THE EFFECTS OF BORON COMPOUNDS AND DIFFERENT MELAMINE CONTENTS IN MUF RESINS ON SOME PROPERTIES OF MDF PANELS

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The objective of this study was to determine the effects of some boron compounds, which have fire retardant properties, as well as melamine-urea-formaldehyde (MUF) resins having different melamine contents (10%, 15%, and 20%) on some physical and mechanical properties of medium density fiberboard (MDF) panels. It was found that the water absorption (WA) and thickness swelling (TS) of MDF panels increased depending on types and concentrations of boron compounds. However, the WA and TS values of MDF panels decreased with increasing melamine content in MUF resins. It was also found that the modulus of rupture (MOR) and internal bond strength (IB) of MDF panels showed different trends depending on the experimental parameters. Boron compounds showed some negative effects on the MOR and IB values. However, these effects decreased with increasing melamine content in MUF resins. The best results were obtained in MDF panels manufactured with an MUF resin having 20% melamine content. Consequently, increasing melamine content in MUF resins showed positive effects on some properties of MDF panels.

Keywords: Boron compounds; MUF; MDF; Physical properties; Mechanical properties

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INTRODUCTION

In recent years, the use of wood-based panels such as fiberboard, particleboard, oriented strandboard, plywood, etc., for several applications has been gaining great importance due to their suitable properties.

In particular, wood-based panels are widely used as structural parts for the construction of buildings, many furniture applications, and transport industries. Products to serve these applications are in the form of panels and boards (Grexa et al. 2003). However, it is known that wood has some undesired properties. It is a biodegradable and combustible material. Furthermore, its dimensions change with varying moisture contents (Rowell et al. 1984). The flammability of wood is one of the main drawbacks, affecting many common applications of wood-based products in everyday life.

A number of effective protective chemicals and various methods are widely used to achieve desired properties in wood and wood-based panels, and minimize their drawbacks. One of the important classes of protective chemicals is boron compounds, and they have been known as effective fire retardant (FR) chemicals (Yalinkilic 2000). In particular, borates such as boric acid, borax, or disodium octaborate tetrahydrate have strong efficiency as wood preservatives. Furthermore, boron compounds have been used for many years to protect wood against to wood-destroying organisms such as insects and fungi (Lyon et al. 2007). They have been preferred due to their favorable properties such as cost-effectiveness, lack of color, lack of odor, ease of application, non-flammability, non-corrosiveness, neutral pH, preservative effectiveness, less effect on the mechanical properties than some other fire retardant chemicals, low mammalian toxicity, low volatility, minimal effects on environment, and fire retardancy properties at higher loading (Yalinkilic 2000; Kartal et al. 2004, 2007; Baysal and Yalinkilic 2005; Baysal et al. 2007a).

Many studies have been carried out referring to the effects of boron compounds on the mechanical, physical, biological properties and fire resistance of wood and wood-based panels (Laks and Manning 1995; Tsunoda et al. 2002; Akbulut et al. 2004; Donmez 2005; Ayrilmis 2007; Kartal et al. 2007; Baysal et al. 2007a,b; Usta et al. 2009).

It was reported that some fire retardant (FR) treatments and chemicals may cause undesirable secondary effects in wood. The reduced strength properties and increased moisture content are some of these undesirable effects. Especially, acidic fire retardant chemicals may cause strength losses in wood at elevated temperatures (LeVan and Winandy 1990). Furthermore, it was reported that traditional fire retardant chemicals are inorganic salts and they cause high moisture contents in wood products (Östman et al. 2001). Wood treated with inorganic salts is more hygroscopic than untreated wood. The hygroscopic behavior changes depending on some factors such as type of chemical, level of chemical retention, and species of wood (Tomak et al. 2011).

When the wood-based panels are subjected to the fire retardant treatments there should also be considered the influence of these fire retardant chemicals on the bonding performance of the resin used. It was reported that water-soluble borates have some effects on the bonding properties of resins. They show some or no effect on the bonding performance of urea formaldehyde, but, on the contrary, show adverse effect on the bonding performance of phenol-formaldehyde resins (Laks and Manning 1995).

Urea-formaldehyde (UF) resin is one of the important resins for wood-based panels because of its low cost, perfect intrinsic cohesion, ease of application. However, it has also main drawbacks such as their lack of water and weather resistance (Aydin et al. 2006). To overcome the drawbacks of UF resins, some different approaches have been applied over the years. The most widely preferred of these approaches is the preparation of melamine-urea-formaldehyde (MUF) resins (Prestifilippo et al. 1996). MUF resins have been used successfully in the manufacturing of wood-based panels such as particleboard, medium density fiberboard, plywood, etc. They have higher bond qualities than those of urea-formaldehyde resins (Tohmura et al. 2001).

The objectives of this study were: (1) to determine the changes occurring in the physical and mechanical properties of MDF panels manufactured with some boron compounds that are widely used as fire retardant chemicals, and MUF resins having different melamine contents, (2) to reduce some adverse effects caused by the use of boron compounds at high concentration, (3) to evaluate the possible synergistic effects between boron compounds and MUF resins, and (4) to determine the efficacy of sodium perborate tetrahydrate (SPT), which is generally used in bleaching formulations and added to detergents directly or by converting to another form (Yüksel et al. 1996), on some properties of MDF panels.

MATERIALS AND METHODS

Chemicals and Experimental Parameters

Boric acid (BA), borax (BX), zinc borate (ZB), and sodium perborate tetrahydrate (SPT), a mixture of boric acid-borax (BA+BX), and a mixture of boric acid-sodium perborate tetrahydrate (BA+SPT) were used as chemical agents in this study. BX and SPT were supplied by Eti Mine Works General Management (Turkey). The melamine-urea-formaldehyde (MUF) resins were provided by Polisan Company (Turkey).

In this study, all boron compounds were used at 5%, 10%, and 15% concentrations. The mixture of BA+BX, and mixture of BA+SPT were prepared in the ratio of 60:40(w/w). Three different melamine-urea-formaldehyde resins (MUF-1: having 10% melamine, MUF-2: having 15% melamine, and MUF-3: having 20% melamine) were used for panel manufacturing. These MUF resins were chosen according to some pre-experiments.

In this study, three experimental panel groups were designed with MUF-1, MUF-2, and MUF-3 resins. Only a 10% concentration of boron compounds was taken as a constant concentration for MUF-1 and MUF-2 groups in order to evaluate the effects of boron compounds and types of MUF resins on some physical and mechanical properties of MDF panels.

Three different concentrations (5%, 10%, and 15%) of boron compounds were taken for only MUF-3 resin group, and the comparisons were made for this group to see the effects of concentrations of chemicals on some properties of MDF panels. These conditions were chosen because this resin group gave the best results on the mechanical and physical tests of MDF panels in pre-experiments. Control panels for all groups were manufactured without boron compounds.

Manufacturing of MDF Panels

In this study, the mixture of hardwood fibers (%90) and softwood fibers (10%) supplied from Çamsan A.Ş. (Turkey) were used as raw material. Three different concentrations (5%, 10%, and 15%) of boron compounds based on oven-dry fiber weight were used for treatments. The solutions of chemicals were prepared, and fibers were kept in these solutions until they had completely absorbed the solutions. Then, the wet fibers were dried in a laboratory oven until reaching 2-3% moisture contents. After drying, the fibers were glued with melamine-urea-formaldehyde resins, and MDF mats were formed. These mats were pressed at a temperature of 190 ^{o}C for 8 min in a computer-controlled press and conditioned in a climate room until they reached equilibrium moisture content. After climatizing of MDF panels, the test and control specimens were cut from these panels to determine some physical and mechanical properties.

Physical and Mechanical Properties

Some physical and mechanical properties of MDF panels were tested according to EN 310 (1993), EN 317 (1993), and EN 319 (1993) standards. The experiment results were evaluated with ANOVA and Duncan test using SPSS-program.

RESULTS AND DISCUSSIONS

Physical Properties

The average values and statistical evaluation of physical properties results (water absorption and thickness swelling for 24 h) of MDF panels are represented in Tables 1 and 2. Furthermore, the density values of control panels and MDF panels manufactured with boron compounds were found to be between 800 and 829 kg/m^3 for MUF-1, MUF-2, MUF-3 resin groups.

Table 1. Average Values, Statistical Test Results for Water Absorption (WA) of MDF Panels

WA(%)							
	MUF-1	MUF-2	MUF-3			**Statistical Evaluation**	
Chemicals	10% conc. of chemicals		5% conc. of chem.	10% conc. of chem.	15% conc. of chem.	**Effects**	**Sign.**
BA	38.10(b)	38.07(c)	30.65(a)	34.85(b)	48.47(d)	Types of chemicals (X)	***
BX	44.04(d)	40.95(d)	35.47(b)	39.46(c)	56.58(e)	Conc. of chemicals (Y)	***
SPT	45.55(e)	41.55(d)	37.81(c)	40.94(c)	55.51(e)	Interaction(X-Y)	***
ZB	56.39(f)	55.53(e)	42.57(d)	54.27(d)	62.47(f)		
BA+BX	38.29(b)	36.85(b)	31.13(a)	34.25(b)	44.98(b)	Types of chemicals (X)	***
BA+SPT	40.01(c)	38.59(c)	30.80(a)	35.40(b)	46.68(c)	Types of MUF (Z)	***
Control	33.38(a)	32.59(a)	30.93(a)	30.93(a)	30.93 (a)	Interaction (X-Z)	***

"a-f" represent a ranking from lowest to highest values for homogeneity groups (HG).
HG groups indicate significant difference by Duncan's mean separation test.
*** Significant at P<0.001, ns: not significant

As can be seen from Table 1, the WA values of MDF panels manufactured with boron compounds were found to be higher than the WA values of control panels. The WA values showed differences depending on types of boron compounds, concentrations of boron compounds, and types of MUF resins. When comparing the WA values of MDF panels manufactured with 10% concentration of boron compounds for all MUF resin groups, it was found that the highest WA values were obtained with ZB chemical, and MUF-1 resin. The WA values were slightly reduced for MDF panels manufactured with MUF-2 and MUF-3 resins. The WA values especially showed a decrease with increasing melamine content in MUF resins. The best results were obtained with MUF-3 resin for 10% concentration of chemicals.

Table 1 also shows the changes in the WA values of MDF panels for MUF-3 resin group based on concentrations of chemicals. The WA values for this resin group increased depending on increasing chemical concentrations. Actually, this is an expected result due to structure of boron compounds. It is known that the hygroscopic nature of some boron salts used as wood preservative chemicals may show negative effect on dimensional stability of wood. They can likely increase water sorption of wood (Yalinkilic 2000; Baysal et al. 2007b). Therefore, the higher the application amounts of boron salts, the greater were the increases in the WA values of MDF panels. The higher values were determined at 15% concentration of ZB, BX, and SPT chemicals for MUF-3 resin group.

Furthermore, according to the statistical evaluation results represented in Table 1, the effects of types of chemicals, concentrations of chemicals, types of MUF resins, and interactions between these parameters on the water absorption of MDF panels were found to be statistically significant.

Table 2. Average Values, Statistical Test Results for Thickness Swelling (TS) of MDF Panels

TS(%)							
	MUF-1	MUF-2	MUF-3			**Statistical Evaluation**	
Chemicals	10% conc. of chemicals		5% conc. of chem.	10% conc. of chem.	15% conc. of chem.	**Effects**	**Sign.**
BA	15.74(b)	14.66(b)	9.42(a)	15.72(b)	25.42(c)	Types of chemicals (X)	***
BX	20.11(d)	19.27(d)	14.10(b)	17.27(c)	28.05(d)	Conc. of chemicals(Y)	***
SPT	22.00(e)	19.26(d)	13.84(b)	18.15(d)	30.16(e)	Interaction(X-Y)	***
ZB	29.46(f)	26.05(e)	17.15(c)	21.61(e)	34.17(f)		
BA+BX	15.32(b)	14.84(b)	9.57(a)	16.34(b)	22.03(b)	Types of chemicals(X)	***
BA+SPT	17.74(c)	16.55(c)	10.22(a)	15.74(b)	22.77(b)	Types of MUF (Z)	***
Control	10.53(a)	10.26(a)	9.59(a)	9.59(a)	9.59(a)	Interaction (X-Z)	***

Table 2 shows that the TS values of MDF panels had a similar trend to WA values of MDF panels. The TS values of MDF panels manufactured with boron compounds were found to be higher than the TS values of control panels. When comparing the TS values of MUF-1, MUF-2, and MUF-3 resin groups for 10% concentration of boron compounds, it was found that the highest values were obtained from MUF-1 group, and with ZB chemical. Some decreases were observed in the TS values of MDF panels with increasing melamine content in MUF resins. The lowest values were obtained from the MUF-3 resin group for 10% concentration of chemicals. It may be clearly seen that increasing melamine content provided a positive effect on the TS values.

Table 2 also shows the changes of TS values based on concentrations of chemicals for MUF-3 resin group. It may be observed that the TS values for MUF-3 resin group increased with increasing chemical concentrations. The highest TS values were determined at 15% concentration of ZB chemical, whereas the lowest TS values were obtained at 5% chemical concentration of BA chemical.

According to the statistical results in Table 2, it was found that the effects of all experimental parameters on the thickness swelling of MDF panels were statistically significant.

The WA and TS values were evaluated together; it was found that the WA and TS values of MDF panels manufactured with boron compounds were higher than the values of control panels. The WA and TS values increased with increasing chemical concentrations. The results in this study were found to be consistent with the explanations stated in literature. It has been reported that most fire retardant chemicals are inorganic salts and water-soluble (LeVan and Winandy 1990; Denizli 1997). These salts are hygroscopic. Therefore, they increase moisture contents in wood products (Östman et al. 2001). The types of chemical, levels of chemical retention are some of affecting factors for increasing equilibrium moisture content (EMC) of wood (Denizli 1997). Akbulut et al. (2004) found that the WA and TS values of fiberboard specimens treated with boric acid, and borax were higher than those of control specimens.

In this study, even though all chemicals caused some increases in the WA and TS values of MDF panels, these increases were not found to be considerably higher. It is noticeable that some improvements occurred in these values depending on increasing melamine content in MUF resins. These results could be attributed to properties of melamine, and possible positive interactions between experimental parameters.

It has been reported that the addition of melamine to UF resins provide better performance to improve the low resistance of UF bonds to humidity and water (Dunky 1998; Aydin et. al 2006).

Mechanical Properties

The average values and statistical test results for the modulus of rupture (MOR) and internal bond strength (IB) of MDF panels are represented in Tables 3 and 4, respectively.

Table 3. Average Values, Statistical Test Results for Modulus of Rupture (MOR) of MDF Panels

MOR(N/mm^2)							
	MUF-1	MUF-2	MUF-3			**Statistical Evaluation**	
Chemicals	10% conc. of chemicals		5% conc. of chem.	10% conc. of chem.	15% conc. of chem.	**Effects**	**Sign.**
BA	30.23(c)	31.02(b)	30.85(bc)	30.52(b)	32.95(c)	Types of chemicals (X)	***
BX	27.89(b)	27.61(a)	29.99(ab)	30.14(b)	24.41(b)	Conc. of chemicals (Y)	***
SPT	25.96(a)	26.67(a)	27.98(a)	26.75(a)	22.15(a)	Interaction(X-Y)	***
ZB	25.66(a)	28.27(a)	31.01(bc)	30.74(b)	22.61(ab)		
BA+BX	33.33(e)	34.01(c)	33.11(cd)	34.36(c)	36.13(d)	Types of chemicals (X)	***
BA+SPT	31.35(d)	33.05(c)	32.72(cd)	33.94(c)	34.70(cd)	Types of MUF (Z)	***
Control	31.25(d)	32.73(c)	34.59(d)	34.59(c)	34.59(cd)	Interaction (X-Z)	***

As can be seen in Table 3, the MOR values changed depending on types of chemicals, concentrations of chemicals, and types of MUF resins. All MOR values of MDF panels manufactured with boron compounds were found to be between 22.15 and 36.13 N/mm^2. The control values were found as 31.25 N/mm^2 for MUF-1, 32.73 N/mm^2 for MUF-2, and 34.59 N/mm^2 for MUF-3 resin groups (in Table 3). Most of MOR values for MDF panels manufactured with boron compounds were found to be lower than the MOR values of control panels. This could be due to structural properties of the chemicals used in this study. The fire retardant chemicals are inorganic salts. These salts are acidic or basic. Therefore, some reductions in wood strength properties may occur through cellulose hydrolysis (Rowell et al. 1984).

When comparing the effects of MUF resins, it was observed that the MOR values of MDF panels manufactured with 10% concentration of boron compounds improved with increasing melamine content in MUF resins. The best results were obtained with MUF-3 resin. Specifically, the highest values were determined from MDF panels manufactured with mixture of BA+BX. Similarly, the highest values were obtained with a mixture of BA+SPT. This trend could be based on good interactions between boron compounds used in these mixtures, and MUF resins.

Returning to data in Table 3, it can be seen that the MOR values of MDF panels manufactured with 5%, 10%, and 15% concentrations of boron compounds for MUF-3 resin group were found similar to, or lower than the control values. The MOR values showed variability depending on concentrations of chemicals and types of chemicals. It was found that some of these values declined with increasing chemical concentrations.

As can be seen from Table 3, the effects of types of chemicals, concentrations of chemicals, types of MUF resins, and interactions between these parameters on the modulus of rupture of MDF panels were found to be statistically significant.

Table 4. Average Values, Statistical Test Results for Internal Bond Strength (IB) of MDF Panels

IB(N/mm^2)							
	MUF-1	MUF-2	MUF-3			**Statistical Evaluation**	
Chemicals	10% conc. of chemicals		5% conc. of chem.	10% conc. of chem.	15% conc. of chem.	**Effects**	**Sign.**
BA	0.77(b)	0.83(b)	0.82(b)	0.87(bc)	0.83(d)	Types of chemicals (X)	***
BX	0.68(a)	0.71(a)	0.74(a)	0.71(a)	0.64(c)	Conc. of chemicals (Y)	***
SPT	0.69(a)	0.73(a)	0.74(a)	0.73(a)	0.59(b)	Interaction(X-Y)	***
ZB	0.65(a)	0.71(a)	0.75(a)	0.72(a)	0.54(a)		
BA+BX	0.85(c)	0.87(b)	0.85(bc)	0.90(c)	0.89(e)	Types of chemicals (X)	***
BA+SPT	0.84(c)	0.83(b)	0.82(b)	0.84(b)	0.83(d)	Types of MUF (Z)	***
Control	0.82(c)	0.87(b)	0.88(c)	0.88(c)	0.88(e)	Interaction (X-Z)	ns

As can be seen from Table 4, the IB values changed depending on types of MUF resins, and types of chemicals. It was found that, generally, the IB values of MDF panels manufactured with boron compounds were lower than the IB values of control groups. When comparing the IB values of MUF resin groups for 10% concentration of boron compounds, it was found that these values slightly increased with increasing melamine content in MUF resins. The higher values were obtained with the MUF-3 resin group.

Furthermore, it may be observed from Table 4 that the IB values of MDF panels for MUF-3 resin group changed depending on concentrations of chemicals. Increasing concentrations of boron compounds negatively affected the IB values, and some values slightly decreased. The lower IB values were obtained with 15% concentration of ZB, SPT, and BX chemicals.

As can be seen from the statistical results in Table 4, the effects of types of chemicals, concentrations of chemicals, and their interaction on the internal bond strength of MDF panels were found to be statistically significant, but interaction between types of chemicals and types of MUF resins was found to be statistically insignificant.

In literature, it has been reported that fire retardant chemicals can cause some reductions in strength properties of wood (Rowell et al. 1984; LeVan and Winandy

1990). Our results were found consistent with the literature reports. In this study, some reductions were observed in the mechanical properties. The MOR and IB values of MDF panels decreased to some extent with increasing concentrations of boron compounds.

Laks and Manning (1995) reported that the IB values of waferboard bonded with pMDI adhesive reduced with increasing content of zinc borate used in the board manufacture. Donmez (2005) found that the MOR and IB values of oriented strandboard (OSB) manufactured with two different adhesives decreased with increasing amount of zinc borate in the treated OSB panels.

In this study, even though boron compounds were used especially at high concentrations for treatments, it was found that the values of mechanical properties did not drastically decrease. Some improvements in the mechanical properties were obtained with increasing melamine content in MUF resins. This could be related to properties of melamine, and coherent interactions between boron compounds and MUF resins.

CONCLUSIONS

1. The types of boron compounds, concentrations of boron compounds, types of MUF resins, and interactions between these parameters were found to have considerable effects on some physical and mechanical properties of MDF panels.
2. The WA and TS values of MDF panels, in particular, increased with increasing chemical concentrations. The WA and TS values of MDF panels were found to be higher than the values of control panels. The highest WA and TS values were obtained with ZB chemical. Some improvements in these values were provided with increasing melamine content in MUF resins. In particular, the best results were determined at 5% and 10% concentrations of chemicals for the MUF-3 resin group.
3. The MOR and IB values of MDF panels showed different trends depending on experimental parameters. These values generally decreased with increasing chemical concentrations and decreasing melamine content. The lowest MOR and IB values were obtained from the MUF-1 resin group for 10% concentration of boron compounds. However, these values slightly improved with increasing melamine content in MUF resins. In particular, good results were obtained with mixture of BA+BX, and mixture of BA+SPT at 5%, and 10% concentrations for MUF-3 resin group.
4. Furthermore, the efficacy of SPT for the physical and mechanical properties was found similar to the efficacy of BX, and the close values were obtained with both these chemicals.
5. Consequently, the negative effects of boron compounds used at high concentration were reduced to some extent, and some improvements were achieved on some physical and mechanical properties of MDF panels with increasing melamine content in MUF resins. Positive synergistic effects were obtained between experimental parameters.

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