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PHYSICAL AND MECHANICAL PROPERTIES OF DENSIFIED BEECH WOOD PLASTICIZED BY AMMONIA

Petr PAŘIL
Mendel University in Brno – Department of wood science
Address: Zemědělská 1, Brno, Czech Republic
E-mail: xparil1@mendelu.cz

Martin BRABEC
Mendel University in Brno – Department of wood science
Address: Zemědělská 1, Brno, Czech Republic
E-mail: xparil1@mendelu.cz

Radim ROUSEK
Mendel University in Brno – Department of wood science
Address: Zemědělská 1, Brno, Czech Republic
E-mail: xparil1@mendelu.cz

Ondřej MAŇÁK
Mendel University in Brno – Department of wood science
Address: Zemědělská 1, Brno, Czech Republic
E-mail: xparil1@mendelu.cz

Peter RADEMACHER
Mendel University in Brno – Department of wood science
Address: Zemědělská 1, Brno, Czech Republic
E-mail: xparil1@mendelu.cz

Petr ČERMÁK
Mendel University in Brno – Department of wood science
Address: Zemědělská 1, Brno, Czech Republic
E-mail: xparil1@mendelu.cz

Aleš DEJMAL
Mendel University in Brno – Department of wood science
Address: Zemědělská 1, Brno, Czech Republic
E-mail: xparil1@mendelu.cz

Abstract:
Gaseous ammonia treatment in combination with densification of wood has been known since several decades, but nowadays there is no industrial production of thus modified material; also little research is published in this area of wood science. Selected physical and mechanical properties, i.e., density profile, bending strength, hardness and equilibrium moisture content were investigated for Lignamon material, which was obtained from Czech industrial production. Densitometry showed a large variability of the density profile. It is due to the position of the sample in the original block and is affected by the loading of the sample in tests (direction orthogonal or parallel to compressing). Strength properties, hardness and moisture exclusion efficiency of Lignamon are enhanced. Further investigation will be carried out with self-produced samples.

Key words: *ammonia treatment; densified wood; Fagus sylvatica L.; plasticization; density; hardness; modulus of rupture; modulus of elasticity; Lignamon.*

INTRODUCTION

Scheuerch (1963) started a series of experimental work focused on the plasticizing effect of ammonia on wood. It has been shown that this process is suitable to allow the creation of extreme shapes or complicated structures without decreasing the strength (Berzins 1972, Davidson and Baumgardt 1970, Kalnins *et al*. 1967, Pandey *et al*. 1991). Plasticization by ammonia has lower energy consumption than conventional steaming and does not require long time for the process settings (Strauss 1995, Wienhaus *et al*. 1978). Wood is a natural heterogeneous material consisting of three main components: cellulose, hemicellulose and lignin. These components are responsible for most physical and mechanical properties of materials (Banks and Gibson 1988). Ammonia has a relative high affinity to main components of wood. Ammonia can penetrate into the crystal structure of cellulose. It leads to disruption of lignin-carbohydrate complex. Increases of the nitrogen content and the number of amide bonds can occur with a decrease in ester bonds. Ammonia treatment of wood has no significant influence on the content of cellulose, hemicellulose and lignin; plasticizing effect is temporary, the wood returns to its solid form even with a more compressed / compact structure after evaporation of ammonia (Berzings and Rocens 1970).

Ammonia treatment may significantly upgrade the decorative value of wood (Weigl *et al*. 2012) and improve colour stabilization (constant colour of wood over time) and dimensional stability (Weigl *et al*. 2009).

Rosca *et al*. (2002) published a study about the effect of ammonia treatment on physical and mechanical properties of wood. The authors confirmed that the deformation of ammonia plasticized wood increased when the initial moisture content of the material is higher than 20%. The results showed reduction of modulus of elasticity and modulus of rupture in compression which leads to increasing of deformability (Rocens 1976).

The changes are effective in the whole structure or only in the surface layers of wood with respect to the processing time, the dimensions of the material and the concentration of ammonia. Tannins are perceived as wood extracts that are responsible for reacting with ammonia, which lead to the required colour changes of wood (Tinkler 1921, Bariska 1969). The content of extractives in solid beech is about 2% (Wagenführ 1966).

A commercially applied process of wood plasticized by ammonia is so-called Lignamon. It is technology using mainly beech wood, which is plasticized by ammonia gas and then densified in the orthogonal direction to wood fibres (up to 60%). Some tropical or subtropical wood species can be replaced by this modified material (improved hardness, colour etc.).

This technology was developed in the 1960. The research was carried out by the Institute of Wood Chemistry on Scientific Academy in Latvia (Riga), led by I. Y. Kalninsha (Czerny and Valasek 1974). The methodology for process industrialization was developed in cooperation with A. Stojčev (Stojčev 1979). Unique semi industrial line for ammonia modified wood was in 1974 produced in cooperation with the Czechoslovak Institute (VVÚD - Timber Research Institute in Prague). Analyses have shown that processes designed by these authors are much more effective than the processes developed in the USA, Austria and other countries. The patent for that modified wood is owned by Soviet and Czechoslovak researchers.

OBJECTIVES

The aim of this paper is to investigate selected physical and mechanical properties of ammonia treated and compressed beech wood (Lignamon). Influence of ammonia and compression on investigated wood properties is given and obtained results are discussed. This study is the first step for further research, which will be the development of ammonification unit and determine optimal parameters for plasticization, compression and thermal modification.

METHOD, MATERIALS AND EQUIPMENTS

Material called Lignamon was obtained from industrial production (closed in 2010) in the Czech Republic. Material for samples were from one process and thus ensure beech from a consistent source. Lignamon is the trade name for thermally, chemically and mechanically modified wood. Modification process is initiated by chemical modification of gaseous ammonia using vacuum and pressure. The transversal compression of wood was carried out in the radial or tangential direction. Modification process is completed by a thermal stabilization of ammonia in the structure of wood. The whole process is carried out in a single device with built-in compressing plates. The total duration of the process is up to 30 hours depending on the material and the parameters of the modification. Modification process was optimized for wood of European beech (*Fagus sylvatica* L.). The moisture content of the wood entering this process is 18%. Finally material with modified physical, mechanical and chemical properties of wood with various levels of compression (15%, 30% and 50%) is produced.

15 prisms with dimensions of 35×80×740mm^3 were available from old production process. Different compress directions of each sample were found. Most of samples were made from the side boards so the compress direction was not purely tangential or radial. Only a few of them were compressed strictly in the tangential direction. Density range was from 885kg/m^3 to 1185kg/m^3 (conditioned in 65% rH and 20°C).

Sample size for densitometry and hardness was 50×50 × thickness mm^3. Transverse dimensions of bending samples were $14×14mm^2$ and length was 210mm. The ratio of these dimensions followed the British Standard BS 373-1957. The material was cut on a circular saw and machined using a cylindrical grinder. Sanding used a p150 grit size. Each sample was weighed and sorted by weight, then marked with a code defining the order position in the original block and the direction of compression in the production. 147 quality bending samples were obtained.

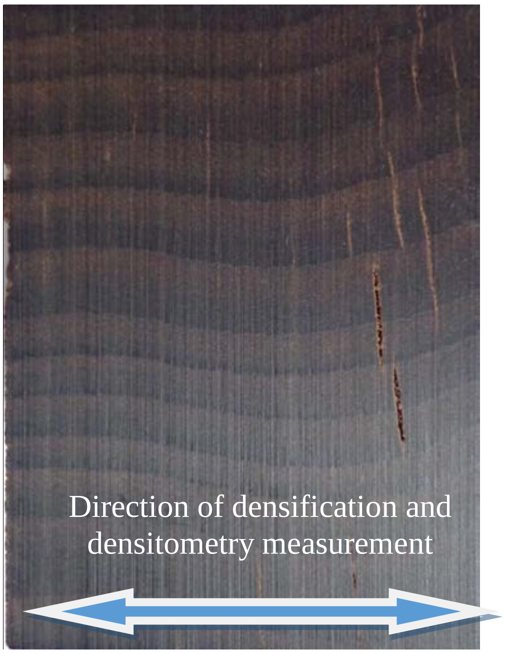

Fig. 1
Cross-section of sample.

The aim for x-ray densitometry measurement was to obtain density profile of available Lignamon blocks. This gave information in which direction the lignamon blocks were pressed, and if the blocks were cut after. The measurement was performed on X – RAY DENSE – LAB densitometer. Principle of measurement is sending of an x-ray beam through the sample and its intensity is measured on the other side. This scan is carried out in a single line in the direction of thickness, with steps of 0,01mm. Direction of x-ray beam is orthogonal to sample width. Computational software analyses outputs and matches average intensity with average specific density, so the intensity of each measured position can be converted to specific density.

The static bending test was carried out according to British Standards BS 373-1957. Bending test specimens were conditioned in a conditioning chamber (SANYO MTH 2400) at 20±3°C and relative humidity of 65±2%. The process of conditioning was controlled by weighting 40 pieces of representative samples at regular intervals. Modulus of rupture (MOR) and modulus of elasticity (MOE) were determined during the static bending test by central loading method (three-point bend). The span of supports was 196mm. For the calculation of MOE a linear part of stress-strain curve was used; limit values were 0,1 × F_{max} and 0,4 × F_{max} with corresponding deflections. Input data for TestXpert application were obtained by weighing and measuring of each sample dimension. Each sample was placed between bending grips of the universal testing device (ZWICK Z 050) and the loading was launched. Samples marked with odd numbers were loaded parallel to direction of compression. The other 50% of samples were loaded orthogonal to the direction of compression. The density of each sample was calculated by using its weight and dimensions. Small samples were made immediately after testing for determination of moisture content (MC) of wood. The small samples were dried to 0% MC in a laboratory drying kiln (SANYO MOV 112) at a temperature of 103±2°C. Afterwards, each sample was weighed and measured. The obtained data were used to calculate the density of each sample.

Janka hardness method in combination with Brinell hardness method was used to determine the hardness. Hardness test specimens were conditioned in a conditioning chamber (SANYO MTH 2400) at 20±3°C and relative humidity of 65±2%. Testing of hardness was carried out on universal testing device (ZWICK Z 050) according to Czech Standard ČSN 49 0136. Impress depth was reduced to 2.82mm because of small dimensions of the sample in case of Janka method. The force was 1000N for Brinell method. Hardness of Lignamon was measured on the side surface in the compressed direction and orthogonal to the pressing direction.

The moisture absorption test was done by placing the specimens into the conditioning chamber (65% rH and 20°C). The EMC and swelling in all directions were measured.

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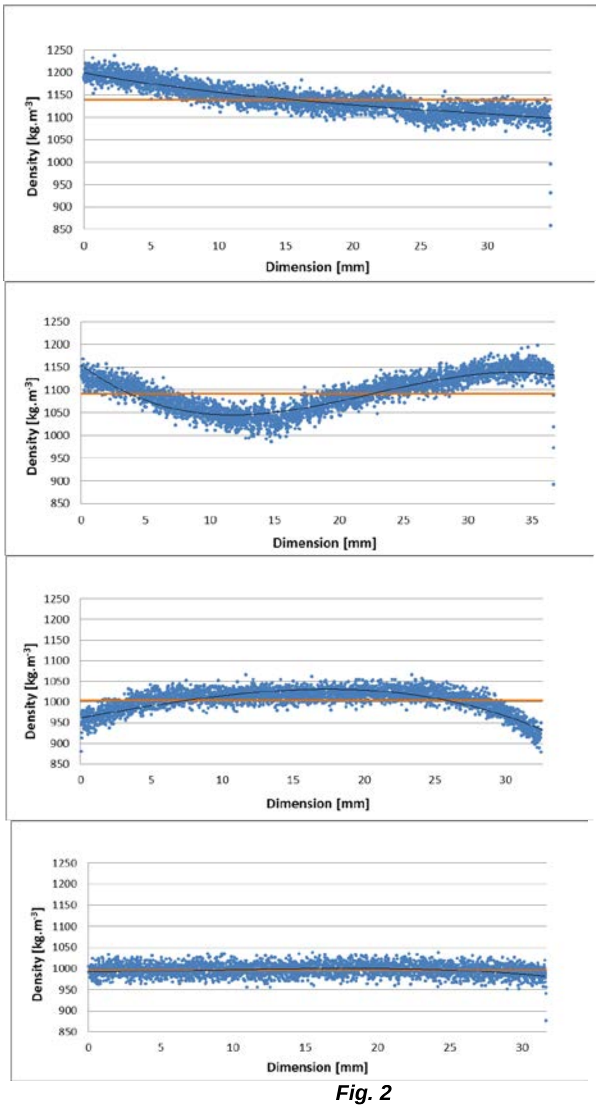
RESULTS

Density

Four types of density profile are shown in Fig. 2. The first has a higher density on the outer sides than in the middle (Fig. 2b). The second has opposite profile than first - lower density on the outer sides than in the middle (Fig. 2c). The third has on the first side a higher density than on the second and the course of the cross-section is more or less linear (Fig. 2a). The final type of profile has nearly constant density in the whole cross section (Fig. 2d). Different density distributions are probably caused and influenced by the compressing direction, level of compressing, anatomical structure of wood and degree of plasticization. Densitometry results gave an image of specific density profile and range for 15 available samples. The idea, that samples were cut after compressing, and that original manufactured dimensions were different, was partially confirmed. Almost uniform specific density and low specific density range in Fig. 2d can be interpreted as the sample was cut from a middle section of compressed wood. But there is still a question of original conditions. Further look at the results suggests that samples with lower average specific density have more uniform density profile (Fig. 2d), and samples with higher average specific density have higher density zones on the edges (Fig. 2a,b). This could lead to the conclusion, that lower compressing level means more uniform density profile. Special case can be seen in Fig. 2c, where probably spring back effect occurs. The range of specific density for each sample varied from 100 to 200kg/m^3, coefficients of variation from 1,4 to 3,7%. Further investigation has to be carried out with self-produced samples. Density of solid beech in the same conditions is about 720kg/m^3 (Požgaj 1997).



a

b

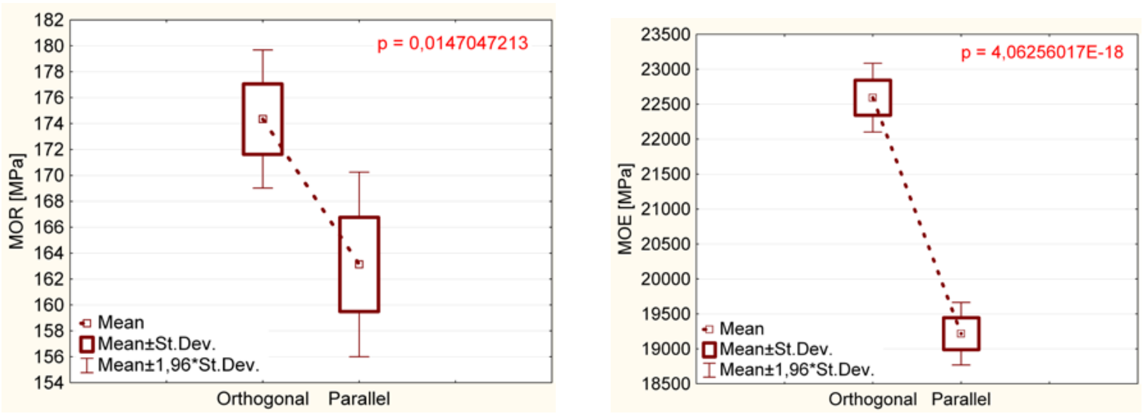
c

d

Fig. 2
Density profile (65% rH, 20°C):
a - profile with a higher density on the first side than on the second; b - profile with higher density on the outer sides than in the middle; c - profile with lower density on the outer sides than in the middle; d - profile with constant density in the whole cross section.

Modulus of rupture and Modulus of elasticity

The MOR measured orthogonal to compressing direction of samples was 174MPa and variation coefficient was 13%. The mean MOR measured parallel to compressing direction of samples was 163MPa with a variability of 19%. The higher MOE as well as MOR were calculated orthogonal to compressing direction and it was 22593MPa. MOE measured parallel to compressing direction of samples was 19217MPa. The variation coefficient of both sets of samples was nearly 10%. The differences between the MOR and MOE (orthogonal and parallel) (i.e. 11MPa, resp. 3376MPa) were assessed by the T-test for independent samples as statistically significant at a level of significance $\alpha = 0.05$ (Fig. 3a,b). These significant differences can be caused by the different longitudinal shear strength of samples during loading orthogonal and parallel to compressing direction. If the sample is loaded parallel to compressing direction, the longitudinal shear strength will be very affected by the uneven density profile (see Fig. 2). The differences in density through thickness of sample probably cause lower longitudinal shear strength followed lower MOR and MOE.



a b

Fig. 3
T-test orthogonal vs. parallel:
a - Modulus of rupture; b - Modulus of elasticity.

Fig. 4 is showing improvement of MOR and MOE. Also impact bending strength can be improved by ammonia treatment in combination with densification as showed Kalnins et al. (1967). The MOR increased by about 35% and MOE about 50%. The MOR and MOE improvement of Lignamon material can be seen in Fig. 4. Lignamon shows a significant improvement of strength properties. Modulus of rupture and modulus of elasticity are the highest in the orthogonal direction to the compressing. Also impact bending strength can be improved by ammonia treatment in combination with densification as showed Kalnins *et al*. (1967). The MOR increased by 35% and MOE by 45% comparing to solid beech. These results can be explained by the densification during compression of raw wood material. In addition, temporary character of ammonia plasticization effect is also confirmed. This effect is disappearing by the exhaustion of ammonia after compression in production. Increased MOR and MOE allow using a modified wood in applications where a natural wood failed.

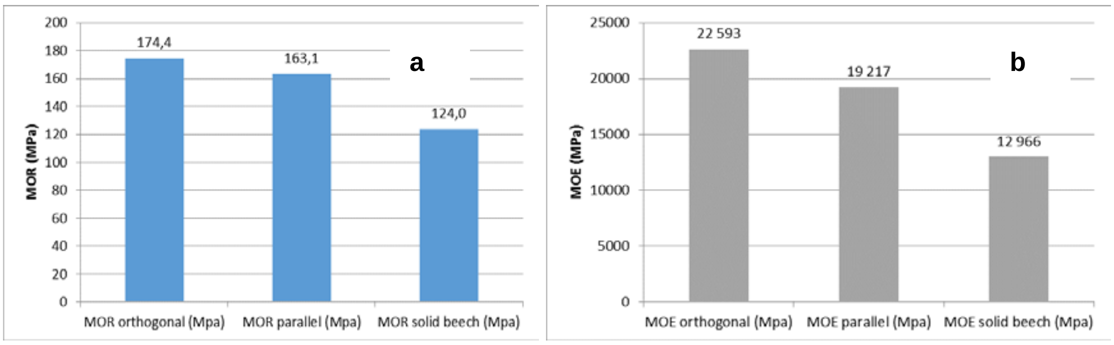


Fig. 4
Comparison of MOR (a) and MOE (b) to solid beech and direction of compression (Požgaj 1997) MOR (a) and MOE (b) of lignamon samples with average density of 1092kg·m^{-3} and MOR (a) and MOE (b) of solid beech samples with average density of 720kg·m^{-3}.

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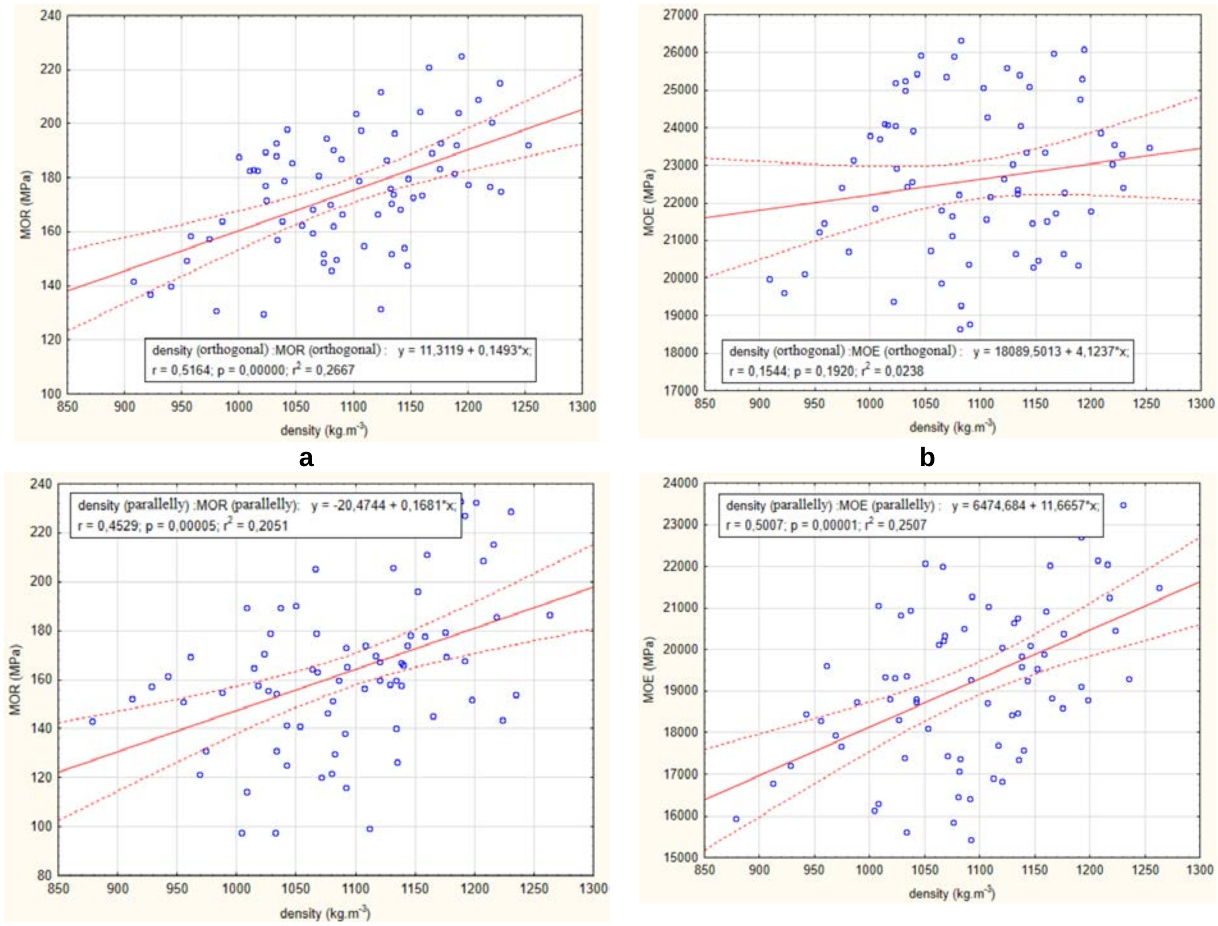
Influences of the density on bending strength are shown in Fig. 5. For evaluation of influence of density on the strength and stiffness linear regression analyse was used. In the case of MOR orthogonal and parallel to pressing direction of samples, the values of the correlation coefficient were around 0.5 (Figs. 5a,c). These correlation coefficients are on the threshold of high influence and show that increases of MOR are due to an increase of density. The use of the proposed regression model would make the estimation of the correlated quantity more precise as compared to arithmetic mean. Similar correlation coefficient as in case of MOR was determined in case of MOE parallel to pressing direction, i.e. approximately 0.5. Influence of density on the MOE (orthogonal to pressing direction) is not significant (Fig. 5b). This correlation coefficient was very low → 0.15. It can be potentially caused by structural abnormalities, which affect very much stiffness but not much strength (e.g. variable proportion of spring and summer wood in the annual ring). Results from linear regression analyse confirmed that density affects strength and stiffness of described modified wood similar like natural wood.

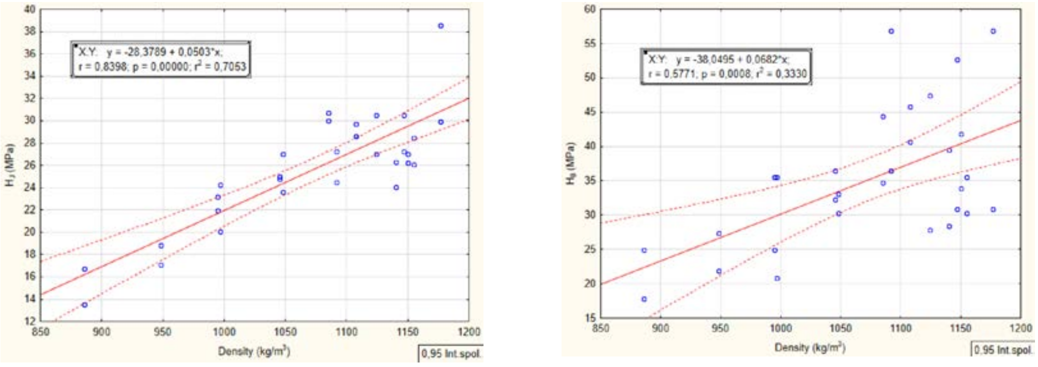


c d

Fig. 5
Regression analysis:
a,b - Influence of density on MOR and MOE in orthogonal to the compression direction; c,d - Influence of density on MOR and MOE in the compression direction.

Hardness and equilibrium moisture content

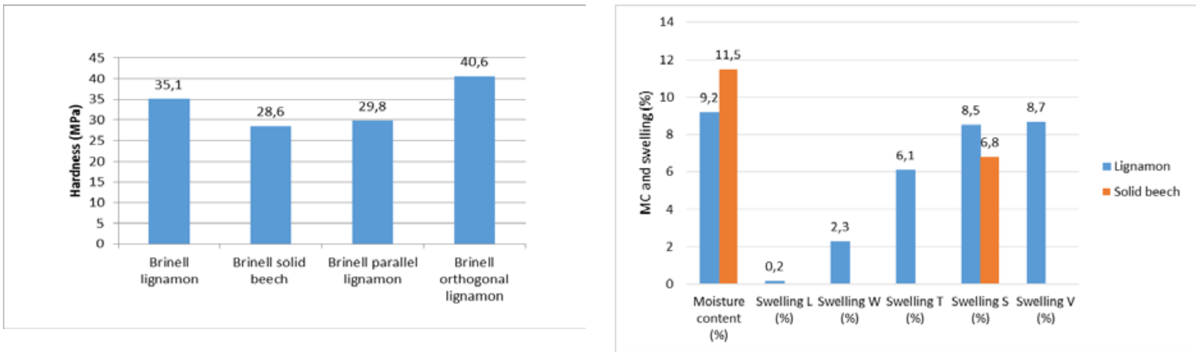
Influence of the density on hardness is shown in Fig. 6. Correlation coefficient in case of Janka harness is very high (0,84) and Brinell hardness has lower value, but it is still above the 0.5. Janka and Brinell hardness are significantly influenced by the density, with higher density hardness increases.



a b

Fig. 6
Influence of the density on Janka (a) and Brinell (b) hardness (65% rH, 20°C).

Hardness of lignamon is increased about 23% comparing to solid beech. There is no difference between hardness of solid beech and hardness of lignamon in compressing direction, but a big difference between hardness in parallel or orthogonal compress direction. The biggest hardness is in orthogonal compressing direction (about 41% comparing to solid beech). The values of moisture content and swelling after conditioning (65%, 20°C) are shown in Fig. 7b. The equilibrium moisture content of Lignamon is almost 3% lower than EMC of solid beech. Lignamon swells lower in orthogonal to the compressing direction (swelling W) than in the compressing direction (swelling T). Swelling in the longitudinal direction (swelling L) has properties similar to solid beech.



a b

Fig. 7
Hardness, equilibrium moisture content and swelling (Požgaj 1997):
a - Comparison of hardness to solid wood and influence of compression direction; b - Swelling and moisture content after conditioning (65% rH, 20°C).

CONCLUSIONS

Density investigation showed that Lignamon samples with lower average specific density have more uniform density profile and samples with higher average specific density have higher density zones on the edges. Samples with low specific density were probably cut from a middle section of densified wood. The differences in density profile probably result in low longitudinal shear strength followed lower MOR and MOE. Samples loaded parallel to compressing direction are very affected by the uneven density profile. Lignamon showed an improvement of strength properties (the highest values were in the orthogonal direction to the compresion). Hardness of Lignamon is enhanced primarily in orthogonal to the compressing direction. The moisture exclusion efficiency is also improved. Swelling of Lignamon is lower in orthogonal direction compared to swelling in the compression direction. The success of the method we suggest attributable to a mechanism involving no-fracture buckling of the cell walls and forming of new secondary bonds between the cellulose chains. The potential applications for Lignamon could be sailboats, exterior cladding, terrace decking etc.

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