Failure of Multimaterial Fusion Bonding Interface Generated during Over-Injection Molding/Thermoforming Hybrid Process

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ABSTRACT: The effects of processing parameters on the strength of the fusion bonding interface have been investigated. The interface was generated when an isotactic polypropylene homopolymer was injection molded on a solid self-reinforced polypropylene substrate. The interface strength was measured in shear configuration, and the melting behavior of substrate was studied using differential scanning calorimetry. The results show that strong bonding interface can be achieved when the correct processing parameters are chosen. The interfacial strength is largely improved if the interface temperature is higher than the melting temperature of the substrate layer, and in these specimens failure does not take place at the interface. Furthermore, for a fixed interface temperature, interface strength increases with thermal gradient. Finally, in the analyzed holding pressure range, pressure apparently has no effect on strength. © 2006 Wiley Periodicals, Inc. J Appl Polym Sci 102: 261–265, 2006

Key words: failure; strength; interfaces; injection molding; fusion bonding

INTRODUCTION

There are several applications (automotive, home electrical appliance, etc) with structural requirements, parts meeting complex geometries, large area, and small thickness. Current processing technologies are not well adapted to this kind of parts: processes adapted to complex geometries (injection molding) are not adequate for parts with large projected area. However, processes adapted to large area parts (thermoforming, GMT, metal sheet conformation) do not allow complex geometry and, thus, several parts and assembling operations are necessary. To develop advanced, low cost, low-weight components based on polymer, a new injection-molding/thermoforming hybrid process is proposed.

The principle (Fig. 1) is based on a thermoformed substrate, which fills the esthetical, isolation, ergonomic, and geometric functions, and an over-injected skeleton composed by a ribbed structure, which fills the structural requirements.

As polymer interfaces play a critical role in controlling the properties and reliability of a broad range of products, many different investigations have been carried out.[1–4] Establishing strong bonded interfaces between different materials, or between components with different thermomechanical histories is essential to the success of the proposed approach, and is the motivation of this research, where the main objective is to establish the effects of the processing parameters on the strength of the substrate/skeleton interface.

EXPERIMENTAL

Materials and specimen preparation

For this investigation, polypropylene (PP) has been selected. PP is an extremely versatile thermoplastic and is available in many grades and forms (homopolymer, copolymer, blend, fiber, film, foam, etc.).[5]

For the over-injection skeleton, an injection grade isotactic polypropylene (PP) homopolymer (SM6100, Montell) was used, whereas for the thermoformed substrate, CURV™ (from BP Amocco) was selected. This material is composed of high-modulus polypropylene fibers which are compacted to form a self-reinforced, thermoformable, 100% polypropylene sheet.

The process in which there is some degree of interpenetration between the macromolecules of both sides of the interface and cocrystallization is known as cicatrisation. Cicatrisation process is composed of four main steps[6]: (1) the contact between the molten polymer and the substrate; (2) heat transfer from the mol-

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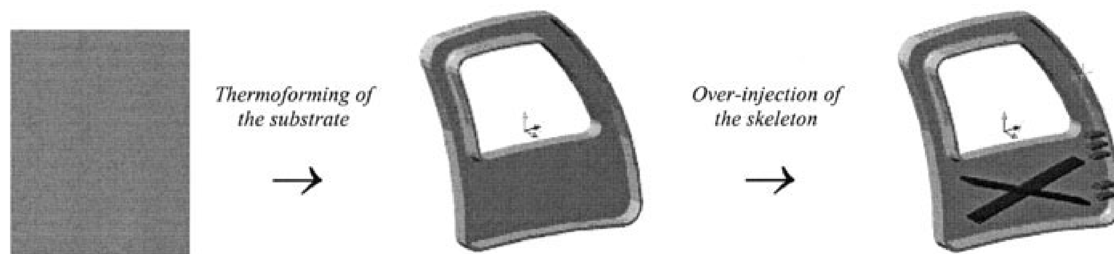

Figure 1 Principle of the new injection-molding/thermoforming hybrid process.

ten polymer to the substrate, and eventually the melting of the latter; (3) molecular interpenetration across the interface; (4) solidification of the interface.

The over-injection molded specimens have been obtained in a Battenfeld BA 600CDC injection-molding machine. From the multiple processing parameters involved during the hybrid process of over-injection molding/thermoforming, three have been analyzed in this work. The chosen parameters are the over-injection molding temperature (T_o), the substrate temperature (T_s), and the holding pressure of injection (P). Three different temperatures have been selected for T_o (210, 230, and 250°C) and for T_s (80, 100, and 120°C). The substrate was temperate in a fan-assisted oven for 5 min before being inserted into the mold. The combination of these T_o and T_s values gives different interface temperatures (defined as the average of both T_o and T_s temperatures, $T_i = (T_o + T_s)/2$) and thermal gradients ($\Delta T = T_o - T_s$). Table I shows the obtained T_i and ΔT values resulting from the combination of T_o and T_s temperature values.

To analyze the influence of holding pressures (P), two different values (247 and 371 bar) were used for each combination of temperatures shown in Table I.

The rest of over-injection parameters have been kept constant and their values are the following: the injection time and the screw rotation rate used were 0.43 s and 200 rpm respectively, and a cooling time of 20 s.

Over-injection test specimen geometry was composed of a CURV™ substrate sheet (2 × 15 × 150 mm^3) and a prismatic over-injection molded bar (4 × 13 × 135 mm^3).

TABLE I
Interfacial Temperature (T_i) and Gradient (ΔT, in parenthesis) Obtained by the Combination of the Over-Injection Molding Temperature (T_o) and Substrate Temperature (T_s)

	T_o (°C)		
T_s(°C)	210	230	250
80	145 (130)	155 (150)	165 (170)
100	155 (110)	165 (130)	175 (150)
120	165 (90)	175 (110)	185 (130)

Strength characterization test

In a previous work,[7] the type of solicitation and stress level at which interfaces are subjected have been analyzed by finite element simulation. The main conclusion, for the case studied, was that bonding interface should withstand during service of a shear stress of 2.2 MPa. Basically, the test method is a tensile test, but the substrate and skeleton have been machined, leaving a limited bonding interface length of 10 mm (Fig. 2), so that the interface works under shear conditions. Strength characterization tests were performed on an Instron 4206 standard testing machine at a crosshead speed of 50 mm/min and at room temperature.

The results obtained on each test were the mode of failure and shear strength of the interface, which is calculated by dividing the failure force by the bonding interface area (10 × 13 mm^2). Given values of strength are calculated as the average of a minimum of eight specimens.

Melting behavior of the substrate

Differential scanning calorimetry (DSC) was carried out with a Perkin–Elmer DSC7 thermal analyzer under nitrogen atmosphere. According to ASTM D-3417–97 the samples typically weighed between 5.5 and 6 mg. DSC samples were heated from 50 to 250°C at a heating rate of 10°C/min, with the aim of determining the melting behavior of the substrate (melting temperature range and peak temperature, T_m).

Evolution of interface temperature during cicatrisation

To study the dependence of interface temperature on time, finite element simulations (COSMOS™) have

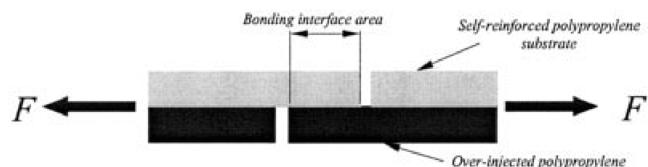

Figure 2 Schema of the test for interface strength characterization.

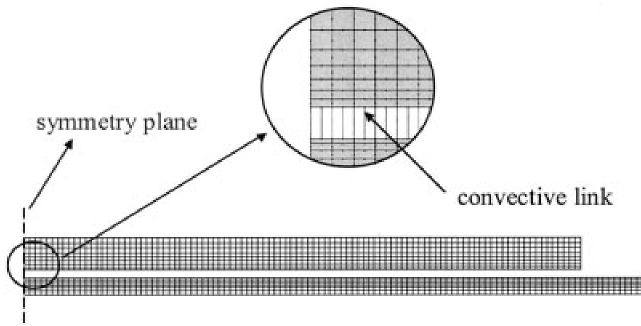

Figure 3 Schematic representation of the interface model for FEA simulations.

TABLE II
Specific Thermal Capacity (c_p) and Density (ρ) as a Function of the Temperature

Temperature (°C)	Property	
	ρ (g/cm^3)[a]	c_p (J/kg°C)[b]
25	0.905	1623
50	0.893	1754
100	0.879	2117
150	0.833	2511
200	0.759	2712
250	0.741	2892

[a]Values taken from ref. 8.
[b]Values taken from ref. 9.

been carried out. Because of the over-injected specimen configuration, bidimensional heat transfer model and adiabatic symmetry have been chosen to simulate the interface cicatrisation process (Fig. 3). Thermoformed skin and over-injected skeleton have been respectively discretized by 111 × 6 and 100 × 8 quadrilaterals elements of 8 nodes. The contact between them (8.775×10^{-4} m2) is modelized by convective heat transfer links.

The simulations are limited to the earlier instants of the interface generation, so heat transfer is supposed to occur only across the contact interface. Consequently, the temperature in the bulk of the specimens is considered to be constant and the specimen/mold interface are modelized adiabatically.

Specific thermal capacity (c_p) and density (ρ) as a function of the temperature (Table II), heat transfer coefficient (10^5 W/m^2 °C), and specific latent heat of

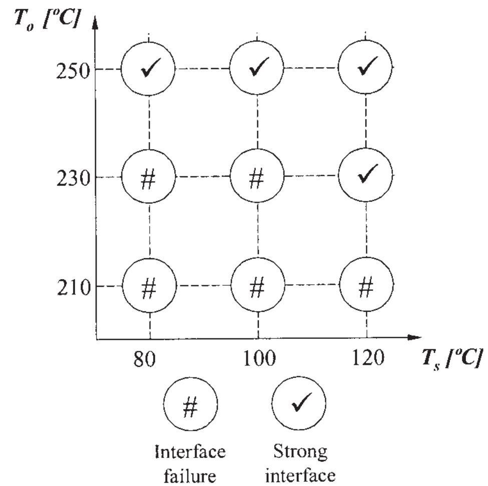

Figure 5 Map of failure modes depending on over-injection (T_o) and substrate temperatures (T_s). Failure of the interface is indicated by the symbol #, whereas failure of the substrate is identified by $\surd$.

fusion (209×10^3 J/kg) have been taken from the literature.[8,9] Values of thermal conductivity (k) of PP go from 0.22 to 0.11 W/m °C, at 25 and 250°C, respectively. As the thermal diffusivity ($\alpha = k/\rho c_p$) of the PP is low, k is considered to vary linearly with temperature between these two values.

RESULTS AND DISCUSSION

Failure mode analysis

As it has been mentioned in the introduction, establishing strong bonded interface between the thermoformed substrate and over-injection molded skeleton is essential to the success of the proposed approach. Of immediate practical interest is to know whether the failure develops at the interface or not, and which are the critical processing parameters. As a result of mechanical tests, two failure modes have been developed: (1) failure of the interface by shearing [Fig. 4(a)], or (2) delamination of the substrate [Fig. 4(b)].

The effects of the processing parameters on failure mode are summarized in the Figure 5. As can be seen, it is possible to obtain strong interfaces (marked with the symbol $\surd$) with the combination of high substrate and over-injection temperatures, but not all the pro-

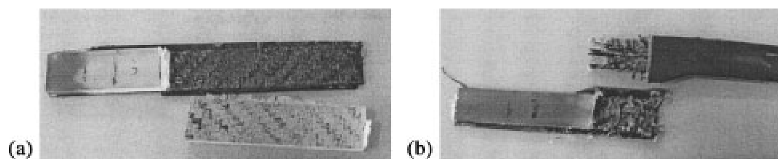

Figure 4 Failure modes of over-injection molded specimens: (a) failure of the interface by shearing, and (b) delamination of the substrate.

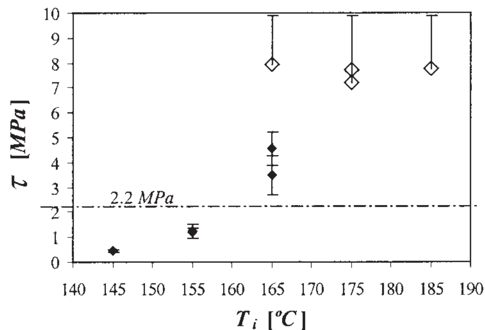

Figure 6 Shear strength of the interface (τ) as a function of the interface temperature (T_i).

cessing parameters ensure the integrity of the specimen, since the failure can also develop at the interface (marked with the symbol #).

Interface strength

To study in a more quantitative way the effects of processing parameters, the shear strength (τ) of the interface has been analyzed as a function of the interface temperature (T_i) (Fig. 6). In specimens where failure developed at the substrate, the shear strength of the interface could not be calculated, but it was assumed higher than the shear stress at the failure point. As can be seen, there are processing conditions for which strength is higher than the required shear stress (2.2 MPa), assuring the structural integrity of the component. Even when the failure develops at the interface, there are several specimens that fulfill this requirement.

Furthermore, three different $\tau - T_i$ dependences could be distinguished in Figure 6:

1. For T_i lower than 165°C the strength increases slightly with T_i, but it takes the lowest values.
2. At 165°C different strength values can be achieved.
3. For Ti higher than 165°C the interface is stronger even than the substrate.

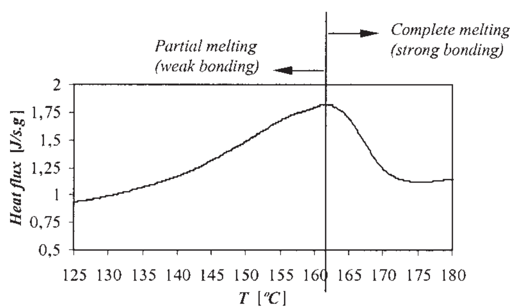

Figure 7 Heating DSC curves for substrate material (CURV™).

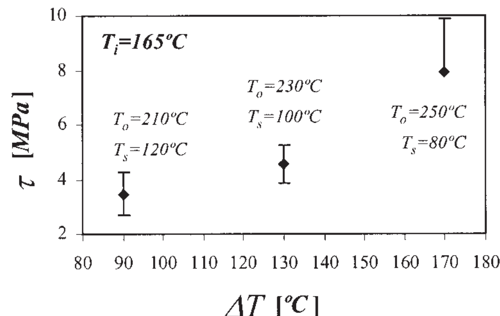

Figure 8 Shear strength of the interface (τ) as a function of the thermal gradient at the interface (ΔT) for T_i = 165°C.

To understand the dependence of the strength on the interface temperature is necessary to analyze the melting behavior of the substrate material (Curv™). Heating DSC curve (Fig. 7) shows a single broad endothermic peak around 163°C. In contrast to pure, low molecular weight materials that melt at exactly well-determined temperature, the melting of semicrystalline polymers such as PP takes place over a range of temperatures.[10] Polymers generally crystallize under nonequilibrium conditions with considerable supercooling, consequently nonequilibrium structures are formed. The further crystallization conditions are from the equilibrium, the less perfect is the structure produced (lower degree of crystallinity, smaller size crystallites, lower melting point) implying a lower structural stability. So the crystallite-size distribution generated during the nonequilibrium crystallization originates the range of melting temperature. Crystallites with smaller dimensions (thinner) melt at lower temperatures, whereas those with larger dimensions (thicker) melt at higher temperatures. Thus, when T_i is lower than 163°C, the substrate only melts partially, and consequently the bonding is not strong. However, when T_i is higher than the melting peak temperature, the substrate melts completely, the molecular diffusivity across the interface is higher, and a strong bonding interface can be generated. From literature data on diffusion rates for the molecular weights[11] corresponding to commercial polymers, it can be stated that no appreciable transport of center of mass will occur across the interface within the processing time used for injection molding (0.43 s for injection and 20 s for cooling). Therefore, the thickness of melted solid layer next to the interface need to be only a few radii of gyration.

As it has been mentioned earlier (Fig. 6), there are three specimens with identical interface temperature (165°C) obtained with different thermal gradients ($\Delta T = T_o - T_s$). The strength plotted versus the thermal gradient (Fig. 8) shows that strength increases with ΔT. Thus, it is not necessary to preheat the thermoformed substrate layer well above its melting temperature to obtain strong bonded interfaces.

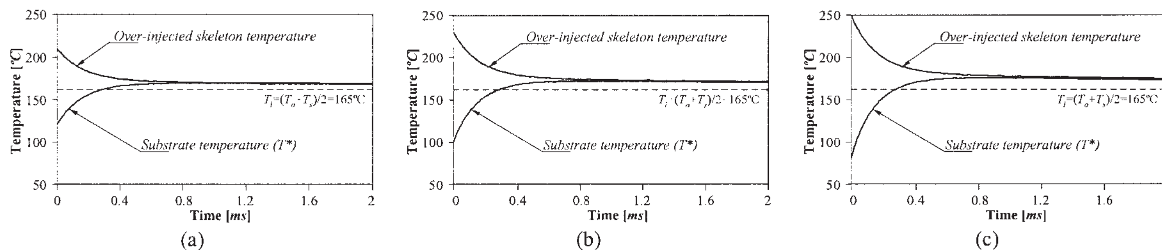

Figure 9 Thermal simulation results of the evolution in time of the temperature at the interface on the substrate (T^*) for different processing parameters: (a) T_o = 120°C/T_s = 210°C, (b) T_o = 100°C/T_s = 230°C and (c) T_o = 80°C/T_s = 250°C.

One possible contributing factor to the observed increase in strength with ΔT for fixed T_i is that calculated T_i is not a good measure of the interface temperature, specially at the earlier stage of bonding. Because of the low thermal conductivity of polypropylene, the earlier stage of cicatrisation can be supposed to develop under adiabatic conditions. Furthermore, heat capacity of the molten polypropylene is higher than that of solid one.[9] Thus, the temperature at the interface on the substrate (T^*) goes through a peak higher than T_i before reaching the steady state. Finite element simulations of the temperature evolution at the interface on the substrate (T^*) for different processing parameters (Fig. 9) show that this peak temperature increases with thermal gradient (169.3, 172.2, and 175.8°C, respectively). And thus, this justifies the higher strength of the specimens obtained with larger gradient, as a consequence of the larger molecular diffusivity across the interface.

The importance of the thermal gradient has also been pointed out in some other investigations[2,12] where, for a given interface temperature above melting temperature, it has been shown that most rapid bonding is obtained for the highest thermal gradients.

The strength differences obtained with identical temperatures (T_o = 230°C and T_s = 100°C) and different holding pressures (P) are small (3.5 ± 0.8 MPa for 371 bar and 3.1 ± 1.0 MPa for 247 bar), so it can be concluded that the pressure has no influence in the range analyzed. Similar results have been obtained[12] in compression molding, where at pressures higher than 200 bar bonding strength depends weakly on pressure.

CONCLUSIONS

The effects of processing parameters on the strength of the fusion bonding interface have been investigated. The interface was generated when an isotactic polypropylene homopolymer was injection molded on a solid self-reinforced polypropylene substrate. By varying over-injection molding temperature, substrate temperature and holding pressure of injection processing map has been established, aimed to providing guidelines for the optimization of PP/CURV™ bonding.

The results show that strong bonding interface can be achieved if the correct processing parameters are chosen, and that in these specimens failure does not take place at the interface. Heating DSC curve of the substrate shows a single broad endothermic peak around 163°C. When interface temperature (T_i) is lower than 163°C the substrate only melts partially, and consequently the bonding is not strong. However, when T_i is higher than the melting peak temperature, the substrate melts completely, the molecular diffusivity across the interface is higher, and a strong bonding interface can be generated.

Furthermore, for a fixed interface temperature, interface strength increases with thermal gradient. Finally, in the analyzed holding pressure range, pressure apparently has no effect on strength.

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