Thermoplastic Polymers as Modifiers for Urea-Formaldehyde (UF) Wood Adhesives. II. Procedures for the Preparation and Characterization of Thermoplastic-Modified UF Wood Composites

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ABSTRACT: Wood composites were prepared by using wood flour (sugar maple, *Acer saccharum* March) and thermoplastic-modified urea-formaldehyde (UF) suspensions. Thermoplastic (5–10% w/v) was introduced into the UF suspension as an aqueous solution, a self-stabilized dispersion in water, or as a surfactant-stabilized latex. The modified suspension was blended with wood flour, and the blend was cured by using a cure cycle that was suitable for all the thermoplastic-modified UF formulations and unmodified UF controls. The wood flour composites were tested by using a notched Izod impact strength test. All formulations containing surfactant decreased the impact strength by ~ 30–40% relative to the unmodified UF control, whereas the water-soluble thermoplastic had no effect on the impact strength. The formulations with self-dispersed thermoplastics all increased the notched Izod impact strength, with the greatest increase being 69% more than the UF control, except in a single instance when the molecular weight of the thermoplastic was very high, which decreased resin flow. Increasing the thermoplastic content from 5 to 10% w/v did not further improve the impact test results. Scanning electron microscopy of the fracture surfaces showed morphological differences in the systems that varied with the thermoplastic and method of thermoplastic addition to the UF suspension. © 2002 Wiley Periodicals, Inc. J Appl Polym Sci 87: 898–907, 2003

Key words: thermoplastic; composite; adhesive

INTRODUCTION

The use of wood composites has increased as the demand for wood products grows and as the properties and potential applications for wood-composite products have improved. Urea–formaldehyde (UF) is the least expensive of the major thermoset wood adhesives, but its use is limited because its mechanical properties and moisture resistance are inferior to those of more expensive adhesives such as phenol-formaldehyde (PF) and melamine-formaldehyde (MF). The potential applications for UF resins would increase substantially if the moisture resistance and mechanical properties of UF could be brought closer to those of MF and PF while still maintaining a cost advantage over these resins. If a modified UF was developed with improved properties and was less expensive than PF and MF, then even if the properties do not equal those of MF and PF, additional applications for this UF would still be likely.

We have been investigating the use of thermoplastic polymers to improve the properties of UF wood adhesive. Our objective is to develop thermoplastics and methods of adding thermoplastics to the aqueous UF suspension that do not increase the viscosity of the suspension beyond that which can be accommodated with conventional industrial spray equipment and do not significantly alter the cure cycle of the UF, that will improve both the toughness and the moisture resistance of UF wood adhesive, and that can do so while maintaining a cost savings relative to MF and PF. Our initial investigation[1] looked at different methods of adding thermoplastic copolymers to the UF suspension and studying the effect of the modifier on the suspension viscosity. We studied hydrophobic and hydrophilic thermoplastics that were prepared via free-radical polymerization. The most hydrophobic thermoplastics were introduced into the UF suspension as a surfactant-stabilized latex and the most hydrophilic thermoplastic was introduced as an aqueous solution. Those thermoplastics that possessed intermediate hydrophilicity were made by using surfactant-free emulsion polymerization or in solution in an acetone–water mixture, but were able to form stable dispersions in water without the aid of surfactant. These were termed self-dispersed or self-stabilized dispersions. All of the thermoplastics used, regardless

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of the method used to introduce them into the aqueous UF suspension, did increase the viscosity of the UF suspension, but the increase was not excessive. At a thermoplastic loading level of 5% w/v of thermoplastic in the UF suspension (58% total solids), the modified suspension viscosity remained well within our target range of 75 to 200 cP at 30°C and was typically around 100 ± 25 cP. When the thermoplastic loading level was at 10% w/v (63% total solids), the modified suspension viscosity slightly exceeded our maximum desired value of 200 cP, but adjusting molecular weight or surfactant amounts would be sufficient to bring the viscosity back into our target range.

Collectively, these methods afforded sufficient versatility to permit us to introduce thermoplastics with different degrees of hydrophilicity into the UF suspension and to keep the viscosity of the modified suspension within a range that could be processed with conventional spray equipment. The second goal of this project was to investigate the effect of the thermoplastic modifiers on the mechanical properties of wood composites. At this early stage of the research, we decided to make wood flour composite specimens (by using *Acer saccharum*, sugar maple) and to use a notched Izod impact test. Although wood flour composites are not a commercial product and notched Izod impact tests are not typical mechanical tests, we selected this for our preliminary study for several reasons. First, the smallest operational scale we could employ for particleboard was 10 pounds, whereas we could make the wood flour composites in small batches of ~ 100 mL of modified suspension with ~ 50 g of wood flour and obtain 25 test specimens to evaluate a formulation. Second, although impact data are not directly related to more common wood composite mechanical properties such as internal bond (IB) strength or modulus of rupture (MOR), impact tests might serve as a rough screening tool for modified UF formulations. That is because for a wood composite to have a high impact strength, the adhesive must be able to dissipate energy rapidly and must also have a good adhesive bond to the wood. For a good IB strength and MOR, the adhesive must be able to dissipate energy, although not necessarily on a rapid time scale, and also have a good adhesive bond to the wood. Therefore, although some adhesives might be able to perform well in IB strength or MOR tests that would not perform well in impact strength tests, it seemed likely that an adhesive that gave a good impact test result would also perform well in more standard wood composite tests. Consequently, for these very early studies, the small batch size and ease of preparation made this test method an attractive screening tool for us. Also, in these early studies, we looked at the fracture surfaces of the wood flour composites to study the effect of the method of thermoplastic addition (with and without surfactant) and thermoplastic composition on the fracture surface in an attempt to relate these variables to the impact strength results.

This article reports the relationships between the molecular weight of a thermoplastic, the thermoplastic loading, and the methods of introducing a thermoplastic into UF, and the resulting notched Izod impact strength of wood flour composites prepared with the thermoplastic-modified UF.

METHODS

Materials

The monomers, methylmethacrylate (MMA), acrylamide (AM), acrylic acid (AA), 1-vinyl-2-pyrrolidinone (VP), ethyl acrylate (EA), and vinyl acetate (VA), were purchased from Aldrich Chemical Co. (Milwaukee, WI). The initiator 2,2-azobis(2-methyl propionamide dichloride) (V50) and the surfactant Tween40 were purchased from Aldrich. Acetone was purchased from Fisher Chemical Fisher Scientific (Fair Lawn, NJ). Urea formaldehyde (U : F ratio 1 : 1.18) was donated by Southeastern Adhesives Co. (Lenoi, NC) and was 60% (w) solids. Mold release agent (tetrafluoroethylene telomer/CO_2 type) was purchased from Miller-Stephenson (Danbury, CT). Wood flour (*A. saccharum* Marsh, sugar maple) was donated by Horner Flooring Co. (Dollar Bay, MI).

Instrumentation

Wood composite specimens were compression molded by using a 12-101 T Wabash Hydraulic Press (Wabash Metal Products Co., Inc., Wabash, IN). Impact tests (ASTM D256, 1997) were performed on wood composite specimens that were first notched by using a CS-93E sample notcher (Custom Scientific Instruments, Cedar Knolls, NY) and then tested on a Tinius Olsen Model 92 Impact Tester for Plastics (Tinius Olsen Testing Machine Co., Willow Grove, PA). A JEOL 35C scanning electron microscope (SEM) (JEOL Ltd., Tokyo, Japan) was used at 20 kV to observe the fracture surfaces of wood composites that had been used as impact test specimens. The SEM specimens were gold coated prior to observation by using an E5000 sputter coater (Edwards High Vacuum, BOC Ltd., Crawley, Sussex, U.K.).

Preparation of thermoplastic-modified UF suspensions

The procedure for the synthesis of the thermoplastic modifiers was described in a preceding article.[1] Different thermoplastics were added into the aqueous UF suspension as a concentrated aqueous solution, a concentrated self-stabilized dispersion, or a concentrated

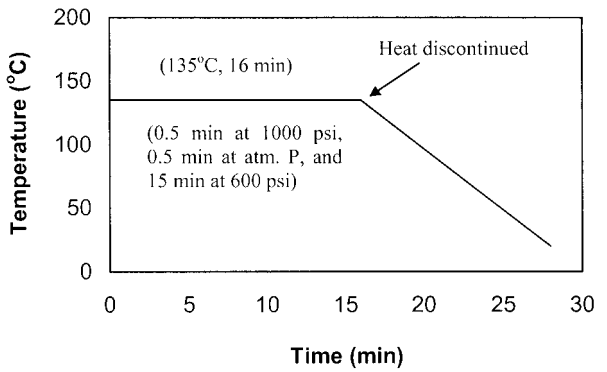

Figure 1 Cure cycle used to prepare wood composites for impact testing.

surfactant-stabilized latex. The mixture was blended together and then the NH_4Cl catalyst was added (0.1 g, 0.1% w/v UF suspension). Sufficient water was then removed under reduced pressure to afford a UF suspension with ~ 58% total solids (5% thermoplastic) or 63% total solids (10% thermoplastic) in the modified UF suspension.

Molding of wood flour composites

Wood composite specimens were prepared by blending wood flour (50 g) into the aqueous UF suspension (100 mL, with or without thermoplastic) containing NH_4Cl catalyst (0.1 g, 0.1% w/v UF suspension). The mixture (~ 45% wood by mass) was immediately transferred into molds (64-mm-long, 12.7-mm-wide, and 3.8-mm-thick that had been sprayed with release agent and allowed to dry before use) and cured in a hot press. A single cure cycle was used for all the thermoplastic-modified UF suspensions as well as the unmodified UF adhesive control. This cycle is shown in Figure 1. The cure cycle appeared to allow a uniform cure and yielded crack-free specimens. After removing the specimens from the mold, the edges and surfaces were buffed with 40-grit paper followed by buffing with 20-grit paper and then notched. The procedure was in accordance with ASTM D256.

Mechanical testing of wood composites

Notched wood flour composites were placed in a vertical clamp in the impact tester. The pendulum was allowed to strike through the specimen and the breaking energy (B.E.) was read from the instrument panel. The impact strength was calculated as:

$$\text{Impact strength} = \text{Breaking energy (J)}/\text{Width of specimen (mm)}$$

A minimum of 20 test specimens was used to generate each data point and standard deviation. The statistical significance of the data was determined by using *t*-test analysis. Data points were compared to control values and to one another to confirm their statistical significance.

RESULTS AND DISCUSSION

Wood flour composites were prepared by using maple wood flour and UF resin in a weight ratio of resin to wood of ~60 : 50, or ~ 45% wood by weight. This ratio afforded good quality composites and was selected to allow resin properties to dominate the composite mechanical properties and allow modifier effects to be evident. A single cure cycle was used for the unmodified UF control and all modified UF formulations. The notched Izod impact strength of the wood composites was studied as a function of the method of introduction of the thermoplastic into the UF suspension (solution, self-stabilized dispersion, surfactant-stabilized latex), the thermoplastic composition, thermoplastic molecular weight, and thermoplastic loading in the suspension. The morphology of the composite specimens was observed by using SEM and was compared with the SEM of the wood flour (Fig. 2) and the unmodified UF control (Fig. 3). Comparison of the micrograph of the unmodified UF control with that of the wood flour shows that the unmodified UF has thoroughly coated the wood surface and that it adheres well to the wood because even after fracture the wood structure is no longer seen. The SEMs of the modified UF composites are compared with the unmodified UF control, and inability to see the wood surfaces is taken as an indication of good adhesion to the wood.

Effects of thermoplastic composition and blending method

The thermoplastic composition influences the method of blending thermoplastic into the UF because if the thermoplastic is not water-soluble, methods must be employed to stabilize it in water as a dispersion so that it can be blended into the aqueous UF suspension. Because the thermoplastic composition influences how a thermoplastic can be introduced into the UF, the method of blending the thermoplastic and its composition are interrelated. Hydrophobic thermoplastic might require the assistance of surfactant to form a stable dispersion in water. However, some thermoplastics that are not water-soluble can be stabilized in water with the use of ionic end groups (from initiator fragments) or suitable hydrophilic comonomers in the backbone, or some combination of these two approaches. Here we employed both approaches. A free-radical initiator, V50, gave ionic end groups that helped stabilize the copolymers in water and might interact chemically with the UF. We also employed

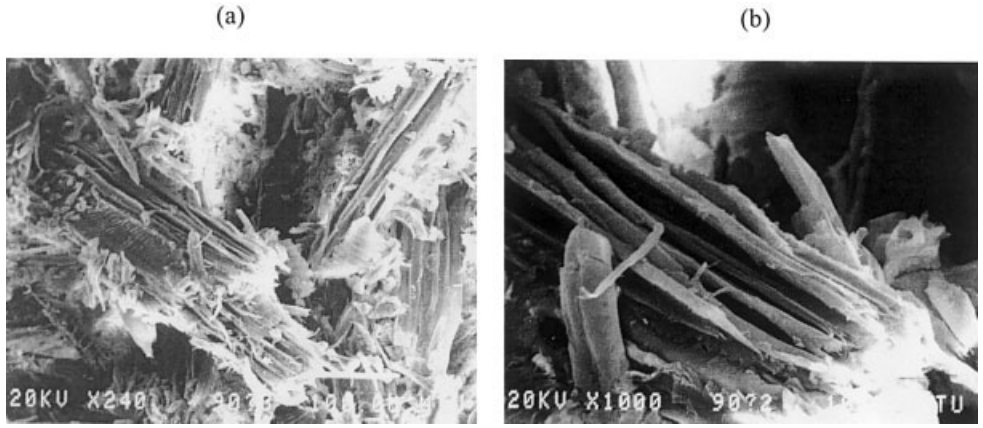

Figure 2 SEM micrographs of sugar maple wood flour at (a) ×240 magnification and at (b) ×1000 magnification.

acrylamide, a hydrophilic comonomer, that also might react with the UF to bind the thermoplastic to the thermoset network.

Several thermoplastic formulations with different degrees of hydrophilicity were evaluated. VP : AM (2 : 10) was water-soluble (WS) and introduced into the UF as a solution. MMA : AM (2 : 10) and (5 : 10) were prepared via solution polymerization in acetone/water, isolated, and then self-stabilized dispersions in water (SD) were blended with the UF suspension. Hydrophobic thermoplastic formulations, MMA : EA : AM : VA (30 : 35 : 25 : 10) and EA : AM : VA (65 : 25 : 10), were introduced as surfactant-stabilized lattices (SS). To confirm the effect of surfactant, the MMA : AM (2 : 10) was also prepared via emulsion polymerization and introduced as a surfactant-stabilized latex (SS), and the EA : AM : VA (65 : 25 : 10) was prepared via a surfactant-free emulsion polymerization with V50 initiator and introduced as a self-stabilized dispersion (SD). The impact strengths of the wood flour composites prepared with these methods and thermoplastic modifiers are shown in Table I.

The water-soluble VP : AM (10 : 2) does not increase the impact strength of the wood composite. The fracture surfaces of cured resin plaques of UF modified with VP : AM (10 : 2) were observed earlier by SEM and found that this thermoplastic does not phase separate from the UF matrix.[1] Observation by SEM of the wood composite prepared with UF modified with 5% w/v of VP : AM (10 : 2) also shows no evidence of phase separation of the thermoplastic within the UF phase (Fig. 4). The fracture surface of this composite appears to be uniformly coated with resin and to adhere well to the fibers and resembles the fracture surface of the unmodified UF control wood composite (Fig. 3). Generally phase separation and interfacial adhesion are important factors for improved toughness in thermoplastic-modified thermosets.[2–5] Consequently, a lack of thermoplastic phase separation may explain these results, and because the modifier does not hinder the adhesion of the UF to the wood, it is probably not surprising that the impact strength of the modified UF does not differ from that of the unmodified control.

Water-soluble thermoplastics might be easily mixed with the UF suspension but they might also be too compatible with the hydrophilic UF to phase separate

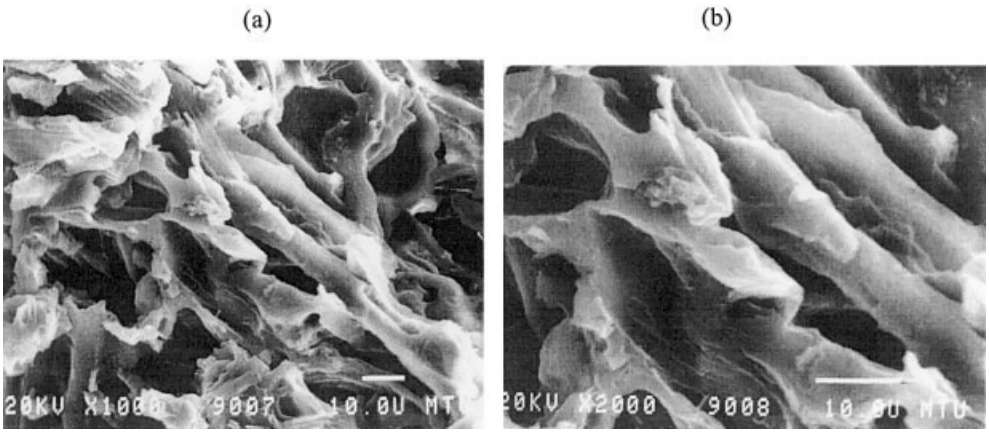

Figure 3 SEM micrographs of the fracture surface of impact test specimens of unmodified UF wood flour composites at (a) ×1000 magnification and (b) ×2000 magnification.

TABLE I
Effect of Thermoplastic (5% w/v) on Notched Izod Impact Strength

Thermoplastics	Blending methods	Impact strength (J/m)	Δ Impact strength (%)
UF control		3.1 ± 0.9	—
VP : AM (10 : 2)	WS	3.0 ± 2.0	0
Low MW MMA : AM (2 : 10)	SD	3.9 ± 0.5	+23
MMA : AM (2 : 10)	SD	5.3 ± 0.9	+69
MMA : AM (5 : 10)	SD	5.0 ± 1.0	+61
Low MW MMA : AM (2 : 10)	SS	2.0 ± 0.8	−36
MMA : AM (2 : 10)	SS	1.8 ± 0.8	−42
EA : AM : VA (65 : 25 : 10)	SD	3.8 ± 0.9	+22
EA : AM : VA (65 : 25 : 10)	SS	2.1 ± 0.7	−33
MMA : EA : AM : VA (30 : 35 : 25 : 10)	SS	2.5 ± 0.9	−21

from it. The self-dispersed thermoplastics are easily blended with the suspension and are less hydrophilic, so phase separation is more favored. In fact, MMA : AM (2 : 10) was clearly observed as phase-separated domains within a UF continuous phase in cured neat resin plaques when fracture surfaces were observed by SEM.[1] The thermoplastic was observed as white domains within a continuous UF matrix that appeared brown. The fracture surfaces of the wood flour composites of UF modified with MMA : AM (2 : 10) and MMA : AM (5 : 10) [Fig. 5(a) and (b), respectively] showed that the modified UF adhesive adhered well to the wood fibers. No sign of the wood-fiber structure is seen in the fracture surface of the composite from UF modified with MMA : AM (2 : 10), seen in Figure 5(a), indicating excellent adhesion. The thermoplastic is thought to be the resin that appears similar to white protuberances in the micrograph (some are indicated in the figure). The fracture surface of the composite prepared with UF modified with MMA : AM (5 : 10), seen in Figure 5(b), differs from that of the UF modified with MMA : AM (2 : 10). In this micrograph, the wood-fiber structure is evident. Fractured wood lumens are seen, although it appears that the wood structure is coated with resin.

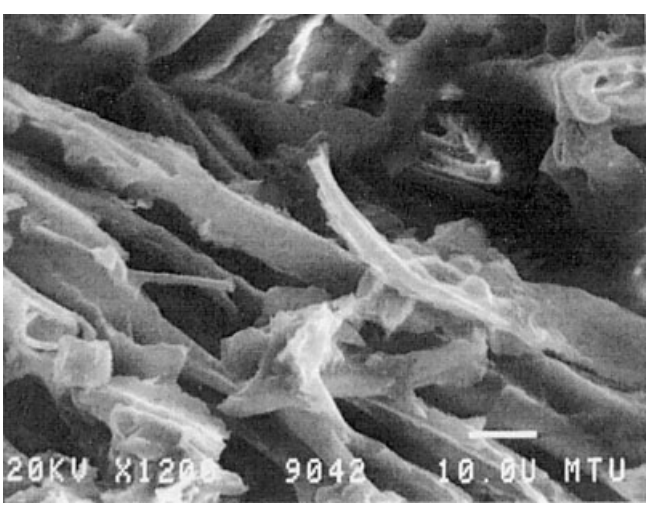

Figure 4 SEM micrographs of the fracture surface of wood composite impact test specimens prepared from UF modified with VP : AM (10 : 2) 5% w/v UF observed at ×1200.

Although the fracture surfaces appear quite different, it is difficult to determine the physical significance of these differences, because both composites showed excellent increases in the notched Izod impact strength. The MMA : AM (2 : 10) formulation afforded a 69% increase in the notched Izod impact strength of the composite, whereas the MMA : AM (5 : 10) gave a 61% increase in impact strength (Table I). A *t*-test statistical analysis showed these values were not significantly different from one another when they were compared (95% probability), but both values were statistically significant when compared to the impact strength of the unmodified control. As stated earlier, notched Izod impact strength does not correlate directly with more standard composite mechanical properties such as IB strength, but the increases in impact strength are sufficiently large that these modified resins are candidates for future investigation with particleboard. The data show that the increase in MMA content from MMA : AM (2 : 10) to (5 : 10) did not significantly change the impact strength but the morphology of the two composites was quite different. The significance of the different morphology is not clear. Typically, materials with different morphologies would be expected to have some behavioral differences, but at this point, no conclusions are being made until more work is done with these adhesives. This is because although the regions shown were selected as representative of the composite areas viewed, it is difficult to be certain that they represent the macroscopic morphology.

The hydrophobic thermoplastics MMA : EA : AM : VA (30 : 35 : 25 : 10) and EA : AM : VA (65 : 25 : 10) were added to UF as surfactant-stabilized lattices. The impact strengths of composites prepared from these adhesives decreased by 21 and 33%, respectively, relative to the UF control (Table I). A *t*-test analysis confirmed that the decreases were statistically significant relative to the unmodified UF control. However, the differences in impact strength of the two thermoplastic-modified UF resins were not statistically sig-

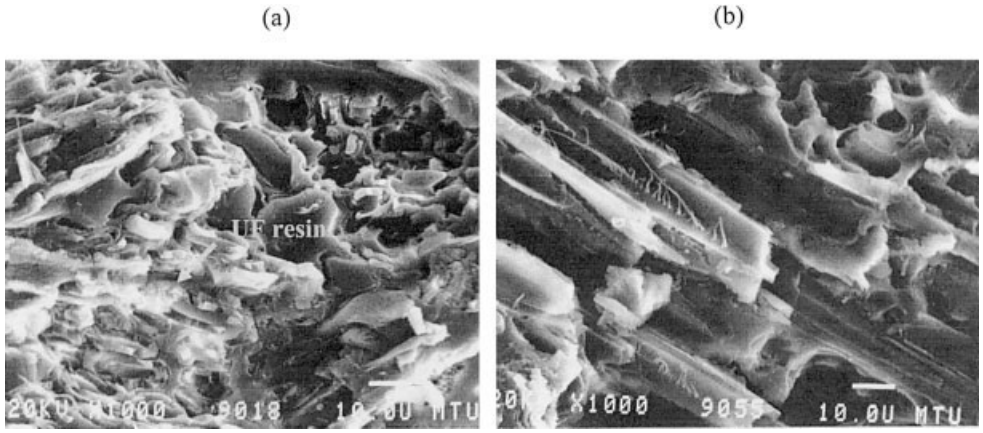

Figure 5 SEM micrographs of the fracture surface of wood composite impact test specimens prepared from UF modified with (a) MMA : AM (2 : 10) 5% w/v UF observed at ×1000 magnification; (b) MMA : AM (5 : 10) 5% w/v observed at ×1000 magnification.

nificant when compared to each other according to the *t*-test analysis. To determine if the surfactant was the reason the modified adhesive afforded composites with decreased impact strength relative to the unmodified control, MMA : AM (2 : 10) was introduced into the UF as a surfactant-stabilized latex, by using both a low molecular weight thermoplastic and the benchmark molecular weight. The notched Izod impact strength of the composites (Table I) decreased by 36 and 42%, respectively, relative to the unmodified UF control. Again, a *t*-test analysis showed the decreases were significant relative to the control but not from one another. The benchmark molecular weight MMA : AM (2 : 10) afforded a 69% increase in the impact strength relative to the unmodified control, ~ 110% greater than the UF modified with the same thermoplastic introduced as a surfactant-stabilized latex. Therefore, the surfactant is detrimental to the impact strength of the composite.

The detrimental effect of surfactant was further corroborated when the hydrophobic EA : AM : VA (65 : 25 : 10) was prepared by using a surfactant-free emulsion polymerization method that allowed it to form a self-stabilized dispersion in water. The dispersion was used to modify the UF and the notched Izod impact strength of the composite from this modified UF was 22% greater than that of the unmodified control, compared with a 33% decrease when this thermoplastic was introduced as a surfactant-stabilized latex. A *t*-test analysis confirmed the 22% increase was statistically significant compared to the unmodified control but the increase in impact strength was less than that measured for the MMA : AM-modified UF when the thermoplastic was introduced as a self-dispersion.

It is evident that surfactant is detrimental to the impact strength of the composites, and presumably all mechanical properties, and the surfactant effectively obscures any compositional effects associated with the thermoplastic modifier. The reason for this is probably desorption of the surfactant from the latex onto the wood surface, hindering the ability of the adhesive to wet and adhere to the wood surfaces. This hypothesis is supported by observation of the composite-fracture surfaces. Representative SEM micrographs of composites prepared from UF modified with surfactant-stabilized lattices are shown in Figure 6 (a)–(d). The wood surfaces are clearly visible with little resin on the wood filaments and little evidence of resin adhesion to the wood.

By contrast, the fracture surface of the composite prepared from UF modified with self-dispersed EA : AM : VA (65 : 25 : 10), shown in Figure 7, is distinctly different from that of the surfactant-stabilized EA : AM : VA (65 : 25 : 10) shown in Figure 6(a,b). Although the modified UF appears to have aggregated with little resin coating the wood fibers when surfactant was present, the modified UF resin without surfactant appears to have coated the wood. This is probably why the impact strength for this composite was improved relative to the unmodified UF control but was decreased for the surfactant-stabilized version of the thermoplastic.

The differences in the fracture surfaces of the composites from UF modified with self-dispersed EA : AM : VA (65 : 25 : 10) and the UF modified with MMA : AM (2 : 10 and 5 : 10) may also explain why the hydrophobic EA : AM : VA did not afford the same level of improvement in impact strength afforded by the more polar MMA : AM thermoplastics. The fracture surfaces of the composites containing the polar MMA : AM (2 : 10 and 5 : 10) both show that the wood fibers are completely coated with resin, and in fact, the wood structure in Figure 5(a) is completely obscured. However, the differences between the surfaces of the composites from UF modified with polar MMA : AM (5 : 10) and the hydrophobic EA : AM : VA

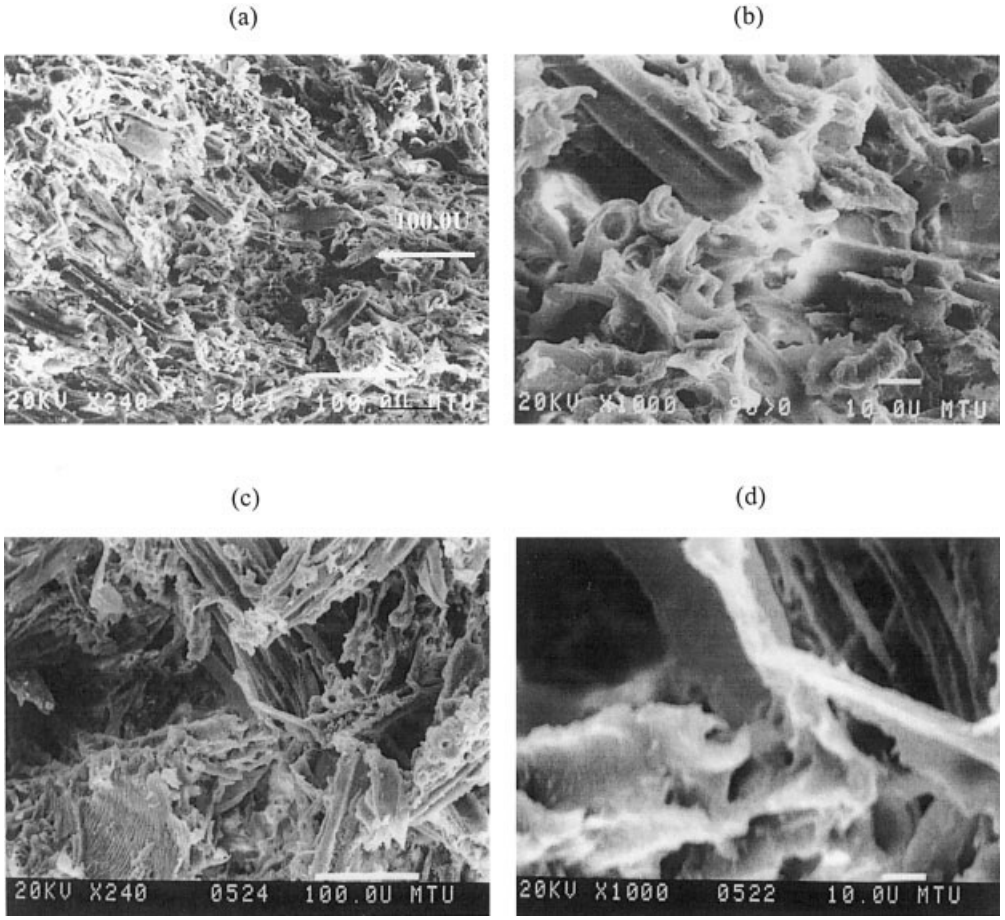

Figure 6 SEM micrographs of the fracture surface of wood composite impact test specimens prepared from UF modified with surfactant-stabilized EA : AM : VA (65 : 25 : 10) 5% w/v UF observed at (a) ×240 magnification and (b) at ×1000 magnification as well as specimens prepared from UF modified with surfactant-stabilized MMA : AM (2 : 10) 5% w/v UF observed at (c) ×240 magnification and (d) at ×1000 magnification.

(65 : 25 : 10) are less dramatic [Figs. 5(b) and 7, respectively]. The wood structure is clearly evident in both composites, and because even though the micrographs shown attempt to show representative features of the composite, the data and numbers of samples observed at this point prevent conclusions from being made.

Therefore, the data are insufficient to explain why the hydrophobic resin did not afford the same increase in impact strength as the composites prepared with the more hydrophilic MMA : AM (2 : 10 and 5 : 10)-modified UF. The data do, however, clearly show that any thermoplastic must be introduced without surfactant to result in improved properties. Also, based on the results with the single water-soluble thermoplastic and from numerous observations made in the toughened-thermoset literature,[6] the modifier should phase separate during the cure to improve toughness.

Effect of thermoplastic molecular weight

The effect of thermoplastic molecular weight on the impact strength of the modified UF wood composites was evaluated by using a series of MMA : AM (2 : 10) thermoplastics. The molecular weight was not measured but was designated as low molecular weight (LMW, prepared with 4 mol % initiator) or high molecular weight (HMW, prepared with 1 mol % initiator) relative to the benchmark molecular weight MMA : AM (prepared with 2 mol % initiator). All other reaction conditions were kept the same to help ensure that the copolymers' composition and sequence distribution were similar.

The impact strengths of wood composites prepared from UF modified with self-dispersed MMA : AM (2 : 10) thermoplastic having different molecular weights are shown in Table II. The composite prepared with UF modified with benchmark MMA : AM possessed an impact strength that was 69% greater than that measured for the unmodified UF control, whereas the impact strength of the composite having LMW thermoplastic was 23% greater than the impact strength of the control. The impact strength of composites prepared from UF modified with the HMW thermoplastic was less than that of the unmodified UF control, al-

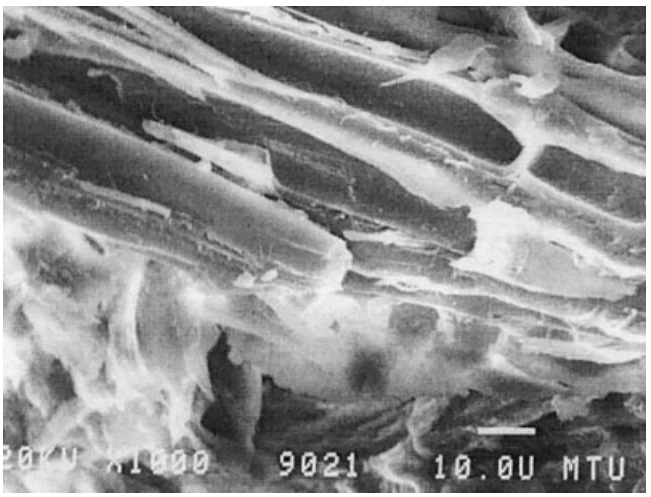

Figure 7 SEM micrographs of the fracture surface of wood composite impact test specimens prepared from UF modified with self-dispersed EA : AM : VA (65 : 25 : 10) observed at ×1000 magnification.

though a *t*-test analysis compared this value with that of the UF control and found that the difference was not statistically significant. However, the impact strength of the wood composites prepared from UF modified with the benchmark and the LMW MMA : AM (2 : 10) were statistically significant when compared to unmodified control and when compared to each other.

These results show that the impact strength of the composites is affected by the molecular weight of the thermoplastic modifier. However, the effect of molecular weight on impact strength appears to be complex. Mechanical properties generally increase with molecular weight up to some critical value and then level off and no additional increases are attained with further increases in molecular weight. If LMW thermoplastic affords smaller improvements in impact strength because the chains are too small to dissipate energy effectively before sample fracture but the benchmark molecular weight chains effectively dissipate energy, then HMW thermoplastic should be more effective or as effective at energy dissipation as the benchmark chains. Therefore, the effect of thermoplastic molecular weight on the adhesive viscosity might also be playing a role. The viscosity (30°C, 58% solids) of the UF modified with HMW thermoplastic was ~ 188 cP, compared with ~ 114 cP for the UF modified with the benchmark molecular weight thermoplastic, and was only ~ 60 cP for the LMW thermoplastic.[1] This is not an excessive increase in viscosity, but the viscosity also increases rapidly as the solids content increases.[1] Therefore, the viscosity increase may have altered the ability of the adhesive to wet and flow across the wood surface and affected the impact strength. A lesser degree of flow does appear to be evident in the SEM micrograph of the fracture surface of the HMW-modified UF [Fig. 8(a)], where not only is no evidence of wood structure seen but also the resin surface is very rough. Again, it is difficult to make macroscopic conclusions on the basis of the small area observed by SEM. However, if the resin had undergone significant flow under pressure, the surface would be smooth. Inadequate flow would result in thick bond lines in some parts of the composite but the overall area of bonding would decrease, which is detrimental to the overall properties of the composite. By contrast, the micrograph of the benchmark-modified UF composite shows smooth resin surfaces indicative of flow, and wood surfaces are not exposed, which would indicate poor adhesion or starved glue lines are seen. The wood surfaces are clearly seen in the fracture surface [Fig. 8(b)] of the composite from UF modified with LMW thermoplastic, which may indicate a starved glue line from excessive resin flow.

The data are not conclusive, but it would have been necessary to modify molding cycles to alter the flow to prove this hypothesis, and that was beyond the scope of the project at this early stage in the investigation. However, the data do suggest the impact strength is being affected by the thermoplastic in multiple ways, and modification of the adhesive viscosity will need to be considered at a later stage in the research.

Effects of thermoplastic loading

The MMA : AM (2 : 10) thermoplastic was introduced into the UF prepolymer as an aqueous dispersion at a loading of 5 and 10% w/v UF suspension. This corresponded to 8.3 and 16.7% thermoplastic weight with respect to the weight of the UF solids. Increases in impact strength of 69 and 57%, respectively, were measured (Table III). A *t*-test analysis confirmed that the differences were statistically significant relative to the unmodified UF control but were not statistically significant when compared to one another. The fracture surface of the composite having the UF containing 10% w/v thermoplastic (Fig. 9) differed from that of the composite modified with UF containing 5% w/v thermoplastic [Fig. 5(a)]. However, although the wood

TABLE II
Effect of Thermoplastic Molecular Weight (5% w/v) on Notched Izod Impact Strength

Thermoplastics	Types	Impact strength (J/m)	Δ Impact strength (%)
UF control	—	3.1 ± 0.9	—
Low MW MMA : AM (2 : 10)	SD	3.9 ± 0.5	+23
MMA : AM (2 : 10)	SD	5.3 ± 0.9	+69
High MW MMA : AM (2 : 10)	SD	2.7 ± 0.7	−12

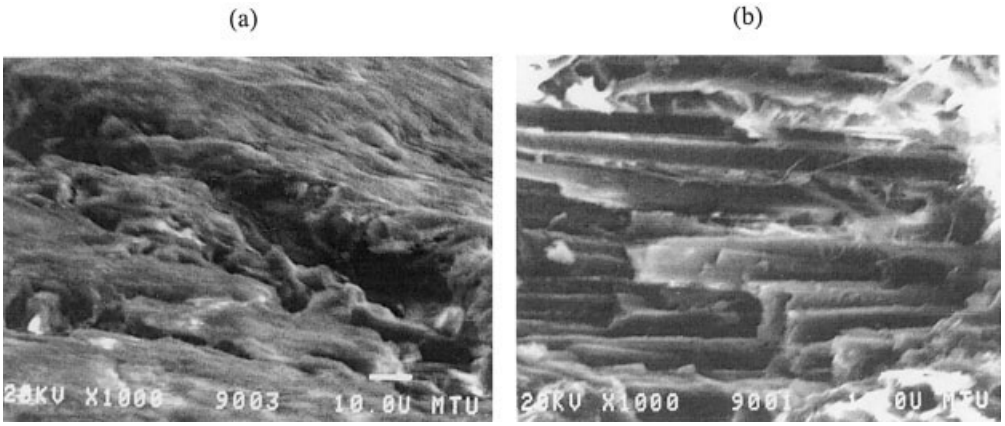

Figure 8 SEM micrographs of the fracture surface of wood composite impact test specimens prepared from UF modified with (a) high molecular weight MMA : AM (2 : 10) 5% w/v UF observed at ×1000 magnification; (b) low molecular weight MMA : AM (2 : 10) 5% w/v observed at ×1000 magnification.

surfaces were thoroughly coated with adhesive, the resin did appear to flow, and the surfaces did not have the roughened appearance seen in Figure 8(a). Again, this is difficult to explain because prior work showed this adhesive possessed higher viscosity than that modified with only 5% of the HMW thermoplastic.[1] The critical role of viscosity in adhesive bonding is well established. For example, Marra described several stages in the formation of an adhesive bond. He stated that, during the press cycle, the resin must flow in a controlled manner: it must wet the substrate; it must transfer to the opposite surface during pressing; it must penetrate the wood pores; and finally, it must set to a solid.[7] Clearly, viscosity plays a role in several of these steps, but the data are not to be explained by viscosity effects alone. At this point in time, the only conclusion that can be made is that increasing the thermoplastic loading level to 10% w/v does not improve the impact strength, but does increase the cost both in the cost of the modifier itself and in making the adhesive more difficult to spray. Therefore, it seems that no more than 5% thermoplastic seems to be required.

CONCLUSION

Thermoplastic copolymers were prepared via radical polymerization and introduced into a UF suspension as an aqueous solution, a surfactant-stabilized latex, and a surfactant-free self-stabilized dispersion. The thermoplastic-modified UF suspensions were mixed with sugar maple wood flour and cured into wood composite specimens by using a single cure cycle for all the formulations, including the unmodified UF controls. The wood composites were tested by using notched Izod impact strength tests. Use of surfactant resulted in decreasing the notched Izod impact strength of all the wood composites regardless of the identity of the thermoplastic, which was attributed to surfactant hindering the adhesion of resin to the wood fibers. Modification of the UF with a single water-soluble resin had no effect on the notched Izod impact strength of the composite. SEM micrographs showed that the modified resin adhered well to the wood but that this particular thermoplastic failed to phase separate within the UF matrix, which is often considered to be important for improving the mechanical properties of a thermoset. Modifying the UF suspension with self-stabilized dispersions of thermoplastic gave significant increases in the notched Izod impact strength, relative to the unmodified UF control, ranging from 22% for a hydrophobic thermoplastic EA : AM : VA (65 : 25 : 10), to 69% for hydrophilic MMA : AM (2 : 10). The only self-dispersed resin that gave no increase in the impact strength was HMW MMA : AM (2 : 10). This may have been due in part to decreased resin flow, but the reason for the lack of effect of the HMW thermoplastic was not completely clear. When thermoplastic content in the UF was increased from 5% w/v UF suspension to 10% w/v

TABLE III
Effect of Thermoplastic Loading on Notched Izod Impact Strength

Thermoplastics	Forms	Thermoplastic loading (%)	Impact strength (J/m)	Δ Impact strength (%)
Control	—	—	3.1 ± 0.9	—
MMA : AM (2 : 10)	SD	5	5.3 ± 0.9	+69
MMA : AM (2 : 10)	SD	10	4.9 ± 0.9	+57

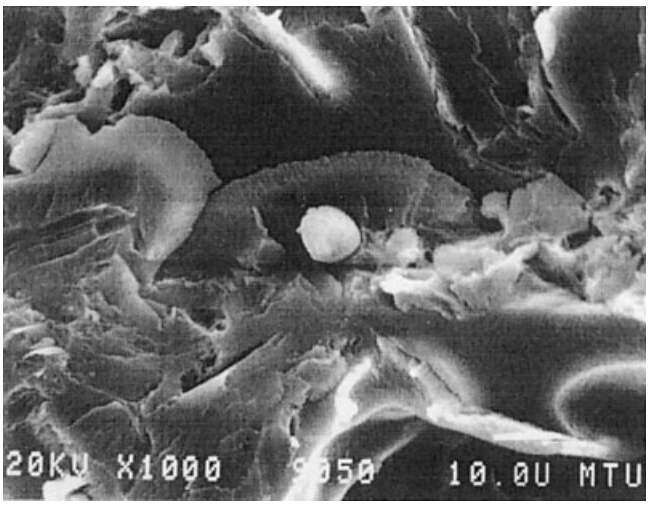

Figure 9 SEM micrographs of the fracture surface of wood composite impact test specimens prepared from UF modified with MMA : AM (2 : 10) 10% w/v UF observed at ×1000 magnification.

UF suspension, no additional increase in the impact strength of the wood composites was found. Consequently, 5% thermoplastic modification, which is less costly and easier to spray, seems to be the preferred modification level.

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