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Influence of refiner fibre quality and fibre modification treatments on properties of injection-moulded beech wood–plastic composites

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ABSTRACT

Thermo-mechanical pulp (TMP) fibres made from beech wood were produced using increasing refiner gap widths and thus with increasing fibre length and coarseness. Fibres (60% by weight) were compounded in an internal kneading mixer using high-density polyethylene as the matrix and injection-moulded. Fibre lengths and length/width ratios were determined (a) before processing and (b) after injection-moulding and Soxhlet extraction using the optical FibreShape system. An increase in fibre length resulted in a decrease in water absorption and an improvement in flexural strength and modulus of elasticity of the wood–plastic composites (WPC). However, flexural strength of the WPC with TMP fibres was not improved compared to WPC with wood flour when maleic anhydride-grafted polyethylene (MAPE) was used as a coupling agent. After injection-moulding, differences in length of the various TMP fibre types were minor. Fibre geometry before processing strongly influences the water absorption and flexural properties of the composite. Fibre treatment with emulsified methylene diphenyl diisocyanate (EMDI) resin before compounding was shown to be equally efficient in reducing water absorption and improving flexural strength as the addition of MAPE during the compounding step.

1. Introduction

In the current prEN 15534-1 [1] the term wood–plastic composites or wood–polymer composites (WPC) refers to materials or products consisting of one or more lignocellulosic fibres or flours and one or a mixture of polymer(s). Various wood and natural fibres have been used in the processing of WPC, for example hemp [2], flax [3], and cellulosic fibres [4–6]. Recently, there has been a commercial interest in the use of thermo-mechanical pulp (TMP) fibres for manufacturing of WPC [7–10]. These fibres are produced in a refining process where wood chips are continuously added to a digester and steamed at temperatures between 150 °C and 180 °C and between 3 and 5 bar pressure for several minutes. After softening, the wood is ground between the refiner discs to produce fibres which are discharged through a blow-line where resin is applied, usually, urea–formaldehyde or isocyanate-based resin in the case of medium-density fibreboard (MDF) production. Finally, fibres are dried, collected and formed for hot-pressing of panels. A major advantage of the refiner fibre process is that fibre processing, treatment with additives (for example, emulsified coupling agents), and drying can be achieved in a continuous, cost-efficient process. In addition, refiner fibres display higher aspect ratios than the commonly used wood flour [11] and can therefore potentially provide reinforcement to the thermoplastic matrix in WPC. In contrast to chemically pulped wood, thermo-mechanically pulped or refined wood still contains most of the lignin.

For TMP fibres produced from radiata pine, it has been shown that their surfaces are covered with lignin [12]. Therefore, these fibres were claimed to possess a high potential for bonding with the hydrophobic thermoplastic polyolefins. TMP fibres are slightly weaker than kraft pulp fibres, cheaper to produce and more amenable to surface modification [13]. Regarding TMP fibres made from beech wood, little information pertaining to their surface characteristics is available while research has been focused on properties of MDF panels made with such fibres [14]. Wulf et al. [15] demonstrated that MDF made from beech wood showed predominantly better wetting behavior than MDF panels made from spruce, based on contact angle measurements. X-ray photoelectron spectroscopy (XPS) was also used to corroborate these findings [16].

At present, to the best of our knowledge, no information is available on the influence of processing parameters during the refining process, for example, shape of the refiner discs, gap width of refiner discs, temperature and pressure, on the properties of WPC made with such fibres. Stark and Rowlands [11] used TMP

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fibres in their investigations, however, without varying the processing conditions during refining. Wood chips were processed in a laboratory-scale 300-mm single-disk Sprout-Bauer pressurized refiner with incoming steam pressure of 414 kPa and a constant plate gap width of 0.127 mm.

Processing parameters during refining are expected to have a strong influence on WPC properties as well as on the process ability of fibres, specifically feeding into compounders and extruders. From the MDF production process, it is well known that with increasing the refiner disc gap, coarser fibres are produced. It is also known that with increasing the temperature during thermomechanical pulping of pine wood, fibre length and fibre diameter is decreased while fines content is increased [17]. In general, during commercial MDF production, quality control of the refiner fibres (determination of fibre length and width, fines content) is usually not performed while the quality of the resulting MDF panels is monitored.

In Europe, industrial MDF production is focussed based on softwoods such as pine and spruce. However, beech wood and other hardwood species will become more available in central Europe in the near future; therefore, in the present investigation beech was used as wood species for manufacturing WPC. It was the objective of this investigation to determine the influence of beech wood particle geometry and various modification treatments on resulting WPC properties. Wood flour, wood particles and refiner fibres were prepared from beech wood and processed using a laboratory kneading mixer. The resulting mixtures were granulated and injection-moulded. Modification treatments were chosen based on their applicability in an industrial-scale blow-line in a MDF refiner plant. Modification using emulsifiable isocyanates was identified as one promising route. It has been shown that the isocyanate group can react with the acid groups of maleated polyolefins, and benefits have been obtained with wood flour-reinforced PP by using them together [18]. Pickering and Ji [19] also investigated combinations of isocyanates and maleic anhydride-grafted polypropylene (MAPP). They found that benefits were obtained by combining isocyanates and MAPP with long chain lengths which can be explained by isocyanate bonding with hydroxyl groups not accessible to further coupling with MAPP once saturation is achieved.

In addition, it was our objective to determine the influence of processing on the wood fibre geometry and WPC properties. It was expected that only if high fibre aspect ratio can be maintained during processing, matrix reinforcement is provided. Unfortunately, it is difficult to characterize MDF fibres using traditional fibre fractionation methods (vibrating screen and air jet sieve analysis) because the fibres tend to agglomerate strongly. However, MDF fibre length, width and shape factors can be determined using the Fibrehape system [20–22]. Fibreshape has also been used to determine fibre length and thickness of flax fibres which were isolated from a PP matrix [3]. Measurable size range of the FibreShape system is from approx. 30 μm in fibre width to 30 cm in fibre length but depends on the method and settings of the image acquisition. The particles to be measured are distributed on a transparent foil and automatically scanned using a high-resolution flat-bed scanner. Projection areas are then measured using contour measurement methods (image analysis) and statistically evaluated.

2. Materials and methods

2.1. Materials

Beech (*Fagus sylvatica*) wood particles sourced from Northern Germany were used in this study. Processing is described in the methods section.

For wood particle and TMP fibre treatment, the following materials were used:

- Emulsifiable methylene diphenyl diisocyanate (MDI) resin (I-BOND MDF EM 4330, Huntsman, Everberg, Belgium; 100% solids content); the abbreviation EMDI will be used from now on.
- Water-dilutable, phenol-based resin (Bakelite PF 1279 HW, Bakelite AG, Duisburg-Meiderich, Germany; solids content approximately 48%); the abbreviation PF will be used from now on; used for the production of the middle layer of particleboards type V100 (moisture-resistant).
- Water-repellent wax emulsion (Hydrowax 730, Sasol Wax GmbH, Hamburg, Germany; 55% solids content; anionic/non-ionic emulsifier); abbreviation: Hydrowax.
- Water-based wood oil (Koralan Holzöl, Kurt Obermeier GmbH & Co. KG, Bad Berleburg, Germany; 13% solids content); abbreviation: Koralan.

High-density polyethylene (HDPE; type BorPEX HE2590; melt flow index of 8.5 g/10 min.; density of 0.944 g/cm^3; melting point approximately 160 °C) from Borealis AG was used as the polymer matrix for wood–plastic composites.

A maleic anhydride-modified, carboxylated, linear low density polyethylene (Scona TPPE 1102 PALL, BYK Kometra GmbH, Schkopau, Germany) with a total maleic anhydride content of more than 1.5% (wt.) was used as coupling agent (abbreviation: MAPE).

An oxidized polyethylene wax (Licolub H12 fine grain, Clariant Produkte (Deutschland) GmbH, Gersthofen, Germany) was used as lubricant.

2.2. Methods

2.2.1. Preparation of wood particles and wood flour

Debarked log sections were processed into wood chips using a drum mill (Klöckner, type 120 × 400 H2WT). Wood chips were processed in a cross hammer mill (Condux, type HM 45/60L) with a knife protrusion of 0.45 mm and discharged using a cyclone. The drum mill was then equipped with a sieve insert (sieve openings 8 mm × 60 mm), and the wood particles were sieved and discharged again using a cyclone. The result was a mixture of wood particles, wood flour and wood dust. This mixture was manually sieved and separated into a wood flour fraction (particle size less than 400 μm) and a wood particle section (400–800 μm).

2.2.2. Preparation of thermomechanical pulp (TMP) fibres

Thermomechanical pulp (TMP) wood fibres were processed using a 12 in. (30.5 cm) double disc refiner (Andritz, Graz, Austria). Beech wood chips were steamed at 160 °C for 5 min at 5 bar pressure. Four fibre classes (Fig. 1) were generated by varying the disc refiner distance between 0.15 mm, 0.4 mm, 0.7 mm and 1.0 mm (the terms F0.15, F0.4, F0.7 and F1.0 will be used from now on). Length and width of the fibres increases with refiner disc distance. Fibres were discharged through a blow-line without resin application and dried in a tube drier. Wood moisture content of the fibres after drying was between 8% and 13% (wt.), depending on the fibre class. To separate the dust fraction from the fibres, fibres were sieved manually (sieve openings 630 μm). Fibres were then dried to 3–4% wood moisture content using a drying oven.

2.2.3. Wood particle and TMP fibre treatment

Wood particles and TMP fibres (F1.0 only) were treated with 10% (wt.) of EMDI, PF or Koralan each and with 1% of Hydrowax, taking into account the respective solids contents of the liquids. Due to the small amounts of liquid to be applied to the fibres, the liquids were diluted with 200 ml of water and could then be