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Weld lines in injection-moulded immiscible blends: model predictions and experimental results

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In this work, a theoretical approach based on diffusion and the Flory–Huggins theory of the free energy of mixing is considered to predict the weld-line strength in injection-moulded polycarbonate (PC) and its blend with high-density polyethylene (PE) as a function of melt temperature and contact time. A comparison between the model predictions and experimental results was also performed for pure PC as well as for its blend. Different melt temperatures ranging from 220 to 300°C and a constant total cooling time of 15 s were used. Two types of specimen were produced, one with a dual-gate mould cavity and another with a single gate. In the case of PC, the model predicts satisfactorily the weld-line strength as a function of injection temperature. High-density PE was also moulded in the same conditions but in a lower temperature range (145 to 220°C). The results showed that the tensile strength of PE remains unchanged in the presence of weld lines. For the PC/PE blend, the results showed a good agreement between the model predictions and the experimental results. It was also possible to predict the weld-line strength of the blend as a function of its composition and temperature.

(Keywords: immiscible blends; injection moulding; weld-line strength)

INTRODUCTION

Injection moulding is one of the most attractive polymer processes in industry owing to its high production rate, short cycle times and low percentage of scrap, with special mould design, and to the versatility of the moulds that can be used. In addition, this process is used for almost all polymeric materials with no serious restrictions regarding their properties[1,2]. However, moulding bulky parts and parts containing inserts may generate serious difficulties in terms of mould filling[3–6]. In fact, moulding of such pieces usually produces weld lines once the melt fronts have joined either by impingement flow or around an insert. Moulding bulky parts requires multi-gate moulds through which the polymer melt flows, and the mould is filled in a shorter time, avoiding short shots and increasing the production rate[3]. On the other hand, whenever separate flow fronts of a material in the liquid state meet, a contact area is formed after cooling or after a phase change (*Figure 1*). For polymeric materials, this area, known as a weld line or a knit line, can be visible to the naked eye in some cases and exhibits different properties and morphologies from the bulk. It is also observed in other processes such as extrusion[7]. In injection moulding, a flow pattern similar to that observed for extrusion occurs in the presence of inserts inside the mould. Weld lines can also be formed if jetting occurs inside the cavity[7,8].

The presence of weld lines has been observed to reduce the mechanical properties of mouldings of various materials[9–12]. The effect of processing parameters on the morphology and mechanical properties of particular polymers has already been studied[13–18]. They were found to play an important role in minimizing weld-line effects, among them the increase of melt and mould temperatures, screw back-up pressure and injection speed or enlarging the gate to reduce flow resistance.

In the case of multiphase systems, the presence of weld lines has an even larger impact on the properties of the

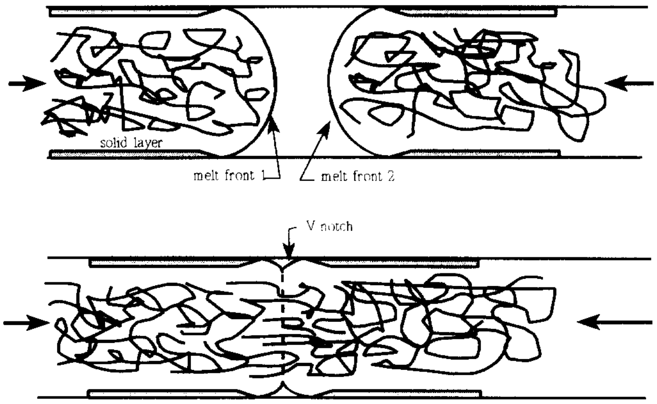

Figure 1 Formation of the weld line in injection moulding

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blends owing to the incompatibility of most polymers[19-21]. In addition, the morphology obtained near the weld line also has a great effect on the final properties of the blends[9,22-24].

From a theoretical standpoint a model based on the molecular theory of reptation was derived using Fick's law of diffusion combined with the Flory–Huggins free energy of mixing[25]. The model predictions for weld-line strength were compared to experimental results obtained for injection-moulded polystyrene bars[25]. However, for polymer blends, and to the best of our knowledge, no-one has reported a theoretical approach for the prediction of the strength of weld lines in injection moulding, especially in the case of immiscible blends, even though a lot of work has been reported for filled polymers and composites[26-29].

In this work, a new approach has been considered for the prediction of weld-line strength of immiscible systems in injection moulding. This approach takes into consideration three different diffusion coefficients corresponding to the welding in each phase of the blend. The original model has been used first to predict the weld-line strength of injection-moulded polycarbonate (PC). In a second step, the new model was used for the prediction of weld-line strength of polycarbonate and high-density polyethylene (PE) blend. In this case, the weld-line strength depends on the blend composition, injection temperature and contact time. The results of these predictions have been compared to experimental results obtained in the particular case of PC/PE 80/20.

THEORY

Kim and Suh have developed a model for the prediction of weld-line strength as a function of temperature and cooling time for glassy and amorphous polymers[25]. Assuming that diffusion at the knit line is only due to pure diffusion of the molecules across the interface, the strength of the weld line is given in terms of the degree of bonding defined as the ratio of the unbonded to the bonded areas at time t by:

$$\sigma_w = \sigma_b\left(\frac{A_0 - A_N}{A_0}\right) \tag{1}$$

where σ_w and σ_b represent the tensile strength of the moulded part with and without weld line respectively, A_N is the non-bonded area, and $A_0 - A_N$ is the bonded area at a specific time t.

The potential of adhesion by diffusion at a time t is represented by the free-energy difference ΔG between the intermediate adhesion step and perfect adhesion:

$$\Delta G = G_i - G_f \tag{2}$$

where G_i is the free energy at time t_i and G_f is the free energy for a complete adhesion (at t_f).

The rate of diffusion at the interface is given by Fick's law:

$$\frac{dA_N}{dt} = C\frac{D\Delta G}{kT} \tag{3}$$

where C is the limit of equation (3) when T tends towards T_g, D is the diffusion constant and k is the Boltzmann constant.

The free-energy difference ΔG can also be expressed in terms of the sum of the surface energy of the unbonded area and the configurational entropy of mixing:

$$\Delta G = -2\gamma A_N - T\Delta S_m \tag{4}$$

in which γ is the surface tension and ΔS_m is the entropy of mixing determined from the Flory–Huggins theory of lattices[30]. Combining the area ratio and Fick's law together with the entropy of mixing, we obtain the expression for the degree of bonding as a function of temperature and contact time. Assuming isothermal diffusion, one obtains[25]:

$$\frac{\sigma_w}{\sigma_b} = 1 - \exp\left(\frac{CD}{kT}\left[-2\gamma + kT\,\delta x_0\,\delta n_0 \ln(1/2)\right]t\right) \tag{5}$$

where δx_0 is the diffusion thickness and δn_0 the number of lattices per unit volume. The rate of diffusion of polymeric chains is estimated by the self-diffusion coefficient of the polymer. In the case of long chains, the segments of the chain jump from one position to another due to chain entanglements in the neighbourhood (reptation theory).

There exist several methods for measuring the self-diffusion constant of polymers at temperatures higher than the glass transition temperature[31]. Graessley[32] has analysed the Doi–Edwards theory and showed that the diffusion coefficient for self-diffusion of a polymer in the melt state can be estimated from viscoelastic functions:

$$D = \frac{G_0}{135}\left(\rho\frac{RT}{G_0}\right)^2\left(\frac{R_e^2}{M_w}\right)\left(\frac{M_{cr}}{M_w^2\eta_{0,cr}}\right) \tag{6}$$

where R_e^2/M_w is the ratio of the mean-square end-to-end distance to average molecular weight, $\eta_{0,cr}$ is the zero-shear viscosity at the critical molecular weight for entanglements, M_{cr} is the critical molecular weight, R is the universal gas constant, G_0 is the plateau modulus and ρ is the density.

In the case of immiscible polymer blends and depending on the viscosity ratio, the predominant morphology of the blend is characterized by spherical particles dispersed in a continuous phase for a low concentration of one of the phases. Experimentally, in the weld-line region, both melt fronts present a similar morphology with an equal ratio of particle surface to the cross-sectional area of the sample (1:10^6) and also a random distribution of the minor phase. In this case, the overall diffusion between the two fronts is essentially dependent on the individual diffusion in each phase. Statistically, all cases are possible, i.e. diffusion between PC and PC, PE and PE, and a heterogeneous diffusion between PC and PE. Based on these assumptions, it is possible to calculate, for surface element δs, the probability of having a contact between two identical phases of PC or PE and between two different phases PC–PE (*Figure 2*). For a weight fraction of polycarbonate of Φ_A and of polyethylene of Φ_B so that $\Phi_A + \Phi_B = 1$, the probability of having a PC–PC contact is Φ_A^2 and the probability of a PE–PE contact is Φ_B^2. Therefore the probability of a heterogeneous contact PC–PE will be $1 - \Phi_A^2 - \Phi_B^2$. In the particular case of the PC/PE 80/20 blend the probability of having each diffusion is 0.64 for PC–PC, 0.04 for PE–PE and 0.32 for PC–PE. In each of the PC and PE phases the rate of

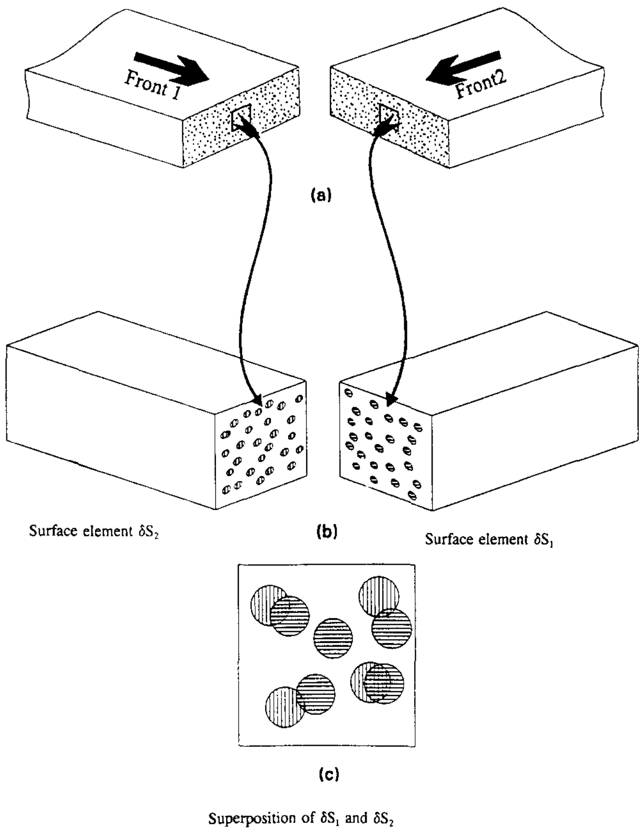

Figure 2 Melt front morphology of the PC/PE 80/20 blend

diffusion can be expressed in terms of:

$$\frac{\mathrm{d}A_{\mathrm{N}i}}{A_{\mathrm{N}i}} = -\frac{C_i D_i}{kT}[2\gamma_i - kT\,\delta x_{0i}\,\delta n_{0i}\ln(1/2)]\,\mathrm{d}t \tag{7}$$

For the heterogeneous contact PC–PE, the situation differs in the fact that the free energy in such a system at a specific time of diffusion is given by:

$$\Delta G_{\mathrm{AB}} = (\gamma_{\mathrm{A}} + \gamma_{\mathrm{B}})A_{\mathrm{NAB}} + \Delta G_{\mathrm{mix}} \tag{8}$$

where γ_{A} and γ_{B} are the surface tensions of phase A and phase B, A_{AB} being the non-bonded area in the PC–PE diffusion. ΔG_{mix} is the free energy of mixing of the two phases that have diffused into each other[30]:

$$\Delta G_{\mathrm{mix}} = RTV\left(\frac{\rho_{\mathrm{A}}}{M_{\mathrm{A}}}\phi'_{\mathrm{A}}\ln\phi'_{\mathrm{A}} + \frac{\rho_{\mathrm{B}}}{M_{\mathrm{B}}}\phi'_{\mathrm{B}}\ln\phi'_{\mathrm{B}} + \chi_{\mathrm{AB}}\right) \tag{9}$$

Here V is the total volume of diffusion, ρ_{A} and ρ_{B} are the densities and M_{A} and M_{B} are the average molecular weights of each phase respectively. χ_{AB} is the Flory–Huggins interaction parameter and ϕ'_{A} and ϕ'_{B} are the volume fraction of each phase A and B that have contributed to the diffusion in the PC–PE phase. Therefore, the free-energy term can be used to calculate the rate of diffusion in the PC–PE phase through:

$$\frac{\mathrm{d}A_{\mathrm{NAB}}}{A_{\mathrm{NAB}}} = -\frac{C_{\mathrm{AB}}D_{\mathrm{AB}}}{kT}\Bigg[\gamma_{\mathrm{A}} + \gamma_{\mathrm{B}} + RT\,\delta x_{0\mathrm{AB}} \times\left(\frac{\rho_{\mathrm{A}}}{M_{\mathrm{A}}}\phi'_{\mathrm{A}}\ln\phi'_{\mathrm{A}} + \frac{\rho_{\mathrm{B}}}{M_{\mathrm{B}}}\phi'_{\mathrm{B}}\ln\phi'_{\mathrm{B}} + \chi_{\mathrm{AB}}\right)\Bigg]\mathrm{d}t \tag{10}$$

Integrating equation (7) one obtains for PC and PE:

$$\frac{A_{\mathrm{N}i}}{A_{\mathrm{T}i}} = \exp\left(-\frac{C_i D_i}{kT}[2\gamma_i - kT\,\delta x_{0i}\,\delta n_{0i}\ln(1/2)]t\right) \tag{11}$$

For PC–PE diffusion the integration of equation (10) leads to:

$$\frac{A_{\mathrm{NAB}}}{A_{\mathrm{TAB}}} = \exp\left\{-\frac{C_{\mathrm{AB}}D_{\mathrm{AB}}}{kT}\Bigg[\gamma_{\mathrm{A}} + \gamma_{\mathrm{B}} + RT\,\delta x_{0\mathrm{AB}} \times\left(\frac{\rho_{\mathrm{A}}}{M_{\mathrm{A}}}\phi'_{\mathrm{A}}\ln\phi'_{\mathrm{A}} + \frac{\rho_{\mathrm{B}}}{M_{\mathrm{B}}}\phi'_{\mathrm{B}}\ln\phi'_{\mathrm{B}} + \chi_{\mathrm{AB}}\right)\Bigg]t\right\} \tag{12}$$

Knowing that the contact surfaces in each of the phases are:

$$\text{PC–PC} \qquad A_{\mathrm{AT}} = \phi_{\mathrm{A}}^2 A_{\mathrm{T}} \tag{13}$$

$$\text{PE–PE} \qquad A_{\mathrm{BT}} = \phi_{\mathrm{B}}^2 A_{\mathrm{T}} \tag{14}$$

$$\text{PC–PE} \qquad A_{\mathrm{ABT}} = (1 - \phi_{\mathrm{A}}^2 - \phi_{\mathrm{B}}^2)A_{\mathrm{T}} \tag{15}$$

and the total non-bonded area A_{NT} is:

$$A_{\mathrm{NT}} = A_{\mathrm{NA}} + A_{\mathrm{NB}} + A_{\mathrm{NAB}} \tag{16}$$

on integrating the ratio of the non-bonded area to the total area we obtain:

$$\begin{aligned}\frac{\sigma_{\mathrm{w}}}{\sigma_{\mathrm{b}}} = 1 &- \phi_{\mathrm{A}}^2\exp\left(\frac{-C_{\mathrm{A}}D_{\mathrm{A}}}{kT}[2\gamma_{\mathrm{A}} - kT\,\delta x_{0\mathrm{A}}\,\delta n_{0\mathrm{A}}\ln(1/2)]t\right)\\ &- \phi_{\mathrm{B}}^2\exp\left(\frac{-C_{\mathrm{B}}D_{\mathrm{B}}}{kT}[2\gamma_{\mathrm{B}} - kT\,\delta x_{0\mathrm{B}}\,\delta n_{0\mathrm{B}}\ln(1/2)]t\right)\\ &- (1 - \phi_{\mathrm{A}}^2 - \phi_{\mathrm{B}}^2)\exp\left\{\frac{-C_{\mathrm{AB}}D_{\mathrm{AB}}}{kT}(\gamma_{\mathrm{A}} + \gamma_{\mathrm{B}})\right.\\ &\left.+\left[RT\,\delta x_{0\mathrm{AB}}\left(\frac{\rho_{\mathrm{A}}}{M_{\mathrm{A}}}\phi'_{\mathrm{A}}\ln\phi'_{\mathrm{A}} + \frac{\rho_{\mathrm{B}}}{M_{\mathrm{B}}}\phi'_{\mathrm{B}}\ln\phi'_{\mathrm{B}} + \chi_{\mathrm{AB}}\right)\right]t\right\}\end{aligned} \tag{17}$$

The diffusion coefficient (D_{AB}) in the third term is defined by the following relation[33]:

$$D_{\mathrm{AB}} = \Phi(1 - \Phi)\Lambda_0\frac{N_{\mathrm{e}}}{N}\left(\frac{1}{N\Phi} + \frac{1}{N(1 - \Phi)} + 2\chi_{\mathrm{AB}}\right)kT \tag{18}$$

in which Φ represents the weight fraction of one of the components, polycarbonate in this case, Λ_0 is the mobility coefficient, N_{e} is the degree of polymerization for entanglements and N is the degree of polymerization. This model is valid if we assume that the diffusion occurs in the same medium[33]. It is based on the Doi and Edwards theory for displacement of long chains in reptation inside a tube and assumes the same degree of polymerization and molecular weights for both phases. In a more general case where the molecular weight of each component is higher than the critical molecular weight, the diffusion coefficient is given by[33]:

$$D_{\mathrm{AB}} = D_0\frac{\Phi_1\Phi_2}{\Phi_1 + f\Phi_2} \tag{19}$$

with:

$$D_0 = 2\chi_{12}kT\Lambda_{01}\frac{N_{\mathrm{e}}}{N_2} \tag{20}$$

and:

$$f = \frac{\Lambda_{02}}{\Lambda_{01}}\frac{N_1}{N_2} \tag{21}$$

Table 1 Some characteristics of the materials used

Materials	*MFI*[a] (g/10 min)	Density[a] (g cm^{-3})	Power-law index, *n*[b]	Water absorption[a] (%)
HDPE	0.3[c]	0.91–0.97	0.75	–
PC	11.5[d]	1.2	0.60	0.35[e]

[a] Supplied by manufacturer
[b] Measured at 270°C
[c] 190°C/5 kg
[d] ASTM D-1238
[e] ASTM D-570 (23°C/24 h)

where Λ_{01} and Λ_{02} are the mobility coefficient and N_1 and N_2 are the degree of polymerization of phase A and B respectively.

According to Rouse theory[30], the friction coefficient can be related to the viscosity by:

$$\xi_0 = \frac{1}{\Lambda_0} = \frac{36 M_0}{\rho R_e^2 N_A}\eta_{0\mathrm{cr}} \tag{22}$$

where ξ_0 is the friction coefficient, N_A is the Avogadro number, R_e^2 is the mean-square end-to-end distance by a unit monomer and M_0 the molar mass of a monomer unit. Introducing the friction coefficients in the diffusion equation it is then possible to calculate D_{AB} as a function of the critical viscosity.

EXPERIMENTAL

Materials

The materials used in this study are polycarbonate (PC, Lexan 141L-112) supplied by General Electric Co., USA, and high-density polyethylene (HDPE, 32060C) provided by Dow Chemicals, Canada. Some characteristics of these materials are reported in *Table 1*.

Blending

Polycarbonate was dried prior to blending at 120°C for 16 h to remove all the moisture. The composition prepared (80/20 by weight PC/HDPE) was selected from a previous systematic study on this particular blend owing to its appreciable properties compared to polycarbonate[34].

Blending of polycarbonate and polyethylene was carried out in a Werner & Pfleider twin-screw extruder at a screw speed of 250 rpm and a temperature profile ranging from 220°C near the hopper to 240°C at the die exit. The extrudate was then continuously cooled in a water trough, air dried and pelletized prior to injection moulding.

Injection moulding

All specimens were injection moulded using a Nissei fully hydraulic injection moulding machine (model PS-60E9ASE), with a maximum injection capacity of 114 cm^3/shot and a maximum clamping force of 62 tons ($\sim$550 kN). The moulding conditions were selected after a moulding area was established for both polycarbonate and polyethylene. The injection rate was fixed at 75 cm^3 s^{-1} and the mould temperature was kept at 30°C for all experiments. The injection temperature was varied from 220 to 300°C for polycarbonate and the blend, and from 145 to 220°C for polyethylene. A maximum injection pressure of 124.1 MPa was used and the packing pressure was kept at 55.1 MPa. The maximum injection time and screw speed were fixed at 10 s and 80 rmp respectively. All specimens had a cooling time of 15 s. The mould contained two ASTM standard dogbone-shaped cavities for mechanical testing for both welded and non-welded specimens similar to those shown in *Figure 3*.

Mechanical testing

The specimens were tested under the same conditions using an Instron tensile testing machine at a cross-head speed of 20 mm min^{-1} at room temperature, using an ASTM D-638 standard method.

Phase and surface morphology

Scanning electron microscopy investigation was carried out on the different blend specimens broken in liquid nitrogen at the weld line. The broken specimens were then covered with a gold–palladium alloy prior to the investigation. The equipment used was a JEOL JSM scanning electron microscope model 25S-III. The investigation was mainly focused on the whole cross-sections of the specimen in order to examine the difference between the skin and the core, and also on the core of the specimen moulded at 250 and 300°C in the presence of weld lines. Micrographs were also taken on the top surfaces of the weld lines for PC, PE and PC/PE 80/20 moulded at 220, 145 and 220°C respectively.

MODEL PREDICTIONS

Homopolymers: PC

Figure 4 shows the predictions for the weld-line degree of bonding as a function of contact time at different injection temperatures for pure polycarbonate having an average molecular weight M_w of 30 000 g mol^{-1} and a critical molecular weight M_{cr} of 3000 g mol^{-1}. According to this graph a total bonding can be achieved at 300°C within 10 s. However, below 220°C there is almost no diffusion and the degree of bonding is almost zero.

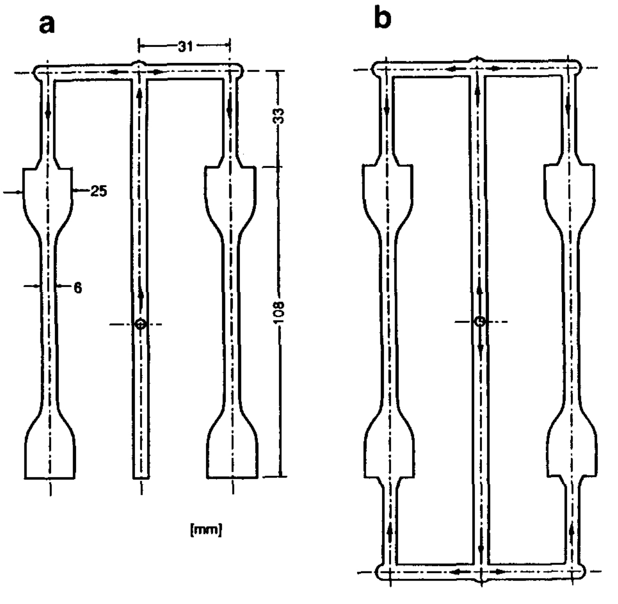

Figure 3 Mould cavity used for the study of weld lines: (a) no weld lines; (b) with weld lines

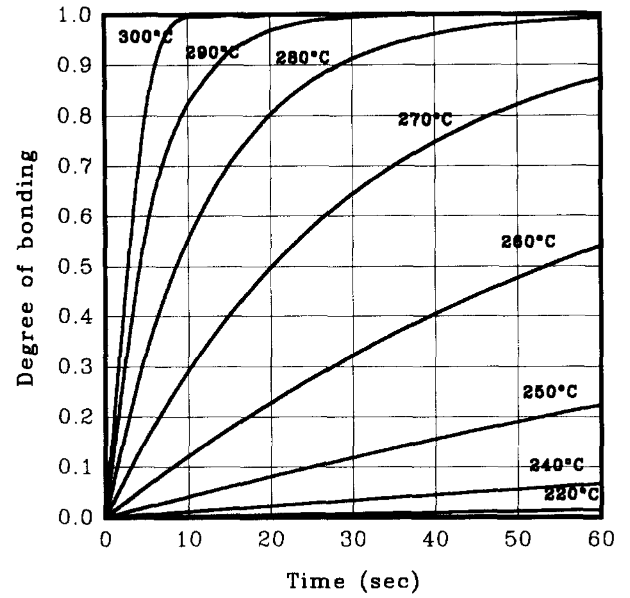

Figure 4 Theoretical predictions for the degree of bonding of polycarbonate as a function of contact time and injection temperature

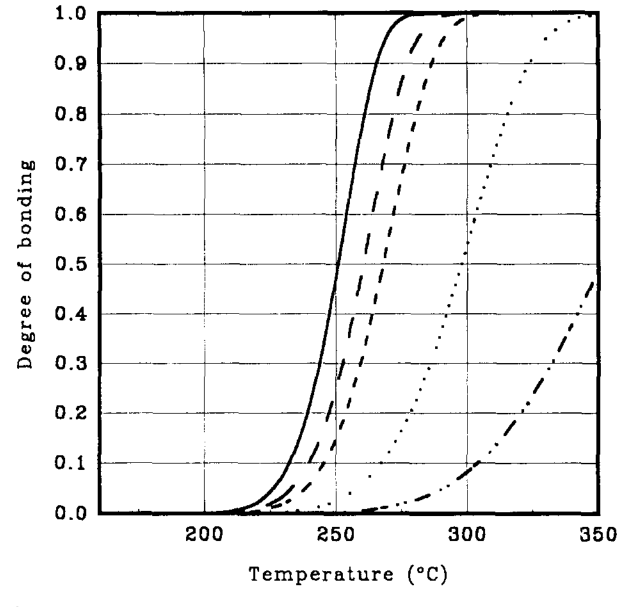

Figure 5 Theoretical predictions of the degree of bonding of polycarbonate as a function of temperature: effect of average molecular weight, $M_w = 2 \times 10^4$ (——), 3×10^4 (— —), 4×10^4 (– – – –), 1×10^5 (········) and 3×10^5 g mol^{-1} (—··—)

Therefore, the degree of bonding varies between almost zero and unity for a temperature range of 220 to 300°C. On the other hand, the temperature considered here is that of the melt fronts, which is lower than the injection temperature owing to the heat exchange with the cold mould. Based on these considerations, we have fixed the total cooling time at 15 s for the whole range of injection temperature.

Effect of molecular weight M$_w$. The effect of molecular weight (M_w) on the degree of bonding of polycarbonate is shown in *Figure 5*. An increase in the average molecular weight leads to a reduction in the degree of bonding owing to the higher viscosity and lower diffusion across the weld line. In fact, at 250°C the degree of bonding decreases from 0.5 to 0.15 for an increase of the molecular weight by twofold (i.e. 2×10^4 to 4×10^4). This is in good agreement with the physics of diffusion. According to equation (6), the diffusion coefficient decreases with an increase of the molecular weight. It is therefore better to use a low-molecular-weight polycarbonate grade for an easier processing at low temperatures and a better diffusion.

Effect of critical molecular weight M$_{cr}$. We now consider the effect of the critical molecular weight (M_{cr}) on the degree of bonding; three values were used in addition to a few arbitrary values. Mercier *et al.*[35] and Seitz[36] have calculated a M_{cr} of 2400 g mol^{-1}, whereas Wu[37] has found a value of 3560 g mol^{-1} compared to that reported by Van Krevelen[38] of 3000 g mol^{-1}. In all cases, the values used for the illustration are still lower than the average molecular weight (30 000 g mol^{-1}). Therefore, for $M_{cr} < M_w$, the variation of the plateau viscosity with the average molecular weight is linear and equation (A.3) reported in Appendix 2 applies with exponent 1.0 instead of 3.4. Theoretically, the critical molecular weight is a molecular property of the material; hence, it is supposed to be constant for the same material (in our case polycarbonate of bisphenol A type). It is therefore interesting to study how sensitive is the degree of bonding of polycarbonate to this parameter. The results reported in *Figure 6* show that this parameter has an appreciable effect and should be as accurate as possible in order to predict with accuracy the degree of bonding, especially in the low-temperature region (<280°C).

Considering polyethylene, the situation becomes different because one needs to consider the crystallization effect. The results are not shown for the sake of simplicity. The theoretical model applied to PE has shown a similar behaviour as for PC with a shift in temperature towards the crystallization temperature of PE and with complete bonding around 200°C.

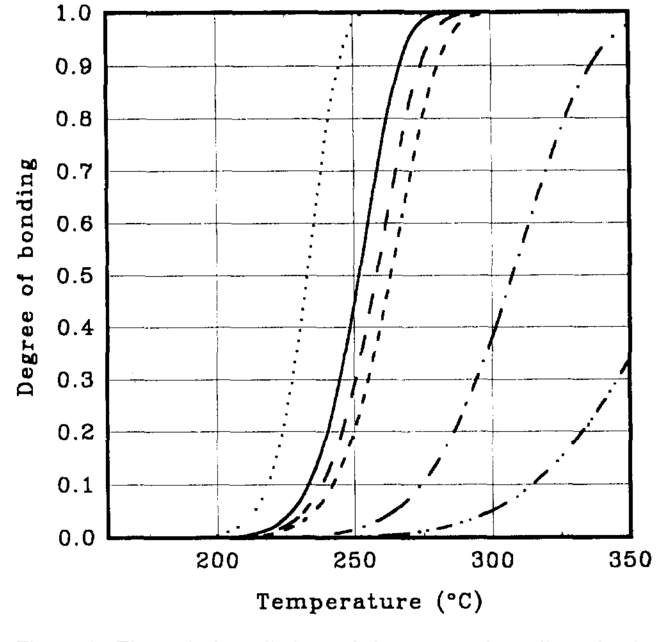

Figure 6 Theoretical predictions of the degree of bonding of polycarbonate as a function of temperature: effect of critical molecular weight, $M_{cr} = 1 \times 10^3$ (········), 2.4×10^3 (——), 3×10^3 (— —), 3.56×10^3 (– – – –), 1×10^4 (—·—) and 2.5×10^4 g mol^{-1} (—··—)

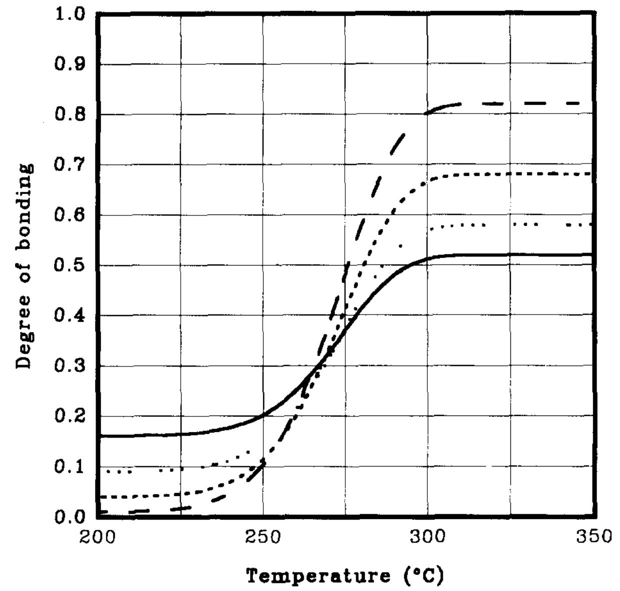

Figure 7 Theoretical predictions of the weld-line strength of PC/PE blend as a function of temperature: effect of blend composition, PC/PE 90/10 (— —), 80/20 (- - - -), 70/30 (······) and 60/40 (—)

Immiscible blends: PC/PE

Effect of blend composition. It is possible to predict the weld-line strength expressed by the degree of bonding for the blend at different composition. Model predictions obtained using equations (17) and (22) are shown in *Figure 7* for different compositions. Model parameters are those of PC and PE. For these compositions, we have obtained a morphology where polyethylene is dispersed as particles in the polycarbonate matrix; this is valid for compositions lower than 30% by weight^{24}. In general, the degree of bonding increases with temperature for all blend compositions. For temperatures lower than 250°C, the addition of polyethylene to polycarbonate leads to an increase of the degree of bonding with the PE concentration. This effect is expected since, in this temperature range, only polyethylene contributes to the adhesion of the melt fronts. For temperatures higher than 275°C, the contribution of polycarbonate becomes more predominant with increasing temperature and decreasing polyethylene content. We also notice that the model predicts a maximum degree of bonding for temperatures higher than 300°C. This maximum becomes higher when the PE content is lowered. A similar behaviour is observed at temperatures lower than 225°C. However, in this temperature range, we should reach a zero degree of bonding close to the melting point of polyethylene. This region has no practical use in our case.

In summary, these results are based on theoretical calculations using data available in the open literature and are reported in Appendix 2. In the case of pure PC, the results were found to agree with those reported by Kim and Suh^{25} for injection-moulded polystyrene in the presence of weld lines.

EXPERIMENTAL RESULTS

The moulding area was defined by short shots and flushing in terms of injection rate, melting and/or softening point of the resins and thermal degradation for the injection temperature, and, for mould temperature, between room temperature and a higher temperature for easy ejection, and low warpage.

Homopolymers (PC and PE)

Figure 8 shows the degree of bonding as a function of injection temperature for polycarbonate and the blend. The results show that the degree of bonding is strongly dependent on temperature. The degree of bonding characterized by the ratio of the tensile strength of the welded specimen to that of the non-welded specimen increases with temperature for both materials.

For polyethylene, the experimental results obtained were not surprising. In fact, the samples could not be broken even at high extension rates, which is in agreement with the results reported by other authors for semi-crystalline materials^{9,11,12,22,39}. This was attributed to the fact that, within the temperature range considered, the viscosity of polyethylene is very low and therefore the diffusion at the interface is favoured.

Immiscible blends (PC/PE)

For the PC/PE 80/20 blend, the presence of polyethylene in the PC matrix reduces the degree of bonding for the whole experimental temperature range. This is due to the incompatibility of the two components on the one hand, and to the nature of the interface, which exhibits a poor adhesion between the two phases, on the other hand. However, the degree of bonding increases on increasing the injection temperature between 220 and 300°C. For the high moulding temperature, the injected volume at the screw tip was corrected in order to avoid flushing of the material between the mould plates.

Phase and surface morphology

Figures 9a, 9b and *9c* show the morphology of the surface near the weld line for polycarbonate (PC), polyethylene (PE) and the PC/PE 80/20 blend respectively. These micrographs were taken on samples moulded at the lowest injection temperature in each case. Qualitatively,

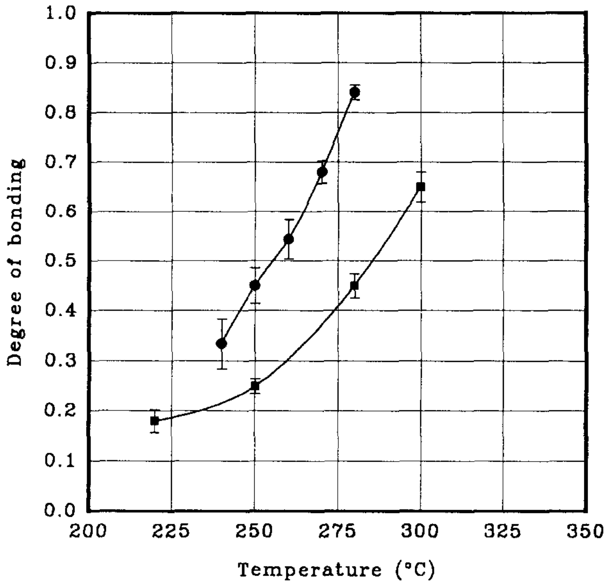

Figure 8 Experimental results for the weld-line strength of polycarbonate (●) and PC/PE 80/20 blend (■) as a function of injection temperature

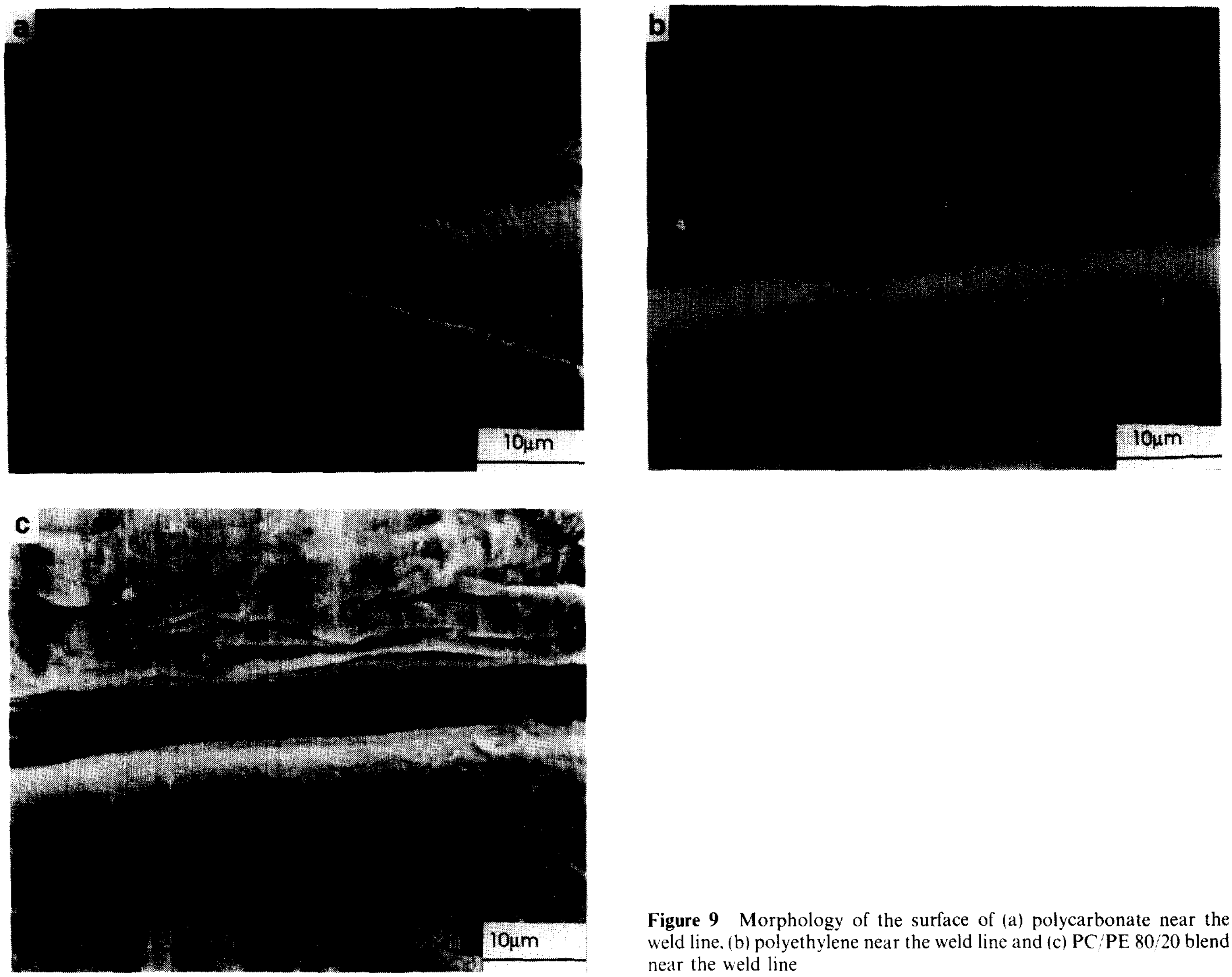

Figure 9 Morphology of the surface of (a) polycarbonate near the weld line, (b) polyethylene near the weld line and (c) PC/PE 80/20 blend near the weld line

for polycarbonate, the amplitude of the notch present at the interface seems to be less important than that observed for PE. The presence of the notch across the sample is only due to the air trapped during cavity filling, and can be partially eliminated by providing venting close to the weld line[40]. For the PC/PE 80/20 blend, it is clear that the nature of the interface is different from those observed for the homopolymers. In fact, a deeper notch is observed. In addition, an incomplete adhesion of one of the phases is observed.

Effect of injection temperature. The morphology of the blend observed perpendicular to the surface of the interface is shown in *Figures 10a–d* at the extreme injection temperatures respectively (250 and 300°C). These micrographs show the difference between the core and the skin of the samples as a function of temperature. For the former, the morphology is quite the same except perhaps for a larger particle size of the dispersed phase with increasing temperature, indicating significant effects of coalescence. For the skin, it can be observed that the outer layer close to the cavity wall can be thinner or thicker whether the temperature is increased or decreased. These micrographs indicate, however, that the core region is more important than the skin region. Moreover, the core region is predominantly spherical, which corresponds to the assumption used in the model.

Comparison with model predictions

Homopolymers: PC. The mechanical properties of polycarbonate characterized by the predicted degree of bonding as a function of injection temperature are compared to those obtained experimentally in *Figure 11*. The predicted values were determined by the diffusion model given by equation (5). The results show that a total adhesion can be obtained at an injection temperature of 300°C within 15 s of contact time. This means that the molecular chains have sufficient energy to cross the interface. However, below 150°C, which is close to the glass transition temperature of PC (154°C) measured by d.s.c.[41], there is no physical diffusion and the degree of bonding is assumed to be zero. Therefore it is apparent that the degree of bonding varies between zero and unity for a temperature ranging from T_g to 300°C depending upon the processing conditions such as mould temperature and injection speed. However, it is not necessary to study the effect of each parameter individually since they all have a direct effect on the contact time. The predictions are in good agreement with the experimental data, with an overestimation of the degree of bonding at high

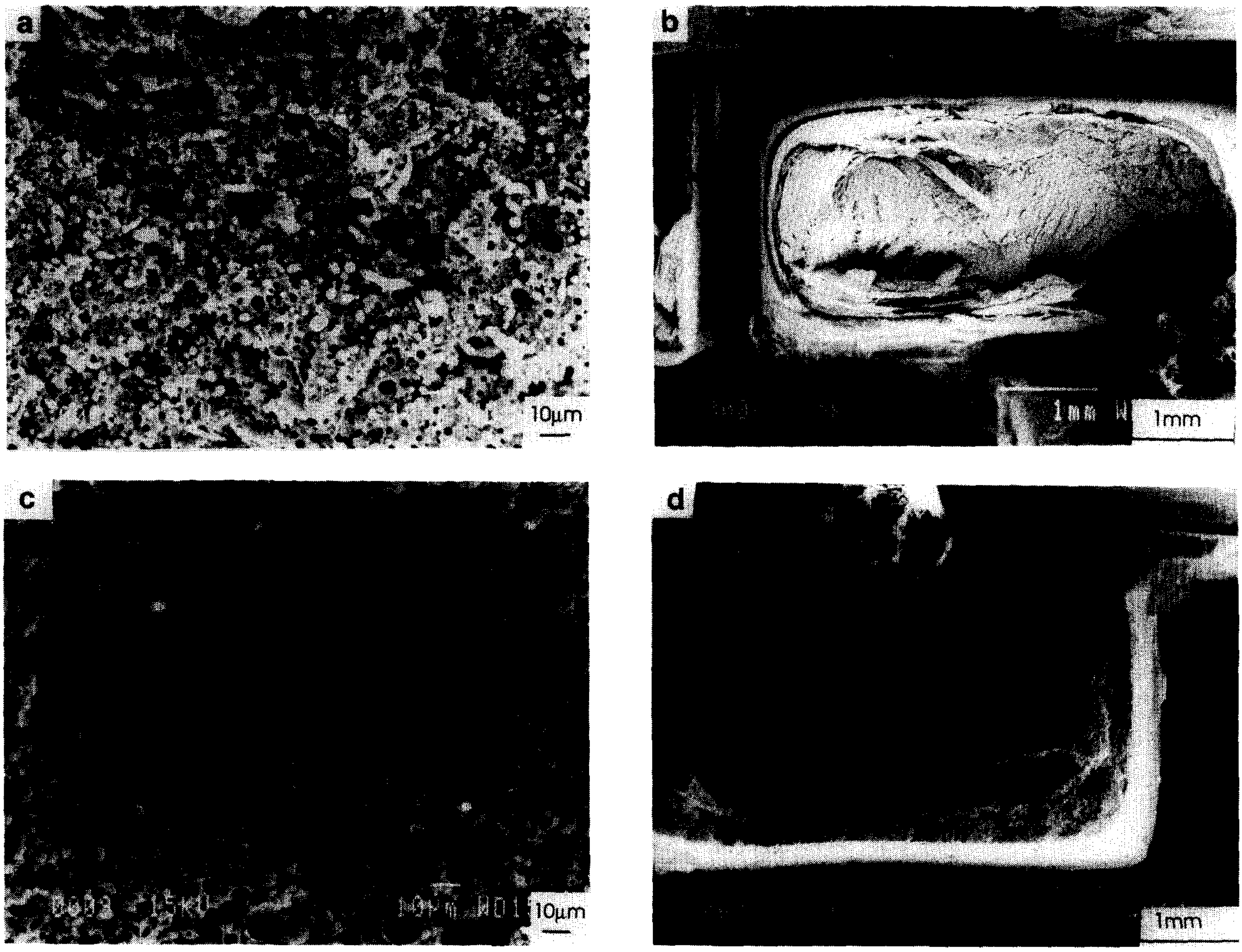

Figure 10 Morphology of the PC/PE 80/20 (a) at the core at 250°C, (b) at the skin at 250°C, (c) at the core at 300°C and (d) at the skin at 300°C

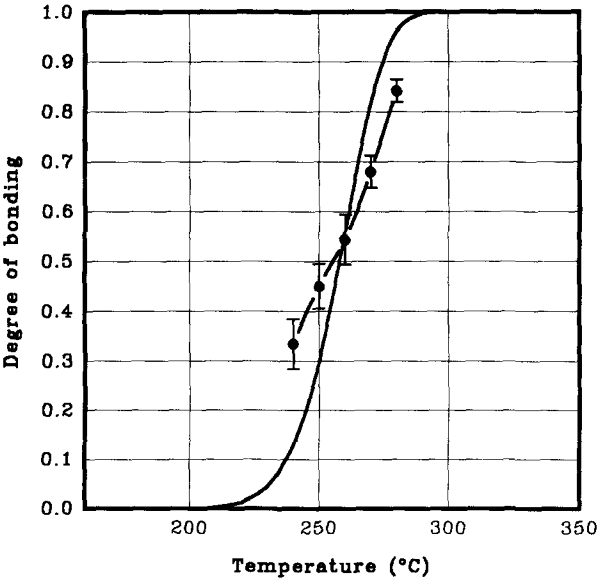

Figure 11 Comparison between the model predictions (——) and the experimental data (—●—) of the weld-line strength of polycarbonate

temperatures. Taking into consideration the approximate values of the plateau viscosity, the plateau modulus and the critical viscosity, it can be concluded that the superposition is satisfactory especially if we consider the standard deviations of the experimental data. In addition, it was assumed that the adhesion at the interface is only due to pure diffusion (Fick's law), whereas the presence of the V-notch as well as the effect of the molecular orientation were not considered.

Immiscible blends: PC/PE. For the particular case of the PC/PE 80/20 blend, the superposition of the theoretical predictions of the weld-line strength with the experimental data is reported in *Figure 12*. The Flory–Huggins interaction parameter was fixed to 0.5, which is comparable to the values reported in the literature for immiscible systems[42]. Therefore, the superposition of the model predictions with the experimental data relies essentially on one adjusting constant C_{AB} since C_A and C_B used are those determined from the superposition of the model predictions and experimental results of the homopolymers. *Figure 12* shows that a very good

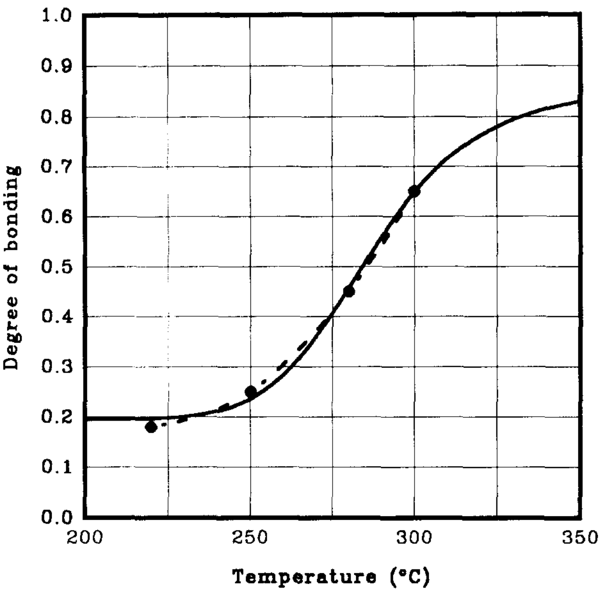

Figure 12 Comparison of the model predictions (——) and the experimental data (– – ● – –) of the weld-line strength of PC/PE 80/20 blend

agreement is obtained between the predicted and the measured weld-line strength in the case of PC/PE 80/20 blend. However, for a different system containing both phases with a degree of bonding lower than unity, a third constant C_B should be calculated based on the experimental data. Several other features of the weld line in the case of immiscible blends can be obtained from the model.

CONCLUSIONS

A model based on the lattice theory of Flory has been used to predict the weld-line strength of injection-moulded parts. This model is expressed in terms of a diffusion coefficient, on the one hand, and processing conditions (temperature and contact time), on the other hand.

In the case of the homopolymers and particularly for polycarbonate, the results are in good agreement with those reported in the literature. The superposition of the model predictions with the experimental data was found to be satisfactory, whereas it was not possible to predict the weld-line strength in the case of polyethylene owing to the fast crystallization during cooling and to the fact that it was not possible to break it even at high testing speeds.

For immiscible systems such as that of PC/PE, the model has been modified for a multiphase system by introducing additional terms representing the free energy of each phase and that of the blend. It was possible to study the effect of blend composition and to investigate the contribution of each phase to the degree of bonding. This model was shown to predict satisfactorily the degree of bonding in the particular system of PC/PE 80/20 in a temperature range of 220 to 300°C.

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APPENDIX 1

Nomenclature

A_N	non-bonded area
A_0	cross-sectional area
A_{nt}	total non-bonded area
C	constant
C_A	constant
C_{AB}	constant
C_B	constant
D	diffusion coefficient
D_A	diffusion coefficient in phase A
D_{AB}	diffusion coefficient of phase A in phase B
D_B	diffusion coefficient in phase B
G_0	plateau modulus
G_f	final free energy
G_i	initial free energy
k	Boltzmann constant
MFI	melt flow index
M_A	average molecular weight of phase A
M_B	average molecular weight of phase B
M_{cr}	critical molecular weight
M_e	critical molecular weight for entanglements
M_w	average molecular weight
N	degree of polymerization
N_e	degree of polymerization for entanglements
R	universal gas constant
R_e	end-to-end distance
t	time
T	temperature
T_{cr}	critical temperature (1000 K)
T_g	glass transition temperature
V	molar volume
γ	surface tension
γ_0	surface tension at 0 K
γ_A	surface tension of phase A
γ_B	surface tension of phase B
ΔE	activation energy
ΔG	free-energy difference
δn_0	number of lattices per unit volume
ΔS_m	entropy of mixing
δs	surface element
δx_0	diffusion thickness
η_{0cr}	zero-shear viscosity at critical molecular weight for entanglements
η_{Tg}	viscosity at glass transition temperature
Λ_0	mobility coefficient
ξ_A	friction coefficient of phase A
ξ_B	friction coefficient of phase B
ρ	density
ρ_A	density of phase A
ρ_B	density of phase B
σ_b	tensile strength of the bulk material
σ_w	tensile strength of the welded specimen
Φ	weight fraction of one of the components
ϕ'_A	volume fraction of phase A that has contributed to mixing
ϕ'_B	volume fraction of phase B that has contributed to mixing
ϕ_A	volume fraction of phase A
ϕ_B	volume fraction of phase B
χ_{AB}	Flory–Huggins interaction parameter

APPENDIX 2

Reported predictions

For the theoretical prediction, it might be useful to show that the physical parameters that are temperature-dependent (viscosity, surface tension and diffusion coefficient) have been estimated using theoretical and experimental predictions. The variation of viscosity as a function of temperature was estimated from the Williams–Landel–Ferry (WLF) model for temperatures $T_g < T < T_g + 100$, and from the Arrhenius model for temperatures $T > T_g + 100$, using:

$$\text{WLF} \qquad \log \eta_0 = \log \eta_{Tg} - \frac{17.44(T-T_g)}{51.6+(T-T_g)} \qquad \text{(A.1)}$$

$$\text{Arrhenius} \qquad \eta_0 = \eta_\infty \exp\left(\frac{\Delta E}{RT}\right) \qquad \text{(A.2)}$$

ΔE is the activation energy. The calculated viscosities were then used to estimate the critical viscosity, which in turn depends on the average molecular weight and the critical molecular weight using the following equation:

$$\eta_{0,cr} = \eta_0 \left(\frac{M_{cr}}{M_w}\right)^{3.4} \qquad \text{(A.3)}$$

On the other hand the prediction of the surface tension as a function of temperature has been determined from an experimental model given by equation (10)[43]:

$$\gamma = \gamma_0 \left(1 - \frac{T}{T_{cr}}\right)^{11/9} \qquad \text{(A.4)}$$

where T_{cr} is the critical temperature (1000 K for most polymers), and γ_0 is the surface tension at 0 K. The self-diffusion coefficient is also dependent on temperature and on other viscoelastic properties such as the plateau modulus G_0. This factor is considered as a constant even if it has a temperature dependence:

$$G_0 = \rho \frac{RT}{M_e} \qquad \text{(A.5)}$$

with:

$$M_e = 2M_{cr} \qquad \text{(A.6)}$$

The variation of the plateau modulus with temperature has been found to be around 12% by Zoller[44], who measured the variation of the specific volume of polycarbonate with temperature at different pressures. Therefore, the plateau modulus can be considered as constant.