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Full Length Research Paper

Determination of bonding performance of several modified wood adhesives

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We have assessed the effect on bonding performance of modifying synthetic wood adhesives. Wood samples were prepared from Scots pine (*Pinus sylvestris* L.) and oriental beech (*Fagus orientalis* L.). Samples were bonded using poly(vinyl acetate) (PVAc) and urea-formaldehyde (UF) adhesives modified with melamine-formaldehyde (MF) and phenol-formaldehyde (PF). Modified formulations included PVAc alone, PVAc + UF (at 15, 35 and 50% UF), UF alone, UF + MF (at 15, 35 and 50%), and UF + PF (at 15, 35 and 50% PF). Samples were exposed to three experimental environments: Standard atmosphere, cold water, and boiling water. For bonding strength determination, we used DIN 53 255 standard. We conclude that the adhesive modifications tested generally have no significant effect on bonding performance under standard atmospheric conditions, but do increase bonding performance under cold and boiling-water conditions. The best adhesive bonding strength was found using oriental beech samples with the UF + MF (50%) adhesive: 9.24 N/mm^2 bonding strength under standard atmosphere conditions, 6.06 N/mm^2 in cold water, and 4.39 N/mm^2 in boiling water.

Key words: Adhesive modification, poly(vinyl acetate) (PVAc), urea-formaldehyde (UF), melamine-formaldehyde (MF), phenol-formaldehyde (PF), bonding strength, bonding performance.

INTRODUCTION

Animal-based adhesives have been widely used in assorted industries for more than 300 years. Synthetic adhesives provide very important technical advantages to the woodworking industry. Synthetic adhesives are typically either thermoplastic or duroplastic in nature. Thermoplastic adhesives as PVAc are applied at ambient temperature (TS EN 205). Duroplastic adhesives are applied hot, in plasticized state, same as the thermoplastic adhesives, but they are not cross-linked at the time of application. Contrary to thermoplastics, these adhesives are not plasticized again when they are reheated. The process is not reversible.

Studies have shown that the bonding characteristics of synthetic adhesives can be altered when modified with certain compounds. Plywood resistant to cold water can be produced using Wikol (super lackleim 308) and modified Wikol adhesives (DIN 53 255). Medium-density fibreboard (MDF) and plywood (between 3 and 9 mm thick), have been covered with oak, elm, teak, and Paulownia wood panels (0.25 mm thick) using poly(vinyl acetate) (PVAc), urea-formaldehyde (UF), and PVA/UF adhesive mixtures. The highest bonding strengths obtained were from PVA/UF adhesives (Gos et al., 1987).

Samples bonded with UF adhesives modified with poly(vinyl alcohol) (1 to 2%) and amylum (10 to 15%) were tension tested and showed increased bonding strength (Suh et al., 1989). The UF adhesive was modified with poly(vinyl alcohol) and amylum to increase water resistance. The results indicated that the amount of free formaldehyde decreased and bonding strength

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Table 1. Modified adhesive mixture combinations and proportions.

Modified adhesive	Adhesive type	Mixture proportion values as a percentage of total mass (g)			
		0%	15%	35%	50%
PVA + UF	PVA	100	85	65	50
	UF	0	15	35	50
UF + MF	UF	100	85	65	50
	MF	0	15	35	50
UF + PF	UF	100	85	65	50
	PF	0	15	35	50

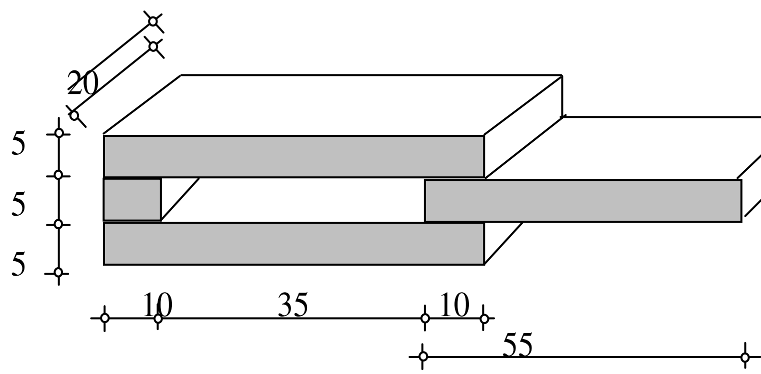

Figure 1. General test sample configuration (units: mm).

increased for the modified adhesive (Liu, 1997). Tensile strength tests were conducted on samples of pine, cedar, acacia, and oak bonded with ST-10 (a PVA-based, hardened, transparent adhesive), and ST-10 + UF (at 10, 20 and 30% UF). The highest shear strength recorded was found in oak bonded with ST-10 + UF (20%) under standard atmosphere conditions; the lowest shear strength was found in acacia bonded with ST-10 + UF (20%) under boiling-water conditions (Shen, 1997).

In a similar study, PVA and UF-based adhesives were modified with melamine-formaldehyde (MF). Beech samples bonded with UF + MF (50%) provided the highest bonding strength in standard atmosphere conditions, whereas pine bonded with UF + and phenol-formaldehyde (PF; 15%) provided the lowest bonding strength under boiling-water conditions (Altinok et al., 1999). Pine, cedar, acacia, and oak wood samples were bonded with VB20 (a PVA-based, hardened, transparent, and water resistant adhesive) and VB20 + UF (at 10 and 20% UF). Oak bonded with VB20 + UF (20%) produced the highest shear strength, whereas acacia bonded with VB-20 alone produced the lowest shear strength (Kocaturk, 2000; Pizzi and Mittal, 2003; Frihart, 2005; Šernek, 2007; Altinok et al., 2000).

EXPERIMENTAL

For this study, we tested the bonding strength of Scots pine (*Pinus sylvestris* L.) and Oriental beech (*Fagus orientalis* L.) samples bonded with PVAc and UF adhesives. Both adhesive types are unsuited for use in exterior conditions, so for this study, they were modified with melamine-formaldehyde (MF) and phenol-formaldehyde (PF), both of which maintain strong bonds under exterior conditions. Scots pine and oriental beech are species widely used in the furniture industry. Wood samples were randomly selected from timber markets in Ankara, Turkey.

The PVAc-based adhesive was purchased from Polisan (Izmit, Turkey). Viscosity at 20°C was 500 ± 10 cP, pH was 5 (20°C), and density was 1.1 g/cm^3. The UF-based adhesive was composed of 60% resin, 20% wheat flour, 10% water, and 10% ammonium sulphate (a hardener). Viscosity of the UF adhesive solution at 20°C was 480 ± 10 cP, pH was 8 (20°C), and density was 1.22 g/cm^3. The PF-based adhesive was composed of 85% liquid material and 15% hardener (p-toluenesulfonic acid). Viscosity of the PF adhesive solution at 20°C was 450 ± 10 cp, pH was 9.5 (20°C), and density was 1.22 g/cm^3. The MF-based adhesive was composed of 85% liquid material and 15% hardener (ammonium chloride). Viscosity of the MF adhesive solution at 20°C was 450 ± 10 cP, pH was 9.5 (20°C), and density was 1.22 g/cm^3. The PVA, UF, MF, and PF adhesives were combined using different proportions into 12 different adhesive test mixtures (Table 1).

Preparation of experimental samples

Ten replicates of the combination of two wood types, three modified adhesives, four modification mixture ratios, and three test conditions (holding in standard atmosphere, cold water and boiling water), for a total of 720 samples (10 × 2 × 3 × 4 × 3), were prepared and tested (Figure 1).

Rough samples were cut from sapwood and conditioned in a

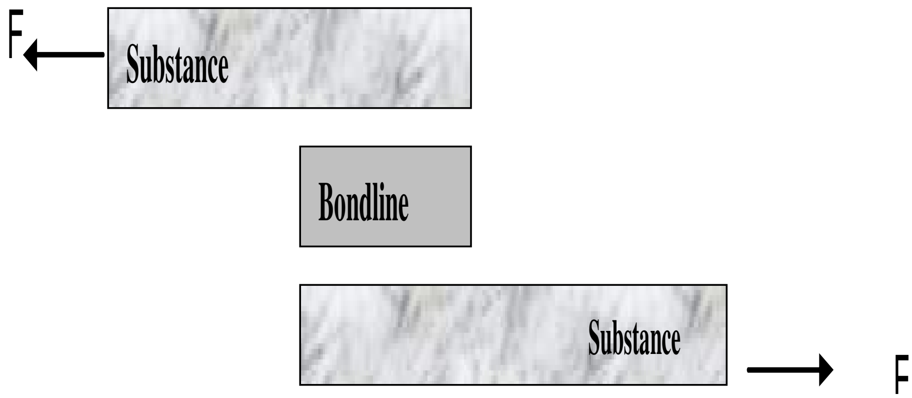

Figure 2. Tensile experiment apparatus.

Table 2. Mean adhesive bond strength results (in N/mm²).

Wood type and test condition	Modified adhesive mixtures											
	PVA + UF (%)				UF + MF (%)				UF + PF (%)			
	0	15	35	50	0	15	35	50	0	15	35	50
I + Sa	70.97	70.25	70.28	70.26	80.64	70.61	80.47	90.24	80.64	50.79	70.61	80.28
I + Cw	00.00	10.06	10.19	10.14	50.13	40.64	60.01	60.06	50.13	10.15	20.29	30.50
I + Bw	00.00	00.00	00.00	00.00	00.23	10.26	30.62	40.39	00.00	00.00	00.00	00.00
II + Sa	50.06	50.21	40.14	40.62	40.26	40.44	30.98	40.44	40.26	40.37	40.30	40.06
II + Cw	00.00	10.74	20.14	20.10	30.31	30.44	30.56	40.16	30.31	20.54	20.91	30.47
II + Bw	00.00	00.00	00.00	00.00	00.23	10.26	30.22	30.57	00.00	00.33	00.43	00.91

I - Oriental beech; II - scots pine; Sa - standard atmosphere; Cw - cold water; Bw - boiling water; 0.00 indicates test failure.

chamber at 20 ± 2°C and 65 ± 3% relative humidity until their weight stabilized (TS EN 205 standard). Samples were cut to final test dimensions when their moisture content was 12% (TS 2471 standard). The modified adhesives were applied to the samples using the DIN 53 252 standard and the samples were assembled as shown in Figure 1 (DIN 53252, 1964). Press temperature was 60°C for PVAc and 80°C for the other mixtures, with a press time of 4 min at a compression pressure of 0.2 N/mm^2 (Ors, 1987).

Replicates of each sample type were exposed to three different environmental conditions (TS EN 205 standard): 1) Holding in standard atmosphere (Sa), 7 days at a temperature of 20 ± 2°C with a relative humidity of 65% ± 3%; 2) Holding in cold water (Cw), 7 days in standard atmospheric conditions, then 4 days in cold water; and 3); holding in boiling water (Bw), 7 days in standard atmospheric conditions, then 4 days in boiling water, followed by 2 h in cold water.

Bonding strength (tensile shear strength) test

Bonding strength tests were carried out using a standard machine in compliance with the DIN 53 255 standard procedures. The loading rate was 50 mm/min (Figure 2).

Loading continued until complete separation or a break in the surface of the test sample occurred. Bonding strength (**σ_k**) was calculated as:

$$\sigma_k = F_{max} / 2A \Rightarrow \sigma_k = F_{max} / 2(a \times b),$$

where F_{max} is maximum load at the break point (N), a is glued face width (10 mm), and b is glued face length (20 mm).

RESULTS AND DISCUSSION

Multivariate analyses were performed to determine the primary and interaction effects on bonding strength mean. Duncan's multiple comparisons procedure was used to determine mean differences among the 720 samples (Table 2).

The results indicated that, in general, modification of the base PVAc and UF adhesives only slightly decreased the bonding strength for both wood types under Sa conditions but slightly increased bonding strength under Cw conditions. Considerable bonding strength was obtained on both wood types under Bw conditions for all of the UF/MF adhesive mixtures. This result is likely explained by the fact that UF and MF are both aminoplastic resins, and urea resin and melamine resin have similar molecular structures, though melamine resin has a higher resistance against both cold water and boiling-water conditions. Multivariate analysis results regarding the effect of wood type, modified adhesive

Table 3. Analysis of variance by wood type, modified adhesives type and mixture ratio.

Source of variance	DF	Sum of squares	Mean square	F–value	5%
A	1	270.149	270.149	1262.181	0.000
B	2	467.922	233.961	1093.104	0.000
AB	2	56.817	28.408	132.728	0.000
C	2	3380.859	1690.430	7897.966	0.000
AC	2	412.806	206.403	964.348	0.000
BC	4	237.939	59.485	277.922	0.000
ABC	4	28.458	7.115	33.240	0.000
D	3	73.543	24.514	114.534	0.000
AD	3	22.980	7.660	35.789	0.000
BD	6	102.399	17.066	79.737	0.000
ABD	6	25.592	4.265	19.928	0.000
CD	6	39.780	6.630	30.976	0.000
ACD	6	18.265	3.044	14.222	0.000
BCD	12	107.793	8.983	41.968	0.000
ABCD	12	9.888	0.824	3.849	0.000
Error	648	138.694	0.214	-	-
Total	719	5393.882	-	-	-

A - wood type; B - adhesive type; C - test condition; D - mixture ratio; DF - degrees of freedom.

type, mixture ratio, and interactions with test conditions are presented in Table 3.

Differences among groups with respect to the effect of variance source on adhesive bonding strength were statistically significant ($p = 0.05$). Duncan's mean comparison test results were conducted to determine the importance of differences among the test groups (Table 4).

The highest adhesive bonding strength observed was for the UF + MF 50% mixture on beech, with a value of 9.24 N/mm^2 under Sa conditions, 6.06 N/mm^2 under Cw conditions, and 4.39 N/mm^2 under Bw conditions (Figures 3 and 4). This result may be due to the higher density of beech (0.63 g/cm^3) compared with pine (0.49 g/cm^3), which allows the UF/MF mixture to adhere more strongly. The lowest adhesive bonding strength observed occurred on both wood types using any of the PVAc/UF and UF/PF mixtures under Cw and Bw conditions. This is likely because vinyl acetate resin is very easily softened under Cw and Bw conditions. Phenol resin is not as strong as melamine resin in water or under exterior conditions. Pine contains more natural resin than does beech, and has a lower density. Natural resins and other fatty extractives impair bonding between wood cellulose and adhesive molecules.

Unmodified PVAc adhesive maintained a bond with both wood types only under Sa conditions. Because poly(vinyl acetate) resin is hardened physically (thermoplastic), the Cw and Bw conditions altered the structure and caused a failure. PVAc/UF mixtures had lower bonding strength under Cw conditions than under Sa conditions but did not fail, as was the case for Bw conditions. Other research has noted that boiling water has a greater effect on PVA than does cold water (Gos et al., 1987).

Unmodified UF adhesive bonding strength on beech was much reduced compared with that on pine. Adhesive bonding strengths of the UF/MF mixtures (35 and 50% ratio) increased under both Ca and Bw conditions for both wood types. All UF/PF mixtures tested under Bw conditions failed on beech and had very low bonding strength on pine. Melamine resin and phenolic resin are hardened by a chemical reaction (duroplastic), and the hardened bonds should be resistant to both water and heat. Our results indicate that melamine resin mixtures offer strength under Bw conditions, and both offer strength under Cw conditions (Suh et al., 1989; Liu, 1997).

Beech wood bonded with a UF + MF 50% adhesive mixture under Sa conditions had the highest measured adhesive bonding strength. The lowest adhesive bonding strength measured was with the combination of pine, UF + PF 15% adhesive mixture, and Bw conditions. Beech, which has higher density, contains lower extractives, and has a more homogeneous structure than pine, allowed for stronger adhesion to the wood surface for both UF/MF and UF/PF mixtures.

Differences between PVA, UF, and both types modified with MF and PF (at all mixture ratios) were not statistically significant under Sa conditions for both wood types 7, 8, 9). Unmodified PVA adhesives failed under Cw and Bw conditions. Unmodified UF adhesives had lower bonding performance under Cw and Bw conditions (Table 5).

Table 4. Duncan's mean comparison test results (in N/mm^2).

Wood material + modified adhesive + test condition + rate of mod.	X	HG	Wood material + modified adhesive + test condition+ rate of mod	X	HG
I+[UF+MF 50%]+Sa	9.24	A	II+[UF+MF 15%]+Cw	3.44	L
I+[UF+PF 0%]+Sa	8.53	B	II+[UF+MF 0%]+Cw	3.31	LM
I+[UF+MF 35%]+Sa	8.47	B	II+[UF+PF 0%]+Cw	3.31	LM
I+[UF+PF 50%]+Sa	8.28	BC	II+[UF+MF 35%]+Bw	3.22	LM
I+[PVA+UF 0%]+Sa	7.98	CD	II+[UF+PF 35%]+Cw	2.91	MN
I+[UF+PF 0%]+Sa	7.81	D	II+[UF+PF 15%]+Cw	2.54	NO
I+[UF+PF 35%]+Sa	7.61	DE	I+[UF+PF 35%]+Cw	2.29	OP
I+[UF+MF 15%]+Sa	7.61	DE	II+[PVA+UF 35%]+Cw	2.14	OPQ
I+[PVA+UF 35%]+Sa	7.28	E	II+[PVA+UF 50%]+Cw	2.10	PQ
I+[PVA+UF 50%]+Sa	7.26	E	II+[PVA+UF 15%]+Cw	1.74	QR
I+[PVA+UF 15%]+Sa	7.25	E	I+[PVA+UF 50%]+Cw	1.46	RS
I+[UF+MF 50%]+Cw	6.06	F	II+[UF+MF 15%]+Bw	1.28	ST
I+[UF+MF 35%]+Cw	6.01	F	I+[UF+MF 15%]+Bw	1.26	ST
I+[UF+PF 15%]+Sa	5.99	F	I+[PVA+UF 15%]+Cw	1.19	ST
II+[PVA+UF 15%]+Sa	5.21	G	I+[UF+PF 15%]+Cw	1.15	STU
I+[UF+MF 0%]+Cw	5.13	G	I+[PVA+UF 15%]+Cw	1.06	STU
I+[UF+PF 0%]+Cw	5.13	G	II+[UF+PF 50%]+Bw	0.92	TU
II+[PVA+UF 0%]+Sa	5.06	G	II+[UF+MF 0%]+Bw	0.74	UV
I+[UF+MF 15%]+Cw	4.64	H	II+[UF+PF 35%]+Bw	0.43	VW
II+[PVA+UF 50%]+Sa	4.62	HI	II+[UF+PF 15%]+Bw	0.33	VW
II+[UF+MF 15%]+Sa	4.44	HIJ	I+[UF+PF 0%]+Bw	0.23	W
II+[UF+MF 50%]+Sa	4.44	HIJ	I+[PVA+UF 0%]+Bw	0.00	W
I+[UF+MF 50%]+Bw	4.39	HIJ	I+[PVA+UF 15%]+Bw	0.00	W
II+[UF+PF 15%]+Sa	4.37	HIJ	I+[PVA+UF 35%]+Bw	0.00	W
II+[UF+PF 35%]+Sa	4.30	HIJ	I+[PVA+UF 50%]+Bw	0.00	W
II+[UF+MF 0%]+Sa	4.26	HIJ	I+[UF+PF 0%]+Bw	0.00	W
II+[UF+PF 0%]+Sa	4.26	HIJ	I+[UF+PF 15%]+Bw	0.00	W
II+[UF+MF 50%]+Cw	4.16	HIJ	I+[UF+PF 35%]+Bw	0.00	W
II+[PVA+UF 35%]+Sa	4.14	IJ	I+[UF+PF 50%]+Bw	0.00	W
II+[UF+PF 50%]+Sa	4.06	J	I+[PVA+UF 0%]+Cw	0.00	W
II+[UF+MF 35%]+Sa	3.98	JK	II+[PVA+UF 0%]+Cw	0.00	W
I+[UF+MF 35%]+Bw	3.62	KL	II+[PVA+UF 0%]+Bw	0.00	W
II+[UF+MF 50%]+Bw	3.57	KL	II+[PVA+UF 15%]+Bw	0.00	W
II+[UF+MF 35%]+Cw	3.56	KL	II+[PVA+UF 35%]+Bw	0.00	W
I+[UF+PF 50%]+Cw	3.50	L	II+[PVA+UF 50%]+Bw	0.00	W
II+[UF+PF 50%]+Cw	3.47	L	II+[UF+PF 0%]+Bw	0.00	W

LSD ± 0.4062; 0.00 implies test failure; X: Mean value; HG: Homogeneity group.

Conclusion

Unmodified PVA adhesives could be recommended for use with either wood type under standard atmospheric conditions. PVA adhesives modified with UF (35 and 50% ratios) could be recommended for use with pine under low relative humidity and exterior conditions. PVA adhesives should not be recommended for use in furniture production where wet, hot conditions can occur. It is suggested that unmodified UF adhesives can be used for both wood types under standard atmospheric conditions. UF adhesives modified with MF and PF (35 and 50% ratios) can be recommended for use under relatively humid conditions. UF adhesives modified with MF (50% ratio) can be used under hot, humid conditions on both pine and beech.

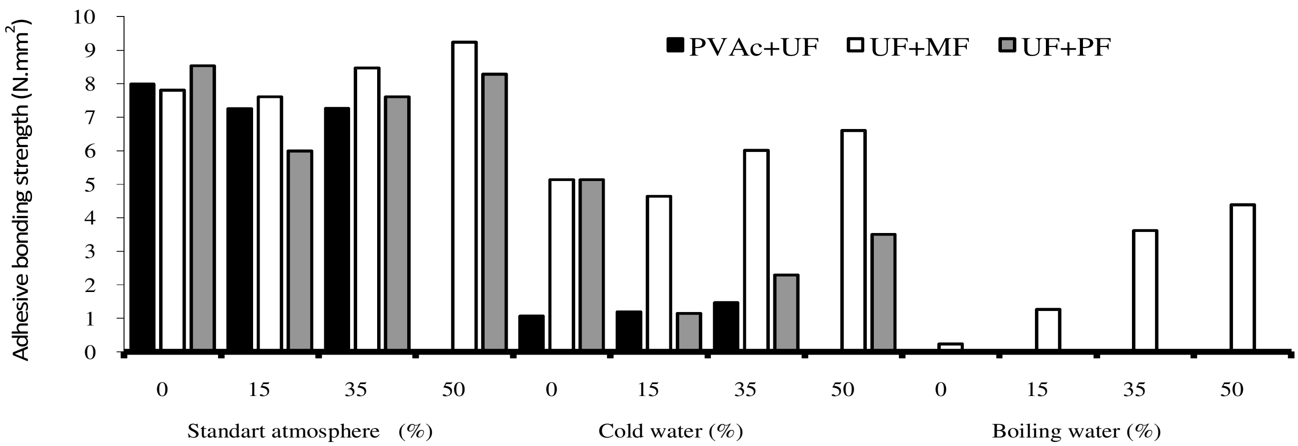

Figure 3. Adhesive bonding strength of modified adhesives on beech.

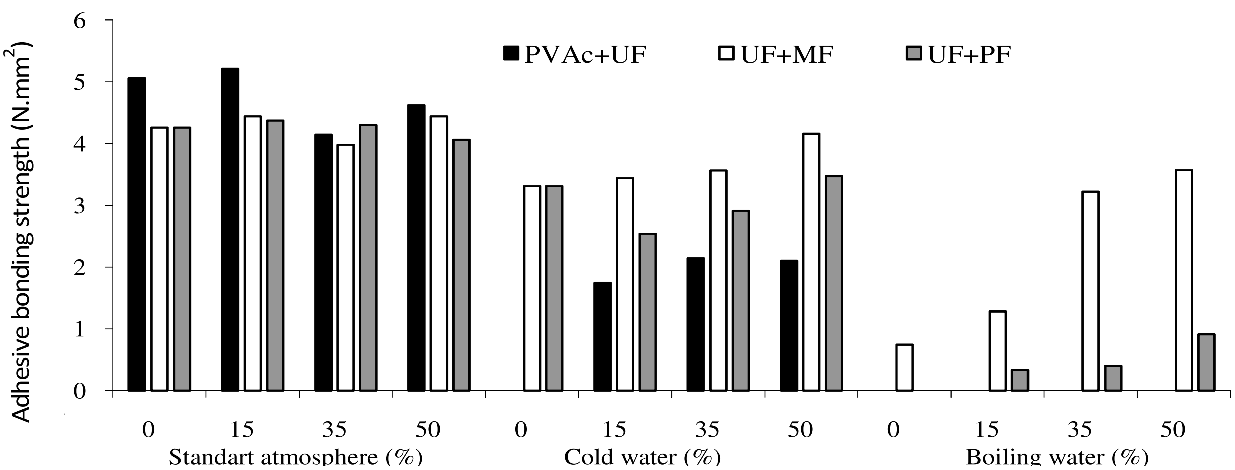

Figure 4. Adhesive bonding strength of modified adhesives on pine.

Table 5. Bonding performance (N/mm²) of modified adhesives under cold water and boiling-water conditions.

		PVA + UF (%)				UF + MF (%)				UF + PF (%)			
		0	**15**	**35**	**50**	**0**	**15**	**35**	**50**	**0**	**15**	**35**	**50**
I	Cw Bw	0	14	16	15	59	61	71	65	60	20	30	42
		0	0	0	0	3	16	43	47	0	0	0	0
II	Cw Bw	0	33	51	45	77	77	89	94	78	58	68	85
		0	0	0	0	5	28	81	80	0	8	10	22

I – beech; II – pine; 0 – test failure.

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