The Effect of Acetylation on Heat Transfer Mechanism During Hot Pressing and Mechanical Properties of Wood Based Composites

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Heat transfer is one of the most important factors in wood based composite hot pressing. Acetylation have been utilized to improve wood composites properties. Present research was conducted to investigate the effect of acetylation on heat transfer from the press plates to particleboard mat and the mechanical properties of wood based composites. After 12h soaking in acetic anhydride, particles were heated in oven at 1200C for 60 and 180 min, in order to achieve two levels of weight percentage gain, 7 and 18% respectively. During hot pressing, variation of the temperature was measured with thermocouples in the thickness of the mat. The particleboards were subjected to mechanical testing. Results of current study revealed that the acetylation significantly affected the heat transfer in core layer of the sample boards. However the surface layer was not influenced by the acetylation. The core layers reached lower temperature than the surface layers. Also, results of mechanical testing indicate that the modulus of rupture (MOR) and the modulus of elasticity (MOE) of the specimens were decreased due to the particles acetylation. Generally, Internal bonds (IB) were higher for untreated boards than acetylated boards. It was concluded that acetylation reduced the flow of water vapor through the porous structure of the higher density board, and consequently reduced the rate of heat transfer. In the case of acetylated particleboard mechanical properties, the majority of failure was due to the resin and not to the wood. There by, the acetylated chips required longer time to achieve higher temperatures.

Keywords: Acetylation; Heat transfer; Hot-Pressing; Particleboard; Populus nigra

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INTRODUCTION

Many research groups have concentrated on improving the properties of particleboards from various raw materials. Among this researches, acetylation as a chemical modification technique has been used to enhance some properties especially dimensional stability (Rowell, 1975). This process increases the bulk density of wood (Sander et al., 2003), and makes it hydrophobic toward moisture which is necessary for biodeterioration (Takahashi 1996; Mohebby and Militz 2002; Brield and Westin 2007; Mohebby 2003) and dimensional changes (Youngquist et al. 1986; Gomes-Bueso et al. 1999). However, acetylation deteriorates mechanical properties of particleboard such as modulus of rupture, elasticity and internal bonding (Youngquist et al. 1986; Khosravani 2006; Rowell 1996; Fuwape and Oyagade 2000; Mahlberg et al. 2001). There are several arguments to explain the reasons of mechanical properties deterioration caused by the acetylation; e.g. type of the adhesive (Vick et al. 1991), less press pressure (Rowell 1996; Rowell et al. 1989; Rowell et al. 1988) and springback after conditioning (Mohebby et al. 2009) because of the debonding of the wood elements and stress relief (River 1994).

In addition, woods would be less permeable to the water by acetylation (Mohebby and Hadjhassani 2008; Kays and Crawford 1993). Therefore, it is possible that during pressing time the heat transfer to the panel core for resin curing changes in accordance with acetylation level.

To our knowledge, there is no report about heat transfer mechanism during hot pressing of acetylated particles. Therefore, this research was conducted to investigate the effect of acetylation on heat transfer from hot press plates to the surface and core layers of wood acetylated particle mat made with the acetylated wood particles. Because of the access problems to acetic anhydride, and due to the fact that previous research projects proved the effectiveness of this modification near to these levels (Mohebby et al. 2009), we decided not to use of higher WPG.

EXPERIMENTAL

Acetylation

Chips were prepared using a laboratory chipper from poplar wood (*Populus nigra*) and dried in an oven for 24 h at 103± 2˚C. The oven dried chips were reacted with acetic anhydride at 120˚C to achieve different weight percent gains (WPGs). Acetylation of chips was done by soaking them in the acetic anhydride solution for 12h. Then after evacuation of solution, the chips were heated to 120˚C for 60 and 180 minutes in acetylation reaction vessel. Treated chips were washed in distilled water to remove excess acetic acid, the reaction by products, and un-reacted acetic anhydride. Acetylated wood particles were oven dried for 24h. Weight percent gains (WPG) were calculated according to Eq. 1. The WPG of 7 and 18% were reached in acetylated chips.

Eq. 1 $$WPG= [(W_{act} - W_{unt})/ W_{unt}] \times 100$$

Where WPG indicates the weight percent gain (%), Wact and Wunt are oven dry weights after and before the acetylation (g), respectively.

Determination of heat transfer

Treated and untreated chips were blended with urea formaldehyde (UF) resin (10% based on the dry weight) and transferred into a mold to prepare the mat. Nominal dimension of specimens was 400×400×15 mm with 5 replications for each treatment. Three thermocouples were inserted into the mat at 5, 25 and 50% of the board thickness from top surface (Fig. 1). Mat was placed in a laboratory press and compressed applying 30 bar pressure for 5min at temperature of 175˚C. Target density of the boards was 0.75g/cm^3. Measured temperatures by the thermocouples were recorded from ambient up to the target pressing temperature (175˚C) for each board, at 30 seconds intervals after the press closure.

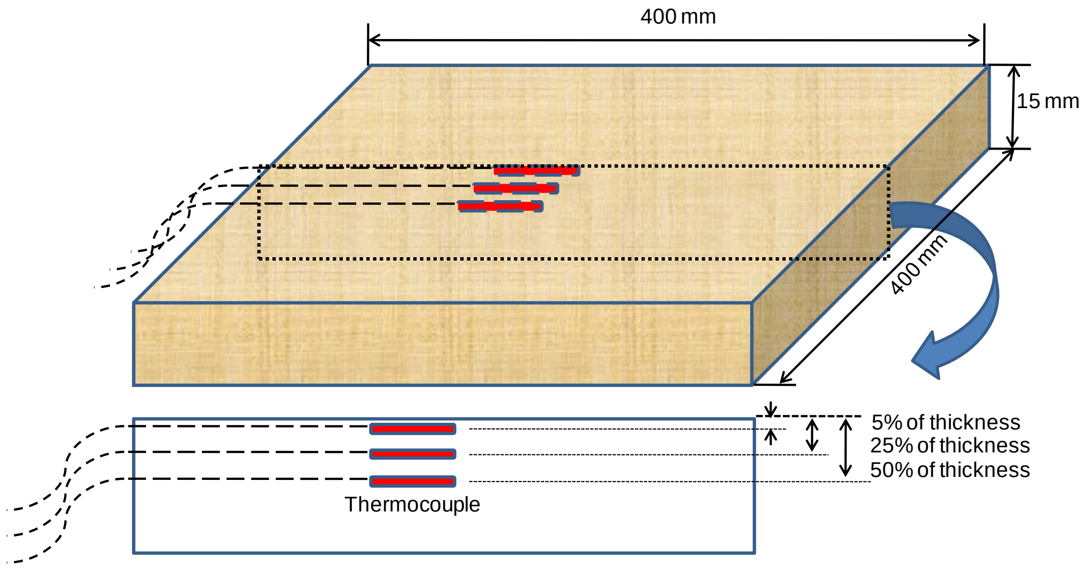

Fig. 1. Position of thermocouples in board's mat**.**

Mechanical test

Prior to the mechanical test, the boards were conditioned at 65% relative humidity (RH) and 20±2˚**C** for two weeks until the panels reached equilibrium moisture content.

A static bending test was performed on fifteen specimens of each treatment level. The specimens' size was 50×150×4 mm. The distance between the supports was 100 mm; loading speed was 10 mm/min. The modulus of rupture (MOR) and the modulus of elasticity (MOE) were calculated according to European Standard EN 310. The internal bond (IB) strength of the test boards were determined according to the European Standard EN 319 on at least fifteen specimens of each treatment level (three specimens from five individual boards). Specimen size was 50×50×4 mm.

RESULTS AND DISCUSSION

Heat transfer

There were no significant differences between surface layer temperatures of the boards with different WPG (Fig. 2). However, as the WPG rose in the mat, the temperature of core layer decreased (Fig. 3). At the end of the pressing time (300s), the core layer temperatures of the untreated boards were 103˚C; while, when the particles with 18% WPG were used, it was 98˚C.

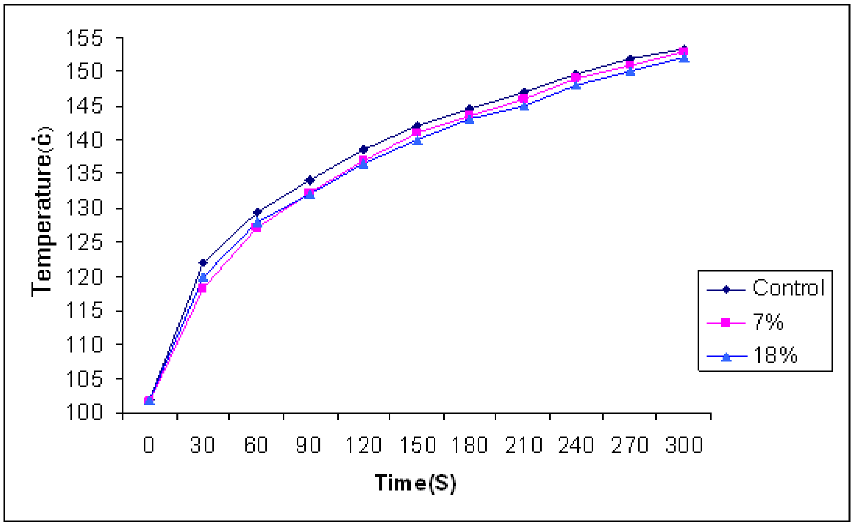

Fig. 2. Board's surface temperature variation versus press time**.**

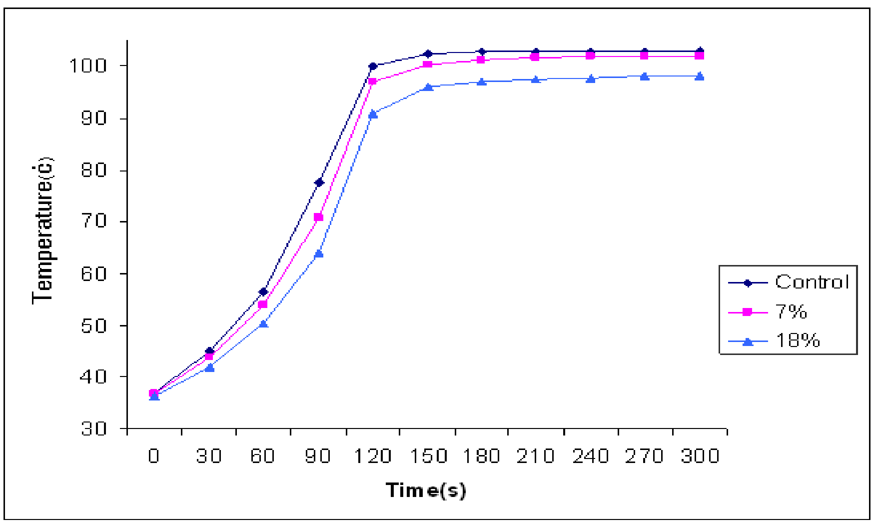

Fig. 3. Board's core layer temperature variation versus press time**.**

At the surface layer, temperature increases as a quadratic model; while due to the hot steam bulk flow, variation of the core layer temperature followed as a cubic model (Table 1). As the board's surface layers were in direct contact with the hot plates, by conductive mechanism they heated up rapidly (Fig. 2). However, due to the position of the core layer, with a delay, its temperature increased in three steps (Fig. 3). First, during the initial 60 seconds of the press time the temperature increased very slowly. Then, the temperature jumped to higher level during the period of 60-120s. Finally, the temperature stabilized and reached the highest possible level based on the press time (5min). The core layer was heated by diffusion vapor from the surface layers. Since, the wood moisture evaporated and migrated from the surface to the middle layers of the mat after the press closure.

Table 1. Fitted models for board's temperatures versus press time in acetylated particleboard.

Layer	WPG (%)	b0	b1	b2	b3	Model: Quadratic $Y=b0+b1X+b2X^2$
Surface	0	108.192	0.322	-0.0001	-	$Y = 108.192 + 0.322X - 0.0001X^2$
	7	106.415	0.321	-0.0001	-	$Y = 106.415 + 0.321X - 0.0001 X^2$
	18	107.509	0.306	-0.0001	-	$Y = 107.509 + 0.306X - 0.0001X^2$
						Cubic $Y = b0 + b1X + b2X^2 + b3X^3$
Core	0	31.363	0.651	-0.01	-1.518E-07	$Y = 31.363 + 0.65X - 0.013X2 - 0.0000001X^3$
	7	31.620	0.548	-0.0001	-1.6E-06	$Y = 31.620 + 0.54X - 0.0001X2 - 0.000001X^3$
	18	31.658	0.440	-0.00004	-2.72E-06	$Y = 31.658 + 0.44X - 0.00004X2 - 0.000002X^3$

Y: Temperature (°C); X: Press time (s)

Variation of the core layer temperature versus the WPG indicated a reverse correlation (Fig. 4). It has been indicated that higher WPG reduce core temperature significantly (R^2=0.74). It is likely that the acetylation affected the heat transfer from the surface to the core layer.

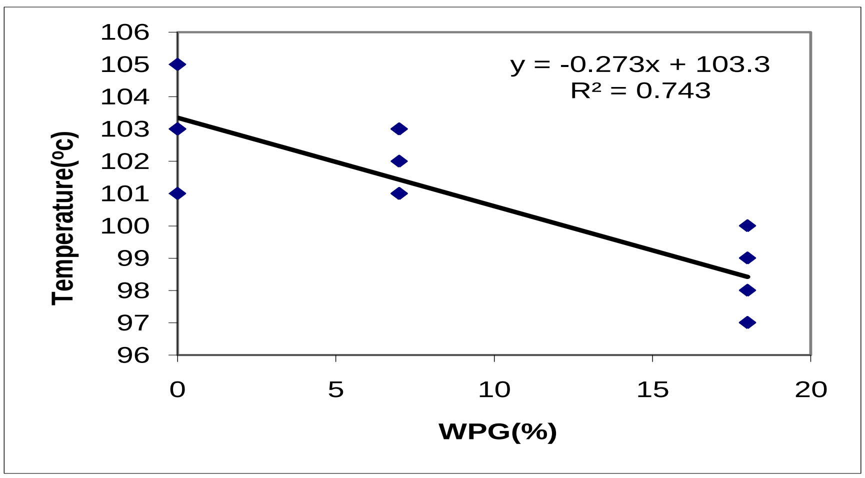

Fig. 4. The effect of the acetylation level on the final temperature in the core layer**.**

Mechanical properties

The mechanical properties values of the experimental particleboards made from acetylated wood chips are summarized in Table 2. The statistical analysis proved that significant changes ($P<0.05$) due to acetylation took place in the modulus of rupture. The bending strength values of the acetylated boards were 81% (The lower acetylation level) and 28% (The higher acetylation level) of control board values.

Table 2. Average values of mechanical properties of acetylated and control aspen particleboard.

Type of particles	MOR(MPa)	MOE(MPa)	IB(MPa)
Untreated	20.93(A)	2501.7(A)	1.14(A)
Acetylated (7%)	17.15(B)	2297.5(A)	0.71(B)
Acetylated (18%)	5.90(C)	785.9(B)	0.043(C)

Acetylation level of 18% showed significant decreasing effect on MOE (Table 2). The 7% level treatment, although lower in value than the control samples, didn't show significant decrease ($P>0.05$).

From Table 2, it can be seen that acetylation of chips resulted in significant decrease in internal bonding strength. These values for the modified particleboards in low and high acetylation level were about 0.60 and 0.04% of untreated values, respectively.

Discussion

Results of current study showed that acetylation reduced the temperature of the core layer to the level lower than 100°C, which is required for curing of the UF resin (Hood 2004). Hence, this modification affects the mechanical properties of the boards. Reduction of the mechanical strengths of acetylated wood based composites (Khosravani 2006; Rowell 1996; Fuwape and Oyagade 2000; Mahlberg et al. 2001; Fuwape and Oyagade 2000; Chow et al. 1996; Papadopoulos and Traboulay 2002; Dreher et al. 1964) because the hydrophobic nature of the modified substrate and poor adhesive penetration, might be related to the incomplete curing of the resin in the core layer, too. It was reported that failure in control boards occurred in the wood, whereas in the acetylated boards, the majority of failure was due to the resin and not to wood (Papadopoulos and Traboulay 2002).

The heat transfer is occurred due to variety of mechanisms, but generally it is believed that conduction and convection are the most important ones (Rowell 2005). The conductive mechanism takes place in wood cell wall materials; while, the convective takes place through the cell cavities and the void spaces between the wood particles. The rate of heat transfer is important, because the adhesives mostly require curing temperature above 100˚C for the polymerization. If the resin is not fully cured during the process, the board could debond due to expanding internal vapor pressure (Hood 2004). During the hot-pressing, steam generated from bound water in the wood particles and the water in the resin is driven to the core layer of the mat (Vick and Rowell 1990), and the thermosetting adhesives are polymerized throughout the panel. Rate of the heat transfer to the panel core is controlled by several factors such as particle geometry, moisture content, density, permeability etc.

The mat moisture content is an extremely critical factor for the total press time and board formation. Reports indicate that, surface moisture evaporates and migrates to the mat, which causes the heat transfer to the core (convection), and allows the adhesive to react more quickly than heat moved by conduction through the wood and air spaces (Kelly 1977). It appears that mass flow of the water vapor (convection) is responsible for the initial increase in the heating rate of the core (Kayihan and Johnson 1983). Increasing the moisture content of mat, will raise the pressure gradient, which results in an accelerated rate of convection heat transfer by vapor flow (Zombori 2001). Acetylation affects hydrophilicity of the lignocellulosic material and alters it to hydrophobic nature (Militz and Beckers 1994). The conductive mechanism through the wood cell wall materials is excluded. Also, in the acetylated boards, it might be expressed that surface layers moisture reduced due to higher WPG and caused lower vapor migration and diffusion through the voids and cavities to the core layers. Volume of water vapor as well as its temperature affects the temperature of the core layer directly.

Density is directly related to the available void spaces in the mat, and influences the tortuosity of flow path (Garcia 2002). Higher densities reduce flow of the water vapor through the porous structure of the mat, consequently reducing the rate of the conductive heat transfer (Zombori 2001). Rate of the convective heat transfer to the panel core is controlled by its permeability. Permeability in the panel also controls the flow of vapor to the panel edges, thereby influencing the potential for panel "blowing" (Hood 2004). Therefore, permeability in the acetylated wood might be decreased, because acetylated materials are denser (Mohebby et al. 2009) and bulkier (Sander et al. 2003). This might be another reason for lower volume of water vapor bulk flow from the surface layer to the core layer that resulted in reduction of the temperature in the core layer.

Indeed, acetylation increases density (Kumar et al. 1979) and bulk and decreases moisture content, porosity and permeability. Because higher density reduces the flow of water vapor through the porous structure of the board, the rate of the heat transfers is consequently reduced (Zombori 2001). Thereby, the acetylated chips required longer time to achieve higher temperatures. Therefore, the press time (300s) was not sufficient to reach sufficient temperature at the core layer.

Other press time with different treatment level per wood particles, as well as their effects on the other aspects of composite boards, are yet to be studied in further studies to come to a final conclusion on the optimum press time for each level treatment with regard to the all properties of wood based composites.

CONCLUSIONS

Results of current research showed that:

1. Heat transfer follows two different models in surface and core layers of the particle board mat.
2. Increase in weight gains due to the acetylation does not affect the heat transfer in the surface layer; while the core layer's temperature was decreased as the WPG increased.
3. Acetylation resulted in a remarkable loss in modulus of rupture, elasticity and internal bonding values for the poplar particleboard.

REFERENCES CITED

Brield, PL and Westin, M. (2007). “Acetylated wood–results from long- term field tests,” In: Proceedings of the European Conference on Wood Modification, Pp. 71–78.

Chow, P. Boo, Z, Youngquist, J. A., Rowell R. M., Muehl, J. H., Krzysisk, A. M. (1996). "Properties of hardboards made from acetylated Aspen and southern pine," *Wood fiber Science* 20, 252-258.
Dreher, W. A., Goldstein, I. S., Cramer, G. R. (1964). "Mechanical properties of acetylated wood," *Forest products Journal* 14, 66–68.
Fuwape, J. A. and Oyagade, A. O. (2000). "Strength and dimensional stability of acetylated Gmelina and Sitka spruce particleboard," *Tropical Forest Prod*. 6, 184-189.
Fuwape, J. A and Oyagade, A. O. (2000). "Strength and dimensional stability of acetylated Gamelina and spruce particleboard," *Journal of Tropical Forest Products* 6, 184–189.
Garcia, P. (2002). "Three-dimensional heat and mass transfer during oriented strandboard hot-pressing," *Ph.D. Thesis, University of British Columbia, Vancouver.*
Gomes-bueso, J., Westin, M., Torgilsson, R., Olesen, P. O., Simonson, R. (1999). "Composites made from acetylated lignocellulosic fibers of different origin. Part II: The effect of nonwoven fiber mat composition upon molding ability," *Holzals Roh-und Werkstoff* 57, 178-184.
Hood, J. P. (2004). "Changes in oriented strand board permeability during Hot-pressing," *M.Sc. Thesis, Virginia polytechnic Institute and state University, Blacksburg.*
Kavihan, F. and Johnson, J. A. (1983). "Heat and moisture movement in wood composite materials during the press operation- a simplified model. Numerical methods in heat transfer," Volume II. John Wiley & Sons, Pp.511-531.
Kavs, M. W. and Crawford M. E. (1993). Convective heat and mass transfer. McGraw-Hill, New York, Pp.319.
Kelly M. W. (1977). "Critical literature review of relationships between processing parameters and physical properties of particleboard," Forest Products Laboratory, Madison. Wisconsin, Pp. 64.
Khosravani, Y. (2006). "Effect of acetylation on physical and mechanical properties of particleboard," M. Sc. Thesis, Tehran University.
Kumar, S., Singh, S. P. and Sharma, M. (1979). "Effect of acetylation on permeability of wood." *Journals of Timber Development Association* 25, 5–9.
Mahlberg, R., Paajanen, L., Nurmi, A., Kivisto, A. and Rowell, R. M. (2001). "Effect of chemical modification of wood on the mechanical and adhesion properties of wood fiber/polypropylene fiber and polypropylene/veneer composites," Holz als Rohund Werkstoff, 59, 319–326.
Militz, H., Beckers, E. P. (1994). "Process for acetylation solid wood," European patent Application, 85850268.5.
Mohebby, B., Ghorbani-Kokandeh, M and Soltani, M. (2009). "Springback in acetylated wood based composites," Construction Building Materials 23, 3103-3106.
Mohebby, B. and Militz, H. (2002). "Soft rot decay in acetylated wood: Microcalorimetry: Chemical and anatomical changes in decayed wood," *International Research group on wood preservation,* IRG/WP 02-40231.
Mohebby, B. (2003). "Biological attack of acetylated wood," *Ph.D. Thesis, Göttingen University, Göttingen, Germany*,167.

Mohhebby, B. and Hadjhassani, R. (2008). "Moisture repellent effect of acetylation on poplar fibers," *Journal of Agricultural Science Technology* 10, 157–163.

Papadopoulos, A. N., and Traboulay, E. (2002). "Dimensional stability of OSB made from acetylated fir strands," *Holz Roh- Werkstoff* 60, 84-87.

River, B. H. (1994). "Outdoor aging of wood-based panels and correlation with laboratory aging," *Forest Products Journal* 44, 55–65.

Rowell, R. M. (2005). Chemical modification of wood. USDA, Forest Service, Forest products Laboratory and Department of Biological Systems Engineering. Pp. 487, University of Wisconsin, Madison, WI. Handbook of wood chemistry and wood composites.

Rowell, R. M., Kawai, S. I. and Norimoto, M. (1989). "Dimensional stability, decay resistance and mechanical properties of veneer-faced low-density particleboards made from acetylated wood," *Wood Fiber Science* 21, 67–79.

Rowell, R. M., Youngquist, J. A. and Imamura, Y. (1988). "Strength test on acetylated aspen flakeboards exposed to a brown rot fungus," *Wood Fiber Science* 20, 266–271.

Rowell, R. M. (1975). "Chemical modification of wood: Advantages and disadvantages," Pp. 1-10 in Proceedings of American Wood-Preservation Association, 71.

Rowell, R. M. (1996). Physical and mechanical properties of chemically modified wood. In:Hon DNS, editor. Chemical modification of lignocellulosic materials, Pp. 295-310. New York,Marcel Dekker.

Sander, C., Beckers, E. P., Militz, H., and Veenendaal, W. (2003). "Analysis of acetylated wood by electron microscopy," *Wood Science and Technology* 37, 39-46.

Takahashi, M. (1996). Biological properties of chemically modified wood. In: Hon DNS, editor. Chemical modification of lignocellulosic materials; Marcel Dekker, New York, Pp. 339-369.

Vick, C. B. and Rowell, R. M. (1990). "Adhesive bonding of acetylated wood," *International Journal of Adhesion and Adhesives* 10, 263–272.

Vick, C. B., Krzysik, A. and Wood, J. E. (1991). "Acetylated, isocyanate-bonded flakeboards after accelerated aging: dimensional stability and mechanical properties," Holz als Roh- und Werkstoff,49:221–228.

Youngquist, J. A., Krzysik, A. and Rowell R. M. (1986). "Dimensional stability of aspen flakeboard," *Wood Fiber Science* 18, 90-98.

Zombori, B. G. (2001). "Modeling the transient effects during the Hot-pressing of wood-based composites," *Ph.D.* Thesis, Department of Wood Science and Forest products, Virginia Polytechnic Institute and State University, Blacksburg.

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