

Research article

Preliminary studies of soy meal additives for use in polypropylene wood composites

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Abstract. We have discovered that water-insoluble proteins (IM) isolated from whole soy meal (WM) have the capacity to improve the thermal stability of polypropylene (PP) wood composites without negatively impacting mechanical properties. We compared an undiluted composite to composites diluted with 5% by weight of a 3/1 w/w mixture of stearic acid and paraffin wax (SA/wax), 5% by weight IM, 5% by weight WM, 5% by weight of a 50/50 w/w mixture of IM in combination with SA/wax, and 5% by weight WM in combination with SA/wax. Water contact angle (CA) measurements and Fourier transform infrared spectroscopy (FTIR) revealed a synergy between IM and SA/wax, resulting in enhanced surface hydrophobicity. Thermogravimetric analysis (TGA) showed lower weight loss in composites made with IM up to the onset of the degradation of PP, and reduced rates of lignin degradation, independently of the presence of SA/wax. Dynamic mechanical analysis (DMA) revealed higher modulus and lower viscoelastic loss resulting from the removal of water-soluble soy components from WM. Total reflectance ultraviolet spectroscopy (UV) showed that IM absorbs UV radiation strongly in the UV-B and UV-C regions. Collectively, these findings suggest that IM has the potential to become a multi-purpose additive for use in PP wood composites.

Keywords: particle-reinforced composites, additive, polyolefins, bioadditive, biopolymer, renewable resource biopolymer, thermogravimetric analysis, differential scanning calorimetry, dynamic mechanical analysis, hydrophobicity, spectroscopy

1. Introduction

Polymer/wood composites have found use in industries ranging from automotive and construction to packaging and transportation [1, 2], where end-use advantages include high strength and stiffness, low thermal conductivity, durability, and recyclability [1]. However, challenges include maintaining moisture resistance and mechanical properties under harsh environmental conditions (*e.g.*, exposure to extreme temperatures, high humidity, water, and ultraviolet light) [2]. Moreover, manufacturing challenges include achieving adequate lubrication during processing to mitigate polymer degradation and chain scission, while simultaneously achieving good

particle wetting, dispersion, and homogeneity for optimal mechanical properties [1, 3].

It is well known that the mechanical properties of composites are influenced by interactive contributions from each of the individual components [4–6]. For example, the modulus of a composite [6] is influenced by the modulus and volume fraction of the matrix phase, the modulus and volume fraction of the filler/reinforcing phase, the modulus and volume fraction of the meso phase (*i.e.*, the volume element that constitutes the radial fraction of matrix phase material and other components that are constrained by wetting and adhesion to the filler/reinforcing phase), and the fraction of macro void volume within

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the composite structure, which is influenced by the homogeneity of dispersion and the efficiency of the wetting of the filler/reinforcing phase by the matrix phase [4–6].

The homogeneity of dispersion and the adhesion between the matrix phase and the filler/reinforcing phase is influenced by interfacial contributions (*e.g.*, dispersion forces, instantaneous dipole interactions, permanent dipolar interactions, hydrogen bonding and sometimes covalent bonding) [4–6], and by the physical property contributions of component members (*e.g.*, the viscoelastic and energy absorbing characteristics of the individual component members and the coupled meso phase) [4–6]. Moreover, successful interfacial penetration can result in phase mixing at the boundary between the meso phase and the filler/reinforcing phase, leading to the development of a mixed interphase with its own unique set of mechanical properties [4, 6, 7]. Of course, other factors include the shape (*e.g.*, aspect ratio), the size distribution and the total surface area of the filler/reinforcing phase [4, 6, 7].

Examples of polymers that have been used in polymer/wood composites include high-density polyethylene (HDPE), polypropylene (PP), polyvinyl chloride (PVC), polyethylene terephthalate (PET), polylactic acid (PLA) and polyurethanes (PU) [2], as well as recycled plastics like polyvinylchloride and polypropylene [3]. Internal and external lubricants are typically used to improve processability (*e.g.*, waxes, stearic acid, Zn-stearate, silicone elastomers) [1]. Amphiphilic polymer-to-wood coupling agents are also used in polymer–wood composites as surface compatibilizers between polar cellulosic wood particulates and non-polar polymer binders for the purpose of improving homogeneity and mechanical properties [1]. For example, maleated polypropylene (MAPP) is used as a coupling agent in PP/wood composites because it can wet wood components through its acid/anhydride functionality (*e.g.*, via hydrogen bonding and dipolar interactions) and simultaneously entangle with the bulk PP segments through its lipophilic PP (*e.g.*, via dispersion forces) [8, 9].

Soy-based products have been used in multiple non-food applications, ranging from release agents and emulsifiers (*e.g.*, lecithin) [10] to biodegradable films [10, 11] and wood adhesives [12]. Soy-based natural products have also been explored for use in polymer blends [11], where difficulties have arisen due to poor phase compatibility, inadequate dispersion, and

process limitations, arising in part from soy flour's susceptibility to thermal degradation at temperatures near 200 °C [11]. For example, in studies of soy flour blends with PP and low-density polyethylene, the use of conventional single-step melt processing methods led to inadequate dispersion of the soy flour particulates in the polymer matrix, resulting in poor wetting, particle agglomeration, and impaired physical properties [13–15]. Formulation additives (*e.g.*, plasticizers, coupling agents and chemical modifiers) were used to improve the dispersion and interfacial adhesion between the soy particles and the polymer matrix phase, thereby facilitating improvements in properties [13–15]. In a separate study, a two-step single-screw extrusion process was used in combination with solid-state shear pulverization (*i.e.*, three process steps in total) to overcome the shear-related deficiencies associated with conventional single-step melt processing, leading to better dispersion and improved properties without the use of additional formulation additives [16].

Although process and formulation modifications have led to substantial improvements in the properties of PP/soy flour blends [13, 14, 16], no attempts have been made to discern whether the underlying composition of the soy flour itself might also affect the efficiency of blending, matrix phase wetting, agglomeration or physical properties. Given that soy flour and soy meal are inherently comprised of highly polar water-soluble components like carbohydrates [17, 18], it stands to reason that these components might interfere with matrix phase wetting and dispersion, particularly when blended with non-polar polymers like polypropylene. Thus, from a chemical perspective, it makes sense to consider whether the removal of these components might lead to improvements in end-use properties.

From another perspective, the incorporation of wood particulates into a molten polymer is known to increase shear viscosity during conventional melt mixing [1]. For the case of polypropylene (PP), this can lead to shear degradation, which is often mitigated by the incorporation of antioxidants (*e.g.*, hindered phenols) and lubricants like stearic acid (SA) and wax [1]. Although higher shear forces can lead to the degradation of PP and chain scission during melt mixing [1, 3], prior studies [16] have shown that the higher melt viscosities afforded by the incorporation of wood flour could be advantageous for co-blending soy flour or soy meal particulates with PP, particularly

if the higher shear forces can help to overcome the aforementioned problems with dispersion and homogeneity. Thus, provided that the thermal degradative stability of the soy additive is not adversely affected, the higher shear forces afforded by the incorporation of wood particulates could be beneficial when it comes to achieving reduced agglomeration, improved homogeneity, and improved mechanical properties.

Given that amphiphilic plant proteins like those isolated from soy contain polar as well as non-polar amino acid side-chain residues [19], it stands to reason that they might also be able to function as surface compatibilizing agents in polymer/wood composites. For example, amphiphilic natural proteins like those isolated from soybean meal and canola meal have been observed to exhibit the ability to disperse oils in water, leading to moisture-resistant binders for particle board and oriented strand board composites [20]. Likewise, adhesives based on amphiphilic, water-dispersible soy protein isolates have displayed improved moisture resistance in laminated wood composites when the proteins were denatured to expose non-polar hydrophobic moieties [12, 21]. If these types of proteins can wet polar wood surfaces and facilitate the encapsulation and distribution of the wood particles within a non-polar polymer matrix, then at minimum, it might be possible to realize economic benefits by extending the synthetic polymer phase in a polymer–wood composite with natural products like those derived from whole soybean meal. At best, improvements in end-use properties might also be realized.

Soy proteins are comprised of several amino acid residues that coincide with a subset of those that have been independently shown to exhibit antioxidant properties [22], including tryptophan, methionine, histidine, lysine, cysteine, arginine, and tyrosine [23]. Two of these contain aromatic moieties (*e.g.*, tryptophan, and tyrosine), which are also known to exhibit the capacity to absorb UV light [24]. In addition, soy protein contains phenylalanine [23], which is also a strong UV absorbing compound [24]. Given that polymers like PP are prone to photooxidative degradation in the presence of UV light [25], and given that synthetic UV absorbing stabilizers are required to stabilize many types of polymer/wood composites [26], it stands to reason that soy proteins might also have the capacity to impart a photooxidative stabilizing function, which could

render them even more beneficial for use in polymer/wood composites.

With these possibilities in mind, we have undertaken a preliminary investigation of PP/wood composites made both with and without two natural soybean meal additives, one comprising whole soybean meal (WM), and the second comprising a water-insoluble extract from whole soybean meal (IM). In the latter case, we purposely removed the water-soluble components from WM to determine whether the presence or absence of these components would influence mechanical properties, thermal degradative properties and other properties like moisture resistance, especially given that the presence of water-soluble components (*e.g.*, carbohydrates) was previously observed to have a negative impact on the moisture resistance of laminated wood composites [27]. In this initial study, we present preliminary results pertaining to the effects of water-soluble components on surface hydrophobicity, dynamic mechanical properties, crystallinity, and thermal stability. In addition, we also present the UV absorption characteristics of IM and WM. Additional studies on mechanical properties, morphology, rheology, and environmental exposure testing will be reported in future communications.

2. Experimental

2.1. The preparation of WM and IM samples

Whole soy meal (WM) was obtained from CHS, Inc. (Soybean Meal, CAS# 68308-36-1, Inver Grove Heights, MN, USA), and was ground to an average particle size diameter of 45 μm with a surface area of 0.74 m^2/g with the use of a Rotormill 1500 air classifier mill (Prater Industries, Inc., Bolingbrook, IL, USA) operated by the International Process Equipment Company (IPEC, Pennsauken, NJ, USA). A proteinaceous water-insoluble fraction (IM) was isolated from WM with a 15-cycle water extraction–washing protocol followed by oven drying. In the first step, 12 g aliquots of WM powder were placed into individual 80 mL HDPE centrifuge tubes together with 63 g aliquots of distilled water. The contents were then shaken by hand, and the resulting dispersions (16% w/w solids) were placed into a gravity oven at 45 °C for 12 h, where they were allowed to settle under static conditions to assist in the dissolution of water-soluble components [27]. The resulting sediments were then redispersed by shaking and were then centrifuged at 3400 rpm for 60 min at

25 °C. Upon completion of the first centrifuge cycle, the low-haze supernatant solutions were decanted from the washed sediments, and fresh distilled water was added to the 70 mL mark of each tube. The tubes were then shaken by hand to redisperse the sediments, marking the completion of the first extraction–wash cycle. The extraction–wash cycle was then repeated fourteen additional times (15 cycles in total) to yield a viscous paste comprised of 23.5% (w/w) IM solids in water. The resulting IM dispersions were then allowed to dry in open-air for 24 h at 80 °C in a gravity oven. Upon drying, the yield of IM was gravimetrically determined to be approximately 61.7% by weight of the starting WM powder (*i.e.*, WM consisted of ~38.3% water-soluble extractable solids by weight). The dried IM solids were then ground with a mortar and pestle, which yielded a fine powder for subsequent use.

2.2. PP/wood composite materials and preparations

Polymer composite compounding was performed at the University of Akron polymer applications laboratory in Akron, OH (USA) with a Brabender twin blade batch mixer equipped with roller blades and an REE6 PlastiCorder controller (C. W. Brabender Instruments, Inc., a subsidiary of Anton Paar, South Hackensack, NJ, USA). A batch size of 47.55 g was mixed at 30 rpm with a set point temperature of 175 °C, regulated with 3-zone heating and air cooling. Materials were added stepwise over a 16 min mastication cycle. In the first step, a 50/50 w/w blend of PP pellets with melt-flow indices of 4 and 12 were masticated for 6 min, which allowed for melt clarification (polypropylene, melt index: 4g/10 min, CAS 9003-07-0, isotactic, glass transition temperature: $T_g = -26$ °C, crystalline melting temperature: $T_m = 160$ °C, density: 0.9 g/cm³, cat. #130, Scientific Polymer Products, Ontario, New

York, USA; polypropylene melt index: 12 g/10 min, CAS 9003-07-0, isotactic, number average molar mass: $M_n = 67\,000$ amu, mass average molar mass: $M_w = 250\,000$ amu, $T_m = 160$ – 165 °C, density: 0.9 g/cm³, cat. #427888, Sigma Aldrich, Inc., St. Louis, MO, USA). In the second step ($t = 6$ min), one-third of the total maple wood flour was added (maple wood flour 8010, 60/35 w/w mesh sizes 40 and 60, CAS 9004-34-6, moisture content ~4%, American Wood Fibers, Columbia, MD, USA). In the third step ($t = 7.5$ min), a premixed aliquot of additives was added, in accord with the formulation recipes in Table 1. Additives included IM and WM (prepared as noted above), Irganox 1010 (pentaerythritol tetrakis[3,5-di-*tert*-butyl-4-hydroxyhydrocinnamate] 98%, CAS 6683-19-8, $T_m = 115$ – 118 °C, cat. #441783 Sigma Aldrich, Inc., St. Louis, MO, USA), paraffin wax (CAS 8002-74-2, $T_m = 54$ – 57 °C, cat. #W1018, Spectrum Chemical, New Brunswick, NJ, USA), and stearic acid (CAS 57-11-4, $T_m = 67$ – 72 °C, 95%, cat. #W303518, Sigma Aldrich, Inc., St. Louis, MO, USA). In the fourth step ($t = 8.5$ min) a second third of the total wood flour aliquot was added. In the fifth step ($t = 9.5$ min), the final third of the total wood flour aliquot was added, completing all raw material additions. The mix was allowed to masticate until the end of the mix cycle at $t = 16$ min. Note that between $t = 11$ and $t = 16$ min, the mixes were jogged with reverse rotation (twice) to assist in removing and mixing dead-spot material near the front and rear plates of the mixer. The material was discharged at the end of the cycle and was allowed to cool under static ambient conditions so that all formulations had the same thermal history.

2.3. Fourier transform infrared spectroscopy (FTIR) studies

Solid-state surface attenuated total reflectance Fourier transform infrared spectroscopy (SATR-FTIR)

Table 1. PP/wood composite formulations (weight percent). The weight ratio of stearic acid to wax (SA/wax) was maintained at a level of 3/1 w/w, consistent with a mixture that was previously determined to be ideal for internal and external lubrication of similar formulations [1, 3].

Additive type	Sample ID	PP [%]	Maple wood flour [%]	Irganox 1010 [%]	WM [%]	IM [%]	SA [%]	Wax [%]	Total [%]
IM/SA/Wax	4	60.99	33.65	0.32	0.00	2.52	1.89	0.63	100.00
WM/SA/Wax	6	60.99	33.65	0.32	2.52	0.00	1.89	0.63	100.00
IM	5	60.99	33.65	0.32	0.00	5.04	0.00	0.00	100.00
WM	10	60.99	33.65	0.32	5.04	0.00	0.00	0.00	100.00
SA/wax	14	60.99	33.65	0.32	0.00	0.00	3.78	1.26	100.00
No additives	15	64.24	35.44	0.32	0.00	0.00	0.00	0.00	100.00

studies were performed with a Bruker Alpha-P spectrometer (Bruker Instruments, Billerica, MA, USA) equipped with a diamond-cell attenuated total reflectance (ATR) accessory. Anvil pressure was applied to ensure intimate contact between the samples and the diamond crystal. Single-bounce spectra, referenced against atmospheric background spectra, were obtained under ambient conditions by averaging the signal of 24 scans at 4 cm^{-1} resolution over the spectral range of 4000 to 650 cm^{-1} .

2.4. Thermogravimetric analysis

Thermogravimetric analysis (TGA) was performed on samples weighing approximately 18–20 mg, in platinum pans at a heating rate of $10\text{ }^{\circ}\text{C}/\text{min}$ from 25 to $600\text{ }^{\circ}\text{C}$ under a dry air purge in a TA Instruments Model 550 TGA (TA Instruments, New Castle, DE, USA) at the University of Akron polymer applications laboratory in Akron, OH (USA).

2.5. Contact angle measurements

Compounded PP/wood composite formulations were individually pressed between two Teflon-lined aluminum plates at $180\text{ }^{\circ}\text{C}$ for 4 min under a pressure of 1055 kPa in a hot press (Carver Auto Series 13.6 Metric-Ton Laboratory Press, Carver, Inc., Wabash, IN, USA), tapering to zero pressure as regulated by aluminum spacers to a final thickness of 0.3 mm. The hot assemblies were then removed from the press and were allowed to cool to ambient temperature before the aluminum plates were removed. Water contact angle (CA) measurements were performed with a goniometer Model PG1 from Gardco (Paul N. Gardner Company, Columbia, MD, USA). CA was measured at $25\text{ }^{\circ}\text{C}$ with distilled water. Initial CA measurements ($t = 0\text{ min}$) were made within 5 s of releasing a water drop onto the horizontal surface of each sample. Additional CA measurements were then performed at time increments of 2, 3, 5, and 10 min. Five replicates of each type of sample were individually measured. Analysis of variance (ANOVA) was performed on the six groups of samples at each of the independent CA time increments. We used Fisher's least significant difference (LSD) comparisons to evaluate population differences at the 95% confidence level ($p = 0.05$). Statistical analyses were performed with KaleidaGraph data analysis software (Synergy Software, Reading, PA, USA).

2.6. Ultraviolet spectroscopy (UV)

Ultraviolet reflection-absorption measurements were performed on IM and WM solid powders with a Shimadzu UV3600i UV-VIS spectrometer equipped with an ISR-603 integrating sphere attachment (Shimadzu Scientific Instruments, Inc., Columbia, MD, USA) at the Duke University shared materials instrumentation facility in Durham, NC (USA). For comparative purposes, additional analyses were also performed on a rutile titanium dioxide pigment powder (Ti-Pure R-902+, CAS 13463-67-7, Chemours, Wilmington, DE, USA). Samples were measured in total diffuse reflectance mode [28] with a specular reflectance attachment and a barium sulfate powder as a relative standard (barium sulfate white analytical standard, CAS 7727-43-7, cat. #101748, Sigma Aldrich, Inc., St. Louis, MO, USA). Samples were analyzed over a wavelength range of 220–410 nm with a slit width of 5 nm and a scan rate set to medium.

2.7. Dynamic mechanical analysis (DMA)

Samples of the PP/wood composites were cut to dimensions of 9.0 to 10.5 mm in width, 17.5 mm in length, and 3.0 to 4.0 mm in thickness. The samples were analyzed with a TA Instruments Discovery 850 dynamic mechanical analyzer (TA Instruments, New Castle, DE, USA) at the University of Maine advanced structures and composites center in Orono, Maine (USA). Samples were run in single cantilever mode with an initial preload force of 0.1 N at a fixed frequency of 1 Hz from -80 to $170\text{ }^{\circ}\text{C}$ at a heating rate of $5\text{ }^{\circ}\text{C}/\text{min}$, with an oscillation strain amplitude of 0.01%. Note that prior to each run, the samples were cooled from 25 to $-80\text{ }^{\circ}\text{C}$ at $20\text{ }^{\circ}\text{C}/\text{min}$, and were allowed to equilibrate for 5 min.

2.8. Differential scanning calorimetry (DSC)

DSC samples were tested with a TA Instruments Model 2500 DSC (TA Instruments, New Castle, DE, USA) at the University of Maine advanced structures and composites center in Orono, Maine (USA). Samples weighing approximately 8–10 mg were placed in aluminum pans with lids containing pinholes for degassing. The samples were pre-cooled from 30 to $-80\text{ }^{\circ}\text{C}$ at a cooling rate of $-20\text{ }^{\circ}\text{C}/\text{min}$ under a dry nitrogen purge and then were run from -80 to $180\text{ }^{\circ}\text{C}$ at a heating rate of $10\text{ }^{\circ}\text{C}/\text{min}$.

3. Results and discussion

3.1. Contact angle results

CA results are presented in box plots (Figure 1). The area within each box encompasses 50% of the CA data from each population set ($n = 5$), where the span between the top and bottom of each box represents a distribution of CA values equivalent to $\pm 25\%$ of the mean for each set. The bar lines extending from the top and bottom of each box denote the minimum and maximum values within each population set. Qualitatively, the trends indicate that CA decreased with time in each case, where the most hydrophobic composite was the one made with IM in combination with SA and wax (sample #4). Fisher's LSD results (Table 2) indicate that the composite made with IM in combination with SA/wax (sample #4) was significantly more hydrophobic than all other composites at the 95% confidence level ($p = 0.05$). Indeed, the removal of the water-soluble fraction led to a statistically significant improvement in hydrophobicity, but only when the resulting IM fraction was blended with SA/wax (*i.e.*, sample #6 vs. sample #4 as shown in Figure 1 and as compared in Table 2). In

the absence of SA/wax, the removal of the water-soluble fraction from WM (*i.e.*, to yield IM) did not significantly improve hydrophobicity (*i.e.*, sample #10 vs. sample #5 as shown in Figure 1). These results indicate that the removal of the water-soluble components from WM resulted in improved hydrophobicity, as first hypothesized [27], but only when the resulting IM additive was used in combination with SA/wax.

3.2. FTIR results

SATR-FTIR scans of the WM and IM powders are provided in Figure 2 and Figure 3. The water extraction process led to the isolation of a protein-rich component (IM) as indicated by the higher relative absorbance intensity of the carbonyl amide stretching band at 1630 cm^{-1} with respect to the common C–O stretching band centered at 1040 cm^{-1} [17, 27]. The water extraction process also resulted in the removal of carbohydrate components associated with the C–O stretching absorption band at $\sim 990\text{ cm}^{-1}$ [17, 27]. This band was previously linked to components comprising a moisture-sensitive boundary layer that became the locus of water-induced delamination

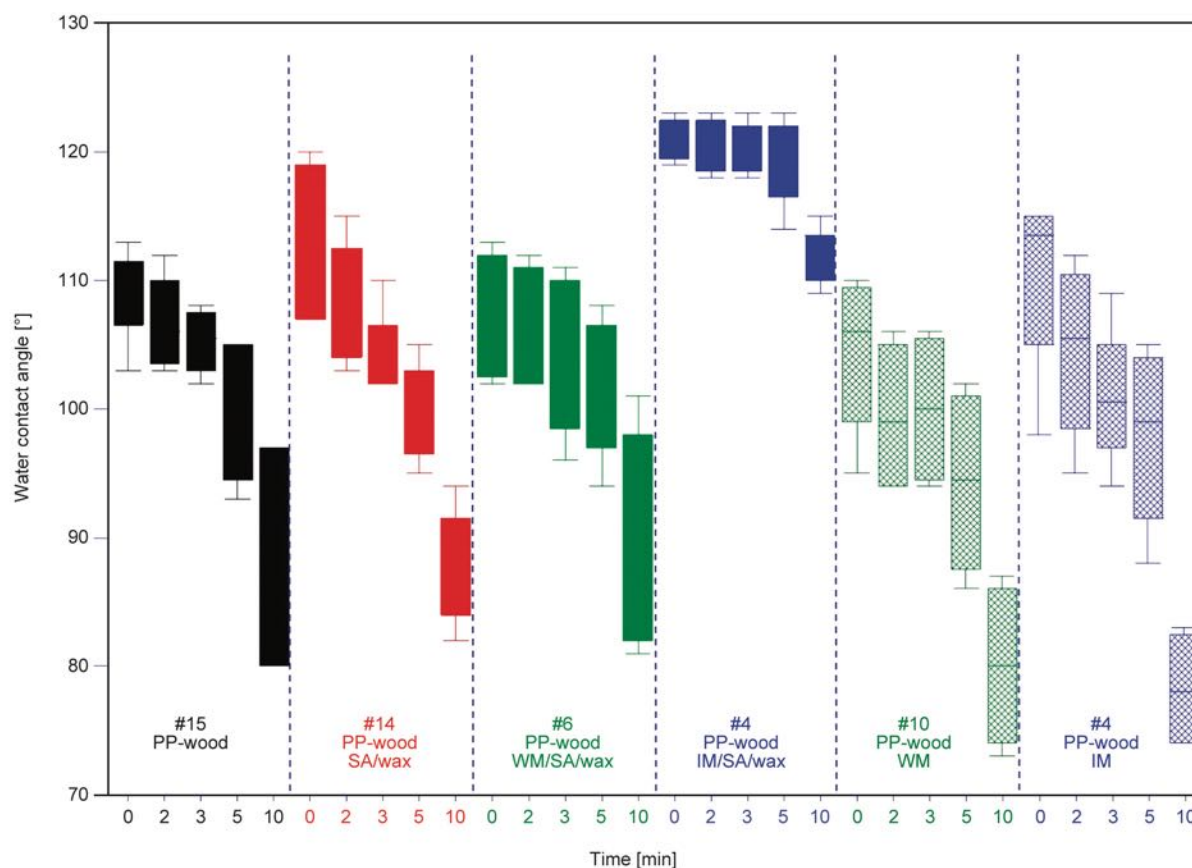


Figure 1. Water contact angle results for PP/wood composites as a function of contact time at 25°C . Box plots show distributions for $n = 5$ replicates at each CA time increment.

Table 2. One-way ANOVA was conducted with the use of water CA data from each type of PP/wood composite at each CA time increment (*i.e.*, ANOVA was conducted on six groups of CA data at each of the five independent time increments). We performed Fisher's least significant difference (LSD) comparisons to compare groups for differences at the 95% confidence level ($p = 0.05$). The p -values were reported for the comparisons where the differences in the means satisfied the criterion of p being less than or equal to 0.05. For cases where $p > 0.05$, the differences were reported as not significant (NS). Note that the comparisons are provided only for cases where the population differences were deemed significant at one or more of the CA time increments. For example, the comparison of samples #10 and #5 were determined to be NS at each of the CA time increments and were therefore not reported.

PP/wood composite paired comparisons	$t = 0$ min	$t = 2$ min	$t = 3$ min	$t = 5$ min	$t = 10$ min
Sample #4 vs. sample #15	0.01	0.002	0.0006	0.0003	0.0002
Sample #4 vs. sample #14	NS	0.005	0.0003	0.0003	0.0001
Sample #4 vs. sample #6	0.004	0.002	0.0003	0.0009	0.0003
Sample #4 vs. sample #10	0.0009	0.0001	0.0001	0.0001	0.0001
Sample #4 vs. sample #5	0.02	0.0006	0.0001	0.0001	0.0001
Sample #6 vs. sample #5	NS	NS	NS	NS	0.02
Sample #6 vs. sample #10	NS	NS	NS	NS	0.05
Sample #15 vs. sample #5	NS	NS	NS	NS	0.05
Sample #14 vs. sample #10	0.05	0.03	NS	NS	NS

failures in wood laminates made with soy-based adhesives [27].

One of the hypotheses that was tested in the present study was based on the premise that the removal of water-soluble components from WM to yield the protein-rich IM fraction might lead to improved moisture resistance in polymer/wood composites, analogous to improvements that have been observed in other types of wood composites [20]. Indeed, as discussed in the prior section, this hypothesis appears

to be true, but only when IM was used in combination with SA/wax. To shed more light on the underlying reasons for this unexpected finding, we performed SATR measurements on the surfaces of the PP/wood composites for the purpose of probing for near-surface compositional differences that might correlate with the CA results. As Figure 4 shows, the relative level of stearic acid near the surface (as determined by comparing the absorbance intensity ratio of the stearic acid carbonyl [29] at ~ 1702 cm^{-1} to the

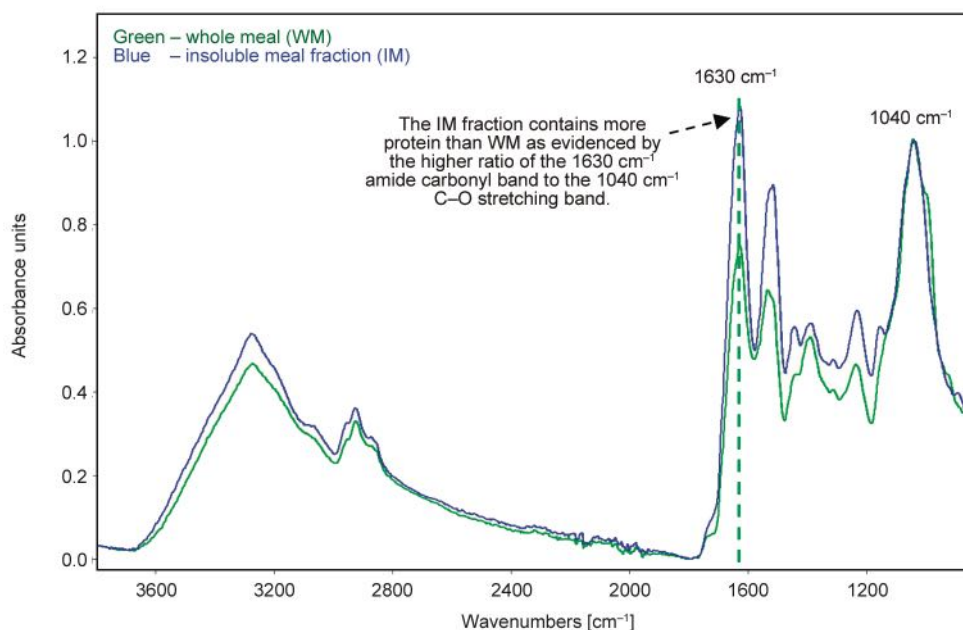


Figure 2. SATR-FTIR absorption spectra over the spectral range 830 to 3800 cm^{-1} for WM and IM, illustrating the higher relative intensity of the IM carbonyl amide stretching band at 1630 cm^{-1} with respect to the common C–O stretching band centered at 1040 cm^{-1} . Spectra were normalized to the common band at 1040 cm^{-1} and were vertically maximized to a common vertical scale.

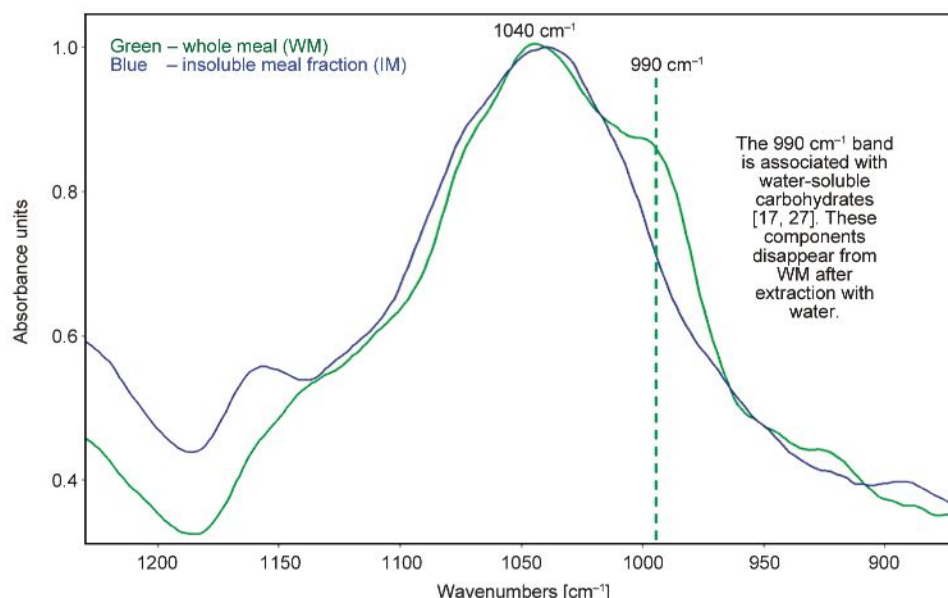


Figure 3. SATR-FTIR absorption spectra over the spectral range 870 to 1230 cm^{-1} for WM and IM, illustrating the differences in the C–O stretch region associated with the presence of water-soluble carbohydrates in WM, and their absence in the IM fraction. Spectra were normalized to the common band at 1040 cm^{-1} and were vertically maximized to a common vertical scale.

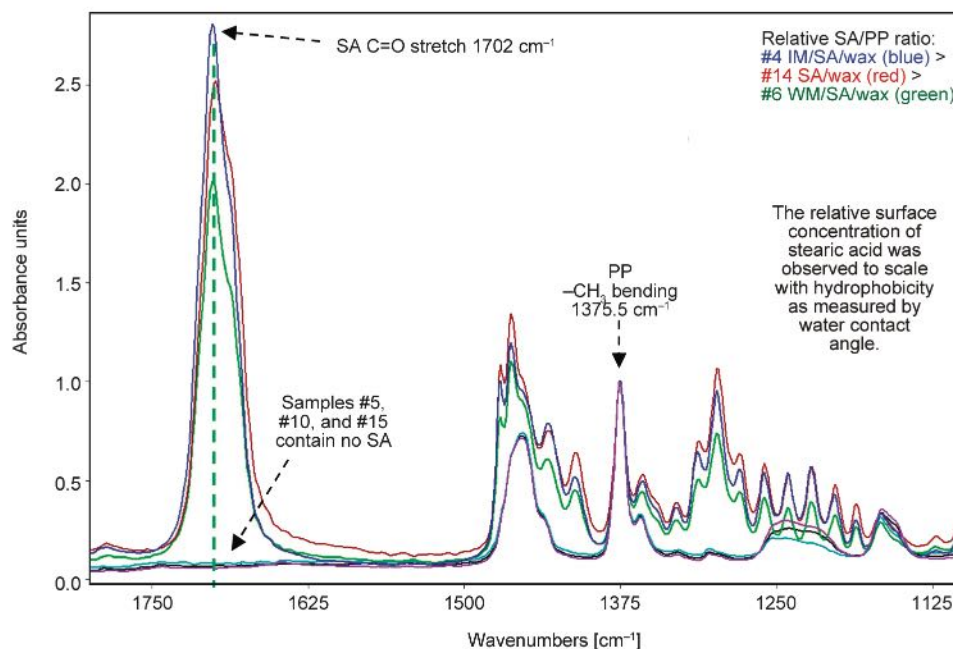


Figure 4. SATR-FTIR spectral overlay of PP/wood composite samples over the spectral range 1100 to 1800 cm^{-1} , showing the higher relative absorbance intensity of the carbonyl stretching band for stearic acid at 1702 cm^{-1} on the surface of sample #4 (IM/SA/wax, blue), and the lower concentration on the surface of sample #6 (WM/SA/wax, green). Spectra were normalized to the polypropylene $-\text{CH}_3$ bending band at 1375.5 cm^{-1} and were vertically maximized to a common scale.

PP methyl bending band [30] at $\sim 1375.5 \text{ cm}^{-1}$), was the highest in sample #4 (IM/SA/wax), and the lowest in sample #6 (WM/SA/wax). Moreover, the relative surface concentration of SA in sample #4 was higher than that observed in the control sample #14, even though the control contained twice the level of

SA (*i.e.*, 3.78% in sample #14 vs. 1.89% by weight in sample #4).

From a phenomenological perspective, these results correlate with the surface hydrophobicity trends, suggesting that the higher concentration of stearic acid at the air/composite interface is likely responsible for

the enhanced hydrophobicity of sample #4. From a compatibility perspective, the results also indicate that the phase separation of SA from the bulk composite matrix was favored by the presence of IM. By contrast, the degree of SA phase separation was reduced in the presence of WM, suggesting that the water-soluble components within soy meal (*e.g.*, carbohydrates) facilitate the compatibilization of SA within the composite matrix. In addition, given that WM consists of ~61.7% by weight of the IM component (*i.e.*, in combination with ~38.3% by weight water-soluble extractable solids), the lower degree of SA phase separation in the composite made with WM/SA/wax could also be related to the lower effective level of insoluble proteins within composites made with WM when compared to the level of insoluble proteins in composites made with neat IM/SA/wax.

A possible explanation for the higher degree of SA phase separation in the presence of IM could be competitive wood particle surface adsorption, whereby SA and denatured soy proteins compete for wood particle surface sites, analogous to the competition that has been hypothesized to occur between SA and MAPP in similar PP/wood composites [1]. It is plausible that like MAPP, soy proteins might facilitate wetting between wood particulates and PP, where carboxylic and/or amine groups from denatured proteins become adsorbed onto the hydroxyl-rich surfaces of cellulosic wood particulates, and where the non-polar/lipophilic protein moieties from surface-tethered peptide chains become selectively wetted by the non-polar PP phase. This type of amphiphilic wetting between soy proteins and wood has been hypothesized to occur in other types of cellulosic composites [19, 21].

Given that SA carboxylate groups can also become attached to the surfaces of wood particulates [1] and given that the analogous moieties of soy proteins have been observed to do the same [19, 21], it stands to reason that SA might compete with protein adsorption. Depending on the IM protein's relative affinity for wood surface sites, the competition between SA and denatured protein could lead to mixed surface adsorption, which would alter the balance between the relative fractions of free and surface-adsorbed species of both types. Consequently, under competitive conditions, the fraction of desorbed SA could be higher in composites made with SA/protein mixtures than in those made with neat SA alone.

Alternatively, it is also possible that IM displaces and occludes free SA from the amorphous domains of the bulk PP matrix phase, resulting in the migration of excess SA to the composite surface (*i.e.*, to the air interface). The viability of this hypothesis would depend on whether IM exhibits some degree of bulk compatibility with PP. However, as will be discussed below, the DMA results do not support the PP compatibility hypothesis, leaving the competitive surface adsorption hypothesis as the more viable alternative explanation.

3.3. TGA results

Figure 5 provides an overlay of TGA weight loss curves for neat IM and WM. The onset temperatures for the degradation of IM and WM were approximately 240 and 210 °C, respectively, indicating that IM has better thermal stability than WM. Note that the onset of the degradation of IM was in close agreement with the reported onset value for other soy protein isolates [31]. The data also indicate that the water-soluble components in WM (*e.g.*, carbohydrates) [27] have a deleterious effect on thermal stability.

The thermograms in Figure 5 also show a weight loss of ~7% at 100 °C for both IM and WM, attributable to the volatilization of entrapped moisture [31], which indicates that both IM and WM contained similar moisture contents prior to compounding. This is important because soy proteins with a

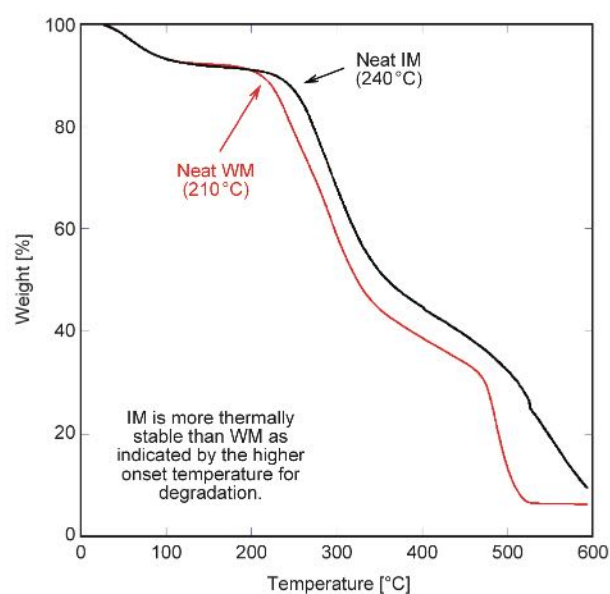


Figure 5. TGA thermograms of neat WM and IM powders. The thermo-oxidative stability of IM exceeds that of WM.

moisture content of 7% are reported to have a denaturing temperature of approximately 160–170 °C [31], which is lower than the compounding temperature that was used for the preparation of the PP/wood composites (*i.e.*, 175 °C). Thus, it is likely that the IM proteins became denatured during processing.

Figure 6 provides an overlay of the TGA weight loss curves for the PP/wood composites, illustrating the % weight loss from 25 °C to the onset of the temperature of PP degradation (280 °C, as reported for PP in similar wood composites) [3]. Note that the degree of weight loss at 280 was observed to qualitatively scale with the levels of IM and SA/wax in the composites.

The composite made with 5% SA/wax (sample #14) exhibited the highest degree of weight loss below 280 °C, most likely due to the combined volatilization of SA and wax components, while the sample made with 5% neat IM (sample #5) exhibited the lowest degree of weight loss. The percent weight loss at 280 °C was ranked from highest to lowest as follows: 5% w/w SA/wax (sample #14) > 2.5% w/w SA/wax with 2.5% w/w WM (sample #6) > 2.5% w/w SA/wax with 2.5% w/w IM (sample #4) > 5% WM (sample #10) = no additives (sample #15) > 5% w/w IM (sample #5). Note that this ranking was also in agreement with the ranking of the temperatures at which the samples exhibited 5% weight loss (see Table 3).

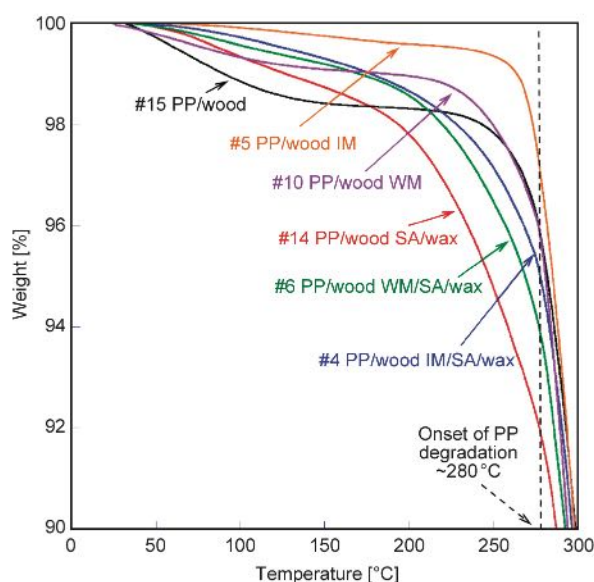


Figure 6. Overlay of TGA weight loss thermograms for PP/wood composites, illustrating the % weight loss from 25 °C to the onset of the degradation of PP (280 °C as reported for PP in similar PP/wood composites) [3].

Each composite also exhibited a small but measurable degree of weight loss below 100 °C (*i.e.*, <1.5%), most likely due to the volatilization of water that was entrapped within the wood particulates [3] or within the meal additives. The extent of this weight loss was highest for the composite made without additives (sample #15) and was lowest for the composite made with neat IM (sample #5). Composites containing neat WM, neat SA/wax, and mixtures of SA/wax with IM or WM exhibited intermediate degrees of weight loss. The degree of weight loss at 100 °C was ranked from highest to lowest as follows: no additives (sample #15) > 5% w/w SA/wax (sample #14) = 5% WM (sample #10) > 2.5% w/w SA/wax with 2.5% w/w WM (sample #6) > 2.5% w/w SA/wax with 2.5% w/w IM (sample #4) > 5% w/w IM (sample #5).

Given that the composites were masticated and processed at 175 °C, it is likely that most of the wood and protein-entrapped moisture was removed during processing. Thus, it follows that the weight loss trend at 100 °C might instead be associated with the susceptibility of the composites to atmospheric moisture ingress during storage. Given that SA has been hypothesized to become surface adsorbed onto wood particulates [1], it follows that SA might serve as a hydrophobic protective barrier to help mitigate atmospheric moisture ingress into the hygroscopic wood particulates. Also, given that WM is more hygroscopic than IM due to the presence of water-soluble carbohydrates [20, 27], it follows that the absence of carbohydrates in IM could also lead to a slower rate of moisture ingress. Finally, given that the presence of IM was observed to produce a lower degree of weight loss up to the onset of PP degradation, it follows that heat-denatured IM proteins might have become chemisorbed onto the surfaces of wood particulates [19] as hypothesized earlier, leading to a hydrophobic protective barrier analogous to that provided by SA [1], and possibly leading to improved matrix-phase wetting, analogous to that provided by MAPP in similar PP/wood composites [8, 9]. Experiments are underway to test these hypotheses, including studies of water swelling, moisture ingress via gravimetric tests and nuclear magnetic resonance (NMR) imaging, as well as studies of morphology and matrix-phase coupling via solid-state NMR techniques [27], scanning electron microscopy techniques [1], and mechanical techniques [1, 32]. Referring to the thermal degradation of other components at temperatures greater than 280 °C, TGA

studies of similar PP/wood composites [3] have led to the assignment of first-derivative peak temperatures (*i.e.*, from the first derivatives of weight loss *vs.* temperature curves) to the thermal degradation processes associated with specific components, including cellulose, PP and lignin [3]. We also observed derivative peaks associated with each of these components (Figure 7 and Figure 8). The derivative peak temperatures (*i.e.*, T at $(dW/dT)_{\max}$) associated with PP and lignin are provided in Table 3, along with the temperatures associated with 5% weight loss from each composite, as mentioned previously.

Regarding the cellulose components, we did not detect discernable differences in dW/dT peak temperatures, except for the composite made with neat IM, which exhibited a slightly lower peak temperature for cellulose degradation than the other composites. By contrast, however, we found discernable differ-

ences for the PP and lignin components. The peak temperatures for PP degradation spanned a range of 363–378 °C, which was within the range observed in prior TGA studies of similar composites, where the variation in peak temperatures was attributed to the degree of PP shear degradation during processing [3]. The maximum rate of PP degradation shifted to higher temperatures in composites made with SA/wax and was highest for the composite made with SA/wax alone. Previous studies of PP/wood composites revealed that the maximum degradation rate for PP was correlated with melt viscosity during mixing [3], where melt viscosity was affected by the level of SA/wax, as well as by the ratio of SA to wax. Further, it was hypothesized that shear degradation from higher melt viscosity during mixing produced PP chain defect sites, which in turn led to lower activation barriers for PP chain scission, and to lower peak derivative temperatures [3].

Table 3. TGA data from PP/wood composites, including the temperature at 5% weight loss, and the temperature at the peak rate of weight loss (*i.e.*, T at $(dW/dT)_{\max}$) associated with the degradation of the PP and lignin components [3].

Sample ID	Meal and SA/wax additives (~5% by wt)	Temperature at 5% weight loss [°C]	dW/dT peak temperature for PP [°C]	dW/dT peak temperature for lignin [°C]
15	0	283	373	459
14	SA/wax	249	378	459
4	IM/SA/wax	277	373	467
6	WM/SA/wax	269	371	450
5	IM	285	363	460
10	WM	281	365	445

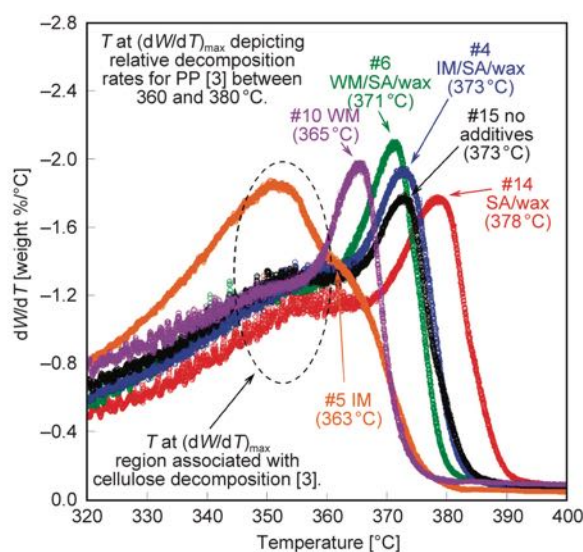


Figure 7. Derivative of TGA weight loss curves (dW/dT) depicting the peak temperatures associated with the maximum rate of weight loss (*i.e.*, T at $(dW/dT)_{\max}$) for the PP and cellulose components [3] in PP/wood composites.

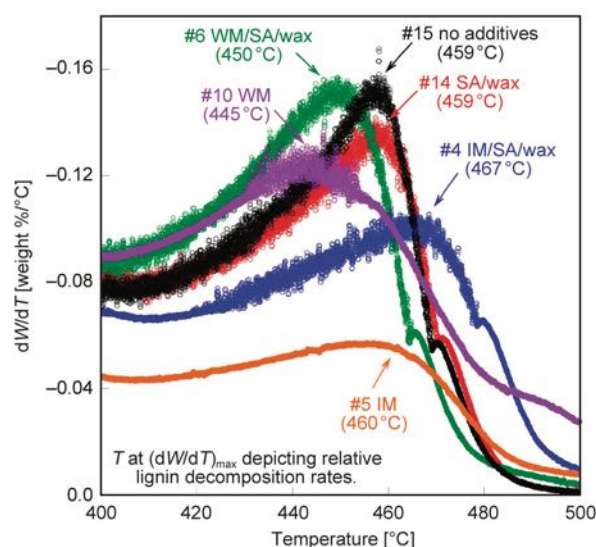


Figure 8. Derivative of TGA weight loss curves (dW/dT) depicting the peak temperatures associated with the maximum rate of weight loss (*i.e.*, T at $(dW/dT)_{\max}$) for lignin [3] in PP/wood composites.

The dW/dT trends from the present study suggest that partial or complete replacement of SA/wax with meal additives may have led to a higher degree of shear degradation of PP during mixing, perhaps due to an increase in melt viscosity [1, 3]. This hypothesis is currently under investigation. However, it nevertheless remains apparent that the effect was minimal for the IM/SA/wax blend (sample #4), which exhibited a T at $(dW/dT)_{\max}$ for PP that was similar to that of the composite made without additives (sample #15), and was closest to that of the composite made with SA/wax alone (sample #14).

The composite made with IM/SA/wax also facilitated slightly better thermal oxidative stability of PP than the composite made with WM/SA/wax. It is likely that this improved stability is partially attributable to the absence of water-soluble carbohydrate components, as supported by the thermograms for the neat WM and IM powders (Figure 5).

The IM meal additive also increased the peak temperature associated with the maximum thermo-oxidative decomposition rate of the lignin component. The maximum rate of lignin degradation decreased and shifted to higher temperatures in composites made with IM, independently of the presence of SA/wax. This result is suggestive of a chemical interaction between the two components. Indeed, chemical interactions are plausible given that lignin and soy proteins both contain aromatic free-radical stabilizing entities, including polyphenolic compounds as found in lignin [33], and aromatic side-chain residues as found

in soy proteins [23, 34]. The thermo-oxidative degradation of lignin has been observed to produce volatile degradation products (*e.g.*, substituted phenols) as well as non-volatile char-like structures [35]. It is plausible that aromatic side chain moieties like those found in soy protein could react with volatile lignin components to produce higher yields of non-volatile char-like structures, consistent with the reduced volatility and slower lignin degradation rates as measured by TGA in the composites made with IM proteins.

3.4. UV spectroscopy results

The % total reflectance curves for the IM, WM, and TiO_2 solid powders, relative to a non-absorbing barium sulfate standard, are provided in Figures 9 and 10, with each depicting the % reflectance over the ranges spanning 220–410, and 220–320 nm, respectively. The % total reflectance for solid powders encompasses both specular and diffuse components of the reflected incident light, where a lower % reflectance is indicative of higher UV absorption [28].

The results indicate that neat IM absorbs more UV light than WM over the entire UV range. This observation is consistent with the lower concentration of IM proteins within WM when compared to neat IM (*i.e.*, WM is comprised of 61.7% IM, diluted by 38.3% water-soluble components by weight). The similar shapes of the WM and IM absorbance curves indicate that the IM proteins are the chromophores

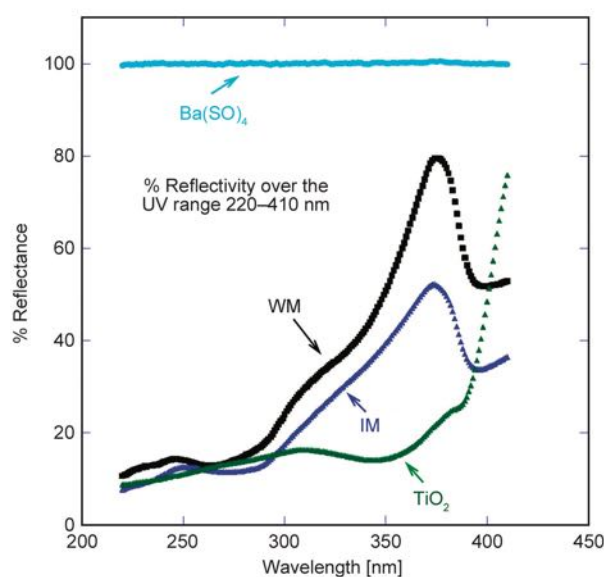


Figure 9. % total reflectance curves for IM, WM, and TiO_2 solid powders, relative to a barium sulfate standard over the ranges spanning 220–410 nm.

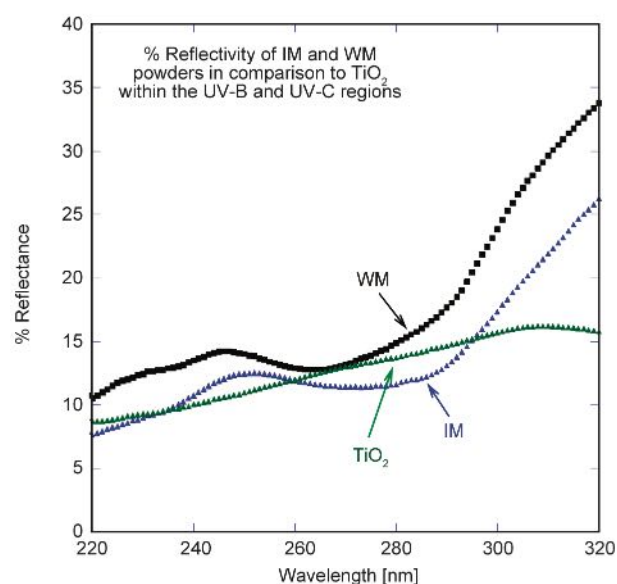


Figure 10. % total reflectance curves for IM, WM, and TiO_2 solid powders, relative to a barium sulfate standard over the range 220–320 nm.

that are primarily responsible for UV absorption, and that the soluble carbohydrate fraction in WM [17, 27] contributes little to the UV absorbance characteristics.

The IM proteins exhibited strong absorbance, particularly below 300 nm, coincident with the absorbance regions associated with tryptophan, tyrosine, and phenylalanine side chains residues [24, 36, 37], all of which are known to be components of soy proteins [23]. It is interesting to note that the absorbance characteristics below 300 nm are comparable to rutile TiO₂ powder, which is known to be a strong UV-absorbing pigment that provides UV stabilization to many types of coatings and polymers [38], including polypropylene [39]. This similarity suggests that the IM powder might also have the potential to function as a UV-stabilizing additive for polymers like PP, particularly since PP is susceptible to UV degradation in the UV-B (290–320 nm) and UV-C (200–290 nm) ranges [39], both of which happen to overlap with strong IM absorption. Although this possibility remains to be studied, it is also worth noting that an analogous UV-protection scheme has been previously proposed by others, whereby neat soy protein isolate (SPI) films have been studied for use as UV-protective coatings [40]. However, no one to our knowledge has successfully used soy protein powders as polymer additives for this purpose.

3.5. DMA results

Fixed frequency storage modulus data (E') are provided in Figure 11. We observed that dilution with 5% w/w neat WM (sample #10) led to a reduction in the storage modulus across the entire temperature range when compared to the composite made without additives (*i.e.*, sample #15), and when compared to the composite made with neat IM (sample #5). This result indicates that the water-soluble components in WM had a deleterious effect on mechanical properties. By contrast, when the PP/wood composite was diluted with 5% w/w neat IM (*i.e.*, in the absence of water-soluble components), the effects on modulus were minimal, despite the lower degree of crystallinity as measured by DSC (to be discussed below), and despite the lower levels of wood and PP in the composite (see Table 1). Note that the incorporation of SA/wax also led to a reduction in storage modulus across the entire temperature range, scaling with the percentage of SA/wax in the formulation.

Fixed frequency loss modulus curves (E'') are provided in Figure 12. The loss modulus maxima were labeled to indicate the temperatures for three types of transitions, including: the onset of the PP melt transition (*i.e.*, ~147 to 150 °C, which is consistent with DSC results); an α -transition [32], which has been previously associated with the presence of a constrained amorphous phase within PP [41]; and a

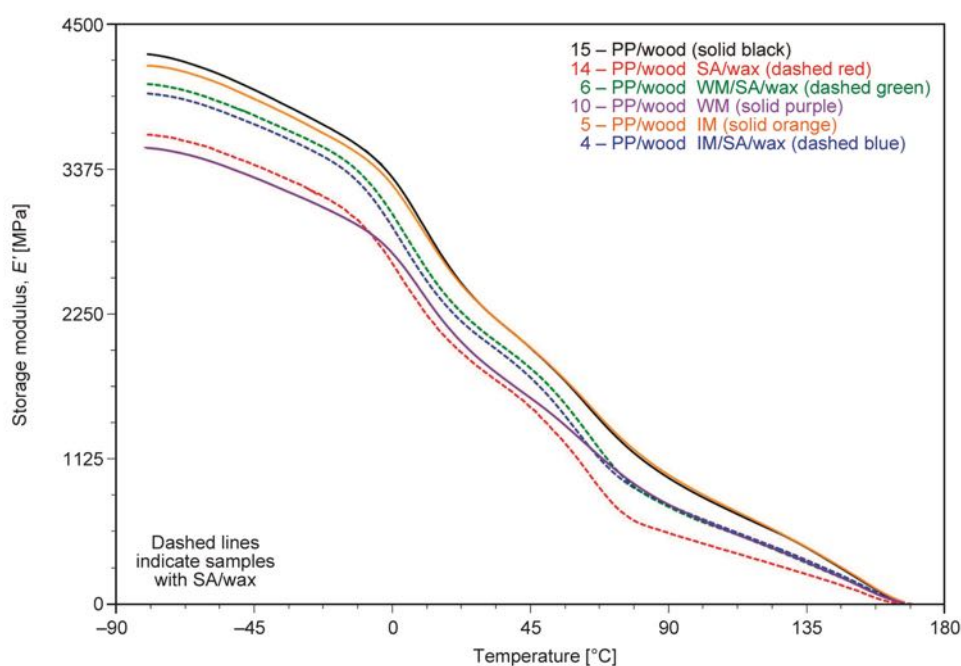


Figure 11. Overlay of DMA storage modulus (E') curves at 1 Hz for PP/wood composites, illustrating the effects of IM and WM additives on modulus in the presence and absence of SA/wax.

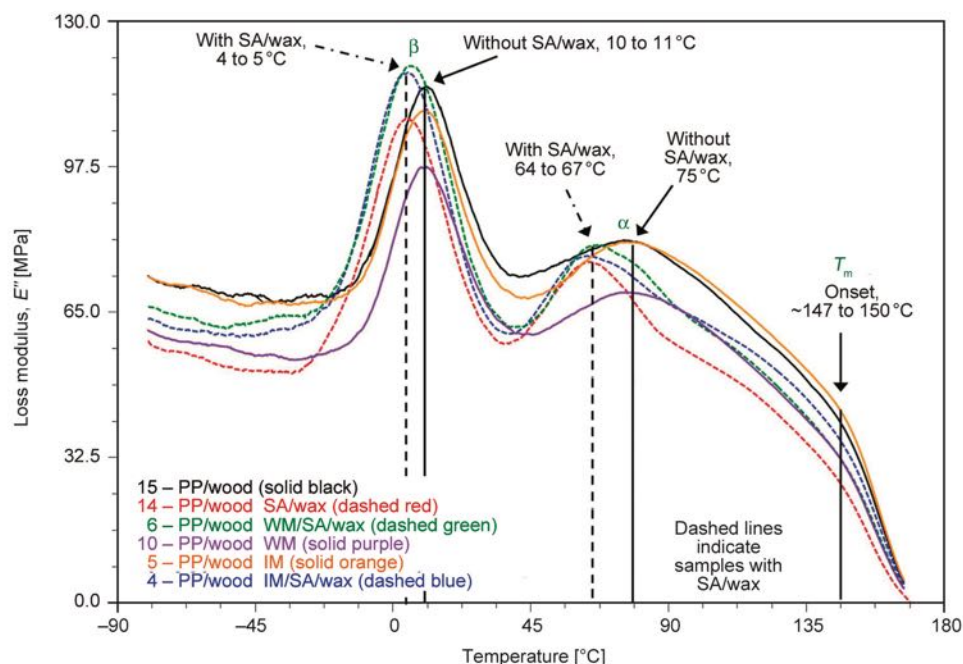


Figure 12. Overlay of DMA loss modulus (E'') curves at 1 Hz for PP/wood composites, illustrating the effects of IM and WM additives on transition behavior as measured by the temperatures associated with loss modulus maxima for PP in the presence and absence of SA/wax.

β -transition [32], which is consistent with the previously assigned glass transition temperature (T_g) for the free amorphous phase [41]. The temperatures associated with the α - and β -transitions were unaffected by the presence of the neat meal additives. However, the presence of SA/wax led to a reduction in α - and β -transition temperatures, which is consistent with amorphous phase plasticization [42]. Specifically, the addition of SA/wax led to approximately a 6 °C reduction in the β -transition temperature, independent of the presence or absence of meal additives. Although the α -transition temperature was similarly reduced by approximately –10 °C, the dilution of SA/wax with meal additives led to the retention of a higher temperature shoulder at 75 °C, which overlapped with the position of the α -transition temperature in the composites that were made without SA/wax.

Tan δ curves (E''/E') for the composites are provided in Figures 13 and 14. Studies of similar PP composites have shown that lower tan δ values are associated with less viscoelastic energy loss, and with improved wetting/coupling between the dispersed and matrix phase components [32, 41, 43]. As Figure 13 shows, tan δ increased with the addition of SA/wax, indicating that SA/wax interferes with wetting/coupling [32, 41, 43, 44].

By contrast, the composites made without SA/wax exhibited lower tan δ values than those made with SA/wax. More specifically, as Figure 14 shows, the composite made with neat IM (sample #5) exhibited less viscoelastic loss than the composite made with neat WM (sample #10), indicating that the water-soluble components in WM interfere with wood to matrix phase wetting/coupling [32, 41, 43]. The composite made with neat IM also exhibited slightly less viscoelastic loss than the composite made without meal additives (sample #15). Thus, at minimum, we can say that IM does not interfere with surface wetting interactions between the wood particulates and the PP matrix phase; and at best, it remains possible that IM has a positive impact on PP/wood wetting interactions. Additional studies are underway to test this hypothesis, including measurements of stress and strain at failure, scanning electron microscopy (SEM) analyses of pre-strained and post-strained failure sites, measurements of motional dynamics via solid-state nuclear magnetic resonance (NMR) techniques, and surface FTIR microscope analysis of the resulting failure sites.

3.6. DSC results

Figure 15 shows the PP melting transitions for the various composites. Note that the reported heats of

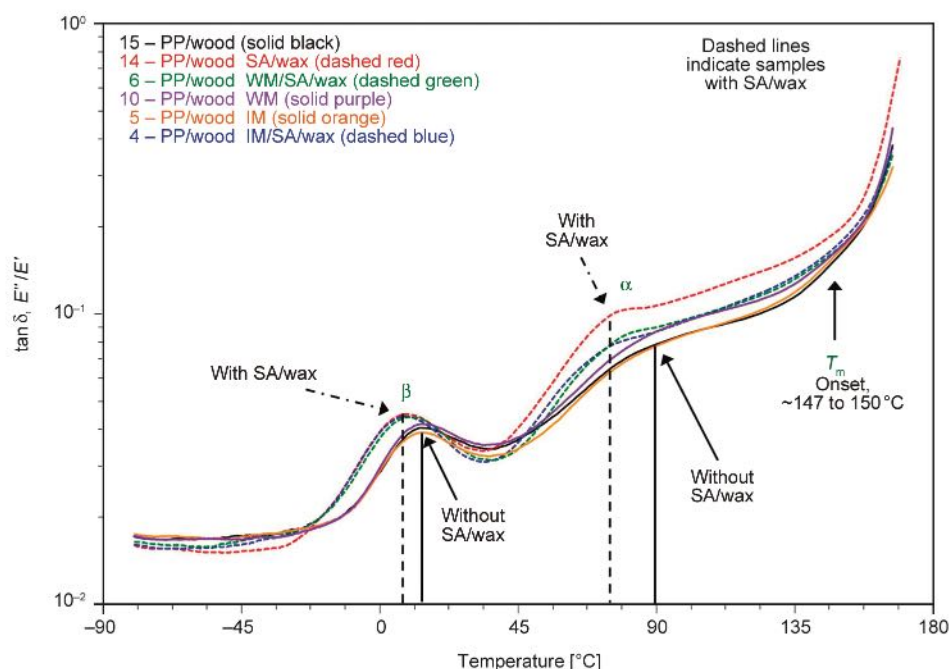


Figure 13. Overlay of DMA $\tan \delta$ (E''/E') curves at 1 Hz for PP/wood composites, illustrating the effects of IM and WM additives on the damping behavior (viscoelastic loss) in the presence and absence of SA/wax.

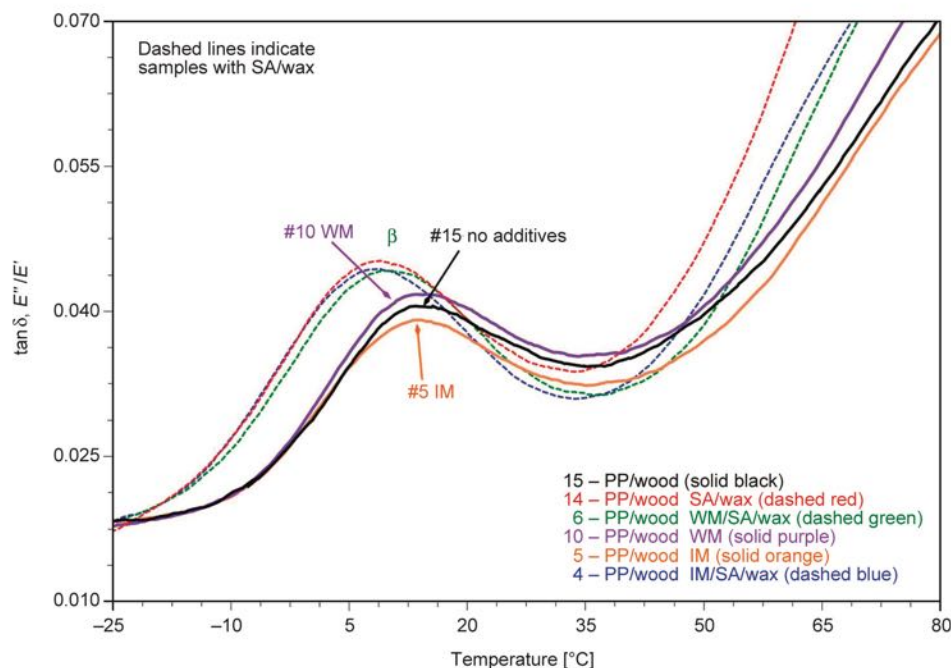


Figure 14. Expansion of Figure 13 depicting an overlay of DMA $\tan \delta$ (E''/E') curves at 1 Hz for PP/wood composites, illustrating the effects of IM and WM additives on damping behavior (viscoelastic loss) in the presence and absence of SA/wax over the temperature range -25 to 80 °C.

fusion were weight-corrected to account for the weight percentage of PP in the composites with the use of the data in Table 1. The composites containing IM and WM exhibited lower heats of fusion than the comparable composites that were made without these additives. This result is indicative of reduced PP crystallinity [45].

The lower heats of fusion for the composites made with neat IM and with neat WM are indicative of a higher fraction of amorphous PP, which implies that the meal additives impeded the rate of re-crystallization during post-fabrication cooling, possibly due to amorphous phase mixing between protein moieties and PP. However, the possibility of amorphous phase

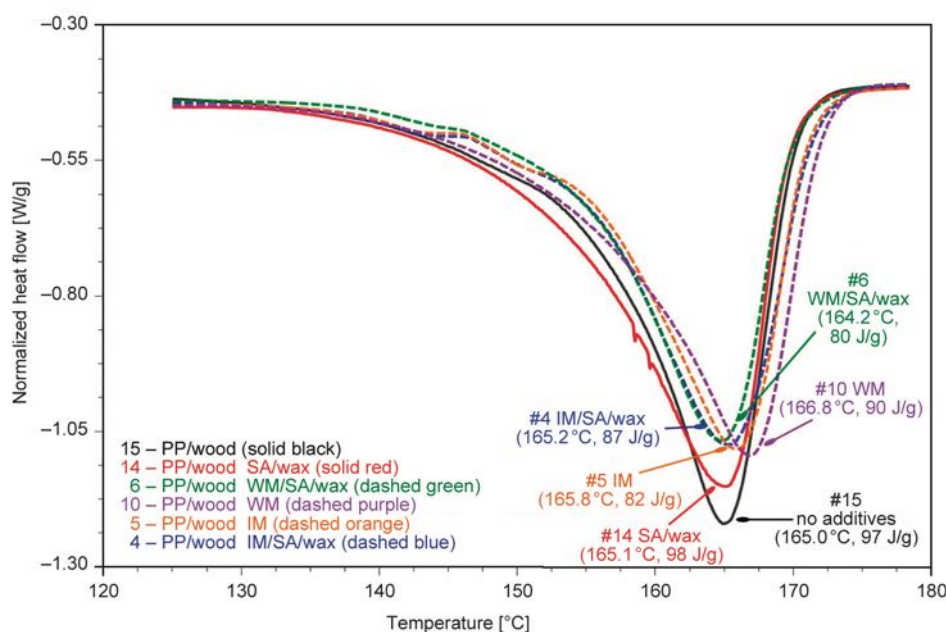


Figure 15. Overlay of DSC thermogram curves for PP/wood composites, showing the effects of IM and WM additives on the relative heats of fusion and endotherm minima for the PP melt transition. The heats of fusion were weight-corrected to account for the weight percentage of PP in the composites with the use of the data in Table 1.

mixing is not supported by the DMA loss modulus data, since the α - and β -transition temperatures were not affected by the presence of the neat meal additives. Moreover, neat soy protein isolate has been reported to exhibit a 1 Hz loss modulus maximum at approximately 125 °C [31]; and because there were no new loss modulus transitions near this temperature, we cannot presently say whether or not a fraction of the WM and IM additives remained dispersed within the PP matrix (*i.e.*, as independent morphological domains). Although DMA provides no evidence for phase separation or for matrix phase mixing, SEM and NMR studies are planned to probe further for this possibility.

Independently of the reason for the higher amorphous content in the IM and WM composites, it remains important to note that the composite with neat IM had a lower relative percentage of PP crystallinity (sample #5, heat of fusion: 82 J/g) than the comparable composite that was made with neat WM (sample #10, heat of fusion: 90 J/g). Given that the modulus of the PP matrix phase would be expected to scale with the percentage of PP crystallinity [46], one would expect that the modulus of the PP matrix phase in the IM composite would be lower than that of the PP matrix phase in the WM composite [46].

If we consider the factors that contribute to the overall modulus of a composite [4–7], and if we assume

that all factors are equal except for the contribution of the modulus of the PP matrix phase, the expectation would be that the modulus of the WM composite would be higher than the overall modulus of the IM composite. However, we observed the opposite trend (*i.e.*, WM led to a decrease in the overall composite modulus). Thus, it follows that the decrease in the modulus of the WM composite cannot be attributed to a decrease in the modulus of the PP matrix phase; instead, it can only be attributed to WM having a negative impact on one or more of the other contributing factors (*e.g.*, homogeneity of dispersion, wetting and adhesion between particulates and matrix phase, properties of the meso phase, *etc.*). Additionally, given that the temperature-dependent modulus of the composite that was made with neat IM was higher than that of the composite made with neat WM, and given that its $\tan \delta$ values were lower, it follows from the logic presented above that IM's positive impact on mechanical properties is also consistent with improved wetting between the PP matrix phase and the dispersed wood phase, and/or with improved homogeneity of wood dispersion in the matrix phase.

Analogous reasoning can be used when comparing the composite without additives (sample #15) to the composite that was diluted with 5% w/w neat IM (sample #5). Namely, the composite with neat IM had a lower relative percentage of PP crystallinity

(sample #5, heat of fusion: 82 J/g) than the comparable composite without additives (sample #15, heat of fusion: 97 J/g). Again, given that the modulus of the PP matrix phase would be expected to scale with the percentage of PP crystallinity [46], one would expect that the contribution of the PP matrix phase to the overall modulus would be lower in the IM composite than in the additive-free composite [46]. If we consider the factors that contribute to the overall modulus of a composite [4–7], and if we assume that all factors are equal except for the contribution of the modulus of the PP matrix phase, the expectation would be that the modulus of the additive-free composite would be higher than that of the IM composite. However, although the glassy-state modulus for the additive-free composite was slightly higher, the two composites exhibited similar modulus values over the remainder of the temperature range (Figure 11). Given that the temperature dependent modulus of the composites were similar over most of the temperature range, and given that the $\tan \delta$ values for the IM composite were lower despite the lower concentration of PP and wood, it follows that the contribution of the PP matrix phase to the modulus of the IM composite was overshadowed by IM's positive impact on one or more of the other contributing factors [6], such as improved wetting between the PP matrix phase and the dispersed wood phase, and/or improved homogeneity of particulate dispersion in the matrix phase. Again, these possibilities remain under investigation.

Reverting to the FTIR and CA data, we are presently left with competitive surface adsorption between SA and IM as being the most viable explanation for IM-induced surface segregation of SA. Given the absence of a loss modulus phase transition associated with independent IM domains, and given that the amorphous phase transition temperatures for PP were not affected by the meal additives, we are left with the hypothesis that the IM-induced surface separation of SA (*i.e.*, as measured via FTIR) is associated with competitive surface adsorption and displacement of SA from the surfaces of the wood particulates by IM, as proposed earlier.

Finally, the results of these studies collectively indicate that the water-soluble fraction in WM has a negative impact on thermal stability, dynamic mechanical properties, and UV absorptivity. In addition, unlike IM, WM had no effect on the composite's surface hydrophobicity. On the other hand, the combination

of IM and SA led to a synergistic enhancement in surface hydrophobicity at reduced levels of SA. Said differently, the composite made with 2.5% IM in combination with 2.5% SA/wax led to enhanced hydrophobicity even though the concentration of SA was half that used in the comparable composite made with 5% w/w neat SA/wax (sample #14). Given that SA interferes with coupling and deteriorates mechanical properties [1], the synergy between SA and IM affords an opportunity to achieve enhanced hydrophobicity at lower SA levels, thereby minimizing SA's negative impact on mechanical properties, while still providing the formulation latitude to impart lubricity during melt processing to the extent required (*i.e.*, if the need presents itself to mitigate shear degradation associated with PP chain scission). Although the combined data seem to collectively align with the idea that IM can promote matrix phase wetting of wood particulates, conclusive proof of the mechanisms will only come through further studies of mechanical properties and morphology, as mentioned previously, and through studies of interfacial compositions (*e.g.*, via FTIR microscopic analyses of failure interfaces). However, from a practical and phenomenological perspective, the removal of water-soluble components leads to improvements in all properties. Thus, regardless of the underlying reasons, these discoveries indicate that IM is deserving of further consideration for use as an additive in PP wood composites, and perhaps for use as a multi-purpose additive in other polyolefin applications.

4. Conclusions

We have discovered that the removal of water-soluble components (*e.g.*, carbohydrates) from WM leads to an IM protein fraction which has the potential to function as a multi-purpose additive for use in polymer/wood composites. For example, the incorporation of IM into PP/wood composites led to improved surface hydrophobicity when it was used in combination with SA, as well as to improved thermal oxidative stability (*e.g.*, reduced rates of lignin degradation and reduced levels of thermal weight loss below the onset of PP degradation). In addition, IM exhibited strong UV-absorption characteristics in the UV-B and UV-C regions, suggesting that the IM proteins might also have the potential to impart UV and photo-oxidative stabilization characteristics. The incorporation of 5% w/w neat IM also improved mechanical properties when compared to 5% neat WM

and 5% SA/wax alone. Moreover, dilution with 5% w/w neat IM had little to no impact on mechanical properties when compared to an undiluted composite, despite the lower levels of PP and wood in the case of the diluted composite. Although the effects of WM and IM on the mechanisms of wetting and coupling remain uncertain, we have nevertheless observed that the presence of water-soluble components leads to inferior performance, including reduced surface hydrophobicity, worse thermal stability, reduced composite modulus, higher viscoelastic loss, and reduced UV absorptivity. Thus, the removal of these components from WM appears to be a necessary precursor for the optimization of IM as a polymer additive.

No one to our knowledge has attempted to use this type of soy protein isolate (*i.e.*, IM devoid of water-soluble components) as an additive for polymer composites, let alone for the purpose of imparting hydrophobicity, thermal stability, or UV-absorption characteristics. Although we have hypothesized possible mechanistic reasons for these attributes (*e.g.*, protein isolation and denaturing to achieve improved polymer-to-wood wetting/coupling, competitive wood particle surface adsorption leading to SA displacement and phase separation, protein side-chains with the capacity to absorb UV radiation and to react under thermal-oxidative conditions), a great deal of work remains to be done, not only to test our hypotheses, but also to study the process and end-use property characteristics of polymers and composites made with IM additives. To this end, additional studies are underway, including: a) studies aimed at testing the effects of IM on melt viscosity and on chain defect formation during compounding [1, 3]; b) studies aimed at determining whether IM can be made to provide polymer to wood coupling analogous to that imparted by MAPP in similar PP/wood composites [8, 9]; and c) studies aimed at determining whether the UV-absorption characteristics of IM can lead to improved photooxidative stability in PP/wood composites.

In addition, we are also developing surface-treated versions of IM, one of which is showing promise as an additive for enhancing the moisture resistance of laminated wood composites made with soy-based adhesives. These surface treatments will soon be evaluated for their ability to impart lubricity during polymer/wood composite processing. Thus, our goal

is to optimize an all-natural, multi-faceted additive for general use in the polymer industry, either as a replacement for traditional synthetic additives, or as a co-additive for specific process and end-use purposes (*e.g.*, lubricity, hydrophobicity, thermal stabilization, UV stabilization).

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