Holz als Roh- und Werkstoff 49 (1991) 221--228 Springer-Verlag 1991

Forschung und Praxis Holz als Roh- und Werkstoff

Acetylated, isocyanate-bonded flakeboards after accelerated aging

Dimensional stability and mechanical properties

C.B. Vick, A. Krzysik, and J.E. Wood Jr.

USDA Forest Service, Forest Products Laboratory, One Gifford Pinchot Drive, Madison, Wisconsin 53705-2398, USA

To improve flakeboard dimensional stability and mechanical properties for long-term exterior exposure, we fabricated and tested a highly durable isocyanate-bonded acetylated flakeboard. Flakeboards were tested for internal bond strength, thickness swell, modulus of elasticity, modulus of rupture, and breaking strength in bending, after two severe accelerated aging tests. Flake design, flakeboard density, resin content, and their interactions were critical to the performance of acetylated flakeboards in the aging tests. In bending tests, the longer disc-cut flakes produced much higher strength values than did the ring-cut flakes. In internal bond strength tests, the ring-cut flakes performed comparably to the disc-cut flakes. However,. ring-cut flakes contributed somewhat more to reducing thickness swell than did disc-cut flakes. Acetylated flakeboards fabricated at high density were much stronger than those fabricated at low density. However, the high-density boards developed more thickness swell than did the low-density boards. The 5-% resin content contributed little to increased strength values in the bending tests, but this level of resin significantly improved flakeboard performance in internal bond strength and thickness swell tests. Acetylation did not interfere to any practical degree with the isocyanate resin, and all property values were enhanced by acetylation, particularly internal bond strength and resistance to thickness swell.

Acetylierte, Isocyanat-gebundene Spanplatten nach beschleunigter Alterung: Dimensionsstabilität und mechanische Eigenschaften

Um die Dimensionsstabilität und die mechanischen Eigenschaften fur die Außenanwendung zu verbessern, wurden Isocyanat-gebundene Spanplatten aus acetylierten Spänen hergestellt. Getestet wurden Querzugfestigkeit, Dickenquellung, Elastizätsmodul, Bruchspannung und Biegefestigkeit nach zweimaliger beschleunigter Alterung. Spandimensionen, Plattendichte und Festharzanteil erwiesen sich als die wichtigsten Einflußgrößen für das Testverhalten bei der beschleunigten Alterung. Späne aus dem Flachscheiben-Zerspaner führten zu höheren Festigkeitswerten als Späne nach Messerring-Zerspanung, während die Querzugfestigkeit bei beiden Spantypen vergleichbar war. Die Späne aus dem Ringzerspaner hatten jedoch einen größeren Einfluß auf die Reduzierung der Dickenquellung. Hochverdichtete Platten wiesen höhere Festigkeitswerte auf als Platten geringerer Dichte, allerdings war bei ihnen gleichzeitig die Dikkenquellung ausgeprägter. Der Festharzanteil von 5% trug wenig dazu bei, die Biegefestigkeit zu erhöhen, verbesserte aber wesentlich Dickenquellung und Querzugfestigkeit. Die Acetylierung hatte keine erkennbare Störung der Isocyanat-Verleimung zur Folge. Alle Plattenkennwerte wurden durch die Acetlierung verbessert, insbesondere Querzugfestigkeit und Dickenquellung.

Introduction

Wood composite panel products will command a greater share of the sheathing and siding market in the 1990s when they meet ever-increasing consumer demands for higher quality and better performance. The fabrication of composite panels at moisture levels much less than those experienced in service results in excessive expansion (Wellons 1990). Improved dimensional stability could minimize consumer complaints of excessive thickness swell at panel edges and of buckling from excessive linear expansion. Acetylation of wood flakes with acetic anhydride greatly improves the dimensional stability of aspen flakeboards as indicated by significant reductions in irrecoverable springback, thickness swell, and water absorption in water-immersion tests (Youngquist et al. 1986a, b). Although acetylated flakeboards are not completely resistant to subterranean termites, they have superior resistance to attack by brown-, white- and soft-rot fungi and tunneling bacteria (Rowell et al. 1987a; Imamura et al. 1987). Flakes can now be acetylated with acetic anhydride by a simple dip procedure that requires neither cosolvent nor catalyst and has a shortened reaction time of 2 to 4 h (Rowell et al. 1986).

Acetylation generally interferes with the adhesion of most aqueous-based adhesives used to bond wood, depending on the level of acetylation, and high-temperature-curing phenol-formaldehyde (PF) resins are particularly limited (Vick and Rowell 1990). In humidity and water-immersion soak tests, acetylated flakeboards made with aspen flakes and bonded with PF resin (6% resin solids) had only one-sixth and one-seventh the thickness swell, respectively, of unacetylated flakeboards made with aspen (Youngquist 1986 a). Mechanical tests on these flakeboards after accelerated aging indicated that acetylation reduced initial internal bond strength and modulus of rupture by 90% and 80%, respectively (Youngquist et al. 1986 b). A scanning electron microscopic study indicated that decreased wetting, flow, and penetration of acetylated flakes by PF resin were the causes of poor adhesion (Rowell et al. 1987). An isocyanate proved to be a better adhesive for acetylated aspen flakes (3% resin) than did PF resin (5% resin solids), in terms of less irreversible thickness swell in both liquid water soak and water vapor tests. However, isocyanate-bonded flakeboards had only slight improvements in internal bond strength, modulus of rupture, and modulus of elasticity when compared with these properties of PF-bonded flakeboards. Mechanical tests were not conducted on isocyanate-bonded acetylated flakeboards that had been subjected to accelerated aging tests [Youngquist and Rowell (in press)].

We know from the previously mentioned research that acetylation of flakes sharply reduces irreversible thickness swell. Our preliminary research indicated that resin type and amount, flake design and thickness, and flakeboard density can further improve flakeboard dimensional stability and mechanical properties, particularly for withstanding severe accelerated aging. The purpose of this study was to fabricate a highly durable flakeboard for exterior service taht would remain dimensionally stable and decay resistant and retain high levels of strength properties and surface integrity for long-term exposure.

2 Materials

2.1 Adhesive

The adhesive was a polymeric diphenylmethane diisocyanate (MDI) called Mondur E-541 (Mobay Chemical Corporation, Pittsburgh, Pennsylvania [1]. According to the technical literature, E-541 consists of 45% to 55% (weight basis) diphenylmethane diisocyanate and 45% to 55% of higher oligomers. It has a weight-averaged molecular weight of 350 and viscosity of 200 mPa s at 25° C.

2.2 Flakes

Half the flakes were ring-cut with a Pallman flaker from 19-mm-long pulp chips of quaking aspen. The ring-cut flakes were 0.25 to 0.30 mm thick, in random widths, and up to 19 mm long. The remaining flakes were disc-cut from 6.4-mm-thick, rotary-cut veneer of quaking aspen. The disc-cut flakes were 0.25 to 0.30 mm thick, 6.4 mm wide, and 38 mm long.

All flakes were dried to approximately 2 percent moisture content (MC) and then screened to remove fines £0.80 mm. We allowed the moisture content of control (unacetylated) flakes to increase to 5% to 6% before the flakes were blended.

2.3 Acetylation

Both ring-cut and disc-cut flakes wen: acetylated to approximately 17% weight grain, based on the original ovendry weight of untreated flakes. The acetylation procedure included soaking ovendried flakes in acetic anhydride for 3 h at atmospheric pressure, draining, then reacting at 120° C for approximately 3 h. After reaction, the flakes were dried for 24 h at 105° C. The acetylated flakes from all acetylation runs were blended before fabrication to obtain uniform distribution of acetyl and moisture content in all flakeboads. At the time of blending, the moisture content of the flakes increased to 2% to 3%.

3 Experimentalmethods

3.1 Experimental design

This experiment was designed to determine what combination of variables could produce an acetylated flakeboard of exceptional strength and durability capable of withstanding longterm exterior exposure. The variables were two flake designs, two MDI resin contents, and two flakeboard density levels. Effectiveness of adhesion of MDI to acetylated flakes at various combinations of fabrication variables were meansured in terms of breaking strength in bending (BSB), modulus of rupture (MOR), modulus of elasticity (MOE), internal bond strength (IB), and thickness swell (TS). These variables were measured before and after exposure of flakeboards to two severe accelerated aging tests: the American Plywood Association (APA)Test Method D-5 six-cycle test (APA 1988) and the American Society for Testing and Materials (ASTM) Test Method D1037 six-cycle test (ASTM 1987). The independent and dependent variables and their respective treatment combinations and test methods arc: listed in Table 1.

Table 1. Independent and dependent variable in experimental design

Independent variables	Levels of treatment
Flake design	Ring-cut pulp chips Disc-cut veneer
Resin content	3% 5%
Flakeboard density	672 kg/m^3 833 kg/m^3
Control	Disc-cut flakes 3-% resin content 672 kg/m^3 density
Dependent variables	**Levels of test**
Internal bond, breaking	Unaged
modulus of elasticity, modulus of rupture, and thickness swell	ASTM D1037 six-cycle test

The experimental design was completely randomized with a factorial arrangement of (2) flake designs × (2) MDI resin contents × (2) flakeboard density levels to yield eight treatment combinations. Each treatment combination was replicated five times. From each replicate, one observation was made for MOE and MOR for each of the three aging conditions: unaged, APA D-5 sic-cycle test, and ASTM D 1037 sic-cycle test. Two observations were made for IB, BSB, and TS from each replicate for all three test conditions. These two observations were averaged within each replicate. Forty acetylated flakeboards were fabricated for the eight combinations of treatments.

Previous research had shown the merits of acetylation; therefore, comparisons of mechanical and physical properties of acetylated and unacetylated flakeboards were not an interest in study. However, for reference and for convenience to the reader, a set of unacetylated controls was prepared to show how selected fabrication variables could improve certain properties when flakeboards were acetylated. Five replicates of unacetylated flakeboards were fabricated with a resin content of 3% , density of 672 kg/m^3, and disc-cut flakes. These unacetylated controls could bd not be included in the factorial experiment; however, they are shown as non-statistical comparisons in Figs. 1 through 5.

3.2 Flakeboard fabrication

Acetylated flakes at 2% to 3% MC and untreated flakes at 5% to 6% MC were sprayed and blended with MDI resin to the prescribed 3 or 5% resin content (based on ovendry weight of flakes). The resin-coated flakes were randomly oriented and hand-formed into 406-mm-square mats before loading into the hot-press. Acetylated and unacetylated

[1] The use of trade or firm names in this publication is for reader information and does not imply endorsement by the US Department of Agriculture of any products or service

flakeboards were made in density levels of 672 and 833 kg/m^3 and thickness of 12.7 mm. A fully automatic, programmable press with an automatic data acquisition system was used to press the flakeboards. The press was accurately controlled by an electrical, servo-activated pump, actuated by pressure and position-monitoring transducers. The pump was time-programmed to operate to specific press openings or pressures.

The press temperature was maintained at 177° C with the respective press closing times and press times of 45 s and 6 min, respectively, for the 672 kg/m^3 board and 90 s and 8 min, respectively, for the 833 kg/m^3 board. Pressures for making acetylated and unacetylated control flakeboards were 6.21 to 7.58 MPa for the 672-kg/m^3 board and 10.34 MPa for the 833-kg/m^3 acetylated board.

3.3 *Accelerated aging procedures*

ASTM Method D1037:
The six-cycle ASTM D1037 procedure consisted of the following events:

1. Immersion in water at 49° C for 1 h
2. Exposure to steam at 93° C for 3 h
3. Storage at - 12° C for 20 h
4. Heating at 99° C in dry air for 3 h
5. Exposure to steam at 93° C for 3 h
6. Heating in dry air at 99° C for 18 h

These events were repeated five times, thus constituting the six-cycle 'procedure.

APA Method D-5:
The six-cycle APA D-5 procedure consisted of the following events:

1. Immersion in a vacuum-pressure vessel in water at 66° C with a vacuum of 635 mm Hg for 30 min
2. Soaking under a pressure of 448 kPA for 30 min with no additional heating of water
3. Drying at 82° C with forced-air circulation for 6 h
4. Repeating events 1 and 2
5. Drying at 82° C with forced-air circulation for 15 h

These events were repeated two times, thus constituting the six-cycle APA test.

Test on unaged specimens:

All unaged specimens and those subjected to accelerated aging procedures were conditioned to equilibrium at 27° C and 65% relative humidity before physical measurements for mechanical tests were conducted.

3.4 *Specimen design and testing*

Breaking strength in bending:

Breaking strength in bending specimens was 25.4 mm wide, 127 mm long, and 12.7 mm thick. The breaking load of each specimen was tested as a beam on edge over a 101.6-mm span, as described in APA Test Method S-6 (APA 1988).

Moduli of rupture and elasticity:

The flakeboard had a 12.7-mm nominal thickness; therefore, according to ASTM Method D1037 (ASTM 1987), the static bending specimen must be 76 mm wide by 356 mm long. After accelerated aging and conditioning as previously described, specimens for MOE and MOR were tested in static bending according to ASTM Method D1037 (ASTM 1987).

Internal bond strength:

Internal bond specimens were 12.7 mm thick and 50.8 mm square. They were prepared and tested for IB strength according to ASTM Method D1037 (ASTM 1987). Because most specimens had exceptionally high IB strength values, it was necessary to use special surface preparation procedures to ensure that the bond between the IB specimens and the steel-loading blocks exceeded the IB strength values of the specimens.

Thickness swelling:

The MOE-MOR specimens were used for TS measurements. After appropriate conditioning, specimens were measured before and after accelerated aging tests. Measurements of TS were made along each of the four sides at a point midway along the side and 25 mm from the edge. The four measurements were averaged for each specimen.

4 Results and Discussion

4.1 *Breaking strength in bending*

Flake design, resin level, and density were highly significant main effects on the BSB of unaged specimens and on those specimens subjected to APA and ASTM accelerated aging tests. No significant interactions occurred within the aging tests (Table 2).

A comparison of average BSB values in Fig. 1 shows that disc-cut flakes (D) produced much higher breaking loads than did ring-cut flakes (R). The average increase in breaking load resulting from flake design over all treatment combinations (except unacetylated controls) was 48.1 kg in the unaged specimens, 44.4 kg in the APA-aged specimens, and 40.4 kg in the ASTM-aged specimens. The disc-cut flakes were longer and wider (38 mm by 6.4 mm), more uniform in size, and fewer in number than the ring-cut flakes, which were 19 mm long, were random width, and had a large number of small particles. Thus, the disc-cut flakes were more efficient in sharing and transferring the breaking loads within the flakeboard than were the smaller ring-cut flakes.

The high density level (833 kg/m^3) also produced much stronger flakeboards than did the low density level (672 kg/m^3), with across-treatment average increases of 40.4 kg in the unaged specimens, 41.3 kg in the APA-aged specimens, and 33.1 kg in the ASTM-aged specimens, (Fig. 1). Interestingly, bond strength deterioration, which normally accompanies springback in high-density flakeboards, was not apparent in the relatively steady 41.3- and 33.1-kg strength increases in the high-density flakeboards in both accelerated aging tests. These strength increases were comparable to the 40.4-kg strength increase in the unaged specimens.

Increasing resin content from 3% to 5% produced a relatively small increase in BSB values - 10.9 kg in the test on unaged specimens, 12.2 kg in the APA test, and 8.2 kg in the ASTM test. In terms of BSB, a resin content increase to 5% was not worthwhile.

Table 2. Effect of independent variables on mechanical properties and dimensional stability of unaged and aged acetylated flakeboardsa

Significance of interaction on various properties [a]

variable[b]	Breaking strength in bending			Modulus of rupture			Modulus of elasticity			Internal bond			Thickness swell	
	UNA	APA	ASTM	UNA	APA	ASTM	UNA	APA	ASTM	UNA	APA	ASTM	APA	ASTM
Flake design (ring-cut and disc-cut)	***	***	***	***	***	***	***	***	***	--	--	***	***	*
Resin content (3% and 5%)	***	***	***	***	***	***	***	***	***	***	***	***	***	***
Flakeboard density (low and high)	***	***	***	***	***	***	***	***	***	***	***	***	***	***
Ring-cut and disc-cut flakes × 3% and 5% resin contents	--	--	--	--	--	--	--	--	--	--	--	--	*	--
Ring-cut and disc-cut flakes low and high density	--	--	--	--	--	--	--	--	--	--	***	***	***	***
3% and 5% resin contents × low and high density	--	--	--	--	--	--	--	--	--	*	--	--	--	--
Ring-cut and disc-cut flakes × 3% and 5% resin contents × low and high density	--	--	--	--	--	--	--	--	--	--	*	**	--	--

[a] UNA is unaged; APA is APA D-5 six-cycled test; ASTM is ASTM D1037 six-cycle test.
[b] All sources of variation based on 1 degree of freedom.
[c] Results of ANOVA. Dash (--) indicates no level of significance. Asterisks indicate significance at varying levels of probability; * 0.05, ** 0/01, and *** 0/001.

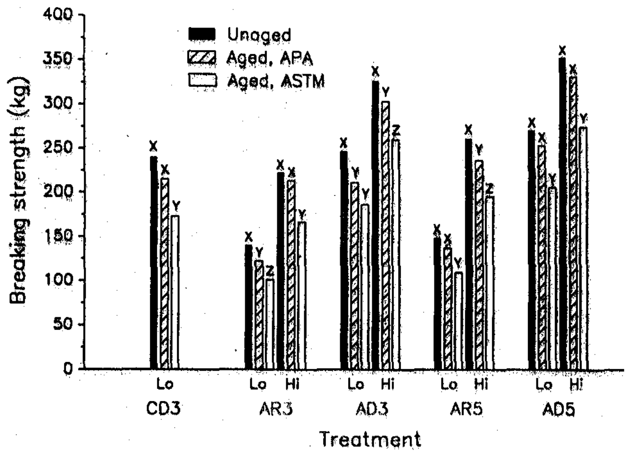

Fig. 1. Breaking strength in bending of aceylated (A) and unacelated (C) flakeboards as affected by flake design (D disc-cut, R ring-cut), resin content (3% or 5%), and flakeboard density (Lo, Hi), before and after APA and ASTM accelerated aging tests. In comparisions of three aging tests, means with the different letters X, Y or Z above bars are significantly different than the 0.05 level of probability

Bild 1. Biegefestigkeit acetylierter (A) und nicht acetylierter (C) Spanplatten in Abhängigkeit von der Zerspanung (D = Scheibenzerspaner; R = Messerringzerspaner), vom Festharzanteil (3% oder 5%) und von der Plattendichte (Lo = miedrig; Hi = hoch) vor und nach beschleunigter Alterung gemäß dem APA- und ASTM-Standard. Nach Auswertung von jeweils drei Alterungstests sind die Unterschiede der mit verschiedenen Buschstaben (X, Y oder Z) bezeichneten Mittelwerte signifikant auf dem 0,05%-Niveau

When the BSB values of acetylated flakeboards were compared to the low-density, disc-cut flake, 3% resin unacetylated controls, there was little difference in values of unaged and aged specimens (Fig. 1.). No evidence was found that acetylation caued a reduction in breaking load. Acetylated flakeboards actually performed much better in both aging tests when disc-cut flakes were pressed at high density and 3% resin content. Increasing resin resin content to 5% produced only small increases in BSB values in the APA and ASTM test (Fig. 1).

The ASTM Method D-1037 was the most severe of the two accelerated aging tests, causing significant reductions in BSB of aged specimens when compared to the unaged specimens. With only one exception, BSB values after the ASTM test were also significantly less than BSB values after the APA test. The APA test caused significant reductions in breaking load in less than half the treatment combinations.

4.2 Modulus of rupture

The pattern of responses in MOR to the main effects of flake design, resin content and density was essentially a duplicate of the responses in MOE (discussed later). All three main effects were very highly significant, and no significant interactions occured (Table 2).

Flake design was the most important of the three main effects in determining MOR, with the disc-cut flake producing much higher values than the ring-cut flake. Tn the test on unaged specimens, the average MOR increase was 16.5 MPa. This high level of contribution to strength was maintained throughout the two aging tests; the average increase was 16.3 MPa in the APA test and 15.5 MPa in the ASTM test. Again, the longer and wider flake is evidently more efficient than the smaller flake in sharing and transferring bending stresses within the flakeboard (Fig. 2).

High density was also a major determinant of high MOR values, although as the accelerated aging tests became more severe, high density contributed less to high MOR values. Average increases in MOR resulting from high density were 15.2 MPa in the test on unaged specimens, 11.0 MPa in the APA test, and 6.0 MPa in the ASTM test (Fig. 2).

The 5-% resin content contributed to high MOR values that were highly significant in all three tests; the more severe the accelerated aging tests became, the more the resin content contributed to increased MOR values. However, the increase in resin content from 3% to 5% contributed less to increases in MOR than did the disc-cut flake and the high density of flakeboard. Average increases in MOR as a result of the 5-% resin content were 4.5 MPa in test on unaged specimens, 6.0 MPa in the APA test, and 7.1 MPa in the ASTM test (Fig. 2).

Acetylated flakeboards made with 5-% resin content and disc-cut flakes had extremely high MOR values after the APA and ASTM aging tests (Fig. 2). The low-density acetylated flakeboards retained 97% and 85% MOR after the APA and ASTM tests, respectively, whereas the high-density flakeboards retained 87% and 71 Yo MOR after these respective tests. This provides evidence that disproportionate reductions in MOR values after accelerated aging were attributable to the increased springback of the high-density flakeboards (note TS in Fig. 5).

At comparable conditions of board fabrication, acetylated flakeboards had superior MOR values after accelerated aging relative to unacetylated flakeboards. After the APA test, the acetylated flakeboards made with disc-cut flakes at the low density and 3% resin content were 5.8 MPa higher in MOR than were the controls; 91% strength was retained in the acetylated flakeboards compared to only 67% in the controls (Fig. 2). After the ASTM test, MOR was 4.1 MPa higher in the acetylated flakeboards; 75% MOR was retained in the acetylated boards compared to only 56% in controls.

4.3 Modulus elasticity

Flake design, density, and resin content were again highly significant main effects on MOE within each of the aging tests, with no interactions indicated within any aging test (Table 2).

Flakeboards made with disk-cut flakes had consistently high average MOE values–1.93GPa in the test on unaged specimens, 1.86 GPa in the APA test, and 1.95 GPa in the ASTM test, as was the case with the MOR values. High density also contributed to high MOE, although the difference in density contributed less to MOE increases as the aging tests became more severe. This trend to lower MOE with higher density was consistent with the trend in MOR values. Average MOE differences resulting from density declined from 1.58 GPa in unaged specimens to 1.02 GPa in the APA test to 0.48 GPa in the ASTM test. However, these results contrast with the previously mentioned BSB tests where increased breaking loads were consistently maintained in high-density specimens even in the severe APA test and the more severe ASTM test. The 5% resin content also produced higher MOE values than the 3-% level, although this factor contributed less overall than did either the disc-cut flakes or the high density. However, as the aging tests became more severe, the 5-'YO resin content played a more important role in increasing the MOE differences resulting from resin content; that is, MOE differences were 0.47 GPa in the unaged specimens, 0.68 GPa in the APA tests, and 0.88 GPa in the

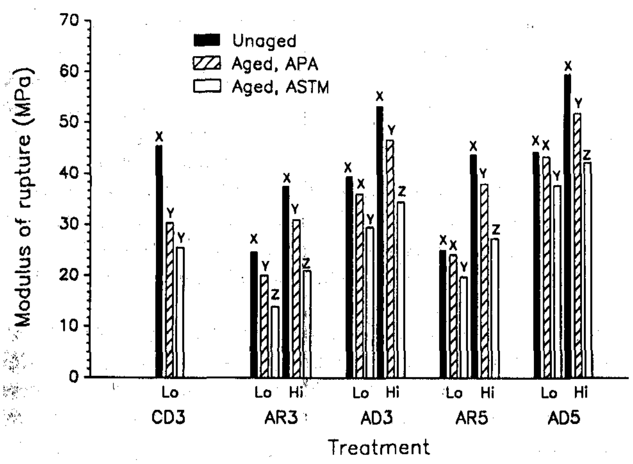

Fig. 2. Modulus of rupture of acetylated (A) and unacetylated (C) flakeboards as affected by flake design (D disc-cut, R ring-cut). resin content (3% or 5%), and flakeboard density (Lo, Hi) before and after APA and ASTM accelerated aging tests. In comparisons of three aging tests, means with the different letters X, Y, or Z above bars are significantly different at the 0.05 level of probability

Bild 2. Bruchspannung acetylierter (A) und nicht acetylierter (C) Spanplatten in Abhängigkeit von der Zerspanung (D = Scheibenzerspaner; R = Messerringzerspaner), vom Festharzanteil (3% oder 5%) und von der Plattendichte (Lo = niedrig; Hi = hoch) vor und nach beschleunigter Alterung gemäß dem APA- und ASTM-Standard. Nach Auswertung von jeweils drei Alterungstests sind die Unterschiede der mit verschiedenen Buchstaben (X, Y oder Z) bezeichneten Mittelwerte signifikant auf dem 0,05%-Niveau

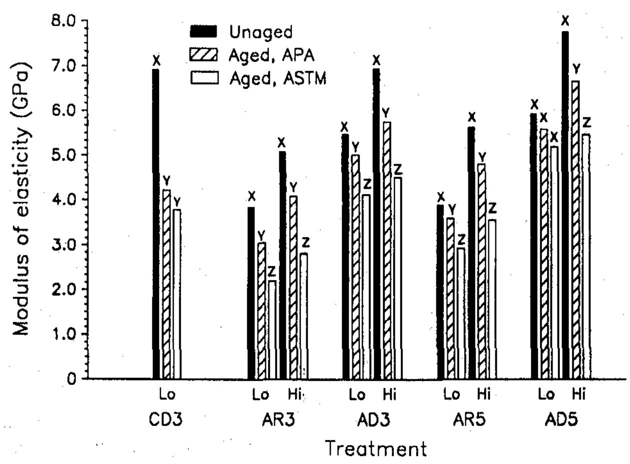

Fig. 3. Modulus of elasticity of acetylated (A) and unacetylated (C) flakeboards as affected by flake design (D disc-cut, R ring-cut), resin content (3% or 5%), and flakeboard density (Lo, Hi) before and after APA and ASTM accelerated aging tests. In comparisons of three aging tests, means with the different letters X, Y, or Z above bars are significantly different at the 0.05 level of probability

Bild 3. Bruchspannung acetylierter (A) und nicht acetylierter (C) Spanplatten in Abhängigkeit von der Zerspanung (D = Scheibenzerspaner; R = Messerringzerspaner), vom Festharzanteil (3% oder 5%) und von der Plattendichte (Lo = niedrig; Hi = hoch) vor und nach beschleunigter Alterung gemäß dem APA- und ASTM-Standard. Nach Auswertung von jeweils drei Alterungstests sind die Unterschiede der mit verschiedenen Buchstaben (X, Y oder Z) bezeichneten Mittelwerte signifikant auf dem 0,05%-Niveau

ASTM tests. All these effects can be observed in the relationship shown in Fig. 3.

It is interesting to compare the MOE values of acetylated and unacetylated flakeboards made with disc-cut flakes at low density and 3% resin. The MOE values of the APA- and ASTM-aged acetylated flakeboards were somewhat higher and the acetylated flakeboards retained 91% and 75%, respectively, of the MOE values of their unaged counterparts. The unacetylated flakeboards retained only 61% and 55% MOE, respectively, of their unaged counterparts. Even better MOE performance can be obtained in severe aging tests by increasing density. Although the increase in MOE performance could not be justified by the cost of the additional resin, the 5-% resin content did improve performance in the accelerated aging tests. The unaged acetylated flakeboards made with disc-cut flakes at low density and 5% resin retained 94% and 87% MOE after the APA and ASTM tests, respectively.

In all but one instance, the ASTM aging test caused significant decreases in MOE relative to the test on unaged specimens (Fig. 3). No significant MOE differences occurred among tests of the acetylated flakeboards made with disc-cut flakes at low density and 5% resin. In general, both acclerated aging tests caused a significant decrease in MOE when compared to the test on unaged specimens.

4.4 *Internal bond strength*

Resin content and flakeboard density were highly significant main effects in ail three aging tests, but flake design was significant only in the ASTM test. However, significant interactions occurred in all three tests; therefore, a more detailed analysis was indicated for IB strength tests (Table 2).

Even though flake design was not a significant main effect in the APA test, a study of interactions showed that for the ring-cut flakeboards, the increase in IB strength produced by high densify was highly significant - 0.45 MPa at the 3-% resin level and 0.73 MPa at the 5-% level. In these flakeboards, the 5-% resin content produced 0.42 MPa and 0.70 MPa increases in IB strength at the low and high density levels , respectively. Within the disc-cut flakeboards, the high-density flakeboards were 0.28 MPa higher in IB strength than were the low-density boards, and the 5% resin content produced IB strength averaging 0.59 MPa higher than the 3-% resin boards. In Fig. 4, note the extraordinarily high IB strength values of acetylated flakeboards after the APA aging test, particularly among the highdensity flakeboards; none of the IB strength values is less than 1.08 MPa or as high as 1.82 MPa. After the APA aging test, those flakeboards retained a minimum of 75% and as much as 97% of their original IB strength.

In regard to the interaction of flake design and density in flakeboards made at 3-% resin content, only the high density produced a significant IB increase (0.39 MPa). However, at the 5-% resin level, the disc-cut flake contributed to an IB increase of 0.29 MPa at the low density, and at the high density, the ring-cut flab produced an IB stength increase of 0.21 MPa. Perhaps this is an anomaly, because this apparent disparity was not statistically significant at the 0.05 level of probability.

After specimens were aged by the ASTM test, all three main effects were highly significant along with interactions of flake design × density and the three-way interaction of flake design × density × resin content (Table 2).

For the acetylated flakeboards made with the disc-cut flakes, density was not a significant factor but resin content was – the 0.45-MPa increase in IB strength was attributable to resin increase. In contrast, flakeboards made with the ring-cut flakes had significant and large increase in IB strength values attributable to density –0.23 MPa at the 3-% resin level and 0.48 MPa at the 5-% level. Further, more the ring-cut flakeboards had IB strength increases also attributable to an increase in resincontent – 0.43 MPa at low density and 0.68 MPa at high density. Note the large IB strength increase of 1.30 MPa in the high-density, 5-% resin acetylated flakeboards after the severe ASTM test (Fig. 4). These flakeboards retain 64% of their original IB strength.

At 3-% resincontent, the boards with ring-cut flakes produced significant but only slightly increased IB strength values. At the same resin level, high density increased strength 0.18 MPa. At 5-% resin content, high density increased IB strength 0.48 MPa in the flakeboards made with ring-cut flakes, but the difference resulting from density level was not significant in boards made with disc-cut flakes. At the same resin level, the ring-cut flakes increased IB strength by 0.44 at high density. These effectscan be observed in Fig. 4.

Both APA and ASTM tests caused significant deterioration in IB strength values relative to each other and to the unaged specimens at all treatment combinations, with one exception (Fig. 4). The ASTM test contained 36 h of steam treatment that caused severe reductions in IB strength among all flakeboards regardless of fabrication variables. The acetylated flakeboards made with ring-cut flakes with 5-% resin at high density had an average IB strength of 1.30 MPa and retained 64% of their stength after the ASTM test. Figure 4 shows quite clearly that the increase in resin content to 5% was the key to the greatly increased IB strength values and strength retentions among acetylated flakeboards subjected to the ASTM aging test.

The improvement in aging capability produced by acetylation can be seen in limited way by comparing IB strength values of the unacetylated controls with the comparable

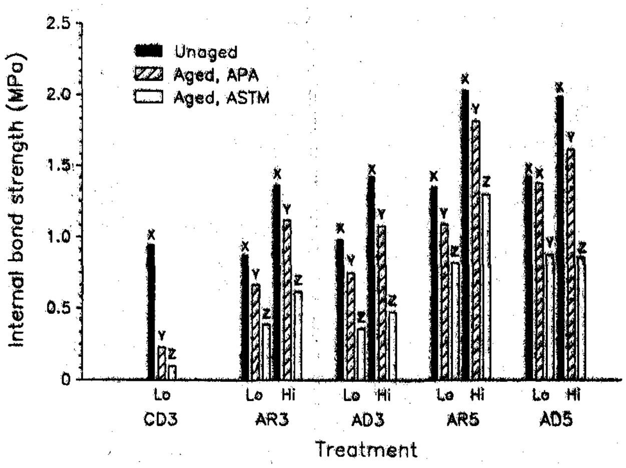

Fig. 4. Internal bond strength of acetylated (A) and unacetylated (C) flakeboards as affected by flake design (D disc-cut, R ring-cut), resin content (3% or 5%), and flakeboard density (Lo, Hi) before and after APA and ASTM accelerated aging tests. In comparisons of three aging tests, means with the different leters X, Y, or Z above bars are significantly different at the 0.05 level of probability

Bild 4. Quezugfestigkeit acetylierter (A) und nicht acetylierter (C) Spanplatten in Abhängigkeit von der Zerspanung (D = Scheibenzerspaner; R = Messerringzerspaner), vom Festharzanteil (3% oder 5%) und von der Plattendichte (Lo = niedrig; Hi = hoch)vor und nach beschleunigter Alterung gemäß dem APA- und ASTM-Standard. Nach Auswertung von jeweils drei Alterungstests sind die Unterschiede der mit verschidenen Buschstaben (X, Y oder Z) bezeichneten Mittelwerte signifikant auf dem 0,05%-Niveau values of fabricated acetylated flakeboards. After the APA test, acetylated flakeboards retained 75% of their strength and unacetylated boards retained only 25% of their strength. After the ASTM test, acetylated flakeboards retained 36% of their strength and unacetylated boards, 10%. The twelve 3-h steam treatments caused severe deterioration in IB strength, relative to the other variables tested in acetylated and unacetylated flakeboards. The acetylated flakeboards were much less affected by the six cycles in the APA test.

4.5 *Thickness swell*

Flake design, resin content, and flakeboard density were significant variables affecting TS percentages in the APA and ASTM aging tests. However, there were significant interactions of flake design × density in both tests and an interaction of flake design × resin content in the APA test (Table 2).

Note that high density in the flakeboards caused a larger increase in TS than did low density. This commonly known and undesirable effect, called springback, must be weighed against the desirable increases in IB, MOE, MOR, and BSB that were produced by high density. The interaction of density and flake design was important in both aging tests. In the APA test, high-density flakeboards made with ring-cut and disc-cut flakes had an average TS increase of 2.2 and 4.1%, respectively. The effect of density was even more pronounced in the ASTM test: TS increased 4.0% and 8.5% in boards made with ring-cut and disc-cut flakes, respectively. The ring-cut flakes had less influence on TS because they were much smaller and more variable in size than the disc-cut flakes. Consequently, the ring-cut flakes were able to conform and connect to adjacent particles with less residual stress. The disc-cut flakes contained essentially no fine particles after particles <0.80 mm were screened out.

As shown in Fig. 5, the 5-% resin content had a greater effect on TS than did the 3% resin, as expected. In the APA test, the lowest percentage of TS (4.5) occurred in low-density flakeboards made with ring-cut flakes and 5% resin. At 3% resin, TS increased to 8.1%. In the ASTM test, TS in comparable flakeboards was 7.9% at the 5% resin level and 15.3% at the 3-% resin level. Even with 5-% resin level, 36 h of steaming in the ASTM test sharply reduced resistance to TS, particularly among the high-density flakeboards.

It is interesting to compare TS of the unacetylated controls with that of comparable acetylated flakeboards. In the APA test, TS averaged 16.5% in the controls, whereas acetylation reduced TS to 7.6%. However, in the ASTM test, steaming helped increase TS to 12.9% in the acetylated flakeboards compared with 17.6% in the unacetylated controls.

From the standpoint of minimizing TS in an exterior composite where raised surface flakes and fibers might be objectionable, the best surface can be obtained by fabricating boards with 5% resin and acetylated, ring-cut flakes pressed to a conventional density of 672 kg/m^3. The next best surface can be obtained by using 3% resin, low density, and acetylated disc-cut or ring-cut flakes?

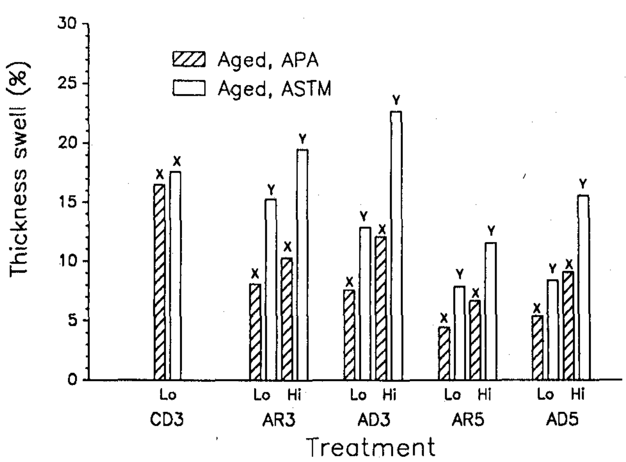

Fig.5. Thickness swell of acetylated (A) and unacetylated (C) flakeboards as affected by flake design (D disc-cut, R ring-cut), resin content (3 or 5%), and flakeboard density (Lo, Hi) after APA and ASTM accelerated aging tests. In comparisons of two aging tests, means with the different letters X or Y above bars are significantly different at the 0.05 level of probability

Bild 5. Dickenquellung acetylierter (A) und nicht acetylierter (C) Spanplatten in Abhängigkeit von der Zerspanung (D = Scheibenzerspaner; R = Messerringzerspaner), vom Festharzanteil (3% oder 5%) und von der Plattendichte (Lo = niedrig; Hi = hoch) vor und nach beschleunigter Alterung gemäß dem APA- und ASTM-Standard. Nach Auswertung von jeweils drei Alterungstests sind die Unterschiede der mit verschiedenen Buchstaben (X, Y oder Z) bezeichneten Mittelwerte signifikant auf dem 0,05%-Niveau

5 Concluding remarks

Flake design, flakeboard density, resin content, and their interactions were critical to the performance of acetylated flakeboards in APA and ASTM accelerated aging tests. as indicated by breaking strength in bending, modulus of rupture, modulus of elasticity, internal bond strength, and thickness swell. The longer disc-cut flakes produced much higher strength values than did the ring-cut flakes in the three bending tests. The ring-cut flakes performed comparably to the disc-cut flakes in internal bond tests; however, ring-cut flakes contributed somewhat more to reducing thickness swell than did disc-cut flakes. Acetylated flakeboards fabricated at high density were much stronger than those fabricated at low density in all four mechanical property tests. However, the high-density boards developed more thickness swell than did the low-density boards. The 5-% resin content contributed little to increased strength values in the bending tests, but this level of resin significantly improved flakeboard performance in internal bond and thickness swell tests.

The APA six-cycle, vacuum-pressure soak and dry procedure was a severe aging test, but it caused only small strength reductions and minimal thickness swell in the acetylated flakeboards. The ASTM aging test with its twelve 3-h steam cycles caused significantly greater reductions in strength properties and greater thickness swell than did the APA aging test.

The improved aging capability produced by acetylation is readily apparent when the mechanical properties and thickness swell of the acetylated flakeboards are compared with that of comparably fabricated unacetylated controls. The acetylated flakeboards retained much higher levels of strength and resistance to thickness swell after the APA and ASTM aging tests. It is also apparent from all tests that acetylation did not interfere to any practical degree with the adhesion of MDI (polymeric diphenylmethane diisocyante) resin to acetylated flakes. All property values were enhanced by acetylation, particularly internal bond strength and resistance to thickness swell.

Acknowledgments. We express our gratitude to Wesley Rork and Ben Henderson for preparation of flakes, to Jeffrey Rowell and Ben Henderson for acetylation of flakes, to Bill Kreul for conducting the accelerated aging tests, to Jill Wennesheimer and Mary Doran for conducting the mechanical and physical property tests, and to Steve Verrill for conducting the statistical analyses. We also thank Daryle Layton of Mobay Chemical Company for his consultations and for supplying the adhesive for this study.

6 Literature

APA. 1988: Manual of APA test methods for structural-use panels. Performance standards and policies for structural-use panels. In: APA PRP-108. American Plywood Association, Tacoma, WA, pp. 32-70

ASTM 1987: Standard methods of evaluating the properties of wood-base fiber and particle panel materials. American Society for Testing and Materials Designation D1037-87. In: Annual Book of ASTM Standards 1987. Vol. 04.09 Wood, pp. 226-272

Imamura, Y.; Nishimoto, K.; Rowell, R. M. 1987: Internal bond strength of acetylated flakeboard exposed to decay hazards. Mokuzai Gekkaishi 33(12):986-991

Rowell, R.M.; Tillman, A.-M.; Simonson, R. 1986: A simplified procedure for acetylation of hardwood and softwood flakes for flakeboard production. J. Wood Chem. Technol. 6(3):427-448

Rowell, R.M.; Esenther, G.R.; Nicolas, D.D; Nilsson, T. 1987a: Biological resistance of southern pine and aspen flakeboards made from acetylated flakes. J. Wood Chem. Technol. 7(3):427-440

Rowell, R.M.; Youngquist, J. A.; Sachs, I.B. 1987b: Adhesive bonding of acetylated aspen flakes, Part 1. Surface changes, hydrophobicity adhesive penetration and strength. Int. J. Adhesion and Adhesives 7(4):183-188

Vick, C. B.; Rowell, R.M. 1990: Adhesive bonding of acetylated wood. Int. J. Adhesive and Adhesives 10(4):263-272

Wellons, J. D. 1990: Wood adhesives: Research needs for the 1990s. Keynote address for Wood Adhesives 1990. Symposium sponsored by the USDA Forest Service, Forest Products Laboratory, and Forest Products Research Society, Madison, WI

Youngquist, J.A.; Rowell, R.M.; Adhesive bonding of acetylated aspen flakes, Part 3. Adhesion with isocyanates. Accepted by International Journal of Adhesion and Adhesives (in press)

Youngquist, John A.; Krzysik, Andrzej; Rowell, Roger M. 1986a: Dimensional stability of acetylated aspen flakeboard. Wood Fiber Sci. 18(1):90-98

Youngquist, J. A.; Rowell, R.M; Krzysik, A. 1986b: Mechanical properties and dimensional stability of acetylated aspen flakeboard. Holz. Roh- Werkstoff 44(12):453-457