EFFECT OF BORON ADDITION TO ADHESIVE AND/OR SURFACE COATING ON FIRE-RETARDANT PROPERTIES OF PARTICLEBOARD

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ABSTRACT

Boron added to urea formaldehyde (UF) resin during manufacture of board from waste tea leaves was combined with a fire-retardant coating system containing boron to further reduce combustibility. Boric acid (BA) and borax (BX) were used as boron compounds separately or in a mixture of 5:1 (BA:BX, w/w) in aqueous solutions of UF resin. A BA+BX mixture was also added to a 40% aqueous solution of trimethylolmelamine (TMM) (as a binding agent) and applied to the board surface. Fire-retardant properties were determined according to the Japanese Industrial Standard JIS A 1322.

Thickness swelling and water absorption levels were not changed remarkably by boron addition to UF resin, while static bending and internal bond strengths were reduced to some extent. However, BX addition suppressed the adverse effect of BA on board strength, probably by buffering its acidic pH to almost neutral levels, while BA reduced the glowing or smoldering effect of BX. Panels with or without added boron showed no ignition after the flame source was cut off after 13 min. Surface coating with the boron-added TMM improved fire retardance. To limit the reduction of strength properties due to boron addition to the resin, fire-retardant surface coating can be applied to impart a required level of fire protection.

Keywords: Boron, bio-based composite, fire retardance, surface coating, particleboard, UF, waste tea leaves.

INTRODUCTION

Carbon-based, cellulosic materials, such as wood- and bio-based composites, will combust, pyrolize, or otherwise burn when subjected to either radiant energy (as from a fire a short distance away) or sufficient direct heat (as from a flame in direct contact). The designer seeking to enhance the fire resistance of a structure or assembly built with either sawn wood or bio-based engineered composite products can employ several techniques, including coating members with approved fire-proofing materials, or using fire-retardant-

treated sawn wood and engineered bio-based material products (Slifka 1997).

The borate chemicals offer substantial advantages for wood protection, providing fire resistance as well as efficacy against both fungi and insects, low cost, ease of handling and treatment. There is growing interest in their low mammalian toxicity and environmental acceptability (Laks et al. 1994). Adding boron compounds to the adhesives during board production seems one practical treatment. However, interference by the added chemicals in bond formation, which results in poor strength properties, limits this kind of application. Instead, raw materials can be immersed in a treatment solution, chemicals can be sprayed onto particles, and/or the end product can be surface coated. In the first two cases, chemical interference in the bonding is still likely to occur, while surface coating of boards with a fire retardant is not expected to impart long-term protection (Laks and Palardy 1990; White and Sweet 1992). On the other hand, borate loading sufficient to prevent damage by fungi and insects was claimed to improve fire resistance, but does not impart fire retardancy (Barnes and Amburgey 1989). Therefore, a combination of both boron addition to the adhesive and surface coating of the board might improve combustion resistance and some other important board properties. Compatibility of adhesive with boron is also important. The water-soluble borates have a strong negative effect on the bonding performance of phenol formaldehyde (PF) resins, but, in contrast, there was little or no effect with the urea formaldehyde (UF) (Laks et al. 1988; Laks and Manning 1994). Moreover, the UF-boron combination was promising in terms of flame retardancy (Kozlowski et al. 1992).

Agro-based fibers are of growing importance as a potential resource for composite products of the future. High-performance composite materials with uniform densities, durability in adverse environments, and high strength can be produced by using these fiber sources (Rowell et al. 1993; Rowell 1996). Wood is currently the major raw material source in the board industry, but non-wood lignocellulosic sources can work just as well. Residual tea leaves of tea factories provide an abundant fiber source in tea-growing countries (Kacar 1991; Örs and Kalaycioglu 1991; Memed et al. 1992; Kalay et al. 1993). Particleboard manufactured from the industrial wastes of tea leaves (WTLB) has recently shown high biological resistance, possibly due to the high phenolic extractive content of tea leaves (Yalinkiliç et al. 1997a; Kacar 1991). Previous studies on the matter have been conducted by Su et al. (1997a, b) and Yalinkiliç et al. (1997b).

The objective of this study was to determine the effects of boron, either added to the adhesive or as a surface treatment, on fire retardance and other properties of composite board made from waste tea leaves.

MATERIALS AND METHODS

Board manufacture and boron treatment

Waste leaves of *Camelia sinensis* L. were obtained from the biggest public sector tea-producing plant of Turkey (CAYKUR). Urea formaldehyde adhesive containing 55% solid was used with the ratio of 10% (w/w), based on the oven-dry weight of the particles. In the first of three series of treatments, boron was added to the UF as boric acid (BA) and borax (BX) at final concentrations of 0.25, 1.00, and 4.70% (w/v) and a mixture of 5:1 (BA:BX, w/w) with the same concentration levels, representing low, moderate, and high loading of the chemicals as generally required for fungal, insecticidal, and fire resistance (Yalinkiliç et al. 1996; Drysdale 1994). Ammonium chloride and paraffin wax were also added to the glue at 1% from the 33% and 40% concentrations of original solutions, respectively. Control boards were manufactured at three density levels, 0.55, 0.65, and 0.75 g/cm³, while boron added boards were of medium density (0.65 g/cm³). Boards made with the boron addition to UF resin were labeled "boron-added" boards. The thickness of all boards was adjusted to 8 mm at the press. Pressing was ap-

TABLE 1. *Codes of the treatment groups at experimental design.*

Boron compound added to UF resin	Final concentration in the resin solution %	Board density g/cm³	Surface coating type		
			Non-coated	TMM-coated	TMM + boron-coated
Untreated	—	0.55	1-1/2-1/3-1	1-2/2-2/3-2	1-3/2-3/3-3
	—	0.65	4-1/5-1/6-1	4-2/5-2/6-2	4-3/5-3/6-3
	—	0.75	7-1/8-1/9-1	7-2/8-2/9-2	7-3/8-3/9-3
Boric acid (BA)	0.25	0.65	A-1	A-2	A-3
Borax (BX)	0.25	0.65	B-1	B-2	B-3
BA + BX	0.25	0.65	C-1	C-2	C-3
BA	1.00	0.65	D-1	D-2	D-3
BX	1.00	0.65	E-1	E-2	E-3
BA + BX	1.00	0.65	F-1	F-2	F-3
BA	4.70	0.65	G-1	G-2	G-3
BX	4.70	0.65	H-1	H-2	H-3
BA + BX	4.70	0.65	I-1	I-2	I-3

Note: Each code represents 4 boards manufactured with a size of 565 × 565 mm and 8-mm thickness; TMM: trimethylolmelamine.

plied by 20–22 kp/cm² at 150 °C for 5 min. Experimental design is summarized in Table 1.

In the second series of treatments, trimethylolmelamine (TMM) was applied to the untreated and boron-added boards' surfaces by brushing. Accordingly, an amount corresponding to 50 g/m² 40% aqueous TMM solution was brushed onto the boards' surface; and after one day's air drying at ambient temperature, curing was done at 120°C overnight.

In the third series of treatments, BA+BX mixture (5:1, w/w) was added to TMM solution at the 4.70% concentration level (w/v). The amount of the applied surface coating solution was adjusted by continuous weighing of the specimens during brushing. Higher density boards needed an extra brushing for the same amount of coating.

Testing of fire retardancy

A number of WTLB panels were tested according to the Japanese Industrial Standard JIS A 1322 (1982). 210-mm-long and 70-mm-wide board panels 8 mm thick were clamped with steel holders and exposed to the heating flame at an angle of 45 degrees (Fig. 1). Liquefied petroleum gas was used as the heat source. The heating flame was adjusted to a height of 65 mm, and was applied for 13 min, based on preliminary tests in which the flame penetrated the other surface of the control pan-

els. The times for the panels to be charred and ignited were recorded. The post-flaming time (i.e., the duration of continued burning after the removal of flame source) was recorded. Flame penetration through the board, weight loss due to combustion, and char length were determined. Char length was calculated as the mean value of measurements of the exposed and unexposed surface charring dimensions by width and length of each specimen. Char was considered as decomposed parts of test boards after combustion, in this study, rather than superficial charred area, since almost all boron-added boards showed this kind of early charring.

Exposed surfaces of some representative panels were photographed after combustion to illustrate their general appearance.

Thickness swelling (TS), water absorption (WA), and mechanical tests

To reveal the effect of boron addition to UF resin on TS and WA levels of WTLB, boron-added and non-added boards were subjected to relevant tests. All the boards were conditioned at 18–22°C and 60–70 RH for 6 weeks. The TS and WA of boards were determined according to the Japanese Industrial Standard JIS A 5908 (1994) for 2 and 24 h. Specimen size was 50 × 50 mm with the given thickness. Twelve replications were made for each

FIG. 1.   Testing of fire retardance according to JIS A 1322.

board type for TS and WA determinations. Specimens' edges were sealed with an epoxide resin prior to these tests. British Standard BS 5669 (1979) was followed for the static bending and internal bond resistance (SB and IB) tests by using 300-mm-long and 50-mm-wide and, 50- × 50-mm sized specimens, respectively, with the given thickness. Six replications were made for each board type for SB and IB determinations.

RESULTS AND DISCUSSION

Fire retardancy

Post-flaming and flame penetration.—All the tested WTLB panels interestingly did not show post-flaming regardless of boron treatment. This was probably due to urea in the UF adhesive or the inherent combustion behavior of the waste tea leaves owing to high tannin content. Vegetable tannin is used as corrosion and a heat-resistant coating material for metallic surfaces, especially aluminum sheets, in combination with other supplemental chemicals (Reghi 1982; Millet 1985). Interestingly, tea leaves contain almost 10 times as much Al

as higher plants (Kacar 1991), so that possible tannin-Al interaction may also be of importance in the fire-retardant properties of WTLB. Table 2 includes observations of flame penetration through the board, from the exposed to the unexposed surface, at the end of 13-min flaming time.

Flame penetrated the unexposed surface of almost all the untreated boards at three density levels and boron-added boards at the fungal and insecticidal loading concentrations unless coated with the surface fire retardants. Boron-added boards showed great resistance to flame penetration after TMM coating, whereas untreated boards at the same density had no resistance. The TMM delayed the ignition time almost twofold, and char length and WL decreased remarkably with BA (Su et al. 1997a and b; Yalinkiliç et al. 1997b). Relatively late penetration of flame was observed for the untreated boards of 0.75 g/cm³ density, although flame penetrated through all the untreated panels after 13-min exposure. Flame penetration on coating with the TMM-boron mixture and, to a lesser degree with TMM of boards of 0.65

TABLE 2. *Flame penetration of each treatment group after flaming.*

Treat-ment codes	Chemical in the UF solution	Board density g/cm³	Flame-penetrated number of specimens		
			Non-coated	TMM-coated	TMM + boron-coated
1 to 3	—	0.55	All	All	All
4 to 6	—	0.65	All	All	Two third in number
7 to 9	—	0.75	All	All	One third in number
A	BA (0.25%)	0.65	All	All	Two third in number
B	BX (0.25%)	0.65	All	One third in number	None
C	BA + BX (0.25%)	0.65	All	None	Two third in number
D	BA (1.00%)	0.65	All	All	All
E	BX (1.00%)	0.65	All	Two third in number	One third in number
F	BA + BX (1.00%)	0.65	All	One third in number	None
G	BA (4.70%)	0.65	None	None	None
H	BX (4.70%)	0.65	Half in number	None	None
I	BA + BX (4.70%)	0.65	Half in number	One third in number	None

Note: Results reflect observations of 6 specimens belonging to each treatment group subjected to the test (54 × 6 = 324 specimens). For abbreviations refer to Table 1.

and 0.75 g/cm³ density, was reduced. Boron-added boards with surface coatings showed excellent resistance against flame penetration (Table 2). The TMM-boron mixture was much more effective than TMM alone even in boron-added boards. As a consequence, neither separate surface coating nor sole boron addition to the adhesive yielded sufficient flame-proof property to the WTLB panels. Therefore, a combination of both treatments appeared to be necessary to improve flame-resistance.

Glowing

Glowing time (GT) duration after closing off the flame source is given in Tables 3 and 4. Glowing time decreased with increasing board density (Table 3). At lower density, hollow spaces in the board might provide sufficient air for internal combustion, though air is

TABLE 3. *Glowing time after 13 min flaming of the WTLB manufactured without boron addition to UF resin.*

Board density (g/cm³)	Glowing time (min)		
	Non-coated Mean ± SD	TMM-coated Mean ± SD	TMM + boron-coated Mean ± SD
0.55	41.6 ± 5.5	32.2 ± 3.5	24.7 ± 11.9
0.65	12.6 ± 4.7	11.7 ± 2.9	16.3 ± 5.1
0.75	8.7 ± 1.2	13.4 ± 2.0	12.3 ± 1.1

Note: SD; Standard deviation. For abbreviations refer to Table 1.

known as a good insulator against heat transfer in large-scale structural boards (Slifka 1997). In the present case, boards at high densities did not allow further glowing under the charred layer. Void spaces within the boards are not desired for decay resistance. Mycelial mass of decay fungi fills such spaces and begins to severely damage surrounding parts (Imamura 1993). However, one advantage of the low density (0.55 g/cm³) that appeared in this study is the easy application of surface coating by quick absorption and drying of the brushed paste, which enables use of more coatings when required and fixation of a film layer with deeper penetration. Glowing time

TABLE 4. *Glowing time after 13 min flaming of the WTLB manufactured with boron addition to UF resin.*

Board type	Glowing time (min)		
	Non-coated Mean ± SD	TMM-coated Mean ± SD	TMM + boron-coated Mean ± SD
Control	9.3 ± 3.3	10.0 ± 2.8	15.0 ± 6.0
A	6.4 ± 2.0	15.0 ± 3.5	11.0 ± 2.8
B	14.3 ± 4.9	11.0 ± 3.0	6.0 ± 3.4
C	10.0 ± 0.6	7.0 ± 1.0	6.0 ± 1.3
D	10.0 ± 0.9	20.0 ± 0.2	12.0 ± 0.8
E	27.0 ± 2.8	25.0 ± 1.0	23.0 ± 1.5
F	13.0 ± 1.6	10.0 ± 1.9	12.0 ± 3.0
G	11.0 ± 4.4	23.0 ± 3.0	13.0 ± 3.7
H	34.5 ± 6.2	23.0 ± 3.8	23.0 ± 3.1
I	14.3 ± 1.0	9.0 ± 1.5	5.0 ± 0.4

Note: For codes and abbreviations refer to Tables 1 and 2.

TABLE 5. *Weight loss and char length caused by combustion of the WTLB manufactured without boron addition to UF resin.*

Board density (g/cm³)	Weight loss (%)			Char length (mm)		
	Non-coated Mean ± SD	TMM-coated Mean ± SD	TMM + boron-coated Mean ± SD	Non-coated Mean ± SD	TMM-coated Mean ± SD	TMM + boron-coated Mean ± SD
0.55	23.3 ± 2.2	18.7 ± 3.4	18.6 ± 4.0	74.1 ± 7.9	63.4 ± 3.7	59.1 ± 10.6
0.65	17.8 ± 3.0	16.4 ± 1.6	13.1 ± 5.6	44.6 ± 2.6	43.8 ± 8.7	33.8 ± 21.3
0.75	17.8 ± 0.6	14.4 ± 0.9	13.5 ± 0.8	48.8 ± 2.6	43.2 ± 7.9	24.2 ± 2.3

Note: For codes and abbreviations refer to Tables 1 and 2.

was considerably shorter for TMM and TMM-boron coated boards than non-coated ones. This result is consistent with those of a previous study (Su et al. 1997a). As for the boron-added boards, BA acted to shorten the GT, while BX tended to extend it (Table 4). Borax generally reduces flame spread but promotes smoldering or glowing and BA suppresses smoldering but has little effect on flame spread (LeVan and Tran 1990; Yalinkiliç et al. 1997b, c). The mixture of BA and BX reduced glowing after flame in the present study.

On the other hand, surface coating of boron-added boards reduced the GT. Trimethylol-melamine and TMM-boron appeared more effective when they were applied on the boards manufactured with the BA+BX addition. Since they are also effective in slowing down boron leaching from coated plywood surface (Su et al. 1997b), a combination of boron addition to resin solution with the boron containing surface coating would be preferable.

Weight loss (WL) and char length

Weight loss and char length due to combustion are given in Tables 5 and 6. Lower density boards (0.55 g/cm³) without boron addition lost 20–25% their original weights (Table 5). With reference to the WL levels of the boards without boron addition but coated, TMM-boron mixture coating was more effective than only TMM for the 0.65 and 0.75 g/cm³ density boards. High-density boards with the TMM-boron coating yielded the least char formations (Table 5). However, WL could only be reduced by 5 to 10% with the untreated high-density boards compared to the non-coated ones.

Boron addition to the resin, as expected, resulted in a remarkable decrease in WL of the boards (Table 6). More importantly, BA-added boards were least affected by combustion followed by BA+BX. Borax addition could not keep WL levels much lower than those of con-

TABLE 6. *Weight loss and char length caused by combustion of the WTLB manufactured with boron addition to UF resin.*

Board type	Weight loss (%)			Char length (mm)		
	Non-coated Mean ± SD	TMM-coated Mean ± SD	TMM + boron-coated Mean ± SD	Non-coated Mean ± SD	TMM-coated Mean ± SD	TMM + boron-coated Mean ± SD
Control	20.0 ± 3.8	16.2 ± 4.9	17.7 ± 3.2	43.1 ± 4.9	38.5 ± 8.0	54.0 ± 13.6
A	13.4 ± 4.4	12.4 ± 1.1	12.7 ± 5.0	46.8 ± 6.3	32.7 ± 8.6	26.8 ± 11.0
B	13.8 ± 6.5	9.8 ± 1.2	11.0 ± 4.1	43.4 ± 16.0	17.1 ± 5.5	11.0 ± 3.3
C	13.4 ± 3.5	11.2 ± 0.6	14.9 ± 4.0	34.5 ± 5.4	5.0 ± 0.6	20.0 ± 8.8
D	12.3 ± 3.5	13.0 ± 1.9	14.3 ± 6.8	34.5 ± 11.0	31.0 ± 4.3	28.0 ± 2.7
E	14.7 ± 4.2	12.8 ± 1.0	13.9 ± 5.5	49.2 ± 3.5	48.4 ± 4.8	25.1 ± 4.2
F	14.6 ± 2.8	11.0 ± 2.0	11.8 ± 2.0	42.1 ± 1.8	30.2 ± 2.2	3.0 ± 1.0
G	11.1 ± 1.0	12.7 ± 3.8	12.6 ± 4.7	3.0 ± 1.5	3.0 ± 1.8	3.0 ± 1.2
H	16.4 ± 2.5	16.9 ± 1.3	13.9 ± 6.0	30.9 ± 8.7	15.0 ± 4.9	5.2 ± 1.7
I	12.6 ± 0.4	11.3 ± 2.6	11.3 ± 1.5	12.6 ± 3.3	7.0 ± 2.7	3.0 ± 1.0

Note: For codes and abbreviations refer to Tables 1 and 2.