EFFECTS OF SELECTED WOOD SPECIES AND MOISTURE CONTENT ON PMDI RESIN APPLICATION AND PANEL PROPERTIES

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Abstract. This study investigated the effect of selected species and moisture conditions on resin distribution and composite panel properties. Flakeboard composites were made from aspen, pine, or poplar flakes that were equilibrated to 4, 8, or 12% moisture content (MC). Resin droplet size (resin "footprint") was measured, as was the percentage of the flake surface that was covered by resin. Aspen flakes showed higher resin coverage and also larger resin droplet sizes. Conversely, pine and poplar flakes had smaller resin droplets and lower resin coverage per flake surface, suggesting greater pMDI penetration. Internal bond (IB) testing revealed optimal performance for aspen flakes at 12% precure MC, and poplar and pine flakes at 8% precure MC. Modulus of rupture and modulus of elasticity results correlated with IB results. Aspen panels bonded at 8 and 12% MC had minimal thickness swell.

Keywords: pMDI, aspen, poplar, pine, composites, resin droplet size, internal bond strength, thickness swell.

INTRODUCTION

Polymeric diphenylmethane diisocyanate (pMDI) binders are commonly used in structural wood composites. These resins exhibit excellent moisture resistance even at relatively low resin loadings, and due to the inherent reactivity of isocyanate with water, completely drying the wood flakes is not necessary. The underlying reasons for the excellent performance of pMDI resins continue to be researched, with significant

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effort devoted to detecting resin-wood covalent bonds. While spectral peaks consistent with urethanes have been detected via nuclear magnetic resonance spectroscopy (Zhou and Frazier 2001), research has also suggested that common industrial manufacturing conditions lead to a chemically complex polyurea-based network (Bao et al 2003). Recently, the excellent durability of pMDI resin has been attributed to its unique morphology (Bao et al 2003; Frazier 2003). Specifically, the low molecular weight of pMDI at application allows it to effectively diffuse into the wood cell wall. This impacts the relaxation behavior of wood polymers, as evidenced by Marcinko et al (1998, 1999). The existence of a cured, interpenetrating network (IPN) morphology is a conceptual possibility, and if it exists, this morphology could explain some of the unique performance attributes of pMDI.

As noted previously, MC impacts pMDI-wood cure chemistry, but another important factor is the species of wood being adhered (Johns et al 1982, 1985; Gruver and Brown 2006; Das et al 2007). Johns et al conducted two studies (1982, 1985) on the effect of species, MC (MC), and assembly time on bonding composite panels. Selected species included Douglas-fir, white fir, red oak, hickory, and loblolly pine; the various species formed panels with unique performance attributes. Their second study reported the effects of five hardwoods species (aspen, green ash, paper birch, red maple, and northern red oak) on panel properties. Work by Gruver and Brown (2006) and Das et al (2007) have also probed isocyanate-species interactions, but on solid wood substrates.

In this paper, we further explore the species and moisture-dependent performance of pMDI in wood composites made from selected species. Of particular interest here is identifying resin distribution differences among the species, and potentially correlating resin distribution with panel properties. Aspen, poplar, and pine flakes, equilibrated to 4, 8, or 12% precure MC were used. These species were selected due to their widespread use in industry as well as their

prevalence in previously reported research. Panel properties including density, IB strength, thickness swell, modulus of elasticity, and modulus of rupture were all measured. Image analysis provided insight regarding resin distribution on wood flakes.

<div align="center">EXPERIMENTAL</div>

Board fabrication

Wood composite manufacturing processes include a number of variables. Here, many of these variables (resin loading, flake dimension, flake orientation, wax loading, release agents, atomizer features and settings, and press features and settings) were kept constant to isolate the effects of species and MC. Bigtooth aspen (*Populus grandidentata*), yellow-poplar (*Liriodendron tulipifera*), and southern pine (*Pinus spp.*) wood flakes were prepared from kiln-dried boards which were pressure-soaked in water prior to flaking. All flakes were prepared with a laboratory disc-flaker and were kiln-dried to 3% MC. Flake dimensions were approximately $100 \times 30 \times 0.8$ mm. The flakes were equilibrated to 4, 8, or 12% MC precure MC. Equilibrated flakes for a given species and MC condition (combinations will be designated "species*MC") were placed in a tumbling blender fitted with an EL-3 disc atomizer. Commercial slack wax was applied to a target of 0.5% per OD furnish weight. Bayer Mondur® 541 Light pMDI resin was then applied to a target of 3.0% per OD furnish weight. After blending, mats were formed by hand in a random orientation within a deckle box. The 0.8- $\times$ 0.8-m mats were formed on caul plates presprayed with a release agent. The mat was then placed in a hot press preset at 204°C with a total press cycle of 150 s and final panel thickness target of 13 mm. Three panels were produced for each species*MC combination; a total of 27 panels were produced and tested.

Board testing

From each panel, ten 50- $\times$ 50-mm IB samples, two 150- $\times$ 150-mm thickness swell (TS)

samples, and four 80- × 360-mm samples for density, modulus of elasticity (MOE), and modulus of rupture (MOR) were cut. All specimens were tested in accordance with ASTM D 1037–06a (ASTM 2006). MOR, MOE, and IB samples were tested on an Instron Testing Machine.

Image analysis

Image analysis samples were randomly selected from the blender following wax and resin application. Thirty flakes were taken from each species*moisture condition. These samples were analyzed using Bayer MaterialScience's portable Mondur® Monitor (patent application submitted) image analyzer. The instrument was calibrated prior to use by examining flakes with given resin coverage. The Mondur® Monitor image analysis software was used to calculate percentage resin coverage and resin droplet size. Outliers (points outside of the distribution by more than 1.5 times the interquartile range) were discarded from the data set. In all species*moisture conditions, more than 20 data points remained.

Statistical analysis

The statistical model for predicting the least squared means (Minitab, General Linear Model) was expressed as a function of wood species and MC, plus their two-way interaction (species*MC). Analyses of variance (ANOVA) with Tukey multiple comparison tests were used to evaluate the data, where the criterion for significance was $\alpha = 0.10$. Covariance analyses were also performed to account for the impact of panel density on the panel physical properties. Throughout the paper, interval plots are used to illustrate the data. Mean values for a given combination of species and MC are illustrated by a symbol. The error bars represent 90% confidence intervals about each mean. Mean values for a given species are joined by line segments to illustrate trends in the data.

RESULTS

Characteristics of resin coverage on flakes

A study by Smith (2003) revealed that adhesive droplet size and adhesive coverage on the flake surface could give rise to differences in board properties and performance. The Bayer Mondur® Monitor (patent application in progress) was used to both: 1) probe the percentage of the flake surface that was covered by resin, and 2) identify individual resin droplet areas. Note that the term "resin droplet" refers to the observed footprint of the resin on the flake surface. As such, it is not only a consequence of resin atomization, but is also impacted by flake-flake contacts during blending, as well as the complex interaction of resin wetting and penetration during the time interval between resin application and analysis.

Figure 1 reveals the mean percentage of flake surfaces that were covered by resin, whereas Fig 2 indicates mean resin droplet size on the flake surface. Statistical modeling determined that both wood species and MC have significant effects on resin droplet size and percent resin coverage.

Resin coverage changed significantly as a function of moisture conditions (Fig 1). Pine flakes equilibrated to 4% MC had the greatest resin
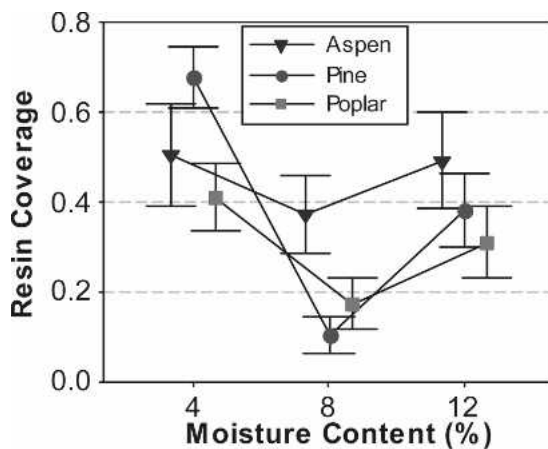

FIGURE 1.   Fraction of the flake surfaces that are covered with pMDI resin for each species*MC condition.
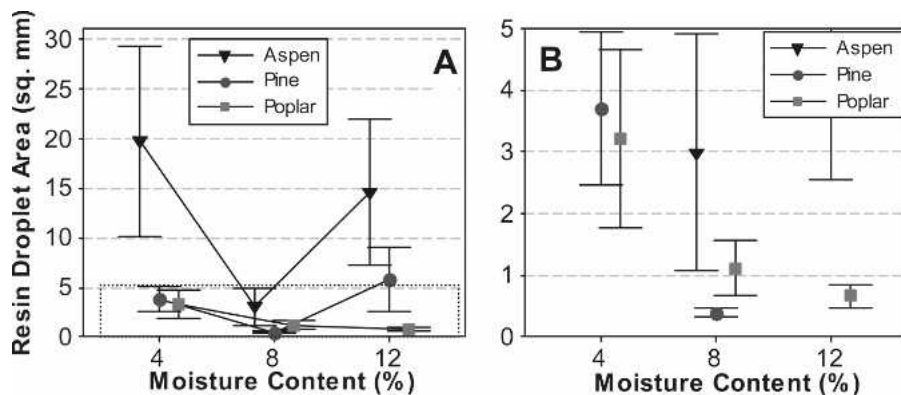
FIGURE 2. Resin droplet sizes as observed on the wood flakes. Dashed area within A (left) is replotted in B (right). Lines connecting means were deleted in B.

coverage, while resin coverage dropped at 8%, and recovered moderately at 12% MC. The same overall trend was true for poplar flakes (lowest coverage at 8% MC, highest coverage at 4% MC). For aspen, minimum coverage was again observed at 8% MC, and yet this value far exceeded minimum coverage values in pine and poplar.

Like resin coverage, resin droplet size (Fig 2) differed according to species and MC. The range of droplet sizes was substantial, particularly on aspen flakes. The largest droplets were found on aspen flakes equilibrated to either 4 or 12% MC. Smaller resin droplets were detected on flakes equilibrated to 8% MC. It is interesting to note that this corresponds with a minimum in observed resin coverage (Fig 1). A number of changes occur as MC increases from 4–12%, including increasing wood tissue permeability, increasing surface free energy, and increasing potential for pMDI-moisture reactions. The unique behavior at 8% MC suggests minimal spreading, perhaps due to pMDI penetration; however, this was not measured experimentally.

In terms of the magnitude of the observed droplet sizes and the level of flake coverage, Kamke et al (1996) observed significantly smaller droplets (average pMDI droplet size was 917 μm^2) and lower resin coverage ($\leq 1\%$) in flakes blended under similar conditions, but using a different type of atomizer. The Mondur® Monitor requires the transfer of resin from the flake surface to paper. The paper is then analyzed, thus eliminating interference from lignin fluorescence. Clearly, the pressure applied during resin transfer step influences the resulting data. Both the Mondur® Monitor technique and the fluorescence microscopy approach used by Kamke et al are informative. The Mondur® Monitor data mimics resin coverage on pressed strands and analyzes a relatively broad section of the flake surface (625 mm^2); Kamke's approach more precisely investigates initial droplet sizes (before any pressure is applied) over a much smaller region of the sample (1.65 mm^2).

Physical properties of panels

Pressing parameters were maintained throughout the experiment to identify the effects of species and flake MC. Panel physical properties measured here are in line with data recently published by Papadopoulus et al (2002).

Internal bond. Wood species and MC gave rise to differences in IB strength (Fig 3). IB tests are indicative of both adhesion and panel density. Excellent correlations were found between IB and panel density for yellow-poplar specimens ($R^2 = 0.9999$); the correlation was less notable for aspen and pine (R^2 values = 0.8383 and 0.7917, respectively). The finding that 4% MC led to poor performance in aspen and pine is not
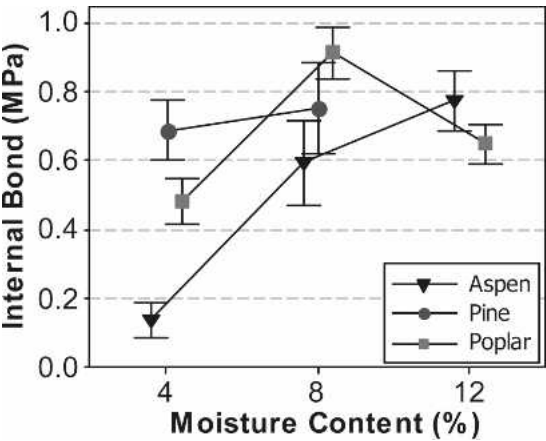
FIGURE 3.   IB performance plotted as a function of species and MC.

TABLE 1.   *Mean panel physical properties as tested by ASTM D-1037 (ASTM 2006).*

Wood Species	Pre-cure Flake MC (%)	Panel Density (kg/m³)	Modulus of Elasticity (MPa)	Modulus of Rupture (MPa)
Aspen	4	718.6	3578	17.98
	8	672.1	5684	41.70
	12	681.6	6346	46.48
Pine	4	705.9	4801	32.09
	8	706.7	4955	34.02
	12	668.2	4135	27.37
Poplar	4	658.2	5087	35.48
	8	704.5	6060	42.92
	12	677.4	4983	33.95

surprising; two factors likely contributed to this result. Moisture plays an important role in building the polyurea network, so limited moisture suggests poor evolution of the resin network structure. Secondly, limited water vapor (steam) results in poorer heat transfer to the panel core, thus affecting resin cure. While poplar and pine showed maximum performance when flakes were equilibrated to 8% moisture, the performance of aspen panels was best at the highest moisture level.

Poor performance at 4% MC is also consistent with the presence of relatively large resin droplets (Fig 2). Smith (2003) showed that strands with larger droplets gave rise to relatively poor performance (via lap shear tests) as compared with strands coated with smaller droplets. In poplar and pine, optimal IB performance was observed at 8% MC, which corresponds with small resin droplets and low resin coverage.

MOR, MOE, and density. Correlations between MOR and MOE (raw data in Table 1) were very strong (R^2 values for MOR vs MOE for aspen = 0.9946, poplar = 0.9948, pine = 0.9883; data not shown). Statistically significant correlations were also evident between MOR and panel density, as well as MOE and density. Finally, aspen and pine exhibited very strong correlations between IB and MOR ($R^2 = 0.9852$ and

0.9794, respectively; poplar $R^2 = 0.7027$) and between IB and MOE (0.9977 and 0.9375, respectively; poplar $R^2 = 0.7663$). In all cases, increased MOR or MOE correlated with increased IB. For aspen, increased MC led to increased MOR and MOE. In pine and poplar, MOR and MOE peaked at 8% MC.

Thickness swell. After exposure to a 24-h water soak, sample dimensions were measured to determine thickness swell (Fig 4). At low precure MC, poplar panels had better resistance to thickness swell than aspen. At 8 and 12% precure MC, aspen showed optimal resistance to thickness swell. Pine had no statistical differences in thickness swell when precure flake MC was changed. Some notable similarities exist be-
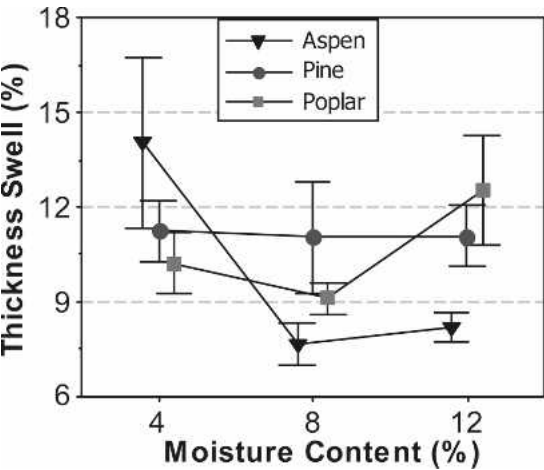

FIGURE 4.   Interaction plot of species*MC for thickness swell.

tween the thickness swell and the IB data, including the 4%-aspen being among the worst performers, and the 12%-aspen and 8%-poplar being among the best performers. The relatively constant performance of pine was not expected.

CONCLUSIONS

Wood composite panels prepared with pMDI binder exhibited properties that depended on both wood species and MC. Precure MC affected the performance of each wood species differently. The role of isocyanate cure chemistry at the various MC likely contributed to observed performance differences. Aspen IB values peaked at 12% MC, while poplar and pine IB values peaked at 8% MC. Thickness swell results indicated that aspen had optimal resistance to thickness swell at 8 and 12% MC, while poplar yielded the best results at 4 and 8% MC. Pine exhibited similar thickness swell regardless of precure MC. Image analysis of resin droplet size and resin coverage suggested distinct differences for the species and moisture conditions. All wood species displayed a slight drop in resin coverage and droplet size at 8% MC, suggesting greater pMDI penetration into wood at this moisture level. Correlations between resin coverage and performance were not statistically significant when all conditions were included. However, the optimal IB of poplar and pine at 8% MC does correspond to low coverage and small droplets, confirming the results of Smith (2003). In sum, these results suggest that the wood species and MC affect resin droplet size and flake coverage as well as panel properties.

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