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Abstract of the Ph.D. thesis

Investigation of the influence of steaming on selected mechanical and physical properties of two European and two tropical hardwood species

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1. Antecedent, importance and objectives of the research topic

Wood, contrary to other materials like gold, petroleum or natural gas, is a renewable raw material resource coming more and more to the front again recently. This tendency is not only due to its reproductable nature. Wood is one of the extraordinary materials that takes breath, gains or loses moisture even after building in, turning into part of a construction or furniture, and delights each and every people with no exception through the diversity of its appearance. However, there are fields of application requiring special wood characteristics, just as wood species with physical and mechanical properties that do not fit for a given application or will suit only after certain alterations. Technologies effecting this kind of changes are called modification.

Regarding research and development activities as well as secondary wood processing, modification of wood became a separate speciality in the last decades. Different goals require diverse technologies; some of them use chemicals or high temperature values, others apply light irradiation with different wavelengths.

During steaming as a modifying hydrothermal treatment, wood is exposed to heat through vaporized water. Structure and constituents of natural wood undergoes a transformation due to the heat transmitted by steam. As a result, some of the wood properties alter temporarily or permanently. In principal, temporary changes of properties promote an easier processing, while permanent changes ensure a more gentle timber.

As for industrial technologies, steaming has a tradition only in case of certain wood species (black locust, beech for instance). The number of scientific reports on laboratory experiments done in frame of research and development activities, and that of wood species involved increases constantly.

The aim of this scientific research is to investigate certain physical and mechanical properties of steamed (with different process parameters) and untreated test specimens of two European and two tropical wood species. In case of oak (*Quercus robur* L.) as a native European wood species, the goal of the steaming can be the improvement of the adhesion performance and the change of aesthetical appearance. Although hydrothermal treatment of black locust (*Robinia pseudoacacia*) – which is not a native genus in Europe but its productivity is remarkable – has a tradition in excess of 60 years, there are numerous surface characteristics that may have high significance from the viewpoint of processing (coating, gluing for instance) but have not been tested yet. The two tropical species, merbau (*Intsia bijuga*) and sapupira (*Angelim pedra*) are widely used even in Europe thanks to their prosperous mechanical properties and exceptional appearance. Certain technological problems, such as gluing and coating difficulties arising during processing of these two species, can be solved by steaming.

Based on the above, the following research aims were appointed:

I) Influence and significance of steaming parameters (time and temperature) on the colour of the investigated wood species will be examined. The most sensible species from the viewpoint of colour change will be detected.

II) UV light induced discolouration of steamed and untreated test specimens will be examined. Coated, as well as uncoated surfaces will be irradiated.

III) Assuming that mechanical properties of the investigated wood species deteriorate during steaming, connections will be detected between these changes and the steaming parameters (time and temperature).

IV) Gluing difficulties deriving from the highly reactive surface chemistry will be examined from two points of view: on the one hand surface free energy will be calculated from the measured contact angle values; on the other hand shear strength of glued joints will be determined. These measurements will be performed on untreated as well as steam treated timber.

2. Materials and methods

2.1. The applied technology

Before processing, raw materials were stored at normal climate (20°C, 65% relative humidity) for at least two weeks. Precise sizes were shaped with saw followed by planning. Test specimens used for the experiments were then assorted carefully based on the colour, fibre and annual ring structure. Since the volume of the load is restricted by the size of the cylindrical pressure chamber, extreme material properties had to be avoided in order to realize a low number of samples. After shaping, test specimens were stored at normal climate for another two weeks.

Parameters (temperature and time) of the steaming processes and the number of repeats were as follows:

a) For black locust, merbau and sapupira:

	3 hours	7.5 hours	20 hours
108°C	x		x
115°C		xxx	
122°C	x		x

b) For oak:

	3 hours	7.5 hours	20 hours
92°C	x		x
100°C		xxx	
108°C	x		x

Steaming temperatures of oak had to be reduced because of the weaker mechanical properties and frequent ray checks. The other three wood species are less sensitive therefore overpressure was applied in the whole temperature range.

Test specimens were put in a metal basket which was placed then in the chamber. Heating up of the system took two hours. The control equipment connected to a computer allowed of to keep the required temperature value in the range of ±0.5°C and to follow up the temperature changes during the entire process. Based on the graphs, the test specimens followed well the temperature changes of the environment (in the chamber) in all cases. The time values in the tables above show the steaming time without heating up and cooling down time values. After reaching the atmospheric pressure (under 100°C), the pressure chamber can be opened so the cooling down took 10-20 minutes. According to my observations, a longer cooling down period is unnecessary because the ray checks usually come up during the effective steaming process, not after that. Test specimens were stored at normal climate for one week after treatment.

2.2. Determination of the bending strength

The bending strength of samples was measured with 3-point loading parallel to the grain, according to the DIN 52186 standard. Test specimens with a 20x20 mm cross section were examined using Zwick Z020 testing machine with a support distance of 300 mm. Only the specimens with homogeneous, uniform fibre structure were tested. The annual ring angle was 25-45°.

2.3. Hardness measurement

During the hardness measurements according to Janka, half of a polished steel bullet with a diameter of 11.284 mm was pushed into the sample specimen. The force needed for pushing in gives the hardness directly. In the course of the tests, the side hardness of specimens with a cross section of 40x70 mm was measured along the two larger sides using Zwick Z020 testing machine.

2.4. Colour measurement

Objective measurement of the colour change caused by steaming was performed using Minolta CM 2600d spectrophotometer. Sample specimens with a cross section of 40x70 mm were cut in half parallel with their larger side after steam treatment. Fresh-cut surfaces were then planed. Lightness (L*) and hue (a* and b*) values were measured in the CIELab system. The sensor head was 10 mm in diameter. Measurements were made using a D65 illuminant and a 10° standard observer. This method is very fast since the spectrophotometer directly indicates the colour co-ordinates so there is no need of additional calculation. The colour difference (ΔE*), that is the distance of two colour point, is given by the spatial Pythagorean theorem. The colour change is demonstrated well by the a*(t), b*(t) and L*(t) diagrams.

In the course of my investigations, short-term irradiation of untreated as well as steamed wood samples was also performed using QUV accelerated weathering tester equipped with UVA-340 lamp. The irradiance ranges from 295 nm to 390 nm with a peak intensity of 0.7 W/m^2. Before the test, two sample specimens with a surface of 40x350 mm were shaped from each steaming series. Four different parts were composed on the surface of each specimen: uncoated, coated with transparent lacquer, treated with pigmented stain and coated with white opaque dye. After coating, specimens were conditioned at 23°C, 50% RH, the surface colour was measured and then irradiated with UV light. The surface colour was measured after 1, 2, 7 and 14 days of irradiation.

2.5. Determination of the contact angle and the surface free energy

Surface free energy of untreated and steam treated timber surfaces was calculated using their contact angle data measured on fresh-cut, planed surfaces with the sessile drop method. Components of surface free energy were calculated using the contact angle of diiodomethane, formamide, and water.

2.6. Shear strength of glued joints

Sample materials for the adhesion test were modified in one piece. After steaming, samples were shaped according to the EN 205 1998 standard (*Figure 1*). Two 5 mm thick panels were glued with ordinary PVAC adhesive, and then pressed. After hardening, a set of samples was stored under normal climate for one week and another one in cold water for 24 hours. Adhesion performance of untreated and steamed samples was then evaluated by calculating the shear strength of glued joints:

$$\tau = F/A,$$

where F indicates the shear force, and A indicates the glued area.

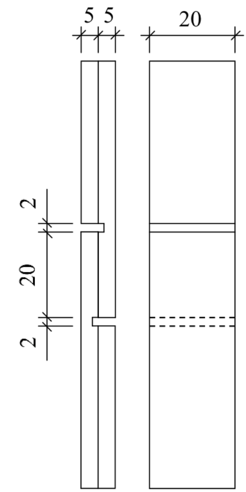

Fig. 1 Test specimens for shear strength measurements

3. *Summary of the new scientific results*

I) It was found, that black locust is the most sensitive among the investigated wood species as regard the colour change caused by steaming; this change can be described by the lightness (L*) and yellow hue (b*) co-ordinates. The colour achieved is determined by the two parameters of the steaming process jointly (temperature and time). Red (a*) and yellow (b*) hue of merbau is not affected by the steaming parameters; lightness is determined by the applied steaming temperature. Lightness and hue of sapupira and oak did not change significantly even after a longer treatment time at higher temperature.

II) It was shown, that the UV light induced discolouration of black locust timber can be reduced significantly by hydrothermal treatment; mean colour difference (ΔE*) value of untreated test specimens calculated from the colour co-ordinates measured before irradiation and after one week UV irradiation is 18.55; applying a pre-treatment at 108°C for 20 hours, this value is 10.33. Red hue shift caused by UV irradiation is nearly zero in case of long term (20 hours) steam pre-treatment at high temperature level (122°C). Colour stability is influenced in the same degree by the steaming temperature and the steaming time. The colour of heat-treated oak (108°C, 20 hours) is more stable because of the higher stability of the yellow hue after steaming. Steam treatment of oak followed by short term UV irradiation might be an interesting combined technology in order to emphasize the structure of the wood surface. Colour shift of steam treated merbau and sapupira samples was similar to that of untreated specimens.

Discolouration of wood samples treated with the investigated coating materials was moderate during UV irradiation. Colour stability of black locust coated with transparent lacquer can be improved by steaming. In case of merbau and sapupira, discolouration of coated test specimens was the same regardless of steam pre-treatment. Therefore, heat treatment cannot be a solution for the problem of colour stability. Discolouration can be reduced to a minimum applying pigmented coating; similar colour differences (ΔE*) of different wood species prove the discolouration of the coating.

III) During the research work, mean moisture content of each series of specimens was also determined as a percentage of the sample's dry mass. There were significant differences found however between the net moisture content of untreated samples and that of samples intended to be steamed. This discrepancy can be explained by the differences of the dry mass values, since the steamed samples were dried after the treatment to mass stability. This difference indicates the dry matter decrease caused by the steaming. Based on this value, the amount of water-soluble extractives outgoing during steaming can be calculated assuming identical starting equilibrium moisture content. The differential value is influenced by the steaming parameters jointly: longer treatment time and/or higher temperature results in getting loose of more extraneous materials. The differential value depends also on wood species: large differences were found among the moisture content values of merbau. The lowest value was measured in case of sapupira.

IV) It was demonstrated that the investigated mechanical properties of the four wood species deteriorated under heat treatment. Bending strength decrease of the tropical species was more gentle compared to that of black locust and oak. The most dramatic changes were observed in case of black locust but, thanks to the very high original values, its bending strength is still higher than that of untreated oak even after a long-term (20 hours) treatment at higher temperature level (122°C). It was found that steaming temperature is not significant from the viewpoint of bending strength of the investigated wood species. In the investigated range of parameters, bending strength is influenced by treatment time rather than temperature. The explanation is that the temperature under 200°C is not sufficient condition in itself for the degradation of the main substances of wood, namely the lignin and cellulose. Through a longer treatment time however the structure of these chemical compounds will be altered.

V) My experiments showed that the contact angle between wood and water depends on wood species and changes during steaming. Contact angle between black locust wood and water diminishes during a 3 hours long treatment at 108°C. Longer heat treatment and/or higher steaming temperature resulted in higher contact angle value. Contact angle between sapupira wood and water did not change significantly during steaming. The relatively high contact angle of oak can be decreased by steam treatment. No connection was found between the evolved angel value and the treatment parameters. Contact angle of merbau surface increased during steaming.

Surface free energy is influenced by both of the process parameters independently of wood species. Surface free energy of merbau decreases to the half of the original value during a 20 hours long heat treatment at 122°C. Surface free energy of oak can be increased with short term (3 hours) modification at lower temperature (108°C) while the surface energy of sapupira rises with a long term (20 hours) treatment at high temperature (122°C).

VI) Adhesion tests performed using PVAC adhesive demonstrated that adhesion performance of black locust and sapupira wood can be improved by steaming with proper process parameters; contact angle between black locust wood and water decreased during short term (3 hours), low temperature (108°C) steaming (statement V), therefore the strength of the joint made of water soluble PVAC adhesive increased. In case of sapupira, a 7.5 hours long steam treatment can be proposed at medium temperature (115°C). Adhesion performance of merbau did not change significantly, while that of oak deteriorated at any applied set of parameters.

4. Publications related to the topic of the Ph.D. thesis

English articles published in foreign countries:

László Tolvaj, Dénes Varga & Szabolcs Komán (2002): Colour modification of dried black locust and beech woods by steaming. Wood Structure and Properties ’02 109-113

Varga, D., van der Zee, M. E. (2008): Influence of steaming on selected wood properties of four hardwood species. Holz als Roh- und Werkstoff (will be publish in print in issue 66/1 February 2008)

Articles in Hungarian:

Tolvaj L., Varga D., Molnár S., Pál A. (2001): A gőzölés színváltoztató hatása fehér és színes gesztű bükk faanyag esetében. Faipar 2001/4 11-12

Dr. Tolvaj L., Varga D. (2002): Az akácgőzölés színváltoztató hatása. Intarzia 2002/6 19-22

Horváth-Szováti E., Varga D. (2000): Az akác faanyag gőzölése során bekövetkező színváltozás vizsgálata II. A 105, 100 és 115°C-on történő gőzölés eredményei, javaslat az ipari hasznosításra. Faipar (4) 11-13

Tolvaj L., Molnár S., Takáts P., Varga D. (2004): Az akác (*Robinia pseudoacacia* L.) faanyag színének változása a gőzölési idő és hőmérséklet függvényében. Faipar 2004/4 9-14

Tolvaj L., Molnár S., Takáts P., Varga D. (2005): Az akác faanyag színének homogenizálása gőzöléssel. Faipar 2005/1 13-15

Conference appearances:

Tolvaj L., Varga D. (2001): Az akác faanyag színének változtatása gőzöléssel. Konferencia, Székelyudvarhely, Románia, 2001. május 18.

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Varga D., Takáts P., Tolvaj L. (2003): Új lehetőségek az akác faanyag hidrotermikus kezelésénél. 5. Magyar Szárítási Szimpózium, Szeged 2003. október 21-22.

Varga D., van der Zee, M. E., Nienhouis, J., Tolvaj L. (2006): Influence of steaming on selected wood properties of four hardwood species. 56th Annual Meeting of the Japan Wood Research Society, August 8-10, 2006, Akita JAPAN

Varga D., van der Zee, M. E. (2006): Influence of Hydrothermal Treatment on UV-light Induced Discolouration of *Robinia pseudoacacia* L. and *Quercus robur* L. Timber Surfaces. In the Proceedings of the JSPS Japan and Hungary Research Cooperative Program / Joint Seminar, October 16-19, 2006, Noshiro JAPAN

Varga D., van der Zee, M. E. (2007): Influence of hydrothermal treatment on UV-light induced discolouration of *Robinia pseudoacacia* L. and *Quercus robur* L. timber surfaces. In the Proceedings of the 3rd European Hardwood Conference, Sopron, September 3, 2007.