Weld-Line Sensitivity of Injected Amorphous Polymers

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ABSTRACT: The effects of the processing parameters on the weld-line mechanical properties of polystyrene (PS) and polycarbonate (PC) were investigated. PS was very sensitive to the presence of a weld line, showing property reductions of up to 70%. However, this sensitivity was mainly connected to the surface notch at the weld line. When this notch was removed, behavior close to that of unwelded specimens was obtained. The injection temperature was the main processing parameter because it affected the macromolecular diffusion speed and, therefore, influenced the weld quality. A direct relationship between the distance of molecular diffusion and the fracture mechanism was established. PC had a low weld-line sensitivity, despite being an amorphous polymer like PS. The difference between these materials was connected to the different sizes of the surface defects and to the different entanglement densities, which influenced the relaxation time and the global behavior (brittle–ductile). © 2004 Wiley Periodicals, Inc. J Appl Polym Sci 93: 644–650, 2004

Key words: diffusion; mechanical properties; polycarbonates; polystyrene

INTRODUCTION

Injection molding is one of the most common ways of processing thermoplastic polymers. Very versatile, this method is suitable for the mass production of tiny parts as well as large elements, such as watch components and car instrument panels. The complexity or size of the parts can require the use of inserts or multigated molds. Unfortunately, these elements generate an important defect, called a weld line or knit line, due to the splitting and recombination of the melt flow. The result is a serious reduction in the mechanical properties of many materials.

Weld lines are complex features that are influenced by many parameters, such as the material (e.g., viscosity and crystallinity), the processing conditions (e.g., temperature and pressure), and the tooling (e.g., mold roughness and venting). This problem is not specific but can appear in most polymers, amorphous or semicrystalline, neat, blended, or reinforced.[1–4] Moreover, the always increasing aesthetic requirements are turning weld lines into undesirable surface defects.

The origin of the significant weakening of many materials at the weld line is mainly related to two factors: the intrinsic weld quality and the surface flaw (notch or groove).

The weld quality depends on the way in which the flow fronts meet and bond to each other. If the processing conditions are adequate, the macromolecular chains diffuse through the original interface and give the weld a strength close to that of the bulk (cohesive bond). If the conditions are not adequate (e.g., the temperature is too low), the diffusion is not sufficient for good healing of the interface (adhesive bonding), and this leads to a dramatic reduction in the mechanical properties. The intrinsic properties of the polymer (e.g., molecular weight) influence this aspect through the coefficient of diffusion and the relaxation time. Some fillers also play an important role. For example, weld lines in fiber-reinforced polymers are very harmful to the mechanical properties because fibers do not easily cross the interface and remain oriented in the plane of the weld.[4]

The surface defect associated with weld lines has two origins: an incompletely filled cavity and differential shrinkage during solidification and cooling. For Piccarolo and Saiu,[5] the unfilled mold is the dominant mechanism explaining the notch on the surface of amorphous polymers. They did not observe mold machining marks in the weld-line notch on polystyrene (PS); this means that the material solidified before reaching the mold surface. As a result, parameters acting on the flow front (e.g., mold temperature and injection speed) can modify the notch size. In the case of a semicrystalline polymer, the surface notch or bump is attributed to differential shrinkage during solidification due to a different microstructure between the weld area and the rest of the part.[6] Machin-

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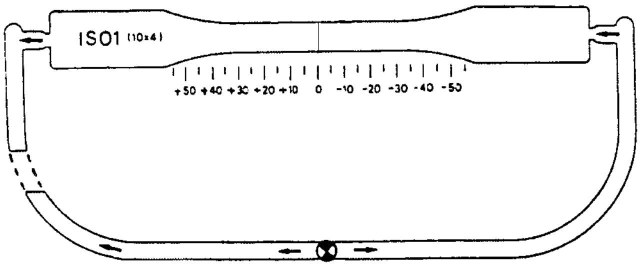

Figure 1 Mold layout (4 mm thick).

ing marks were observed in the weld-line notch on polyamide 6, and this means that the melt first completely filled the cavity and then shrank.

The influence of a weld line on PS, a typical brittle and amorphous polymer, has been widely studied.[3,5,7–12] Globally, it has been shown that the presence of a weld line dramatically reduces material properties, such as the stress and strain at break or fracture toughness (reduction up to 70%). Processing parameters generally have a limited influence on the mechanical properties of the weld line, except for the injection temperature and, to a lesser extent, the mold temperature and injection speed. Piccarolo and Saiu[7] did not notice any relationship between the strength and processing parameters. They explained this situation by the flaw generated at the surface of the weld line, which controls the failure.

Investigations performed on polycarbonate (PC), a ductile and amorphous polymer, showed a behavior very different from that of PS. The presence of a weld line does not influence properties such as the yield stress, Young's modulus, or fracture toughness.[1,11,12] However, properties at break are very sensitive to it. Criens and Mosle[11] reported an approximately 50% reduction in these properties. Tomari et al.[12] measured decreases in the stress and strain at break of about 27 and 50%, respectively. No influence of the processing conditions on this reduction has been reported. Criens et al.[11] showed that the processing temperature is not a significant parameter for temperatures above 300°C. This loss of properties is due to the initiation of a crack at the weld line that stops the necking phenomenon and leads to failure. Considering this mode of failure, Haufe et al.[13] showed that, for statistical reasons, the average elongation at break is only 3/8 of that of specimens without weld lines. The values are strongly scattered between 0 and 0.5 times the elongation of the reference specimen.

In this article, the effects of some processing parameters on the mechanical properties of PS are succinctly presented as a starting point for a detailed analysis of the two main features controlling the weld-line strength (i.e., surface geometry and processing temperature). An approach based on the calculation of the molecular diffusion length at the weld line is used to explain the fracture mechanisms. Finally, the behavior of PC is briefly contrasted to that of PS. Conclusions drawn from the investigation of PS are used to introduce hypotheses explaining the differences between these two amorphous polymers.

EXPERIMENTAL

Two materials were used for this investigation: PS supplied by BP Chemicals (HH 111) and PC from Bayer (Makrolon 2405). Dog-bone ISO 1 specimens were molded with a Billion H280/90 injection-molding machine (Oyonnax, France). Modular equipment was used to choose between single-gated and double-gated injection (Fig. 1). Thus, specimens with and without weld lines were produced.

Injected specimens were characterized in tension and bending (standards NF T 51-034 and 001) with an Instron 1185 test machine at a crosshead speed of 5 mm/min. The impact toughness was determined with Charpy tests. These tests were performed on a Zwick 5101 impact device loaded with a 7.5-J pendulum (standard NF T 51-035) (Ulm, Germany). The central part of the ISO 1 specimens was used for bending and Charpy tests. All the mechanical tests were performed at room temperature (23°C). The fracture surfaces were observed with a Philips 505 scanning electron microscope.

The most significant processing parameters reported in the literature[8–11] were considered in this study. For PS, they were the injection and mold temperatures and the injection speed. The influence of these parameters on the weld-line strength was studied with a two-level Taguchi methodology (Fig. 2). Constant processing parameters were used with PC because of their limited influence on the weld-line strength reported in the literature. An injection temperature of 320°C and a mold temperature of 80°C were used in agreement with the specifications of the material supplier.

To highlight the notch effect on PS and the real influence of some processing parameters, we performed tensile tests on specimens for which the notch had been ground off with sand paper. Charpy tests

	Level 0	Level 1
Injection speed	8 mm/s	32 mm/s
Mold temperature	40°C	60°C
Injection temperature	210°C	290°C
Holding time	15 s	
Holding pressure	50 bars	

Figure 2 Processing parameters used for the Taguchi analysis of PS.

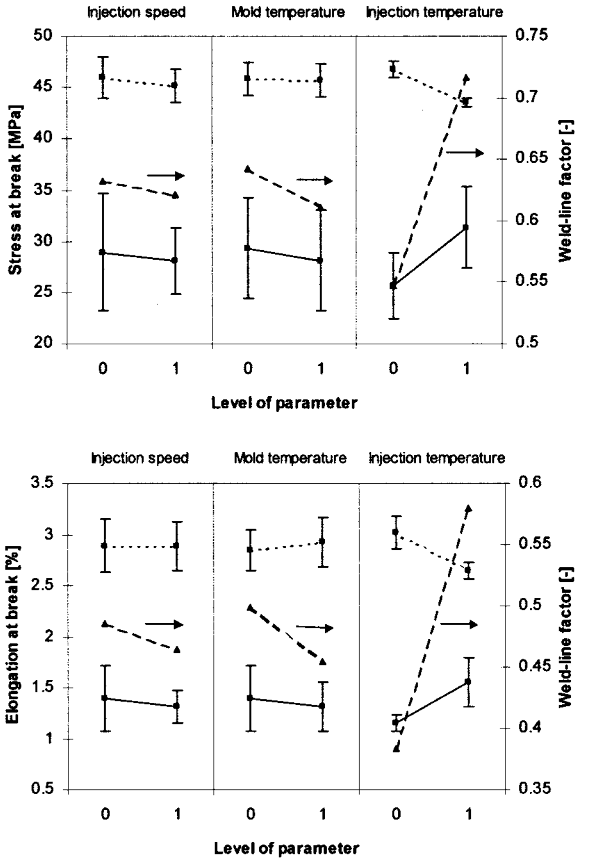

Figure 3 Effects of the processing parameters on the tensile properties of PS: (—) with a weld line, (· · ·) without a weld line, and (- - -) weld-line factor.

were also carried out on specimens without weld lines but with an artificial notch on each face made with a razor blade or a milling machine (bit diameter = 0.25 mm)

RESULTS AND DISCUSSION

PS

Mechanical properties and processing parameters

Figure 3 shows the influence of the processing parameters on the tensile properties (two-level Taguchi analysis). The weld-line factor is the ratio of the property magnitude when a weld line is present to that without a weld line. The results for the bending tests are not reported here because they appeared identical to the ones for tension. We can see that the presence of a weld line considerably weakens the strength of the material, with reductions in properties of up to 60% (70% for bending) in comparison with specimens without a weld line.

A statistical analysis of the tests has shown that the injection temperature has a significant influence on the weld-line strength, whereas the mold temperature and injection speed do not. These aspects are discussed later.

Finally, it has been observed that the failure always occurs at the weld line for all processing conditions and test methods. The surface defect (notch) acts as an initiation site for the damage.

Notch effect

Because of the important role of weld lines in the initiation of damage leading to failure, the influence of the surface notch has been studied in detail.

First, it appears that the notch width does not influence the mechanical properties. As reported by Piccarolo and Saiu[5] and Debondue,[14] the mold temperature and injection speed are the most important parameters influencing the notch width of PS and high-impact PS, respectively. The aforementioned results show that these parameters do not have a significant influence on the strength of the weld line even though the width can strongly vary with the different processing conditions used in this study. Therefore, the behavior is due to the defect generated by the notch, not to its macroscopic size. After initiation, the crack propagates through the plane of the weld, the strength of which depends on the injection temperature. This is why the mechanical properties are temperature-dependent, as reported previously.

The elimination of the notch brings a 25% improvement in the tensile strength (Fig. 4). The use of a high injection temperature (290°C) gives the part a strength close to that of a specimen without a weld line. In this case, failures outside the weld area occur. The global tensile behavior also changes. The material is still brittle, but a loss of linearity can be seen as in specimens without a weld line. This is due to the crazing of the material observed during the tests. At a lower temperature or when the notch is not removed, the behavior

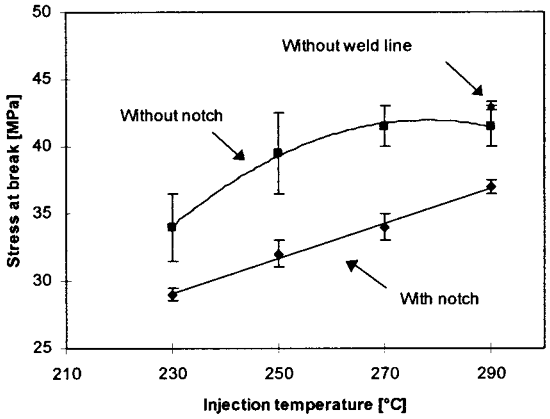

Figure 4 Evolution of the tensile strength when the weld-line notch is eliminated (PS).

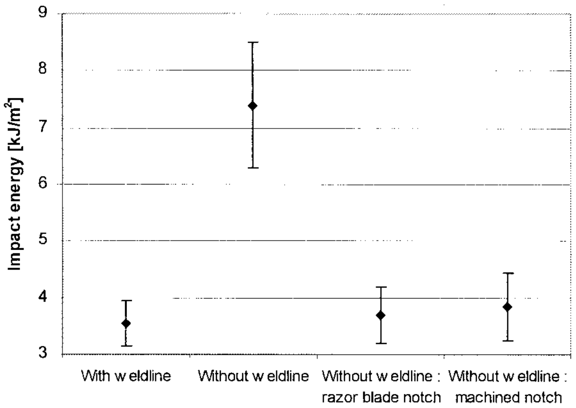

Figure 5 Impact toughness of PS as a function of the notch type (injection temperature = 290°C).

is purely brittle without a loss of linearity (failure at the weld line).

At a high injection temperature, an artificial notch on a specimen without a weld line has a deleterious effect: there is a 50% decrease in the impact toughness (Fig. 5). The loss of properties is similar to that shown by specimens with a weld line, and it is not sensitive to the notch radius (razor blade or milling machine). This observation explains the lack of a correlation between the strength and width of the weld line.

Effect of the injection temperature

Figure 4 shows that both the surface notch and processing temperature influence the strength. In this section, the effect of the temperature is analyzed.

A study of the fracture surfaces at different injection temperatures reveals very different morphologies. Figure 6 shows the evolution of the fracture surface of specimens for which the notch has been eliminated. At low temperatures, the surface has a rough aspect due to a quick propagation of the crack. A similar aspect has been observed for all the specimens when the notch has not been removed. When the temperature is increased, a smooth, mirrorlike area appears in the center of the specimen and progressively covers the whole surface. At high temperatures, the surface becomes similar to that of specimens without a weld line (Fig. 7). The smooth zone corresponds to the propagation of the crack by the rupture of the crazes. The surface is made of broken fibrils with an orientation perpendicular to the surface of the rupture. Therefore, we can conclude that the mirrorlike zone is related to perfect welding. For intermediate temperatures, there should be a macromolecular interdiffusion gradient throughout the part, with strong entanglement in the center and low entanglement close to the sides, insufficient for good welding.

To check this hypothesis, we have calculated the distance of diffusion for these processing conditions according to the reptation theory.[15] The quadratic distance of diffusion ($\langle l^2 \rangle$) is

$$\langle l^2 \rangle = 2Dt \qquad (1)$$

where D is the coefficient of diffusion and t is the time of diffusion. Equation (2) gives D as a function of the viscoelastic parameters of the material:[16,17]

$$D = \frac{(\rho RT)^2 R_g^2 M_c(T)}{135 G_N^0 M_w^3 \eta_{0,M_c}(T)} \qquad (2)$$

where G_N^0 is the plateau shear modulus (equal to the modulus of conservation when tan δ is minimum in dynamic rheology), ρ is the density of the polymer, R

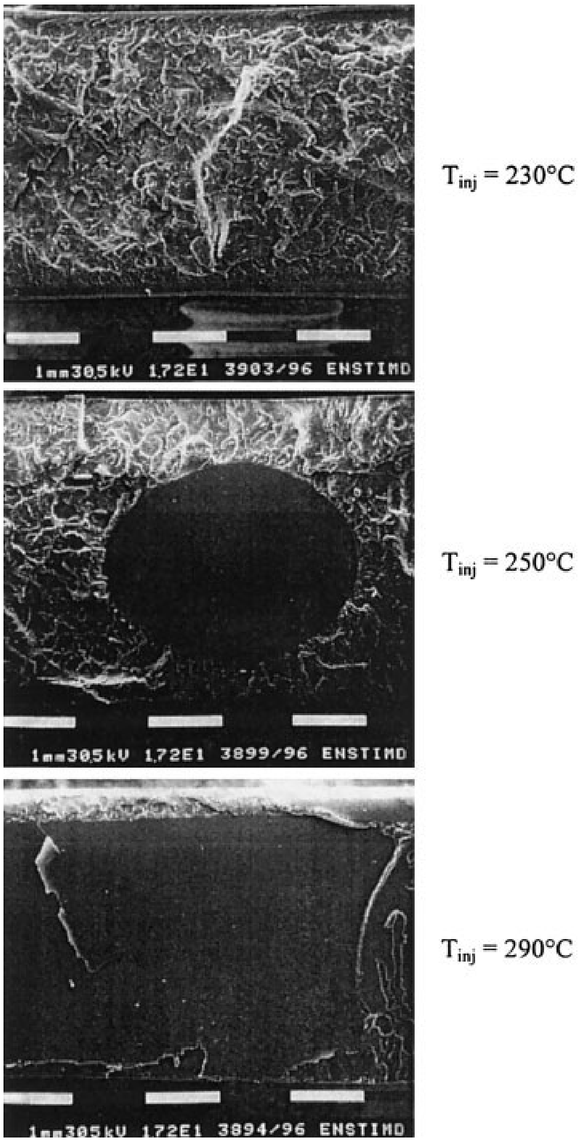

Figure 6 Evolution of the fracture surface of specimens with a weld line (notch removed) when the injection temperature (T_{inj}) is increased (PS; 1 dash = 1 mm).

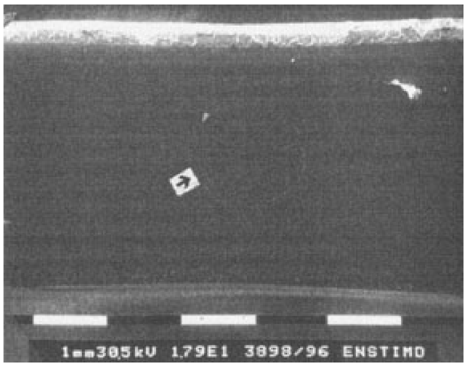

Figure 7 Fracture surface of a specimen without a weld line (PS; 1 dash = 1 mm).

is the universal gas constant, T is the material temperature, R_g is the radius of gyration, $\eta_{0,M_c}(T)$ is the zero-shear viscosity at the critical molecular weight and temperature T, M_w is the molecular weight, $M_c(T)$ is the critical molecular weight at temperature T ($M_c = 2M_e = 2\rho RT/G_N^0$), and M_e is the average molecular weight between entanglements. The parameters appearing in eq. (2) are easily accessible by experience with mainly rheological tests.[14]

D depends on the temperature, which is time- and position-dependent because of the nonisothermal injection process. The evolution of the temperature with time and position can be obtained through the solution of the heat equation. This equation has been simplified with an infinite-plate hypothesis:

$$\frac{dT}{dt} = \frac{k}{\rho C_p}\Delta T \cong \frac{k}{\rho C_p}\frac{\partial^2 T}{\partial z^2} \tag{3}$$

where k is the thermal conductivity, ρ is the density, and C_p is the specific heat of the material.

With the solution of eq. (3), D as a function of time and position can be then obtained from eq. (2). Considering that diffusion occurs until the temperature reaches the glass-transition temperature (T_g), we can calculate the average quadratic distance of diffusion ($\langle l^2 \rangle_z$) in any location:

$$\langle l^2 \rangle_z = 2\int_0^{t_{\mathrm{diff}}^z} D(t)dt \tag{4}$$

where t_{diff}^z is the time of diffusion (time to reach T_g) at distance z from the medium plane of the cavity.

The evolution of the distance of diffusion given by these calculations is displayed in Figure 8 for different injection temperatures. These results show that an increase in the processing temperature considerably raises the distance of diffusion. Moreover, this distance is strongly dependent on the location within the specimen. As suggested by Pecorini and Seo,[17] R_g has been used as a reference for the transition from adhesive to cohesive weld. Figure 8 shows that at 250°C the distance of diffusion is larger than R_g in only a part of the section. A comparison with Figure 4 shows an evident correlation, with the existence of two zones with different morphologies corresponding to two different failure modes and, therefore, two interface qualities. Furthermore, there is a good size match between the zones defined by R_g (Fig. 8) and the actual zones appearing in Figure 6.

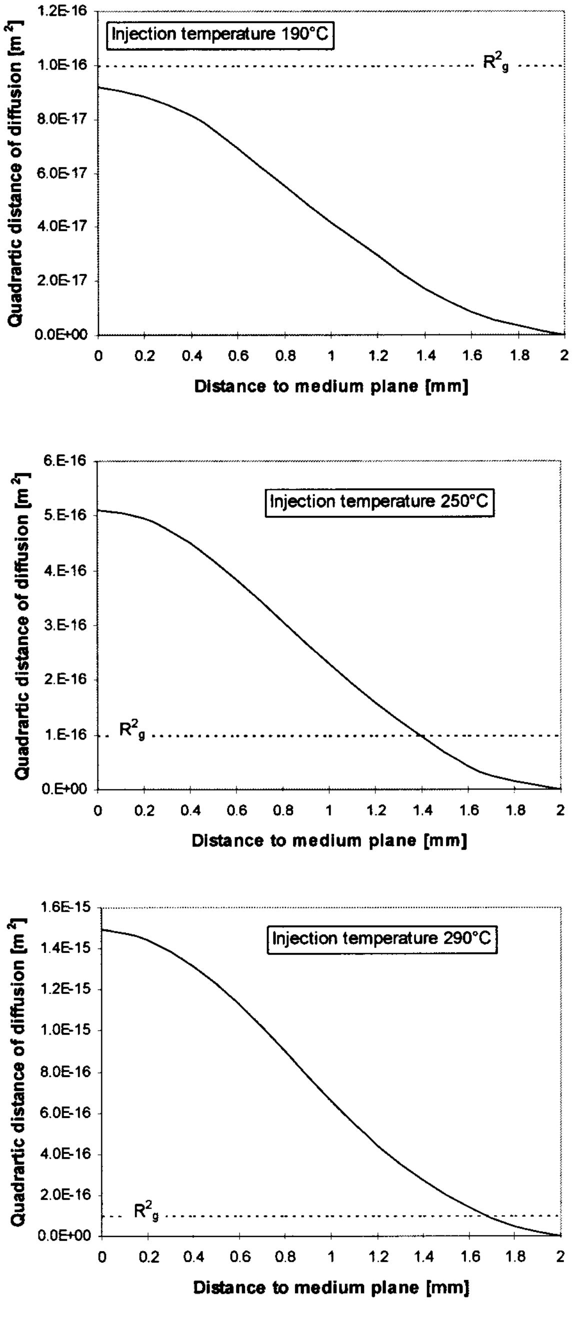

Figure 8 Quadratic distance of diffusion for different injection temperatures (PS).

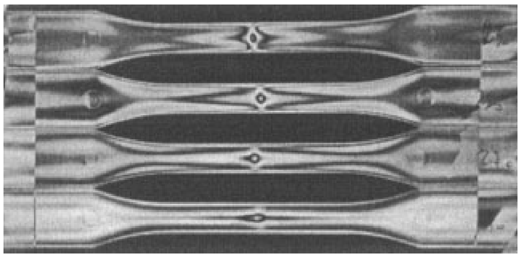

Figure 9 Image under polarized light of PS specimens with a weld line. The injection temperatures, from top to bottom, were 230, 250, 270, and 290°C.

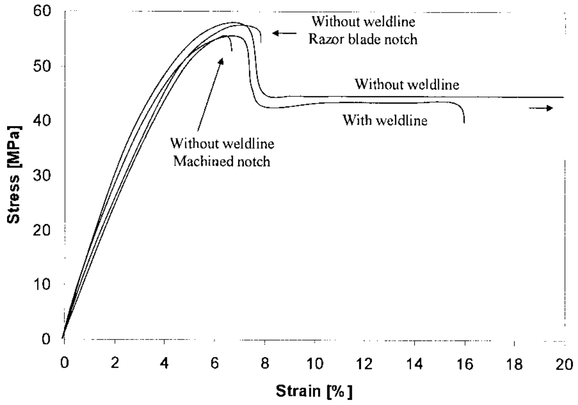

Figure 10 Tensile behavior of PC with and without weld lines and with artificial notches.

The specimens injected at high and low temperatures have confirmed this observation because they correspond to the extreme cases in which R_g is larger or lower, respectively, than the distance of diffusion through the whole section. In these cases, the fracture surface shows a homogeneous morphology. We can deduce from these results that a distance of diffusion larger than R_g is necessary for a cohesive weld.

The observation of specimens under polarized light (Fig. 9) rounds off the analysis of the temperature effect. The disrupted area around the weld line disappears progressively when the processing temperature is increased. The internal structure of the specimens approaches that of specimens without a weld line, and so do the mechanical properties (when the surface notch is removed).

The aforementioned results show that the problem of a weld line on a brittle, amorphous polymer such as PS is essentially controlled by the surface defect, as long as the weld is cohesive (an effect of the injection temperature). Therefore, the loss of properties is mainly connected to the notch sensitivity of the material and not to the intrinsic weld quality.

PC

Even though PS and PC are both amorphous polymers, their sensitivity toward weld lines is very different, as discussed next. The following paragraphs also provide some general hypotheses based on the results obtained with PS to explain this discrepancy.

Tests performed on PC (Fig. 10) show that the tensile behavior in the early stage of the tests (up to 15% strain) is very similar for specimens with and without weld lines. The necking starts outside the weld area, and properties such as Young's modulus and the yield stress and strain are not significantly influenced by the presence of weld lines.

However, as mentioned in the introduction, the stress and strain at break are sensibly reduced by a weld line. Even though this reduction has not been quantified in this study, its origin has been clearly observed. It is due to the interruption of the necking phenomenon connected to the formation of a crack at the flaw on the surface of the weld line. When the necking reaches this point, a crack starts opening and leads to failure after slow propagation through the weld area.

Figure 11 shows a PC specimen with a weld line under polarized light. The weld line is hardly visible, and this indicates that the interface between the two flows is cohesive (similar to PS injected at a high temperature; Fig. 9).

PC, like PS, is known as a notch-sensitive material. Its behavior can change from ductile to brittle with a reduction in the radius of the notch or the test temperature or with an increase in the test speed, as reported by Inberg and Gaymans.[18] Tests performed on specimens without a weld line but with an artificial notch made with a razor blade or a milling machine have shown that PC becomes brittle when a notch is present (Fig. 10). The necking starts in the vicinity of the notch, and the failure occurs just afterward. From this behavior, we can conclude that weld lines in PC are related to surface defects that are small enough to have little effect on the global behavior (ductile behavior with necking outside the weld zone as in unwelded specimens). However, this defect remains sufficient to lead to an early failure of the specimens.

These observations can partly explain the difference in behavior toward weld lines between PS and PC. Considering that both materials are intrinsically notch-sensitive, we can explain the different weld-line sensitivities by a large flaw size discrepancy. Tomari et al.[12] showed that the weld-line depth of PS is 0.2–

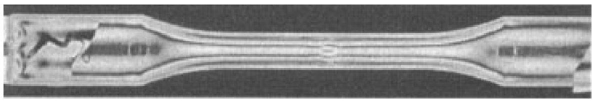

Figure 11 Image under polarized light of a PC specimen containing a weld line.

0.3 mm, whereas PC has a surface flaw of a negligible size. From a comparison of the behaviors with artificial notches and weld lines (Figs. 5 and 10), it can be concluded that the flaw in PS is large enough to act as a notch and a stress concentration site, whereas it is too small in PC. As a result, PS is weld-line-sensitive, and PC is not. The precise origin of this observation needs to be further investigated.

The difference between PS and PC can also be explained by different densities of entanglement, PS being less entangled than PC.[19] Therefore, the relaxation time is shorter for PC, and a thermodynamic equilibrium is quickly reached when the melt fronts meet. That is why PC is not sensitive to the processing temperature, unlike PS. Furthermore, the ductility of PC, associated with a high density of entanglement, allows the development of a plastic zone at the tip of the crack, reducing in this way the impact of defects. PS cannot dissipate energy through plastic deformation because of its low density of entanglement and its subsequent brittleness, which makes the material much more sensitive to defects such as weld lines.

CONCLUSIONS

The experiments carried out in this study show that the weld-line sensitivity of PS is mainly related to the conjugated effect of the surface notch and processing temperature. The first acts as a stress concentration site and is predominant, whereas the second influences the intrinsic quality of the weld. Calculations of the molecular diffusion have established a direct relationship between the distance of diffusion and the fracture mechanisms. R_g appears to be a pertinent indicator for determining the distance of diffusion at which the transition from adhesive to cohesive weld takes place.

Comparisons between PS and PC have shown very different behaviors, PS being much more sensitive to weld lines than PC, even though both are amorphous polymers. Considering the observations made for PS, we find that the differences in the surface flaw size and density of molecular entanglement can be used to explain this situation.

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