

Friction Welding of Similar Materials: Polypropylene, High-Density Polyethylene, and Nylon-6

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ABSTRACT: The effects of spin-friction pressure and timing on the friction welding between parts of the same materials, PP, HDPE, and nylon-6, are discussed. The heat-affected zone is divided into three regions: the plasticized region (Zpl), the partly plasticized region (Zpd), and the undeformed region (Zud). The tensile fracture morphology has three main sections: the nonplasticized central section (Fud), the plasticized peripheral section (Fpl), and the partly plasticized section between Fud and Fpl (Fpd). Friction pres-

ures (spinning timing) of 1.17 MPa (16 s) in PP–PP welding, 1.77 MPa (8 s) in HDPE–HDPE welding, and 0.98 MPa (8, 10, and 12 s) in nylon-6–nylon-6 welding attains better bonding strength because their components have larger Fpd and Fpl regions, which have no Fud in the central part. © 2004 Wiley Periodicals, Inc. *J Appl Polym Sci* 91: 2771–2780, 2004

Key words: friction welding; polypropylene (PP); high-density polyethylene (HDPE); nylon-6; bonding strength

INTRODUCTION

Mechanical locking,¹ adhesive bonding,² solvent welding,³ and welding⁴ are all commonly used methods for joining thermoplastics. Thermoplastic welding mainly involves eight methods: (1) hot-plate welding⁵; (2) hot-gas welding⁶; (3) infrared-heating welding⁷; (4) resistive-implant welding⁸; (5) spin-friction welding⁹; (6) vibration or linear-friction welding¹⁰; (7) ultrasonic welding¹¹; and (8) laser welding.¹² Spin-friction welding is a process in which one substrate is fixed whereas the other is given a controlled angular velocity and brought into contact with the former to cause friction heating and agitation to form a bond. Advantages of spin-friction welding are high welding quality, simplicity, speed, and reproducibility.

Wool and O'Connor¹³ reported five steps for thermoplastic welding: surface rearrangement, surface approach, wetting, molecular chain interdiffusion, and randomization. The latter two steps formed the main parameters that control welding strength, and by the end of the wetting step, potential barriers that combined with inhomogeneities in the interface disappear and molecular chains are free to move across the interface in the subsequent steps of interdiffusion and randomization. According to Kim and Wool¹⁴ molec-

ular chain interdiffusion and randomization could be illustrated through the reptation model,¹⁵ proposed by de Gennes.¹⁶ According to Kausuh and Jud¹⁷ welding strength was attributed to entanglement of molecular chains after interdiffusion. According to Wool et al.¹⁸ the operating temperature should be higher than the glass-transition temperature (T_g) during hot-press welding because with the help of thermal activity minor chains could interdiffuse easily by untying entangled molecular chains. If the operating temperature were lower than T_g , the welding phenomenon would not occur.¹⁹

The present research was aimed at understanding the effects of spin-friction pressure and timing on friction welding between parts of the same material, for example, PP with PP, HDPE with HDPE, and nylon-6 with nylon-6.

EXPERIMENTAL

Materials preparation

PP rods (Clap Co.; 8400R), HDPE rods (USI Co.; LS 606), and nylon-6 rods (Missisipi Co.; 1010C) were cut for welding specimens whose standard is shown in Figure 1. Carbimet paper (1200 grit) was used for polishing the welding surfaces, and finally polishing with 1 μm Al_2O_3 slurries followed. Polishing was performed during friction pressure for two specimens to have close contact. Then, the specimens were kept in an air furnace at $50 \pm 2^\circ\text{C}$ for 24 h for annealing, and then the annealed specimens were cooled to room temperature to eliminate residual stress on the welding surface.

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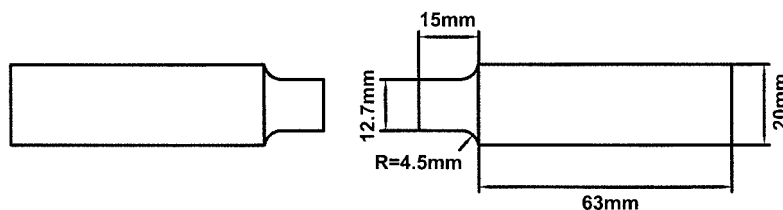


Figure 1 Schematic of specimens.

Spin-friction welding

An apparatus for spin-friction welding of PP with PP, HDPE with HDPE, and nylon-6 with nylon-6 is shown in Figure 2. Welding surfaces of the work pieces were degreased before welding. During spin-friction welding, one specimen was fixed while the other remained stationary. Three different spin-friction pressures (0.49, 1.18, and 1.37 MPa) were used for PP-PP welding with an ac motor of 460 rpm for 16, 18, and 20 s to closely join the two specimens. Then, 5 s of forge pressure, the same as the spin friction pressure, was immediately applied to complete the welding. The same process as for PP-PP welding was used for HDPE-HDPE welding with 0.98, 1.37, and 1.77 MPa friction pressures exerted on the specimens for 8, 10, and 12 s; and nylon-6-nylon-6 welding with 0.49, 0.784, and 0.98 MPa friction pressures exerted on the specimens for 12, 16, and 20 s.

Metallographic preparation

The welded specimen sections were polished with 1200-grit carbimet paper and suspension polishing liquid of 1 μm Al_2O_3 particulates. Then, the microstruc-

ture of the heat-affected zone was observed through an Optiphot-100 Nikon optical microscope (Nikon, Tokyo, Japan).

Tensile test

Tensile tests were performed at room temperature at a crosshead speed of 1 mm/min on an Instron tensile machine (Instron, Canton, MA), after fringes of welded specimens were machined. The bonding strengths were calculated from the load at failure, divided by the original cross-sectional area. Three measurements were taken for each of the bonding strengths to calculate their average data. After 4 min of gilding of the tensile fracture surface, tensile fracture morphology was observed through a JEOL-JSM 840A SEM (JEOL, Tokyo, Japan).

RESULTS AND DISCUSSION

Heat-affected zone

The schematic diagram of the heat-affected zone (HAZ) of welded PP with PP is shown in Figure 3. Three regions that are distinctly evident in the plasti-

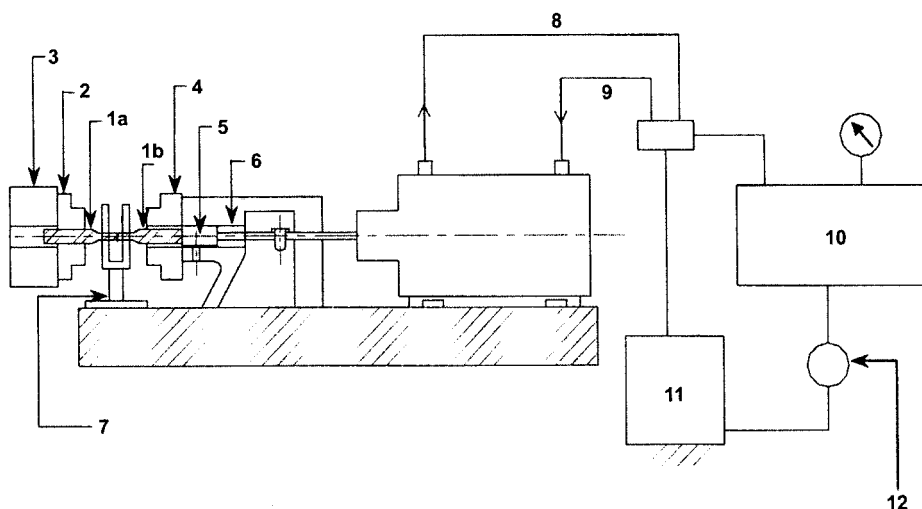


Figure 2 Schematic of spin-friction welding system: (1a, 1b): specimen; (2) rotational clip; (3) brake system; (4) fixed clip; (5) piston; (6) cylinder; (7) supporter; (8) oil outlet; (9) oil inlet; (10) oil pressure adjuster; (11) oil tank; (12) oil pressure pump.

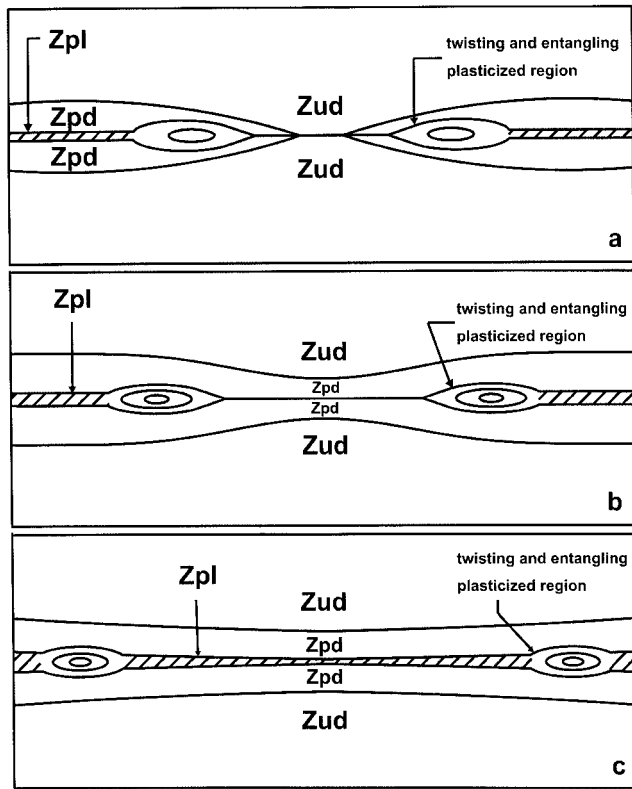


Figure 3 Schematic of heat-affected zone of welded PP with PP.

cized region (Zpl), the undeformed region (Zud), and the partly plasticized region (Zpd), which the friction heat and thermal transients experienced in the Zpl and the Zpd zone, are sufficiently high to dissolve and soften the polymer matrix, respectively. The width of

the Zpd zone and the area of the Zpl zone are increased with increasing spinning time. Under the spinning process, the heat-affected zone produces larger plasticization near the central zone, and deviation tends to arise in spinning, which causes the active molecular chains to become a “twisting and entangling plasticized region” around both sides of the central zone, after which they gradually spun along the radius to the peripheral zone, a phenomenon that increased with spinning time.

When the HAZ of welded PP with PP was under 0.98 MPa friction pressure with 16, 18, and 20 s spinning timing, the Zpd zone was formed in the central zone. The area of the Zpd and Zpl zone and the distance of both the “twisted and entangled plasticized regions” in the central zone increased with increasing spinning time, as shown in Figure 4(a) and (b). A friction pressure of 1.17 MPa with a spinning time of 16 s produced an apparent Zpl zone in the central zone, as shown in Figure 4(c). For spinning times of 18 and 20 s, only the Zpd zone was formed in the central zone, and the area of the Zpd zone decreased with increasing spinning time. At a friction pressure of 1.37 MPa with spinning times of 16 and 18 s, a smaller Zpd zone was formed in the central zone. For a spinning time of 20 s, formation of the Zpd zone cannot be observed in the central zone, as shown in Figure 4(d).

The schematic diagram of the HAZ of welded HDPE with HDPE is shown in Figure 5. It is also divided into three main regions: the Zud zone, the Zpd zone, and the Zpl zone. Although the friction heat in the central zone is small at the beginning stage of spinning, the friction temperature still exceeded the

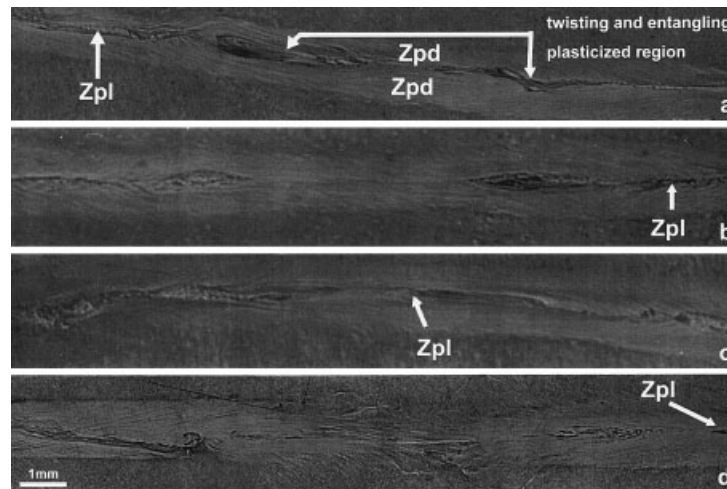


Figure 4 OM morphologies for heat-affected zone of welded PP with PP, under 0.98 MPa friction pressure with (a) 18 s and (b) 20 s of spinning timing; (c) under 1.17 MPa friction pressure with 16 s of spinning time; (d) under 1.37 MPa friction pressure with 18 s of spinning time.

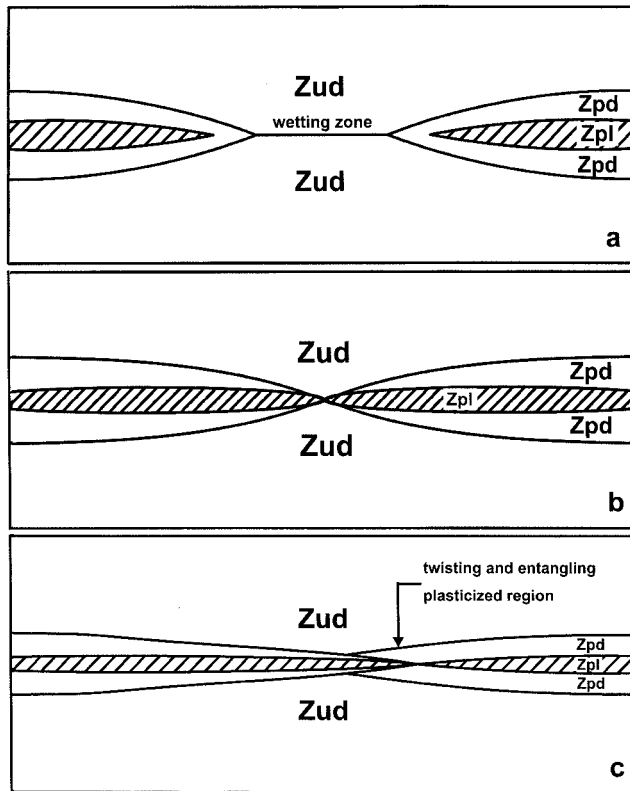


Figure 5 Schematic of heat-affected zone of welded HDPE with HDPE.

glass-transition temperature of HDPE ($\sim -105^{\circ}\text{C}$); thus the active minor chains that thrust through one side of the interface easily wet each other in the central zone, as shown in Figure 5(a). Areas of the Zpd and the Zpl zones increased with the spinning time, as shown in Figure 5(a) and (b). When the central zone produced larger plasticization, deviation tends to arise in spinning, which causes active molecular chains to become entangled around the central zone. Consequently, a "twisting and entangling plasticized re-

gion" was formed. At this time, melting of the polymer formed a tacky layer in the Zpl zone, and while the forge pressure was being applied to the HAZ, the Zpl zone was easily squeezed because of the forging action, after which the total width of the Zpd and Zpl zones was decreased, as shown in Figure 5(c).

In the HAZ of welded HDPE with HDPE under 0.98 MPa friction pressure, for spinning times of both 8 and 10 s, only a small Zpd zone was formed in the central zone, as shown in Figure 6(a). For a spinning time of 12 s, only the wetting zone was formed in the central zone, as shown in Figure 6(b). Under a friction pressure of 1.37 MPa, for spinning times of both 8 and 10 s, the Zpl zone was apparently formed in the central zone, and the area of the Zpl zone decreased with increasing spinning time. For a spinning time of 12 s, only the wetting zone was formed in the central zone. Under a friction pressure of 1.77 MPa, for spinning times of both 8 and 10 s, a "twisting and entangling plasticized region" was formed in the central zone, as shown in Figure 6(c). For a spinning time of 20 s, only a small Zpd zone was formed in the central zone.

The schematic diagram of the HAZ of welded nylon-6 with nylon-6 is shown in Figure 7. It is also divided into three regions: the Zud zone, the Zpd zone, and the Zpl zone. When the central zone produced apparent plasticization, the active molecular chain became entangled around the central zone because the tangent velocity of the central zone is smaller. Consequently, a larger "twisting and entangling plasticized region" was formed in the central zone. As this time, the flow of the melting polymer formed a tacky layer in the Zpl zone, which was squeezed while the forge pressure was being applied, thus causing both Zpd zones to undergo mutual wetting, as shown in Figure 7(a). The plasticization of the "twisting and entangling plasticized region" increased with increasing spinning time. Finally, the "twisting and entangling plasti-

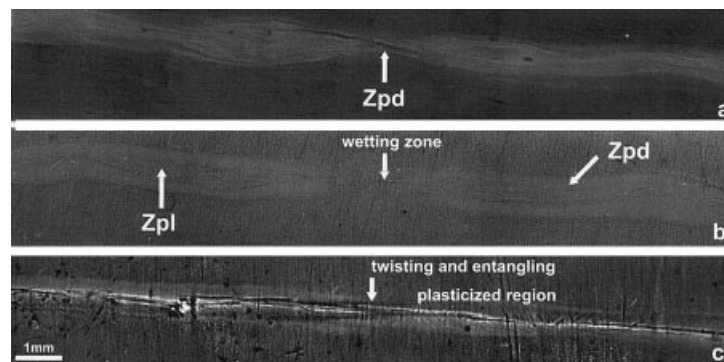


Figure 6 OM morphologies for heat-affected zone of welded HDPE with HDPE, under 0.98 MPa friction pressure with (a) 10 s and (b) 12 s of spinning timing; (c) under 1.77 MPa friction pressure with 10 s of spinning time.

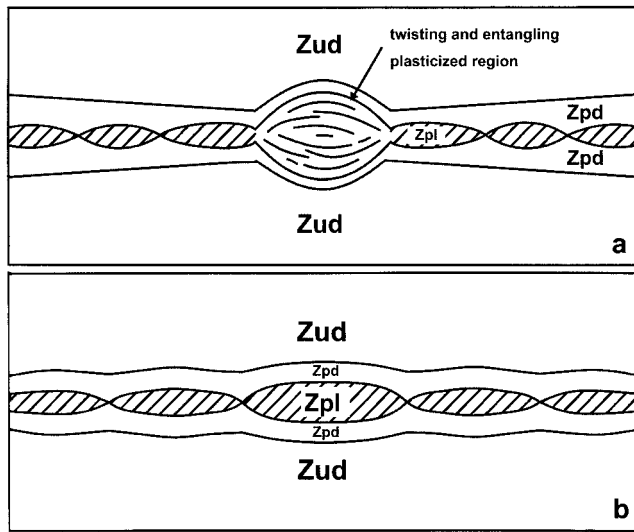


Figure 7 Schematic of heat-affected zone of welded nylon-6 with nylon-6.

cized region" converted completely into the Zpl zone, as shown in Figure 7(b).

In the HAZ of welded nylon-6–nylon-6 under a friction pressure of 0.49 MPa, for spinning times of 12, 16, and 20 s, a larger "twisting and entangling plasticized region" was formed in the central zone, as shown in Figure 8(a). Under a friction pressure of 0.78 MPa, for spinning times of 12, 16, and 20 s, the Zpl zone was apparently formed and both of the Zpd zones underwent mutual wetting, as shown in Figure 8(b). Comparing with Figure 8(b), under a friction pressure of 0.98 MPa, for spinning times of 12, 16, and 20 s, a larger Zpl zone was formed in the heat-affected zone, as shown in Figure 8(c).

Tensile fracture morphology

The tensile fracture morphology of welded PP with PP under different spin friction pressures and spinning

times is shown in Figure 9. It has three main sections: (1) the central zone (Fud), which has no plasticized "wearing surface" morphology; (2) the peripheral zone (Fpl), which has a plasticized "dimple" fracture morphology; and (3) the middle zone (Fpd), which has a partly plasticized "dot" fracture morphology. The dimensions of the three zones change with different friction pressures and spinning times. The result for forming the "dot" fracture morphology may arise for two reasons: (1) the PP is a semicrystalline polymer with a spherulitic structure, and the local crystallization often produces a mechanically interlocked interface; (2) the tensile process is accommodated by elongation at entanglement welded points. Molecular chains of the welded points are oriented along the tensile direction, so when the area of local welded points could not support the tensile stress, fracturing occurred at this strained welding "dot." With a tensile fracture morphology of welded PP with PP under a friction pressure of 0.98 MPa, the area of the Fud zone decreased with increasing spinning times; the tensile fracture morphology at a spinning time of 20 s is shown in Figure 9(a). The tensile fracture morphology under a friction pressure of 1.17 MPa for a spinning time of 16 s is shown in Figure 9(b). It was found that only the Fpd zone formed in the fracture surface. For spinning times of 18 and 20 s, the Fud zone was formed in the central zone and the other region was the Fpd zone [shown in Fig. 9(c)], although the area of the Fpd zone at a spinning time of 18 s was larger than that at a spinning time of 20 s. For the tensile fracture morphology under a friction pressure of 1.37 MPa for spinning times of 16, 18, and 20 s, the Fud zone was formed in the central zone, and the other zone was the Fpd zone, in which the Fud zone area is broadened with spinning time.

The tensile fracture morphology of welded HDPE with HDPE under different spin friction pressures and spinning times is shown in Figure 10. It also has three main regions: (1) the central section (Fud), which has

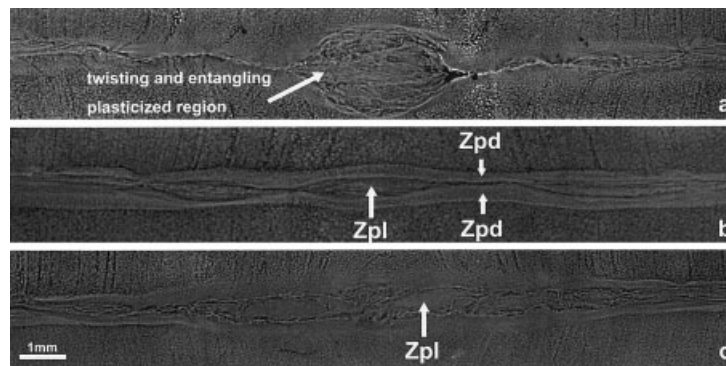


Figure 8 OM morphologies for heat-affected zone of welded nylon-6 with nylon-6, under (a) 0.49 MPa; (b) 0.784 MPa; and (c) 0.98 MPa friction pressure with 20 s of spinning time.

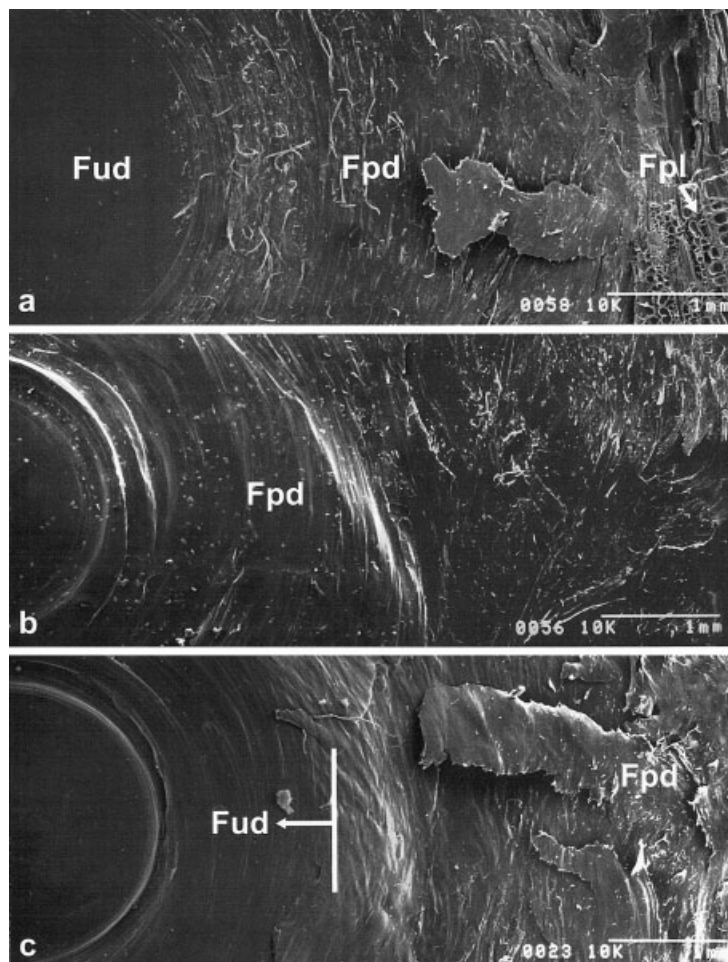


Figure 9 SEM micrographs of tensile fracture morphologies of welded PP with PP: (a) under 0.98 MPa friction pressure with 20 s of spinning timing; (b) under 1.17 MPa friction pressure with 16 s of spinning time; (c) under 1.37 MPa friction pressure with 20 s of spinning time.

no plasticized “wearing surface” morphology; (2) the peripheral section (Fpl), which has a plasticized “patch” fracture morphology; (3) the middle section (Fpd), which has a partly plasticized “mixed fibrillate with ribbon” fracture morphology. Formation of the “fibrillate” or “ribbon” may be attributed to the following. The HDPE is a linear semicrystalline polymer and the molecular chains are mutually entangled rather deeply in the Zpl zone. Under the tensile process, the extensive fibrils were pulled from the welded plane, resulting in a fine fibrillated or ribbon fracture morphology. The tensile fracture morphology of welded HDPE with HDPE under a friction pressure of 0.98 MPa, a small Fud zone and an Fpl zone were formed in the central zone and near the peripheral zone, respectively; the greater area was the Fpd zone, as shown in Figure 10(a), and the Fud zone area increases with increasing spinning time, as shown in Figure 10(b). Tensile fracture morphologies, under a friction pressure of 1.37 MPa, for the small Fud zone

and the Fpd zone were formed in the central zone and near the central zone, respectively. Moreover, it was found that the greater of the Zpl zones gradually spun along the radius to the peripheral zone, as shown in Figure 10(c); the area of the Fpl zone under a spinning time of 10 s was smaller than that under a spinning time of 8 s. For a spinning time of 12 s, the greater Fud zone was formed in the central zone. With respect to the tensile fracture morphology under a friction pressure of 1.77 MPa [shown in Fig. 10(d)], the greater tensile fracture surface was in the Fpl zone and the smaller Fud zone, and the Fpl zone [shown in Fig. 10(c)] was the area in which the Fud zone increased with increasing spinning times.

The tensile fracture morphology of welded nylon-6 with nylon-6 under different spin-friction pressures and spinning times has three main regions: (1) the central section (Fud), which has no plasticized “wearing surface” fracture morphology; (2) the peripheral section (Fpl), which has a plasticized “ripple” fracture

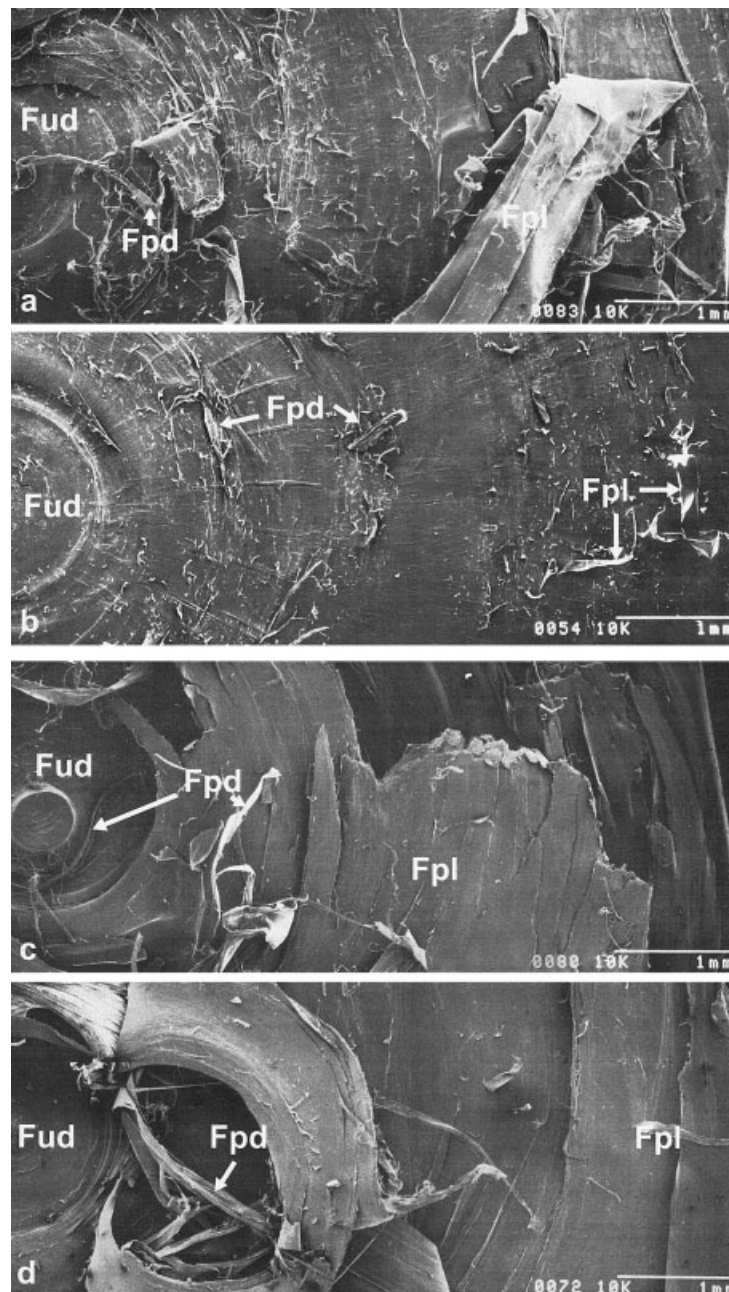


Figure 10 SEM micrographs of tensile fracture morphologies of welded HDPE with HDPE under 0.98 MPa friction pressure with: (a) 10 s of spinning time; (b) 12 s of spinning time; (c) under 1.37 MPa friction pressure with 8 s of spinning time; (d) under 1.77 MPa friction pressure with 12 s of spinning time.

morphology; and (3) the middle section (Fpd), which has a partly plasticized “dot” fracture morphology. With respect to the tensile fracture morphology under a friction pressure of 0.49 MPa with 12, 16, and 20 s spinning times, it was found that the larger Fud zone was formed in the central zone, as shown in Figure 11(a); for the tensile fracture morphology under a friction pressure of 0.78 MPa with 12, 16, and 20 s spinning times, a smaller Fud zone was formed in the

central zone, as shown in Figure 11(b). For the tensile fracture morphology under a friction pressure of 0.98 MPa with 12, 16, and 20 s spinning times, no Fud zone (an unwetting zone) was formed in the central zone, as shown in Figure 11(c). A comparison of Figure 11(a) with 11(c) shows that the area of the Fpd zone increases with increasing friction pressure, but the area of the Fpl zone has less effect on the friction pressure.

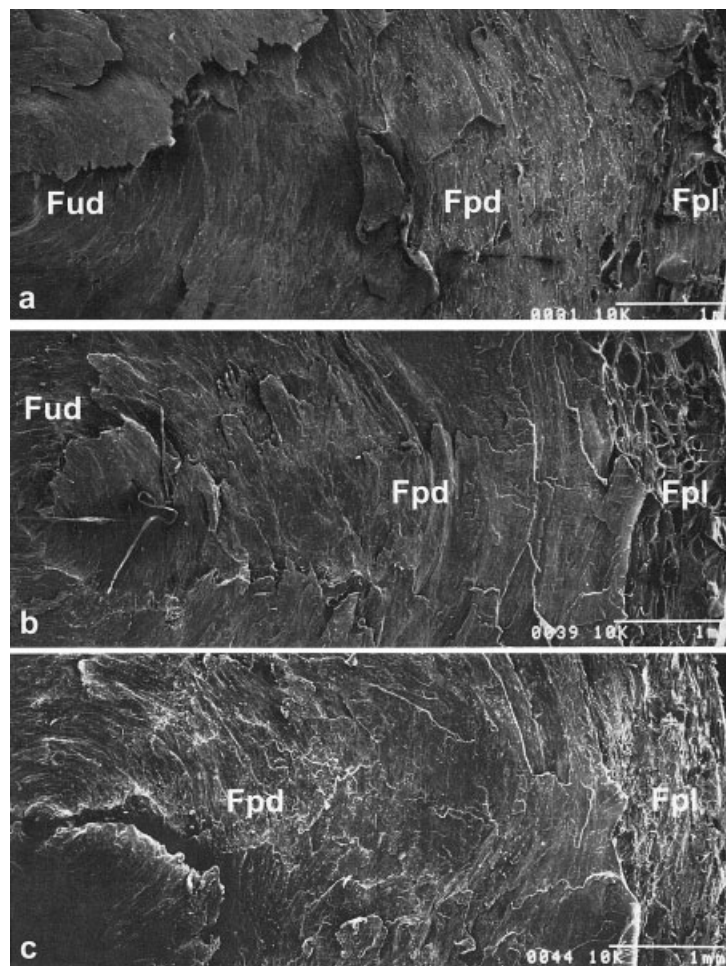


Figure 11 SEM micrographs of tensile fracture morphologies of welded nylon-6 with nylon-6 under (a) 0.49 MPa, (b) 0.784 MPa, and (c) 0.98 MPa friction pressure with 20 s of spinning time.

Bonding strength

Bonding strengths of welded PP with PP under different spin-friction pressures and spinning times are shown in Table I. A friction pressure of 1.17 MPa with

TABLE I
Bonding Strength of Welded PP-PP Under Different Spin Friction Pressures and Times^a

Friction pressure (MPa)	Friction time (s)	Bonding strength (MPa)
0.98	16	5.54
	18	6.03
	20	6.50
1.17	16	8.05
	18	6.23
	20	4.04
1.37	16	3.88
	18	2.76
	20	1.23

^a Friction pressure = forge pressure; forge time = 5 s.

16 s of spinning time can obtain the best bonding strength (indicated in bold) because it produced a larger plasticized region, which has no unplasticized region (the Fud zone) in the welding zone, as shown in Figure 9(b). At a friction pressure of 1.37 MPa with 20 s of spinning time, because cracks tend to propagate in the unwelding zone (the Fud zone) as shown in Figure 9(c), a lower bonding strength was achieved because the welded part was in an internal circular crack (the Fud zone) in the central zone. When the externally applied tensile stress exceeded the fracture stress of the welded part, the internal circular crack grew as long as the tensile stress was applied, until such time as the welded part's cross section could no longer support the stress put on the welded part; in other words, the greater the value of the Fud zone (the internal crack), the greater the increase in the stress at the crack tip. When we consider the problem of bonding strength or the mechanical energy development as two pieces of polymer are brought into contact, wet-

ting and interdiffusion of molecule chains segments, back and forth across the welded interface, are required to separate the welded polymer as a function of friction pressure, friction time, friction temperature, forge time, forge pressure, and molecular weight. Friction time is very important among these parameters because plasticization occurs first from the periphery to the central zone, and the plasticized area is increasingly broadened with the friction time. When the central zone produced an apparent plasticization, the plasticization region gradually spun along the radius to the peripheral zone, increasing with spinning time, and an unplasticization zone left the central zone. At this time, if the forge pressure is immediately applied to the welding, the welded part was in an internal circular crack in the central zone, which decreased the bonding strength. For these reasons, to achieve the best friction-weld bonding, the optimum friction pressure must be applied collaboratively with the optimum friction timing.

The bonding strength of welded HDPE with HDPE under different spin-friction pressures and spinning times is shown in Table II. A friction pressure of 1.77 MPa at a spinning time of 8 s obtains the best bonding strength (indicated in bold) because it produces a larger plasticized zone. Under a friction pressure of 0.98 MPa with a spinning time of 12 s [as shown in Fig. 10(b)], the plasticized region in the Fud was totally spun into the Fpd zone, and the bonding strength are smaller because cracks tend to propagate in the unwelded zone in Fud. For each friction pressure, the bonding strength with spinning times of 8 and 10 s was always greater than that with a spinning time of 12 s. Additionally, the bonding strength increases with increasing friction pressure. Bonding strengths of nylon-6 with nylon-6 under different spin friction pressures and different spinning times are shown in Table III. Pressures of 0.98 MPa (12, 16, and 20 s) obtained the best bonding strength because it produces a larger plasticized region, which has no unplasticized regions

TABLE II
Bonding Strength of Welded HDPE–HDPE Under Different Spin Friction Pressures and Times^a

Friction pressure (MPa)	Friction time (s)	Bonding strength (MPa)
0.98	8	5.98
	10	5.23
	12	2.55
1.37	8	7.55
	10	6.67
	12	3.43
1.77	8	8.63
	10	7.73
	12	4.51

^a Friction pressure = forge pressure; forge time = 5 s.

TABLE III
Bonding Strength of Welded Nylon-6–Nylon-6 Under Different Spin Friction Pressures and Times^a

Friction pressure (MPa)	Friction time (s)	Bonding strength (MPa)
0.49	12	10.83
	16	10.69
	20	10.33
0.78	12	11.82
	16	11.59
	20	11.02
0.98	12	14.97
	16	14.92
	20	14.34

^a Forge pressure = 15.7 MPa; forge time = 5 s.

(the Fud zone) in the central part, as shown in Figure 11(c). A friction pressure of 0.49 MPa (12, 16, and 20 s) obtains a lower bonding strength because cracks tend to propagate in the unwelded zone (the Fud zone), as shown in Figure 11(a). A comparison of Figure 11(a) with 11(c) shows that the bonding strength increases with increasing friction pressure, attributed to the plasticized area under 0.98 MPa friction pressure, which is greater than 0.78 and 0.49 MPa.

CONCLUSIONS

The effects of spin-friction pressure and timing on the properties of PP with PP, HDPE with HDPE, and nylon-6 with nylon-6 spin-friction welding were investigated. The bonding tensile strength was measured and related fracture morphologies were examined. Some results are summarized below:

1. The heat-affected zone has three main regions: the plasticized region (Zpl), the undeformed region (Zud), and the partly plasticized region (Zpd).
2. The tensile fracture morphology of welded PP with PP, HDPE with HDPE, and nylon-6 with nylon-6 has three main sections: the central zone (Fud), which has a “wearing surface” fracture morphology; the peripheral zone (Fpl), which has “dimple,” “patch,” and “ripple” fracture morphology, respectively; and the middle zone (Fpd), which has partly plasticized “dot,” “mixed fibril with ribbon,” and “dot” fracture morphology, respectively.
3. The greater plasticized Fpd and Fpl zones, with no Fud zone in the central zone—1.17 MPa (16 s) in PP–PP welding, 1.77 MPa (8 s) in HDPE–HDPE welding, and 0.98 MPa (8, 10, and 12 s) in nylon-6–nylon-6 welding—can achieve the best bonding strengths.

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References

1. Borroff, E. M.; Wake, W. C. *Trans Inst Rubber Ind* 1949, 25, 199.
2. Landrock, H. A. *Adhesives Technology Handbook*; Noyes Publications: Park Ridge, NJ, 1985.
3. Yue, C. Y.; Cherry, B. W. *J Adhes* 1987, 24, 127.
4. Kinlock, A. J. *Adhesion and Adhesives: Science and Technology*; Chapman & Hall: New York, 1987.
5. Rapai, J. *Annu Tech Conf Soc Plast Eng An Quim* 1997, 3, 3674.
6. Sims, J.; Ellwood, P. A.; Taylor, H. J. *Annu Tech Conf Soc Plast Eng An Quim* 1994, 1269.
7. Grim, R. A. *Regional Tech Conf Soc Plast Eng An Quim* 1996, 11.
8. Howie, I.; Gillespie, J. W., Jr.; Smiley, A. J. *J Thermoplast Compos Mater* 1993, 6, 205.
9. Tappe, P.; Potente, H. *Polym Eng Sci* 1989, 29, 1655.
10. Potente, H.; Uebbing, M. *Polym Eng Sci* 1997, 37, 726.
11. Shields, J. *Adhesives Handbook*, 3rd ed.; Butterworth: London, 1984.
12. Grimm, R. A. *Adv Mater Proc* 1995, 3, 27.
13. Wool, R. P.; O'Connor, K. M. *J Appl Phys* 1981, 52, 5953.
14. Kim, Y. H.; Wool, R. P. *Macromolecules* 1983, 16, 1115.
15. de Gennes, P. G. *Hebd Seances Acad Sci Ser B* 1980, 291, 219.
16. de Gennes, P. G. *J Chem Phys* 1971, 55, 572.
17. Kausch, H. H.; Jud, K. *Plast Rubber Compos Process Appl* 1982, 2, 265.
18. Wool, R. P.; O'Connor, K. M. *Polym Eng Sci* 1981, 21, 970.
19. Kim, H. J.; Lee, K.-J.; Lee, H. H. *Polymer* 1996, 37, 4593.