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Vol 2

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CONTENTS

Editorial

Each volume gathers contributions on specific topics:

- **Vol 1. Industrial applications**
- **Vol 2. Material science**
- **Vol 3. Material and Structural Behavior – Simulation & Testing**
- **Vol 4. Experimental techniques**
- **Vol 5. Manufacturing**
- **Vol 6. Multifunctional and smart composites**
- **Vol 7. Life cycle performance**
- **Vol 8. Special Sessions**

This collection contains the proceedings of the 21st European Conference on Composite Materials (ECCM21), held in Nantes, France, July 2-5, 2024. ECCM21 is the 21st in a series of conferences organized every two years by the members of the European Society of Composite Materials (ESCM). As some of the papers in this collection show, this conference reaches far beyond the borders of Europe.

The ECCM21 conference was organized by the Nantes Université and the Ecole Centrale de Nantes, with the support of the Research Institute in Civil and Mechanical Engineering (GeM).

Nantes, the birthplace of the novelist Jules Verne, is at the heart of this edition, as are the imagination and vision that accompany the development of composite materials. They are embodied in the work of numerous participants from the academic world, but also of the many industrialists who are making a major contribution to the development of composite materials. Industry is well represented, reflecting the strong presence of composites in many application areas.

With a total of 1,064 oral and poster presentations and over 1,300 participants, the 4-day event enabled fruitful exchanges on all aspects of composites. The topics that traditionally attracted the most contributions were fracture and damage, multiscale modeling, durability, aging, process modeling and simulation and additive manufacturing.

However, the issues of energy and environmental transition, and more generally the sustainability of composite solutions, logically appear in this issue as important contextual elements guiding the work being carried out. This includes bio-sourced composites, material recycling and reuse of parts, the environmental impact of solutions, etc.

We appreciated the high level of research presented at the conference and the quality of the submissions, some of which are included in this collection. We hope that all those interested in the progress of European composites research in 2024 will find in this publication sources of inspiration and answers to their questions.

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Table of Contents

Click on **CONTENTS** on the right side to return to the table of contents

Table of Contents

Table of Contents

Table of Contents

Table of Contents

Table of Contents

Table of Contents

FIBRILLATION - IMPROVING THE FIBRE/MATRIX ADHESION OF LYOCELL FIBRES FOR USE IN SHORT FIBRE-REINFORCED AND 3D PRINTED COMPOSITES

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Keywords: regenerated cellulose, fibrillation; fibre/matrix adhesion; additive manufacturing

Abstract

This study investigates the influence of fibrillation of lyocell fibres on the mechanical properties of compression moulded polylactide (PLA), polypropylene (PP) composites, and 3D printed PLA composites. Fibrillation was shown to reduce the strength and elongation at break of the fibres without affecting the Young's modulus compared to untreated fibres. Nevertheless, fibrillation in composites resulted in a 1.15 higher strength for PP composites and 1.62 for PLA composites. Young’s modulus and impact strength were increased by factors of 1.41 and 1.38 for PP composites and 1.2 and 1.23 for PLA composites. Applying the fibrillated fibres in 3D printed PLA shows a significant increase in the mechanical properties. For example, with a fibre mass fraction of 30%, the tensile strength of the composites with fibrillated fibres was increased by a factor of 1.18 compared to composites with untreated fibres. The use of maleic anhydride in the PLA matrix in combination with composite heat treatment further increased the strength by a factor of 1.46. With a strength of 85 MPa, a Young's modulus of 7.2 GPa and an elongation at break of 3.2%, these are some of the highest values reported for this kind of 3D printed materials.

1. Introduction

Using regenerated cellulose fibres, especially lyocell fibres, in composite applications is not new. Lyocell fibres are particularly characterised by their high toughness compared to plant bast fibres (see, e.g., [1]). Improving the adhesion between the generally hydrophobic matrix and the hydrophilic fibre is important. Besides using adhesion promoters [2], alternative approaches involve modifying the fibre surface, e.g. by plasma treatment, enzymatic processes, ultrasound or chemical treatment methods. One method for significantly increasing the specific surface area and, thus, the bonding surface between the matrix and the fibre is fibrillation. While fibrillation is unwanted in textile applications and makes processing more difficult, this process can offer distinct advantages in fibre-reinforced composites. Similar to a plant root system, the nanofibrils can intertwine with the matrix, making them more resistant to detaching from the matrix when force is applied [3].

In this study, fibrillation was deliberately induced to improve the reinforcing effect of short and randomly oriented lyocell fibres in a PLA and PP matrix produced by compression moulding [4] and to improve the properties of short fibre-reinforced, 3D printed PLA materials [5]. In a previous study, it was shown that improved adhesion affects especially short fibre-reinforced materials. Material extrusion processes, particularly fused deposition modelling (FDM), are one of the most commonly used methods for 3D printing with polymers or polymer composites due to their simplicity and low operating costs [6]. Therefore, the present study investigated compression moulded and 3D printed composites with short fibre reinforcement and random fibre orientation. Additionally, the use of maleic anhydride (MA) on the properties of PLA and as a potential adhesion promoter between cellulose fibre and matrix is being investigated. This approach to improve fibre/matrix adhesion has already been tested [7]. Some materials were also subjected to heat treatment to increase the crystallinity of the PLA.

2. Materials & methods

Lyocell fibres with a fineness of 15.0 dtex and a length of 90 mm were used to produce the compression-moulded samples (Lenzing AG, Lenzing, Austria). The matrices used were PLA (SLN 2660 D Ingeo fibres from Eastern Textile Ltd. (Taipei, Taiwan) with a fibre fineness of 6.0 dtex and a staple fibre length of 64 mm) and PP fibres (lot no. PP-N08 with a diameter of approx. 35 µm without adhesion promoters supplied by Nafgo GmbH, Dötlingen-Neerstedt, Germany). For the 3D printed materials, lyocell fibres with a fineness of 1.3 dtex (FCP400) with a nominal length of 400 µm (Lenzing AG, Lenzing, Austria) and as matrix PLA (grade 2003D from NatureWorks®) were used.

The fibres were fibrillated in an ultrasonic bath (Emmi-H22, EMAG AG, Mörfelden-Walldorf, Germany, operating frequency of 40 kHz and an ultrasonic power of 120 W) in a multi-stage treatment with demineralised water for 30 min, isohexane for 60 min, acetone for 60 min and demineralised water for 15 min. The fibres were dripped onto cellulose paper for 5 min between the treatment steps. To measure the fibre width of lyocell 15.0 dtex fibres and the length of lyocell 1.3 dtex fibres, the FibreShape software (IST AG, Vilters, Switzerland) was used. Fibres were conditioned at 20 °C and 65% relative humidity. For untreated and fibrillated fibres, 3 slide frames of each sample (Gepe Produkte AG, Zug, Switzerland) were prepared with fibre snippets and scanned with a slide scanner (CanoScan FS 4000US, Canon Deutschland GmbH, Krefeld, Germany; transmitted light mode with a resolution of 4000 dpi). The influence of fibrillation on the mechanical properties of the lyocell fibres was analysed using 15.0 dtex lyocell fibres in fibre tensile tests (clamping length 20 mm, test speed 10 mm/min, test conditions: 20 °C, 65% rel. humidity; test device Fafegraph M testing machine, Textechno, Mönchengladbach, Germany; equipped with a pneumatic clamping system and a 100 cN load cell). The influence of fibrillation on fibre/matrix adhesion was investigated using microbond tests. For this purpose, individual lyocell fibres with a fineness of 15.0 dtex were placed in a perforated aluminium frame, and, in the case of the PLA matrix, 2-3 PLA fibres were knotted around a single lyocell fibre; for PP, one fibre was knotted around one lyocell fibre. The ends of the polymer fibres were cut off close to the lyocell fibre, and the aluminium frame was treated in an oven for 5 minutes at 185 °C to melt the polymers. The embedding length of the fibres was measured under a microscope and the fibres were then pulled out of the matrix in a universal testing machine at a test speed of 1 mm/min and a free gauge length of 5 mm with a self-developed test device for a Zwick/Roell Z020 universal test machine (Zwick/Roell GmbH, Ulm, Germany; load cell 5 N). The forces were measured to determine the shear strength (test conditions: 23 °C, 50% rel. humidity).

For the compression moulded composites, lyocell 15.0 dtex fibres were carded with PLA or PP fibres to blend them using a manual roller card (Standard 46 tpi, fleece size 78 x 19 cm², Louët, Lochem, The Netherlands). The fibre content was set to 30 vol.-%. The multilayer webs were cut to a size of approx. 200 x 200 mm² and dried for 18 h in an oven at 60 °C. The fibres were then pressed to a thickness of 1 mm using 1 mm thick spacers at a temperature of 185 °C and a pressure of 15 bar for 2 minutes (LaboPress P200S, Vogt Labormaschinen GmbH, Berlin, Germany). The sheets were shredded (shredder type EBA 2326C, Krug & Priester GmbH & Co. KG, Balingen, Germany) into pellets and then pressed again to a thickness of 1 mm. In order to ensure a higher homogeneity of the random fibre-oriented composites, a further shredding process was applied. Afterwards, the granules were pressed to a thickness of 2 mm using 2 mm thick spacers at a temperature of 185 °C and a pressure of 15 bar for 5 min. The test specimens for the tensile and impact tests were produced from these sheets with a band saw.

A fibre content of 30 mass% was used for the 3D printing materials. The PLA matrix and fibres were blended at 180-195 °C with a Sigma blade-type compounder for 10 min. Compounds were produced with and without maleic anhydride (MA). For the materials with MA, 2% MA in relation to the PLA mass and 10% dicumyl peroxide in relation to the MA mass were added during the compounding process. 3D printing wires with a diameter of 1.7 mm were produced from the granulates manufactured. After compounding, the composites were granulated into particles of < 4 mm using a Moretto GR knife mill (Mercer County, PA, USA). The particles were vacuum dried at 60 °C for 4 h and extruded using a Filabot EX2 single screw extruder (Barre, VT, USA) at 180-185 °C. An air cooling system was used to cool the thermoplastic wire. The 3D printing was carried out using a fused deposition modeling (FDM) process with a MakerGear™ M2 desktop 3D printer (Beachwood, OH, USA) using the Simplify 3D® software package for slicing the CAD files and controlling the 3D printer. Before printing, all the wires were vacuum-dried at 50 °C for 2 h. All samples were printed using a perimeter (shell) of one printing line (approx. 0.75 mm). After 3D printing, one set of samples for each formulation was heat treated at 105 ± 2 °C for 2 h in a laboratory oven.

Tensile characteristics of compression moulded composites (n=6; dimensions 200 x 15 x 2 mm³) were tested at a clamping length of 125 mm and a test speed of 2 mm/min with a universal testing machine type Zwick Z020 (Zwick/Roell GmbH, Ulm, Germany) equipped with a 20 kN load cell and a pneumatic clamping system (clamping pressure: 1 - 2 bar) at a crosshead speed of 2 mm/min. The elongation was determined using a video extensometer (VideoXtens, Zwick/Roell GmbH, Ulm, Germany; measuring mark distance 80 mm). The impact properties were investigated using unnotched Charpy impact tests (n=8; dimensions 80 x 10 x 2 mm³; pendulum device type 5102, Zwick GmbH, Ulm, Germany) with a bearing distance of 40 mm and broadside impact.

The 3D-printed tensile test specimens (n=5; ASTM D638 type V samples, dimensions 10.96 x 3.18 x 1.2 mm³) were tested in an Instron® 5982 universal testing machine (Norwood, MA, USA) equipped with a 5 kN loadcell) using a crosshead speed of 2 mm/min. The elongation was measured with a 10 mm clip-on extensometer.

SEM investigations were done with a JSM 6510 scanning electron microscope (Jeol, Eching, Germany) operating with secondary electrons. Prior to SEM investigations, the samples were sputtered with a gold layer for 90 s under a current of 56 mA using a Bal-Tec sputter coater type SCD 005 (Bal-Tec, Liechtenstein).

More details on the materials and methods used are described in previous publications [4, 5].

3. Results & discussion

3.1 Effect of fibrillation on fibre surface and mechanical characteristics

Figure 1 shows the influence of the surface treatment on the lyocell fibres with a fineness of 15.0 dtex. The fibres are clearly fibrillated due to swelling and ultrasonic treatment. It becomes apparent that the fibres are only fibrillated in the outer areas, whereas the fibre core has remained intact. The measured nanofibrils have widths between ~100 and ~400 nm. It is to be expected that the fibrils lead to better bonding to the matrix in a composite material due to the increase in surface area, similar to a plant root system in the soil. The fibre width did not change significantly as a result of the treatment and resulted in mean values of 36.0 ± 5.6 µm (median value: 36.0 µm) for the untreated fibre and 35.7 ± 5.4 µm (median value: 35.6 µm) for the fibrillated fibre. Results show good agreement with the calculated equivalent diameter of 35.7 µm.

It was shown that fibrillation slightly reduces the tensile strength and elongation at break of the fibres, while it does not affect Young´s modulus. A median value of 251 MPa was determined for the tensile strength of the untreated fibre and 192 MPa for the fibrillated fibre. The fibrillation did not significantly influence Young's modulus, as it is determined in the linear-elastic initial slope, and the influence of possible defects is lower. The Young´s modulus resulted in a median value of 6.47 GPa for the untreated fibre and 6.25 GPa for the fibrillated fibre. The elongation at break was reduced by the defects on the fibre surface, from a median value of 12.1% for the untreated fibre to 8.5% for the fibrillated fibre.

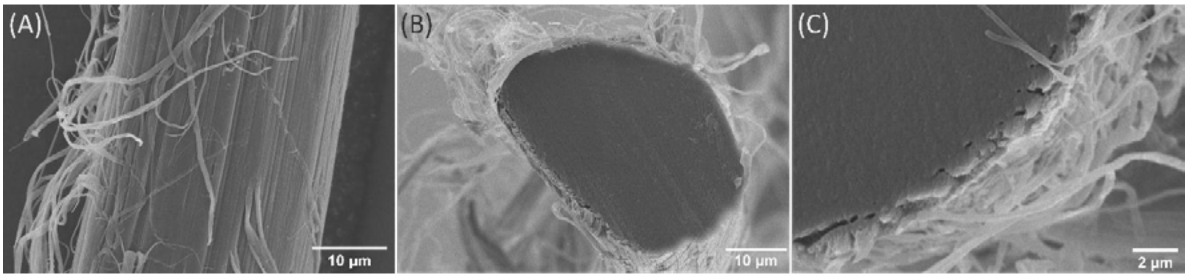

Figure 1: Fibrils on a lyocell fibre surface (A) in cross-section (B) and in detail in cross-section (C)

3.2 Influence of fibrillation on the fibre/matrix adhesion in a PLA and PP matrix and

The fibre/matrix adhesion of lyocell fibres with a fineness of 15.0 dtex was tested in a PLA and PP matrix using microbond tests. The median interfacial shear strength could be increased from 3.3 to 4.4 MPa in a PP matrix and from 7.1 to 8.2 MPa in a PLA matrix using fibrillated fibres compared to untreated fibres (Figure 2 A&B). Better adhesion of lyocell in PLA than in a PP matrix is achieved. The improvement in IFSS is not significant from a statistical point of view. It should be noted that only individual fibres are tested in the microbond test, whereas a large number of fibres are present in a composite, and the effects of the improved adhesion are more pronounced. It is assumed that individual fibrils anchor themselves in the matrix and thus, lead to a larger specific bonding area between fibre and matrix, which improves the shear strength. In addition, fibrillation leads to a higher roughness of the fibre surface, which can increase friction and mechanical interlocking. At the same time, fibrillation significantly reduces the critical fibre length, which means that a shorter fibre length can have a reinforcing effect in a composite. Analogous to the better adhesion in the PLA matrix, it was found that the critical fibre length for PLA composites is lower than for PP-based composites (Figure 2 C&D). This is particularly interesting when short fibres with lengths around the critical fibre length are used, e.g. in injection-moulded or 3D printed composites.

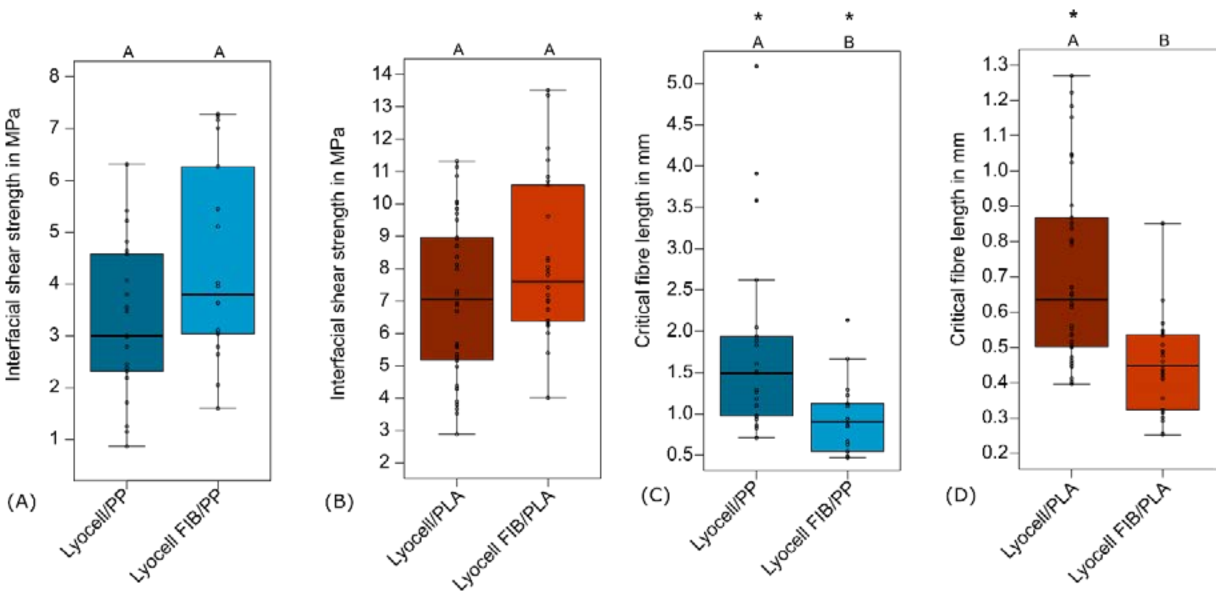

Figure 2: Box-Whisker plots of the interfacial shear strength of lyocell fibres embedded in a PP matrix (A) and a PLA matrix (B) and the critical fibre length in a PP matrix (C) and PLA matrix (D) (results that do not comply with a normal distribution are marked with an asterisk, significant differences between median values are labelled with different letters; figure reproduced from [4])

3.3 Influence of fibrillation on compression moulded short fibre-reinforced PLA and PP composites

Composites with short fibres and random fibre orientation were produced by compression moulding to evaluate whether fibrillated lyocell fibres can improve the mechanical properties in a PP and PLA matrix. Figure 1 shows that fibrillation only takes place on the fibre surface, and the core of the fibre is hardly affected. Despite the lower tensile strength of the fibrillated lyocell fibres, the strength of the composites was increased by a factor of 1.15 for PP and 1.62 for PLA compared to the composites produced with untreated fibres (Figure 3 A&D). The Young´s modulus of the composites was increased by a factor of 1.41 for PP and 1.20 for PLA with fibrillated fibres (Figure 3 B&E). The increase in strength can be explained by an improvement in adhesion due to anchoring effects caused by the larger specific fibre surface area as described by Karlsson et al. [3].

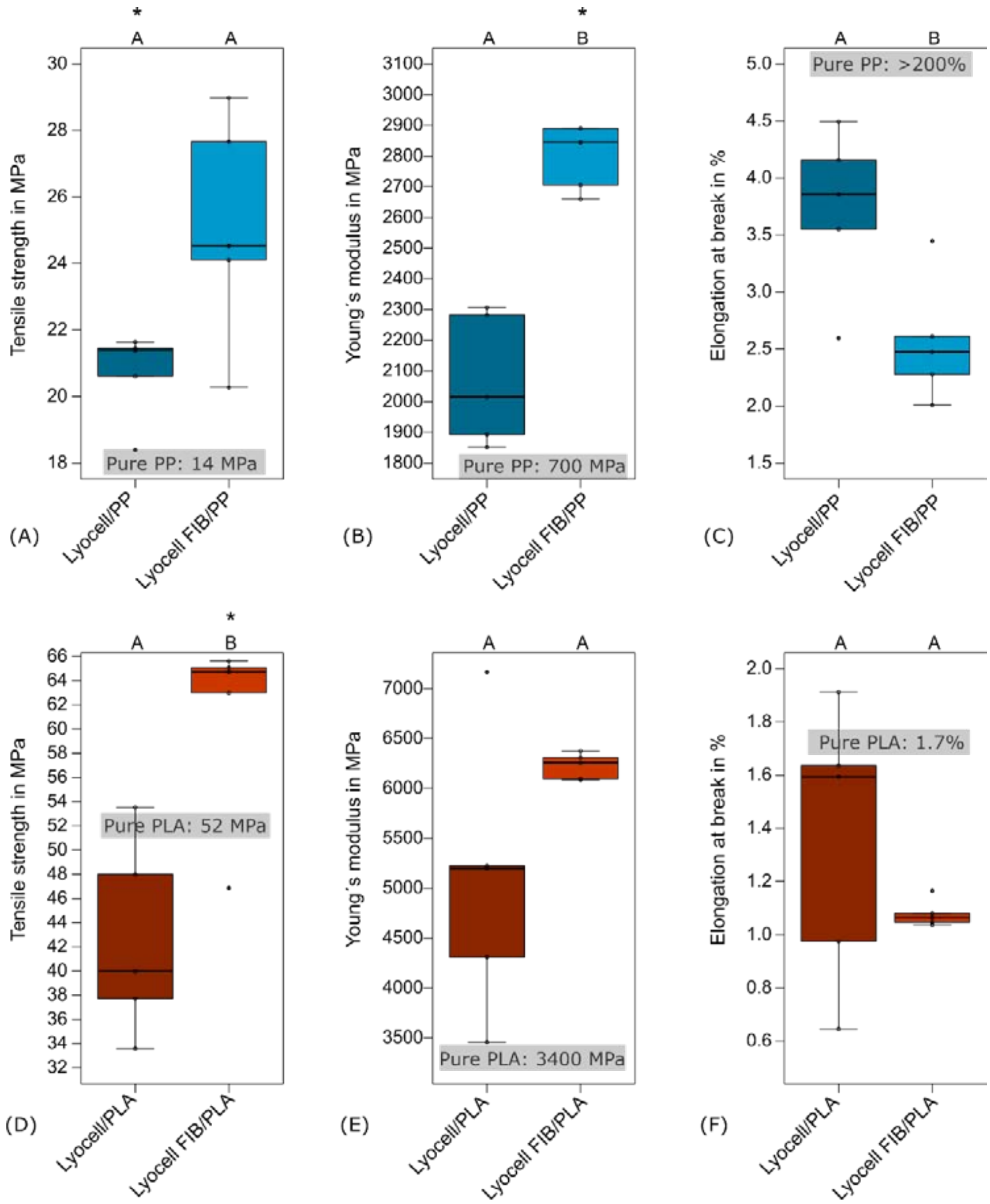

Figure 3: Box-Whisker plots of the tensile characteristics of lyocell/PP composites (A-C) and lyocell/PLA composites (D-F) (results that do not comply with a normal distribution are marked with an asterisk, significant differences between median values are labelled with different letters; figure reproduced from [4])

SEM analyses indicate that the improved adhesion between the fibrillated lyocell fibres and the PP and PLA matrices, as demonstrated by the results of the microbonding tests, can be transferred to the composites. Figure 4 shows an example of a fracture surface of a composite made with fibrillated lyocell fibres. A ripped lyocell fibre can be seen on which fibrils are visible; a broken lyocell fibre coated with PP matrix is visible on the right. At the point of breakage, fibrils protrude from the matrix and bridge the crack, delaying it. The fibrils serve as a kind of anchor within the matrix. Since the Young's modulus of the fibres was not significantly influenced by the fibrillation, structural effects must be a reason for the increased Young's modulus of the composites. Only the main fibre is tested when measuring the fibre Young's modulus. The nanofibrils cannot be measured and, therefore, do not influence the measured Young's modulus. However, in a composite, they are embedded in the matrix and contribute significantly to the stress transfer from the matrix to the fibre. It is assumed that the individual nanofibrils embedded in the matrix contribute to an increase in Young's modulus. Due to the increased adhesion of the nanofibrils, the elongation at break of the composite is reduced. While the elongation at break of the PLA matrix is barely affected by the lyocell fibres, the very high elongation at break of the PP matrix (>200%) is significantly reduced by the considerably lower elongation of the fibres (Figure 3 C&F).

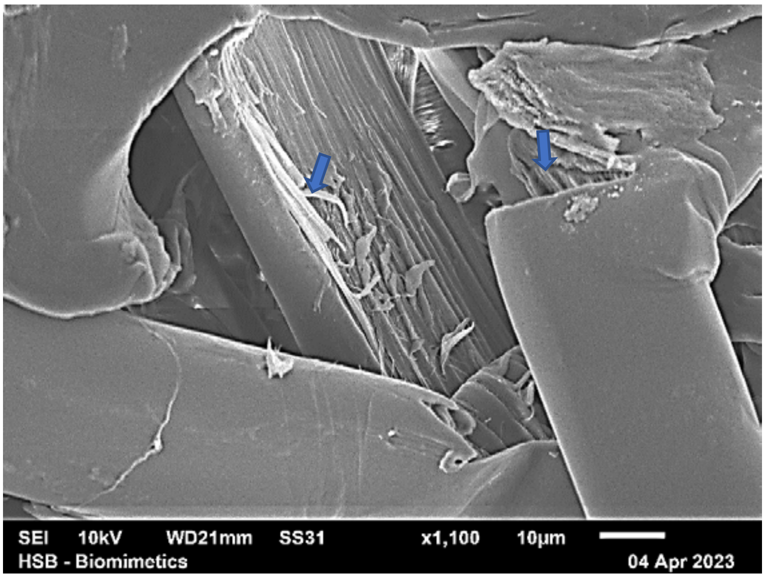

Figure 4: Fracture surface of a lyocell/PP composite (fibrils are marked with blue arrows)

For both the PLA and the PP matrix, the impact strength could not be improved with the lyocell fibres. The reason for this is the use of short fibres with random orientation. These fibres prevent the energy-absorbing fibre pull-outs. Using longer and oriented fibres leads to significantly improved energy absorption by fibre pull-outs when the material breaks. However, the use of fibrillated lyocell fibres results in a significant increase in impact strength compared to untreated fibres. The unnotched Charpy impact strength was improved by a factor of 1.38 for PP composites and 1.23 for PLA composites by using fibrillated fibres.

3.4 Influence of fibrillation on 3D printed short fibre-reinforce PLA composites

As improved fibre/matrix adhesion has a more significant effect on the mechanical properties of short fibre-reinforced materials compared to long fibre-reinforced composites, fibrillated lyocell fibres were used to produce 3D printed PLA composites [5]. In order to increase the aspect ratio of the short lyocell fibres, 1.3 dtex fine lyocell fibres were used instead of the coarse lyocell 15.0 dtex fibres. Fibrillation was carried out as described for the 15.0 dtex lyocell fibres. An analysis of the fibre lengths (extracted fibres from composites) after compounding, wire production and 3D printing revealed fibre lengths for the different formulations between 116 and 134 µm and, thus, similar fibre length distributions. An influence of different fibre lengths can therefore be excluded [5].

The tensile properties of 3D-printed composites are summarised in Table 1. The formulation of fibrillated lyocell fibres and PLA show an improvement by factor 1.18 for tensile strength and 1.14 for Young's modulus compared to the composite with unmodified fibres and has similar tensile properties to the lyocell-reinforced maleic anhydride modified PLA (PLA-MA). The combination of the fibrillated fibres and the MA-modified PLA further improved the tensile properties, resulting in a tensile strength of 76.6 MPa, Young's modulus of 6.44 GPa, and elongation at break of 3.7%. These results represent an improvement of 1.31 in tensile strength and 1.28 in Young's modulus compared to the composite with untreated lyocell fibres and unmodified PLA matrix.

Table 1: Tensile properties of lyocell-reinforced composites produced by 3D printing

Composite	*Tensile strength in MPa*	*Young´s modulus in MPa*	*Elongation at break in %*
PLA	65.3	3460	6.7
PLA-MA	57.2	3320	4.0
30% lyocell 1.3 dtex/PLA	58.4	5050	2.9
30% fibrillated lyocell 1.3 dtex/PLA	69.2	5760	3.4
30% fibrillated lyocell 1.3 dtex/MA-PLA	76.6	6440	3.7
30% fibrillated lyocell 1.3 dtex/MA-PLA + heat treatment	85.0	7180	3.2

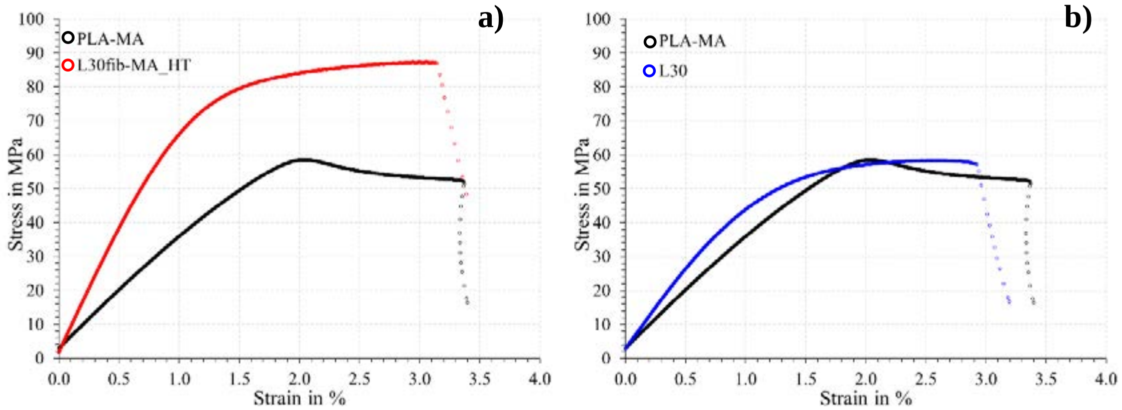

Figure 5: Stress-strain curves of the 3D printed composites produced from fibrillated lyocell/PLA (L30fib-MA_HT) after heat treatment (HT) (a) and untreated lyocell/PLA (L30) (b) samples in comparison with PLA-MA (figure reproduced from [5])

The mechanical properties of the composites with a high proportion of lyocell fibres can be further improved by increasing the PLA crystallinity through heat treatment (HT). Heat treatment was beneficial for all formulations, and the combination of fibre fibrillation, matrix modification (MA treatment) and heat treatment can achieve very promising mechanical properties such as a tensile strength of 85 MPa, a Young's modulus of 7.18 GPa and an elongation at break of 3.2% (see Table 1 and Figure *5*). These values correspond to an improvement by factor 1.30 in tensile strength and by 2.11 in Young's modulus compared to 3D printed pure PLA (1.49 and 2.12, respectively, compared to PLA-MA), without significantly affecting the elongation at break. Additionally, it has been demonstrated that increasing PLA crystallinity, when combined with reinforcing lyocell fibres, significantly enhances the thermo-mechanical stability of the composite. This is evidenced by a storage modulus at 80 °C that is up to 200 times higher than that of neat PLA [5].

4. Conclusions

The results show that the fibrillation of lyocell fibres positively affects adhesion in both a PP and a PLA matrix. The fibrillation process reduced the strength and elongation at break of the lyocell fibres while the Young's modulus remained constant. Despite the lower fibre strength, the tensile strength of compression moulded short fibre-reinforced PLA and PP composites was significantly increased by using fibrillated lyocell fibres in both matrices compared to the use of non-fibrillated fibres. The tensile modulus of the composites was also improved. Despite reducing the elongation at break of the composites, the fibrillated lyocell fibres significantly improved the impact strength. The use of fibrillated lyocell fibres in 3D printed PLA composites was tested, as it has been shown that an improvement in adhesion has a particular effect on short fibre-reinforced composites. The shear forces during extrusion and 3D printing allow the short fibres to partially align and split off nanofibrils from the main fibre, resulting in a significant increase in fibre surface area and fibre aspect ratio, which could significantly increase the reinforcing effect. The micro- and nanofibrils can anchor themselves in the matrix, similar to a plant root system. Fibrillated lyocell fibres could be successfully processed with a PLA matrix in 3D printing and showed a considerably increased tensile strength compared to materials made from untreated lyocell fibres. The mechanical properties were significantly increased by using maleic anhydride and heat treatment resulting in a tensile strength of 85 MPa, a Young's modulus of 7.2 GPa and an elongation at break of 3.2%. This class of materials has some of the highest characteristic values currently known in the literature for short fibre-reinforced 3D printing (fused deposition modeling - FDM) materials.

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[4] N. Graupner, S. Schmidt, C. Gauss, and J. Müssig. Making positive use of the fibrillation of lyocell fibres in composite materials. *Composites Part C: Open Access*, 11:100359, 2023.

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Fibrillation – improving the fibre/matrix adhesion of lyocell fibres for use in short fibre-reinforced and 3D printed composites

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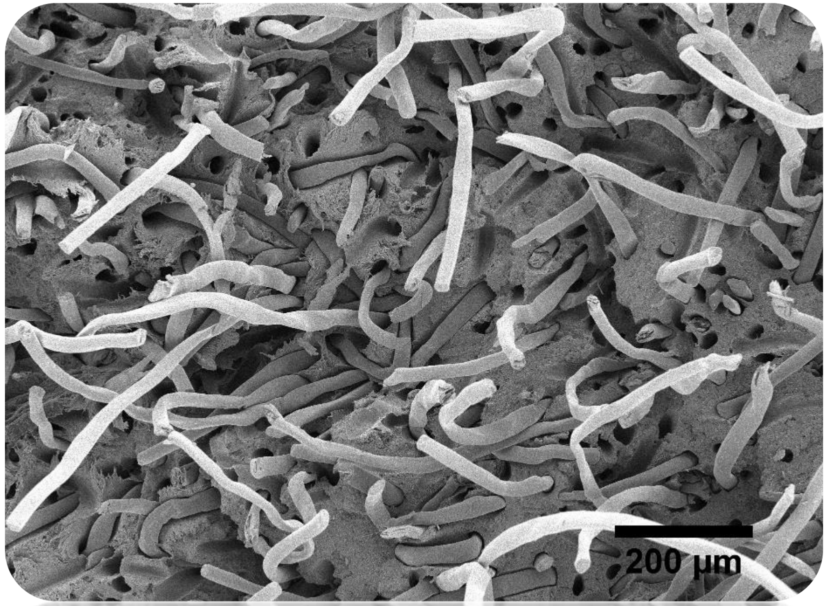
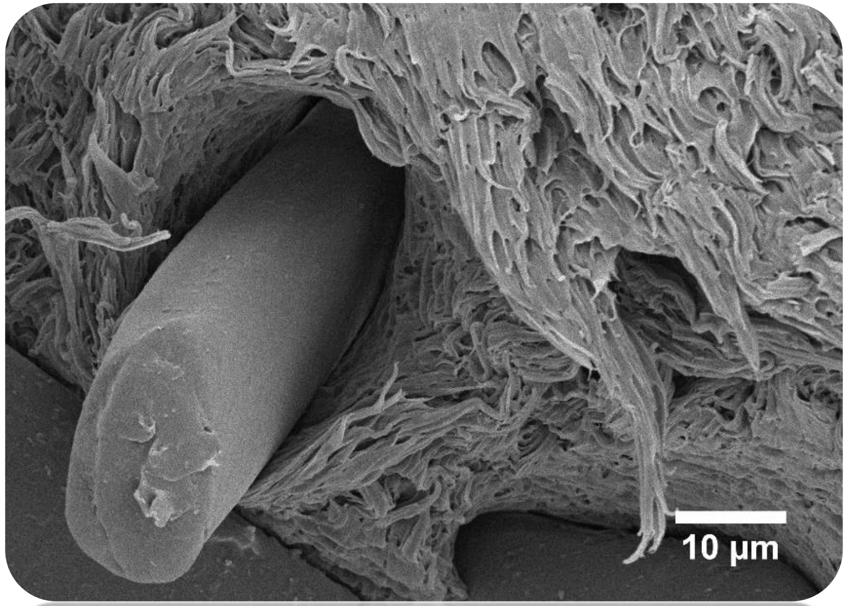
Motivation

- The adhesion of regenerated cellulose (lyocell) in plastic matrices is generally poor
- The fibres normally have smooth surfaces, resulting in fewer anchor points in the matrix
- In this way, particularly short fibres can absorb little energy when the materials break





Solution approach to improve fibre/matrix adhesion

- Solution: fibrillation
  - Fibrillation process according to Karlsson et al., 1996
  - Enlargement of the specific fibre surface
  - Improved pull-out resistance when the composite breaks
- Hypothesis: Increase in strength and toughness, especially for short fibre-reinforced composites

Plant root system

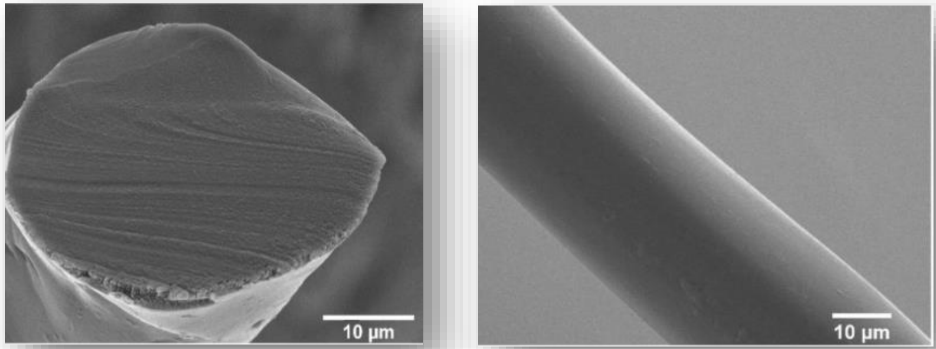
Fibrillated lyocell fibre

Karlsson, J. O. / Blachot, J.-F. / Peguy, A. / Gatenholm, P. (1996): **Improvement of adhesion between polyethylene and regenerated cellulose fibers by surface fibrillation**. *Polymer Composites* , 17(2): p. 300-304

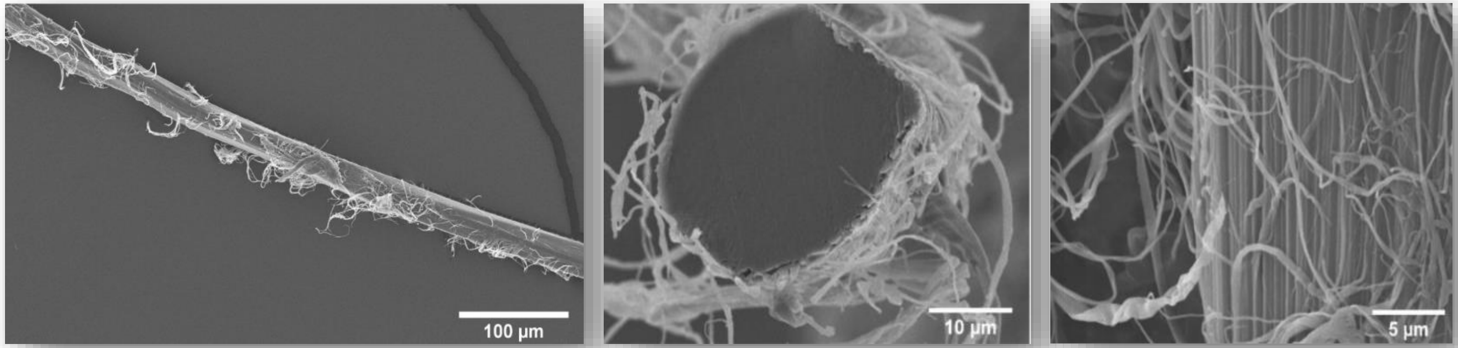
Fibrillation process

Fibrillation protocol:

- Fibrillation in an ultrasonic bath of lyocell 15.0 dtex fibres
  - Demineralised water for 30 min
  - Isohexane for 60 min
  - Acetone for 60 min
  - Demineralised water for 15 min
  - The fibres were dripped onto cellulose paper for 5 min between the treatment steps



Untreated lyocell fibre

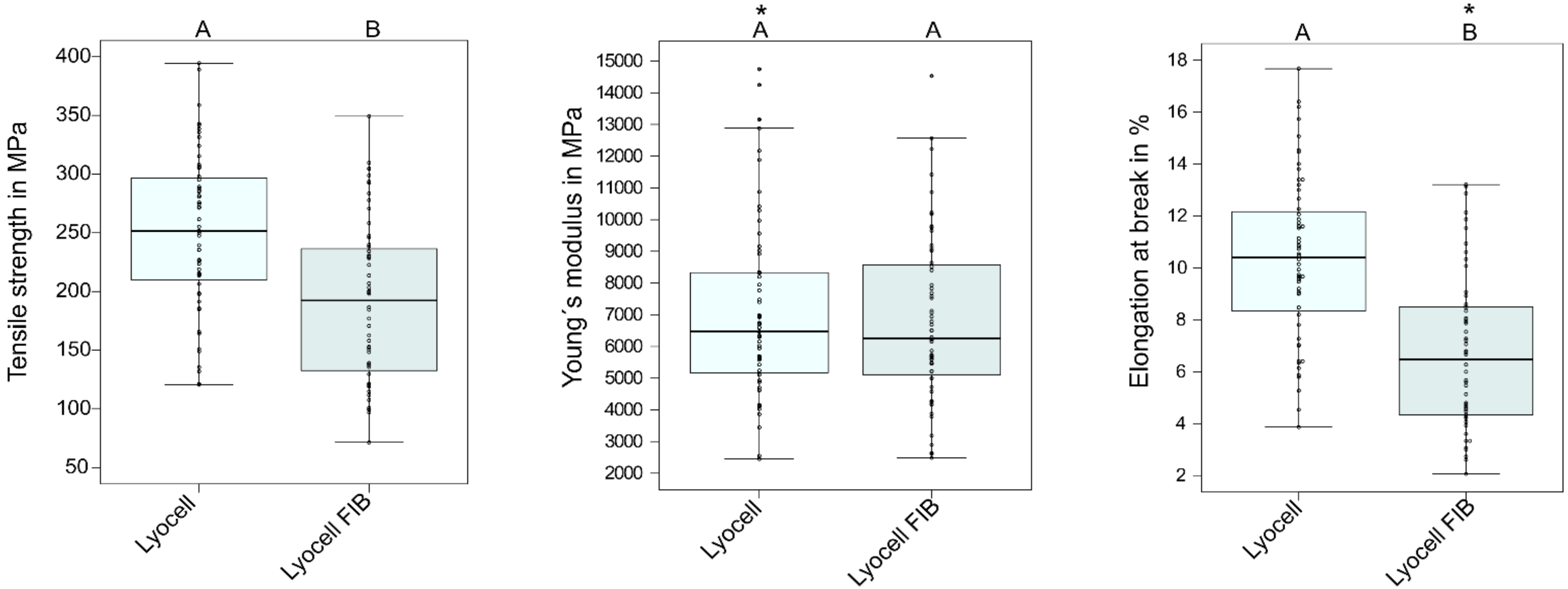


Fibrillated lyocell fibre

Graupner, N. / Schmidt, S. / Gauss, C. / Müssig, J. (2023): **Making positive use of the fibrillation of lyocell fibres in composite materials.** *Composites Part C: Open Access* , 11: 100359

Influence of fibrillation on the properties of lyocell fibres

- Tensile characteristics (Fafegraph M, gauge length 20 mm, test speed 10 mm/min)

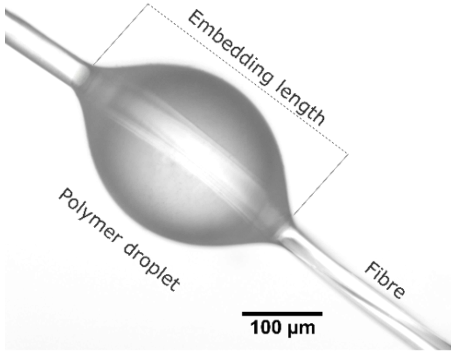


⇨ The fibrillation causes a significant reduction in strength and elongation at break and does not appear to have any influence on the Young´s modulus

Graupner, N. / Schmidt, S. / Gauss, C. / Müssig, J. (2023): **Making positive use of the fibrillation of lyocell fibres in composite materials.** *Composites Part C: Open Access* , 11: 100359

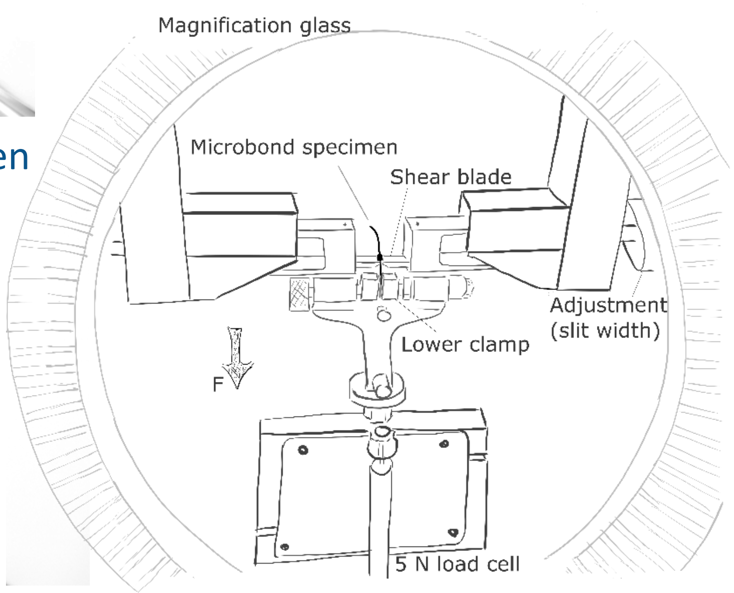
Interfacial shear strength measured with microbond tests

- Preparation and testing of lyocell/PLA

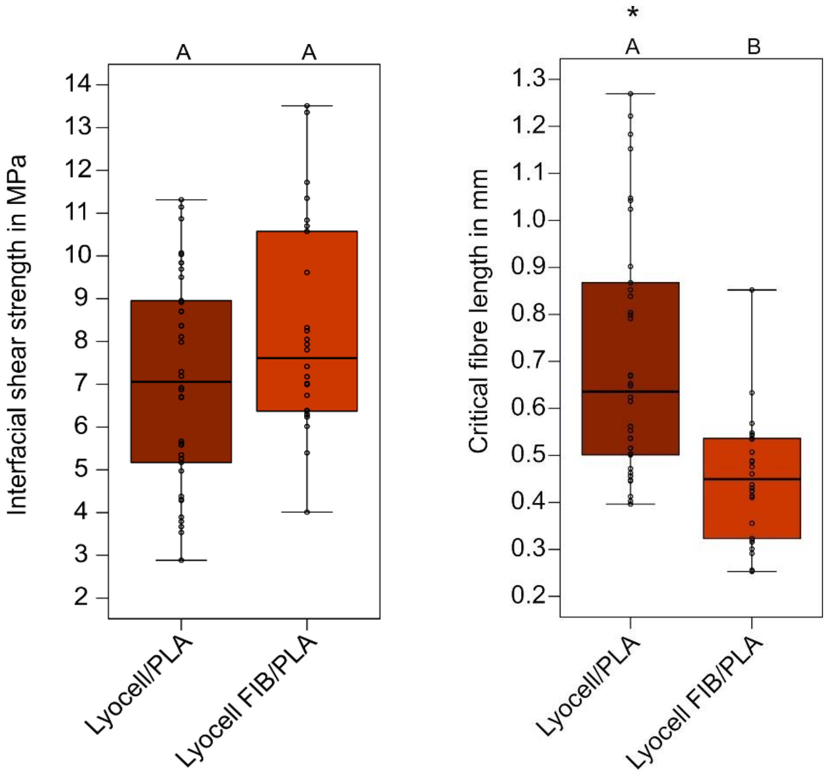


Microbond test specimen

Microbond test



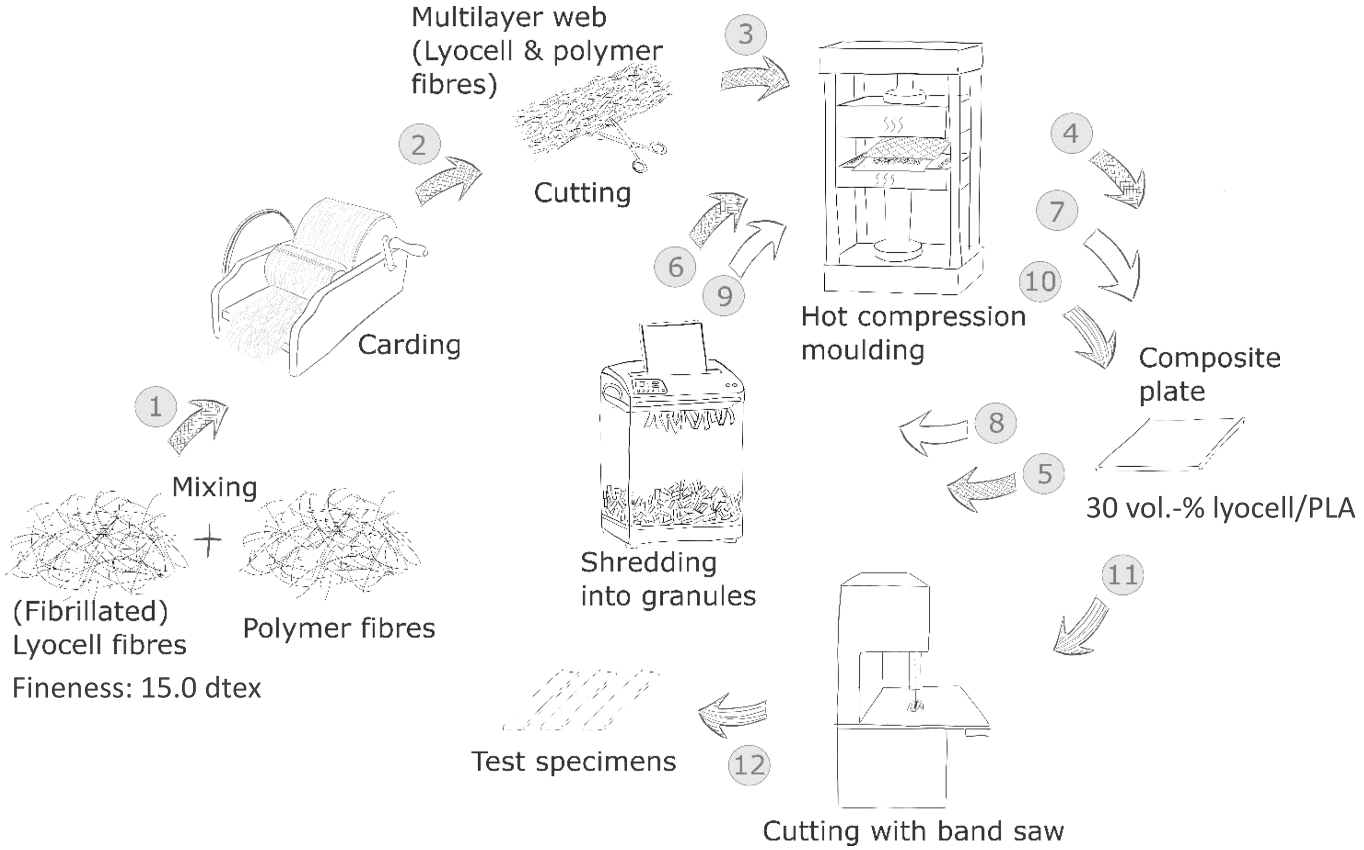
⇨ Results



Graupner, N. / Schmidt, S. / Gauss, C. / Müssig, J. (2023): **Making positive use of the fibrillation of lyocell fibres in composite materials.** *Composites Part C: Open Access* , 11: 100359

Use of short fibrillated lyocell fibre for PLA based composites

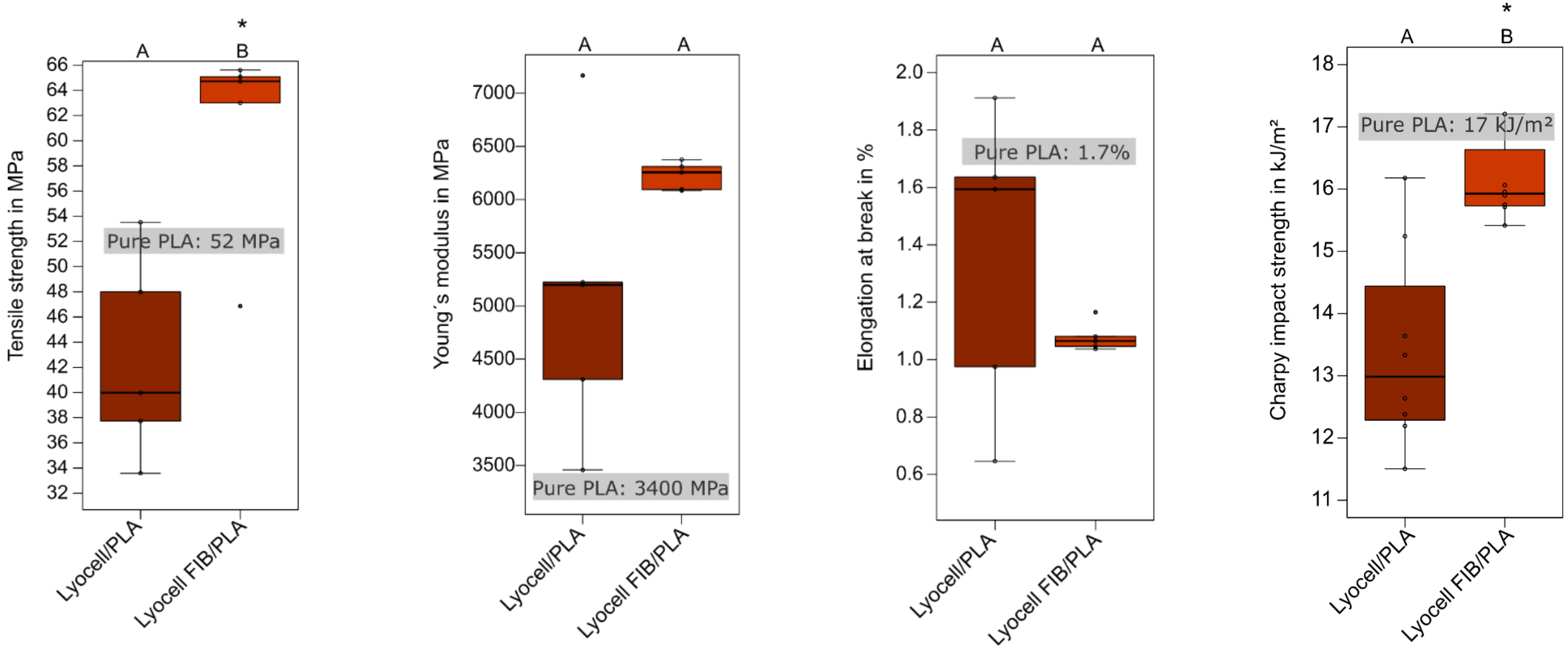
- Composite production by compression moulding



Lyocell/PLA granules

Graupner, N. / Schmidt, S. / Gauss, C. / Müssig, J. (2023): **Making positive use of the fibrillation of lyocell fibres in composite materials.** *Composites Part C: Open Access* , 11: 100359

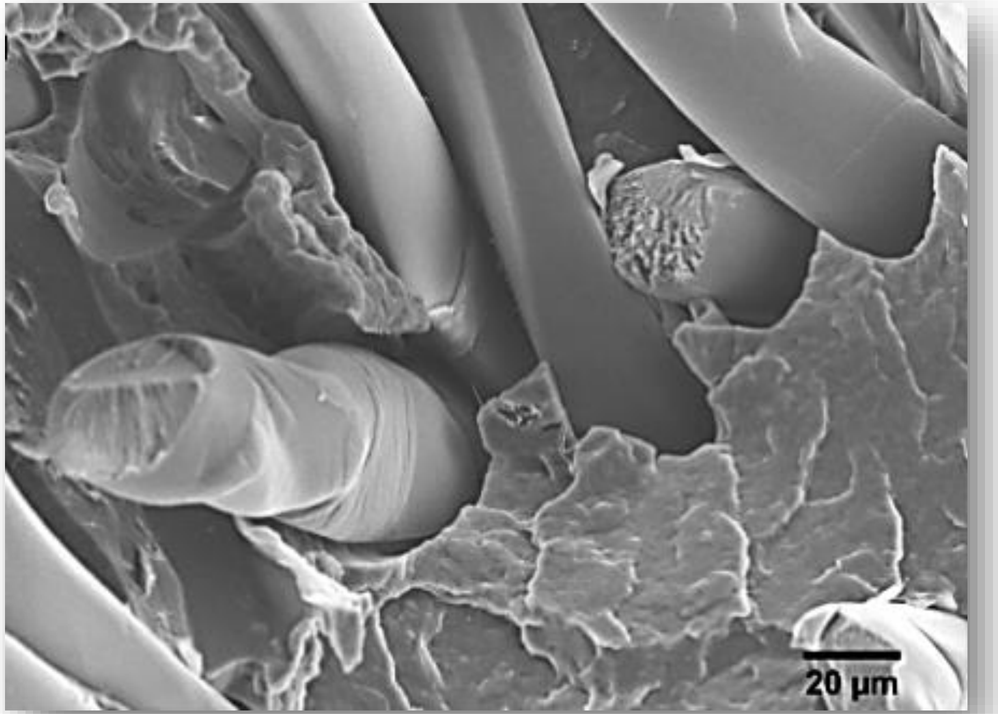
Mechanical properties of compression moulded 30 vol.% lyocell/PLA composites



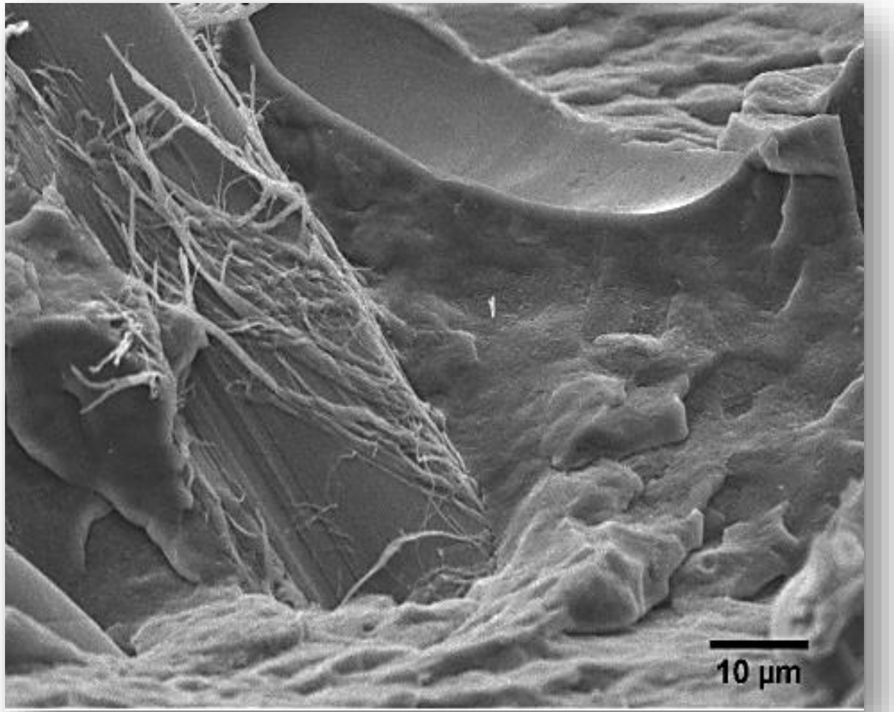
- ⇨ Tensile strength and Young´s modulus could be clearly increased by fibrillation
- ⇨ Elongation at break was not significantly affected by fibrillation
- ⇨ Unnotched Charpy impact strength could be significantly increased by fibrillation

Graupner, N. / Schmidt, S. / Gauss, C. / Müssig, J. (2023): **Making positive use of the fibrillation of lyocell fibres in composite materials.** *Composites Part C: Open Access* , 11: 100359

SEM investigations of fracture surfaces



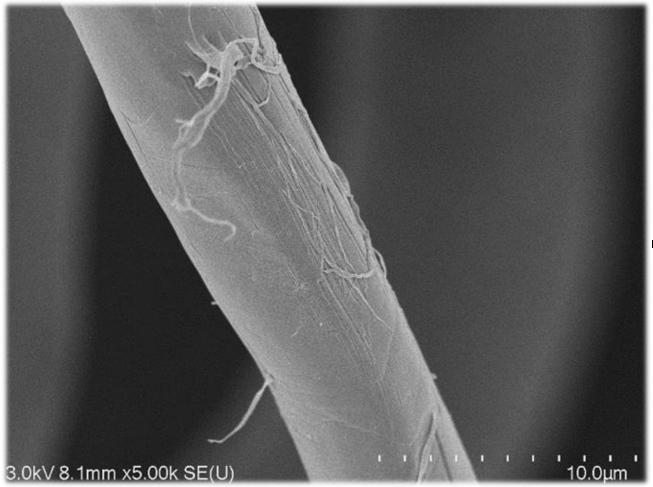
Untreated lyocell/PLA composite



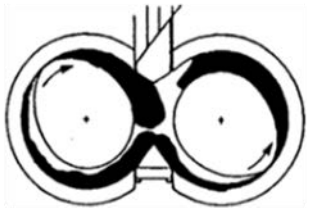
Fibrillated lyocell/PLA composite

⇨ The fibrils act as an anchoring system, similar to a plant root system, and contribute to improved adhesion of the fibres in the matrix

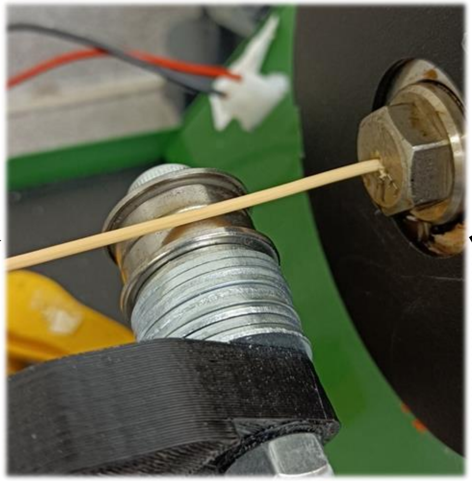
3D printing of lyocell/PLA and fibrillated lyocell/PLA composites



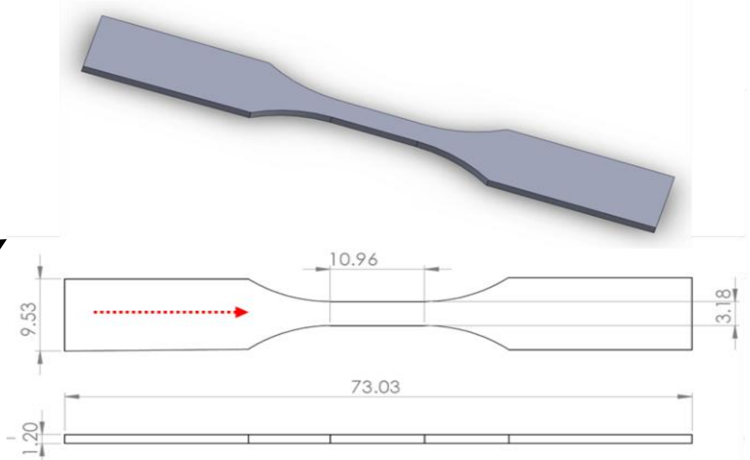
Fibrillated lyocell fibres (fineness 1.3 dtex)



Blending PLA matrix and with and without maleic anhydride (MA) (fibre content of 30 mass%)



3D printing filaments (≈ 1.7mm)



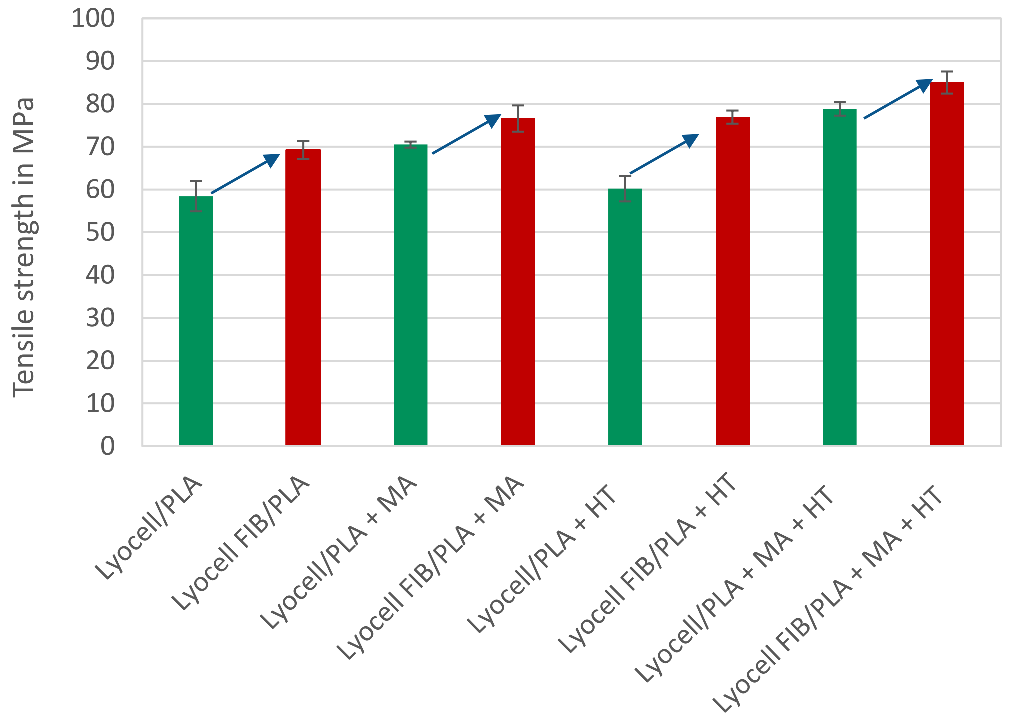
3D printed test specimen

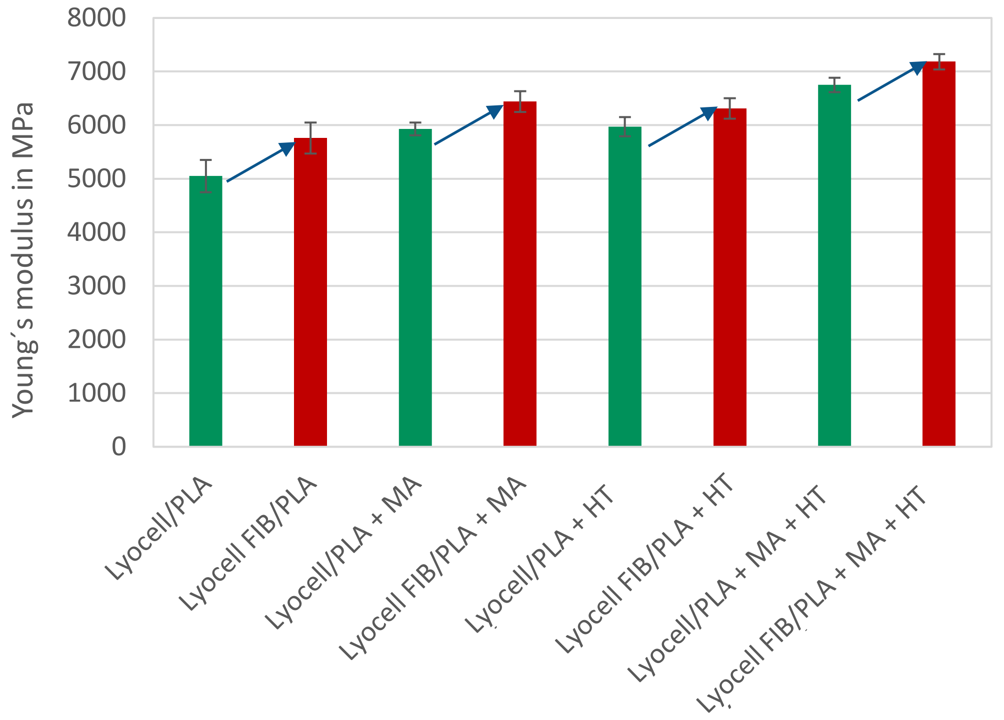
3D printing (FDM) with a MakerGear™ M2 desktop 3D printer

Perimeter (shell) of one printing line (approx. 0.75 mm), raster angle of 0° (parallel to loading direction), and nozzle of 0.75 mm

Gauss, C. / Pickering, K. L. / Graupner, N. / Müssig, J. (2023): **3d-Printed Polylactide Composites Reinforced with Short Lyocell Fibres - Enhanced Mechanical Properties Based on Bio-Inspired Fibre Fibrillation and Post-Print Annealing**. *Additive Manufacturing* , 77: 103806

Tensile characteristics of 3D printed 30 mass-% lyocell/PLA and fibrillated lyocell/PLA composites

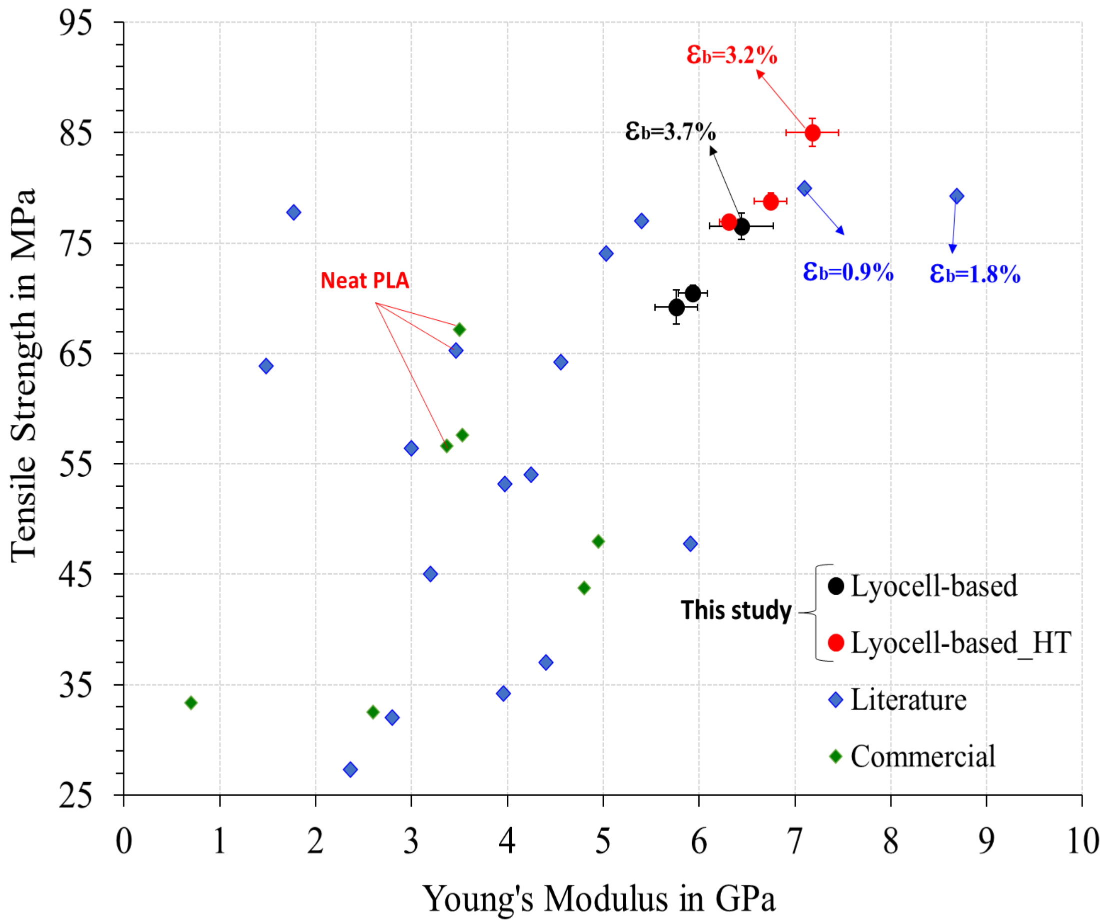




⇨ The use of fibrillated lyocell fibres leads to a significant increase in strength and tensile modulus compared to untreated lyocell fibres

Gauss, C. / Pickering, K. L. / Graupner, N. / Müssig, J. (2023): **3d-Printed Polylactide Composites Reinforced with Short Lyocell Fibres - Enhanced Mechanical Properties Based on Bio-Inspired Fibre Fibrillation and Post-Print Annealing**. *Additive Manufacturing* , 77: 103806

3D printed short fibre-reinforced PLA composites – literature comparison



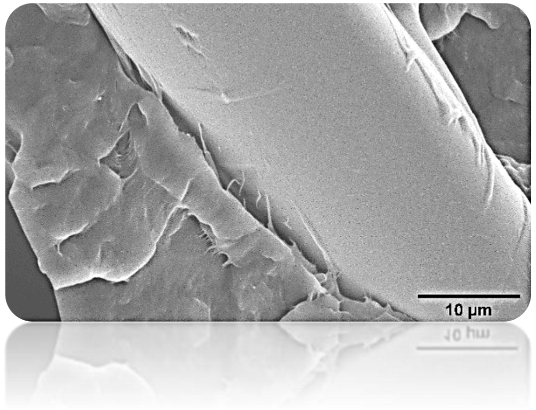
A comparison with literature data on other short fibre-reinforced PLA composites shows that the fibrillated lyocell fibres in combination with heat and MA treatment can lead to very high mechanical properties

Tensile properties of PLA/lyocell composites compared to other 3D-printed PLA-based composites (hemp, microcystalline cellulose, nanofibrillated cellulose, flax, wood, basalt, bamboo, lyocell, harakeke, curaua, carbon, heneken fibres)

Gauss, C. / Pickering, K. L. / Graupner, N. / Müssig, J. (2023): **3d-Printed Polylactide Composites Reinforced with Short Lyocell Fibres - Enhanced Mechanical Properties Based on Bio-Inspired Fibre Fibrillation and Post-Print Annealing**. *Additive Manufacturing* , 77: 103806

Conclusions

- The fibrillation of the lyocell fibres leads to a slight reduction in fibre tensile strength and elongation at break, Young's modulus was not affected
- Fibre/matrix adhesion has been significantly improved by fibrillation
- Tensile strength, Young's modulus and impact strength have been significantly increased by using fibrillated fibres in compression-moulded PLA composites
- Fibrillation also led to a significant increase in tensile strength and Young's modulus in 3D printed PLA composites
- The hypothesis that fibrillated lyocell fibres can anchor themselves in the matrix like a root system can therefore not be rejected



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Thank you very much vor your attention!

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