WOOD RESEARCH
52 (1): 2007
87-98

EFFECTS OF SOME IMPREGNATION CHEMICALS AND WATER REPELLENTS ON THE HYGROSCOPICITY OF BEECH WOOD

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ABSTRACT

Salt based chemicals tend to increase in hygroscopicity of wood. This phenomenon is known as one of the disadvantages of these salts in wood preservation in spite of their protective effectiveness against biological agents and fire. This study was designed to determine the rate of hygroscopicity of both some impregnation chemicals treated and untreated beech wood. Impregnation solutions of boric acid (BA) and borax (BX) were prepared by water and polyethylene glycol (PEG-400). Additionally, some water repellents (WR) such as paraffin wax (P), Styrene (St), methylmethacrylate (MMA) and icocyanate (ICO) were used to decrease the hygroscopicity level of beech wood.

Results indicated that paraffin wax and water repellents were considerably lowered the hygroscopicity level of beech wood. Four homogeneity groups were formed form the statistical analysis as follows (from the most hygroscopic treatments to lesser, respectively): 1. PEG-400 2. PEG-400+BA 3. WR +BA 4. P+BA+BX. Except for PEG-400, all the treatment chemicals decreased the hygroscopicity levels of beech wood compare to that of untreated control specimens.

KEY WORDS: impregnation, boron, wood preservation, hygroscopicity, beech wood

INTRODUCTION

In order to provide the possible longest usage time of wood as protecting it against biological and non-biological harmful factors, it needs to be impregnated by chemical substances changing with respect to usage area. The existence of poisonous effect of chemical protection substances for wood against harm may result in negative effects in other living creatures (Kurtoglu 1988, Willconson 1979, Richardson 1987).

As it is known that usage of wood material without protection is impossible, it is inevitable not to use these chemical substances for protection. In recent years, the researchers have been carried out about the environmentally-friendly impregnation substances. It is known that flourier, applied

by diffusion method, have high poisonous effect against mammals and they show volatile property in the form of bi-fluorides. Therefore, the researchers have been conducted in boron compounds (Murphy 1990, Greavers 1990).

In the time, the impregnation substances, soluble in water, are leaching and taken away from the wood completely. Due to this reason, they are not able to provide protection for a long time. So, the elaborating the impregnation compounds, remaining in the wood firmly, includes chemical and thermal reactions, aiming to make insoluble compounds in the wood (Assenault 1973, Murpphy et al. 1990). The concentration of impregnation substances and the application time of this process are very important in leachin the impregnation particles out of the wood. In addition, it is stated that there are numerous factors affecting leaching rates significantly, like the type of wood, extractive substance rate, pH value of washing liquid (Assenault 1973).

Baysal (1994) indicated that in order to prevent leaching the boron from wood, a secondary impregnation is applied by using water repellent substances, keeping the water away from the wood. It is observed that these substances make physical obstacles against the boron, held inside the wood. In addition to prevent leaching boron from the wood, They causes shrinkage and decrease in expansion of wood, which increases dimensional stability. After performing the fire retardant application in the timber parts, used in the exterior, leaching the impregnation salts out of the wood can be prevented in some degree by surface processes. But, due to hygroscopic properties of fire retardant (FR) substances in general, they have tendency to contain much more moisture and get wet with respect to the normal wood. This causes deterioration in the top surface process of the film layer quickly. In order to provide, non-hygroscopic structure in the wooden material, the polymer systems have been developed and so that the flow of the moisture in the wooden material has been decreased about 45-95 % (Norimatu et al. 1992, Yildiz 1992, Yalinkilic and Alma 1992).

Yalinkilic et al. (1995) reported that hygroscopicity of Scots pine wood were increased treated with PEG-400 significant level. Therefore, it is concluded that the increase in hygroscopicity can be eliminated out if PEG 400 reacts with WR.

Hafizoglu et al. (1994) found that hygroscopicity of douglas wood treated with mono-ammonium phosphate and pyresote were increased in maximum degree.

Nikolov and Encev (1967) proved that the hygroscopicity of the beech wood in the corresponding criteria decreases in case the degree of the temperature in east beech wood is increased as time dependent.

Stejnberg (1962) treated the Scots pine by various types of chemical substances, and while the wood samples are exposed to high temperature and various time periods (12 hours- 10 days) - (20°-110°-120°-130°), he searched for their properties, like hygroscopicity, swelling and shrinkage. It is observed that hygroscopicity varies with respect to time-temperature-type of wood and chemical substances.

Sadoh et al. (1960) impregnated *Betula nikoensis* and *Hama ecyparis* woods with formaldehyde. They searched for their dimensional stability and change in hygroscopicity. As a result of these researches, it is observed that formaldehyde affects both properties in positive manner.

Kenoga et al. (1959) applied gamma rays into "Ponderosa" Pine wood, it is seen that the hygroscopicity, shrinkage and swelling are affected in insignificant level.

Shibamato and Inove (1954) searched for water absorption and hygroscopic properties of bamboo wood, newly cut. After reaching 7% of moisture in wood, it is observed that there is a quick rate of change in evaporation and hygroscopicity.

Kadita and Nakato (1949) impregnated *Fagus Crenata* and *Chamaecyparis-obsusa* woods with benzen and alcohol. These substances significantly increase the hygroscopicity in both types of woods.

This study was designed to determine hygroscopicity levels of beech wood treated with some borates and water repellents.

MATERIAL AND METHODS

The wood specimens , used within the research, are prepared by East beech wood. For this purpose, substance, called as ANTIBLUE, is applied into the profile surfaces of logs, obtained from the Forest Department of the Regional Forest Office in accordance with the Turkish Standard 345 (TS 345 1974), in order to prevent to be coloured.

In order to perform the impregnation of experiment samples, the boron compounds are brought from Etibank Bandırma Borax and Acid Factory Department; "Vinilmonomerler" are gotten from Petkim- İzmit Rafinery and Polisan Chemical Industrial Incorporated Company. Other substances are obtained from PEG-400 Shell Petroleum Co and from other commercial companies. The wood specimens were prepared according to TS 345 (1974).

Three different impregnation substances were used:

1. Boron compounds
   a) Water solutions (In impregnation, having one action), "Boric acid (BA), Borax(BX), BA+BX
2. Water solutions + Water repellent (WR): (BA+BX)+St, (BA+BX)+MMA, (BA+BX)+ICO, (BA+BX)+(St+MMA)
2. WR: Styren (St), Methylmetacrylate (MMA), Isocyanate (ICO), (St+MMA)
3. Commercial impregnation substances: PEG 400, Tanalith -CBC, Vacsol (V), ammonium-sulphate, di-ammoniumphosphate.

The plan of the impregnation experiment is given below in Tab. 1:

Impregnation Method

The impregnation of wood samples is carried out in accordance with the conditions in ASTM 1413-76 (1976) in a metal mechanism, having vacuum holders of gap of the window. In this method, after applying a vacuum, equal to 760cm Hg^{-1} for 60 minutes, the samples are left to the diffusion in the solution in atmospheric pressure for 60 minutes. The weight of samples is measured before and after impregnation. In order to determine the retention ratios of the impregnation substance, kg/m^3 after impregnation and completely dried wood weight ratio, the samples are dried up to the moisture degree prior to the experiment. The help of formulations, given below calculates kg/m^3 and % retention. Impregnation experiment mechanism is shown in Fig. 1.

$$\text{Retention} = \frac{G \times C}{V} \times 10 \ (kg/m^3)$$

$$\text{Retention} = \frac{M_{ai} - M_{bi}}{M_{bi}} \times 100 \ (\%)$$

In the formulations,

$G = T_2 - T_1$

T_2= the weight of sample after impregnation (kg)

T_1= the weight of sample before impregnation (kg)

M_{ai}= after impregnation, completely dry sample weight (kg)
M_{bi}= before impregnation, completely dry sample weight (kg)
C: solution concentration
U: sample volume

Tab. 1: The plan of impregnation

Groups of Chemical	Ingredients	Imp. Test No	Imp. Process Number	I.Process	II. Process	Concentration of solution (%) I. Imp.	II .Imp.	Solvent I. Imp.	II .Imp.
I. Group Commercial impregnation chemicals	Boron compounds	1	1	Tanalith-CBC	-	13	-	DW	-
	Ammonium compounds	2	1	Ammonium sulphate	-	13	-	DW	-
	Containing phosphorus compounds	3	1	Di-ammonium phosphate. (DAP)	-	13	-	DW	-
	Organic solvents	4	1	Vacsol (V)	-	100	-	-	-
II. Group Boron compounds	Fire retardants, insecticides and fungisits	5	1	Boric acid (BA)	-	13	-	DW	-
		6	1	Borax (BX)	-	13	-	DW	-
		7	1	BA+BX (7:3. W:W)	-	13	-	DW	-
		8	2	BA+BX (7:3)*	Stiren (St)	13	100	DW	-
		9	2	..	MMA	13	100	DW	-
		10	2	..	St+ MMA	13	70:30	DW	-
		11	2	..	ICO	13	100	DW	-
		12	2	..	ICO	13	100	DW	-
		13	1	P+BA+BX	-	13	-	DW	-
III. Group PEG-400		14	1	PEG-400	-	100	-	-	-
		15	2	PEG-400	St	100	100	-	-
		16	2	PEG-400	MMA	100	100	-	-
		17	2	PEG-400	St+MMA	100	70:30	-	-
		18	2	PEG-400	ICO	100	100	-	-
IV. Group WR compounds	Water repellent	19	1	St	-	100	-	-	-
		20	1	MMA	-	100	-	-	-
		21	1	St+MMA	-	70:30	-	-	-
		22	1	ICO	-	100	-	-	-

Determination of hygroscopicity of wood specimens

The hygroscopicity measurements of wood specimens were made with respect to ASTM D 3201-86 (1986) standard. According to the corresponding standard, after keeping the samples in an environment, having 90 ± 3 % relative moisture and 27 ± 2°C temperature, for 7 days, their weights are determined and so, the change percentages in hygroscopicity of wood, caused by chemical substances, are calculated by using the formula, given below:

$$\text{Moisture increment} = \frac{R_{last} - R_{first}}{R_{first}} \times 100 \ (\%)$$

$$\text{Change in hygroscopicity} = \frac{R_{last(test)} - R_{first(kontrol)}}{R_{first(kontrol)}} \times 100 \ (\%)$$

R_{last} = sample moisture after impregnation (%)
R_{first} = sample moisture before impregnation(%)

The variance analysis with 95 % reliability and DUNCAN Tests (DT) of all results, obtained at the end of the studies, are discussed by using statistical software STATGRAF.

RESULTS

Results about properties of impregnation solutions

The properties of the solutions, used in impregnation of wood specimens are given in Tab. 2 in accordance with the impregnation experiment number.

According to the Tab. 2:

1. There is no change in pH values and density of solutions, measured before and after the impregnation. To use fresh solution in every impregnation may result in this situation.
2. In the solutions, where BA is used alone, and especially, in 13 % solution of commercial Tanalith-CBC, pH values, being in acidic region, strengthen the possibility that these solutions affect the polysacharides of the wood negatively. When the borax is used alone, the borax being in base region, should be taken account that bases have lignin base extractive solvent effect. In this case, resistance properties are under consideration. pH values of 7.81-7.95 in the solutions, prepared by mixing BA and BX in the ratio of 7:3 (weight/weight), are so close to neuter. Therefore, it is assumed that the chemical compounds in the wood are affected so little.
3. The pure solution of PEG-400, being in acidic region (5.60-5.67), may result in a small amount of decrease in the bending strength.
4. WR substances such as; St, ICO and MMA. Their water abducent activity values are high and they also make the mechanical properties better in the wood. It is determined that pH values of St and ICO are in the acidic region whereas the one of MMA is so close to the neuter. In this situation, it is expected that MMA affects the mechanical properties in positive manner. In the mixture of ST and MMA(70:30), the pH value of the mixture(5.65-5.70) is found by taking the average of their individual pH values.

Tab. 2: Peculiarities of solution used for impregnation of wood specimens

Group No	Imp. Test No	Sequence of Process	Solvent	Concentration of solution (%)	pH		Density		%	
					BI	AI	BI	AI	BI	AI
I	1	1. Tanalith-CBC	DW	13	2.48	2.79	1.08	1.08	48.2	48.2
	2	1.AS	DW	13	4.55	4.06	1.07	1.07	-	-
	3	1.DAP	DW	13	6.64	6.70	1.07	1.07	-	-
	4	1.V	-	100	5.91	6.00	0.81	0.81	-	-
II	5	1.BA	DW	13	4.60	4.64	1.02	1.02	35.3	35.3
	6	1.Bx	DW	13	11.2	11.3	1.02	1.02	33.4	33.4
	7	1.BA-BX	DW	13	7.86	7.91	1.11	1.11	62.3	62.3
	8	1.BA-BX 2.St	DW -	13 100	7.86 4.14	7.91 4.10	1.11 0.91	1.11 0.91	62.3 -	66.8 -
	9	1.BA-BX 2.MMA	DW -	13 100	7.86 7.41	7.91 7.85	1.11 1.22	1.11 1.22	62.3 -	66.8 -
	10	1.BA+BX 2. St+MMA	DW -	13 70:30	7.86 5.70	7.91 5.73	0.91 1.12	0.91 1.12	62.3 -	66.8 -
	11	1.BA+BX 2. ICO	DW -	13 100	7.82 4.60	7.92 4.62	0.91 1.21	0.91 1.21	62.3 -	66.8 -
	12	"	"	"	"	"	"	"	"	"
	13	1.P+BA+BX	DW+E+T	13	8.12	8.07	1.03	1.03	-	-
III	14	1. PEG-400	-	100	5.67	5.60	1.12	1.12	-	-
	15	1.PEG-400 2.St	-	100 100	5.67 4.14	5.60 4.10	1.12 0.91	1.12 0.91	-	-
	16	1.PEG-400	-	100	5.67	5.60	1.12	1.12	-	-
	17	1.PEG-400 2. St+MMA	-	100 70:30	5.67 5.70	5.60 5.65	1.12 1.12	1.12 1.12	-	-
	18	1.PEG-400 2.ICO	-	100 100	5.67 4.60	5.60 4.60	1.12 1.21	1.12 1.21	-	-
IV	19	1.St	-	100	4.14	4.10	0.91	0.91	-	-
	20	1.MMA	-	100	7.41	7.85	1.22	1.22	-	-
	21	1.St+MMA	-	70:30	5.70	5.65	1.12	1.12	-	-
	22	1. ICO	-	100	4.60	4.60	1.21	1.21	-	-

AI: After impregnation, BI: Before impregnation, E: Emulgator, T: Triethylenamin

Retention rates of (kg/m^3) the beech wood is given in Tab. 3.

Tab. 3: Amount of retention for beech wood (kg/m³)

Group No	Test no Number of process and sequence		Amount of retention %						
			I. Impregnation		II. Impregnation		Total		
			Average	Standard Deviation.	Average	Standard Deviation	Average	Standard Deviation	HG*
I	1	1. Tanalith-CBC	32.36	1.80	-	-	32.36	1.80	a
	2	1.AS	70.93	12.18	-	-	70.93	12.18	a
	3	1.DAP	81.83	9.12	-	-	81.83	9.12	a
	4	1.Vacsol	343.25	24.10	-	-	343.25	24.10	def
II	5	1.BA	98.00	2.94	-	-	98.00	2.94	a
	6	1.BX	88.25	6.60	-	-	88.25	6.60	a
	7	1.BA+BX	80.19	10.57	-	-	80.19	10.57	a
	8	1.BA+BX 2.St	68.69 -	20.78 -	- 406.28	- 40.03	474.73 -	30.30 -	f
	9	1.BA+BX 2.MMA	70.78 -	7.60 -	- 396.01	- 20.11	466.16 -	87.35 -	f
	10	1.BA+Bx 2. St+MMA	80.00 -	11.15 -	- 401.61	- 48.07	472.13 -	51.83 -	fg
	11	1.BA+BX 2. ICO	89.14 -	5.77 -	- 400.60	- 90.36	480.48 -	89.38 -	fg
	12	1. BA+BX 2. ICO	68.25 -	9.72 -	- 163.00	- 9.74	252.56 -	7.32 -	de
	13	1.P+BA+BX	237.00	22.88	-	-	68.25	22.28	cd
III	14	1. PEG-400	227.70	4.50	-	-	237.00	4.50	b
	15	1.PEG-400 2.St	251.71 -	3.59 -	- 124.08	- 19.86	352.00 -	21.92 -	def
	16	1.PEG-400 2.MMA	240.00 -	29.92 -	- 163.03	- 25.35	419.76 -	8.03 -	ef
	17	1.PEG-400 2. St+MMA	248.12 -	6.85 -	- 169.18	- 47.34	409.89 -	52.05 -	def
	18	1.PEG-400 2.ICO	320.75 -	29.65 -	- 297.54	- 150.30	546.34 -	162.41 -	g
IV	19	1.St	305.75	74.87	-	-	320.75	74.87	d
	20	1.MMA	305.75	28.55	-	-	305.75	28.55	cd
	21	1.St+MMA	204.00	118.00	-	-	204.00	118.00	bc
	22	1. ICO	327.00	14.46	-	-	327.00	14.46	de

* Group of homogeneity (P≤0.05)

According to the Tab. 3:

1.) The highest retention rates in impregnation of +WR and P4+WR, the lowest retention rates in the application of the impregnation, having one action, of salts are observed respectively.

2) According to the results of simple variance analysis and DT, it is determined that the retentions make homogeneous groups, given below, in descending order for beech wood.
   a. PEG-400+ICO
   b. (BA+BX)+(St+MMA),(BA+BX)+ICO, (BA+BX)+St, (BA+BX)+MMA, V, PEG-400+St, PEG-400+(St+MMA)
   c. (BA+BX)+ICO ,ICO, MMA, St, P+BA+BX
   d. PEG-400
   e. Tanalith-CBC, AS, DAP, BA, BX, BA+BX

The retention % for beech wood is given in Tab. 4.

Tab. 4: Rate of retention for beech wood

Group No	Test no Number of process and sequence		Rate of retention %						
			1. Impregnation		II. Impregnation		Total		
			Average	Standard Deviation.	Average	Standard Deviation	Average	Standard Deviation	HG
I	1	1. Tanalith-CBC	13.11	1.33	-	-	13.11	1.33	a
	2	1.AS	20.45	1.98	-	-	20.45	1.98	abcde
	3	1.DAP	23.00	0.84	-	-	23.00	0.84	abcde
	4	1.Vacsol	28.29	3.57	-	-	28.29	3.57	abcdef
II	5	1.BA	70.62	14.15	-	-	70.62	14.15	abcdef
	6	1.BX	68.25	9.97	-	-	68.25	9.94	abcdef
	7	1.BA+BX	48.14	11.87	-	-	48.14	11.87	gh
	8	1.BA+BX 2.St	35.08 -	6.52 -	- 39.03	- 5.91	74.56 -	3.91 -	h
	9	1.BA+BX 2.MMA	28.68 -	2.78 -	- 30.43	- 10.86	59.10 -	8.01 -	efg
	10	1.BA+BX 2. St+MMA	27.12 -	6.72 -	- 21.03	- 3.33	48.16 -	4.36 -	fgh
	11	1.BA+BX 2. ICO	45.26 -	13.44 -	- 29.24	- 12.70	73.63 -	4.29 -	fgh
	12	1. BA+BX 2. ICO	110.00 -	11.98 -	- 7.35	- 4.03	117.37	12.89	abc
	13	1.P+BA+BX	25.08	8.97	-	-	25.08	8 .97	abcdef
III	14	1. PEG-400	15.14	4.37	-	-	14.14	4.37	a
	15	1.PEG-400 2.St	21.62 -	1.41 -	- 3.98	- 1.63	25.38 -	4.27 -	abcdef
	16	1.PEG-400 2.MMA	20.70 -	1.93 -	- 10.21	- 4.25	30.92 -	2.85 -	bcdef
	17	1.PEG-400 2. St+MMA	18.81 -	0.97 -	- 9.38	- 2.48	28.19 -	3.00 -	bcdef
	18	1.PEG-400 2.ICO	19.82 -	2..81 -	- 16.26	- 2.27	36.01 -	2.43 -	abcdef
IV	19	1.St	32.67	8.40	-	-	32.67	8.40	cdefg
	20	1.MMA	23.37	14.36	-	-	28.37	14.36	abcdef
	21	1.St+MMA	15.19	0.83	-	-	15.19	0.83	ab
	22	1. ICO	18.36	2.48	-	-	18.36	2.48	abc

According to the Tab. 4:

1. While % retention rates of chemical substances change between 13.40 % and 73.62 %, the highest retention is obtained by the impregnation of BA, BX, BA+BX and (BA+BX)+ICO whereas the lowest one is gotten by commercial impregnation substances and the solutions with PEG-400.
2. % retention rate of beech wood treated with St, is significantly higher than St+MMA impregnated beech wood..
3. High retention rates, gotten by the salts with boron, show exactly that the beech wood can be obtained with these salts.
4. From the statistical interpretations, in the impregnation of the beech wood with the related chemical substances, they make following homogeneous groups, given in descending order, in terms of c.d.w.w.r % retention.

a) (BA+BX)+ICO, BA, BX
b) BA+BX, (BA+BX)+St, (BA+BX)+(St+MMA)
c) (BA+BX)+MMA,St,V,P+BA+BX,PEG-400 St, P4+MMA, P4+(St+MMA), P4+ISO, MMA, DAP
d) AS, (BA+MMA), PEG-400+ICO, MMA, DAP

Hygroscopicity level and the change of hygroscopicity level of wood specimens compared to control specimen, are given in Tab. 5.

Tab. 5: Hygroscopicity levels of beech wood treated with boron and water repellents

Group No	Impregnation Trial No	Chemicals	Hygroscopicity levels		Differences compared to untreated control specimen	
			Ort.*	**St.Sp**		**(±)**
I	1	1. Tanalith-CBC	13.46	2.12	17.99	(-)
	2	1.AS	10.96	3.75	15.03	(-)
	3	1.DAP	11.13	2.00	12.66	(-)
	4	1.Vacsol	14.42	2.01	23.04	(-)
II	5	1.PEG-400	26.33	6.12	35.42	(+)
	6	1.PEG-400 2.St	18.20	7.56	38.10	(-)
	7	1.PEG-400 2.MMA	15.03	3.78	33.45	(-)
	8	1.PEG-400 2.St+MMA	13.42	4.00	39.55	(-)
	9	1.PEG-400 2.ICO	10.12	2.95	42.25	(-)
III	10	1.BA	6.12	1.45	13.50	(-)
	11	1.BX	8.96	0.75	9.15	(+)
	12	1. BA+BX	12.00	3.45	11.50	(-)
	13	1.BA+BX 2. St	17.15	1.38	31.78	(-)
	14	1. BA+BX 2.MMA	14.26	2.90	28.70	(-)
	15	1.BA+BX 2.St+MMA	15.10	3.98	33.56	(-)
	16	1.BA+BX 2.St+MMA	16.23	5.00	25.10	(-)
	17	1.BA+BX 2.ICO	17.27	1.25	22.88	(-)
	18	1.P+BA+BX	4.56	1.55	85.13	(-)
IV	19	1.St	10.51	2.78	63.05	(-)
	20	1.MMA	13.44	3.15	58.18	(-)
	21	1.St+MMA	16.42	4.56	70.56	(-)
	22	1. ICO	14.50	2.80	55.12	

Note:
* Each trials consist of 5 wood specimens
** In each trial, two hygroscopicity groups, having 24 pieces, are included
*** (-) differences show that hygroscopicity decreases with respect to control
(+) differences show that hygroscopicity increases with respect to control

According to the Tab. 5:

1) The substance, showing the most increase in the hygroscopicity, is PEG-400
2) The most important impregnation substance, decreasing the hygroscopicity, is P+BA+BX (85.13 %).
3) In applications of both salts and boron compounds with WR the hygroscopic properties of salts decrease due to substances of WR.
4) Hygroscopicity convenience order is given as follow; P+BA+BX>WR >Salt based chemicals+WR>PEG-400+WR >Commercial impregnation substances

CONCLUSIONS

This study was designed to determine hygroscopicity levels of beech wood treated with some impregnation chemicals and water repellents

According to the results, the most determinative factor in retention rates of impregnation substances under applied conditions in this study is solvent substance. In addition, while the substance with PEG provides high absorption, the salts dependent to the applied method, give low retention rates.

Although, hygroscopicity level of beech wood treated with PEG-400 were increased compare to that of untreated control specimens; other all chemicals decreased the hygroscopicity levels of beech wood compare to that of untreated control specimen. WR chemicals considerably lowered hygroscopicity level of beech wood. The hygroscopicity level of beech wood was the lowest impregnated with P+BA+BX. Also, other WR chemicals remarkably lowered the hygroscopicity levels of beech wood when applied sole treatment or secondary treatment on borate.

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