THE EFFECT OF SPECIMEN THICKNESS AND MORPHOLOGY ON FRACTURE TOUGHNESS OF THERMOPLASTIC POLYMERS

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ABSTRACT

Fracture mechanical experiments were conducted using different types of polymers with glassy (PS), coarse spherulitic (PP), and fine spherulitic (PE) morphology at 20°C and constant strain rate. The increase (PE) or decrease (PP, PS) of fracture toughness towards very small specimen diametre was explained using microscopic observations of the fracture behaviour. Results on anisotropic fracture behaviour of stretched foils were interpreted in a similar manner.

KEYWORDS

Thermoplastic polymers, morphology, stretching, specimen thickness, limits of fracture machanics, microscopy of plastic zone, craze distribution, inter-spherulitic fracture.

INTRODUCTION

Thermoplastic polymers are often used as thin films i.e. as sheets with a thickness << 1mm. Crack propagation in such films is an important problem. It is however well known that the applicability of fracture mechanics is limited for small specimen thicknesses. This is even more to be expected for materials with a large ability for plastic deformation and work hardening- as thermoplastic polymers in an intermediate range of temperature and strain rate.

The critical stress intensity K_c usually increases from K_{Ic} with decreasing thickness as a consequence of the transition from plane strain to three-dimensional strain. There is however an uncertainty about the continuation of this behaviour at very small thicknesses (Schwalbe, 1975). It is the purpose of this work to explore this situation for PS (glassy), PE and PP (both semi-crystalline) at ambient temperature. Besides thickness, the effect of morphology and stretching is considered in these investigations (fig. 1).

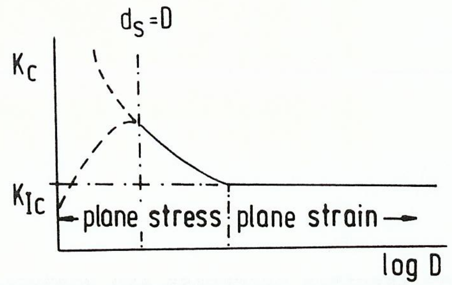

Fig. 1. Variation of critical stress intensity K_c with specimen thickness. Anomalous decrease for spherulitic diametres $d_s > D$, schematic.

MATERIALS AND EXPERIMENTAL CONDITIONS

Materials and experimental conditions are indicated in tables 1 and 2.

TABLE 1

	glass transition point T_g (°C)	yield stress N/mm^2	E-modulus N/mm^2	elongation % at fracture
PS	+ 100	30-60	3500	3-4
PE	- 110	10-25	2000-15000	200-1200
PP	- 18	25-35	11000-15000	700-1200

TABLE 2 Specimen dimensions and testing conditions

sample	width B/mm	initial crack length/mm	strain rate mm/min	test-temperature °C
single edge notched	40	10	0,1	20

Exploratory experiments show a strong dependence of critical K-values on specimen dimensions at ambient temperature. Strain rates in the range of 0,05 - 2,5 mm/min, as well as single edge or central notched specimen did not show a significant difference in the experimental results. In this work these conditions were left constant (table 2) and emphasis was put on the differences due to the molecular structure, morphology and specimen thickness of the polymer. For determination of K-values in all cases position of the crack tip and not the dimension of any plastic zone was used. The mechanical tests were supplemented by investigations of the molecular arrangements in the plastic zone by transmission-,light-, and electron microscopy as well as by X-ray diffraction.

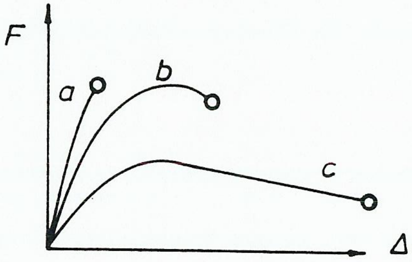

Fig. 2. Typical force-extension-diagrammes
a) cast PS-films, coarse-spherulitic PP
b) biaxially stretched PS
c) fine-spherulitic PE, PP.

In figure 2 several typical force-deformation curves of precracked films are shown. It is evident that it is difficult to obtain comparable critical stresses, if continuous straining and work hardening occurs. In all cases the maximum force was used to obtain K_c or K_{Ic}. The experimental results from plates and sheets of the three polymers in three morphological conditions indicate that only fine-spherulitic PE behaves normal, while coarse-spherulitic PP and sheets produced from a 10%-solution of PS in xylene show an additional decrease of fracture toughness for small thickness D. For such small dimensions it was impossible to obtain significant values with PE, because general yielding ($\sigma > R_{p\ 0,2}$) occured before the crack became critical (fig. 3).

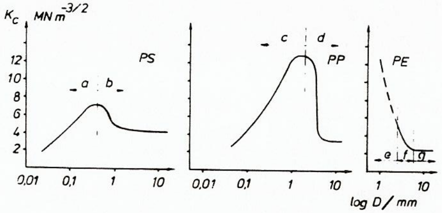

Fig. 3. Measurement of K_c in three polymers (see table 1 and 2), structure of the plastic zone:
a) single craze crack path
b) multiple craze
c) inter-spherulitic fracture
d) additional transspherulitic crack path
e) general yielding
f) supercraze
g) multiple craze

The behaviour of the materials could be modified further by mono-, or biaxial stretching (fig. 4). For primary cracks oriented perpendicular to the direction of stretching the resistance to crack growth increased as a function of molecular rearrangements, while a transition to a quasi-brittle behaviour occurs after high amounts of stretching and a precrack oriented parallel to the molecular alignement.

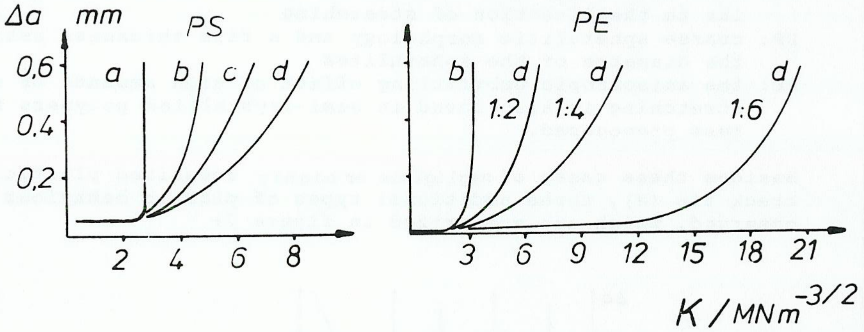

Fig. 4. The effect stretching on subcritical static crack growth
- a) uniaxial, parallel 1 : 2,5
- b) unstretched
- c) biaxial 2,5 : 2,5
- d) uniaxial, perpendicular 1 : 2,5

In order to distinguish between critical und subcritical crack growth, it is useful to plot the measurements as functions $\Delta a = f(K)$ (fig.4), or $da/dK = f(K)$ (see fig. 7).

In figure 5 typical results of the microscopic investigations of the mechanisms of seperation at the crack tip are shown.

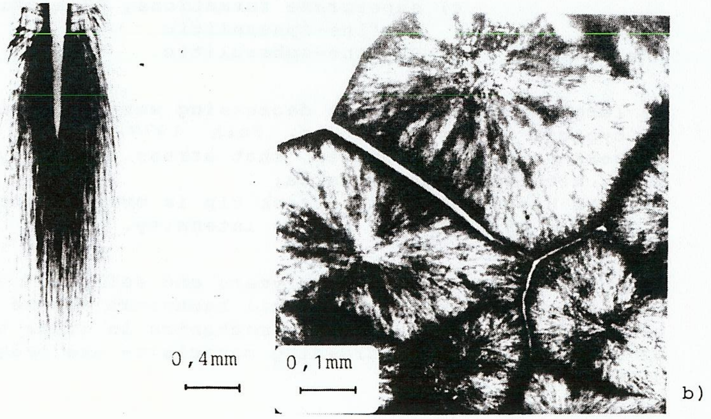

Fig. 5. a) PS, D = 1mm, multiple crazing
b) PP, $D < d_s$, interspherulitic crack path

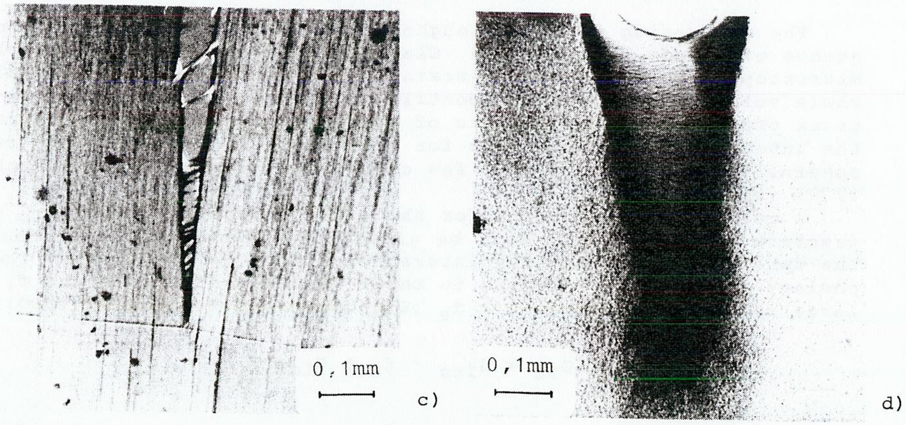

Fig. 5. c) PE, stretched 1:6, cleavage along aligned molecules
d) PE, unstretched, super-craze.

CONCLUSIONS

Two features of these experimental results shall be discussed:

a) The anomalously low fracture toughness of thin films with certain morphological conditions,

b) The limits of applicability of classical fracture mechanics.

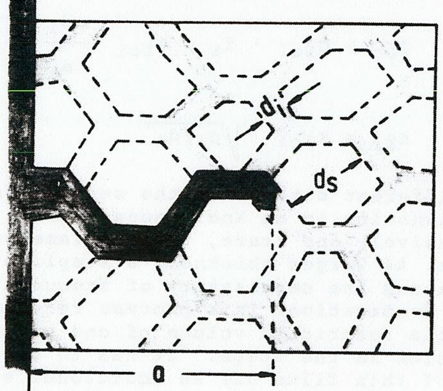

Fig. 6. Crack propagation restricted to the interspherulitic portions of the structure, schematic.

The decreasing fracture toughness of PP- and PS-films is a consequence of strain localisation. Classical fracture mechanics implies microscopically homogeneous strain at the crack tip (Kausch, 1978). The whole volume is supposed to contribute to dissipation of energy during crack propagation. The process of separation is however restricted to the interspherulitic portions for thin films of PP containing coarse spherulites, and to one or a few crazes for glassy PS (Friedrich, 1979) (fig.6).

A quantitative relation for the crack extension energy G_{Ic} or the fracture toughness K_{Ic}, will be given for the example of PP. Both the spherulitic (s) and the interspherulitic (i) portions of the morphology contribute according to their volume portions f_s and f_i for large specimen diametres $D > d_s$ (Hornbogen and Friedrich, 1980):

$$G_{Ic} = G_{Ics} \cdot f_s + G_{Ici} \cdot f_i \qquad (1)$$

If the thickness D decreases below the diametre of the spherulites d_s, separation is observed to take place exclusively in the interspherulitic zone with a thickness of d_i. The volume portion f_i is approximately given by

$$f_i = \frac{d_i}{d_s + d_i} \approx \frac{d_i}{d_s} \qquad (2)$$

The first term of the equation 1 becomes zero, if energy is only dissipated in the small volume portion of interspherulitic matter (glassy, impure, small molecular weight). This provides for conditions under which separation takes place at a smaller stress (but usually not at a smaller local strain) than inside the spherulites:

$$G_{Ic} = G_{Ici} \cdot f_i = G_{Ici} \frac{d_i}{d_s} \qquad (3a)$$

$$K_{Ic} = K_{Ici} \sqrt{d_i/d_s} \qquad (3b)$$

In the glassy PS different details of the separation mechanism can be used for an explanation in an analogeous manner. In thin films the crack follows exclusively one craze, if the diametre of the sheet is less than $D < 0{,}1$mm. At larger thickness a steplike fracture surface is observed, indicating the necessarity of frequent changes of the crack plane during propagation. This process requires additional energy, i.e. not only the restricted volume of one craze dissipates energy by plastic deformation in the steps. It has to be mentioned that surface roughness of thin films has an additional effect in lowering fracture toughness. It is significant for coarse spherulitic material as well as for as-cast amorphous films.

Stretching prior to the fracture experiment changes this behaviour of PS. If the crack propagates in direction of the alignements of the molecules, fracture toughness is further reduced, because splitting occurs between the weakly bonded molecules. Perpendicular to this direction, however, fracture stress increases. The most favourable conditions were obtained with biaxially stretched sheets. The fracture stress was particularly high, because the molecules were plastically re-oriented into the direction of the applied stress in a so-called "supercraze".

Which are the conditions under which fracture mechanics can be used in sheets for the three materials at ambient temperatures?

PS: cast films, but not in stretched sheets after loading perpendicular to the direction of stretching
PP: coarse spherulitic morphology and a film thickness smaller than the diametre of the spherulites
PE: the anisotropic embrittling effect of high amounts of uniaxial stretching is also found in semi-crystalline polymers but it is less pronounced.

Besides these cases of negligible or highly localized plasticity at the crack tip (a), three additional types of plastic behaviour have been observed, which are summarized in figure 7:

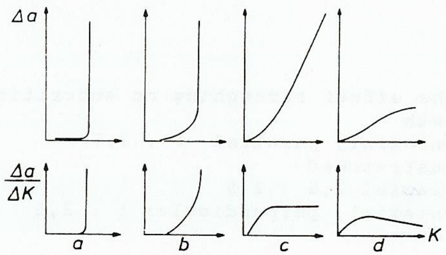

Fig. 7. basic possibilities for crack growth in precracked polymer films
a) PS, as-cast films, PP coarse-spherulitic
b) PS, plates, multiple crazing
c) supercraze formations, PS biaxially stretched, PE fine-spherulitic
d) PP fine-spherulitic.

b) large deformation, but decreasing work hardening coefficient (also typical for tough metals, Shih 1979);
c) work hardening provides, that stress intensity K increases linearly with crack progress Δa;
d) work hardening at the crack tip is overcompensating the contribution of Δa to the stress intensity.

It is characteristic for polymers and still more so for thin films that these particular mechanical behaviours can be observed. Attempts to expand classical fracture mechanics in order to cover the behaviour of such highly work hardening materials- are presently under consideration.

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