Alternative Particleboards Based on Treated and Untreated Hay

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Agricultural resources have a great potential to be a supplement or replacement for wood, especially in countries lacking wood resources, or during times of economic turmoil of wood markets, for manufacturing panel products. Previous research has focused on various sources including wheat straw, rice straw, rapeseed, or oil palm stems, but so far hay was not considered. Hay consists of cut and dried grasses, legumes, or other herbaceous plants. It has similar structure to wheat straw with a typical waxy surface layer and poor bondability. Soaking in NaOH was employed to improve the bondability of used full-length hay, or hay particles for urea formaldehyde (UF) resin. A comparison of the physical and mechanical properties was assessed. The vertical density profile of hay panels made from treated hay reached smaller differences between surface and core density. Full-length hay panels reached the higher average values of the equilibrium moisture content (EMC), due to the structure. The bending properties of panels made of treated hay particles showed improvement, with modulus of rupture being 3.5 times higher, and modulus of elasticity 2.6 times higher than that of the untreated hay particles. Thickness swelling after 48 hours decreased for the NaOH-pretreated hay panels.

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INTRODUCTION

The growing demand for wood-based panels for buildings and furniture and the increasing worldwide concern for reducing the pressure on forest resources require alternatives to wood raw materials. Agricultural resources as supplementing or replacing materials for wood in composites have been revealed for some time (Bower and Stockman 2001; Zhu *et al.* 2012; Klímek *et al.* 2018). Earlier research has focused on the manufacturing of particleboards, using agricultural resources such as wheat straw (Mo *et al.* 2003), rice straw (Gerardi *et al.* 1998; Li *et al.* 2010; Yasin *et al.* 2012), rapeseed (Hýsek *et al.* 2018), reed canary grass (Trischler and Sandberg 2014), cotton stalks (Guler and Ozen 2004), bamboo (Lee *et al.* 1996), sorghum stalks (Kimura 1996), miscanthus stalks (Park *et al.* 2012; Klímek *et al.* 2018), or oil palm stems (Yahya 1994). Technical issues with the utilization of these materials have been identified, *i.e.*, prevalent seasonality, storage issues, decentralized availability of sources, or poor bondability (Rowell and Rowell 1996). Among, the unsolved problem of poor bondability is especially critical when using urea-formaldehyde resins (Zucaro 1995).

Hay is a renewable biomass produced worldwide, and consists of cut and dried grasses, comprising many grass species. Hay is utilized almost exclusively in the agricultural industry. Other possibilities exist for the use of hay as a raw material in the wood-based panel industry. Despite possible added-value uses, a vast amount of dried and cut grasses remains on the fields, is burned, or is used as a landfill.

There are two options for manufacturing panels made from herbaceous plant stalks. The first option is to use polymeric diphenylmethane diisocyanate (pMDI) resin, and the second option is the utilization of urea-formaldehyde (UF) resin (Dalen 1996; Shen *et al.* 2011). The high price of pMDI, along with the minor use in the particleboard industry are seen as disadvantages. The UF resin, as the most used adhesive in the particleboard industry, is very affordable. When using plant stalks as a raw material to make particleboards, some treatments such as soaking in acidic or alkaline solutions are necessary to reach appropriate bonding level (Han *et al.* 2001). Alkaline treatment, *i.e.* NaOH, has been known as an effective pretreatment method for straw biomasses, because both lignin and hemicellulose are partly dissolved (Wan *et al.* 2011). The NaOH treatment breaks down ester linkages between lignocellulose and the cuticular wax layer present on the exterior grass stalk surfaces (Binod *et al.* 2010).

The structure and components of the many straws of herbaceous plants are quite similar, but research has been focusing mainly on wheat straws due to the higher amounts available. The main problem of wheat straw is the epicuticular wax layer that contributes to poor bondability with UF (Xia and Fang 1993; Wu and Zhou 2002; Wu and Zhou 2003; Cheng *et al.* 2004; Dai *et al.* 2004; Halvarsson *et al.* 2010). Several pre-treatment methods have been looked at, including the application of silane coupling agents, spraying/soaking with NaOH, spraying with H_2O_2, or enzymatic pre-treatments (Han *et al.* 1998, 2001; Zhang *et al.* 2003; Mingzhu *et al.* 2007; Shen *et al.* 2011; Cao *et al.* 2017). Treatments did improve the penetration of the adhesive into the straw fine structure, which affected also the physical and mechanical properties (Han *et al.* 1998, 2001).

Overall, strawboards have shown acceptable performance in many studies, demonstrating the potential to substitute for wood in particleboards (PB) or medium-density fiberboards (MDF) (Grigoriou 2000). Strawboards, reedboards, and hayboards could be attractive in countries that have a lack of wood resources, while having readily available agricultural biomass resources (Bower and Stockman 2001). Many of the currently existing strawboard mills are almost exclusively focused on wheat straw use (Dalen 1996; Zhou and Zhang 2007; Zhu *et al.* 2012). The authors are not aware of manufacturing sites that are concentrating on the use of hay to produce panels manufacturing or use hay as an addition in otherwise wood-based PB production. In this study, we hypothesize that the utilization of the hay particles can reach competitive physical and mechanical panel properties. The study has focused on the additional treatment of hay particles and full-length hay stalks, for the manufacturing of PB with the utilization of the UF resin.

EXPERIMENTAL

Manufacturing

Hay was collected in the area of Stare Hamry, Beskydy (Czech Republic). This included bulbous oat grass (*Arrhenatherum elatius* L., J. Presl, & C. Presl), meadow foxtail

(*Alopecurus pratensis* L.), orchard grass (*Dactylis glomerata* L.), golden oat grass (*Trisetum flavescens* (L.) P. Beauv.), black bent (*Agrostis gigantean* Roth.), westerwolds ryegrass (*Lolium multiflorum* Lam.), and other grasses.

Two different hay fractions were used: (1) full length-uncut hay, and (2) hay particles cut with the cutting mill SM 300 (Retsch, Hann, Germany) mill. For the latter, the particles were screened, and the fractions smaller than 1.0 mm and bigger than 5.0 mm were excluded. Subsequently, some of the hay was soaked in 5% NaOH solution for 15 min and 60 min, at 20 °C, followed by a washing in 5 batches of clean water for 2 min, after which all of the material was dried under 60 °C to 6% MC.

Six different groups of panels were made: The first group contained 100% core wood particles Norway spruce (*Picea abies* L. Karst), which served as the control (PB-C). The second group was produced with full-length untreated hay (FL-C), while the third group contained untreated hay particles (CP-C). The fourth group included full-length hay soaked in NaOH for 15 min (FL-15). The fifth group contained hay particles soaked for 15 min in NaOH (CP-15), while for the sixth group hay particles soaked for 60 min in NaOH were used (CP-60).

Table 1. Prepared Panel Groups for Testing

Group 1: Spruce particleboard (control)	PB-C
Group 2: Full-length hay (untreated)	FL-C
Group 3: Hay particles (untreated)	CP-C
Group 4: Full-length hay (15 min soaked in NaOH)	FL-15
Group 5: Hay particles (15 min soaked in NaOH)	CP-15
Group 6: Hay particles (60 min soaked in NaOH)	CP-60

The panels were made using 49% urea-formaldehyde (UF) resin solid with 12% Kronores CB1635 (Dukol Ostrava s.r.o., Ostrava, Czech Republic) by weight mixed with the catalyst, and 0.2% wax added. The resin was sprayed through nozzles into the rotary blender. Two 12-mm thick panels with dimensions of 600 mm × 600 mm were prepared. The target oven-dry density of the panels was 580 kg/m^3. The panels were pressed at 180 °C, with 30 s for closing, 120 s holding at the pressed position, and 30 s for opening.

Testing Procedure

Images of the hay particles were acquired using the Sympatec QicPic (Sympatec GmbH, Germany) device, equipped with the Gradis dispersion system, at the frequency of 500 frames per second. Measuring range was between 17 µm and 33,792 µm.

The pH value of the hay and control particles was measured after drying, a solution of 2 g of particles in 100 g of water was mixed for about 10 min.

Contact angle was measured on hay and spruce particles at room temperature using the Surface Energy Evaluation System (Advex Instruments, Brno, Czech Republic). Contact angle of the droplet is an angle between the droplet and the surface of the wood. Contact angles of 12 droplets of 1 µL volume were measured.

Physical and mechanical testing

Specimens were conditioned at 20 °C and 65% relative humidity (RH), prior to mechanical and physical property testings. All these properties were tested according to European standards unless noted otherwise.

Density and density profile

The density EN 323 (1994) was determined on 10 specimens cut from each panel, and the density profiles were measured on three 50 mm × 50 mm × 12 mm specimens cut from each panel. Density profiles were obtained at a linear resolution of 0.01 mm through the sample thickness by using an X-ray Density Profile Analyzer (DPX300-LTE, IMAL, Italy), and the average density profile was calculated.

Wood-water relations

The moisture content (MC) EN 322 (1994) was determined on 10 specimens with dimensions of 50 mm × 50 mm. The thickness swelling (TS) EN 317 (1996) was measured after 24 h and 48 h, respectively, using 10 specimens with the same dimensions.

Bending properties

The mechanical testing was carried out on a Zwick ®Z050 universal testing machine with testXpert v11.02 software and a 50 kN load cell (Zwick GmbH & Co. KG, Ulm, Germany). Three-point bending tests EN 310 (1995) were conducted to determine the modulus of elasticity (MOE) and the modulus of rupture (MOR) on 10 specimens of each panel type, having the dimensions of 290 mm × 50 mm, with the span 240 mm.

Statistical Analysis

Data were processed using STATISTICA 10 software (StatSoft Inc., USA), applying a one-factor analysis of variance (ANOVA), followed by Tukey's honest significance post-hoc test (HSD test).

RESULTS AND DISCUSSION

Particle size data are shown in Fig. 1, revealing that there are higher amounts of thinner and longer particles for hay, compared to spruce particles.

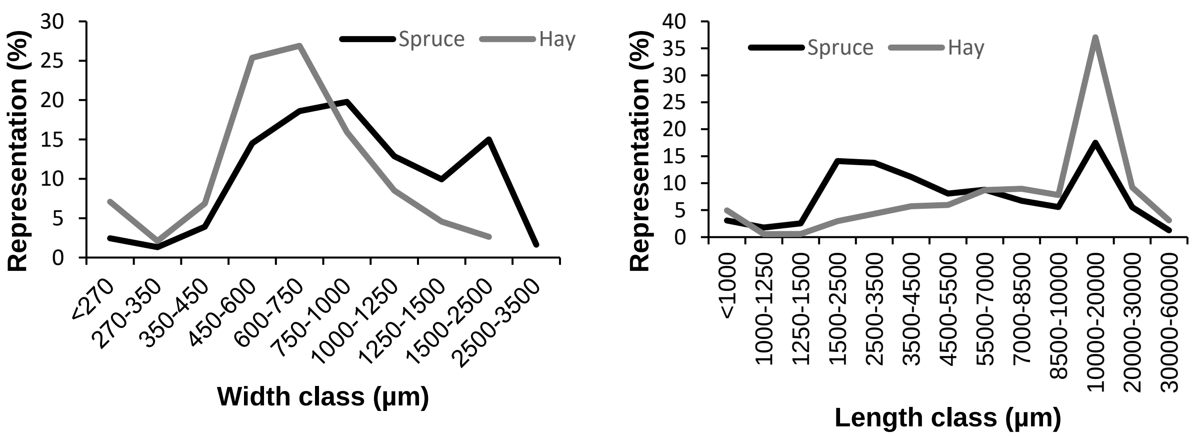

Fig. 1. Particle size distribution for hay and spruce

The pH value for spruce particles was 5.53, and for hay it was 6.04. For hay after 15 min in NaOH the pH increased to 9.43, while 60 min in NaOH the pH was 9.65. The results from other research (Zhu *et al.* 2012) showed similar values, such as for wheat straw where the pH without treatment was 6.10, and 7.86 after an alkali treatment.

The contact angle for spruce particles was 63.8°, whereas for hay it was 92.2°. For hay after 15 min in NaOH the contact angle was 63°, and for hay after 60 min in NaOH it was 61.9°. Liu *et al.* (2004) stated that the cuticular wax layer gets chemically roughened after a NaOH treatment, called "chemically etched", and the wettability of binders gets potentially improved by the treatment. Therefore, the removal of cuticular wax layer is lowering the contact angle with the bonding getting increased (Kurokochi and Sato 2015).

The average density obtained was 589 kg/m^3, and the density of the panels did not show significant differences across the tested groups. The usual u-shaped density profile was observed for all panel types (Kelly 1977; Wong *et al.* 1998; Wong 1999), with the density maxima located near the surface layers. The hay control panels (FL-C and CP-C) showed higher differences between surface and core density, compared to the alkaline treated ones. The specimens CP-15 and CP-60 have shown density profiles being almost identical, with only minor differences in the core layers (Fig. 1). The density profile is highly responsible for differences in physical and mechanical properties, especially for the improvement of the hay panels treated with NaOH.

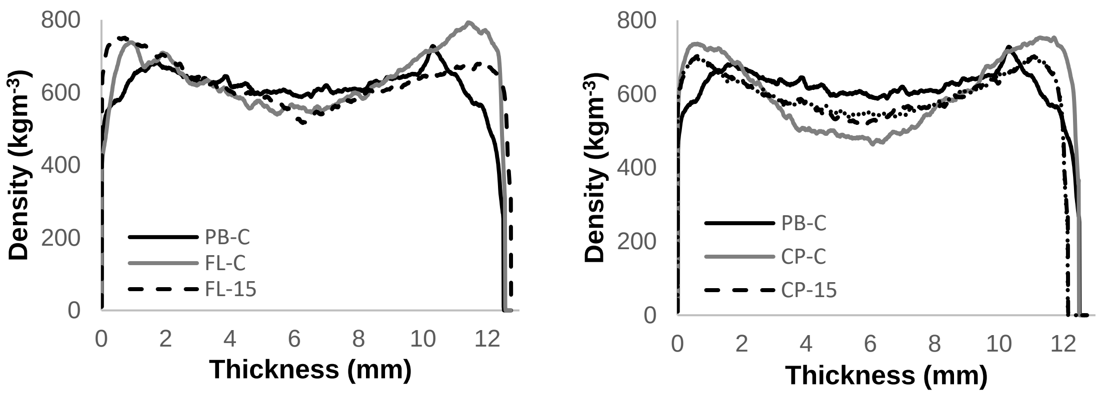

Fig. 2. Density profile for the panel groups PB-C, FL-C, CP-C, FL-15, CP-15, and CP-60 at 20 °C and 65% RH (each profile averaged from three scans), see table 1 for group indicators

Panels made from full length hay stalks (FL-C, FL-15) reached higher equilibrium moisture contents (EMC). A lower EMC observed for hay particle-made panels is most likely caused by the less porous panel structure, with a better particle-to-particle contact (Han *et al.* 1998). Similar EMC values for spruce and hay particleboards corresponded with the trends of average EMC values, ranging between 7.5% and 7.8% in an earlier study (Grigoriou 2000).

Control panels made from spruce particles reached the MOR value of 5.8 MPa, which is comparable with cut hay particles soaked in the NaOH CP-15 and CP-60, 4.5 MPa and 5.0 MPa respectively. Comparison of the CP-15 and CP-60 with control panel CP-C showed that treatment of the hay particles raised MOR by about 3.5 times, which is in accordance with (Zhu *et al.* 2012), indicating that utilization of the NaOH improves the mechanical properties of panels. Full-length hay composite showed no statistically significant difference between control and NaOH treated panel with higher variability, which was caused by the dimensions of the hay.

The samples CP-15 and CP-60 reached the highest MOE about 1700 MPa, which was higher than CP-C (651 MPa) and particleboard panel (1095 MPa). Full-length panels with and without treatment reached the same values of the MOE as PB-C without any significant difference. The configuration and particle size have a great effect on bending properties (Brumbaugh 1960).

The improvement of MOR and MOE and of alkaline-treated panels might be not only due to the "chemically etched" phenomenon, but probably also due to an increased crystallinity, as shown by Cao et al. (2017). An NaOH treatment destroys amorphous cellulose regions (Zheng et al. 2012), thus crystallinity of the cellulose increases, which is in turn has improved the bending properties.

Table 2. Average Values of EMC and Bending Properties of PB-C, FL-C, CP-C, FL-15, CP-15, CP-60 Panels at 20 °C and 65% RH

	EMC (%)	MOR (MPa)	MOE (MPa)
PB-C	8.7 (0.1) A	5.8 (2.1) D	1095 (289) B
FL-C	9.3 (0.6) B	3.7 (1.8) B, C	1341 (249) B
CP-C	8.7 (0.2) A	1.3 (0.5) A	651 (92) A
FL-15	9.4 (0.1) B	2.8 (1.2) A, B	1119 (373) B
CP-15	8.7 (0.2) A	4.5 (1.0) B, C, D	1710 (233) C
CP-60	8.7 (0.2) A	5.0 (0.9) C, D	1703 (239) C

Means with the same letter in column do not differ statistically by Tukey´s test (α = 0.05). Numbers in parentheses represent standard deviation

Full length hay stalk panels (FL-15) showed higher TS values after 24 h in comparison with PB-C. The trends for higher average values of TS after 24 h for hay composites in this research was comparable with a previous study by Grigoriou (2000), which was on wheat straw showing TS 23.9% for wood particles, and 31.3% for a mixture of 75% wheat particles, and 25% wood particles. After 48 h there was a significant increase of the TS compared to the PB-C, but it was also observed that alkaline treatment had a decreasing effect on the TS values, which opposes Kurokochi and Sato (2015) who have found greater 24h TS values for NaOH treated straw panels.

The trends for TS after 48 h for the hay particles and full-length hay stalk panels corresponded with other research (Han *et al.* 1998), which stated that closer structure of the fine particles tended to decrease the TS in comparison to the panels made from coarse particles. It is well known that TS is affected by the quality of bonding and adhesive properties (Mo *et al.* 2003).

Overall, potential for future research in the field of hay treatments is evident, especially for particleboards with improved water properties. A next step in this research could be hay-spruce mixtures, which might be easier to get applied in particleboard manufacturing.

Table 3. Thickness Swelling of Tested Specimens after 24 h and 48 h of Water Immersion

	24 h TS [%]	48 h TS [%]
PB-C	20.0 (3.0) A, B	24.2 (3.0) A
FL-C	25.1 (8.8) A, B, C	73.6 (11.4) D
CP-C	23.2 (4.2) A, B, C	60.5 (18.3) C, D
FL-15	29.7 (10.2) C	57.8 (8.4) C
CP-15	26.6 (6.4) B, C	42.0 (5.4) B
CP-60	17.1 (7.6) A	47.2 (9.7) B, C

Means with the same letter in column do not differ statistically by the Tukey´s test (α = 0.05). Numbers in parentheses represent standard deviation

CONCLUSIONS

1. We were able to approve the hypothesis that hay particles and full-length hay stalks could be successfully used for particleboards reaching competitive physical and mechanical properties.
2. Sodium hydroxide treated hay particles and full-length hay stalks, respectively, showed significant potential as an alternative particleboard material.
3. The hay particle treatment led to higher bending properties, compared to untreated hay particleboards, due to “chemical etching” effect as well as higher crystallinity. Further, compared to classic spruce-made particleboards similar MOR and a higher MOE value could be achieved.
4. The sodium hydroxide treatment of hay has also reduced thickness swelling, after 24 h well as 48 h, with values close to the ones obtained for the spruce-made control.

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