

Hot-Melt Adhesion and Wettability between Polyethylene and Other Polymers in the Vicinity of the Adherend Melting Point

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Synopsis

The effect of bonding temperature on the bonding strength of polyethylene to polypropylene or other polymers was investigated. It became clear that the interfacial bonding strength reached maximum strength at a bonding temperature in the vicinity of the melting point of the adherend (i.e., the polymer having the higher melting point). The effect of temperature on the contact angle of the molten polymer having the lower melting point on the adherend surface was also investigated. The wettability of the adherend by the molten polymer drop was considerably reduced at the temperature near the melting point of the adherend. The relationship of increasing adhesiveness to decreasing wettability was found out.

INTRODUCTION

If different kinds of thermoplastic polymers in contact with each other are heated, adhesion occurs between the two when the polymer having the lower melting point melts. The bonding strength normally increases with bonding temperature.¹⁻³ However, if the polymer having the higher melting point also melts, the resulting interfacial bonding behavior has not been made clear. Yamakawa³ and Sung and Suh¹ have shown an effect of bonding temperature on the bonding strength of a joint of two polymers which were bonded at the temperature exceeding the melting point of the adherend. However, the interfacial bonding strength was not made clear because the failure mode of the joint was not interfacial but cohesive. In the present study, hot-melt adhesion was performed at several bonding temperatures, including the melting point of adherend and the effect of bonding temperature on the interfacial bonding strength was investigated. This report employs the term "adherend" for one of a pair of polymers having the higher melting point. To make the plane of separation be at the interface, the interfacial bonding strength should not be too high. Hence, no pressure other than a contact pressure and a pressure of 1 atmosphere of argon was applied to the test specimens during the bonding process, and untreated polyethylene was used for one of the polymer pairs because it cannot be bonded to give high bond strength for most applications.⁴

The formation of a good adhesive joint requires intimate contact between the adhesive and the adherend,⁵ that is, the extent of interfacial contact is a major factor in establishing the performance.⁶ Such contact can be termed the wetting of the adherend by the adhesive.⁵

Then, wetting of adherend by molten adhesive drop was measured for the same combination of polymers as used for the bonding test.

TABLE I
Test Materials

Sample	Material and supplier	Melting point (°C) (DSC peak)	Density (g/cm ³)	Tensile strength (kg/cm ²)
PE	Low density polyethylene (16P: Mitsui Poly-chemical Co.)	110	0.92	100
PP	Polypropylene (J109G: Ube Industries Co.)	162	0.91	> 360
EVA-1	Ethylene-vinyl acetate copolymer (UE634: Toyosoda Manufacturing Co.)	79	0.95	130
EVA-2	Ethylene-vinyl acetate copolymer (EVAL-E: Kuraray Co.)	164	1.14	470
NYLON6-66	copolymer nylon (5013B: Ube Industries Co.)	198	1.14	> 270
EPOXY	Epoxy resin (Epikote 828: Shell Chemical Co.)/ 2-ethyl-4-methyl-imidazol in 100/4 ratio	—	1.18	850

EXPERIMENTAL

Materials

Low density polyethylene (PE), polypropylene (PP), nylon (NYLON:6-66 copolymer), two kinds of ethylene-vinyl acetate copolymer (EVA-1, EVA-2), and epoxy resin (EPOXY) were used for test materials. All these polymers, except EPOXY, supplied in the form of pellets were molded into the form of a sheet having 2 mm thickness except in the case of PE. The thickness of PE sheet was 0.20 or 0.25 mm. PE contained no additives. The test materials are shown in Table I together with their properties; melting point (differential scanning calorimetry, DSC peak), density, and tensile strength.

Hot-Melt Adhesion

Hot-melt adhesion of the systems of PE/PP, PE/EVA-2, PE/NYLON, and EVA-1/PE was performed. A polymer sheet having the lower melting point was laid over another polymer sheet. The pair was placed in an oven maintained at a predetermined bonding temperature. Then, the air in the oven was rapidly pumped out. At the moment when the polymer having the lower melting point began to melt, the oven was filled to overflowing with argon gas. By this operation, the formation of a gas bubble at the interface between two sheets was prevented. The specimen was kept at the bonding temperature for 2 h for the PE/PP system, 1 h for the PE/NYLON and PE/EVA-2 system, and 30 min for the EVA-1/PE system.

Adhesion of the EPOXY/PE system was also examined. Placing a PE sheet on the bottom of a vessel, the vessel was put into the thermostated oven and kept at a predetermined bonding temperature after the air in the vessel was replaced with argon gas. Then EPOXY consisting of 100 parts by weight of epoxy resin and 4 parts by weight of imidazol was poured on the PE sheet. The result was an EPOXY layer, 3 mm thick. The test materials were kept at

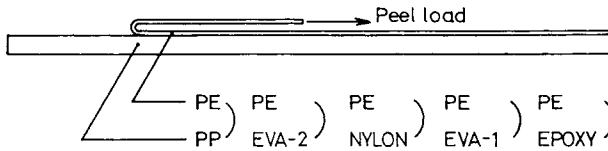


Fig. 1. Schematic drawing of peeling test.

this bonding temperature for 1 h. The EPOXY had cured within about 30 min after being poured.

These specimens were subjected to 180° peel test at 23°C, in which the PE sheet was peeled off, as shown in Figure 1. The cross head speed was 50 mm/min. The average force was taken as the peel strength.

Contact Angle Measurement

A spherical piece of about 1.5 mm in diameter was prepared from one of a pair of polymers (EVA-1/PE, PE/PP, or PE/EVA-2) having the lower melting point and placed on a surface of the other polymer of the pair and premelted in an environmental oven to adhere to the substrate surface. The test surface with the spherical piece was placed upside down in an environmental chamber. This retards or prevents the spherical piece from penetrating into the surface due to its weight, which was softened at a temperature near the melting point of the substrate. The chamber was heated and maintained in an oven at a certain temperature monitored by a chromel-alumel thermocouple. When the shape of the piece melted and spread over the substrate, it was photographed with a camera from the horizontal angle. The contact angle of the molten drop on the photograph was measured by a protractor.

The effect of gravity on contact angle of the 1.5 mm diameter molten drop used in this experiment was estimated to be negligible, because the lineal dimension of the drop L is smaller than $(2\gamma/\rho g)^{1/2}$, where (γ) is the surface tension of the molten polymer, (ρ) the density, and (g) the gravitational acceleration.⁷

RESULTS AND DISCUSSION

Effect of Bonding Temperature on Bonding Strength

Figure 2 shows the relationships between bonding temperature and peel strength for the PE/PP and PE/EVA-2 systems, and Figure 3 gives the results of the PE/NYLON system. In Figure 3, the PE sheet of the specimen bonded at the temperature near the melting point of NYLON (198°C) was broken by peeling. As shown in these results, the peel strength reaches its maximum at bonding temperature in the vicinity for the melting point of each adherend and is reduced when the bonding temperature exceeds it.

The results of the EVA-1/PE and EPOXY/PE systems are shown in Figure 4. It is evident that the peel strength for the EVA-1/PE system has its maximum at the bonding temperature near the melting point of PE, 110°C. The solidifying process of the EPOXY/PE system is different from those of the other systems because EPOXY has finished to cure while heating. A similar result is obtained in EPOXY/PE specimens. Visual inspection for the

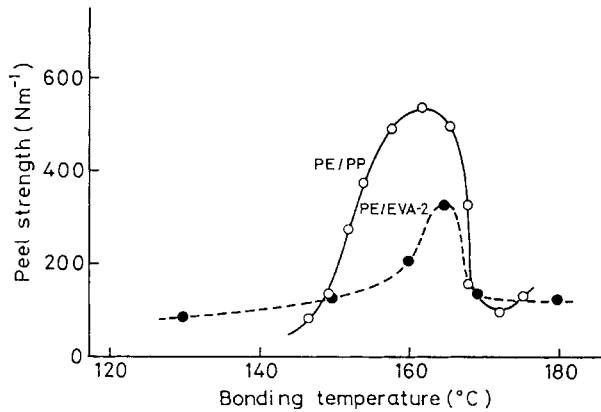


Fig. 2. Effect of bonding temperature on peel strength for PE/PP (O—O) and PE/EVA-2 (●---●) systems.

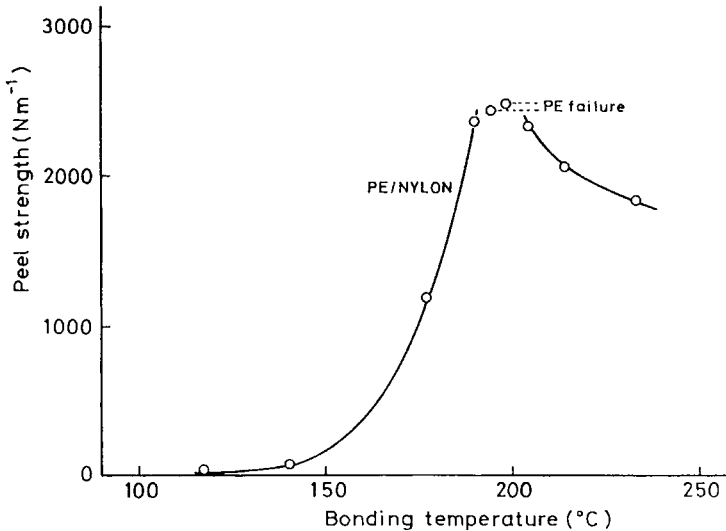


Fig. 3. Effect of bonding temperature on peel strength for PE/NYLON system (O---, PE failure).

specimen surfaces obtained by peeling revealed that fractures occurred because of interfacial failure except for the case of PE/NYLON specimens bonded at a temperature close to the melting point of NYLON.

It is also obvious from the following that the failure mode was interfacial. PE is considered to have the lowest strength among those polymers used in this experiment. Hence, if the failure mode is not interfacial, the failure has to occur in the PE layer and the observed peel strength should depend on the strength of PE. However, the peel strength was unrelated to the strength of PE. Therefore, the peel strength corresponds to interfacial bonding strength.

The influences of the bonding pressure and the bonding time on the peel strength were examined for the PE/PP systems by shortening the bonding time to 10 min and by applying a bonding pressure to a maximum of 2.3 MPa.

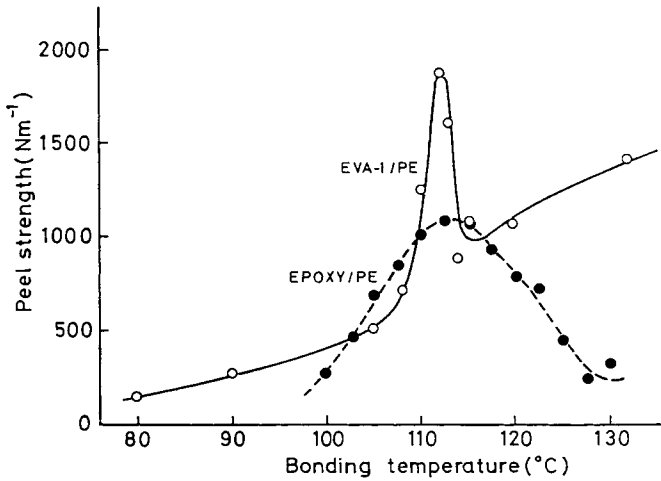


Fig. 4. Effect of bonding temperature on peel strength for EVA-1/PE (○—○) and EPOXY/PE (●---●) systems.

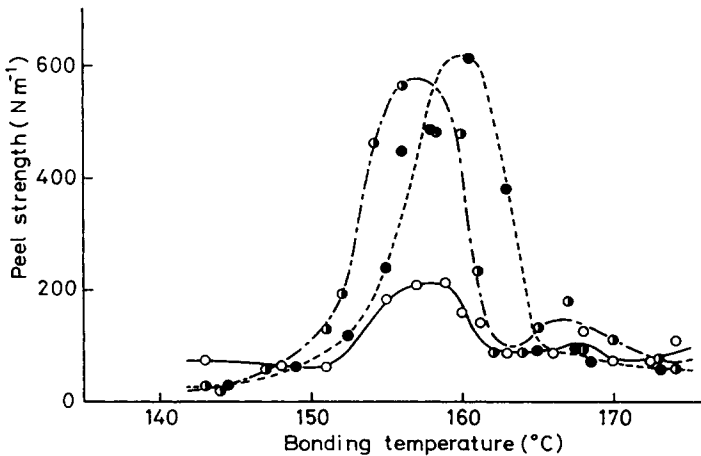


Fig. 5. Effect of bonding pressure (○—○) 0 MPa; (○---○) 1.1 MPa; (●---●) 2.3 MPa) on peel strength for PE/PP system.

The results are shown in Figure 5. It is evident that even when the bonding time is short and regardless of whether bonding pressure is applied or not, the peel strength has its maximum at a temperature in the vicinity of the melting of PP. In conclusion, the bonding strength between two polymers reaches its maximum at the temperature where the adherend in the solid state changes to the liquid state.

Interfacial Topography

Bonding strength may be affected by the interfacial topography.⁸ Then, the tendency for peel strength to vary with bonding temperature was checked if it was caused by the interfacial topography or roughness.

When the adherend having a higher melting point has been solidified during bonding, the polymer having a lower melting point is still molten. Hence, the

interfacial topography of the bonded specimen is considered to depend on the topography of the adherend surface and can be estimated by heat treating only the same polymer as the adherend under the same conditions as in the bonding procedures and observing its surface.

The surfaces of EVA-2, PP, and PE which were the same polymers as the adherend used in the PE/EVA-2, PE/PP, and EVA-1/PE systems, respectively, were heat-treated and observed by an electron microscope.

There was no remarkable difference in surface roughness between the surface of EVA-2 heat-treated at 164°C, which was indicative of the interface of the specimen with high peel strength and that heat-treated at 175°C, which was indicative of the interface with a lower peel strength.

The surface of PE corresponding to the adherend of the EVA-1/PE specimen was tested the same way, but, it was difficult to find in the roughness a clear cause of the difference in peel strength.

The surface of PP gradually became rougher after the temperature exceeded 140°C but showed a smooth condition after the temperature exceeded 170°C. The same tendency was observed with the surface of the PP adherend after the PE sheet was peeled off. It is probable that interfacial surface roughness had some effect on the peel strength in the case of PE/PP system. However, taking into consideration the surface conditions shown by adherends other than PP, it is difficult to explain the common phenomenon that the peel strength has a peak value only by surface roughness.

Wettability

For every system, the contact angle reached an equilibrium after a time lapse of about 2×10^3 min. Here, the cosine of contact angle, extrapolated to infinite time, is used as wettability.

Figure 6 shows the relationship between the temperature and the wettability of the PP surface by the molten PE drop. The wettability is considerably reduced at a temperature near the melting point of PP and increased to some extent at a temperature above the melting point.

At about 170°C where PP is completely molten, the bottom of the PE drop sinks into the PP substrate and the contact angle cannot be measured.

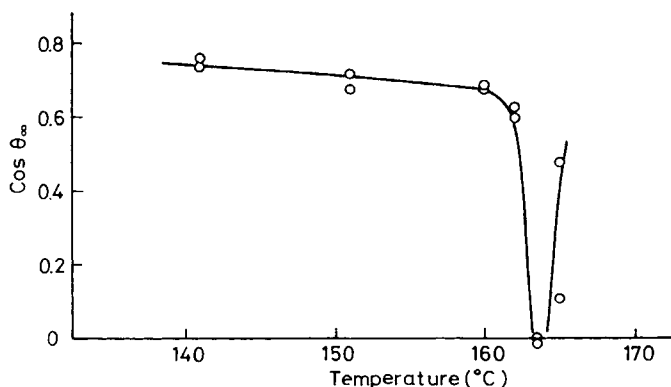


Fig. 6. Relation between temperature and wettability of PP surface by molten PE drop.

The wettability of the EVA-2 surface by the molten PE drop was reduced at a temperature close to the melting point of the EVA-2. This tendency of the wettability being reduced near the melting point of the substrate was observed too, in the case of the EVA-1/PE system. Thus, it is clear that the wettability becomes worst at a temperature near the melting point of the substrate, and the maximum bonding strength is obtained at that temperature.

Reports that good wettability produces good bonding are often found. For instance, Barbarisi⁵ reported that the bonding strength between epoxy adhesive and PE with different degrees of surface treatment was proportional to the wettability of the PE surface by epoxy adhesive drop. Raraty and Tabor⁹ recognized that smaller contact angle of water drops on various solid surfaces brings better adhesion of the ice to the surface.

In the present study, the inverse relation to these instances has been recognized. Therefore it is difficult for the adhesive strength and the wettability to be directly related to each other.

CONCLUSION

Effect of bonding temperature and the interfacial bonding strength of polyethylene to polypropylene and other polymers was investigated. Polyethylene, which has a poor bonding force, was used for one of a pair of polymers to make the plane of separation be at the interface.

The interaction between polyethylene and the other polymers is considered to differ with each system. As a result, the interfacial bonding strength differed with each system. But it has been shown in all systems that the bonding strength had its maximum at the bonding temperature near the melting point (DSC peak) of the adherend, that is, the bonding between two kinds of polymers proceeded most completely at a temperature where the adherend changes from a solid phase to a liquid phase. The wettability clearly becomes worst at the same temperature mentioned above. The results obtained in this study are the opposite of other instances where good wettability produces good bonding, and accordingly the interfacial bonding strength is considered to be determined by some factor other than wettability.

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Received June 5, 1985

Accepted March 3, 1987