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From postconsumer cotton towels to new cotton textile fibers

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Abstract

The chemical recycling of cotton towels via the Ioncell® technique is demonstrated. Cotton is the most common natural fiber. The season's value jumped 31% to 54.3 billion US$ in 2020/2021, and annually the average value in quota-free periods accounts to 46.3 billion US$. Consequently, enormous amounts of cotton wastes are emerging. Especially, European countries are forced by the new legislation of the union to develop new recycling strategies. Due to uncountable cotton applications, various types of garments exist, which require different recycling strategies. The recycling of an additional cotton waste side stream of Lindström Oy white pre and dyed and white postconsumer cotton roll towels was pursued. The mechanical properties of the fibers and yarns have been evaluated. Thereby, the following elongations and tenacities of conditioned fibers produced at DR11 have been achieved: 10.4%/59.5 cN/tex for preconsumer and 10.6%/60.4 cN/tex for postconsumer white cotton, 10.4%/60.0 cN/tex for postconsumer blue cotton. The achieved elongations at break are close to values reported in literature (7%–14%), however, the tenacities exceed reported values (40–58 cN/tex). Highly oriented fibers of high quality have been produced and with regards to the mechanical properties, a technique to perform fiber-to-fiber upcycling is illustrated.

KEYWORDS

cellulose products, cotton recycling by means of Ioncell®, dry-jet wet fiber spinning technique, fibers, high quality recycled cotton fibers, mechanical properties, rheology, x-ray

1 | INTRODUCTION

According to the book, the fiber year 2022, the world fiber and spunlaid market reached a value of 127.1 million tonnes in 2021. Thereby, manmade fibers including synthetic- and cellulose-based fibers dominate the market and reached a value of 88.2 million tonnes, followed by natural fibers mainly cotton (30.3 million tonnes) and spunlaid output (8.6 million tonnes).[1] The cultivation of cotton crops requires enormous amounts of arable land, plastics for mulching, water, pesticides, and fertilizers. In 2012/2013, an average cotton lint yield of 769 kg/ha was

reached. Depending on the used technologies, the lint yields vary between the cultivation countries. China for instance produced 1438 kg/ha in 2012/2013.[2] With regards to the lint yield of China, around 21 million hectares of arable land are required to meet the current demands for staple fibers and are exposed to soil pollution. Per year, a European citizen uses around 26 kg of textiles and 11 kg is in average disposed per year by each person. Almost 87% of these textile litters are incinerated or end-up in landfill sites. Due to inadequate technologies, less than 1% of textiles are recycled and turned into new products worldwide. Consequently, the potential of certain high-quality resources, like cotton, are entirely unexploited and are destructed prior to reaching the end of service life. By the European Commission, a strategy plan to implement a circular economy was presented in March 2022.[3]

To support this strategy and to counteract the depletion of crucial resources, the possibility of the utilization of cotton roll towels as a raw material for fiber production is presented. By means of the Lyocell type dry-jet wet spinning technology, Ioncell®, the cotton-based material is chemically recycled and turned into new high-performance fibers. Thereby, the material is pretreated to meet the demands of the process. Impurities such as inorganic components are removed before use, and the degree of polymerization (DP) is adjusted to increase the production capacity through higher polymer concentration and ensure trouble-free fiber production. The material is dissolved in an ionic liquid (IL) and processed into second generation fibers by dry-jet wet spinning.[4] The produced fibers are turned into yarn, which is used to produce a dress (Figure 1).

In comparison to commercially available fiber spinning techniques, Ioncell® is a gentle treatment for chemical recycling and allows an upcycling with regards to the fiber strength.[5] The cellulose dissolution is implemented using suitable superbase-based ILs like 1,5-diazabicyclo [4.3.0]non-5-enium acetate ([DBNH][OAc]), 7-methyl-1,5,7-triazabicyclo[4.4.0] dec-5-enium acetate ([mTBDH][OAc]) or 1,8-diazabicyclo[5.4.0]undec-7-enium acetate ([DBUH][OAc]) at adequate temperatures (80–95°C), followed by the generation of fibers at temperatures between 80 and 95°C and the regeneration of the cellulose in cold water (12°C).[6] Ioncell fibers belong to the category of Lyocell fibers. Lyocell fibers are produced by utilizing *N*-methylmorpholine *N*-oxide (NMMO), a direct solvent associated with certain stability issues. The reached tenacities of NMMO fibers are moderately below the values reached by superbase-based fibers, however, the fibers revealed similar elongations.[6]

Many approaches for the recycling of cotton waste have been developed in the past decades. However, even under the best conditions, most of the recycling techniques cause a reduction of the strength and quality of the original cotton fibers. Consequently, the fibers are downcycled not recycled. The yarn quality can be maintained by blending the recycled cotton fibers with synthetic fibers or virgin cotton.[7–10] Because of coloring, impurities, and degradation, the recycling of postconsumer waste is more challenging compared with preconsumer waste.[4,11,12]

FIGURE 1 Dress designed by Kasia Gorniak, Elina Onkinen, and Kirsi Niinimäki (picture by D. Luganski). [Color figure can be viewed at wileyonlinelibrary.com]

In 2016, the recycling of cotton hospital bed sheets provided by the Uusimaa Hospital Laundry (Uudenmaan sairaalapesula Oy, Finland) by Asaadi et al., and in 2019, the recycling of white preconsumer cotton purchased from Iisalmen Kangastukku (Finland) and white postconsumer hospital bed sheets supplied by Uusimaa Hospital Laundry (Uudenmaan sairaalapesulo Oy, Finland) dyed with reactive dyes to develop the recycling of colored postconsumer textile waste by Haslinger et al. were investigated. Thereby, great mechanical fiber properties of 40–58 cN/tex (tenacity, conditioned) and 7%–14% (elongation, conditioned) have been achieved.[4,7]

Now, to face the challenges of postconsumer waste recycling in more detail, the recycling of preconsumer (white) and postconsumer (white and blue) cotton roll towels supplied by Lindström Oy via the Ioncell® technology is presented by a comprehensive study. The fiber properties have been analyzed in detail by means of diverse methods (e.g., x-ray diffraction [XRD], sugar analysis, molar mass distribution, FTIR, TGA), compared with literature values and the initial material. The achieved results demonstrate a fiber-to-fiber upcycling. Chemically recycled fibers were spun into yarns, and finally a knitted demo product was manufactured (Figure 1).

2 | RESULTS AND DISCUSSION

2.1 | Sample preparation

Various white pre and postconsumer as well as blue postconsumer cotton roll towels have been provided by Lindström Oy. Towels from all stages of lifetime, from the beginning till the end, were collected. In compliance with the age, the towels experienced 0 up to 50 washing cycles, which caused degradation, (de)colorization, accumulation of impurities, and partly the removal of finishing substances. To mimic a real textile waste situation, the towels of different ages have been mixed up. At the very first, the determination of the amount of the inorganic impurities, which were accumulated or washed out in the expired lifespan, was of interest. Therefore, the ash content of all the samples was determined and a reduction of the amount of impurities from 0.42% ± 0.03% (preconsumer cotton towels) to 0.31% ± 0.14% (postconsumer white cotton towels), and 0.17% ± 0.04% (blue postconsumer cotton towels) is shown (Table 1 and Table S1).[13] The increased value of the preconsumer cotton towel might be caused by the presence of textile finishing substances, and after several laundry cycles, a considerable amount of inorganics is lost.[14] However, compared with common dry cotton fibers (ash content: 1.1%–3.1%), the amount of inorganics is less pronounced.[15]

In analogy to Ioelovich and Leykin (2008), a high cellulose content of ~96% for white and of 98% for blue cotton towels was found (Table 1).[15] Only small amounts of noncellulosic saccharides (0.83%–1.3%) and nonsaccharides (0.87%–2.7%) are present. Thereby, the number of nonsaccharides includes all acid insoluble and soluble nonsaccharides like components derived from, for example dyes, textile finishing substances, or detergents. The trends of noncellulosic components are in compliance with the age of the garments. Aged garments (postconsumer blue cotton mix) show an increased cellulose, but

TABLE 1 Summary of white pre/postconsumer and blue postconsumer cotton samples studied including their ash content (%), the sugar and nonsaccharide content (%), and the intrinsic viscosity (mL/g).

			Preconsumer white cotton (sample 1)	Postconsumer white cotton mix (sample 2)	Postconsumer blue cotton mix (sample 3)
Ash content (%)		Initial	0.42 ± 0.03	0.31 ± 0.14	0.17 ± 0.04
Saccharides and nonsaccharides	Cellulose (%)	Initial	96 ± 0.03	96 ± 4.0	98 ± 0.58
	Noncellulosic saccharides (%)	Initial	1.3 ± 0.00041	0.88 ± 0.40	0.83 ± 0.12
	Nonsaccharides (%)	Initial	2.7 ± 0.20	3.7 ± 3.9	0.87 ± 0.40
Intrinsic viscosity (mL/g)		Initial	1126 ± 18	1005 ± 4	739 ± 4
		CO after acidic washing	1086 ± 8	946 ± 3	717 ± 12
		CO after acidic treatment	497 ± 2	472 ± 6	433 ± 8

a reduced noncellulosic saccharides value compared with preconsumer white cotton, induced by an extraction of the noncellulosic saccharides during laundry. Vice versa, laundry causes an accumulation of nonsaccharides (postconsumer white cotton) but due to harsh dyeing conditions an abrupt reduction (postconsumer blue cotton). The strength properties of cotton lint fibers depend on the fiber structure and shape. Thus, the length of the polymer chain defined by the DP illustrated by the intrinsic viscosity (η) varies a lot and is affected by the growth process and the processing chain conditions like sunlight, microorganisms, or chemicals.[16] However, in comparison with wood pulp, cotton fibers are high viscosity fibers.[17] The intrinsic viscosity declines within a lifetime of a fabric, due to numerous washing cycles and heavy usage.[11] Consequently, the preconsumer white cotton sample (1126 ± 18 mL/g) showed an intrinsic viscosity higher compared with the postconsumer textiles (white: 1005 ± 4 mL/g; blue: 739 ± 4 mL/g) (Table 1) but low compared to literature values (Haslinger et al.: 2032 mL/g).[4] To remove the accumulated impurities and to reduce the intrinsic viscosity to the requirements of the Ioncell® technology, a two-step sulfuric acid treatment was performed. First, the samples have been purified by a gentle acidic treatment (0.01 M sulfuric acid) causing a reduction of the intrinsic viscosities to 1086 ± 8 mL/g (preconsumer white cotton), 946 ± 3 mL/g (postconsumer white cotton mix), and 717 ± 12 mL/g (postconsumer blue cotton mix), respectively. Next, by means of 0.07 M and 0.05 M aqueous sulfuric acid solutions, the viscosities of the samples were adjusted to the requirements of the system (400–500 mL/g).[5,7]

2.2 | Fiber to textile processing

Because of the intrinsic viscosities of postconsumer white cotton mix (472 ± 6 mL/g) and postconsumer blue cotton mix (433 ± 8 mL/g), spinning solutions of 13% consistency and in the case of the increased intrinsic viscosity of preconsumer white cotton (497 ± 2 mL/g) a spinning soltuion of 12% consistency have been produced at 80°C in combination with the IL [DBNH][OAc].

The viscoelastic properties have been determined and compared with literature values (Figure S1). At 60°C, the dopes preconsumer white cotton and postconsumer blue cotton mix illustrated a zero-shear viscosity (η_0*) of 60,808 and 43,565 Pa s and a crossover modulus ($G' = G''$) of 4808 and 5230 Pa, respectively, and the postconsumer white cotton mix of 55,843 Pa s and 5949 Pa at 65°C (Table S2). In previous studies, optimum spinning of prehydrolized kraft (PHK) pulp was achieved with zero shear viscosity of 25,000–35,000 Pa s and a crossover point at around 4000 Pa.[18] However, Haslinger et al. presented a successful spinning of dopes made of postconsumer cotton of significantly higher zero-shear viscosities of 32,700–78,200 Pa s and crossover points between 3131 and 6996 Pa, which is in line with the achieved results.[4] The dopes were turned into fibers of high quality via dry-jet wet spinning at 60°C (preconsumer white cotton mix, postconsumer blue cotton mix) and 65°C (postconsumer white cotton mix), respectively. Because of the differences in the intrinsic viscosities and amount of impurities of the pulps and the cellulose dope consistencies, the dopes needed to be processed at different temperatures. Thereby, staple fibers for further processing have been collected at draw ratio (DR) 11 and for tensile characterization purposes at the maximum processing DRs, which are close to the reported maximum DRs of PHK pulp: DR18 for preconsumer white cotton, DR11 for postconsumer white cotton mix, and DR17 for postconsumer blue cotton mix.[18]

The color of the produced white Ioncell® CO fibers can be described as off-white or beige, whereas the initial preconsumer cotton is white and the postconsumer cotton depending on the age white to gray. This change in color was illustrated by the determination of the CIE Lab (Table S3) color space values reaching high values for L* (90.55–90.71), a* (0.48–0.40), and b* (0.63–0.60) for the initial CO and reduced values of 90.38–88.56 for L*, 0.17–1.45 for a* and an increased value for b* of around 8.55–7.35 for the produced Ioncell® CO fibers.[19] The postconsumer blue cottons have been dyed by a mixture of three reactive dyes, Drimaren Navy blue Cl-R, Drimaren Blue Cl-BR, and Drimaren Red CL-5B, to prolong the lifetime of the towel by covering the stains. Depending on the lifetime of the garment, the color varies from dark blue to royal purple presented by a L* value of 45.27, a* value of −0.52, and a b* value of −18.66. Due to the cellulose dissolution, the color changed from blue to magenta caused by leaching of one dye component or more into the spin bath, which is a common effect.[4] The color change is expressed by an increase of the L*, a*, and b* values (Table S3).

From all staple fibers, ring-spun yarn was produced and a showpiece was designed (Figure 1).[5]

2.3 | Chemical structure

The effect of the treatment steps on the polymer structure from the start till the end was traced via the determination of the change in the molar mass distribution (Figure 2 and Table S4).

The molar mass (M_w) distribution of the initial sample is broad and reaches values of 465, 366, and 343 kDa,

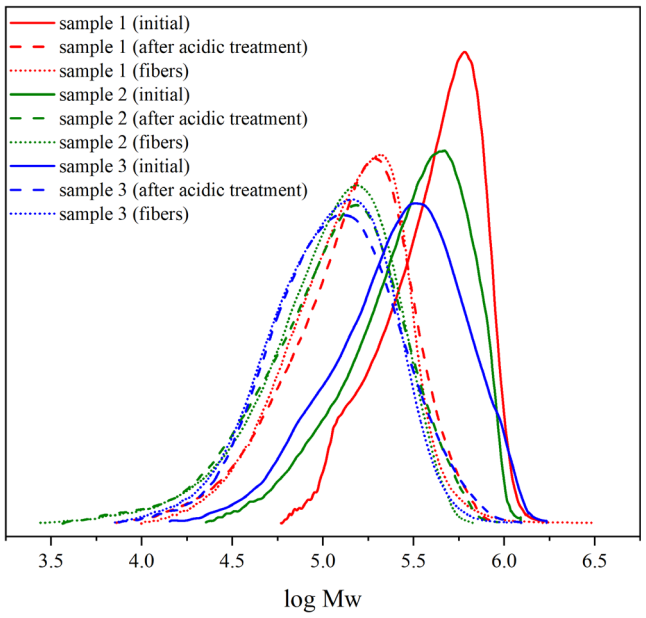

FIGURE 2 Logarithmic illustration of the molar mass distribution change in log(Da) of all samples after each treatment step (initial, after acidic treatment, fibers). [Color figure can be viewed at wileyonlinelibrary.com]

respectively. Due to the limitation of the gel permeation chromatography (GPC) column, the molecular chains are not separated adequately and everything around 6 log M_w is eluated abruptly at the same time, resulting in an asymmetric shape of the distribution. After the acidic treatment (washing and viscosity adjustment), the distribution gets uniform and the values are reduced to 266, 151, and 154 kDa. Because of the chemical recycling via dry-jet wet spinning (e.g., heating time and temperature), the polymer length is further reduced and reaches 169, 146, and 137 kDa, respectively. Under the consideration of the standard deviations, the poly dispersity index (PDI) remains the same in the course of all treatment steps. The initial sample shows no low molecular weight fractions (DP < 100) but many high molecular weight fractions (DP > 2000). This ratio changes from starting material to fibers and only 8.60%, 5.76%, and 5.20% of (DP > 2000) can be found in the end. The low PDI illustrates the uniform length of the cellulose molecules compared with fibers made from wood pulp, facilitating the high strength of the fibers. Similar trends have been reported by Rissanen et al., Haslinger et al., and Michud et al.[4,5,20]

The chemical structure of all components (initial and fibers) has been evaluated via FTIR measurements. All materials showed an O—H stretching band at around 3300 cm^{-1}, a prominent C—O stretching band between 1022 and 1032 cm^{-1}, the group C1 frequency between 890 and 897 cm^{-1} and the C—O—C asymmetric stretching between 1151 and 1164 cm^{-1} (Figure S2), which is in compliance with values reported in literature.[21,22] In case of the initial cotton sample, the group C1 frequency and the C—O—C asymmetric stretching are shifted to higher values compared with the corresponding fiber samples caused by the transformation of cellulose I to cellulose II.[22]

The initial samples (1, 2, and 3) show a great thermal stability (Figure S3). The minor mass loss observed between 50 and 110°C is caused by the vaporization of water attached to the surface. At around 220°C, the degradation starts till it ends at a temperature of around 380°C. Thereby, a mass loss of 80%–85% is illustrated, which is similar to the trends reported by Corradini et al.[23] The purification via acidic treatment affects the stabilities (degradation start at 240–280°C) and the mass losses (87%) and leads to a shift to higher values. In compliance with Xu et al., the produced fibers show a similar thermal behavior with an increased degradation speed.[24]

The chemical recycling causes a change in crystalline structure from cellulose I (initial, after acidic treatment) to cellulose II (fibers) (Figure S4). Thereby, the cellulose I pattern is illustrated by the reflections 1–10 (15° 2θ), 110 (17° 2θ), 200 (23° 2θ), and 004 (35° 2θ) and cellulose II by 1–10 (12° 2θ), 110 (20° 2θ), 020 (22° 2θ), and 004 (35° 2θ).[25] The initial starting materials showed a crystallinity index (CI) of 42% (sample 1), 41% (sample 2), and 44% (sample 3) (Table S4). After the acidic treatment, the amorphous parts have been removed. Consequently, the CI increases and reaches values of 48% and 50% for cellulose I, respectively. The chemical recycling turns cellulose I fibers into cellulose II fibers reaching CIs of 39% (sample 1), 40% (sample 2) and 38% (sample 3), which are in line with literature values (34%–35%).[5,18]

2.4 | Fiber properties

The conditioned and wet tensile properties of the produced fibers (sample 1: DR11, 18; sample 2: DR11; sample 3: DR11, 17) were determined from 10 cm specimens. The fibers produced at DR11 revealed similar linear densities of 1.3 dtex, tenacities of around 60 cN/tex (conditioned), and 55 cN/tex (wet) and elongations at break of 10% (conditioned) and 12%–13% (wet) (Table 2). DR17 (sample 3) and DR18 (sample 1) samples showed reduced linear densities of 0.9 and 0.7 dtex and elongations at break (conditioned: 9%–10%, wet: 11%–12%), respectively. With regards to sample 3, an increase of the DR to DR17 caused a reduction of the tenacity to 58 cN/tex (conditioned) and 54 cN/tex (wet). However, sample 1 DR18 fibers show increased tenacity values of 62 cN/tex (conditioned) and 59 cN/tex (wet). The achieved linear densities

TABLE 2 Linear density and mechanical properties of all produced fibers at DR11 and the corresponding maximum DR's in conditioned and wet state and the total orientation f_t.

Regenerated fiber/properties	Linear density (dtex)	Tenacity (cN/tex)	Elongation (%)	Young's modulus (GPa)	Modulus of toughness (MPa)	Orientation f_t
Preconsumer white cotton (sample 1) (DR11, conditioned)	1.3 ± 0.1	59.5 ± 3.9	10.4 ± 1.0	24.9 ± 2.1	54.6 ± 9.5	0.723 ± 0.087
Preconsumer white cotton (sample 1) (DR11, wet)		56.2 ± 3.4	13.1 ± 1.0	9.2 ± 0.6	61.7 ± 8.5	
Postconsumer white cotton mix (sample 2) (DR 11, conditioned)	1.3 ± 0.1	60.4 ± 2.8	10.6 ± 1.2	27.4 ± 3.1	57.6 ± 7.2	0.728 ± 0.031
Postconsumer white cotton mix (sample 2) (DR 11, wet)		54.3 ± 3.8	13.1 ± 1.1	8.6 ± 0.8	58.6 ± 8.7	
Postconsumer blue cotton mix (sample 3) (DR11, conditioned)	1.3 ± 0.1	60.0 ± 3.2	10.4 ± 0.8	23.8 ± 3.2	55.4 ± 5.2	0.751 ± 0.034
Postconsumer blue cotton mix (sample 3) (DR11, wet)		55.3 ± 3.9	12.2 ± 1.0	9.6 ± 0.8	55.8 ± 8.3	
Postconsumer blue cotton mix (sample 3) (DR17, conditioned)	0.9 ± 0.1	58.1 ± 6.5	9.0 ± 1.3	25.1 ± 5.4	46.0 ± 10.2	0.655 ± 0.035
Postconsumer blue cotton mix (sample 3) (DR17, wet)		54.4 ± 3.5	11.2 ± 1.0	10.5 ± 1.2	49.7 ± 8.1	
Preconsumer white cotton (sample 1) (DR18, conditioned)	0.7 ± 0.1	62.3 ± 2.9	10.0 ± 1.0	27.2 ± 3.5	53.6 ± 8.2	0.702 ± 0.119
Preconsumer white cotton (sample 1) (DR18, wet)		58.7 ± 3.6	11.6 ± 1.2	11.2 ± 0.8	56.5 ± 9.3	

Abbreviation: DR, draw ratio.

(0.7–1.3 dtex) and elongations at break (10%–13%) are close to the values reported in literature (IL-based lyocell fibers from cotton waste: 1.0–1.4 dtex, 7%–14%,[4] NMMO-based lyocell fibers from cotton waste: 1.3 dtex, 10%[26,27]), but the reached tenacities exceed the recently reported values (NMMO and IL-based lyocell fibers from cotton waste: 40–58 cN/tex).[4,26,27] The fibers are highly oriented resulting in a strong structure due to enhanced hydrogen bond formation.[4,5]

The produced fibers show a homogeneous, circular cross-sectional structure and are tensed (Figure 3). No defects can be found on the surface. In previous studies, similar fiber shapes have been reported.[5,28,29]

2.5 | Yarn properties

All the produced white (474 g) and blue (50 g) staple fibers have been combined by color, spin finished to control the antistatic and cohesion properties and turned into white (430 g) and colored yarn (43 g).

The used staple fibers have been even in length and linear density (Table 3). Consequently, the yarn produced via ring spinning was also even with small standard deviations (32.1 ± 0.5 tex, 30.4 ± 0.6 tex). The tenacities of white and blue yarns from regenerated fibers are remarkably higher than cotton yarns. Elongation at break of yarns from regenerated fibers is slightly higher than that of cotton yarns. Compared with literature values, the produced yarns are on the same level or slightly higher.[4,5] In this study, tenacity and elongation of break values of postconsumer cotton yarns were lower than values of virgin cotton yarns in literature showing the degradation of yarns during laundering and wearing.[30,31]

3 | CONCLUSION

Within this work, the upcycling of cotton roll towels via the IL ([DBNH][OAc]) based dry-jet wet spinning technique, Ioncell®, was successfully demonstrated. Thereby, white pre, post and blue postconsumer cotton towels have been grinded, pretreated, dissolved, turned into 2nd generation staple fibers of high strength and further into yarn and a final demonstrator. The success is illustrated by enormous high wet/conditioned fiber tenacities (54–62 cN/tex) and adequate wet/conditioned elongations at break (9%–13%). The high orientation causes an

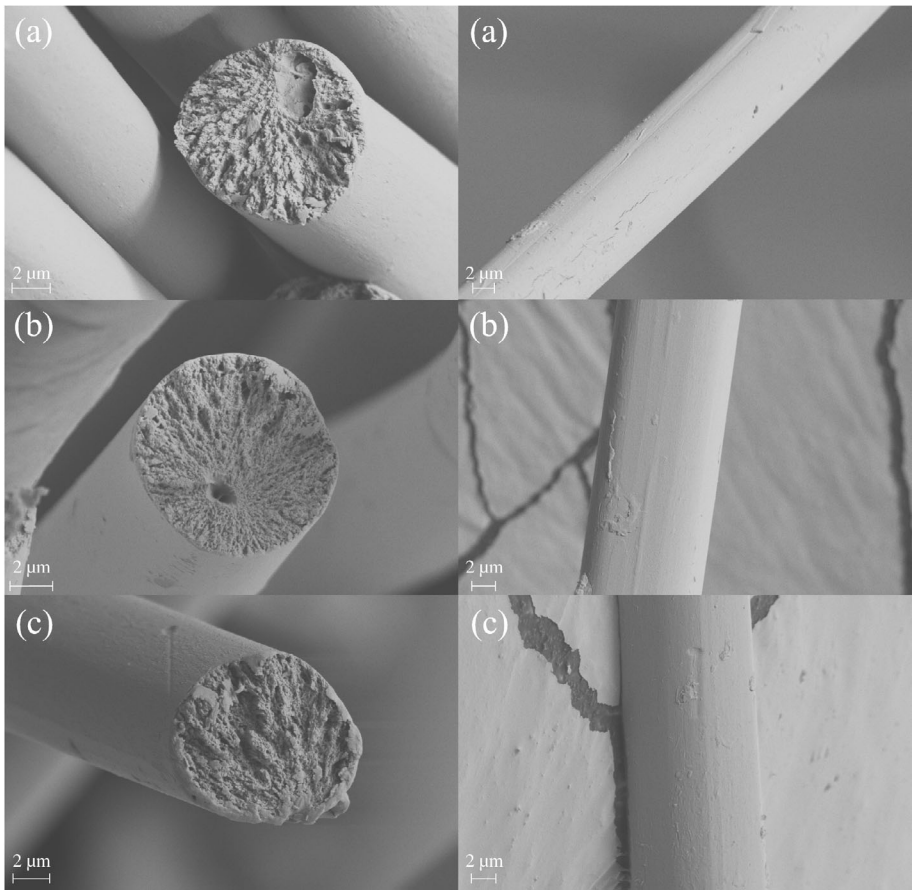

FIGURE 3 Scanning electron microscopy images from the cross section (left) and the surface (right) of the regenerated fibers (a) sample 1, (b) sample 2, and (c) sample 3.

TABLE 3 Linear density and mechanical properties of the initial cotton and all produced yarns from DR11 fibers.

Yarn/properties	Linear density (tex)	Tenacity (cN/tex)	Elongation (%)
White cotton warp yarn unraveled from towel	32.6 ± 2.0	11.8 ± 1.2	5.3 ± 0.5
Blue cotton warp yarn unraveled from towel	31.2 ± 2.9	11.1 ± 3.7	4.2 ± 0.9
White yarn from regenerated fibers (DR 11)	32.1 ± 0.5	32.5 ± 3.9	7.2 ± 0.6
Blue yarn from regenerated fibers (DR 11)	30.4 ± 0.6	30.7 ± 2.9	6.6 ± 0.9

Abbreviation: DR, draw ratio.

enhanced hydrogen bond formation and leads to a reduced flexibility and consequently, reduced elongations at break.[5] During spinning, the blue colored cotton fibers turned into magenta caused by the leaching of one or more dye components into the spin bath. The produced yarn reached high tenacities (31–33 cN/tex) and it was easy to turn into a demonstrator. The process might be optimized with regards to the DP adjustment by means of sulfuric acid. The implementation of an electron beam (e-beam) procedure is pursued.[32] Besides, future investigations will focus on the upscaling of the process and the use of the upcycled cotton fibers for advanced applications (e.g. production of high quality composite materials or carbon fibers).[33,34]

4 | EXPERIMENTAL SECTION

4.1 | Materials and methods

Preconsumer white, postconsumer white, and blue cotton roll towels, dyed with Drimaren Navy blue Cl-R (EU number: 241-164-5, CAS number: 17095-24-8, molecular weight: 991.8 g/mol, empirical formula: $C_{26}H_{21}N_5Na_4O_{19}S_6$, synonym: reactive black 5), Drimaren Blue Cl-BR (EU number: 219-949-9, CAS number: 2580-78-1, molecular weight: 626.5 g/mol, empirical formula: $C_{23}H_{16}N_2Na_2O_{11}S_3$, synonym: Reactive Blue 19/Cl 61,200 anthraquinone class) and Drimaren Red CL-5B (CAS number: 89157-03-09, molecular weight: 1136.3 g/mol, empirical formula:

$C_{31}H_{19}ClN_7Na_5O_{19}S_6$, synonym: reactive red 241/195) have been provided by Lindström Oy, and all other chemicals have been purchased from commercial suppliers. The IL [DBNH][OAc] was prepared according to Rissanen et al.[5] By means of a Wiley Mill M02 (Arthur H. Thomas Company), the textiles have been grinded. The ash content was determined by heating the samples to 575°C by a Naber L51/S muffle oven for 4 h.[13] Following the standard SCAN-CM 15:88 for Scandinavian pulp, paper, and board testing the intrinsic viscosity (η) was examined. The amount of saccharides and nonsaccharides was analyzed according to the Technical Report NREL/TP-510-42618 (revised July 2011) of the National Renewable Energy Laboratory by Sluiter et al.[35] By means of GPC, the molar mass distribution was determined via a Dionex Ultimate 3000 HPLC module (Thermo Fisher Scientific Inc., Waltham, USA) equipped with a Shodex DRI (RI-101) (Showa Denko, Ogimachi, Japan), and Viscotek/Malvern SEC/MALS 20 (multiangle light-scattering, MALS) detector (Malvern Panalytical Ltd., Malvern, UK) system according to Pitkänen and Sixta.[36] The starting materials, the viscosity adjusted intermediate products, and the final fibers have been determined in triplicate. 50 ± 5 mg of each sample was beforehand activated via water, acetone, and dimethylacetamide, next dissolved in a LiCl/DMAc solution (90 g/L), filtrated and diluted to a final concentration of 1.0 mg/mL.[5,37] Hundred microliters of the dissolved sample was injected to the GPC system. FTIR analysis was pursued via a spectrum two FTIR spectrometer from Perkin Elmer. Thermal analysis was performed using a Netzsch STA 449 F3 Jupiter. Each sample (initial cotton and fiber samples) was heated with a heating rate of 10 K/min under helium atmosphere from 40 to 500°C. The XRD measurements of sample 1, 2, and 3 (initial, after acidic treatment, and fibers) were conducted by means of a Smart Lab RIGAKU CuK_α x-ray instrument. The device was operated at 45 kV and at 200 mA ($\lambda = 1.5418$ Å). Each sample was grinded and 100 mg pellets were formed by using a Perkin Elmer Hydraulic Press (4 MPa). Data were collected from each sample at three different spots with a $\theta/2\theta$ geometry ranging from 5° to 60° 2θ. A background correction was performed by subtracting an air scattering profile from the intensity profiles of the samples, and the data were corrected by inelastic scattering. The amorphous cellulose contribution was subtracted from the scattering profiles to determine the CI. The crystal widths (CW_{hkl}) were calculated by the Scherrer equation.[38] Via scanning electron microscopy (Sigma VP Zeiss) the morphology of the regenerated fibers was evaluated. To improve the conductivity, the sample surfaces were sputter coated with Au/Pd (80/20) for 90s with a Q 150 S plus (Quorum) sputter. The fiber tenacity and elongation at break were determined with a Favigraph tensile tester (Textechno GmbH, Germany) in analogy to Rissanen et al.[5] 20 individual fibers of each samples have been tested at conditioned (20 ± 2°C and 65% ± 2% relative humidity [RH]) and wet state (10 s wetting prior to the testing) using a 20 mm gauge length and a 20 mm/min speed. From the stress–strain curves the young's modulus and the modulus of toughness were calculated. The CIE Lab color space was detected by GretagMacbeth™ SpectroScan (Germany) system using a 10° observer and a standard illuminant of D65 according to Haslinger et al.[4] Using the optical properties of fibers (Birefringence), the total orientation of the cellulose was evaluated with a polarizing light microscope (Zeiss Axio Scope, Zeiss AG, Germany) and a tilting compensator according to Rissanen et al.[5] The fiber diameter was calculated using the linear densities and the density (1.5 g/cm^3) and the achieved birefringence was divided by the maximum birefringence value of cellulose (0.062).[39]

4.2 | Experimental procedure

4.2.1 | Sample pretreatment

In total 18 cotton towels have been received, grinded, and analyzed (Table S1: ash content and analysis of saccharides and nonsaccharides). Roll towel 4 was used as preconsumer white cotton (sample 1). An aliquot portion of all white towels (roll towel 1–15) was taken and mixed (postconsumer white cotton mix; sample 2). Additionally, from the blue samples (roll towel 16–18) an equivalent amount was combined (postconsumer blue cotton mix, sample 3). Next, the sample preconsumer white cotton (sample 1) and the mixtures postconsumer white cotton mix (sample 2) and postconsumer blue cotton mix (sample 3) have been washed with 0.01 M sulfuric acid (H_2SO_4), rinsed with excess of deionized water and dried in an oven at 105°C overnight.

4.2.2 | Viscosity adjustment

The viscosity of the samples was adjusted according to the following acidic treatment procedures, respectively: Preconsumer white cotton treated with 0.07 mol/L sulfuric acid (80°C, 60 min, 1:3 solid to liquid ratio), Postconsumer white cotton treated with 0.05 mol/L sulfuric acid (80°C, 60 min, 1:3 solid to liquid ratio), Postconsumer blue cotton with 0.05 mol/L sulfuric acid (80°C, 30 min, 1:3 solid to liquid ratio). The samples have been washed with excess of deionized water and 1 M NaOH was added

till a neutral pH was reached. The excess water was removed by spinning and the samples were dried in an oven at 105°C overnight.[5]

4.2.3 | Chemical recycling by dry-jet wet fiber spinning

For opening purposes, the cotton samples (preconsumer white cotton, postconsumer white and blue cotton mix) have been milled. Dopes of specific cotton consistencies (preconsumer white cotton: 12%, postconsumer white cotton mix: 13%, postconsumer blue cotton mix: 13%) were produced by mixing the pretreated cotton samples with [DBNH][OAc] in a vertical kneader (30 rpm) at 80°C at reduced pressure (90–110 mbar) for 2 ½ hours. The produced dopes have been filtered with a vertical filter press using a GDK Ymax 2 metal filter at 200 bar. The viscoelastic properties of the solutions have been determined using an Anton Paar Physica MCR 302 rheometer. Thereby the complex viscosity (η^*) and the dynamic moduli (G', G'') as a function of the angular frequency (ω) were determined using a dynamic frequency sweep test (100–0.1 s^{-1}) at a temperature range from 60 to 65°C. Under the assumption of the validity of the Cox-Merz rule the zero-shear viscosity (η_0^*) and the crossover point ($G' = G''$) were analyzed using the curve/crossover/G', G'' (omega) and the regression/eta/cross function.[5] By means of a customized dry-jet wet spinning system (Fourné Polymertechnik, Germany), from the preconsumer white cotton and the postconsumer blue cotton mix dopes fibers have been generated at a spinning temperature of 60°C and from the postconsumer white cotton mix at 65°C in analogy to Rissanen et al.[5] Thereby, the spin solution was extruded through a 400-hole spinneret (0.1 mm diameter, length-to-diameter ratio of 0.2). The generated filaments were stretched into an airgap of 0.5 cm, immersed in a water bath (<11°C), collected at different DRs ($DR = v_{take\text{-}up}/v_{extrusion}$) for testing (preconsumer white cotton: DR11, DR18; postconsumer white cotton mix: DR11; postconsumer blue cotton mix: DR11, 17), and at DR 11 for staple fiber processing. The produced fibers have been washed with hot water (60°C) and dried at room temperature.

4.2.4 | Yarn processing and fabric knitting

Following the procedure of Rissanen et al., the staple fibers have been spin finished using Afilan CVS and Leomin PN provided by Archroma with a liquore ratio of 1:20, opened via a Trash Analyzer 281C (Mesdan Lab, Mesdan SpA, Italy), carded by means of a Carding machine 337A (Mesdan Lab, Mesdan SpA, Italy), drafted two times and shaped into a preyarn by Stiro Roving Lab 3371 (Mesdan Lab, Mesdan SpA, Italy), and finally ring-spun with a Ring Lab 82BA (SER.MA.TES srl, Italy) device into a 30-tex (tex = gram/1000 m) yarn (Z torsion, 600 twists per meter).[5] The spun yarn was ply twisted into a 2-ply yarn without a ply twist. The showpiece garment was weft knitted (Stoll CMS ADF 32W E7.3 multi gauge). The linear densities of spun yarn were determined from 10 m skeins ($n = 4$) and expressed in tex. The linear density of cotton yarn was determined from the mass and the length of unraveled warp yarns (white cotton yarn $n = 20$; blue cotton yarn $n = 30$). Weft cotton yarns were too short for the testing. The yarn tenacities ($n = 20$–30) were determined according to Rissanen et al.[5] Yarns were conditioned (20 ± 2°C and 65% ± 2% RH) overnight and tensile testing was performed by MTS400 tensile tester equipped with the 50 N load cell. The testing speed was 250 mm/min and the gauge length was 250 mm.

AUTHOR CONTRIBUTIONS

Inge Schlapp-Hackl: Conceptualization (lead); data curation (lead); formal analysis (lead); investigation (lead); methodology (lead); validation (lead); visualization (lead); writing – original draft (lead). **Marja Rissanen:** Conceptualization (equal); data curation (equal); formal analysis (equal); investigation (equal); methodology (equal); validation (equal); visualization (equal); writing – original draft (supporting). **Krishna Ojha:** Data curation (equal); formal analysis (equal); investigation (equal); methodology (equal); validation (equal); visualization (equal). **Daisuke Sawada:** Data curation (equal); methodology (equal). **Kasia Gorniak:** Resources (supporting). **Elina Onkinen:** Resources (supporting). **Kirsi Niinimäki:** Resources (supporting). **Herbert Sixta:** Conceptualization (lead); funding acquisition (lead); project administration (lead); supervision (lead); writing – review and editing (lead).

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DATA AVAILABILITY STATEMENT

The data that support the findings of this study are available in the supplementary material of this article.

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