Bio-resin Bonded Acetylated OSB

Emma Östmark[1,2], Mark Lawther[3], Rune Ziethén[4], Petra Nordqvist[2], Farideh Khabbaz[5], Eva Malmström[2] and Mats Westin[4]

[1]SP Technical Research Institute of Sweden, SE-114 86 Stockholm, Sweden [email: emma.ostmark@sp.se]

[2]KTH Royal Institute of Technology, Fibre and Polymer Technology, SE-100 44 Stockholm, Sweden [email: petranor@kth.se, mavem@kth.se]

[3]TLA Development, DK-4000 Roskilde, Denmark [email: ml@tla-development.com]

[4]SP Technical Research Institute of Sweden, SE-501 15 Borås, Sweden [email: rune.ziethen@sp.se, mats.westin@sp.se]

[5]Casco Adhesives AB, SE-100 61 Stockholm, Sweden [email: farideh.khabbaz@akzonobel.com]

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ABSTRACT

In this study two types of bio-based resins have been used for the production of oriented strand boards (OSB) from acetylated wood strands. The resins are a commercially available furan binder from TransFuran Chemicals and an experimental protein-based resin developed by the authors. This resin is at an early stage of development and was synthesised using enzymatically treated cereal protein fractions. A commercially available phenol-formaldehyde resin from Casco Adhesives AB was used for production of unmodified boards, as reference. The acetylated boards show a much lower thickness swelling compared to the boards made from unmodified wood. In particular the protein-based binder, since the unmodified boards disintegrated during water soak, whereas the acetylated ones showed a low swelling of 6.4 %. The internal bond strength, evaluated according to EN 319, of the acetylated board was lower than that of the unmodified wood for the furan binder, but the binder generally performed well. The strength retention after water soak and reconditioning of these acetylated boards was very good. The boards produced using the protein-based binder showed reversed behaviour and the acetylated boards showed higher IB than that of the unmodified wood, but the IB was barely acceptable even for the acetylated boards.

INTRODUCTION

Oriented strand board (OSB) is a board type made from wood strands consolidated by heat, pressure and a suitable adhesive. The areas of use for OSBs comprise applications demanding high strength combined with low weight. The North American market has previously been the dominating one for OSB, but currently, OSBs are finding larger markets in Europe. To expand the uses of OSB, an improved exterior durability is required since the poor dimensional stability of the boards due to moisture swelling usually limits their application areas. Wood modifications, for example wood acetylation, are useful methods for improving the dimensional stability and increasing the biological resistance of wood (Rowell *et al.* 1986, Larsson-Brelid 1998).

OSB has previously been fabricated using modified wood. For example heat-treated wood strands have been used for OSB production and the boards showed improved dimensional stability and lower thickness swelling than boards made from unmodified wood strands (Paul *et al.* 2006). In addition, the internal bond strength (IB) was unaffected by the treatment. Further, acetylated Norway spruce (*Picea abies)* with two different degrees of acetylation, has been used to produce OSB in combination with a phenol-resorcinol-formaldehyde type resin (Papadopoulus and Traboulay 2002). It was found that the dimensional stability was improved compared to similar boards of unmodified spruce and that the biological resistance was improved if modified wood with a high (WPG) (20.4 %) was used for the boards (Papadopoulos 2006). A lower WPG of 11 % was observed to be affected by microorganisms after 36 months ground stakes test. However, the internal bond strength was found to be lower for the boards produced with acetylated spruce. The IB of particle boards and OSB made form acetylated wood were affected by the type of cross-linker used, diisocyanate or formaldehyde (Papadopoulos *et al.* 2006). The diisocyanate showed higher IB when used with modified wood strands in OSB and this was explained by an improved wetting and a less pH dependent cross-linking system. Commonly, petroleum-based adhesives relying on formaldehyde cross-linking chemistry, such as phenol-formaldehyde or urea-formaldehyde, are used as wood board binders. To further improve the eco-efficiency of boards from modified wood, it would be desirable to use binders derived from renewable resources. A need for reduction of the use of petroleum-based raw materials and an urge to substitute formaldehyde with low-emitting, less irritating substances has initiated the search for new resin systems. Attempts to mix lignin or lignosulphonates with formaldehyde-cured resins have been made (Pizzi 2006). Also, phenol-formaldehyde substitutes from biomass, for instance lignin derivatives used for phenol substitution, have been explored (Effendi *et al.* 2008). However, for acetylated wood the phenol-formaldehyde resol type system is known to be low-performing, due to low wettability of acetylated wood by the resin (Rowell *et al.* 1987). Other types of binders, preferably from renewable resources, with satisfactory wetting of acetylated wood therefore need to be developed. Protein-based adhesives, commonly from soybean proteins in combination with other components, are currently being widely investigated. The soybean based adhesives see a large potential as wood adhesives and the excess of soy, primarily on the North American market, encourage their use (Pizzi 2006). However, the inherent water sensitivity of the proteins indicated the need for protein modification in order to be appropriate as wood adhesive (Sun and Bian 1999). Another interesting type of biobased material, obtained from bagasse carbohydrate waste, is the furan-type binders, which are already commercially available. This study introduces two biobased binders for use in production of OSB from acetylated wood, an already commercially available furan-type binder and an experimental, prototype cereal protein binder developed by the authors. The dimensional stability and the IB of the produced boards and controls will be presented and discussed.

EXPERIMENTAL

Materials

The biobased furan binder Biorez® (050915-A) was supplied by TransFuran Chemicals, Belgium and the phenol-formaldehyde resin (PF1550) was supplied by Casco Adhesives AB, Sweden. Cereal protein was provided by Lantmännen, Sweden. Hexamethylenediisocyanate and sodium bisulphite were supplied by Aldrich and the water-soluble, blocked diisocyanate was produced according to a literature procedure (Lin-Gibson *et al.* 2003). Scots pine (*Pinus Sylvestris*) sapwood was used for all boards.

Methods

Binder preparation

The Biorez was mixed with 5% (dry weight) of the M-type catalyst and was diluted with methanol to a dry content of 60%. The protein was fractionated and one specific fraction was utilised and subjected to enzymatic hydrolysis. The hydrolysate was concentrated to a dry content of 34% and was mixed with 2.5% (dry/dry) of the cross-linking diisocyanate before use.

Acetylation

The Scots pine was acetylated in SP's microwave heated pilot plant according to a literature procedure (Larsson-Brelid 1998). The degree of acetylation, expressed as acetyl content, was about 21%.

Fabrication of OSB

OSB strands were produced in a Bezner OSB flaker from the unmodified and acetylated wood. The acetylated wood was boiled in water 1 h prior to the flaking in order to avoid brittle failure of the wood. Size fractionation of the strands was performed in a OSB sieve and the largest fractions were used for the boards. Wood strands that had been stored at ambient conditions, 607.5 g of dry wood mass, were put in a rotary drum blender. Binder, 67.5 g (10% of dry wood mass), was added with a spray gun to the strands in a rotating resination drum for even distribution of the binder (extra 10 % was added to cover up for losses of strands and binder in the drum). The aimed wet weight of the resinated strands was taken out of the drum (750 kg/m^3) and the boards were hand-formed in a box (50x300x300 mm^3). A temperature probe was placed in the middle of the board and the boards were pressed in a hot press at 76 kN. Two distance bars of 10 mm height were placed between plates of the press to regulate the board thickness. The board pressing parameters are displayed in Table 1. The measured temperature was used as measuring point and the steam release when the temperature passed 110ºC in order to avoid steam bubbles in the board. The boards were cooled down under press to ambient temperature in order to avoid warping of the panels.

Table 1: Fabrication parameters for OSB:s

Binder	Press temp. [ºC]	Pressing time [min]	No of boards of unmodified wood	No of boards of acetylated wood
Biorez	150	10	3	2
Protein adhesive	160	20	3	2
Phenol-formaldehyde	185	9	2	0

Density measurements

Specimens 50x50 mm from the boards were sawn and the density was measured. The density profiles were measured using an ATR density profilometer.

Internal bond strength

Internal bond strength (IB) was evaluated according to EN 319. The IB was evaluated on differently treated samples: samples conditioned at 20 ºC and 65% RH, samples boiled in water during 2h that were allowed to cool down to 20 ºC during 1h, and samples that had been water soaked and reconditioned. The water soak cycle consisted of 20 ºC and 65% RH during 24 h, soak in water of 20 ºC during 24 h, and reconditioning in 20 ºC and 65% RH during 1 week.

Thickness swelling

Specimens were allowed to swell in water at 20 ºC during 24 h and reconditioned at 20ºC and 65% RH during 24 h. The weights and thicknesses were recorded prior to, and after the swelling.

Decay resistance test

Decay resistance according to the mini block test was assessed (Bravery 1979) and mass loss was calculated according to EN 113. However, the full results can not yet be reported due to the continuation of the testing period.

RESULTS AND DISCUSSION

Oriented strand boards (OSB:s) of unmodified and acetylated Scots pine could successfully be made using two different bio-based resins. The two bio-based resins were a commercially available furan-type binder, Biorez, and an experimental protein hydrolysate, produced by the authors. As a reference, a phenol-formaldehyde (PF) resin was included for fabrication of boards from unmodified pine. It has earlier been shown that boards of acetylated aspen flakes glued with PF resins of resol type show inferior strength compared to boards made from unmodified wood (Rowell *et al.* 1987), probably due to a too low wetting of the acetylated wood with this type of binder. However, it has been demonstrated that a PF novolak type resin, which is a more hydrophobic prepolymer system that was cross-linked using hexamethyltetramine, showed increased wettability of acetylated wood (Gomez-Bueso *et al.* 1999). Consequently, no such PF control board was included in this study. The boards produced were hand-formed and hot-pressed in laboratory. Due to colour differences of the resins, the boards produced from the furan resin and the PF resin were darker, while the use of protein resin resulted in boards where the wood could be seen through the binder. The pressing parameters of the boards can be found in Table 1 in the experimental section.

Density measurements

Test specimens of 50x50 mm of the boards were sawn and the density was measured. The aimed density of the boards was 750 kg/m^3 and the average measured densities are shown in Table 2.

Table 2: Results of density measurements on OSB:s

Binder	Density, boards of unmodified wood [kg/m^3]	Density, boards of acetylated wood [kg/m^3]
Biorez	759 ± 15	820 ± 22
Protein adhesive	797 ± 57	826 ± 34
Phenol-formaldehyde	877 ± 53	-

As can be seen in Table 2, the achieved densities are all higher than the target level of 750 kg/m^3 and they show some variations (759-826 kg/m^3), which might be due to the possibility that some of the resin solvent (methanol or water) evaporated during the spraying and / or a result of the hand-forming procedure. Due to difficulties during the application of the protein adhesive, depending on an initial too high viscosity, an unknown mass of the protein binder was lost; however, this seems to be not reflected in the measured densities. The density profile of the boards was measured and it could be observed that the density was larger in the centre parts of the boards, which is likely an effect of the hand forming procedure. The density profiles of selected boards with the furan binder and the protein binder are shown in Figure 1.

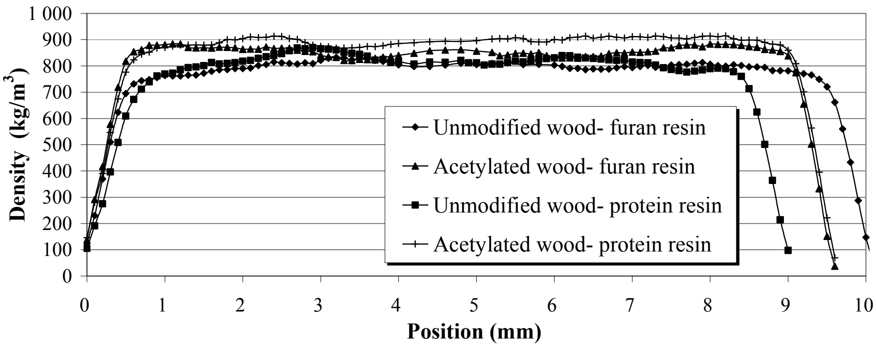

Figure 1. Density profiles of the boards produced using Biorez and the protein adhesive

Thickness swelling

The swelling tests are very revealing due to the large differences in swelling between the unmodified wood and the acetylated wood that was observed. The percentage swelling after soak in water during 24 h (20 ºC) is displayed in Table 3 and the boards after swelling are shown in Figure 2. The boards produced from the unmodified wood show high degrees of swelling for all the binders (exceeds 30%). The boards of unmodified wood produced using the protein adhesive could not withstand the water soak and these boards disintegrated during the test. However, the boards produced from acetylated wood show 5-8 times lower swelling than the unmodified ones. The Biorez and the protein adhesive both show very low swelling with just a few percent. The higher dimensional stability of the boards is certainly due to the acetylation procedure, since this type of modified wood is much less hydrophilic than unmodified wood and resists water absorption to a larger extent (Rowell *et al.* 1986). This behaviour is very important for use of OSBs in exterior applications, since a low moisture uptake is related to a slower decay induced by microorganisms.

Table 3: Results of swelling measurements on OSB:s

Binder	**Swelling, boards of unmodified wood [%]**	**Swelling, boards of acetylated wood [%]**
Biorez	33 ± 2	3.9 ± 0.5
Protein adhesive	[a]	6.4 ± 1.0
Phenol-formaldehyde	34 ± 3	-

[a]*The boards disintegrated in water*

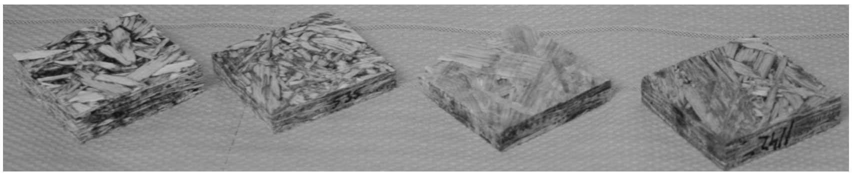

Figure 2: Typical samples after swelling test (from left to right): Unmodified wood - Biorez binder, acetylated wood - Biorez binder, acetylated wood - protein binder, and unmodified wood - PF binder

Internal Bond Strength

The internal bond strength (IB) was evaluated according to EN 319, which gives the tensile strength perpendicular to the plane of the board. The results of the measurements for conditioned samples, samples boiled in water, and samples that were reconditioned after water soak are displayed in Table 4.

Table 4: Results of internal bond strength (IB) evaluation on OSB:s

Binder	**IB, boards of unmodified wood [N/mm^2]**			**IB, boards of acetylated wood [N/mm^2]**		
	Conditioned	After boiling	After water soak	Conditioned	After boiling	After water soak
Biorez	0.80	0.21	0.57	0.59	0.10	0.56
Protein adhesive	0.11	[a]	[a]	0.37	[a]	0.15
Phenol-formaldehyde	0.85	0.28	0.67	-	-	-

[a]*The boards disintegrated in water*

Conditioned samples

In Table 4 it can be seen that the highest IB for the conditioned boards, 0.85 N/mm^2, is obtained when the PF binder is used for unmodified wood strands (used as control). This is not unexpected and this binder is commonly used in commercial boards. Also, the Biorez binder used for unmodified wood strands is performing well and shows an IB of 0.80 N/mm^2. However, the protein adhesive gave an extremely low IB of 0.11 N/mm^2 for the unmodified wood boards, which could be partly due to the loss of protein binder during the resination. It could also be partially attributed to a lack of cross-linking within the glue matrix, perhaps caused by a low level of isocyanate de-blocking during the hot-pressing process. This low IB is still somewhat expected, since the protein hydrolysate was designed for the more hydrophobic, acetylated boards, though such a large effect could not be predicted. In the case of the acetylated boards, a decrease in strength for the Biorez bonded boards compared to the unmodified board can be observed. It is possible that another type of Biorez, having a more hydrophobic character, would show improved wetting of the acetylated boards and perform better in the IB evaluation. However, the IB of the protein hydrolysate binder is significantly higher, 0.37 N/mm^2, and passes the demands of EN 300, which requires 0.32 N/mm^2 for OSB 3. The increased IB is in accordance with what would be expected for more hydrophobic wood strands and the protein hydrolysate. For the acetylated boards with the protein binder it is possible that the inherent acidity of the acetylated wood strands can have accelerated the cross-linking of the diisocyanate, resulting in a higher degree of cross-linking and thereby a higher IB. However, this hypothesis needs further investigation.

Boiled samples

The boards made from unmodified wood subjected to boiling kept some of their strength, except for the protein adhesive boards, which disintegrated during boiling. The acetylated boards lost nearly all strength and the boards made from the protein binder were completely destroyed during boiling. It has earlier been reported that boards made from acetylated fibres were significantly reduced in IB after boiling (Korai 2001) and this was explained by a low bondability of acetylated fibres. However, novolak PF resin bonded fibreboards from acetylated fibres retained almost all their strength after a boil test, unlike the boards from unmodified fibres, demonstrating the importance of choosing a proper resin with good wetting of acetylated wood surfaces (Westin 1998).

Water-soaked samples

It is noteworthy that the acetylated boards with furan binder virtually retains its IB after soak in water and reconditioning as it changes from 0.59 to 0.56 N/mm^2 (Table 4). As a comparison, the boards with unmodified wood and the furan binder loses 29% of its strength after water soak and reconditioning. However, the acetylated board with the protein binder loses 59% of its IB after water soak and reconditioning, but is still intact after the procedure. This implies that there is a large difference in IB reduction if the protein binder is subjected to hot or cold water, which probably could depend on that secondary chemical bonds in the protein binder break when a high temperature is applied, but they are unaffected at ambient temperature.

Decay resistance (Mini-block test)

The final results can not yet be reported. However, visual inspection indicates that the acetylated boards are not decayed by the brown rot fungi whereas the unmodified boards are severely decayed. The full results will be presented during the conference.

CONCLUSIONS

Oriented strand boards (OSB) of unmodified wood and acetylated wood strands could be obtained with the use of two biobased resins, a furan-type binder and a cereal protein-based binder. The OSBs made from acetylated wood strands showed improved dimensional stability, exhibiting significantly lower thickness swelling (5-8 times lower), than the boards made from unmodified wood. The furan-type binder-based OSB showed an internal bond strength (IB) of 0.80 N/mm^2 for unmodified wood-based OSB, which is comparable to the performance of the phenol-formaldehyde-glued control, and 0.59 N/mm^2 for the acetylated ones. The IB of the protein-glued acetylated OSB was lower, but fulfilled the criterion in EN 300 for OSB 3 of 0.32 N/mm^2. The unmodified wood board glued with the protein binder was very weak and disintegrated during water soak.

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REFERENCES

Bravery, A.F. (1979). A miniaturised wood-block test for the rapid evaluation of wood preservative fungicides. *Swedish Wood Preservation Institute*, Stockholm, p 136

Effendi, A., Gerhauser, H. and Bridgwater, A.V. (2007). Production of renewable phenolic resins by thermochemical conversion of biomass: A review. *Renewable and Sustainable Energy reviews*, **12**, 2092–2116.

EN 113 Wood preservatives – Test method for determining the protective effectiveness against wood destroying basidiomycetes – Determination of the toxic values

EN 319 Particleboards and fibreboards – Determination of tensile strength perpendicular to the plane of the board

Gomez-Bueso, J., Westin, M., Torgilsson, R., Olesen, P.O. and Simonson, R. (1999). Composites made from acetylated lignocellulosic fibers of different origin. *Holz als Roh- und Werkstoff*, **57**, 433-438.

Korai, H. (2001). Effects of low bondability of acetylated fibers on mechanical properties and dimensional stability of fibreboard. *Journal of Wood Science*, **47**, 430-436.

Larsson-Brelid, P. (1998). Acetylation of solid wood – wood properties and process development. *PhD Thesis, Chalmers University of Technology, Gothenburg, Sweden.*

Lin-Gibson, S., Walls, H.J., Kennedy, S.B. and Welsh, E.R. (2003). Reaction kinetics and gel properties of blocked diisocyanate crosslinked chitosan hydrogels. *Carbohydrate Polymers*, **54**, 193-199.

Papadopoulos, A.N., Ntalos, G.A.. Soutsas, K., Tantos, V. (2006). Bonding behaviour of chemically modified wood particles for board production. *Holz als Roh- und Werkstoff*, **64**, 21-23.

Papadopoulos, A.N. (2006). Decay resistance in ground stake test of acetylated OSB. *Holz als Roh- und Werkstoff*, **64**, 245-246.

Paul, W., Ohlmeyer, M., Leithoff, H., Boonstra, M.J., Pizzi, A. (2006). Optimising the properties of OSB by a one-step heat pre-treatment process. *Holz als Roh- und Werkstoff*, **64**, 227-234.

Pizzi, A. (2006). Recent developments in eco-efficient bio-based adhesives for wood bonding: opportunities and issues. *Journal of Adhesion Science and Technology*, **20**(8), 829-846.

Rowell, R., Tilman, A.M. and Simonson, R. (1986). A simplified procedure for the acetylation of hardwood and softwood flakes for flakeboard production. *Journal of Wood Chemistry and Technology*, **6**(3), 427-448.

Rowell, R.M., Youngquist, J.A. and Sachs, I.B. (1987). Adhesive bonding of acetylated aspen flakes, Part 1. Surfaces changes, hydrophobicity, adhesive penetration and strength. *International Journal of Adhesion and Adhesives*, **4**, 183-188.

Sun, X. and Bian, K. (1999). Shear strength and water resistance of modified soy protein adhesives. *Journal of the American Oil Chemists Society*, **76**, 977-980.

Westin, M. (1998). High-performance composites from modified wood fiber. *PhD Thesis, Chalmers University of Technology, Gothenburg, Sweden.*