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High-soy-containing water-durable adhesives

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Abstract—Water-resistant bonds are important in many wood products and have been hard to obtain with many bio-based adhesives. Using a three-step process, water-soluble soy flour has been converted into adhesives that cure into an insoluble material for water-durable adhesives. The process consists of denaturation of soy flour, followed by modification with formaldehyde and then conversion *via* co-polymerization with a suitable cross-linking agent into an insoluble material. Both formaldehyde and phenol-formaldehyde are used as cross-linking agents. The extent of conversion into the cross-linked copolymer was assessed with a 24-h water extraction procedure and via elemental analysis. Soy-based resins with 44–86% conversion of soluble soy flour have been successfully prepared with up to complete conversion of the protein component. These resins were also used to prepare strandboards of comparable performance to a control commercial phenol-formaldehyde resin. A direct relationship between the percentage of soy flour incorporated and the final board performance was obtained.

Keywords: Soy; soybeans; phenol; formaldehyde; wood adhesive; durable adhesive.

1. INTRODUCTION

The use of soy-based adhesives is not new to the chemical world. Traditional soy-based systems were common many years ago, and they were studied extensively in the late 1920s and 1930s for their use as adhesives [1]. Soy flour, produced by grinding the meal after removal of the more valuable oil from the soybean, is high in protein. This protein is considered to be the main adhesive material, although carbohydrates and the carbohydrate–protein Maillard reaction should not be overlooked for their contributions [2]. Early work mainly involved exploring different means of denaturing the soy protein to expose the amide functional groups for maximizing adhesion. It was generally believed that the best adhesive was obtained if the soy flour was mixed with a caustic solution [3–5]. However,

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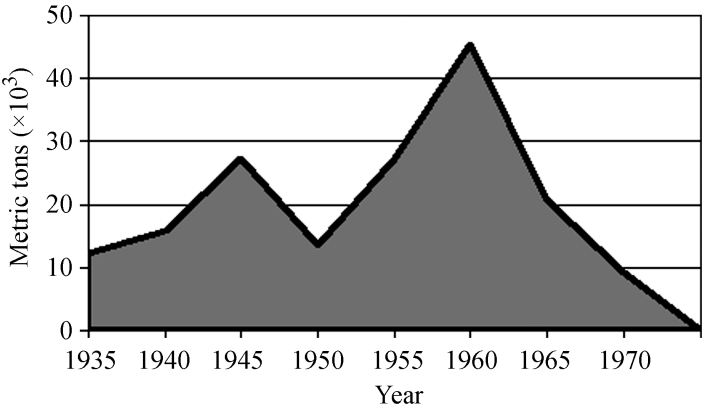


Figure 1. Soy adhesive usage in the United States (F. Trocino, personal communication).

these materials were greatly limited by their very short pot-lives (room-temperature stability), poor biological stability, low solids content, slow press times and, most importantly, poor water resistance. The last characteristic limited the use of these adhesives to mainly interior applications. Petroleum-based adhesives entered the market in the 1940s and soon demonstrated that they were far superior to traditional soy-based adhesives in terms of durability, viscosity, and pot-life. Moreover, by the 1960s, they even offered a lower price. Eventually, these factors led to the nearly complete replacement of soy-based adhesives by petroleum-based adhesives in the wood-bonding arena (Fig. 1) (F. Trocino, personal communication).

Today, phenol-formaldehyde (PF) resins enjoy a dominant place in the resin market for exterior wood composites, and urea-formaldehyde (UF) resins are equally as dominant in the interior wood composites market. With recent increases in petroleum prices, along with formaldehyde emission concerns and general phenol safety issues, the use of naturally renewable soy is again being evaluated as a viable exterior-grade adhesive. Soy flour could also be used for interior applications; however, it is believed that the very-low-cost, fast-curing UF resins would be much harder to displace.

There have been some recent successes in the development of durable soy-based adhesives. Kreibich *et al*. [6] were able to demonstrate the viability of soy flour technology in the finger joining of green lumber. This process involves the use of the much more costly soy protein isolate. In this technology, hydrolyzed soy isolate and a phenol-resorcinol resin are applied to separate pieces of the finger joints, which are then joined together in what is now known as the 'honeymoon' process. Clay *et al*. [7] expanded upon this work and demonstrated that soy flour could also be used in this process. The problem with the honeymoon technology is that it requires storing the soy portion separate from the phenolic or resorcinolic-based binder or cross-linking agent. The premise of this technology is obviously the high reactivity between the two components; thus, a blended one-component resin would offer a limited pot-life. More recently, Hse *et al*. [8] developed a one-

component soy flour/phenol-formaldehyde system for panel boards. However, this work involved using large amounts of caustic NaOH, which resulted in very high pH values and also used low soy levels (30 wt% maximum phenol replacement by soy). Kuo *et al*. [9] have also worked in this area, but this technology often leads to resins with very high viscosities, low solids content, and very short pot-lives. Conner [10] has written a general review of some other efforts.

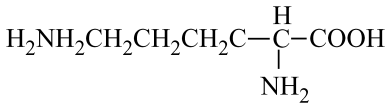
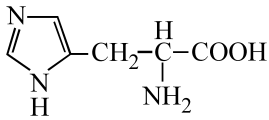
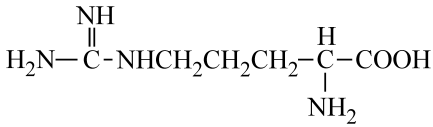
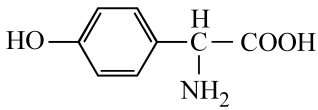
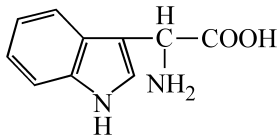
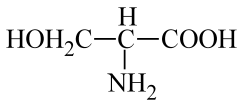
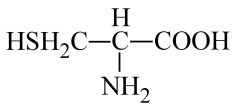
We have overcome the above-mentioned issues by developing a simple three-step process for making durable soy-based adhesives. These steps are as follows: (1) denaturation, (2) modification and (3) co-polymerization.

These steps may be individual operations or combined into a one-pot process. One commonality in all of the previous soy-based adhesive work has been the importance of denaturing or hydrolyzing the soy flour. This has been well studied [11, 12] and is generally recognized as the most critical step in producing protein adhesives that are capable of producing strong bonds with wood, mainly a result of exposing the hydrophilic groups during the uncoiling of the protein. The second step, modification, is carried out with formaldehyde to prepare the soy flour for co-polymerization with suitable cross-linkers, mainly PF resole type resins. Additionally, the formaldehyde modification step is important to reduce or eliminate the biological degradation of the soy flour as observed through an improved resistance to fungal attack, as well as to provide the necessary room temperature stability after the initial co-polymerization with phenol.

Soy protein has been shown to cross-link with formaldehyde, but this reaction is often very easily reversed and does not result in the formation of a durable bond unless a large excess of formaldehyde is utilized [13]. As a result, non-co-polymerized soy adhesive formulations typically do not form water-stable or insoluble dried adhesives. This epitomizes the primary problem with traditional soy adhesive, as it is very likely that the soy does not lose its water solubility after the curing/drying process has taken place. As a result, when subjected to moisture, the soluble adhesive is returned to solution; thus, bond failure is likely at high moisture levels. The authors also realized that other failure modes may still occur, even with a totally water-insoluble adhesive, and are only advocating that water insolubility is one requirement. We believe that when a water-based resin is cured it must form a water-resistant material to offer sufficient bond durability. Typically, this occurs by the irreversible formation of a cross-linked network, resulting in a thermosetting adhesive. This behavior is believed to be responsible for the excellent water resistance offered by many commercial resins today, including phenol-formaldehyde (PF), melamine–urea-formaldehyde (MUF) and isocyanates (often polymeric diphenylmethane diisocyanate, pMDI). We feel that this approach is also possible with soy flour.

Soy flour contains a large amount of proteins that have many potentially reactive side-chain amino-acid groups (Table 1) [14]. Many of these amino acids have been shown to react with formaldehyde and to undergo further condensation [15, 16]. It is this reactive nature that provides soy resin with the ability to form thermoset-

Table 1.
Potentially reactive side-chain amino acids in soy protein [13]

Amino acid	Structure	wt%
Lysine		6.8
Histidine		3.4
Arginine		7.7
Tyrosine		4.2
Tryptophan		1.3
Serine		5.4
Cysteine		2.5
Total		31.3

ting networks with suitable cross-linking agents. In effect, these reactive side groups of the amino acids allow the soy to be converted to insoluble material by incorporating them into a thermoset network. However, because a condensation reaction is believed to occur between lysine and tyrosine [17], it is expected that a similar reaction can be realized with lysine and phenol. Notably, the limited flexibility of peptide chains, especially when partially cross-linked, may limit the degree of conversion. Furthermore, not only can the protein fraction of soy flour react with PF-type cross-linking agents, but the carbohydrate fraction may also contribute to additional durability through co-polymerization, either by a Maillard type reaction or *via* re-

action with phenol. This is important as it allows the use of soy flour rather than the high-priced protein isolates for the preparation of these novel durable adhesives.

The objective of this research was to prepare a soy flour adhesive, which was rendered water-insoluble through co-polymerization with PF resins, such that durable composite panels could be made which were capable of meeting high-performance standards, such as the American Plywood Association's (APA) PRP-108.

2. EXPERIMENTAL

2.1. Materials

Soy flour was supplied by Oelwein Custom Commodities (Oelwein, IA, USA). The flour was ground such that 80 wt% passed through a 100-mesh screen. The composition of the flour was found to be 44 wt% protein, 10 wt% oil and 5 wt% ash on a dry basis, with the remainder being mainly carbohydrates. Phenol (99 wt%) and formaldehyde (37 wt%, 8–9 wt% MeOH) were purchased from Aldrich (Milwaukee, WI, USA). Sodium hydroxide (99 wt%) was purchased from Fisher Scientific (Fair Lawn, NJ, USA). Commercial PF was donated by an oriented strandboard (OSB) manufacturer.

2.2. Furnish

The furnish used for the strandboard study comprised black gum, southern yellow pine, and soft maple, with trace amounts of red oak. Strand size was nominally 7.5 cm long by 1.5 cm wide by 0.08 cm high, with a standard deviation of around 20%.

2.3. Soy-D (denatured only)

In a three-neck round-bottom flask equipped with a mechanical stirrer, thermometer and condenser, tap water (778.0 g), NaOH (28.0 g) and the phase-transfer agent ethylene glycol (5.3 g) were added while mixing and heated to 70°C. Soy flour (350.0 g) was then charged to the rapidly stirring solution at an average rate of 5%/min (or as rapidly as possible while ensuring proper dispersion and no clumping). The mixture was then heated to 90°C over 15 min, with rapid agitation, and held between 88 and 92°C for 2 h. After the reaction, the vessel was cooled to 35°C with an ice bath and filtered through a course screen (35 mesh).

2.4. Soy-F (denatured and treated with excess formaldehyde)

In a three-neck round-bottom flask equipped with a stirring rod, thermometer and condenser, tap water (364.0 g), sodium hydroxide (14.4 g) and ethylene glycol (2.7 g) were combined while mixing and heated to 70°C. Soy flour (180.0 g) was then added at an average rate of 5%/min (or as rapidly as possible while ensuring

proper dispersion and no clumping) to the rapidly stirring solution. The mixture was then heated to 90°C over 15 min, with rapid agitation, and held between 88 and 92°C for 1 h. After the denaturing reaction, 37% formaldehyde (104.4 g) was added through an addition funnel over 15 min. The mixture was held at 90°C for 5 h, then cooled to 35°C with an ice bath and filtered through a coarse screen (35 mesh).

2.5. Soy-PF-XX (XX is % soy)

Several soy/PF resins were made using the following procedure. The 40/60 soy/PF (Soy-PF40) example is described in detail. All other examples differ only in the stoichiometry. In a three-neck round-bottom flask equipped with a stirring rod, thermometer and condenser, tap water (270.2 g), NaOH (10.0 g) and ethylene glycol (1.9 g) were added while mixing and heated to 70°C. Soy flour (125.0 g) was then added at an average rate of 5%/min (or as rapidly as possible while ensuring proper dispersion and no clumping) to the rapidly stirring solution. The mixture was then heated to 90°C over 15 min, with rapid agitation, and held between 88 and 92°C for 1 h. After the denaturing reaction, the heat source (mantle) was removed and 37% formaldehyde (115.4 g) was charged over 10 min through an addition funnel to the rapidly stirring denatured soy solution, allowing the temperature to cool to 75°C. The temperature was held at 75°C for an additional 20 min after the addition was completed. Solid phenol (128.6 g) was then added over 10 min. As soon as the phenol addition was completed, NaOH (5.5 g) was added and the mixture was allowed to stir for 20 min, while maintaining a temperature of 75°C. This was followed by a second addition of 37% formaldehyde (115.4 g) over 10 min. After this addition was completed, NaOH (2.7g) was added, followed 10 min later by another addition of NaOH (2.7 g), while always maintaining a temperature of 75°C. The mixture was then held for an additional 90 min at 75°C, then cooled to 35°C with an ice bath and filtered through a coarse screen (35 mesh) and stored at 5°C.

2.6. Analysis

The percentage of non-volatile solids was measured by heating a 1–2-g sample of the resin solution in a small aluminum pan in an oven at 150°C for 1 h. Viscosity was measured at 25°C using a Brookfield LVT viscometer with the #2 or #4 spindles at both 60 and 30 rpm. The specific spindle selections were based on the recommended viscosity ranges for each spindle. The multiple speed measurements were deemed important for proper evaluation of the shear-thinning properties of soy resins. The free formaldehyde level was determined using a hydroxylamine hydrochloride back titration method [18]. The extraction of the neat resins was conducted as follows. Neat cured resin was prepared *via* the same procedure used to measure the percentage of non-volatile solids. A Soxhlet extraction was performed using 2.5–3.0 g of lightly ground cured resin solids. Using water as the solvent, the extraction was allowed to proceed for 24 h. The residue in the thimble was then oven dried for at least 2 h at 150°C, then it was removed from the thimble and weighed.

The percentage of residue was calculated as 100 × gram residue/gram starting resin solids. The amount of soy converted to insoluble material was calculated as follows:

$$\text{Total residue} = \text{Soy residue} + \text{PF residue}. \tag{1}$$

Thus,

$$\text{Soy residue} = \text{Total residue} - \text{PF residue}, \tag{2}$$

where the soy residue, i.e., the total soy-insoluble soy fraction after curing, is defined as

$$\text{Soy residue} = \text{Soy converted} + \text{Native insoluble soy}, \tag{3}$$

where the native insoluble soy is 10% of this soy flour. It is the amount of soy flour that is water insoluble without additional crosslinking. This was determined by extraction of oven dried, non-crosslinked, denatured soy flour.

For the PF component, when subjected to the oven solids method, it is assumed that the phenol and formaldehyde will condense to 75% hydroxymethyl to methylene conversion (based on correlation of measured solids with theoretical solids) and that only the added NaOH will be removed via the extraction procedure. Thus, the PF residue is represented as:

$$\text{PF residue} = \text{PF solids (calculated at 75\% conversion)} - \text{NaOH (in PF solids)}. \tag{4}$$

The amount of soy converted is then calculated from the algebraic manipulation of the four equations.

$$\text{Soy converted} = \text{Total residue} - (\text{PF solids} - \text{NaOH}) - 0.10 \times \text{Total soy}, \tag{5}$$

where Total soy is defined as the dry soy weight percentage of the total resin solids. From this, the percentage of soy converted can be calculated as:

$$\%\ \text{Soy conversion} = 100 \times \text{Soy converted}/(\text{Total soy} - 0.10 \times \text{Total soy}). \tag{6}$$

3. RESULTS AND DISCUSSION

3.1. Soy conversion to insoluble material

Soy flour comprises a large amount of carbohydrates (40 to 45%) in addition to the mainly water-soluble protein. These carbohydrates exist in a variety of forms consisting of soluble oligosaccharides (mainly sucrose) and insoluble cellulose, hemi-cellulose, pectin and starch. Liu [14] has presented an excellent discussion of all the soy flour components. Some of these carbohydrate fractions and some proteins are water insoluble. Thus, prior to drying/curing, even if a sample of soy flour is denatured, it will still contain a measurable amount of water-insoluble material. We termed this the 'native insoluble soy' in this paper to help differentiate

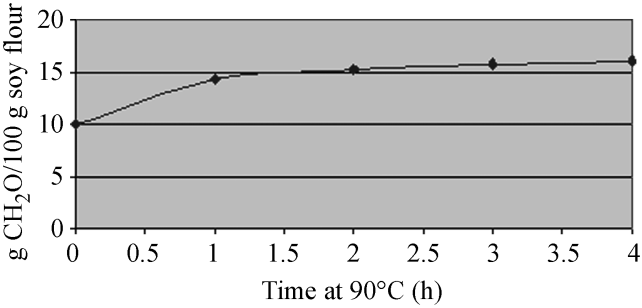


Figure 2. Formaldehyde uptake by soy flour.

it from the soy that has been converted to insoluble material as a result of co-polymerization or cross-linking. Typically, the amount of native insoluble soy will range from 5 to 15%, depending on whether the soy flour has been de-hulled and on the quality of the flour and grind. The soy flour used in this study has a native insoluble soy level of 10%. The amount of soluble soy that is converted to insoluble soy can only be determined if the native insoluble value is known (see the discussion on the extraction procedure in the Experimental section for more details).

The cross-linking of soy protein can occur by a number of different reactions. As it is well accepted that soy protein must be decoiled (denatured) to afford any reasonable adhesive ability, all of these reactions will be studied on denatured material. The first possible reaction can be considered a self-cross-linking reaction and is often observed with the 'heat curing' of proteins [19]. This reaction is described as:

$$\text{Soy} + \text{Soy} \rightarrow \text{Soy–Soy (network)}. \tag{7}$$

This is considered to be a hydrolytically very unstable product and was probably the cause for the poor durability of many of the early soy adhesives.

This reaction is represented best by the Soy-D example in Table 2. In this example, no formaldehyde or phenol cross-linker is present. Thus, upon heating/curing at 150°C for 1 h in an oven, only heat curing is realized. As expected, this reaction did not result in any water durability as indicated by the fact that the percentage of residue was equivalent to the amount of native insoluble soy. This is an excellent demonstration of the typical durability problem associated with many soy adhesives.

The second reaction involves the cross-linking of a formaldehyde-modified soy or the methylolated soy [15, 20]. One possible self-condensation reaction of methylolated soy is shown as:

$$\text{Soy–CH}_2\text{OH} + \text{Soy–CH}_2\text{OH} \rightarrow \text{Soy–CH}_2\text{–Soy (network)}. \tag{8}$$

Soy flour reacts with formaldehyde and reaches a saturated formaldehyde state very quickly. The amount of formaldehyde that is required to saturate the soy is dependent upon the amount of protein present and on the particle size of the flour. The formaldehyde uptake by the soy used in this study is shown in Fig. 2. With this

Table 2.
Physical properties and characteristics of soy-based resins

Resin	Soy (wt%)	pH	Solids (wt%)	Viscosity (cP) at 60/30 rpm	Gel time (min)	Extract[a] (wt%)	Conversion[b] (%)
Soy-D	100	12.2	30.2	1650/2340	>90	90	0
Soy-F	90	8.1	30.4	1690/2270		56	35
Control PF	0	11.2	53.3	240	24.6	24	N/A
Soy-PF30	30	9.9	39.0	100/105	60.3	10	86
Soy-PF40	40	9.9	38.9	220/245	38.9	14	78
Soy-PF50	50	10.1	39.1	500/610	52.6	18	75
Soy-PF70	70	10.3	38.6	5050/6380	83	34	55

[a] Water-soluble extract after 24-h water Soxhlet extraction.
[b] As calculated from equation (6).

particular soy, a 14% uptake is realized after 1 h at 90°C, with only a small increase over the next 3 h. The increased uptake observed after the initial hour is at least partially explained by the occurrence of the Cannizzaro reaction under such alkaline conditions [21], although it is likely that slow reacting amides are also consuming some formaldehyde under these conditions. Because the reaction described in equation (8) is considered reversible unless large amounts of excess formaldehyde are present, we studied this reaction under excess formaldehyde conditions. For resin Soy-F in Table 2, the amount of free formaldehyde was measured to be 6 wt% after the denatured soy was saturated with formaldehyde. Interestingly, with excess formaldehyde present and under these reaction conditions, the soy is able to form some new water-insoluble material. In effect, drying and curing are taking place simultaneously. The amount of soluble soy converted to insoluble soy as a result of this process was calculated to be 35%. The problem with this approach in preparing durable soy-based adhesives is two-fold: first, the conversion to water-insoluble resin is still quite low; second, the amount of free formaldehyde required to achieve this conversion is unacceptable in commercial applications due to the large amount of formaldehyde emissions.

The ability of the denatured, modified soy flour to cross-link with PF resins and form a water-insoluble thermoset system is the main characteristic studied in this paper. One possible reaction can be depicted as

$$\text{Soy–CH}_2\text{OH or Soy} + \text{PF} \rightarrow \text{Soy–CH}_2\text{–PF}. \tag{9}$$

Although much research has been conducted on reaction of proteins with formaldehyde, we have not been able to find any work involving an alkaline co-polymerization with phenol or PF resins. Because of the high reactivity of the hindered tyrosine with methylolated lysine [17], we believed that these reactions were possible.

The formation of the above product (equation (9)) at high conversions should lead to the development of a highly durable, irreversible, soy–PF cross-linked network capable of bonding wood of exterior grade quality. To test this, several reactions were carried out using a range of soy/PF ratios to determine the ability of the soy to

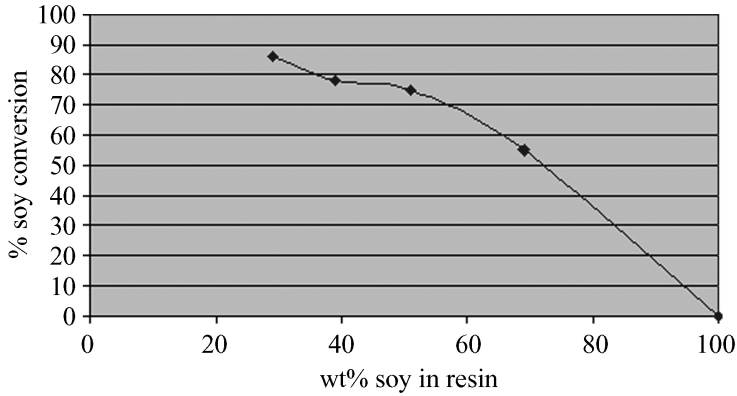


Figure 3. Soy conversion from water-soluble to water-insoluble resin as a function of soy level in the formulation.

co-polymerize in these soy–PF systems. In this study, all the soy was denatured for 1 h at 90°C in the presence of 8% (w/w soy) NaOH. The denatured soy was then treated with ample amounts of formaldehyde to saturate the soy, followed by the addition of phenol and more formaldehyde to produce highly methylolated phenol *in situ* at 75°C. The 75°C reaction temperature was selected to yield large conversions of protein amine groups to methylol with a minimal amount of condensation of these methylols to methylene. The reactions were characterized by separating the individual hydroxymethyl phenol molecules *via* high-pressure liquid chromatography (HPLC) [22]. With the use of model compounds, we were able to identify many of the hydroxymethyl phenol components in the resin solutions. The levels of these compounds were found to decrease at very slow rates over the course of the reaction, suggesting that, under these conditions, there was minimal conversion of hydroxylmethyl phenol to methlyene or other condensed products. A constant level of 2.08 mol formaldehyde per mol phenol was used with 0.2 mol NaOH per mole phenol in all of these experiments. However, it is apparent that the effective F/P ratio (defined as the average number of methylol groups per phenol molecule, as determined by HPLC) of the final adhesive will decrease with increasing soy level as a result of the first-step consumption of formaldehyde by the denatured soy. This too is probably a factor in the co-polymerization reaction and definitely part of the reason why the gel times are generally lengthened as the amount of soy is increased. The longer than expected gel time for the Soy-PF30 warrants further investigation and could suggest that an optimum region exists for the stoichiometry between the soy and PF. Although not yet reported, we have been able to show marked decreases in gel time when higher effective F/P levels are realized with high soy containing resins.

These soy–PF resins demonstrate a direct relationship between the amount of PF cross-linking agent and the percentage of soy converted. This suggests that the cross-linking efficiency may be low and that the soy is, perhaps, more anchored into

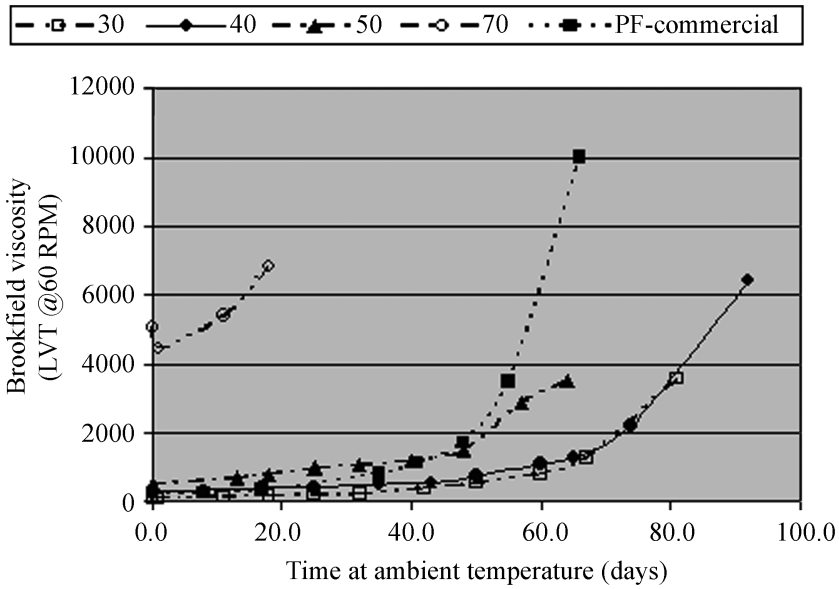


Figure 4. Room temperature storage stability of soy-based resins.

the insoluble resin network and not incorporated with a high cross-link density. Nonetheless, we have been successful at reducing the water solubility of the final soy adhesive at all soy levels. Figure 3 shows the relationship between soy conversion and soy load. Based on this graph, it appears that the soy reacts to high conversion (>70%) with soy loads of 50% or less. However, the 70% soy resin still demonstrates >50% conversion of soy and should still offer improved durability as compared to that of previous soy adhesives.

Two important property characteristics of soy-based technology that have raised concerns in the past are the higher viscosities and short pot-life (stability). Although soy-based resins often have higher Brookfield viscosity values than do many commercial PF resins, it should be recognized that soy-based resins are shear thinning. This implies that Brookfield viscosity measurements may be misleading with respect to the spraying ability of the final adhesive. Therefore, a soy-based resin at 500 cP is actually much less viscous than is a Newtonian PF resin at 500 cP when subjected to a high shear environment like a spraying process. In our history of applying these resins either by air or spinning disk atomization, we have never observed difficulties obtaining adequate resin distribution. Soy loads of 50% or less offer resins with reasonable viscosities (<700 cP). Only the highest soy resin, Soy-PF70, resulted in an unacceptably high viscosity value.

Many soy-based resins also suffer from poor room-temperature stability and, thus, have very short useful pot-lives. This present technology takes into account the importance of room-temperature stability. The order of addition in preparing these resins, most notably the formaldehyde modification step, is critical to preparing a resin of good stability. Figure 4 shows the room-temperature viscosity stability of

Table 3.
Elemental analysis of Soy-PF40 oven-dried solids and post-extraction residue

Material	N (wt%)		C (wt%)		H (wt%)		Extraction (wt%)	
	Theor.	Exp.	Theor.	Exp.	Theor.	Exp.	Theor.	Exp.
Oven-dried solids	2.9	2.8	60.5	58.9	5.8	5.4		
Post-extraction residue	3.1	3.2	65.1	64.6	5.8	5.7	85.7	85.7

Theoretical calculations were made assuming that no protein, all of the NaOH and 40% of carbohydrates and oils were extracted.

these resins when compared to that of a commercial PF resin. All of the soy–PF resins, with the exception of the high soy containing soy-PF70, offer comparable or better room-temperature viscosity stability when compared to that of a commercial PF resin.

3.2. Soy conversion analysis

The question still remains as to what percentage of the protein from soy flour is actually converted to insoluble material *via* co-polymerization with PF. We employed elemental analysis to answer this question. Because the only source of nitrogen within our soy–PF resin systems was from the protein, elemental analysis (C,H,N by Minnesota Valley Testing Laboratories, New Ulm, MN, USA) was used to evaluate pre- and post-extracted solids (residue) to determine the actual amounts of protein consumed by comparing the relative levels of elemental nitrogen. The samples were finely ground to increase the surface area and to reduce the extent of potential resin entrapment that could lead to erroneously high conversion levels. We have found this procedure to be very quantitative in extracting other water-soluble non-proteinaceous materials. This study was carried out on the Soy-PF40 sample, which contained 40% soy. The results demonstrate a good correlation with the theoretical value of the starting solids (Table 3). More importantly, these results show that essentially none of the nitrogen is extracted from the cured solid sample. This suggests that a soy–PF resin prepared under these conditions will have a complete conversion of the protein components to insoluble material. Additionally, approximately 60% of the other soluble components (oil/carbohydrate) is also converted. These results are consistent with the near 100% conversion of protein observed in similar experiments with soy isolates in our laboratory.

3.3. Adhesive performance

The purpose of our study is ultimately the development of durable adhesives for exterior-grade panel production through the preparation of highly water-insoluble soy–PF resins. The prior data in this paper related to making the soy flour more water-insoluble do not indicate whether the polymer network has suitable strength for utility as a durable adhesive. A sampling of results is shown in this paper.

Table 4.
Strandboard panel preparation parameters

Component	Value
Formed mat size	40.6 by 40.6 cm
Trimmed board size	35.6 by 35.6 cm
Starting furnish moisture (face and core)	2.0 wt%
Furnish type	Mixed hard/soft
Face/core ratio	55 : 45
Final thickness	11 mm
Final target density	673 kg/m^3
Face resin	3.26 wt%
Face wax (emulsion)	1.31 wt%
Core resin	3.89 wt% (always PF control)
Core wax (emulsion)	1.39 wt%
Application method	Air atomization
Press temperature	200°C
Press soak times (time at target thickness)	330 s
Press close time (mat contact to target thickness)	40 to 50 s

Table 5.
Properties of soy-PF40 and commercial phenol-formaldehyde containing random strand panels[a]

Face resin	Density (kg/m^3)	Thickness swell (%)				Internal bond strength (kPa)	
		2-h	Boil	24-h	RT	Dry	Wet
PF control	678	62.8	(4.8)	15.2	(1.5)	600 (56)	56 (10)
Soy–PF40	671	65.1	(3.6)	14.5	(1.7)	620 (60)	60 (40)

[a] ASTM 1037. Wet internal bond is determined for center portion of test panel oven dried after 2-h boil. Values in parentheses represent one standard deviation of the data.

We previously reported other panel results from similar resins [23]. Random strandboards were prepared using the resin Soy–PF40 (Table 2) and compared to a commercial PF resin. These resins were used only in the face section of the panels. Panel preparation details are presented in Table 4. We recognize that other variables, such as resin distribution, resin penetration, flake orientation and press conditions, are also of great importance in preparing quality wood composites, and that these too would need to be evaluated in the future. A direct comparison of our soy resins to a commercial resin for this type of OSB furnish was considered the most valid measurement of performance. The soy flour resin (Soy–PF40) produced boards of the same quality as that of the commercial resin prepared panel (Table 5), most notably, excellent low thickness swell at both room temperature and the very aggressive 2-h boil compared to thickness swell of the commercial resin. To our knowledge, no soy-based adhesives, with such high levels of soy flour, which can withstand a 2-h boil test have been produced. These results are consistent with our belief that soy resins can result in durable adhesives when they

are properly modified and copolymerized to convert them into a water-insoluble material. The raw material cost of such a resin is estimated to be 30–40% less than that of a commercial PF resin [22]. These savings could offer huge cost reduction opportunities to panel manufactures without compromising quality.

4. CONCLUSIONS

The preparation of a novel adhesive with high soy content has been described. Based on water extraction and elemental analysis results, we have successfully converted 55–86% of previously water-soluble soy flour into a water-insoluble material *via* co-polymerization with a resole phenol-formaldehyde (PF) resin. These resins were successfully used as a face resin for the preparation of random strandboards with no significant performance differences compared with a control commercial PF resin. Thus, these co-polymerized resins are viable candidates for the manufacturing of many durable composite panels. Moreover, the soy-based resins offer substantial cost savings along with excellent durability. This technology offers a great opportunity for panel manufactures to reduce the cost of face resin by 30–40% by replacing phenol and formaldehyde with non-hazardous soybean flour.

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