

CALIBRATION OF A COHESIVE ZONE FOR CRAZING IN GLASSY POLYMERS

N. Saad, R. Estevez, C. Olagnon, R. Séguela.
GEMPPM INSA Lyon, 20 av A. Einstein, 69621 Villeurbanne cedex, France

ABSTRACT

Failure of glassy polymers involves two mechanisms: shear yielding and crazing. Shear yielding refers to (visco-)plasticity localized in shear bands and is related to the bulk constitutive law with intrinsic softening upon yielding followed by re-hardening at continued deformation. Crazing is the mechanism preceding crack nucleation. It is also a viscoplastic process but at a smaller scale and consists in three steps: initiation, thickening and breakdown resulting in crack nucleation.

A finite element [1] analysis featuring shear yielding and crazing has shown that the toughness is governed by the competition between these two mechanisms and its evolution with loading rate appears consistent with experimental observations. In particular, a description of crazing with a viscoplastic cohesive zone [2] is able to capture the main features of glassy polymers fracture, at least qualitatively.

The present experimental study aims to identify the parameters involved in this FE analysis for quantitative predictions. Those involved in the constitutive law are necessary to describe shear yielding and are derived from uniaxial compression tests. The calibration of the parameters of the cohesive zone requires separate analyses of the three stages of crazing. Initiation is assumed to occur for a critical stress state which is derived from experiments in which a stress gradient is introduced. Once initiated, a craze thickens up to a critical opening that can be measured by interferometry. The energy dissipated during the viscoplastic craze thickening corresponds to the energy for crack nucleation. Therefore, the calibration of the viscoplastic craze thickening is based on measurements of the evolution of the energy release rate with loading rate, using specimens with sharp notches. Details of the preparation of the specimens and a protocol for the calibration of the parameters are presented for PMMA.

1 INTRODUCTION

Failure of glassy polymers is generally thought to be ductile when shear yielding takes place while crazing is associated to a brittle response. Shear yielding refers to (visco-)plasticity localized in shear bands and is related to the intrinsic softening upon yielding followed by re-hardening at continued deformation of the bulk. Crazing precedes crack nucleation and is also a viscoplastic process but at a smaller scale. It consists in three steps: initiation, thickening and breakdown for a critical thickness resulting in crack nucleation.

In a recent modeling of glassy polymer fracture, Van der Giessen and coworkers [1,2] have incorporated in a finite element framework a viscoplastic description for shear yielding and a cohesive surface formulation for crazing. A computational analysis presented in [1] has shown that the toughness is governed by the competition between these two mechanisms. The predicted evolution of toughness with loading rate is qualitatively in agreement with experimental observations. In particular, the model is able to capture the ductile to brittle transition observed at low loading rates and also a slight increase of the toughness with increasing loading rate, when the bulk is elastic.

This paper deals with an experimental analysis of glassy polymer fracture within the framework of the above mentioned modeling. The aim is to define a complete protocol to calibrate the parameters involved in the description of both processes. Since the condition for craze breakdown has been extensively investigated by interferometry with data available for instance in [3, 4], the present study focuses on the characterization of the first two stages of crazing: initiation and thickening. A setup with a specimen subjected to a stress gradient is employed for the analysis of craze initiation and the identification of the more appropriate criterion in terms of critical stress

state. Once initiated, the craze thickens up to a critical value for which a crack is nucleated. The energy related to the process of thickening corresponds to that necessary to nucleate a crack, which identifies to the energy release rate when the bulk response is elastic. As craze thickening is a viscoplastic mechanism, the energy for crack nucleation is rate dependent. Therefore, the characterization of the parameters involved in the description for craze thickening is based on measurements of the toughness at various loading rates. In order to illustrate the methodology, a calibration of the parameters of crazing for PMMA is reported.

2 COHESIVE ZONE FORMULATION FOR CRAZING

Motivated by the Kramer and Berger [5] description of the crazing process, Tijssens et al. [2] proposed a viscoplastic description for crazing within the framework of a cohesive zone methodology. The traction-separation law proposed in [2] consists of three parts corresponding to initiation, thickening and breakdown of a craze.

The physical mechanism for craze initiation is not yet clearly identified and various criteria have been proposed depending on the assumed mechanism and length scale (see [6] for a review). A craze is supposed to initiate by a stress and temperature controlled mechanism. Therefore, Sternstein and Ongchin [7] have proposed a stress bias criterion for initiation at a given temperature which is reformulated in [1] for the cohesive surface as

$$\sigma_n \geq \sigma_m - \frac{A^0}{2} + \frac{B^0}{6\sigma_m}, \quad (1)$$

under plane strain conditions. Here, σ_n and σ_m correspond to the normal and mean stresses at the plane of initiation. The coefficients A^0 and B^0 are material parameters and are temperature dependent [8]. Since craze thickening involves an intense viscoplastic activity within the active zone [5], the process is taken to be time and temperature dependent. The craze thickening rate $\dot{\Delta}_n^c$ is taken as [1,2]

$$\dot{\Delta}_n^c = \dot{\Delta}_0 \exp\left\{\frac{-A^c \sigma^c}{T} \left(1 - \frac{\sigma_n}{\sigma^c}\right)\right\}, \quad (2)$$

with $\dot{\Delta}_0$, A^c and σ^c being material parameters. Craze breakdown is supposed to occur when the craze thickness reaches a critical value Δ_n^{cr} . This parameter is of major importance since it governs the onset of crack propagation when craze fibrils break down.

The three stages of the crazing process are combined in the traction-opening law

$$\dot{\sigma}_n = k_n (\dot{\Delta}_n - \dot{\Delta}_n^c), \quad (3)$$

with $\dot{\Delta}_n$ the normal opening rate of the cohesive surface, $\dot{\Delta}_n^c$ the thickening rate of the craze according to Eq. (2) and k_n an elastic stiffness. The traction-opening law in Eq. (3) is used for the three stages of the crazing process. Prior to craze initiation, $\dot{\Delta}_n^c$ is not relevant and Eq. (3) reduces to $\dot{\sigma}_n = k_n \dot{\Delta}_n$ in which the stiffness k_n has to be 'infinite' to ensure the elastic opening to remain small and do not significantly affect the continuity of the fields. The expression in Eq. (3) is used during craze thickening. At the onset of craze fibrils breakdown, the normal stress vanishes as

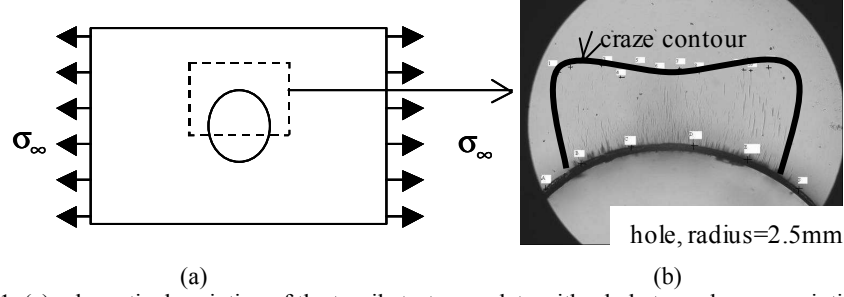


Figure 1: (a) schematic description of the tensile test on a plate with a hole to analyse craze initiation; (b) optical micrograph of the region with crazes. The contour corresponds to the local critical stress state for initiation. The craze plane is perpendicular to the direction of the major principal stress

a crack has nucleated. For an elastic material with crazing only, the energy release rate is derived from the traction-opening law in Eq. (3) as

$$G_c = \int_0^{\Delta_n^c} \sigma_n(\dot{\Delta}_n^c) d\Delta_n. \quad (4)$$

For a given loading rate, G_c is a measure of the energy dissipated during craze thickening.

3 EXPERIMENTAL CALIBRATION

The material under consideration is a commercial PMMA (Perspex) supplied in the form of plates with a thickness of 10 mm. The specimens used for the analysis of craze initiation and craze thickening are from the same plate. The molecular weight measured by GPC results in $M_n = 864 \text{ kg/mol}$ and $M_w = 1843 \text{ kg/mol}$. Döll [3] has reported an influence of the molecular weight on the development of a stable craze. For PMMA, a critical value about 300 kg/mol is evidenced and the mechanism of crazing depicted in Sec. 2 operates for molecular weight larger than this value, as considered here. The principal and secondary relaxations were characterized by dynamic mechanical spectroscopy. The glass transition identified as the principal relaxation at 1 Hz occurs at 410 K while the β relaxation appears at 300K.

Craze initiation

The experiments are similar to those presented by Sternstein and Ongchin [7] with a plate containing a circular hole subjected to a remote and constant tensile stress (see Fig. 1). The loading is applied during one minute and maintained at a constant value. The crazes initiate at the equator of the hole. The contour of the crazed region is observed to be stable for a loading time larger than thirty minutes. From the elastic solution of the stress distribution, we estimate the parameters of the criterion in Eq. (1) to be $A^0 = -31 \text{ MPa}$ and $B = 680 \text{ MPa}^2$, for an applied remote tension of 30 MPa. Therefore, a craze stress for uniaxial tension of 46 MPa is predicted, which is reasonable for PMMA.

It should be noted that the original observations of crazing for PMMA of Sternstein were performed at temperature ranging from 50°C to 70°C, well above T_β . The validity of their formulation was not checked at room temperature and our results indicate that there is sufficient molecular mobility to promote crazing. The initiation of crazing for lower temperatures is questionable since craze nucleation requires the formation of micro-voids and involves some plastic deformation, even at a very localized scale. Therefore, the use of the criterion adopted here

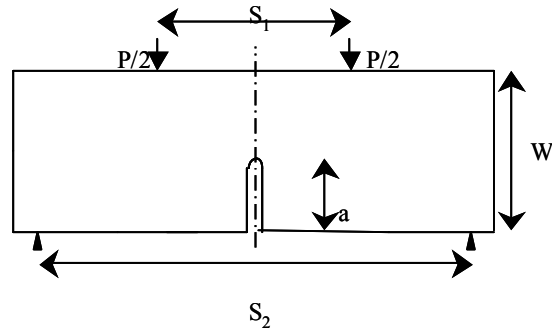


Figure 2: four points bending device to analyze mode I fracture of a PMMA specimen with a sharp crack. The dimensions are $S_1=40\text{mm}$, $S_2=90\text{mm}$, $W=20\text{mm}$, $a/W\approx 1/2$ and the thickness is 10 mm.

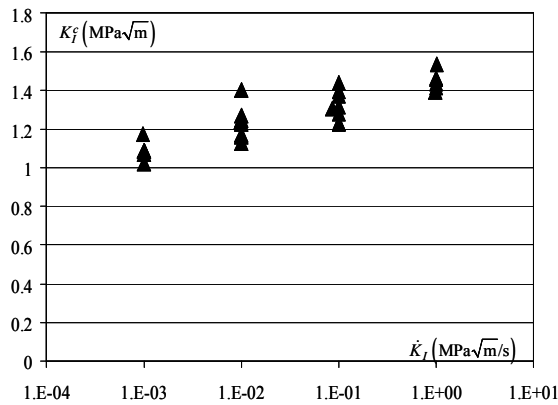


Figure 3: measurements of the critical stress intensity factor of PMMA with loading rate. The onset of crack propagation corresponds to an unstable crack advance.

at lower temperatures is questionable as the critical shear defined in Eq. (1) is expected to be related to some local plastic deformation.

Craze breakdown

The development of a single craze at the tip of a notch specimen subjected to mode I loading resembles to a Dugdale zone and this configuration has been analyzed extensively par interferometry [3, 4]. Crack propagation is assumed to occur for a critical thickness Δ_n^{cr} . This critical value is about three microns for a wide class of PMMA as reported in [3, 4]. Based on these observations, a value of three microns for Δ_n^{cr} is adopted here but we are carrying out our own interferometer to estimate this parameter for the material under investigation.

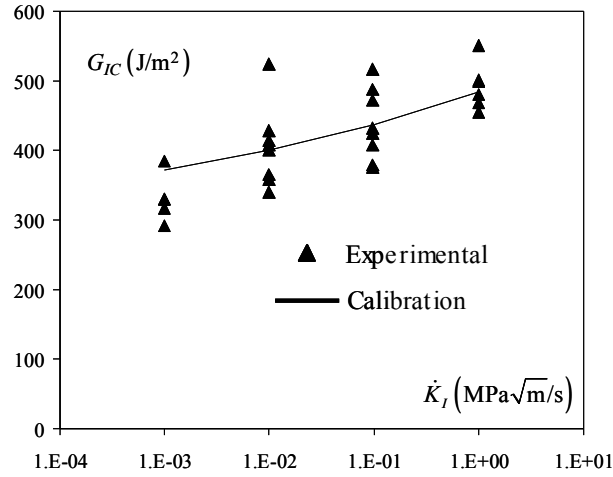


Figure 4: measurements of the energy release rate with loading rate for PMMA, under mode I loading.

Craze thickening

The calibration of the parameters $\dot{\Delta}_0$, A^c and σ^c involved in Eq. (2) are derived from fracture experiments from which the energy release rate is derived. For this purpose, a sharp crack was made by tapping with an instrumented device a razor blade at the tip of the notched specimen. This procedure is performed at room temperature and results in reproducible sharp crack, with a length at least four times larger than the radius of the pre-notch, following the requirement of the ESIS-TC4 technical committee [9]. The samples are loaded under pure bending by using the device presented in Fig. 2. In Fig. 3 is reported the evolution of the toughness in terms of stress intensity factor at failure with loading rate, represented by the rate of the stress intensity factor. In order to verify the conditions for small scale yielding and plane strain, the yield stress of the material has to be estimated. The yield stress of the bulk is measured from uniaxial compression tests conducted at various loading rates. The characteristic time scale for plasticity of the bulk without crazes can be taken as $t_0 = \sigma_y / \dot{\sigma}$ and that for failure as $t_{failure} = K_I^{cr} / \dot{K}_I$. By using $t_{failure}$ equal to t_0 , an estimate of the yield stress under uniaxial compression is derived. Following the recommendations in [9], the yield stress under tension is thought to be 70% of this value is used to account for the characteristic pressure sensitivity of glassy polymers. The dimensions of the specimens used in the fracture tests are actually sufficiently large to ensure small scale yielding and plane strain conditions. At the onset of crack propagation, unstable crack advance takes place and the related stress intensity factor corresponds to the critical toughness.

The estimate of the energy release rate is derived from $G_c = K_I^{cr} / E'$, with $E' = E / (1 - \nu^2)$ for plane strain conditions.

Prior to the viscoplastic yield stress, glassy polymers exhibit some viscoelasticity which needs to be accounted for to estimate G_c . We incorporate this effect by using the secant Young modulus at the stress level for failure. The value is extracted from four-points bending test of un-notched beams subjected to the same loading conditions as the fracture specimens, loaded up to the stress at failure.

The secant modulus evolves from $E = 3.22\text{GPa}$ at $\dot{K}_I = 1 \times 10^{-3} \text{MPa}\sqrt{\text{m/s}}$ to $E = 3.82\text{GPa}$ at $\dot{K}_I = 1 \text{MPa}\sqrt{\text{m/s}}$. By accounting for this time dependence and by assuming that the Poisson ratio remains constant, the evolution of the energy release rate is calculated and reported in Fig. 4. The increase of G_c observed with increasing loading rate in Fig. 4 indicates that the process for failure is time dependent.

The experimental data reported in Fig. 4 are then used to calibrate the parameters involved in the description of crazing presented in Sec. 2. Those of the criterion for craze initiation in Eq. (1) are derived from the analysis of crazing observed during the tension of a plate with a centered hole. With the value of the critical thickness of three microns for PMMA [3, 4], the calibration of the parameters (A^c , $\dot{\Delta}_0$ and σ^c) presented in Fig. 4 is performed as follow. The variable A^c represents the temperature dependence of craze thickening which involved some viscoplasticity in a thin layer between the bulk and the craze fibrils [5]. Its value is taken equal to that estimated for the bulk subjected to uniaxial compression tests which is 60 K/MPa. The two parameters ($\dot{\Delta}_0$, σ^c) have to be identified to fit the experimental data reported in Fig. 4. This is performed by using the FE model presented in [1,2]. The parameters of the bulk identified for PMMA are used and a cohesive zone is inserted along the symmetry plane of a pre-cracked specimen. By assuming small scale yielding, a mode I, boundary layer approach is used for the loading so that the prescribed remote displacements are those of the elastic K -fields. An initial blunted crack with radius of five microns is considered to represent the notch introduced by tapping. The use of a smaller radius did not affect the calibration.

It was shown in [1] that the onset of the first fibril breakdown at the critical thickness Δ_n^{cr} triggers unstable crack propagation and the corresponding loading level is used as the critical toughness. The related energy release rate is compared to the experimental data. Following a try-and-error procedure, the pair ($\dot{\Delta}_0$, σ^c) which better fits the experiments is found to range from 0.001 mm/s to 0.05 mm/s for $\dot{\Delta}_0$ and 110 MPa to 130 MPa for σ^c .

4. CONCLUSION

An experimental procedure to calibrate the parameters of a cohesive zone used to describe crazing of glassy polymers is presented and exemplified for PMMA. The model is able to capture the evolution of the energy release rate with loading rate for a specimen containing a sharp crack. Others experiments consisting in configurations with larger notch radii are under progress to test the validity of the present calibration.

REFERENCES

1. Estevez R, Tijssens MGA, Van der Giessen E., 2000, J Mech Phys Sol, 48:2585.
2. Tijssens MGA, Van der Giessen E., Sluys LJ, 2000, Mech. Mat, 32:19
3. Döll W, 1983, Adv Polym Sci 52-53:106
4. Döll W, Könczöl L, 1990, Adv Polym Sci 91-92:138
5. Kramer EJ, Berger LL, 1990, Adv Polym Sci 91-92:1
6. Kausch HH, 1987, Polymer Fracture, 2nd edn. Springer Berlin Heidelberg New York
7. Sternstein SS, Ongchin L, 1969, Polym Preprints 10:1117
8. Sternstein SS, Myers FA (1973) J Macromol Sci -Phys B 8:539
9. Williams JG, Pavan A, 2003, Fracture of Polymers, Composites and Adhesives II, ESIS Publications