Mechanical Properties of Regenerated Cellulose Fibres for Composites

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Summary: A broad variety of regenerated cellulose fibres was subjected to single fibre tensile tests in order to determine the modulus of elasticity, tensile strength, and failure strain. The results were compared to glass fibres and flax fibres, which are considered the most important technical and natural fibres, respectively. With regard to their modulus of elasticity and tensile strength, regenerated cellulose fibres showed clearly lower values than glass fibres, even when their low density was taken into account. The average modulus of elasticity and tensile strength of regenerated cellulose fibres was also lower than the values measured for flax fibres, but when variability was considered, both fibres performed similarly. In terms of interfacial shear strength with polypropylene, lyocell fibres performed significantly less well than sized glass fibre and ramie fibre. The most important difference between regenerated cellulose fibres and both glass and flax fibres is their high failure strain and thus high work to fracture. The high work to fracture of regenerated cellulose fibres makes them particularly useful for composite applications where high fracture toughness is required.

Keywords: cellulose; fibres; interfacial shear strength; tensile properties

Introduction

Cellulose combines excellent mechanical properties with the advantage of being a natural, fully renewable and biodegradable material. Due to the rise in importance of the latter properties, research on the use of cellulose for the reinforcement of polymer composites is increasing in importance, as demonstrated in reviews on natural fibre reinforced composites[1,2] and cellulose nanocomposites.[3] Also regenerated cellulose fibres, which are produced by the dissolution of pulp in a suitable solvent and subsequent spinning, profit from this rise in interest as documented by recent publications.[4–8] Concerning the reinforcement of polymer composites, tensile strength, modulus of elasticity, elongation at break, and interfacial shear strength in combination with a polymer matrix are important fibre properties, as they govern the strength, stiffness, and toughness of composites. The mechanical performance of regenerated cellulose fibres is inferior to glass fibres, the most widely used reinforcement fibre, but cellulose gains in competitiveness when its low density of 1.5 g cm^{-3} compared to 2.5 g cm^{-3} for glass is taken into account. The non-abrasiveness of cellulose compared to glass fibre is a strong advantage of cellulose in processes such as extrusion and injection moulding. On the market, regenerated cellulose fibres also compete with flax fibre, which is the currently most important natural reinforcement fibre. It is well known that the properties of flax fibres, though very good on average, are highly

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variable due to inherent natural variability and partly damaging processing methods.[9] In comparison, the properties of regenerated cellulose fibres may be tuned to specific value within a wide range and narrow limits.[10–13]

Besides basic mechanical fibre properties, the interaction of fibre and matrix polymer in terms of adhesion is crucial to composite performance. While thermoset resins usually show good adhesion, thermoplastics are often not compatible with cellulose, which is highly hydrophilic due to accessible OH groups, because of their hydrophobicity. Therefore, surface modification of cellulosic fibres is performed in order to improve adhesion to thermoplastic matrices.[14–16]

In the present study, a large number of regenerated cellulose fibres was characterised by single-fibre tensile tests and compared to glass fibre and flax. In addition, the interfacial shear strength between regenerated cellulose fibres and polypropylene droplets was determined. Based on the experimental results obtained, an assessment of the suitability of different types of regenerated cellulose fibres for the reinforcement of polymer composites is given.

Materials and Methods

Regenerated cellulose fibres (viscose staple fibre, modal staple fibre, lyocell staple fibre) with linear density from 0.8 dtex to 1.3 dtex were obtained from Lenzing R&D (Lenzing, Austria). Rayon tire cord with a linear density of 1.85 dtex was provided by Cordenka (Obernburg, Germany). Flax fibres, ramie fibres, and glass fibres were purchased (Lotteraner, Vienna, Austria). The fibres were mounted to paper frames as described in a companion paper[17] and tested at a speed of 1 mm min^{-1} on a Zwick-Roell 20 kN universal testing machine equipped with a 50 N load cell. For regenerated cellulose fibres the fibre cross section was determined from the linear density and the density of cellulose, whereas the diameter of flax, ramie, and glass fibres was measured in a Zeiss light microscope equipped with image analysis software. The modulus of elasticity was determined from the recorded stress-strain curves by fitting a linear regression to the initial linear-elastic part of the curve. The total strain energy (plastic and elastic), taken-up by the fibre immediately before fracture (work to fracture $W_{\varepsilon f}$), was determined by numerical integration of the area under the stress-strain curve. For the determination of interfacial shear strength, one type of lyocell fibre as well as glass and ramie were selected. Same as for single fibre tensile testing, the fibres were mounted to paper frames. A polypropylene (PP) film with a thickness of 0.1 mm was obtained from Goodfellow (PP301400, www.goodfellow.com). Small pieces of PP film were cut to a trouser-like shape and placed onto the fibres. The fibres with the PP were then heated above a laboratory heating plate until the PP started to melt and formed a drop, which was then cooled by removing the sample from above the heating plate. Before testing, the length of contact l and the fibre diameter d were measured in a ZEISS light microscope equipped with a video camera and a digital image analysis system (Figure 1).

Subsequently, the fibre was mounted to the universal testing machine as shown in Figure 1. By means of adjustable carbide cutter blades, the PP drop was gripped and pulled from the fibre. A typical force against displacement from such a test is shown in Figure 2. The force on the fibre rises continuously with displacement until the PP drop abruptly debonds from the fibre surface indicated by a sharp force drop in Figure 2.

After debonding the drop glides along the fibre as indicated by more or less constant force against displacement and finally the fibre end slips through the drop, indicated by a force drop to zero. The peak force reached before debonding F_{peak} is used to calculate the interfacial shear

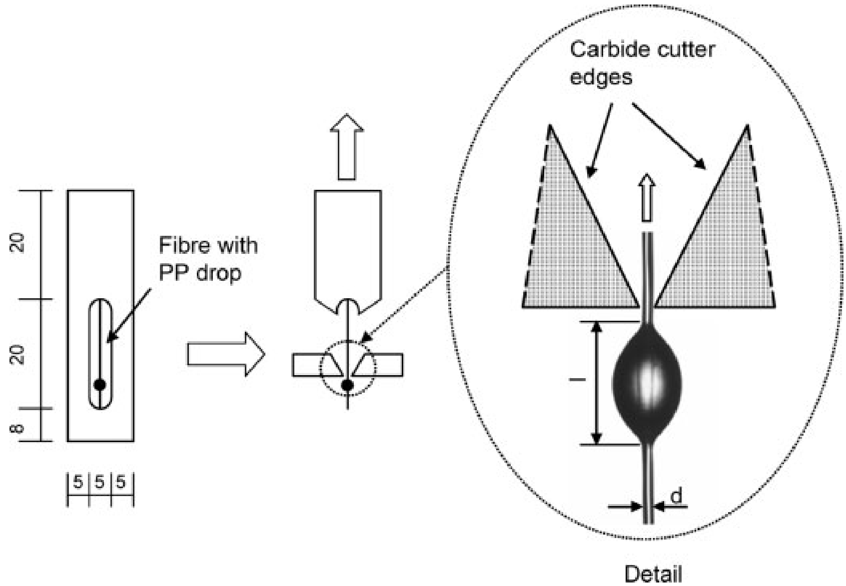

Figure 1.
Set-up for the determination of interfacial shear strength by drop pull-off testing. The fibre with polypropylene (PP) droplet is mounted to the testing machine in a paper frame, which is cut-open thereafter. Upon testing, the droplet is pulled from the fibre by means of adjustable carbide cutter edges. The embedded detail (right) shows a light microscope image of an actual PP droplet on a lyocell fibre.

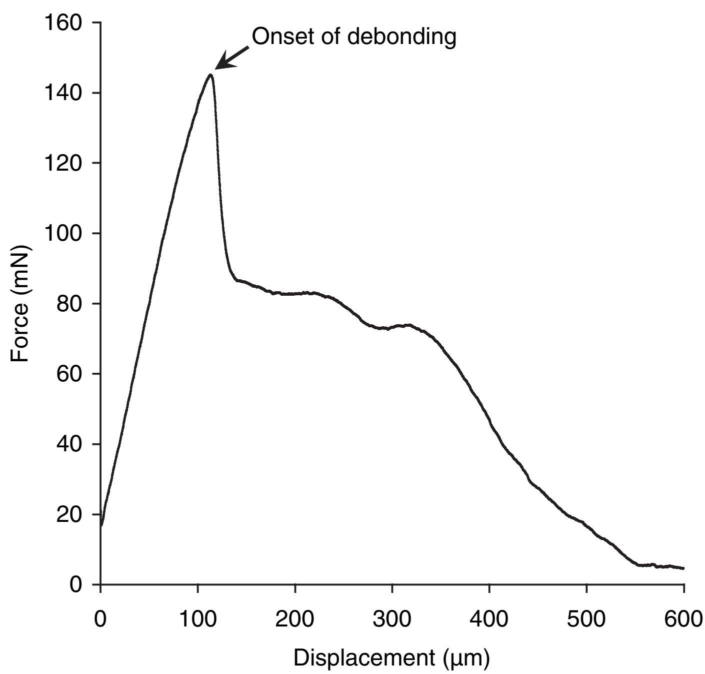

Figure 2.
Typical force-displacement curve from a drop pull-off test with a polypropylene drop on a lyocell fibre. The peak force indicating the onset of debonding was used for the calculation of interfacial shear strength.

strength τ_{IF} according to equation 1.

$$\tau_{IF} = \frac{F_{peak}}{dl\pi} \qquad (1)$$

Results and Discussion

The results of the mechanical characterisation of various regenerated cellulose fibres in comparison with flax and glass fibre are shown in Figure 3 and Table 1. Representative stress-strain curves shown in Figure 3 demonstrate highly variable mechanical behaviour for the different regenerated cellulose fibres tested, similar to results recently published by Ganster and Fink.[7] While all regenerated cellulose fibres show an initial linear elastic phase up to a yield strain of approximately 1% and a second phase of plastic behaviour, the measured modulus of elasticity, tensile strength, and elongation at break differ significantly in dependence of the fibre type. Viscose fibre produced for use in textiles showed lowest strength and stiffness while being highly extensible. Average mechanical properties improved significantly for modal fibres and lyocell. Top values for fibre strength among regenerated cellulose fibres were measured for rayon tirecord (Table 1). However, the modulus of elasticity was slightly higher for average lyocell fibres, and top values for lyocell clearly surpassed the modulus of tire cord while showing comparable tensile strength (Table 1). In comparison to flax and glass fibre, the most obvious difference of regenerated cellulose fibres is their high extensibility. Both flax and glass show essentially linear-elastic behaviour up to fracture (Figure 3, Table 1).

An assessment of the competitiveness of regenerated cellulose fibres with glass fibre in terms of single-fibre mechanical performance leads to different results depending on which property most emphasis is

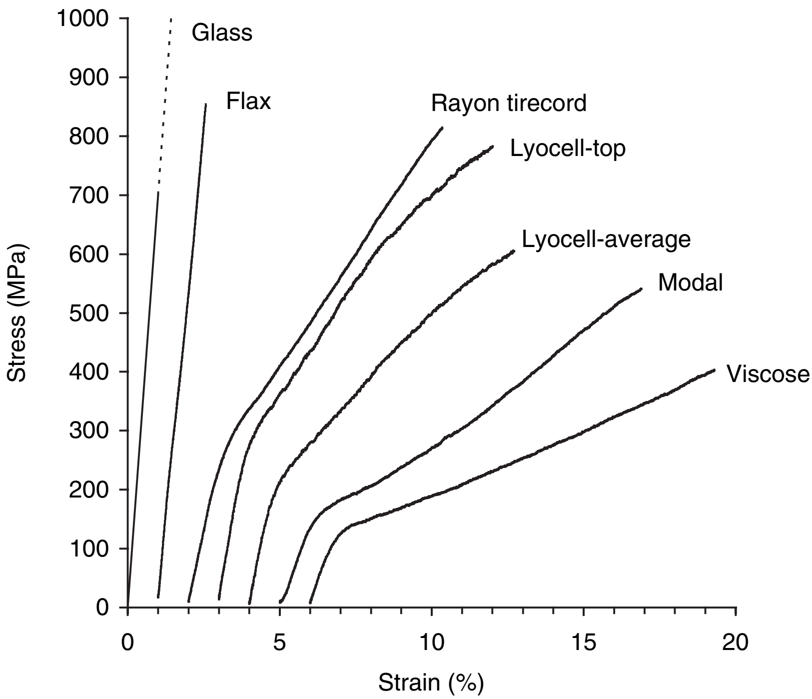

Figure 3.
Representative stress-strain curves of various regenerated cellulose fibres in comparison to flax and glass fibre (the stress-strain curve of glass fibre extending up to 3000 MPa is not fully shown and curves are off-set by 1% strain for better viewing).

Table 1.
Average tensile properties of regenerated cellulose and reference fibres (E – modulus of elasticity, σ_f – tensile strength, ε_f – elongation at break, $W_{\varepsilon f}$ – work to fracture, for lyocell top values are given in brackets).

Fibre	E (GPa)	σ_f (MPa)	ε_f (%)	$W_{\varepsilon f}$ (J^*10^{-3} mm^{-3})
Viscose	10.8 ± 2.5	340 ± 73	15.4 ± 2.2	32.7
Modal	13.2 ± 2.2	437 ± 69	10.4 ± 1.8	37.2
Lyocell	23.4 (30.5) ± 3.9	556 (790) ± 78	8.7 ± 1.6	34.5 (47.1)
Rayon tirecord	22.2 ± 1.0	778 ± 62	10.7 ± 1.4	40.8
Flax	40.0 ± 19.2	904 ± 326	1.4 ± 0.2	6.7
Glass	70.0 ± 9.3	3000 ± 356	4.3±	54.3

attributed to. It is obvious that none of the regenerated cellulose fibres can compete with the strength of glass fibre. Even on a weight basis, taking into account the lower density of cellulose (1.5 g cm^{-3}) compared to glass (2.5 g cm^{-3}), competitiveness is not given. It is remarkable however that the work to fracture taken-up by the best-quality lyocell fibres and rayon tire cord tested in this study is comparable to glass fibre in absolute terms (Table 1) and surpasses glass fibre on a weight basis.

Compared to flax, the natural fibre chosen for reference, neither rayon tire cord nor lyocell reach comparable average values of tensile strength and modulus of elasticity (Table 1). However it has to be considered that the average values obtained for flax are affected by high variability as seen in Figure 4. When a composite is designed to bear a certain load, characteristic values taking into account variability have to be used instead of average values. Due to the comparably

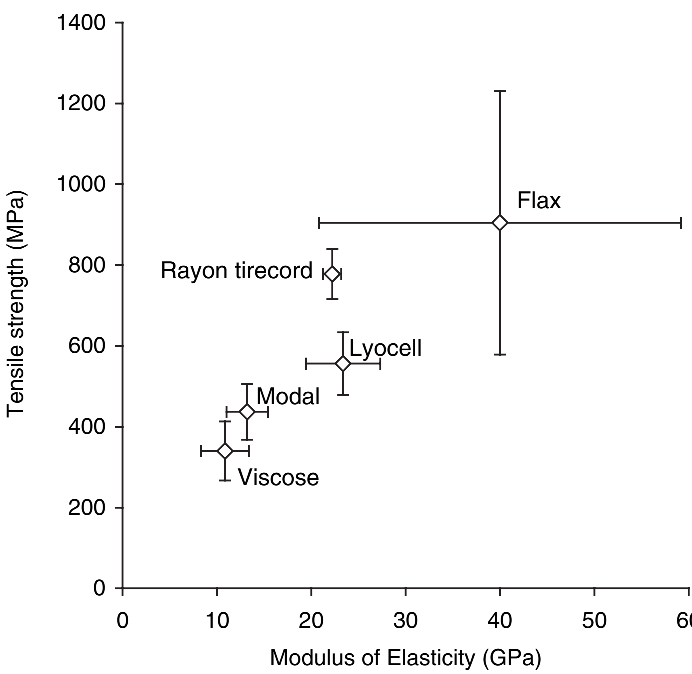

Figure 4.
Average values and standard deviation of modulus of elasticity and tensile strength for different cellulosic fibres.

low variability observed for their mechanical properties, rayon tirecord and lyocell are both well competitive with flax fibre. It has to be noted here that flax fibres may be considered composites themselves, since they consist of cellulose microfibrils embedded in a matrix of hemicellulose and pectin. Natural variability in cellulose content and the orientation of cellulose microfibrils in the flax fibre cell wall therefore accounts for a part of the observed variability in mechanical properties. As already mentioned in the introduction paragraph, current processing methods also contribute significantly to varying mechanical properties.[9] Similar as observed in comparison to glass fibre, the work to fracture shows the biggest difference between regenerated cellulose fibres and flax (Table 1). The work to fracture of cellulose fibres surpasses the value of flax by a factor of 5 (viscose) to 7 (lyocell).

In addition to the tensile properties of single fibres, the adhesion between matrix polymer and fibre is crucial to the mechanical performance of a composite. Since problems with adhesion are infrequent with thermoset resins, polypropylene, which is frequently used as matrix polymer in extrusion and injection moulding, was selected for the present study. Two main fibre properties governing adhesion are surface chemistry and surface roughness.[18] Suitable surface chemical properties lead to good wetting and development of adhesion forces, while surface roughness provides mechanical interlocking between fibre and polymer. PP drop pull-off tests revealed significant differences between the interfacial shear strength values of glass, ramie, and lyocell fibres (Figure 5).

Both cellulose fibres show lower interfacial shear strength than (sized) glass fibre, presumably due to the different chemical character of cellulose (polar, hydrophilic) and PP (non-polar, hydrophobic). Ganster et al.[8] recently showed that this disadvantage of regenerated cellulose fibres can be overcome by the addition of a suitable coupling agent to the polymer matrix. In

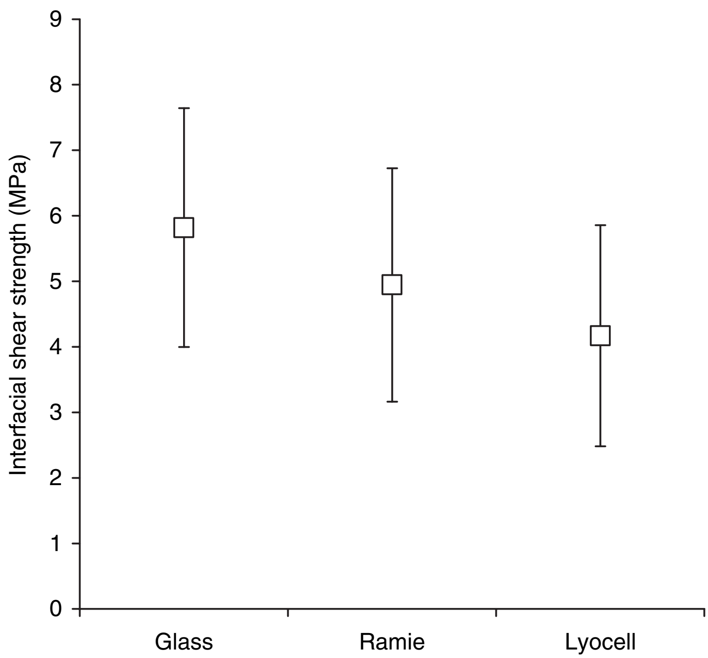

Figure 5.
Average values and standard deviation of interfacial shear strength measured for different fibres in PP drop pull-off tests.

spite of being chemically very similar to lyocell as they consist almost exclusively of cellulose, ramie fibres show higher interfacial shear strength than lyocell, presumably due to the higher surface roughness observed for natural fibres.

The importance of surface roughness was demonstrated by Karlsson et al.[19] In a study on the interfacial shear strength of lyocell fibres with different roughness due to fibrillation, they showed that adhesion between fibre and low density polyethylene is significantly improved by increasing surface roughness. Also, it was repeatedly demonstrated that surface modification of cellulosic fibres is capable of significantly improving composite performance.[1,8,20–22]

Conclusion

The experimental data presented above show that among a number of different regenerated cellulose fibres tested in tension, rayon tire cord and lyocell have highest strength while lyocell has the highest modulus of elasticity. In comparison to glass fibre, regenerated cellulose fibres performed less well regarding strength and modulus, but well in terms of work to fracture. In comparison to flax fibre, regenerated cellulose fibre performed equally well in strength and modulus when their different variability was considered. The work to fracture of regenerated cellulose fibres was by far superior to flax. Interfacial shear strength between cellulose fibres and PP was worse than for glass fibre. Considering all this, two important conclusions may be drawn:

- when using an non-polar matrix polymer, surface modification of regenerated cellulose fibres or the addition of a suitable coupling agent to the polymer matrix is necessary, and
- regenerated cellulose fibres perform very well in terms of work to fracture, which makes them perfectly suitable reinforcement fibres for composite applications where high fracture toughness is required.

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