On September 24-26, 2014, the Forest Products Society European section (FPS Europe) partook as a professional association partner in a successfully carried out third edition of the **International Conference on Processing Technologies for the Forest and Biobased Products Industries**.

This high-level meeting was hosted by the organizing institution, Salzburg University of Applied Sciences, Campus Kuchl, in partnership with other scientific and supporting professional associations, including COST FP1006, University of Tennessee (Knoxville), University for Natural Sciences (Tulln), "Transilvania" University of Brasov , and IUFRO.

149 participants from 31 countries and 5 continents, including many FPS members, attended the event, which incorporated 79 oral presentations within 2 days in 11 parallel session and 23 posters.

The Society applauds the newest local section's significant constirbutions to this important event, and the Board is inspired by their efforts to further advance the Society's vision of being the international leader for advancing the sustainable use of renewable cellulosic resources through science and technology.

Proceedings from the International Conference on Processing Technologies for the Forest and Biobased Products Industries

ELECTRON BEAM IRRADIATION OF WOOD: AN EXPERIMENTAL PARAMETER STUDY

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Key words: artificial weathering, color measurement, fir, larch, Norway spruce, wood modification

ABSTRACT

Surface modification by electron beam irradiation is one of the most advanced industrial and scientific techniques for improving surface properties of a polymer. This modification process may beneficially influence various functional and structural properties of wood.

In this study, different wood species and process parameters (e.g. irradiation dosage) were used. The changes physical properties (e.g. discoloration and weathering behavior) were analyzed.

The results of colour measurement show different effects of the various wood species and irradiation dosages during an artificial weathering test. For specific applications, this wood surface modification process can contribute to improving the functional and structural properties of wood. These results provide a helpful basis for further fundamental research.

1 INTRODUCTION

The modification of wood and wood surface properties has been a long tradition and has focused on ion irradiations (e.g. plasma treatment) to develop new modification processes (Hill 2006, Militz 2012, Petrič 2013). Surface modification is one of the most advanced scientific techniques that may lead to the goal of very precise applied surfaces and can enhance polymer properties on the surface. Polymer modification of materials can be described here creating beneficial changes of the properties, whereas two interesting technologies can be used. On the one hand a reactive, excited-state or partly ionizing gas (e.g. plasma treatment) alters the surfaces (Ellinghorst 2005, Sheton and Stevens 2001, Wolkenhauer et al. 2008). On the other hand electron beam irradiation (ionizing radiation) can be applied to modify polymers (Jarović 1990). Also, the electron beam irradiation process may beneficially influence various functional and structural properties of wood.

Wood is susceptible to degradation by UV-light and in exterior application undergoes various changes in properties during its life time. The ageing mechanisms are very complex and are influenced by many factors (e.g. rain, solar radiation and temperature) (Evans et al. 1992, Feist and Hon 1983, Hon 2001). These studies show that chemical bonds break and the lignin content of wood decreases as a consequence of photo-degradation. New chromophores form as exposure time is increased (George et al. 2005, Müller et al. 2003, Pandey 2005, Tolvaj and Faix 1995). Furthermore, the surface color of wood varies rapidly when it is exposed to light (Hon and Minemura 2001). If the surface is wetted by water (e.g. rain) then the degradation products from wood components (e.g. lignin) can be leached out (Evans et al. 1992, Kishino and Nakano 2004).

To improve the resistance to weathering ion irradiation is one possible method to reach this objective; however, a systematic study of the effects of high energy electron beam radiation on wood surfaces to influence the weathering behaviour is not available.

Fengel and Wegener (2003) give a literature overview of the effects on wood materials of ionizing rays (gamma-rays) at high dose levels resulting in changes of the properties of wood in structural and chemical behaviour. High energy irradiation of cellulose causes depolymerisation, a reduction in crystallinity and extensive decomposition with increasing dose levels (Charlesby 1955, Saeman et al. 1952). Fischer et al. (1985) described a pre-processing of pulp with ionizing radiation for the production of regenerated cellulose. In dependence on the dose level (10 to 100 kGy) the degree of polymerization changes to lower values, simultaneously the number of functional groups increases in the cellulose. Also the side chains of xylan (hemicellulose) are subject to cleavages resulting from oxidative reactions (Seifert 1964). Irradiation of lignin (MWL) does not change the aromatic hydroxyl groups and conjugated carbonyls; however changes take place on the aliphatic hydroxyl groups and guajacyl residues, which were decreased (Fischer and Goldberg 1987). They have assumed that condensation reactions between the aromatic nucleus and the side chain occur during the irradiation.

This proposed condensation reaction may improve the stability of wood against weathering effects. Therefore, the objectives of the current study are the analysis of the effect of electron irradiation doses on weathering behaviour of larch, fir and Norway spruce wood samples.

2 MATERIAL AND METHODS

2.1 Wood species

In this study, three samples from fir, larch and Norway spruce wood were cut to dimension of 150 x 75 x 20 mm³ according to EN 927-6 (2006). The untreated and treated samples were stored in a climatic chamber (20°C and 65 % relative humidity) until the weathering test.

2.2 Electron beam irradiation (EBI)

For the E-beam treatment at doses of 25, 50, 100 and 200 kGy an Electrocurtain® LAB Unit CB 175/15/10L electron accelerator was used with radiation energy of 150 keV. Five spruce samples were irradiated under nitrogen atmosphere for each EBI doses. Higher electron beam doses than 50 kGy were reached by repeating the process with 50 kGy. Moreover, three larch and three fir samples were only irradiated with 50 kGy.

2.3 Artificial weathering

The artificial weathering procedure was applied according to EN 927-6 (2006) with test cycles of water condensation (40 °C temperature), light irradiation with UVA-340 bulbs (0.89 W/m²/nm and 60 °C temperature) and water spray (6-7 litres/min) with an exposure period of 950 hours. For the deter-

mination of color and chemical changes before and after the weathering test the samples were always collected after a 2 hours light irradiation cycle and cooled down in an ambient climate room (temperature 20 °C and relative humidity 65 %).

2.4 Colour Measurement

Wood colour was measured with a Mercury 2000 spectrophotometer (Datacolor) and the selected diameter for measurement was 11 mm. Colour is expressed according to the Commission International de l'Eclairage (CIE) L*a*b* colour space (abbreviation CIELAB) with a standard illuminant D_{65} and a 10° standard observer. The L* parameter represents lightness where the values L vary from 0 (black) to 100 (white). The a* and b* parameters describe the chromatic coordinates on green-red (a*) and blue-yellow (b*) axes. The ΔL*, Δa* and Δb* values of the colour differences from measurements were taken before, during and after artificial weathering. In this study, the positive values of Δa* and Δb* indicate an increased in intensity of red and yellow colour tone, while the negative values of Δa* and Δb* reflect a desaturation of red and yellow colour tone. The mean of three measurements per sample was used for colour analysis.

3 RESULTS AND DISCUSSIONS

The results of the color measurements before and after 24 hours of the electron beam irradiations of all used wood samples did not show a statistical significant difference. Therefore, it could be assumed that the irradiated has not visible effect of the wood color.

However, it is a fact that the wood color changes during the exterior applications, while varying effects of weather show different consequences on the wood surfaces (Anderson et al. 1991). Therefore, it is not surprising that the tendency of change in colour of the wood samples used in this study was similar to other investigations.

The ΔL*, Δa* and Δb* values show differences between the unirradiated and electron beam irradiated Norway spruce samples (Figure 1). In particular the ΔL* and Δb* values were varied after the first 320 and 530 hours. All irradiated samples show lower changes in lightness (ΔL*), however, higher shifts in yellow color tone (Δb*). After 740 hours the influence of the electron beam irradiations became less important for the Norway spruce samples.

With regard to all three parameters L*, a* and b*, the color of the different sample groups became uniform to the end of the weathering test at 950 hours.

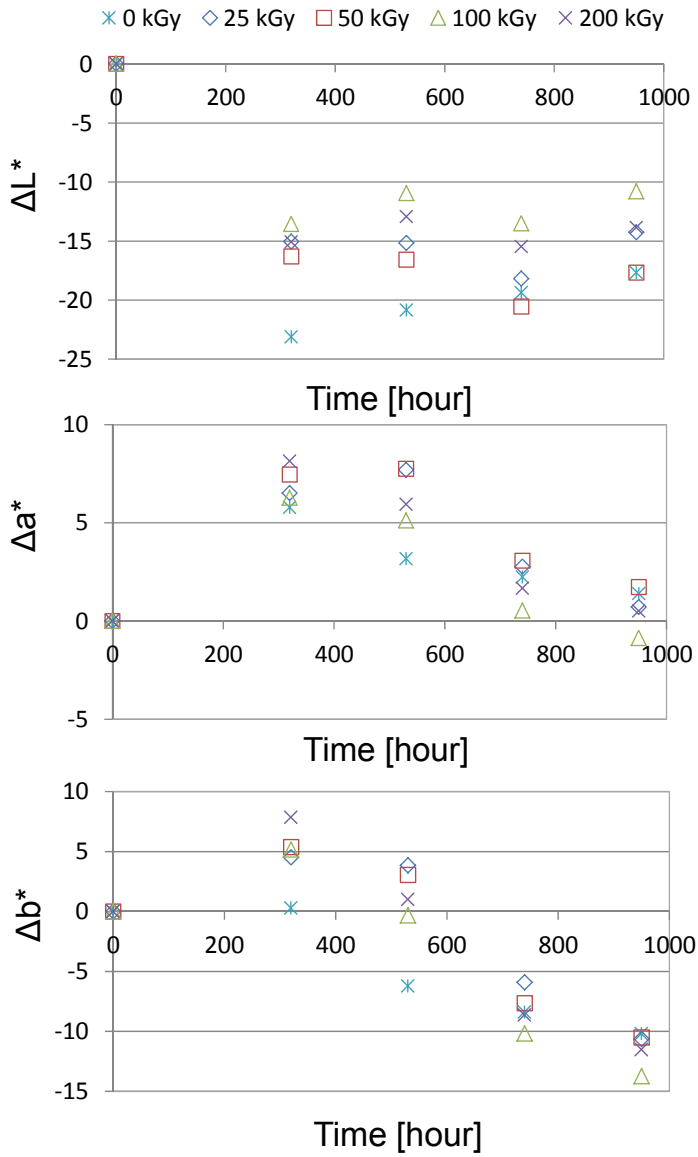

Figure 1. Estimated mean of ΔL*, Δa* and Δb* values of Norway spruce sample with various EBI treatments of various exposure time

The ΔL*, Δa* and Δb* values of the unirradiated and irradiated fir samples changes in equal tendency, however, the fir samples after the electron beam irradiation with 50 kGy show lower discoloration after the first 320 hours of the weathering test (Figure 2). Then the ΔL* values of the irradiated samples changed without a trend, the values were stable between -11 and – 8. Whereas the ΔL* of the untreated fir samples decreased until to the 740 exposure hours. This behavior of a very slight increase up to the end of the artificial weathering test was also found by Temiz et al. (2005). They concluded that this phenomena may generated by removing and loosening fibers.

The Δa* and Δb* values of the EBI treated samples showed a faster decrease in intensity of the color. The red and yellow color tone faded. This behaviour is consistent with the investigation of discoloration of wood surfaces due to natural and artificial weathering test by Hon (2001), Kishino and Nakano (2004) and Schnabel et al. (2009).

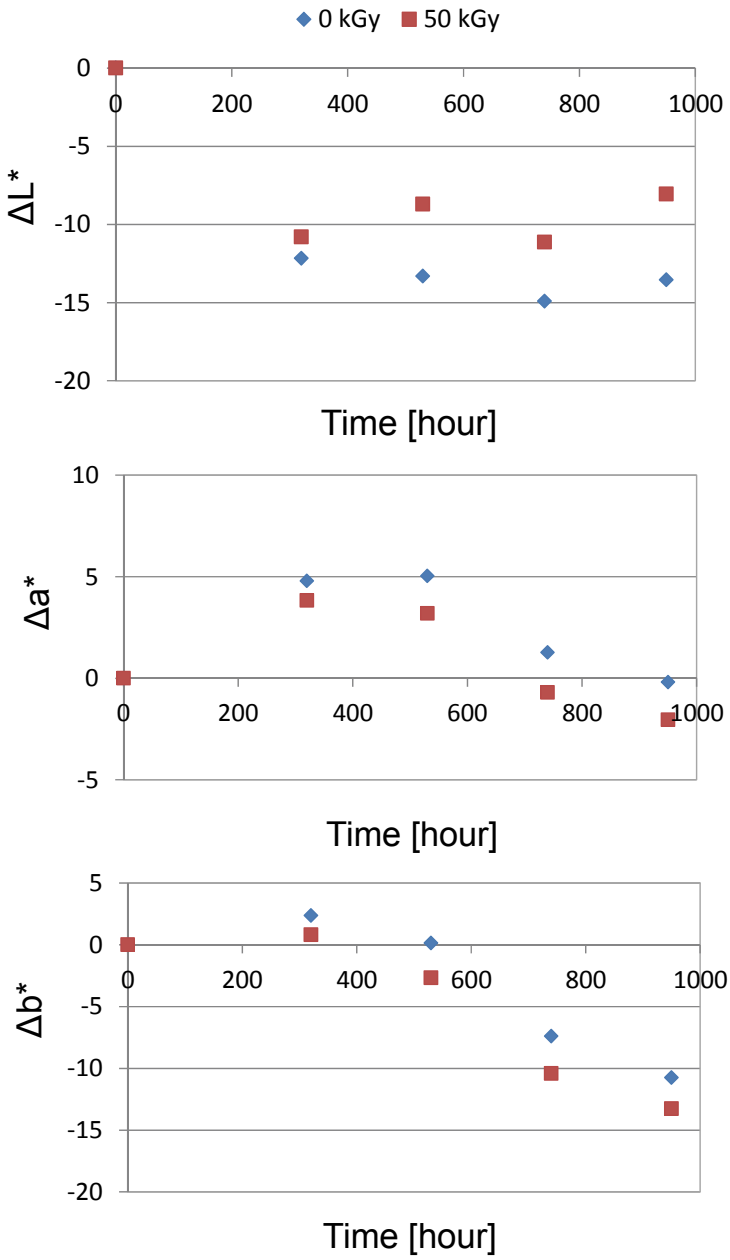

Figure 2. Estimated mean ΔL*, Δa* and Δb* values of untreated and treated fir sample of various exposure time

The Δa* and Δb* values show the highest differences between unirradiated and electron beam irradiated samples (Figure 3), while the changes in ΔL* values (lightness) do not differentiate significantly between those samples.

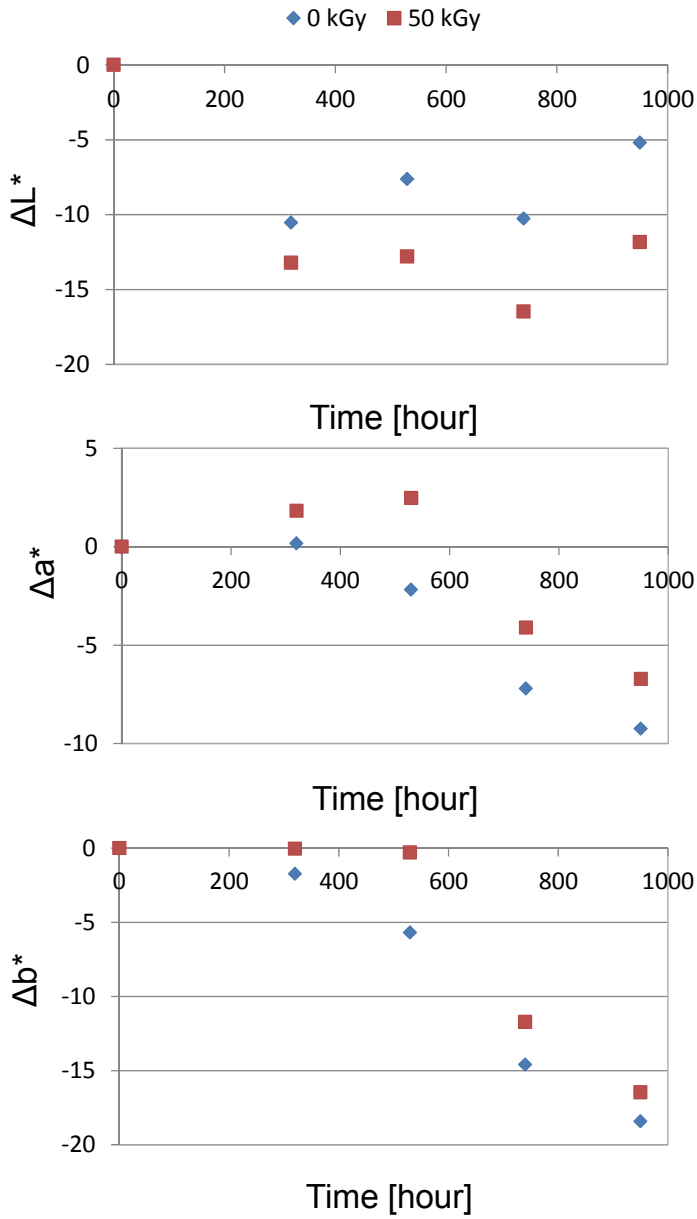

Figure 3. Estimated mean ΔL*, Δa* and Δb* values of untreated and treated larch sample of various exposure time

The Δa* values of untreated samples decreased after the first 320 hours considerable until the end of the weathering test, while the Δb* values of untreated samples decreased until the 950 hours of the weathering test, during this period the color intensity was faded.

The larch samples treated with electron beam irradiation using 50 kGy irradiation doses show a slighter increase in Δa* values during the first 530 hours compared to the untreated sample. Afterwards the decrease in Δa* can further be observed until the 950 hours of the weathering test. Moreover, the changes in the mean of Δb* values of the irradiated samples behave different compared to the Δa* values. The intensity did not change significantly in the first 530 hours. Then the Δb* values decreased until the end of the weathering test. Also the intensity of the color faded. However, with regard to all three parameters L*, a* and b*, the color of the different sample groups were different at the end of the weathering test.

The increase in the b* values is influenced on the photo-yellowing of the photo-degradation of phenolic products (Kishino and Nakano 2004, Müller et al. 2003, Pandey 2005), and the decrease in the b* values is estimated by the leaching effect of photo-degradation products (Kishino and Nakano 2004). Changes in wood colour attend with chemical changes in components due to photo-

degradation (Kishino and Nakano 2004, Müller et al. 2003, Pandey 2005, Tolvaj and Faix 1995). Therefore, it could be assumed that different discoloration processes reflect varied photo-chemical reactions on the wood surfaces during the weathering test. The electron beam irradiation affects not only the color changes during the artificial weathering but also the photo-degradation of wood components (Schnabel et al. 2013, Schnabel and Huber 2014).

4 CONCLUSIONS

The unirradiated and irradiated samples showed different intensities in discolorations during the artificial weathering test. The electron beam irradiations influence differently the weathering behavior of the various wood species. This result may give some indication of condensation reactions of different wood components and improvement the colour stability of larch wood samples in the first 530 hours of the weathering test. For Norway spruce and fir wood samples the differences in discoloration between unirradiated and irradiated samples were smaller compared to the larch samples.

These results show that the electron beam processing modified the weathering behaviour of larch wood samples. Even the improvement of the colour stability may serve to extend the service life performance of wood products. However, further studies on the effect of low radiation dose on wood materials were helpful to prevent undesirable reactions of only wood component cleavage.

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ANALYSIS OF THE DEFORMATION OF WOOD DURING THE DRYING PROCESS: AN EXPERIMENTAL APPROACH

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Key words: w timber drying, MC distribution, prong samples

ABSTRACT

Deformations in samples can tell us more about internal stress in wood due to the drying process. This study deals with an approach to model and simulate the realistic deformations in wood samples during two laboratory drying devices.

In order to minimize the sample preparation work and to avoid destroying parts of the drying batch by cutting out samples for prong or slicing tests, we want to predict the resulting deformation and therefore the drying quality concerning casehardening mathematically via the moisture distribution of the samples. This moisture distribution can by examined using the kilns moisture measurement equipment placing the electrodes in different depths over the cross section of a sample. Another positive effect could be that the moisture gradient over the cross section during the drying process may be traced as well.

In the first studies we used beech wood and could predict the deformation (bending) of the prong samples fairly good. The used model for the deformation intensities is suitable for the convectional and the vacuum drying process, respectively. A good comparison between the realistic and simulated changes in deformation was found for both drying process. Further investigation shall prove the results for other wood species.

1 INTRODUCTION

The optimum drying process should be the main objective of industrial and scientific members and actually are focused on saving energy (e.g. short drying times and reasonable costs) and high product quality. Therefore, a homogenous moisture content distribution and low drying stresses in wood boards are very important (EDG-guide line).

The vacuum drying process is one suitable drying method to fulfil the quality and cost requirements (Malmquist and Noack 1960, Simpson 1987, Chen 1997, Teischinger et al. 2013). This process is based on the fact that the boiling point of water is lowered by reducing the atmospheric pressure. Therefore, the temperature to produce water vapour is rapidly reached, and the increasing wood permeability are the dominate factors of the controlling moisture movement (Siau 1984, Perre 2007). This behavior reduces drying times and produces a possible higher end-product quality of the wood.

Different studies deal with the vacuum drying technology to optimize the drying times, the discolorations and avoiding surfaces and honeycomb checks (He et al. 2013, Sandoval-Torres et al. 2009, Altun et al. 2011, Sattho and Yamsaengsung 2005, Yamsaengsung and Buaphud 2006, Ressel 2003, Hansmann et al. 2008). Also, the fundamental driving forces in wood when vacuum drying were analyzed (Chen 1997, Siau 1984, Sebastian and Turner 1994,) and different models can be generated for drying process (Perré 2010, 2011). This model can describe the physics of wood-water relations and interactions with the vacuum dryer (Sandoval-Torres et al. 2011). This can be used to simulate the vacuum drying process. However, information on the moisture content distribution, deformations and stress development during vacuum drying processes is limited. Therefore additional methods are necessary to analyze especially drying stress and casehardening.

Figure 1. Cross section of a sample dried at a high drying gradient for three hours– deformation, discoloration and checks are well apparent (red rectangle is original cross section)

The objectives of the current study are the analysis of the moisture content distribution and deformation development in wood drying with two different laboratory drying chambers. A standard climate chamber was used for the reference process. The data are applied to model and simulate the shrinkage deformations of wood in order to predict casehardening and possibly checking. Therefore, two different drying methods were analysed to simulate the shrinkage deformations in order to test the ability of the algorithm in general.

2 MATERIAL AND METHODS

2.1 Wood species

In this study, beech wood samples with dimensions of 400 mm long x 80 wide x 35 mm were used. The initial moisture content was approximately 80 %. The end grains of the samples were coated using an aluminium foil bonded to the wood surface with PUR resin to prevent too fast drying on these sides.

2.2 Drying conditions

Twenty eight samples of beech with red heartwood wood were dried simultaneously at constant climate conditions. To simulate a convectional drying process a standard climate chamber (Binder KBF 240) was used drying fourteen samples at 85 °C and 15 % relative humidity (RH). To simulate a vacuum drying process a vacuum drying oven (Binder VDL 115) was used drying fourteen samples at 65 °C and at an atmospheric pressure lower than 100 mbar.

2.3 Sample preparation

After 1, 3, 6, 10, 24, 34 and 48 hours of drying time two samples of each laboratory drying oven were selected and removed to measure deformation (figure 1) and to determine checking and moisture distribution (figure 3).

The prong slices were cut out in the middle of the length of the dried samples. Before cutting the prongs a grid with a mesh with of 5 mm was sprayed on the end grain (Figure 2). After taking a picture of the labelled end grain a prong sample was cut and again a picture was taken (Figure 4). The MC for each sample was obtained applying the oven dry method.

The moisture distribution of the cross section was determined by cutting off another slice of the end grain next to the prong slice and split it into 25 sections (5 x 5 pieces) to get detailed information on it (figure 3). The MC of each section was determined applying the oven dry method.

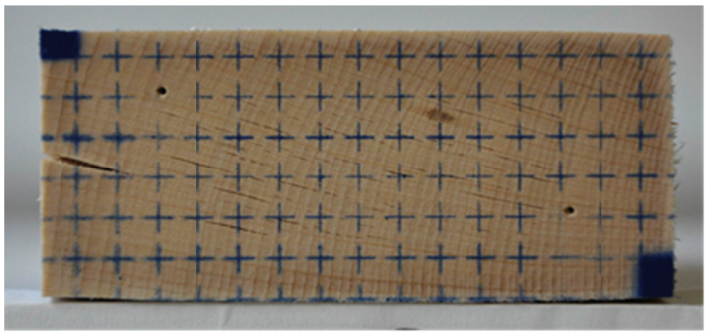

Figure 2. cross section of a sample with sprayed grid to determine the deformation before and after cutting the prongs.

2.4 Modelling and Simulation of the shrinkage deformation

Matlab© R2010a software packages was used for the analysis of the data and the calculation of the deformation during the different wood drying process.

The deformation process was simulated based on the moisture distribution data of each sample at various drying times. The samples were divided into finite elements to model the two dimensional deformations according to the real changes in moisture content of the elements. The size of each element correlated with the real section size of the moisture distribution sample.

3 RESULTS AND DISCUSSIONS

The complete analysis on the moisture distribution and the behaviour of deformations was performed for beech wood samples in different drying conditions at various drying times. To get a qualitative impression of the variation of the measured moisture contents and their distribution over the cross section see Figure 3.

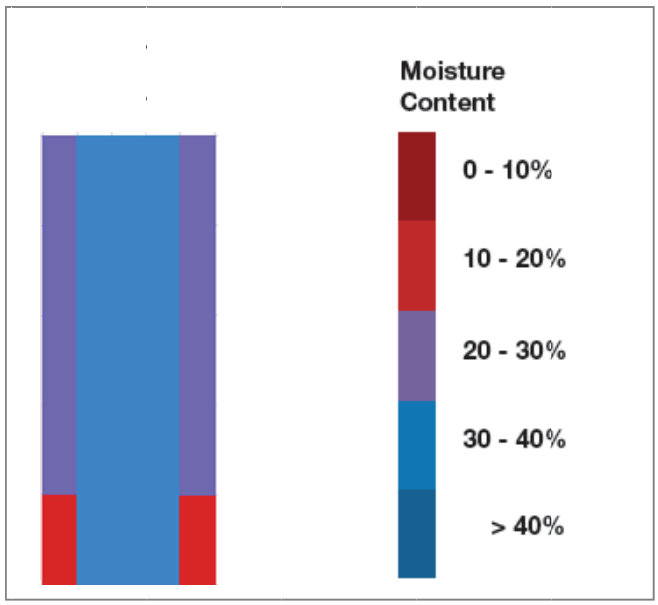

Figure 3. Moisture distribution of one sample after 3 hours dying in the climate chamber at 85° C and 15 % RH

The deformation of the prongs can be seen in the Figure 4. After having cut the prong sample a slight deformation can be observed comparing Figure 2 and Figure 4, a). At this drying stage with a MC above FSP in the core layers of the sample, the prongs should bend out, but presumably due to the relatively high drying temperature (85° C) some ductile set could have been occurred and some internal stresses could have been relieved by cracking, see Figure 2.

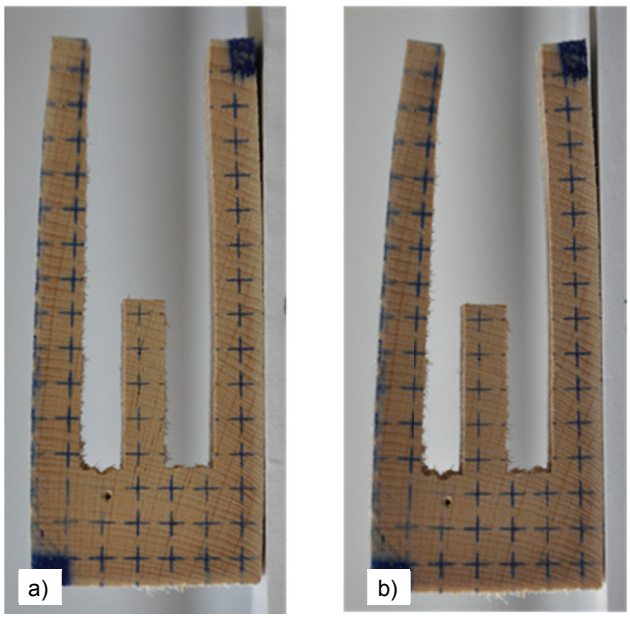

Figure 4. Detail of prong samples a) direct after cutting and b) after 24 hours storage to equalize the MC

After 24 hours storage of the prong samples according to EDG-guide clearly deformations due to the equalization of the MC are apparent (figure 4, b)). The deformation of the prongs is related to internal stresses due to the drying process and is used to define the quality of the wood drying process and the drying quality of the timber. As a recommendation prong deformation (PR) for drying quality class one and class two should not exceed 1,5 % and 2 % respectively. There are no limitations for the level of drying stresses for the third class. For our study the quality results are class three.

To validate our approach, the comparison between the real deformation of the prongs and the calculated deformation due to the moisture distribution is shown in Figure 5. It is recognizable that the simulation of the prong deformation and the actual deformation due to the drying process are comparable. However, there are still some challenges to optimize the modelling and simulation method (e.g. algorithm). Nevertheless, the aim of this study was fulfilled, to show the potential for a 2 dimensional simulation model of realistic deformation due to the wood drying process.

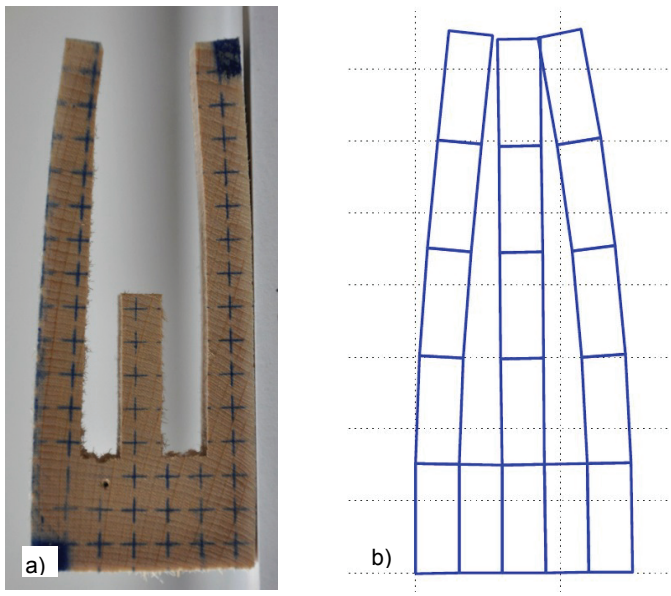

Figure 5. a) prong deformation after 24 hours storage, b) calculated prong deformation due to moisture distribution

4 CONCLUSIONS

The algorithm to simulate prong deformation on basis of a moisture distribution works promising for the first tests with beech for both drying processes. Further studies on different drying rates and different wood species should be carried out to proof and improve prediction accuracy.

Applying accurate moisture measurement equipment and measuring design could provide the adequate information on the moisture content distribution already during the drying process to predict the drying quality with the possibility to alter the drying process parameters if required.

Nevertheless a non-destroying prediction of the prong deformation (drying quality regarding internal stresses) can save material and time as there is no need to cut samples and to wait for 24 hours to get the results.

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USE OF TREE BARK AS INSULATION MATERIAL

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Key words: Tree bark, insulation material, porosity

ABSTRACT

The general trend towards energy efficient building constructions has led to an increased need of insulation materials. Basically, organic (hydrocarbons) and inorganic insulation materials can be distinguished whereby the first group is dominated by petrol based products (polystyrene) and the second one is mainly mineral wool, which both have a rather poor ecological performance because of non-renewable resources or an energy intensive production process.

An option would be low density bark based panels which have proved to be a promising insulation material and are especially interesting because it can be made use of the natural 'tree insulation material', namely bark. Bark has advantageous properties like a low density, a relatively high resistance against microorganisms and a low thermal conductivity whilst also having a high heat storage capacity. Also from an economic point of view the material is interesting, as bark is a

traditional by-product of timber manufacturing and is rarely used for products with a higher value added.

Therefore, insulation boards made out of differently fractionated bark particles and a tannin-hexamine resin were produced. These panels were manufactured with a density lower than 500 kg/m³. Furthermore, the mechanical and thermal properties of the bark panels have been evaluated.

Measurements showed that the mechanical properties (MOR, MOE, internal bond, tensile strength and compressive resistance) of the bark-based panels are comparable with those of commonly available insulation boards. Only water absorption and thickness swell after 24 h are marginally higher than those of conventional insulation materials.

In terms of the thermal characteristics, these bark based panels showed thermal conductivity values higher than those of light insulation boards. Nevertheless, the application as insulation material is possible. By analyzing the thermal diffusivity (material property describing the rate at which heat flows through a material) of bark based panels, their performance is clearly superior to existing insulation boards.

The bark boards' structure has been analyzed by the means of X-ray computed tomography and it has been found that the pore structure can be satisfactorily influenced in the pressing process, which is promising for a selective adjustment of the boards' thermal properties.

On the one hand a raw material source for a booming industry was found, on the other hand the study of a natural thermal insulator could lead to improved materials for future requirements.

1 INTRODUCTION

The building industry requires a rising amount of insulation materials due to a general trend towards energy efficient buildings.

The insulation materials market is dominated by polystyrene and mineral wool, which both are based on non-renewable resources.

An option could be low density bark based panels which proved to be a promising insulation material and are especially interesting because one makes use of the natural 'tree insulation material', namely bark. Globally roughly 1.6 billion solid m³ of wood are industrially used, although this accounts only for 43 % of the total cuts as the majority is directly burned (Barbu et al. 2014). Considering that the average bark content of a tree is approximately 10 %, the whole bark volume is roughly 160 million m³.

Even though tree bark is already used in products like bark mulch, absorption materials, raw materials for tannin production and fertilizers, there is a call for alternative products with a higher value added (Naundorf et al. 2004).

Bark is the boundary layer of a tree and protects it from physical and biological exterior attacks (Figure 1). Therefore, it has ideal properties, such as a low density, a high concentration of extracts, very good thermal insulation properties and a relatively low flammability (Fengel and Wegener 2003).

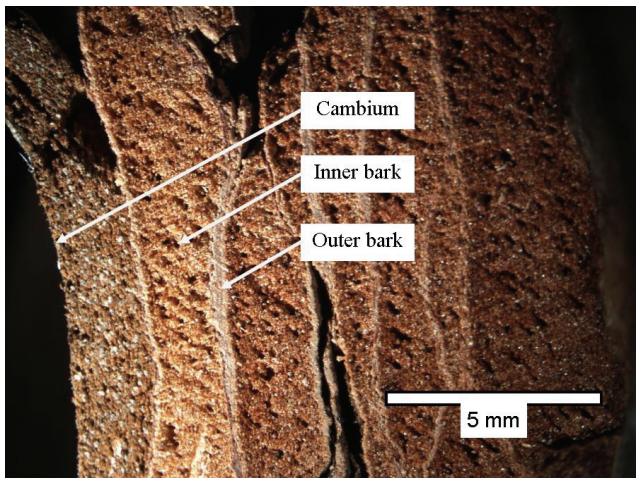

Figure 1. Bark cross section from the cambium to the outside

There is a long history of bark use in various wood-based products (e.g. Volz 1973, Nemli and Colakoglu 2005, Kraft 2007, Xing et al. 2007, Yemele et al. 2008). These studies concentrated on adding bark constituents to standard wood based products finding that increasing bark ratio goes along with poorer mechanical properties.

The aim of this study was to assess whether low weight (density < 500 kg/m³) bark panels can be produced and whether those are suitable as a thermal insulation material (Kain et al. 2012, 2013a).

2 MATERIALS AND METHODS

2.1 Material

The insulation boards are based on larch (Larix decidua Mill.) bark, which was collected in a sawmill in Unternberg, Salzburg, Austria.

The bark was crushed in a four-spindle shredder (RS40) and successively sieved so that only the fraction of diameter 6-10 mm was selected for the production of the insulation boards. This fraction was dried with a vacuum dryer at 60 °C and an absolute pressure of 200-250 mbar until a final average moisture content (MC) of 9 to 10 % was reached.

Quebracho-tannin-extract (Colatan GT 10) from Markmann GmbH, hexamethylenetetramine from Merck Schuchardt OHG (≥ 99 %) and sodium hydroxide solution (≥ 32 %) from Carl Roth GmbH & Co. KG were used to prepare the resin.

2.2 Panel production

The resin was obtained mixing 50 weight-% of tannin-extract powder and 50 weight-% of water with a mechanical stirrer at variable stirring rate in the range between 700 and 1500 rpm. 10 % of hexamethylenetetramine (whe/wt) was added using a 33 % solution and adjusting the pH to 9.0 with the solution of sodium hydroxide.

The bark particles were mixed for 60 seconds at around 200 rpm with the tannin-based adhesive in a ploughshare mixer. The resin coated particles were then discharged and spread in different quantities in a mould of 240 x 350 mm and manually pre-compacted. Then the boards were pressed with a laboratory press HLOP 280 of Höfer GmbH for 8 minutes at a plate temperature of 180 °C fixing the final height of the panel at 20 mm. Eleven different boards were produced with two replicates each. The effect of density and resin amount was studied according to the experimental design depicted in Table 1.

2.3 Panel testing

The mechanical [modulus of rupture (MOR), modulus of elasticity (MOE), internal bond (IB), tensile strength (T), thickness swell (TS), water absorption (WA) after 24h] and thermal properties (thermal conductivity) of the bark panels were evaluated. Ongoing the bark panels were scanned with a CT scanner and the obtained scans were analyzed by the means of digital image analysis.

Table 1. Experimental design with the factors density and resin content (based on the oven-dried weight of bark particles) (2 specimens for each combination)

Target density (kg/m³)	Tannin resin content (%)	Mass of resinated particles (g)
500	15	996
	10	972
	5	948
400	15	797
	10	778
	8	770
350	15	697
	10	680
300	15	597
250	20	510
	15	498

3 RESULTS

Some examples of the insulation boards obtained are shown in Figure 2.

Figure 2. Tannin-resin bonded bark insulation panel (thickness = 20 mm, density = 400 kg/m³)

An overview of the mechanical panel properties can be seen in Table 2.

Variations in panel properties could be satisfactorily explained by the regression models which was shown by coefficients of determination greater than 0.70 (Table 3) and statistically highly significant ($p<0.001$) models for all properties apart from thermal conductivity where the model is only very significant ($p<0.01$). Panel density and resin content do have a significant influence on mechanical board properties, which is known from various studies on mechanical board properties (e.g. Gupta et al. 2011).

Table 2: Mechanical board characteristics with standard deviations in brackets

Target Density (kg/m³)	Amount of Adhesive (%)	Density MOR/MOE (kg/m³)	MOR (N/mm²)	MOE (N/mm²)	Density IB (kg/m³)	IB (N/mm²)	Density TS/WA (kg/m³)	TS 2 h (%)	TS 24 h (%)	WA 2 h (%)	WA 24 h (%)
500	15	565 (21.21)	2.94 (0.43)	502.02 (73.43)	507.65 (75.70)	0.32 (0.10)	514.87 (61.08)	8.41 (1.54)	12.83 (1.12)	34.95 (7.22)	58.69 (8.69)
500	10	565 (21.21)	2.19 (0.18)	384.46 (33.83)	514.77 (65.51)	0.21 (0.08)	534.19 (74.12)	13.46 (2.17)	21.08 (1.65)	45.41 (9.24)	70.11 (8.94)
500	5	555 (7.07)	1.72 (0.12)	254.54 (43.44)	533.38 (23.99)	0.16 (0.01)	535.10 (29.50)	20.52 (1.11)	27.10 (1.72)	56.23 (3.08)	77.17 (2.96)
400	15	475 (7.07)	1.71 (0.23)	239.55 (10.42)	457.63 (31.05)	0.24 (0.02)	452.62 (23.25)	8.97 (1.52)	12.16 (1.79)	45.64 (1.99)	67.13 (2.32)
400	10	435 (7.07)	1.09 (0.21)	165.55 (6.67)	402.27 (58.29)	0.14 (0.06)	408.16 (67.69)	12.68 (0.93)	16.80 (2.05)	52.12 (6.52)	79.12 (4.57)
400	8	455 (7.07)	0.85 (0.02)	140.11 (0.28)	440.34 (14.93)	0.14 (0.01)	442.45 (9.75)	15.56 (1.27)	19.76 (2.02)	56.76 (1.31)	80.39 (0.78)
350	15	405 (7.07)	0.86 (0.11)	149.70 (27.62)	384.68 (22.91)	0.16 (0.03)	386.37 (40.10)	10.39 (1.86)	14.04 (2.98)	51.11 (3.62)	78.32 (3.62)
350	10	400 (14.14)	0.67 (0.11)	121.46 (9.59)	387.29 (23.24)	0.12 (0.02)	387.09 (39.00)	13.37 (1.10)	17.34 (1.95)	54.93 (3.07)	82.60 (4.20)
300	15	345 (7.07)	0.45 (0.04)	67.80 (8.45)	340.64 (28.42)	0.10 (0.05)	325.21 (35.79)	10.27 (0.79)	12.21 (1.07)	57.68 (1.45)	83.06 (2.62)
250	20	320 (14.14)	0.37 (0.01)	41.94 (6.21)	293.24 (23.73)	0.07 (0.03)	293.83 (31.12)	9.62 (1.07)	11.81 (1.06)	55.87 (1.82)	83.79 (2.05)
250	15	305 (21.12)	0.27 (0.01)	46.39 (15.32)	293.53 (29.84)	0.06 (0.03)	282.14 (31.50)	8.80 (2.12)	12.31 (3.35)	59.81 (2.32)	85.96 (10.24)

MOR = modulus of rupture, MOE = modulus of elasticity, IB = internal bond, TS = thickness swelling, WA = water absorption

Table 3. Regression model (Beta-values for the coefficients with significance) for the board properties including the regressors resin content and panel density

	MOR	MOE	IB	TS 2 h	TS 24 h	WA 2 h	WA 24 h	Lambda
Coefficient of determination	0.949**	0.921**	0.867**	0.699**	0.729**	0.788**	0.824**	0.820*
Significance level	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.001
Resin content	0.399**	0.392**	0.532**	-0.896**	-0.763**	-0.686**	-0.592**	0.092
	(0.000)	(0.000)	(0.000)	(0.000)	(0.000)	(0.000)	(0.000)	(0.650)
Density	1.140**	1.123**	1.092**	-0.125	0.151	-1.042**	-1.076**	0.962*
	(0.000)	(0.000)	(0.000)	(0.154)	(0.070)	(0.000)	(0.000)	(0.001)
Sample size	22	22	59	60	60	60	60	11

MOR = modulus of rupture, MOE = modulus of elasticity, IB = internal bond, TS = thickness swelling, WA = water absorption, Lambda = thermal conductivity

This context was also proved for the investigated bark-based tannin glued panels. For MOR, MOE, IB, WA both density and resin content have a highly significant influence, whilst TS is only statistically significantly influenced by resin content and thermal conductivity only by density. MOR is nearly three times stronger influenced by the panel density than by the resin content (the same is true for MOE). The MOR lies between 0.32 N/mm² (standard deviation (sd) = 0.06 N/mm²) on average for the 250 kg/m³ boards and 2.28 N/mm² (sd = 0.59 N/mm²) on average for boards with a density of 500 kg/m³. The MOE ranges between 44.2 N/mm² (sd = 9.88 N/mm²) and 380.3 N/mm² (sd = 118.08 N/mm²) for the same boards.

Focusing on IB it was found that it is twice as much influenced by density than by resin content. The 250 kg/m³ boards showed an IB of 0.06 N/mm² (sd = 0.03 N/mm²) whilst the 500 kg/m³ boards accounted for 0.23 N/mm² (sd = 0.10 N/mm²).

Thickness swell is statistically highly significantly influenced by resin content, but not by density. The highest thickness swell was observed with panels with a resin content of 5 % with 20.52 % on average (sd = 1.11 %) after 2 h water immersion and 27.10 % (sd = 1.72 %) after 24 h water immersion.

Water absorption after 2 h is 1.5 times and after 24 h 1.8 times stronger influenced by panel density than by resin content. Water absorption was highest with the lightest boards (250 kg/m³) with 57.8 % (sd = 2.86 %) after 2 h and 84.88 % (sd = 7.06 %) after 24 h water immersion (a similar trend was observed for 2 hours water immersion).

With regard to thermal conductivity a very significant ($p<0.01$) positive correlation between panel density and thermal conductivity could be detected (coefficient of determination = 0.82). A target density of 250 kg/m³ led to an average thermal conductivity of 0.069 W/(m*K) (sd=0.0007 W/(m*K) and the heavier boards (500 kg/m³) showed a thermal conductivity of 0.093 W/(m*K) (sd=0.002 W/(m*K)).

CT scanning and subsequent digital image analyses enabled the evaluation of structure and pore size distribution of the bark based insulation panels. It was found that pores smaller than 1 mm² are predominantly pores within the bark particles whilst the larger pores are caused by imperfect particle compression (Figure 3).

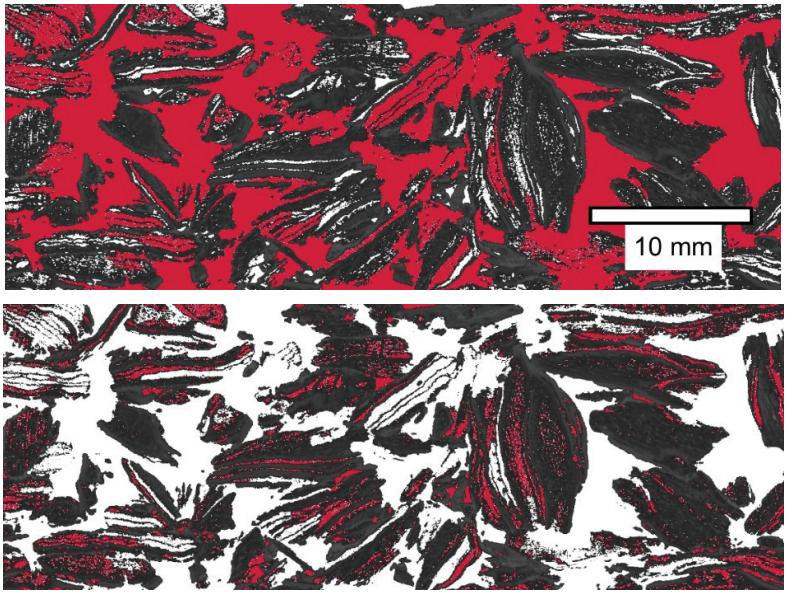

Figure 3. CT-tomogram of a bark insulation panel (thickness = 20 mm, density = 250 kg/m³), first pores > 1 mm² highlighted (red), second pores < 1 mm² highlighted

4 DISCUSSION

Low weight bark based panels glued with a condensation resin (8 to 12 % UF based on the oven-dried weight of particles) were found to show sufficient mechanical strength (MOR, IB, CR (compressive resistance), T (tensile strength), TS) for insulation purposes (Kain et al. 2013a). The boards glued with tannin were on the one hand partly much lighter than the boards studied before and on the other hand were found to have similar properties than the UF resin bark boards (Kain et al. 2014).

The studied panels had a density between 250 and 550 kg/m³.

The MOR of a standard wood fiber insulation panel with a density between 230 and 400 kg/m³ and a thickness of more than 19 mm is defined to be more than 0.8 N/mm² according to EN 622-4 (EN 2010). This requirement was met with panels with a density of 350 kg/m³ glued with 15 % of tannin resin (average 0.86 N/mm², standard deviation 0.11 N/mm²). The heavier boards were even better, whilst the lighter boards didn't meet the requirements and their potential usage has to be limited to applications where they are not subjected to bending.

Focusing on IB (most important for insulation panels as it is the characteristic describing the adhesion of single particles especially in the core) the studied boards showed properties superior to most of the other commonly available insulation panels, of course also because of the comparatively higher panel density (Ashby-chart in Figure 4).

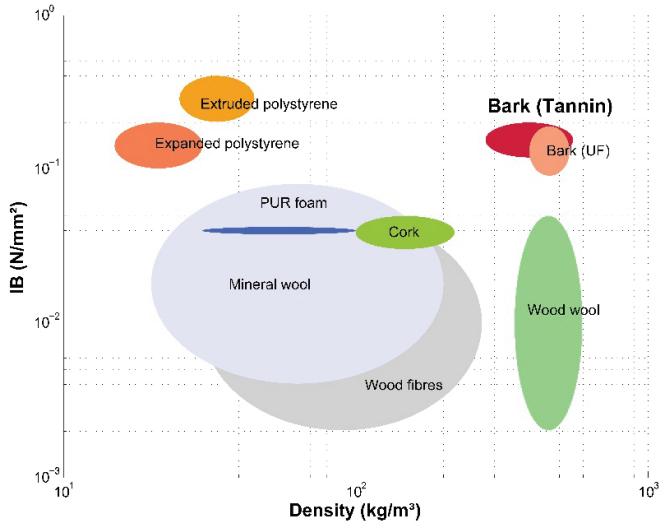

Figure 4. Internal bond of insulation materials by comparison (data apart that of bark according to Pfundstein et al. 2007, p. 13)

Schwemmer (2010) developed insulation panels out of reed mace and limited the TS after 24 hours of water storage at 20 °C with 15 %. This requirement was achieved with a resination of 15 and 20 %, whilst a resination of only 10 % resulted on average in 18.4 % (SD = 0.6 %) thickness swell.

With regard to thermal conductivity the bark based tannin glued panels showed exactly the same context between density and thermal conductivity than the bark (*Pinus sylvestris*) panels pressed by Kain et al. (2012). Comparing them to other insulation materials, their thermal conductivity is with 0.069 W/(m*K) (sd SD = 0.00070 W/(m*K)) for the 250 kg/m³ panels and 0.093 W/(m*K) (sd = 0.0021 W/(m*K)) for the 500 kg/m³ panels not as low as with the very light insulation materials (e.g. polystyrene and mineral wool). Nevertheless is still competitive with wood wool or heavier wood fiber materials (Ashby-chart in Figure 5).

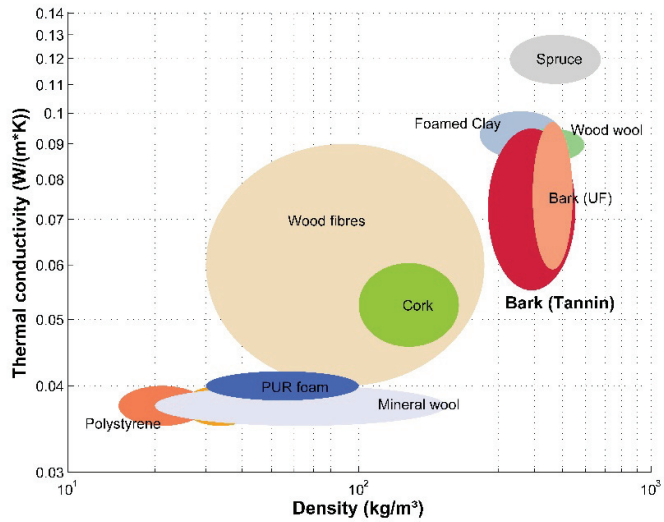

Figure 5. Thermal conductivity of insulation materials by comparison (data apart that from bark according to Pfundstein et al. 2007, p. 9)

In terms of pore size distribution it was found that the heavier panels have less very large pores (> 5 mm²) but also have a higher thermal conductivity. An optimization task for future research activities

is therefore to find the optimal combination of particle size and panel density in order to produce an adequately stable and insulating panel.

5 CONCLUSIONS

With the presented study it could be shown that larch bark (*Larix decidua*) is a suitable raw material for insulation panels. More important a highly natural adhesive system based on tannin resin could be successfully applied for particle bonding which enables manufacturers to produce synthetic resin-free insulation panels.

As expected density and resin content significantly influence the mechanical board properties (MOR, MOE, IB), but interestingly resin content only explains a third of the variance that can be explained by panel density and has therefore much less explanatory power than expected. These circumstances raise the question which chemical bondings are prevalent in the particle resin matrix and if reactive groups within the bark plays a significant role in particle bonding. Therefore further investigations should focus on that assumption.

Furthermore the heat transfer mechanisms within the presented boards have to be studied in detail as large cavities in the relatively thin boards (20 mm) are likely to cause worse results in thermal conductivity measurements than those which would be measured with thicker boards. Therefore, ongoing scientific work might focus on a test run with thicker boards. In addition, the interactions between particle size, panel density and thermal conductivity might be an issue for further research emphasis.

In this respect the knowledge of panel structure, especially of the pore size distribution could enhance the opportunities in the optimization of thermal conductivity.

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ADVANCED TANNIN BASED WOOD PRESERVATIVES

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Key words: Tannin, Caprolactam, Wood preservative, Durability

ABSTRACT

Protecting timber with natural products is one of the most significant challenge in wood technology since decades, but recently also the political institutions show the need for this challenge to be win. Tannin-based wood preservatives were studied for a long time (Laks et al. 1988), but only combining the knowledge in tannin chemistry developed in more recent years (Pichelin et al. 1999) it was possible to produce tannin-based formulations that enabled to fix in the timber structure resisting leaching (Thevenon et al. 2009).

The first generation of water resistant tannin-based wood preservative presented very good mechanical properties, enhancement of fire resistance and also outstanding biocide properties (Tondi et al. 2012). Unfortunately, the weathering tests underlined the limit of these innovative formulations: On the one hand the dark color of the treated sample brings to higher absorption of UV-light with the consequence of breaking down the tannin polymer and on the other hand the rigid tannin polymer did not resist the continuous dimensional changes of wood (Tondi et al. 2013).

With this work we would like to present a second generation of tannin wood preservative in which the effect of polyethylene glycol and caprolactam have been studied. The latter have been added to the tannin-based formulation in order to increase the elasticity of the tannin polymer whether with a blend or with a real copolymer.

The results of these tests have shown a considerable improvement of the properties of the sample treated with tannin-caprolactam solution especially in terms of resistance against water and biological attacks. Some improvements have been registered also for the dimensional stability and for the weathering even if these enhancements do not solve the problem completely yet.

1 INTRODUCTION

In the previous centuries, universal solutions for preserving wood were used. From the very beginning, solutions based on oils and waxes were recognized for most of the applications until creosote first, and heavy metal based formulations such as chromate-copper-arsenate (CCA) after, were dominating the market warranting very long service life (Richardson 1993, Ruddick 2011). More recently, restrictions on the use of creosote and heavy-metal based wood preservatives have focused public and government attention on technological developments in the wood protection area, specifically on the availability of more natural alternatives (Evans 2003, Schultz et al. 2007, 98/8/EC, 2011/71/EU). Thus the availability and use of environmentally friendly wood preservatives is nowadays strictly required.

Nowadays, several research groups are working with the objective of finding natural solutions for protecting wood and a very interesting overview on this topic has been recently published by Singh and Singh (2012). In addition of these researches also other groups have developed interesting wood preservatives.

In the very wide research field which proposes to protect wood with natural substances, a considerable work is carried by the researcher that considers tannins as the ideal solution. Following the original work of Laks (Laks et al.1988) other research groups have decided to follow a similar investigation line (Yamaguchi and Okuda, 1998; Taylor et al. 2006; Tascioglu et al.2012).

However, thanks to the discovery of the group of Prof. Pizzi (Pizzi 1980), (Pichelin et al.1999) related to the polymerization of poly-flavonoids, it has been possible to combat the long-standing leaching problem of tannin.

The technology that matches to this tannin property consists in infiltrate the activated oligomers of flavonoid in the wood with a successive in-situ polymerization catalyzed by heat (Thevenon et al. 2009).

The original formulations have shown very good preservation properties against biologic attacks and fire and they have also shown improved mechanical properties (Tondi et al., 2012). Unfortunately the tannin-treated timber is still quite highly affected by artificial and natural weathering exposures. The tannin polymers, being relatively rigid and dark carry to two drawbacks: (i) to mechanically cracking of the polymer during the continuous dimensional changes occurring when the wood is exposed outdoors and (ii) to higher radical degradation due UV-lights (Tondi et al. 2013).

The idea of using PEG and caprolactam for rendering the polymers more elastic and clearer is presented in this article.

2 EXPERIMENTAL

2.1 Materials

Mimosa (Acacia Mearnsii) tannin extract was supplied by Silva Chimica (Cuneo, Italy), while hexamethylenetetramine (hexamine), boric acid sodium hydroxide, ε-caprolactam and polyethylene glycol 400 (PEG) were provided by Sigma-Aldrich. The scots pine (Pinus Sylvestris) boards were provided by two different sawmills from Kuchl (A).

2.2 Methods

2.2.1 Impregnation method

Scots pine samples of various dimensions were cut from stabilized sapwood portions and successively oven-dried at 103 °C until constant mass. All treatments were performed using a single vacuum impregnation. In the treatments, the vacuum cycles consisted of 20 min at 8 mbar in a treatment desiccator. When the pressure was slowly increased up to ambient pressure, the specimens were kept in the tannin-based solution for 24 hours. After the impregnation treatment, the excess solution of specimen surface was removed with some blotting paper. The weights of the wet samples were recorded and the retention of tannin boric acid uptake on wood blocks was calculated.

These impregnated test specimens were kept at least 12 hours in the oven at 103 °C to allow the tannin resin to harden. The weight of the dry samples was further registered and then the impregnated wood blocks were then reconditioned at 20 °C and 65% RH until stabilization before testing.

The tannin formulations tested were prepared according to table 1 and following the method reported by Tondi (Tondi et al. 2013).

Table 1. Composition by weight percentage of tannin-based formulations for the impregnation of Scots pine

Formulation	Tannin (%)	Boric acid (%)	NaOH (%)	Hexamine	Additive (%)	Water (%)
Tannin 10%	9,0	0,5	0,5	0,6	-	89,4
Tannin 20%	18,0	1,0	1,0	1,2	-	78,8
Tan.10%+ □-Caprolactam	7,6	0,4	0,4	0,5	13,1	78,0
Tan.10%+ PEG	9,0	0,5	0,5	0,6	19,5	69,9

2.2.2 Testing methods

Various tests have been done in order to evaluate the efficacy of the new, more elastic treatment.

Dimensional stability

Specifically cut samples of 20 (t) x 20 (r) x 10 (l) mm were impregnated with the tannin formulations and cured. When the specimens were fully dried, they were exposed to constantly increasing humidity condition for one week for each step. The three dimensions were collected at 20°C with relative humidity of 0, 10, 20, 30, 40, 50, 60, 70, 80, 90 and 95%.

Leaching

Treated samples of 50 x 25 x 15 mm dimensions were undergone to the ENV1250-2 leaching methods. This leaching process was performed dipping the treated samples under water under continuous stirring with exchanging the water at 1, 2, 4, 8, 16 and 48 hours.

The tannin released was spectrometric quantified measuring the absorbance of the leaching water at the wavelength of 340 nm in which tannin present a maximum of absorbance. The concentration of the solutions was calculated according to the calibration curve obtained with solution to known strength of tannin (pH 9,0). The data were successively treated so that the sum of the released tannin was reported at each leaching step.

Biological tests

Sample of 50 x 25 x 15 mm³ were exposed to a mono culture decay test conducted according to EN 113 (1996) using the brown-rot “Antrodia” species strain CIRAD/2304/1. Sterile culture medium (65 ml), prepared from malt (40 g) and agar (20 g) in distilled water (1 l), was placed in a culture flask, inoculated with a small piece of freshly grown pure mycelium and incubated for 2 weeks to allow full colonization of the medium by the mycelium. Tested blocks were sterilized by gamma radiation after measuring their oven-dried weights. Two blocks (one treated and one control) were placed directly on malt agar medium in each culture flask under sterile conditions. Incubation of Antrodia species and were carried out at 27 °C, 75% RH in a climatic chamber for 16 weeks. Six replicates per treatment were exposed to the fungi.

Once the fungal exposure was finished, the blocks were removed from the test bottles, and the mycelium was carefully brushed off the samples, and the blocks were dried at 103°C and weighed. Controls used only untreated pine sapwood samples to check the virulence of the fungus. The extent of the fungal attack was determined based on the percentage of mass loss. Weight loss (WL) was expressed as a percentage of the initial oven dried weight of the sample (eq.1).

$$WL\ (\%) = ((Wb-Wa)/Wb) \times 100 \quad (1)$$

Where Wb and Wa are the weight before and after the biologic attack, respectively.

Artificial Weathering

Series of 5 samples of treated and untreated Scots pine sample of 100 x75 x20 mm³ (l,t,r) were exposed to a QUV/spray accelerated weathering tester (Q-Panel Lab Products Cleveland, USA) according to the guidelines of EN 927-6 (2006).

The color of the surface was monitored with the Mercury 2000 spectrophotometer (Datacolor) after regular intervals of exposure. Six measurements for sample were recorded. The diameter of the spotlight used for measurement was 11 mm. The wood color was determined according to the CIELAB space with CIE standard illuminates D65 and a 10° standard observer.

3 RESULTS AND DISCUSSION

The addition of PEG and caprolactam to increase the elasticity of the formulation after polymerization have shown one immediate difference: The sample treated with PEG presented a thick layer of PEG which could not fully penetrate in the wooden structure, while the addition of caprolactam showed a pleasant lighter color and a homogeneous penetration. After cleaning the surface, all the samples were investigated against the four properties described in the following sub-chapters.

3.1 Dimensional stability

It is well known that the longitudinal swelling is very minor compared to the radial and tangential swelling. In our tests it was observed that swelling registered in radial and tangential direction were rather similar and therefore only the tangential swelling is described (Fig.1)

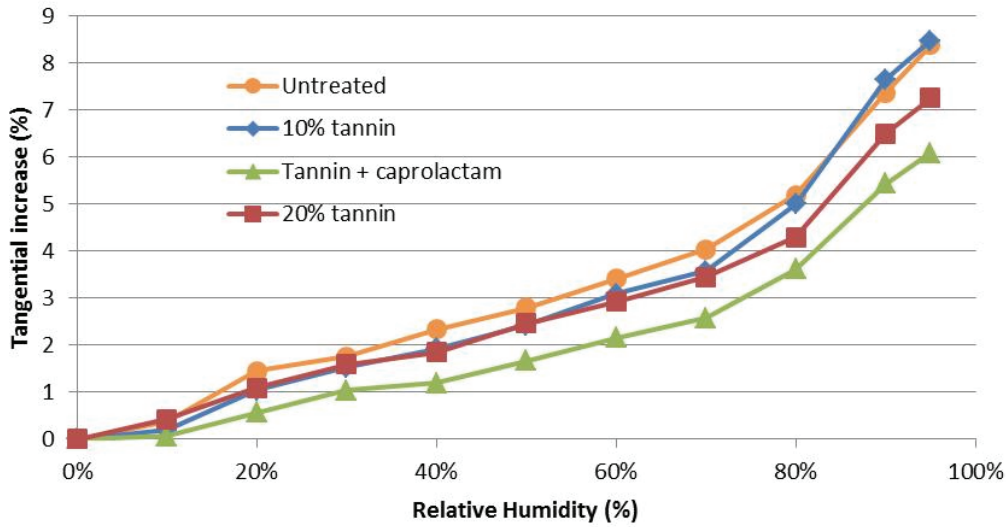

Figure 1. Tangential swelling of untreated, 10% and 20% tannin treated scots pine specimens compared with Tannin-caprolactam treated samples under different RH conditions.

As already underlined in previous works, the presence of tannin polymers contains the swelling of wood. However, the effect of caprolactam show a significant decrease in swelling which suggests a tighter polymerization between tannin molecules and caprolactam. This property suggest an increased of internal stress of the structure and therefore a higher mechanical strength of the formulation.

The sample treated with PEG did not get reliable results, because the gelly patina produced by PEG in presence of water molecule embedded the correct evaluation of the swelling

3.2 Leaching tests

The leaching of tannin according to the leaching test ENV1250-2 is reported in fig.2.

The loss of tannin during leaching is generally very limited for PEG treated samples. This phenomenon is mostly due to the protective effect of PEG layer which is found all over the wood surface. The appearance of the specimens at the end of the leaching present a thick slimy film of swelled PEG that renders the sample impossible to handle.

The caprolactam added formulations also show improved leaching resistance. The leaching of tannin for the sample treated with this formulation was around 40% less than that shown by the samples treated with the classic 10% formulation.

This result supports the ones obtained for the swelling: The polymer infiltrated in the wood is more strongly reticulated bringing to a more hydrophobic surface. The leaching is than less effective because of two reasons. On the one hand more tannin molecules are cross-linked, and on the other hand the penetration of water becomes consequently harder decreasing the leaching of the few free tannin molecules.

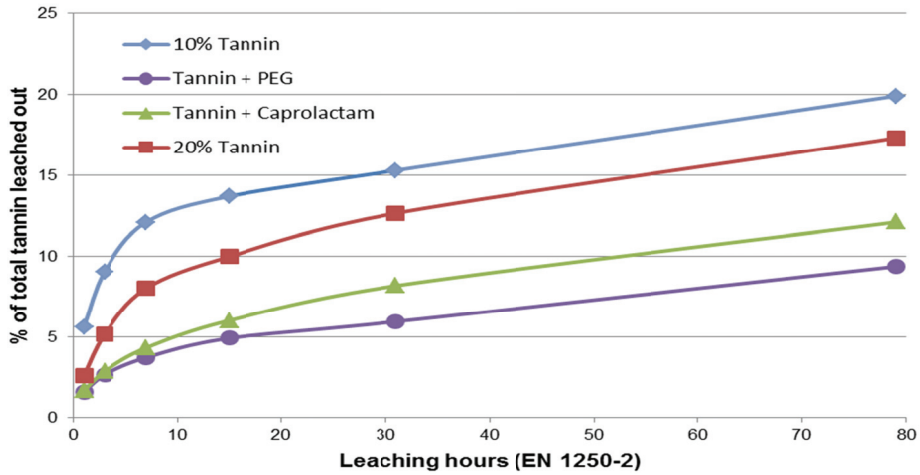

Figure 2. Percentage of total tannin released during the ENV 1250-2 leaching process.

3.3 Biological tests

The biocidal effect of the tannin formulations can be observed on table 2.

Table 2. Weight loss of the Scots pine samples exposed to Antrodia**.**

Treatment	Leaching	Weight loss (%)	Stand.Dev.
Untreated		56,75	6
Tannin 10%		1,31	0,23
Tannin 10%	x	3,35	0,84
Tannin 10% + PEG		1	0,07
Tannin 10% + PEG	x	1,63	0,16
Tannin 10% + caprolactam		1,65	0,25
Tannin 10% + caprolactam	x	0,69	0,16

The efficacy of the treatments with tannin and boric acid is fully confirmed also with the addition of the two additives. In case of PEG the limited weight loss is mostly due to the impossibility for the fungi to reach the wood surface, while in case of caprolactam the slightly higher degradation is due to the lower concentration of tannin and boron in the samples. Conversely, after leaching, the PEG added samples offers a higher free wood surface that can be more easily attacked. In the meantime, the samples added with caprolactam offer an enhanced wood protection due to the higher resistance of the tannin-caprolactam polymer against leaching.

3.4 Artificial weathering tests

The more significant parameter to observe the modification of the surface for tannin treated wood species is L*. Lightness, indeed, show how light/dark is the surface and therefor it gives the immediate information on how much the samples are degraded by artificial weathering. In Fig. 3 the L* value at different exposure time are reported.

It can be observed that at the beginning of the experiment the caprolactam modified samples have clearer color than the other tannin treated specimens.

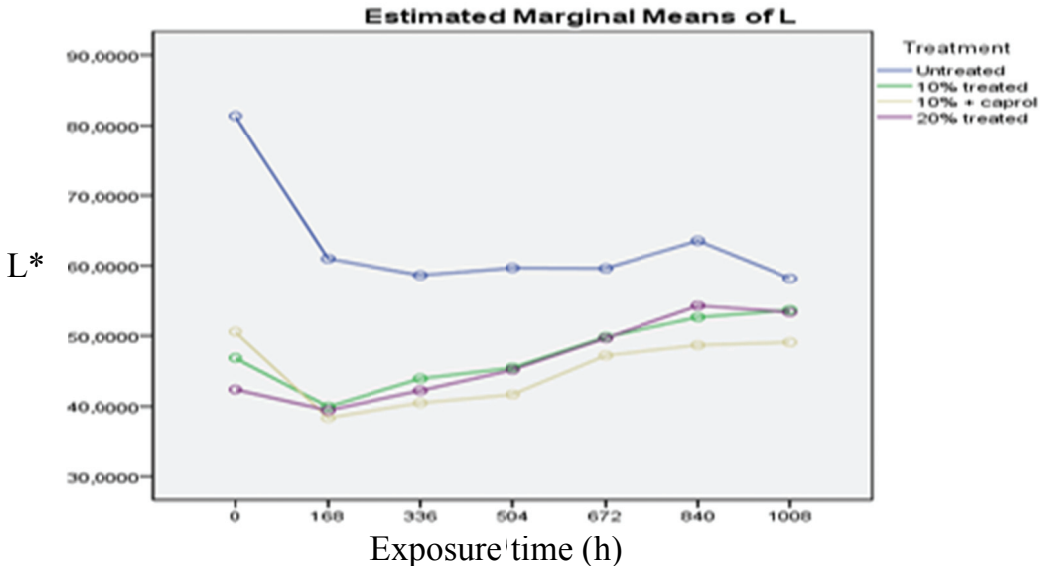

Figure 3. Trend of the color coordinate L* (lightness) during artificial weathering test.

After 168 hours exposure, the color turns to darker and become very similar to that of the classic formulations. Then the greying begins and it can be observed that the caprolactam-modified samples converge also to the grey level of untreated wood but more slowly than the samples treated with the classic formulations.

These observations suggest that the polymer of tannin containing caprolactam resists better the weathering than the classic formulations even if the surface degradation process still occurs.

4 CONCLUSIONS

The caprolactam-added tannin-based advanced formulations have shown a significant improvement for all the parameter considered in this study. The new polymer of tannin and caprolactam allows to produce wooden samples with:

higher dimensional stability due to a higher polymerization;

lower release of unreacted tannin during leaching;

high biocidal properties;

Increased resistance against artificial weathering.

Even if the weathering degradation still considerably stress the polymer structure, these new wood preservative show enhanced properties which can be considered highly interesting for the setting up of natural-based sustainable wood preservatives.

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SANDWICH PANELS WITH 100% NATURAL TANNIN FURANIC FOAM CORE

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Key words: Core foams, Lightweight boards, natural resources, Tannin polymer

ABSTRACT

Tannin foams are innovative materials obtained exploiting the capacity of tannin for undergoing poly-addition reactions and that of furfuryl alcohol to polymerize under acid catalysis. These materials are known for many years, but only recently they have been systematically studied and many of its properties have been evaluated.

The first generation of the tannin foams have shown good insulation properties and very significant resistance against fire but unfortunately they were produced with around 5% of formaldehyde needed for keeping the network stable and variable amounts of diethyl ether for allowing the blowing at room temperature. These two drawbacks have hindered the development of the foam for industrialization because formaldehyde is almost completely forbidden for inner application while diethyl ether is an easy-burning VOC.

In this study an important upgrading of the original formulation is presented. The synthesis of 100% natural formulation only catalyzed by heat is described. Due to this innovative approach, the toxic chemicals have been replaced with other bio-based resources so that the interest of the insulation industry is arisen again.

In particular, these second generation of foams have confirmed many of the properties of the first generation one with the significant advantages of being easily exploited as core material for many a wide range lightweight sandwich panels. The versatility of the new tannin-furanic formulations allows to produce composite materials with a broad set of decking materials according to performance required for the specific final application.

1 INTRODUCTION

In very recent years, an innovative material has taken the attention of many scientists all over Europe: Tannin-based foams. This material initially studied from the group of Prof. Pizzi (Meikleham & Pizzi 1994) has started a deeper investigation line in the final years of the last decade (2009a,b). The results obtained in the first years of investigation were extremely interesting for the industrial applications. This material, indeed, presented a highly natural skeletal structure combined with good physical properties and therefore a great amount of research work in this direction was and is still done. For instance, Lacoste et al. were able to produce foams out of pine tannin (2013), Li et al. prepared elastic foams (2013) while Basso et al. (2011) developed a method to produce room-temperature tannin-foams increasing the amount of furanic derivative and consequently the energy of the polymerization process. In the same years the tannin foams were also investigatedfocusing in the more applicative purpose of producing lightweight panels based on formaldehyde-free tannin foams. Various formulations of formaldehyde-free tannin foams were developed for insulation purposes (Link et al. 2011, Kolbitsch et al. 2012, Tondi et al. 2014).

A general review of these works is presented in this paper.

2 EXPERIMENTAL

2.1 Materials

The formaldehyde-free tannin foams were produced with tannin powder and furanic derivatives as main constituent of the skeleton. The tannin used was condensed ones and in particular Mimosa (Acacia Mearnsii) and Quebracho (Schinopsis Balancae) were the industrial extracts effectively used. Furfuryl alcohol was the co-reactant more suited. Various blowing agents (e.g. Diethylether, Pentane, Ethanol); catalysts (e.g. p-toluensulphonic acid and sulphuric acid); additives (e.g. plastifiers, extenders) were also used.

2.2 Methods

The tannin foams were produced discharging the blowing-polymerizing mixture already added of the catalyst in boxes made of different decking materials. The system then underwent energy sources such as a hot-press, microwaves and infrared radiation. The co-polymer between tannin oligomers and furanic moieties foamed (due to the evaporation of the blowing agent) and hardened (thanks to the increasing of cross-linkages between the co-monomers of the lightweight structure.

The foamed polymer solidifies in short time and then the filled box can be cut and stabilized at 20°C and 65% RH before characterization of the material.

2.3 Testing

Many tests have been done so that this new material could be considered for a broad range of applications. The way in which the tests were realized are reported in our previous research paper as well as in the Master theses of Kolbitsch and Link (2013). The tests discussed in this paper are the following: Density, homogeneity, mechanical resistance and thermal conductivity.

3 RESULTS AND DISCUSSION

3.1 Decking materials

During the process of synthesis of tannin-furanic foams, a great number of cross-linkage occurs. This material, indeed, can easily react with wood-based substrates but also with organic plastics and therefore the connections between the foam and the molds ware always established without any further adhesive.

In table 1 several decking materials were considered in order to produce a batch of composite material with different application domains.

Decking the foams with classic engineered wood allows to produce suggestive composite materials with properties which are strongly dependent on that of the decking. The presence of the tannin-furanic foam impart to the sandwich materials mainly: (i) lighter weight; (ii) improved fire properties; (iii) improved thermal resistance. The composite of table 1 become then interesting for wall and door thermal insulation; lightweight furniture production and semi-structural applications.

In particular the sandwich panels with HDF present very good stability problem and a homogeneous development of the foams which will also warranty more reliable and repeatable results. Another interesting panel is that produced with recycled paper because of the low cost and the

improved handling of the brittle foam. The latter could be the tannin-furanic composite with more interesting perspective because lighter, highly fire resistant and easy to handle. When the tannin-foams in the core are produced with higher density, also the composites with laminate become immediately very interesting because of combining the good optical and surface properties of the laminate with the lightness and mechanical resistance of the core layer.

Table 1. Possible applications of tannin-furanic foam composites with different decking materials

Decking material	Density (kg/m³)	Thickness (mm)	Costs (€/m²)	Application for the foamed composites
HDF	850	4	2,75	Core material for indoor insulation. Average mechanical properties.
Particleboards	650	12	6,3	Insulation and mild structural applications. With laminate also in furniture and doors.
OSB	650	14	7,8	Insulation and mild structural applications. Higher surface properties.
Plywood	680	15	22,8	Material for application requiring good mechanical properties. Furniture, doors.
Laminate	1400	0,5	10,4	Material for application requiring good surface, fire resistant and aesthetical properties.
High quality paper	20	0,05	0,06	Core for insulation walls. Light and cheap but easy to transport. Higher surface properties.
Recycled paper	15	0,05	0,02	Core for insulation walls. Light and cheap but easy to transport. Lower surface properties.

In Figure 1 are shown some example of tannin-furanic based composite materials.

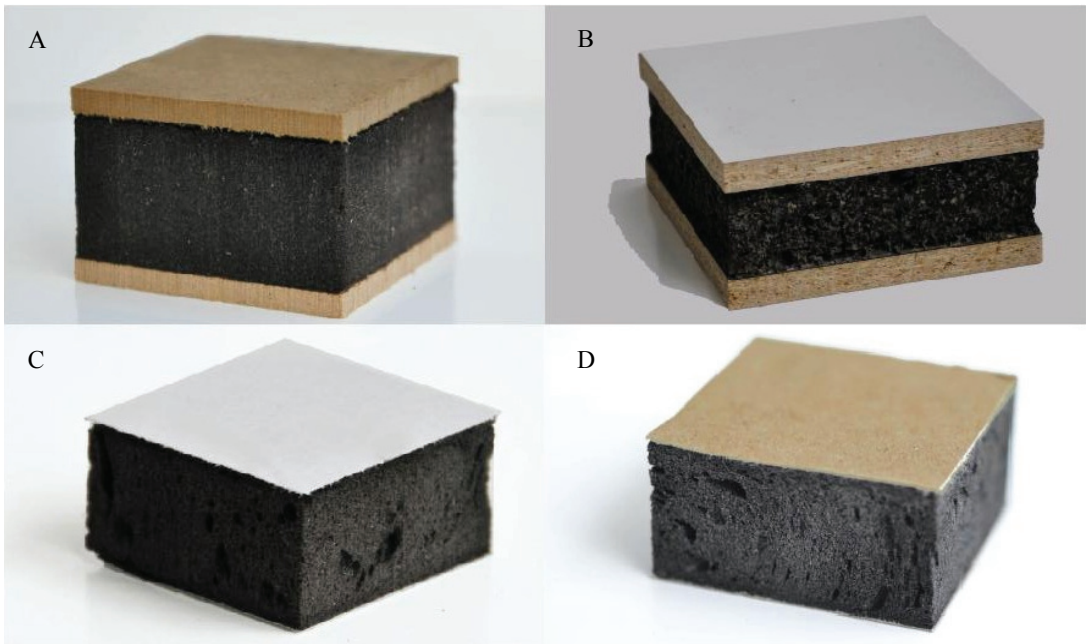

Figure 1. Example of tannin-based lightweight panels with different deckings: a) HDF, b) laminated particleboards, c) paper and d) recycling paper.

3.2 Specific formulations

Different amount of chemicals and specific additives have brought to various interesting results that are resumed in table 2.

The background colors of the tabs show positive and negative features for a foam. Ideally a foam extremely light, high homogeneous, highly mechanical resistant and with low thermal conductivity would be the perfect material for most of the applications. The different formulations show positive and negative trend according to properties considered. For example the foam containing more furfuryl alcohol are lighter and more homogeneous, but mechanically and thermally less performing than the standard formulations. The foam blown with ethanol instead of diethyl ether are mechanically more resistant, but heavier and less thermally insulating. The effect of loads carries to denser foams with increased mechanical properties. It has to be noticed that there is a direct proportionality between density and mechanical resistance that was reported in the following equation:

$$M.R. = 0{,}0018\,d - 0{,}0183 \quad (1)$$

Where M.R. is the mechanical resistance to compression and d is the density of the foam. The R^2= 0,86 confirm the acceptable proportionality.

Table 2. Features modification occurring by blowing different tannin-foams formulations.

Formulation modification	Density	Homogeneity	Mechanical	Thermal
Standard	100 kg/m³	Good	0,15 N/m²	42 mW/m.K
Co-monomer: Furfuryl Alcohol (60% more)	Lower	Higher	Lower	Higher
Blow. Agent: Ethanol	Higher	Lower	Higher	Higher
Load: Wood chips (f=1-2mm)	Higher	Similar	Higher	Higher
Load: Foam powder	Higher	Similar	Higher	Similar
Additive: Black liquor	Higher	Higher	Higher	Similar
Additive: PolyVinylAlcohol	Similar	Higher	Higher	-

The presence of additives such as PVA increases the mechanical properties without affecting too much the density. The presence of already structured macromolecules such as that of lignin and PVA allows the production of foams with improved homogeneity because of the increased original viscosity of the blowing mixture and therefore a better distribution of the blowing vapors. This improved homogeneity carries to enhanced mechanical properties without affecting the thermal conductivity.

3.3 Production methods

The formaldehyde-free tannin foams can be produced with external heating source. Four methods have been developed in the last years: The hot-press, the ventilated oven, the microwaves and the infrared heater. In table 3 the summary of the results is presented.

Table 3. Synthesis of tannin foams with different energy sources.

Heating system	Properties obtained
Hot-press	Homogeneous foams, high repeatability, low orthotropicity
Ventilated oven	Lower homogeneity, faster surface hardening, limited repeatability
Microwaves	Fast process, bigger cells, inverted orthotropicity
Infrared heater	Fast process, faster surface hardening, good homogeneity

According to the different applications all the processes can be applied to develop tailored tannin foams. The hot-press system ensures a better heat distribution that lead to highly homogeneous foams but it requires longer hardening time. The process with ventilated oven and Infrared heated tends to produce foams with faster surface hardening which can lead to lower repeatability. However in case of infrared heater the blowing/curing process occurs in a limited time. Sensibly different is the production of the tannin foams with microwaves because in this case the process occurs in a much

shorter time and producing much bigger cells oriented according to the wave. This process is also particularly interesting in case of industrial synthesis of the material.

According to the graphic of Ashby (1988) the tannin foams in general respect the relation between density and modulus of elasticity of classic foams and in particular it covers a range of very light foams with limited mechanical resistance. The combination of the tannin-foam as core material with different decking improves strongly the mechanical and surface properties carrying to a highly interesting engineered material. The formaldehyde-free tannin-based lightweight panels.

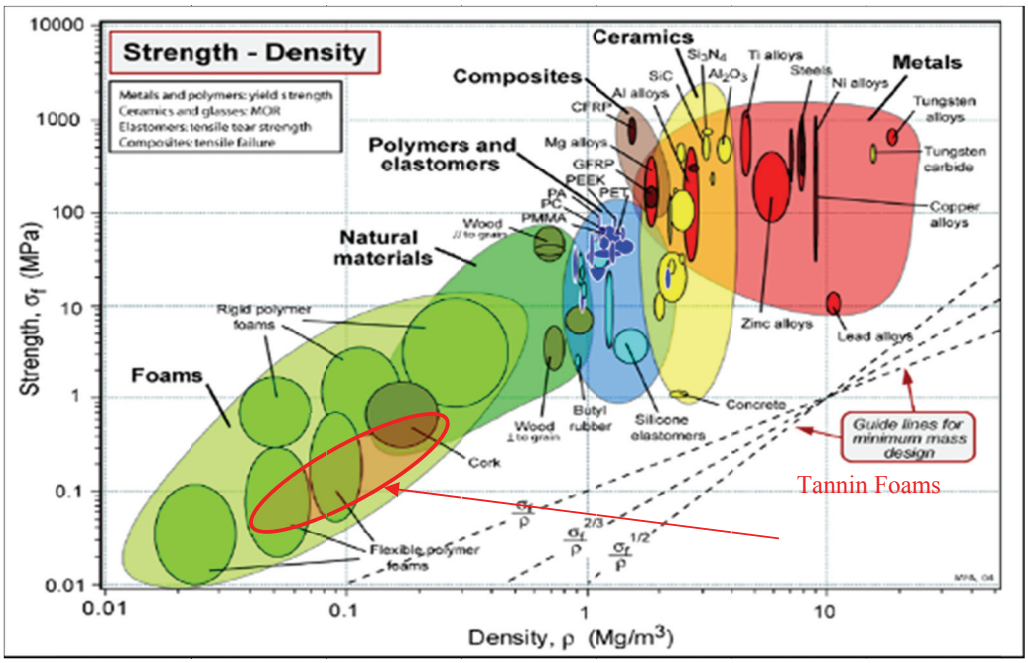

Figure 2. Diagram of Ashby included with the tannin foam material (in red). (Ashby, 1988)

4 CONCLUSIONS

Sandwich panels with formaldehyde-free tannin-furanic foams as core layer were developed. The self-gluing composite material present improved mechanical and surface properties as soon as the decking is applied. These composite materials can be tailored in order to be suitable for a wide range of applications: from insulation material for indoor walls up to structural component for doors and furniture. The great versatility in terms of formulation and production process allows to consider various industrial solutions for the processing of this bio-based innovative material.

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COMPARISON BETWEEN HB AND HDF MADE FROM WASTE LEATHER

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Key words: Wood – leather composites, wet process, Wet White, Wet Blue

ABSTRACT

Due to the ever-increasing scarcity of resources and raw materials in the wood panels industry, it is imperative to look for suitable alternatives to the established resources. Therefore a combination of the traditionally used and newly explored sources may reveal highly innovative ways.

The goal of this study is to provide an insight into the behaviour a wood-leather-composite material and its possible new applications.

Wet White (WW) and Wet Blue (WB) leather particles were used for this analysis and mixed with the wood fibres of spruce and beech. Wood – leather panels' combinations with 0%, 15%, 25%, 35%, and 50% leather amount were produced in two different production processes (wet and dry discontinuous process). To evaluate how the differences of the two production processes influence the mechanical properties of the panels, investigations concerning the internal bond (IB), bending strength (MOR) and modulus of elasticity (MOE) were conducted.

The leather content of the panels and the production process affected the results of the material properties. In the case of urea-formaldehyde (UF) bonded high-density fibreboards (4,5 mm, 900 kg/m^3) the results show a non-linear improvement of the IBs with increasing leather content, whereas MOE and MOR decrease. The hardboards, (3,5 mm, 900 kg/m^3) which were produced in the wet process with 1% phenol formaldehyde resin, show a similar behaviour in MOE and MOR to the HDF panels, but differ in tensile strength. The results give an overview how the leather amount infuences the board properties of two different panel types. Furthermore the here published results enable a convincing comparison between the mechanical properties of the two wood-leather composite board types.

1 INTRODUCTION

An analysis of the European wood market shows, that raising the netto imports of round wood can only increase the production of wood products (Barbu et al., 2014). However, untapped timber reserves such as bark need to be mobilized from the Austrian Forest (Schwarzbauer, 2005, Kain, 2012). Based on the current market situation, both options seem to be quite unlikely to reach. The only remaining possibility is to develop new sources of raw materials or to increase the efficiency of the existing industrial use of wood (Barbu et al., 2014).

According to the current market situation and the growing interests in Europe to obtain energy by burning biomass (Mantau et al., 2010), investigations in nearly every section of new material resources have been made through the passed years. Many of these studies set their focus on the up- or recycling of by-products, to find possible supplements to wood fibers or particles for wood based panel production (Deppe et al., 1996). Therefore, some research have been made to incorporate lingo-cellulosic materials such as wheat and reed (Han et al., 2001; Halvarsson et al., 2009), industrial hemp fibres (Crowley, 2001), bamboo and rice straw (Hiziroglu et al., 2008) or more exotic materials as coconut fibres (Van Dam et al., 2004) and stream exploded banana bunch fibres (Quintana et al., 2009) in fibre- and particleboards. A forward-looking way of reusing by-products seems to be an upcycling of waste materials, which occur in the meat production, as chicken feathers and leather shavings (Parkinson, 1998, LMC International, 2007). Most of these by-products are currently recovered thermally or getting landfilled in depositories in Central Europe (Kangaraj et al., 2006, Schröter, 1995). Investigations by Ostrowski (2012) and Grünewald (2012) describe the mechanical and physical influence of leather shavings in medium density fibreboards (MDF) and in insulation mats. Furthermore, Grünewald (2012) presented results according to the distribution of wood fibres and leather particles in panels and determine the structure with a Raman spectroscopy. Wieland et al. (2012) and Stöckl (2013) determined an increasing fire resistance with increasing leather-amount in MDF panles. Rindler et al. (2014) described the mechanical properties of high-density fibreboards (HDF) made from waste leather and wood, which is issued by Gerald Lackinger (2009). The corpus of published research, which dealed with fibrematerial, concerned about the usage of new materials produced under laboratory conditions using a dry discontinous process. As the raw material leather incurres with a moisture content >45% out of the leather production process, a different way of fibreboard production could be more suitable for this material.

This work deals with the comparison between the influence of the leather amount to the mechanical properties of hard-density fibreboards made from wood fibres and leathershavings in a wet process (HB) and high-density fibreboards produced in a dry process (HDF). Investigations concerning the MOE, MOR and IB properties of both boardtypes have been made to determine the behaviour of leather in those processtypes.

2 MATERIAL AND METHODS

2.1 USED MATERIAL

The leather material for the presented tests was specified and provided by Gerald Lackinger Consulting (Salzburg, Austria). The leather particles accrued during the thickness shaving process of cattlehides preparation.

For this investigation two different leather types, which are defined as Wet White (WW) and Wet Blue (WB), were used. The tanning process of Wet White uses a synthetically non-chromatic tanning, whereas in Wet Blue chromium is connected with the proteins of the hides in a stable bond. Both leather types were pre-fractionated (particles < 2 mm) at a moisture content of > 45%.

For the dry process the leather particles got dried to average moisture content of 8 ± 1%.

The unglued wood fibres (Picea Abies) for the dry process where provided by MDF Hallein GmbH with a moisture content of 8 ± 1%. For the wet process wood fibres (⅓ bark residuals and ⅔ Fagus Sylvatica fibers) with moisture content of 45% were used.

2.2 PRODUCTION OF THE PANEL

Fibreboards with dimensions 450 x 450 x 4.5 mm (dry process) and 450 x 450 x 3.5 mm (wet process) were produced under laboratory conditions. Both panel types were manufactured with different leather types Wet White (WW) and Wet Blue (WB) and varying leather-fibre ratios (leater amount in the board: 0%, 15%, 25%, 35%, and 50%). To obtain a significant comparison, both board types were manufactured with the same average density of 900 kg/m^3.

For the dry process an amount of 10% urea-formaldehyde resin (UF) was used. The adhesion occurred in a plowshare blender with a two-substance nozzle upright section to obtain a homogenious glue application. For this process a nozzle with a hole diameter of 2.3 mm and a pneumatic pressure of 2 bar was used. After the adhesion of the fibres a conventional vacuum cleaner "Bosch GAS 50-M" was used to soak the glued material of the blender and thereby counteract the agglomeration behaviour of the glued fibres. Further the fibres were distributed manually in a frame and finally pressed in a Hoefer HLPO 280 automated hot press at 80°C with a pressing factor of 1 min/mm.

For the wet processed leather-fibreboards beech wood fibres (u = 45%) and leather particles, Wet Blue (u = 69%) and Wet White (u = 45%), were used. The material got mixed in 10 l white water from the fiberboard production process. During the blending process 1% of phenol formaldehyde resin was added. The fibre-leather liquid got filled in a dewatering press with an operating pressure of 100 bar. Subsequently the pre-compressed fibre mat was pressed for 4 min and 120 bar at a temperature of 200°C in a Bürkle hotpress.

In total 10 panels were produced (Table 1).

Table 1: Total overview of the pressed boards

Leather/Fibre Ratio	Leather Type	Production Process
0/100	WW	Dry
		Wet
	WB	Dry
		Wet
15/85	WW	Dry
		Wet
	WB	Dry
		Wet
25/75	WW	Dry
		Wet
	WB	Dry
		Wet
35/65	WW	Dry
		Wet
	WB	Dry
		Wet
50/50	WW	Dry
		Wet
	WB	Dry
		Wet

2.3 SPECIMEN PREPERATION

After the pressing process the samples from the wet and the dry production process, were conditioned in a standard climate at 20 ± 1°C and 65 ± 1% relative humidity, until the equilibrium moisture was reached.

To eliminate the density variation in the edge region, the panels were trimmed to a dimension of 400 x 400 mm. Subsequently the test pieces were cut out of the boards according to the EN 326-1:2005, to ensure randomised test results.

The testing samples for the modulus of rupture (MOR) and modulus of elasticity (MOE) were prepared by the following equation (1), which is in accordance to the OENORM EN 310:2005.

$$l = 20t + 50 \quad (1)$$

l…length, t…thickness

Based on this formula, the samples for MOE and MOR test were 50 x 140 mm for the dry process panels and 50 x 120 mm for the wet process.

To analyse the internal bond (IB) the test was conducted according to OENORM EN 319:2005 and therefore the testing samples were prepared in the dimensions 50 x 50 mm. To counteract a weak performance of the adviced hot melt glue (EN 319) in the glue layer during the IB testing, PUR (Kleiberit 501 PUR-Leim D4) resin was used to fix the HB and HDF samples to the testing crosshead.

2.4 MECHANICAL TESTS

The determination of the mechanical properties of the panels was performed on a Zwick Roell Z 250 universal testing machine. For all mechanical tests, a total of five samples (n=5) of each combination and process were tested.

2.4.1 Internal Bond

The IB strength was determined according to the OENORM EN 319:2005. The force was applied with a constant speed to achieve a breaking of the specimen within 60 ± 30 seconds.

2.4.2 Modulus of Rupture and Modulus of Elasticity

The MOR and MOE were evaluated according to OENORM EN 310:2005. The samples were tested for each of the two similar panels of each combination. The specimens were tested with a continuous crosshead-speed to achieve a breaking within 60 ± 30 seconds. The MOE was obtained from the linear values between 10 to 40% of the maximal load.

2.5 DATA EVALUATION

To evaluate the data of the mechanical tests, an Analysis of Variance (ANOVA) with the density as a covariate was used to determine the explanatory effect of each parameter on the model. To determine the statistical influence of the leather amount, the leather type and the process type on the tested data, a one-way Welch-ANOVA was used to compensate the unequal variance. The general level of significance was 0.05.

3 RESULTS AND DISCUSSION

3.1 INFLUENCING FACTORS ON THE MECHANICAL PROPERTIES

The following tables give an overview of the obtained values of the mechanical properties (IB, MOE and MOR) and the measured density. Table 2 displays the calculated averages of the HDF boards and Tabel 3 shows the means of the HB boards with WW and WB particles. The upper value in the box represents the mean of the tested panel and the data in brace below their standard deviation (sd). The number of specimens of IB, MOE, and MOR was 5 and the number of density measurements 10.

Table 2: Estimated mean and standard deviation (sd) of the mechanical properties of WW and WB HDF-boards

Type [%]	Density [g/cm³]	IB [N/mm²]	MOE [N/mm²]	MOR [N/mm²]
	n=10	n=5	n=5	n=5
Wood HDF	0.80 (.04)	0.92 (.19)	2083.3 (110.1)	29.8 (3.8)
WW15	0.92 (.08)	1.23 (.46)	2697.8 (358.0)	37.1 (5.1)
WW25	0.97 (.07)	1.52 (.19)	3230.2 (806.1)	47.8 (9.9)
WW35	0.97 (.04)	1.39 (.14)	2599.5 (222.5)	35.2 (4.2)
WW50	0.88 (.10)	0.98 (.34)	1902.7 (414.4)	25.3 (6.9)
Wood HDF	0.80 (.04)	0.92 (.19)	2083.3 (110.1)	29.8 (3.8)
WB15	0.95 (.06)	0.74 (.20)	2988.5 (384.9)	38.0 (5.1)
WB25	0.87 (.04)	0.85 (.18)	2508.0 (163.5)	33.4 (3.4)
WB35	0.92 (.05)	1.04 (.11)	2361.2 (558.4)	31.8 (9.1)
WB50	0.92 (.07)	1.65 (.16)	1863.2 (260.3)	28.5 (4.4)

As clearly visible in table 2 all WW and WB blending ratios of HDF panels obtained similar density means. According to results of the mechanical properties it is to say, that the WW HDF panels reached overall higher means compared to the WB HDF panels with the same leather ratio. Only WB 50 achieved higher results in case of IB compared to WW 50%. In case of MOE and MOR the panels with 15% Wet Blue amount show better properties than the WW 15% panel.

Table 3: Estimated mean and standard deviation (sd) of the mechanical properties of WW and WB HB-boards

Type [%]	Density [g/cm^3]	IB [N/mm^2]	MOE [N/mm^2]	MOR [N/mm^2]
	n=10	n=5	n=5	n=5
Wood HB	0.97 (.02)	0.27 (.07)	3232.2 (186.4)	37.5 (2.3)
WW 15	0.96 (.03)	0.19 (.17)	2756.6 (223.5)	32.8 (3.4)
WW 25	0.98 (.03)	0.22 (.17)	2737.7 (134.3)	33.1 (2.0)
WW 35	0.98 (.04)	0.26 (.23)	2557.7 (223.4)	33.7 (3.3)
WW 50	0.96 (.02)	0.33 (.16)	1799.8 (268.7)	25.5 (3.6)
Wood HB	0.97 (.02)	0.27 (.07)	3232.2 (186.4)	37.5 (2.3)
WB 15	0.95 (.03)	0.22 (.24)	2772.2 (290.7)	31.1 (5.4)
WB 25	0.95 (.01)	0.33 (.05)	2436.9 (84.8)	31.3 (1.8)
WB 35	0.94 (.01)	0.33 (.06)	2366.2 (100.6)	28.2 (2.4)
WB 50	0.95 (.03)	0.22 (.11)	1919.9 (140.5)	24.1 (2.3)

In table 3 it can be seen, that all WW and WB blending ratios of HB boards reached similar density means. According to results of the mechanical properties it is to say, that the WB-HB panels achieved overall higher means compared to the WW-HB boards with the same leather ratio (expect WB 50%). In case of MOE and MOR there is no explicit difference between Wet White and Wet Blue noticeable.

3.1.1 Internal Bond

Figure 1 and 2 show the medians (line in the box) and the interquartile range (whole box) of the IB results, which indicate a relation between transverse tensile strength and leather content in the panels. The range of the wiskas shows 95% of the measured values. Figure 1 indicates the test results of the fibreboards, which were produced, in the dry process and figure 2 displays the wet process.

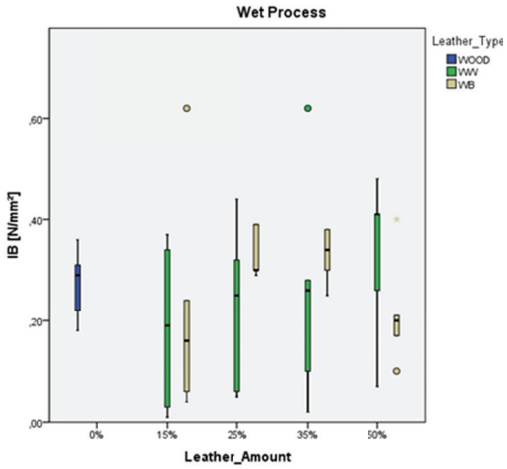

Figure 1: IB of fibreboards from dry process

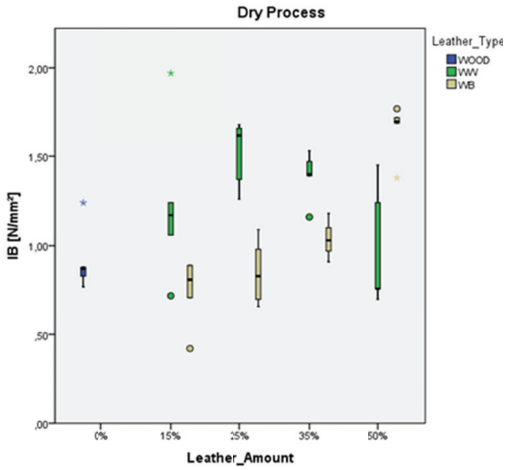

Figure 2: IB of fibreboards from wet process

Allover it is to say that the IB values are increasing with rising leather content in the dry production process. Only WB 15%, WB 25% and WW 50% show a lower value than the pure wood panel. As visible in both figures (1,2) it can be seen, that the medians of the boards from the dry process reached IB between 0.7 and 1.7 N/mm^2 and the wet produced panels only between 0.15 to 0.5 N/mm^2. However, the medians of WB 50 dry process and WW 50% wet process represent the strongest tensile strength properties. The analysis of variance (p=0.05) shows that the amount as well as the type of leather and the process type, highly significant, influences the IB.

3.1.2 Modulus of Elasticity

Figure 3 and 4 show the medians and standard deviations of the MOE results. Figure 3 indicates the dry process, while figure 4 displays the wet process.

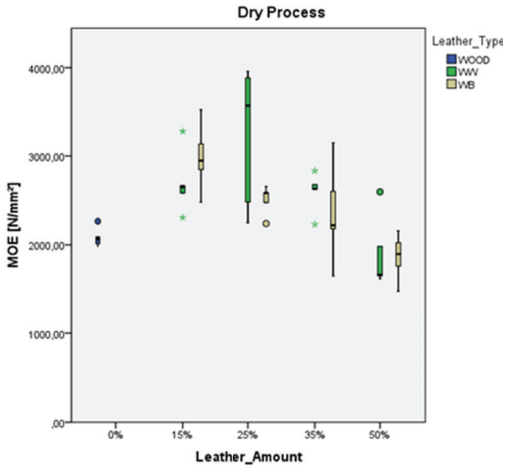

Figure 3: MOE of fibreboards from dry production process

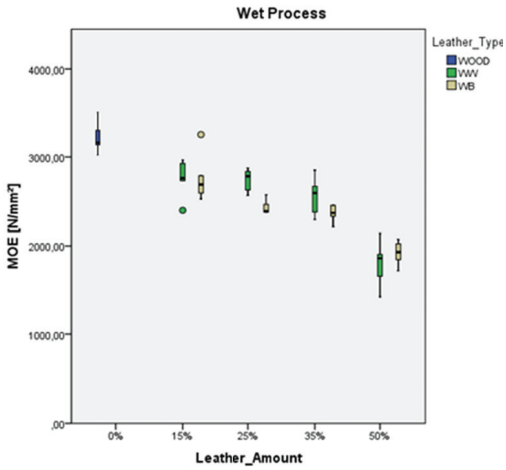

Figure 4: MOE of fibreboards from wet production process

Interestingly both leather types (WW, WB) show a lower MOE value with a leather amount of 50% compared to the pure wood panels in the dry process. All other blending ratios of the dry process performed with better MOE medians. According to the wet process it is to say, that decrease of elasticity appears in a more continous way than the behaviour of the dry process.

The comparison of the medians show, that no median of the blending ratios could achieve higher results than the pure wood panel. Comparing the influence of the leather amount between dry and wet process, it is to say, that the medians of MOE are in both cases between 1800 and 3500 N/mm^2. According to the analysis of variance (p=0.05) it is to say that the amount of leather is highly significant for both fibreboards, but the type of leather shows only a significant influence on HDF and HB. Further the type of process is highly significant.

3.1.3 Modulus of Rupture

Figure 5 and 6 show the medians and standard deviations of the MOR results for the dry and the wet production process.

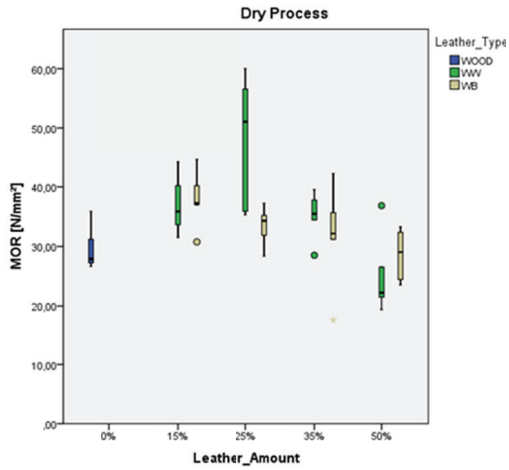

Figure 5: MOR of fibreboards from dry production process

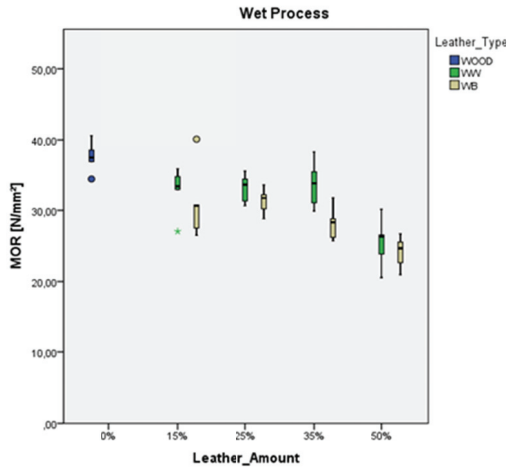

Figure 6: MOR of fibreboards from wet production process

The results for the MOR are similar to the MOE values. Also here the same bahaviour of material with increasing leather content is visible. While in the dry process only the 50% leather blendings obtain lower values than the pure wood panel, in the wet process the pure wood board performed the best. The analysis of variance (p=0.05) shows that the amount as well as the type of leather and the process type influences the IB highly significant.

4 CONCLUSIONS

The results for the internal bond (IB) showed interesting insights into the importance of the process. The hardboards showed a weaker performance than the high-density fibreboards. A reason for this could be the, in 2.3 mentioned, problem with the weaker screen side of the hardboards. Although the used PUR adhesive performed well, the sieve surface obviously was too unregular to perform better. The second reason could be the low adhesion amount in the hardboards. It seems that the amount of 1% PF is to low with rising leather content, as pure leatherparticles do not possess the attribute of a self-bonding character.

The values of the modulus of rupture and elasticity investigations were highly satisfying and showed good performance for both pressing types. A significant difference could be noticed between

the standard deviations. The results in the dry process showed a significantly higher standard deviation than the wet process. This reveals to the forming of the fibremat before the pressing. The forming occurs much more homogenious in the wet process than in the dry laboratory process.

From the mechanical point of view it is definitely worth to produce hardboards with a leather amount up to 50%. The huge advantage of the wet process is economically by leaving a prior drying process of the raw material and using less adhesive.

To the opinion of the author further research concerning the physical properties of leather hardboards should be made to obtain a better understanding of probable future applications.

5 ACKNOWLEDGEMENT

The authors gratefully acknowledge for the support of the Austrian Research Promotion Agency (FFG) under grant no.836988.

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BAMBOO - A FUNCTIONALLY GRADED COMPOSITE MATERIAL

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Key words: Bamboo, Composite material, Microstructure, Fiber volume fraction

ABSTRACT

Bamboo is one of the fastest growing plants and it is abundantly growing in most of the tropical regions. It is considered as a composite material because its structure is composed of long and aligned cellulose fibers embedded in a lignin matrix. Moreover, the bamboo culm shows a composite type of structure on both of the macroscopic and microscopic levels. Additionally, bamboo properties vary significantly along the culm length and culm wall thickness. According to these characteristics, the bamboo culm is a functionally graded natural composite material.

This paper deals with the characterization of the micro-structure of bamboo culms (Dendrocalamus asper) by Digital Image Analysis. The variation of the fiber volume fraction and microstructure across transversal section of the bamboo culm were studied. Furthermore, bamboo properties which influence the specific gravity and bending strength were analyzed along the culm length.

The results show that bamboo culms in transverse section composed of numerous fibers embedded in the parenchymatous ground tissue. The distribution of fiber shows a specific pattern within the culm, both horizontally and vertically. The parenchyma and conducting cells are more frequent in the inner one-third of the wall, while the percentage of fiber is higher in the outer part of the wall. In vertical direction, the fiber area percentage increases from bottom to top with the decreasing parenchyma content. The number of vascular bundles is high and the vascular bundles are dense in the outer part compared to the inner part. The vascular bundles near inner part are almost circular while they are more of elliptical shape near the outer part of the culm wall. The specific gravity of D. asper rangs from 0.58 to 0.71 while Modulus of Rupture (MOR) and Modulus of Elasticity (MOE) range from 132 to 181 MPa and from 9,465 to 15,115 MPa, respectively. These values are increasing from the bottom to the top of the culm. The results indicated a correlation between microstructure and properties of bamboo material. The specific gravity and bending strength increase linearly with the volume fraction of fiber.

From the microscopic point of view, bamboo is a functionally gradient material with a hierarchical structure. The decrease of fiber volume fraction from the outer to the inner part of the culm wall changes density and properties which have to be considered for bamboo. Moreover, the volume fraction of fiber increases from the bottom to the top of the culm. Subsequently, bamboo can distribute and resist the bending stress generated by wind load. These characteristics make bamboo a good substitute for raw material in construction if the variation of properties is considered in design of products. Bamboo being the fastest growing plant on earth opens a good opportunity for environmental friendly and sustainable resource supply.

1 INTRODUCTION

Bamboo is a fast growing monocotyledon which is mostly distributed in the tropical and subtropical regions. It is a major building material in many countries, particularly in Asia, Africa and South America, because of its strong characteristics, light weight and flexible properties. It can be used for almost all parts of houses, including posts, roofs, walls, floors, beams and trusses.Bamboo is natural lignocellulosic composite. Moreover, it is supposed to be the functionally gradient composite material because bamboo structure is composed of fibers (sclerenchyma tissues in vascular bundles) embedded in matrix (parenchymatous tissues which are thin walled cells around vascular bundles). The fibers are the main part determining the mechanical characteristics of bamboo while the parenchymatous tissues pass loads and take the role of a composite matrix. Then, bamboo's properties depend on its components such as the volume fraction and distribution of fibers and the interface properties of bamboo components (Shao et al. 2010). Several researches have established that the distribution of bamboo components along the horizontal and vertical direction shows a gradient. They reported that the amount of fibers decreases from outer to inner side of the bamboo culm. Furthermore, the size of the vascular bundle is large in the inner and middle layer but smaller and denser in the outer layer. Many studies have been published on the anatomical features of bamboo which directly affect the physical and mechanical properties. It is expected that these features may affect the final application of bamboo.

In this study, the microstructure of Dendrocalamus asper was characterized by Digital Image Analysis. The variations of microstructure and fiber volume fraction across transverse section of bamboo culm were studied. Additionally, the physical and mechanical properties of bamboo culm in relation to its anatomical characteristics were also evaluated to assess it possible use in furniture, building and general construction. Furthermore, bamboo properties which influence the specific gravity and bending strength were analyzed along the culm length. These knowledge will supplementary provide as a basic guide and also promote its use for commercial purposes.

2 MATERIALS AND METHODS

Bamboo culms with three years old were cut into each internode along their length. The specimens were randomly selected and cut into the specific size for the analysis of microstructure, physical and mechanical properties. All specimens were placed in a conditioning chamber, at a temperature of 20°C and relative air humidity of 65% until the moisture content reached 12%.

For microstructure characterization, bamboo culms were divided into three parts (bottom, middle and top) each of 6 m lengths. The samples polished up the surface in order to observe and photograph their cross sections in an optical microscope, as present in figure 1. The image of bamboo cross section was analyzed with Image Proplus software. The cross section of each sample was divided evenly into tree layers horizontally (outer, middle and inner zones) and the vascular bundle size and number were measured.

From the following internode, three specimens were randomly selected and cut into the specific size. The testing method for bending strength was performed in accordance with the ASTM D 143-94 standard. The MOR and MOE were calculated. Subsequently, the specimens were cut and determined for specific gravity, moisture content and fiber distribution. Image processing and analysis for fiber distribution determination was developed under a commercial software package Mathematica. The cross section of each sample was viewed under the optical microscope and the image was captured using a digital camera. The volume fraction of fiber was measured.

3 RESULTS AND DISCUSSION

3.1 Variation in vascular bundle concentration

The concentration of vascular bundles varies with different height and thickness, as present in table 1. The result suggests that the concentration of vascular bundles increase with increasing height of a culm. The vascular bundle concentration is the highest on the top part. The higher density of vascular bundles at the top part of bamboo culm has been explained by Grosser and Liese (1971) as a result of the decrease in culm wall thickness. The highest vascular bundle concentration was found on the top at the outer part of bamboo culm with 3 bundles/mm2, while the lowest vascular bundle concentration was found on the bottom at the inner part of bamboo culm with 0.5 bundles/mm2. This finding was also in agreement with Fujji (1985). He reported that 50-80% of the vascular bundles are located in the outer third of the wall, 10-35% in the middle, and only 10-20% in the inner third.

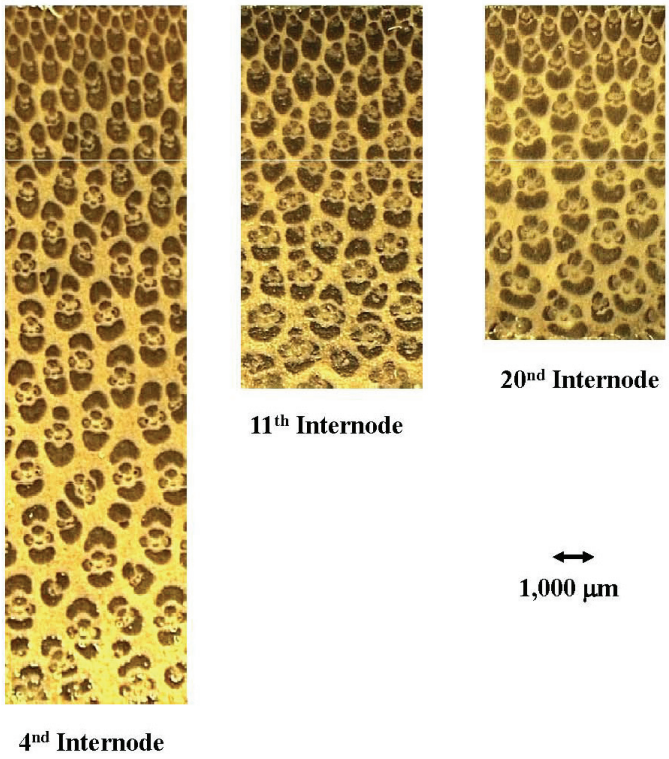


Figure 1. Changes in the microstructure at the 4nd, 11th and 20th internodes along the culm length in *Dendrocalamus asper*.

Table 1. The variation in vascular bundle characteristics with respect to the location of bamboo culm.

Height	Thickness	Vascular bundle concentration (n/mm^2)	Radial length (μm)	Tangential width (μm)	R/T ratio
Bottom	Inner	0.70 (0.10)[1]	1,221.56 (113.61)	947.33 (88.03)	1.31 (0.15)
	Middle	0.81 (0.17)	1,340.25 (124.93)	732.15 (50.56)	1.84 (0.15)
	Outer	2.19 (0.21)	957.54 (116.13)	490.57 (33.12)	1.97 (0.26)
Middle	Inner	0.81 (0.13)	1,166.25 (157.70)	933.52 (67.04)	1.26 (0.18)
	Middle	0.95 (0.13)	1,174.44 (166.22)	677.38 (48.39)	1.75 (0.26)
	Outer	2.54 (0.25)	847.23 (75.92)	460.75 (32.29)	1.86 (0.21)
Top	Inner	0.96 (0.07)	1,025.81 (138.28)	981.56 (60.05)	1.06 (0.19)
	Middle	1.02 (0.08)	1,115.56 (123.11)	650.22 (35.96)	1.73 (0.24)
	Outer	2.73 (0.18)	881.37 (69.20)	468.34 (32.33)	1.89 (0.15)

[1] Number in parenthesis is associated to standard deviation

3.2 Variation in vascular bundles size

The variation in vascular bundles size (i.e.; radial length, tangential width and radial/tangential ratio) with different height and thickness is present in table 1. The redial/tangential ration is termed of the ratio of radial length to tangential width of vascular bundles (Grosser and Liese 1971). This result suggests that the ratio significantly increases from the bottom to the top of culm and from the inner zone towards the outer zone. It indicates that the vascular bundles near inner part are almost circular while they are more of elliptical shape near the outer part of the culm wall. This finding is also in agreement with Liese (1985) and Wenyue et al. (1981). They reported that a vascular bundle is longest and smallest at the outer zone but shorter and bigger towards the inner zone. The variation in microscopic characteristic of bamboo directly affects the physical and mechanical properties. It is expected that these features may affect the bamboo processing and utilization.

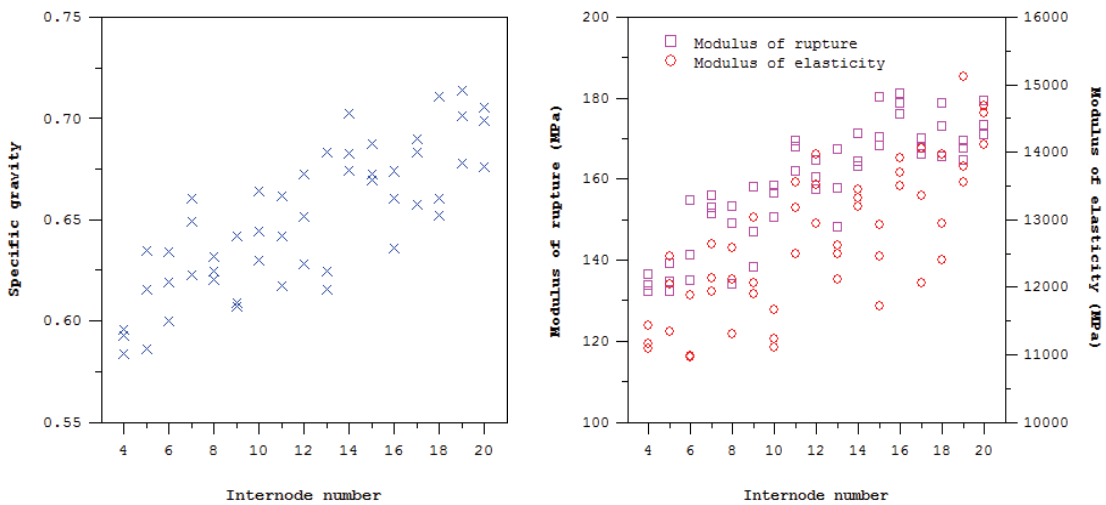
3.3 Variation in physical and mechanical properties

The specific gravity of D. asper at 12% MC is in the range of 0.58 to 0.71, as presented in Figure 2(a). The result indicates that the location along the bamboo culm is significant for the specific gravity value. The value slightly increases from the bottom to the top of the culm. The increasing of specific gravity with height is probably due to the increasing fiber density. The amount of these fibers strongly increases from the bottom to the top, as mentioned below.

Figure 2(b) shows the variation of MOR and MOE along the culm height. The values are in the range of 132 to 181 MPa and 9,465 to 15,115 MPa, respectively. The result demonstrates that MOR and MOE vary in the relation to culm height location. This result is confirmed by the findings of Abd. Latif et al. (1990 and 1994). They found that the mechanical properties of bamboos vary significantly with culm height. Furthermore, MOR and MOE are strongly related to specific gravity, as shown in figure 3. The linear regression equations and coefficient of determinations (R2) indicate the relationship between the two properties. This observation is consistent with Abd. Razak et al. (1995), Anwar et al. (2005) and Yu et al. (2008) who reported that the bamboo strengths depend on its specific gravity. It has been postulated that the bamboo specific gravity is closely related to the relative proportions of vascular bundles and ground tissues; especially as the proportion of thick-walled sclerenchyma cells within the culm.

3.4 Analysis of fiber distribution

Figure 4 shows the variation of the volume fraction of fibers along the culm length. The result shows that the volume fraction of fiber linearly increases from about 14% to about 32% from the bottom to the top part of the culm. Interestingly, the distribution of fiber correlates with the SG and bending properties generated within the culm at each height. It can be presented by the plotting graph and correlation value (R2) between fiber volume fraction against SG and bending properties presented in Figure 5(a) and 4(b), respectively.



(a) (b)

Figure 2. Variation of (a) specific gravity and (b) bending properties along the bamboo culm length.

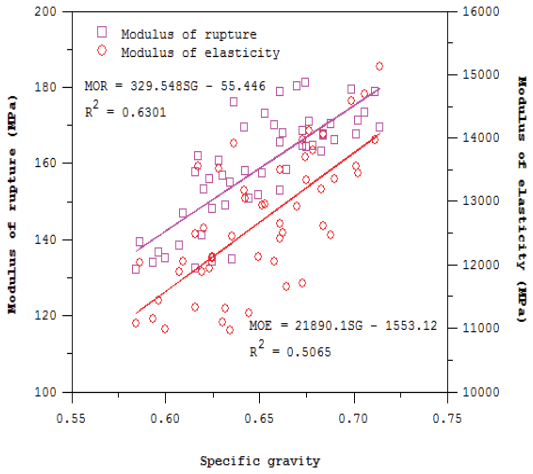


Figure 3. The relation between bending properties and specific gravity.

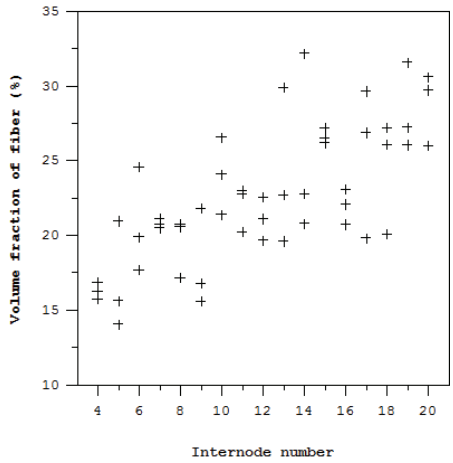


Figure 4. Variation of volume fraction of fibers along the bamboo culm length.

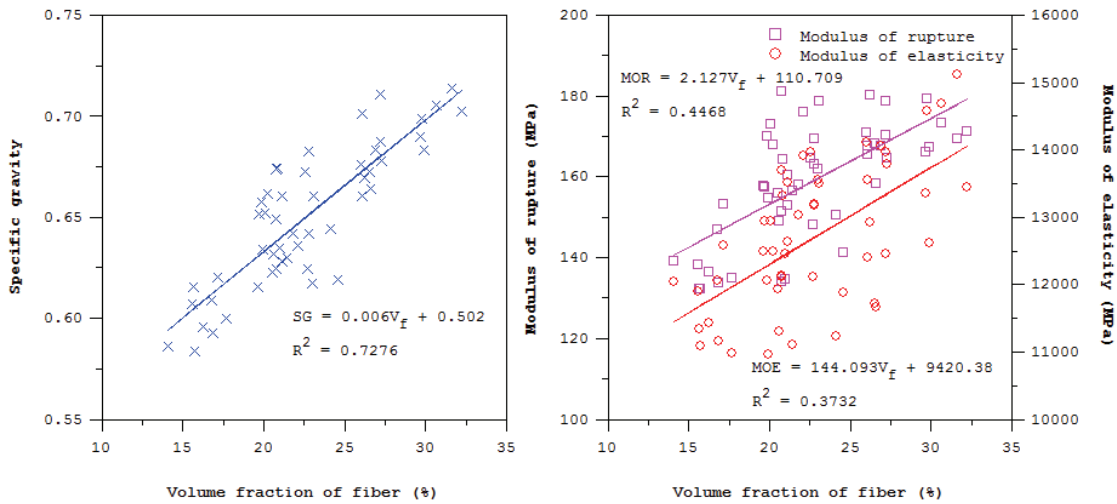


Figure 5. The relation between volume fraction of fibers and (a) specific gravity, (b) bending properties.

4 CONCLUSIONS

The variations of microstructure and properties along the culm length of *D. asper* have been analyzed. The following conclusions can be drawn from this part of the study:

1. The concentration of vascular bundles varies with different height and thickness. The concentration of vascular bundles increase with increasing height of a culm. The vascular bundle concentration is the highest on the top portion.
2. The vascular bundles size varies with different height and thickness. The vascular bundles near inner part are almost circular while they are more of elliptical shape near the outer part of the culm wall.
3. The specific gravity and bending properties vary with height location. The top part of the culm has a consistently higher SG, MOR and MOE than the bottom part.
4. The volume fraction of fiber linearly increases from the bottom to the top part of the culm.
5. The specific gravity and bending strength increase linearly with the volume fraction of fiber.

5 ACKNOWLEDGEMENT

The authors would like also to express our thanks to the Wood Science and Engineering Research Unit, Walailak University, Thailand for providing spaces and facilities for the experimental work.

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THERMAL INSULATION PANELS FROM CELLULOSIC FIBRES

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Key words: cellulosic fibres, insulation panel, thermal conductivity

ABSTRACT

*Recycling of paper is a relatively old but important process. During breakdown of recycled paper cellulosic fibres with different morphology (size) are generated. An important aspect that was considered was the impact of fibre size on thermal insulation. For the research cellulosic fibres where separated into three different fractions, from which panels with target density of 50 kg/m3 and thickness 80 mm were made. Thermal conductivity of panels was determined by steady-state technique at temperature differences of 10, 25 and 40°C. It was determined that size of fibres used has little impact on thermal conductivity. The lowest thermal conductivity was determined when boards were made from coarser fibres, while the highest was achieved when fines were used. Thermal conductivity was between 0.045 and 0.066 W/m*K. An increase in thermal conductivity with regard to temperature difference was determined.*

1 INTRODUCTION

The choice of insulation material is nowadays of great importance when smart building is aimed. Available insulation materials have a wide range from synthetic to natural, from foam to fibrous and panel types. Considering sustainability the obvious choice for insulation, the natural based materials are sustainable but request further developments. These are breathable (improving air quality), hygroscopic (regulating moisture), good insulation properties as well as high phase shift (Korjanic et al., 2011; Zach et al., 2013). One of the materials that is interesting for insulation purposes is also paper or the fibres obtained from recycled paper by re-pulping and/or shredding. The use of cellulose fibres as insulation started in 1970´s in USA, respectively 1980´s in Europe. According to Siddiquai (1989), Nicolajsen (2005), Vėjelis et al. (2006), Montažna gradnja Tadej Zimic s.p. (2013), Cellulose Insulation Manufacturers Association (2013) insulation materials made from cellulose fibres have good fire resistance, good sound insulation, due to sorption behaviour of fibres, better microclimate and low thermal conductivity. Usual application of cellulose fibres for insulation is by blowing these into the wall construction. So-called loose-fill insulation has a density between 20 and 70 kg/m3 (Nicolajsen, 2005; Vėjelis et al., 2006). Despite low thermal conductivity, application of cellulose fibres by means of blow-

ing has several disadvantages like settling and compaction hence slightly increase in thermal conductivity. One solution to overcome settling and compaction is the production of cellulose insulation in form of panels (Medved et al. 2013).

During production flakes/fibres of different size are generated. The fines are removed (Siddiqui, 1989) in small part also during blowing.

Aim of this paper is to present the impact of fibre size on thermal insulation of cellulosic based panels (matrisses).

2 MATERIALS AND METHODS

For the purpose of this experiment fibres made from recycled newspapers was used. The fibres were made in Slovenian company Montažna gradnja Tadej Zimic s.p. (figure 1).

Figure1. Cellulose fibers from recycled paper

The separation of fibres according to size were done by the means of sieve analysis where 300 g of cellulose fibres were shaken for 10 minute through the set of sieves. The set of sieves used is present in table 1

Table 1. Set of sieves used for sieve analysis and size classification

Sieve number	Sieve opening in mm	Size classification
9	6.0	
8	4.0	C (coarse)
7	2.0	
6	1.5	
5	1.27	B (middle size)
4	1.0	
3	0.6	
2	0.237	A (fines)
1	Bottom	

After the separation of flakes/fibres per size class (min. 2500 g) appropriate mass of flakes/fibres was put into blending machine, sprayed and mixed with MUF adhesive. Blending ratio was 15%. Blended flakes/fibres were formed per hand into a box (500×500 mm). Target density for panels was 50 kg/m3. This density was chosen due to the fact that in most cases when cellulose fibers are in use, the average density requested for blowing of fibers into a wall was set to 50 kg/m3. The thickness of panels was 80 mm. The mat was pressed at 190°C. Pressing time was 6 minutes. Pressing pressure was set to 2 N/mm2 but distance bars were also used. After pressing the panels were cooled at room temperature for 60 minutes and afterwards conditioned at 20°C and 65% relative humidity until constant mass was achieved. From the panel with dimensions 300×300 mm the samples were cutted out. Two panels per flake/fiber size were produced.

The determination of thermal conductivity was done in accordance to EN 12667 at temperature differences of 10, 25 and 40°C (table 2) on conditioned samples.

Table 2. Temperature set for determination of thermal conductivity

Set	Lower plate °C	Upper plate °C	Tdifference °C
1	2.50	17.50	10
2	17.50	32.50	25
3	32.50	47.50	40

Thermal conductivity determination was done by the Heat Flow Meter LM.305 (Stirolab, Sežana, Slovenia). The device is shown in figure 2

Figure 2. Device for the determination of thermal conductivity

3 RESULTS AND DISCUSSION

During breakdown of paper the highest share is presented by the coarser constituents (figure 3) that are more or less in shape of flakes (figure 4c), compared to middle class and fines for which constituents are more fibrous shape (figure 4b and 4a).

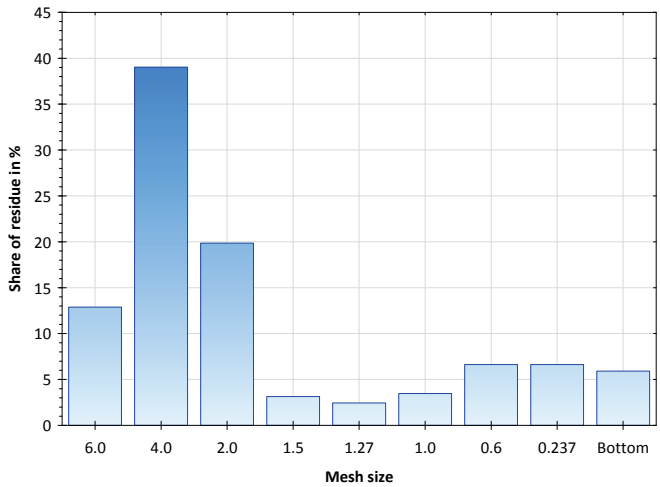

Figure 3. Results of sieve analysis

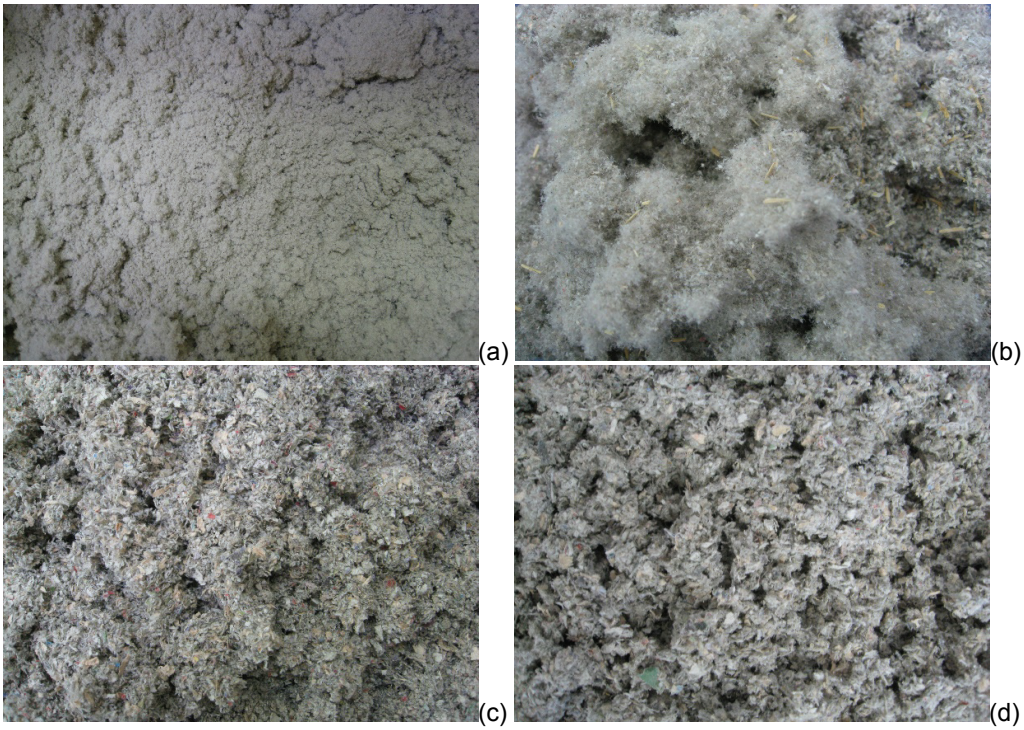

Figure 4. Mixed (d), coarse (c), middle size (b) and fine (a) constituents

Analyzing the panels, with regard to density, thickness and moisture content, the results for thickness and moisture content were evident, while the differences in density are minimal (table 3).

Table 3. Thickness, density and moisture content with regard to the panel

Panel type	Size class	Thicknessn mm	Density g/cm3	Moisture content %
Control	Mixture	83.15	0.0481	9.85
A	Fines	82.23	0.0486	9,78
B	Middle	82.90	0.0483	10.03
C	Coarse	83.70	0.0478	10,16

During experiment it was determined that constituent size influences thermal conductivity as it is shown in figure 5.

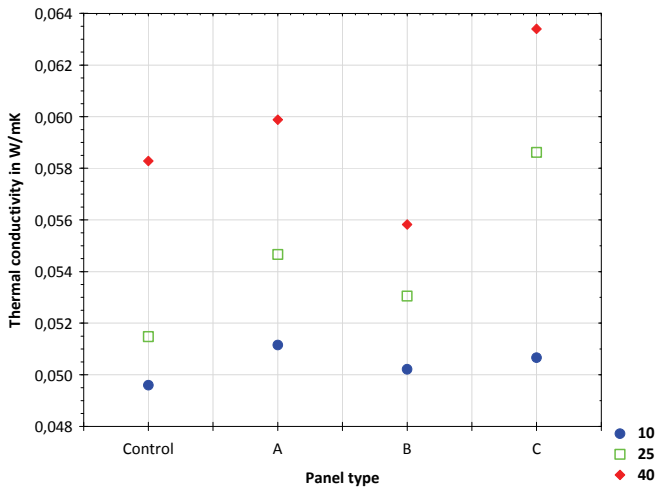

Figure 5. Thermal conductivity with regard to average temperature and panel type

Thermal conductivity varies with regard to the constituent size as well as on the testing conditions. In all cases it can be determined that thermal conductivity increases with the temperature difference (15°C steps). The smallest differences are at panel B, while the biggest are at panel with coarser constituents (C). As it can be observed the lowest average thermal conductivity was determined at panel B where middle size constituents were used (table 4).

Table 4. Average thermal conductivity with regard to the panel

Panel type	Size class	Average thermal conductivity W/mK
Control	Mixture	0.05313
A	Fines	0.05523
B	Middle	0.05303
C	Coarse	0.05756

Comparing the results from sieve analysis and thermal conductivity of panel an interesting behaviour can be observed. The results from control panel relates more to middle size constituents rather to the coarser constituents which have the highest share in mixture. Possible reason could be related to the:

- position and overlapping of constituents in panel,
- uniformity of constituent distribution in panel
- porosity of the panel.

4 CONCLUSIONS

The results obtained in this research show the importance of constituent size. Fines and coarser constituent resulted in high thermal conductivity, while middle size constituents resulted in much lower thermal conductivity. Thermal conductivity also depends on the testing condition, namely with increasing temperature differences the thermal conductivity increases. Still further research regarding the treatment of constituents for a better dimensional stability should be conducted.

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WOOD-BASED PANELS WITH IMPROVED SURFACE PROPERTIES

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Keywords: Laminated wood-based panels, Particleboards, MDF, Water-repellency and Oil-repellency

ABSTRACT

Responding to the market trends, CHIMAR HELLAS S.A., a Greek SME company serving the wood-based panels' industry, has developed resins and impregnation syrups that offer improved water and oil repellency properties to wood-based panels. For this achievement, CHIMAR HELLAS S.A., has been cooperated with the pioneering nanotechnology company NanoPhos S.A., located in Lavrio, Greece. NanoPhos developed nanomaterials tailor made for CHIMAR products, while CHIMAR modified the synthesis process of its resins and syrups in order to fit with the special properties of the NanoPhos products. CHIMAR has used the nanomaterials as additives in the glue mixture of Urea-Formaldehyde (UF) resin suitable for the manufacturing of particleboards, as covering materials of particleboards and as additives in Melamine – Formaldehyde (MF) impregnation syrups for lamination papers. Tests have been carried out both at lab and industrial scale and the results show that the nanoadditive enhanced CHIMAR products can offer wood-based panels with improved oil and water repellency surfaces. Such products, with new attractive and easy care properties, are expected to find high appreciation in the market.

1 INTRODUCTION

Wood-based panels with non-wettable surfaces from liquids enjoy special interest from consumers because they offer products with longer life and greater ease in everyday use. A traditional method used for imparting waterproof properties to wooden surfaces is the coating of the finished product with paint or oil from fossil or natural resources. However, this solution has transient effect while needs frequent maintenance. The use of polymeric diphenylmethane diisocyanate (pMDI) as binder of wood for the production of wood-based panels results also in products with good mechanical and water resistant properties. However, pMDI has great affinity to metal causing problems during the pressing of panels since the glued fibres or particles adhere on the metallic surface of the press and belts. Moreover, stringent work safety measures have to be provided and complied with when handling pMDI. In the recent years, scientists have investigated various other methods for offering oil and water resistant properties to wood surfaces like acetylation and silylation treatment (1, 2), fluorination treatment (3), fluorinated silica nanocoating (4), mild pyrolysis in inert atmosphere (5) and covalent grafting of polymer (6). Tran V. Chu et al. (7) have reported the improvement of hydrophobicity of

wood when treated with TiO2 gel under hydrothermal conditions, while Mojtaba Soltani et al (8) have reported relative achievements with the use of nano-zinc oxide (nano-ZnO).

It is widely accepted that it is harder to achieve surfaces with oleophobic than hydrophobic properties because most oils have low surface tension. Additionally, the most common method to give oleophobicity to a surface is to enhance the hydrophilic properties of it. In the case of wood-based panels, both resistances to oil and water wettability are required, so such solution is not acceptable.

In the current work, CHIMAR HELLAS S.A. has studied various methods for the improvement of the hydrophobic and oleophobic properties of surfaces of wood-based panels through the use of nanomaterials provided by the company NanoPhos S.A.

2 OBJECTIVES

The main objective of this study was the improvement of the water and oil repellency properties of wood-based panels, like particleboards and MDFs, without impairing their mechanical properties and/or physical appearance. The methods followed were aiming to provide these properties across the board or just on their surface. Tests were carried out with nanomaterials both by adding them in the glue mixture of a Urea–Formaldehyde (UF) resin suitable for the manufacturing of particleboards and by using them as coating material targeting the creation of protective film on the surface of particleboards.

Additionally, and since many wood-based panels are used laminated with paper, usually impregnated with a melamine-based resin, tests were carried out in order to study the effect of the addition of nanomaterials in the glue mix of Melamine–Formaldehyde (MF) resin. The target was to create surfaces with improved water and oil repellency properties without affecting the physical appearance of the panels.

This study may be considered as new, because there are no literature references for products that can impart both hydrophobic and oleophobic properties to wood, while any literature citation for the use of nanoadditives in the production of wood-based panels is related with the improvement of other properties than the studied ones, like the lowering of formaldehyde emissions (9).

3 MATERIALS AND METHODS

3.1 Materials

The adhesives used in this study were Urea-Formaldehyde (UF) resin and Melamine–Formaldehyde (MF) resin prepared by CHIMAR HELLAS S.A. with suitable modifications of proprietary technology and using technical grade raw materials. All tested nanomaterials were in the form of emulsions and were prepared by NanoPhos SA. Wood chips and grey uni-colour paper of 80g/m^2 were supplied by SHELMAN S.A., Greece.

3.2 Methods

Totally two groups of nanomaterials were tested. The first group was tested in particleboard while the second group was tested in MDF. In particular, the nanomaterials of the first group were tested for their effect to the waterproof property of particleboards, either by adding them in the glue mixture of the resin, replacing the conventionally used petrochemical paraffin (wax), or by using them as covering material of the finished panel. The nanomaterials of the other group, were evaluated for the oleophobic and hydrophobic result they can render to the surface of paper-laminated MDF panels, by contact angle measurements and visual tests.

The testing process followed in each case is described below:

3.2.1 Use of nanomaterials as additives in the glue mix of UF resin for the production of particleboards

Three samples of nanomaterials with the code names A, B and C were provided by NanoPhos S.A.

They were used as substitutes of wax in the glue mix formulations of Urea-Formaldehyde (UF) resin with molar ratio (F/U) 1.07/1 at the level of 0.5% solid/dry wood. For control, a glue mixture with petrochemical paraffin (wax) instead of nanomaterials was prepared. All mixtures were used in the production of single-layer particleboards from fine chips. The panels had dimensions 35.0x35.0x1.6 cm and density 650 kg/m^3 while their production process was simulating the industrial practice. Their testing and evaluation was carried out according to the European standards used by the wood-based

industry (EN 317:1993, EN 319:1993, EN 322:1993, EN 120:1992). Next, the most promising candidates were studied in comparison with the control for their waterproofing properties by contact angle measurements, artificial weathering testing (QUV) according to the European standard EN11507 and evaluation tests for the effectiveness of water repellent coatings on wood, according to the ASTM D5401 standard ("*Evaluation of Clear Water Repellent Coatings on Wood*").

For the contact angle measurements, a water droplet was placed on the surface of the panels and the angle formed by the droplet and surface was measured both in static and dynamic conditions. The measurements took place every minute for 20 minutes period of time.

The QUV test contained repeatable cycles of 4 hours exposure to condensation (50°C), 4 hours UVB and 15 minutes of water spraying. The samples were exposed for 300 hours in the accelerated weather test and the results were visually inspected.

For testing the water repellency efficiency (WRE) of panels according to the ASTM D5401, the specimens were placed in a container of water at 23°C and were allowed to float for 15 min. Then, they were turned over and were allowed to float for another 15 min to give a total water contact time of 30 min. Then, the water repellency efficiency (WRE), expressed in percentage, was calculated for each sample.

The water repellent efficiency, (WRE) in percent, was calculated for each specimen as follows:

$$WRE = 100 \bullet [(A - B) - (C - D)]/(A - B)$$

where:

A = weight of the untreated specimen after water contact, g,

B = weight of the untreated specimen before water contact, g,

C = weight of the treated specimen after water contact, g, and

D = weight of the treated specimen before water contract, g.

3.2.2 Use of nanomaterials as coating of particleboards

The above nanomaterials (A, B, C) as well as a new one (D) were tested as coating materials of commercial particleboards with dimensions 15x15cm. Approximately 4.0–4.5 g of the samples A, B & C and about 16-20g of the sample D were applied on the surfaces (15x15cm) of the particleboards in order to employ the same quantity of solid nanomaterial per cm^2 for each sample. The coatings were allowed to dry at room temperature and after 15h they were subjected to tests for surface water absorption (standard AS 4266.12), and thickness swelling (after immersion in water of 20°C for 2h and 24h).

3.2.3 Use of nanomaterials as additives in the glue mix of MF syrup for paper impregnation

Five nanomaterials with the code numbers 1, 2, 3, 4 & 5 were used as additives in the glue mix of a MF resin at the level of 5% w/w. Two paper sheets were also prepared without any additives to be used as reference. Double resin impregnation was performed on the papers for each sample. After each impregnation cycle, the papers were dried in oven of 130°C with air circulation, for 2min. 16mm MDF commercial boards of 36 x 36 cm were selected as substrate for the lamination. The laminated MDF panels were produced at CHIMAR premises. NanoPhos tested the final panels for their water and oil repellency properties through contact angle measurements and visual inspection.

4 RESULTS

4.1 Use of nanomaterials as additives in the glue mix of UF resin for the production of particleboards

It was found that the use of different nanomaterials result in different performance of the panel. The results of the testing of particleboards for the determination of their physical properties are shown in the table 1.

Table 1. Testing results of particleboards

Formula			1	2	3	4
Resin Type			UF	UF	UF	UF
Sample No			Wax	A	B	C
	Unit	Value				
Internal Bond (EN319:1993)	N/mm2	Ave	0.58	0.60	0.57	0.67
		SD	0.06	0.07	0.04	0.07
Thickness Swelling 2h, 20oC	%	Ave	22.67	20.28	31.69	30.69
		SD	1.79	1.51	1.12	1.58
Thickness Swelling 24h, 20oC (EN317:1993)	%	Ave	41.94	37.97	42.25	38.48
		SD	2.80	1.83	2.64	2.20
Absorption 24h, 20oC	%	Ave	104.99	113.88	115.00	116.28
		SD	3.44	7.38	6.14	5.98
Moisture content (EN322:1993)	%		5.82	5.15	5.38	5.34
Perforator value (EN120:1992)	mg/100g		4.00	3.99	4.10	3.69
Formaldehyde content 6.5% MC	mg/100g		4.35	4.69	4.69	4.24

Among the nanomaterials used as wax substitutes in the production of particleboards, the sample A resulted the lowest thickness swelling after immersion in water of 20°C for 2h and 24h, while its overall performance was comparable to the panel with wax. However, all panels produced with the NanoPhos samples had higher water absorption values (24h, 20°C) than the control (Formula 1). The mechanical properties and the formaldehyde content of the panels remained practically unaffected.

The results of the measurements for the determination of the water resistance efficiency of the panels are cited in the following graphs and images (numbers).

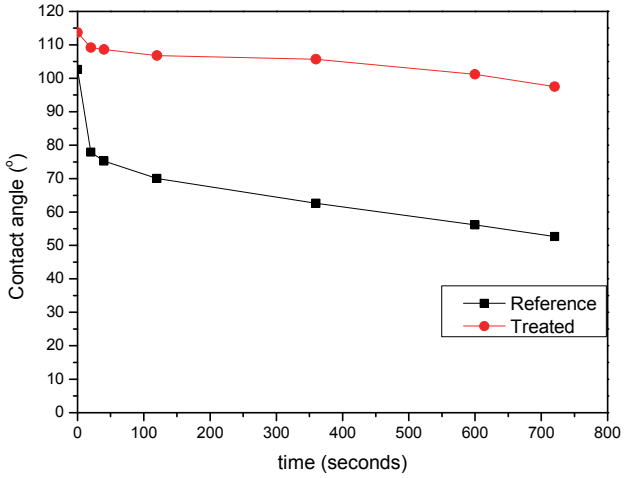


Figure 1: Contact angle measurements of reference panel and the treated panel.

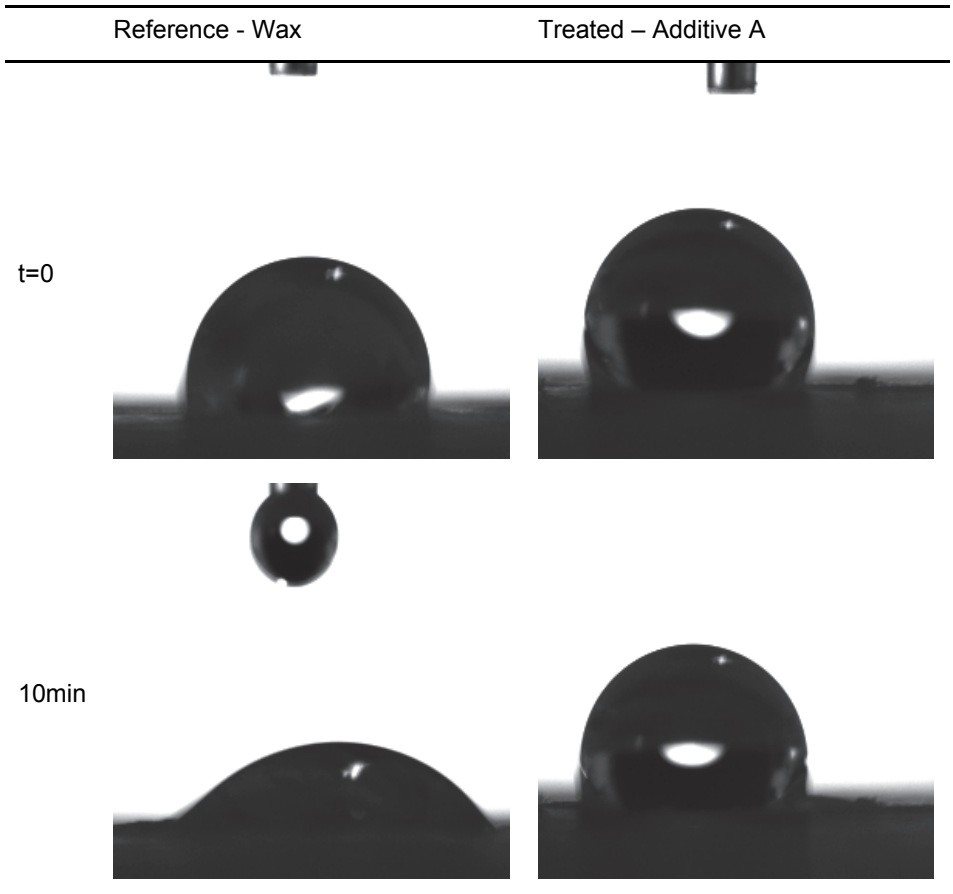


Figure 2: Photos of the water droplet during the measurement.

The water beading is easily observed on the treated panel. The larger the angle the larger the water repelling force. It is obvious that the water repelling forces of the surface are extremely high (>90°C). The contact angle on the treated panel with the additive A is higher than 110°C. It means that the surface of the panel with additive A is superhydrophobic.

The QUV test showed that the panel with wax (control-formula 1) is extensively damaged, while the swelling of the other sample with the additive A is in acceptable range of values.

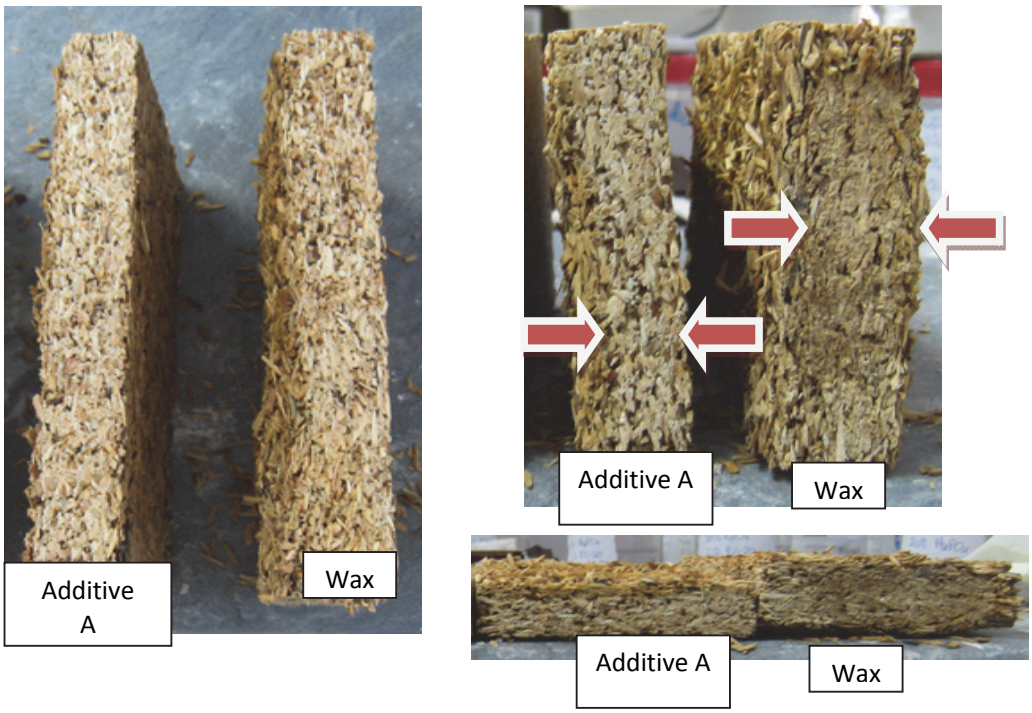


Figure 3: Photos of the panels after 300 hours exposure in QUV.

The results of the water repellency efficiency tests of the panels according to the ASTM D5401 are presented in the table 3. It is obvious that the A nanoadditive increases the WRE of the panel.

Table 2: Water Repellent Efficiency measurement.

	mass before floating (gr)	mass after floating (gr)
Reference (Wax)	48,8	54,28
Treated Additive A	46,38	51,59
	WRE =	4,93%

4.2 Use of nanomaterials as coating of particleboards

The results of the particleboards coated with the nanomaterials are presented in table 4 and 5.

Table 3:. Test of 2h and 24h Swelling & absorption

Sample	2h swell (%)	24h swell (%)	Absorption (%)	Density (kg/m³)
A	18.13	21.65	97.07	653
B	18.68	22.26	96.24	662
C	19.28	22.69	96.24	667
D	20.65	25.02	94.66	691
Blank	21.96	25.70	96.95	689

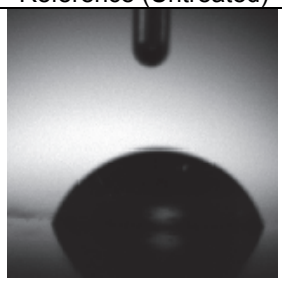
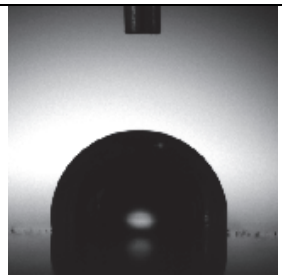
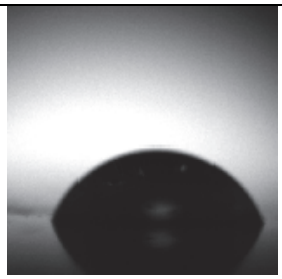
Table 4: Surface absorption test (115min each side at 0.01226 m2 surface)

Sample	Face	Weight before (g)	Weight after (g)	Surface Absorption (g/m2)
A	1	207.42	229.23	1,778.96
	2	229.23	233.53	350.73
B	1	204.14	216.7	1,022.84
	2	216.68	312.2	7,787.93
C	1	206.75	227.0	1,651.71
	2	225.02	319.7	7,725.94
D	1	204.42	205.8	109.30
	2	205.76	209.9	336.87
Blank	1	218.95	242.7	1,940.46
(no covering)	2	242.74	279.2	2,970.64

In the case where the various samples were used as covering materials, all of them had close performance when tested for thickness swelling after either 2h or 24h (table 4). On the other hand, in the surface absorption test (table 5), the sample D had the best performance with values significantly lower than the other samples. This might happened because the sample D had lower solids and therefore larger liquid amount was used to cover the panel with the same quantity of samples on solid basis. Most probably, this has led to the formation of a better film on the surface of the panel.

4.3 Use of nanomaterials as additives in the glue mix of MF syrup for paper impregnation

For testing the water resistance of the laminated MDF panels, a water droplet was placed on the surface and the angle formed by the tangent of the droplet and the surface at the three phase point was measured at different times (t=0, 1min, 10min and 20min). Images of the best performed sample (No 4) and the reference are shown in the following image 3.

	Reference (Untreated)	Treated
t=0		
1min		

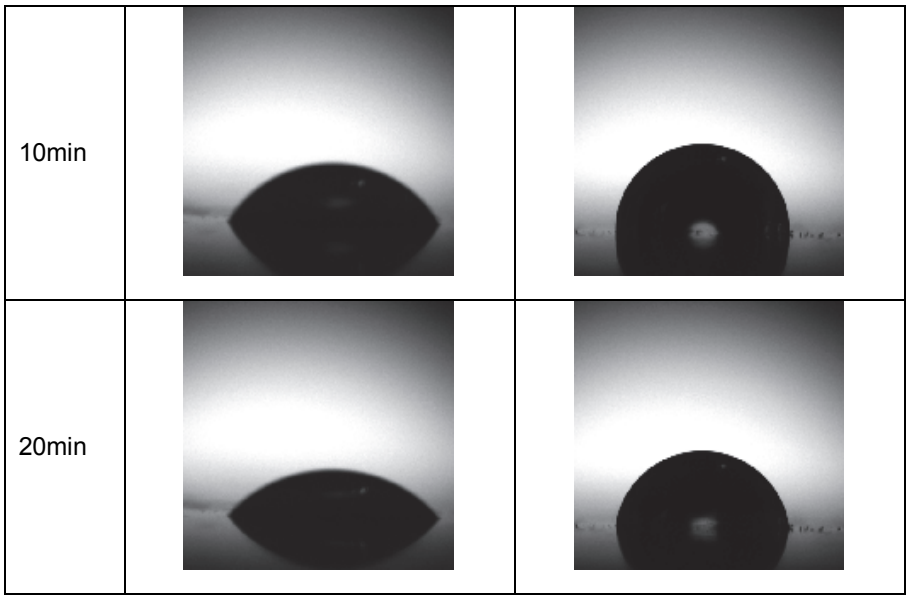


Image 3. Photos of the water droplet during the measurement of the untreated and treated panel.

Table 6. Contact angle measurements of water droplets.

	time (min)						
	0	1	2	5	10	15	20
Reference	69.32	67.35	66.11	62.72	58.4	49.44	46.81
Treated (sample No 4)	102.41	101.37	99.04	97.00	93.31	89.25	83.45

The water beading is easily observed on the treated panel. The larger the angle the larger the water repelling forces. The contact angle on the treated panel is higher than 90^{o}. As a result, the surface of the panel is hydrophobic.

Similarly, for the evaluation of the oleophobicity of the surface of the laminated MDF, an oil droplet was placed on their surface and the angle formed by the droplet and the surface was measured at different times (t=0, 1min, 10min and 20min).

Table 7. Contact angle measurements of oil droplet.

	time (min)			
	0	1	10	20
Reference	13.32	13.21	10.74	8.28
Treated (sample No 4)	67.00	64.84	63.89	61.08

From the table 7, it is obvious that the additive improves the oil repellency of the panels. The contact angle of the oil droplet is a very important value, but an empirical test (demonstrated below) is more representative in the evaluation of the elimination of oil residue on the panel. For the test, an oil droplet (10μm) was placed on the surface of the reference panel and another droplet was placed on the surface of the panel with the sample 4 nanoadditive treated surface. The oil droplet was spread by hand. The samples were visually observed for the effectiveness of the surface to repel oil without leaving oil residue.

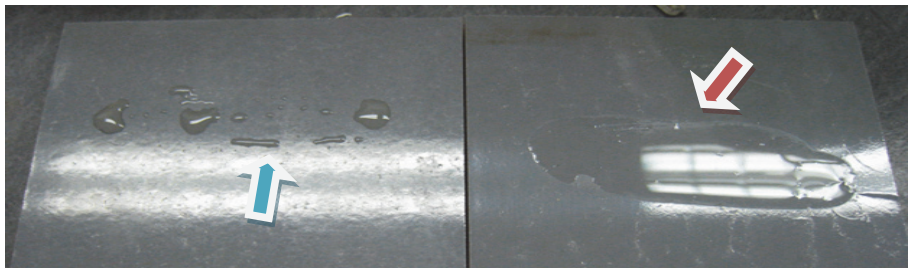


Image 4. Photos of the treated surface (left) and untreated surface (right).

It is observed that oil droplet on the surface of the reference panel is leaving oil residue. On the other hand, the treated panel repels the oil and the oil shrinks without leaving oil residue. As a result, the surface becomes easy to clean. The following image presents the results of the same testing at large scale.

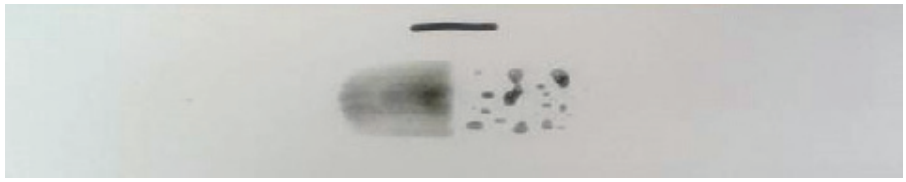


Image 5. Photos of untreated surface (left) and treated surface (right).

5 CONCLUSIONS

The nanoadditives when used either in the UF resin glue mixture for the production of particleboards or as coating materials, can improve the water repellency capability of the surface of the panels. Moreover, it was found out that nanoadditives when added in suitably modified MF syrups for impregnation of paper can impart oleophobic and hydrophobic properties to the surface of MDF panels laminated with such papers.

All the above results drive to the conclusion that the production of wood-based panels with improved water and oil repellency surface properties is practically easy and feasible. The nanoadditives used in this study may be easily incorporated into suitably modified UF resins and MF syrups while they can be used either for surface or throughout panel treatments. Such systems resins/syrups – nano additives are ready for the market.

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WHAT IS SUPERB WOOD SURFACE? DEFINING USER PREFERENCES AND SERVICE LIFE EXPECTATIONS

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Key words: hedonistic test, user expectations, service life, aesthetics

ABSTRACT

The superior properties and the natural beauty of wood make it a desired material for various applications including construction, interior/exterior design or other uses. Unfortunately, wood as any other material is a subject of deterioration due to several factors, including among others; weathering, oxidation, biodegradation, wear or decay. It is important therefore to assure the performance of wood products during their functional service life. On the other hand, not just the functional performance is an issue, but it is extremely important to consider also aesthetical service life. The goal of this work was to study how the progress of surface changes affects customer perception of the wood. The effect of gender, age, education and nationality has been included in the research. A dedicated software tool has been developed within the frame of SWORFISH project and in collaboration with COST Action FP1006 „Bringing new functions to wood through surface modification". The set of 256 respondents representing different social groups has been requested to choose between images of wooden surfaces exposed to natural weathering and displayed on the computer desktop. The choice was related to the personal "end of aesthetical service life" and based only on aesthetical impression of the respondent. The same test has been repeated providing additional information defining the time of samples exposition to weathering. It was possible, after analysis of responses, to define the time when the wood surface is not anymore acceptable and requires maintenance. The statistical evaluation highlighted difference between respondent groups but also change of tolerance for surface imperfection after realizing the efforts related to frequent repairs.

1 INTRODUCTION

Service life is a period of time after construction, during which an object/part exceeds/meets the performance requirements. During their functional life, wooden elements are exposed to several conditions including weathering (UV radiation, temperature variation, moisture stress) and biological factors (moulds and fungi). Under extreme conditions, timber may deform, check, split and/or pull away from fasteners. The formation of discontinuities on the wooden surface can activate penetration of the wood-decaying biological agents into the material structure, thus affecting also mechanical performances of wooden members. Wood colour and appearance significantly change affecting esthetical perception (Sandak et al. 2013).

Declaration of Performance (DoP) is the key concept for the Construction Products Regulation (CPR). CPR was established in order to ensure reliable information on construction products in relation to their performances. The DoP gives the manufacturer the opportunity to provide the information about the essential characteristics of his product directed to the market. Moreover, DoP assumes the responsibility for the conformity of the construction product with the declared performance. Often used Life Cycle Assessment (LCA) approach serves to evaluate wooden materials regarding their production taking into account the affiliated energy and environmental impacts (Asif et al. 2002). Reliable experimental data (period of time) regarding service life are also crucial for Service Life Planning (SLP) and Whole Life Cost (WLC). Service Life Planning provides guidance in determining the design life necessary to meet the business service requirements and building's life cycle performance. The total costs of ownership over the life of an asset are called Whole Life Cost, (or Life-Cycle Cost (LCC)). That includes the financial cost as well environmental and social costs. Therefore expenses related to planning, design, construction and acquisition, operations, maintenance, renewal and rehabilitation, depreciation and cost of finance and replacement or disposal should be considered.

Nowadays trends for maximization in the use of renewable resources, improvement of the sustainability and optimization of the long-term functionalities of building materials have become very significant. A lot of efforts are recently dedicated to increase the service life of wooden structures by implementing new finishing technologies and innovative products (Fufa et al. 2012). Accurate service life time prediction, service life costing and aesthetical performance models of recently available wooden building materials are within interest of producers and final users. It is assumed that by knowing wood characteristics and how these characteristics change according to specific processes, it is possible to predict the product properties and foresee the structure service life. Several researchers are developing models of service life and durability of wooden structures or elements (van de Kuilen 2007, Viitanen et al 2011, Thelandersson et al. 2011). On the other hand empirical aesthetics, even if considered as relatively a young science is old subject of human interest (Carbon et al. 2012). Aesthetical aspects of service life, specific consumer preferences, as well as the functionality of wooden building assemblies should be therefore within focus of currently performed research. It is foreseen that consumer demands and preferences, which might serve as limit states to develop service life prediction and performance models, will consider aesthetical aspects as well as the functionality of building assemblies.

The goals of this work were:

- to develop a simple software tool for "assessing user preferences"
- to investigate what is acceptance limit for surface defects in various groups of respondents (perception/tolerance of imperfection)
- to define the critical limits (as a factor of aesthetics).

2 MATERIALS AND METHODS

2.1 The test

Dedicated software tool has been developed in LabView 2013 (National Instruments) in order to asses the preferences of respondents. The user interface was simplified to assure focus on the inquiries and avoid any disturbing factors. The whole test consisted of seven questions. The average time needed for answer all questions in the test was ~226 seconds. The test initiated with the plain questionnaire where information regarding age, gender, nationality, education level and expertise in wood technology were collected. Series of carefully selected images were shown following the questionnaire. Photographs (or scans) of woody surfaces, presenting various aspects of the aesthetical perception were obtained from the experts in the field and/or collections available at CNR-IVALSA. No any help or supervision was provided to respondents during the hedonistic test, with exception of few

cases where respondents did not understand the exercise. Only one portable computer has been used for visualization of the sample images during whole experiment (HP Pavilion HDX, 20' display size, resolution 1680x1050 pixels) in order to minimize effect of image size variations and/or differentiations of colour as viewed on various displays. Each person was requested to read the question, look at the set of images, decide the response and express selection by clicking appropriate image. The expression of the first impression was requested and only in a few cases respondents needed extended time for decision making.

Analysis of one (within seven) questions is described within this report. The goal of that test was to determine a tolerance level for the surface weathering, assuming application of wood in windows. The user interface was basic and included text explaining context of the test and four images. The text was "at which stage are you willing to renovate your window frame" and instruction of action to be performed by respondent "(click once on the image)". The four pictures presented images of wood surface of the same sample but exposed to natural weathering for 0, 1, 2 and 4 years. The last information (the period of exposition) was not shown to the respondent. The wood sample was Scotch pine (Pinus silvestris) coated with transparent coating and exposed to the south direction on the roof of CNR-IVALSA with the inclination of 45°. The sample has been collected from the exposition stand and after conditioning the colour picture was taken once a year. The print screen of the user interface as presented to the respondents is shown in Figure 1.

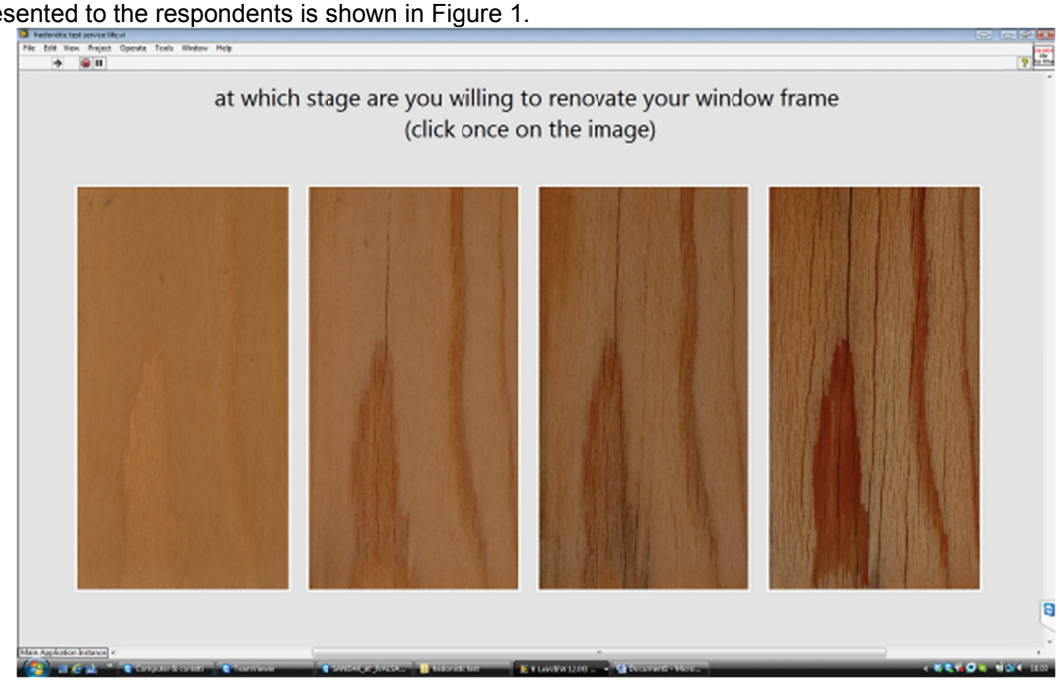

Figure 1. User interface of the preference test

2.2 Respondents

Total number of 256 respondents contributed to the hedonistic test. Several age groups, education levels and nationalities were represented within the set. The youngest respondents were junior high school students (12 years old) where the oldest were retired pensioners. Some most important contributions, in term of respondents' number were:

- members of COST Action FP1006 (and FP0904)
- students and staff from University of Life Sciences in Poznan (Poland) and University of Trento (Italy)
- students and teachers from Scuola Media in Mezzocorona and Istituto di Formazione Professionale "Sandro Pertini" in Trento
- carpenters from association San Patrignano (Italy)
- staff of the GraphiTech foundation in Trento
- staff and visitors of CNR-IVALSA/CNR San Michele and Florence
- others

As a result, citizens of 24 countries contributed to the survey. The list of countries include: Austria, Belgium, Canada, Croatia, Egypt, Eritrea, Finland, France, Germany, Ghana, Italy, Macedonia, Mo-

rocco, Netherlands, Norway, Pakistan, Poland, Portugal, Romania, Serbia, Slovenia, Spain, Switzerland and Thailand. The most frequent representation was from Italy (203) as shown in Figure 2.

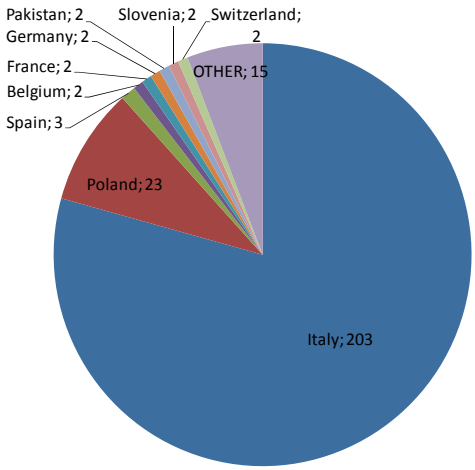

Figure 2. Distribution of the country of residence for respondents of the preference test

3 RESULTS AND DISCUSSION

An example of the survey results in regard to aesthetical preferences as depending of the country of origin is presented in Figure 3. Histograms show number of respondents selecting images taken from the samples after certain years of exposition. It has to be pointed that respondents were not aware of that time while selecting surface state suitable for renovation. Very different trends of preferences can be derived from these data. Several respondents selected brand new surface (before weathering) as that to be renovated. It was especially frequent within respondents from all counties other than Poland. The reasons for such selection were: not clear understanding of the phenomenon of wood degradation/weathering (in case of secondary school students) and overall dislike of using wood in windows (as declared by several adult respondents). The most frequent selection of the renovation time was two years after exposition. Again, respondents from Poland presented different opinion and most respondents selected one year old surface as requiring maintenance. In general, not many respondents tolerated wood surface as weathered for 4 years, with exception of Italy where more than 50 votes signify relatively high tolerance to weathered wood surface defects.

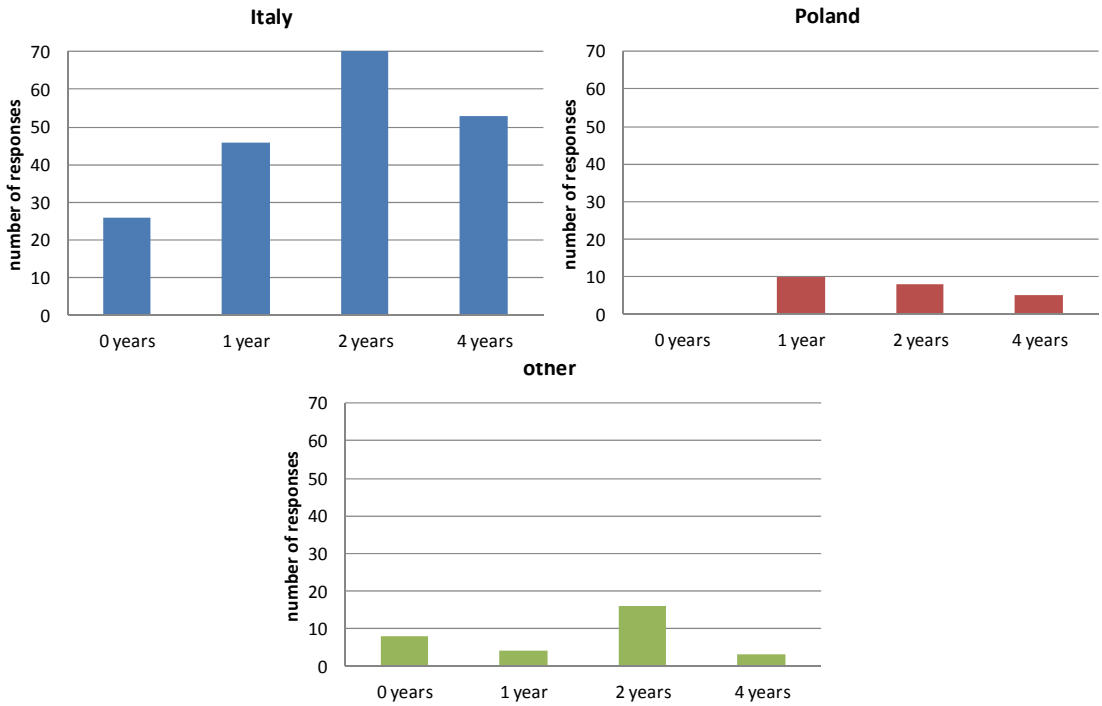

Figure 3. Histograms of responses in relation to the country of residence

The summary of the preference test can be simply presented in the chart below (Figure 4) where each respondent vote is indicated as a small circle. The preferred time for surface renovation is plotted here against the age of respondent. The trend (thick black line) indicates a "tolerance" to the surface imperfections as related to the age. It is clear that such "tolerance" increases slightly with an age of respondent.

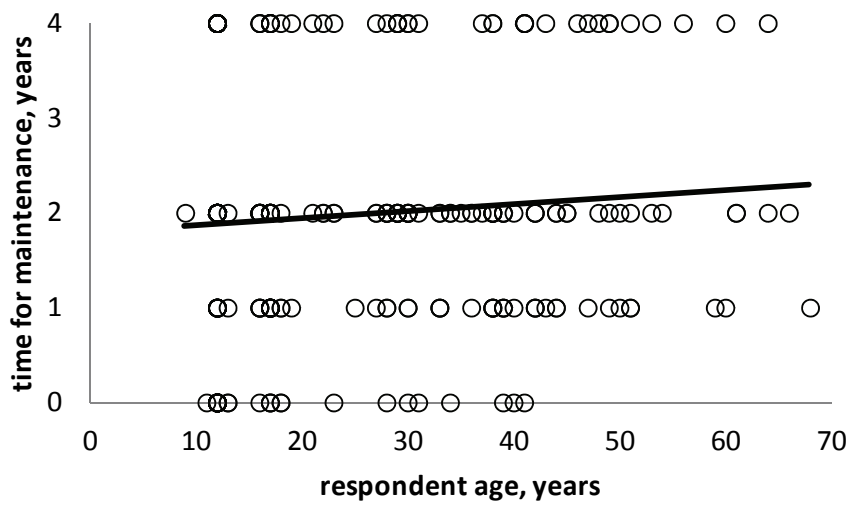

Figure 4. Preferred time for maintenance of the wood surface in relation to the respondent age

4 CONCLUSIONS

Wood is an amazing material, possessing several advantages over alternative resources. Natural beauty makes it very attractive for several applications providing superior aesthetical impressions, microclimate comfort and environmental friendliness. The performance of wood within house interior is usually straightforward. In the exterior use, however, wood products may loose visual appeal (due to staining, oxidation, and discoloration, etc.) leading to a perceived need for replacement even if they are far from the functional failure. The purpose of regular maintenance is, thus, to maintain the life of the wood product by postponing obsolescence in terms of both aesthetic and technical requirements. As a consequence, it is critical to re-define the "service life" as not only related to the technical performance but also to include a factor of aesthetical perception/tolerance. It is relatively simple to define the critical limits when wood is loosing its technical characteristics (due to decay, mechanical strength decrease, lost of dimensional tolerances, etc.). It is extremely difficult, however to define the critical limits as regarding the aesthetics - the question raises "how to measure beauty?"

The work presented here is a trial to generate routine tools capable to determine customer preferences related to outlook of the wooden products. An example discussed above can be used for scheduling regular maintenance programs, to be optionally optimized depending on the target group. It was shown that most respondents considered 2 years as the due moment to renovate window frame. The period seems to be relatively short (considering state-of-the-art wooden windows guarantying far longer maintenance-free periods). On the other hand it was intention to test the most challenging use of wood outside (softwood, coated with transparent coating, directly exposed to sunlight and not protected from rain). Some trends as related to the citizenship of respondents can be derived, even if the group size may not be representative. Moreover, no any statistical significance indicators were computed yet, as the overall project is still ongoing.

The most important conclusion of these studies, as well as driving force for further development of further research, is the necessity to "not underestimate aesthetical performance!"

5 ACKNOWLEDGMENTS

Part of this work has been conducted within the framework of the project SWORFISH (team 2009 incoming (CALL 2) and Trentino—PCOFUND-GA-2008-226070) co-financed by Provincia Autonoma di Trento. Part of this work has been conducted within collaboration of COST Action FP1006. Martino Negri, Ignazia Cuccui and Sandro Bonfa for providing some images used in the hedonistic tests.

Authors are grateful for all respondents to our test for their time and engagement, with a special thanks to members of COST Actions FP1006 and FP0904, students and teachers from Scuola Media in Mezzocorona and Istituto "Sandro Pertini" in Trento, University of Life Sciences in Poznan and University of Trento, Fondazione Graphitech in Trento as well as carpenters from association SanPatrignano.

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PROCESSING PELLETS TOWARDS LOW EMISSIONS

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Key words: pellets, VOC emissions, aldehydes, terpenes, blue stain fungi, Ceratocystis coerulescens, pine, spruce, pilot plant station

ABSTRACT

Fuel pellets are usually produced from raw materials with high lignocellulosic content like bio-mass from wood, straw etc. with a huge variety of the raw material quality grades. While high calorific value is the most desired end-product property, other product characteristics are also of interest. Gas emissions could be crucial for the subjective acceptance of pellets as "green product" and could lead to higher indoor concentrations of unwanted volatile substances if the pellets are stored inside the premises.

VOC (volatile organic compounds) emissions from pellets produced from pine and spruce were evaluated at the early stages of the product lifecycle - from wood shredding through transport and storage of the freshly produced pellets. It was found out that the VOC emission profile depends on wood type and lifecycle stage. The emitted VOC belonged exclusively to the substance classes of terpenes and aldehydes. The observations showed that spruce- and pinewood based pellets differentiated mainly in their aldehyde emissions whereby pinewood based pellets emitted substantially more aldehydes. The treatment of pinewood chips with the blue stain fungus Ceratocystis coerulescens for four weeks led to a reduction in aldehyde emissions of the produced pellets with more than 80%.Thus the profile of the VOC emissions of treated pinewood based pellets was much more similar to that of spruce based pellets.

1 INTRODUCTION AND PROJECT BACKGROUND

Conversion of energy systems towards sustainable concepts is a mayor task nowadays. Reduced production of greenhouse gases is ecologically and legally desired. Expansion of the bio based energy sector supports this strategy and further helps to assure availability of energy as well as to increase local use of resources. However, there is a constant conflict concerning utilisation of such resources for solid applications such as for paper products or wood based products on the one hand, and energy carriers such as pellets on the other. At the same time there is a global conflict about the utilisation of agricultural and other spaces for breeding of food plants, and plants for solid or energetic use.

Three members from Austrian Cooperative Research (ACR), i.e. Holzforschung Austria (HFA), OFI Technologie & Innovation GmbH and Österreichischer Kachelofenverband (KOV), joined for a closer cooperation (www.BioUp.at), in order to gain further insight in processing and optimization of production steps and product utilities of pellets. One of the major tasks within this consortium is the procession of nowadays unutilized bio-based resources as well as the further engineering of existing production processes. Fresh wood as a typical industrial processed material is often understood as a typical reference material. This further helps to better understand functional principles such as impact of processing parameter on material friction or product properties. However, the BioUp is in general open for any kind of research task related to raw materials processing, process- and product engineering.

The present study is dedicated to the problem of emissions from the end product (fuel pellets). Off-gassing is nowadays widely discussed. Carbon monoxide (CO)-emissions from bio-based pellets are of major interest due to human health risk. The process of CO-emissions is widely unclear. Currently no distinct answer can be given which factors influence CO formation from stored pellets. However, it is hypothesized, that a reduction of VOC (volatile organic compounds)-emissions from the product might be associated with a reduced risk of CO-emissions (Rossner et al. 2013).

Processes influencing the VOC emission rate might occur undetected, such as due to production process stages, material aging while storage or generation of new surfaces due to manipulation. Especially process settings such as temperature or moisture content can be assumed to show significant influence on VOC emissions. On the other hand, the VOC emission spectrum might be influenced on purpose, if such processes are better understood. Furthermore, the VOC emission rate might also be reduced by additional processes, such as material modification. One promising wood modification process could be the application of blue-stain fungi.

The blue-stain fungus Ceratocystis coerulescens is known as an effective decomposer of unsaturated fatty acids in wood. Since unsaturated fatty acids are known to be precursors of volatile aldehydes (Svedberg et al. 2004) their reduction in the raw material can be expected to cause a reduction in the VOC emissions from the end product since aldehydes are known to be major constituents of the total volatile organic compounds (TVOC) emitted from pinewood based products. Blue stain fungi are particularly suited for selective decomposition of only the extractives content in softwood without degrading its lignocellulosic backbone (Stratev et al. 2011). Albino strains of blue staining fungi are commercially used as resin and fatty acids decomposers in the pulp production (Farrell et al. 1993). Similar applications are also known for solid wood products as well as for wood based panel products.

2 OBJECTIVES

Fuel pellets are usually produced from raw materials with high lignocellulosic content like bio-mass from wood, straw etc. with a huge variety of the raw material quality grades. While high calorific value is the most desired end-product property, other product characteristics are also of interest. Gas emissions could be crucial for the subjective acceptance of pellets as “green product” and could lead to higher indoor concentrations of unwanted volatile substances if the pellets are stored inside the premises.

The objectives of this research were 1) to characterize the VOC emissions from two different softwood based pellet types, 2) to demonstrate the impact of several production processes on VOC emissions from the processed material, 3) to evaluate the potential of fungal pretreatment of raw material (wood chips) in order to reduce the VOC emissions from pellets, and 4) to evaluate the changes in VOC emissions after pellets transportation and during their storage.

3 MATERIAL AND METHODS

Pellet production was performed within the BioUp pilot plant.

3.1 Pellets production through processing of native wood species

Bark-free chips from Scotts pine (Pinus sylvestris) and Norway spruce (Picea abies) were selected as raw materials. TMP-woodchips (i.e. thermo-mechanical pulping chips) were obtained from two industrial saw-mills. Both chip species contained sapwood as well as heartwood or mature wood respectively. Figure 1 illustrates the processing and analytical pathways.

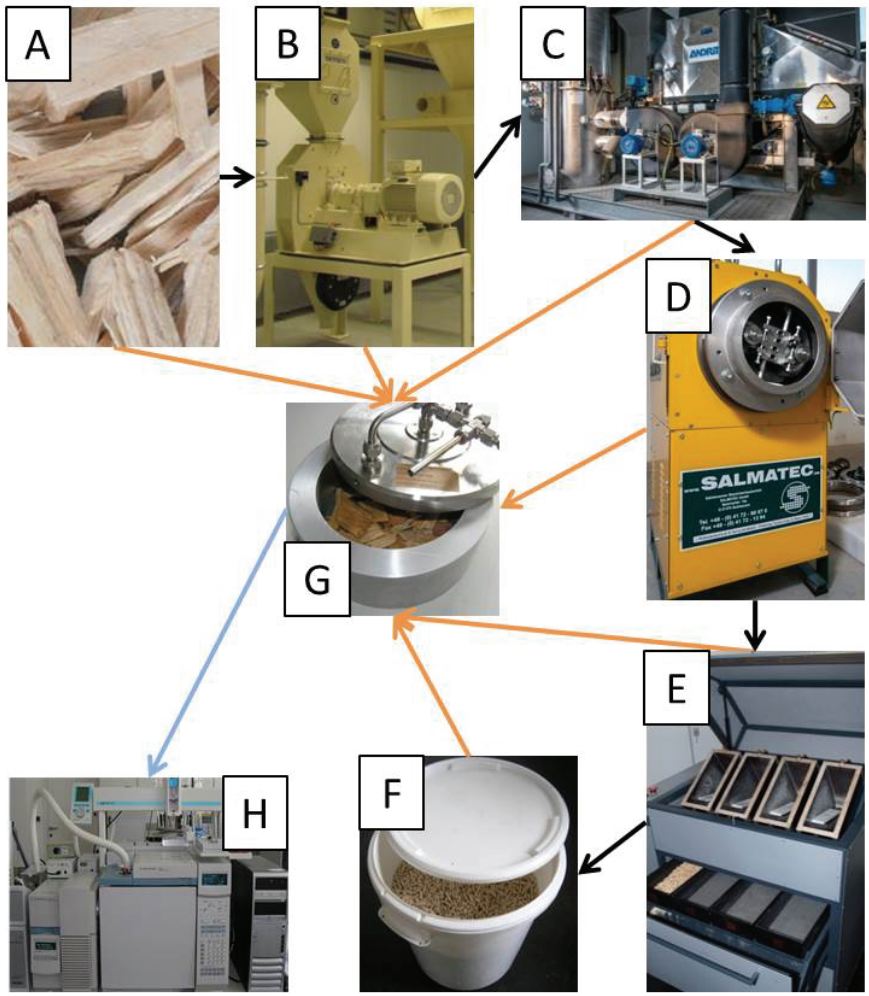

Figure 1. Pathways for material processing (dark arrows) and sampling (bright arrows): A) wood chips, B) hammer mill, C) belt dryer, D) ring die pellet press, E) tumbler, F) storage in closed buckets; F) modified FLEC including Tenax® sampling tube, G) TD-GC/MS

Wood chips (stage A) from pine and spruce were separately milled within a hammer mill (to stage B) and technically dried (to stage C) by using a belt dryer in order to gain optimum particle size and water content for further pelletizing. Pelletizing was conducted within a ring die pellet press (stage D). Freshly produced pellets were further processed (to stage E) within a tumbler loading pellets with mechanical forces thus simulating transportation processes (see below). The pellets were then stored in closed buckets (stage F) for up to six weeks in order to simulate aging. VOC (volatile organic compounds) emissions from all stages were evaluated through exposition of material sample aliquots in a modified FLEC cell (Field and Laboratory Emission Cell; by defined temperature and air flow rate) for a certain time interval and by trapping the volatiles in Tenax® sampling tube (stage G).

3.2 Pellets production through processing of fungal modified wood

Blue stain fungus-treated (*Ceratocystis coerulescens*) pine chips were also used as feedstock (stage A*) for the production of treated pellets (stage D*) in order to investigate the potential of the selected fungi for reducing VOC emissions. The fungal incubation period was chosen to be four weeks in order to ensure enough time for the fungus to metabolize the unsaturated fatty acids in the wood. The exact incubation conditions as well the inoculation scheme will be presented in a following extended article.

3.3 Examination of the influence of mechanical transport on VOC emissions

Mechanical as well as pneumatic transportation of pellets could also have impact on VOC emissions throughout product life time. Manipulation of pellets always yields formation of new product surfaces due to cross-sectional breaking and axial abrasion. Furthermore, while movement in the transportation sleeve the surface of the pellets is subjected to friction and as a result the material temperature rises. Primary VOC (*i.e.* volatile substances that evaporate directly from the material without any alteration) will tend to evacuate the pellets material faster than at neutral conditions. The pneumatic air ventilation at higher temperatures increases the oxidative potential of the pellets environment. Thus the autoxidative process which normally takes place in the fresh wood material (Roffael, 2006) and the corresponding secondary VOC Emissions (*i.e.* volatile substances that are formed from nonvolatile precursors as a result of oxidative processes) will be intensified.

For the examination of the impact of pneumatic transportation on VOC emissions a part of the freshly produced pellets (stage D) were not put into the tumbler thus forming the stage of the non-transported pellets (E*). These pellets were separately stored (to stages F*) and characterized regarding their VOC emissions and the results were compared against the “transported” pellets.

3.4 Time schedule of the experiments and labeling of the different stages

In virtue of limitations in the capacities of the production- and measurement- systems it was not possible that all pellet variances (spruce, untreated pine, treated pine, transported spruce etc.) were produced and examined simultaneously. This is usually the biggest drawback by conducting VOC related experiments, because during processing of one material fraction there are changes which occur in the VOC potential of the unprocessed material fractions. Therefore it is of importance to know the time schedule of the experiments in order to interpret the observed differences accordingly.

In the present study, the major time gap occurred due to fungal treatment of pine wood chips. This process took four weeks. Reference material was stored throughout this time under comparable conditions without any fungal treatment. However, this reference material was subjected to natural processes. Hence, the VOC emission spectrum changed within that time. In order to evaluate the impact of fungal treatment, a reference value for non-treated pine wood chips after four weeks storage was determined additionally.

3.5 VOC emissions determination

Aliquots of 25 g material were used for the VOC emission determinations. The material samples were added without any further processing (like drying) into a cylindrical aluminum chamber with an internal diameter of 150 mm and a volume of 1,68 l. A FLEC® cell (www.chematec.com) was used as lid of the chamber as its inlet was connected to the air supply of the test chamber. VOC sampling was conducted by connecting Tenax® sorption tubes (www.markes.com) at the FLEC® cell outlet. The air flow and the sampling volume were chosen to be 100 ml/min and 1,5 l.

Loaded Tenax®-tubes were subjected to VOC-measurement following the thermal desorption measurement principle described in DIN ISO 16000-6 (2004) by means of TD-GC/MS (thermal desorption gas chromatography coupled with mass spectrometry: Markes Ultra TD - Agilent 6890N/5973 GC/MS). Qualitative and quantitative interpretation of derived chromatograms was based on a calibration covering more than 70 single VOCs. If possible, detected substances were evaluated based on the calibration for the identified substance or for a similar substance if missing from the calibration. The portion of substances that had to be evaluated based on toluene d8 equivalents was lower than 5 % compared to the TVOC throughout the whole research. Due to reasons of simplicity, results presented here will represent the sum of single substances belonging to two different substance classes, i.e. terpenes and aldehydes.

4 RESULTS

Native pine wood showed the highest total amount of VOC emissions (TVOC) throughout the whole experiment. Figure 2 illustrates the alteration of VOC composition among terpenes (green) and aldehydes (red) throughout the experiment. While terpenes dominated the emissions (>95%) of chips and fresh powder, aldehydes from autoxidation of fatty acids appeared after technical drying. Throughout further processing, aldehydes dominated the emission spectrum.

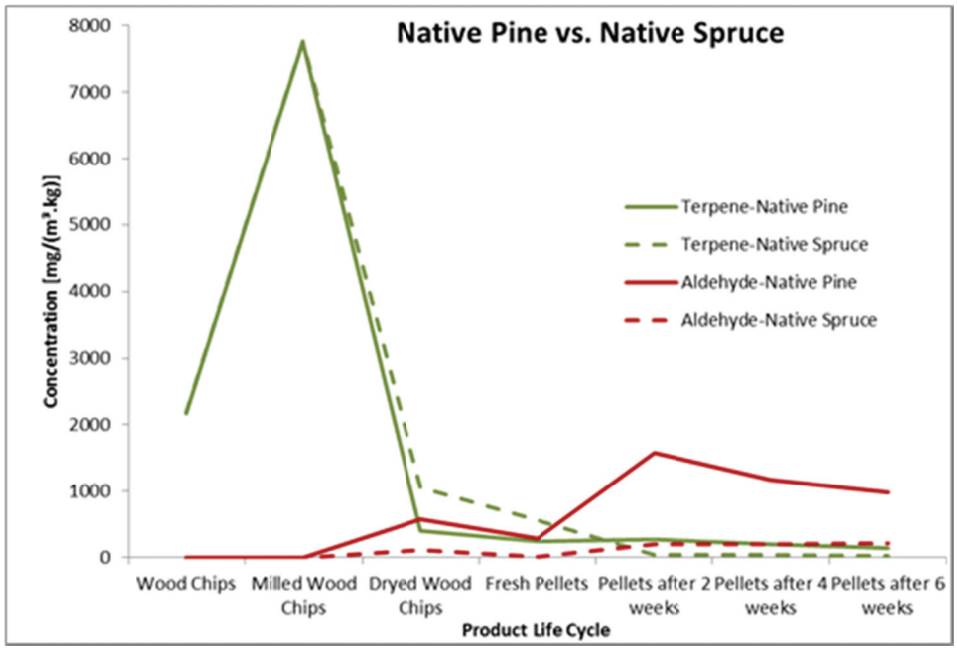

Figure 2. Comparison of native pine vs. native spruce: VOC measurement results at several product life cycle stages for fresh wood chips (stage A), after milling (stage B), after drying (stage C), directly after pelletizing (stage D) as well as after storage (stage F). (terpenes = green, aldehydes = red, native pine = full line, native spruce = dashed line)

Comparison of native pine wood and blue-stain treated pine wood is given in Figure 3. By means of fungal treatment (dashed and dotted line), this characteristic VOC profile of pinewood based products was significantly altered. Due to natural alterations within the sored material, a reference value was taken for non-treated pine chips. This reverence value is about 2000 mg/(m³.kg) lower compared to native pine wood chips without storage. Hence, strong alterations actually occur in this stage with respect to primary VOCs. This fact must be seen as a potential overlaying factor concerning VOC emissions for fungal treated wood. However, formation of secondary VOCs occurs in later stages of the product life cycle.

After technical drying, TVOC of fungal treated material decreased substantially due to the reduction of aldehyde emissions. Even for aged pellets, a considerable reduction of aldehyde emissions was observed due to fungal treatment.

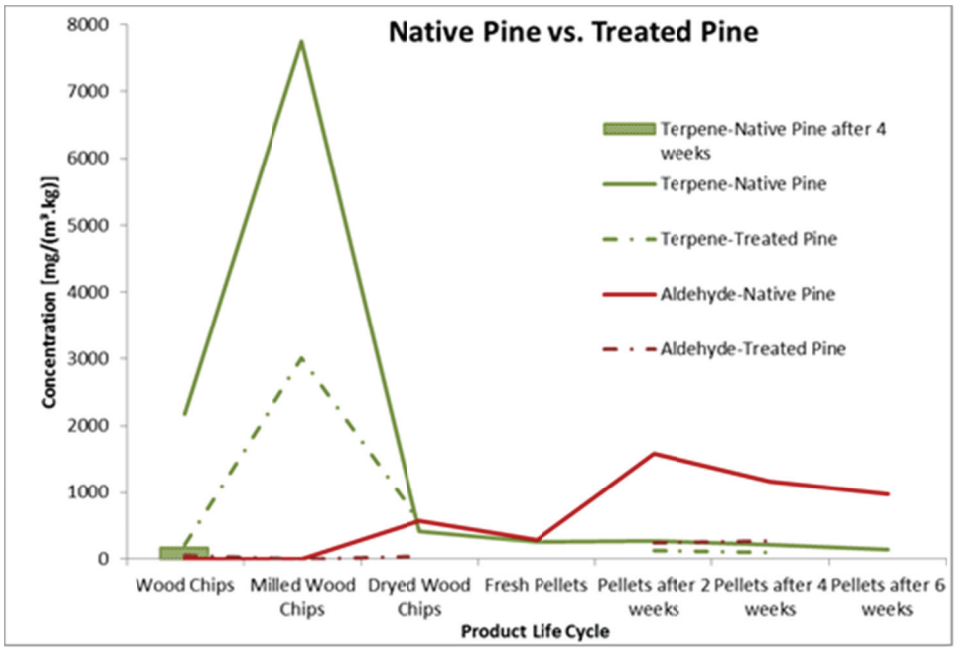

Figure 3. Comparison of native vs. treated pine: VOC measurement results at several product life cycle stages for fresh wood chips (stage A, A*), after milling (stage B, B*), after drying (stage C, C*), directly after pelletizing (stage D) as well as after storage (stage F, F*). (terpenes = green, aldehydes = red, native pine = full line, treated pine = dashed and dotted line, native pine chips after storage = box)

Comparison of fungal treated pine with native spruce demonstrated, that fungal treatment is suitable for the production of low emitting pine pellets, reaching emission levels such as spruce pellets (Fig. 4). Once again, the offset for terpene emissions due to storage must be considered here.

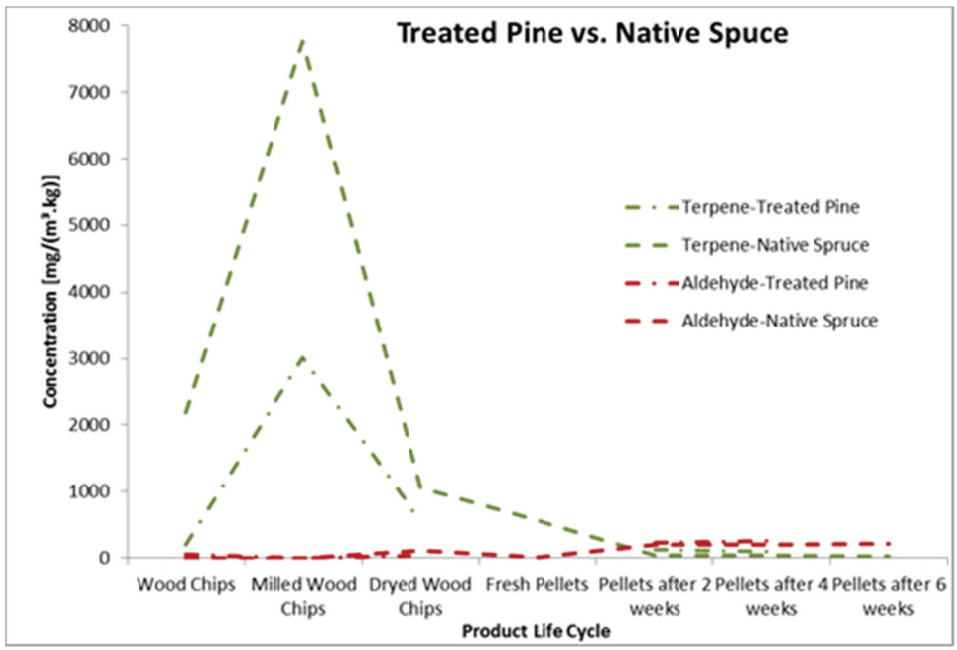

Figure 4. Comparison of treated pine vs. native spruce: VOC measurement results at several product life cycle stages for fresh wood chips (stage A), after milling (stage B, B*), after drying (stage C, C*), directly after pelletizing (stage D) as well as after storage (stage F, F*). (terpenes = green, aldehydes = red, native spruce = dashed line, treated pine = dashed and dotted line)

Transportation simulations (stage E) also showed a VOC-reduction, whereas this effect was most pronounced for native pine pellets of low age (Fig. 5). Due to abrasive forces while transportation, new surfaces were generated leading to increased emissions during transport, and hence led to lower primary VOC emissions thereafter. However, VOC reduction by means of abrasion is not worthwhile and less effective than fungal treatment.

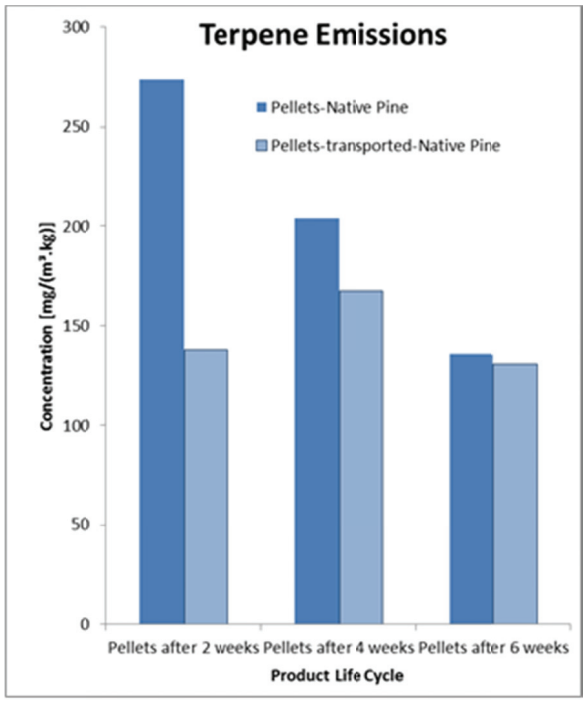

***Figure 5**. Impact of transportation simulation on terpene emissions from pellets produced from native pine wood.*

5 DICUSSION AND CONCLUSION

During the pellets production the highest emission rates were found for milled wood chips, as particle surface was increased leading to easier evaporation of volatiles from the material matrix. At this stage the emitted VOC belonged almost exclusively to the group of monoterpenes. Terpene emissions showed a distinct decline during the following stages. Such trend is known for terpenes from other applications, representing a typical behavior of primary VOC. On the other hand, emissions of secondary VOC such as aldehydes arose as their formation was catalyzed by processing conditions.

The highest VOC emissions from pellets were found for native pine wood. Pellets from native spruce wood showed much lower aldehyde emissions compared to native pine wood and comparable terpene emissions.

Treating native pine wood with blue-stain fungi for four weeks led to reduction in the VOC emissions of the raw material and in the end product. The emission profile for treated pine wood turned out to be similar to the profile of native spruce wood.

Pellet transportation turned out to reduce terpene emissions most probably as a result of the temperature increase due to friction.

The formation of secondary VOCs should be decreased. The most pronounced increase was found for pellets produced from native pine while storage. Application of spruce wood as well as blue-stain fungi treated pine wood can reduce such undesired effects.

It is concluded that the formation of undesired emissions from wood pellets can be reduced by means of controlled processing and raw material selection, including wood modification. Treatment of fresh wood chips by means of blue-stain fungi seems to be very promising in this context. Further research trials were actually started within BioUp aiming to better understand interactions between processed materials and applied processes, in order to reduce VOC emissions on the one hand, and to eliminate the risk of CO formation on the other.

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QUANTIFYING THE NATURAL VARIATION OF FORMALDEHYDE EMISSIONS FOR WOOD COMPOSITE PANELS

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Key words: Formaldehyde emissions, wood composites costs, Taguchi Loss Function

ABSTRACT

Quantifying the natural variation of formaldehyde (HCHO) emissions during the manufacture of wood composite panels (e.g., particleboard, medium density fiberboard or MDF, etc.) and estimating the costs incurred by controlling the natural variation of HCHO emissions are the goals of this study. Natural variation of HCHO emissions from wood composite panels in the presence of regulatory limits directly influences the operating target that manufacturers' must maintain, i.e., larger natural variation of HCHO emissions requires lower operating targets for HCHO emissions which increases the costs of manufacture. Taguchi's "Loss Function" is used to estimate the costs incurred from controlling variability in HCHO emissions. Taguchi's key premise is that costs in a manufacturing process increase at a non-linear rate for deviations from the operating target (two-sided for lower and upper specifications) or specification limit (one-sided for only one specification limit). In this study, the one-sided loss function was used to estimate the costs incurred due to variation in HCHO emissions. Statistical models are developed of HCHO emissions as a function of multiple independent process variables. Statistical models with R2 > 0.70 are used to estimate the total variance of HCHO emissions from particleboard manufacturing processes. Examples of Taguchi loss analyses for producers with a range of variability in HCHO emissions will be reviewed.

1 INTRODUCTION

Quantifying the natural variation of formaldehyde (HCHO) emissions during the manufacture of wood composite panels (e.g., particleboard, medium density fiberboard or MDF, etc.) and estimating the costs incurred by controlling the natural variation of HCHO emissions were the goals of this study. Natural variation is present in all manufacturing processes and influences the costs of manufactured product (Deming 1986, 1993, Young and Winistorfer 1999). Natural variation of HCHO emissions from wood composite panels in the presence of regulatory limits directly influences the operating target that manufacturers must maintain, i.e., larger natural variation of HCHO emissions requires lower operating targets for HCHO emissions which increases the costs of manufacture (Young et al. 2014). Taguchi (1993, 1995) developed the universally-accepted "Taguchi Loss Function" for businesses to quantify the costs (or loss) from natural variation of manufacturing processes.

The California Air Resources Board (CARB) enforces an Airborne Toxic Control Measure (ACTM) for HCHO emissions of 0.09 ppm for particleboard and 0.11 ppm for MDF and other composite panel products (currently known as CARB II). In 2010, President Obama of the USA signed into law the "Formaldehyde Standards for Composite Wood Products Act1," which establishes the same limits as CARB II for the entire United States. HCHO is contained in many resins that are used as a binding agent for wood composite products.2

A common characteristic of plants that manufacture wood composite panels is the presence of automated processes. Such processes are usually the most important factor in determining the efficiency of consumption of raw materials, in maintaining the integrity of product design parameters during production, maximizing the level of product quality, and maintaining environmental emission standards, such as HCHO emissions. A key process in wood composite manufacture is the "pressing operation," which is the site of the thermal bonding of HCHO-based resins with cellulosic fibers by the application of heat and pressure in a highly controlled manner. HCHO emissions from manufactured panels are a by-product of the manufacturing process and the natural variation of HCHO emissions is a function of the natural variation of manufactured product. That is, a highly efficient manufacturing process will have less natural variation in manufactured product and therefore less variation in HCHO emissions from manufactured panels; to the contrary inefficient manufacturing processes will have more variation in HCHO emissions (Singh et al. 2010). The wood composite panel industry in the USA has a large variety of manufacturing technologies with varying age of facilities with varying types of feedstocks and resin technologies (André and Young 2013). Therefore, the industry has considerable differences in the natural variation of manufactured product and resulting HCHO emissions.

2 METHODS

Quantifying variance is fundamental to the concept of reducing variability and improving industrial processes (Young et al. 2014). Variability in a wood composite manufacturing process propagates as a "series" and the variability (or variance) of final manufactured product is the summation of the variances of the subcomponents of the system (Galton 1869), see equation (1) for a two component simple-system example,

$$Var_{TOT}(aX + bY) = a^2VarX + b^2VarY \pm 2abCov(X,Y) \quad (1)$$

Where:
VarTOT = Total process variance,
VarX = process variance component X,
VarY = process variance component Y,
Cov (X,Y) = covariance of components X and Y,
a = scale influence of process component X,
b = scale influence of process component Y.

As mentioned earlier, the variability of manufactured products (and therefore variability in HCHO emissions) varies significantly across the wood composites panel industry given the differences in manufacturing technologies, diversity of feedstocks and chemical additives, and age of facilities. In this study, HCHO emissions are modeled as a function of the independent process variables (e.g.,

[1] http://www.govtrack.us/congress/bill.xpd?bill=s111-1660

[2] HCHO is synthesized by the oxidation of methanol. It is among the 25 most abundantly produced chemicals in the world and is employed mainly in the production of resins that are used as adhesives and binders for wood products, pulp and paper, and insulation materials.

press cycle times, amount of resin applications, etc.) for multiple wood composite manufacturers that represent the diversity across the industry. The natural variability in HCHO emissions are estimated from the statistical models.

Taguchi's "Loss Function" is used to estimate the costs incurred from controlling variability in HCHO emissions. Taguchi's key premise is that costs in a manufacturing process increase at a non-linear rate for deviations from the operating target (two-sided for lower and upper specifications) or specification limit (one-sided for only one specification limit), see Taguchi et al. (2005). In this study, the one-sided loss function was used to estimate the costs incurred due to variation in HCHO emissions. If a wood composites panel manufacturer theoretically had zero variance in HCHO emissions, it could operate at the CARB II upper specification limit (USL) and incur no additional costs due to variability. However, as variability in HCHO emissions increase, the manufacturer must run a lower planned operating target in HCHO emissions to comply with the CARB II USL (i.e., must operate a HCHO target to the left of the USL). The estimate of loss for the one-sided Taguchi Loss Function is given in equation (2)

$$Loss = c \cdot (USL - 6 \cdot \hat{\sigma})^2 \quad (2)$$

Where

c = cost coefficient per ppm reduction in HCHO emissions,

$\hat{\sigma}$ = estimated standard deviation of HCHO emissions,

USL = CARB II upper specification limit.

The lower the operating target for HCHO emissions, given no reduction in HCHO variation, the higher the cost, e.g., slower pressing cycles, more expensive resins and chemical additives, higher panel density, etc. (Figure 1). If variation is reduced in HCHO emissions, an operating target can be attained that is closer to the USL, therefore, incurring less financial loss (Figure 2). This is the direct rational for variation reduction as means of improving industrial processes and lowering operating costs (Young et al. 2014).

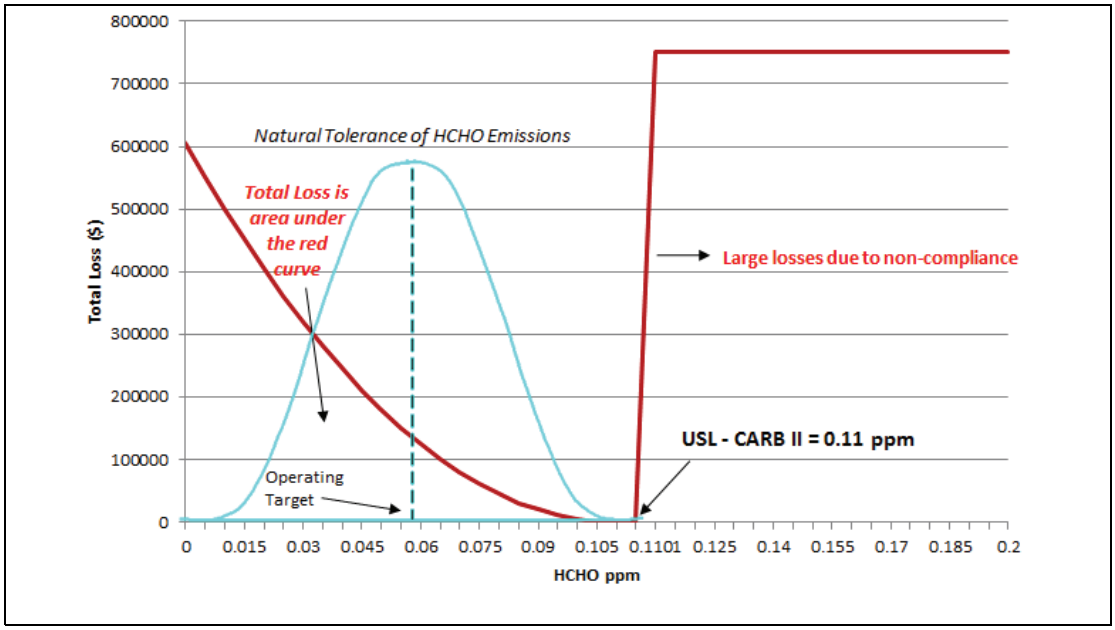

Figure 1. Illustration of one-sided Taguchi Loss Function on an annual basis for particleboard HCHO emissions variability.

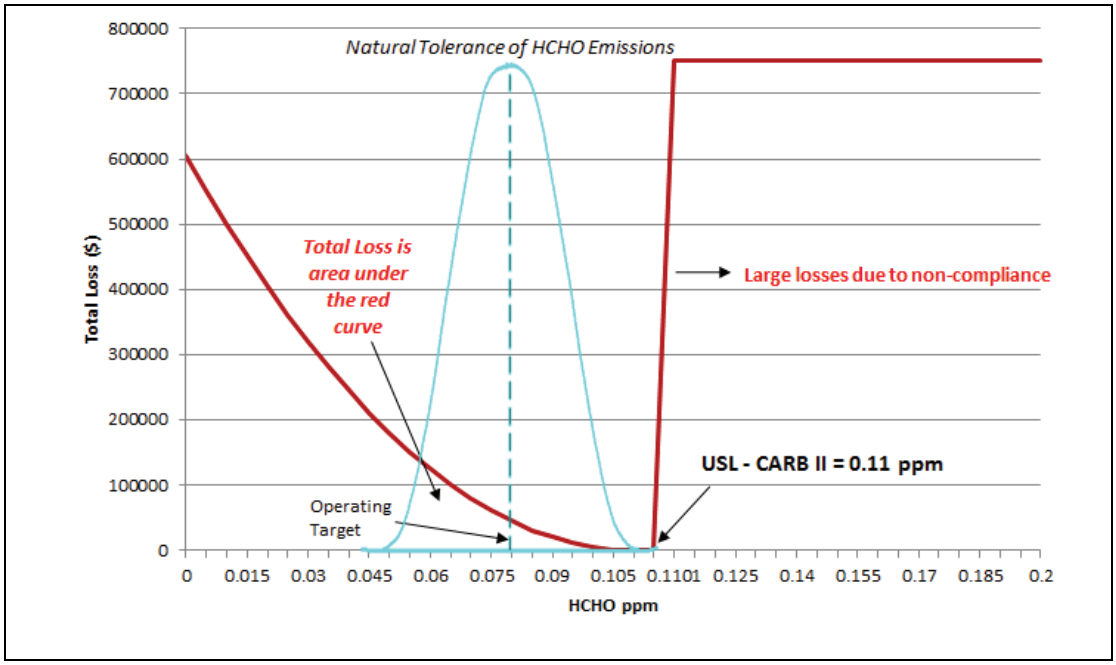

Figure 2. Illustration of one-sided Taguchi Loss Function on an annual basis for particleboard HCHO emissions with reduced variability.

3 RESULTS

Statistical models were developed of HCHO emissions as a function of multiple independent process variables. Actual data are obtained from several particleboard manufacturers. Statistical models with R2 > 0.70 are used to estimate the total variance of HCHO emissions from particleboard manufacturing processes, recall equation [1]. Based on the total HCHO emission variance estimates, the natural tolerance for HCHO emissions is derived using the model parameters and Monte Carlo simulations (n = 1000) of natural variability in HCHO emissions. Since the HCHO emissions data are from two-hour DMC (GP® Dynamic Microchamber) 3 chamber estimates, results are presented in the context of the scale of DMC units of measure. The operational upper specification limit (USL) for HCHO emissions based on DMC measurements will vary by particleboard producer and the producer's strength of correlation to large chamber testing to meet the general CARB II USL, e.g., 0.09 ppm for particleboard, 0.11 ppm for MDF.

Results vary based on the process variability of a particleboard producer, i.e., not all wood composite manufacturers have the same natural tolerances (natural tolerance = six standard deviations). A producer that has low variability in HCHO emissions has an operational target closer to the USL. In this study, the data from a low variability particleboard producer indicates DMC natural tolerances of approximately 0.10 ppm suggesting an operational DMC target of 0.16 ppm with a process mean of ± 0.01 ppm around the target (Table 1). The process mean and operational target differ given the assumption of a "wandering mean" associated with most continuous processes. The "typical" particleboard producer has an estimated DMC natural tolerance of 0.15 ppm in HCHO emissions suggesting an operational DMC target of 0.14 ppm with a process mean of ± 0.03 ppm around the target. The high variability particleboard producer has an estimated DMC natural tolerance of 0.20 ppm in HCHO emissions suggesting an operational DMC target of 0.12 ppm with a process mean of ± 0.05 ppm around the target (Table 1).

Taguchi loss analyses indicate a loss for a low variability producer of approximately $4.32 per thousand lineal feet ($4.32/M) given an operational DMC target of 0.16 ppm. For 100 MM annual production, the total loss per year due to HCHO emissions variability and the resulting target is $432,000 for this low variability producer. For a typical producer the loss is $7.26/M given an operational DMC target of 0.14 ppm. The typical producer has an estimated total annual loss of $726,000 for this target. For a high variability producer the loss is $11.13/M given an operational DMC target of 0.12 ppm. The estimated annual loss for the high variability producer is $1,113,000.

[3] DMC was the GP® Dynamic Microchamber.

Table 1. Losses for low, typical, and high variance wood composite panel producers.

Wood Composite Panel Producer Scenario	Process Mean from Target (ppm)	Operating Target (ppm)*	Natural Tolerance of HCHO (ppm)	CARB II USL/Producer Target (ppm)**	Loss per unit at Operating target***	Total Annual Loss****
Low HCHO variance	± 0.01	0.16	0.10	0.09/0.06	$4.32/M	$432,000
Typical HCHO variance	± 0.03	0.14	0.15	0.09/0.04	$7.26/M	$726,000
High HCHO variance	± 0.05	0.12	0.20	0.09/0.02	$11.13/M	$1,113,000

**Using two-hour DMC values and based on the QCL correlating to 0.09 ppm large chamber testing.*
***USL and estimated producer target for particleboard for CARB II. DMC USL for CARB II would be higher and vary based on a mill's correlation to large chamber testing.*
****Costs per thousand are for one standard deviation of variation which was necessary to scale-up estimates for 100 MM of annual production. Costs differ given the natural variation produced from the Monte Carlo simulations and the strength of correlation in the statistical model predicting HCHO emissions.*
*****Based on a 100 MM per year particleboard mill.*

4 CONCLUSION

Study results based on actual data obtained from particleboard producers' suggest the following:

- Particleboard manufacturers have different levels of variability in their processes, products, and HCHO emissions;
- Variability within continuous wood composite processes propagates throughout the process, resulting in variability in HCHO emissions from manufactured panels;
- HCHO emissions variability result in differences in the natural tolerances of HCHO emission (0.10 to 0.20 ppm in DMC values) and influence the operational targets necessary to meet the CARB II requirements;
- Lower operational targets in HCHO emissions result in higher costs of production, *i.e.,* low operational targets require higher cost resins, slow line speeds, slow press cycle times, higher density targets, etc.;
- Costs due to controlling the variability in HCHO emissions vary from $430,000 to $1,100,000 per year for any given wood composite producer.

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