Bond Strength of Different Wood-Plastic Hybrid Components Prepared through Back Injection Moulding

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Golf tees with a plastic head and wooden shaft were prepared by back injection moulding of a beech wood (Fagus sylvatica) shaft with different polymers (i.e. ionomer, polypropylene, and polyamide). In order to facilitate adhesion between the polymer melt and the wood surface, the wooden shafts were pre-treated with different primer substances, including a commercially available primer for ABS edges, a 10% solution of alkyl ketene dimer (AKD) in toluene and tumbling lacquer. The mechanical strength of the wood-plastic interphase was characterized by applying a pull-out test. Bond strength values of more than 9 N/mm² were observed for polyamide, whereas ionomer and polypropylene specimens achieved values between 0.7 and 3.8 N/mm². Surprisingly, the used primers failed to improve interfacial adhesion with the exception of the ionomer sample pre-treated with the commercial ABS-primer. Although light microscopy and SEM revealed some differences in the penetration behaviour of the different polymers as well as in the extent of plastic wood deformation imposed during injection moulding, the chemical nature of the polymer seems to be the most important determinant for the bond strength of wood-plastic hybrid components.

Keywords: Bond strength; Pull-out test; Wood-plastic components; Wood-plastic interface

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INTRODUCTION

Free shaping, the bonding of wood to non-wood materials as well as compounds and material combinations, are required for new component designs and concepts in the wood technology sphere. Wood plastic composites (WPCs) are used in a variety of applications (Schwarzkopf and Burnard 2016). However, most of the WPCs are not designed for load-bearing constructions because of their limited strength properties and creeping behaviour. In regards to bending, tension, and compression, wood shows excellent mechanical characteristics in the axial direction. When properly applied, modern wood composites are competitive to metals and fibre-reinforced materials. Wood is an abundant, carbon-neutral, and renewable resource. Additionally, raw material costs are low and especially in Europe, high quality wood material is widely available. Decades of experience in aeronautical, nautical, and even in automotive engineering provide abundant proof that wood and wood composites are a reliable construction and engineering material. Wood can broaden the material portfolio in automotive engineering and can help improving the CO₂-balance with a lower weight and lower cost (Leitgeb et al. 2016; Müller et al. 2017). The application of wood in automotive engineering includes the development
of wood-based hybrid materials and components. Hybrid-materials or components are composites consisting of two or more different constituents. Bonding and joining of such hybrid-components, specifically the interface between those components, is assumed to be a key issue in the future development of wood-based hybrid materials. Besides the bonding and gluing of metal or plastic inserts, simple injection moulding, back injection moulding, or over-moulding processes are thought to be an appropriate technology to integrate engineered wood products (EWP) and/or engineered wood components (EWC) into wood-based hybrid material structures.

When joining wood and plastic in EWP or EWC, a load transfer needs to take place from the wood element into the surrounding elements. In the case of wood-plastic hybrid elements, interfacial bond strength as well as interpenetration of the plastics into the hierarchical cell structure of the wood are important research questions. For wood-plastic composites (WPCs), the mechanical stress transfer has already been investigated on macroscopic (Gacitua and Wolcott 2009) and microscopic levels (Sretenovic et al. 2006). Micrographs of the extruded WPCs clearly show that the plastic material is interpenetrating the wood particles to a large degree. This is due to the high pressure and high temperature within the front part of the extruder, where the low viscosity polymer melt is compressing the wood cells and penetrating the inner wood structure. Penetration of the molten plastic into the cell structure and deformation of the wooden cells are influencing the mechanical stress transfer and the mechanical interlocking that is taking place (Gacitua and Wolcott 2009).

The treatment of wood fibres by maleated polyolefin coupling agents and maleic anhydride were investigated by Correa et al. (2007) and Keener et al. (2004). The two studies found that the couplers increase the tensile strength and impact properties of the wood fibre, polyethylene composite, to a large degree. Furthermore, the interfacial bonding between wood fibres and PP-matrix can be improved by using maleic anhydride couplers. Consequently, mechanical properties of the composite might be increased (Karnani et al. 1997; Thwe and Liao 2003; Kuo et al. 2009).

Therefore, the mechanical properties of WPCs are not only determined by the content, length, and alignment of fibres and the mechanical properties of the plastic matrix, but also by the interfacial behaviour and stress transfer from the plastic matrix to the fibres (Sretenovic et al. 2006). This stress transfer is influenced by coupling agents used in WPCs to improve bonding between the hydrophobic and nonpolar matrix material and the hydrophilic surface of fibres. In addition, the stress transfer is determined by the interpenetration of the plastic material into the porous structure of wood, and therefore mechanical interlocking. Furthermore, the penetration behaviour of the thermoplastic matrix into the wood structure is influenced by the plastic deformation of the cell walls (densification of the surface layer), which also affects the mechanical and physical behaviour of the wood fibres (Gacitua and Wolcott 2009). Mechanical properties of the WPCs are well studied. However, the stress transfer between wood and plastic elements, as occurring in over-moulded wooden inserts and outserts, has hardly been investigated so far.

In this study, golf tees with a plastic head and wooden shaft were produced. In the process, wooden shafts were over-moulded with different plastics in an injection mould (Fig. 1). The interfacial bond strength of three wood-plastic components over-moulded with three different plastics was determined by applying a pull-out test. Before the injection moulding, the wooden rods were pre-treated with different coupling agents. Incident light microscopy and SEM (scanning electron microscopy) were used to investigate the interface
between the wooden shaft and the plastic head. The interpenetration of the plastic into the cell structure, and also the fractured surface of the wooden rods after testing, were examined. During the production process, the wooden rods were heavily densified due to the high pressure in the injection mould. Different mechanisms are responsible for the interfacial shear strength of the plastic and wood elements. On the one hand, it is assumed that mechanical anchoring plays a role through the penetration of the plastic into the wood structure. On the other hand, it is assumed that the high compaction, and resulting enlargement of the surface, also contributes positively to the adhesive strength.

EXPERIMENTAL

Industrially manufactured beech wood (*Fagus sylvatica*) rods with a total length of 52.1 mm and a diameter of 4.5 mm were used for the experiments (Fig. 1). The surfaces of the wood rods were treated with different primers: a) reference – untreated; b) primer A: AKD-toluene solution with 10% alkylketene dimer (Kemira Chemie, Krems, Austria); primer B: Intrafol L1140/31 (H.B. Fuller, Wels, Austria); and primer C: conventional solvent based tumbling lacquer (confidential company property, IB Steiner, Spielberg, Austria).

For injection moulding experiments, the following plastic materials were used: polypropylene (Daplen KSR 4525, Borealis, Vienna, Austria), polyamide 6 (Grilon BZ 3/2, EMS-Chemie AG, Domat, Switzerland), and ionomer (Surlyn 1652, DuPont, Berlin, Germany).

![Fig. 1. Top view and longitudinal section of the golf tee used in the experiments](image)

Pre-Treatment and Injection Moulding

The pre-treatment of the specimens was performed only for primers A and B. The lacquered wooden rods (primer C) were provided by an external company. Before pre-treatment, the wooden rods were stored in a standard climate of 20 °C and 65% relative humidity. Each primer was filled in a beaker to a liquid level of 15 mm from the bottom of the beaker. The wood rods were dipped into the primer solutions with their blunt end for exactly 3 s. To guarantee that the same quantity was applied, the weight of 7 selected samples was examined with a lab balance (Sartorius, CPA225D, Göttingen, Germany) before and exactly 60 s after treatment. For drying and curing, the treated rods were put into a styrofoam plate. The average mass for primer A was 0.04 + 0.003 g, and the average mass for primer B was 0.09 + 0.007 g, which consists of an applied amount of 199.1 g/m²
(primer A) and 486.3 g/m² (primer B). Since the lacquered rods were received already in their finished state, no application quantity can be provided for primer C.

The injection moulding process runs fully automated. In the first step, the geometry of the rods was checked by a camera system and automatically oriented fully to ensure that the moulding tool was in the correct position. In the second step, the shafts were transported to a transfer plate where they were picked up from a robot that inserted them directly into the moulding tool. After the mould was closed, the wooden shaft was over-moulded through the use of an ENGEL VC 200/50 (Schwertberg, Austria) injection moulding machine (screw diameter of 25 mm) with the handling robot (ENGEL ERC 23/0-F, Schwertberg, Austria). For the material ionomer the temperature of the hot runners was set to an average value of +203 °C. The volumetric flow rate was 15 cm³/s to 40 cm³/s at an injection pressure of 550 bar. The cycle time for the ionomer was 39 s. The golf tees with polypropylene (PP) were produced with an average hot runner temperature of +250 °C. The volumetric flow rate was 15 cm³/s at an injection pressure of 300 bar. The cycle time was 41 s. For polyamide (PA 6), the average hot runner temperature was set to +256 °C. The volumetric flow rate was 40 cm³/s at an injection pressure of 410 bar. The cycle time for PA 6 was 40 s.

![Fig. 2. Aluminium bracket (50 x 50 x 20 mm³) with a negative form of the plastic cup in the centre to fix the golf tee](image)

**Mechanical Experiments**

All mechanical experiments were performed on a universal testing machine (Zwick/Roell Z100, Ulm, Germany). An aluminium bracket (50 x 50 x 20 mm³) with a negative form of the plastic cup in the centre was manufactured by means of a CNC machine (Homag BMD110/Venture 115 B, Herzebrock-Clarholz, Germany) (Fig. 2). Prior to the mechanical testing, the bottom of the wooden shaft (opposite the plastic head) was glued into a pre-drilled beech wood bracket (50 x 50 x 20 mm³; 15 mm depth of the drill hole) by means of a polyurethane glue from Henkel CEE GmbH (LOCTITE HB S309 PURBOND, Sembach, Switzerland). Clamps originally designed for testing internal bond strength of the particle and fibre boards (ÖNORM EN 319 2003) were used to attach the
aluminium and wooden bracket to the machine. The clamps were equipped with a cardan joint to guarantee that no bending moments were induced during the mechanical experiments. A pre-force of 15 N was applied before the pull-out test was started. The constant cross head speed of 5.75 mm/min was chosen to ensure a maximum load within 60 +/- 30 seconds. After a 12 mm deformation the pull-out test was stopped. Bond strength (BS) was calculated as follows,

\[ BS = \frac{F}{A} \]  

(1)

where \( F \) corresponds to the peak force (N) and \( A \) is the calculated overall interface area (mm\(^2\)) of the wooden shaft embedded into the plastic head (Fig. 1).

**Scanning Electron Microscopy (SEM) and Light Microscopy**

After mechanical experiments, two samples per combination of the material were used for microscopic inspection by means of SEM (Hitachi TM3030, Tokyo, Japan). The fracture surface was examined at the wooden rods after the plastic head had been removed in the mechanical experiment. For high resolution cross section images and for determining the densified cross-sectional area, the golf tees were cut with a double-bladed circular saw perpendicular to the wooden shaft as shown in Fig. 3. The cutting section was afterwards cut with a razor blade to get a smooth surface.

The wooden rods were strongly compressed during the injection moulding process. To specify this densification after the injection moulding, the reduced cross-sectional area was divided by the original cross-sectional area. For the determination of the densification level, seven samples were selected of each type of plastic. The cross-sectional area of these samples was photographed using an incident light microscope from Carl Zeiss Microscopy GmbH at a 50x magnification (Stemi 2000-C, Jena, Germany). The cross-sectional areas were then determined with the image evaluation software ImageJ (ImageJ 1.52a; National Institutes of Health, Bethesda, USA). The compaction was calculated as follows,

\[ D = 100 - \left( \frac{A_{om}}{A_{real}} \right) * 100 \]  

(2)

where \( D \) (%) is the densification, \( A_{om} \) (mm\(^2\)) is the measured cross section of the rod in the over-moulded area as determined from the microscopic images, and \( A_{real} \) (mm\(^2\)) is the calculated cross section of the same rod assuming a circular diameter.

**Fig. 3.** Cut beech wood rods perpendicular to the grain for high resolution images and for determining the cross-sectional area for densification
RESULTS AND DISCUSSION

Figure 4 shows the differences in interfacial shear strength of wood-plastic hybrid components prepared in this study. For all three polymer types, remarkably high strength values were observed for reference samples lacking a surface treatment. Only for the ionomer, the treatment with Intrafol (2.36 + 0.17 N/mm²) resulted in a minor, but statistically significant, strength improvement as compared to the reference samples (2.13 + 0.13 N/mm²). Treatment with AKD and coating with tumbling lacquer resulted in a decreased interfacial bond strength of 1.62 + 0.08 N/mm² and 1.69 + 0.11 N/mm², respectively. Although PP is a very hydrophobic material, the interfacial bond strength of the reference samples demonstrated that sound bonding to the hydrophilic wood surface was taking place (3.46 + 0.24 N/mm²). The highest interfacial shear strength was observed for wood reference specimens over-moulded with the polyamide material (9.01 + 0.63 N/mm²). The excellent adhesive strength can be explained by the hydrophilic character and high strength of this kind of plastic material. The treatment of the wood surface with Intrafol (6.33 + 1.25 N/mm²) and coating with tumbling lacquer (7.19 + 1.22 N/mm²) caused a significant strength reduction. Lowest adhesion between polyamide and wood surface was observed for AKD treated wooden shafts (5.84 + 0.3 N/mm²).

![Graph showing interfacial bond strength](image)

**Fig. 4.** Shear strength of the wood-plastic interface of the different golf tees with beech wood rods treated with AKD, Intrafol, and tumbling lacquer and polypropylene, polyamide, and ionomer plastic head

Figure 5 shows a cross section of a sample consisting of an untreated wooden rod over-moulded with ionomer. The two different materials are separated by a small interphase. This means that in the over-moulding process, some plastic material is interpenetrating the outer cellular structure of the wood through sliced vessels and compressed wooden cells. In addition, the SEM analysis showed that the melt flow was mainly in the tangential direction, which is consistent with the results of Gacitua and Wolcott (2009). Therefore, minor mechanical interlocking can be achieved.

In the case of WPCs, where the wooden particles and the plastic material are usually compounded in a first production step, a much better penetration of the plastic matrix into the lumens can be observed. Due to the low viscosity, high pressure, and residence time in the extruder, the melted thermoplastic is interpenetrating the wood structure (Balasuriya et al. 2002; Gacitua and Wolcott 2009). Sretenovic et al. (2006) showed that the mechanical
interlocking of the plastic into the wood structure has an important effect on the stress transfer from the plastic matrix to the wooden particles. Especially at the end grain surfaces of the particles, the plastic is deeply penetrating the wood structure. Additionally, mechanical properties and creeping behaviour of the WPCs can be influenced to a large degree by varying the content and sort of the coupling agents.

![SEM micrograph of the cross section at the wood-plastic (ionomer) interface. Cell lumens filled with ionomer are indicated by white arrows.](image)

Fig. 5. SEM micrograph of the cross section at the wood-plastic (ionomer) interface. Cell lumens filled with ionomer are indicated by white arrows

It is assumed that in the case of back injection and over-moulding, completely different mechanisms are acting in comparison to the extrusion of WPCs. The different surface treatments used in this study were meant to act as a coupling agent to improve the interfacial adhesion between the plastic and the wooden component. However, the different treatments resulted in a significant reduction of the interfacial bond strength in almost any case. With all the plastics used, the highest bond strengths were achieved on untreated wood surfaces, excepting ionomer, where the variant primed with Intrafol showed the highest strength values.

The high temperature and pressure applied during the injection moulding results in a heavy plastic deformation of the wood structure as seen in Figs. 5 and 6. With the over-moulding of the wooden rods by using ionomer, PP and PA resulted in a reduction of the cross sectional area of 18.43 + 6.35%, 9.31 + 2.44%, and 34.93 + 3.81%. It has to be assumed that the viscosity of the liquid plastic melt and the conditions in the mould (temperature and pressure) are responsible for the different degrees of densification of the wood elements. However, no direct relation between pressure (Ionomer 550 bar, PP 300 bar, and PA 410 bar) in the mould and densification was observed.
It is clearly shown in Fig. 5 that especially the vessels of the beech wood were highly compressed perpendicular to the grain, for the ionomer and PA. Plastic deformation of beech wood to this extend corresponds to a pressure level of more than 10 N/mm² at room temperature (Müller 2003). The softening of wood is strongly related to the glass transition of lignin (Back and Salmén 1982). Regardless of the exact environmental conditions and the heat transfer from the molten plastic into the wood, an increase in pressure in the injection mould and a simultaneous increase in temperature leads to increasing compression. Therefore, higher densification levels can be expected at the same pressure at higher temperatures. Due to the high pressure of the moulding process, the plastic material is tightly pressed onto the wood surface. Wood rays are oriented perpendicular to the grain and are acting as reinforcing elements. In between the wood rays, the vessels and tracheids are heavily compressed, whereas in the area of wood rays, hardly any densification was observed. Therefore, the wood rods are irregularly deformed, which results in increasing of the roughness of the wooden surface (see Fig. 5). It is assumed that an increase of the surface roughness has also had an effect on the mechanical properties of the wood-plastic element.

High densification by applying pressure (e.g. during wood machining or gluing together) is causing a so-called weak boundary layer. The formation of a weak boundary layer was also expected for the wood rods in the injection moulding process. However, a microscopic analysis of the fracture surface identified no shallow wood failure, which would be expected if a weak boundary layer, but exclusively at the wood-plastic interface.

![Fig. 6. Plastic deformation of the golf tees with beech wood rods and ionomer, polypropylene, and polyamide plastic head](image)

Figure 7 shows a comparison of SEM images with all types of plastics and primers after the pull-out test without the plastic head. The penetration into the cell lumens was observed only for untreated rods over-moulded with the ionomer and PP. By contrast, for PA and for treated rods no penetration of the plastic material into the wood cell structure was observed. Beech wood rods treated with Intrafol showed similar results to the lacquered surfaces. Tumbling lacquer acts like a sealing of the wooden surface and Intrafol seems to be a filler and penetrate into the porous wood structure. After curing, the primers prevent the melt from penetrating the wooden structure, which might lead to a decrease in the bond strength. Anyhow, the different primers form a separate layer between the wood and plastic material. These layers prevent the penetration of the polymers into the wood surface. Smith et al. (2002) also found, that thermoplastics penetrate into the wooden structure and the mechanical interlocking depends on the roughness and porosity of the
wood surface, as well as on the viscosity of the molten thermoplastic. Also, a much better interlocking can be achieved with a longer process time, which means, the molten plastic should be left longer on the wooden surface.

Slightly lower densification levels were observed for the rods treated with primers. One possible explanation to this observation is that heat transfer from the plastic melt to the wood surface is altered by the primer. A lack of mechanical interlocking and a smoother surface are possible explanations for lower mechanical properties of the treated samples in comparison to untreated ones. It is possible that the boundary layer formed by the primer acts as a sliding surface, which would also explain a reduction in strength.

Untreated wood surfaces are much more reactive than treated ones. In the case of PA, the chemical interaction in the form of secondary valence forces such as hydrogen bonds could be responsible for the better adhesion of untreated samples.

**Fig. 7.** SEM images of wood-plastic components after the pull-out test, a) ionomer untreated, b) PP untreated, c) PA untreated, d) ionomer with AKD, e) PP with AKD, f) PA with AKD, g) ionomer with Intrafol, h) PP with Intrafol, i) PA with Intrafol, j) ionomer with tumbling lacquer, k) PP with tumbling lacquer, l) PA with tumbling lacquer
In general, the treatment with AKD and tumbling lacquer showed the lowest values in the bond strength. Although it is known that AKD is able to covalently attach to cellulose via a beta-keto ester bond (Kumar et al. 2016), this reaction usually requires the presence of a catalyst, elevated temperatures, and a reaction time of several hours in order to achieve a substantial degree of substitution. Due to the short cycle time of 40 s on average, these conditions were not fulfilled in the present study. Therefore, it is assumed that the AKD did not, or only to a small extend, chemically react with the wood substrate. This means, that AKD was only physically attached to the wood surface.

Due to its molecular structure comprising of the long alkyl chains, AKD is inherently hydrophobic, which limits the formation of secondary valence forces between the individual AKD molecules to very weak London forces. As a result, AKD shows a soft and waxy consistency in a dry state, thus providing an inert, hydrophobic surface layer on the wood substrate, which the plastic melts are largely unable to adhere to during injection moulding. Furthermore, Zhang et al. (2007) reported that AKD decomposed into fatty acids when heated to a temperature of 100 °C for a few minutes. Due to the high temperatures of the polymer melts used in the present study (approx. 200 °C for ionomer and 250 °C for PP and PA), thermal decomposition of AKD may have occurred as well. However, this was not analysed in more detail.

Additionally, another hypothesis for increasing the BS at a higher densified wood is the swelling pressure. Increased temperature in the mould during the injection process causes a reduction in moisture content. Especially, at the surface of the wooden rods, where the temperature of the plastic melt reached more than 200 °C, a reduction of the moisture content must be assumed. The change in moisture content was not measured. However, it can be assumed that after a cooling down and conditioning of the rods in the climate chamber before mechanical testing, the wooden rods took up moisture. Water uptake of wood is always accompanied with swelling. The tip of the highly densified wooden rods is surrounded by the plastic heads. The plastic heads will block free thickness swelling of the wooden rods, which causes a swelling pressure. Swelling pressure of wood increases with the density (Niemz et al. 2002). The same is assumed for the highly densified wood. Therefore, high swelling pressure can ensure a high contact pressure between wood and plastic. Higher densification was observed for ionomer than for the PP. This indicates a higher swelling pressure and therefore higher bond strength for rods over-moulded with ionomer. However, PP showed higher BS. On the one hand, differences in BS between the ionomer and PP were low. On the other hand, this can probably be explained by the lower rigidity of the ionomer, which has a low E-modulus of 0.16 GPa compared to the PP with a much higher E-modulus of 1.01 GPa.

However, mechanical performance is thought to be mainly influenced by interfacial adhesion between the plastic and the wooden component and the plastic deformation of the over-moulded wood structure.

CONCLUSIONS

1. The over-moulding process is a useful procedure to join wood-plastic hybrid components. Interpenetration of the plastic melt into the wooden structure can explain the mechanical properties of the wood plastic elements only to a small extend.
2. Primers used in the study did not improve the mechanical properties. This can be explained by chemical and mechanical effects. To improve bond strength by means of a primer, longer reaction times and/or other chemical substances are demanded. The highest bond strength was achieved using wooden rods without a surface treatment for all of the plastic materials investigated.

3. Rods over-moulded with polyamide showed the highest bond strength. This can be explained by the high strength of the polyamide and the chemical interaction between this plastic and wood surface. Ductile deformation and change of the surface morphology of the wooden elements (increase of the surface) and swelling pressures are thought to have additional effects on the bond strength.

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