Doctoral Thesis

THE EFFECT OF OIL-HEAT-TREATMENT ON SOME MAJOR PROPERTIES OF POPLAR WOOD

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1. Introduction and objectives

The different heat-treatment processes give the opportunity to improve the applicability of wood materials, which due to their lower biological resitance and/or weather resistance and/or dimension stability have a limited application field. In Hungary these are mainly plantation grown species, for instance black locust (*Robinia pseudoacacia* L.) and different poplar species (*Populus* × *euramericana*). Particularly poplar species deserve attention, because there is a large quantity available of them. However its application is limited to some area (mainly fruit boxes and pallets, plywood, fiber- and particle board). By improving of the properties its application could be widened by substituting more valuable wood species. For that reason I have chosen as topic of my thesis the heat treatment of Pannonia poplar (*Populus* × *euramericana cv.* Pannonia) in vegetable oils, which is the most widespread Hungarian poplar species. Investigation of heat treatment in different vegetable oils is stated with the fact that in wood occur various chemical transformations during the procedure. Vegetable oils have partially different chemical content, which may have some influence on the changes of the wood properties.

In the literature are detailed studies about the effects of different heat treatment methods on wood. However investigations about oil-heat-treatment are remarkable limited for all its favourable properties. Considering the fact that a process is already commercialised in Germany (industrial scale process) it is especially interesting. The limited information available in literature and the favourable properties discussed later stated to choose this method as a topic of my thesis. One of the advantages of the method is its high intensity, thus it is a fast treatment. Other, in the literature mentioned and even industrially used methods are using long warm up and cooling periods, whereas by oil-heat-treatment wood is directly placed into the hot oil bath and after the treatment it is placed on room temperature without a special cooling period in the chamber. According to literature the long warm up and cooling periods are used to avoid cracks and deformations. By poplar wood the application of these cycles is not necessary, because of its relatively homogenous texture with open pores. According to my experiences, no damages occur by the oil-heat-treatment of poplar wood. Owing to the reasons mentioned above the treatment time can be significantly shortened. Further advantage of the method is that in case of continuous processing the heating medium and its heat is recyclable. By other methods the heating medium and its heat gets lost, or needs special heat recovery system at the end of the cycles.

In this thesis selected wood properties were investigated which are important by the specification of the application fields of the treated material. Certainly, it is important to uncover the effects of the treatment on the mechanical properties and the description of the changes in colour properties. After gaining this knowledge, basics of the technical and aesthetical utilizations are given. Because of the hygroscopic properties of wood material, investigation of wood-water relationships is necessary. One of the main goals of heat treatment methods is to improve the decay resistance of wood, thus durability aspects are investigated as well. Because of the character of the treated wood primarily are outdoor applications are considered, therefore weathering investigations are needed. The thesis provides a comprehensive view about the most important properties of the oil-heat-treatment of poplar wood. Its necessity is justified, because heat treatment changes the chemical composition of the wood, which results in a new material. Investigations about oil-heat-treatment – especially for poplar wood – are quite rare. Therefore, results of this study can be useful by facilitate the use of oil-heat-treated poplar wood. In my thesis specific utilization examples or product developments are not discussed, however the presented results make this possible.

2. Materials and methods

To the investigations Pannonia poplar wood (*Populus × euramericana cv*. Pannonia) from the wood-mill of the Kisalföldi Forestry plc in Kapuvár was used. Heat treated samples were cut from 50 mm thick unedged boards. The boards were air dried on normal climate to ca. 10±2% moisture content (oven dry based). The boards were then cut into smaller laths with the dimension of (18mm×40mm×220mm), which was determined by the size of the equipment used for the hot oil treatment. After the treatment the laths were cut into smaller specimen, according to the further investigations. For each schedule 20 laths were treated, and the laths were taken randomly from more boards, thus the results include the variability of properties between logs as well. The samples were treated in 3 different vegetable oils. Different oils were used in order to clarify whether the oil type used has any effect on the results of the heat treatment. Therefore three different type of oils were chosen. Linseed oil is a drying oil (linolen acid group), sunflower oil is semi-drying oil (linol acid group) and rapeseed oil is a not- or slightly drying oil (eruka acid group). All of these oils are easily obtainable and commonly used. Linseed oil is used for wood coating long ago, sunflower oil is used mainly for eating and rapeseed oil is a well known raw material for biodiesel production.

According to the literature and own experiments, the wood samples were treated at 2 different temperatures. 160°C were chosen, because under this temperature heat treatment cannot change the chemical composition and structure of wood effectively, therefore this is considered as a lower limit of heat treatments. 200°C is generally noted as the lower limit for improve the durability, nevertheless mechanical properties could decrease above that temperature. As Poplar wood has limited strength properties, therefore treatments above 200°C cannot be recommended. Three different durations were used (2h, 4h and 6h), thus as a variation of these parameters considering the three different oils resulted in 18 different schedules or treatment methods, which were then applied. (Table 1.).

Table 1. Applied heat treatment schedules

Oil	Sunflower						Linseed						Rapeseed					
T [°C]	160			200			160			200			160			200		
Time [h]	2	4	6	2	4	6	2	4	6	2	4	6	2	4	6	2	4	6

The laths were put directly into the hot oil bath, and after the treatment time (2h, 4h and 6h), the specimen were taken out from the bath and placed to normal climate (T=20°C; φ=65%). To the static mechanical properties (compressive-, tensile- and bending strength) universal Instron 4208 testing machine was used. To the dynamic property (impact bending) the Charpy's pendulum (100 Joule) was used. The colour coordinates were measured in CIELab system with a Konica Minolta CM-2600d device.

Changes in density and in wood-water relationships (swelling, equilibrium moisture content, moisture uptake properties) due to heat treatment were investigated. Characterisation of the colour change due to heat treatment and the colour change of heat treated material during weathering were carried out according to the CIELab system. Also the effect of heat treatment on decay resistance against the white rot fungus *Pleurotus ostreatus* and the brown rot fungus *Poria placenta* were investigated.

Investigations were made according to the relevant standards if a standard was available for the method used.

3. Conclusions

Every treatment parameter has an effect on the complex changes in wood during heat treatment, thus investigation of wood properties is always necessary. In case of oil-heat-treatment the exclusion (minimising) of oxygen in air is solved by the characteristic of the treatment medium.

Mechanical properties of wood are strongly dependent on the quality and quantity of the main cell wall components, furthermore on the structure in nano-, micro- and macro scale. However, the woods native composition and structure is changing due to the high temperatures. Elevated temperatures usually results in mass loss, which is an indication for thermal degradation. In spite of that, mechanical properties are increasing in some cases. For example compressive strength increased after all treatments and also bending strength increased after 160°C treatment. However, tensile- and impact bending strength decreased significantly.

Weight percentage gain (WPG) of wood (uptake of treating medium) is mostly an important factor by estimation of the effectiveness of modification treatments. By oil-heat-treatment the determination of WPG was not calculated for more reasons. The main reason is, that oil uptake cannot be determined exactly, because it takes place simultaneously with the thermal degradation of wood. However, results show that poplar wood has large oil uptake in the case of 160°C treatments while in the case of 200°C treatments oil uptake is much smaller. Due to the differences in oil uptake the density of wood treated at the lower temperature increased considerably, while the density of wood treated at higher temperature remained almost unchanged.

It is well known long ago that hydroxyl groups on the surface of the cell wall components are responsible for the bounding of water. Oil-heat-treatment decreased moisture uptake significantly which was proved at all investigated climates as equilibrium moisture content was decreased (desorption). As a consequence of that, moisture induced swelling decreased as well in both radial and tangential directions, which resulted in improved dimensional stability. Swelling decreased in tangential direction more pronounced compared to radial. This result shows that although the swelling anisotropy decreases, it will not fully disappear. However, maximal swelling of heat treated wood was smaller, the percentile swelling caused by 1% change in moisture content (swelling coefficient) did not after due to heat treatment.

During the service life of a product the surrounding climate is regularly changing and the changes are often fluctuating in. All of the investigated heat treatment schedules decreased the moisture uptake rate and equilibrium moisture content. According to that, heat-treated samples absorb less moisture during the same time than the untreated ones. However, decrease in moisture uptake rate was only apparent, because equilibrium moisture content was reached by all samples in the same time. Accordingly, saturation rate of wood remained unchanged and the apparent decrease in moisture uptake rate is therefore due to the reduction in the amount of sites (functional OH-groups), which are able to bound water molecules. Namely, the water bounding capacity decreases, not the water bounding capability.

Modification of the main cell wall components has considerable effect on the colour properties as well. Heat treatment of wood material resulted in significant colour change, which was mainly darkening of the colour. The red hue of the poplar wood – which has in natural condition bright yellowish colour – increased significantly; the same was observed for the yellow hue. Accordingly, also colour saturation increased, primarily at the lower treatment temperature. Parallel to that the originally inconspicuous texture of poplar wood appeared much more pronounced, which enhanced the aesthetical value. Heat treated poplar wood is well separable on the basis of the colour from the natural wood, because the total colour change was considerable by all the investigated schedules.

One of the important goals of heat treatment is to make wood applicable for outdoor using, but beside of that in many instances colour has also great importance. Poplar wood has in natural condition homogeneous appearance and application possibilities are influenced on whether the improved aesthetical value achieved by heat treatment can be maintained. During the 3 years of weathering all of the investigated wood material became remarkably darker. The red- and yellow hue of natural and with different schedules heat treated poplar wood decreased as well, which is equal to the well known greying effect of wood under outdoor conditions. Accordingly, colour saturation decreased as well. The different heat treatments resulted in different total colour changes. Summing up, colour of the heat treated materials is not more durable than that of the untreated material, because colour change occurred in the same time interval. Natural and heat treated material were turning grey as well, consequently heat treatment do not have any effect on colour stability of poplar wood under outdoor conditions. However colour stability remained unchanged heat treatment at 200°C resulted in smaller total colour change compared to untreated wood, which is favourable in the course of outdoor application. Whereas heat treatment at 160°C is clearly disadvantageous in this respect, because the total colour change of poplar wood heat treated at 160°C is superior to that of the natural wood.

Decay resistance is usually a problem by application of Poplar wood, because it is one of the least durable wood species. Durability of the poplar wood was different against the investigated white- and brown rot fungi. However, heat treatment at 160°C was ineffective against both of white- and brown rot, as well as treatments at 200°C with 2 hours duration. Only the two most intensive treatments (200°C, 4 and 6 hours) resulted in significant, 30-35% improvement (mass loss reduction) of durability against both of the investigated fungi.

No differences could be proven between the investigated oils regarding the investigated properties. Accordingly, consideration of other factors is needed to select the oil for heat treatment. Most important factors are current price and purchasing possibilities. During processing of heat treated wood and utilization of products made of heat treated wood is the presence of liquid oil unfavourable, therefore drying ability of the oil used is an important factor. Accordingly, linseed oil should be preferred to the other oils, because it is a drying oil, while sunflower oil is semi-drying oil and rapeseed oil is a not- or slightly drying oil. If heat treated wood is used in living space or its toward, also smell can be an important factor, which could limit the use of linseed oil, because of its penetrating smell. Under outdoor conditions the smell due to heat treatment fades in short time.

4. Theses of the dissertation

1. It is supported by the investigations that in spite of degradation of the cell wall components some mechanical properties increased in case of Pannonia poplar after heat treatment. Compression strength parallel to grain increased by 15-25% as a result of heat treatment, depending on the schedule used. At 160°C also bending strength increased by 5-10% as a result of heat treatment however it decreased if treated at 200°C by 5-20%. Tensile strength parallel to grain and impact-bending strength decreased by 15-55% and by 10-55% respectively depending on the schedule used. Explicit increase in compressive strength is explained by the heat stability of lignin. During treatment lignin remains almost unchanged, thus the positive effect of the lower equilibrium moisture content is more pronounced than the negative effect of the thermal degradation. The other three investigated mechanical properties are primarily depending on the structural integrity of the hemicelluloses and cellulose. As this components are partly degraded, the the positive effect of the lower equilibrium moisture content cannot compensate the strength loss.

2. It was experimentally proved that heat treatment in vegetable oils decreases the equilibrium moisture content of Pannonia poplar at all climate condition. Heat treatments in vegetable oil at 160°C and 200°C decrease equilibrium moisture content by 40-50% and 50-60% respectively, irrespective of the climate conditions. Thus equilibrium moisture content of heat treated Pannonia poplar at normal climate ($T=20°C$; $\varphi=65\%$) decreased between 4-6% depending on the treatment method used.

3. It was revealed that heat treatment in vegetable oils resulted in significant decrease in swelling of Pannonia poplar. Depending on the heat treatment schedule used tangential and radial swelling decreased 30-45% and 20-40% respectively. Results verified that although the swelling anisotropy was slightly reduced, it did not disappear. Heat treatment in vegetable oils did not have any effect on the swelling coefficient of Pannonia poplar.

4. It has been found that heat treatment in vegetable oils decreases the moisture uptake rate of Pannonia poplar only apparently. To corroborate that equilibrium moisture content (EMC)-ratio was introduced in my work, which shows the ratio of the momentary moisture content and the EMC of wood at constant climate and starting from absolute dry state.

   Definition of EMC-ratio is with the following equation possible:

$$S_{ENT} = \frac{U_T}{U_{ENT}} \cdot 100[\%]$$

   where: S_{ENT}: EMC-ratio [%]
   U_T: momentary moisture content [%]
   U_{ENT}: EMC at normal climate (T=20°C; φ=65%) [%]

   It was proved by investigations that EMC-ratio of natural and in vegetable oils heat treated Pannonia poplar is throughout equal, namely EMC is reached in the same time, and accordingly relative moisture uptake rate is equal. Therefore, only the water bounding capacity of wood decreases, not the water bounding capability. This result corroborates the supposition that decrease in moisture uptake is due to the reduction in the amount of sites (functional groups), which are able to bound water molecules.

5. It was proved that heat treatment in vegetable oils decreased the lightness (ΔL*=15-45) of Pannonia poplar remarkably, dependent on the treatment parameters. Furthermore, red hue (Δa*=5-10) and yellow hue (Δb*=4-15) was increased. With increasing treatment time and duration colour saturation increased inversely proportional (ΔC*= 5-18). Whereas total colour change increased consistently (ΔE*=20-45) with increasing treatment time and duration. Extractive content of poplar wood in natural condition is very low, thus colour change is mainly owing to the degradation of hemicelluloses as well as the arising degradation products.

6. The results available in the literature concerning the colour change in outdoor by the dry heat treated woods could be verified for the oil heat treatment as well. The colour change of wood occurs under outdoor conditions mainly during the first 12 months. The surface became grey and dark as weathering tests for by oil-heat-treated Pannonia poplar showed. Results proved that the colour stability of oil-heat-treated poplar wood is not better compared to that of the untreated wood, because colour change, which means the greying of the surface occur in the same amount of time. It was shown that heat treatment carried out at 200°C resulted in lower total colour change (ΔE^*=21-42) of Pannonia poplar compared to the untreated wood (ΔE^*=45) after 3 years weathering. However, 160°C heat treatment resulted in higher total colour change (ΔE^*=47-60).

7. Laboratory tests showed that heat treatment of Pannonia poplar at 160°C did not reduced the decay (mass loss) of the white rot fungus *Pleurotus ostreatus* either the brown rot fungus *Poria placenta* during 16 weeks incubation time. Heat treatment at 200°C only improved the decay resistance (by 30-35%), if treatment duration was at least 4 hours long.

5. Publications in the research field

Publications in edited scientific journals written in foreign language

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9. Bak, M.; Németh, R. (2012) Modification of wood by Oil Heat Treatment. *In: Neményi, M.; Heil, B.; Kovács, J.A.; Facskó, F. (szerk.) International Scientific Conference on Sustainable Development & Ecological Footprint, The Impact of Urbanization, Industrial and Agricultural Technologies on the Natural Environment. Sopron, 2012.03.26-27.* Paper 11., pp. 1-5., (ISBN:978-963-334-047-9)

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10. Bak, M.; Németh, R.; Tolvaj, L; Molnár, S. (2008) Ültetvényes termesztésből származó fafajok anyagának hőkezelése növényi olajban. *Faipar,* **56**(különszám), pp. 22-26.

11. Németh, R.; Ábrahám, J.; Bak, M.; Molnár, S. (2011) Faanyagok modifikálása az NymE Faipari Mérnöki Karán. *Magyar Asztalos és Faipar,* **20**(1) pp. 64-65.

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2. Németh, R.; Tolvaj, L.; Molnár, S.; Horváth, N.; Ábrahám, J.; Bak, M. (2008): Új eljárások az ültetvényes faanyag minőségének javítására, fanemesítés. *In: Az ültetvényes fagazdálkodás fejlesztése. Ed: Molnár, S.; Führer, E.; Tóth, B.; NyME,* ISBN 978-963-9883-21-5, pp. 61-66.

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