Influence of the Geometry on the Essential Work of Fracture of Polypropylene Materials

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Keywords: Essential work of fracture, geometrical requirements, polypropylene materials, structure-property relationships.

Abstract. For reasonable application of the essential-work-of-fracture (EWF) concept to polymers some geometrical requirements has to be fulfilled, whereas these conditions, especially that of plane state of stress and self-similarity of the load–displacement diagrams are often handled very non-critical in the literature. A brief discussion of minimum and maximum valid ligament length and the influence of specimen thickness on toughness has been given, therefore, by comparing data empirically determined with the predictions. Furthermore, the applications of small-sized specimens has been shown on example of ethylen-propylene copolymer.

Introduction

Based on fundamental studies of Broberg [1] as well Cotterell and Reddel [2], the method of essential work of fracture (EWF) as one of the main concepts of the 'Post-Yield' Fracture Mechanics has been first applied to polymers by Mai and Cotterell [3]. At present, the EWF methodology is widely used for highly ductile materials, mostly polymers but also other materials such as metals, paper or ductile ceramics, which are prepared in form of films or thin plates. The continuous success of the method, also manifested in a standard draft of the European Structural Integrity Society (ESIS) [4], is caused by its relative simple experimental preconditions compared to other approaches in fracture mechanics as well as the low consumption of materials and time. Recently, well-founded correlations between molecular and fracture mechanics parameters (EWF) have been detected by Chen and Wu [5] for polymers, likewise by Halary et al. [6] as well as Lach and Grellmann [7] in the case of Linear-Elastic and Elastic–Plastic Fracture Mechanics, which can provide the basis of understanding the underlying physics of polymer toughening.

The EWF concept is based on the assumption that the total work of fracture W can be divided into the component W_e, scaling with the ligament area B·l (B – specimen thickness, l – length of unnotched ligament), dissipated in the inner or fracture process zone, and the component W_p, scaling with the volume $B \cdot l^2$, dissipated in the outer or plastic zone:

$$W = W_e + W_p = EWF \cdot Bl + \beta w_p \cdot Bl^2 . \quad (1)$$

After divided W by the ligament area, the specific work of fracture w is obtained:

$$w = EWF + \beta w_p \cdot l , \quad (2)$$

where w_p is the non-essential work of fracture and β is the shape factor of the plastic zone. EWF was found to be independent on specimen configuration such as single or double edge notched tensile specimens (SENT or DENT specimens) etc. for given thickness, whereas βw_p is a function of the plastic constraint. EWF has the meaning of a 'crack-moving force' comparable to that of the

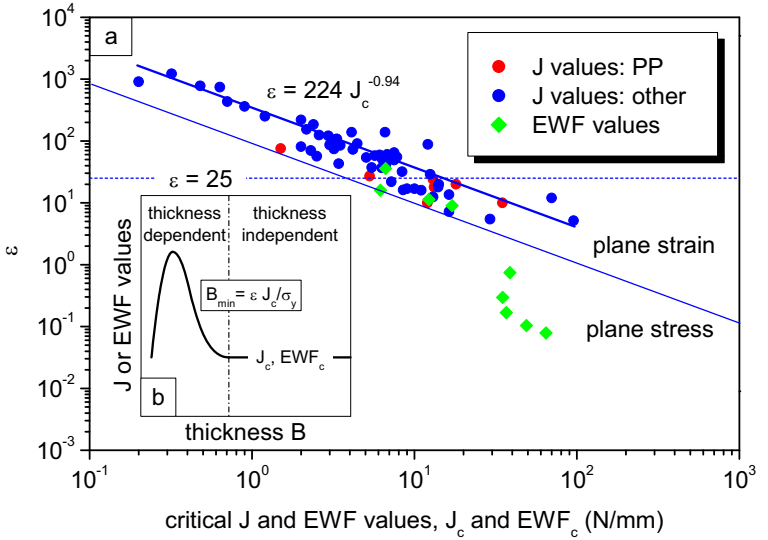

Fig. 1. (a) Placement of critical EWF, EWF_c, independent on specimen thickness in the geometry factor–toughness diagram, $\varepsilon = f(J_c)$ [9] (EWF_c–ε data from analyzing results of the literature [11], J_c–ε data from [10]). (b) Generalized thickness dependence of fracture mechanics parameters.

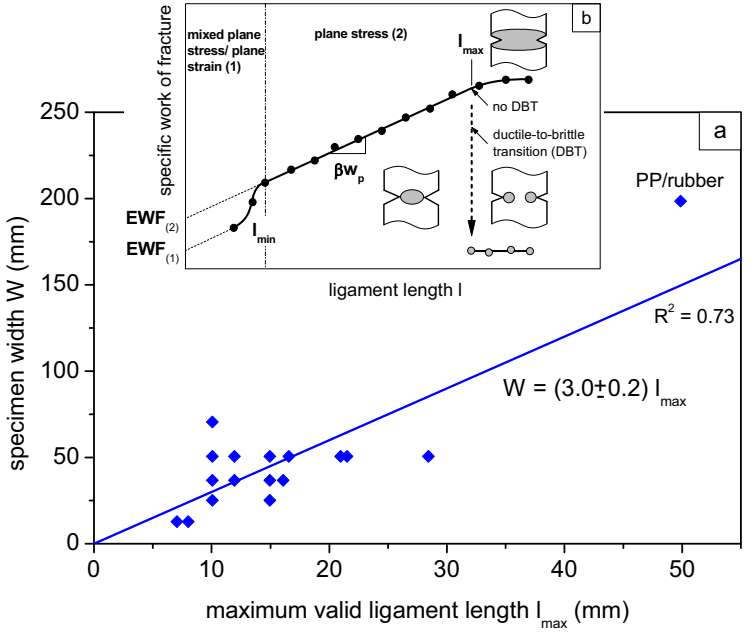

Fig. 2. (a) Comparison of the maximum ligament length l_{max} depending on specimen width W [9] (data extracted from the literature [3,12]). (b) Scheme to define minimum and maximum ligament length, l_{min} and l_{max} (the gray areas in the inserts represent the formation of the plastic zone; EWF – essential work of fracture, w_p – non-essential work of fracture, β – shape factor of the plastic zone).

physical crack initiation values J_{Ic} if the J-integral concept is valid, where EWF ~ J_{Ic}, and βw_p is a measure of the resistance against stable crack propagation such as the slope dJ/da of the crack resistance curves, where $\beta w_p \sim$ ¼·dJ/da for DENT specimens. These relations were experimentally, numerically and theoretically validated by different authors [3,8].

For application of the EWF concept a couple of geometrical conditions, such as (i) crack initiation after plastification of the ligament, (ii) plane state of stress and (iii) self-similarity of the load–displacement diagrams, must be fulfilled which often are handled very non-critically also in the standard draft of ESIS. So they have to be shortly discussed below.

Discussion of the Geometrical Conditions of the EWF Concept

Plane State of Stress. Eq. 1 and 2 are fully valid only if predominant plane state of stress is applied, although the EWF is also defined for plane state of strain or mixed plane stress/plane strain state. (compare Fig. 2b). Because the plane strain state only exists for small ligament-to-thickness ratios, i.e. for $l/B \leq 1$, for l/B ratios much higher than one, say 3–5 as generally used in the literature, one can feel certain to measure at pure plane stress state. However, the l/B ratios related to the plane strain/plane stress transition, where $l = l_{min}$ (see Fig. 2b), have been experimentally found to vary from 1.3 to 56 [9], so that the use of l/B = 3–5 leads to a pronounced over- or underestimation on notched depth. Only for films or thin plates thinner than 2 mm, the minimum valid ligament length has empirically determined always to l > 3–5B.

The EWF values have often been observed to be independent of the specimen thickness for thickness values higher than a critical one ($B \geq B_{min}$) [10]. Just as in the geometry criterion of the J-integral concept, a factor ε can be determined by

$$\varepsilon = B_{min} \frac{\sigma_y}{EWF_c}, \tag{3}$$

where EWF_c is the critical EWF value for $B \geq B_{min}$ and σ_y is the yield stress (Fig. 1b). Because the EWF_c values are generally comparable to the critical J values J_c, both sets of data, $\varepsilon = f(J_c)$ [11] and $\varepsilon = f(EWF_c)$, are plotted in the same diagram (Fig. 1a) [9]. From Fig. 1a it can be drawn that the independence of EWF values on thickness is not a sufficient criterion for plane strain state. Only data measured using compact specimens with B ≥ 3 mm scatter close to the general relationship

$$\varepsilon(J_c) = 224 \cdot J_c^{-0.94}, \tag{4}$$

i.e. in ranges where the pure plane strain state is valid (the relationship $\varepsilon(J_c) = 25$ often used to split ranges of plane strain and plane stress does not work for polymers, see Fig. 1a [11]). All other data are lying clearly below the scatter band of $\varepsilon(J_c)$ indicating a pure plane stress state typical found for thinner films (B ≤ 1 mm). The latter has been also confirmed by the independence of flow stress (i.e. the net section stress σ_n, $\sigma_n = m \cdot \sigma_y$, where m is the plastic constraint factor with m = 1.15 for DENT specimens and plane stress state) on ligament length. The net section stress has to check at any rate, therefore, by the method of Clutton [4], for example.

Self-similarity of the Load–displacement Diagrams. The self-similarity of the load–displacement diagrams fulfilled by both the maximum load and the maximum displacement have to be proportional to the ligament length is limited by a maximum ligament length l_{max} (see Fig. 2b) through interaction of the plastic zone with the specimen boundary or by a ductile-to-brittle transition (DBT) [3,12]. The DBT is characterized by a pronounced drop in ligament-length dependent specific work of fracture as found for polypropylene (PP) random copolymer [13].

In the case that the maximum ligament length cannot be determined experimentally, l_{max} may be estimated by $l_{max} = 2r_p$ or $l_{max} = W/3$ (r_p – size of the plastic zone, W – specimen width). However, a

critical evaluation of the experimental data clearly demonstrates that a correlation between r_p (determined using Dugdale's formula, for example) und l_{max} does not exist, i.e. the l_{max}/r_p ratio scatters from 0.5 to 4.6 [9]. In comparison to that the relation $l_{max} = W/3$ has been empirically confirmed despite high scatter of the data (Fig. 2a).

Motivation of Fracture Mechanics Investigations of PP Materials using the EWF Approach

Notwithstanding about 60 papers in scientific journals dealing with the application of the essential-work-of-fracture approach on PP materials were published up to 2008, a very few investigations using EWF parameters as a function of crystalline structure [14], the molecular weight [15] and the specimen thickness [13,16] are available for PP material. But investigations dealing with a combination of such influencing factors i.e. parameters of the molecular architecture (tacticity, molecular weight), the morphology (degree of crystallinity, crystalline phases, size of spherulites) and the specimen geometry (thickness, width) as in the present study opens a new field.

Due to confinement of the formation of spherulitic structure and possible nucleation effects in thin films the morphology of semicrystalline polymers such as PP is never independent of the size of the semi-finished products (films and plates) used for preparation of the specimens also in cases of identical thermal treatment and the same molecular architecture. Therefore to separate the influence of specimens size from that of the structure, only one parameter has to be varied where all other ones have to keep constant. In the first stage, PP homopolymers are used for fracture mechanics investigations varying the molecular architecture, the morphology as well as the size of the specimen:

- Variation of the film thickness B (typically from several ten micrometer up to about 1 mm) of DENT specimens having width; application of small-sized specimens to show the limits of miniaturisation.
- Variation of the crystalline structure; non-nucleated and beta- and alpha-nucleated isotactic PP homopolymer (standard molecular weight): The alpha-PP/beta-PP ratio are varied by controlled thermal treatment (monoclinic alpha-phase, hexagonal beta-phase, mixed alpha/beta-phase). The smectic phase are produced by quenching. Additionally, the pure smectic phase are used as a basic material for annealing using different temperatures to obtain alpha-PP with less ordered crystallites. The size of spherulites and the degree of crystallinity have to kept approximately constant.
- Variation of the tacticity of standard-molecular-weight PP homopolymer (from isotactic PP to syndiotactic PP).
- Variation of the molecular weight of PP homopolymer including PP type with standard molecular weight and PP types with molecular weights lower and higher than that of the standard PP.

Furthermore, PP copolymers and PP/clay nanocomposites are also involved into the research program.

Application of Specimens with Reduced Width

In the first stages of the synthesis of new polymers, for instances, only very limited amount of material (sub-gram) is often available less than necessary to enable the determination of fracture mechanics parameters using bulk samples or EWF specimens having a usual width W of 20–200 mm. Furthermore, sometimes the applications of EWF approach is advantageous to characterize the crack propagation behaviour also at plane strain conditions. In both cases the use of small-sized samples may be favoured.

As shown in Fig. 3 taking the crack propagation in PP copolymer films having a thickness of about 0.17 mm as an example, both the values of EWF and βw_p has found to be the same for

samples with W = 12 mm and a gauge length Z = 20 mm compared to that with W = 30 mm and Z = 40 mm.

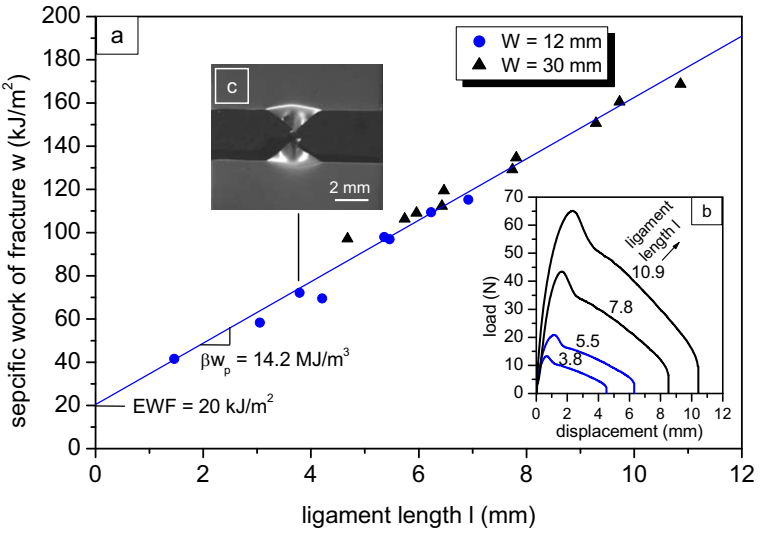

Fig. 3. Specific work of fracture (a) and load–displacement diagrams (b) as a function of the ligament length l and the specimens width W (blue: W = 12 mm, black: W = 30 mm) for PP copolymer. (c) Formation of the plastic zone and stable crack growth in small-sized specimens (l = 3.8 mm).

Summary

The EWF values have often been observed to be independent of the specimen thickness for thickness values higher than a critical one. Just as in the geometry criterion of the J-integral concept, a factor ε has been determined which shoes that the independence of EWF values on thickness is not a sufficient criterion for plane strain state. Therefore, the constancy of the net section stress as a sufficient criterion for plane stress has to be checked at any rate.

The rather accepted guidelines often given in the literature, to determine the minimum and maximum valid ligament length, does either not work or only with some reservations. So, the minimum valid ligament length has empirically determined always to be higher than 3–5 times of the thickness only for samples thinner than 2 mm. Furthermore, of all predictions used to estimate the maximum valid ligament length, the only one which has been empirically confirmed is that, where the ligament length has to be smaller than one-third of the specimens width.

Finally, small-sized samples with reduced width can be used if limited amount of material is available.

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