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IMPACT OF SILICON-BASED CHEMICALS ON SELECTED PHYSICAL AND MECHANICAL PROPERTIES OF WOOD

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ABSTRACT

This study deals with the impact of silicon-based chemicals on selected physical and mechanical properties of wood. Wood of European beech and Scots pine was the testing material used for impregnation using water glass and commercial product Lukofob EVO 50. The impact of the treatment on dimensional stability, bending strength and modulus of elasticity was tested. Wood density was also evaluated. Although the modification using silicon-based staffs resulted in a statistically significant decrease in swelling for both of the tested species, the positive effect of the treatment was accompanied by a decrease in the strength and stiffness of wood. Water glass had a stronger effect on the tested properties from the chemicals we used in our research.

KEYWORDS: Silicones, chemical modification, properties, swelling, modulus of elasticity, strength.

INTRODUCTION

Due to its properties, wood is a natural material that has been utilized since antiquity. Its positive features mainly include its high strength and elasticity at low weight. Its negative properties include dimensional instability and low resistance against insects, fungi and fire. Such negative characteristics can be limited by modifying wood. Modified wood can be defined as wood with a deliberately altered structure and purposefully improved properties such as resistance to biotic and abiotic damage, water repellency or increased dimensional stability. Mechanical, chemical, thermal and biological methods, as well as combinations thereof, can be used to modify wood. Chemical modification of wood is based on treating wood with chemicals without a direct biocidal or other protective effect (Hon and Shiraishi 2000, Reinprecht and Grznárik 2014).

One way to improve the technical properties of wood, and thereby the lifespan of wooden structures, is to impregnate wood with silicon, which is based on the principle of mineralization. The process of impregnating plant tissues with silicon dioxide is generally considered one of the most common and also the most enduring fossilization processes. Mineralized wood first occurs through the decomposition of the cell wall and the subsequent deposition of silicon dioxide on these walls (Kim et al. 2009). Natural mineralization may take millions of years, but it can be successfully done faster under special conditions (Fengel 1991, Snelling 1995, Akahane et al. 2004, Kim et al. 2009). Artificial oversaturation of wood with silicates leads to structural changes similar to those in natural mineralization. The mineral solution penetrates into the wood, where it remains in lumens, or penetrates the cell walls where it can chemically react with some of the components (Hill 2006). The first attempt to carry out mineralization under laboratory conditions was published in 1968 (Akahane et al. 2004).

Of the various types of organic silicones, those best suited for modification of wood are monomeric silicones (e.g. alkyl trialkoxy silicone) and polymeric silicones. Although small silicone molecules penetrate well into the wood structure (Donath et al. 2006), silicone molecules are not stable and they therefore first react with water molecules and produce silanes, which subsequently reacts with the OH groups of wood (Hill 2006). Experiments were also carried out using water glass (Na_2SiO_3), for which the storing of sodium silicate in an amorphous opal on the outside of the cell wall was typical (Mamoňová et al. 2015).

Water glass (aqueous sodium silicate solution) is widespread and a commonly used substance in practice. It is made of glass sand at a temperature of 1400 - 1600°C using alkaline fluxing agents.

This is a clear or slightly cloudy viscous liquid with no odour. The density is in the range of 1120 - 1680 $kg{\bullet}m^{-3}$ and pH in the range of 11 to 12. Water glass is used as an additive for degreasing, washing and cleaning agents, and as a binder in the production of welding wires or sand moulds in foundries. It is also used for demanding applications such as industrial floors, special fire-resistant materials, colours, as an additive for refractory fireclay mixtures for repairs of stove and fireplace linings, or to protect wood against rot and fire (Svoboda et al. 2013). Another substance used in practice that utilizes the unique properties of silicon is a product known as Lukofob in the commercial sector. Lukofob is a silicone-based liquid that is used for the hydrophobicisation of porous silicate construction materials. Their application limits absorption of rainwater into the surface, reduces dirtiness and washout of soluble components, there are no acid rain effects, and the thermal insulation of brick silicate materials (plaster, concrete, ceramic roofing, etc.) is maintained. In this study, specifically Lukofob EVO 50 was used, which has the alkylsiloxane active ingredient. Due to its small size, the active ingredient in the preparation gets deep inside the treated material. The colour of the substance is milky white, but it cannot be seen after it is applied to the material. The density ranges from 900 – 950 $kg{\bullet}m^{-3}$ and pH is in the range of 8-10 (Lukofob EVO 50 – technical sheet).

Studies utilizing modification of wood with silicone are mostly concerned with the impact of the modification on resistance against fungi or other biotic factors. Impregnation of wood with NH_2 R- silicone colloidal solution increased the resistance of the treated wood against the effects of wood decaying insects (Predieri et al. 2011). Wood impregnated by alkoxysilanes showed greater resistance to fire (Saka and Ueno 1997), or increased resistance against soil microorganisms and termites (Donath et al. 2004, Feci et al. 2009). Very few studies are devoted to the impact of modification on the properties of wood (Brebner and Schneider 1985).

Based on an analysis of the literature, it can be stated that the authors are mostly devoted to wood impregnation using silicon-based solutions in terms of chemical dependence on wood mass and its subsequent use against the effects of biotic and abiotic factors. However, with regard to the subsequent use of modified wood, particularly in structures, it is also important to assess the impact of the modification on physical properties and strength and elasticity characteristics. This study examines the influence of chemical modification of wood using commonlyavailable silicon-based substances on selected physical and mechanical properties of representatives of common commercial deciduous and coniferous trees in the Czech Republic.

MATERIAL AND METHODS

Beech wood (*Fagus sylvatica* L.) was used to produce test specimens as a representative of one of the most important native hardwoods. Scots pine (*Pinus sylvestris* L.) was used as a representative of one of major native softwoods. The test material was free of defects (false heartwood in beech). In the case of the pine sapwood was used. Both types of wood belong to easily-impregnable species. Test specimens with dimensions of 20x20x30 mm (tangential x radial x longitudinal) were produced in order to determine physical properties (swelling and density). Test specimens 20x20x300 mm (tangential x radial x longitudinal) were made for mechanical tests (bending strength, static modulus of elasticity).

The silicone-based chemicals used for modifying wood were water glass and a substance with the commercial name Lukofob EVO 50. In our case the passive chemical modification was used that deals with introducing modifying substances into lumens, or partially into the cell walls of wood, and these purposefully do no create chemical bonds with wood components, or these are bonds with low stability. Impregnation via dipping was carried out for 24 hours at atmospheric pressure and under standard laboratory conditions, i.e. at 20°C. A 10 % concentration was used for both of the chemicals. Test specimens were divided into four parts. The first series of test specimens was impregnated using Lukofob EVO 50, the second quarter of the samples was impregnated with water glass, and the third quarter was impregnated with a mixture of Lukofob EVO 50 and water glass. The last series was left as a reference. There were 30 samples for each series.

After the test specimens were removed from the solutions they were conditioned to equilibrium moisture content. Samples intended for determining swelling were dried in a kiln at 103± 2°C to zero moisture content. The relevant tests were then carried out on the modified samples.

Determination of physical properties

Density at 12 % wood moisture content was calculated according to the following Eq. 1 (ČSN 49 0108):

$$\rho_{12} = \frac{m_{12}}{V_{12}} \qquad (1)$$

where: ρ_{12} - wood density at 12 % moisture content ($kg \cdot m^{-3}$),
m_{12} - weight of the wood at 12 % moisture content (kg),
V_{12} - wood volume at 12 % moisture content (m^3).

Basic wood density ρ_k was also determined, which is calculated according to the following Eq. 2:

$$\rho_k = \frac{m_0}{V_{max}} \qquad (2)$$

where: ρ_k - basic wood density ($kg \cdot m^{-3}$),

m_0 - weight of the wood in an oven dry state (kg),
V_{max} - wood volume at a moisture content above the fibre saturation point (m^3).

Volumetric swelling was determined according to the following formula (ČSN 49 0126):

$$\alpha_{V\,max} = \frac{V_{max} - V_{min}}{V_{min}} \cdot 100 \qquad (3)$$

where: $\alpha_{v\,max}$ - maximum volumetric swelling (%),
V_{max} - volume of the specimen at a moisture content at or above the fibre saturation point (cm^{-3}),
V_{min} - volume of the specimen in an oven dry state (cm^3).

Determination of mechanical properties

Tests were carried out on a TIRA 50 kN testing machine using 240 mm support spacing, i.e. 12 times the height of the specimen. All of the tests were determined for 12 % wood moisture content.

The modulus of elasticity was calculated according to the following formula (ČSN 49 0115):

$$E_{ohw} = \frac{1}{4} \cdot \frac{\Delta F \cdot l_0^3}{b \cdot h^3 \cdot \Delta y} \qquad (4)$$

where: E_{ohw} - the modulus of elasticity (MPa),
ΔF - the force difference between the upper and lower load threshold (N),
l_0 - the distance between the supports in mm, b, h is the width and height of the specimen (mm),
Δy - the bend of the test specimen in a zone of pure bending equal to the difference in bend values corresponding to the upper and lower load threshold (mm).

Bending strength was calculated according to the following formula (ČSN 49 0116):

$$\sigma_{pohw} = \frac{3 \cdot F_{max} \cdot l_0}{2 \cdot b \cdot h^2} \qquad (5)$$

where: σ_{pohw} - bending strength (MPa),
F_{max} - the force corresponding to the breaking strength in (N),
l_0 - the distance between the supports (mm),
b - the width of the specimen (mm),
h - the height of the specimen (mm).

A so-called qualitative number (Požgaj et al. 1993) was used as another indicator of the effect of the modification on the quality of the testing material. The qualitative number represents theproportion of adequate strength and density. This indicator is better way of informing about the impact of the modification on the practical usability of the material.

Analysis of variance (ANOVA) was used in order to assess the difference between the evaluated sets. Duncan's test was used for subsequent testing of a hypothesis about the conformity of the mean values of two independent sets and $\alpha = 0.05$ was used as the level of significance.

RESULTS AND DISCUSSION

Although the foregoing impregnating substances caused a statistically significant decrease in wood swelling in both of the assessed woods, i.e. beech and pine, the positive effect of such modification was accompanied by a decrease in the strength and elasticity properties of wood, i.e. the strength and modulus of elasticity (Tabs. 1-3). In contrast to literature (Ghosh et al. 2008; Reinprecht and Grznárik 2014), our tests confirmed a statistically significant decrease not only in strength, but also in the modulus of elasticity, even more significantly than in the final strength. The impact of the relevant modification on wood absorption has not been evaluated, as the assumption of higher water absorption values for such modified wood is obvious, and in terms of its practical use, the free water content in wood is not relevant.

Tab. 1: Basic statistical analyses of the physical and mechanical properties for untreated and treated beech wood.

	Treatment degree	Mean	Median	Minimum	Maximum	Standard deviation	Coefficient of variation (%)
Density ($kg.m^{-3}$)	1	746	747	708	768	14	1.9
	2	749	752	716	779	16	2.2
	3	738	762	740	781	13	1.7
	4	764	764	729	804	15	2.0
Basic wood density ($kg.m^{-3}$)	1	581	590	506	603	25	4.4
	2	589	600	523	619	27	4.6
	3	589	595	507	617	26	4.5
	4	609	609	578	641	19	3.1
Volumetric swelling (%)	1	16.8	16.9	14.9	18.4	1.1	6.3
	2	19.1	18.9	16.5	23.0	1.5	7.7
	3	17.3	17.3	16.0	18.4	0.7	3.9
	4	20.8	21.6	18.3	23.0	1.5	7.0
Modulus of elasticity (MPa)	1	11700	11800	10200	13000	574	4.9
	2	12200	12200	9500	13800	876	7.2
	3	11600	11600	7900	12800	869	7.5
	4	13000	13200	7700	14900	1100	8.8
Modulus of rupture - bending strength (MPa)	1	117	118	88	132	10	8.8
	2	122	122	111	132	6	4.8
	3	114	114	86	130	9	8.0
	4	123	125	56	135	13	10.6
Qualitative number (km)	1	1.57	1.58	1.25	1.77	1.3	8.2
	2	1.63	1.63	1.46	1.73	0.7	4.4
	3	1.50	1.50	1.15	1.70	1.2	7.9
	4	1.61	1.64	7.3	1.76	1.7	10.5

Treatment degree: 1 = water glass, 2 = Lukofob EVO 50, 3 = water glass + Lukofob EVO 50,
4 = reference, with no treatment.

Tab. 2: Basic statistical analyses of the physical and mechanical properties for untreated and treated pine wood.

	Treatment degree	Mean	Median	Minimum	Maximum	Standard deviation	Coefficient of variation (%)
Density ($kg.m^{-3}$)	1	569	556	513	651	41	7.3
	2	571	559	504	729	50	8.7
	3	561	559	499	636	30	5.4
	4	575	579	469	691	46	8.0
Basic wood density ($kg.m^{-3}$)	1	475	473	423	541	30	6.4
	2	485	471	449	611	44	9.0
	3	485	490	389	543	36	7.4
	4	492	467	411	639	62	12.7
Volumetric swelling (%)	1	9.7	9.8	8.0	11.2	1.0	10.4
	2	10.3	10.4	8.0	12.2	1.2	11.9
	3	9.9	10.1	7.4	11.5	1.4	14.4
	4	11.4	11.4	8.6	13.4	1.4	12.5
Modulus of elasticity (MPa)	1	8600	8800	7000	10300	1000	11.5
	2	8700	8600	6900	10700	1000	11.9
	3	8200	8100	6000	10500	1100	13.3
	4	9600	9900	7400	11600	1400	14.1
Modulus of rupture - bending strength (MPa)	1	87	88	64	99	7	8.1
	2	90	91	72	106	8	9.2
	3	85	83	73	109	8	9.3
	4	93	94	75	109	9	9.6
Qualitative number (km)	1	15.3	15.5	11.3	16.9	1.1	7.4
	2	15.9	15.6	12.2	19.3	1.8	11.4
	3	15.2	14.9	13.2	20.0	1.4	9.0
	4	16.2	16.3	11.6	21.1	2.0	12.3

Treatment degree: 1 = water glass, 2 = Lukofob EVO 50, 3 = water glass + Lukofob EVO 50, 4 = reference, with no treatment.

It is evident from the obtained results that the wood became lighter after the impregnation, which was caused by dissolving the substances in the wood, as the impregnating substances were strong alkalis. With reference to the literature (Reinprecht et al. 2013; Mamoňová et al. 2015), we can therefore conclude that there was also a weakening of the cell walls, in particular the attenuations that provide functions for the transport of water and organic substances with subsequent permeability for water molecules. During impregnation with Lukofob 39, it was revealed that silanolate penetrates into the secondary cell wall, which is reflected by the peeling of the secondary cell wall S3 and the formation of cracks in the penetration zone. The use of water glass exhibits similar effects. It is therefore evident from the results that after impregnation the wood became less strong and brittle, which is caused by the fact that the polyphenol (lignin) poorly resists alkalis.

Tab. 3: Changes in wood property of treated wood in comparison to the reference (untreated) wood (%).

	Treatment degree	Beech	Pine
Density	1/4	-2.4	-1.0
	2/4	-2.0	-0.7
	3/4	-3.4	-2.4
Basic wood density	1/4	-4.6	-3.5
	2/4	-3.3	-1.4
	3/4	-3.3	-1.4
Volumetric swelling	1/4	-19.2	-14.9
	2/4	-8.2	-9.6
	3/4	-16.8	-13.2
Modulus of elasticity	1/4	-9.5	-10.4
	2/4	-6.0	-8.6
	3/4	-10.5	-13.9
Modulus of rupture - bending strength	1/4	-4.9	-6.5
	2/4	-0.8	-3.2
	3/4	-7.3	-8.6
Qualitative number	1/4	-2.9	-6.1
	2/4	1.0	-1.9
	3/4	-6.7	-6.5

1/4 = water glass vs. reference, with no treatment, 2/4 = Lukofob EVO 50 vs. reference, with no treatment, 3/4 = water glass + Lukofob EVO 50 vs. reference, with no treatment.

A more significant impact on all of the assessed properties was demonstrated when using water glass, and it can be stated that the positive impact of these modifications on reducing wood swelling (by as much as 19.2 % for beech when using water glass) overcame the negative effect on strength reduction (by as much as 8.6 % for pine using both substances together) and wood elasticity in bending (by as much as 13.9 % for pine using both substances together). From the perspective of qualitative numbers, which is the proportion of strength to density, this effect is of course much less noticeable due to a decrease in the density of the modified wood. Furthermore, it is necessary to state that although the wood modified in this manner is visibly more brittle, it is also harder.

The obtained data were subjected to a factor analysis (Tabs. 4-9 and Fig. 1). The paper includes the results of a two-factor analysis - the influence of the tree species and modification on all of the assessed properties.

Tab. 4: Analysis of variance for density.

Monitored factor	Sum of squares	Degree of freedom	Variance	Fisher´s F-test	Significance level
Intercept	109359083	1	109359083	38614.75	$P< 0.01$
1 - Wood species	2049059	1	2049059	723.52	$P< 0.01$
2 - Treatment degree	12534	3	4178	1.48	$P = 0.22$
1*2	1624	3	541	0.19	$P = 0.90$
Error	691021	244	2832		

Significance was accepted at $P < 0.01$.

Tab. 5: Analysis of variance for basic wood density.

Monitored factor	Sum of squares	Degree of freedom	Variance	Fisher´s F-test	Significance level
Intercept	34758032	1	34758032	26764.30	$P < 0.01$
1 - Wood species	347871	1	347871	267.87	$P < 0.01$
2 - Treatment degree	7941	3	2647	2.04	$P = 0.11$
1*2	935	3	312	0.24	$P = 0.87$
Error	145451	244	1299		

Significance was accepted at P < 0.01.

Tab. 6: Analysis of variance for volumetric swelling.

Monitored factor	Sum of squares	Degree of freedom	Variance	Fisher´s F-test	Significance level
Intercept	24917.77	1	24917.77	16019.14	$P < 0.01$
1 - Wood species	2007.37	1	2007.37	1290.50	$P < 0.01$
2 - Treatment degree	146.96	3	48.99	31.49	$P < 0.01$
1*2	24.36	3	8.12	5.22	$P < 0.01$
Error	174.22	244	1.56		

Significance was accepted at P < 0.01.

Tab. 7: Analysis of variance for modulus of rupture.

Monitored factor	Sum of squares	Degree of freedom	Variance	Fisher´s F-test	Significance level
Intercept	2719215	1	2719215	32865.98	$P < 0.01$
1 - Wood species	57345	1	57345	693.10	$P < 0.01$
2 - Treatment degree	2825	3	942	11.38	$P < 0.01$
1*2	54	3	18	0.22	$P = 0.89$
Error	20188	244	83		

Significance was accepted at P < 0.01.

Tab. 8: Analysis of variance for static modulus of elasticity.

Monitored factor	Sum of squares	Degree of freedom	Variance	Fisher´s F-test	Significance level
Intercept	2.749801E+10	1	2.749801E+10	26490.20	$P < 0.01$
1 - Wood species	7.060102E+08	1	7.060102E+08	680.13	$P < 0.01$
2 - Treatment degree	6.712384E+07	3	2.237461E+07	21.55	$P < 0.01$
1*2	7.173548E+05	3	2.391183E+05	0.23	$P = 0.88$
Error	2.532829E+08	244	1.038045E+06		

Significance was accepted at P < 0.01.

Tab. 9: Analysis of variance for qualitative number.

Monitored factor	Sum of squares	Degree of freedom	Variance	Fisher´s F-test	Significance level
Intercept	62196.51	1	62196.51	29236.32	P< 0.01
1 - Wood species	0.91	1	0.91	0.43	P = 0.51
2 - Treatment degree	50.09	3	16.70	7.85	P< 0.01
1*2	4.13	3	1.38	0.65	P = 0.59
Error	519.08	244	2.13		

Significance was accepted at P < 0.01.

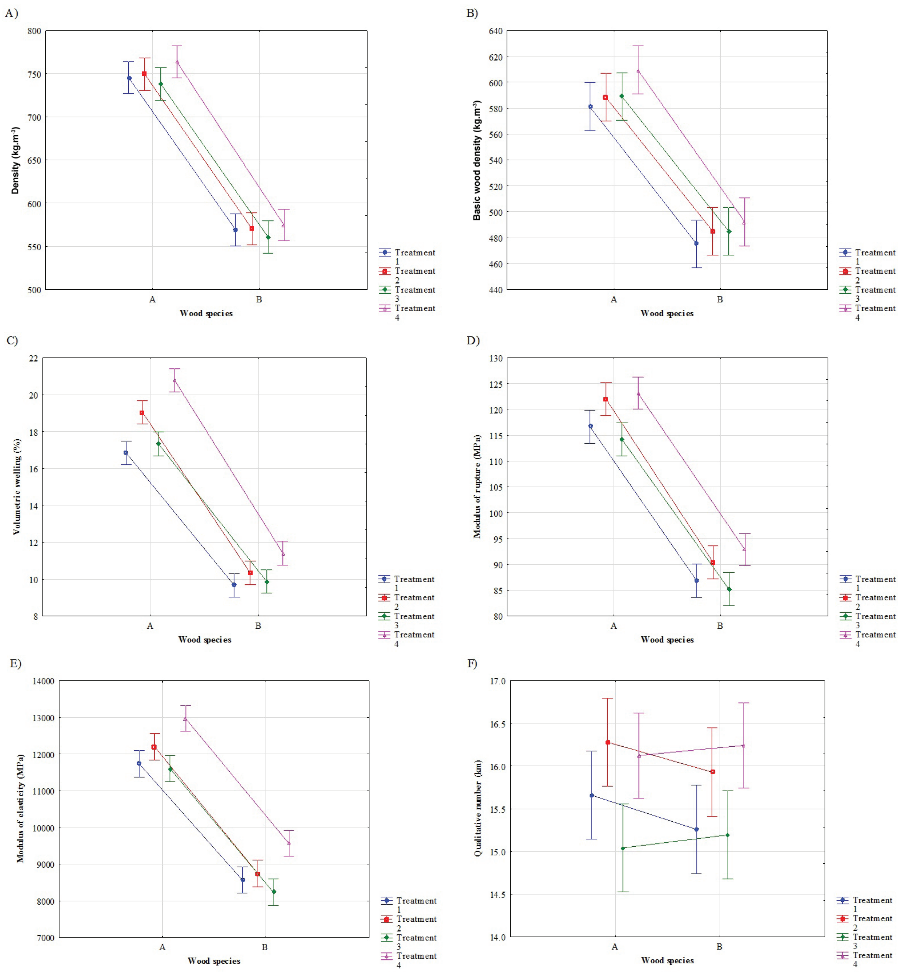

Fig. 1: Graphic visualization of the effect of wood species and treatment on A) oven dry density, B) basic dry density, C) volumetric swelling, D) modulus of rupture, E) modulus of elasticity and F) qualitative number, at a 95 % significance level. X-axis: A = beech and B = pine. Treatment degree: 1 = water glass, 2 = Lukofob EVO 50, 3 = water glass + Lukofob EVO 50, 4 = reference, with no treatment.

Statistical factor analyses showed that the impact of tree species on all of the assessed properties is of course statistically significant at the chosen level of significance, with the exception of the qualitative number, which is logical because a decrease in density also led to a decrease in strength. As a factor, the modification is statistically significant in all cases.

A more detailed Duncan's test showed us that the differences between the individual modifications are statistically significant for pine, and only in some cases for beech. The combination of both factors is negligible in all cases.

CONCLUSIONS

It was found that passive chemical impregnation of wood using silicon-based modifying substances, i.e. water glass and Lukofob EVO 50, which are stronger alkalis, reduced swelling, but this also decreased the wood's strength and modulus of elasticity. The decrease in values was statistically significant for all of the modifications. There proved to be a more significant difference between individual modifications in pine than in beech. The modification is therefore a change to the structure of wood cell wall in a manner that negatively affects its mechanical properties. Thus, such modified wood is particularly suitable for exterior cladding due to lower shape and dimensional changes. It is not very suitable for use in constructions, in particular for lower values of elasticity and strength characteristics.

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