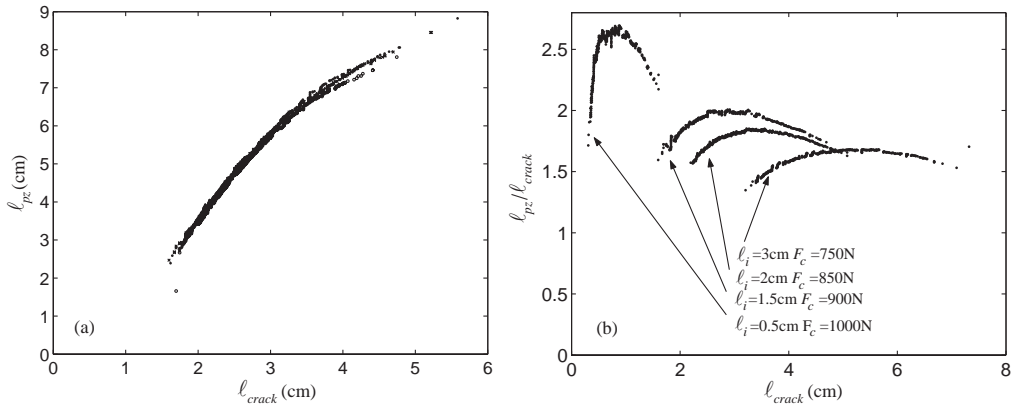
Fig. 4 – On the left: process zone length as a function of the crack length for three experiments performed in the same experimental conditions ($\ell_i = 1.5$ cm and $F_c = 900$ N); on the right: ℓ_{pz}/ℓ_{crack} ratio for four experiments performed with different experimental conditions as a function of the crack length.

between ℓ_{crack} and ℓ_{pz}. Using analytical expressions for these SIFs in the case of an infinite elastic sample, Dugdale finds a proportionnality dependence of ℓ_{pz} on ℓ_{crack}: $\ell_{pz}/\ell_{crack} = 1/\cos(\frac{\pi}{2}\sigma/\sigma_y)$. This model was successfully compared with experimental data in metals [6]. However, for many polymers, it does not predict the correct order of magnitude for the process zone size if one uses the yield stress constant of the material as the Dugdale stress constant [9–11]. In fig. 4b) it appears clearly that the ℓ_{pz}/ℓ_{crack} ratio is not constant during a constant stress experiment, so that the ℓ_{pz} against ℓ_{crack} relation is not a proportionality relation.

To make a more quantitative comparison with the model, we represent, in fig. 5, the ℓ_{pz}/ℓ_{crack} ratio as a function of the applied stress σ for four experiments performed with different applied stresses. The dispersion of the data for each experiment corresponds to the important discrepancy from a proportionality law between the two lengths. For each of the four
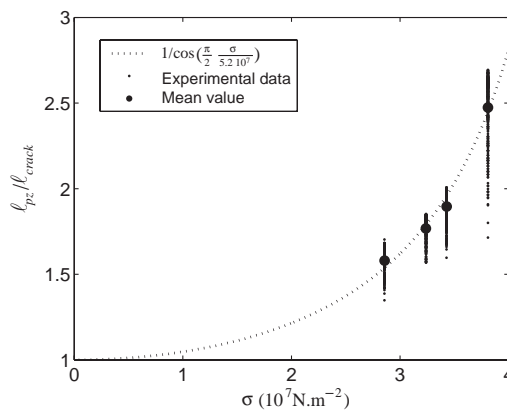

Fig. 5 – ℓ_{pz}/ℓ_{crack} ratio as a function of the applied stress σ for four experiments performed with different experimental conditions and the fit of its mean values (the larger points correspond to the mean value of the data for each σ) by the Dugdale-Barenblatt law.
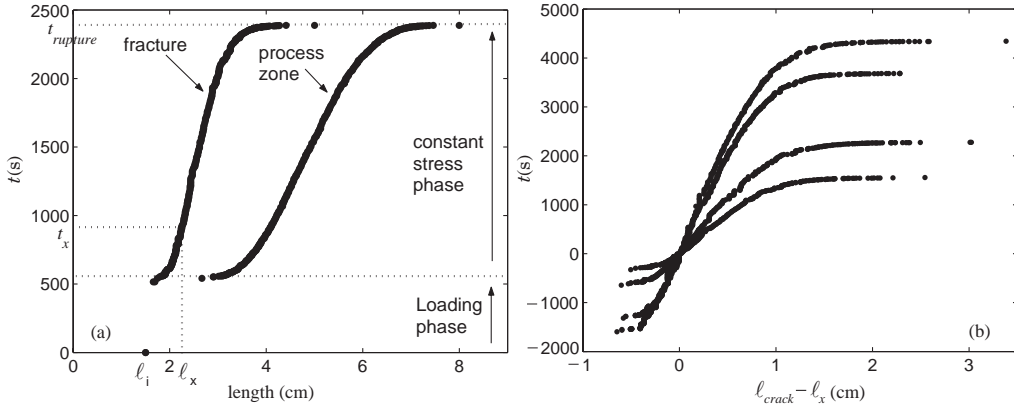
Fig. 6 – On the left: time as a function of both the crack and process zone lengths for an imposed stress experiment ($\ell_i = 1.5\,\mathrm{cm}$, $F_c = 900\,\mathrm{N}$); on the right: time as a function of $\ell_{crack} - \ell_x$ for four experiments performed in the same experimental conditions ($\ell_i = 1.5\,\mathrm{cm}$, $F_c = 900\,\mathrm{N}$) with a change in time and length origins.

experiments, the mean value of the ℓ_{pz}/ℓ_{crack} ratio is superimposed on the data. Fitting these mean values against σ with the Dugdale-Barenblatt law, $\ell_{pz}/\ell_{crack} = 1/\cos(\frac{\pi}{2}\sigma/\sigma_y)$, leads to an estimate of the Dugdale stress constant: $\sigma_y = 5.2\,10^7\,\mathrm{N\,m^{-2}}$. It is striking how the value found here for σ_y is close to the plastic peak stress σ_p of the rheology curve of fig. 1. So, even if the Dugdale-Barenblatt model does not predict the non-linear dependence of ℓ_{pz} on ℓ_{crack}, it predicts the correct order of magnitude for the ratio ℓ_{pz}/ℓ_{crack}, as a function of σ, using a plastic stress constant σ_y that corresponds very well to the maximum reachable stress σ_p obtained from mechanical tests (cf. fig. 1). We insist that it is not usually the case for polymers. We believe that the reason it works reasonably well is that, for polycarbonate, plasticity remains confined close to the crack tip while for many other polymers plasticity is much more diffuse.

Regarding the non-proportionality of the curves in fig. 4a), finite-size corrections to the Dugdale-Barenblatt model lead to an opposite curvature [11] to the one observed experimentally. An explanation for the experimental curvature, at the beginning or at the end of the experiment, could be dynamical effects that bring the process zone out of equilibrium. However, for the main part of the data, crack growth is slow and process zone equilibrium models need to be improved to explain the observed non-linearity.

Breaking time and crack growth curves. – The measured breaking times for a given experimental condition (ℓ_i, F_c) are statistical. Actually, we commonly observe a factor five between the smallest and the largest breaking times.

Typical growth curves of the fracture and process zone are shown in fig. 6a). Both curves show a quite similar smooth shape. We observe, once the loading phase is finished, at the beginning of the constant stress phase, large velocities of the fracture and the process zone tips which decrease till reaching quasi-constant values before increasing back dramatically till the final rupture. So, the first part of the growth curves in the constant stress phase corresponds to a deceleration of the fracture tips. We think this phenomenon is caused by delays in deformation accumulated during the loading phase. In other words, this deceleration corresponds to the time needed by the viscous effects to relax all those delays to quasi-equilibrium. The final acceleration corresponds to a transition to fast crack dynamics.

We can point out some important scaling properties of these growth curves. Actually,
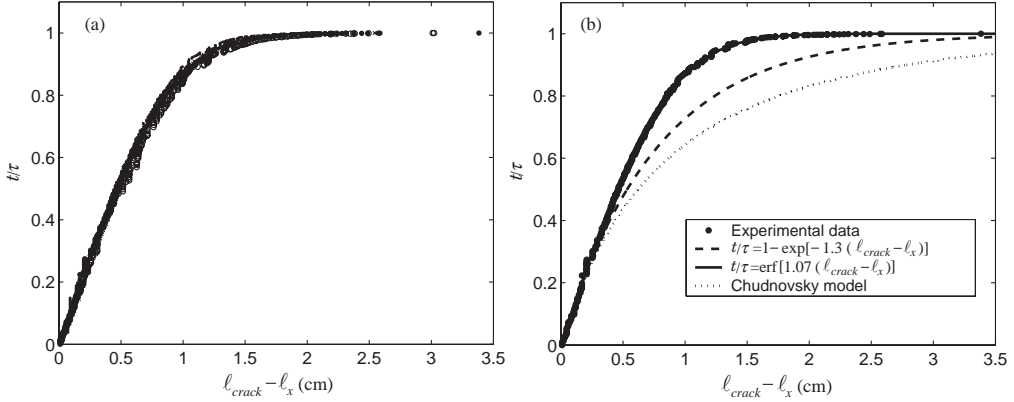
Fig. 7 – On the left: time rescaled by the time left to rupture as a function of $\ell_{crack} - \ell_x$ for four experiments performed in the same experimental conditions ($\ell_i = 1.5\,\text{cm}$, $F_c = 900\,\text{N}$, same experiments as for fig. 6b)) with a change in time and length origins; on the right: time rescaled by the time left to rupture as a function of $\ell_{crack} - \ell_x$ for an imposed stress experiment ($\ell_i = 1.5\,\text{cm}$, $F_c = 900\,\text{N}$) and fits by the Chudnovsky model, the thermal activation model and an error function adjusting the initial velocity.

by redefining the origin of time and length using the coordinates of the inflexion point (ℓ_x and t_x) of the fracture curve (cf. fig. 6b)) and rescaling the time by the time left to break τ ($\tau = t_{rupture} - t_x$), all the growth curves corresponding to identical experimental conditions fall on a master curve (cf. fig. 7a)). By introducing a rescaling factor on the lengths, we can as well make all growth curves, for different experimental conditions, fall on the same master curve. This rescaling property implies that the dynamics of the final part (the part after the inflexion point) of the crack growth follows:

$$t = \tau(\sigma, \ell_i, \text{realization})\, g\left(\frac{\ell - \ell_x}{\lambda}\right),\tag{1}$$

where g is the functional form of the master curve. λ and ℓ_x are *a priori* functions of σ and ℓ_i. In this formula, all the statistics is included in τ. No conclusive study of $\lambda(\ell_i, \sigma)$ has been possible due to the limited ranges of ℓ_i and F_c experimentally accessible.

In searching for an analytical expression for g, we first tried to use previously established models. Santucci *et al.* [2, 12] developed a thermal activation model that can explain sub-critical single crack growth in elastic brittle films. This model that has been successfully faced with experimental data for slow crack growth in paper [2, 3], predicts an exponential growth law: $t = \tau[1 - \exp[-\frac{\ell - \ell_i}{\xi}]]$. In fig. 7b), we can see that this thermal activation law (dashed line) does not work with experimental data for polycarbonate. Obviously, it is not a surprise, since polycarbonate is far from being simply an elastic medium at least because of the macroscopic plastic process zone. There are very few models giving an equation of motion for crack growth in viscoplastic media. A tentative model is the one of Chudnovsky [13] that has been developed first for stick-slip crack growth description in polyethylene films. This model introduces an aging process to describe the creep of the process zone. By using the Dugdale relation for process zone size, Chudnovsky reached a continuous expression for the crack velocity as a function of the elastic stress intensity factor. The curve obtained by a numerical integration of this equation is shown in fig. 7b) (dotted line). It also does not fit the experimental data well. Without any theoretical arguments, it appears that the error function

is a quite good guess candidate for an analytical description of g as we can see on the data fit of fig. 7b) (solid line).

Conclusion. – The experimental study of the process zone of polycarbonate film cracks allowed us to analyze the relevance of the Dugdale-Barenblatt model. We have shown that the mean ratio between the process zone and crack lengths, as a function of the stress, agrees with this model. We have also found a characteristic growth curve for crack in polycarbonate films which cannot be explained by recent models based either on thermally activated rupture or on surface energy aging due to viscoplastic flow. It is important to introduce a more precise description of the rupture mechanisms in the process zone in order to model crack growth in polycarbonate. One direction is to consider the general framework of viscoplastic rupture introduced by Kaminskii [14, 15]. Work is in progress to test the applicability of this model to crack growth in polycarbonate.

REFERENCES

[1] ZHURKOV S. N., *Int. J. Fract. Mech.*, **1** (1965) 311.
[2] SANTUCCI S., VANEL L. and CILIBERTO S., *Phys. Rev. Lett.*, **93** (2004) 095505.
[3] SANTUCCI S., CORTET P. P., VANEL L. and CILIBERTO S., *Sub-critical crack growth in paper: a thermally activated process*, in preparation.
[4] LU J. and RAVI-CHANDAR K., *Int. J. Solids Struct.*, **36** (1999) 391.
[5] DONALD A. M. and KRAMER E. J., *J. Mater. Sci.*, **16** (1981) 2967.
[6] DUGDALE D. S., *J. Mech. Phys. Solids*, **8** (1960) 100.
[7] BARENBLATT G. I., *Adv. Appl. Mech.*, **7** (1962) 55.
[8] At a distance r from a crack tip, the local stress behaves as $K/\sqrt{r}$, where K is the stress intensity factor.
[9] STOJIMIROVIC A., KADOTA K. and CHUDNOVSKY A., *J. Appl. Polym. Sci.*, **46** (1992) 1051.
[10] HADDAOUI N., CHUDNOVSKY A. and MOET A., *Polymer*, **27** (1986) 1377.
[11] STOJIMIROVIC A. and CHUDNOVSKY A., *Int. J. Fract.*, **57** (1992) 281.
[12] SANTUCCI S. *et al.*, *Europhys. Lett.*, **62** (2003) 320.
[13] CHUDNOVSKY A. and SHULKIN Y., *Int. J. Fract.*, **97** (1999) 83.
[14] KAMINSKII A. A., *Sov. Appl. Mech.*, **15** (1979) 1078.
[15] KAMINSKII A. A., *Int. Appl. Mech.*, **40** (2004) 829.