#### INTERFACE SCIENCE

# Study of the adhesion strength on overmoulded plastic materials using the essential work of interfacial fracture (EWIF) concept

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Received: 7 July 2007/Accepted: 21 April 2008/Published online: 13 May 2008 © Springer Science+Business Media, LLC 2008

**Abstract** The influence of the overmoulding processing conditions (melt and mould temperatures, holding pressure and injection rate) on the adhesion strength between two polymers in a hard-soft part was studied. The used materials were thermoplastic vulcanized elastomer (TPV) and polypropylene (PP). The study of the adhesion strength was made by measuring the essential work of interfacial fracture (EWIF) concept using a single edge-notched tension (SENT) geometry. It was observed that, the data distribution was good since an adequate correlation between the total work of interfacial fracture and the ligament length was obtained. It was verified that, when the melt temperature, and in some cases, the mould temperature are increased, a higher adhesion strength is obtained. These parameters affect in higher degree the adhesion strength, since they may improve the interdiffusion and the wetting process of the PP substrate by the molten material (TPV), causing as a result a good contact between them. A little influence was observed in the case of the holding pressure and injection rate.

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#### Introduction

Recently, new injection moulding methods have been developed, the "Multicomponent/Multimaterial injection moulding" being one of these novel techniques, also known as: "overmoulding" or "two-shot" moulding. This kind of technique is widely used in the manufacturing of hard-soft parts for several industrial sectors (automotive, packaging and medical), where an elastomer (soft) is joined to a rigid (hard) polymer without needing adhesives. It is a variation of the conventional injection moulding, and consists of moulding an initial portion of the component and then, when it is solidified, the part is rotated or transferred to another cavity and the second material is injected. This process needs at least two injection units to "overmould" the materials [1]. In the last decade, hard-soft overmoulding has radically changed the look, tactile, texture and functions of parts. Products like toothbrushes, razors, power tools, cameras, kitchen items and automobile-interior parts.

In the hard–soft parts' fabrication a thermoplastic elastomer (TPE) is injection moulded over or around a compatible (or at least miscible) substrate using either insert or multi-shot process. Nowadays, the followings are use as TPE's soft components: Olefin-based TPE, thermoplastic polyurethanes (TPU), thermoplastic vulcanizates (TPV), styrene–ethylene/butylenes–styrene block copolymers (SEBS). As substrates or rigid components the range is wider, and among them: acrylonytrile-butadiene-styrene terpolymers (ABS), polycarbonates (PC), polyamides (PA) and olefin-based polymers (polypropylene (PP), polyethylenes (PE)) can be found [2].

In automotive parts, one of the most used combinations for hard-soft parts is olefin-based TPV/PP. TPV are an especial class of TPE produced by dynamic vulcanization techniques, which consists in elastomer vulcanization during its blending with a thermoplastic polymer using an internal mixer (i.e. Banbury) or twin-screw extruder. This way, a dispersion of elastomer particles within a thermoplastic matrix is obtained, offering many of the product features of crosslinked rubbers, coupled with the processability of thermoplastic polymers. They are outstanding in their low compression set and high extensibility, in addition, to their solvent resistance. TPV have properties that occur between those of TPE and TPU in performance, but they are only slightly higher in price than TPE [3].

#### Adhesion joint models

An universal model does not exist to explain the different types of adhesion joints. In general, each adhesion joint may be explained considering several models such as mechanical adhesion, electrical model, diffusion model, thermodynamic adsorption model, chemical model, acid– base model, rheological model and interphase layers of weak cohesion mechanism [4].

The diffusion model is essentially the only one that allows explaining auto-adhesion between similar polymers. It states that there is a migration of polymeric chains that are soluble, generating an increase in the degree of cohesion at the interface. Due to that, to achieve an optimal diffusion, polymers used should be miscible (soluble) between them. This miscibility may be quantified by their solubility parameters ( $\delta$ ), which should be as close as possible between them. However, this model shows possible deviations when the considered polymers are semicrystallines and/or they have strong chemical interactions [5].

Using this model to the overmoulding TPV/PP injection process, two key stages may be identified: wetting of the second molten material over the substrate (solidified hard component), assuring an intimate contact among them; and interdifussion of polymer chains between the components, favoured by a "partial" melt of the substrate in the interface. After solidification the joint between both plastic materials will take place. It is important to mention that this interdifussion may be affected by the migration of additives contained in the TPV and/or substrate formulations [5–7].

In accordance with the foregoing, as general criteria for choosing convenient material combination may be considered the similarity of the chemical structure and solubility parameter ( $\delta$ ), and in some cases the possibility of some kind of reactivity between them. However, this approach does not assure a good adhesive strength between them, as those interactions could be influenced by TPV's rheological characteristics when it makes contact with the substrate. Thus, it should be considered, temperature at the TPE's flow front in the cavity, the cooling temperature applied, holding pressure, among others variables.

Based on these statements, the main idea of this work was to study the influence of the overmoulding process conditions (melt and mould temperatures, injection rate and holding pressure) over the adhesion strength between a TPV over a PP substrate, applying the essential work of interfacial fracture concept.

The essential work of interfacial fracture (EWIF)

For adhesion strength evaluation it is common to use the typical configuration of T-peeling test [8]. However, some problems could arise when working with materials that show high extensibility, like the case of TPE. In a previous study made by Candal et al. [9] it was evidenced that this test configuration may not be applied for the analysis of the effect of the processing conditions, as in many of the injection moulding conditions TPV showed tearing before interfacial separation. Furthermore, the obtained value depends on the selected size of the adhered surface.

As an alternative, Laüke and Schüller [10] suggested the use of the essential work of interfacial fracture (EWIF) analysis, based on the essential work of fracture (EWF) concept. A hard-hard join (PP/PA 70/30 blend-PP copolymer) was used for the study. Briefly, the postulates of the EWF provide that in systems where crack propagation begins after a full plastic collapse has occurred, the total work of fracture ( $W_F$ ) may be separated into two components [8]:

$$W_{\rm F} = W_{\rm e} + W_{\rm p} \tag{1}$$

where  $W_e$  is the essential work of fracture and includes both: the applied work to generate the constriction at the crack tip and the necessary work to generate a free surface;  $W_p$  is the non-essential work of fracture and represents the consumed energy in any dissipative deformation process (usually plastic) of the network of material, that surrounds the fracture process zone (crack propagation plane), also known as the "outer process zone (OPZ)". Considering the ligament area, Eq. 1 may be written using specific terms:

$$w_{\rm F} = w_{\rm e} + \beta \ w_{\rm p} L \tag{2}$$

where  $w_e$  is the specific essential work of fracture or the  $W_e$ by the sectional ligament area  $(L \times B)$  and it may be considered as a typical parameter of the material under test,  $w_p$  is the specific non-essential work of fracture by volume of deformed material in the OPZ and  $\beta$  is a geometrical factor with a value <1 that quantifies how much this volume deviates from having an ideal volume equal to  $L^2B$ , in which *B* is the specimen thickness and *L* is ligament length.

Lauke and Schüller [10] have modified Eq. 1 with reference to the EWIF concept. Considering that part of the applied work during the mechanical solicitation is converted into energy for the creation of new surface and that crack propagation will proceed at the interface of two phases, the total work of interfacial fracture ( $W_{\rm IF}$ ) may be partitioned into two contributions:

$$W_{\rm IF} = W_{\rm I} + W_{\rm NI} \tag{3}$$

where  $W_{\rm I}$  represents the essential work of interfacial fracture and  $W_{\rm NI}$  is the non-essential work of interfacial fracture. Similar to Eq. 2,  $W_{\rm IF}$  may be expressed by their specific terms:

$$w_{\rm IF} = w + (\beta_1 w_{\rm NI,1} + \beta_2 w_{,2})L. \tag{4}$$

Notice that in this case the non-essential work of interfacial fracture  $(\beta_x w_{NI,x})$  is decomposed in two additives terms, referring each one to the components joined at the interface, where crack propagation is supposed to proceed. This splitting should be done to consider the possible differences regarding the volume of plastic deformation in each component, as in the case of the present study.

It is necessary to underline, that during the separation process of the two components, a break of the adhesion in the interface formed occurs, that caused a break of the adhesion formed in the interface between the two overmoulded materials. The resistance or strength to separate is known as cohesive resistance. It is important to mention, that during the testing, the applied work is converted into energy for the creation of new fracture surfaces. When the two adhered surfaces of the PP and the TPV are separated, the material plastically deformed stores the potential energy that is responsible for the growth of the crack precisely done in the interface of both materials. It is important to say that since it is a polymer-polymer interface, what may be determined is the  $w_{\rm I}$  and in this case, it corresponds to the used energy for separating both materials. Considering this, it is assumed that  $w_{I}$  is a measure of the joint resistance and higher  $w_{\rm I}$  results in greater adhesion.

## **Experimental part**

## Materials

Polypropylene (PP) homopolymer *Moplen HP501H* from BASELL POLYOLEFINS and TPV *Santoprene 8211-55* from ADVANCED ELASTOMER SYSTEMS were the used materials (Table 1). According to a preliminary characterization by Fourier transformed Infrared Spectroscopy (FT-IR) and Differential Scanning Calorimetry (DSC), *Santoprene* is a PP/EPR-based TPV with a high percentage of olefinic paraffin, commonly used to enhance processability and a Table 1 Main properties of polymers used in this study

Properties	Value		
	PP (HP501H)	TPV (8211-55)	
Density (g/cm <sup>3</sup> )	0.9	0.93	
Hardness (Shore D)	70		
Hardness (Shore A)		55	
Melt flow index (230/2.16) <sup>a</sup> (dg/min)	2.1	_	
Rupture elongation (%)	700	480	
Solubility parameter, $\delta^{b}$ (MPa <sup>1/2</sup> )	18.8		
Solubility parameter, $\delta^{c}$ (MPa <sup>1/2</sup> )		19.0	
Young modulus (MPa)	1450		
Tensile stress at 100% (MPa)		2.1	

<sup>a</sup> ASTM D1238-04c

<sup>b</sup> See Ref. [11]

<sup>c</sup> See Ref. [12]

silica as a filler. The solubility parameter ( $\delta$ ) of TPV, a lightly crosslinked polymer, was indirectly determined by a treatment, with a number of solvents with different known  $\delta$ . The  $\delta$  value of the best solvent (which swells the specimen the most) is considered as the nominal  $\delta$  value of TPV [5, 13].

To simulate the rheological behaviour of TPV during the cavity filling stage of injection moulding, the C-MOLD 2000.7 was used, drawing the half plate with 3000 nodes. The TPV's numerical parameters of constitutive equations ( $\eta_0$  (Newtonian viscosity);  $\tau^*$  (critical stress level at which  $\eta$  transitions from the Newtonian plateau); n (Power law index);  $A_1$ ,  $A_2$ ,  $D_1$ ,  $D_2$  and  $D_3$  (constants)) used in the software (Cross exponential, cross exponential/WLF and hydraulic loss [12]) were determined by experimental rheological curves (apparent viscosity versus shear strain rate). Capillar rheometry tests were performed in a GÖTTF-ERT Rheograph 2000 at four temperatures (190, 200, 220 and 230 °C) using three dies with different length-todiameter ratio (L/D = 30/1, 20/1 and 10/1), to make the respective Bagley and Rabinowitsh's correction. Numerical results are reported in Table 2.

## Overmoulded specimens

Test specimens for mechanical testing were extracted from double edge-notched tension (DENT) specimens directly overmoulded in a two-step process. Initially, PP half plates  $(100 \times 50 \times 1 \text{ mm}^3)$  were injection moulded. One side of these PP plates was partially wrapped with aluminium foil (thickness =  $0.015 \pm 0.001 \text{ mm}$ ) leaving a certain length of "free PP" between the covered zones. This assembly was used as an insert in the mould cavity  $(100 \times 100 \times 1 \text{ mm}^3)$  during the injection moulding of TPV. This way, sets of DENT specimens with about 14

Table 2 TPV's rheological parameters according to WLF model

Rheological parameter	Value	
<i>n</i> (–)	0.212	
$\tau^*$ (Pa)	$2.038 \times 10^{4}$	
D <sub>1</sub> (Pa s)	$2.79 \times 10^{16}$	
<i>D</i> <sub>2</sub> (K)	223.15	
<i>D</i> <sub>3</sub> (K/Pa)	0	
A <sub>1</sub> (-)	58.96	
<i>A</i> <sub>2</sub> (K)	246	

different ligament lengths (L) ranging from 6 to 32 mm and very sharp notches were obtained. After cutting them in half, single edge-notched tension (SENT) specimens were obtained (Fig. 1) and they were used to evaluate the adhesion strength.

The injection moulding machine used was a MATEU & SOLÉ *Meteor 440/90*. The process conditions for PP halves were: melt temperature  $(T_m)$  at 220 °C, injection rate  $(R_i)$  of 20.7 g/s, holding pressure  $(P_H)$  of 50 MPa, holding pressure time  $(t_H)$  of 8 s and mould cavity temperature  $(T_{mld})$  at 40 °C. For TPV several process conditions have been used, summarized and codified in Table 3. The TPV's processing conditions considered as variables along this study were:  $T_m$  (low L = 190 °C; medium M = 210 °C and high H = 230 °C),  $R_i$  (6, 11 and 32 g/s),  $T_{mld}$  (40, 65 and 90 °C) and  $P_H$  (20 and 35 MPa), remaining constant  $t_H$ .



Fig. 1 TPV/PP SENT specimen geometry

 Table 3
 Processing conditions and codes used in the overmoulding of TPV

Conditions Code <sup>a</sup>	Melt temperature, $T_{\rm m}$ (°C)	Mould temperat T <sub>mld</sub> (°C	Holdir ure, pressu ) $P_{\rm H}$ (M	ng Injection re, rate, IPa) $R_i$ (g/s)
L1	190	40	20	6
L2		40	20	11
L3		40	35	6
L4		40	35	11
L5		40	35	32
L6		65	35	6
L7		65	35	11
L8		65	35	32
L9		90	35	6
L10		90	35	11
L11		90	35	32
M1	210	40	20	11
M2		40	35	6
M3		40	35	11
M4		40	35	32
M5		65	35	11
M6		90	35	6
M7		90	35	11
M8		90	35	32
H1	230	40	20	11
H2		40	35	6
H3		40	35	11
H4		40	35	32
H5		65	35	11
H6		90	35	6
H7		90	35	11
H8		90	35	32

Melt temperature: L = 190 °C, M = 210 °C and H = 230 °C

### Mechanical testing

While the DENT configuration is often used (and recommended) for the EWF analysis, in a previous study made by Candal et al. [9] the results in this configuration showed high dispersion due to the asymmetric crack propagation. Despite this, SENT geometry configuration has been chosen.

Tests have been performed in a LLOYD universal testing machine using a crosshead speed of 20 mm/min, a distance between grips of 50 mm and at  $23 \pm 1$  °C. For each ligament length (3–16 mm, after being cut), load–displacements (*P*–*d*) curves were used to calculate the essential work of interfacial fracture (*w*<sub>I</sub>) parameter following the same procedure as for EWF analysis, described elsewhere [14–16].

Using a stereoscopic MEDILAB microscope ( $\pm 0.0001 \text{ mm}$ ), all the real ligament lengths from the fracture surface have been determined after testing. Additionally, and with the help of image analyser software, the mean surface area of adhered rubber was estimated on each tested specimen.

#### **Results and discussion**

Figure 2 shows typical Load–displacement (P-d) curves registered with SENT specimens. It can be observed that regardless of the ligament length, curves show the same characteristics. The load increases rapidly until a maximum is reached, where the onset of crack propagation was observed. After that point, a smooth load reduction was observed, which coincided with the stable interface crack propagation. In some of the displacement values, a sudden drop occurred, due to complete interface separation. This similarity of curves, as observed and reported by other works on EWF analysis, should assure that the form of the outer process zone is non-dependant on ligament length and good linear fitting of Eq. 3 would be expected [14–16]. As Fig. 3 and Table 4 show, a low dispersion on linear fitting of Eq. 4 was observed.

It is important to mention that, although the adaptation of EWF was performed with all the criteria involved in its application following ESIS's protocol [8], the requirement of fully yielded ligament prior to crack propagation was not satisfied. In the outer process zone (surrounding the interface) and at the moment where propagation was initiated, no-plastic deformation (or even elastic) was observed on PP substrate. The greater amount of deformation fit to nonlinear elastic (highly recoverable and time dependant) of TPV half.

On one hand, this implies that Hill's plasticity criterion loses importance and verification of pure stress state situation may not be assured by this way. On the other hand,



**Fig. 2** Typical load–displacement (*P*–*d*) curves of overmoulded SENT specimens at different ligament lengths (*L*). Condition M3



Fig. 3 Typical plots  $w_{IF}$  versus L at injection temperature of 190, 210 and 230 °C ( $P_H = 35$  MPa,  $T_{mld} = 40$  °C and  $R_i = 11$  g/s)

and the most important thing, is that the non-essential term of the analysis may be very difficult to interpret hence low values are observed. However, this aspect should be carefully analysed and it is a subject of study by the authors. Analysing Eq. 4, whole non-essential term of the analysis will be up to TPV half, so:

$$w_{\rm IF} = w_{\rm I} + \beta_{\rm TPV} \ w_{\rm NI, TPV} L \tag{5}$$

Since no sign of plastic or permanent deformation may be detected or even measured,  $\beta_{\text{TPV}}$  should be very low. Thus, this term may be associated to the typical elastomeric hysteresis loop.

Additionally, this situation would imply that  $w_I$  shows some contribution of the stored energy during extension of TPV. Some hysteresis tests have been performed on TPV half, and the results showed that no significant variations on the loop, in the processing conditions studied were observed.

However, it should be kept in mind that during the test, a separation of the two overmoulded materials occurred, breaking the adhesion formed in the interface between them. Due to that, the work applied is transformed into energy for new free surface's creation regarding  $w_{\rm I}$ , which may be associated with the cohesive resistance.

In view of all this, it was decided to use the essential parameter  $(w_{\rm I})$  as a quantitative adhesion energetic parameter for comparative purposes between processing conditions.

Influence of the melt temperature  $(T_m)$ 

As it can be evidenced in Fig. 4, when melt temperature  $(T_m)$  increases a higher  $w_I$  can be observed, and as a result there is adhesion improvement between the two materials. These

 
 Table 4
 Essential and non-essential work of interfacial fracture for each condition

Conditions Code	Essential work of interfacial fracture, w <sub>I</sub> (kJ/m <sup>2</sup> )	Non-Essential work of interfacial fracture $\beta_{\text{TPV}} w_{\text{NI,TPV}} (\text{MJ/m}^3)$	$R^2$
L1	$1.4 \pm 0.2$	$0.28\pm0.02$	0.990
L2	$3.0 \pm 0.1$	$0.18\pm0.01$	0.995
L3	$0.43 \pm 0.09$	$0.15\pm0.01$	0.994
L4	$1.5 \pm 0.2$	$0.40\pm0.01$	0.991
L5	$0.14 \pm 0.09$	$0.30\pm0.01$	0.995
L6	$1.2 \pm 0.1$	$0.15\pm0.01$	0.991
L7	$3.1 \pm 0.2$	$0.32\pm0.01$	0.991
L8	$0.2 \pm 0.1$	$0.40\pm0.01$	0.998
L9	$0.6 \pm 0.2$	$0.34\pm0.02$	0.992
L10	$3.5\pm0.2$	$0.39\pm0.01$	0.991
L11	$0.32\pm0.09$	$0.48\pm0.01$	0.998
M1	$6.2 \pm 0.2$	$0.28\pm0.01$	0.990
M2	$1.3 \pm 0.2$	$0.35\pm0.02$	0.990
M3	$2.3\pm0.2$	$0.52\pm0.02$	0.994
M4	$2.0 \pm 0.2$	$0.33\pm0.01$	0.993
M5	$5.0 \pm 0.2$	$0.41 \pm 0.02$	0.991
M6	$1.5 \pm 0.2$	$0.35\pm0.02$	0.991
M7	$2.9\pm0.2$	$0.55\pm0.02$	0.995
M8	$2.4 \pm 0.3$	$0.70\pm0.03$	0.994
H1	$6.5 \pm 0.2$	$0.35\pm0.02$	0.991
H2	$2.5 \pm 0.2$	$0.49\pm0.01$	0.997
H3	$6.1 \pm 0.2$	$0.49\pm0.02$	0.993
H4	$3.2 \pm 0.3$	$0.53\pm0.03$	0.990
H5	$5.1 \pm 0.2$	$0.45\pm0.02$	0.990
H6	$1.9 \pm 0.3$	$0.66\pm0.02$	0.993
H7	$5.8 \pm 0.2$	$0.59\pm0.02$	0.993
H8	$4.0 \pm 0.3$	$0.62\pm0.03$	0.994

results coincide with previous ones reported by Candal et al. [13] regarding adhesion strength measured by tensile tests as adhesion force ( $F_{adh}$ ). It is expected that increasing  $T_m$ , more intimate contact between molten material (TPV) and the substrate (PP) is promoted, generating greater wetting, fusion and diffusion of TPV through PP, which creates a molecular entanglement between them [7].

These results coincide with Kim and Suh [17] observations. They reported that,  $T_m$  is the processing condition that affects weldline strength the most. That fact is easily comparable with adhesion strength between two overmoulded materials. Using the theory of chains' diffusion, they expressed that weldline strength depends on the degree of adhesion between both cooled down fronts. On the other hand, McKelvey and Strome [18], when working with sealability of polyethylene films, pointed out that increasing the second extruded film temperature, the molecular diffusion



**Fig. 4** Essential work of interfacial fracture  $(w_i)$  variation with melt temperature  $(T_m)$  grouped by similar process conditions  $(P_H, T_{mld} \text{ and } R_i)$ . See Table 3 for codes

rate between two materials will increase and help to observe better weldline structure properties.

Huang and Chen [19] studied the effects of processing parameters, such as melt and mould temperatures and cooling time, on the bonding strength of the interface in a sequential two-staged injection moulding process of polystyrene (PS). A theoretical bonding strength model, which accounts for the cooling profile through the thickness of the part and the interpenetration depth of polymer chains across the interface, was proposed.

Considering what Weng et al. [7] proposed, it may be stated that, since TPV and PP have similar solubility parameter ( $\delta$ ), a higher degree of interaction (van der Waals type) among them could be formed. When molten TPV makes contact with the surface of PP substrate, there is an interdiffusion. TPV molecules may diffuse through solidified PP, and PP molecules could diffuse into TPV due to a partial and localized melting, thus forming entanglements between them. Once TPV solidifies, an interface with TPV/PP gradient of concentration could be formed, creating strong adhesion between them.

Analysing the fracture surface of the specimen (Fig. 5), the pattern generated corresponds to the sticky type: separation of the interface leaving small rest of TPV on PP surface. None of the evaluated cases showed a cohesive rupture type, since TPV never broke in the test conditions. Measurements of the total area of residual rubber adhered to the substrate as a function of ligament length allows verifying the obtained results. The higher the  $T_m$  is, higher is the covered area by TPV residual over PP (Fig. 6). It may be analysed not only by the absolute area value, but also regarding the relation with the ligament length or length of substrate if the slope of the representation is given in Fig. 6.



6

Rubber Adhered x 10<sup>-4</sup> [mm<sup>2</sup> Surface Area of Residual



Fig. 6 Surface area of residual rubber adhered to substrate variation with melt temperature. Process conditions L4, M3 and H3. See Table 3

4

Effect of the mould temperature  $(T_{mld})$ 

2

0

T.: 190°C

T\_: 210°C

T\_: 230°C

At first, it might be expected that the higher the  $T_{mld}$ , greater is the adhesion between both materials, since the temperature at the end of TPV's flow path will be higher, generating an overheating substrate (PP) favouring the interdiffusion process. Nevertheless, this trend is only observed at low  $T_{\rm m}$ , as may be seen in Fig. 7 (conditions L), lost to higher temperatures. Thus, mould temperature does not seem to affect the adhesion strength when higher melt temperatures are used.

Effect of the holding pressure  $(P_{\rm H})$ 

As can be observed in Fig. 8, increasing  $P_{\rm H}$  from 20 to 35 MPa decreases the value of  $w_{I}$ . Theoretically, it would be expected that for higher  $P_{\rm H}$  diffusion should be favoured increasing the adhesion strength [7], but that did not take place. Using the commercial software C-Mold 2000.7 the evolution of  $P_{\rm H}$  with time in the join zone was estimated (Fig. 9). For higher  $P_{\rm H}$  (conditions M3 and M4 vs. M1), the pressure is maintained locally effective for a longer time interval, even beyond the solidification of TPV. This situation may induce residual stress resulting from local

**Fig. 7**  $w_i$  variation with mould temperature  $(T_{mld})$ , grouped by  $T_i$  (L: 190 °C, M: 210 °C and H: 230 °C).  $P_{\rm H} = 35$  MPa. See Table 3 for codes



Fig. 8  $w_i$  variation with holding pressure ( $P_H$ ) grouped by  $T_i$  (L: 190 °C, M: 210 °C and H: 230 °C). See Table 3 for codes

molecular orientation and changing crystalline textures, which may explain inconsistencies in the theory [7].

Another aspect to consider is the high component paraffinic content, with very low molecular weight, in the TPV



Fig. 9 Estimated evolution with time of cavity pressure traces of TPV at the interface between the two plastic materials during the overmolding process

formulation. An increase on pressure could force higher diffusion of this component, increasing its concentration at the interface and hindering the adhesion between substrate and TPV [4]. However, it should be emphasized that  $w_{\rm I}$  values seem to be less affected by  $P_{\rm H}$  than by  $T_{\rm m}$ . These results agree with Bhatt and Extrand [20] who underlined the poor influence of the holding pressure on final properties of two overmoulded materials.

Effect of the injection rate  $(R_i)$ 

As shown in Fig. 10, as  $R_i$  is increased from 6 to 32 g/s,  $w_I$  takes its maximum value at 11 g/s. It may be attributed to the combined effect of two factors. On the one hand, with increasing injection rate the overmoulded material has less time to cool down before getting the surface of the



**Fig. 10**  $w_i$  variation with injection rate ( $R_i$ ), grouped by  $T_i$  (L: 190 °C, M: 210 °C and H: 230 °C).  $P_H = 35$  MPa. See Table 3 for codes

substrate. Therefore, an equivalent effect to higher melt temperature is achieved. On the other hand, this increase on  $R_i$  also reports an increase of the injection pressure which entails a decrease in  $w_I$  as it was pointed out in previous section (Fig. 9).

## Conclusions

The essential work of interfacial fracture (EWIF) based on the essential work of fracture concept, seems to be a powerful tool for quantitative studies dealing with adhesion strength in overmoulded (bi-material) parts. According to this concept and analysing the obtained results along this work, it could be stated that:

- The diffusion of the polymeric materials takes place as soon as the melt wets the substrate. Once the interface is formed by the intimate contact between both materials, the polymeric chains will spread through it and an entanglement between the chains of both materials will take place. After melt solidification, an interface with a gradient concentration of both materials will be obtained. Thus, for the diffusion to take place, it is necessary that join materials are compatible (similar  $\delta$  values). Nevertheless, low molecular additives (i.e. paraffin) in the TPV compound could migrate towards the interface and spread towards the PP, making difficult the adhesion between both materials, favoured by an increase on the holding pressure effectiveness ( $P_{\rm H}$ ).
- The adhesion strength between both materials seems to be more affected by the melt temperature than by the mould temperature due to the favoured interdiffusion.
- An increase on holding pressure beside favouring the low molecular weight additives, could promote residual stress in the interface, creating a loss in adhesion strength. However, its influence over adhesion strength seems to be insignificant.
- Increasing injection rate  $(R_i)$  seems to operate two opposed phenomena: higher melt temperature at the moment of contact with the substrate (favouring interdiffusion) and at the same time increasing the holding pressure effectiveness (favouring migration and residual stress).
- Finally, the solidification of the molten material takes place, when both materials are strongly adhered. This joint between both materials, as it has been demonstrated previously, occurs mainly due to the increase in the melt temperature of the TPV, and in lower degree, due to the effect of the mould temperature in some cases. In the present case, the  $R_i$  may have a double effect, in some cases generate an increase of the

adhesion and in others cases a diminution, whereas holding pressure does not significantly influence the adhesion between the TPV and the substrate of PP.

Acknowledgements Candal wishes to thank "Dirección de Desarrollo Profesoral" of the UNIVERSIDAD SIMÓN BOLÍVAR (USB) and the FONDO NACIONAL DE CIENCIA, TECNOLOGÍA E INVESTIGACIÓN (FONA-CIT) from Venezuela for the financial support for the trips to Barcelona, Spain, to work on her Ph.D. thesis at the UNIVERSITAT PO-LITÈCNICA DE CATALUNYA (UPC). She specially would like to thank institutional relations of the USB and UPC for exoneration of payment of her Ph.D. Authors would like to thank the "Decanato de Investigación y Desarrollo" from Universidad Simón Bolívar for generous funding through grant DID-GID-02. Also they thank the CENTRE CATALA DEL PLASTIC and the LABORATORY "E" (USB) for allowing them to use their facilities and equipments that made possible the development of this investigation.

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