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Copy paper as a source of reinforcement for biodegradable composites – Influence of fibre loading, processing method and layer arrangement – An overview

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A B S T R A C T

The study investigates the use of copy paper in poly (butylene adipate terephthalate)-PBAT and polylactide-PLA composites. The influence of paper volume fraction (21, 30 and 48%) on the void content and the composite properties was analysed by using two different compression moulding methods. An increasing fibre content lead to significantly improved tensile characteristics, while the Charpy impact strength showed a toughness decrease for PBAT and an increase for PLA composites. Additionally, layered composites, where either two stiff PLA composite layers enclosed a ductile PBAT composite core or vice versa, were produced with the aim to increase the toughness. Due to the high elongation of PBAT, the insufficient fibre/matrix adhesion and the low fibre length, this goal could not be reached. However, copy paper represents a good reinforcing component achieving a maximum strength of 120 MPa and a tensile modulus of 10.2 GPa at the highest paper volume fraction in PLA.

1. Introduction

Paper is a fibre-made product, where the fibres can be obtained from a variety of plants, such as: deciduous and conifers, straw and grasses. Wood fibres are most frequently used for paper production [1]. The quality and the characteristics of paper are highly dependent on the fibre dimensions, lignin and cellulose content [2]. However, as paper consists of chemically processed fibres, the properties within one paper type are leading to a homogeneous fibre quality which is an advantage over other natural fibres used for composite production. The use of recycled paper fibres as polymer reinforcement could also be a promising approach to reduce paper waste. In Europe, about 54% of used paper is recycled and the rest goes to landfill or is incinerated. Here, the option of establishing an additional recycling route for waste paper has great potential [3]. There are many studies dealing with the processing of wood fibres or recycled fibres from paper and cardboard to composites. Some of them investigated the use of biodegradable matrices. Georgiopoulos & Kontou [4] investigated, for example, the use of different commercial available wood fibres such as Arbocel® and Lignocel® in a biodegradable polyester matrix 'EcovioVR' (BASF; blend of poly (butylene adipate terephthalate) (PBAT) copolyester and polylactide (PLA). Another approach of using biodegradable matrices was carried out by Lopez et al. [5]. The authors produced composites from TPS (thermoplastic starch), reinforced with cellulosic fibres from recovered newspaper and found increased tensile strength and Young's modulus values. A new technique to prepare cellulose fibre-reinforced composites with fibre contents between 10 and 50% is the Aquacomp™ paper-like production process. This processing technique allows a high orientation of fibres with a low degradation compared to composites produced by a dry-mixing process [6,7].

Apart from biobased and/or biodegradable matrices, the most frequently used thermoplastic matrix for wood and paper fibre-reinforced composites is polypropylene (PP). Due to the hydrophobic properties of PP and the hydrophilic properties of cellulose-based fibres, the fibre/matrix adhesion is generally insufficient and delamination can occur. This results in poor composite properties such as low tensile strength values. However, fibre/matrix adhesion can be modified by using different compatibiliser. For example Burgstaller et al. [8], Kazayawoko et al. [9] and Schirp & Stender [10] found a clear improvement of mechanical characteristics of wood fibre-reinforced PP using maleic

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anhydride (maleic-anhydride grafted PP – MAPP) as adhesion promotor.

In today's world, recycling becomes increasingly important as resources become scarcer. Vilaseca et al. [11] recycled kraft fibres from old paper sacks for the preparation of PP composites using MAPP as coupling agent. The results showed an improved incorporation of the fibres, which led to an increase of stress transfer from matrix to fibre. By incorporating 50 mass-% recycled kraft fibres, the tensile strength was doubled, compared to the neat matrix. Baraoulaki et al. [12] investigated the dispersion of recycled paper fibres in a hot polymer solution. Composites were produced by compression moulding with fibre contents ranging from 10 to 40 mass-%. The tensile strength of high density polyethylene (HDPE) and PP composites exhibited a slight decrease with increasing filler content while no clear effect on the Youngś modulus could be observed. Beg & Pickering [13] recycled whole composite structures based on radiata pine (*Pinus radiata*) fibres in a MAPP matrix. Composites, containing 40 mass-% fibres were reprocessed up to eight times. The tensile strength and Young's modulus decreased with the number of reprocessing cycles by 25% and 17%, respectively. However, the mechanical properties were still significantly higher than the ones for the unreinforced PP matrix. Paper and cardboard are a great source for the recovery of wood fibres. Thus, some studies deal with the processing of fibres obtained from old newspapers to composites. Some authors have shown that improved strength values were achieved when MAPP was added to the PP matrix and the fibres led to a clear reinforcing effect [14–17].

Besides fibre quality and choice of matrix material, the characteristics of the composites are additionally influenced by the manufacturing process, and in particular by the applied process parameters. For example, previous studies for regenerated cellulose fibre-reinforced PLA composites have shown that the choice of process parameters in the compression moulding process significantly influences the mechanical properties. There are studies available about PLA composites reinforced with lyocell/PLA hybrid fibre multilayer webs, which were processed on the one hand with a compression moulding technique resulting in poor compaction of the composites [18]. On the other hand the same semi-finished product was processed with an optimised pressing process resulting in well-compacted materials [19]. The poorly compacted samples resulted in a tensile strength of 35 MPa for 20% lyocell 6.7dtex/PLA and 36 MPa for 40% lyocell/PLA, whereas the well compacted samples reached a tensile strength of 72 MPa and 97 MPa, respectively. These results show the importance of well-selected processing conditions.

Only a few approaches are published regarding the use of whole paper sheets as reinforcement in composites. Nordin & Varna [20], for example, investigated the compressive behaviour of phenol–formaldehyde paper composites for their creep and strain recovery and observed a nonlinear viscoelastic behaviour with irreversible strains. Prambauer et al. [21] have shown that commercially available papers such as copy paper, filter paper and newspaper are suitable reinforcing components in compression moulded thermoplastic matrices. A significant improvement of the mechanical characteristics, such as tensile strength, flexural strength and flexural modulus was achieved by using copy paper and newspaper with an increasing paper volume fraction up to 40%. The use of MAPP led to a significant improvement of the fibre/matrix adhesion, compared to unmodified PP [22].

Previous studies have shown that bioinspired composite structures consisting of stiff and more ductile layers lead to improved impact properties [23]. These different layers were produced either by using different matrices or different fibres [23–25]. In the first case more ductile outer layers made from PP/regenerated cellulose fibres enclosed a core of stiff PLA/regenerated cellulose fibres. The impact strength was improved by approx. 36% compared to a composite consisting of a mixture of these materials [24]. In the second case different fibres were used for the different layers in combination with a PLA matrix: long ramie fibres were used as stiff outer layers enclosing a core of long ductile regenerated cellulose fibres. This bio-inspired layer arrangement led to an increase in impact strength of 55% as compared to the layered composite [25].

Within the scope of the investigation in the present study, thermoplastic laminate composites made of PLA and PBAT reinforced with copy paper were produced, and the mechanical composite characteristics were analysed as a function of the fibre volume fraction (21–48%). In addition, the influence of layered structures with different characteristics on the mechanical properties was investigated. The layers were built up from paper/PLA and paper/PBAT and different arrangements were tested with the aim of optimising the mechanical properties.

Tensile properties, impact strength, interlaminar shear strength as well as density and fibre volume fraction were analysed for all composites. Moreover, two different compression moulding techniques were used to produce composites with a different compaction to investigate the influence of processing parameters on paper composites.

The following hypotheses were investigated:

(1) Strength and modulus increase with increasing fibre fraction due to higher strength and modulus values of paper fibres compared to the PLA and PBAT matrices.
(2) The impact strength of PBAT composites decreases while that of PLA increases with an increasing fibre fraction due to the more or less brittle character of paper fibres compared to the matrices.
(3) PLA based composites show higher strength and modulus values due to the higher values of the matrix compared to PBAT and a better assumed fibre/matrix adhesion between paper and PLA.
(4) The layered composite structure consisting of a more ductile core of paper and PBAT enclosed by stiffer outer layers of paper and PLA shows improved impact characteristics.
(5) A lower compaction of the paper layers in the composites results in lower strength and stiffness, and in higher impact strength.

2. Materials & methods

2.1. Materials

For composite production standard copy paper (Xerox Transit, Xerox Austria GmbH, Wien, AT) with a grammage of 80 g/m^2 produced from kraft pulp fibres was chosen. The mean thickness of the paper was determined to be 0.098 mm, which was measured with a micrometer (Micromaster® IP54 from Tesa Technology, Renens, CH). Polymer foils were produced with a thickness of either 100 or 140 µm with a width of 18 cm from polylactide - PLA (type Ingeo 2003D, Nature Works® LLC, Minnetonka, USA; density 1.24 g/cm^3) and poly(butylene adipate-terephthalate) – PBAT (type ecoflex® F Blend C1200, BASF, Ludwigshafen, DE; density 1.25–1.27 g/cm^3). Ecoflex® F Blend C1200 is a statistical, aliphatic-aromatic copolyester which is based on 1.4-butanediol, adipic acid and terephthalic acid monomers in the polymer chain [26]. PLA and PBAT were extruded to foils on a flat-film extrusion line (PM30 from Plastik-Maschinenbau Geng-Mayer GmbH, Kelberg, DE) having a screw diameter of 35 mm, a L/D ratio of 18 and a throughput of 15 kg/h. The six barrel temperature zones of the extruder were heated in the following order: Zone 1 to 200 °C, zone 2 to 210 °C, zone 3 to 220 °C, zone 4 to 230 °C, zone 5 to 240 °C and zone 6 to 240 °C.

2.2. Paper testing

Specimens for tensile testing were produced from the copy paper with a cutter in the paper sheet's machine direction, so that most fibres were orientated in longitudinal direction in the specimen. Size effects were investigated by testing different specimen sizes (big samples – type 1A according to DIN EN ISO 527-2 [27] and small samples – type CP2 according to DIN EN ISO 20753 [28]). Tensile tests of 20 samples per specimen type were carried out with a Zwick/Roell Z020 universal

Table 1
Composition and layer arrangement of paper composites (light grey lines display paper layers, black lines and black dotted lines display layers of either PLA or PBAT polymer matrices).

Composite	No. of paper layers	No. of polymer layers (thickness in brackets)	Layer arrangement	Paper volume fraction in %
PLA or PBAT	9	24 (140 µm)		20.8
PLA or PBAT	13	21 (140 µm)		30.2
PLA or PBAT	20	21 (100 µm)		48.3
PLA/PBAT/PLA or PBAT/PLA/PBAT	9	6/12/6 (140 µm)		20.8
PLA/PBAT/PLA or PBAT/PLA/PBAT	13	5/11/5 (140 µm)		30.2
PLA/PBAT/PLA or PBAT/PLA/PBAT	20	5/11/5 (100 µm)		48.3

testing machine (500 N load cell, 8135/20 N clip specimen holder with a vulcolan coating; Zwick/Roell GmbH & Co. KG, Ulm, DE). A gauge length of 100 mm was used for specimen type 1A and of 25 mm for specimen type CP2. The test was started with a testing speed of 1 mm/min up to 0.5% elongation and was then accelerated to 5 mm/min. The calculation of the Youngś modulus was done between 0.25 and 0.5% strain which was determined from the traverse path via linear regression. All test specimens were pre-dried in a Vötsch VCL 4003 forced air oven (Vötsch Industrietechnik GmbH, Reiskirchen-Lindenstruth, DE) for at least 16 h at 80 °C and stored in a desiccator prior testing.

2.3. Composite production

Composites were produced by a film-stacking method. Copy paper and polymer foils were cut in dimensions of $150 \times 80\,mm^2$ and were layered as shown in Table 1. In this way PLA and PBAT composites with different fibre volume fractions of 21, 30 and 48% were obtained. Additional, mixed layered composites were produced from copy paper, PLA and PBAT with stiff PLA composite layers enclosing a ductile core of PBAT and vice versa. After layer arrangement, the semi-finished products were dried in a forced-air oven at 80 °C for at least 16 h.

The first compression moulding process (CM1) took place in a miniature press ($150 \times 80\,mm^2$) with shearing edges usable for a Zwick/Roell Z020 universal testing machine with a 20 kN load cell. No spacers were used for composite production. The press plates were heated with heating cartridges. The dried semi-finished material which had been wrapped in Teflon foil (foil type 0903 AS; Böhme Kunststofftechnik GmbH + Co KG, Schwarzenbek, DE) was heated for a residence time of 30 s with a tool spacing of 1 cm and a temperature of 190 °C. Subsequently, a pressure of 10 bar was applied for 5 min and finally the pressure was boosted to 15 bar for 10 s. The composites were then removed from the press and cooled between two aluminium plates with a mass of 965 g ($30 \times 12 \times 1\,cm^3$) at room temperature.

In order to investigate the influence of the compression moulding method on the composite properties, a second compression moulding technique (CM2) was used. Composites with a fibre volume fraction of 48.3% and dimensions of $350 \times 250\,mm^2$ were produced in a Wickert WLP 80/4/3 hot press (Wickert Maschinenbau GmbH, Landau in der Pfalz, DE). The paper and polymer layers were manually stacked and placed into the pre-heated steel mould. The sample thickness was controlled by means of a spacer frame with 3 mm thickness. The filled mould was then placed into the hot press and the polymer layers were molten for 10 min without applied pressure. Afterwards, a pressure of 30 bar was applied for 10 min, followed by a 10 s final consolidation step at 50 bar. For PBAT composites the last pressure step was omitted due to rupture of the paper layers at the high pressure. The mould was then removed from the hot press and placed into a cooling press (type Höfer H10, Höfer Presstechnik GmbH, Taiskirchen im Innkreis, AT) with a water cooling cycle (water temperature 4 °C) and an applied pressure of 50 bar during the cooling step. After 15 min the mould was cooled down and the composite plate was removed.

In both compression moulding processes (CM1 and CM2) a fast cooling of the composites was carried out without annealing at a specific temperature. It is therefore assumed that the differences in crystallinity are negligible.

Test specimens produced by the different techniques (CM1: compression moulding technique with shearing edges and CM2: compression moulding technique with spacers) were sawn (saw type Proxxon Micromot MBS 240/E, Proxxon GmbH, Föhren, DE) from the composite plates for the corresponding tests parallel to the predominant fibre orientation. The thickness of the composites ranged between 2.4 and 3.6 mm.

2.4. Composite testing

Prior testing, all test specimens were conditioned according to the standard DIN EN ISO 291 [29].

2.4.1. Tensile tests

At least five tensile test specimens (type 1A according to DIN EN ISO 527-2 [27]) were prepared from the composite plate in direction of the main fibre orientation with a jigsaw and a metal frame. Afterwards, the edges were sanded (sandpaper, grain size 400). A displacement speed of 1 mm/min was used for the determination of the Youngś modulus up to an elongation of 0.25%, and was then accelerated to 5 mm/min. A gauge length of 100 mm, and a preload of 0.5 N were adjusted, using the Z020 Zwick/Roell universal testing machine with a 20 kN load cell and a pneumatic clamping system (applied pressure: 1.5 bar). The elongation was measured with a VideoXtens extensometer (Zwick/Roell GmbH & Co. KG, Ulm, DE) between two markers spaced 50 mm apart. The Young's modulus was calculated according to DIN EN ISO 527-2 [27].

2.4.2. Charpy impact tests

The unnotched Charpy impact strength was measured with a Zwick 5102 (Zwick, Ulm, DE) device with a pendulum of 2 J according to DIN EN ISO 179 [30]. At least 5 specimens with dimensions of $80 \times 10\,mm^2$ were prepared for each type of composite.

2.4.3. Interlaminar shear strength

The interlaminar shear strength (ILSS) of at least three specimens was determined with a double-notched tensile test. Specimens were produced from the composite laminates in the dimensions of 60 × 15 mm². Two opposed notches (one on each flat side of the specimen) with a distance of 12 mm were produced with a hand saw. The depth of the notches was chosen as a function of sample thickness and was always deeper than half of the sample depth. The samples were tensile tested at the Z020 universal testing machine with a testing speed of 2 mm/min at a gauge length of 40 mm.

2.4.4. Density and fibre volume fraction

Density and fibre volume fraction were determined for the rectangular Charpy impact test specimens after a conditioned state (to DIN EN ISO 291 [29]). Thickness, length and width of each specimen were measured with an accuracy of 0.01 mm at three different positions with a caliper (Absolute Digimatic, Typ CD-6″CX, Mitutoyo Corporation, Kanagawa, JP). The mass of the samples was weighed with a scale (type 440-35N; Kern & Sohn GmbH, Balingen, DE) with an accuracy of 0.01 g. The density was then calculated from the calculated volume and the measured mass.

The fibre volume fraction of each specimen was calculated from the mass and the grammage (80 g/m²) of the paper assuming a mean paper thickness of 0.098 mm. Considering the amount of paper layers, the specimen dimensions and the density of the PLA (1.24 g/cm³) and PBAT matrices (1.26 g/cm³) the fibre volume fraction was calculated. Due to compression moulding the paper thickness could be changed leading to a possible slight variation of the paper volume fraction.

2.5. Statistical evaluation

The results were statistically evaluated with the "R" software (http://www.r-project.org/). The results were analysed by means of a Shapiro test, in order to investigate if the obtained results follow a normal distribution. Significant differences between results of different samples were determined with a Tukey-test (normally distributed data) or a Wilcoxon test (data which do not follow a normal distribution). All tests were carried out with a significance level $\alpha = 0.05$. The word "significantly" is only used when the results have shown significant differences.

3. Results and discussion

3.1. Tensile characteristics of paper

The tensile strength of paper specimens shows significantly higher values for small samples with 74.4 ± 8.1 MPa compared to the big samples with 56.4 ± 9.5 MPa, indicating a strong size effect as already described, e.g., by Sutherland et al. [31], Heckel & Köhler [32] or Weibull [33] for steel, ceramics, glass and carbon fibres as well as their composites, and wood, respectively. At the same time a higher tensile modulus of 6039 ± 585 MPa was measured for the small samples as compared to the big samples (4426 ± 650 MPa), while the elongation at break does not show significant differences with a mean value of 1.5%. In a study of Prambauer et al. [21] a tensile strength of 62 MPa and a tensile modulus of 5468 MPa for the same copy paper was reported. The dimensions of test specimens as well as the results published by Prambauer et al. [21] were in between our big and small samples, confirming the above mentioned size effects.

Mark [34] reported a Younǵs modulus of around 12 GPa for virgin paper fibres, which were extracted from a paper sheet. This value is significantly higher compared to our results, where whole paper sheets were tested. When measuring the plain paper sheets, the adhesion between the individual fibres and fibre bundles is measured and not the strength of the individual wood fibre. Therefore, the actual strength and modulus of the paper fibres are significantly higher than the properties determined for paper sheets. By embedding the paper sheets in a matrix, the properties of the individual fibres are more important than the properties of the paper. This aspect must be taken into account when evaluating the results of composites.

3.2. Fibre volume fraction of composites

The mentioned fibre volume fractions were not achieved in the case of 20% and 30% CM1 composites. The actual fibre volume fractions are clearly higher with values between 26% and 30% for a composite with an aimed volume fraction of 20%. Also for the 30% volume fraction composites, a higher fibre content of 37% and 38% was obtained. This is attributed to the processing technique. In the CM1 process, the composites are not pressed to a minimum thickness but with maximum pressure, so that the polymer is exhausted, resulting in a higher fibre volume fraction. For the composites with a targeted fibre volume fraction of 50%, the deviation between the theoretical and the practical value is smaller. The actual fibre volume fraction resulted in values between 47 and 54%. Due to the higher paper content less matrix flew out. It is noted that the density of the 50% samples produced with CM1 tends to be higher than that of the CM2 samples (e.g. 1.30 g/cm³ versus 1.25 g/cm³ for PBAT composites and 1.33 g/cm³ versus 1.18 g/cm³ for PLA composites) indicating a lower compaction of the CM2 samples due to the use of a spacer frame which often results in lower density and higher void content in compression moulded composites when the thickness is not exactly calculated (compare e.g. [18]).

3.3. Tensile characteristics of composites

A significant increase of modulus values with increasing fibre volume fraction could be demonstrated for all CM1 composites, as described by Prambauer et al. [21] for copy paper/MAPP composites. The comparison of different composites with a paper volume fraction of 50% produced with CM1 and CM2 shows that the values of the CM2 samples in the pure PBAT and PLA composites are on the level of the 20% CM1 samples (see Fig. 1). The layered 50% CM2 samples show somewhat higher values than the CM1 composites reinforced with 20% paper. However, statistically, the data of the 50% CM2 composites are not significantly different from those of the 20% CM1 samples. It can be concluded that CM2 resulted in composites with a low compaction and in an insufficient stress transfer from the matrix to the fibre.

Fig. 1 shows considerably higher moduli for PLA composites than for PBAT samples, due to the higher matrix properties of PLA. The tensile modulus is specified with 3500 MPa for PLA [35] and 44 MPa for PBAT [36]. It is also visible that the layer arrangement does not affect the Younǵs modulus; the values for the mixed layered matrix composites range between those determined for PLA and PBAT composites.

Similarly, to the Younǵs modulus the tensile strength increases significantly for all CM1 composites with increasing fibre volume fraction.

When the tensile strengths of the 50% fibre volume fraction composites are compared, one can see that the moulding technique CM1 leads to significant higher values than CM2. In the case of PBAT and PLA composites the tensile strength of composites produced by CM2 is on the level of the CM1 composite with a fibre volume fraction of 20% (see Fig. 2). The layered CM2 composites show strength values in between the 20 and 30% composites produced by CM1. The lower differences compared to the composites produced with either PLA or PBAT may be explained with the degree of compaction. In contrast to PBAT or PLA composites the differences in density between CM1 and CM2 composites are lower for the layered composites (1.29 g/cm³ versus 1.28 g/cm³ for PBAT/PLA/PBAT and 1.33 g/cm³ versus 1.30 g/cm³ for PLA/PBAT/PLA). This statement could be confirmed by examining composite fracture surfaces from tensile tests. As visible in Fig. 3, the samples prepared from PLA or PBAT with the CM2 technique show delaminations and voids between paper layers and matrices. In the PLA

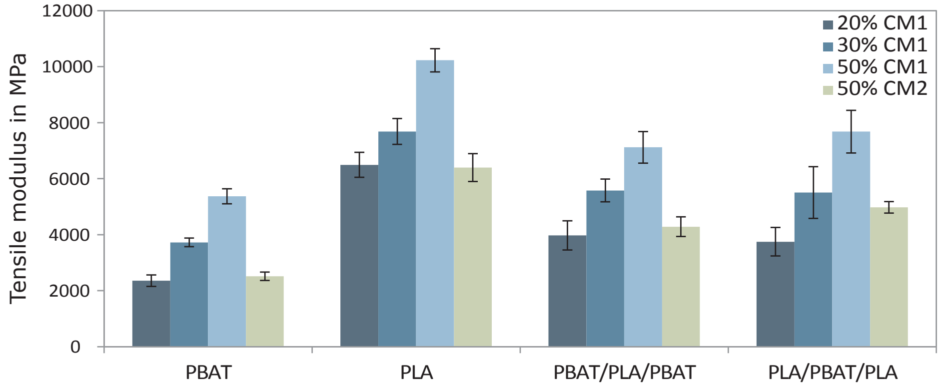

Fig. 1. Mean tensile modulus with confidence intervals as error bars of all tested paper composites produced from different matrices and with different layer arrangement (tensile modulus of pure PLA = 3500 MPa [35], tensile modulus of pure PBAT = 44 MPa [36]).

sample the paper layers appear to be rippled which leads to a reorientation and damage of fibres; in the case of the PBAT sample an extreme deformation and pull-out of PBAT layers could be observed. This indicates a poor adhesion between paper and PBAT. The layered samples PLA/PBAT/PLA and PBAT/PLA/PBAT, on the other hand, show better compaction. The fracture surfaces show no clear differences between samples produced with CM1 and CM2. Nevertheless, strength and density of the layered CM2 samples are lower.

It is hypothesised that a better penetration of the matrix into the paper layers occurred during the CM1 process leading to better tensile properties. Considering the tensile strength of layered composites, no significant differences were found as a function of layer arrangement. Tensile strength values are in between PLA and PBAT composites (see Fig. 2). This confirms the hypothesis that the layer arrangement has no effect on the tensile strength as stated in previous studies for bio-inspired composites (compare [23,25]).

It is striking that some composites significantly exceed the strength of copy paper, which was determined to be 74 MPa for the big paper specimens. This statement is valid for all PLA composites and for layered CM1 composites with a paper volume fraction of 50%. This phenomenon can be explained with the above mentioned fact that by paper tensile testing, rather the adhesion between the paper fibres than the strength of the paper fibre was tested. The single cellulose fibres in the paper sheet are mainly kept together by cohesive forces such as hydrogen bonding or friction, which are much smaller than the tensile strength of the fibre itself. Thus, when the paper sheet is under tensile stress, the fibres are rather pulled apart than ruptured. When paper is embedded in the matrix, the properties of the individual fibres and fibre bundles play a more important role. Burgert et al. [37] showed that the strength of a single wood cell (*Picea abies* [L.] Karst.) ranges between 750 and 850 MPa. The strength of the individual fibres and fibre bundles in the copy paper is higher than the value of the paper sheet, which explains the strong reinforcing effect of copy paper when the single fibre or single fibre bundle is embedded by the polymer.

Table 2 gives an overview about the tensile characteristics of wood, pulp and recycled paper fibre-reinforced thermoplastic matrices. PBAT composites investigated in the present study have clearly higher values than e.g. a wood fibre-reinforced Ecovio matrix [4] and are comparable with bleached and unbleached kraft pulp fibre-reinforced MAPP [8,9], recycled newspaper or paper sack fibre-reinforced PP or MAPP [11,17]. Paper/PLA composites show clearly higher strength and modulus values compared to the listed composites in Table 2. With a tensile strength of 120 MPa and a tensile modulus of 10,230 MPa at a fibre volume fraction of 50%, this composite is comparable with the characteristics of a long regenerated cellulose fibre-reinforced PLA composite with a fibre mass fraction of 40% [19]. Furthermore, the obtained values are even higher than for a glass fibre-reinforced PP laminate with a fibre mass fraction of 40% and 60% [38], and double as high as for a 25 vol.-% short carbon fibre-reinforced PP composite [39]. Comparing paper/PLA composites with injection moulded wood fibre-reinforced PLA from literature, the tensile strength was determined to be 120.4 MPa for paper/PLA (this study) and 54.9 MPa for wood fibre/PLA [41] at a fibre volume fraction of around 50%. Prambauer et al. [21,22,40] produced paper-reinforced composites made of different kinds of paper and matrices in a similar manner. From these results, it is known that copy paper provides the best tensile characteristics when used as reinforcement, followed by newspaper. Comparing copy paper reinforced composites, it is striking that PBAT leads to the lowest strength and stiffness compared to other matrices like MAPP, PA 12, PA 6 or PS investigated by Prambauer et al. [21,22,40]. PLA composites, on the other hand, show the highest values, and even a significantly higher strength than a copy paper/PA 6 composite. In the whole consideration

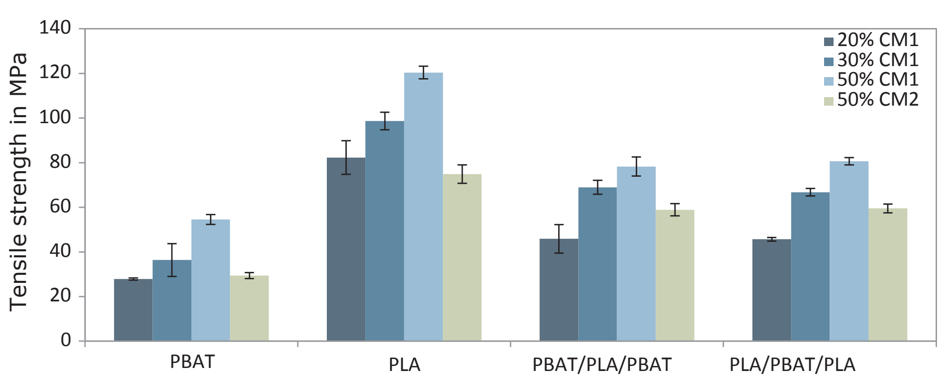

Fig. 2. Mean tensile strength of paper composites produced from different matrices and with different layer arrangement with confidence intervals as error bars of all tested samples (tensile strength of pure PLA = 53 MPa [35], tensile strength of pure PBAT = 14 MPa [36]).

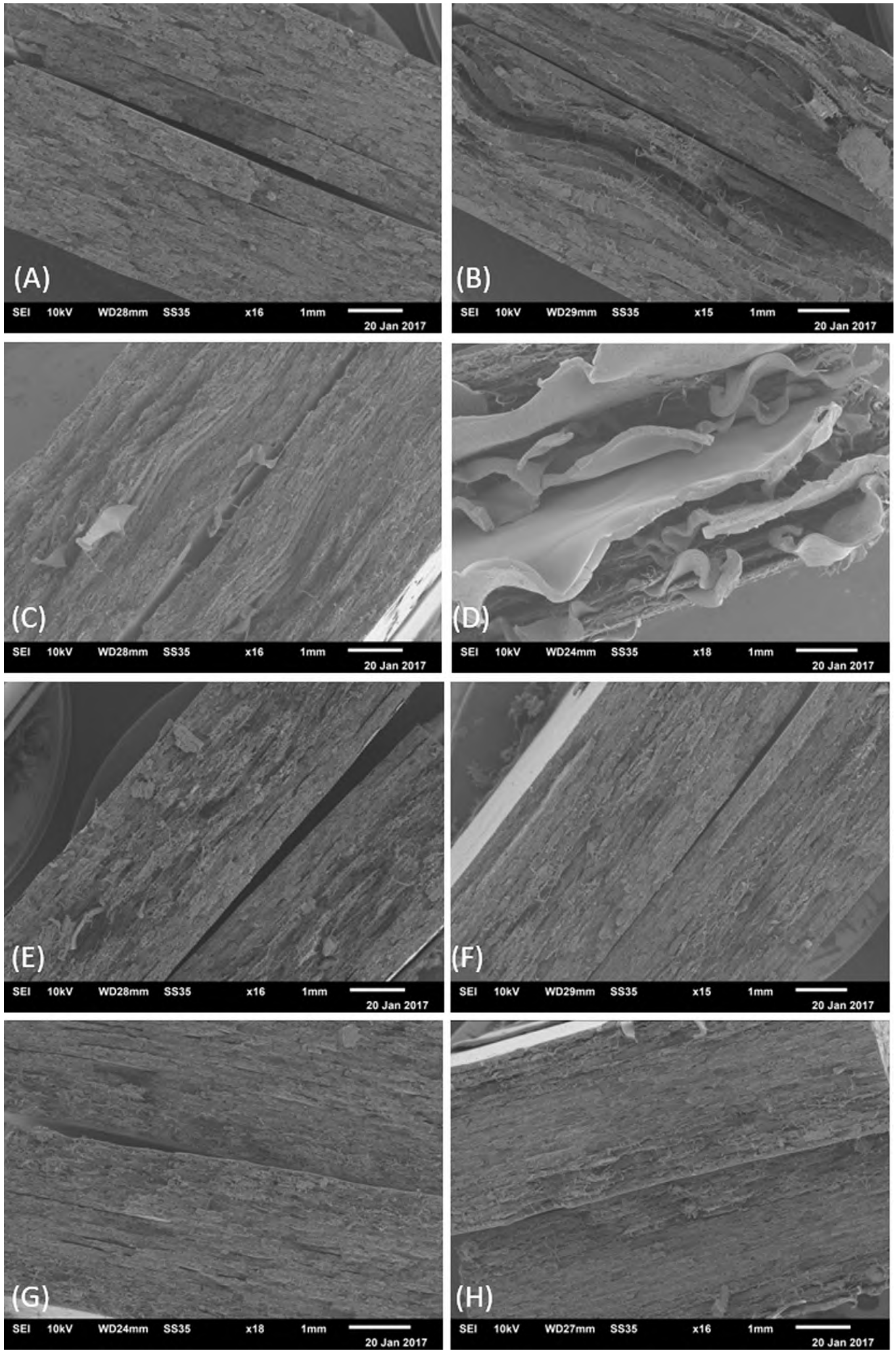

Fig. 3. SEM micrographs of tensile fracture surfaces of paper/PLA (A & B), paper/PBAT (C & D), paper/PLA/PBAT/PLA (E & F) and paper/PBAT/PLA/PBAT (G & H) composites with a fibre volume fraction of 50%. Composites produced with CM1 are shown on the left side, composites produced with CM2 on the right side. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Table 2
Tensile characteristics of wood and recycled paper fibre-reinforced composites. Characteristics of neat matrices are given in brackets. (Ecovio® is a commercial available blend of PLA and PBAT; CM-G: compression moulding from granules; IM: injection moulding; TM: thermomechanically separated; WP: wet prepared; DP: dry prepared, n.a.: not analysed).

Type of reinforcement/matrix	Fibre load	Tensile strength in MPa	Youngs modulus in MPa	Production procedure	Reference
Wood and pulp fibre					
Lignocel® BK 40/90/Ecovio®	20 mass-%	15 (17)	1281 (414)	CM-G	[4]
Lignocel® BK 40/90/Ecovio®	30 mass-%	15 (17)	1154 (414)	CM-G	[4]
Arbocel® C-320/Ecovio®	20 mass-%	16 (17)	1141 (414)	CM-G	[4]
Arbocel® C-320/Ecovio®	30 mass-%	18 (17)	1313 (414)	CM-G	[4]
Arbocel® C-100/Ecovio®	20 mass-%	16 (17)	1173 (414)	CM-G	[4]
Arbocel® C-100/Ecovio®	30 mass-%	17 (17)	1480 (414)	CM-G	[4]
Bleached kraft pulp/PP	30 mass-%	33 (n.a.)	2190 (n.a.)	IM	[9]
Bleached kraft pulp/MAPP	30 mass-%	43 (n.a.)	2330 (n.a.)	IM	[9]
Unbleached TM pulp/PP	30 mass-%	35 (n.a.)	2450 (n.a.)	IM	[9]
Unbleached TM pulp/MAPP	30 mass-%	48 (n.a.)	2410 (n.a.)	IM	[9]
Unbleached kraft pulp/MAPP	30 mass-%	50 (n.a.)	3400 (n.a.)	IM	[9]
Unbleached kraft pulp/MAPP	45 mass-%	55 (n.a.)	5000 (n.a.)	IM	[9]
Recycled paper fibre (source of reinforcement)					
Newspaper (TM)/80% TPS + 10% PBAT	20 mass-%	11 (1)	13 (4)	CM-G	[5]
Newspaper (WP)/PP	20 mass-%	27 (34)	1500 (1000)	CM-G	[15]
Newspaper (WP)/PP	30 mass-%	26 (34)	1500 (1000)	CM-G	[15]
Newspaper (WP)/PP	40 mass-%	24 (34)	1800 (1000)	CM-G	[15]
Newspaper (WP)/PP	50 mass-%	25 (34)	1900 (1000)	CM-G	[15]
Newspaper/recycled PP	55 mass-%	11 (n.a.)	530 (n.a.)	CM-G	[14]
Newspaper/recycled MAPP	55 mass-%	12 (n.a.)	590 (n.a.)	CM-G	[14]
Newspaper/recycled HDPE	55 mass-%	11 (n.a.)	510 (n.a.)	CM-G	[14]
Newspaper/recycled HDPE + MAPP	55 mass-%	13 (n.a.)	650 (n.a.)	CM-G	[14]
Newspaper (DP)/PP	20 mass-%	25 (31)	351 (483)	CM-G	[12]
Newspaper (DP)/PP	30 mass-%	17 (31)	367 (483)	CM-G	[12]
Newspaper (DP)/PP	40 mass-%	12 (31)	385 (483)	CM-G	[12]
Newspaper (WP)/PP	20 mass-%	35 (28)	2800 (1500)	IM	[17]
Newspaper (WP)/MAPP	20 mass-%	39 (28)	2800 (1500)	IM	[17]
Newspaper (WP)/PP	30 mass-%	36 (28)	3700 (1500)	IM	[17]
Newspaper (WP)/MAPP	30 mass-%	44 (28)	3800 (1500)	IM	[17]
Newspaper (WP)/PP	40 mass-%	32 (28)	4300 (1500)	IM	[17]
Newspaper (WP)/MAPP	40 mass-%	45 (28)	4200 (1500)	IM	[17]
Newspaper (WP)/PP	50 mass-%	30 (28)	5300 (1500)	IM	[17]
Newspaper (WP)/MAPP	50 mass-%	50 (28)	5300 (1500)	IM	[17]
Newspaper (DP)/MAPP	20 mass-%	40 (28)	2900 (1500)	IM	[17]
Newspaper (DP)/MAPP	30 mass-%	45 (28)	3700 (1500)	IM	[17]
Newspaper (DP)/MAPP	40 mass-%	49 (28)	4500 (1500)	IM	[17]
Newspaper (DP)/MAPP	50 mass-%	55 (28)	5400 (1500)	IM	[17]
Newspaper (DP)/PP	40 mass-%	34 (n.a.)	4420 (n.a.)	IM	[16]
Newspaper (DP)/MAPP	40 mass-%	52 (n.a.)	4120 (n.a.)	IM	[16]
Paper sacks (WP)/PP	20 mass-%	30 (28)	2000 (1100)	IM	[11]
Paper sacks (WP)/PP	30 mass-%	32 (28)	2510 (1100)	IM	[11]
Paper sacks (WP)/PP	40 mass-%	34 (28)	3080 (1100)	IM	[11]
Paper sacks (WP)/PP	50 mass-%	33 (28)	3680 (1100)	IM	[11]
Paper sacks (WP)/MAPP	20 mass-%	37 (28)	2100 (1100)	IM	[11]
Paper sacks (WP)/MAPP	30 mass-%	41 (28)	2540 (1100)	IM	[11]
Paper sacks (WP)/MAPP	40 mass-%	49 (28)	3150 (1100)	IM	[11]
Paper sacks (WP)/MAPP	50 mass-%	55 (28)	3700 (1100)	IM	[11]

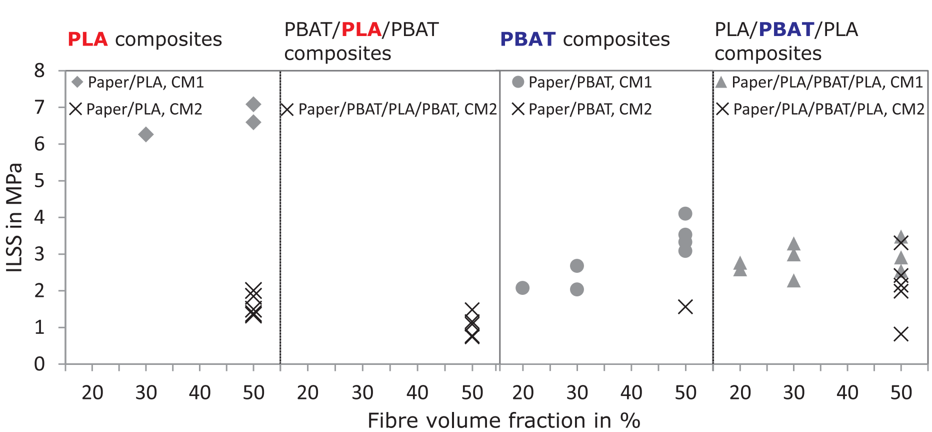

Fig. 4. Interlaminar shear strength (ILSS) of paper composites produced from different matrices and layers as a function of fibre volume fraction. Due to the low amount of valid measurements individual results of successful tests are shown. The bold names of the polymers indicate the shear area. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

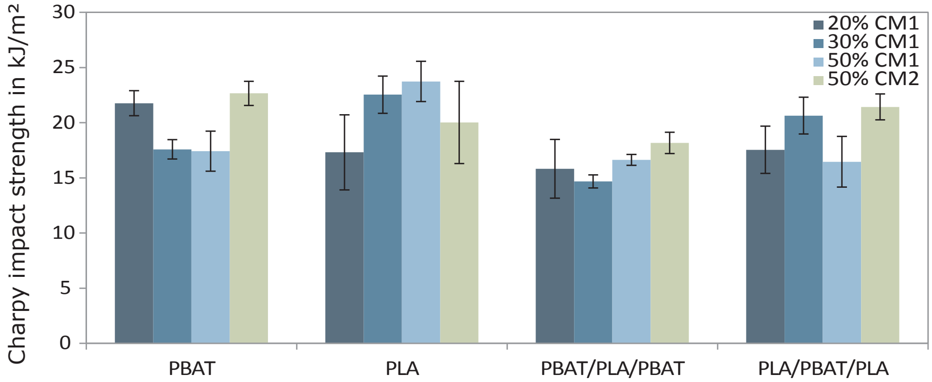

Fig. 5. Mean unnotched Charpy impact strength with confidence intervals as error bars of all tested paper composites produced from different matrices with different layer arrangement. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

it can be concluded that the combination copy paper/PLA has the best potential to be used in mid to high performing components where an appropriate strength with a high stiffness is required.

3.4. *Interfacial adhesion*

The tensile test results have shown a significant lower tensile strength for PBAT composites than for PLA composites (compare Fig. 4). Reason for these significant differences is among other things the lower strength of PBAT (PLA: 53 MPa [35], PBAT: 14 MPa [36]). Additionally, the low strength values of PBAT composites may be explained with the low adhesion between paper and matrix resulting in a suboptimal stress transfer from the matrix to the fibres under tensile load (compare Fig. 3C & D). In contrast to this, Sykacek et al. [41] have found a good fibre/matrix adhesion between wood fibres and a PBAT matrix (Ecoflex® F BX 7011) processed via injection moulding. In the mentioned study the strength increased from 8.6 MPa of neat PBAT to 15.1 MPa, 18.1 MPa and 22.3 MPa by using a wood fibre load of 20, 30 and 50 mass-%, respectively. Due to a higher strength of the neat matrix used in our study, and an assumed higher fibre orientation in testing direction, the strength increased up to 54.5 MPa at a paper volume fraction of 50%.

The better apparent adhesion between wood fibres and PBAT as reported by Sykacek et al. [41] may be based on the process technique. Due to shear and mixing processes in the injection moulding machine a better distribution and wetting of the fibres may take place as compared to the compression moulding technique. The mechanical interlocking between fibre and matrix may be better and could result in higher friction between fibre and matrix, leading to an improved apparent adhesion.

From a chemical point of view PLA and PBAT contain polar and non-polar groups while PBAT contains additional polarisable groups. The main components of paper fibres are cellulose, hemicellulose, inorganic fillers and processing adds such as paper sizings. Most of the wood's lignin content is removed by pulping and bleaching processes during paper production. While cellulose and hemicellulose have a polar character, paper sizing agents are rather nonpolar and have a more hydrophobic character, in order to improve the printability. The nonpolar sizing can interact with the matrices via van der-Waals forces by their non-polar groups. Both matrices, PLA and PBAT, might also interact with cellulose through their carbonyl groups (polar interaction) [42]. Paper sizings are frequently made of starch derivatives or modified natural resins, which means that they also exhibit some polar oxygen groups where the matrix polymers can interact with. It was not investigated if the sizings stay on the paper surface or migrate into the polymer melt during compression moulding. However, it is assumed that the characteristics of cellulose dominate the nature of interactions throughout the paper sheet. More polar interactions are possible between paper fibres and PLA, meaning that the bond between PLA and paper fibres might be stronger due to a more hydrophilic character of PLA compared to PBAT. A study by Svidler et al. [43] analysed the surface tension of PLA Ingeo 4043D and Bioflex F1110 (a blend of PBAT (Ecoflex) and less than 10 mass-% PLA). The results of the study stated a surface tension of 37.1 mN/m for PLA and 22.5 nN/m for Bioflex F1110 as well as a polar surface tension of 2.8 mN/m for PLA and 0.3 mN/m for Bioflex. The greater the surface tension the better the wettability [43]. That means that PBAT has a lower polarity leading to weaker fibre/matrix adhesion than PLA. These statements could be confirmed by own contact angle measurements. Small deionised water droplets were placed on the PLA and PBAT sheets and 10 contact angle measurements were carried out on each matrix. A small contact angle is an indication for good wettability. The results showed a contact angle of $59 \pm 7°$ for PLA and $82 \pm 18°$ for PBAT.

The adhesion between paper and matrix in our study was quantified with double notched tensile tests. Some specimens failed in the region of the notches, which prevented a shear failure of the samples. These results were excluded from the evaluation, and only the valid individual results of measurements are plotted in Fig. 4. Due to the arrangement of the notches, the shear plane is located in the middle of the specimen (inner layer). In the case of a PBAT/PLA/PBAT or PLA/PBAT/PLA composite the shear plane is located in the PLA or PBAT layer, respectively. Most valid readings were obtained for PBAT and PLA/PBAT/PLA composites (shear plane in the PBAT layer). It can be seen that the determined ILSS of the two different composites is in the same range. The values obtained for the samples with a volume fraction of 50% produced with the CM2 technique are at a similar level independent of the layer where the shearing took place. The difference between CM1 and CM2 composites with a fibre volume fraction of 50% is much more pronounced with PLA in the shear plane than with PBAT. It can be concluded that the fibre/matrix adhesion in PLA is more influenced by the moulding technique compared to PBAT. The suspected weak fibre/matrix adhesion between PBAT and paper can thus be confirmed and should be optimised to improve the tensile properties of the composite.

3.5. *Charpy impact strength*

In comparison to tensile strength and Younǵs modulus data, the unnotched Charpy impact strength shows different trends (Fig. 5): the impact strength of PBAT decreases while it was improved for PLA composites with increasing paper content. Similar trends were reported by Le Digabel et al. [44] for hydrolised wheat straw/PBAT composites with a fibre content between 1 and 30% or Bax & Müssig [45] for flax fibre-reinforced PLA with fibre mass-fractions from 10 to 30%.

The samples with 50% paper-reinforcement prepared with CM2 show significantly higher values for PBAT, and lower values for PLA compared to the CM1 samples resulting in comparable values to the

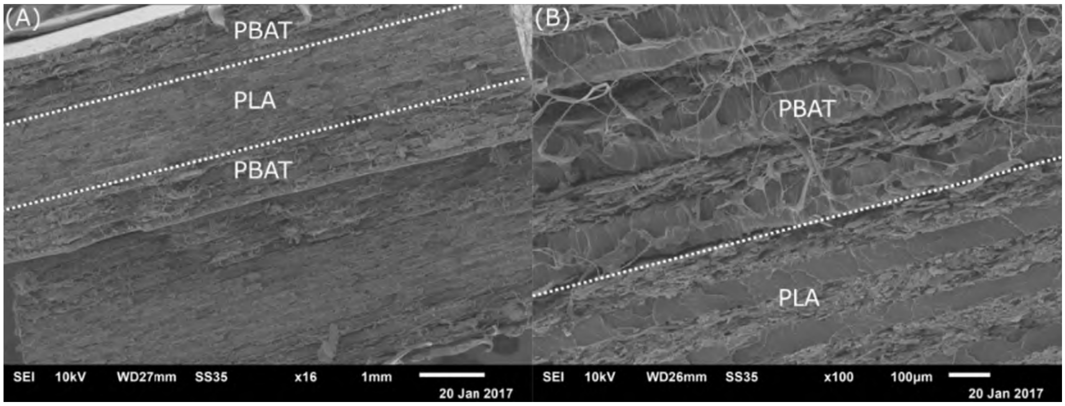

Fig. 6. SEM micrographs of a fracture surface of a 50% PBAT/PLA/PBAT composite produced with CM1. (A) Overview, (B) Detailed micrograph.

20% CM1 composites. The different material behaviour is based on the mechanical properties of the matrix and the reinforcing component. The elongation at break of PLA and PBAT is specified to be 6.0% [35] and 560% [26], respectively. The elongation of the paper sheet was determined to be 1.5 ± 0.2%. Since paper fibres are arranged in sheets, the mechanical properties of a paper sheet are not only determined by the properties of the cellulose fibres, but also by the hydrogen bonds between the cellulose fibres which are influenced by the composition of the paper (fillers and additives), by structural characteristics such as fibre length, agglomeration and the microfibril angle. Burgert et al. [46] tested individual wood cells and were able to show that the elongation is decisively dependent on the development stage and the origin of the wood fibres. Depending on the origin – early wood, latewood, juvenile wood, opposite wood or compression wood – the fibril angle of the fibre cells varies. In general, the higher the fibril angle, the higher the elongation. In addition, processing (mechanical or chemical) affects the stress-strain behaviour of wood fibres significantly. Therefore, the elongation of the fibres can vary between 1% and > 30%. It is assumed that different types of fibres of variable stress-strain characteristics are combined in copy paper. Due to the significantly lower elongation of the fibres compared to PBAT, the impact strength decreases with increasing paper content since the elongation properties as well as the fibre/matrix adhesion are directly related with the toughness of a composite. The lower compaction of the CM2 sample leads to remarkable delaminations, and thus to an increased energy absorption compared to the CM1 reference sample (compare Fig. 3C & D). The PBAT layers were considerably pulled out from the multi-layer composite. The fracture behaviour becomes more brittle with increasing paper content. However, it is surprising that the ductile PBAT composites tend to have lower values than the brittle PLA composites. A further explanation for this phenomenon could be based on a combination of the insufficient fibre/matrix adhesion and the very high elongation of the PBAT layers. Observations have shown that PBAT layers can be elongated and pulled-out from the multilayer composite. After separation and fracture of the paper layers the energy absorption is solely done by the polymer. Due to the fact that the matrix layers are very thin, the bending stiffness is low and the layers can hardly absorb the impact energy. As a result, the low strength of PBAT leads to less impact resistance.

PLA composites show an opposite trend. It is supposed that the average elongation of the fibres in the paper sheet is higher than that of PLA leading to improved toughness of the composite. The fracture behaviour becomes more ductile with increasing paper content. Usually a lower fibre/matrix adhesion may lead to a better impact performance of a composite unless the compaction is insufficient and short fibres are used as reinforcement. Fig. 3A shows a brittle failure of the PLA layers. In the case of the 50% CM2 sample (Fig. 3B), the short paper fibres absorb less energy during fracture due to the poor fibre/matrix adhesion. The inferior fibre/matrix adhesion of the CM2 PLA sample compared to the CM1 sample could be demonstrated with double-notched tensile tests (see Fig. 4). With values between 17 kJ/m^2 and 24 kJ/m^2 PLA and PBAT composites display comparable impact strength to e.g. bast fibre-reinforced PP, flax and hemp fibre-reinforced epoxy or wood fibre-reinforced acrylate [47,48].

As shown in previous studies [23,25] a layered structure with stiff outer layers and a more ductile core layer may lead to a significant improvement of impact strength. Unfortunately, this concept did not work for paper/PLA/PBAT/PLA composites, which do not show any significant differences to the controversial layered PBAT/PLA/PBAT composite (Fig. 5). As a trend, the mixed layered composites show even lower values than the PLA composites. In the previous study [25] long fibres (> 30 mm) were used as reinforcement. Stiff and less ductile bast fibres (elongation < 5%) were used for the outer layers while more ductile regenerated cellulose fibres (elongation > 12%) were used as core layers in a PLA matrix (elongation approx. 2%). The difference in ductility is thus clearly lower and the gradient between the different layers is finer. With this approach an impact strength of 41 kJ/m^2 was achieved which is an increase of more than 55% compared to a composite produced from a mixture of the same materials. It was stated that the fineness of a gradient from layer to layer can significantly affect the toughness [23]. Another approach for a layered composite was implemented by varying the matrix. Long and ductile regenerated cellulose fibres (elongation > 12%) were used as reinforcement. The matrix used for the outer layers was PP (elongation ~ 200%) and PLA (elongation approx. 2%) was used for the core. This approach also demonstrated a significant increase in impact strength up to a value of 84 kJ/m^2 which means an improvement of approx. 36% compared to a composite consisting of a mixture of the same materials, of 53% compared to a brittle PLA composite, and even of 15% compared to a ductile PP composite [24].

With an elongation of 6% for PLA and of 560% for PBAT, this matrix combination displays an extreme variation in ductility. It is assumed that the elongation of PBAT is too high and the adhesion to the paper fibres not sufficient. When PBAT is located in the impacted outer layer, delaminations occur between paper and PBAT layers. Due to the high elongation and the low strength of PBAT, energy is absorbed insufficiently on the tensile loaded specimen side, and is passed on to the brittle PLA layer. When the PLA layer is located on the impact loaded side, some energy is absorbed by the fracture. However, there is the same problem as described for the reverse layered composite: due to the high elongation, less energy absorption takes place by PBAT, which also results in a low toughness for this material. An additional aspect could be the low fibre length. In the case of an insufficient fibre/matrix adhesion, fibres do not absorb enough energy even in the case of fibre pull-out.

Fig. 6 shows the different fracture behaviour of the PBAT and PLA layers. Clearly more fibre pull-outs are visible in the PBAT layers. Moreover, gaps between PBAT and paper layers are recognisable, which indicates a poor adhesion between paper and matrix. However, the PLA layer fails brittle. To investigate the effect of the layer arrangement, layered composites were produced in which PBAT and PLA were alternately stacked with paper (PLA/PBAT/PLA/PBAT…). The samples were tested for impact strength. However, no positive effect on the toughness could be achieved and the results have shown no significant differences to the values obtained with the layered PLA/PBAT/PLA and PBAT/PLA/PBAT composites. In order to find out whether the short fibre length or the extremely different elongation properties of the matrices have a higher influence, further test series should be carried out. Long fibres with the same combination of matrices should be investigated, and matrices should be used which have a lower difference in their ductile properties. For this purpose, the influence of short and long fibres should be verified.

4. Conclusions

The results show that copy paper is very well suited to reinforce thermoplastic polymers. The paper-making process allows the preferential fibre orientation to be utilised in composite production. The five hypotheses stated in the introduction may be answered as follows:

(1) With regard to the tensile properties, a significant increase in strength and modulus could be achieved for all samples examined as a function of the fibre volume fraction. The layering of different polymers had no influence on the tensile properties.
(2) The impact strength of PBAT was reduced with increasing fibre volume fraction because of the lower ductility of paper or fibres as compared to the matrix. In contrast to this the impact strength of PLA was improved at higher fibre volume fractions due to the assumed higher ductility of the fibres compared to neat PLA.
(3) It could be confirmed that PLA composites have higher tensile strength and Youngś modulus values due to the higher strength and stiffness of neat PLA compared to PBAT as well as the better fibre/matrix adhesion between paper fibres and PLA.
(4) The arrangement of different polymer layers did not lead to the desired increase in impact strength. This is attributed to the very high elongation and low strength of the PBAT matrix compared to PLA, the poor adhesion between PBAT and paper, and the low fibre length of the paper fibres. In future research, an improvement in adhesion between PBAT and paper should be achieved. In addition, the trials of the layered composites should be repeated with long fibres to investigate the influence of fibre length, and/or additionally, two matrices should be used showing lower differences in their ductility in which one matrix should have a higher strength and stiffness, and the other one a higher ductility.
(5) The use of different moulding methods demonstrated the importance for precisely matched production parameters. The tensile properties were considerably reduced by a low compaction. The impact strength increased for PBAT composites with a lower compaction while a low compaction did not improve the impact strength of PLA composites.

Our results show new application areas for the paper making industry. However, even if there are still some optimisation possibilities, it can be concluded that copy paper, particularly in combination with a PLA matrix, is very suitable for increasing the tensile properties, and thus the use in laminar large components with a moderate degree of deformation. A big advantage of using paper for composites is the possibility of a continuous production technique based on paper roll goods.

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