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Investigation into the supramolecular properties of fibres regenerated from cotton based waste garments

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ABSTRACT

This paper investigated the supramolecular properties and accessibility of fibres regenerated from cotton-based waste garments and compared to typical lyocell fibres.

The supramolecular and accessibility properties of the cotton-based waste garments fibres regenerated from three sources (waste denim garments, easy care finished cotton fabrics and a blend of cotton-based waste garment with wood pulp) were analysed and compared to the lyocell fibres. The Attenuated Total Reflectance Fourier Transform Infrared (ATR-FTIR) spectroscopy and Wide Angle X-ray Diffraction (WAXD) analyses indicated that the fibres from cotton waste garments had supramolecular properties similar to the typical lyocell fibres. The exception was spun from the cotton pulp reclaimed from easy care treated cotton fabrics and maybe related to increased amorphous cellulose content in its structure. The fibre's accessibility by reagents behaviour correlated well with the supramolecular properties. The results indicate that the waste garment purification process may affect the properties of the pulp and hence the supramolecular properties of the resultant fibres. Further research on the purification and regeneration of fibres from waste garments may lead to the use of cotton waste garments as an alternative feedstock source to the lyocell process.

1. Introduction

The increase in the number of fashion seasons has resulted in an increase in the amount of waste garments generated, particularly in the developed countries. It is reported that at least 50% of the waste apparel collected by developed countries is transported to developing countries to be used as second hand clothing (Ekström & Salomonson, 2014). In addition this second hand clothing business hinders the growth of textile and fashion industries in the developing countries, while the solid waste management cost in the developed source countries is also increasing (Amankwah-Amoah, 2015).

Cellulosic fibres contribute significantly in fashion textiles due to the ability of these fibres to provide comfort to the users. The main sources of cellulosic fibres are cotton and wood pulp with the cotton fibres harvested from the cotton plant, while the wood pulp is used for the regeneration of viscose and lyocell fibres by chemical derivatisation and physical dissolution of the pulp, respectively. The viscose process is characterised by a high consumption of water and the generation of by-products which may pollute the environment. In contrast the lyocell process is more environmental friendly with a "closed" processing structure that generates little effluent. However while the viscose and lyocell fibre making industries become ever more efficient the challenge is to source cheap, readily available raw materials.

In order to globally combat the impact of high fashion affluence and the shortage of the raw material for fibre making, closed loop recycling has been suggested (Danish Fashion Institute, 2012). As part of closed loop recycling, garments and bottles made from polyethylene terephthalate can be depolymerised into monomers and commercially re-polymerised into new filaments for textile application (DEFRA, 2009; Shen, Worrell, & Patel, 2010).

Recent research on closed loop recycling of cellulosic waste garments has suggested the use of the garments as feedstock for regeneration of fibres via the lyocell process (Haule, Carr & Rigout, 2014; Haule, Carr, & Rigout, 2016b). These studies demonstrated the approach to remove the easy care finishes from the cotton waste garments and subsequent regeneration of fibres via the lyocell process. Further the mechanical properties of the fibres reclaimed from cotton based waste garments were found to be superior to the traditional lyocell fibres (Haule, Carr, & Rigout, 2016a). In addition

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the work demonstrated that wood pulp can be blended with the pulp from cotton waste garments and regenerated into fibres with improved mechanical properties. The work reported in this study builds on the previous investigations by studying the supramolecular properties in order to characterize the structure of the fibres reclaimed from cotton waste garments and compare the fibre properties with traditional lyocell fibres. The supramolecular properties relate to the interaction of the cellulose polymer chains above the molecular level. In particular the supramolecular properties investigated were the interactions of the cellulose polymer molecules within the polymers chains and with neighbouring chains, the degree of order of the chains and the proportion of crystalline material in the fibres. Associated with this characterisation the relative fibre accessibility by reagents of a range of fibres with respect to the supramolecular properties was investigated.

The influence of structural properties on the fibre accessibility by reagents was also discussed. The fibres were characterized by the use of Attenuated Total Reflectance-Fourier Transform Infrared (ATR-FTIR) spectroscopy and the results correlated with Wide Angle X-ray Diffraction (WAXD) analyses.

2. Methodology

2.1. Preparation and purification of the fabrics

The material for the spinning of the fibres was prepared as per previously reported procedures (Haule et al., 2016a) and described hereunder.

In order to simulate the effect of extended washing during domestic usage the plain woven cotton fabric was washed 50 times with ECE-phosphate based detergent in a Wascator FOM-71 machine, (Haule, Rigout, Carr & Jones, 2012), and this fabric was the source material for deconstruction into pulp and spinning of regenerated lyocell (*ReCell-1*) fibres. Similarly in order to prepare cross-linked, crease resistant fabrics the plain woven cotton fabric was treated with 100 g/L Dimethylol dihydroxyethylene urea (DMDHEU) easy care finish (Haule et al., 2012) and the easy care finished cotton fabric was then Wascator washed 50 times with ECE-phosphate based detergent and subsequently purified in acid/alkali solutions to produce a component source for the *ReCell-2* fibres (Haule et al., 2014). The *ReCell-2* fibres were prepared from a blend of 20% cellulose recovered by purification of the DMDHEU treated cotton fabrics and 80% wood pulp. In order to prepare the waste indigo dyed denim garments for deconstruction into pulp for spinning, 5 pairs of indigo dyed waste denim were washed once with European Colourfastness Establishment (ECE)-phosphate based detergent, tumble dried, zippers, buttons and threads removed manually and considered the source of *ReCell-Denim* fibres.

2.2. Dissolution and spinning of fibres

The dissolution and spinning of the fibres were as per previously reported methods (Haule et al., 2016a) and involving preparing the requisite spinning dope by mixing 300 g of 50% *N*-methylmorpholine *N*-oxide (NMMO) solution with 27 g pulp and 0.2 g *n*-propyl gallate using a mechanical kneader. The dissolution process was made possible by mixing the pulp and NMMO solution at increasing temperature and vacuum at suitable intervals until the final spinning dope was composed of 9–13% cellulose, 10–13% water and 77–78% NMMO. For every sample the dissolution dope was checked for fibre solubility using a light microscope. The fibres were then spun in filament form from a laboratory scale spinning machine at Lenzing AG, Austria. The spinneret used had 19 holes of 100 μm in diameter and the spinning temperature was 115 °C. The dope throughput was 0.03 g/min per hole and the air gap conditions were set at 30 mm, 24 °C and 53% relative humidity. The fibre fineness and molecular weight were determined as per previously reported methods (Haule et al., 2016a) are presented in Table 1.

2.3. ATR-FTIR analysis of the fibres

The ATR-FTIR spectra were collected on a Nicolet 5700 instrument with a diamond crystal. Scanning was performed from 4000 to 600 cm^{-1} with 128 scan repetitions and a resolution of 8 cm^{-1}. For consistency and reproducibility all spectra were normalized against the C—O—C asymmetric stretching vibration at 1155 cm^{-1}.

The ATR-FTIR crystallinity indices of the ReCell fibres were determined in accordance to the method proposed by Nelson and O'Connor (1964a, 1964b). The method was specifically proposed and applied to cellulose polymers with either crystalline cellulose I or II, or mixtures of both cellulose I and II forms. Infrared absorption results in molecules in the crystalline fractions vibrating at a different frequency to similar molecules in the amorphous fraction and the resultant vibrational intensities of the crystalline and amorphous fractions can be used to estimate the total crystallinity index (TCI) and lateral order index (LOI) of the fibres. The TCI provides information about the crystallinity degree of the cellulose II while the LOI gives an indication of the degree of order of the cellulose II structure. Nelson and O'Connor defined the TCI and the LOI as the ratios of the FTIR 1372/2900 cm^{-1} and 1420/893 cm^{-1} peak intensities, respectively and in this work similar absorption intensities were observed at 1375 cm^{-1}, 2892 cm^{-1}, 1420 cm^{-1} and 894 cm^{-1}, respectively. Previous research indicated that absorption band in the 1372–1375 cm^{-1} region was related to crystallinity in cellulose, and the ratio of this band to the absorption band at 2900 cm^{-1} could provide useful TCI data (Nelson & O'Connor, 1964a). The ratio of 1420 cm^{-1} and 894 cm^{-1} absorption intensities was used for the determination of LOI because the two are related to the amount of crystalline structure and the amorphous region in cellulose II, respectively. Therefore in this work the TCI and LOI were determined at the 1375/2892 cm^{-1} and 1420/894 cm^{-1} absorption bands, respectively.

The ATR-FTIR data were processed using Origin Pro 8.1 SR3 application software.

2.4. Wide Angle X-ray Diffraction (WAXD) analysis of the fibres

The Xpert Phillips (Power 45 kV and current 40 mA) instrument with a copper anode with an X-ray wavelength of 1.54060 Å was used for WAXD analysis. Scans were recorded from 1 to 30° 2θ with a step size of 0.07 with both equatorial scans acquired for aligned fibres, taking the meridional axis as the orientation reference.

In order to account for instrument broadening contribution to the experimental peak width, the same experiment geometry was applied to a powdered silicon standard with the (1 1 1) reflection at 28.40° (2θ). The crystallinity of the fibres regenerated from cotton based waste garments and lyocell fibres were determined from the wide-angle X-ray diffraction patterns recorded perpendicular to the fibres axis. The percentage of crystalline material was calculated by the first determination of the peak height of the diffractograms at the position of the (0 0 2) plane at $2\theta = 21.7°$ (I_{002}) and the peak height of the amorphous background at $2\theta = 16°$ (I_{am}) (Nelson & O'Connor, 1964a) and crystallinity was calculated as Eq. (1):

$$\text{crystallinity} = \frac{I_{002} - I_{am}}{I_{002}} \times 100 \quad (1)$$