CRACK PROPAGATION KINETICS OF POLYMERS AT QUASISTATIC AND MODERATE IMPACT LOADING

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ABSTRACT

This study gives a contribution to understand the crack toughness as resistance against the intrinsic rate of fracture mechanics parameters, which is rarely investigated at moderate impact loading conditions up to now. An adequate description of the whole fracture process of polymers as typical non-linear viscoelastic materials associated with a pronounced time- and temperature-dependent mechanical behaviour implies the application of various concepts of elastic-plastic fracture mechanics. One of the more popular is the crack resistance curve (R curve) concept that can be utilized if higher amounts of stable crack growth exist. Based on experimental investigations using this concept the following conclusions can be done:

(1) Crack resistance curves for polymers at impact loading conditions can only adequately determined using the stop block method in multiple-specimen technique.

(2) The CTOD rate is a measure of the rate sensibility of stable fracture process depending on "matrix" deformation delivers insight into micromechanics and activation mechanisms of the fracture process.

(3) The stable crack propagation can been understand as 3-phase process including crack-tip blunting/crack initiation, non-stationary stable crack growth and steady-state stable crack growth (an equilibrium state).

(4) Steady-state stable crack propagation can been described as a kinetic phenomena.

Furthermore, crack resistance curves are measured in situ at quasistatic loading conditions using video technique. The crack propagation is also characterized by crack-tip blunting/crack initiation and non-stationary stable crack growth as found at impact loading. Steady-state stable crack growth is observed for materials having limited plastic deformations compared to the size of the specimen.

1 INTRODUCTION

In contrast to static, fatigue or high-speed loading [1], the crack toughness as resistance against the intrinsic rate of fracture mechanics parameters such as in the form of the crack-tip opening displacement rate (CTOD rate) is rarely investigated under moderate impact loading conditions (i.e. in the 1 m/s range).

Polymers are materials combining clear inertia effects at high loading rates due to low sound velocities and relatively low intrinsic toughness in comparison to metals with pronounced non-linear viscoelastic behaviour especially at low loading rates or at temperatures closed to the glass or phase transition temperatures. Based on that, different aspects of time-dependent fracture phenomena of polymers can be derived which results in various concepts and parameters. Theories developed to describe crack initiation and propagation in viscoelastic materials are based on different fracture criteria, either local such as the criterion introduced by Schapery [2] or global (Christensen's criterion) [3] (for detailed information on these theories see also [4]). But, none of these theories is widely used in polymer science and engineering due to the high complexity in the general form of some of the results [5].

The idea that fracture in time-dependent materials such as polymers can be described by a kinetic theory was reactivated by Zhurkov and Bueche [6] in the middle of the 20th century. The basic principles of this theory and some experimental results are summarized by Kausch [7].

2 R-CURVE DETERMINATION FOR POLYMERS UNDER IMPACT LOADING CONDITIONS

An adequate description of the whole fracture process of polymers as typical non-linear viscoelastic materials associated with a pronounced time- and temperature-dependent mechanical behaviour, where different stages of crack kinetics such as crack formation, crack-tip blunting, stable crack initiation and propagation, and unstable crack propagation can be observed, implies the application of various concepts of elastic-plastic fracture mechanics. One of the more popular is the crack resistance curve (R curve) concept that can be utilized if higher amounts of stable crack growth exist. These R curves as functions of loading parameters (such as J integral and crack-tip opening displacement) versus stable crack growth Δa can be measured at impact loading conditions (instrumented Charpy impact test) by the use of different experimental procedures [8].

On example of TPU/ABS blends made from a polyurethane based thermoplastic elastomer (TPU) and an acrylonitrile-butadiene-styrene copolymer (ABS) and other polymers [8] it has been shown that only by use of the stop block method in multiple-specimen technique it is possible to determine R curves those correspond to homogenous kinetics of crack propagation, i.e. the crack-tip opening displacement rate $\dot{\delta}$ defined as $d\delta/dt$ is constant excepting small Δa values.

The stop block technique leads to lower crack initiation and propagation values (Fig. 1a). But, compared to the stop block technique, $\dot{\delta}$ values determined by using the low-blow method increase with increasing Δa-values (i.e. with increasing pendulum hammer speed v_H) (Fig. 1b), because the ratio $\dot{\delta}/v_H$ is material-specific as noted below. The increasing crack-tip blunting area observed by using the (quasi) single-specimen technique leads to a decreasing crack-tip opening displacement rate $\dot{\delta}$ (Fig. 1d), that is a measure for the local strain rate ahead the crack tip. Comparing to the multiple-specimen technique, this behaviour is combined with higher energy dissipation as a function of Δa (Fig. 1c).
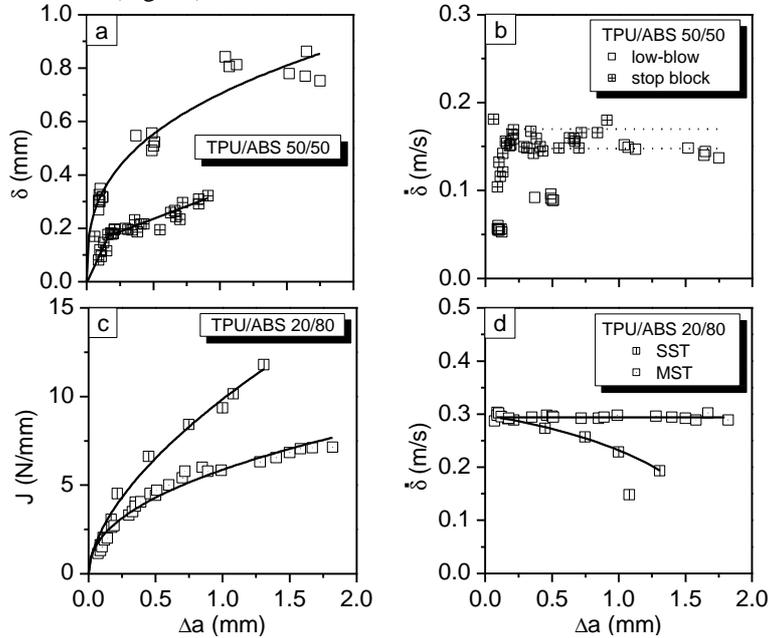
Figure 1: δ-R (a) and J-R curves (c) measured using various methods (SST − single-specimen technique, MST − multiple-specimen technique) corresponding to crack-tip opening displacement rate for TPU/ABS blends (b,d).
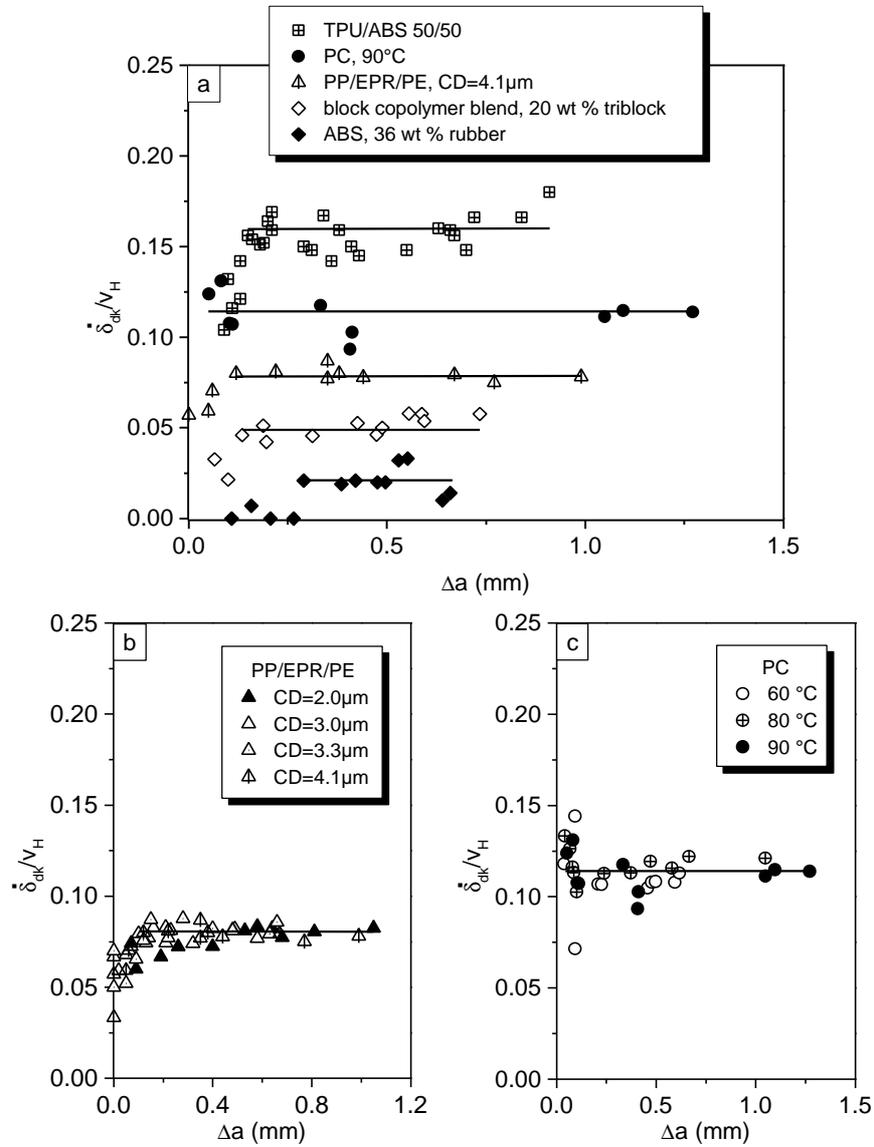
Figure 7: Normalized CTOD rate for different polymer materials (a), and as a function of particle centre-to-centre distance (CD) for PP/EPR/PE (b) and temperature for PC (c).

3 CRACK PROPAGATION KINETICS OF POLYMERS UNDER IMPACT LOADING CONDITIONS

For most polymers investigated in this study such as PC (T > 40 °C), PP/EPR/PE, PP, PP/glass-fibre composites, PE-HD, ABS, TPU/ABS blends and SB block copolymer blends a 3-phase be-haviour could been found experimentally where a threshold value $\dot{\delta}_{lim}$ was observed after rapid convergence of the CTOD rate $\dot{\delta}_{dk}$ as a function of Δa (or time): steady-state stable crack propa-gation behaviour (phase III) after crack-tip blunting/crack initiation (phase I) and non-stationary

stable crack propagation (phase II) [9]. In principle, it has been shown that the normalized steady-state CTOD rate $\dot{\delta}_{lim} / v_H$ (v_H – pendulum hammer speed) yields typical values (Fig 1a) which are a function of the matrix or major phase for a given material group such as materials with a PP matrix (Fig. 1b), but also PE-HD and ABS, and independent on the temperature in a well-defined range of temperature (PC: Fig 1c).

The cause that the maximum CTOD of PP materials rate lies in the range of that of pure PP (80 mm/s at $v_H = 1$ m/s) is that the CTOD is a measure of the deformation capacity close to the crack tip. Consequently, the CTOD rate is only related to deformations, which occur in the matrix or the major phase but not in the particles or the minor phase, or at interfaces, respectively.

4 STABLE CRACK PROPAGATION AS A KINETIC PHENOMENON

Whereas the crack speed indicates the compliance of the whole mechanical system and, consequently, it is dependent on the brittleness of cracked specimens (shape and size of specimens, materials), the internal stiffness of the testing devise and the type of loading (strain-rate or stress-rate controlled loading etc., bending or tensile loading etc.), the CTOD rate is also related to "matrix" deformation processes because the CTOD is a measure of the deformation of the material close to the crack tip. Therefore, the threshold of the CTOD rate as a measure of the rate sensitivity of the stable fracture processes depends on the type of plastic deformation and delivers insight into micromechanics and activation mechanisms of the fracture process.

As a result, a relationship between the threshold values $\dot{\delta}_{lim} / v_H$ and the activation energy of deformation ΔH^* associated with the crack propagation process was found (Fig. 3a).

As demonstrated for polymers such as PC, PE and PTFE [10], non-linear–viscoelastic deformation processes including also fracture are characterized by the same activation energies ΔH^* as deformation in the linear response range despite different deformation amplitudes. The latter is probably caused by comparable molecular motions underlying all macroscopic deformations (fluctuation-dissipation theorem). Another clear evidence for universal relations between fracture and small-strain behaviour is that stress intensity factor-versus-crack speed data as a function of temperature or test speed could be brought into superposition (mastering) by using Arrhenius-type shift factors derived from small-strain deformation measurements [4,5,11].

Therefore, results from dynamic mechanical analysis (DMA) may also be used in this study for determining ΔH^*. The "material" parameter ΔH^* is also in fact a function of temperature T, $\Delta H^* = \Delta H^*(T)$, and speed (or frequency) but nearly constant for well-defined material-dependent "temperature-speed" windows.

This relation in Fig. 3a can be nearly characterized assuming that the process of stable crack propagation can be thermodynamically described as a kinetic phenomenon on the basis of a two-potential model with ΔH as generalized activation energy. The resulting proportionality between the CTOD rate and $\exp(-\Delta H/RT)$ (R – universal gas constant) can be rewritten in the normalized form as

$$\dot{\delta}_{lim} / v_H = \exp\left(-\frac{\Delta H^*(T) - \Delta H_0^*}{\Delta H_0^*}\right) = \exp\left(1 - \frac{\Delta H^*(T)}{\Delta H_0^*}\right) \tag{1}$$

$$\dot{\delta}_{lim} / v_? \approx \frac{\Delta H^*(T)}{\Delta H_0^*} . \tag{2}$$

ΔH_0^*, the non-thermal part, is the activation energy for an elementary process depending little on the materials, which is exemplary combined with highly localized motions of side groups etc., but also those where crack propagation instability occurs in the $\dot{\delta}_{lim}$ -ΔH^* diagram. Such instability can be observed for materials those are intrinsically brittle such as PS or at very low temperatures near zero degree in general. Therefore, values of ΔH_0^* determined by using equation (2) and by linear approximation of log(ΔH^*)-temperature plots from DMA to zero degree [12] (Fig. 3b) are in the same order (~10 kJ/mol) due to minimum crack propagation stability at very low temperatures.
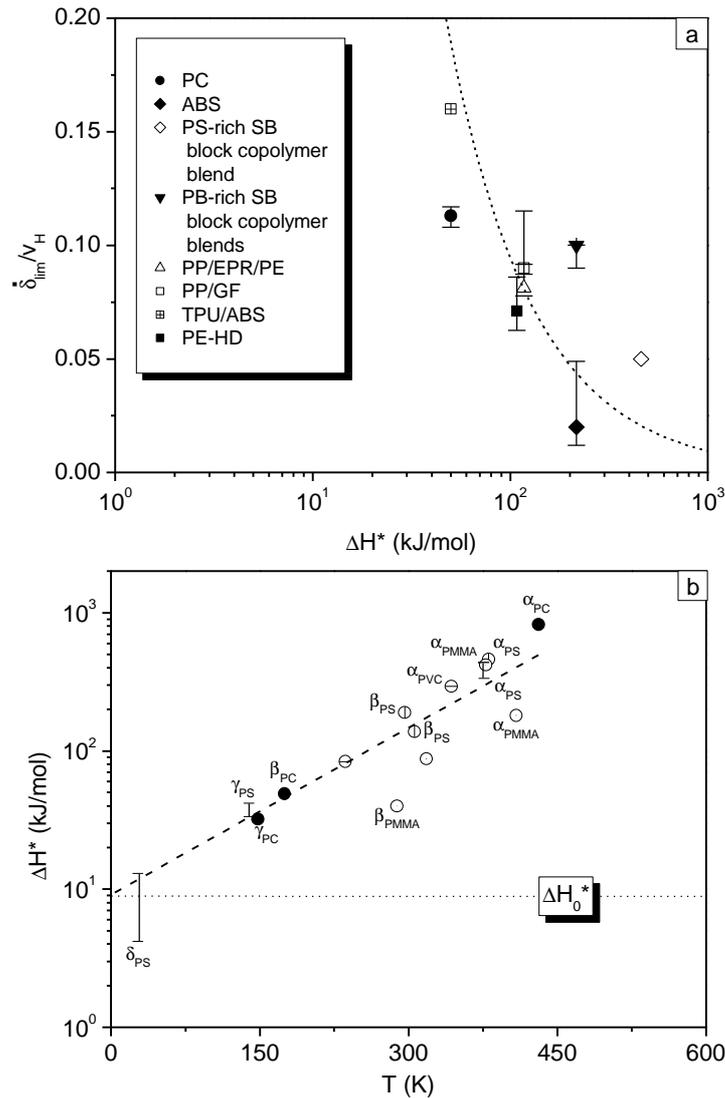
Figure 3: Normalized steady-state CTOD rate as a function of ΔH^* for different polymer materials; the dashed line is plotted using equation (2) with $\Delta H_0^* \sim 10$ kJ/mol (a); determination of the activation energy ΔH_0^* for an elementary process using DMA (frequency = 1 Hz), α – dynamic glass transition, β – secondary relaxation process, γ, δ – local modes, own data and data from [12] (b).

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