Effects of raw materials on the properties of wood fiber-polyethylene composites—Part 3: Effect of a compatibilizer and wood adhesive on the interfacial adhesion of wood/plastic composites

Chin-yin Hwang
Chung-yun Hse
Todd F. Shupe*

Abstract

The objective of this study was to examine the effect of maleated polypropylene compatabilizer on the interfacial properties of wood and polyolefins. Birch wood dowels containing an adhesive applied on the surface were embedded in molten plastic matrices using specially designed jigs. The three plastics investigated included low density polyethylene (LFPE), linear low density polyethylene (LLDPE), and polypropylene (PP). The four adhesives used were urea formaldehyde (UF), phenol formaldehyde (PF), isocyanate (ISO), and a control group that did not contain any adhesive. A low molecular weight emulsion type MAPP, Epolene (E-43) was used at concentrations of 0 (control), 10 percent, 20 percent, 30 percent, and 40 percent. Modified single-fiber pull-out tests were performed to investigate the interrelationships between a four-component system of wood-adhesive-compatibilizer-plastic, simulating the interphase of a wood-thermoplastic composite. The interfacial shear strength as measured by the modified single-fiber pull-out test indicated that the overall average interfacial shear strength was highest in PP and lowest in LDPE. ISO-bonded LDPE without E-43 treatment showed the highest interfacial shear strength among all other LDPE composites. The E-43-treated wood surfaces without adhesive displayed overall superior performance than all adhesive-bonded LLDPE. Addition of E-43 greatly enhanced the interfacial shear strength for all plastic types, except for ISO-bonded LDPE and LLDPE. The effect of E-43 was most pronounced in PP and less in LDPE and LLDPE. Adhesive failure prevailed in LDPE and LLDPE composites while PP showed predominately cohesive failures in the wood dowel or in the plastic matrix.

Wood flour and fibers are excellent fillers for thermoplastics because of their low density, low cost, high strength and stiffness, desirable fiber aspect ratio, flexibility during processing, and biodegradability (Felix and Gatenholm 1991, Collier et al. 1995). However, satisfactory dispersion of wood fillers in the matrices of thermoplastics has long been a problem due to the hydrophilic nature of bio-based fibers and the hydrophobic nature of plastic (Felix et al. 1994, Luo et al. 2002, Tze et al. 2004). The chemical incompatibility prohibits the formation of a durable interface in the plastic composites, causing failure in stress transfer from one phase to another. Therefore, to enhance the affinity between these two components, the surface properties of one must be modified.

The compatibility of wood and plastic can be improved by introducing a compatibilizer into the system. Bifunctionality of compatibilizers is advocated to increase the adhesion between the interface of wood and plastic, and thus enhance stress transfer. Maleated polypropylene (MAPP) is the most commonly used compatibilizer for polypropylene and polyethylene polymers. The carboxylic groups in MAPP provides

---

The authors are, respectively, Senior Scientist and Head, Forest Chemistry Division, Taiwan Forestry Research Inst., Taipei, Taiwan (chinyin@mail.tfri.gov.tw); Principal Wood Scientist, Southern Research Sta., USDA Forest Serv., Pineville, Louisiana (chse@fs.fed.us); and Professor, Louisiana Forest Products Development Center, School of Renewable Natural Resources, Louisiana State Univ. Agri. Center, Baton Rouge, Louisiana (tshupe@agcenter.lsu.edu).
This paper (No. 06-40-0206) is published with the approval of the Director of the Louisiana Agri. Expt. Sta. This paper was received for publication in September 2007. Article No. 10403.
*Forest Products Soc. Member.

Forest Prod. J. 58(5):66-72.

Table 1. — Basic physical properties for the experimental plastics.

Plastic type	Melt index[a]	Density[b]	Characteristic point[c]
	(g/10 min)	(g/cm^3)	(°C)
PD729 N E7 (PP)	4.0	0.900	95
LL-3001 (LLDPE)	1.0	0.917	125
LD-200 (LDPE)	7.5	0.917	104

[a] Test methods are based on ASTM D 1238 (ASTM 2004a).
[b] Test methods are based on ASTM D 792 (ASTM 2004b).
[c] Temperatures listed are deflection temperature at 66 psi using ASTM D 638 (ASTM 2004c).

covalent and hydrogen bonding to the hydroxyl groups of cellulosic fiber surface. The long chains of MAPP, on the other hand, can bind with the polypropylene matrix which allows segmental crystallization. MAPP can be used as a low molecular-weight additive (compatibilizer) in a polymer blend system and a high molecular weight substitute for polypropylene. Numerous studies have shown mostly positive effects by adding MAPP to a multiphase system (Luo et al. 2002, Spear et al. 2004).

Several methods have been developed to directly characterize the interface of reinforced composites, namely, the single-fiber fragmentation test (Tai et al. 1992, Shaler 1993, Felix and Gatenholm 1994), the single-fiber pull-out test (Westerlind et al. 1984), and the microbond test (Miller et al. 1987, Sanadi et al. 1992, Tao et al. 1993, Liu 1994). These three techniques have been widely used to approximate the interfacial shear strength of fiber-reinforced thermoplastic composites using a model composite. In this study, a modified single-fiber pull-out test by using wood dowels, instead of wood fibers, was applied to assess the “bulk” interfacial shear strength. The wood dowels were selected because of their relatively uniform surfaces and their relative ease to prepare and test. The objective of this study was to examine the effect of maleated polypropylene compatabilizer on the interfacial properties of wood and polyolefins. This objective was achieved by examining interfacial shear strength and failure mode. This study is part of a series of experiments to determine the effects of recycled materials on the properties of wood fiber-polyethylene composite panels. Earlier studies have investigated the effect of (1) recycled fiber on fiberboard panel properties and (Hwang et al. 2005) and (2) compatibilizer on the wettability of birch plywood and polyoefins (Hwang et al. 2007). Earlier studies by Wu et al. (1994) and Hwang (1998) found surprisingly favorable results when binding wood plastic composites with thermoset resins. Therefore, this paper was also a follow-up to these and other earlier studies.

Materials and methods

Nominal 3/8-in. birch wood dowels were obtained from a local mill. The three types of polyolefin polymers used in this study were supplied by the Exxon-Mobil Chemical Co. They were polypropylene (PP), linear low-density polyethylene (LLDPE), and low-density polyethylene (LDPE) (**Table 1**). Three thermo-set wood adhesives used in the study were urea formaldehyde (UF), phenol formaldehyde (PF), and isocyanate (ISO). Both urea-formaldehyde (Casco resin TD-33C) and phenol-formaldehyde (Cascophen 1770 to 3) resins were supplied by Hexion Speciality Chemicals, Inc. The typical viscosity at 25 °C is 175 cps for UF and 130 cps for PF, and the solids content are 65 percent and 53 percent, respectively. The isocyanate used in this study was ISOBOND, a product of Dow Plastics, which has 30.9 percent NCO content and 185 cps viscosity at 25 °C. The compatabilizer was a low molecular weight emulsion type MAPP, Epolene E-43 received as a 40 percent solution, from the Eastman Chemical Co. Birch dowels were cut into 2-3/8-in-long samples, and then oven dried at 103° ± 3 °C until a constant weight was obtained. These ovendried dowels were then transferred to desiccators over anhydrous $CaSO_4$ to cool to room temperature. About 30 g of plastic pellets were melted in TEFLON® containers at various temperature for 2 hours. The temperature settings for LDPE, LLDPE, and PP were 140, 160, and 200 °C, respectively.

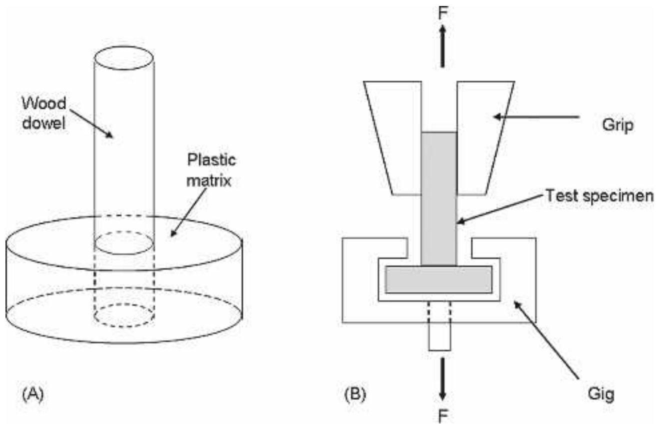

Figure 1. — Schematic diagram of the interfacial shear strength test procedure (A) test specimen and (B) specimen and fixture layout.

Wood dowels were embedded in the molten plastic matrices using specially designed jigs and remained in the oven for 30 minutes Two hours before embedding, the adhesive was applied onto the surface of the dowels at the interfacial area and then set to dry in a desiccator. The resin solid contents coated on the interfacial area were approximately 10 lbs per 1000 ft^2 for UF and PF and 4 lbs per ft^2 for ISO. In a similar manner, the compatibilizer was applied 1 hour before the embedding procedure.

To minimize shrinkage of the plastics, especially PP, test specimens were cooled gradually by turning off the oven and allowing them to remain in the oven until room temperature was reached. Interfacial shear strength tests were conducted with a Instron Universal Testing Machine at a load cell speed of 0.2 in/min., using the tension fixture on the dowel end and the internal bond fixture on the plastic matrix end. The schematic diagram for the testing procedure is shown in **Figure 1**. Bulk interfacial shear strength (T) was determined as follows:

$$T = \frac{F}{\Pi \times d \times l} \qquad [1]$$

where F is the debonding force, d is the fiber diameter, and l is he fiber embedded length.

This experiment had a 3 × 4 × 5 factorial treatment arrangement (**Table 2**). Data were analyzed by analysis of variance and simple linear regression. Mean separation tests were performed using the Tukey test at alpha = 0.05 (SAS 1999).

Results and discussion

Interfacial shear strength

The mean interfacial shear strengths (ISS) (Eq. [**1**]) for all treatment combinations showed that interfacial shear strength between birch dowels and polyolefin polymers increased drastically with the addition of E-43, except for ISO-bonded LDPE and LLDPE which tended to decrease (**Table 3**). In the control groups (0% E-43 concentration) within each of the three plastic types, ISO had the highest interfacial shear strength value among all other adhesives in both LDPE (410 vs. 112 to 186 psi) and LLDPE (400 vs. 140 to 253 psi). Moreover, in LDPE, ISO-0 (ISO-bonded with 0 percent E-43) had the highest interfacial shear strength (410 psi) among all other treatment combinations (112 to 342 psi). The addition of E-43 inhibited the interfacial shear strengths of ISO-bonded LDPE and LLDPE composites and enhanced the interfacial shear strengths for composites bonded with the other three adhesives. However, the most favorable treatment combinations for LLDPE were E-43 treated composites without adhesive, i.e., NO-20 (562 psi) and NO-30 (531 psi). In PP, the composites with no adhesive (NO) showed the highest mean value of interfacial shear strength among all control groups (416 vs. 256 to 316 psi). With the addition of E-43 to PP, all the four adhesives had a 2- to 3-fold increment in interfacial shear strengths.

Table 2. — Experimental design for testing bulk interfacial shear strength of birch dowels and polyolefin polymers.

Variable	Level	Description
Plastic type (PT)	3	Low density polyethylene (LFPE)
		Linear low density plyethylene (LLDPE)
		Polypropylene (PP)
Adhesive type (AT)	4	Control (NO)
		Urea-formaldehyde (UF)
		Phenol-formaldehyde (PF)
		Isocyanate (ISO)
E-43 concentration (CON)[a]	5	0 (control), 10 %, 20 %, 30 %, 40%
Replication	6	

[a]Based on solids content.

Table 3. — Interfacial shear strength between plastic matrices and birch wood dowels treated with various E-43 concentration (CON) and adhesives.

Plastic type[a]/CON	Adhesive type[b]				
	NO	UF	ISO	PF	MEAN
(percent)	------------------------------------	------------------------------------ (psi)	------------------------------------	------------------------------------	
LDPE					
0	176.9 (14.6)[c]	186.5 (35.3)	410.3 (33.3)	111.9 (12.4)	
10	279.4 (34.4)	235.0 (49.4)	341.9 (52.6)	287.5 (60.1)	
20	259.2 (30.0)	228.0 (36.8)	316.7 (53.2)	240.2 (82.3)	
30	219.6 (38.3)	211.4 (33.0)	335.9 (57.0)	170.1 (33.1)	
40	178.3 (29.5)	151.9 (29.4)	262.8 (15.7)	153.9 (51.9)	
Mean	222.7	202.6	333.5	192.7	237.9(C)[d]
LLDPE					
0	253.1 (15.8)	163.2 (16.9)	400.7 (69.1)	140.5 (13.5)	
10	492.9 (29.8)	430.8 (58.4)	313.4 (39.2)	513.0 (47.8)	
20	561.7 (83.3)	430.9 (82.2)	286.6 (40.7)	408.2 (62.3)	
30	531.0 (40.7)	424.0 (96.4)	321.7 (25.3)	272.9 (34.1)	
40	377.8 (78.6)	339.0 (69.8)	259.4 (27.0)	278.1 (38.8)	
Mean	443.3	357.6	316.4	322.5	359.9(B)
PP					
0	416.1 (62.4)	316.2 (40.7)	311.1 (41.2)	255.5 (31.3)	
10	906.8 (106.8)	100l.3 (126.1)	904.8 (123.7)	767.9 (149.3)	
20	967.5 (138.6)	895.9 (92.4)	861.3 (l38.4)	744.1 (83.5)	
30	918.1 (74.9)	895.9 (160.7)	663.9 (94.3)	566.3 (97.6)	
40	818.0 (101.1)	769.7 (110.1)	649.8 (90.0)	524.5 (72.6)	
Mean	805.3	775.8	678.2	571.7	707.7(A)
Grand mean	490.4(A)	445.5(B)	442.7(B)	362.3(C)	

[a]LDPE = low-density polyethylene, LLDPE = linear low-density polyethylene, and PP = polypropylene.
[b]NO = no adhesive (control), UF = urea-formaldehyde, ISO = isocyanate, and PF = phenol-formaldehyde.
[c]Numbers in parentheses are SDs. Sample size = 6.
[d]Means with the sample letter are not significantly different according to the Tukey mean separation test at alpha = 0.05.

The ANOVA in **Table 4** reveals that all sources of variation were highly significant. The effect of plastic type on interfacial shear strength (**Table 3**) indicates an order of PP>LLDPE>LDPE, which was the same order as observed for the contact angle measurement, i.e., the opposite of wettability (Hwang et al. 2007). The average interfacial shear strength of PP (708 psi) is 2 to 3 times stronger than those of LDPE (238 psi) and LLDPE (360 psi). Though MAPP may react identically with wood at the interface of wood-polyolefin composites, its reaction to plastic matrices may be different. The attraction between MAPP and polyolefin polymer is mostly van der Waals physical interaction. Since MAPP is chemically similar to PP, the attraction between MAPP and PP may be stronger than its counterparts, therefore exhibiting higher interfacial shear strength. Moreover, the differences among the three plastic types also coincided with their degrees of crystallinity. Other studies have also shown that MAPP is more compatible with PP-based composites than PE-based composites (Bledzki et al. 1996, Snijder and Bos 2000).

The interfacial shear strength values using LLDPE in this study are of the same order of magnitude as from a modified single fiber pull-out test by Sanadi et al. (1993). The report by Sanadi et al. (1993) reported the interfacial shear strength between low-molecular-weight polyethylene and 2-mm birch dowel to be 290 psi for untreated and 450 psi for MAPP-coated model composites. However, Felix and Gatenholm (1994) showed an appreciably higher value of 885 psi, as compared to 416 psi in the control group in this study. This difference may be due to the presence of air bubbles in the interphase of the wood and the matrix interfering with stress transfer between the two bulk phases. Although the birch dowels were carefully ovendried, the application of adhesive and E-43 on the

Table 4. — ANOVA showing the effects of plastic type (PT), replication (REP), adhesive type (AT), and E-43 concentration (CON) on interfacial shear strength of birch dowels and plastic matrices.

Source	DF	MS	F	P value
PT	2	7137437.69	537.78	0.0001[a]
REP(PT) [b]	15	13272.04	2.82	0.0004**
AT	3	255766.70	54.44	0.0001**
CON	4	895722.40	190.66	0.0001**
AT*CON	12	44440.78	9.46	0.0001**
PT*AT	6	154844.51	32.96	0.0001**
PT*CON	8	283293.55	60.30	0.0001**
PT*AT*CON	24	18815.65	4.00	0.0001**
ERROR	285	4698.11	--	--

[a] Denotes significance at alpha = 0.01.
[b] REP(PT) indicates replicates (REP) within plastic type (PT).

Table 5. — Solid E-43 content (SCC) and solid content (SRC) for wood dowels and plastic matrices with plastic type combined.

	Adhesive type[a]			
CON	NO	UF	ISO	PF
(percent)	(lb per 1000 sq. ft)			
SCC				
0	--	--	--	--
10	2.16 (0.55)[b]	0.58 (0.23)	0.84 (0.21)	1.13 (0.31)
20	4.93 (1.64)	1.37 (0.84)	1.96 (0.44)	2.19 (0.63)
30	5.72 (1.33)	1.85 (0.55)	3.03 (0.67)	3.37 (1.31)
40	8.73 (1.47)	3.64 (1.24)	4.90 (0.71)	5.21 (0.98)
SRS				
0	--	7.84 (1.84)	3.95 (0.68)	5.66 (1.24)
10	--	7.51 (1.69)	3.77 (0.46)	5.42 (1.22)
20	--	8.22 (1.49)	4.15 (0.95)	5.39 (0.83)
30	--	8.18 (1.19)	3.80 (0.89)	5.43 (0.59)
40	--	8.11 (1.48)	3.92 (0.99)	5.64 (1.20)

[a] ISO, PF, UF and NO denote isocyanate, phenol formaldehyde, urea formaldehyde, and no adhesive (control), respectively.
[b] Numbers in parentheses are SDs.

contact area before embedding into the matrix polymer could cause an increase in MC on the interface. Even though the coated specimens were stored in desiccators, they might not have been sufficiently dry to eliminate excess moisture. Moreover, condensation water upon curing of UF and PF resins might also be responsible for the presence of air bubbles. It is also possible that any adhesive/chemical coated on the wood dowel will diffuse in the molten plastic during embedding thus diluting the effect of this treatment.

The effect of the adhesive type on the interfacial sheer strength indicates that if other factors were combined, wood-polyolefin composites without adhesive as a binder (NO) showed superior interfacial shear strength, whereas PF performed poorly in the interphase (**Table 3**). This finding does not agree with the results of contact angle measurements in wood or plastic (Hwang et al. 2007). Conversely, the effect of E-43 concentration on interfacial shear strength (**Table 3**) somewhat coincided to the wettability of plastics (Hwang et al. 2007), since the interfacial shear strength decreased with increasing contact angle (i.e., decreasing wettability).

The three-way interaction (PT*AT*CON) was analyzed individually by plastic type. For LDPE (**Table 3**), the performance of ISO was superior to the other three adhesives at all E-43 concentration levels. Unlike the other three adhesives which trended toward and then downward with increasing E-43 concentration, ISO showed a downward trend with increasing E-43 concentration despite some minor fluctuations. UF-bonded composites exhibited comparable mean interfacial shear strength with that of no-adhesive-bonded composites at 0 percent E-43 concentration, and had slightly lower values at all other concentration levels. The increment in interfacial shear strength was most pronounced in PF from 0 percent to 10 percent E-43 concentration. However, the reduction in interfacial shear strength from 10 percent to 40 percent E-43 concentration level was most dramatic in PF. Interfacial shear strength of PF was comparable with no adhesive (NO) at 10 percent E-43 concentration and comparable with UF at 40 percent E-43 concentration.

Similar to LDPE, interfacial shear strength of LLDPE bonded with ISO trended downward with increasing E-43 concentration (**Table 3**). The addition of E-43 showed a negative effect in the interfacial shear strength of ISO-bonded LDPE and LLDPE. For the other adhesives in LLDPE, the trends were also similar to those of the three adhesives in LDPE. The improvement upon addition of E-43 was most apparent in PF-bonded LLDPE, in which interfacial shear strength increased by approximately 3.5 times (140 psi for PF with 0 percent E-43 vs. 513 psi for PF with 10 percent E-43). However, it was most sensitive to increases in E-43 concentration. PF had equivalently high interfacial shear strength with no-adhesive bonded LLDPE at the 10 percent concentration level, and it had equally low interfacial shear strength with ISO at the 40 percent E-43 concentration level. The most favorable E-43 concentration for NO, UF, and PF was in the range between 0 percent to 10 percent.

In PP, the responses of interfacial shear strength to different adhesive types and concentration levels are shown in **Table 3**. All four types of adhesives showed similar trends in which interfacial shear strength trended upward and then downward with increasing E-43 concentration. UF showed superior interfacial shear strength to PF at all concentration levels, and superior to ISO upon the addition of E-43. The interfacial shear strengths for UF, PF and ISO showed a negative response when E-43 concentration level exceeded 10 percent. Conversely, the interfacial shear strength of the composites without adhesive (NO) increased slightly with an increase in concentration from 0 to 20 percent then fell beyond 20 percent concentration.

The most appropriate treatment combinations for interfacial shear strength were ISO-0 (ISO-bonded with 0 percent E-43 concentration) for LDPE, NO-20 (no-adhesive-bonded with 20 percent E-43 concentration) for LLDPE, and UF-10 (UF-bonded with 10 percent E-43 concentration) for PP. Except for ISO-bonded LDPE and LLDPE, the addition of E-43 showed a positive effect on interfacial shear strength but lower concentrations yielded more favorable results. The highest efficiency in PP was with E-43 (**Table 3**).

Solid E-43 content (SCC) and solid resin content (SRC) were also calculated for each specimen. Results of solid E-43 content and solid resin content with plastic type combined are listed in **Table 5**. Due to the manual application of

Table 6. — Effect of different plastic and adhesive types as denoted by the model $Y = a + b_1X_1 + b_2X_2$.

Plastic[a]/ adhesive type[b]	Statistics			Partial coefficient		
	F	p	R^2	b_0	b_1	b_2
LDPE						
ISO	19.734	0.0001	0.4134	383.478	−22.221	--
NO	8.466	0.0014	0.3854	199.435	23.798	−2.739
UF	4.882	0.0155	0.2656	197.675	25.631	−7.601
LLDPE						
ISO	11.533	0.0002	0.4607	382.965	−55.477	6.669
NO	15.659	0.0001	0.5370	290.320	87.697	−7.679
PF	3.646	0.0397	0.2126	246.591	90.820	--
UF	11.548	0.0002	0.4610	229.218	191.694	−39.239
PP						
ISO	4.361	0.0228	0.2442	515.730	202.645	−34.933
NO	38.567	0.0001	0.7407	480.765	189.800	−17.229
PF	5.711	0.0085	0.2973	400.041	184.755	−29.105
UF	7.200	0.0031	0.3478	582.903	353.379	−70.582

[a] LDPE = low density polyethylene, LLDPE = linear low-density polyethylene, and PP = polypropylene.

[b] ISO, PF, UF and NO denote isocyanate, phenol-formaldehyde, urea-formaldehyde, and no adhesive (control), respectively.

E-43, solid E-43 content showed some variations with different adhesives. Specimens with no adhesive (NO), received and absorbed more E-43 than those which had adhesive. The application of E-43 after an adhesive was brushed on the interface area around the dowel may be the reason for higher variations in solid E-43 content. The overall solid E-43 contents for all concentration levels were 1.18, 2.61, 3.49, and 5.62 lb/1,000 ft^2 from the 10 percent through 40 percent levels, respectively. In solid resin content, there were minimal variations among all E-43 concentration levels within each adhesive type but greater variations among adhesive types. UF had the highest average solid resin content, whereas ISO had the lowest. With all other factors pooled, solid resin contents were 3.92, 5.51, 7.97 lb/1,000 ft^2 for ISO, PF, and UF, respectively. All plastic types were assumed to obtain the same loading of solid E-43 content and solid resin content.

From regression analyses, interfacial shear strength was found to be independent of solid resin content but dependent on solid E-43 content. Therefore, solid resin content was excluded from the model fitting. Regression analyses for solid E-43 content for all treatment combinations are shown in **Table 6**. Polynomial regression models best fit most of the treatment combinations and only interfacial shear strength of ISO-bonded LDPE showed linear dependence on solid E-43 content. The partial coefficients also indicate that, except for the ISO-bonded LDPE and LLDPE, interfacial shear strength increases with an initial addition of E-43 and decreases as solid E-43 content further increases. The coefficient of determination (R^2) of all models showed that solid E-43 content accounted for approximately 20 percent to 74 percent of the variability of interfacial shear strengths in wood dowel-polyolefin composites. This finding indicates that some other factors may also contribute to the variation in interfacial shear strength. The interaction between adhesive and E-43 may be responsible for this difference.

Table 7. — Frequencies of failure mode on the interface of wood-plastic model composites.

Adhesive type[a]	NO				UF				ISO				PF			
Failure mode[b]	0	1	2	3	0	1	2	3	0	1	2	3	0	1	2	3
	Plastic type/CON[c]															
LDPE (percent)																
0	6	0	0	0	6	0	0	0	1	1	4	0	6	0	0	0
10	5	0	1	0	6	0	0	0	2	1	3	0	6	0	0	0
20	5	0	1	0	6	0	0	0	2	1	3	0	6	0	0	0
30	6	0	0	0	6	0	0	0	2	1	3	0	6	0	0	0
40	6	0	0	0	6	0	0	0	2	1	3	0	6	0	0	0
LLDPE																
0	6	0	0	0	6	0	0	0	5	0	1	0	6	0	0	0
10	6	0	0	0	6	0	0	0	5	0	1	0	6	0	0	0
20	6	0	0	0	6	0	0	0	6	0	0	0	6	0	0	0
30	6	0	0	0	6	0	0	0	6	0	0	0	6	0	0	0
40	6	0	0	0	6	0	0	0	6	0	0	0	6	0	0	0
PP																
0	6	0	0	0	4	0	2	0	3	0	3	0	3	0	3	0
10	0	1	1	4	0	0	2	4	0	0	1	5	0	0	5	1
20	0	0	0	6	0	0	2	4	0	0	2	4	0	0	6	0
30	0	0	0	6	0	0	3	3	2	0	1	3	0	0	6	0
40	1	0	1	4	1	1	3	1	1	0	5	0	0	0	6	0

[a] ISO, PF, UF, and NO denote isocyanate, phenol-formaldehyde, urea-formaldehyde, and no adhesive (control), respectively.

[b] 0 = clean surface, 1 = wood failure, 2 = plastic failure, 3 = undermined due to broken dowel.

[c] CON: E-43 concentration.

Failure mode

Failure mode on the interface was recorded and presence of air bubbles was examined following the fiber pull-out test. The frequencies of failure mode are presented in **Table 7**. E-43 showed extremely low affinities for LDPE and LLDPE, as their failure modes were exclusively "clean interface" due to weak adhesive force. Plastic and wood failure occurred in more than two thirds of ISO-bonded LDPE model composites, indicating better interface adhesion with stronger adhesion forces than cohesion forces. These results are consistent with the average interfacial shear strength magnitudes in **Table 3**. The addition of E-43 did not show any distinguishable differences among the four adhesive types in LDPE and LLDPE, but the addition of E-43 caused a much higher percentage of "undetermined" failure modes in NO-, UF-, and ISO-bonded PP model composites. Undetermined failure mode is caused by dowel failure at the grip end while the interface remains intact. Dowel failure was due to loss of strength from thermal degradation when wood dowels were subjected to 200 °C for 30 minutes during the manufacture of the PP composites. When the adhesive bonding between the wood dowel and PP is not very strong, as was the case in 0 percent E-43 concentration, the wood dowel can be pulled out before it breaks. On the other hand, if the adhesion force on the interface exceeds the dowel strength, the dowel breaks. Thus the intrinsic interfacial shear strength values should be higher in such PP model composites.

The relationship between the percentage of adhesive failure and plastic type (**Fig. 2**) shows that over 80 percent of LDPE, 98 percent of LLDPE, and 17 percent of PP composites had

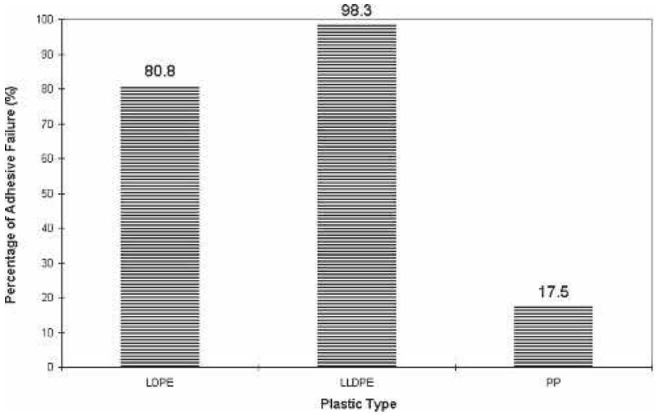

Figure 2. — Percentage of adhesive failure in three wood-polyolefin composites.

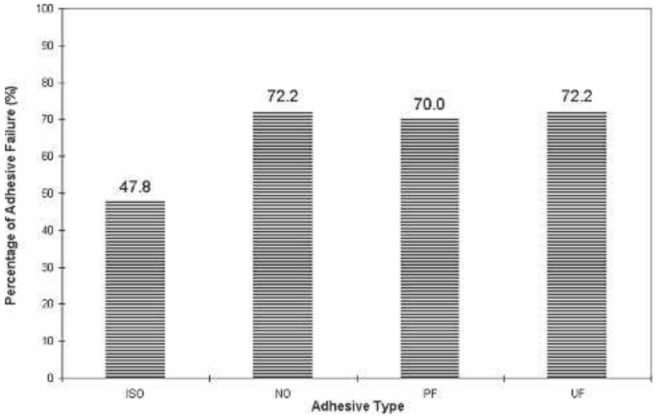

Figure 3. — Percentage of adhesive failure in wood-polyolefin model composites by adhesive type.

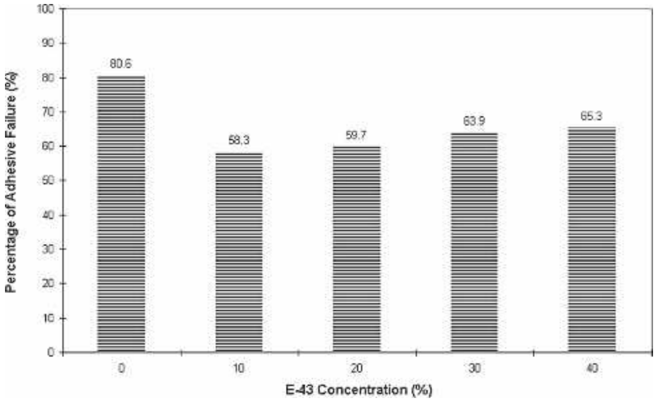

Figure 4. — Percentage of adhesive failure in wood-polyolefin composites by E-43 concentration.

adhesive failure. The relative ranking of adhesive failure in LDPE and LLDPE was consistent with that of interfacial shear strength. The relationship of interfacial shear strength with percentage adhesive failure however, was not consistent with respect to PP due to the high frequency of undetermined mode failures. The effect of adhesive type (**Fig. 3**) reveals that ISO exhibited the lowest percentage of adhesive failure, and the frequencies of adhesive failure for NO (no adhesive), PF, and UF were similar to one another. The effect of E-43 concentration on failure mode (**Fig. 4**) indicates that adhesive failure was highest at 0 percent E-43 concentration, decreased appreciably when E-43 was added to the interface, and slightly increased with an increase in concentration level. This trend coincided with the results of interfacial shear strength (**Table 3**).

Conclusions

Interfacial shear strength measured by a modified single-fiber pull-out test indicated that the overall average interfacial shear strength was highest in PP and lowest in LDPE. ISO-bonded LDPE without E-43 treatment showed the highest interfacial shear strength among all other LDPE model composites. The E-43-treated wood surfaces without addition of adhesive displayed overall superior performance than all other adhesive-bonded LLDPE. Addition of E-43 greatly enhanced the interfacial shear strength for all plastic types, except for ISO-bonded LDPE and LLDPE. The effect of E-43 was most pronounced in PP, and less in LDPE and LLDPE. However, increasing E-43 concentration from 10 percent to 40 percent had a negative effect on interfacial shear strength. Regression analyses for the interfacial shear strength on E-43 solid content for all adhesive and plastic type combinations showed that interfacial shear strength could be partly explained (20 to 75%) by solid E-43 content, except in PF-bonded LDPE. There was no correlation between interfacial shear strength and solid resin content for all adhesive types.

Adhesive failure prevailed in LDPE and LLDPE model composites while PP showed predominately cohesive failures either in the wood dowel or in the plastic matrix. About one third of PP model composites exhibited dowel failures at the grip end, resulting in undetermined failure mode; therefore, their true interfacial shear strengths are believed to be higher than the apparent interfacial shear strengths. Results of failure mode were in good agreement with interfacial shear strengths. This modified single-fiber pull-out test showed great feasibility for measuring interfacial shear strength. However, air bubbles resided at the interfaces of approximately 80 percent of the model composites which might impair stress transfer. Further improvement in this method would increase its applicability.

Literature cited

American Soc. for Testing and Materials (ASTM). 2004a. Standard test method for melt flow rates of thermoplastics by extrusion plastometer. D1238-04c. Annual Book of Standards, Vol. 08.01. ASTM, West Conshohocken, Pennsylvania.

__________. 2004b. Standard test methods for density and specific gravity (relative density) of plastics by displacement. Annual Book of Standards, Vol. 08.01. ASTM, West Conshohocken, Pennsylvania.

__________. 2004c. Standard test method for tensile properties of plastics. D638-03. Annual Book of Standards, Vol. 08.01. ASTM, West Conshohocken, Pennsylvania.

Bledzki, A.K., S. Reihmane, and J. Gassan. 1996. Properties and modification methods for vegetable fibers for natural fiber composites. J. Appl. Polym. Sci. 59:1329-1336.

Collier, J.R., M. Lu, M. Fahrurrozi, and B.J. Collier. 1995. Reactive composite systems. *In*: Woodfiber-Plastic Composites: Virgin and Recycled Wood Fiber and Polymers for Composites. D.F. Caulfield, R.M. Rowell, and J.A. Youngquist, Eds. Forest Products Soc., Madison, Wisconsin. pp. 67-73.

Felix, J.M. and P. Gatenholm. 1991. The nature of adhesion in composites of modified cellulose fibers and polypropylene. J. Appl. Poly. Sci. 42:609-620.

__________ and __________. 1994. Effect of transcrystalline morphology on interfacial adhesion in cellulose/polypropylene composites. J. Mater. Sci. 29:3043-3049.

__________, __________, and H.P. Schreiber. 1994. Plasma modification of cellulose fibers: Effect of some polymer composite properties. J. Appl. Poly. Sci. 51:285-295.

Hwang, C.Y. 1998. Effect of recycled fiber, compatibilizer and preformed fiber handsheet on the performance of wood-polyolefin composites. PhD. dissertation, Louisiana State Univ. 194 pp.

__________, C.Y. Hse, and T.F. Shupe. 2005. Effects of recycled fiber on the properties of fiberboard panels. Forest Prod. J. 55(11):61-64.

__________, __________, and __________. 2007. Effects of recycled materials on the properties of wood fiber-polyethylene composites—Part 2: Effect of a compatibilizer on the wettability of birch plywood and polyolefins. Forest Prod. J. 57(11):80-84.

Liu, F.P. 1994. Charactering interfacial adhesion between wood fibers and a thermoplastic matrix. PhD. dissertation. West Virginia Univ., Morganton, West Virginia.

Luo, X., R.S. Benson, K.M. Kit, and M. Dever. 2002. Kudzu fiber-reinforced polypropylene composites. *In*: Proc. Sixth Intl. Conf. on Woodfiber-Plastic Composites. Forest Products Soc., Madison, Wisconsin. pp. 35-42.

Miller, B., P. Muri, and L. Rebenfeld. 1987. A microbond method for determination of the shear strength of a fiber/resin interface. Comp. Sci. Tech. 28:17-32.

Sanadi, A.R., R.M. Rowell, and R.A. Young. 1992. Estimation of fiber-matrix inter-facial shear strengths in lignocellulosic-thermoplastic composites. *In*: Materials Interactions Relevant to Recycling of Wood-Based Materials: Proc. Materials Res. Soc. Symp., April 27-29, 1992. R.M. Rowell, T.L. Laufenberg, and J.K. Rowell, Eds. Materials Res. Soc., San Francisco, California. 266:81-92.

__________, __________, and __________. 1993. Evaluation of wood-thermoplastic-interphase shear strengths. J. Mater. Sci. 28:6347-6352.

SAS. 1999. SAS/STAT® User's Guide, Version 8. SAS Inst. Inc., Cary, North Carolina. 3884 pp.

Shaler, S.M. 1993. Mechanics of the interface indiscontinuous wood fiber composites. *In*: Wood-Fiber/Polymer Composites: Fundamental Concepts, Processes, and Material Options. Proc. First Wood Fiber-Plastic Composite Conf., M.P. Wolcott, Ed. Forest Products Soc., Madison, Wisconsin. pp. 9-14.

Spear, M., C. Hill, and J. Tomkinson. 2004. Transcrystalline interphases in polypropylene composites: Exploring the possibilities. *In*: Proc. Seventh Intl. Conf. on Woodfiber-Plastic Composites. Forest Products Soc., Madison, Wisconsin. pp. 49-57.

Snijder, M.H.B. and H.L. Bos. 2000. Reinforcement of commodity plastics by annual plant fibers: Optimization of the coupling agent efficiency. *In*: Proc. Fifth Intl. Conf. on Woodfiber-Plastic Composites. Forest Products Soc., Madison, Wisconsin. pp. 123-129.

Tai, W.C., S.L. Quarles, and T.G. Rials. 1992. The effect of compatibilizers on interfacial bonding in lignocellulosic fiber/polyethylene composites. *In*: Cellulosics: Chemical, Biochemical and Material Aspects. J.F. Kennedy, G.O. Phillips, and P.A. Williams, Eds. Ellis Horwood, New York. pp. 507-512.

Tao, W., J.R. Collier, and B.J. Collier. 1993. Evaluation of interfacial adhesion in sheath/core composite fibers. J. Appl. Polym. Sci. 47: 1115-1122.

Tze, W.T.Y., S.C. O'Neill, D.J. Gardner, C.P. Tripp, and S.M. Shaler. 2004. Coupling polystyrene and cellulose fibers with hydrophilic and hydrophobic silanes: Effects on interfacial properties. *In*: Proc. Seventh Intl. Conf. on Woodfiber-Plastic Composites. Forest Products Soc., Madison, Wisconsin. pp. 29-35.

Westerlind, B., M. Rigdahl, H. Hollmark, and A. De Ruvo. 1984. Interfacial properties of regenerated cellulose fiber and thermoplastic system. J. Appl. Polym. Sci. 29:175-185.

Wu, Y., C.Y. Hse, E.T. Choong, and C.Y. Hwang. 1994. Effect of resin variables and plastic components on wood-plastic composites. *In*: Proc. 2nd Pacific Rim Bio-Based Composites Symp. P.R. Steiner, Ed. Univ. of British Columbia, Vancouver, Canada. pp. 64-71.