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**Mechanical performance of biocomposites based on PLA and PHBV reinforced
with natural fibres – a comparative study to PP**

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Abstract: In the given research paper, the effects of reinforcing polylactid (PLA) and poly(3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV) biopolymers on the mechanical performance were studied. Both PLA and PHBV were compounded with man-made cellulose, jute and abaca fibres. The test bar specimens were processed via injection moulding. Various testing methods, including tensile and impact tests, were used to investigate the composites' mechanical performance. Scanning electron microscopy was carried out to study the fibre-matrix interphasial adhesion. To determine the fibre size distribution, optical microscopy was used. Finally, the obtained results were compared to composites on PP basis with the same reinforcing fibres.

The reinforcing with fibres increased the tensile stiffness and strength significantly; however, depending on the fibre type, different improvements of the mechanical parameters were achieved. The main enhancement was realised in impact and tensile strength by reinforcing biopolymers with man-made cellulose fibres. SEM photographs show a largely differing fibre/matrix bonding for PLA and PHBV compared to PP composites. The broadest fibre size distribution can be found in abaca composites.

Keywords: A. short-fibre composites, B. mechanical properties, B. impact behaviour, biocomposites

1. Introduction

The first reports concerning the syntheses of biopolymers date back to the early 1930s, i.e. the french micro-biologist Lemoigne described polyhydroxybutyrate (PHB) in 1925 [1,2]. The synthesis of high molecular weight polylactic acid (PLA) was carried out in 1932 by Carothers et al. [3,4].

New biobased plastics have been available on the European market for a few decades and were mostly used for medical purposes [5], later for packaging applications, e.g. biodegradable bags and loose fill and packaging of 'ecologically produced' food [6]. Nowadays, applications in structural parts are common as well.

Since about 2002, the market for biodegradables in the packaging sector has developed dynamically. The annual production capacity of biopolymers in 2008 was at an estimate of 300,000 t worldwide and has had a continuous and very rapid growth rate in the last years, even though most plants first began production after 2000 [7,8].

In long-established technical areas, the major engineering resin is still the PP based composite, as its cost/performance ratio is one of the best of all. Considering the new 'biocomposites', with both biogenous matrix and reinforcing natural fibre, great attention should be paid to their competitiveness with the existing and well-optimised material systems. To satisfy the market expectations and achieve technical specifications, some fundamental basics must be challenged, for example competitive prices and similar or improved mechanical properties compared to 'PP commodities'.

The prefix 'bio' can only be attractive if material costs are moderate and customer acceptance can be guaranteed. Therefore, material data of biocomposites, which is in competition with PP equivalents, should be referenced to adequate values of polypropylene reinforced with comparable fibres and processed with assimilable processing techniques. For this purpose, the choice of tailor-made reinforcing fibres is of central importance. Although natural fibres are a potential reinforcement for plastics,

their properties can differ greatly depending on type, grade, harvest quality, processing method, yield etc. Only natural fibres of high technical quality guarantee sufficient reproducibility of mechanical characteristics [9]. For example, cellulose based fibres ('man-made cellulose' [10], 'Viscose' or 'Rayon' [11]) are being considered as an option for polymer reinforcement; firstly due to the sustainability of the fibre source (native cellulose), and secondly because of the high reproducibility of the fibre characteristics (e.g. diameter, stiffness, tenacity etc.) [12-14]. As stated above, the selective choice of reinforcing fibres is of the highest importance, as the fibre parameters are arbitrate for the overall composite performance. For that reason, natural fibres used in this study are either man-made cellulose or technical natural fibres.

With regard to poly(hydroxybutyrate-co-valerate) (PHBV) and especially PLA biopolymers, one of the main drawbacks concerning technical applications is their low impact strength. Furthermore, they show some difficulties during processing, e.g. a narrow processing window, and low fibre/matrix interphasial adhesion. For that reason, further focal points of the given study were an improvement in impact strength and processing at conventional processing machines, enabling a comparison to real processing conditions. Moreover, the moulding technologies that have been used are typical for short-fibre composites: compounding on single- and twin-screw extruder and, subsequently, injection moulding.

Finally, the purpose of this study is to show the capability and mechanical performance of biopolymers reinforced with natural fibres compared to common PP with the same reinforcing fibres. Furthermore, the paper at hand characterises biocomposites with improved mechanical parameters which can be used for a wide range of multifunctional technical applications.

1.2. Review of prior studies

Research and development of biopolymers have seen a tremendous increase in activities in the last five years. In many publications, fundamental information concerning the processing and properties of different biopolymers is available [15-21]. Ganster and Fink [22] described the application possibilities of man-made cellulose in PP, PE, PLA and PS and proved that it is possible to enhance the notch impact strength considerably. The influence of nanofillers on the mechanical performance of PLA was researched by Pluta et al. [23] and Mathew [24]. In [25], the effect of the addition of chemically treated short abaca fibres on the mechanical properties was studied.

Because of the poor availability, but also due to difficulties in processing, only a few papers concerning biocomposites based on PHB have been published. Among the polyhydroxyalkanoates (PHA) group, the poly(hydroxybutyrate-co-valerate) has been biotechnologically developed to improve the known weaknesses of PHB, for example its brittleness and poor processability [26].

Singh and Mohanty investigated PHBV with increasing wood flour content, which was processed on a mini-extruder and a mini-injection moulding machine [27]. They discussed a possible interaction between moderate polar ($>C=O$) and polar groups ($-OH$ and $-COOH$) of PHBV and a hydroxyl group of cellulose regarding the opportunity for an improvement of the fibre/matrix interphase. The results show that by increasing the wood flour content, a clear increase in the tensile E-modulus can be witnessed; however, the tensile strength decreases significantly with increasing filler content [27].

Wong et al. studied PHB composites with flax fibres modified with different plasticisers [28]. The authors ascertained that poly(ethylene glycol) and tributyl citrate are the most effective plasticisers for native PHB; however, for PHB/flax, glycerol triacetate seems to have the major influence on shifting the glass transition to lower temperatures.

Interestingly, the storage modulus of unmodified PHB/flax does not change compared to the unreinforced matrix.

A great amount of investigations done with PHBV biocomposites only deal with processing techniques marginally, mostly focusing on compression moulding of thin layers or films [29-31]. Some of the results were obtained from composites manufactured on mini-processing machines [32-33]. Both compression moulding of thin composite sheets and processing on mini-moulders are, unfortunately, only limited methods comparable to real processing conditions. In most cases, the results are not transferable in view of the up-scaling.

Mohanty et al. described the possibility of compatibilization for PHB composites with natural fibres [34]. The processing machines that were used for composite manufacturing were of similar size as the machines used in this study. The addition of 30wt-% fibres increases the E-modulus without any or only a slight impact on the tensile strength. In former studies [35,36], the influence of processing parameter and fibrous reinforcement in PHB and PHBV were investigated. The authors used semi-scale processing equipment like a Buss- and Brabender-Kneader and a twin-screw extruder type ZSK 30. The manufactured compounds were injection moulded on a conventional injection moulding machine. As a result of compounding the composites on the Buss-Kneader, the authors obtained an excellent fibre dispersion compared to the twin-screw extruder. The processing of cellulose with PHB results in dramatic fibre size reduction and a very narrow fibre-size distribution [36]. The fibre lengths were 200, 104 and 60 μm for the fibres compounded on the Buss-Kneader, the twin-screw extruder and the Brabender-Kneader respectively [35]. The initial fibre length was 1100 μm . An investigation of the degradation during processing showed a significant degree of depolymerisation which is typical for the melting behaviour of PHB polymers. After the first processing step, a significant reduction in molecular mass was observed. Due to

crotonic acid development during degradation, fibre hydrolysis and, consequently, fibre defibrillation occurred.

2. Experimental section

2.1. Materials used

All polymers used in this study are characterised in Table 1.

Table 1: Characterisation of polymers used

For polymer reinforcement, four different cellulosic fibres were used.

Abaca: from Manila Cordage (delivered by Rieter Automotive); bundle diameter 150 +/- 50µm, density 1.5 g/cm³, elementary fibre diameter 10-30µm, tensile strength over 900 MPa [37]. The fibre quality according to FIDA (Fiber Industry Development Authority) is S3.

Jute: from J. Schilgen GmbH & Co; twisting 3.6/1, bundle diameter 60-110µm

Man-made cellulose: from Cordenka GmbH, Fibre type Cordenka® 700 Super3, dtex=2440, number of mono-filaments 1350, breaking force 128.6N, elementary fibre diameter 12µm

Man-made cellulose is a chemical fibre of natural origin. The source of the fibre is cellulose pulp from different wood species [38].

All composites were produced in a matrix-to-fibre weight ratio of 70/30. To achieve an appropriate melt stability and better ductility, the PHBV was blended with poly(butylene adipate-*co*-butylene terephthalate) (27,6% by weight Ecoflex) and processing aids (2,4% by weight). The PHBV content in the blend was 70% by weight.

In the PLA- and PHBV biocomposites, no coupling agent was used; to the PP

composites, 5% by weight (in relation to the fibre content) of maleic acid anhydride grafted PP (MAH-g-PP) by Clariant (TP Licocene PP MA 6452) was added.

2.2. Composite processing

All polymers besides PP were pre-dried in a convection oven (at least for 16h at 80°C, moisture content 0.02-0.05%) and then processed on a single- (Schwabenthan, Polytest 30P, L/D=25, D=30mm for PHBV) and a twin-screw extruder (Haake, Rheomex PTW 25/32, L/D=32, D=25mm for PLA).

The addition of continuous filament took place in an extruder fitted with a coating die. The pellets manufactured this way were dried (for 16h at 80°C in a convection oven; moisture content < 0.3%) and, in a second step, compounded on the single-screw extruder to guarantee a better fibre distribution and to improve the bulk density. The pellets were then injection moulded into standard test specimen (Kloeckner Ferromatik FM 85, nominal clamping force 850 kN, screw diameter 40 mm, L/D = 21, screw speed 120 rpm). Additionally, the machine's hopper was heated to 80°C and rinsed with nitrogen gas with approx. 4l/min ($N_2 \geq 99.999\text{vol.}\%$, Air Liquide Deutschland, Düsseldorf/Germany). The processing parameters are described on another place [14].

2.3. Testing methods

Tensile test

The manufactured test specimens were tested in a tensile test according to DIN EN ISO 527. The test was carried out on a Zwick/Roell UPM 1446 universal testing machine.

The testing parameters were as follows: 1mm/min for estimating the tensile E-modulus and 5mm/min for estimating tensile strength and elongations. The test results were summarised with the help of the computer-supported software testXpert®. The

following values represent the average of 10 separate measurements. Standard injection moulded tensile bars type 1A were used.

Notch impact strength

To investigate the toughness of the composites, an A-notch Charpy impact strength test according to DIN EN ISO 179/1eA was carried out. Beforehand, the test specimens had been cut out of the parallel length of the norm 1A test bar. The A-notch was realised with the test specimen notching machine NOTCHVIS by CEAST. All composites were tested at ambient temperature (+23°C) and at -30°C.

Scanning electron microscopy

To test the fibre/matrix adhesion and composite morphology, micrographs in scanning electron microscopy (SEM) were taken. The SEM fracture surfaces were prepared in a cryo-break fracture (the “dog-bone” test specimens were stored in liquid nitrogen for at least 10 minutes, then removed and immediately broken). A CamScan MV 2300 scanning electron microscope with a wolfram cathode emission gun with acceleration voltage of 10kV was used.

Fibre size distribution

The fibre geometry obtained from the injection moulded samples was investigated after dissolving the PLA specimens in chloroform. The solutions were filtered, and the fibres retained on the filter were left under a standard atmosphere (50% relative humidity and +23°C) for drying. The fibre size was determined microscopically on Keyence VHX-600K microscope supported with a VHX analysis software. Approximately 500 measurements of the fibre length and 100 of the fibre diameter were taken. The presented results were obtained from PLA composites.

2.4. Results and discussion

2.4.1. Tensile Test

In Figure 1, the stress-strain deformation graphs of the tested biocomposites are represented. The results show that the reinforcing effect of the natural fibres and especially their influence on the elasticity of the composite are very high compared to the unreinforced matrix. In addition, it is obvious that a higher stiffness is achieved with natural fibres (e.g. PLA/jute shows a value of approx. 285% of that of PLA) than with man-made cellulose fibre.

In man-made cellulose composites, the values are accordingly lower (PLA/cellulose shows a value of 175% of PLA). In other tests with PLA/man-made cellulose [13,39], an increase in the E-Modulus of approx. 150% of PLA was observed.

The increased E-modulus indicates a stress transfer ensured by the fibres which possess a much higher stiffness than the matrix. For example, the E-modulus of a single jute fibre is approx. 10-30GPa [37] compared to 3.3GPa for PLA.

Figure 1: Stress-strain curves of the tested composites; elongation up to 5% only

Considering the values of short-glass-fibre polypropylene composites (PP GF), the stiffness of which is of approx. 4-7 GPa [40], the application of tailor-made technical natural fibres in biopolymers has proved to be a very promising alternative to PP GF as a reinforcing component. Furthermore, much higher E-modulus values were achieved in bio-based biocomposites than in PP NF composites.

The variety of natural fibres has a fundamental influence on tensile strength; however, the reinforcement with man-made cellulose clearly shows higher values compared to other natural fibres.

By adding 30wt-% of man-made cellulose, an increase in tensile strength of up to 50% can be achieved for PLA and PHBV composites. Bax and Muessig [39] achieved an improvement in tensile strength of approx. 30% in PLA/man-made cellulose fibre; Ganster and Fink [13] obtained similar values.

Within the natural fibre group, jute seems to achieve the highest improvement (up to 30% in PLA composites).

The obvious reinforcing effect is caused by the fibre performance. Most of the natural fibres consist of single fibre packages, so called 'fibre bundles' where the cellulose is embedded in lignin which functions as a kind of matrix (Fig. 2). Therefore, a natural fibre (fibre bundle) can be considered as a naturally occurring composite. Attributable to the orientation of the cellulose fibrils along the fibre axis, the fibre strength can differ [37,41,42]. Furthermore, the fibre properties differ depending on the fibre geometry. Moreover, due to the varying chemical composition of natural fibres (e.g. cellulose, hemicellulose, lignin, pectin etc.), the orientation of the cellulose and diverse structural defects along the fibre length, a wide distribution of their mechanical properties can be observed (Tab.2).

Figure 2: A comparison of elementary A) abaca fibre bundle and B) man-made cellulose fibre.

Table 2: Mechanical properties of different natural fibres compared to E-glass fibre [43,44].

Interestingly, the composites with jute show the highest tensile strength within the natural fibre group, although the jute fibre is characterised by lower mechanical values as for example abaca (Tab. 2). Also, the abaca elementary fibre diameter (10-30 μ m)

seems to be more favourable than that of jute (15-35 μm) [37]. The explanation for this untypical behaviour of the composite could be the fibre processing method. Most likely, the abaca fibres were manufactured by using a typical fibre treatment for textiles, e.g. wax or mineral oils. Especially, the application of mineral and/or natural oils during fibre manufacturing is a common technique to increase throughput, production efficiency and the overall fibre performance. Obviously, the oil film on the fibre surface influences the fibre/matrix interphase and, due to affected stress transfer, decreases the mechanical properties of the composite consequently. This untypical behaviour of jute composites has been confirmed by another study [45] whose results correspond to the results presented here.

Considering the fibre bundle diameter, which will be discussed in more detail in the next section, abaca is characterised by a higher average value compared to jute. The averages are 156 μm and 47 μm for abaca and jute respectively. The estimated results for abaca fibre are measurably different, as in the previous study [14]. The values of the investigation in the framework of this paper were obtained using an analysis software, unlike in [14], where the fibre diameter and length were quantified manually on the basis of microphotographs.

It is obvious that an increasing fibre diameter affects the aspect ratio and, thus, decreases the mechanical performance of the composites. Jute fibre bundles undergo separation during processing, allowing a better distribution and a favourable diameter, which can be seen in the results of fibre size distribution.

As stated above, the consideration of the elementary fibre diameter and performance can only lead to false results in view of the mechanical properties of the composites.

There are several factors which have to be taken into account when discussing the mechanical behaviour.

Most of the natural fibres have an average diameter in the bundle of considerably more than 50 μ m while man-made cellulose occurs, in most cases, as an elementary fibre with a diameter of only 12 μ m. The higher aspect ratio (fibre length/fibre diameter; L/D) of the fine cellulose fibres is also the cause for a greater strength in the composite. What is more, the mechanical characteristics of the fibre (Tab. 2) predestine it to composites of higher strength and ductility but slightly lower stiffness. For that reason, the stiffness of biocomposites with man-made cellulose fibre is partially deteriorated compared to composites with natural fibres and the strength is significantly higher (Tab. 3).

Also, the PP composites show mostly comparable, sometimes lower characteristic values. In this case, the increase is, besides the fibre geometry and properties, the result of the coupling agent that basically improves all mechanical values by additionally 20-30% [45]. It may also be deduced that by adding an appropriate coupling agent or compatibilizer, the tensile strength of the bio-composites could be increased by a further 20-30%.

All absolute mechanical values are depicted in Table 3.

Table 3: Mechanical parameters of tested composites.

2.4.2. A-Notch Impact Strength

The values that are shown in Figure 3 represent the A-notch impact strength according to Charpy. The related test was performed at ambient (+23°C) as well as at temperature below freezing (-30°C). All average values are the result of ten separate tests.

The massive increase in impact performance for all man-made cellulose composites is noticeable. It equals more than 500% in PHBV composites, 260% in PLA and 220% in PP (at ambient temperature). In literature, an increase in notch impact strength of PLA reinforced with man-made cellulose of approx. 200% is mentioned [26]. Obviously, the

much lower diameter (higher aspect ratio) and smooth surface of man-made cellulose fibre compared to the irregular natural fibre determines the composite performance under impact stress. It affects the fibre/matrix interaction. In this context, pull-outs appear more often with man-made cellulose than with abaca or jute fibre. Supposedly for this reason, the path length of the propagated crack is enlarged, increasing the energy amount needed to break the sample [14]. The thinner the fibre is, the larger the surface, and, in consequence, the higher the total energy necessary to pull-out the elementary fibre. Furthermore, man-made cellulose fibre shows much higher elongation-at-break, as well as lower stiffness (Tab. 2), indicating increased ductility and flexibility. With increasing fibre deformability, the impact energy needed to break the sample increases, and consequently, the impact strength of the composite. A somewhat different effect occurs with jute and abaca. Besides the fibre diameter, other important factors are fibre composition, manufacture at times the fibre preparation as well as damage throughout the compounding and injection moulding process (such as fibrillation or shortening of the fibre bundle) [14].

Figure 3: Charpy A-notch impact strength; above testing temperature +23°C (A), below testing temperature -30°C (B).

The most noticeable improvement of notch impact strength is achieved in PHBV composites with man-made cellulose (up to 570 %); however, it is only partially caused by fibre reinforcement. PHB has a very narrow processing window; the optimal processing temperature is very close to the decomposition temperature and the processing is only marginally controllable [36]. It can be suspected that PHBV is subject to massive degradation during processing caused by thermal decomposition. Gatenholm and Mathiasson also came to this conclusion while investigating PHB and

PHBV composites with lignocellulose fibres [35,36]. They observed that a significant degree of chain scission had already occurred during compounding and injection moulding of unreinforced PHB. Furthermore, the authors conclude that adding cellulose generates much higher shear stress, which creates local overheating. As a result, polymer degradation occurs, resulting in the development of different acids. The second most volatile degradation product of PHB is crotonic acid; however, carboxylic acids are formed as well. Both acids may affect the structure of the cellulose fibres, causing fibre damage, e.g. due to hydrolytical fibrillation [35].

As stated above, the PHBV fraction in the blend breaks down partially into crotonic acid; thus the impact energy is transferred mainly by the very tough, rubbery poly(butylene adipate-*co*-butylene terephthalate) (Ecoflex), leading to much higher impact values. This behaviour can especially be proved by an impact mechanical analysis like the Charpy test. Interestingly, considering the glass transition temperature (T_g) of Ecoflex (-20°C) and the testing temperature at -30°C , the decrease in impact strength is insignificant. Although Ecoflex is in a glassy state, the impact strength remains at a very high level. As PBAT is characterised by a decreased impact strength below its T_g [46], the stress-transfer mechanism must be another. Certainly, a large amount of the fracture energy is dissipated as the initiated crack meets the fibre. The SEM analysis shows (Fig. 4) that only a very poor adhesion between the fibre and matrix occurs. Thus, it can be concluded that the obtained significant values are not a result of the strong interphase.

A weak interphase affects the fibre/matrix interaction, whereby pull-outs appear more often. As a result, the path length of the initiated crack is extremely enlarged, which increases the energy amount needed to break the sample and, consequently, the impact resistance [14].

2.4.3. SEM

The microstructures made visible by scanning electron microscopy are depicted in Figure 4. It is obvious that PHBV composites show a poor interphasial adhesion compared to PLA and PP. PHBV is a highly crystalline copolymer with a noticeably higher shrinkage than low-crystalline PLA. Post-moulding shrinkage values are approx. 1.5% and <0.5% for PHB and PLA respectively [47,48].

As a result, the existing interphase is damaged in the course of time and, due to secondary crystallisation, the coupling between fibre and matrix is decreased. In consequence, the fibres are not well embedded in the matrix and there is no or only poor adhesion. This kind of interaction is more likely to take place in the very smooth man-made cellulose than in rough natural fibres. The effect of shrinkage on adhesion between PHBV and the paper layer during extrusion coating was investigated by Kuusipalo [49]. He concluded that the weak interphase is a result of reduction in adhesion, which is a reason for the secondary crystallisation effects in PHBV. These results correspond to our observations also, as discussed above.

Furthermore, the PHBV composites in the matrix consist of approx. 27% Ecoflex. As Ecoflex is a thermoplastic with high melt viscosity, the appropriate fibre impregnation during processing is hindered in general.

In the case of man-made cellulose, pull-outs occur more often than with abaca fibre.

This is an additional explanation for the improved impact strength of man-made cellulose composites compared to natural fibres. As discussed in the previous section, the appearing pull-outs contribute to an increase in fracture energy during Charpy test.

In addition, the very smooth surface of man-made cellulose in contrast to abaca, or even jute fibre, predisposes this fibre for pull-outs. Especially abaca fibres show an increased surface roughness. In consequence, the fibres are embedded in the matrix predominantly mechanically by anchoring the matrix on the surface irregularities. Unfortunately, due

to the high melting viscosity, this mechanism is not effective enough to ensure a sufficient impregnation along the entire fibre length.

Figure 4: SEM micrographs of tested biocomposites: PHBV (above), PLA (middle) and PP (below).

PLA composites show a slightly better adhesion than PHBV composites. This is due to their one-component matrix system. Moreover, PLA was solidified into a quasi-amorphous state by rapid quenching in the mould. In consequence, the shrinkage was significantly reduced compared to semi-crystalline PHBV. For that reason, the interphase has not been affected as highly as in the case of PHBV composites. However, some pull-outs can be seen also in PLA composites. Particularly PLA with man-made cellulose shows fibre marks on the fracture surface, indicating fibre pull-out. Noteworthy are also the obvious fibre breakages at the surface. Besides fibre pull-out and debonding, fibre fracture is one of the most common mechanisms of energy dissipation in composites [50].

In PP composites, maleic acid anhydride grafted PP was used as coupling agent. Thus, the fibres show a significantly higher adhesion to the matrix than in the case of biopolymers. Consequently, the fibres are well-coated with the polymer and they often break at the fracture surface, although some pull-outs can also be observed.

It is to overstate that the SEM analysis is only qualitative and to determine the strength of the interphase definitely, other quantitative analysis should be made.

2.4.4. Fibre size distribution

The investigation of the fibre parameters was done for PLA composites only; however the authors assume that the tendency is equal or analogous for each of the composites.

Figure 5 depicts the fibre diameter and length distribution after dissolving the injection moulded specimens and recording their geometry under the microscope. It is obvious that the spectrum of the abaca diameter is very broad compared to jute and man-made cellulose. It explains the lower mechanical values obtained in PLA/abaca composites as the calculated fibre aspect ratio (λ_c) is noticeably lower than in jute and man-made cellulose composites, and amounts 6.7, 7.23 and 10.0 respectively.

Figure 5: Fibre size distribution in PLA composites: length (above) and diameter (below).

Considering the measured aspect ratios (λ), the difference between native natural fibres and man-made cellulose is significantly higher, e.g. for man-made cellulose $\lambda = 189$, indicating higher mechanical parameters. Furthermore, there is a high probability that the abaca fibre bundles have undergone defibrillation, which would explain the frequent occurrence of smaller diameters. The initial average value of the abaca bundle diameter was approx. $150\mu\text{m}$, as reported by the supplier. The evidently broader diameter range and fibre defibrillation of abaca fibre can be seen in Figure 6. On the contrary, jute and man-made cellulose fibres show a narrow distribution. In man-made cellulose, the diameter is almost constant for all measurements and amounts to approx. $12\mu\text{m}$, as it is typical for cellulose staple fibres. For this reason, the occurrence of man-made cellulose diameter is almost perfectly narrow. The average diameter of jute fibre bundle is obviously lower and the diameter size distribution is narrower than for abaca (Figs. 4 and 5). The initial fibre diameter of jute bundle was $110\mu\text{m}$.

The fibre length (L) was estimated as for diameter (D). The initial length of the pellets was set at 3mm during compounding. The distribution of abaca fibre length is the broadest of all. Apart from the diameter, the fibre length is also an indicator for the

mechanical performance of the composite. Thus, PLA composites with abaca are characterised by lower mechanical parameters than PLA / jute and PLA / man-made cellulose. Apparently, the fibre length distribution is less dependent on damage throughout processing than on the fibre diameter, as the distribution of the fibre bundle diameter in comparison to the length distribution is much broader. Regarding the structure of the natural fibre bundle (Fig. 2), the probability of longitudinal fibre fracture is much lesser than the damage in diameter (separating of elementary fibres). Unlike man-made cellulose, natural fibres are obviously subject to defibrillation more often (Fig. 6). It is quite obvious that after processing, fibres are damaged both diagonally and longitudinally; but the damage rather occurs in the diameter when the change in length remains in a narrower range.

The fibre length of man-made cellulose is evidently longer compared to both jute and abaca, due to fibre-fibre interaction during polymer flow. Regenerated cellulose fibres intertwine each other during compounding and injection moulding as well (Fig. 6). As a result, longer fibres occur more often.

Figure 6: Microphotographs of extracted fibres; A) man-made cellulose, B) abaca, C) jute.

Table 4 shows average values of measured diameters and lengths as well as calculative quantities of critical fibre lengths and aspect ratios for PLA composites.

Table 4: Measured and calculated average values of fibre length and diameter for PLA composites.

The following estimations of the fibre parameters were done for PLA matrix only. The critical fibre (bundle) length (L_c) and aspect ratio (λ_c) were estimated in accordance with the equations (3.1. – 3.3.)

For the calculation of the critical fibre length L_c the Kelly-Tyson model is commonly in use for short-fibre composites [14, 20, 51-53]:

$$L_c = \frac{\sigma_f^{\max} D}{2\tau} \quad (3.1)$$

with

D – fibre diameter;

σ_f^{\max} – fibre tensile stress at maximum;

τ – interfacial or matrix shear strength;

τ was determined by using the change of shape hypothesis, which can be applied if the tensile state is exactly known [54-57]:

$$\tau = \frac{\sigma_m^{\max}}{\sqrt{3}} = 0.58\sigma_m^{\max} \quad (3.2)$$

σ_m^{\max} – PLA matrix tensile stress at maximum; 63.5MPa

For estimation the fibre aspect ratio λ_c the equation (3.1) can be rewritten as follows:

$$\lambda_c = \frac{L_c}{D} \quad (3.3)$$

The strengths for jute and man-made cellulose were taken from literature [20,37]. As described in [37], the tensile strength of elementary jute fibre is within a range of 400 – 800MPa. The tensile strength of abaca fibre amounts to 980MPa. In the case of natural fibres, the values of elementary fibres seem to be inadequate for calculating the mechanical properties of the composites. More appropriate are the properties of fibre bundles. Kozłowski [37] estimated the tensile strength of fibre bundles for jute and abaca as 36 and 67cN/tex respectively. For Cordenka 700 Super 3, the breaking tenacity

is 540mN/tex. Taking the equation from 3.4 into account, the tensile strengths for all fibres bundles and man-made cellulose are 525, 1005 and 760MPa for jute, abaca and man-made cellulose.

Calculation of tensile strength of fibre bundles and man-made cellulose σ_f^{\max} [58]

$$\frac{\text{cN}}{\text{dtex}} \cdot 100 \cdot \frac{\text{g}}{\text{cm}^3} = \text{MPa} \quad (3.4)$$

However, for abaca the value of 489MPa was used, as the tensile strength for this fibre was estimated separately, as described elsewhere [14].

It can be seen that the critical fibre length for PLA/abaca composites is the largest (1.04mm), than follow PLA/jute (0.34mm) and PLA/man-made cellulose (0.12mm). Taking the aspect ratio into account, the tendency is similar. The lowest value was measured for abaca, than for jute and man-made cellulose fibre. It is obvious that with an increasing aspect ratio, the composite strength increases as well, as the participated fibre surface is larger and the stress is divided by a larger area. As a result, higher amounts of stress can be transferred without fracture. Also, the critical fibre length for PLA/man-made cellulose is only 0.12mm, compared to 1.04mm for abaca, and the lower critical fibre length ensures higher composite strengths as well. The estimated fibre size distribution is a very important contribution to understanding the mechanical behaviour of the composites. As stated above, mechanical parameter of elementary fibres are only limitedly applicable for calculating the mechanical performance of composites. Moreover, specifying only average values can also lead to false conclusions. It is more adequate to define the fibre size distribution. This property is a function of both processing conditions and fibre-matrix relations, e.g. viscosity, fibre-fibre interaction etc.

The obtained results are confirmed in mechanical tests in which PLA/man-made cellulose composites show the highest values compared to PLA/jute. PLA/abaca composites have slightly lower mechanical parameters in general, except for the impact

test in which composites with abaca show noticeably higher values due to more frequent pull-outs compared to jute (Fig. 3). Moreover, abaca is more ductile and characterised by a strain-to-break twice as high as jute [20]. As discussed in the previous sections, the more bendable or flexible the fibre, the higher the impact strength of the composite.

3. Conclusion

The tested biocomposites clearly show better or comparable characteristic values than the 'common' natural fibre reinforced PP. Besides the typical increase in stiffness, a significant increase in strength (> 50%) and a noticeable improvement in notch impact strength (> 250%) can be observed. This striking optimization of materials and processes is an innovative contribution to composite processing; up to now, properties such as strength and notch impact strength have hardly been affected or only slightly improved. Especially man-made cellulose has proved to be a favourable reinforcing fibre due to an advantageous geometry as well as its overall mechanical performance. The SEM analysis shows strongly differing morphologies for each composite. Poor adhesion was observed for PHBV/Ecoflex composites, as the blend was partially degraded during processing and the interphase was destroyed by the shrinking matrix. For PLA, a moderately strong interphase was observed. Due to the use of maleic-acid anhydride grafted polypropylene, an improved interphasial adhesion in PP composites was noted. An analysis of the fibre size distribution of PLA composites after processing showed a broad spectrum of abaca fibre diameter. In addition, the critical fibre length of abaca is much higher and the aspect ratio lower than those of jute and man-made cellulose. Amongst others, this is the reason for deteriorated mechanical parameters of abaca composites. Man-made cellulose is characterised by the highest aspect ratio. Semi-crystalline PHBV is widely comparable to PP composites and can sometimes substitute it in certain applications. But the main hindrance to overcome is still its

processability. PLA composites show much higher mechanical characteristic values than PP; however, it has to be kept in mind that the material loses the ability to transfer stress as soon as the glass transition temperature is exceeded. Therefore, the combination of thermally untreated PLLA and NF is applicable as engineering material only at temperatures lower than +50°C. Further possibilities are to use thermal tempering to increase the crystallinity of PLA or the synthesis of stereo-complexes made of PLDA- and PLLA, which enables the achievement of achieving heat distortion temperatures above 200°C.

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Table 1: Characterisation of polymers used

Polymer	Manufacturer	Composition*	T _g [°C]	T _m [°C]	MVR	M _w
			(DMA)	(DSC)		
Poly-(3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV)	Tianan Biological Material Co., Ltd., (Ningbo, China)	97% 3HB / 3% 3HV	21	162	61,3 (170 °C / 2,16 kg)	520.000*
Poly(lactid 4042D) (PLA)	NatureWorks LLC (USA)	D-lactid content 4-8%	60	152	5,7 (190 °C / 2,16 kg)	166.000*
Polypropylene 575P (PP)	Sabic Europe (Germany)	homopolymer PP	16	165	5,7 (190 °C / 2,16 kg)	509.000**
Poly(butylene adipate-co-butylene terephthalate) (PBAT) (Ecoflex F)	BASF SE (Germany)	based on the monomers butanediol, adipic acid and terephthalic acid	-20	120	4-8 (190 °C / 2,16 kg)	-

* Size exclusion chromatography, molecular weight determined from PS standards

**As reported by manufacturer

Table 2: Mechanical properties of different natural fibres compared to E-glass fibre [43,44].

Fiber	Density [g/cm ³]	Tensile strength [MPa]	Young's modulus [GPa]	Specific modulus [-]	Elongation to break [%]	Moisture absorption [%]	Diameter of elementary fibre [μm]
E-Glass	2.55	2400	73	29	3	-	15-25
Man-made cellulose	1.49	885	27	18	12	12-15	12
Abaca	1.50	980	27 - 32	18 - 21	-	-	10-30
Jute	1.46	400 - 800	10 - 30	7 - 21	1.8	12	15-35

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Table 3: Mechanical parameters of tested composites.

Matrix	Fibre	Tensile E-Modulus [GPa]	Tensile strength [MPa]	Tensile elongation at break [%]
PLA	-	3.4 ± 0.23	63.5 ± 0.4	3.3 ± 0.5
	Man-made Cellulose	5.8 ± 0.15	92.0 ± 4.7	1.9 ± 0.3
	Abaca	8.0 ± 0.34	74.0 ± 0.7	1.4 ± 0.1
	Jute	9.6 ± 0.36	81.9 ± 2.9	1.8 ± 0.0
PHBV/Ecoflex	-	2.1 ± 0.07	27.3 ± 0.3	7.0 ± 1.1
	Man-made Cellulose	4.4 ± 0.34	41.7 ± 3.8	2.3 ± 1.0
	Abaca	4.4 ± 0.06	28.0 ± 1.3	0.9 ± 0.1
	Jute	7.0 ± 0.26	35.2 ± 1.3	0.8 ± 0.0
PP	-	1.5 ± 0.03	29.2 ± 0.4	13.6 ± 1.0
	Man-made Cellulose	3.7 ± 0.11	71.6 ± 2.7	3.5 ± 0.5
	Abaca	4.9 ± 0.11	42.0 ± 0.5	1.7 ± 0.2
	Jute	5.8 ± 0.47	47.9 ± 2.7	1.4 ± 0.1

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Table 4: Measured and calculated average values of fibre length and diameter for PLA composites.

Parameter	Unit	PLA / Man-made Cellulose	PLA / Abaca	PLA / Jute
Calculated values				
L_c	mm	0.12	1.04	0.34
λ_c	-	10.0	6.7	7.23
Measured values				
L	mm	2.27	1.45	0.46
λ	-	189.2	9.3	9.8
Dimension used for calculating				
σ_f^{\max}	MPa	760	489	525
σ_m^{\max}	MPa	63.5	63.5	63.5
D	mm	0.012	0.156	0.047
τ	MPa	36.6	36.6	36.6

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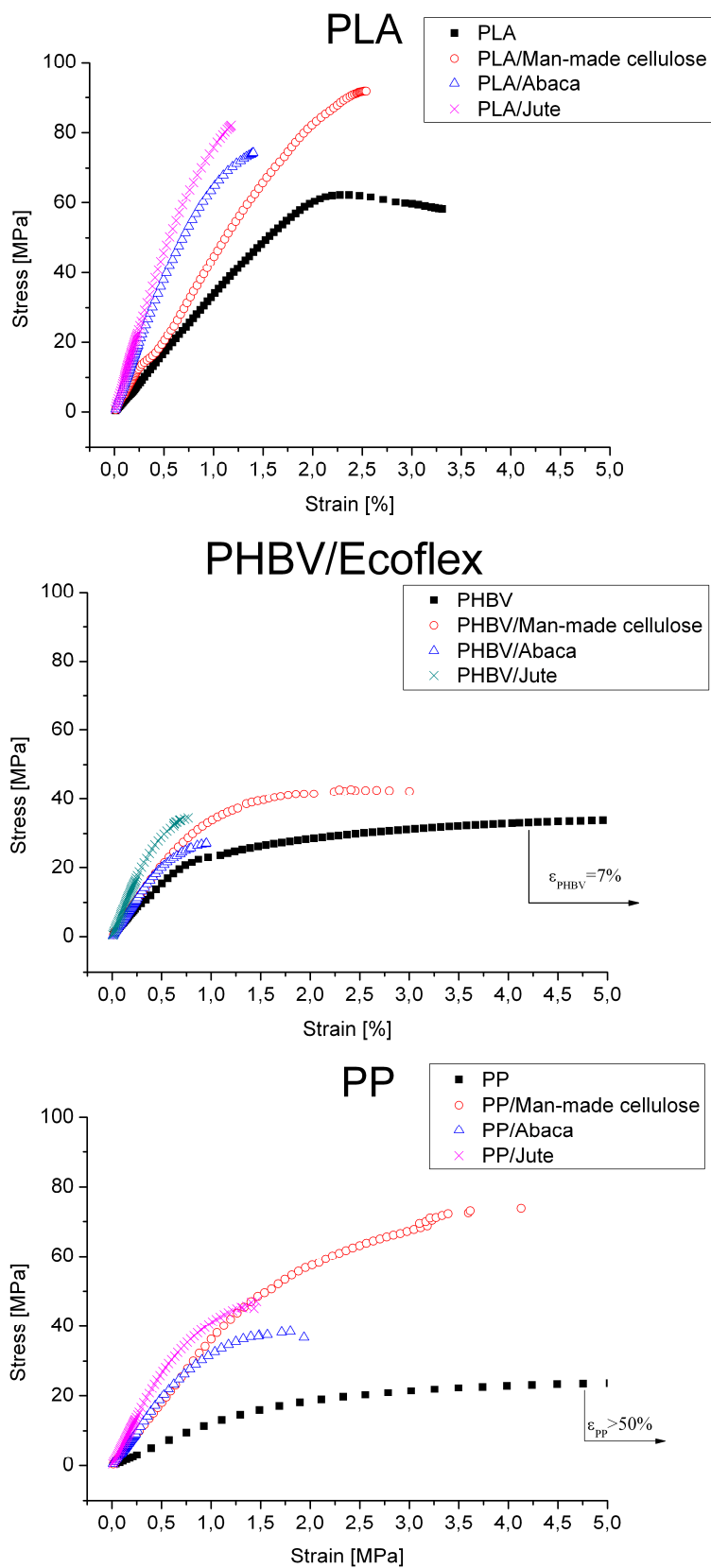


Figure 1: Stress-strain curves of the tested composites; elongation up to 5% only

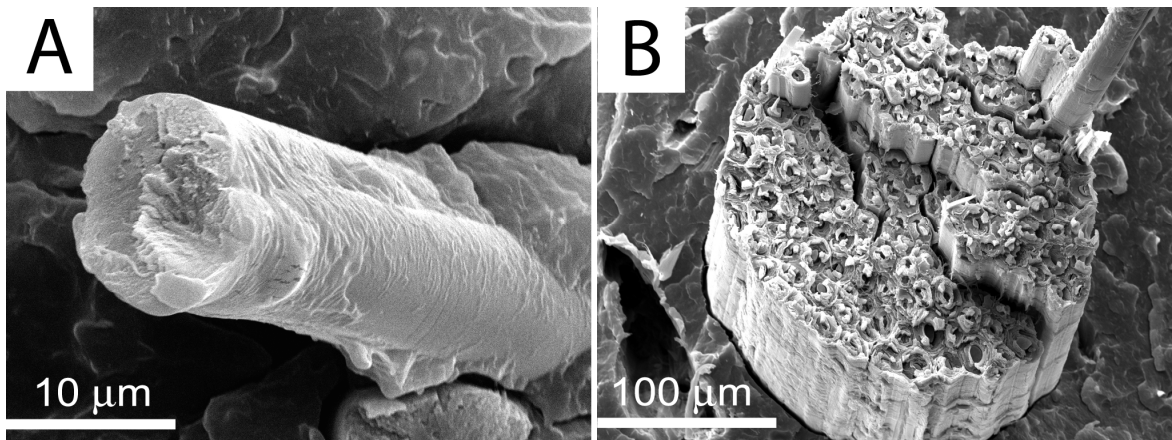


Figure 2: A comparison of elementary A) abaca fibre bundle and B) man-made cellulose.

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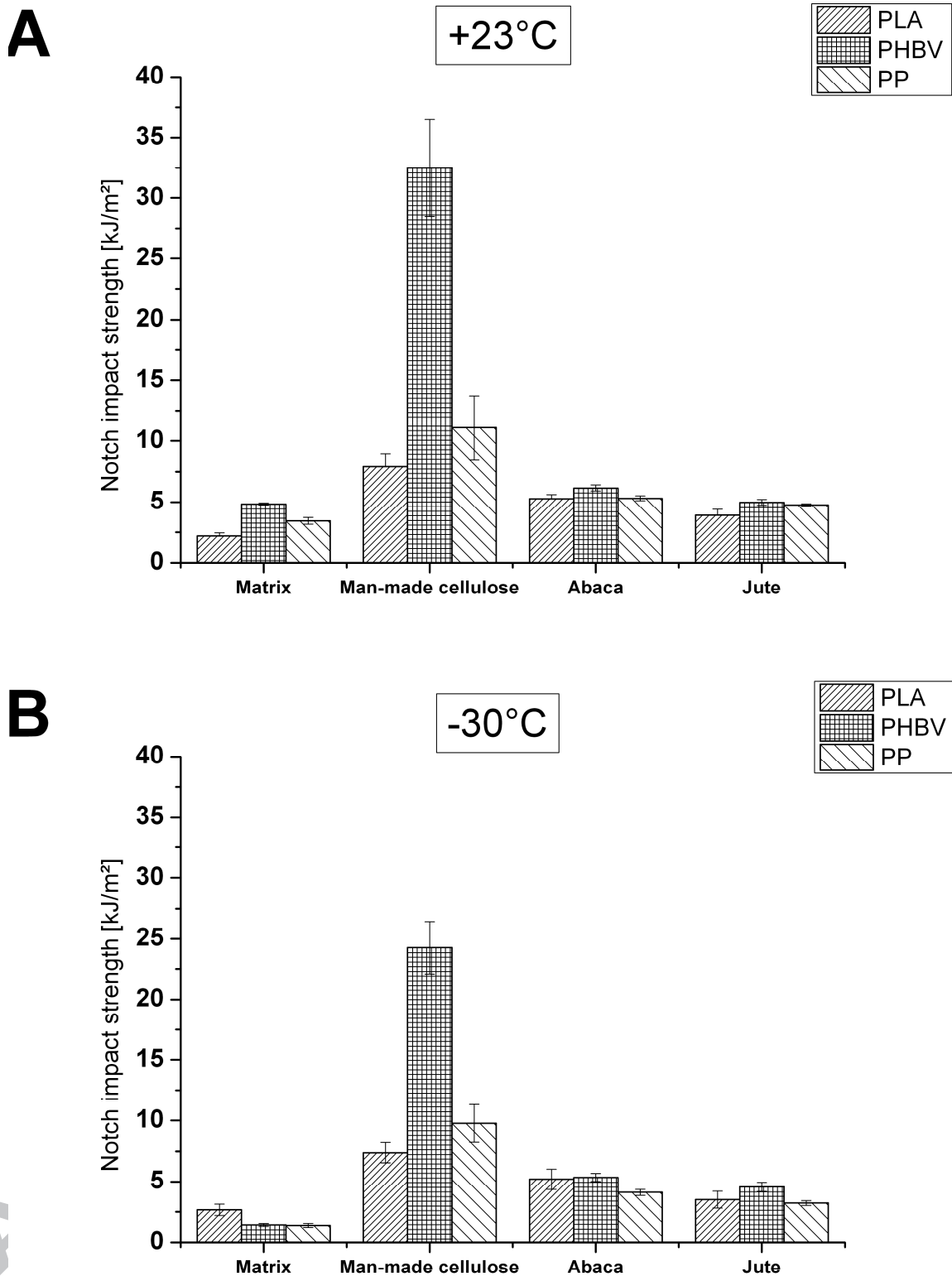


Figure 3: Charpy A-notch impact strength; above testing temperature +23°C (A), below testing temperature -30°C (B).

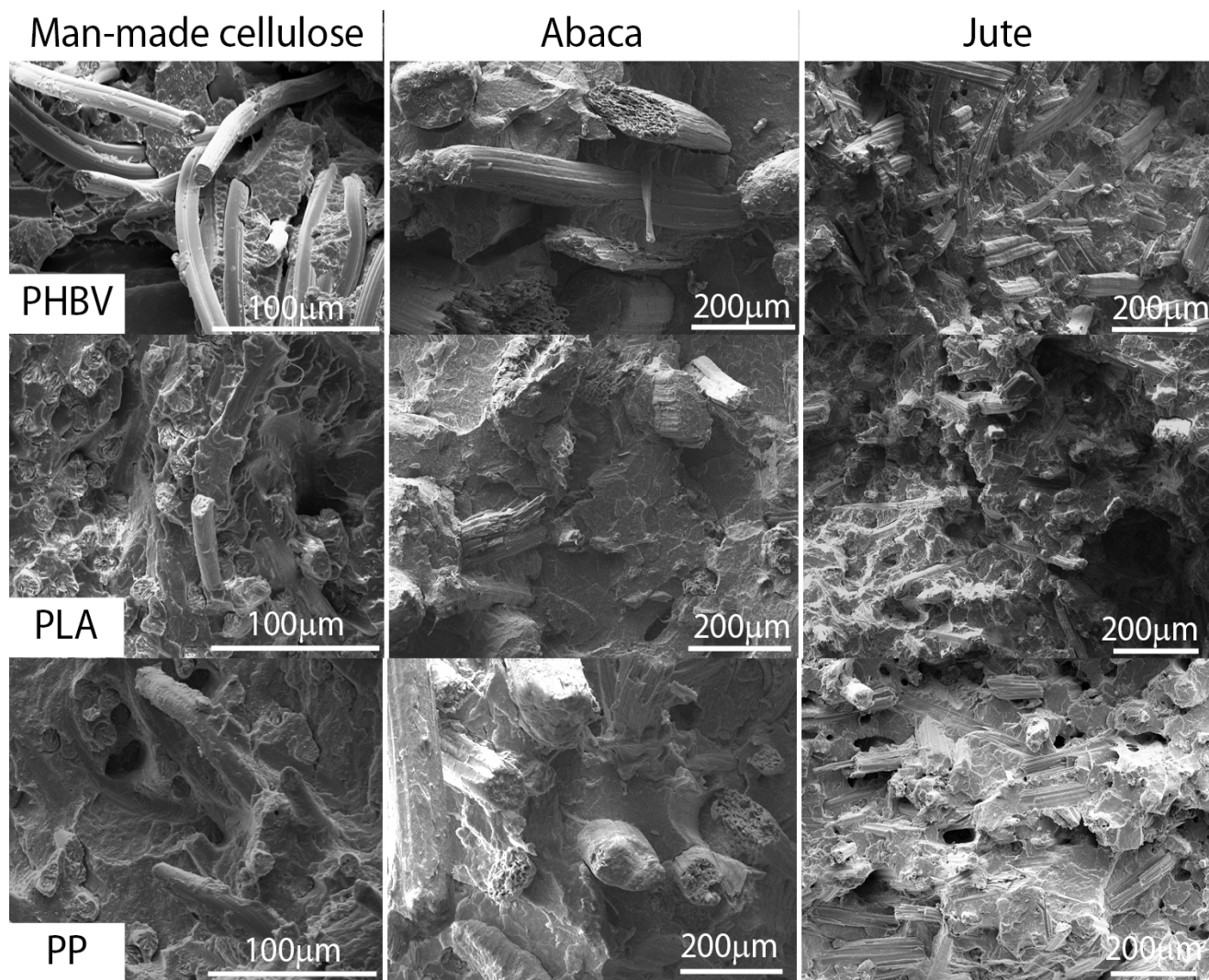


Figure 4: SEM micrographs of tested biocomposites: PHBV (above), PLA (middle) and PP (below).

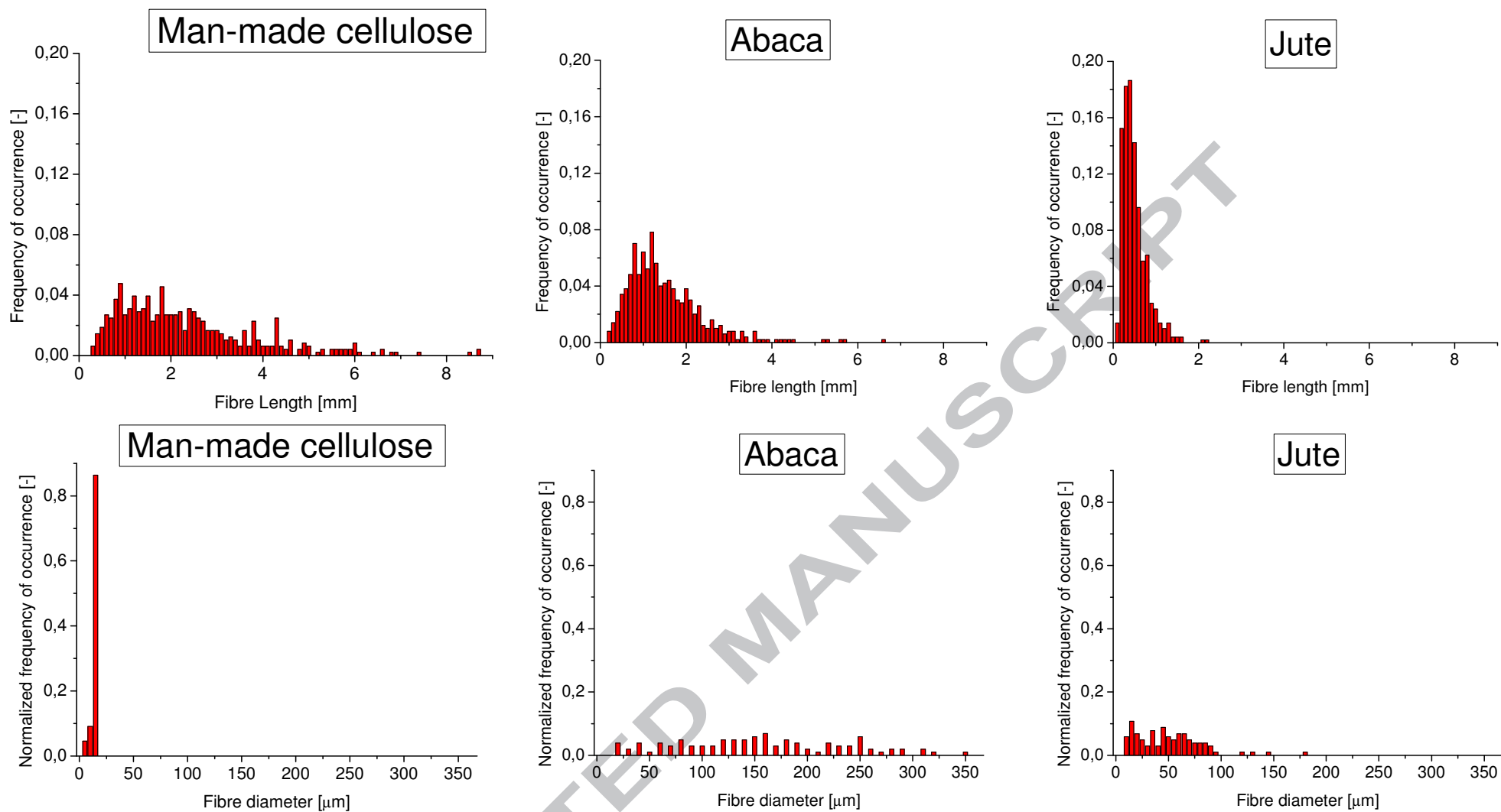


Figure 5: Fibre size distribution in PLA composites: length (above) and diameter (below).

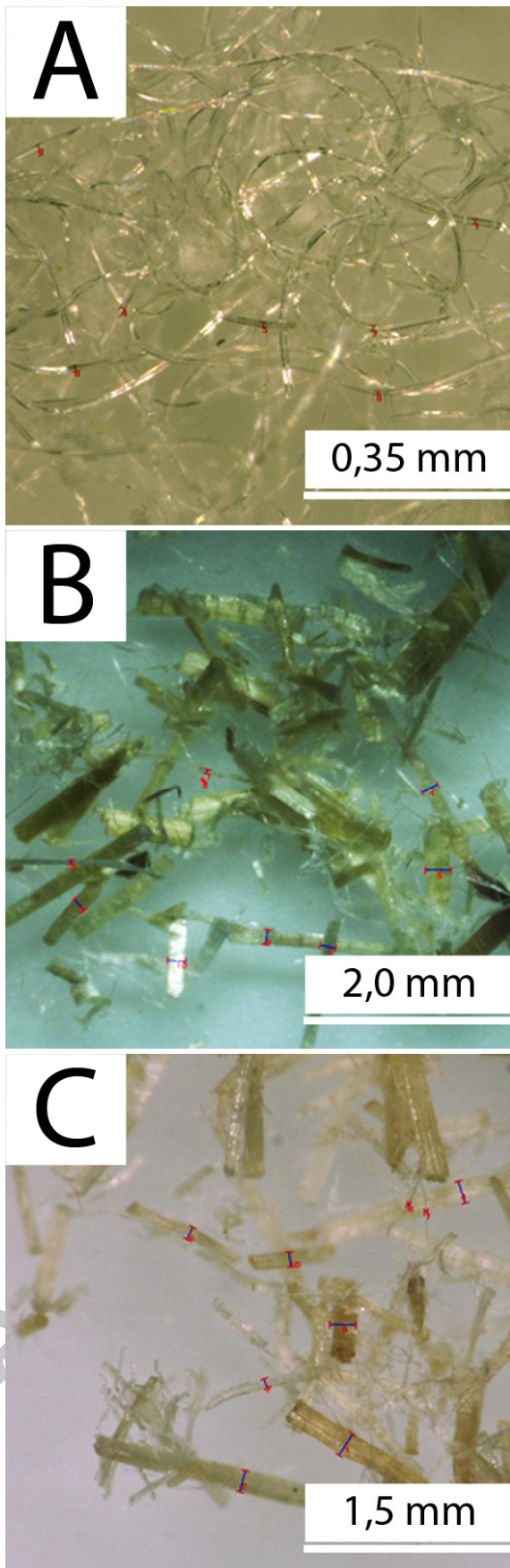


Figure 6: Microphotographs of extracted fibres; A) man-made cellulose, B) abaca, C) jute.