Cellulose fibers and nanofibrils for adhesive reinforcement

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Introduction
In wood industry, adhesive bonding is one of the key process steps. To ensure continuing technological progress in wood industry it is necessary to explore new possibilities of improvement in adhesive bonding. The potential routes for performance improvements suggested by a state of the art report issued by COST Action E13 - Wood Adhesion and Glued Products (Dunky et al. 2002) - focus very much on polymer chemistry, where tremendous improvements have been achieved in past decades. Based on the results achieved in own preliminary experiments, the authors of the present report think that also another well known component of adhesives in wood bonding, usually addressed under the term “filler”, deserves more attention. As is well known from the science of composite materials (e.g. Chawla 1987), the addition of fibrous fillers significantly changes the physical properties of a polymer. Consequently, a large number of studies dealing with the reinforcement of polymers by various types of fibers (e.g. glass fibers, carbon fibers or polymeric fibers) can be found in literature. In contrast, for adhesive bonding of wood, literature on true reinforcing fillers is very scarce.

A recently published study by Richter et al. (2009) reports for the first time on wood adhesives reinforced with cellulose nanofibrils. Two different adhesives, based on polyvinyl acetate (PVAc) and polyurethane (PUR) respectively, were considered. Varying amounts of surface-modified fibrils were added to the adhesive and wood-adhesive assemblies bonded with these modified adhesives were tested. In spite of significant changes observed for cellulose reinforced PVAc composite films characterised in a different experiment in the same study, no significant effects of cellulose nanofibrils on wood-adhesive shear strength were observed neither for PUR nor for PVAc.
In contrast to the study of Richter et al. (2009), the present study focused mainly on the reinforcement of urea-formaldehyde-(UF)-adhesive bonds. UF-adhesives are among the most important adhesives in terms of quantity in wood products industry (Dunky and Niemz 2002, Pizzi 2005, Zeppenfeld 1991). This adhesive type is well known for its pronounced brittleness and tendency to develop microcracks which limits the mechanical performance of UF-bonds. Previous studies on reinforced epoxy, an adhesive typically not used in wood industry, showed that the addition of different fillers primarily improved the toughness of an adhesive bond (e.g. Hunston et al. 1980, Kinloch 2003, Kinloch et al. 2003, Stewart et al. 2007, Soares et al. 2008). Furthermore, a comparative study of a broad variety of wood adhesives (Konnerth et al. 2006a+b, Konnerth et al. 2007) indicated a strong correlation between the toughness of an adhesive and the ultimate shear strength of corresponding wood adhesive bonds. The combination of great industrial importance and limited mechanical performance makes UF an ideal candidate for reinforcement by fibrous fillers. Regarding the latter, pure natural cellulose offers outstanding mechanical properties with a modulus of elasticity of approximately 140 GPa (Sakurada et al., 1962; Nishino et al., 1995) and an estimated strength of up to 10 GPa (Zimmermann et al. 2004). In combination with “green” features such as biodegradability, renewability, sustainability, and favourable carbon-dioxide balance, which all apply to cellulose, there is an evident potential for cellulose-fibers in adhesive reinforcement. This is especially true for nano-scale cellulose fibrils which profit from the full stiffness and strength of pure, almost defect-free cellulose.

Material and methods
For the preparation of cellulose nanofibrils, dissolving grade beech pulp obtained from Lenzing R&D has been used as a starting material. After suspension in water, the pulp was disintegrated mechanically by 20 passes through a laboratory refiner. Fibrillation down to the nano-scale was achieved by running the suspension through an APV-Gaulin high-pressure homogenizer (10 passes at a pressure of 450 bar). Subsequently, the aqueous fibril-suspension was mixed with a urea-formaldehyde-(UF)-adhesive (W-Leim Spezial, Dynea Austria GmbH) present in powder form. To evaluate the effect of fibril diameter on bond strength, the UF-resin was reinforced with both homogenized and untreated pulp fibers respectively. Cellulose content was 5% cellulose per unit weight cured UF in both cases.

In addition to homogenized and untreated beech pulp, regenerated cellulose fibers (Lyocell, Lenzing AG) were used to reinforce a UF-adhesive bond. This material was applied in the form of a non-woven fabric that was impregnated with the adhesive, sandwiched between the two wood adherends, and pressed together. Since the reinforcement by the fabric entailed bond-lines which were about 0.5 mm thick, pure UF-bonds with the same thickness have been prepared as a reference.
Lap joint test specimens according to EN 302-1 (2004) were prepared using both cellulose-reinforced and non-reinforced UF-adhesive. The specimen geometry is shown in Figure 1. Lap-joint testing was performed on a Zwick/Roell Z020 universal testing machine with a testing speed of 1 mm min⁻¹. The overall deformation in the overlapping area of the specimens was recorded by a Zwick Macrosense clip-on deformation sensor.

Fig. 1: Geometry of lap joint specimens according to EN 302-1 (2004)

Results and discussion
As can be seen from figure 2a, the homogenization process leads to the formation of nanofibrillated cellulose with a broad distribution of fibril diameters ranging from about 10-200 nm. The results of lap shear testing are shown in figure 2b. The pure UF-bonds 0.1 mm in thickness showed a lap shear strength of 10.3 ± 0.9 MPa which corresponds well to bond strengths observed for a variety of structural wood adhesives in a previous study (Konnerth et al. 2006a). The addition of 5 wt% untreated pulp fibers had no statistically significant effect on shear strength, whereas the addition of the same amount of homogenized pulp fibers lead to a significant increase of lap shear strength to a value of 13.8 ± 1.4 MPa. At the same time, the reinforced bond-lines showed a significantly higher deformation at failure, suggesting that the UF-adhesive was possibly toughened by the addition of fibrillated cellulose. At a bond-line thickness of 0.5 mm, only reinforcement by the Lyocell fabric resulted in a significant increase in strength compared to non-reinforced UF-bonds.
Bond-line thickness = 0.1 mm
Bond-line thickness = 0.5 mm
None Pulp Pulp hom. Lyocell
Reinforcement

Shear strength (MPa)

Fig. 2a,b: Scanning electron micrograph of beech pulp after high pressure homogenization (a) and lap shear strength of UF-bonds reinforced with different cellulosic materials (b) (None = Pure UF-bond, Pulp/Pulp hom. = UF-bond reinforced with untreated and homogenized pulp respectively, Lyocell = UF-bond reinforced by a non-woven Lyocell fabric). 9 specimens were tested for each group (n = 9).

Conclusions
It was shown that lap shear strength of a UF-bond increased more than 30% due to the addition of 5% homogenized pulp fibers whereas the addition of the same amount of untreated pulp fibers had no statistically significant effect on shear strength. Strength improvement achieved by inserting a non-woven Lyocell fabric into the UF-bond-line amounts to approximately 20% compared to pure UF-bonds. As deformation at failure was also significantly higher for the reinforced specimens, it is assumed that the UF-adhesive was possibly toughened by the addition of fibrillated cellulose and regenerated cellulose fibers respectively. The authors deduce, therefore, that reinforcement of UF-adhesives with cellulosic fibers and nanofibrils presents an up to now little investigated possibility for significant performance improvement, thus opening up new fields of application for UF-adhesives.

References


