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# Fracture behaviour of polypropylene films at different temperatures: assessment of the EWF parameters

D. Ferrer-Balas<sup>a,\*</sup>, M.Ll. Maspoch<sup>a</sup>, A.B. Martinez<sup>b</sup>, E. Ching<sup>c</sup>, R.K.Y. Li<sup>c</sup>, Y.-W. Mai<sup>d</sup>

<sup>a</sup>Departament Ciència dels Materials i Enginyeria Metallúrgica, Universitat Politècnica de Catalunya, Av. Diagonal 647, 08028 Barcelona, Spain <sup>b</sup>Centre Català del Plàstic, C/Colom 114, 08722 Terrassa, Spain

<sup>c</sup>Department of Physics and Materials Science, The City University of Hong-Kong, Tat Chee Avenue, Kowloon, Hong Kong, People's Republic of China <sup>d</sup>Centre for Advanced Materials Technology, The University of Sydney, Sydney NSW 2006, Australia

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## Abstract

The fracture of films of one polypropylene (PP) homopolymer and three ethylene–propylene (EPBC) block copolymers with different ethylene content (EC) at temperatures around their glass transition ( $T_g$ ) is investigated by means of the essential work of fracture (EWF) Method. The fracture behaviour is studied at low speed (2 mm/min) for the different materials, in the range from -40 to +70°C. The materials are tested in the form of extruded films of 90 µm thickness, in a DENT geometry, after being annealed for 1 h at 120°C. The homopolymer is found to be much more temperature-sensitive than the EPBC with lower EWF values at  $T < T_g$  and an opposite trend at  $T > T_g$ . The ductile–brittle transition of the homopolymer that occurs below its  $T_g$  is suppressed for the EPBC in the temperature range studied. The variations of the EWF fracture parameters with T are consistent with those of a previous study in LDPE, and are explained in terms of molecular relaxation, ethylene phase content, and changes in the yield and fracture stresses. Some limitations of the EWF method are found with the more ductile materials, which show the necking phenomenon with DENT specimens at the higher temperature studied. © 2000 Elsevier Science Ltd. All rights reserved.

Keywords: Essential fracture work; Ethylene-propylene block copolymers; Stress-whitening

#### 1. Introduction

Polypropylene (PP) is one of the most used plastics at present in a very wide range of applications [1]. Apart from moulded pieces and fibres, a high percentage of its use is in the form of films and sheets, and it is of most importance to find a reliable method to measure their toughness properties. As polypropylene is brittle at temperatures below its glass transition (from 0 to 20°C) or at impact speeds, ethylene-propylene block copolymers (EPBC) or impact copolymers are used in some applications where an increase in toughness is required. However, a previous study [2] on the fracture behaviour of films by using the essential work of fracture (EWF) concept showed that at room temperature (RT) PP is tougher than EPBC. There is hence a need to understand the fracture toughness behaviour of these toughened systems tested under low molecular relaxation conditions (i.e. low temperatures and/or high strain rates). It is the purpose of this work to contribute to

this understanding by performing and analysing the EWF tests at different temperatures around  $T_g$  (and in the service temperature range) with the selected materials.

# 2. Theory

Recent studies have shown that the EWF procedure can be a very useful tool for studying the fracture properties of thin films and sheets of ductile materials [3-9]. The EWF concept was developed initially by Cotterell and Reddel [10] on the basis of Broberg's idea [11], who suggested that the total work of fracture  $(W_f)$  dissipated in a precracked body could be divided into the work consumed in two distinct zones, the inner and the outer regions. This method of work partitioning gives rise to the essential work of fracture  $(W_e)$  and the non-essential (or plastic) work of fracture  $(W_p)$ , respectively. The former corresponds to the work dissipated in the fracture process zone (FPZ) which is a material property for a given sheet thickness; and the latter to the yielding work in the outer surrounding region or outer plastic zone (OPZ) which depends on the geometry of the specimen tested. Thus, the following

<sup>\*</sup> Corresponding author. Tel./fax: +34-93-401-6706.

E-mail address: dferrer@cmem.upc.es (D. Ferrer-Balas).

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relation can be written:

$$W_{\rm f} = W_{\rm e} + W_{\rm p} = w_{\rm e}lt + w_{\rm p}\beta l^2t \tag{1}$$

where  $w_e$  is the specific essential work of fracture (per unit ligament area),  $w_p$  specific non-essential work of fracture (per unit volume), l the ligament length, t the specimen thickness and  $\beta$  the plastic zone shape factor. The specific work of fracture, is then:

$$w_{\rm f} = W_{\rm f}/lt = w_{\rm e} + \beta w_{\rm p}l \tag{2}$$

According to this equation, the plot of  $w_f$  as a function of l should be a linear relation, whose intercept with the *Y*-axis and slope would give  $w_e$  and  $\beta w_p$ , respectively. Thus, the EWF method consists of testing specimens with different ligament lengths, registering  $W_f$  for each (area under the force–displacement curve), plotting the  $w_f-l$  diagram and calculating the best-fit regression line. More details about the restrictions on the ligament length and its discussion, according to the European Structural Integrity Society (ESIS) EWF protocol [12] are given elsewhere [9] (and in the references therein).

It has already been demonstrated theoretically that  $w_e$  is equivalent to  $J_C$  (and thus to  $G_C$ ) [3], which has also been supported experimentally by different authors [3,13–15]. Thus, the advantage of the EWF method compared to the *J*-Integral procedure is, in many cases, its experimental simplicity.

# 3. Materials

The PP commercial grades chosen were one homopolymer (called H0) and three low-EC block copolymers (EPBC), with 5.5, 7.4 and 12% ethylene (called C1, C2 and C3, respectively), as determined by Fourier transformed infra-red (FTIR) spectroscopy. The material was received as pellets, and cast-extruded to obtain 90  $\mu$ m nominal thickness (*t*) non-oriented films. In order to homogenise the crystal microstructure of the material, which is basically smectic (or "quenched") after rapid cooling [2], the films were annealed for 1 h in a fan-assisted oven at 120°C (controlled to  $\pm 2^{\circ}$ C), producing a transformation of the smectic phase into a monoclinic state.

## 4. Experimental

#### 4.1. Dynamic mechanical analysis

The evolution of the dynamic storage modulus (E') and the loss factor (tan  $\delta$ ) with temperature was studied in tensile mode on a Dynamic Mechanical Analysis TMA Instruments DMA 2980 apparatus, at 1 Hz and 3°C/min, from -100 to 150°C. The sample dimensions were 10 and 5 mm in gauge length and width, respectively.



Fig. 1. DENT specimens used for the EWF tests, indicating the different energy dissipation zones involved.

#### 4.2. Mechanical and fracture tests

Tensile tests were conducted on a universal testing machine (Instron 5567) equipped with a 1000 N load cell and an environmental chamber, at different temperatures in the range between -40 and  $+70^{\circ}$ C ( $\pm 2^{\circ}$ C) and a crosshead speed of 2 mm/min. With the aim of increasing the accuracy of the results, the *dumbbell* specimens, tested according to ASTM D638-91 standard, were individually measured in their thickness with an induction based coating measurer (precision of 1  $\mu$ m). The yield stress ( $\sigma_y$ ), considered as the maximum stress, and the elastic modulus were calculated from the engineering stress–strain curves.

The EWF tests were performed on the same equipment and in the same temperature range as the tensile tests at a crosshead speed of 2 mm/min. Deeply double edge-notched samples (DENT, Mode I) were prepared by cutting the sheets into rectangular coupons of total length  $Z_t = 90 \text{ mm}$  (with a length between the grips of Z = 60 mm) and a width W = 60 mm (Fig. 1). Initial notches were made perpendicular to the tensile direction (which coincided with the extrusion direction) with a fresh razor blade, obtaining for each set at least 20 specimens with ligament lengths varying between 5 and 25 mm. The ligament lengths and the thickness were measured before the test using a travelling binocular lens and the same apparatus described above, respectively. The load-displacement curves were recorded, and the absorbed energy calculated by integration of the area under the curve (Fig. 2). The ligament length range was selected so that the films fractured in a plane-stress state, which was checked by plotting the net-section maximum stress ( $\sigma_{net}$ ) versus ligament length, and observing if the values were consistent with the Hill's prediction of  $\sigma_{net} = 1.15\sigma_v$  in plane-stress



Fig. 2. Effect of test temperature (a) and ethylene content (b) on the load– displacement curves of H0 DENT specimens, with a ligament length of around 14 mm.

[16]. The plastic zone size was measured on the fractured specimens using the travelling binocular lens mentioned.

## 5. Results

## 5.1. Viscoelastic behaviour

In Fig. 3, tan  $\delta$  is plotted against temperature for the four materials studied. Two clear transitions can be observed around -40 and 20°C, and these correspond to the glass transition (or  $\beta$  relaxation) of ethylene ( $T_g^E$ ) and propylene ( $T_g^P$ ) blocks, respectively [17]. It can be observed that the area of the ethylene transition peak increases sharply with the EC, and that of the propylene peak decreases slightly as the EC increases (note, however, that the relative area of both glass transition peaks is not proportional to the ethylene and propylene content, the ethylene transition peak being much more emphasised than the propylene one).



Fig. 3. Evolution of  $tan(\delta)$  as a function of temperature for the four materials studied.

## 5.2. Fracture behaviour

Three types of fracture behaviour were observed during the tests of the DENT samples: *partially brittle, ductile* and *necking with no fracture*, as can be seen in Fig. 4. The *partially brittle* fracture was only observed on the H0 samples tested at  $-40^{\circ}$ C and for the samples with large ligaments tested at  $-20^{\circ}$ C, giving a load–deflection curve with a maximum and a precipitous load-drop (Fig. 2(a)). This behaviour invalidates the use of the EWF for the treatment of the data, since a requirement of the theory is that the ligament is fully yielded before the propagation starts [10]. Clearly, this condition is not met in these samples. Neither is linear elastic fracture mechanics (LEFM) applicable, because the basic requirements of the standard are not satisfied [18]. For the majority of the specimens, the fracture was



Fig. 4. Photomicrographs of the ligament of the fractured specimens as a function of test temperature and material.

stable and *ductile* showing the typical behaviour of polypropylene films in a DENT configuration, already described in our previous works [8,19] as shown in Fig. 2. Crack propagation was observed to occur after ligament yielding in almost all cases (except for H0 at  $-40^{\circ}$ C). Here, the EWF procedure is applicable. From Fig. 2(a), it can be observed that, as the temperature is raised, the load level decreases but the total deformation is increased. The EC has a similar effect, as can be seen in Fig. 2(b). Necking is reflected by the very strong crack-tip blunting mechanism that prohibits crack initiation and propagation. This only occurred in the C2 and C3 samples tested at 70°C and some C3 samples at 50°C (Fig. 4). In these cases, the use of the EWF method of analysis was discarded for samples that showed high degrees of crack-tip blunting. It is noted that similar observations have already been found in PP with high ductility and reported in the literature [2,20].

The plane-stress criterion was checked against Hill's analysis. The results are shown in Fig. 5 and they agree well with the predicted value of  $1.15\sigma_y$  for the four materials studied particularly at low temperatures.

## 5.3. Plastic zone

Another remarkable feature of the fractured specimens is the variations in the different amount of stress-whitening (Fig. 4). It can be seen that this phenomenon only occurs for the copolymers, being more intense as EC increases. Also, low temperatures enhance stress-whitening with a threshold temperature (around 23-50°C for the EPBC) which delimits the range where the phenomenon is visible or not [21]. This temperature seems to increase as EC is higher. Furthermore, the revelation of the stress (or deformation) level at which the transparency is decreased gives the opportunity to observe an interesting feature for the EPBC. At low T, a double plastic zone can be distinguished. A large, diffuse area, that we call the diffuse outer plastic zone (DOPZ), and a more intense and confined, eye-shaped [2] area, that we call the Intense Outer Plastic Zone (IOPZ), are shown in Fig. 6. In fact, both zones are related to strictly different deformation stages. The DOPZ appears for the low-T cases during the initial loading at the end of the elastic regime (when crack propagation has not started yet) and reveals the typical LEFM stress distribution in a doublecrack plate [22]. The size of the zone depends on the temperature (higher as T decreases), and also on the EC (the highest ethylene copolymer shows the largest DOPZ), as can be seen in Fig. 4. Careful observations of the ligament during the test reveal that when all the DOPZ have developed, no variations in the specimen dimensions remain, and the energy absorption is minimal compared to the subsequent processes that are going to take place. Moreover, it can be observed from Fig. 7 that the shape of the DOPZ is independent of the ligament length for the same T, and its size is directly related to  $l^2$ . We believe that this diffuse stress-whitened zone is related to the low molecular



Fig. 5. Plot of  $\sigma_{net}$  against *l* for checking the plane-stress Hill's criterion. (a) H0 at different temperatures (+: 70°C; \*: 50°C;  $\diamond$ : 23°C;  $\bigcirc$ : 0°C;  $\blacksquare$ : -20°C;  $\blacktriangle$ : -40°C). (b) The four materials at 0°C ( $\diamond$ : H0;  $\blacksquare$ : C1;  $\blacktriangle$ : C2;  $\bigcirc$ : C3). Dashed lines indicate  $1.15\sigma_y$  for each test condition obtained from tensile tests.

mobility at these test conditions (low T) which hampers the relaxation processes that normally allow the material to re-order at the molecular level, thus "deleting" the traces of irreversible deformation. In accordance with that supposition, when the same materials are tested at impact rates, the same double plastic zone phenomenon is found, though more work is still being carried out in that direction.

On the other hand, the IOPZ which, unlike the DOPZ, is



Fig. 6. Schematic drawing of the double plastic zone, indicating the different zones (FPZ, fracture process zone; IOPZ, intense outer plastic zone; DOPZ, diffuse outer plastic zone).



Fig. 7. Photomicrographs of fractured C3 DDENT specimens showing double plastic zone, tested at  $-40^{\circ}$ C, with different ligament lengths (indicated in the figure).

always present (independent of T) and is related to the area that is intensely deformed during the whole fracture process. This involves deformation from the initial yielding and necking, and subsequent plastic drawing that occurs during crack propagation. Normally, the limit of the IOPZ is a sudden reduction in thickness (see Fig. 8), whereas the section does not vary at the end of the DOPZ.

# 5.4. Determination of EWF parameters

By plotting the  $w_f$  vs. *l* diagrams of the sets that showed ductile fracture (Fig. 9), very good linear relationships were obtained (except H0 samples at 50°C, where more scatter was observed). From these straight lines, the values of  $w_e$ and  $\beta w_p$  were calculated. A general trend that can be seen is that an increase in *T* produces a progressive raise of the  $w_f$ values, giving an increase of the *Y*-axis intercept ( $w_e$ ) and the slope ( $\beta w_p$ ). However, this trend is much less pronounced as the EC is increased, and the regression lines tend to merge to a single line (see Fig. 9(d)). In the



Fig. 8. Photomicrographs of the section (right figures) obtained from the end of the IOPZ in the ligament region of fractured H0 and C1 specimens, as indicated in the left figure. The arrows on the right figures indicate the limit of the IOPZ, which coincide with the reduction in thickness.

H0 diagram (Fig. 9(a)), the data set at  $-40^{\circ}$ C was not used to calculate the fracture parameters because the samples showed unstable fracture, though they are plotted here for comparison purposes.

With respect to the evaluation of  $w_p$ , the method suggested in the ESIS protocol of 1993 [23] and widely used by different authors [2,7,19,20,24–26] was used. Considering that the shape of the plastic zone shape is the intersection of two parabolas (eye-shaped) [2], one can derive the following expression:

$$h = 3/2\beta l \tag{3}$$

where *h* is the height of the plastic zone (taking into account the two parts of the broken specimen). By plotting *h* vs. *l* (Fig. 10) and calculating the least squares regression line,  $\beta$ is obtained from the slope of the line, and therefore  $w_p$  can be calculated from  $\beta w_p$ . Usually, the line does not pass through the origin as should theoretically be, but the *Y*-axis intercept is neglected (the line is not imposed to pass through the origin). It should be noticed that in the case of the presence of a double plastic zone (IOPZ and DOPZ), the plastic zone that was taken into account for the calculation of  $\beta$  was only the IOPZ, for the following reasons:

- when only one zone is seen, there is no doubt that it is the IOPZ (in the cases of H0, or the EPBC at T ≥ 23°C), so if one wants to compare the values of w<sub>p</sub>, the same plastic zone must be considered;
- the energy dissipated in the process that involves the DOPZ is low compared to the energy for creating the IOPZ;
- even though DOPZ should be measured, many difficulties would be found in determining its limits.

It has to be kept in mind that the EWF theory was initially developed for ductile metals, and as the deformation mechanisms are noticeably different, some important differences may exist in the way as the theory is applied.

The results of the EWF fracture parameters for the four materials as a function of T are summarised in Fig. 11, and listed in Table 1. However, the trends as shown are only approximations and have to be taken carefully. In some



Fig. 9. Plots of  $w_f$  against *l* for the four materials at different test temperatures (+: 70°C; \*: 50°C;  $\bullet: 23°C$ ;  $\odot: 0°C$ ;  $\blacksquare: -20°C$ ;  $\blacktriangle: -40°C$  ( $\Delta:$  brittle)).

cases, the experimental error may be important, especially in the values of  $\beta$  and  $w_p$ , which have been deduced indirectly from the measured values of *h* (notice that *h* has been determined with difficulty in some sets). Hence, the major part of the discussion is focussed on the product  $\beta w_p$  as the confidence is higher. Also, in the literature more discussions are carried in terms of  $\beta w_p$  rather than  $w_p$ .



Fig. 10. Plot of *h* against *l* for H0 at different test temperatures for the determination of  $\beta$  (+: 70°C; \*: 50°C;  $\blacklozenge$ : 23°C;  $\bigcirc$ : 0°C;  $\blacksquare$ : -20°C].

# 5.5. Temperature effect on the specific essential work

It can be seen in Fig. 11(a) that  $w_e$  is very sensitive to the temperature for the homopolymer compared to the copolymers. For H0,  $w_e$  increases sharply with *T* until the material starts to lose its structural cohesion, going through a maximum. Conversely, the copolymers have similar  $w_e$  values (~45 kJ/m<sup>2</sup>) up to a threshold temperature ( $T_{th}$ ). This  $T_{th}$ , above which  $w_e$  starts to decrease, is found to exist in all materials except C2 (possibly attributed to experimental error). It is noted that  $T_{th}$  decreases as the EC increases (i.e. 50–60°C for H0 and C1, and 20–30°C for C3). At  $T < T_g^P$ ,  $w_e$  is clearly higher for the EPBC compared to the H0.

#### 5.6. Temperature effect on the specific non-essential work

The dependence of  $\beta w_p$  on temperature is plotted in Fig. 11(b). For the homopolymer,  $\beta w_p$  increases dramatically as *T* increases, reaching a maximum at around 40°C (similarly to the  $w_e$  results) and dropping beyond that temperature. Obviously, no  $\beta w_p$  data are available for H0 at  $-40^{\circ}$ C, but one could estimate that this term tends to zero as *T* decreases, since the plastic zone is almost non-existent at very low *T* [3]. The steep rise of the data for H0 gradually disappears for EPBC as the EC increases, although the same



Fig. 11. Evolution of  $w_e$  (a),  $\beta w_p$  (b),  $\beta$  (c) and  $w_p$  (d) with temperature for the different materials studied.

trend is apparent but it is clearly less marked. We cannot, however, extrapolate confidently the dependence of  $\beta w_p$  to lower temperatures ( $T < -40^{\circ}$ C), or higher temperatures based on these results. Again, like  $w_e$ , the EPBC show higher values of  $\beta w_p$  than H0 at low *T*.

From the values of  $\beta$  (Fig. 11(c)),  $w_p$  is calculated and shown in Fig. 11(d). H0 presents higher  $w_p$  values than the EPBC at all *T* studied, and in general, this value decreases as the EC is higher at all *T*. As *T* increases,  $w_p$  decreases continuously for the homopolymer. Nevertheless, the EPBC show a totally different dependence. Firstly, there is a maximum that matches perfectly with the tan  $\delta$  peak related to  $T_g^P$ . Secondly, under 0°C, variation in *T* seems not to affect  $w_p$ .

# 6. Discussion

The temperature dependence of the fracture toughness has been treated widely in the literature [27-34]. Sims [29], and particularly Williams and collaborators [30-32] have treated the case of PP systems, basically at impact rates. Much of the work has focussed on the question of

whether or not the energy absorption mechanisms have a direct correlation with the molecular relaxation processes occurring in the material. It should however be realised that adiabatic heating at the crack-tip, which produces a blunting phenomenon [33,34], may also increase the toughness. Nevertheless, in the present work, this second consideration might be ignored, as the test rate and the reduced sample thickness do not allow heat accumulation. Hence, an explanation based on molecular relaxation seems sensible to understand the experimental results.

Concerning H0, the dependence of  $w_e$  can be related to its glass transition. The inflection point in the curve matches fairly well the  $T_g^P$  measured by DMA (note that  $T_g$  is a temperature range, not a fixed value of *T*). Thus, it is convincing to explain the maximum toughness results in terms of the molecular relaxation activated at that temperature. The ductile– brittle transition that occurs at low *T* can be explained by the Ludwik–Davidenkov–Orowan criterion [35], which states that the fracture changes from ductile to brittle if the fracture stress drops below the yield stress. It is known that with decreasing *T* (or increasing test speed), the yield stress increases more rapidly than does the fracture stress.

On the other hand, the copolymers show no increase in  $w_e$ at temperatures near  $T_g^{\rm P}$  or  $T_g^{\rm E}$ , despite the PP blocks (that are nevertheless predominant in the material) and the ethylene segments that show important relaxation at 20 and -40°C, respectively, as seen by DMA. A possible explanation for the absence of a we maximum for the EPBC at 20°C could be that an increase in the energy dissipation due to  $T_g^P$  is offset by a decrease in the toughness attributed to the ethylene phase well above its  $T_g^E$ . A previous EWF study gave a value of  $w_e = 24.3 \text{ kJ/m}^2$  for LDPE films at room temperature and 2 mm/min [36], which is consistent with the results presented here. This same reasoning may explain why at RT the homopolymer shows higher  $w_e$  values than the EPBC. Another interesting feature is that  $T_{\rm th}$  at which  $w_{\rm e}$  starts to decrease clearly depends on the EC, and can be related to the less mechanical resistance to temperature of polyethylene than polypropylene. These results, however, deserve more work than what is in progress.

It can be derived from Fig. 11(b) that the effect of the molecular relaxation on the plastic item  $\beta w_p$  is as important as on  $w_e$  for H0, and even more pronounced for the EPBC. The EPBC present  $\beta w_p$  values that are higher at  $T < T_g^P$  and lower at  $T > T_g^P$  compared to the H0. A competition between the shape factor ( $\beta$ ) and the plastic energy density ( $w_p$ ) should be the key to understand the temperature dependence of the plastic term.

At high *T*, the propylene blocks (which are in majority) have no molecular mobility restriction and govern the plastic behaviour of the material. As the EC increases,  $\beta w_p$  decreases (particularly when the EC of C1 is exceeded). Again, this is in good agreement with previous work (for LDPE films, a value of  $\beta w_p = 5.1$  MJ/m<sup>3</sup> at RT has been reported [36], while in this work this value falls from around 13 to 9 as EC increases from 0 to 12%). The reason for such behaviour can be explained in terms of the variations of the terms included in  $\beta w_p$ . Although the plastic zone shape factor is higher for the EPBC (Fig. 11(c)) than for the H0, the specific plastic work is considerably lower as EC increases (Fig. 11(d)).

At low T, the molecular mobility of propylene segments is restricted, giving clearly a lower total plastic energy dissipation  $(\beta w_p)$  for the H0 with respect to the EPBC. The main reason is that, for the hompolymer, the size of the plastic zone is limited to very low values (Fig. 11(c)), which cannot be compensated by the notable increase of  $w_p$  (Fig. 11(d)). In this situation, much of the energy is stored elastically and may be released suddenly in an unstable fracture. While  $\beta$ increases with the EC, it can be seen that  $w_p$  remains low for all the copolymers, compared to the H0. Hence, the main effect of the ethylene copolymerisation is the drastic increase in  $\beta$ , which is significant enough to offset the reduction in the specific plastic work, particularly at low T. Although a good correlation with T exists between  $w_{\rm p}$ and  $\sigma_{\rm v}$  (Fig. 12) in the whole T range studied for H0, this no longer exists for the EPBC at  $T < T_g^P$ . More efforts are

	$(MJ/m^3)$		6	2	3	2	5
	3 (-) w <sub>p</sub>	1	.139 63.	.100 91.	.313 23.	.290 25.	.252 25.
	$(1/m^3)$	I	0	0	0	0	0
	$\beta w_{\rm p}$ (N	I	8.89	9.12	7.32	7.31	6.42
C3	$w_{\rm e}~({\rm kJ/m^2})$	I	22.6	37.8	46.8	43.2	49.8
	$w_p (MJ/m^3)$	I	65.4	76.1	40.3	40.5	39.8
	β (-)	I	0.144	0.125	0.205	.0197	0.163
	$\beta w_{\rm p}  ({\rm MJ/m^3})$	I	9.42	9.49	8.29	7.33	6.49
C2	$w_{\rm e}~({\rm kJ/m^2})$	I	51.1	43.9	39.9	41.2	45.5
	$w_p (MJ/m^3)$	93.4	75.1	115	48.6	43.4	44.9
	$\beta$ (-)	0.106	0.166	0.117	0.181	0.182	0.14
	$\beta w_{\rm p}  ({\rm MJ/m^3})$	9.90	12.5	13.4	8.79	7.89	6.28
C1	$w_{\rm e}  (\rm kJ/m^2)$	34.0	44.6	43.8	46.8	45.1	49.0
	<sub>p</sub> (MJ/m <sup>3</sup> )	19	36	96	22	46	I
	β (-) μ	0.088 1	0.105 1	0.066 1	0.019 2	0.015 2	I
	$\beta w_{\rm p}  ({\rm MJ/m^3})$	10.5	14.3	12.9	4.23	3.89	I
0H	$v_{\rm e}~({\rm kJ/m^2})$	70.3	33.4	52.0	51.1	25.0	
T (°C) 1	1	70	50 8	23 (	0	-20	-40

Table 1 EWF parameters for the materials studied in function of the test temperature



Fig. 12. Temperature dependence of  $\sigma_{y}$  for the four materials.

being done on the study of the deformation mechanisms which govern the plastic behaviour at high and low T in order to explain the results described, and these results will be given in a future paper.

It is interesting to make an overall comparison of the fracture properties of the copolymers with respect to the homopolymer. As discussed in recent works [2,20], in order to improve the ductile failure behaviour, it is necessary to increase simultaneously  $w_e$  and  $\beta w_p$ , since this brings about an increase of the total fracture work. However, it has been shown that, in some cases, the variation of a morphological characteristic (filler content [30], crystallinity [20] or crystal perfection [2]) produces an opposite effect on the EWF fracture parameters. That is, when one parameter increases the other decreases, and vice-versa. But if we consider EC as the material variable, then this is not the case in this study. On the one hand, it should be observed that at T > RT (and therefore above  $T_g^P$ ), H0 has better fracture properties (both  $w_e$  and  $\beta w_p$ ) than the copolymers. On the other hand, at T lower  $T_g^P$ , the scenario is reversed. In the range of the low temperatures studied, C1 shows the best fracture properties amongst the EPBC studied. Higher EC than that of C1 can be detrimental to other properties ( $\sigma_v$ , E,  $w_e$ , transparency, etc.), hence an optimum EC should therefore be selected to optimise the overall behaviour of PP-based materials. It has to be emphasised that the main interest of the EPBC at low temperatures is that they do not break in a brittle manner, which is a more desirable behaviour for many applications.

# 7. Conclusions

The EWF method has been successfully used to characterise the temperature effect on the fracture properties of PP and EPBC, studied in plane stress conditions as proven by the Hill's analysis of the net section maximum stress. The results show that, as expected, the homopolymer is found to be much more temperature sensitive than the EPBC. Two different ranges delimited by the glass-transition of PP, which is located around RT, are found. At temperatures above RT, the H0 has greater energy dissipation than the EPBC. In contrast, at low T, the homopolymer presents a ductile-brittle transition, which is suppressed in the range studied by copolymerisation with only a small EC. In this T range, the EPBC present higher fracture properties than H0. The specific plastic work and the plastic zone shape factor have been determined from the optical measures of the plastic zone size. Although the experimental error introduced in these calculations can be important, it has been useful to discuss the variation of  $\beta w_{p}$  in terms of  $\beta$  and  $w_{\rm p}$  changes with T. From the results, it follows that the main effect of the ethylene copolymerisation is the dramatic increase of the plastic zone size. The variations of the EWF fracture parameters with T are consistent with those of a previous study in LDPE, and can be explained in terms of molecular relaxation mechanisms, ethylene phase content, and changes in the yield and fracture stresses. However, further work is being done in order to study in depth the relationships between these parameters and the plastic deformation mechanisms involved. Finally, it has to be noted that some limitations of the EWF method were found with the more ductile materials, which showed a necking phenomenon without fracture propagation in DENT specimens at high T.

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