Technology, 14(4), 107-114, (2011)

TECHNOLOGY

EFFECT OF SODIUM BORATE SOLUTION ADDED IN THE ADHESIVE LINE AND USED AS IMPREGNATION MATERIAL ON COMBUSTİON PROPERTIES OF LAMINATED LIMBA WOOD (LVL)

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Abstract

In this study, Limba (*Terminalia superba* Engl. & *Diels*) wood laminated with melamine formaldehyde adhesive containing the additive of sodium borate solution is impregnated with sodium borate solution and its combustion properties are searched. For this aim; papel coatings obtained from Limba wood were pasted with melamine formaldehyde adhesive containing the additive of sodium borate solution in a ratio of 2.5% of the adhesive solid material amount, then test samples were prepared in accordance with the ASTM E 160–50 standard and these prepares samples were impregnated with sodium borate solution in ratios of 5% and 10%, then combustion properties were searched.

Consequently, use of sodium borate solution as an impregnation material by adding the sodium borate solution in the adhesive line has positively affected the combustion properties of all wooden materials. While the highest loss of weight occurred in control samples, the least was obtained from the samples impregnated with 10% solution. In addition, the lowest value in flame sourced combustion (FSC), without flame source combustion (WFSC) and glowing combustion (GC) temperatures was obtained from the samples laminated with modified adhesive and impregnated with 5% sodium borate solution and full collapse did not occur in these samples. As a consequence; sodium borate solution may be used as fire retardant either by being added in melamine formaldehyde adhesive and as an impregnation material in building construction elements with a high fire risk.

Keywords: sodium borate, limba, combustion properties, impregnation

1. Introduction

One of the most negative features of wooden materials is its low resistance against combustion. A number of chemical materials which are combustion preventive and retardant are used together with various efforts in order to remove this negative feature of wooden material. Temperature must increase up to 275 °C to ensure wooden material to burn spontaneously. Moreover, any combustible material may inflame and burn in the existence of a flame source in lower temperatures. Even though wooden material has superior aspects compare to many structural material by means of its structure specific features during a fire, treatment with impregnated materials which increase resistance or retard against combustion become important in terms of ensuring security and preventing or retarding combustion (1,2,3).

Even though there are a number of chemical substances which would increase resistance against combustion or retard inflammation time of wooden material, most of them harms to human health or environment in a certain ratio. For this reason, use of such chemical substances were inhibited or minimized by the pressures of environmental and humanitarian organizations. In addition, alkyl ammonium compounds and boron

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compounds have increasingly become more important as chemical substances which do not harm to both environment and human health. Boron compounds are highly used today due to the characteristics of high degree of effectiveness against biological pests, high solubility with water and applicability, easy penetration in wooden material, requirement of a low cost and easy procurement, very low toxicity impacts against human health and ability of increasing the resistance of wooden material against combustion. (4.5.6.7.)

Laminated material produced from solid wood material is used as an important constructing element and production material in the woodworking industry. It is possible to produce laminated wooden material in desired shape and quality with the lamination technique. Laminated wooden material has significant superiorities in technical and economical sense compare to solid wood material (8).

Wooden material is exposed to environmental factors, biological degradation of bacteria, fungus and insects and chemical deformation effects like fire on site of use. In this regard, impregnation of wooden material with chemical substances is considered obligatory in many places of use in order increase resistance against combustion (9). Wooden material does not burn directly, pyrolysis occurs before combustion. In case the conditions keep on in this way, wooden material continues the pyrolysis until it completely converts into ash as an inorganic residue (10). Temperature must increase up to 275°C to ensure it to inflame spontaneously. Pyrolysis cellulose starts at 350°C. Gases released as a result of pyrolysis react with each other and with oxygen, inflammation and combustion starts after this (11). Resistance against combustion or retard of combustion may be increased in a structure by means of processes to be held in order to make wooden material noncombustible. These processes are also required to prevent spread of flames during a fire (12).

It was determined that aqueous solutions of boric acid and sodium perborate and combinations applied in form of boron salts+water-repellent monomers increase combustion resistance of wood, and water repellent monomers extend the period passed until collapse during combustion (13). It is known that boron compounds improve the combustion properties of wooden material, while it was also exhibited that they succeed in preventing operation of wooden material and washing of impregnated material when used with various resin combinations like melamine formaldehyde (14).

In the study held to determine the effects of sodium borate solution added in the adhesive line on combustion properties in wooden material, papel coatings obtained from Scots Pine (*Pinus sylvestris L.*), Oriental beech (*Fagus orientalis L.*), Sessile Oak (*Quercus petraea Lieble*) and Sweet chestnut (*Castanea sativa Mill*) woods were pasted with Melamine formaldehyde adhesive containing the additive of sodium borate in a ratio of 2.5% of the adhesive solid material amount, then combustion properties were determined. As a result of the study, sodium borate solution added in the adhesive line positively affects the combustion properties of in all wooden materials (15).

Combustion samples prepared from Calabrian Pine (Pinus brutia Ten.) wood were treated with aqueous solutions of calabrian pine bark, valonia oak, sumac leaf and quercus infectoria fruit as vegetative tanning materials and secondly with boron compounds, then their flame sourced, non flame sourced and glowing combustion properties were examined. Consequently, the lowest temeprature level was obtained with 323 ºC in samples treated with the mixture of boric acid and borax in the flame sourced combustion stage, with 404 ºC in samples treated with valonia oak and the mixture of boric acid and borax in the non flame sourced combustion stage and with 107 ºC in samples treated with sumac leaf powder and the mixture of boric acid and borax in the glowing combustion stage (16).

Wooden materials created with combination of beech and poplar trees and laminated with Desmodur-VTKA(Dv) and Polyvinylacetate (PVAc) adhesives were impregnated with Imersol-Aqua (I_{AQUA}), Timbercare-Aqua (T_{AQUA}), Borax(B_X), Boric acid (B_A) and $B_X + B_A$ impregnation substances. Imersol Aqua was impregnated with immersing method, Timbercare-Aqua with brush and Boron compounds with vacuum method, then their combustion properties were examined. As a result of the study, highest temeprature was obtained in Flame sourced combustion (F_{CS}) + T_{AQUA} (528.150 ºC) and the lowest temperature was obtained in non flame sourced combustion $WF_{SC} + B_A$ (391.333 ºC). Consequently, it was reported that the best result is obtained in combustion properties of the samples pasted with Dv adhesive and impregnated with boron compounds and I_{AQUA} (17).

Laminated wooden materials prepared from Uludağ abies as 3 layers with Desmodur-VTKA adhesive were treated with immersing method with sodium perborate, sodium tetraborate, Imersol-WR 2000 and Tanalith-CBC impregnation substances, then their combustion properties were examined. As a result of the study, the

highest loss in mass was determined in solid wood treated with Tanalith- CBC (18).

Samples obtained from scotch pine and beech woods were treated with immersing and pressure method with potassium notrate (KNO_3), zinc sulphate ($ZnSO_4$), sodium tertaborate ($Na_2B_4O_7$ _ $10H_2O$), sodium sulphate (Na_2SO_4) and copper sulphate (Cu_2SO_4) impregnation substances, combustion properties of wooden materials were searched. The most positive results against combustion were obtained from Cu_2SO_4, $ZnSO_4$ and Na_2SO_4 impregnation substances while pressurisation method gives more positive results compare to immersing method (19).

This study aims to determine combustion properties of Limba wood laminated with melamine formaldehyde adhesive modified with sodium borate in a ratio of 2.5% and then impregnated with sodium borate solution in a ratio of 5% - 10%.

2. Material and Method

2.1. Wooden Material

Papel coating with 2 mm thickness obtained by cutting method from imba (Terminalia superba Engl. & Diels) wood with a specific weight of 0.65 g/cm3 in air-dry situation was used as wooden material in this study. Attention was taken to ensure that first class wooden materials selected with random method displayed a steady growth and are riftless, knotless, free of fiber curvature.

2.2. Adhesive

Melamine formaldehyde adhesive commonly used in the industry was used in this study. Sodium borate in a ratio of 2.5% of the dry substance amount of adhesive was added in the adhesive after being solubilized in water.

Melamine formaldehyde adhesive may harden without adding any hardener substance in temperatures of 90–140 ºC. Increase in the solid matter ratio affects resistance against water. Ahesive may display a resistance against boiling water if aving a filling material additive up to 100%, while it displays resistance against water in ambient temperature with an additive between 150-200%. Moroever, resin used in decoration paper impregnation is clear, colorless and in the liquid state, solid matter amount (in weight) is equal to 53±1%, water tolerance is 1/1.2, density is 1.21–1.24 g/cm^3 (20 ^{o}C), viscosity is 40–80 sn/Din cup/4 mm 20 ^{o}C, pH value is 9.0–9.6 (20^{o}C), on-surface application amount is 150–200 g/cm^3, storage period is 15 days, and it hardens in 5-15 minutes at high temperature and press (20).

2.3. Imporegnation material and method of impregnation

Sodium borate (Na2 B10 O16.10H2O) chemical was used as impregnation material in the study. Sodium borate is a chemical containing a high ratio of boron, and has the solubility in a ratio of 16% at 20°C ambient temperature. Being produced as powdered, sodium borate has 700kg /m³ pour density and neutral pH. Its pH may be adjusted in a value between 7–8 if desired. Molecule weight of the compound is 590 g/mol. Boric acid constitutes about 58% of molecular weight of the compound. 5% and 10% distillate water (DW) solutions were prepared in the tests. Chemical substance is ensured to completely dissolve in distillate water without any deposit by means of a mechanical mixer in solution preparations and the solutions were stored in closed cups. Temperature was taken at 20 °C during the course of solution preparation.

2.4. Preparation of Test Samples

Limba papel coatings with 2 mm thickness in air-dry situation were primarily kept in %65±3 relative humidity and 20±2 ºC temeprature to ensure them to reach the equilibrium moist according to TSE–345 (21). Laminated board was obtained by pasting the pieces cut from limba papel coatings in draft in size of 2x200x1030 mm with 2.5% modified and non-modified melamine formaldehyde adhesive in 7 layers. Adhesive was applied on pasting surfaces in average 180 g/m^2. Press compression was applied on test samples at 80°C and 1.4 N/mm^2 pressure for 20 minutes. Then, combustion test samples in a size of 13 x 13 x 76mm were prepared from the drafts in a size of 14x200x1030mm according to the principles of ASTM – E 160 – 50 (22). The test samples prepared were kept in air conditioning cabinet until they reach 12% equilibrium moist.

2.5. Impregnation method

The test samples prepared were impregnated with 5% and 10% sodium borate (Na_2 B_{10} $O_{16}.10H_2O$ or $Na_2O.5(B_2O_3).10H_2O$) solution by complying with the principles of ASTM D 1413–76 (23). For this purpose, the samples were kept in the solution at normal atmospheric pressure for a peirod of 60 minutes after a preliminary vacuum is applied of a period of 60 minutes in a Hg value of 760 mm. The samples with determined weights and sizes in fully-wet situation were dried until they become fully dry at 103±2 ˚C in the drying oven. After the fully-dry measurements of the test samples, Retention (kg/m³) and % Retention rates of the test groups were calculated and combustion tests were applied.

2.6. Conducting the Combustion Test

Combustion test was conducted in accordance with the standard ASTM E 160–50. Test and control samples were brought to the 7% moisture degree recommended in the standard in the air conditioning room where 27 ± 2°C and 30–35% relative humidity atmosphere is regulated before the combustion process. 24 samples were burned by aligning in form of tetragonal prism in 12 layers. Gas pressure was kept steady in a level specified in the standard during the combustion process, combustion test parameters were recorded by measuring for flame sourced, non flame sourced and glowing combustion stages. 15 x 24 = 360 samples were burned in total including 3 groups composed of 24 units as 5 x 3 = 15 groups in each combustion tests.

2.7. Evaluation of the Results

SPSS analysis was applied on the groups among each other in order to determined the effect of impregnation materials on retention amounts, weight losses, combustion temperature and collapse period reported in the combustion test for the samples prepared. DUNCAN test was used in 95% confidence level in order to determine the groups among which the significant differences occurred in results of the variance analysis. The values with the lowest average were displayed in homogeneity groups with "A" letter. The reason of this is that collapse occurs later in the groups with a low weight loss and in low temperature groups.

3. Findings and Discussion

Retention amount of the limba wood as a result of the impregnation applied is given in Table1.

Table1. Retention amount of the limba wood

Type of Tree	Concentration (%)	Kg/m^3 Retention Average	% Retention Average
Laminated limba	5.00	37,845	1,993
	10.00	73,239	6,451

Retention amount was obtained as 73,239 (kg/m^3) in 10% concentration while obtained as 37,845 (kg/m^3) in 5% concentration. When the retention ratios are examined, it occurred as 6.451 (%) in 10% concentration and as 1,993 (%) in 5% concentration. The higher the concentration ratio, the higher the retention amount and ratio has occurred.

Weight losses (%) occurred at the end of the test and standard deviation values for them are given in Table2.

Table2. Weight losses occurred as a result of combustion

Type of process		**Weight loss (%)**	**Standard deviation**	**N**
Adhesive	**Concentration (%)**			
Non modified	Kontrol	87,67	,00577	3
	5.00	86,33	,02082	3
	10.00	68,00	,05196	3
Modified	0.00	83,33	,02309	3
	5.00	81,33	,00577	3
Total		-	,07631	15

When the weight loss results are examined, the highest weight loss occurred as 87,67% in non modified control samples, while the lowest weight loss occurred as 68% in the non modified samples with 10% concentration.

Results of ANOVA conducted according to weight loss results are given in Table3.

Table3. Results of ANOVA conducted according to weight losses

Source of Variance	Sum of Squares	Degrees of Freedom	Average of Squares	F-value	Level of Significance	Level of Importance
Process type	,074	4	,019	24,799	,000	,908
Error	,007	10	,001			
Total	10,004	15				

It was determined that there is a significant relationship between percentage values of ash ratio residued according to process type. It is seen that this relationship is in a high level. Table4 gives the interaction schedule for weight loss, start of collapse, full collapse period and collapse type.

Table4. Results for comparison of weight loss averages according to the interaction of tree type- solution concentration, collapse time and collapse type

Process type		**Weight loss (%)**		**Collapse time (sec)**	**Collapse Type**
Modification type	Concentration (%)	**Ort.**	**HG**		
Non Adhesive Modified	Control	87,7	C	(WFSC) 120*	GC 390 Fully collapsed ***
	5.00	83,3	BC	(WFSC) 210*	Collapse started **
	10.00	68,0	A	GC 270*	Collapse started **
Adhesive Modified	0.00	86,3	BC	(WFSC) 150	GC 450 Fully collapsed ***
	5.00	81,3	B	-	Not collapsed

*First collapse occurred in ... sec beginning from the spontaneous combustion stage.
**Full collapse did not occur even though the collapse started.
***Full collapse occurred in ... sec beginning from the spontaneous combustion stage.

When the table is examined, it is seen that the highest weight losses occured in non modified control samples, and they are respectively followed by modified non impregnated, modified 5% impregnated and non modified 10% concentrations.

It was observed as a result of the study that while the lux (light density) amount is higher in impregnated samples, smoke density is higher and so the lux amount is lower in control samples.

Multiple variance analysis for temperature values in combustion stages is given in Table5.

Table5. Multiple variance analysis for temperature values (°C) in combustion stages

Flame Sourced Combustion						
Source of Variance	**Sum of Squares**	**Degrees of Freedom**	**Average of Squares**	**F-value**	**Level of Significance**	**Level of Importance**
In-group	24416,771	4	6104,193	7,424	,005	,748
Intergroup	8222,683	10	822,268			
Total	32639,453	15				
Without Flame Source Combustion						
Source of Variance	**Sum of Squares**	**Degrees of Freedom**	**Average of Squares**	**F-value**	**Level of Significance**	**Level of Importance**
In-group	52660,150	4	13165,038	6,010	,010	,706
Inter-Groups	21904,408	10	2190,441			
Total	74564,558	15				
Glowing Combustion						
Source of Variance	**Sum of Squares**	**Degrees of Freedom**	**Average of Squares**	**F-value**	**Level of Significance**	**Level of Importance**
In-group	51068,735	4	12767,184	2,463	,113	,496
Inter-Groups	51846,107	10	5184,611			
Total	102914,842	15				

When the table is examined it is seen that type of modification, impregnated solution concentration and dual interactions have significant effects and differences in 95% confidence level on combustion temepratures by the exception of glowing combustion (GC) stage. Average and homogeneity groups are given in Table6 in order to find the factors these differences result from.

Table6. Comparison of combustion stages temeprature (°C) averages and homogeneity groups according to process type interactions

Process Type		**Temeprature (°C)**					
		FSC		**WFSC**		**GC**	
Type of Modification	Concentration (%)	**Avrg.**	**HG**	**Avrg.**	**HG**	**Avrg.**	**HG**
Non adhesive modified	Kontrol	511,4200	B	584,9033	C	337,0700	B
	5.00	425,3033	A	473,2000	AB	210,9933	AB
	10.00	430,7767	A	473,8867	AB	297,4000	AB
Adhesive modified	0.00	424,5267	A	538,3167	BC	286,2633	AB
	5.00	389,3333	A	413,2200	A	178,6700	A

When the table is examined, it is seen that temperatures occurred in flame sourced combustion (FSC) remain between 389-430 °C in all samples other than control samples. This temperature degree caused retard in inflammation. The lowest temperature values were obtained with adhesive modified 5% concentrated laminated wood during the spontaneous combustion (WFSC) stage. It is thought that sodium borate existing in the adhesive between laminated layers is effective in retarding combustion. Temperatures of test samples decreased in control samples in the glowing combustion (GC) stage.

4. Results and Evaluation

In this study; melamine formaldehyde adhesive was modified by adding sodium borate impregnation material in a ratio of 2.5% of the adhesive solid material amount. Then, limba papel coatings were turned into Laminated Board (LVL) by being pasted with modified and non modified melamine formaldehyde adhesive and test samples were prepared in accordance with the standards. Test samples prepared were impregnated with 5% and 10% sodium borate solution, then the tests were conducted in order to determine the physical properties like retention amount, weight loss as a result of combustion and combustion temperatures and periods on the samples.

When the results are evaluated in terms of weight loss, the highest weight loss occurred in control samples. Namely, the samples burned to the greatest extent are control samples. In this respect, use of impregnation material by adding in the adhesive line prevented any weight loss by causing the decline in the period combustion. Destructive effect of fire was eliminated due to the addition of the impregnation material in the adhesive line, so collapse is retarded in the adhesion line. In addition, it was determined that there are disturbances in the adhesive line and degradation occurs in short periods in control and impregnated samples pasted with non modified adhesive. Arslan et al. added sodium borate in adhesive line in scotch pine, sessile oak, sweet chestnut and oriental beech laminated wooden materials and collapse time, weight loss, FSC, WFSC and GC temperature values have parallels with the results found (15).

When collapse results are examined, the best result is given by the sodium borate substance both when added in the adhesive line and used as an impregnation material. Therefore, both adding in the adhesive line and using as impregnation substance would give better results instead of the use of two implementations separately.

The highest values were obtained from control samples in terms of flame sourced combustion (FSC), without flame source combustion (WFSC) and glowing combustion (GC) temepratures. Addition of sodium borate substance in the adhesive line and use of it as an impğregnation substance decreased the temeprature values occurred in the samples. There are a number of studies regarding that boron compounds decrease combustion temepratures, but the results obtained from this study are also compatible with them (24, 25).

One of the factors harming to laminated wooden material used in manu fields in different forms or shapes is fire. For this reason, it is recommended to add sodium borate solution in melamine formaldehyde adhesive in low percentages and use it as an impregnation material and fire retandant in high concentrations while using in fire sensitive areas.

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