

Study on the Influence of Process Parameters on Interface Strength of Film-insert Molding

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Abstract. By the controlling of variable, the influences on the film interface strength that film-line molding process had were investigated. The interfacial bonding strength under the circumstance where various of processing parameters (temperature, film thickness, injecting speed) were combined was measured through a normalized 90° peel test. And we used Super-high magnification lens zoom 3D microscope to observe the Interface structure around the interface and the section of interface was observed too. To profoundly study the interfacial bonding strength mechanism, using the numerical simulation method, we analysed the temperature changes of the interface. And the crystallinities in different processing parameters combination situations were monitored by thermal characterization DSC. And then we calculated the crystallinities of the melt at the interface, drawing a conclusion about the relations between crystallinity and interface strength. The results show that the process conditions are favorable for improving the crystallinity of the interface and have obvious effects on the strength of the interface.

1. Introduction

Film-insert molding (FIM), a technique also known as in-mold decoration, is a relatively new technique in which molten resin is injected to fill the cavity after a preprinted film, which is formed as the shape of the cavity is inserted into the cavity. Recently, FIM has been widely used in injected molded parts such as automotive panels, labels for mobile phone, and household appliance. Compared with traditional injection molding, FIM is one-step process without extra post-processing including screen printing, which can reduce manufacturing time and cost. Furthermore, the product is well protected in harsh abrasive environments with film, so the product is finished with high quality, durability and aesthetic value. Moreover, the hot resin would wet and partially melt the film, which can provide better adhesion of the film to the substrate. It is important to stick to products through the lifetime effectivity and lasting, and the challenge for sufficient adhesion is similar to other processing such as welding, coating[1], and injection overmolding[2-6].

In the past decades, there have been many experimental studies that investigated the effect of material combination and processing parameters on the interface adhesion which determines the properties of final product. Y.W. Leong[7] measured the effect of molding conditions such as barrel temperature, injection speed and holding pressure on the morphological properties between Polypropylene(PP)-Film and PP Matrix. It was found that higher barrel temperature promotes better



interface adhesion. Yamaguchi, S[8] studied the effect of crystallization on interface adhesion between PP film and PP substrate. In the latter work, He studied the adhesion between Polycarbonate (PC) Film and Polycarbonate-Acrylonitrile-Butadiene-Styrene (ABS) Substrate and he found that adhesion increased with thicker film [9]. The effect of molecular orientation induced by various injection speed on the interface was measured [10]

In these studies, fundamental understandings of the interface adhesion have been explained with one or more of following mechanism: interface interlocking, molecular diffusion and entanglement, crystallization, and adsorption theory[11-14]. To obtain the desired film-substrate interface bonding, higher melt temperature is necessary to promote molecular interpenetration, and compatible combination of film and substrate material should also be guaranteed, although the effect of holding pressure and injection speed remain uncertain in terms of different material combination. The final adhesion strength is complicated being the result of a sum of the contributions mentioned above.

For semi-crystalline polymer-polymer adhesion, besides molecular entanglement, the crystallization at or near the interface region also plays an important role in the mechanical properties and attracts a lot of attention[15, 16]. Formation of crystal depends on various factors such as nucleation location, rate of crystallization, transcrystallinity, and molding conditions[17, 18], being more complicated than amorphous polymer. However, the interface region between film and substrate is still in a rudimentary stage. Previous attempts focusing on the effect of processing conditions such as injection speed, melt temperature on the interface crystallization, the understanding of decoration film and mold temperature which determine the interface crystallization is still lacked. Thus, a deep study of the decoration film is necessary. An amorphous PC/ABS resin was in previous investigation of film's effect on adhesion. In this study, semi-crystalline resin polypropylene was used. The influence of film thickness, mold temperature on the interface morphology and adhesion was explored as well as the injection speed. The crystallinities in both the film-substrate and controlled specimen (without film) were monitored by thermal characterization DSC. 90° Peeling test was performed to gauge interface adhesion strength. Thermal analysis and Simulation were performed to get better understanding of the temperature variations at resin-film interface. The relationship between crystallinity and interfacial bond strength was established to enable the control of product properties.

2. Experiment and Methods

2.1 Materials

The substrate resin employed in this study was a commercial grade polypropylene (PP 1215C) with melting point of 165°C, density of 889kg/m³, thermal conductivity of 0.255 W/(m K), and specific heat of 3100 J/(kg K). The material of insert plastic film was polypropylene. Various thickness of the insert film used in the study were 400um, 500um, and 600um. The injection mold was made by P20 steel with density of 7850 kg/m³, thermal conductivity of 31.5 W/(m K), and specific heat of 501.6 J/(kg K).

2.2 Film-insert molding

PP substrate resin was injected to a rectangular shape(size: 140mm×60mm×2mm) using the JSW-180H. The PP film was attached to one side of the mold wall prior to injection. Adhesive tape was used at the both ends to avoid the slide of film. PP resin was injected over the film. Rectangular specimens were cut from the product to perform peel and tensile test. Specimens and product dimensions are shown in Figure. 1(a). One of the PP halves was adhered to thin-layer polyimide film to obtain a nonstick region so that the film could be detached easily from the substrate during the peeling test, the peeling test is shown in Figure. 1(b). Products devoid of the film insert were also molded as control. Injection speed, barrel temperature, film thickness and mold temperature were varied to investigate the interface adhesion. Details of the injection molding conditions are listed in table 1.

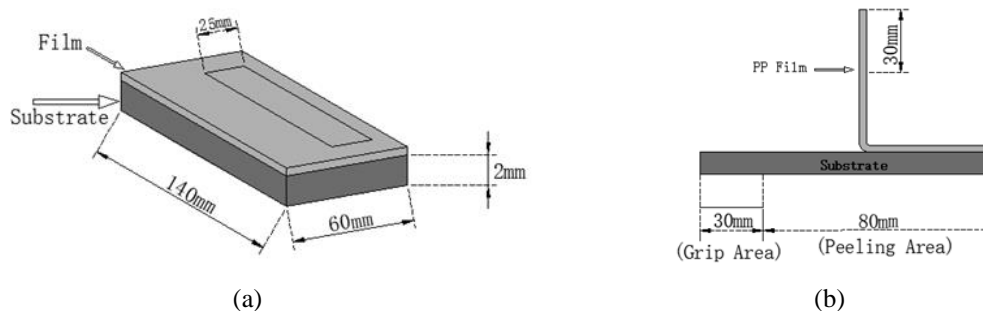


Figure. 1. Injection molding species and peeling sample dimensions
(a)Injection parts; (b)peeling test pieces

Table 1. Film injection processing conditions

Barrel temperature	220°C, 240°C, 260°C
Injection speed	10, 40, 70 mm/s
Mold temperature	30°C, 50°C, 70°C
Film thickness	0.4mm, 0.5mm, 0.6mm
Holding pressure	50 MPa
Holding time	3 s
Cooling time	60 s

2.3 Peeling and tensile test

Adhesion strength between film and substrate was gauged by a normalized 90° peel test using the zwick Proline Testing Machine. The toughness (GA) was determined by

$$G_A = \frac{P}{B}(1 - \cos \theta) \quad (1)$$

where P is the peel force, B is the width of the layer and θ is the peel angle. The configuration and the dimensions of the specimens for the peel test are indicated in Fig.1b. Peeling arm was attached to the grips of a universal machine, while the fixed arm was attached to a moving plane to maintain peeling angle 90°, a peel test speed of 10 mm/min was used. This velocity ensured the test to be realized in a reasonable time period while maintaining the required alignment between specimen arms. All tests were done on at least five specimens to ensure reproducibility and accuracy of results.

2.4 Injection molding simulation

During the film-insert molding, temperature alerts the mechanical properties of final product. It is necessary to track the change of temperature field. Temperature of mold interface and melt interface can be measured by sensors[19]. However, it's difficult to track the change of polymer-film interface temperature and the polymer-film interface temperature is of vital importance to the adhesion. It's difficult to track temperature change. According to processing conditions, we simulate the effects of film on the temperature variation during film-injection molding using analysis software ANSYS and then set similar boundary and initial conditions. As the real injection process is complex, several assumptions are made: a) the initial temperature was barrel temperature, b) there are no temperature gap at the interface. In addition, we simulated the control of mold-polymer interface temperature.

2.5 Thermal and Morphological characterization

For polymer characterization of crystallization, differential scanning calorimeter (DSC) PerkinElmer DSC8000 was performed to assume thermal behavior of the materials. Heating rate of experiment was 10°C/min under the nitrogen atmosphere. We used VHX-1000C Super-high magnification lens zoom 3D microscope to observe the Interface structure around the interface and the section of interface was observed too.

3. Result and discussion

3.1 Interface temperature variation

3.1.1. Influence of mold temperature Figure 2(a) is the interface temperature change simulation in ANSYS software during the cooling period. It can be seen that at the beginning of the cooling, the temperature at the interface drops rapidly below the melting point of PP and then slowly cools down. When the temperature of the mold is 30 °C, cooling time of the interface temperature decreasing rapidly from the cylinder temperature to the melting point of PP is about 0.4s. While for mold temperature of 50°C, the time for the temperature above to decrease to melting point is about 0.5s, longer than mold temperature of 30°C. For mold temperature of 70°C, the time period is 0.9s, twice more than the time of mold temperature 30°C. When the temperature is higher then the PP melting point, PP molecules have a high degree of activity, which can carry out a lot of movement, providing a basis for better interface combination, moreover, during the cooling period, with mold temperature, resin temperatures are respectively higher leading to faster crystallization rate, which is preferred for higher crystallinity.

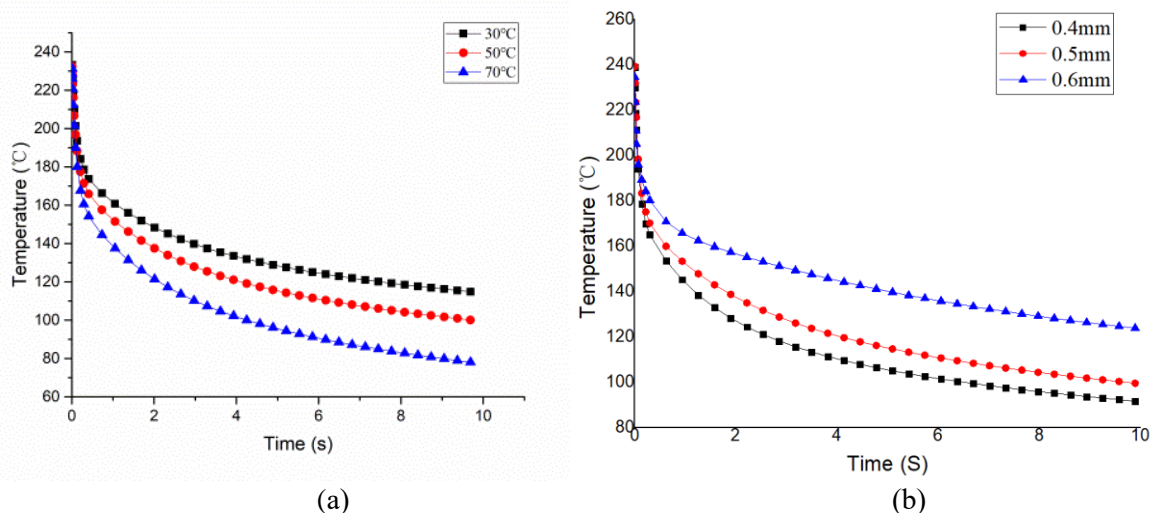


Figure 2. Interface temperature variation for different mold temperature
a) Different mold temperature; b) different film thickness

3.1.2. Influence of film thickness The presence of the film hinders heat transfer resulting in a slower temperature drop at the interface of film and PP resin. Cooling curve of the first few seconds is shown in figure 2(b). Within the first period of cooling, the temperature drop is almost the same. However with the increasing of time, the effect of film is more obvious. For thicker film, there is higher temperature at the interface. What's more, time of interface temperature above PP melting point increases from 0.37s to 0.75s responding to the film thickness from 0.4mm to 0.6mm, which is preferred for better adhesion of interface and higher crystallinity. While for interface temperature below PP melting point, molecular movement is significantly reduced, influencing little on the interface adhesion.

3.2 Interface crystallization

Injection of PP will crystallize during the cooling process. The change of the injection molding process parameters will directly affect the degree of crystallinity and grain size. Crystallization has virtual importance to adhesion strength and grain damage during peeling test absorbs more energy than amorphous structure leading to higher strength. The DSC curves of different mold temperature are shown in figure 3. Calculated crystallinity of injected species is shown in figure 8; the crystallinity of PP interface is relatively lower than core region because the melt temperature at the interface dropped

quickly when coming into contact with film. There is not enough time for melting to crystallize. While for core region, the temperature is higher leading to faster crystallization speed and resulting to higher crystallinity.

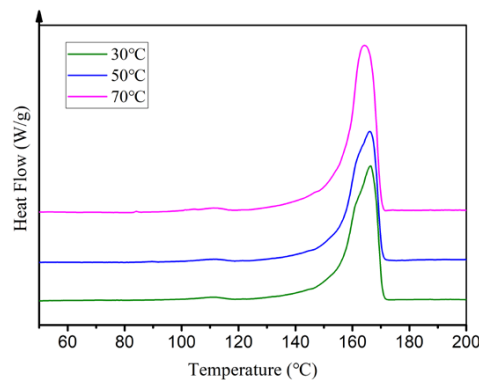


Figure 3. DSC curves of PP at interface

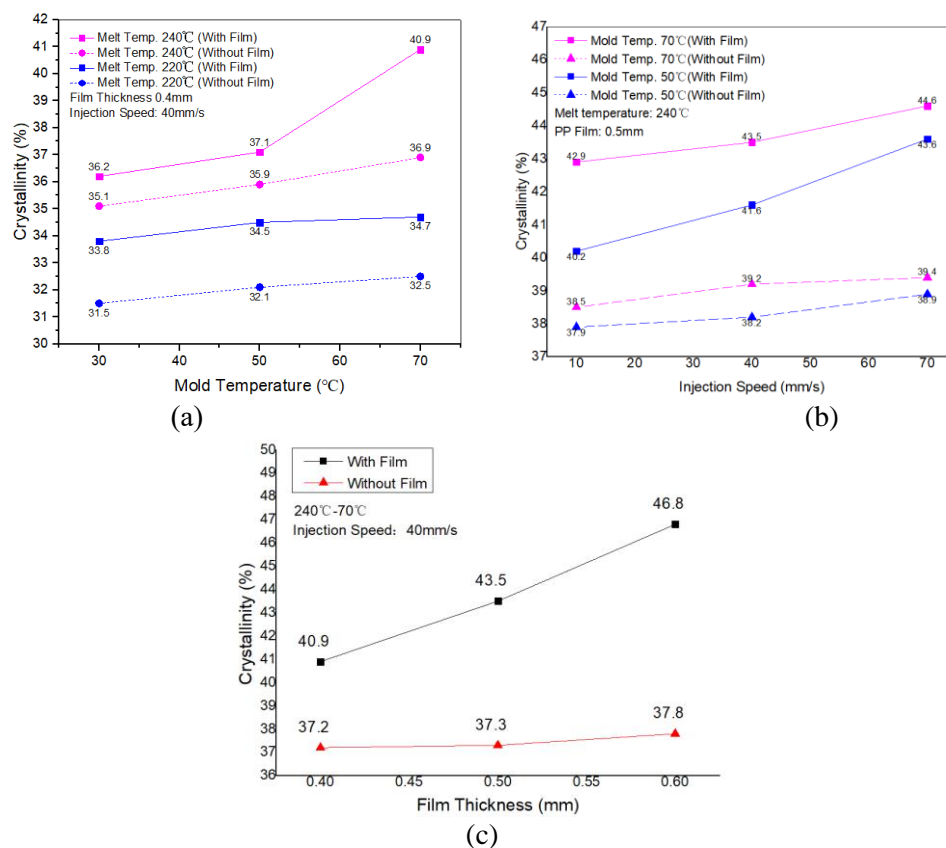


Figure 4. Crystallinity of different injection speed and mold temperature

(a) Influence of temperature on crystallinity; (b) Influence of injection speed on crystallinity;

(c) Influence of film thickness on crystallinity

The surface crystallinities of different mold temperature are shown in figure 4(a). With mold temperature and melt temperature rising, crystallinity rises respectively, because rising of mold temperature and melt temperature accelerate crystallization rate leading to higher crystallinity. Similar observation is shown in figure 4(b): crystallinity rises from 0.42 to 0.46 respectively with injection speed rising, for crystallization induction time is reduced and there is faster crystallization rate. Interestingly, for 240°C-50°C the crystallinity changed little while there is large fluctuation in peeling force, which probably shows that molecular movement effects more when crystallinity is relative high. The crystallinity variations of the molded parts are exhibited in Fig. 4(c) with respect to film

thickness and are compared with the parts molded without inserted film. In this figure we can observe that the crystallinity increase slightly with film thickness. Furthermore, the parts molded with film had larger crystallinity than parts molded without film. The reason can be contributed to the retardation in heat transfer that decreases polymer melt cooling rate. What's more, retardation in heat transfer is effected by film thickness. For thicker film, there is greater retardation leading to higher crystallinity which is preferred for better interface bonding, in correspondence to peeling force of film thickness showed in Fig.6.

3.3 Film–substrate adhesion

The peeling failure was of an adhesive–cohesive type showing a force–displacement peel graph like the one represented in Fig.5(c) curve. As illustrated, there are three kinds of force-displacement curve. Line (I) is relatively smooth and the force is smaller in comparison to others. It can be assumed that this type of interface bonding is of adhesive bonding which the crack generated and extended before film reached the yield stress. Interface morphology of line (I) is shown in Fig. 5(a) curve. Only a small part of film left on the substrate surface indicates the interface bonding in the preliminary stage. For line (II), a very distinct saw-teeth-like curve can be observed in cohesive type which is initiated by simultaneous crazing of the substrate and film surfaces during 90° peel test. Line (II) shows an indication of “stick–slip” phenomenon and it's believed that two factors account for the phenomenon. Firstly, the interfacial bonding is high enough so that the surface of the film would yield until a critical level, after which separation of the yielded regions will occur and the extension of crack will continue; secondly, as soon as the yield region failed, the film would “spring back” hindering crack propagation thus causing peeling force to drop suddenly, then peeling force would rise again for next crack propagation. However, if the interface adhesion is very high, film would undergo plastic deformation losing its ability to “spring back” during peeling test. The load curve of crosshead displacement illustrated as line (III) is gentle without “stick–slip” phenomenon. Morphology of the peeling interface is shown in fig. 5(b) curve, large region of the interface is covered by film, showing that bonding meets the requirements of normal use.

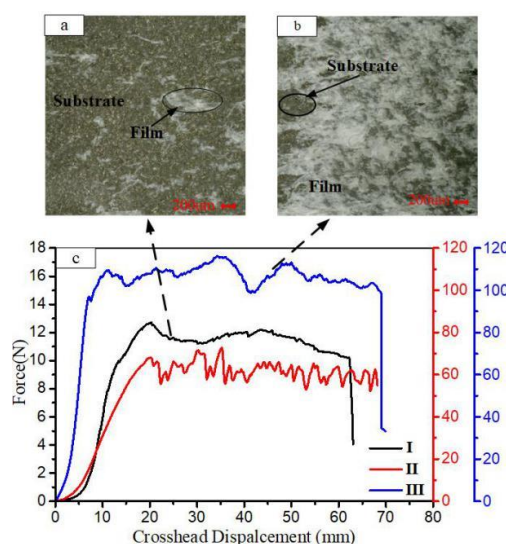


Figure. 5. Exfoliation interface ultra depth image and Peeling curve

The peeling force of different injection process parameters are showed in figure 6 and figure 7 . It shows that adhesion strength rises with melt temperature rising(showed in figure 6). For lower mold temperature the interface adhesion is relatively poor. The reason is probably that there is not efficient molecular movement at lower mold temperature. For higher mold temperature there are longer time period of temperature above PP melting point, providing active molecular entanglement leading to better interface adhesion. Figure 6. also shows that at temperature 240°C-70°C, interface adhesion is much higher than other temperature combination that achieves degree of cohesive type. Relevant

peeling force of mold temperature is shown in fig.6. Compared with Fig.4, it can be assumed that adhesive bonding is effected to some extent by crystallization. A summary of the specimen conditions after the peel test is tabulated as Table 2.

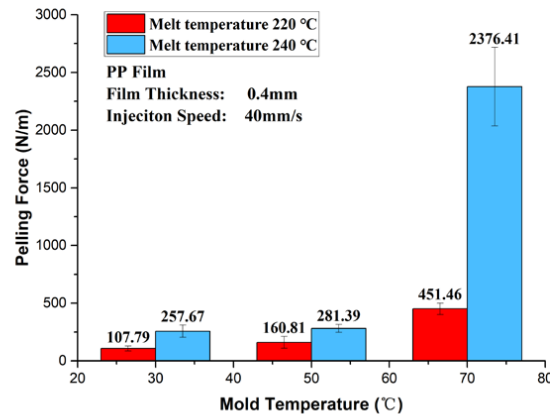


Figure. 6. Interface adhesion of different mold temperature

Table 2. Specimen Conditions after Peel Test

Melt Temp.(°C)	Injection Speed(mm/s)		
	10	40	70
220	◇	◇	◇
240	◇	◇	○

◇ adhesive bonding

○ cohesive bonding

The interfacial bonding strength at different injection speeds as shown in Figure 7. shows that, for the melt die temperature of 240 -50°C, the increase of the injection speed can promote the interfacial bonding strength. According to the results of the study of Figure 4 (b), the lifting injection speed can increase the shear effect, which is beneficial to the crystallization and the small size of the molecule. At the crystal growth, the crystallinity of the interface increases obviously, from 40.2% to 43.6%, the crystal becomes more and more energy is required to destroy the interface. In addition, the injection speed increases the temperature of the plastic melt, the film surface melts more, can also form a thicker diffusion entanglement layer, and the interfacial strength increases.

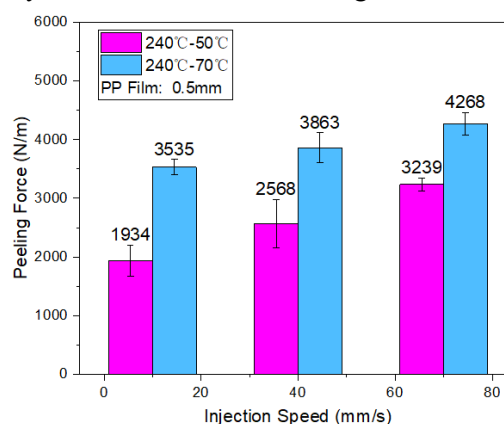


Figure. 7. Peeling force of injection speed

Koch and Mc Carren[20] stated that a thinner film was preferred to enable a firmer film–substrate adhesion in his report. Nevertheless, the findings in our work are contrary to their results and similar to the report of Leong[8]. Figure 8. shows the adhesion strength of different film thickness. It can be concluded that for thicker film there is higher interface adhesion. The existence of PP film hinders

heat conduction of melting resin, causing interface temperature higher leading to blurry boundary and more active molecular movement. For higher melt and mold temperature, the interface strength is as high as ten times of lower melt and mold temperature. It seems that temperature is the primary factor affecting interface bonding strength.

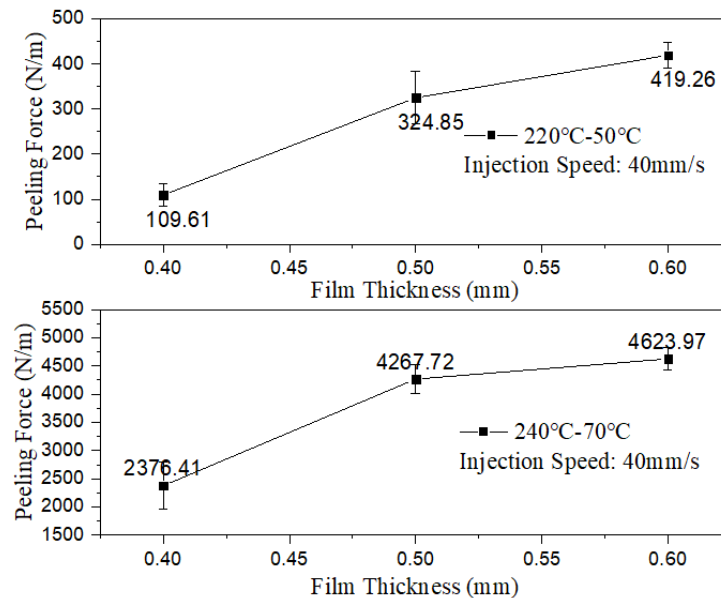


Figure. 8. Peeling force of film thickness

3.4 Interface morphology

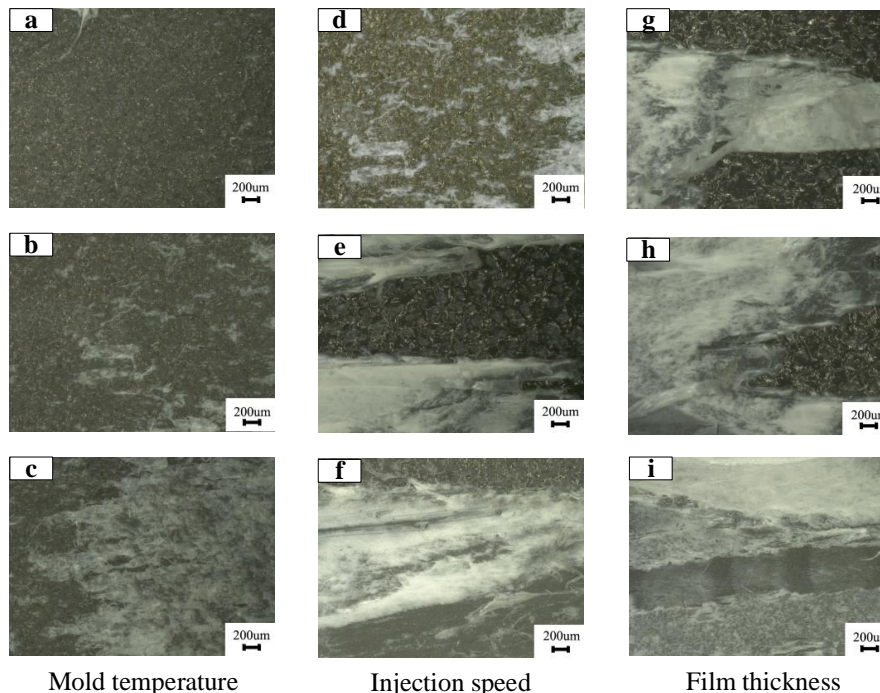


Figure. 9. Interface morphology of the substrate after 90° peeling test (a)240°C-30°C; (b) 240°C-50°C; (c) 240°C-70°C; (d) 10mm/s; (e) 40mm/s; (f) 70mm/s; (g) 0.4mm; (h) 0.5mm; (i) 0.6mm

Figure 9 shows the interface morphology of 90° peeling test under different injection conditions. Figure 9(a)-(c) shows the substrate morphology of different mold temperature with film thickness of 0.4mm. For lower temperature of 30°C there is rarely PP film left on the substrate indicating poor interface adhesion of adhesive type. But for higher temperature of 50°C, there are relatively more PP

film remain after peeling test and the corresponding peeling force in figure 4 rises to 280N/mm. At mold temperature of 70 °C, transition of interface bonding to cohesive type starts. Morphology indicates the mixing mode of two types. Interface morphology of different injection speed is illustrated in Figure 9(d)-(f): peeling force rise from 1729N/mm to 3739 N/mm. large area of PP film is left on the surface showing the bonding is sufficient to meet the requirements of use. In Figure 9(f) the PP film covers most area of the substrate. Figure 9(g)-9(i) is the morphology of different film thickness with melt and mold temperature of 240°C-70°C. Similar discovery is found that peeling force (shown in figure 6) is in positive correlation with cover area of PP film. The larger covered area is, the higher peeling force becomes. The reason is that for cohesive bonding the interface is melt and there are more molecular entanglement and crystallization near the interface leading to higher energy that is needed to break entanglement and crystal.

4. Conclusion

Bi-layer parts composed by PP film/substrate combination have been successfully produced by film insert molding technique. This research performs a CAE simulation investigating the effect of the film used in the IMR process on the temperature field of the mold and its impact on crystallinity as well as mold temperature. Simulation shows that for thicker film and higher mold temperature, there is higher interface temperature due to heat conduction obstruction and lower cooling rate. Results of the study show that interface bonding is significantly affected by melt temperature, injection speed, mold temperature and film thickness. Three types of peeling curves are obtained suggesting different bonding types, namely, adhesive, cohesive and mixed type. It is shown that interface crystallinity of FIM parts is altered by changing film thickness because film acts as an insulator hindering heat conduction from the resin to mold, which leads to lower cooling rate. Effects of mold temperature on crystallinity are more significant due to higher mold temperature and lower cooling rate resulting in higher crystallinity. The interface crystallinity rises linearly with mold temperature rising. With the intrinsic characteristic of viscous fluids, resin temperature rises during injection process leading to faster crystallization rate. For faster injection speed, there is higher crystallinity. Our study shows that crystallinity at the film-substrate interface region influences film-substrate adhesion. For adhesive and transition type, effects of crystallinity are significant. Higher crystallinity of the substrate due to obstructed cooling during injection molding is believed to contribute to interface bonding. Interestingly for interface bonding of cohesive type, the crystallinity rises little with injection molding parameters and molecular entanglement affects interface strength a lot due to more active molecular movement and more effective entanglement. This suggested that crystal plays a decisive role in the initial stage namely adhesive type of interface bonding and leads to bonding type conversion. When it comes to cohesive bonding, molecular entanglements take the leading role dominating interface bonding strength.

References

- [1] Yan, F.Y., et al., Peel-strength behavior of bilayer thermal-sprayed polymer coatings.[J] *Journal of Applied Polymer Science*, 2003. 88(1): p. 214-226.
- [2] Nguyen, S., et al., Adhesion control for injection overmolding of elastomeric propylene copolymers on polypropylene. Effects of block and random microstructures.[J] *International Journal of Adhesion and Adhesives*, 2013. 46: p. 44-55.
- [3] Dondero, M., et al., Adhesion control for injection overmolding of polypropylene with elastomeric ethylene copolymers.[J] *Polymer Engineering & Science*, 2009. 49(10): p. 1886-1893.
- [4] Stan, F. and C. Fetecau. Experimental Investigation of the Adhesion Between Thermoplastic Polyurethane and Acrylonitrile-Butadiene-Styrene Substrate[C]. in *ASME 2014 International Manufacturing Science and Engineering Conference collocated with the JSME 2014 International Conference on Materials and Processing and the 42nd North American Manufacturing Research Conference*. 2014. : American Society of Mechanical Engineers.
- [5] Rossa-Sierra, A., et al., Study of the interface behaviour between MABS/TPU bi-layer structures obtained through over moulding.[J] *Materials & Design*, 2009. 30(10): p. 3979-3988.

- [6] Lee, H.S. and J.R. Park, Experimental study of injection-compression molding of film insert molded plates.[J] *International Journal of Precision Engineering & Manufacturing*, 2014. 15(3): p. 455-461.
- [7] Leong, Y., et al., The effect of molding conditions on mechanical and morphological properties at the interface of film insert injection molded polypropylene-film/polypropylene matrix.[J] *Polymer Engineering & Science*, 2004. 44(12): p. 2327-2334.
- [8] Yamaguchi, S., et al., Effect of crystallization and interface formation mechanism on mechanical properties of film-insert injection-molded poly(propylene) (PP) film/PP substrate.[J] *Journal of Applied Polymer Science*, 2005. 98(1): p. 294-301.
- [9] Leong, Y., et al., Interfacial characteristics of film insert molded polycarbonate film/polycarbonate-acrylonitrile-butadiene-styrene substrate, part 1: Influence of substrate molecular weight and film thickness.[J] *Polymer Engineering & Science*, 2006. 46(12): p. 1674-1683.
- [10] Leong, Y., M. Kotaki, and H. Hamada, Effects of the molecular orientation and crystallization on film-substrate interfacial adhesion in poly (ethylene terephthalate) film-insert moldings.[J] *Journal of Applied Polymer Science*, 2007. 104(4): p. 2100-2107.
- [11] Baldan, A., Adhesion phenomena in bonded joints.[J] *International Journal of Adhesion and Adhesives*, 2012. 38: p. 95-116.
- [12] Awaja, F., et al., Adhesion of polymers.[J] *Progress in Polymer Science*, 2009. 34(9): p. 948-968.
- [13] Creton, C., et al., Adhesion and Fracture of Interfaces Between Immiscible Polymers: from the Molecular to the Continuum Scale, in *Molecular Simulation Fracture Gel Theory*. 2001, Springer. p. 53-136.
- [14] Cole, P.J., R.F. Cook, and C.W. Macosko, Adhesion between immiscible polymers correlated with interfacial entanglements.[J] *Macromolecules*, 2003. 36(8): p. 2808-2815.
- [15] Boucher, E., et al., Enhanced adhesion between polypropylene and polyamide-6: role of interfacial nucleation of the β -crystalline form of polypropylene.[J] *Macromolecules*, 1997. 30(7): p. 2102-2109.
- [16] Laurens, C., et al., Role of the interfacial orientation in adhesion between semicrystalline polymers.[J] *Macromolecules*, 2001. 34(9): p. 2932-2936.
- [17] Lo, C.-T., F.C. Laabs, and B. Narasimhan, Interfacial adhesion mechanisms in incompatible semicrystalline polymer systems.[J] *Journal of Polymer Science Part B: Polymer Physics*, 2004. 42(14): p. 2667-2679.
- [18] Yan, B., et al., Interfacial crystalline structures in injection over-molded polypropylene and bond strength.[J] *ACS applied materials & interfaces*, 2010. 2(11): p. 3023-3036.
- [19] Lin, Y.-H., et al., Effect of asymmetric cooling system on in-mold roller injection molded part warpage.[J] *International Communications in Heat and Mass Transfer*, 2015. 61: p. 111-117.
- [20] Koch, P. and M. McCarren. Developing a Surface Texture with Film Insert Molding (FIM)(916)[C]. in *TECHNICAL PAPERS OF THE ANNUAL TECHNICAL CONFERENCE-SOCIETY OF PLASTICS ENGINEERS INCORPORATED*. 2000.