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Upcycling of cotton polyester blended textile waste to new man-made cellulose fibers

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1. Introduction

Ever since fast fashion has attempted to satisfy the demands of a rapidly growing population (Hämmerle, 2011), the global waste pollution has increased tremendously. Europe disposes 75% (4.3 million tons) of textiles waste annually due to the absence of viable recycling strategies. (Briga-Sá et al., 2013) Interwoven cotton polyester blends represent one of the most prominent mixtures on the market, which are difficult to recycle because of their heterogeneity. Conventional fiber spinning processes can partly reuse pure cellulose waste (Shen and Patel, 2010), but they are unable to handle blended waste garments. Therefore, the recycling of cotton polyester blends has yet demanded either the depolymerization or dissolution of either component. The dissolution of polyester usually involves toxic solvents and elevated temperatures (DMSO), which can influence the properties of the cellulose component (Serad, 1993). Accordingly, polyester is preferably subjected to alcoholysis, hydrolysis, or glycolysis reactions (Barot and Sinha, 2015; Oakley et al., 1992; Palme et al., 2017; Shen et al., 2013).

Similar approaches have also been used to degrade cellulose under acidic conditions (Cowan, 1981; Gruntfest and Turner, 1974; Ouchi et al., 2010; Shen et al., 2013). An example is the

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ABSTRACT

The creation of a circular economy for cellulose based textile waste is supported by the development of an upcycling method for cotton polyester blended waste garments. We present a separation procedure for cotton and polyester using [DBNH] [OAc], a superbase based ionic liquid, which allows the selective dissolution of the cellulose component. After the removal of PET, the resulting solution could be employed to dry-jet wet spin textile grade cellulose fibers down to the microfiber range (0.75–2.95 dtex) with breaking tenacities (27–48 cN/tex) and elongations (7–9%) comparable to commercial Lyocell fibers made from high-purity dissolving pulp. The treatment time in [DBNH] [OAc] was found to reduce the tensile properties (<52%) and the molar mass distribution (<51%) of PET under certain processing conditions. © 2019 The Author(s). Published by Elsevier Ltd. This is an open access article under the CC BY-NC-ND license (http://creativecommons.org/licenses/by-nc-nd/4.0/).

hydrolysis of cotton employing hydrochloric acid to generate a microcrystalline cellulose powder, which can be separated from the remaining polyester fabric. The recovered polyester is re-spun to PET yarns that can then be used to manufacture new textiles (Sun et al., 2018a, 2018b).

Less destructive methods utilize ionic liquids (ILs) or NMMO to dissolve the cotton component. Negulescu et al. prepared a 1-2% cellulose solution from cotton polyester blends, and removed the polyester residue by filtration. Afterwards, the cellulose solution was further concentrated up to 15–17% using cotton material from an alternative feedstock (Negulescu et al., 1998). A recent patent further elaborates this approach. An amine oxide solvent such as NMMO is supposed to dissolve the cellulose component of various blended textile products to yield cellulose solutions of up to 35% (Brinks et al., 2013). Similarly, 1-allyl-3-methylimidazolium chloride ([AMIM] [Cl]) and 1-butyl-3-methylimidazolium acetate ([BMIM] [OAc]) have been described to foster the recycling of blended textile waste. The process steps are comparable to NMMO. The ionic liquid dissolves the cellulose component, while filtration removes the polyester residue, and subsequent coagulation recovers the cellulose (de Silva et al., 2014). Although both NMMO and IL based approaches highlight the potential of the generated cellulose solutions to be used in a film or fiber spinning process, neither of them provides any relevant data that could serve as a proof of concept.





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List of acronyms

[AMIM]	Cl] 1-allyl-3-methylimidazolium chloride
[BMIM] [OAc] 1-butyl-3-methylimidazolium acetate
[DBNH]	OAc] 1,5-diazabicyclo[4.3.0]non-5-enium acetate
[M1]	modified separation experiment 1 using [DBNH] [OAc]
	with excess HOAc
[M2]	modified separation experiment 2 using 2 separation
	experiments to increase the cellulose concentration
[R]	raw-material/reference
[S1]	separation experiment 1 with 1 washing step
İS2İ	separation experiment 2 with 2 washing steps
Â	acid washing
COP	crossover point
CP-MAS	cross-polarized magnetic angle spinning
DBN	1,5-diazobicyclo(4.3.0)non-5-ene
DMAc	N,N-dimethyl acetamide
DMSO	dimethyl sulfoxide
DP	degree of polymerization
DR	draw ratio
Ew	alkaline washing
f _{total}	total orientation
G'	storage modulus, Pa
G"	loss modulus, Pa
GPC	gel permeation chromatography
HFIP	1,1,1,3,3,3-hexafluoro-2-propanol
HOAc	acetic acid

IL LiCl MMCFs MMD M _n	ionic liquid lithium chloride man-made cellulose fibers molar mass distribution number average molar mass, Da
Mw *	mass average molar mass, Da
ղ ₀ Nո	zelo sileal viscosily, Pa s
NMMO	N-methylmorpholine N-oxide
NMR	nuclear magnetic resonance
ŋ*	complex viscosity, Pa s
PDI	polydispersity index
PET	polyethylene terephthalate
RI	refractive index
SEM	scanning electron microscopy
TGA'	thermogravimetric analysis
V _{extr.}	extrusion velocity of the dope, $cm^3 min^{-1}$
V _{take-up}	take-up velocity of the godet rollers to collect the fibers,
	$m min^{-1}$
Z	ozone pretreatment
Δn	birefringence value
ω	angular frequency, s^{-1}

It is evident that there is a strong need to further develop recycling strategies for textile industry. A viable, ecofriendly concept for circular economy should allow quantitative and "green" recycling of all material components without the generation of additional waste streams, or an increase in energy consumption resulting from complex processing steps. In many cases, the approaches discussed so far involve the degradation of either cotton or polyester, and therefore impede the simultaneous upcycling of both components within one process. Even more significantly, however, they fail to show the conversion of textile waste to new-value added products, i.e. to employ textile waste as a feedstock to spin new man-made cellulose fibers (MMCFs).

In previous studies, we demonstrated the usage of [DBNH] [OAc] to convert cellulose based waste material such as cotton, cardboard, and paper to new high-tenacity Ioncell fibers (Asaadi et al., 2016; Ma et al., 2018; Ma et al., 2016). [DBNH] [OAc] does not require the addition of any stabilizers and allows to adjust the process conditions at temperatures 30 °C lower than for spinning solutions prepared from NMMO. In a current patent, we have also been able to show that this solvent enables the selective dissolution of cellulose in cotton polyester blends without any severe degradation of polyester, if adequate process conditions are applied (Haslinger et al., 2017). Utilizing this concept, the research disclosed herein aims to foster the creation of a circular economy for textile industry by transforming blended cellulosic waste material to new textile grade MMCFs. After filtration, the resulting cellulose solution could thus be used directly as a spinning dope for dry-jet wet spinning (Michud et al., 2015b; Parviainen et al., 2015; Sixta et al., 2015; Stepan et al., 2016) to obtain Lyocelltype fibers ranging from 0.7 to 3.0 dtex. The degradation of the recovered PET was studied dependent on treatment time and temperature. A detailed analysis on the further recyclability of PET such as in melt spinning was however beyond the scope of this study.

2. Materials and methods

2.1. Synthesis of [DBNH] [OAc]

The synthesis of [DBNH] [OAc] is a neutralization reaction of an equimolar amount of 1,5-diazabicyclo(4.3.0)non-5-ene (DBN) and glacial acetic acid. During the addition of acetic acid, permanent cooling is necessary because of the exothermic nature of the reaction. The resulting ionic liquid shows a melting point close to 65 °C and therefore requires stirring at elevated temperatures (80 °C, 1 h) in order to ensure a complete conversion.

2.2. Cotton polyester blend

The cotton polyester blend was supplied by SOEX (Germany) consisting of white postconsumer textiles, which were shredded and blended to obtain a mixture with an overall concentration of 50 wt% cotton and 50 wt% polyester. The sample was pretreated by VTT (Finland) using an ozone based sequence Ew-disk refining-Z-P-A. The target of the alkaline washing stage (Ew) was to remove silicate, whereas ozone (Z) and hydrogen peroxide (P) were used to both adjust the viscosity (DP) and to bleach the material. Acid washing (A) aimed to decrease the metal content. Disk refining targeted to homogenize the sample mixture and to improve the reactivity for the following pretreatment and dissolution stages. The molar mass distributions (MMD) of both cellulose and the polyester fractions were determined by size exclusion chromatography, respectively, as described below. All samples were further disintegrated by the means of a Wiley Mill.

2.3. Molecular mass distribution (MMD) and intrinsic viscosity of cellulose

The MMD of the raw material and the spun fibers was determined by gel permeation chromatography (GPC) using a 5 column set up with one precolumn (PLgel Mixed-A, 7.5 × 50 mm) and four analytical columns (4 × PLgel Mixed-A, 7.5 × 300 mm) equipped with an RI-detector (Shodex RI-101). The samples were dissolved in a 90 g l⁻¹ LiCl/N,N-dimethylacetamide (DMAc) solution and diluted with pure DMAc in order to obtain a sample concentration of 1 mg ml⁻¹ in 9 g l⁻¹ LiCl/DMAc. 100 µl of the respective solution were injected at a flow rate of 0.750 ml min⁻¹ (9 g l⁻¹ LiCl/DMAc eluent) at 25 °C. The calibration was conducted with pullulan standards (Mw: 343–708,000 Da) based on a direct-standardcalibration model, in which the molar mass distribution was corrected according to the molar masses of cellulose equivalent pullulan standards (Berggren et al., 2003; Borrega et al., 2013; Michud et al., 2015a).

The limiting viscosity of the raw material before and after its pretreatment was determined according to the SCAN-CM 15:88 standard for viscosity in cupri-ethylenediamine solution.

2.4. Selective dissolution of cotton and recovered PET

The cotton polyester blend was mixed with [DBNH] [OAc] (1 h, 80 °C) using a vertical kneader system (cf. experiments [S1], [S2], and [M2] in Table 1) to yield a cellulose solution of 6.5 w%. For [M1], [DBNH] [OAc] was prepared with an excess amount of HOAc in a ratio of 1:1.3 to reduce the degradation of PET. Subsequently, the undissolved polyester fraction was separated from the cellulose solution via hydraulic pressure filtration (70 °C, 1–8 MPa, metal filter mesh with 5-6 µm absolute fineness, Gebr. Kufferath AG, Germany). In experiment [M2], the filtered solution was transferred back to the kneader system and mixed with an additional amount of the cotton polyester blend (1 h, 80 °C) to obtain a final cellulose concentration of 10.5 wt% after filtration. Whereas the resulting cellulose solutions could be subjected to dry-jet wet spinning directly after solidification (storage at 8 °C for a couple of days), the recovered polyester fibers required further purification as they were still contaminated with residual cellulose. Therefore, the polyester residue was extracted with fresh ionic liquid (1 h, 80 °C, vertical kneader). Afterwards, the solvent was removed by filtration and subsequent washing with deionized water. This step was repeated for experiments [S2], [M1], and [M2].

2.5. Dry-jet wet spinning

The solidified cellulose solution was placed into a customized laboratory dry-jet wet piston spinning unit (Fourné Polymertechnik, Germany) and heated up according to its rheological properties. The melted spin dope was extruded through a multi hole spinneret (36 holes, 0.1 mm diameter, 0.02 mm capillary length) with a constant extrusion velocity of 1.6 cm³ min⁻¹. The resulting filaments were stretched in an 1 cm air gap by varying take up velocities to achieve draw ratios (DR) between 1–8.8 (DR = v_{take-up}/v_{extr}) and coagulated in a water bath (10 °C).

2.6. Rheology of the spinning solution

The shear rheology of all spinning dopes was analyzed with an Anton Paar MCR 300 rheometer with a plate and plate geometry (1 mm gap size, 25 mm plate diameter). Initial dynamic strain sweep tests were performed to define the linear-viscoelastic domain. A strain of 0.5% was chosen for the following frequency tests. All samples were subjected to a dynamic frequency sweep over an angular frequency (ω) range of 0.01–100 s⁻¹ at relevant temperatures (40–100 °C). The complex viscosity (η^*) and dynamic moduli (G', G'') were recorded as a function of the shear rate. The zero shear viscosity (η_0^*) was calculated using the cross model assuming that the Cox-Merz rule was valid (Sammons et al., 2008). The cross over points of storage and loss moduli were also determined.

2.7. Fiber properties and total orientation

Linear density (dtex), elongation at break (%), and tenacity (cN dtex⁻¹) of all spun fibers and of 3.3 dtex PET filaments were measured by a Vibroskop and Vibrodyn 400 set up (Lenzing Instrument, Austria) according to DIN 53816 standard (20 mm gauge length, 5.9 ± 1.2 mN/tex pretension, 20 mm/min test speed). The samples were conditioned prior to the measurement (20 °C, 65% humidity) and were tested in both dry and wet state (fiber count: 10). The young modulus (GPa) was calculated from the slope of the average stress strain curve based on the ASTM standard D2256/D2256.

The total orientation of the spun fibers was determined by a polarized light microscope (Zeiss Axio Scope) with a 5 λ Berek compensator. The birefringence (Δ n) was calculated from the retardation of the polarized light divided by the fiber thickness assuming a density of 1.5 g cm⁻³ for cellulose. In order to obtain the total orientation factor f_t, Δ n was divided by the maximum birefringence value (0.062) of cellulose (Adusumalli et al., 2009).

2.8. Scanning electron microscopy (SEM)

The cross sections of the spun fibers were observed on a Zeiss Sigma VP, Zeiss FE-SEM. An acceleration voltage of 1.5 kV was used and all samples were sputter coated with Au prior to the measurement.

2.9. Degradation experiments of PET filaments

Virgin 3.3 dtex PET filaments (Swerea IVF, Sweden) were treated with [DBNH] [OAc] (6 wt% PET, 0.5–4 h, 80 °C) under permanent stirring. Afterwards, the filaments were rinsed with deionized water and their tensile strength was determined as described in the cellulose section above.

2.10. Gel permeation chromatography of PET

The molar mass distribution (MMD) of polyester was determined by gel permeation chromatography (VTT, Finland) using a 3 column setup with one precolumn (Styragel WATT054405) and two analytical columns (Styragel HR 4E 7.8 \times 300 mm and Styragel HR5 7.8 \times 300 mm) equipped with a RI-detector (40 °C). The analysis was conducted with an 1,1,1,3,3,3-hexafluoro-2-propanol (HFIP)/5mM salt (sodium trifluoroacetate) eluent at a flow rate of

Table 1

Experimental design. Summary of the conducted experiments describing the total concentration of cotton and polyester (CO/PET) before separation, the resulting cellulose concentration (Cellulose) in the spinning dope, the solvent used, and the washing steps employed for the polyester residue.

Exp.	CO/PET (wt%)	Cellulose (wt%)	Solvent	Washing of PET
[S1]	13	6.5	[DBNH] [OAc]	1x [DBNH] [OAc]
[S2]	13	6.5	[DBNH] [OAc]	2x [DBNH] [OAc]
[M1]	13	6.5	[DBNH] [OAc] + HOAc (molar ratio: 1:1.3)	2x [DBNH] [OAc] +HOAc (molar ratio: 1:1.3)
[M2]	21	10.5	[DBNH] [OAc]	2x [DBNH] [OAc]

0.5 ml/min. The system was calibrated with polymethyl methacrylate standards.

2.11. ¹³C CP-MAS NMR

The concentration of the residual cellulose in the recovered polyester fraction was estimated by 13 C CP-MAS NMR. All samples were analyzed on a Bruker AVANCE III spectrometer with a probe size of 4 mm employing an UltraShield Plus 400 MHz magnet running at 100.6 MHz. The spectra were recorded at a spinning speed of 8 kHz using a relaxation delay of 5 s and a CP contact time of 2 ms. 8000 scans with an acquisition time of 34 ms were measured. During contact time, a variable crosspolarization increasing from 50% to a maximum amplitude of 85 kHz was used. A SPINAL-64 ¹H decoupling aimed to decouple the protons during acquisition.

A sigmoidal calibration model was employed to quantify the cellulose content in the polyester residue. The ratios of the peak areas of C1 (cellulose) and C1-8 as well as C2-3-4-5-6 (polyester) were plotted as a function of the cellulose concentration (Haslinger et al., 2019).

2.12. Thermogravimetric analysis (TGA)

The TGAs were performed on a TA Instruments Q500 in a temperature range of 25–800 °C with a heating rate of 10 °C/min under a N_2 atmosphere. 5–10 g of sample were used per measurement.

3. Results and discussion

3.1. Cotton polyester based textile waste as a raw-material

In previous studies, we were able to demonstrate the conversion of pure cellulose waste to new MMCFs, if the material's properties classified it as spinnable (Ma et al., 2018; Ma et al., 2016). Any recycling concept relying on the dry-iet wet spinning of cellulose is challenged by heterogeneous substrates such as textile waste. Besides the removal of residual contaminants, i.e. synthetic fibers and metals, the cellulose fractions need to undergo certain pretreatments to achieve both an adequate average degree of polymerization ([η] \cong 450 ml g⁻¹) and a broad molar mass distribution (MMD) (Michud et al., 2015a). The properties of pre-consumer cotton waste are usually far beyond these criteria due to its high intrinsic viscosity and considerably narrow MMD, which makes pretreatments essential. In postconsumer textiles, laundering induces a certain degree of degradation, leading to an increased polydispersity index (PDI) and a lower intrinsic viscosity, thus allowing milder pretreatments (Palme et al., 2014).

Consequently, the origin of the waste substrate governs the choice of the pretreatment. Whereas enzymes such as endoglucanase hydrolyze cellulose dependent on the amount of amorphous regions in the polymer, acids, e-beam radiation, and hydrogen peroxide cleave cellulose chains with a minor effect on the MMD (Pérez et al., 2002). Moreover, ozone and hydrogen peroxide also enable a bleaching of the waste substrate.

In this work, white postconsumer cotton polyester textiles were treated with an alkaline-disk refining-ozone-peroxide-acid sequence to decrease the intrinsic viscosity [ŋ] of the cellulose fraction from 850 ± 50 ml g⁻¹ to 450 ± 50 ml g⁻¹. Although the latter represent optimum conditions for dry-jet wet spinning, the effect of the pretreatment on PET should not be ignored.

Table 2 displays a summary of Mn, Mw and the PDI of PET throughout the pretreatment sequences.

The PET fraction in the raw-material showed a Mw of 21.5 kDa and a Mn of 9.8 kDa, which remained almost unchanged after the

Table 2

GPC of PET. Number average molar mass M_n , mass average molar mass M_w , and polydispersity index (PDI = $M_w/M_n)$ of polyethylene terephthalate before the pretreatment, and after every pretreatment step.

	Mn (kDa)	Mw (kDa)	PDI
PET before treatment	9.8 ± 0.3	21.4 ± 0.2	2.1 ± 0.2
PET after alkali (E)	9.8 ± 0.3	22.4 ± 0.2	2.3 ± 0.2
PET after disk refining	10.1 ± 0.3	22.5 ± 0.2	2.2 ± 0.2
PET after ozone (Z)	9.4 ± 0.3	21.8 ± 0.2	2.3 ± 0.2
PET after peroxide (P)	9.3 ± 0.3	21.6 ± 0.2	2.3 ± 0.2
PET after acid (A)	9.5 ± 0.3	21.6 ± 0.2	2.3 ± 0.2

last pretreatment step with 21.6 kDa and 9.5 kDa, respectively. The PDI changed from 2.1 to 2.3 implying a slight broadening of the molar mass distribution. These alterations reflect the overall heterogeneity of the waste material, rather than a significant trend, indicating a negligible impact of the pretreatment of cellulose on the properties of PET.

3.2. Dissolution of cellulose and fiber spinning

Under standardized conditions, 13–15 wt% of cellulose is dissolved in [DBNH] [OAc] at 80 °C resulting in a highly viscous spinning dope. Even when prepared from pure cellulose streams, the solutions need to be filtered prior to their spinning due to certain impurities and undissolved residues. The filtration is demanding and usually requires special equipment (i.e. hydraulic press filtration units) to facilitate the purification of a solution in a highly viscous state (Asaadi et al., 2016; Ma et al., 2018, 2016; Michud et al., 2015b). These challenges increase in case of multicomponent materials, such as cotton polyester blends, and therefore demand adjusted processing conditions.

These obstacles have yet tried to be overcome by the choice of either ionic liquids with low viscosities, or considerably low concentrations of cellulose (Negulescu et al., 1998; de Silva et al., 2014). To compromise between facilitated processing and spinnability, we evaluated dope concentration of 6.5 wt% and 10.5 wt% cellulose implying that the same amount of PET remained undissolved in the ionic liquid solution. The concentration of a spinning solution correlates directly with the properties of the spun fibers. Higher concentrations usually lead to fibers with higher tensile strength, but also require higher draw ratios (DRs) to achieve filaments with titers low enough to be used in textiles (t < 3.0 dtex). This implies that a spin dope concentration of 6.5 wt% can already yield microfibers (t < 1.0) at a DR 8, while the same DR for a concentration of 10.5 wt% is just enough to reach a titer of about 2.0 dtex.

As stated before, the employed dissolution parameters mainly targeted to improve the processability of a blended waste substrate on a lab-scale, and to reduce the continuous degradation of PET throughout the process. We used shorter dissolution times for cellulose in [DBNH] [OAc] (1 h, 80 °C), and tried to add an excess amount of acetic acid (30%) to the ionic liquid (cf. [M1]) to prevent a possible degradation of PET (cf. PET section).

Both the concentration of cellulose and the presence of additives in the spinning solution influence the rheological properties of a spin dope. A summary of the zero shear viscosities (η_0^*) and the dynamic moduli (G', G'') at their crossover point (COP) close to the spinning temperature (60 °C) can be found in Table 3. The complex viscosities (η^*) and the dynamic moduli (G', G'') as a function of the shear rate of all tested solutions are depicted in Fig. 1.

At lower concentrations, cellulose solutions in ionic liquids typically show a Newtonian plateau at lower an angular frequency range (Gericke et al., 2009; Yao et al., 2014). As both, shear stress and concentration increase, shear thinning occurs because

Table 3

Rheology data. The zero shear viscosities η_0^* were calculated using the cross model, and the crossover point (COP) of the dynamic moduli (G',G'') was determined as function of the shear rate ω . All values were calculated for 60 °C representing the measured temperature closest to the spinning temperature.

	CO (wt%)	${\mathfrak{\eta}_0}^*$ (Pa s)	ω (s ⁻¹)	G' = G'' (Pa)
[S1]	6.5	1439 ± 12	6.9	1495
[S2]	6.5	686 ± 6	6.9	939
[M1]	6.5	175660 ± 3621	n.a.	n.a.
[M2]	10.5	13294 ± 203	1.4	3224



Fig. 1. Rheology data. Complex viscosity (η^*) and dynamic moduli (G', G'') of the solutions [S1] and [S2] with a concentration of 6.5 wt%, and [M2] with 10.5 wt% in [DBNH] [OAc], as well as [M1] with a cellulose content of 6.5 wt% in a DBN:HOAc mixture with a molar ratio of 1:1.3 as a function of the angular frequency at 60 °C.

entanglements between the polymer chains tend to be disrupted (Chen et al., 2009). Most samples studied herein correspond to this trend. The critical shear rate for shear thinning decreases with a broad MMD and the concentration being raised from 6.5 wt% to 10.5 wt%, while the measured complex viscosities potentiate. Assuming that the Cox-Merz rule is valid for solutions of cellulose in [DBNH] [OAc], the zero shear viscosities (η_0^*) can be determined based on the Carreau and the Cross model (Sammons et al., 2008). The high zero shear viscosity calculated for [M1] implies that the excess amount of acetic acid reduced the ILs power to dissolve cellulose. Throughout the angular frequency range, [M1] also displays higher values for G' than for G" indicating the formation of a gel rather than a solution. The remaining samples show a viscous domain, whose range is dependent on the cellulose content (Chen et al., 2009). The COPs of [S1] and [S2] (6.5 wt%) can be found at considerably high shear rates, whereas the COP of [M2] moves to lower angular frequencies with an increasing concentration (10.5 wt%).

In this context, it is important to note that the spinning conditions are determined by the rheological properties of the solution. In principle, any fluid is spinnable as long as the formed filaments can withstand the applied deformation and surface forces (Ziabicki and Takserman-Krozer, 1964a, 1964b). During spinning, a certain DR (v_{tu}/v_e) is exerted on the extruded filaments in order to align the cellulose chains coaxially to obtain a higher fiber tenacity and a lower fiber diameter (Hauru et al., 2014; Sixta et al., 2015). A cellulose solution in [DBNH] [OAc] should thus display a n_0^* of 20,000– 30,000 Pas and a COP of 2000-5000 Pa to yield optimum spinning conditions (Asaadi et al., 2016; Ma et al., 2018, 2016; Michud et al., 2015a, 2015b; Sixta et al., 2015). Besides varying concentration and MMD, these parameters can be reached through adjusting the spinning temperature. However, the melting point of [DBNH] [OAc] restricts the spinning window of cellulose solutions of lower concentrations and is evident in the values summarized in Table 3.

The spinnability under standardized conditions has been described as follows, DR < 2 non spinnable, 2–8 poor, 8–14 good, 14 < excellent (Asaadi et al., 2016). We redefined these criteria for cellulose solutions of lower concentrations to DR < 2 non spinnable, 2–4 poor, 4–8 good, 8 < excellent, as they require lower DRs to reach fibers with textile grade properties. All samples prepared from pure [DBNH] [OAc], i.e. [S1], [S2], and [M2], showed a good spinnability with maximum DRs of 8, 6, and 8.8, respectively. In case of [S1], the spinning at a DR of 8 led to fibers with a titer of 0.75 dtex, which classifies them as microfibers. The spinning of [M1] was accompanied by the formation of aggregates on the spinneret surface, which led to a maximum DR of 4. As mentioned before, the excess amount of acetic acid reduces the solvent power of the IL and therefore the solubility of cellulose therein, which is consequently reflected in the poor spinnability of [M1].

Besides additives, both the spin dope concentration and the draw ratio influence the tensile properties of dry-jet wet spun fibers. Due to shear forces, the cellulose molecules experience a pre-orientation in the spinneret during extrusion and are further aligned when stretched in the air-gap. As a result, the cohesive forces within the cellulose network increase, which eventually foster the formation of a dense fibrillar cellulose II structure (Fink et al., 2001). A raise in the cellulose content can improve this effect. The properties of the fibers presented in Fig. 2 confirm this principle.

Sample [M2] exhibits the highest tenacity followed by [S1], [S2] and [M1], which also corresponds to the draw ratios and concentrations employed. These properties are comparable in conditioned and wet state, with their elastic moduli (15–20 GPa) exceeding those of commercial Viscose and Tencel fibers as illustrated in the slope of the respective stress strain curves. The higher fiber strength is also explained by a higher total orientation f_{total} of the fibers (s. Table 4) and an increase in the fibrillar density



Fig. 2. Fiber properties and stress strain curves. Tenacity (cN tex⁻¹), elongation at break (%), diameter (dtex), and stress strain curves of the fibers spun from the solutions [S1] and [S2] with a concentration of 6.5 wt% and [M2] with 10.5 wt% in [DBNH] [OAC], as well as [M1] with a cellulose content of 6.5 wt% in a DBN:HOAc mixture with a molar ratio of 1:1.3. All values were taken at their maximum DRs and commercial Viscose and Tencel fibers were added for reference.

(cf. cross sectional images in Fig. S3 in the supporting information) based on the concentration and draw ratio applied.

Compared to commercial Tencel and Viscose fibers, the spun fibers show a lower elongation, because of a missing air gap conditioning on our lab-scale equipment, which could apply moisture as a plasticizer during the spinning process. Despite the difference in the fiber properties obtained, the processing conditions did not have any significant impact on MMD of the cellulose fraction as displayed in Table 4 and Fig. 3a. Evidently, the M_n and M_w of the starting material even seem to show lower values than the final products, which is a good representation of the heterogeneity of the waste substrate studied.

3.3. The effect of [DBNH] [OAc] on PET

In preliminary trials, we could observe a visible degradation of the recovered PET after its processing in [DBNH] [OAc] at 80 °C because of a change in color. PET is usually hydrolyzed under the presence of water and temperatures higher than 100 °C (Buxbaum, 1968). However, several imidazolium based ionic liquids, among them also [BMIM] [OAc], have been described to degrade and partly dissolve PET at elevated temperatures (Wang et al., 2009). These ionic liquids are supposed to catalyze the hydrolysis of PET, whose effectiveness is strongly influenced by the choice of their anion (Liu et al., 2009). Although reported in a number of publications (Brinks et al., 2013; Negulescu et al., 1998; de Silva et al., 2014), a separation of cotton and polyester without any degrading effect on the latter seems questionable.

In initial degradation studies, the decrease in tensile strength of virgin PET filaments (6 wt%) was monitored in [DBNH] [OAc] at $80 \degree C$ (s. Fig. 3b).

Although no substantial weight loss of the PET filaments could be observed, their tenacity was more than cut into half over a treatment time of 4 h. The decrease from 38 to 18 cN tex⁻¹ is exponential, and exhibits the largest drop already after 0.5 h, while the degrading effect of the ionic liquid flattens out with an increased treatment time.

The recyclability of PET fibers strongly depends on its degree of polymerization and purity. Only a high-grade PET substrate is suitable to be re-spun in a melt spinning process, fractions with a

Table 4

Birefringence and molar mass distribution of the spun fibers. The birefringence values Δn and the total orientation f_{total} dependent on the cellulose concentration (CO) and draw ratio (DR) applied, the number average molar mass Mn, the mass average molar mass Mw, and the polydispersity index (PDI = Mw/Mn) of the cellulose fraction before separation [R], and after the spinning trials [S1], [S2], [M1], and [M2].

	CO (wt%)	DR	Δn	f _{total}	Mn (kDa)	Mw (kDa)	PDI
[R]					68.3 ± 0.7	193.2 ± 5.1	2.8 ± 0.04
[S1]	6.5	8	0.0437 ± 0.0050	0.705 ± 0.080	91.1 ± 3.4	219.3 ± 3.2	2.4 ± 0.10
[S2]	6.5	6	0.0375 ± 0.0097	0.605 ± 0.156	71.1 ± 0.1	179.0 ± 3.6	2.5 ± 0.05
[M1]	6.5	4	0.0363 ± 0.0116	0.586 ± 0.187	90.0 ± 0.4	199.2 ± 0.9	2.2 ± 0.02
[M2]	10.5	8.8	0.0417 ± 0.0076	0.673 ± 0.122	84.4 ± 0.7	215.0 ± 0.3	2.5 ± 0.02



Fig. 3. (a) MMD of the cellulose fraction before separation [R], and after the spinning trials with 6.5 wt% ([S1] and [S2]) and 10.5 wt% ([M2]) in [DBNH] [OAc] solution, as well 6.5 wt% ([M1]) in a DBN:HOAc mixture with a molar ratio of 1:1.3. (b) Decrease of tensile strength of virgin 3.3 dtex PET fibers (6 wt%) in [DBNH] [OAc] at 80 °C as function of the treatment time, (c) TGA of the raw-material [R] and the recovered PET fractions after the separation experiments [S1], [S2], [M1], and [M2], (d) Molar mass distribution of PET of the raw-material [R], and after the separation experiments [S1], S2], [M1], and [M2]. For figures b-d, [R] denotes the PET fraction in the raw-material, while [S1] and [S2] respectively describe the PET obtained from 1 separation step and 1 or 2 washing steps with [DBNH] [OAc]. [M1] represents the PET residue from a modified extraction procedure in DBN:HOAc with a molar ratio 1:1.3 (+2 washing steps in the same mixture), whereas [M2] denotes PET resulting from a two-step extraction procedure in [DBNH] [OAc] (+2 washing steps in the same mixture).

lower DP require alternative processing techniques (Venkatachalam et al., 2012). These circumstances inevitably denote the most substantial challenge in the recycling of PET. Any degradation occurring during a separation and recovery process is categorically unwanted. Nevertheless, a fact that yet has often been ignored is that virgin PET, which is converted into textile fibers, already loses its ability to be re-spun during its first melt spinning cycle. Various degradation reactions cause a decrease of the DP of PET before the spun fibers have even been worn once (Samperi et al., 2004). This makes it strictly speaking impossible to recycle PET based textile waste to new fibers without applying any chain extension (Awaja and Pavel, 2005) or depolymerization and repolymerization concepts (Sun et al., 2018a, 2018b). Alternative recycling strategies therefore suggest to separate the PET mechanically from the cellulose component in order to keep a further degradation as a low as possible (Sun et al., 2018b).

However, we still tried to avoid any unnecessary degradation of PET by keeping its treatment time in [DBNH] [OAc] as short as possible during the separation experiments. According to previous results, 1 h appeared effective enough to ensure a complete dissolution of the cotton fraction, and to prevent an extensive degradation of PET (Michud et al., 2015a). To remove any residual cellulose, the PET fraction was filtered off from the cellulose solution and washed with IL and water as described in the experimental section. Table 5 depicts the cellulose content in the recovered PET after the separation experiments. The respective NMR spectra can be found in Fig. S4 in the supporting information.

One washing step in [DBNH] [OAc] (1 h, 80°) was sufficient to decrease the concentration of residual cellulose to 2.5 ± 0.5 wt% (cf. [S1]). The addition of one more washing step further lowered the amount of cellulose to 1.7 ± 0.4 wt% (cf. [S2]). The purity of PET is also confirmed by the TGA shown in Fig. 3c, and does not reveal a major change in the onset temperature T_o of the recovered fractions [S1] = 397 °C, [S2] = 398 °C, [M1] = 393 °C, and [M2] = 397 °C.

As expected, the treatment time in [DBNH] [OAc] affected the molar mass distribution of PET as shown in Fig. 3d and in Table 5. The MMD of the treated substrate noticeably decreased throughout the different treatment steps and time until it almost reached a

Table 5

Residual cellulose in PET and MMD of PET. Amount of cellulose (wt%) in the cotton polyester blend before separation [R], and after the separation experiments [S1], [S2], [M1], and [M2] dependent on their total time of exposure to [DBNH] [OAc] or DBN:HOAc (1:1.3) according to ¹³C CP-MAS NMR, and molar mass distribution of the separated PET fraction including number average molar mass M_n , mass average molar mass M_w , and polydispersity index (PDI = M_w/M_n).

	Total time in IL (h)	Cellulose (wt%)	M _n (kDa)	M _w (kDa)	PDI
[R]	0	49.8 ± 6.7	9.2 ± 0.29	23.5 ± 0.17	2.6 ± 0.2
[S1]	2	2.5 ± 0.5	6.2 ± 0.20	17.4 ± 0.13	2.8 ± 0.2
[S2]	3	1.7 ± 0.4	4.4 ± 0.14	12.0 ± 0.09	2.8 ± 0.2
[M1]	3	2.0 ± 0.4	7.1 ± 0.23	19.5 ± 0.14	2.7 ± 0.2
[M2]	3	1.9 ± 0.5	5.9 ± 0.19	16.5 ± 0.12	2.8 ± 0.2

bisection of M_w and M_n after a total of 3 h in [DBNH] [OAc] (cf. [S1] and [S2]). [M2] exhibits a bimodal MMD, as it consisted of two separate PET fractions, which were added to the IL consecutively. The excess amount of acetic acid added to [M1] seemed to prevent the degradation of PET to some extent. Although the acetic acid caused the formation of a gel-like solution, the cellulose content could be reduced in a similar manner as in the trials that employed pure [DBNH] [OAc]. Neither of the approaches seemed to have a substantial impact on the PDI of the samples, implying that [DBNH] [OAc] most likely induced a uniform cleavage of the polymer chains.

Alternatively, a reduction of the temperature during dissolution to 75 °C or lower might further decrease the degradation of PET, which is favorable especially on an industrial scale as larger kneaders utilize significantly shorter dissolution times (5–10 min).

4. Conclusions

The heterogeneity of textile waste represents both a challenge and an opportunity. The different chemical nature of cotton and polyester enables them be separated efficiently, but also demands very specific processing conditions. [DBNH] [OAc] allows a selective dissolution of the cotton component, and a subsequent dry-jet wet spinning of cellulose to textile grade fibers. The spun fibers were found to have properties similar to Lyocell with linear densities between 0.75-2.95 dtex, breaking tenacities of 27-48 cN/tex, and elongations of 7-9%. The recovered PET fraction showed a very low cellulose content (1.7-2.5 wt%), which could further be reduced on an industrial scale by a multi-stage dissolution process. However, PET undergoes visible degradation once dispersed in [DBNH] [OAc], which becomes evident in a decrease of its MMD (<51%) and tensile properties (<52%). The presented data clearly demonstrate that cellulose based, blended textile waste has the potential to serve as a feedstock for conventional fiber spinning processes. Commercialization, nonetheless, requires the development of efficient, reliable techniques for sorting and material identification to be able to deal with a heterogeneous substrate such as textile waste. Moreover, further optimization of the process conditions, especially in terms of dissolution and filtration, might be necessary to limit the degradation of PET. Whereas a certain degradation of cotton is favorable to reduce the consumption of water and chemicals, a high DP of PET is essential to convert it to textile fibers. Therefore, a further possible pathway to recycle the recovered PET fraction might rather include the manufacture of composites or plastic based packaging materials (Joshi, 2001; Zou et al., 2011).

Author contributions

The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript.

Declaration of Competing Interest

The authors declare no competing financial interest.

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Appendix A. Supplementary material

Supplementary data to this article can be found online at https://doi.org/10.1016/j.wasman.2019.07.040.

References

- Adusumalli, R.-B., Keckes, J., Martinschitz, K.J., Boesecke, P., Weber, H., Roeder, T., Sixta, H., Gindl, W., 2009. Comparison of molecular orientation and mechanical properties of lyocell fibre tow and staple fibres. Cellulose 16, 765–772. https:// doi.org/10.1007/s10570-009-9292-2.
- Asaadi, S., Hummel, M., Hellsten, S., Härkäsalmi, T., Ma, Y., Michud, A., Sixta, H., 2016. Renewable high-performance fibers from the chemical recycling of cotton waste utilizing an ionic liquid. ChemSusChem 9, 3250–3258. https://doi.org/ 10.1002/cssc.201600680.
- Awaja, F., Pavel, D., 2005. Recycling of PET. Eur. Polym. J. 41, 1453–1477. https://doi. org/10.1016/j.eurpolymj.2005.02.005.
- Barot, A.A., Sinha, V.K., 2015. Chemical scavenging of post-consumed clothes. Waste Manage. (New York, N.Y.) 46, 86–93. https://doi.org/10.1016/j.wasman.2015.09.012.
- Berggren, R., Berthold, F., Sjöholm, E., Lindström, M., 2003. Improved methods for evaluating the molar mass distributions of cellulose in kraft pulp. J. Appl. Polym. Sci. 88, 1170–1179. https://doi.org/10.1002/app.11767.
- Borrega, M., Tolonen, L.K., Bardot, F., Testova, L., Sixta, H., 2013. Potential of hot water extraction of birch wood to produce high-purity dissolving pulp after alkaline pulping. Bioresour. Technol. 135, 665–671. https://doi.org/10.1016/j. biortech.2012.11.107.
- Briga-Sá, A., Nascimento, D., Teixeira, N., Pinto, J., Caldeira, F., Varum, H., Paiva, A., 2013. Textile waste as an alternative thermal insulation building material solution. Constr. Build. Mater. 38, 155–160. https://doi.org/10.1016/ j.conbuildmat.2012.08.037.
- Brinks, G.J., Bouwhuis, G.H., Agrawal, P.B., Gooiher, H., 2013. Processing of cottonpolyester waste textile, 11 pp.
- Buxbaum, L.H., 1968. The degradation of poly(ethylene terephthalate). Angew. Chem. Int. Ed. Engl. 7, 182–190. https://doi.org/10.1002/anie.196801821.
- Chen, X., Zhang, Y., Cheng, L., Wang, H., 2009. Rheology of concentrated cellulose solutions in 1-butyl-3-methylimidazolium chloride. J. Polym. Environ. 17, 273– 279. https://doi.org/10.1007/s10924-009-0149-4.
- Cowan, J.C., 1981. Method of recovering polyester fibers and cellulosic powder from polyester/cotton textile waste, 3 pp.
- Fink, H.-P., Weigel, P., Purz, H., Ganster, J., 2001. Structure formation of regenerated cellulose materials from NMMO-solutions. Progr. Polym. Sci. 26, 1473–1524. https://doi.org/10.1016/S0079-6700(01)00025-9.
- Gericke, M., Schlufter, K., Liebert, T., Heinze, T., Budtova, T., 2009. Rheological properties of cellulose/ionic liquid solutions: from dilute to concentrated states. Biomacromol. 10 (5), 1188–1194. https://doi.org/10.1021/bm801430x.
- Gruntfest, I., Turner, R., 1974. Treatment of textile waste formed from polyester and cellulosic fibers, 3 pp.
- Haslinger, S., Hietala, S., Hummel, M., Maunu, S.L., Sixta, H., 2019. Solid-state NMR method for the quantification of cellulose and polyester in textile blends. Carbohydrate Polym. 207, 11–16. https://doi.org/10.1016/j.carbpol.2018.11.052.
- Haslinger, S., Hummel, M., Sixta, H., 2017. Separation and upcycling of cellulosecontaining blended waste, 41 pp.

- Hauru, L.K.J., Hummel, M., Michud, A., Sixta, H., 2014. Dry jet-wet spinning of strong cellulose filaments from ionic liquid solution. Cellulose 21, 4471–4481. https:// doi.org/10.1007/s10570-014-0414-0.
- Hämmerle, F.M., 2011. The cellulose gap: the future of cellulose fibers. Lenzinger Berichte 89, 11–21.
- Joshi, Y.V., 2001. Method for direct recycling of plastic waste, 27 pp.
- Liu, F., Cui, X., Yu, S., Li, Z., Ge, X., 2009. Hydrolysis reaction of poly(ethylene terephthalate) using ionic liquids as solvent and catalyst. J. Appl. Polym. Sci. 114, 3561–3565. https://doi.org/10.1002/app.30981.
- Ma, Y., Hummel, M., Kontro, I., Sixta, H., 2018. High performance man-made cellulosic fibres from recycled newsprint. Green Chem. 20, 160–169. https://doi. org/10.1039/C7GC02896B.
- Ma, Y., Hummel, M., Määttänen, M., Särkilahti, A., Harlin, A., Sixta, H., 2016. Upcycling of waste paper and cardboard to textiles. Green Chem. 18, 858–866. https://doi.org/10.1039/C5GC01679G.
- Michud, A., Hummel, M., Sixta, H., 2015a. Influence of molar mass distribution on the final properties of fibers regenerated from cellulose dissolved in ionic liquid by dry-jet wet spinning. Polymer 75, 1–9. https://doi.org/10.1016/j. polymer.2015.08.017.
- Michud, A., Tanttu, M., Asaadi, S., Ma, Y., Netti, E., Kääriainen, P., Persson, A., Berntsson, A., Hummel, M., Sixta, H., 2015b. Ioncell-F: Ionic liquid-based cellulosic textile fibers as an alternative to viscose and Lyocell. Textile Res. J. 86, 543–552. https://doi.org/10.1177/0040517515591774.
- Negulescu, I.I., Kwon, H., Collier, B.J., Collier, J.R., Pendse, A., 1998. Recycling cotton from cotton/polyester fabrics. Textile Chem. Color. 30, 31–35.
- Oakley, E.O., Gorman, F.J., Mason, J.D., 1992. Process for recycling polyester/cotton blends, 6 pp.
- Ouchi, A., Toida, T., Kumaresan, S., Ando, W., Kato, J., 2010. A new methodology to recycle polyester from fabric blends with cellulose. Cellulose 17, 215–222. https://doi.org/10.1007/s10570-009-9358-1.
- Palme, A., Idström, A., Nordstierna, L., Brelid, H., 2014. Chemical and ultrastructural changes in cotton cellulose induced by laundering and textile use. Cellulose 21, 4681–4691. https://doi.org/10.1007/s10570-014-0434-9.
- Palme, A., Peterson, A., de La Motte, H., Theliander, H., Brelid, H., 2017. Development of an efficient route for combined recycling of PET and cotton from mixed fabrics. Text Cloth Sustain. 3, 53. https://doi.org/10.1186/s40689-017-0026-9.
- Parviainen, A., Wahlström, R., Liimatainen, U., Liitiä, T., Rovio, S., Helminen, J.K.J., Hyväkkö, U., King, A.W.T., Suurnäkki, A., Kilpeläinen, I., 2015. Sustainability of cellulose dissolution and regeneration in 1,5-diazabicyclo[4.3.0]non-5-enium acetate: a batch simulation of the IONCELL-F process. RSC Adv. 5, 69728–69737. https://doi.org/10.1039/C5RA12386K.
- Pérez, J., Muñoz-Dorado, J., de La Rubia, T., Martínez, J., 2002. Biodegradation and biological treatments of cellulose, hemicellulose and lignin: an overview. Int. Microbiol.: Off. J. Spanish Soc. Microbiol. 5, 53–63. https://doi.org/10.1007/ s10123-002-0062-3.

- Sammons, R.J., Collier, J.R., Rials, T.G., Petrovan, S., 2008. Rheology of 1-butyl-3methylimidazolium chloride cellulose solutions. I. Shear rheology. J. Appl. Polym. Sci. 110, 1175–1181. https://doi.org/10.1002/app.28733.
- Samperi, F., Puglisi, C., Alicata, R., Montaudo, G., 2004. Thermal degradation of poly (ethylene terephthalate) at the processing temperature. Polym. Degrad. Stab. 83, 3–10. https://doi.org/10.1016/S0141-3910(03)00166-6.
- Serad, S.L., 1993. Polyester dissolution for polyester/cotton blend recycle, 4 pp.
- Shen, F., Xiao, W., Lin, L., Yang, G., Zhang, Y., Deng, S., 2013. Enzymatic saccharification coupling with polyester recovery from cotton-based waste textiles by phosphoric acid pretreatment. Bioresour. Technol. 130, 248–255. https://doi.org/10.1016/j.biortech.2012.12.025.
- Shen, L., Patel, M.K., 2010. Life cycle assessment of man-made cellulose fibers. Lenzinger Berichte 88, 1–59.
- de Silva, R., Wang, X., Byrne, N., 2014. Recycling textiles: the use of ionic liquids in the separation of cotton polyester blends. RSC Adv. 4, 29094–29098. https://doi.org/10.1039/C4RA04306E.
- Sixta, H., Michud, A., Hauru, L., Asaadi, S., Ma, Y., King, A.W.T., Kilpeläinen, I., Hummel, M., 2015. loncell-F: a high-strength regenerated cellulose fibre. Nordic Pulp & Paper Res. J. 30, 43–57.
- Stepan, A.M., Michud, A., Hellstén, S., Hummel, M., Sixta, H., 2016. IONCELL-P&F: pulp fractionation and fiber spinning with ionic liquids. Ind. Eng. Chem. Res. 55, 8225–8233. https://doi.org/10.1021/acs.iecr.6b00071.
- Sun, C.-H., Chen, X.-P., Zhuo, Q., Zhou, T., 2018a. Recycling and depolymerization of waste polyethylene terephthalate bottles by alcohol alkali hydrolysis. J. Cent. South Univ. 25, 543–549. https://doi.org/10.1007/s11771-018-3759-y.
- Sun, Hongyu, Li, Chunguang, Sheng, C., Wang, J., Deng, H., Jia, R., Xia, Y., Tian, F., Lu, Q., Guo, D., 2018. Method for separating and preparing microcrystalline cellulose from polyester-cotton fabric, 9 pp.
- Wang, H., Li, Z., Liu, Y., Zhang, X., Zhang, S., 2009. Degradation of poly(ethylene terephthalate) using ionic liquids. Green Chem. 11, 1568. https://doi.org/ 10.1039/b906831g.
- Venkatachalam, S., Nayak, S.G., Labde, J.V., Gharal, P.R., Rao, K., Kelkar, A.K., 2012. Degradation and recyclability of poly (ethylene terephthalate). In: Saleh, H.E.-. D. (Ed.), Polyester. InTech.
- Yao, Y., Mukuze, K.S., Zhang, Y., Wang, H., 2014. Rheological behavior of cellulose/ silk fibroin blend solutions with ionic liquid as solvent. Cellulose 21 (1), 675– 684. https://doi.org/10.1007/s10570-013-0117-y.
- Ziabicki, A., Takserman-Krozer, R., 1964a. Effect of rheological factors on the length of liquid threads. Kolloid-Z.u.Z.Polymere 199, 9–13. https://doi.org/10.1007/ BF01499686.
- Ziabicki, A., Takserman-Krozer, R., 1964b. Mechanism of breakage of liquid threads. Kolloid-Z.u.Z.Polymere 198.
- Zou, Y., Reddy, N., Yang, Y., 2011. Reusing polyester/cotton blend fabrics for composites. Compos. Part B: Eng. 42, 763–770. https://doi.org/10.1016/ j.compositesb.2011.01.022.