Cost Savings from Soy Flour Substitution in Methylene Diphenyl Diisocyanate for Bonding Flakes and Particle

Qingzheng Cheng Charles Essien Brian Via Sujit Banerjee

Abstract

Methylene diphenyl diisocyanate (pMDI) adhesive used in the manufacture of oriented strand board and particleboard can be partially substituted with soy flour for significant cost savings. The flour is about one-third of the cost of pMDI. Properties such as internal bond, wet modulus of elasticity and modulus of rupture, and thickness swelling are unaffected by soy flour substitution of up to 20 percent. Adding soy flour to the regular dose of pMDI can improve board properties and reduce delamination.

 \mathbf{S}_{oy} flour and soy protein are commercially used in adhesive formulations in products such as decorative veneer where exposure to water is relatively low (Li et al. 2004, Li 2010). Being hydrophilic, soy products tend to retain water, which potentially causes board distortion or structural failure in a moist environment. The advantage of soy flour over an adhesive such as methylene diphenyl diisocyanate (pMDI) is principally cost; it is about one-third the cost of pMDI. Hence, 20 percent soy flour substitution in pMDI would lower total adhesive costs by 13 percent. There is also a green value attached to the use of soy products. Soy protein is more expensive than pMDI; as a result, the extensive literature (Vnučec et al. 2017) on adhesive formulations with soy protein has yet to find commercial application. This article continues our previous work on the application of soy flour to structural panels (Hand et al. 2017, 2018; Via et al. 2019). It defines the acceptable range of soy flour substitution in pMDI for oriented strand board (OSB) applications and discusses some of the operational factors that must be understood and taken into account before commercial use can be considered. The chemistry of the interaction of soy flour components and pMDI has been discussed elsewhere (Mhike 2014, Hand 2018).

Materials and Methods

Screened pine wood strands (moisture content: 7% to 8%) were donated by J. M. Huber, Louisiana-Pacific, and

Norbord corporations. Wood sawdust particles were obtained from West Fraser and dried to 6 to 7 percent moisture content. Defatted soy flour (7B) was provided by Archer Daniels Midland; pMDI (MONDUR 541) and emulsified wax (Hexion Bord'N-Seal FMH-XD) were provided by Huber Corporation.

The wax was first sprayed on wood strands/particles at 1 percent loading. Then pMDI or mixtures of pMDI and soy flour were heated to 40°C and sprayed on the furnish at 2 and 4 percent loading for OSB and 6 percent loading for particleboard with a paint sprayer powered by an air compressor. The pressure was adjusted to optimize the droplet size distribution. Mats were formed in a 43 by 43-cm frame and then hot pressed for 3 minutes at 213°C and 2 mPa for boards with 2 percent adhesive. The pressure and

doi:10.13073/FPJ-D-18-00050

The authors are, respectively, Research Fellow, Postdoctoral Fellow, and Director of Forest Products Development Center, School of Forestry and Wildlife Sci., Auburn Univ., Auburn, Alabama (qzc0007@auburn.edu, cze0017@tigermail.auburn.edu, brianvia@ auburn.edu [corresponding author]); and Emeritus Professor, School of Chemical and Biomolecular Engineering, Georgia Tech, Atlanta (sb@gatech.edu). This paper was received for publication in December 2018. Article no. 18-00050.

[©]Forest Products Society 2019. Forest Prod. J. 69(2):154–158.

temperature were held constant for the entire 3 minutes. A distance bar was added to reach the target thickness during pressing. Shorter press times were used for boards prepared with 4 percent adhesive. The nominal thickness of the board was 11 mm. The target density was 641 kg/m^3 (40 lb/ft³) and 689 kg/m^3 (43 lb/ft³) for OSB and particleboard, respectively. The OSB had a 50 percent surface-to-core ratio, while the particleboard layer was a single-layer board. The furnish moisture content for all studies was approximately 8 percent.

The size of adhesive droplets (with and without soy flour) in the spray was quantified by spraying on poster paper from a distance of 55 cm. This distance offered the best compromise between over- and underspraying. The former condition leads to overlap of the droplets; the latter makes visualization difficult. The adhesive was kept at 40°C. The paper was then imaged by ImageJ software (Rasband 2018). The droplet diameter was averaged from 400 drops.

The moisture cycle test for bonding performance (single cycle or D4 test) was run according to APA PS2-10 (APA-The Engineered Wood Association 2004). Internal bond (IB), water absorption (WA), and thickness swelling (TS) were measured according to ASTM D1037-12 (ASTM International 2012).

Results and Discussion

Effect of soy flour substitution on adhesive viscosity

The viscosity of the adhesive must be kept below 1,000 mPa/s to ensure smooth spraying (Carvalho et al. 2014). Figure 1 illustrates the viscosities of 9:1 pMDI:soy flour mixtures at various temperatures. Corresponding values for pMDI alone are provided in Table 1; these viscosities vary only by 7 percent over 10 hours and are, therefore, functionally time independent at each temperature. The 25°C values in Figure 1 increase over time, in contrast to the behavior seen at higher temperatures. This has practical



Figure 1.—Viscosities of methylene diphenyl diisocyanate (pMDI) substituted with 10 percent soy flour.

FOREST PRODUCTS JOURNAL VOL. 69, NO. 2

Table 1.— Methylene diphenyl diisocyanate viscosity at various temperatures.

| Temperature (°C) | Viscosity (mPa/s) |
|------------------|-------------------|
| 25 | 299 |
| 40 | 109 |
| 50 | 160 |
| 60 | 38 |

implications because it is difficult to spray fluids of high viscosity. The likely reason for the high viscosity at 25°C is bubble entrapment. Carbon dioxide bubbles are generated from the reaction between pMDI and the water contained in soy flour. At low temperatures where the native viscosity of pMDI is relatively high (Table 1), the bubbles are trapped in the adhesive, which increases the viscosity of the soy/pMDI mixture. At higher temperatures, the pMDI viscosity falls, which facilitates the escape of the CO_2 bubbles. Much more froth was associated with the adhesive mixture at higher temperature, which is consistent with greater bubble release from the adhesive. The presence of bubbles is known to increase viscosity (Albartamani 2000, Abivin et al. 2008, American Chemistry Council 2012) because of flow line distortion around the bubbles (Llewellin et al. 2002). Frothing was not observed from the resinated flakes because the bubbles were unable to coalesce.

Effect of soy flour on adhesive droplet size

The addition of soy flour to pMDI adhesive affects its droplet size on spraying. The results are shown in Table 2. All the values are statistically similar, implying that the soy flour should not affect the spraying operation. Atomizers present in an industrial process will reduce the droplet size further, so any effect of soy flour on droplet size will be further attenuated. However, some nozzle clogging was observed at 20 percent soy flour substitution; the nominally low droplet value of 0.35 mm in Table 2 is a consequence of the larger particles being retained in the nozzle. Hence, 15 percent soy flour substitution appears to be a safe operational maximum at least from this perspective. Spraying at higher temperature should presumably reduce nozzle clogging.

Effect of soy flour substitution on OSB properties

Our initial work with both strand board and particleboard was done with soy flour and adhesive dispensed separately onto the furnish. However, the wet properties were compromised in the presence of soy flour. It is likely that free soy particles on the surface of the wood attracted and retained water. The problem disappeared when the adhesive and soy flour were mixed before application because the soy flour bonded with the pMDI (Hand et al. 2018). The results

Table 2.—Droplet sizes of methylene diphenyl diisocyanate (pMDI) and mixtures of pMDI and soy flour.

| Droplet | Size (mm) |
|----------------|-------------------|
| pMDI | 0.29 ± 0.12 |
| Soy flour, 10% | 0.32 ± 0.11 |
| Soy flour, 15% | 0.39 ± 0.13 |
| Soy flour, 20% | 0.35 ± 0.1325 |

Table 3.—Effect of soy flour substitution on wet oriented strand board panel properties.^a

| | | Percent soy flour substitution | | | |
|-----------------------------------|-----------------|--------------------------------|-----------------|---------------|--|
| | 0 | 10 | 20 | 30 | |
| (a) Soy only in face adhesive | | | | | |
| Wet MOR (mPa) | 12 ± 2 | 11 ± 2 | 12 ± 1 | 8 ± 2 | |
| Wet MOE (mPa) | $1,400 \pm 300$ | $1,300 \pm 300$ | $1,400 \pm 200$ | 800 ± 300 | |
| Thickness swelling (%) | 42 ± 4 | 40 ± 4 | 41 ± 3 | 49 ± 5 | |
| (b) Soy in face and core adhesive | | | | | |
| Wet MOR (mPa) | 12 ± 3 | 11 ± 3 | | | |
| Wet MOE (mPa) | $1,400 \pm 370$ | $1,200 \pm 320$ | | | |
| Thickness swelling (%) | 37 ± 6 | 42 ± 5 | | | |

^a MOR = modulus of rupture; MOE = modulus of elasticity.

presented were all obtained with premixed soy flour and pMDI. In some instances, soy flour was only substituted in the face adhesive.

Two percent adhesive loading.—The effect of substituting pMDI adhesive with soy flour on wet properties is shown in Table 3. Measurements were made with (a) soy flour present only in the face adhesive and (b) in both face and core. Up to 20 percent soy substitution can be tolerated in the face-treated boards. No statistical difference in properties between control and soy treated is evident for condition (b), where the soy was substituted in both face and core layers. However, soy flour substitution was limited to 10 percent in this case.

Four percent adhesive loading.—Boards are usually made under conditions that go beyond minimum specifications to maintain a safety margin. A small substitution of soy flour may not significantly affect the measured properties. Runs were made at press times of 1.5 and 1.75 minutes, which were well below the 3-minute press time used in the rest of the study. It was anticipated that the effect of soy would be more apparent under the shorter press times, where the boards would be weaker. The internal bond results illustrated in Figure 2 show that, as expected, the strength decreased when the pMDI load was reduced from 4 to 3.6 percent pMDI. However, addition of soy flour to 3.6



Figure 2.—Effect of soy flour substitution on internal bond at 1.5- and 1.75-minute press times.

percent pMDI restored the strength back to the value obtained at 4 percent pMDI. Similar trends hold for the wet properties obtained from the D4 test, as shown in Figure 3, where the soy flour improved the properties obtained with 3.6 percent pMDI. The edge swelling is of particular interest because it is a critical property for panels with potential exposure to moisture. These results demonstrate that soy flour can be used to partially substitute pMDI adhesive to reduce cost or be added to the regular adhesive dose to improve board performance.

The edge swelling values in Figure 3 are quite similar; evidently, edge swelling is insensitive to small changes in soy flour substitution. However, a clear difference was observed in board delamination. The soy flour–substituted boards (3.6% pMDI + 0.4% soy) performed almost as well as the 4 percent pMDI boards, whereas the 3.6 percent pMDI samples pressed for 1.5 minutes delaminated to a greater extent.

Insight into the factors that influence edge swelling were obtained by identifying the sample subsets most responsible



Figure 3.—Effect of soy flour substitution on wet properties at 1.5- and 1.75-minute press times. MOE = modulus of elasticity; MOR = modulus of rupture.



Figure 4.—Effect of soy flour on edge swelling.

for edge swelling. Results from a trial run for OSB at 4 percent adhesive load with the soy present only at the surface are presented in Figure 4. The P value for the two sets of data is 7.5E-5, which indicates that they are statistically different but only marginally so from a practical perspective. Fractionating the results into different ranges of edge swelling provides the histogram shown in Figure 5. The 22 to 26 percent fraction is higher for the soy-substituted adhesive, whereas the opposite is true for the other fractions. It is possible that the 22 to 26 percent bin represents fines and smaller chips. This fraction is the weakest element in the board structure, and it is therefore



Figure 5. — Distribution of edge swelling.

Table 4.—Properties of single-layer particleboard made with premixed soy flour and methylene diphenyl diisocyanate.

| Soy (%) | Internal bond (mPa) | Water absorption (%) ^a | Thickness swelling (%) ^a |
|------------|------------------------|-----------------------------------|--|
| 0 | 1.5 ± 0.5 | 17 ± 3 | 15 ± 2 |
| 10 | 1.6 ± 0.5 | 17 ± 3 | 15 ± 2 |
| 20 | 1.4 ± 0.4 | 16 ± 1 | 15 ± 4 |

^a From D4 test.

likely that it will be particularly susceptible to edge swelling.

Effect of soy flour substitution on particleboard properties.—Board properties of soy-substituted boards are compared with those of control boards in Table 4. All the boards were close to the target density and pressed for 3 minutes. There are no statistical differences, and no penalties should be incurred by soy substitution. The overall adhesive loading was 6 percent throughout the board, so the adhesive cost savings will be proportionally greater than that obtained for OSB.

Conclusions

Up to 20 percent of pMDI adhesive can be substituted by soy flour in OSB or particleboard without degrading board properties. The cost savings are significant because soy flour is about three times cheaper than pMDI adhesive. Board properties deteriorate at higher levels of substitution, and the adhesive mixture increases in viscosity, so 20 percent substitution appears to be a practical upper limit at this time. Addition of soy flour to the regular dose of pMDI can improve board performance.

Acknowledgments

This study was funded by the United Soybean Board and the Alabama Farmers Federation. We would like to thank Marina Hornus, Osei Asafu-Adjaye, Alejandro Ariel Cardozo, and Lixia Hu for their help in this study.

Literature Cited

- Abivin, P., I. Hénaut, J-F. Argillier, and M. Moan. 2008. Viscosity behavior of foamy oil: Experimental study and modeling. *Petroleum Sci. Technol.* 26:1545–1558.
- Albartamani, N. S. 2000. Experimental studies on "foamy oil" phenomena. PhD thesis. University of Alberta, Alberta, Canada.
- American Chemistry Council. 2012. Guidance for working with pMDI and polymeric pMDI: Things you should know. https://polyurethane. americanchemistry.com/Resources-and-Document-Library/11364.pdf. Accessed April, 3, 2019.
- APA-The Engineered Wood Association. 2004. Performance standards for wood-based structural-use panels. PS2-04. APA-The Engineered Wood Association, Tacoma, Washington.
- ASTM International (ASTM). 2012. Standard test methods for evaluating properties of wood-base fiber and particle panel materials. ASTM D1037-12. ASTM, Philadelphia.
- Carvalho, A. G., F. A. Mori, R. F. Mendes, A. J. Zanuncio, M. G. da Silva, L. Mendes, and C. L. S. de Oliveira Mori. 2014. Use of tannin adhesive from *Stryphnodendron adstrigens* (Mart.) Coville in the production of OSB panels. *Eur. J. Wood Prod.* 72:425–432.
- Hand, W. 2018. Defatted soy flour substitution in phenol formaldehyde and methylene diphenyl diisocyanate wood adhesives and their curing kinetic behavior. PhD thesis. Auburn University, Auburn, Alabama.
- Hand, W. G., W. R. Ashurst, B. Via, and S. Banerjee. 2018. Mechanism of interaction of soy flour with phenol-formaldehyde and isocyanate adhesives. *Int. J. Adhes. Adhes.* 87:105–108.
- Hand, W., G. Cheng, B. Via, and S. Banerjee. 2017. Soy-substituted

liquid phenol formaldehyde binders for flakeboard. Eur. J. Wood Wood Prod. 75:135–138.

- Li, K. 2010. Formaldehyde-free adhesives and lignocellulosic composites made from the adhesives. US patent 7,722,712.
- Li, K., S. Peshkova, and X. Geng 2004. Investigation of soy proteinkymene adhesive systems for wood composites. J. Am. Oil Chem. Soc. 81:487–491.
- Llewellin, E. W., H. M. Mader, and S. D. R. Wilson. 2002. The rheology of a bubbly liquid. *Proc. R. Soc. Lond. A* 458:987–1016.
- Mhike, M. 2014. Characterization of methylene diphenyl diisocyanate

protein conjugates. PhD thesis. Portland State University, Portland, Oregon.

- Rasband, W. S. 2018. ImageJ. US National Institutes of Health, Bethesda, Maryland. https://imagej.nih.gov/ij. Accessed April 3, 2019.
- Via, B., W. Hand, and S. Banerjee 2019. Use of soy flour in adhesive formulations used to manufacture engineered wood composites. US patent allowed.
- Vnučec D., A. Kutnar, and A. Goršek. 2017. Soy-based adhesives for wood-bonding—A review. J. Adhes. Sci. Technol. 31(8):910–931.