IMPACTS OF IMPREGNATION CHEMICALS ON FINISHING PROCESS AND COMBUSTION PROPERTIES OF ORIENTAL BEECH (*FAGUS ORIENTALIS* L.) WOOD

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Effects of the impregnation materials borax, boric acid, and Tanalith-E on combustion properties of Oriental beech (*Fagus orientalis* Lipsky) were investigated. The immersion method was used for long-term impregnation (24 h). After the impregnation process, polyurethane as well as synthetic, acid hardening, and water-based varnishes were applied on the wood sample surfaces according to company's suggests. The combustion test was performed according to the ASTM-E 69 standard. The mass reduction, release of gasses (CO, NO, O_2), and the temperature differences of samples were determined for each 30 seconds during combustion. It was shown that the most mass reduction occurred when both polyurethane varnish and boric acid were applied.

Keywords: Fire Retardant; Combustion; Varnish; Impregnated; Oriental beech

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INTRODUCTION

The wood house construction sector has recently developed in Turkey. As a construction material, wood has a number of advantages such as good mechanical properties, but shows a low resistance against fire and high temperatures (Bednarek and Kaliszuk 2007).

Wood continues to play an important role as a structural material in today's high-tech society. As lumber and in reconstituted products, wood is commonly used for house siding, trim, decks, fences, and countless other exterior and interior applications. When wood is exposed to sunlight and moisture, special precautions must be taken in structural design as well as in the selection and application of the finish (Cassens and Feist 1991).

The combustion of wood relates to the fuel burn rate (or the reaction rate), the combustion product (or the emissions), the required excess air for complete combustion, and the fire temperatures. These processes are extremely complicated, principally because the wood has a complex physical and chemical composition (Scott 1992). Important reactions to fire parameters in the full scale fire are heat release rate and time of flash-over (Babrauskas and Grayson 1992).

Boric acid and borax are the most common boron compounds that have found many application areas in the wood preservation industry. Both can be used to obtain fire retardant characteristics (Hafızoglu et al. 1994).

Uysal et al. (2008) investigated the effects of finishing materials polyurethane, cellulosic, synthetic, polyester, and acid hardening varnish on combustion properties of Scotch pine. Cellulosic varnish was found to be the most successful varnish according to the CO amounts and mass reduction. According to their results, all of the varnishes used to in their study showed a low resistance against fire and high temperatures (Uysal et al. 2008).

At the same time Uysal and Özçifçi (2000) obtained laminated wood, using Uludağ fir for the outer ply; different veneer materials were used for the core ply, and these were bonded with PVAc. The combustion test was applied to the test samples. While the highest mass reduction and concentration of O_2 were observed in white mulberry, the highest heat increase was found when Scotch pine was used in the core ply).

Uysal and Kurt (2005) studied the impregnation of the oriental spruce (*Picea orientalis* L.) with boron compounds, and the test samples were applied to the combustion test. A borax-boric acid 10% solution was found to be the most successful fire retardant chemical.

The present study investigated the combustion and emission properties of samples after impregnation and varnishing processes. As a test material, oriental beech, used in furniture and construction, was used. Firstly, the test samples that were prepared from oriental beech were impregnated with borax, boric acid, or Tanalith-E by means of a dipping method. Secondly, test samples were varnished with polyurethane, synthetic, acid hardening, and water-based varnishes. After these processes, the combustion properties of samples were observed.

MATERIALS AND METHODS

Material

Oriental beech (*Fagus Orientalis* Lipsky.) was chosen randomly from timber suppliers of Karabuk, Turkey. A blending process was carried out in order to prepare representative control samples and other groups. A special emphasis was put on the selection of the wood material. Accordingly, non-deficient, whole, knotless, normally grown (without zone line, reaction wood, decay, insect or fungal infection) wood materials were selected. Then, test samples were cut roughly ($3x22x1030mm^3$).

In this study the impregnation chemicals borax, boric acid, and Tanalith-E at the 5% concentration were used. Tanalith-E is a commercial impregnation material that is used for the protection of softwood and hardwood timber against the fungal decay, borers, and termites.

As varnish materials, polyurethane, synthetic, acid hardening, and water-based varnishes were used according to the producer's instructions. The selection of type, preparation and application system of using varnish are very important to make varnish layers durable against to various effects. Therefore, using materials in the experiments were stored appropriately until their usage to prevent loss of properties. The varnishes were checked to confirm if they had the specified properties in their descriptions. The technical specifications of the conventional varnishes are given in Table 1

Table 1. Some Technical Properties of the Conventional Varnishes

Technical Properties	Varnish Type			
	Synthetic	Polyurethane	Water-Based	Acid hardening
Density (g/cm^3)	0.94-0.95	0.95-0.96	-	0.94-0.96
Viscosity (second/DIN CUP 4 mm/20°)	18	16	18	18
Amount applied (g/m^2)	100	120	300-600	110
Nozzle gap (mm)	-	1.8-2	1.8	-----
Air pressure (bar)	-	2	3	2
Drying type	Physical	Chemical	Chemical	Chemical
Drying time (20°)	6-8 hour	2-3 hour	4 hour	2-3 hour

Impregnation Process

Impregnation processes were applied on surface according to ASTM D 1413-76(1976), TS 344 (1981), and TS 345 (1974) were applied to the prepared test samples. The long-term dipping method was used. In this method, experimental samples were kept immersed in solutions for 24 hours.

Test samples were oven dried before and after impregnation. The amount of retention (R, kg/m³) and ratio of retention (R, %) were calculated as follows,

$$R = \frac{GxC}{V} 10^6 \tag{1}$$

$$R(\%) = \frac{Mdi - Md}{Md} 100 \tag{2}$$

$$G = M_2 - M_1 \tag{3}$$

where G is the mass of the sample after impregnation (M_2, kg) minus the mass of the sample before impregnation (M_1 , kg), Mdi is the dry mass after impregnation (kg), Md is the dry mass before impregnation (kg), V is the volume of the sample (m^3), and C is the concentration of the solution (%).

Experimental Study

The oversized test samples were climatized until they reached constant weight at 20±2 ^{o}C and 65±3 % relative humidity in a climate room. Following the climatization, they were cut with the dimensions of 3x22x1030 mm^3, and then they were layered (9x19x1016 mm^3) according to the ASTM E – 69-02 (1975), by placing them in solution at 20±3ºC were impregnated with borax, boric acid, or Tanalith-E. After the impregnation process of samples, they were finished with polyurethane, synthetic, acid hardening, or and water-based application. In each test, 10 pieces were manufactured. In total, 260 test samples were prepared.

Execution of Test

The combustion test was carried out according to the ASTM E –69-02 (1975) method. But some changes were made relative to the stardard. A digital balance having 0.01 g sensitivity was used for determination of mass reduction of materials when they were burnt. Butane gas was used to make an ignition flame. The gas flow was standard to give a flame height of 25 cm, and the temperature must be 1000°C.

The distance between the bottoms of the test samples, which were hung inside of the fire tube and the top of the gas pipe, were adjusted as 2.54 cm. During the test, mass reduction, temperature and released gases (CO, NO, O_2) were determined each 30 seconds. The test was made under a chimney where the flow of air blown was drawn with natural draft. At the beginning of combustion the test flame source was used for 4 minutes; then the flame source was taken away and it was continued 6 minutes. In total, the test lasted 10 minutes.

Testo 350 M and XL flue gas analyzers were used for measuring concentration of the released gasses (CO, NO, O_2), and temperature variation. The probe was inserted into the first hole from the top of the fire tube.

Statistical Procedure

To determine the retention in both the prepared natural and the impregnated samples, as well as the effects of impregnation material on combustion with or without flame source, multi-variance analysis was applied. Based on Duncan test's being significant, each test group was compared with one another and itself.

RESULTS AND DISCUSSION

Properties of the solutions used in impregnation process are given in Table 2.

Table 2. Characteristics of Impregnation Chemicals

Impregnation Chemicals	Solvent	Solution (%)	pH		Density (g/cm^3)	
			Before Impreg.	After Impreg.	Before Impreg.	After Impreg.
Borax	Pure water	5	9.26	9.26	1.024	1.024
Boric Acid	Pure water	5	5.13	5.49	1.020	1.023
Tanalith-E	Pure water	5	9.24	8.66	1.060	1.060

As a result of using the fresh solution in every impregnation process, there were no important changes in the acidity and density of the solutions before and after the impregnation. Because the pH values of the boric acid solution was in the acidic zone (5.1), it may have had an effect on the polysaccharides in the wood.

Air-Dry Density

The averages of density are given in Table 3.

Table 3. Average Values of Density (gr/cm^3)

Wood Type	Oven dry density (g/cm^3)	Air dry density (g/cm^3)
Oriental beech	0.66	0.72

The proportion of impregnation chemicals is given in Table 4.

Table 4. Proportion of Retention

Impregnation chemicals	Retention (wt %)
Control	-
Borax	2.37
Boric acid	5.72
Tanalith-E	0.80

The highest retention proportion was observed in boric acid and the lowest in Tanalith-E. Dry film thicknesses and solid amounts of varnish types are given in Table 5.

Table 5. Dry Film Thicknesses of Varnish Types

Varnish Type	Synthetic Varnish	Polyurethane Varnish	Water-Based Varnish	Acid hardening Varnish
Solid Amounts	% 52	% 61	% 31	% 46
Dry film Thickness	100µm	113 µm	87 µm	92 µm

The thickness values of the varnish layers were measured with a comparator with sensitivity of 5 µm. The highest dry film thickness of 113 µm was obtained from polyurethane varnish samples. The lowest dry film thickness of 87 µm was obtained from water-based varnish samples.

The solid amounts of the varnish were measured also. The highest solid amount of 61% was obtained from polyurethane varnish samples. The lowest solid amount of 31% was obtained from water-based varnish samples.

Values of Combustion Attributes

Average values based on impregnation chemicals are given in Table 6. According to the variance analysis, while the effect of impregnation materials on the mass reduction of samples was observed significantly at 95% accuracy level, the type of varnishes and interaction of varnishes and impregnation material not significant. Impregnation material, type of varnish and interaction of them on the gases released the combustion test (NO, O_2, CO) were investigated by variance analysis.

The Duncan test was applied in order to determine which of the differences from variance analysis were significant ($p \leq 0.05$), and the results are shown as different homogenous groups in Table 7.

Table 6. Average Combustion Values of Impregnation Chemicals

Type of impregnation chemicals	Varnish Type	Temp. (°C)	CO (ppm)	NO (ppm)	O_2 (%)	Mass reduct. (%)
Control	Control	292.54	2485.89	31.57	17.44	62.07**
	Polyurethane	291.65	1918.05	33.42	17.70	58.37
	Synthetic	293.81	1347.67	47.38	17.39	60.71
	Acid Hard.	290.52	2445.74	39.72	17.44	59.53
	Water B.	303.71	2800.32	44.64	16.78	60,05
Borax	Control	290.67	2427.69	29.48	17.55	60.09
	Polyurethane	291.74	840.59	39.22	18.10	58.91
	Synthetic	293.81	1347.66	47.38	17.39	56.51
	Acid Hard	288.33	2423.71	58.11	17.34	54.33
	Water B.	327.92	3487.36	72.88	15.34	52.86
Boric Acid	Control	300.14	1512.83	30.13	17.30	57.03
	Polyurethane	323.85	284.52	38.25	17.88	47.99*
	Synthetic	319.39	2405.64	48.86	16.53	48.41
	Acid Hard.	290.11	1432.46	44.26	17.56	52.00
	Water B.	333.76	3612.18	65.58	15.68	50.85
Tanalith-E	Control	281.61	2462.73	51.93	17.07	60.34
	Polyurethane	341.78	573.87	57.61	17.66	56.34
	Synthetic	309.88	1756.03	59.68	17.22	58.17
	Acid Hard.	316.35	2788.88	81.08	16.08	57.89
	Water B.	317.92	4794.89	92.72	15.02	58,54

According to Table 7, while the highest mass reduction was observed in massive control samples, the lowest value was from the polyurethane varnish group for samples impregnated with boric acid. The results connected with these values are shown in Fig. 1.

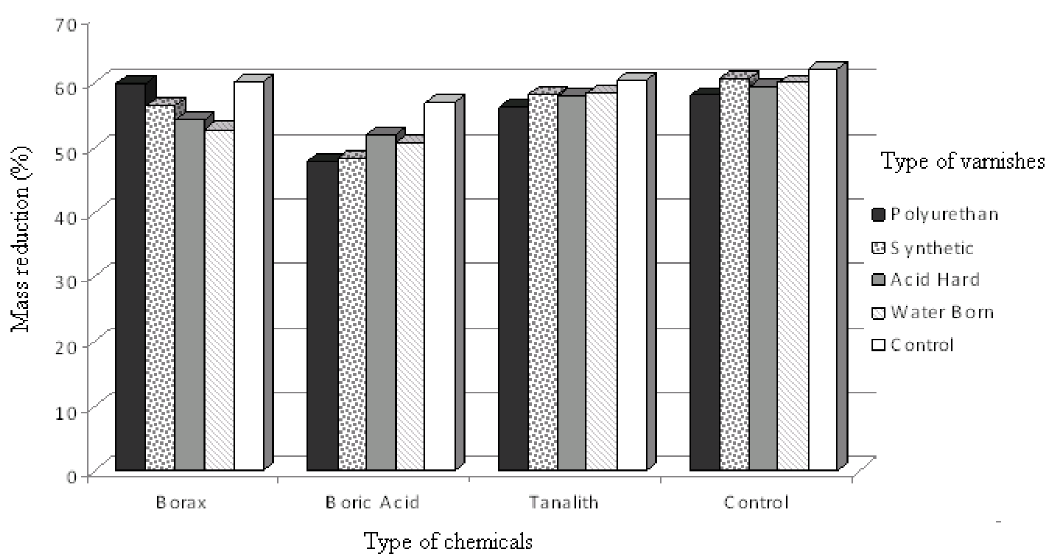

Fig. 1. The Mass reduction % chemical and varnish

Table 7. Duncan Test Results (p≤ 0.05)*

Source of Variance	Temperature (C°)	CO (ppm)		NO (pmm)		O_2 (%)		Mass Reduct (%)
	Mean	Mean	HG	Mean	HG	Mean	HG	Mean
Borax+Polyurethan	291.74	840.59	abc	39.21	abc	18.09	e	58.90
Borax+Synthetic	293.81	1347.66	abcd	47.38	abcd	17.38	cde	56.51
Borax+Acid Hard.	288.33	2423.62	def	58.10	cde	17.34	cde	54.32
Borax+Water B.	327.92	3487.36	f	72.87	ef	15.34	ab	52.85
Borax+Control	290.67	2427.69	def	31.47	a	17.54	cde	59.49
B. Acid+Polyurethan	323.85	284.52	a	38.25	abc	17.88	de	47.98
Boric Acid+Synthetic	319.38	2405.64	def	48.85	abcd	16.53	abcde	48.41
B.Acid+Acid Hard.	290.11	1432.45	abcd	44.25	abc	17.56	cde	51.99
B.Acid+Water B.	333.76	3612.18	f	65.58	def	15.67	abc	50.84
Boric Acid+Control	300.14	1512.83	abcde	30.52	a	17.30	cde	57.02
Tanalith+Polyurethan	341.78	573.86	ab	57.60	cde	17.66	de	56.34
Tanalith+Synthetic	309.87	1756.03	bcde	59.67	cde	17.21	cde	58.17
Tanalith+ Acid Hard.	316.34	2788.88	ef	81.08	fg	16.08	abcd	57.89
Tanalith+Water B.	317.91	4794.89	g	92.71	g	15.01	a	60.05
Tanalith+Control	281.61	2462.72	def	51.93	bcd	17.07	bcde	60.33
Control+Polyurethan	291.65	1918.05	cde	33.41	ab	17.69	de	58.37
Control+Synthetic	293.81	1347.66	abcd	47.38	abc	17.38	cde	60.71
Control+ Acid Hard.	290.51	2445.73	def	39.71	abc	17.44	cde	59.53
Control+Water B.	303.71	2800.31	ef	44.63	abcd	16.77	abcd	58.53
Control	292.53	2485.89	def	30.06	ab	17.44	cde	62.07

* The mean values marked with the same symbol are statistically identical

As a result of combustion, the highest amount of O_2 concentration was measured from the polyurethane varnish group and impregnated with borax. The lowest change of O_2-concentration in combustion was measured in water-based varnish and impregnated with Tanalith-E. It was shown that the change of O_2 values related to the types of varnishes and chemicals used for impregnation (Fig. 2).

The proportion of oxygen in air is normally 21%. So, it was shown that the treated samples had burned poorly because of the decrease in the oxygen ratio. Impregnation chemicals were shown to be effective as fire retardants.

The highest increase in CO concentration was observed in the experiment of water-based varnish and impregnated with Tanalith-E samples. The lowest value of CO concentration was observed in polyurethane varnish samples that had been impregnated with boric acid. The results connected with these values are shown in Fig. 3.

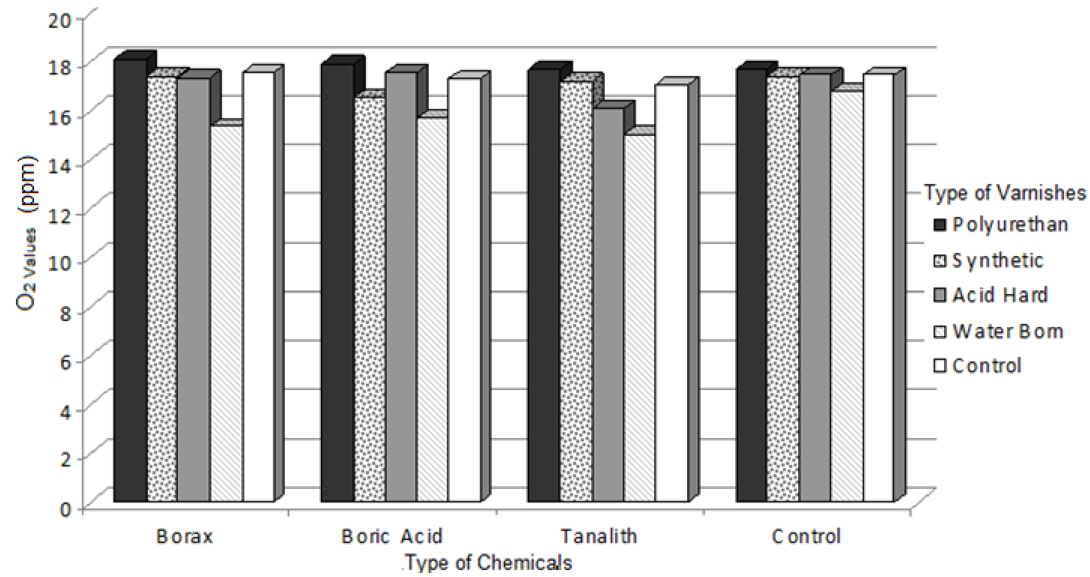

Fig. 2. The changes of O_2 values (ppm)

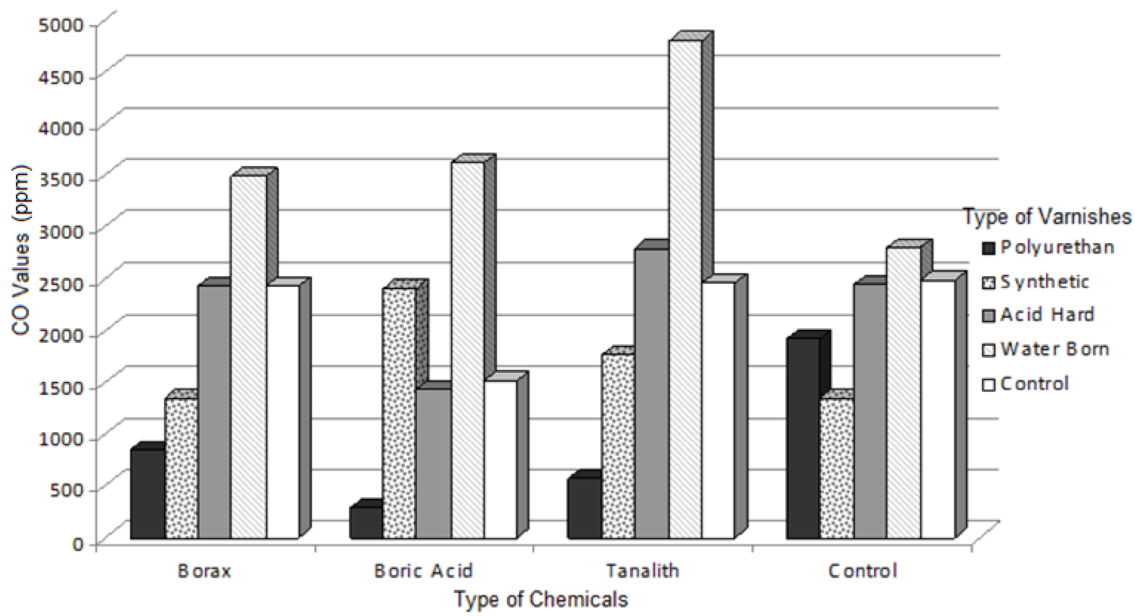

Fig. 3. Average values of CO (ppm) at the end of combustion, depending on the types of impregnation chemical and surface-applied varnish

The highest increase in NO concentration was observed in the experiment of water-based varnish and impregnation processed Tanalith-E samples. The lowest was in those of massive control samples and impregnated with borax. The connected results with these values are shown in Fig. 4. The highest temperature variation was observed in the experiments of the polyurethane varnish group in which the samples had been impregnated with Tanalith-E. The lowest values were observed in those of polyurethane varnish samples that had been impregnated with boric acid. The results associated with these values are shown in Fig. 5.

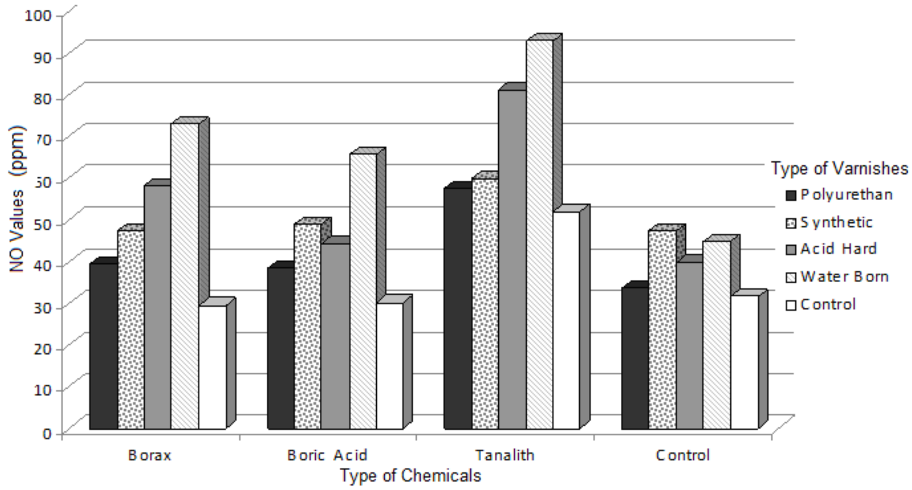

Fig. 4. Average values NO of (ppm) at the end of combustion, depending on the types of impregnation chemical and surface-applied varnish

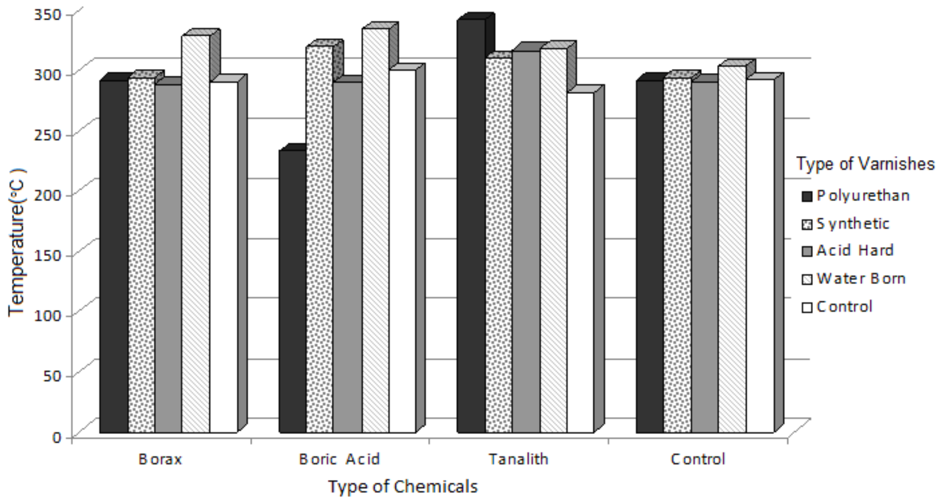

Fig. 5. Average values °C of (ppm) at the end of combustion, depending on the types of impregnation chemical and surface-applied varnish

If flames are present, fire temperatures are high and more oxygen is available from thermally induced convection. The lower temperatures of the smoldering stage result in a lower oxygen supply from diffusion into the fuel bed-gasses, which limits oxidation of the fuel bed during that stage (Lobert and Scharffe 1991).

At the first stage of the combustion, an increase occurred in temperature due to the flame source, and a decrease occurred as a result of the flame source's getting far away from fire tube.

The highest concentrations of O_2 were observed in the polyurethane varnish group impregnated with borax samples, and the lowest were in the case of water-based varnish impregnated with Tanalith-E. The results associated with these values are shown in Fig. 4. According to the control samples it can be said that impregnation chemicals shown the effect of fire retardant.

In the first 4 minutes, the first stage of the experiment, combustion occurred in all the samples nearly at the same time. The highest mass reduction (62.07%) was observed in unprocessed wood (control) samples, the second stage of combustion after the movement of flame source from fire tube. According to Table 6, among the control samples, it was observed that while impregnation chemicals reduced combustion values, borax chemical reduced combustion by nearly 4.15% on average. Boric acid chemical reduced combustion by nearly 8.13% on average, whereas Tanalith-E chemical reduced combustion by nearly 3.28% on average.

Uysal and Kurt (2009) studied impregnation of the oak (*Quercus robur* Lipsky) with di-ammonium phosphate, potassium carbonate, calcium chloride, and zinc chloride chemicals. Also, the test samples were applied to the combustion test. The highest mass reduction was obtained from Laminated Veneer Lumber (LVL), which was bonded with PVAc adhesive and impregnated with aluminums sulphate. The lowest value was from LVL that was bonded with PF adhesive and impregnated with zinc chloride.

CONCLUSIONS

1. At the end of the combustion test, the average O_2 consumption ratio (15.2% O_2) was seen in samples prepared with water-based varnish and impregnated with Tanalith-E. The highest O_2 consumption ratio (18.1% O2) was observed in polyurethane varnish and impregnated with borax.
2. The highest CO ratio (4794 ppm) was observed in samples which were finished with water-based varnish and impregnated with Tanalith-E. The lowest ratio (284 ppm) was observed in samples which had polyurethane varnish and were impregnated with boric acid. As is well known, there are two forms of reaction between C_2 and O_2 during combustion.
3. The combustion ratio of a sample is directly connected to the sum of the amount of CO and CO_2 emissions. The combustion tests are made in an open environment; there is no lack of O_2. Thus, because the amounts of CO and CO_2 emissions of the tests were compared with control samples vs. those impregnated with, borax, boric acid, and Tanalith-E, followed by finishing with polyurethane, synthetic, acid hardening, or water-based varnish, it can be concluded that impregnation chemicals have a fire retardant effect.
4. Due to the presence of a flame source, during the first stage of the combustion test, a linear increase in temperature was observed. The temperature then decreased when the fire source was removed from the fire tube on samples, which both had been impregnated with borax, boric acid, Tanalith-E chemicals and unprocessed wood (control) samples.
5. This situation shows an effect of impregnation, which tended to decrease or end burning phenomena after the fire source was moved away from the fire tube.
6. The highest increase (92.72 ppm) in NO concentration was observed in the samples impregnated with Tanalith-E and then finished with water-based varnish. The lowest one (29.48 ppm) was in unvarnished samples impregnated with borax.

7. As a result, the group of samples finished with water-based varnish and impregnated with boric acid was found to be the most successful in terms of the mass reduction amount. According to the results, all of the varnishes used in the study showed a low resistance against fire and high temperatures.

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