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Effect of temperature during vital gluten adhesive preparation and application on shear-bond strength

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If protein-based adhesives are to become a competitive bio-based alternative to synthetic adhesives, the preparation and application methods have to be considerably improved to reduce process time and thereby improve the economy of the adhesive system. The purpose of this study was to investigate the impact of the temperature during preparation and application on the shear-bond strength of an adhesive based on vital gluten for use in wood applications. Vital gluten was used in its natural form and mixed with water of different temperatures (preparation temperature 0°C or 20°C), and applied on beech veneer at different temperature (application temperature -10°C, 20°C, 60°C and 100°C). Tensile shear-bond strength samples were prepared and tested according to EN 205. The results showed that an increase in veneer temperature during application of the adhesive led to a decrease in the shear-bond strength, but that the preparation temperature of the adhesive had no influence on the strength.

Keywords: application temperature; bond line; preparation temperature; protein adhesive.

Introduction

Proteins or their derivatives have traditionally been used as a basis for adhesives for use in wood applications, but the development of synthetic adhesives has made it more difficult for protein-based adhesives to compete with synthetic adhesives from both an economic and a performance-related perspective [1]. In most cases, the formulations are sensitive to the source of the protein and require time-consuming adjustments leading to low production rates and therefore also high costs [2].

Recently, research has been focused on the use of vegetable proteins because they can be produced in sufficient volume for wood adhesive applications and are reasonable in cost. Although most of the research has been on proteins from soy beans, some work has also been done on other sources, such as canola (rapeseed), wheat
gluten, lupin flour, and zein [2, 3]. Of those, wheat gluten is the cheapest source of protein suitable for the development of wood adhesives [4].

Wheat gluten is derived from whole wheat kernels or flour by a separation of starch and gluten. Vital gluten is gluten isolated and produced at a mild temperature, especially with regard to the drying process, and it therefore still has its ability to form a molecular network with viscoelastic properties [5]. A group of polymeric gluten proteins is called glutenin, and consists of gliadins and glutenins [6]. These molecules are composed of subunits of high molecular weight (HMW) and low molecular weight (LMW), linked together with disulphide bonds [7].

To be used as a wood adhesive, proteins have to be denaturised to achieve an unfolding of the protein and blended with water. The folding and unfolding of proteins depend on local and non-local interactions among the monomers, i.e. the energies and the angles of the peptide bonds [8]. According to the literature, denaturation of proteins in aqueous solution can be achieved with different reagents: guanidine hydrochloride (GuHCl), urea, proteins which are random coils in their native states, salts (other than guanidine hydrochloride), thermal transitions (both reversible and irreversible denaturation), acid and alkaline pH, organic acids, alcohols and dioxane, and detergents.

There is a temperature of maximum “stability” of the protein where the rate of denaturation is a minimum and, with increasing distance from this optimum temperature, the rate of denaturation increases with the increasing temperature [9]. Although from a thermodynamic aspect the effect of an increase in temperature seems quite clear, this is not the case with a decrease in temperature. Earlier studies suggest that this is an effect of hydrophobic side chains which are exposed to solvent water during the unfolding of the protein and of the different solvation of the non-polar groups [9, 10]. Recent studies suggest that the hydrophilic effect is much more important than
the hydrophobic effect in cold denaturation, assuming that the hydrophilic interactions
are the predominant forces for the folding of proteins and enabling protein solubility
[11, 12]. The stability of proteins is probably a dual concept involving both
thermodynamic stability and thermal resistance, and denaturation occurs as thermal
denaturation at high temperatures and cold denaturation at low temperatures [13].

According to the literature, temperature has a strong impact on the properties of
proteins, and this is important when proteins are used as a base for adhesives as it
influences their solubility in aqueous solutions and the reactivity of the adhesive. The
purpose of this study was therefore to investigate the influence on the tensile shear-bond
strength of the temperature during the preparation and application of a vital gluten-
based adhesive aimed for wood applications.

Materials and Methods

Beech (Fagus sylvatica L.) veneers with an initial moisture content (MC) of 6% and a
thickness of 5 mm were conditioned at various temperatures and glued in pairs with a
gluten-based adhesive prepared at different temperatures.

Amygluten powder from Tereos® (Fig. 1) was used as a base for the adhesive.

This gluten was mixed with water in proportions of 1:3 by weight. The temperature of
the solvent during preparation was either 0°C or 20°C, Table 1. The adhesive was
applied within 5 min. after its preparation. The adhesive application was 200 mg/m².

Before application of the adhesive, the veneers were conditioned for 20 min. at
temperature of -10°C, 20°C, 60°C or 100°C. Adhesive was applied to one of the
veneers, and an unglued veneer was then pressed on the top of it for 3 hours under a
pressure of 2.0 MPa at a temperature of 60°C. The moderate setting temperature was
chosen to reduce temperature-related effects such as thermal denaturation of the protein, but at the same time to give a reasonably quick drying of the water.

For comparison, specimens were also glued with a PVAc adhesive (Casco Adhesive 3316). To compare the two adhesives, the PVAc adhesive was diluted to achieve equal dry basis spread amount at comparable tests. It was applied only on samples conditioned at 20°C.

The tensile shear-bond strength specimens were of 500 x 200 x 10 mm in size, prepared according to the standard EN 205 [14]. The bonded area was 20 x 10 mm², Fig. 2.

Before testing, samples were conditioned according to the classifications D1 and D2 of the standard EN 204 [15]. Classification D1 requires one week conditioning at 20°C and 65% relative humidity (RH), and classification D2 requires one week at 20°C and 65% RH, followed by a water-bath at 20°C for 3 hours and a second conditioning one week at 20°C and 65% RH.

For each series, 10 specimens were tested (Table 1). The tensile shear tests were run at a speed of 9 mm/min., and the tensile shear-bond strength and the average amount of wood failure in quartiles of the total area of rupture were evaluated according to EN 205. The average amount of wood failure was determined using an optical microscope.

**Results**

The results of the tensile shear-bond test are presented in Table 1. Fig. 3 shows the median and the upper and lower quartiles of the strength values as a box-and-whiskers plot.
The statistical evaluation shows that there is no significant difference in bond strength between the different test series after conditioned according to the D1 classification (Nos. 1 to 8). Conditioning according to the D2 classification (Nos. 9 to 16) did, however, affect the bond strength. The independent-samples t-test was used to closer examine the results, Table 2.

The statistical analysis shows that there is no significant difference between the D1 and the D2 conditioning. For the series Nos. 1 to 8 conditioned according to classification D1 neither the preparation nor the application temperature affect the shear-bond strength. After a dry-wet-dry conditioning cycle according to classification D2, however, the application temperature does significantly affect the shear-bond strength. Although, the preparation temperature has no significant influence.

The sample conditioned according to classification D1, with the vital gluten-based adhesive showed on average a bonding strength 18% lower than the PVAc adhesive but 21% higher than that required by the standard. In the case of the samples conditioned according to classification D2, the vital gluten-based adhesive showed on average values 40% lower than those given by the PVAc adhesive, at the minimum level required by the standard, see Fig. 3.

**Discussion**

In contrast to prior studies on wheat protein as an adhesive for wood products [6, 16, 17] where the wheat protein was pre-treated before application, vital gluten was used in this study in its natural form and set at a relatively low temperature to avoid changes in its chemical and physical structure not related to the shear-bond preparation and application temperature.
In general, changes in the chemical and physical structures of the protein during preparation are desired, as they improve the bonding strength of the adhesive. In this study, an attempt was made to achieve a denaturation of the protein simple by controlling the temperature under preparation and application of the adhesive. A low temperature should increase solubility and lead to an unfolding of the wheat protein when solved in water. The mild curing conditions after application on wood assume that the adhesive system can be interpreted as being a dispersion adhesive which is not curing but is setting by evaporation of the solvent [18].

The results demonstrate that regulating the temperature during adhesive preparation and application may be essential for the development of protein-based wood adhesives. (1) The time-consuming and energy-extensive pre-treatment processes may thus be reduced [16]. (2) The selection of the protein is important. Vital gluten, for example, can lead to a viscoelastic network [5]. (3) The poor solubility of the wheat protein can be overcome and can be an advantage after setting of the adhesive. (4) Using lower temperature during adhesive preparation and application seems to have no negative impact on the complex protein structure and functionality. The proteins were eventually more unfolded which is important to achieve both strong intra-bonding (within the protein complex) and inter-bonding (between the protein and the substrate). The results showed that a mild temperature during application seemed to have a positive impact on the water-resistance of the bond-line.

Within the different series tested according to classification D1 the variation in the bonding strength was small, and the preparation and application temperatures had no significantly influence on the bonding strength. There was however a large variation in the extent of wood failure within this group. Specimens conditioned according to classification D2 showed a decrease in bonding strength with increasing application
temperature and a smaller variation in the extent of wood failure. The adhesive blended at 20°C performed worse with increasing application temperature regardless of how the specimens were conditioned before the tensile shear test.

The Standard EN 205 requires a minimum shear-strength value for the bond strength for the D1 and D2 classifications of 10 MPa and 8 MPa, respectively. The vital gluten adhesive reached the D1 level for all combinations of adhesive preparation and veneer temperatures. The D2 level was only reached with veneer temperatures of -10°C and 20°C. These results suggest that a decrease in temperature effectuates an unfolding of the protein resulting in the exposure of reactive bonds [11].

In general, the results show a remarkably good bonding strength of vital gluten and the ability to reach the required standard even after water immersion, although the performance is lower than that of PVAc adhesive, Ref1 and Ref2. Nevertheless, the values of bonding strength are comparable to those presented in the literature, e.g. Nordqvist [17]. In the present study, however, vital gluten was used in its natural way or cold-dissolved and curing was avoided, whereas in the studies presented in the literature it was treated before application in different ways and curing was achieved by using higher temperatures [16, 17, 19]. Curing of the protein-based adhesive should lead to polymerisation and thus a greater water resistance of the bond line.

Cold-dissolution and application on a cool substrate seem to give the protein the ability to unfold and develop bonds (inter-bonding) and this performance is much better at higher humidity. At the same time, vital gluten seems not to lose its ability to build a viscoelastic network (intra-bonding) which seems responsible for the good performance regarding the bonding strength in general. These assumptions are strengthened by the fact that a higher temperature during application lead to a strong decrease in bonding strength after water immersion, which indicates that a larger amount of hydrophilic
groups are accessible to water so that the protein applied at a higher temperature was not able to develop proper inter- or intra-bonds.

Conclusions

In this study, the impact of preparation and application temperature on the shear-bond strength of vital gluten blended with water and applied as a dispersion adhesive on beech veneer was studied. The adhesive was prepared at two different temperatures and applied on veneers at temperatures from -10°C to 100°C, and the specimens were thereafter conditioned according to the classifications D1 and D2 in the standard EN 205 before the tensile shear-bond strength was evaluated. The results show that the application temperature influences the bonding strength after a water immersion according to classification D2. Here, an increase in veneer temperature during application of the adhesive considerably decreased the shear-bond strength. The results not only demonstrate that wheat protein has a strong potential for use as a wood adhesive, but also that the use of plant proteins in the area of wood adhesives is still in an initial state.

Acknowledgement

We would like to thank Tereos® for providing the wheat protein Amygluten®. Support from the COST Action FP1407 “ModWoodLife” project (Understanding wood modification through an integrated scientific and environmental impact approach), is gratefully acknowledged.

References


Table 1. Tensile shear-bond strength results for specimens glued with vital gluten adhesive (Nos. 1 to 16) or PVAc adhesive (Ref₁ and Ref₂) at different preparation and application temperatures, conditioned according to EN 204, and the mean shear-bond strength (standard deviation) and wood failure of specimens tested according to EN 205.

<table>
<thead>
<tr>
<th>Series No.</th>
<th>Number of specimens</th>
<th>Temperature of the adhesive [°C]</th>
<th>Temperature of the veneer [°C]</th>
<th>Type of conditioning</th>
<th>Shear-bond strength [N/mm²]</th>
<th>Wood failure [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>10</td>
<td>0</td>
<td>-10</td>
<td>D1</td>
<td>12.5 (1.4)</td>
<td>50</td>
</tr>
<tr>
<td>2</td>
<td>10</td>
<td>20</td>
<td>-10</td>
<td>D1</td>
<td>12.4 (1.1)</td>
<td>75</td>
</tr>
<tr>
<td>3</td>
<td>10</td>
<td>0</td>
<td>20</td>
<td>D1</td>
<td>11.1 (1.4)</td>
<td>75</td>
</tr>
<tr>
<td>4</td>
<td>10</td>
<td>20</td>
<td>20</td>
<td>D1</td>
<td>12.4 (1.3)</td>
<td>0</td>
</tr>
<tr>
<td>5</td>
<td>10</td>
<td>0</td>
<td>60</td>
<td>D1</td>
<td>12.1 (1.5)</td>
<td>25</td>
</tr>
<tr>
<td>6</td>
<td>10</td>
<td>20</td>
<td>60</td>
<td>D1</td>
<td>11.5 (1.1)</td>
<td>0</td>
</tr>
<tr>
<td>7</td>
<td>10</td>
<td>0</td>
<td>100</td>
<td>D1</td>
<td>12.8 (0.9)</td>
<td>25</td>
</tr>
<tr>
<td>8</td>
<td>10</td>
<td>20</td>
<td>100</td>
<td>D1</td>
<td>12.2 (1.7)</td>
<td>25</td>
</tr>
<tr>
<td>Ref₁</td>
<td>10</td>
<td>20</td>
<td>20</td>
<td>D1</td>
<td>14.8 (1.2)</td>
<td>50</td>
</tr>
<tr>
<td>9</td>
<td>10</td>
<td>0</td>
<td>-10</td>
<td>D2</td>
<td>8.2 (1.8)</td>
<td>25</td>
</tr>
<tr>
<td>10</td>
<td>10</td>
<td>20</td>
<td>-10</td>
<td>D2</td>
<td>10.2 (2.1)</td>
<td>25</td>
</tr>
<tr>
<td>11</td>
<td>10</td>
<td>0</td>
<td>20</td>
<td>D2</td>
<td>9.5 (2.1)</td>
<td>25</td>
</tr>
<tr>
<td>12</td>
<td>10</td>
<td>20</td>
<td>20</td>
<td>D2</td>
<td>9.0 (1.9)</td>
<td>0</td>
</tr>
<tr>
<td>13</td>
<td>10</td>
<td>0</td>
<td>60</td>
<td>D2</td>
<td>8.3 (2.5)</td>
<td>0</td>
</tr>
<tr>
<td>14</td>
<td>10</td>
<td>20</td>
<td>60</td>
<td>D2</td>
<td>5.8 (2.1)</td>
<td>0</td>
</tr>
<tr>
<td>15</td>
<td>10</td>
<td>0</td>
<td>100</td>
<td>D2</td>
<td>6.9 (2.2)</td>
<td>25</td>
</tr>
<tr>
<td>16</td>
<td>10</td>
<td>20</td>
<td>100</td>
<td>D2</td>
<td>5.7 (2.0)</td>
<td>25</td>
</tr>
<tr>
<td>Ref₂</td>
<td>10</td>
<td>20</td>
<td>20</td>
<td>D2</td>
<td>13.3 (2.0)</td>
<td>50</td>
</tr>
</tbody>
</table>
Table 2. Statistical analysis of the significance of the effect on the tensile shear-bond strength of the type of conditioning (D1 and D2), the preparation temperature (0°C and 20°C) and the application temperature (>40°C and <40°C).

<table>
<thead>
<tr>
<th>Tested series No.</th>
<th>Independent variable</th>
<th>Sample size</th>
<th>Means</th>
<th>Standard deviations</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 – 8 (class. D1)</td>
<td>Preparation temperature</td>
<td>4/4</td>
<td>12.11/12.11</td>
<td>0.72/0.40</td>
</tr>
<tr>
<td></td>
<td>Application temperature</td>
<td>4/4</td>
<td>8.25/7.66</td>
<td>1.06/2.26</td>
</tr>
<tr>
<td>9 – 16 (class. D2)</td>
<td>Preparation temperature</td>
<td>4/4</td>
<td>12.08/12.14</td>
<td>0.65/0.51</td>
</tr>
<tr>
<td></td>
<td>Application temperature</td>
<td>4/4</td>
<td>9.22/6.69</td>
<td>0.84/1.22</td>
</tr>
</tbody>
</table>
Figure 1. Wheat protein (vital gluten) in its natural form as a white-greyish powder.
Figure 2. Specimen for tensile shear-bond strength test according to the standard EN 205 [14].

- $l = 150 \pm 5$ mm total length of specimen
- $b = 20.0 \pm 0.2$ mm width of specimen
- $l_2 = 10.0 \pm 0.2$ mm length of overlap (length of tested bond line)
- $s = 5.0 \pm 0.1$ mm thickness of the veneer
Figure 3. Box-and-whiskers plot showing the means (red lines) and the upper and lower quartiles (vertical dashed lines) of the shear-bond strength of specimens glued with vital gluten adhesive (Nos. 1 to 16) or PVAc adhesive (Ref₁ and Ref₂). Horizontal green dashed lines show the values of the D₁ and D₂ classifications required by standard.