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Strengthening the retention amount and leaching resistance of boron compounds used as impregnation material

Emprenye maddesi olarak kullanılan borlu bileşiklerin tutunma miktarı ve yıkanma direncinin güçlendirilmesi

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Strengthening The Retention Amount And Leaching Resistance Of Boron Compounds Used As Impregnation Material

Highlights

- *Retention amount of impregnation material higher than samples impregnated with boron compounds.*
- *Colophony addition increases leaching resistance of impregnation materials based boron compounds.*
- *Colophony is preferable additive in order to strengthen basic features of wood material.*
- *Boron Compounds doped colophony may be used in interior and outdoor architecture*

Graphical Abstract

This study is carried out in order to increase the retention performance of synthetic boron compounds. For this purpose, the test samples prepared from Scots pine (Pinus sylvestris Lipsky), European Oak (Quercus petraea Liebl.) and Oriental Beech (Fagus orientalis Lipsky) woods and impregnated with dipping method in accordance with the principles of TS 6193 EN 84 by using 12 different compositions of developed impregnation material.

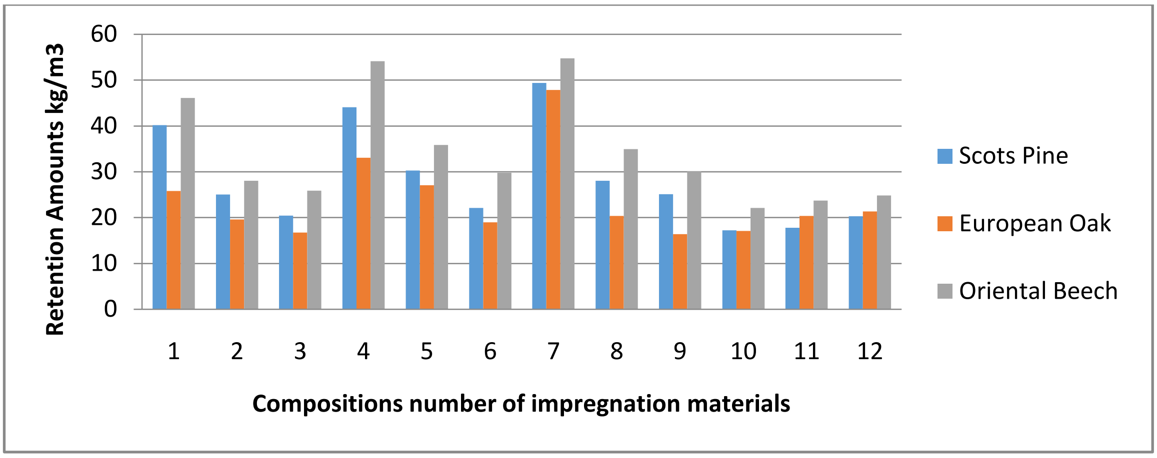

Figure. *Retention amounts of impregnation materials with boron compounds doped colophony*

Aim

The aim of this study to increase retention amount and leaching resistance of Boron compounds based impregnation material.

Design &Methodology

In this study experiemental way was used with different combinations.

Originality

Impact of colophony with different combinations on leaching and retention performance of boron compounds that are used as impregantion material in wood industry, is investigated in this study. From this point of view this study is unique.

Findings

Using colophony doped with boron compounds as impregnation material may increase retention amounts and leaching resistance.

Conclusion

Colophony can be preferred as leaching reducing agent for increasing the retention performances of boron compounds due to its water repellent and leaching resistance characteristics as a result of experiments.

Declaration of Ethical Standards

The authors of this article declare that the materials and methods used in this study do not require ethical committee permission and/or legal-special permission.

Strengthening the Retention Amount and Leaching Resistance of Boron Compounds Used as Impregnation Material

Araştırma Makalesi / Research Article

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ABSTRACT

This study is carried out in order to increase the retention performance of synthetic boron compounds. For this purpose, the test samples prepared from Scots pine (*Pinus sylvestris* Lipsky), European Oak (*Quercus petraea* Liebl.) and Oriental Beech (*Fagus orientalis* Lipsky) woods and impregnated with dipping method in accordance with the principles of TS 6193 EN 84 by using 12 different compositions of developed impregnation material. The impregnated test samples are tested according to the same standards' principles. It has been found that the retention amount and leaching resistance of the impregnation materials approximately 2 times higher than the ones which are impregnated with boron compounds depending on kind of wood. This impregnation material is more environmentally friendly than other toxic impregnation materials which are harmful for human health such as CCA beside its preservation feature against biotic and abiotic factors through boron compounds content. Moreoever the wood materials impregnated with developed impregnation material can be recycled comparatively more easily and in a healthy way at the end of its economical lifetime.

Keywords: Leaching, impregnation, rosin, boron compounds.

Emprenye Maddesi Olarak Kullanılan Borlu Bileşiklerin Tutunma Miktarı ve Yıkanma Direncinin Güçlendirilmesi

ÖZ

Bu çalışma emprenye maddesi olarak kullanılan sentetik borlu bileşiklerin tutunma dirençlerinin artırılması amacıyla yapılmıştır. Bu amaçla hazırlanan Sarıçam (*Pinus sylvestris* Lipsky), Sapsız Meşe (*Quercus petraea* Liebl.) ve Doğu Kayını (*Fagus orientalis* Lipsky) deney örnekleri, 12 farklı bileşimde geliştirilen emprenye maddesi kullanılarak TS 6193 EN 84 prensiplerine uyumlu bir şekilde batırma metodu ile emprenye edilmiştir. Emprenye edilen deney örnekleri aynı standarda göre test edilmiştir. Sonuç olarak geliştirilen emprenye maddesinin, ağaç türüne bağlı olarak, sadece borlu bileşiklerle yapılan emprenye işlemine göre yaklaşık 2 kat daha fazla yıkanma direnci ve tutunma miktarı sağladığı tespit edilmiştir. Geliştirilen emprenye maddesi, borlu bileşik içeriği ile biyotik ve abiyotik faktörlere karşı ağaç malzeme üzerinde koruma sağlarken diğer yandan da CCA gibi insan sağlığı için tehlikeli olan diğer toksik emprenye maddelerinden çok daha çevre dostudur. Ayrıca bu emprenye maddesi ile emprenye edilen ağaç malzeme, ekonomik ömrünü tamamlamasının ardından sağlıklı metodlarla ve çok daha kolayca geri dönüştürülebilmektedir.

Anahtar Kelimeler: Yıkanma, emprenye, kolofan, borlu bileşikler.

1. INTRODUCTION

Impregnation is a major factor in extending the usage lifetime of wooden materials by increasing the resistance against external factors such as biotic and abiotic (physical) influences. Even though developing more resistant impregnation materials through developing technology day by day, they have harmful impacts to human health and environment due to their chemical contents. Permanent damages occur on environment and human that are exposed to heavy influence of impregnating substances both during use and application of these chemicals beside during the disposal of lifetime completed wood materials. At this point, development of less harmful impregnated materials which are comparatively environmental friendly is essential for the protection of the environment and public health. In studies conducted in recent years, boron compounds come to the forefront in the impregnation of wood materials due to not only the toxic effect demonstrated against insects but also to the relatively low impact on human health. Many studies have been carried out in order to determine impacts of exposure of Boron on animals. Studies to detect cancerogenic impact of boron on animals showed that boric acid and boraks are not genotoxic on animals [1]. In addition to the older studies, various more recent studies are available which generally meet current standards. Most studies of borates have been done with boric acid and borax. Borates were found to be neither genotoxic nor carcinogenic. However acute and repeated exposure studies consistently showed the testis as a target organ. This was confirmed in fertility studies.

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Developmental effects were found which are mainly associated with foetotoxic activity [2].

Impregnation of wood material means treating the wood with preservative chemicals that have various protective features and for this purpose, a wide variety of materials have been used and various systems have been developed for decades [3]. The most commonly known imregnating substances are creosote, CCA (copper, chromium, arsenic) and PCP (pentachlorophenol). With the pressure of environment protection associations, useage of creosote and CCA have been restricted recently and PCP was restricted much earlier in many countries. There are serious research results regarding that these substances disrupt the ecological balance and threaten the health of humans and other living creatures. When the impacts of boron compounds like boric acid and borax on the blood of a healthy human are analysed in the laboratory at various ratios, they become beneficial by promoting the antioxidant enzyme activities at low doses. However, although boron compounds create oxidative stress in increasing doses, they have no genotoxic effects even at the highest concentrations [4]. When compared to other traditional imregnating substances, boron compounds are less harmful environmentally, and are toxic at very low levels. For example it is not more toxic than normal table salt for humans and animals and it is colorless and odorless, it has no corrosive properties [5]. Embedding the wood material impregnated with CCA that reaches the end of its lifetime as waste, has become more difficult due to the new regulations. However, the amount of impregnated wood material that reaches the end of its lifetime has significantly increased. For example, the amount of impregnated wood material that reached the end of its lifetime in United States was 1 million m^3 in 1990 and 15 million m^3 in 2010 and this number is estimated to be 18 million m^3 in 2020 [6].

Inorganic boron compounds with aqueous solutions are very effective as fire retardant. A mixture of boric acid and borax show the highest fire resistance [7]. Some physical, biological, mechanical, and fire properties of wood polymer composite (WPC) pretreated with boric acid and borax mixture were investigated. Results showed that except for fire properties; physical, biological, and mechanical properties of WPC were improved compared to untreated control sample [8].

While leaching values are at low level in Eucalyptus wood which is impregnated with commercial and boron substances compared to the fully dry wood, it is obtained significantly higher leaching rates in Eucalyptus wood which is treated with water-repellent substances statistically (WRS)[9]. The leaching amount of the treated wood with the mixture of PEG-400 and borate is remarkably high when compared to the one that is

obtained from the wood treated with aqueous solutions of borate [10].

According to the results of studies conducted to investigate the use of colophony as impregnating agent, dry heat creates some positive changes in the macroscopic properties of wood material with colophony according to the checkedsamples. Given that, it does not stick to the surface of the wood material, annual rings and medullary rays become clear and brightness increases and harmony of color and pattern that do not deteriorate [11]. In scots pine and chestnut woods treated with Boric acid, silica gel process is applied as a second process after treating Boric acid affects the retention amount positively and increases leaching resistance compared to the samples of Scots pine and chestnut wood impregnated with only Boric acid. The amount of retention in chestnut samples its higher in comparison to the Scots pine one. The reason for this is that the number of cell wall of chestnut wood is higher than of Scots pine and that impregnated substance has retention surface at higher amounts [12].

Wood materials which are impregnated with boron compounds need to have different retention rates of boron compounds for rot fungi, insects and fire resistance. While 2.5 kg/m^3 boric acid retention is effective against *Tyromyces palustris* and *Coriolus versicolor* rot fungi, it is needed 42.2 kg/m^3 boric acid retentionagainst *Coptotermes formasanus* termite**[**13]. When wood material is impregnated by adding boron compounds into the poliol substance to prevent boron leachingfrom wood material, boron leachingfrom wood material is significantly reduced as a result of this boron/poliol complex, however, complex compound reduces the resistance against decay of the wood material when compared to the experiment samples impregnated with boron compounds [14].

Alkyl ammonium compounds and conventional boron compounds which are new imregnating substances gain importance increasingly. Boron compounds gain currency due to high biological activities against pests, easy application by dissolving in water, wood diffusion capabilities, being cheap and available, low toxicity that can be neglected against mammals, and increasing the resistance of wood against fire significantly. In addition to this, their use has remained limited to only interiors because of the fact that they are leached from wood easily with the impact of rain [5].

The physical and chemical properties of borates render them suitable for formulation as stand alone preservatives and allows them to make components of more complex formulations in combination with copper, chromium, quaternary ammonium, or organic ligands. Boron-based systems therefore offer a totally flexible option [15].

Improvement of the leaching resistance by increasing the penetration strength of boron compoundsinto the wood material which are used as a kind of comparatively healthy impregnation material in woodworking industry. However it cannot be preferred in many application areas due to the fact that it can be easily diverged from wood material by leaching. Increasing leaching resistancewill expand their useage area and pave the way for a healthy and safe impregnated material. Improving leaching resistance of boron compounds which are comparatively healthy and envorimentally friendly impregnation material by increasing their penetration strength to the wood material using natural resins is aimed through this study.

2.MATERIAL and METHOD

2.1.Wood Materials

Scotspine (*Pinus sylvestris* Lipsky), Europeanoak (*Quercus petraea* Liebl.) and Oriental Beech (*Fagus orientalis* Lipsky) which are commonly used as wood material in forest products industry in Turkey have been used as experiment materials. Wood materials used in the experiments were obtained from local enterprises by completely random method. In the selection of wood materials, it was paid attention to the fact that the timber was healthy, straight grained, knotless, without reaction wood, and had normal growth, no fungus and pests.

2.2. Impregnation Material

In this study, boric acid and borax from boron compounds have been used. Boric acid is obtained from colemanite ore. Borax that has 99.90% purity level, 1.815 g/cm^3 specific gravity and having crystal structure is produced from tincal ore. It is the water whose conductivity value is calculated as 0.055 µS/cm (25ºC) and electrical resistance is 18.2 MΩ-cm. The ionic purity of water may be expressed as electrical conductivity from microsiemens/cm (µS/cm) unit or vice versa it may be expressed as electrical resistance from megaohm-cm unit. Dissolved salts in water also increase the conductivity value by causing the formation of positive and negative charged ions [17]. Distilled water has been used to regulate level of viscosity and acidity of impregnation liquid.

Different kinds of combinations and content of the impregnation material which is produced to be tested in this study are shown in Table 1. Boric acid and Borax have been used in the test due to their easy obtainable, storable features and mostly preferred against pesticides. Pre-test has been carried out and results showed that 10 % of colophony is optimum soluable percentage in solvent. Additional colophony additive causes to residues in the liquid. Here, in order to test colophony's effect on the leaching resistance of wood material, the types formed by the compound concentrations of boron compounds of 7 %, 5 % and 3 % have been compared with only boron compounds of 3 % and their performances have been tested.

Table 1. Content of Developed Impregnation Material used as impregnation material

Imp Mat *Nr.*	Impregnation Materials	Content				
		Borax ($Na_2B_4O_7.10H_2O$) (%)	Boric Acid (H_3BO_3) (%)	Colophony ($C_{19}H_{29}COOH$) (%)	Distilled Water H_2O (%)	Ethanol (C_2H_6O) (%)
E1	BA+COL%7	-	7	10	13	70
E2	BA+COL%5	-	5	10	15	70
E3	BA+COL%3	-	3	10	17	70
E4	BRX+COL%7	7	-	10	13	70
E5	BRX+COL%5	5	-	10	15	70
E6	BRX+COL%3	3	-	10	17	70
E7	BA+BRX+COL%7	3.5	3.5	10	13	70
E8	BA+BRX+COL%5	2.5	2.5	10	15	70
E9	BA+BRX+COL%3	1.5	1.5	10	17	70
E10	BRX%3	3	-	-	97	-
E11	BA%3	-	3	-	97	-
E12	BA+BRX%3	1.5	1.5	-	97	-

BA: Boric Acid, BRX: Borax, COL: Colophony, %: Rate of Boron compunds in the impregnation material composition

Colophony is yellow-colored transparent resin which is obtained by the distillation of tall oil. It is obtained by fractional distillation of tall oil which is by-product of the sulfate method, and obtained also from the wounds on a living tree and from the extraction out of log of tree. It is insoluble in water, solid at room temperature and has a light yellow-brown in color, soluble in ether, alcohol, chlorinated hydrocarbon and hydrocarbons. Color is the most important factor in determining the value of colophony. The color is evaluated with letters from light yellow to dark brown. Naturally resin obtained from pine trees contains 80% colophony 20% turpentine oil. Colophony composition contains 90% resin acids and 10% neutral substances [16].

2.3. Preperation of Test Sampels

11 test samples sized50x25x15 mm and acclimatized at 8-10 % humidity levels were prepared for each test to be used in leaching test in accordance with the norms of TS 6193 EN 84 (TS 6193 EN 84 2014).

Test samples were impregnated by using dipping method. For this purpose, samples were kept in 5 liters special closed containers encoded with related impregnation material number for 24 hours. Prepared test samples were placed carefully into containers and filled with prepared impregnation materials. All test samples have been submerged into the impregnation liquid by using specific cover for each container that can be seen a section in Fig.1.

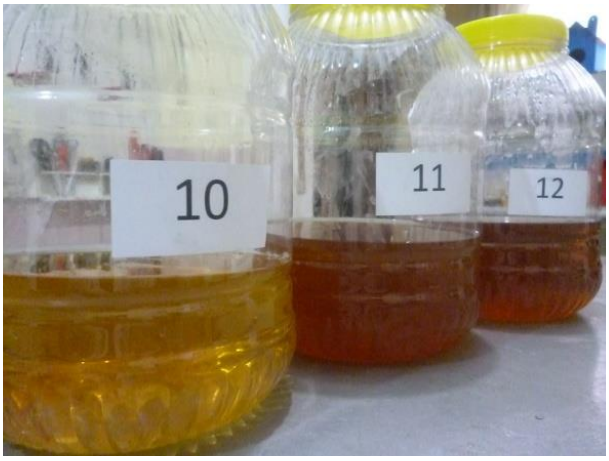

Figure 1. Impregnation process

Moreover the containers have been reversed periodically to keep all test samples in impregnation liquid. Containers whose covers were tightly closed were shakenperiodically to raise the effect of impregnation material. At the end of 24 hours, impregnation material was drained by opening the impregnation containers and impregnation samples were stacked according to the principles of TS 6193 EN 84to be dried at room temperature. Drying process continued at room temperature (20 ^{o}C ± 2^{o}C) until weight stabilization was provided.

2.4. Testing Methods

Leaching test materials prepared from Scots Pine, European Oak and Oriental Beech were subjected to a leaching process by treating with only water in 5-liter special containers. Each sample was exposed to the leaching effect of water with 24-hour dipping process (Fig. 2.)

At the end of the leaching process which was supported by shaking periodically, test samples were kept waiting in room conditions until their wheight stability is provided after impregnation and then their measurement values were recorded as first leaching result, thereafter, a second leaching process was performed by repeating leaching process under the same conditions. Reaction of the test samples and developed impregnation materials against each leaching process was measured. Content of penetrated impregnation material has been determined by elemental analysis of samples. Beside of this, elemental mapping has been done on the sample. Test samples after leaching process are shown in Fig.3.

Following equation (Eq. 1) was used for determination of the retention amount of the developed impregnation liquid:

$$R = \frac{GxC}{V} \text{g/cm}^3$$

Equation 1: Determination of retention amount of developed impregnation material compositions

G= T2 (Complete dry weight after impregnation) – T1 (Complete dry weight before impregnation) (g)

C = Solution concentration (%) Boron compound concentration 3%

V = Sample volume (cm^3) 50x25x15 = 18.75 cm^3

R = Retention Amount (%)

Figure 2. Leaching test

Figure 3. Test samples after leaching

2.5. Scanning Electron Microscope (SEM) Analysis

Elemental analysis was performed for test samples prepared from different tree species in order to determine the boron compound contents that they have after they were resisted to leaching process for the second time. Within this context, samples which will be used in SEM applications has been covered with gold to get an image easily using SEM. In this context, scanning electron microscope which was found in Gazi University Faculty of Technology Department of Material Engineering was used.

2.6. Data analyses

SPSS 22.0 program was used to analyze the data obtained from the test materials. Within the scope of leaching test, multivariate analysis of variance (Manova) was performed for the first and second leaching rates and for retention amounts and rates according to impregnation material. On the other hand, Duncan’s mean separation tests were performed to test the homogeneous subsets in case that there occur differences between the groups as a result of the analysis.

3. RESULTS AND DISCUSSION

3.1. Retention Performance

The retention amount is the most important factor that directly affects the leaching performance of impregnation material. In addition to the quality, the quantitiy of impregnation liquid retained in cell cavities and cell walls of the wood material is also important to save them against external conditions.When impregnated test samples are analyzed, it is seen sludge formation on the surface of impregnated test samples and of impregnation materials that have 7% boron compound concentration (Fig. 4a) It is seen that this sludge is less in the sample with 5% concentration (Fig. 4b)

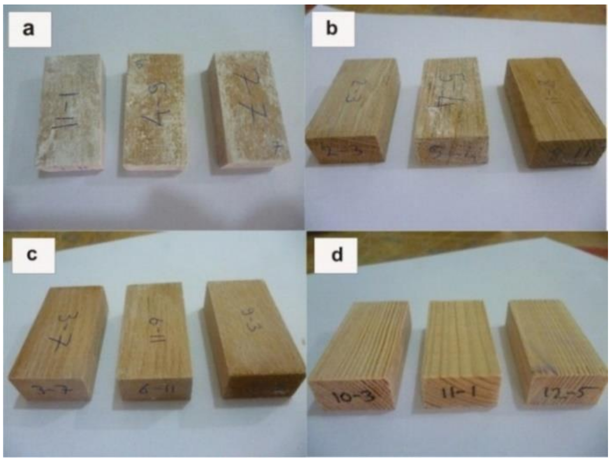

Figure 4. Impregnated test samples: (a) 7% DIL concentration, (b) 5% DIL concentration, (c) 3% DIL concentration, (d) 3% boron compound concentration

It is determined that there is not any sludge on the surface of the material in 3% concentrations (Fig. 4c) On the other hand, any residue formation is not observed on material surface of the test samples impregnated with only boron compounds with 3% concentrations (Fig. 4d).

Sludge formation on the impregnated samples is an undesirable situation in terms of texture and surface treatment of wood. Relations between dependent variables and factors were studied in Manova test that was applied to the samples in order to see impact of factors that are "kind of wood", impregnation material" and kind of wood*impregnation material on dependent variables. Manova test results were seen in Table 2.

Retention amounts and homogeneous subsets of leaching test samples of Scots pine, European oak and Oriental beech impregnated with impregnation material that has 12 different concentrations which were produced in laboratory are shown respectively in Table 3, 4 and 5.

It was seen that impregnation materials create a statistically significant difference on retention amount of Scots pine samples Table 3.

When homogeneous subsets of the test samples on which sludge was not formed was evaluated at the end of Duncan test, after second leaching process it has been seen that the samples numbered 6 (BRX+COL 3 %) and 9 (BA+BRX+COL 3 %) impregnated with impregnation material take part in the same homogenity group and the samples numbered 3 impregnated with impregnation material (BA+COL 3 %) take part in different homogenity group and has less retention amount. It is seen that homogeneous subsets of the test samples impregnated with the impregnation materials numbered 10 (BRX 3 %), 11 (BA 3 %) and 12 (BA+BRX 3 %) remain the same group after two leaching processes.

Retention amount of impregnated Scots pine samples with Boric acid, zinc chloride and aluminum chloride was found 5 kg/m^3 [11]. It was seen that the experimental samples 6 and 9 retention performance was about 3 fold higher than the previous research. This indicates that colophony is an effective solution for increasing the leaching resistance of boron compounds as a result of its cohesive and water repellent structure.

Table 2. MANOVA test results

Multivariate Tests[a]							
Effect		**Value**	**F**	**Hypothesis df**	**Error df**	**Sig.**
Intercept	Pillai's Trace	,999	39866,323[b]	6,000	355,000	,000
	Wilks' Lambda	,001	39866,323[b]	6,000	355,000	,000
	Hotelling's Trace	673,797	39866,323[b]	6,000	355,000	,000
	Roy's Largest Root	673,797	39866,323[b]	6,000	355,000	,000
Kind of wood	Pillai's Trace	1,074	68,766	12,000	712,000	,000
	Wilks' Lambda	,197	74,253[b]	12,000	710,000	,000
	Hotelling's Trace	2,711	79,960	12,000	708,000	,000
	Roy's Largest Root	2,035	120,754[c]	6,000	356,000	,000
Impregnation material	Pillai's Trace	2,720	27,138	66,000	2160,000	,000
	Wilks' Lambda	,005	49,536	66,000	1905,007	,000
	Hotelling's Trace	14,350	76,825	66,000	2120,000	,000
	Roy's Largest Root	7,328	239,836[c]	11,000	360,000	,000
Kind of wood * Impregnation Material	Pillai's Trace	1,093	3,644	132,000	2160,000	,000
	Wilks' Lambda	,279	3,850	132,000	2072,361	,000
	Hotelling's Trace	1,514	4,054	132,000	2120,000	,000
	Roy's Largest Root	,659	10,785[c]	22,000	360,000	,000

a. Design: Intercept + Kind of wood + Impregnation Material + Kind of wood * Impregnation Material
b. Exact statistic
c. The statistic is an upper bound on F that yields a lower bound on the significance level.

Table 3. Duncan test results for leaching test retention amounts of Scots pine samples impregnated with developed impregnation material

		Impregnation			1st Leaching Process			2nd Leaching Process		
Imp Mat *Nr.*	Impregnation Liquid	Retention Amount (kg/m3)	HS[a]	SD[d]	Retention Amount (kg/m3)	HS[b]	SD[e]	Retention Amount (kg/m3)	HS[c]	SD[f]
E10	BRX%3	17.24	A	2.830	6.38	A	0.0136	5.38	A	0.014
E11	BA%3	17.76	A	2.245	5.92	A	0.0128	3.98	A	0.011
E12	BA+BRX%3	20.29	AB	3.796	6.87	A	0.0305	4.75	A	0.019
E3	BA+COL%3	20.48	AB	3.048	14.38	B	0.0498	9.51	B	0.046
E6	BRX+COL%3	22.10	AB	3.235	16.55	BC	0.0482	14.94	CD	0.047
E2	BA+COL%5	25.02	BC	4.711	17.18	BC	0.0846	12.36	BC	0.082
E9	BA+BRX+COL%3	25.09	BC	4.331	17.61	BC	0.0694	16.22	DE	0.068
E8	BA+BRX+COL%5	28.02	CD	3.807	19.12	CD	0.0478	18.00	DE	0.045
E5	BRX+COL%5	30.28	D	5.516	21.50	D	0.0718	19.08	E	0.066
E1	BA+COL%7	40.22	E	6.824	26.64	E	0.1090	23.07	F	0.102
E4	BRX+COL%7	44.11	E	4.576	32.36	E	0.0703	25.88	F	0.063
E7	BA+BRX+COL%7	49.43	F	11.452	36.32	F	0.1689	31.74	G	0.140

[abc] Homogeneous subsets for each level ($p<0.05$), [def] Standard deviations for each level
BA: Boric Acid, BRX: Borax, COL: Colophony, %: Rate of Boron compounds in composition of impregnation material

When retention amount of the test samples on which sludge was not formed at the end of second leaching test was evaluated and it was seen that number 6 (32 %) and number 9 (35 %) test samples impregnated with developed impregnation material showed the best retention performance (Fig.5)

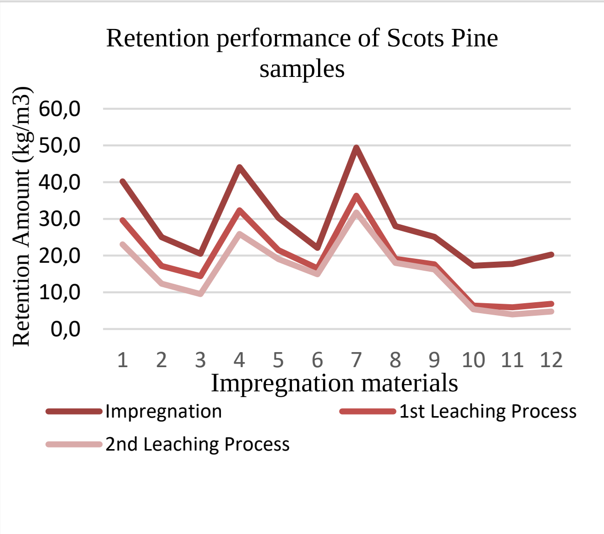

Figure 5. Leaching performance of Scots Pine wood

When retention performances of impregnation materials numbered 6 (BRX+COL 3 %) and 9 (BA+BRX+COL 3 %) which were developed were compared with the retention amounts of the samples numbered 10 (BRX 3 %), 11 (BA 3 %) and 12 (BA+BRX 3 %) that have Borax and Boric acid in their compounds, it was seen that the retention performance of the samples numbered 6 and 9 was 2.22 fold higher than the samples numbered 10, 11 and 12 on average.

It is seen that impregnation materials creates a statistically significant difference on retention amount of European oak samples in Table 4.

Homogeneous subsets of the European Oak samples whose impregnation material concentration is 3% show similarity with Scots pine at the end of Duncan test. When retention amount of the European oak samples on which sludge was not formed at the end of second leaching, test was evaluated, it was seen that number 6 and number 9 samples impregnated with developed impregnation material showed the best retention performance, respectively(43 %) and (44 %) (Fig. 6)

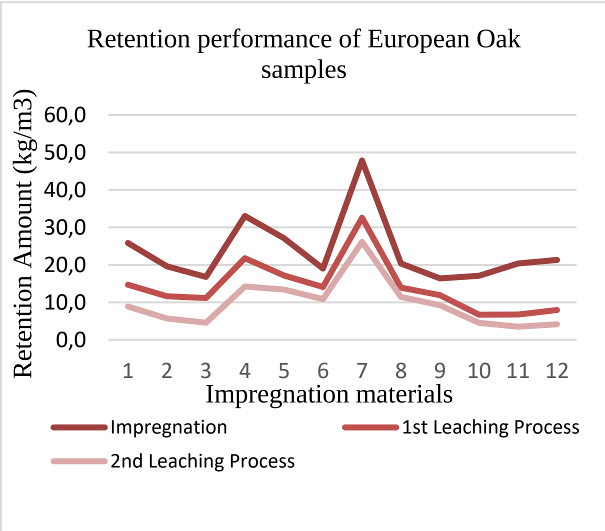

Figure 6. Leaching performance of European Oak wood

Table 4. Duncan test results for leaching test retention amounts of European oak samples impregnated with developed impregnation material

		Impregnation			1st Leaching Process			2nd Leaching Process		
Imp Mat *Nr.*	Impregnation Liquid	Retention Amount (kg/m^3)	HS[a]	SD[d]	Retention Amount (kg/m^3)	HS[b]	SD[e]	Retention Amount (kg/m^3)	HS[c]	SD[f]
E10	BRX%3	17.08	A	2.492	6.70	A	0.024	4.52	A	0.020
E11	BA%3	20.37	A	3.937	6.76	A	0.033	3.49	A	0.032
E12	BA+BRX%3	21.32	A	5.356	7.95	AB	0.016	4.15	A	0.022
E6	BRX+COL%3	18.99	A	4.357	14.10	CD	0.079	10.86	CD	0.090
E2	BA+COL%5	19.59	A	2.701	11.63	BC	0.025	5.65	AB	0.027
E8	BA+BRX+COL%5	20.35	A	3.004	13.92	CD	0.032	11.43	CD	0.030
E9	BA+BRX+COL%3	16.38	A	1.512	11.91	C	0.029	9.24	C	0.027
E3	BA+COL%3	16.77	A	2.404	11.12	BC	0.032	4.54	A	0.030
E1	BA+COL%7	25.83	B	5.904	14.68	CD	0.062	8.89	BC	0.054
E5	BRX+COL%5	27.10	B	3.093	17.17	D	0.021	13.39	D	0.024
E4	BRX+COL%7	33.04	C	4.855	21.76	E	0.074	14.21	D	0.061
E7	BA+BRX+COL%7	47.87	D	12.387	32.57	F	0.232	26.12	E	0.216

[abc] Homogeneous subsets for each level ($p<0.05$), [def] Standard deviations for each level

BA: Boric Acid, BRX: Borax, COL: Colophony, %: Rate of Boron compounds in composition of impregnation material

Number 6 and 9 impregnation material showed the best performance in European oak test samples. It has been inferred that retention performance of the samples numbered 6 and 9 is 1.8 fold higher than the retention performance of the samples numbered 10, 11 and 12 on average. It is seen that impregnation materials creates a statistically significant difference on retention amount of Oriental beech samples in Table 5.

Table 5. Duncan test results for leaching test retention amounts of Oriental beech samples impregnated with developed Impregnation Material

		Impregnation			1st Leaching Process			2nd Leaching Process		
Imp Mat *Nr.*	Impregnation Liquid	Retention Amount (kg/m^3)	HS[a]	SD[d]	Retention Amount (kg/m^3)	HS[b]	SD[e]	Retention Amount (kg/m^3)	HS[c]	SD[f]
E10	BRX%3	22.14	A	3.109	9.33	A	0.021	7.77	A	0.022
E11	BA%3	23.75	A	3.078	9.51	A	0.017	6.71	A	0.019
E12	BA+BRX%3	24.82	A	3.838	10.71	A	0.024	8.02	A	0.014
E3	BA+COL%3	25.87	A	7.464	18.39	B	0.107	12.71	AB	0.110
E2	BA+COL%5	28.07	AB	4.875	19.41	BC	0.085	13.87	AB	0.092
E6	BRX+COL%3	29.77	AB	11.174	23.00	BC	0.187	20.99	CD	0.187
E9	BA+BRX+COL%3	30.08	AB	7.658	20.69	BC	0.118	18.66	BC	0.123
E8	BA+BRX+COL%5	34.92	B	8.633	25.06	BC	0.139	23.38	D	0.143
E5	BRX+COL%5	35.86	B	7.370	26.42	CD	0.136	23.33	D	0.137
E1	BA+COL%7	46.15	C	16.288	30.93	DE	0.261	23.00	D	0.276
E4	BRX+COL%7	54.11	D	9.915	38.18	EF	0.189	30.84	E	0.194
E7	BA+BRX+COL%7	54.78	D	14.257	37.47	F	0.234	33.50	E	0.239

[abc] Homogeneous subsets for each level ($p<0.05$), [def] Standard deviations for each level

BA: Boric Acid, BRX: Borax, COL: Colophony, %: Rate of Boron compounds in composition of impregnation material

Homogeneous subsets of the Oriental beech test samples whose impregnation material concentration is 3 % show similarity with Scots Pine and European Oak at the end of Duncan test. When retention amount of the Oriental beech samples on which sludge was not formed at the end of second leaching test was evaluated, it was seen that number 6 and number 9 test materials impregnated with developed impregnation material showed the best retention performance, respectively(29 %) and (38 %) (Fig. 7).

When retention performances of impregnation materials numbered 6 and 9 were compared with the retention amounts of the test samples numbered 10 (BRX3%), 11 (BA3%) and 12 (BA+BRX%) that have borax and boric acid in their compounds, it was seen that the retention performance of the samples numbered 6 and 9 was 2.02 fold higher than the samples numbered 10, 11 and 12 on average.

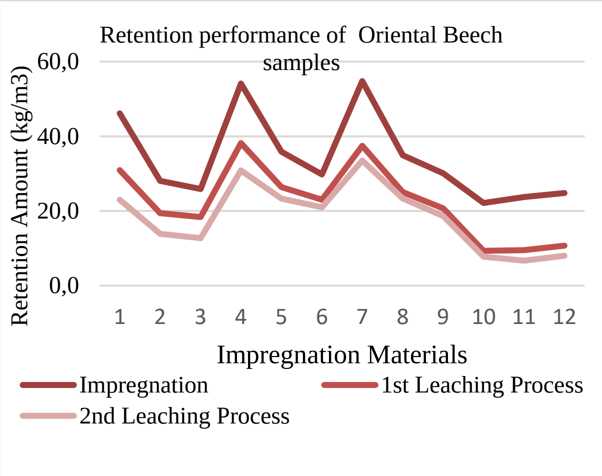

Figure 7. Leaching performance of Oriental Beech wood

When retention amounts were analysed, the highest retention amount was obtained in Oriental beech. The reason for this may be interpreted that Oriental Beech has higher density than Scots pine and European oak and accordingly has more cell wall and thus fixation of more impregnation liquid may be provided.These results were consistent with previous studies[19]. According to Kartal and Imamura, generally 2 kg/m^3 boron compound retention is needed for protection against fungi that color the solid wood material and 3.5 - 7 kg/m^3 boron compound retentionis needed for protection against fungi and pests degrading the wood [3].

When retention amounts are analysed, as a result of leaching process it has been detected that the test samples impregnated with developed impregnation material meet the protective effect limit values in protecting against fungi and pests degrading the wood.

3.2. Leaching Performance

When leaching performance of all test samples are analysed, the highest material loss amount has been determined in the impregnation materials numbered 10, 11 and 12 consisting of only boron compounds. The lowest impregnation material loss that is seen in retention amounts after the leaching process of impregnation materials takes place in the impregnation material numbered 6 that has 3% borax concentration. In addition, there is approximately 2 times more retention in all developed impregnation material combinations compared to the samples impregnated with only boron compounds. This situation shows that all combinations of developed impregnation material are more effective against leaching compared to boron compounds.

In the elemental analysis of samples carried out in Scots Pine samples, 24,695 % boron compound has been observed (Fig. 8c). Wood surface was zoomed in 500 times through SEM (Fig. 8a) to see boron compounds fixed on the wood surface as a proof of resistance against leaching. Furthermore dispersion of Boron compounds on wood surface is shown in Fig. 8b. Fig. 8b is complementary part of Fig. 8a that shooted from same location and zoom from wood material. In Scots pine test materials were exposed to leaching process two times, it is seen that retention of Borax from boron compounds is provided.

It is reported that the addition of montan wax decreased boron leaching from impregnated samples for 20% up to 50% [20]. In contrary to this research when SEM image is examined, it can be seen that the colophony that has resistance against leaching process in Scots pine wood contributes to the retention of borax from boron compounds to the wood material and prevents the leaching considerably.

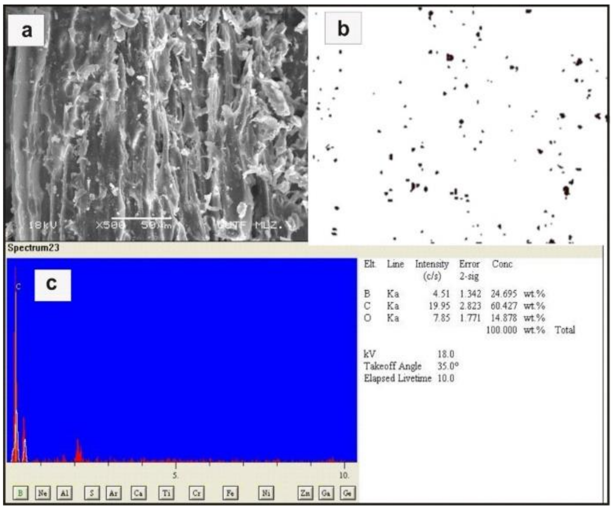

Figure 8. SEM image of Scots pine after impregnation: (a) SEM image; (b) elemental map; (c) Elemental analysis results

The samples impregnated with impregnation material numbered 6 could retain approximately 74 % of impregnation material that it absorbed within the sample as a result of the first leaching process and could retain approximately 57 % of it as a result of the second leaching process. Elemental analysis of the European oak sample is seen in Fig. 9.

According to the Fig. 9a and Fig. 9b show dispersion of boron compound on to the wood surface. Based on the Fig. 9, 13.495 % boron compound is found in the test sample analysed after the second leaching process (Fig. 9c). This situation shows that retention amount within wood material of the developed impregnation material and boron compounds are increased to a significant level despite leaching processes.

Dispersion of boron compounds on to the Oriental beech wood material can be seen in Fig. 10a and Fig. 10b

When elemental analysis is examined within the scope of SEM image obtained from Oriental beech samples (Fig.10c), boron compound in the ratio of 22.063 % has been determined.. In this case, it can be said that fixation of boron is provided substantially after second leaching process.

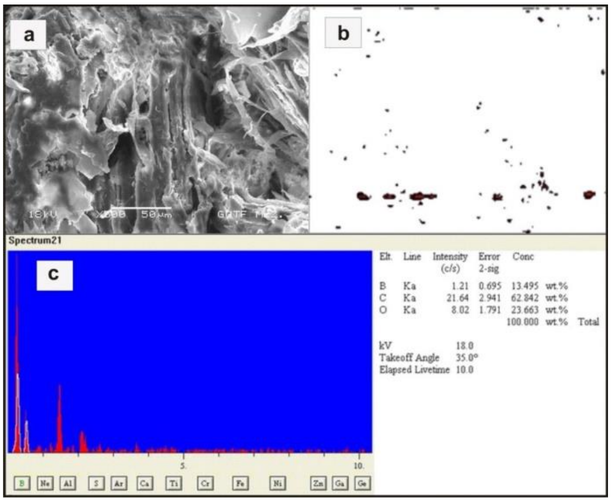

Figure 9. SEM image of European oak after impregnation: (a) SEM image; (b) elemental map; (c) Elemental analysis results

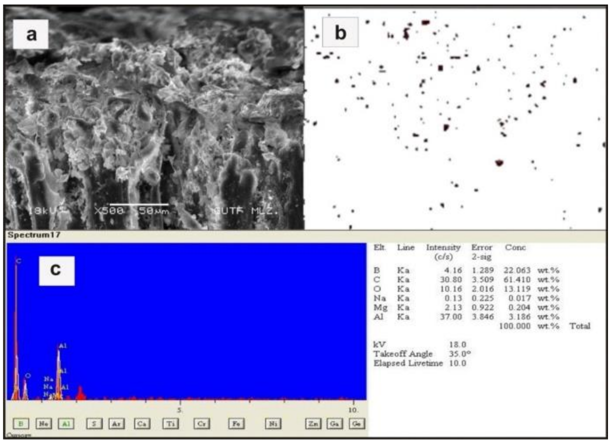

Figure 10. SEM image of oriental beech after impregnation: (a) SEM image; (b) Elemental map; (c) Elemental analysis results

Although it is a harmless impregnation material in terms of human health, the major obstacle in using boron compounds to impregnate wood material used in outdoor areas is that their leaching resistance is low. In this research, it is seen that colophony considerably increases the leaching resistance of boron compounds as fixation agent. Scots Pine, European Oak and Oriental Beech impregnated with developed impregnation material numbered 6 (BRX+COL 3 %) and 9 (BA+BRX+COL 3 %) that has 3 % of concentration of boron compounds can be used in outdoor areas. The wood materials impregnated with developed impregnation material can be use as an industrial design material both indoor and outdoor applications.

4. CONCLUSION

It has been found that the retention amount and leaching resistance of the wood material impregnated with compositions of developed impregnation material that contain colophony and boron compounds with the concentration of 3% are approximately 2 times higher than the ones which are impregnated with only boron compounds. In particular, leaching resistance of developed impregnation material compositions strongly depend on kind of wood. This indicates that colophony can be preferred as leaching reducing agent for increasing the retention performances of boron compounds due to its water repellent and leaching resistance characteristics as a result of experiments. It has been detected that the test materials impregnated with compositions of developed impregnation material meet the protective effect limit values in protecting against fungi and pests degrading the wood.

All components used in developed impregnation material are comparatively environmentally friendly and do not have a negative impact on human health. In addition to this, boron compounds protect the wood material against fungi and pests. Human health can be protected by using developed impregnation material instead of using toxic impregnation materials harmful to human health such as CCA and PCP to protect the wood material against external effects. When wood material is impregnated with developed impregnation material, it can be recycled more easily and in a healthy way at the end of its economic lifetime.

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DECLARATION OF ETHICAL STANDARDS

The author(s) of this article declare that the materials and methods used in this study do not require ethical committee permission and/or legal-special permission.

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