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Vital gluten for particleboard production - effect of wood-particle moisture on board properties --Manuscript Draft--

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Vital gluten for particleboard production – effect of wood-particle moisture on board properties

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The growing environmental awareness is leading to an increased interest in the use of bio-based adhesives and proteins such as vital gluten. The purpose of this study was to investigate the influence of the wood-particle moisture content, water application and press time on the internal bond strength, thickness expansion and thickness swelling of particleboards glued with vital gluten. Green and dried wood particles with similar moisture contents were achieved through drying or water addition and were blended with vital gluten powder and pressed for 1 to 3 minutes. The results show that not only the pressing time and moisture content, but also the way of achieving the moisture content has a strong impact on the performance of the boards. At comparable moisture content, never-dried (green) particles with high moisture content in combination with a dry adhesive application produced boards which performed better than boards made of dry particles with water addition to simulate liquid adhesive application.

Keywords: bio-based adhesive, internal bond strength, particle configuration, thickness swelling

Introduction

A particleboard in its original form is an engineered wood product (EWP) made of wood particles that are hot-pressed in the presence of an adhesive and other additives, to form a flat board (panel). The species of which the particleboard is made, the shape of the particles, their size and distribution within the board have a strong impact on the mechanical properties of the board (Lundqvist and Gardiner 2011). The development of wood-based boards is closely linked to the development of the adhesive as the quality of the bonding is one factor determining the performance and properties of the product. Condensation adhesives are commonly used for particleboard and are delivered in liquid form and consisting of linear or branched oligomers and polymers in aqueous solution or dispersion. As the adhesive cures, the polymers convert to a three-dimensional, cross-linked network which are insoluble and non-meltable (Dunky 2003). Nowadays, the conventional adhesives are 90% amino-plastics such as UMF (urea melamine formaldehyde), 4% phenolic plastics such as PF (phenol formaldehyde), 3% pMDI (polymeric diphenylmethane diisocyanate), and 3% others (Berglund and Rowell 2005, Niemz and Wagenführ 2012, Funk et al. 2017). Additives are substances added to the EWP to bond wood particles together (i.e. adhesives), or to improve the properties of the EWP and facilitate board manufacture (Sandberg 2016).

The use of bio-based adhesives is marginal in volume compared to the use of synthetic adhesives. The problem when using a bio-based adhesive such as protein is that the formulations are sensitive to the source of protein and require time-consuming adjustments leading to low production capacity and therefore higher costs. Furthermore, the mechanical properties of the particleboard glued with protein-based adhesive are positively related to the moisture content (MC) of the mat before pressing and, at comparable moisture content, the boards require higher pressing times than boards

glued with synthetic adhesive (Krug and Tobisch, 2010). The motivation for this research was to reduce this adhesive-related problem in particleboard production and to find ways how to improve the manufacturing process when using one of the emerging bio-based adhesives in the forest products industries, namely vital gluten.

Vital gluten is a form of wheat gluten. Wheat gluten is derived from wheat kernels or their flour by processes leading to the separation of starch and gluten. Vital gluten is gluten isolated and produced under mild temperature conditions, and it therefore still has its ability to form a molecular network with viscoelastic properties (Van Der Borgh et al. 2005).

In general, adhesives based on natural proteins can be classified as dispersion adhesives. Dispersion adhesives are not curing in the way condensation adhesives do but are setting by evaporation of the liquid phase into the wood substrate or atmosphere (Frihart 2013). Therefore, when dispersion adhesives such like protein-based adhesives are used in particleboard production, they often require longer pressing times than condensation adhesives, see e.g. Khosravi et al. (2010) or Lei et al. (2010). The required temperature during the hot-pressing process is at least 130°C which is, according to Metzger (2007), the critical temperature required to achieve melting and polymerization of wheat protein. Polymerization of the protein i.e. curing of the adhesive, leads to a higher water resistance of the bond line.

An aqueous solution of the adhesive is used in particleboard production to achieve a uniform adhesive spread on the wood particles and to provide a uniform wetting. Due to the excess bound water in particles, the wood is often dried to a MC of 2-8%, with the optimum MC depending on the water content of the adhesive used. Too high a MC in the particle mat before pressing leads to longer press times and increases the risk of delamination of the particleboard when the pressure is released after

pressing, due to a high internal vapour pressure (Kelly 1977, Irle and Barbu 2010).

Over-penetration of the adhesive into the wood particles also has to be avoided because it leads to a decrease in the amount of adhesive available for the necessary wood-adhesive and adhesive-adhesive interactions (Frihart 2013).

A certain amount of water is, however, needed to allow for heat transfer from the heating platens to the core of the particleboard during hot-pressing, and to support plasticization of the wood particles. Variations in the MC of the particles in the cross-section of the particle-mat affect the density profile of the finished board, and this in turn affects the mechanical properties of the particleboard (Kelly 1977, Wong et al. 1998).

In summary, a specific amount of water is needed in particleboard production to promote a uniform distribution of the adhesive, to bring the adhesive into close contact with the surface of wood particles, to transfer heat during hot-pressing from the heating plates to the core of the panel, to plasticise the particles, and to regulate the mechanical properties of the panel. With conventional adhesives, water is applied in liquid form on the surface of the particles during adhesive application. When bio-based adhesives such as vital gluten are used in powder form, the MC of the wood particles must be considerably higher than when conventional adhesives are used, and according to Trischler and Sandberg (2015) it is beneficial if the particles and the water in these are frozen when the vital gluten is applied. Such a process has the following advantages:

- The poor solubility of vital gluten in water (Örnebro et al. 2000) will not influence the production process, as it is applied as powder.
- No denaturation of the vital gluten or its derivatives by chemical, mechanical or thermal treatment to improve the solubility before application is needed, see e.g. Nordqvist et al. (2010, 2012, 2013) or D'Amico et al. (2013).

- Problems during the blending of particles with water and the vital gluten due to agglomeration of the protein in the presence of warm water are avoided (Trischler and Sandberg 2015).
- The MC of the wood particles can be regulated independently from the adhesive content, which is advantageous for regulating the pressing time and the cross-sectional density profile, and thus the mechanical properties of the panels (Kelly 1977, Wong et al. 1998).

Objectives

The purpose of this study was to investigate the influence of the wood-particle MC, water application and pressing times on some important properties of particleboards glued with a wheat-protein vital-gluten adhesive.

Materials and Methods

Three-layer particleboards were produced from a mixture of Norway spruce (*Picea abies* Karst.) and Scots pine (*Pinus sylvestris* L.) particles with different initial moisture contents (MC). MC is defined as the weight of water in wood, expressed as a percentage of the oven dry weight of the same sample.

The particles were separated by size into those smaller than 3 mm for the surface layers and those larger than 3 mm (to a maximum of 48 mm) in fibre direction for the middle layer. The mixing proportion was about 80% by weight of Norway spruce and 20% of Scots pine particles; however, an accurate information about the mixture of the particles cannot be done as the particles were received directly from a particleboard mill.

Two types of particles were used for the study: never-dried (green) particles with a mean MC of approximately 50%, and dried and conditioned particles with a

mean MC of 6%. The green particles were dried in a universal drying oven at 103 °C to 15, 20 or 25% MC, and the dry particles were sprayed with water to achieve a mean MC of about the same level, i.e. 15, 20 or 25%, Table 1, in order to study how the origin of the particles (green or dried) and the MC adjustment before particleboard production may influence the properties of the particleboards when vital gluten is used as adhesive. After adjustment of the MC, the particles were stored in sealed packages at -10°C before adhesive application and pressing.

[Table 1]

The wheat protein vital gluten (Amygluten from Tereos®) was applied as a powder on the frozen particles. The amount of vital gluten was fixed at 10% based on the dry weight of the particles.

[Figure 1]

The pressing time, excluding press closure (10 to 20 sec.) and opening (approx. 30 sec.), was varied from 1 to 3 minutes. The press temperature was fixed at 160°C. The target thickness of the boards was 11.5 mm, controlled by a duct spacer, and the target density was 550 kg/m³. The size of the boards was 500 x 500 mm. The target thickness and density together with the thickness expansion (TE) of the particleboard when the press was opened, i.e. the difference between the target thickness and the final thickness in per cent were determined after conditioning at 20°C and 65% RH (relative humidity) for two weeks.

Samples for testing thickness swell (TS) and internal bond strength (IB) were prepared from each board and conditioned at 20°C and 65% RH for a further 7 days. The samples were prepared by trimming the boards from each side (9 cm) to avoid any edge effects. Afterwards they were quartered through the centre, and out of each quarter 9 samples with in plane dimensions of 50 x 50 mm² were cut. Out of these 9 samples, 2

samples for testing IB and 2 samples for testing TS were randomly selected. Four samples were tested according to the following schedules:

- TS according to EN 317: 50 x 50 mm² specimens were immersed in water, maintained at a temperature of 20°C, for 2 and 24 hours (CEN 1993)
- IB according to EN 319: 50 x 50 mm² specimens were tested in a universal testing machine using a loading rate of 10 mm/min (CEN 1993)

Results

Table 2 presents the thickness, thickness extension and density of the particleboards after 2 weeks of conditioning, and the results of the TS test after 2 and 24 hours of immersion in water. The results of the IB test are presented graphically in Fig. 1 in relation to the value required by the standard EN 312. Table 3 shows the effects of particle type and pressing time on the TE, TS and IB of the particleboards.

[Table 2]

The results in Table 2 show that the TE, and therefore also the thickness of the boards, tends to increase with increasing MC at a given pressing time. The results for TS show that most of the swelling occurs within the first two hours and is slightly higher for the boards made of green particles.

[Figure 2]

The IB within the different groups increased with increasing pressing time and was in general higher for the particleboards produced from green particles (Nos. 1 to 7) than for those made of dried particles moistened before adhesive application (Nos. 8 to 14).

The statistical data in Table 3 show that there are differences depending on the type of particles. The reconditioned green particles performed better than those made of

174 dried and moistened particles, but the statistical analysis taking into consideration the
175 scatter of the data shows that these differences between the two groups are not
176 significant.

177 The analysis of the data for 1 minute and 3 minutes pressing time shows a
178 significant influence of the pressing time on IB and TE ($p = 0.011$ and $p = 0.021$
179 respectively), a longer pressing time giving higher IB and TE values, but no significant
180 influence of pressing time on TS.

181 [Table 3]

182 Discussion

183 In general, a low MC during the pressing phase leads to boards with poor mechanical
184 properties, a too high MC again seem to prevent a setting of the dispersion adhesive due
185 to lack of water evaporation and a polymerisation of the vital gluten due to continuous
186 volatilisation of water which lead only to moderate increase of temperature above 100
187 °C in the core of the panel (Metzger 2007, Nordqvist 2012). The results of this study
188 thus agree with the findings of earlier studies by e.g. Kelly (1977) and Khosravi (2015).

189 At a given mean MC the particleboards produced in a conventional way, i.e. dry
190 particles and moistening to adjust the MC to the required level, had a lower IB and TS
191 and a higher TE than the boards produced from green particles. The mean values of IB
192 and TS (2 hours), for example, were respectively 70% and 34% higher when the boards
193 were made from green particles. These results, especially the high TE are an indication
194 for incomplete curing or bonding of the adhesive when applied in liquid form on dry
195 particles.

196 A probable explanation for the poor properties of boards produced from dry
197 particles is that the water applied is located mainly close to the surface of the particles.
198 During the hot-pressing process, this water can evaporate quickly instead of

199 contributing to the softening of the particles and improving the adhesion between
200 particle surfaces. In green particles, this evaporation process into the mat is presumably
201 slower, and the vital gluten can absorb the additional moisture needed for its
202 polymerization.

203 The boards made of green particles with a pressing time of 3 minutes showed an
204 increase in IB with increasing MC. For boards made of dry particles this was not the
205 case, the highest IB was reached at a MC of 20% (series No. 12). The reason may be
206 more incomplete bonding due to the relative high amount of free water. This result is
207 important when developing vital gluten adhesives for the conventional particleboard
208 production process. It seems possible to use green particles with a higher MC as this
209 may be beneficial for the plasticisation of the wood particles and lead to boards with
210 better properties.

211 The results show that it is not possible to reduce the pressing time when using
212 vital gluten by lowering the MC; particleboards made of particles with 15% MC (series
213 Nos. 1-2 and 8-9) showed poor properties. In contrast, the properties of the boards
214 improved with increasing particle MC at a given pressing time. In general, the MC is
215 important to enable moulding of the particle, to regulate the density profile of the final
216 board and to enable heat transfer during pressing (Kelly 1977). When vital gluten is
217 used as the adhesive, the MC in combination with the pressing time have a strong
218 impact on the properties of particleboards as shown in this study and by others, see also
219 Metzger (2007) and Krug and Tobisch (2010).

220 The influence of freezing the green particles with high MC on the properties of
221 the particles itself, and the effects of time and temperature after blending the protein
222 with the particles was not investigated. The temperature and the time span between

blending of the protein with the particles and pressing can also influence the tacking and penetration of the protein-based adhesive system and, therefore, also the bonding.

Conclusion

The influences of wood-particle moisture content (MC), water application, and pressing time on the internal bond strength (IB), thickness expansion (TE) and thickness swelling (TS) were investigated in the manufacture of particleboard with vital gluten powder as adhesive. When using vital gluten as adhesive, which probably involves setting rather than curing, the trade-off between MC within the mat and pressing time becomes crucial. The results show that by using green particles with a high initial MC, the particleboards perform better than those made of dry particles moistened with water to reach the target MC. The results suggest that the successful implementation of vital gluten adhesives in particleboard production may also require changes in the application technique and production process.

Acknowledgment

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Table 1. Design of the experiments. Initial moisture state of particles - type of particles, i.e. green or dried to a MC of about 6%, MC of the particles - moisture content of the particles before pressing, Added water - amount of water added to the particles before adhesive application, Pressing time - effective pressing times without closing and opening time for the particleboard production. Number of samples for each series was 4.

Series No.	Initial moisture state of particles	MC of the particles [%]	Added water [weight%]	Pressing time [min]
1	green	15	0	1
2	green	15	0	3
3	green	20	0	1
4	green	20	0	2
5	green	20	0	3
6	green	25	0	1
7	green	25	0	3
8	dried	6	8	1
9	dried	6	8	3
10	dried	6	15	1
11	dried	6	15	2
12	dried	6	15	3
13	dried	6	20	2
14	dried	6	20	3

Table 2. Particleboard thickness, thickness expansion (TE) and density of the particleboards made of green (Nos. 1 to 7) and dried particles (Nos. 8 to 14), pressed at 160 °C for 1-3 minutes, and thickness swelling (TS) after 2 weeks of conditioning at 20 °C and 65% RH.

Series No.	Thickness after pressing	TE	Density	TS	
				2h	24h
	[mm]	[%]	[kg/m ³]	[%]	[%]
1	12.2	6.3	592	46	51
2	12.1	5.4	578	54	63
3	14.1	22.6	514	36	41
4	12.3	7.0	568	36	41
5	11.9	3.5	598	34	40
6	13.5	17.2	512	26	30
7	12.4	7.4	585	33	41
8	13.0	13.3	474	41	45
9	12.0	4.4	538	44	48
10	13.5	17.2	463	23	25
11	13.5	17.6	471	26	32
12	12.0	4.6	515	23	25
13*	13.4	16.7	496	17	21
14	12.5	8.7	513	24	26

* Pressing time of 2 minutes (the equivalent “series No. 6” has 1 minutes)

Table 3. Statistical analysis to determine the influence of particle type (green or dried) and pressing time (1 or 3 minutes) on internal bond strength (IB, in N/mm²), thickness expansion (TE, in %) and thickness swelling (TS, in %) of the particleboards glued with vital gluten.

Tested series No.	Property	Independent variable	Sample size	Means	Standard deviations
1 – 14	IB	Particle type (green/dried)	7/7	0.22/0.13	0.092/0.083
	TE		7/7	9.9/11.8	7.11/5.85
	TS		7/7	43.7/31.6	10.35/10.55
Without Nos. 4, 11, 13	IB	Pressing time (1/3min)	5/6	0.10/0.24	0.068/0.076
	TE		5/6	15.3/5.7	6.03/1.99
	TS		5/6	38.6/40.2	10.73/14.02

1 331 Figure 1. Vital gluten in its natural form as powder.

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6 333 Figure 2. Internal bond strength (IB) for particleboards made of green particles (series
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8 334 Nos. 1 to 7) and dried particles with water application (series Nos. 8 to 14). The
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10 335 horizontal dashed green lines show the P1 and P2 values of standard EN 312. The box-
11
12 336 and-whiskers plot show median and upper and lower quartiles of the IB.

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Figure 1

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Figure 2