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Ioncell Fibres from Alternative Cellulose Sources for Nonwovens

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Abstract

The textile and nonwoven industries are growing rapidly, rely heavily on non-renewable raw materials and have a low rate of recycling. To change this into a more sustainable direction, textile fibres made from renewable and recycled feedstocks must be developed. A potential solution is to increase usage of man-made cellulosic fibres that combine the renewable nature of natural fibres with added benefits related to synthetic fibres. The main raw material used for man-made cellulosic fibres is wood pulp. However, it is important to consider alternative sources of cellulose to avoid putting too much pressure on forest ecosystems and ensuring a wide enough availability of sustainably produced cellulose to fit growing demands. The Ioncell process is an emerging technology for producing regenerated cellulosic fibres using ionic liquids as solvents. A variety of raw materials have successfully been used for production of fibres with the Ioncell process, including wood pulp and cotton, as well as many recycled materials such as cardboard, newsprint, and textile waste.

This study examined the possibility of using cellulosic raw materials other than wood to produce regenerated fibres with the Ioncell process for production of nonwovens. The studied raw materials were recycled nonwovens and straw pulp. Both raw materials were successfully processed into regenerated fibres and the fibres could be used to produce new nonwovens. The recycled nonwoven could also be upcycled into yarn and a knitted textile sample was produced. Two chemical pre-treatments were applied to the recycled nonwoven to allow sufficient spinnability and improve fibre properties: NaOH for removing binders, and H_2SO_4 for removing a TiO_2 mattifying agent. Sufficient spinnability for production of fibres for nonwovens could be reached by using only the NaOH-treatment, and the H_2SO_4-treatment can be added for production of fibres for knitted or woven textiles or if thinner fibres are required. The produced fibres from both starting materials showed a slightly lowered tenacity compared to standard Ioncell fibres, but still reached competitive values compared to commercially available man-made cellulosic fibres. Thus, it can be concluded that both recycled nonwovens and straw pulp can be processed into regenerated fibres suitable for production of new nonwovens with the Ioncell process.

Keywords Cellulose, Ioncell, nonwovens, straw, textile recycling, textiles

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Tiivistelmä

Tekstiili- ja kuitukangasteollisuudet ovat nopeasti kasvavia ja vahvasti riippuvaisia ei-uusiutuvista raaka-aineista ja niillä on matala kierrätysaste. Jotta tämä voi muuttua kestävämpään suuntaan, tekstiilikuituja täytyy kehittää uusiutuvista raaka-aineista ja kierrätetyistä materiaaleista. Mahdollinen ratkaisu tähän on regeneroidut selluloosakuidut, jotka yhdistävät luonnonkuitujen uusiutuvuuden synteettisten kuitujen hyötyihin. Regeneroitujen selluloosakuitujen yleisin raaka-aine on puupohjainen sellu. On kuitenkin tärkeää harkita vaihtoehtoisia lähteitä selluloosalle, jotta vältetään asettamasta liikaa painetta metsäekosysteemeille ja taataan riittävä saatavuus kestävästi valmistetulle selluloosalle täyttämään sen kasvava kysyntä. Ioncell-menetelmä on lupaava teknologia, jolla valmistetaan regeneroituja selluloosakuituja käyttäen ionisia nesteitä liuottimina. Ioncell-menetelmää on onnistuneesti käytetty kuitujen valmistukseen monista eri raaka-aineista, kuten puupohjaisesta sellusta ja puuvillasta, sekä kierrätetyistä materiaaleista kuten pahvista, sanomalehdistä ja tekstiilijätteestä.

Tässä työssä tutkittiin puulle vaihtoehtoisia selluloosan lähteitä regeneroitujen kuitujen valmistukseen Ioncell-menetelmällä kuitukankaita varten. Tutkittavat materiaalit olivat kierrätetty kuitukangas ja olkisellu. Molemmista materiaaleista oli mahdollista valmistaa regeneroituja kuituja ja niistä pystyttiin valmistamaan kuitukankaita. Kierrätetystä kuitukankaasta oli myös mahdollista valmistaa lankaa ja neulottu kangaspala. Kuitujen kehrättävyyden ja ominaisuuksien parantamiseksi kierrätettyä kuitukangasta muokattiin kahdella esikäsittelyllä. NaOH-käsittelyllä poistettiin kankaasta sidosaineita ja H_2SO_4-käsittelyllä poistettiin mattaisuutta tuovaa TiO_2-pigmenttiä. Pelkällä NaOH-käsittelyllä saavutettiin riittävän hyvä kehrättävyys kuitukankaita varten, ja lisäämällä H_2SO_4-käsittely kehrättävyyttä pystyttiin parantamaan riittävälle tasolle valmistaa ohuempia kuituja ja kuituja langanvalmistusta varten. Molemmista raaka-aineista valmistetut kuidut olivat hieman heikompia kuin tavanomaiset Ioncell-kuidut, mutta ne olivat silti kilpailukykyisiä verrattuna kaupallisesti saatavilla oleviin regeneroituihin selluloosakuituihin. Täten voidaan sanoa, että sekä kierrätetty kuitukangas ja olkisellu sopivat kuitujen valmistukseen Ioncell-menetelmällä ja kuituja voidaan käyttää uusien kuitukankaiden valmistukseen.

Avainsanat Selluloosa, Ioncell, kuitukangas, olki, tekstiilikierrätys, tekstiilit

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Preface and acknowledgements

I am very grateful for all the support I have received during this thesis project. I would like to thank my supervisor prof. Michael Hummel for giving me such an interesting topic to work with and all his valuable feedback along the way. I am grateful for the opportunity to delve deeper into the topic of renewable textile fibres and to contribute to making the textile industry more sustainable and creating positive change. I would like to thank my advisor M.Sc. Nicole Nygren for creating a pleasant, encouraging research atmosphere while providing excellent feedback and guidance throughout the thesis process. I would also like to show my gratitude to the whole Biopolymer Chemistry and Engineering -research group for providing a fun and inspiring environment and offering help whenever it was needed. Special thanks to Dr. Inge Schlapp-Hackl for her outstanding help in planning and conducting the experimental work and overall support in completing this thesis. I am also grateful for the collaboration with VTT and the nonwoven samples they were able to produce from my fibres.

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Emily Hart

Symbols and abbreviations

IL	Ionic liquid
MMCF	Man-made cellulosic fibre
SUP directive	Single-use plastics directive
PP	polypropylene
PE	polyethylene
PET	polyester
PA	polyamide
PC	polycarbonate
NaOH	sodium hydroxide
DP	degree of polymerization
CS_2	carbon disulfide
NMMO	*N*-methyl morpholine *N*-oxide
[DBNH][OAc]	1,5-diazabicyclo[4.3.0]non-5-enium acetate
MWD	Molecular weight distribution
[BMIM][Chc]	1-butyl-3-methylimidazolium cyclohexyl carboxylate
PDI	polydispersity index
DBN	1,5-diazabicyclo[4.3.0]non-5-ene
DMC	dry matter content
CED	cupri-ethylenediamine
GPC	gel permeation chromatography
DMAc	dimethylacetamide
SRS	sugar recovery standard
HPLC	high performance liquid chromatography
FTIR	Fourier Transform Infrared Spectroscopy
DR	draw ratio
STA	simultaneous thermal analysis
TGA	thermogravimetric analysis
DTG	differential thermogravimetric analysis
DSC	differential scanning calorimetry

1 Introduction

The textile industry is one of the fastest growing industries in the world as well as one of the biggest polluters. The global production of textile fibres was almost five times as big in 2022 as it was in 1975, with 113.8 million tons of textile fibres being produced in 2022 (Statista Search Department, 2023). The amount of textiles produced per capita has also been increasing throughout the years. In 1975, 8.4 kg of textile fibres were produced per person in one year, while it is estimated that 17.5 kg of textiles will be produced per person in 2030 (Statista Search Department, 2022). In 2015, the greenhouse gas emissions of textiles production were 1.2 billion tonnes of CO_2 equivalent, which is more than the emissions of all international flights and maritime shipping combined (Ellen MacArthur Foundation, 2017). Furthermore, the textile industry relies heavily on non-renewable raw materials, uses large amounts of water, has a low rate of recycling, and is responsible for microplastics accumulating in the environment (Chen *et al.*, 2021). These factors demonstrate the significant need to develop textile fibres and processing methods that are better for the environment, focus more on the utilization of renewable raw materials, and support recycling.

Like textiles overall, the production of nonwovens has increased in the past years. Global consumption of nonwovens in 2019 was estimated to be 11.2 million tonnes, with an average annual growth rate of 5.8% between 2014 and 2019 (Smithers, 2019). In 2024, the global consumption is predicted to increase to 15.2 million tonnes (Smithers, 2023). According to the 2019 sustainability report of EDANA (2019), the leading global organization of nonwovens, 66% of raw materials used for producing nonwovens are from non-renewable, non-recycled sources. However, the use of renewable and recycled materials is increasing (EDANA, 2019), indicating an opportunity for newly developed nonwovens made from more sustainable materials to enter the market.

Nonwovens are used in a plethora of applications, including hygiene products, wipes, household textiles, construction, and filtration (Albrecht *et al.*, 2006). Many of these are single-use products such as feminine hygiene products, facemasks, diapers, and disposable wipes, that are usually produced from synthetic materials such as polypropylene (PP), polyethylene (PE), polyester (PET), polyamide (PA), and polycarbonate (PC) (Bhat & Parikh, 2010). These materials are not biodegradable, and the short lifespan of the products creates disposability problems (Santos *et al.*, 2021). This poses an issue for the nonwoven industry due to increased environmental consciousness from consumers as well as new restrictive legislations.

The most notable recent legislation affecting the use of nonwovens in Europe is the European Union's (EU) directive on single-use plastics (SUP) (EU Directive 2019/904, 2019). The aim of the directive is to prevent and reduce the effect of plastic products on the environment, with particular focus on marine litter. The directive's goals have been implemented by multiple means, including bans and design requirements on certain plastic products, as well as demands for specific labelling related to proper waste disposal. Some nonwovens, most notably feminine hygiene products and disposable wipes, fall under the scope of the SUP directive and thus particularly require increased utilisation of alternative, non-plastic raw materials. It should be noted that in the guidelines for the SUP directive, released in 2021 (EU 2021/C 216/01, 2021), plastics are defined as chemically modified polymers, thus excluding natural polymers. Regenerated cellulose fibres such as viscose, Lyocell, and Ioncell are considered natural polymers since there is no chemical modification of the cellulose and are thus not restricted by the SUP directive.

Cellulose, a renewable and biodegradable material (Klemm *et al.*, 2005), is a fitting sustainable alternative to synthetic, oil-based materials for nonwoven textiles. Cellulosic fibres also provide many advantageous properties when used as material for nonwovens, such as softness and high

absorbency (Santos *et al.*, 2021). The most common ways of utilizing cellulose in nonwovens are in the form of wood pulp, cotton, and viscose (Santos *et al.*, 2021). Cultivation of cotton requires very large amounts of water and is reaching its maximum capacity because more and more land area is needed for production of food crops (Hämmerle, 2011). At the same time, the need for cellulosic fibres is set to increase by 45% by the year 2030 (Hämmerle, 2011). Less than 60% of this can be covered by use of cotton, creating a so called "cellulose-gap" (Hämmerle, 2011). Thus, the source for cellulosic products should be shifted more towards other resources than cotton. Man-made cellulosic fibres (MMCFs), such as viscose, are one of the solutions to this issue. They utilize renewable resources, while still providing the favourable mechanical properties and possibility for modification of synthetic fibres (Santos *et al.*, 2021). However, the viscose process involves considerable environmental issues, which has driven the development of newer man-made cellulosic fibres that can be manufactured in more sustainable ways (Kreze and Malej, 2003).

One of the possible alternatives to viscose is Ioncell, a man-made cellulosic fibre produced from ionic liquid (IL) solutions (Michud *et al.*, 2016a). The production of Ioncell avoids the negative environmental effects of the viscose process, while possessing mechanical properties superior to other MMCFs and comparable to synthetic fibres (Moriam *et al.*, 2021). Furthermore, Ioncell fibres feel soft and have good breathability (Moriam *et al.*, 2021). As they are cellulose based, the fibres are also biodegradable and do not create microplastics that end up in nature (Moriam *et al.*, 2021).

Man-made cellulosic fibres are mainly produced from wood-derived pulp or cotton linters (Shen *et al.*, 2010). As described above by the "cellulose gap", textile fibres should no longer rely too heavily on cotton as a raw material. In contrast, wood is a more environmentally friendly alternative, as it requires significantly less water and less land area (Hämmerle, 2011). Nonetheless, it is important to consider alternative sources of cellulose also for MMCFs to avoid putting too much pressure on forest ecosystems and

ensuring a wide enough availability of sustainably produced cellulose to fit growing demands. Alternative cellulose sources to wood include non-wood plants and recycled feedstocks. Recycling products decreases the need for new resources and can also lead to energy savings and a diminished environmental footprint (Birat, 2015). Non-wood plants in turn provide a resource that is currently underutilized, particularly if the non-wood plant matter originates from waste streams and by-products of other industries. The most significant non-wood plant suitable for pulping is straw. It is produced as a by-product of cereals grown for food and currently treated as waste that is disposed of by burning (Espinosa *et al.*, 2016). According to the principles of circular economy, the value of resources should be preserved for as long as possible (Suárez-Eiroa *et al.*, 2016). Therefore, utilizing waste and side streams for production of higher-value products is preferable to burning them as waste.

This thesis aims to evaluate the production of man-made cellulosic fibres from alternative sources of cellulose for nonwovens using the Ioncell process. The raw materials examined are straw pulp and recycled nonwoven fabric. Ioncell fibres are produced from each material with pre-treatments applied when necessary, and both the starting materials and the resulting fibres are analysed. The results are used to evaluate whether straw pulp and recycled nonwovens are suitable for producing Ioncell fibres and are these fibres suitable for production of new nonwovens. The possibility of producing knitted fabrics from the produced fibres is also explored. Finally, the thesis will examine how the properties of the starting materials affect the properties of the resulting regenerated fibres and how these properties are changed by introducing various pre-treatments to the process. The thesis aims to answer two main research questions: Can recycled nonwovens and straw pulp be used to produce regenerated fibres with the Ioncell process? Can the produced fibres be used for production of nonwovens?

2 Literature review

2.1 Man-Made Cellulosic Fibres

Man-made cellulosic fibres (MMCFS) are produced by dissolving cellulose and reconstructing fibres from the resulting solution (Parajuli *et al.*, 2021). This way, the advantageous properties of natural textile fibres, such as biodegradability, moisture control, and comfortable feel, are preserved, while adding benefits related to synthetic fibres, such as possible chemical functionalization and tailoring properties like strength and smoothness (Bredereck & Hermanutz, 2005; Parajuli *et al.*, 2021). The dissolution of cellulose is an essential step in producing all MMCFs, since cellulose cannot be processed by melting like other common thermoplastic polymers without disintegrating the cellulose structure (Parajuli *et al.*, 2021). However, the complexity of the inter- and intramolecular hydrogen bonding networks and crystalline structure of cellulose make dissolution rather difficult (Medronho & Lindman, 2015). For this reason, many different methods of dissolving cellulose have been studied. The various methods can be divided into derivatization and direct dissolution (Sayyed *et al.*, 2019). Derivatizing processes require modification of cellulose into a derivative form before dissolution, while direct solvents can dissolve cellulose without this step (Sayyed *et al.*, 2019).

The first man-made cellulosic textile fibre was developed in the 1850s after the discovery of the cellulose derivative cellulose nitrate (Woodings, 2001). The original fibre did not achieve widespread use in the textile industry due to its high flammability, but after a denitration step was introduced, cellulose nitrate became commercially available as “artificial silk” in the late 1800s (Woodings, 2001). Simultaneously, another derivatizing process, involving the reaction of cellulose with alkali and carbon disulphide (CS_2) to create cellulose xanthate, was being developed (Hegan, 1951). The fibres produced using this method came to be known as viscose fibres, and commercial production began in the early 1900s (Woodings, 2001). The production and

profitability of viscose fibres grew rapidly in the 20th century (Woodings, 2001), and viscose is still the most common type of man-made cellulosic fibre today (Jiang *et al.*, 2020). One of the reasons behind the success of viscose is its high versatility. Various modifications are possible at different stages of the viscose process allowing the properties of viscose fibres to be tailored according to specific needs (Bredereck and Hermanutz, 2005).

Despite the extensive use of viscose fibres, the viscose process involves adverse environmental and occupational health effects. These effects are largely due to the use of carbon disulphide, that gets dispersed in exhaust gases, process baths, and solid wastes as various sulphur compounds such as CS_2, H_2S, COS, and SO_2 (Sayyed *et al.*, 2019). These compounds are known to cause air and water pollution (Majumdar *et al.*, 2022), as well as an increased risk of several serious health conditions for the workers in viscose manufacturing, such as cardiovascular disease, reproductive and menstrual disorders, and hypertension (Vigliani, 2001; Sakurai, 1982; Ma *et al.*, 2010; Chang *et al.*, 2007). Therefore, it is important to find alternative methods for producing MMCFs avoiding usage of chemicals and processes that are harmful to the environment or human health.

2.1.1 The Lyocell Process

An alternative method to the viscose process is the Lyocell process. The desire to develop a manufacturing process with a lower environmental impact than the viscose process as well as achieve fibres with better performance for lower costs were the most notable factors driving the development of Lyocell fibres (White, 2001). Both these goals are reached with the Lyocell process, and the resulting fibres are very versatile and suitable for a wide range of applications (White, 2001). While viscose fibres are produced by indirect dissolution of cellulose, Lyocell fibres are made by direct dissolution without the need for a derivatizing step (Borbély, 2008). This can be achieved with different solvents, but the most common one is *N*-methyl morpholine *N*-oxide (NMMO) (Sayyed *et al.*, 2019). NMMO is non-

toxic and it does not produce hazardous effluents (White, 2001). Furthermore, over 99% of the solvent can be recovered and recycled in the process (Sayyed *et al.*, 2019).

Zhang *et al.* (2018) describe the Lyocell process as consisting of five steps. The first step is dissolution, in which cellulose pulp is mixed with an aqueous solution of NMMO. This is followed by filtration to remove coarse particles. Next, the fibres are extruded into a coagulation bath through an air gap to regenerate them. This is known as dry-jet wet spinning (Fink *et al.*, 2001). The fourth step described by Zhang *et al.* (2018) is washing, during which the leftover NMMO is recovered for reuse. The final step is finishing, which includes a variety of possible post-treatments of the fibres including drying and bleaching. Overall, the steps in the Lyocell process are simpler and less time and resource consuming as in the viscose process. The Lyocell process is summarized in Figure 1. Despite its improvements in comparison to the viscose process, the production of Lyocell fibres with NMMO also has some disadvantages. The process includes dangerous side reactions, that can cause degradation of cellulose, discolouration and decreased performance of fibres, decomposition of NMMO, and a high usage of thermal stabilizers (Rosenau *et al.,* 2001). Therefore, the Lyocell process using NMMO as a solvent could still be improved or replaced by another process with limited disadvantages.

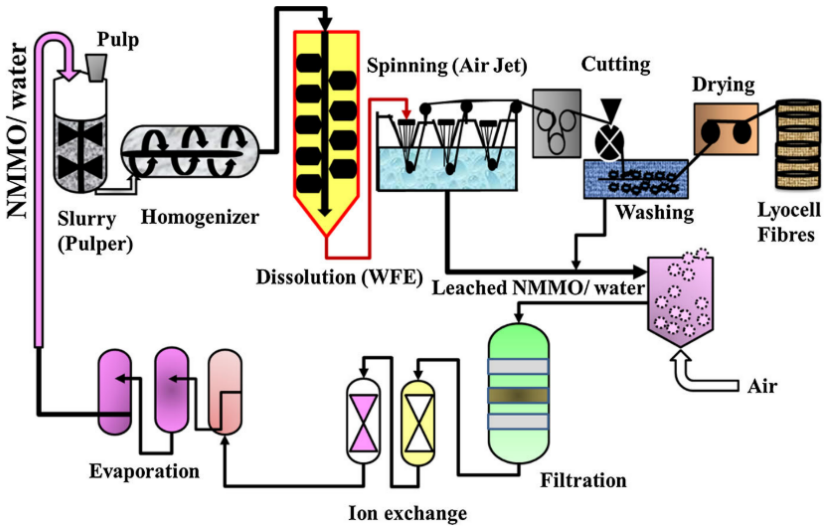

Figure 1. Lyocell process using NMMO as solvent (Sayyed et al. 2019)

2.1.2 The Ioncell Process

The Ioncell process, summarized in Figure 2, is a Lyocell-type process to produce man-made cellulosic fibres using ionic liquids as a direct solvent (Michud *et al.*, 2016a). Ionic liquids are defined as compounds consisting only of ions, with a melting point below 100 ℃ (Lei *et al.*, 2017). They possess unique physicochemical properties such as low vapour pressure, high thermal stability, and high solvation ability making them fitting for MMCF production (Wang *et al.*, 2012). Furthermore, the combination of cations and anions used to create ionic liquids is vast, allowing a wide variety of customizations according to required properties (Parajuli *et al.,* 2021). Most ionic liquids used for dissolving cellulose have been imidazolium-based (Sixta *et al.*, 2015). Despite the inherent advantageous properties of ionic liquids, Bentivoglio *et al.* (2006) show that these imidazolium-compounds require use of a stabilizer to prevent severe cellulose degradation just as NMMO in the Lyocell process. They are also described as corrosive to metal processing equipment and potentially toxic to aquatic environments. Furthermore, the study showed that the recovery of imidazolium-based solvents is significantly more energy consuming and expensive than in the original Lyocell process, due to the need to remove all water. For these reasons, a novel ionic liquid solvent, 1,5-diazabicyclo[4.3.0]non-5-enium acetate ([DBNH] [OAc]) has been developed for the Ioncell process (Parviainen *et al.*, 2013; Michud *et al.*, 2016a). Other ionic liquids can also be used in the Ioncell process, but [DBNH] [OAc] is the most researched and the one used in the experiments of this thesis. The solvent allows fibre spinning at distinctly lower temperatures than in the NMMO-based Lyocell process, thus leading to decreased energy consumption and negligible cellulose degradation that only results from shear forces in the dissolution step (Hummel *et al.*, 2016). The solvent is also noncorrosive and safe to work with (Hauru *et al.*, 2014), improving occupational health in comparison to the traditional Lyocell process as well as preserving processing equipment better than other ionic liquids. Furthermore, the resulting fibres exhibit

excellent mechanical properties, reaching or exceeding the performance of conventional Lyocell fibres (Hauru *et al.*, 2014).

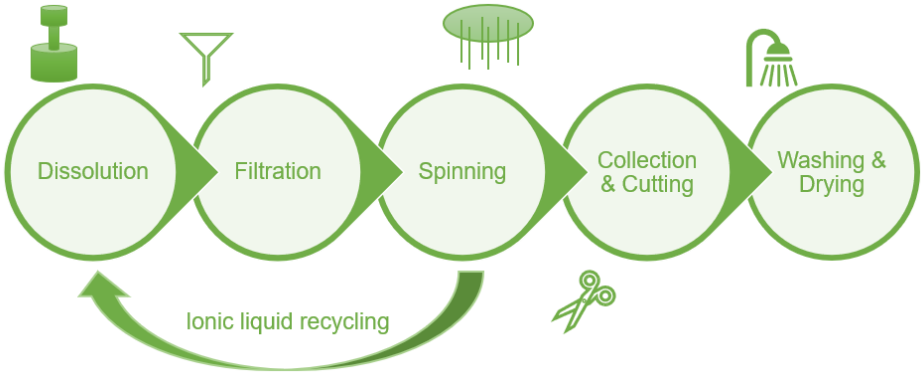

Figure 2. The Ioncell process

The Ioncell process (Figure 2) is very similar to the NMMO-based Lyocell process, as described by Sixta *et al.* (2015) and Hauru *et al.* (2014). Since the melting point of [DBNH] [OAc] is 70 °C, the ionic liquid must first be heated up to this temperature to melt it. It is then mixed with pulp, to produce a suspension generally with 13 wt-% cellulose. The dissolution of cellulose is induced by mixing, heating generally at 80 °C, and operating under vacuum (50 mbar). The dissolution step is followed by filtration to improve solution quality. Next, the solution is spun into filaments through a spinneret into a cooled water coagulation bath via a 1 cm air gap. The unit for this dry-jet wet spinning process is depicted in Figure 3. The filaments are then gathered onto a godet couple. By changing the extrusion speed and the take up velocity of the godet couple, the amount of stretch, or draw ratio (DR), of the fibres can be changed. Finally, the fibres are cut, washed, and dried.

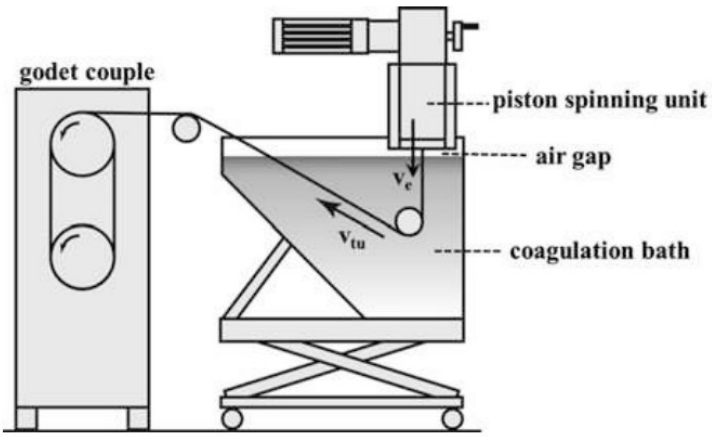

Figure 3. The spinning unit used for dry-jet wet spinning of Ioncell fibres. V_e refers to the speed at which the filaments are extruded into the air gap and then immersed in the water coagulation bath. V_{tu} refers to the take up velocity of the godet couple. (Hummel et al., 2016)

Currently, ionic liquids are a more expensive cellulose solvent than NMMO (Hummel *et al.*, 2016). However, the situation was similar for NMMO when it was being developed and costs dropped as the solvent became commercialized (Sixta *et al.*, 2015). Thus, the same could happen for ionic liquids as well after widespread commercialization. Another crucial aspect in making the Ioncell process economically viable is the recovery and recycling of the ionic liquid used in the process. In the NMMO-based Lyocell process, NMMO is successfully recovered and reused, decreasing the environmental impact of the process as well as decreasing costs (Sixta *et al.*, 2015). A solvent recycling method equal to the NMMO process should be applied to the Ioncell process for it to be able to compete economically with the conventional Lyocell process. This is a concern, because unlike in the recovery of NMMO, water must be removed completely from ionic liquids for them to be used again as solvents, increasing the cost of recycling (Bentivoglio *et al.*, 2006). However, this could be compensated by other aspects of the Ioncell process that decrease overall costs, such as the possibility to use low-cost cellulose waste as raw material, possibility for higher cellulose concentrations, and lack of stabilizers (Sixta *et al.*, 2015). The determination of an ideal recycling method for ionic liquids depends on

the thermal stability of the IL and its interactions with water (Elsayed *et al.*, 2020).

2.2 Nonwovens

Nonwovens are defined as "engineered fibrous assembly, primarily planar, which has been given a designed level of structural integrity by physical and/or chemical means, excluding weaving, knitting or papermaking" (ISO 9092:2019, 2019). This means that nonwovens are produced without a yarn spinning stage, and instead bond staple fibres together into a web-like structure (Wilson, 2022). When the term originated, nonwovens were considered cheap substitutes for traditional textiles, and they were produced from carded staple fibres on converted processing machinery (Wilson, 2010). Since then, the manufacturing of nonwovens has grown into a highly profitable, refined industry with substantial investment in research and development (Wilson, 2022). Many of the applications for nonwovens are relatively inexpensive, single-use products such as wipes and diapers, but nonwovens are also essential for many high-value, multiple-use, and long-life products especially in industrial applications (Wilson, 2022).

The most common categories of nonwoven production methods are dry laying, wet laying, and polymer-laying (Bhat & Malkan, 2006). In general, dry laying methods originate from textile production, wet laying from papermaking, and polymer-laying from polymer extrusion (Wilson, 2010). The general steps in all these methods are web formation, web bonding, and finishing (Albrecht *et al.*, 2006). The main differences between the methods appear in the web formation process as well as the type of raw material that can be used (Albrecht *et al.*, 2006).

2.2.1 Dry Laying

Dry laying is suitable for all staple fibres, that can be carded or dispersed in air (Karthik *et al.*, 2017). The process begins with preparing the fibres for processing by first opening and then blending them (Karthik *et al.*, 2017).

According to Wilson (2010) the web formation process in dry laying can be further divided into two methods: carding and airlaying. These processes are depicted in Figure 4. The primary goal of carding is to separate entangled fibres and to assemble the fibres in the form of a web. This is achieved mechanically, by holding one surface of the fibres while brushing them out. The web can be parallel laid, with the fibres aligned along the machine direction, or randomised. Wilson (2010) continues by describing airlaying as a process with roots in papermaking that relies on air as the force for forming fibres into a web. First, rolls of pulp are fed into a hammermill, which uses small, rotating hammers for separating the pulp into loose fibres. The fibres are then either sifted through a coarse screen or passed through a series of holes in a large cylinder and then placed onto a forming wire with the help of a vacuum. Airlaid nonwovens generally have lower density and higher softness than carded nonwovens (EDANA).

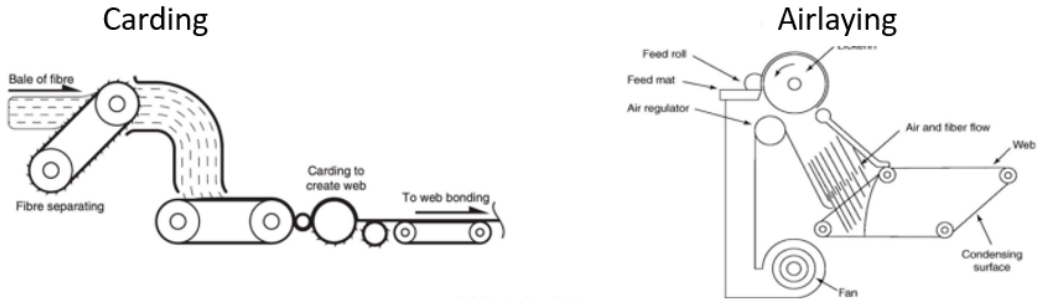

Figure 4. Schematic of the two main dry laying processes: carding and airlaying (Modified from Venkataraman et al., 2023)

2.2.2 Wet Laying

Wetlaid nonwovens are described by Wilson (2010) as nonwovens produced by machinery designed for manipulating fibres suspended in liquid. The raw materials used for wetlaid nonwovens are water and any natural or man-made fibres, along with small amounts of surfactants, binders, and additives (White, 2006). In general, the fibres should have a linear density of 0.1 - 6.0 dtex, preferably no crimp, and relatively short length (0.3-10 mm) (White, 2006), but longer fibres of up to 40 mm can also be included (Koukoulas,

2022). According to White (2006) fibres are prepared for wet laid nonwovens in the same way as they would be prepared for papermaking, by dispersing and swelling them in water. Further treatment can also include beating or refining to increase fibrillation and swelling. Next the fibres are transported on a continuous moving screen through which the water is filtered out (Wilson, 2010). This process differs from the papermaking process by using a more dilute fibre suspension (0.5% for papermaking and 0.0025% for nonwovens), as well as angling the wire at an incline (Wilson, 2010). Wet laying is the least commonly used production method for nonwovens (Koukoulas, 2022). This is due to expensive investments and extensive use of water in the process (Wilson, 2010). However, the main advantage of the process is the ability to process such a wide variety of raw materials (Wilson, 2010). The wet laying process is portrayed in Figure 5.

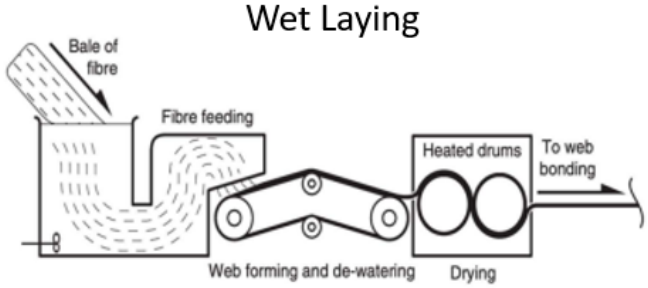

Figure 5. Schematic of the wet laying process (Modified from Venkataraman et al., 2023)

2.2.3 Polymer-laying

Polymer-laid nonwovens are produced by extrusion spinning, in which a web structure is directly formed from molten filaments without need for collecting separate fibres (Bhat & Malkan, 2006). The elimination of certain steps required in other nonwoven production methods leads to a decreased cost and increased production capacity (Bhat & Malkan, 2006). In general, the method consists of sheets of filaments being extruded onto a conveyor and directly forming a random-oriented web (Wilson, 2010). Polymer-laying can further be divided into the spunlaying and meltblowing methods (Bhat &

Malkan, 2006). The basic principles of these methods are the same, but the technology is different (Bhat & Malkan, 2006). The stages of these methods are shown in more detail in Figure 6. Since the processes involve melting of the raw material, only thermoplastic polymers can be used for polymer-laid nonwovens. Thus, cellulose-based fibres are not suitable for polymer-laying.

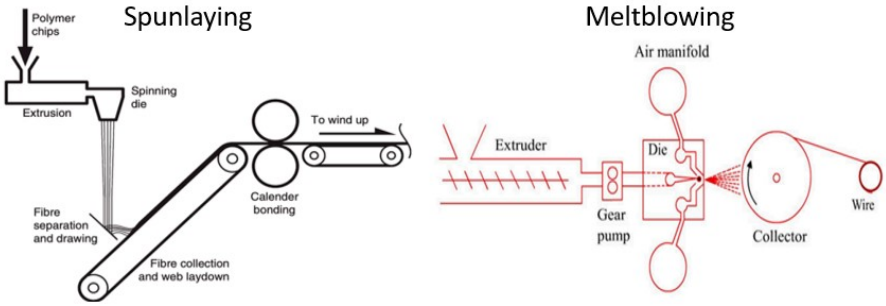

Figure 6. Schematic of the two main polymer-laying processes: spunlaying and meltblowing (Modified from Venkataraman et al., 2023)

2.2.4 Web Bonding

After web formation utilizing any of the previously discussed methods, the fibre web must be bonded by mechanical, chemical, or thermal processes (Wilson, 2010). The web produced in the web formation step is feeble and weak, and the bonding stage imparts strength and stability to the structure (Karthik *et al.*, 2017). The degree of bonding is an important factor determining many of the properties of the nonwoven fabric, such as strength, porosity, flexibility, softness, and density (Wilson, 2010). In principle, mechanical bonding methods use mechanical force to entangle fibres together and form a fabric that stays intact (Karthik *et al.*, 2017). Chemical methods in turn use chemical adhesives to bind the fibres together by saturating, spraying, printing, or foaming (Wilson, 2010). The most used binding agents are acrylate polymers and copolymers, styrene-butadiene copolymers, and vinyl acetate ethylene copolymers (EDANA). Thermal bonding utilizes heat, often in combination with pressure, to make fibres softer and then coalesce without melting them (Wilson, 2010). Thermal bonding processes can be divided into air bonding and calendar bonding

(Karthik *et al.*, 2017). The classification of different processes used for web bonding of nonwovens can be found in more detail in Figure 7.

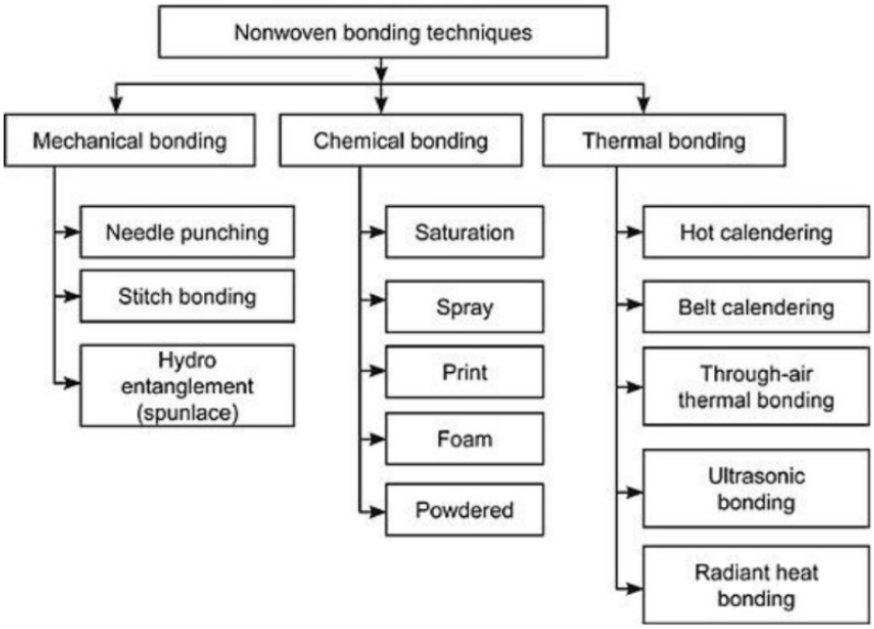

Figure 7. Bonding methods used in the web bonding process for nonwovens (Karthik et al., 2017)

2.3 Cellulose for Regenerated Textile Fibres

2.3.1 Structure and Chemistry of Cellulose

Cellulose is the most abundant organic polymer that occurs in nature (Coffey *et al.*, 1995). The polymer chain consists of anhydroglucose units that are linked together by β-1,4-bonds (Coffey *et al.*, 1995). Every other anhydroglucose unit is rotated 180° in the plane due to the bond angles favoured by the acetal oxygen bridges that connect the units (Klemm *et al.*, 2005). This configuration gives cellulose a linear, rigid structure (Coffey *et al.*, 1995). The molecular structure of cellulose can be seen in Figure 8.

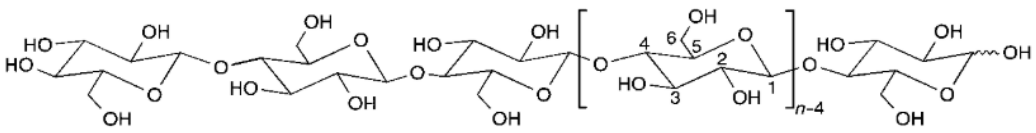

Figure 8. Molecular structure of cellulose (Klemm et al., 2005)

As seen in Figure 8, cellulose molecules have many hydroxyl (OH)-groups that are attached to the C2, C3 and C6 carbon atoms. This allows cellulose to form strong hydrogen bonds both inter- and intramolecularly (Lange Bregado *et al.*, 2019). These hydrogen bond networks form different kinds of crystalline structures in cellulose (Klemm *et al.*, 2005). The OH-groups are also responsible for many of the inherent properties of cellulose, such as hydrophilicity, chirality, degradability, and possibility for chemical modification and variability (Klemm *et al.*, 2005). In the solid state, native cellulose consists of highly ordered crystalline domains and disordered amorphous regions (Gupta *et al.*, 2019). The less ordered regions allow the hydroxyl groups to be more readily available for reaction than the ones in the crystalline regions (Gupta *et al.*, 2019). Therefore, many of the properties of cellulose depend on its hierarchical structure and organization.

There are different ways cellulose can form a crystalline structure. The most studied polymorph is the form of cellulose that is found in nature: cellulose I, also known as native cellulose (Lange Bregado *et al.*, 2019). This polymorph can further be divided into two allomorphs cellulose Iα and Iβ (Lange Bregado *et al.,* 2019). This was initially discovered in the 1980s using ^{13}C-CP/MAS NMR (Atalla and VanderHart, 1984). The study found that both allomorphs are present in native cellulose, and the ratio depends on the source of the cellulose. Cellulose derived from bacteria and fungi have a larger amount of cellulose Iα, while higher plants consist of mostly Iβ. Studies using electron microbeam diffraction (Sugiyama *et al.*, 1991), as well as X-ray and neutron diffraction (Finkenstadt *et al.*, 1998) later revealed that cellulose Iα has a one-chain triclinic unit cell (Figure 9 a,c), while Iβ has two chains in a monoclinic unit cell (Figure 9 b,d). This means that the unit cell of cellulose Iα has three axes all with a different angle, while cellulose Iβ has three axes, two of which are perpendicular to each other and one of which has a differing angle (Hernández-Varela *et al.*, 2021). The differences between these structures can be attributed to the distinct hydrogen bonding networks of each allomorph (Hernández-Varela *et al.*, 2021).

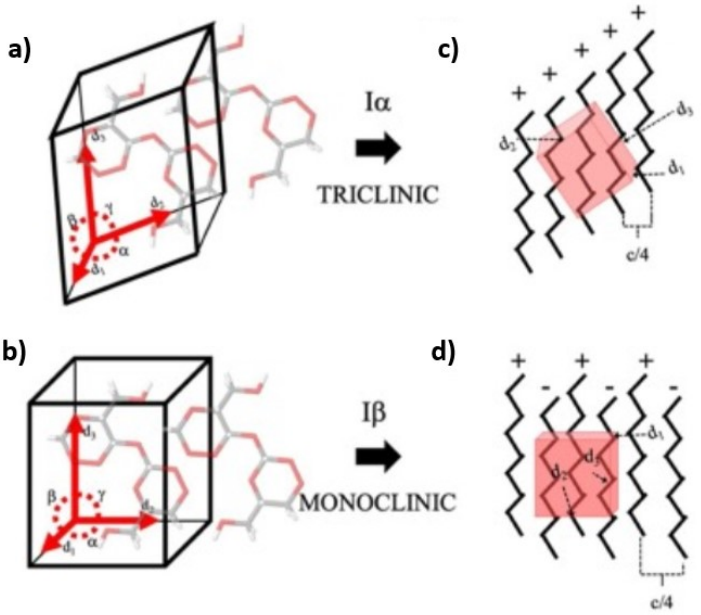

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Figure 9. Representation of the triclinic unit cell of cellulose Iα (a), and monoclinic unit cell of cellulose Iβ (b), and these unit cells represented within corresponding cellulose sheets (c, d). (Modified from Hernández-Varela et al., 2021)

Cellulose can also exist in other crystal structures as cellulose II, III, and IV, of which cellulose II is the most technically relevant (Klemm *et al.*, 2005). It is formed from cellulose I by mercerization, which is a treatment with aqueous sodium hydroxide, or by dissolution and regeneration (Klemm *et al.*, 2005), which is used in formation of man-made cellulosic fibres (Bredereck and Hermanutz, 2005). Like cellulose Iβ, cellulose II is monoclinic, but while cellulose I consists of parallel cellulose chains, the chains in cellulose II are antiparallel (Klemm *et al.*, 2005). This is due to a difference in the hydrogen bonding networks of the two polymorphs, which is demonstrated in Figure 10. As seen in Figure 10, intermolecular hydrogen bonds form between the hydroxyl groups attached to the C6 and C3 atoms in cellulose I and C6 and C2 atoms in cellulose II. Intramolecular bonds in turn form between the hydroxyl groups of C6 and C2 atoms as well as the C3 hydroxyl group and C5 oxygen atom in cellulose I. However, the structure of cellulose II only allows

one type of intramolecular bond to form between the C3 hydroxyl group and C5 oxygen atom. The different crystal structures between cellulose I and II is the reason why mechanical properties are different for native and regenerated cellulose (Bredereck and Hermanutz, 2005). For instance, the shear modulus of cellulose I fibres has been reported as 1.5 GPa and 2.5 GPa for regenerated rayon fibres (Northolt and De Vries, 1985).

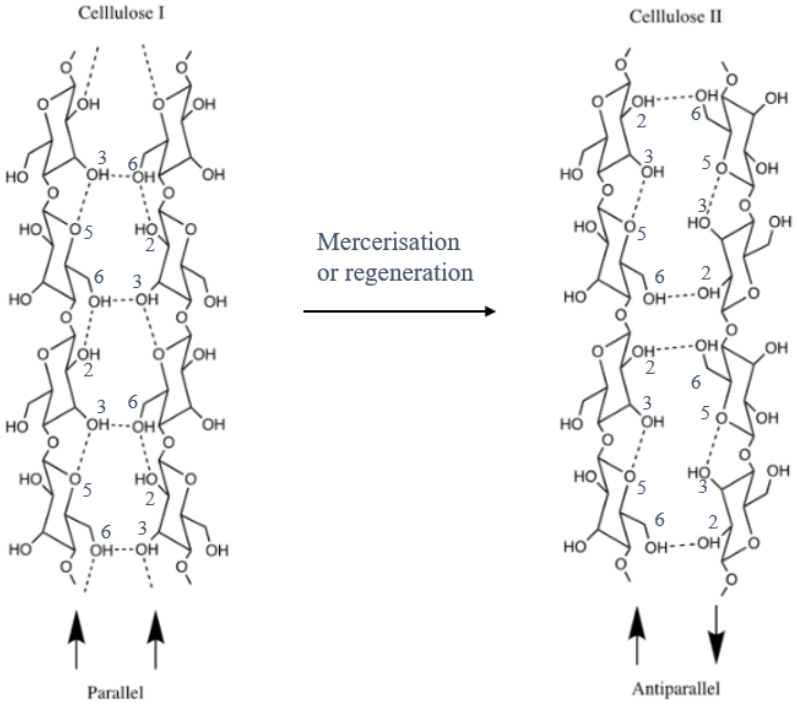

Figure 10. Structures of cellulose I and cellulose II, showing difference between their hydrogen bonding patterns and parallel/antiparallel chain orientation (Modified from Biswas et al., 2021)

2.3.2 Influence of Cellulose Properties on Regenerated Fibres

The performance and properties of MMCFs depends greatly on the properties of the cellulose fibres and pulp used. Furthermore, since the properties of cellulose fibres depend on the source of the cellulose, the origin and type of raw material used for creating regenerated cellulose fibres is of importance. For instance, the degree of polymerization (DP) affects the viscosity of cellulose solutions, thus influencing the spinnability of fibres (Bredereck and Hermanutz, 2005). A higher DP also imparts higher tenacity to fibres (Bredereck and Hermanutz, 2005). Ma *et al.* (2023) demonstrated

these effects when studying the production and properties of Ioncell fibres from pulps with varying DPs and intrinsic viscosities. The strength of the regenerated fibres was strongly dependent on the DP, with tenacity gradually decreasing with decreasing DP and viscosity. This is because a higher DP and viscosity of a pulp correlate to longer crystallites in the cellulose molecules, which also creates longer crystallites in the regenerated fibres. This means there is more internal cohesion of the cellulose molecules, thus creating stronger fibres. Nonetheless, the study showed that Ioncell fibres can successfully be spun from a wide range of intrinsic viscosities as long as they fulfil certain requirements. According to Ma *et al.* (2023), the optimum intrinsic viscosity of pulp is between 360 and 500 ml/g when using a cellulose concentration between 13-15 wt-%.

In addition to the average DP, the spinnability and mechanical properties of regenerated cellulosic fibres are also affected by molecular weight distribution (MWD). A broad MWD correlates to a high fraction of short cellulose chains, which in turn decreases the dynamic modulus of a cellulose solution (Ma *et al.*, 2023). Therefore, pulps with a narrow MWD are favourable for preparing solutions with good spinnability. Furthermore, a narrow molecular weight distribution affects the orientation of fibres in the air-gap during dry-jet wet spinning and thus facilitates the formation of a more favourable inner structure and system of voids between cellulose molecules (Schurz *et al.*, 1995). Michud *et al.* (2015) showed that a narrow MWD is also superior for producing regenerated fibres with high strength.

The inherent properties of the cellulosic material used for production of MMCFS are connected to the cellulose concentration used in the dissolution stage. According to Ma *et al.* (2023), spinning is not feasible if the dynamic modulus of a dope is higher than 6000 Pa·s, which happens at high cellulose concentration with pulps of low viscosity, or lower than 1600 Pa·s, which happens at low cellulose concentration with pulps of high viscosity. The cellulose concentration used must therefore be chosen with consideration of the intrinsic viscosity and DP of the cellulosic starting material. Furthermore,

increasing cellulose concentration in the solution used for producing MMCFs can increase fibre strength, meaning a higher concentration is preferable while still allowing good spinnability (Ma *et al.*, 2023). The improvement in strength could be due to entanglement of the molecular chains in the fibres (Kim *et al.*, 2005), which occurs due to increased intermolecular interactions that decrease the mobility of the cellulose molecules (Michud *et al.*, 2016b). This creates a strong polymer network, that leads to an increased dynamic modulus at the crossover point (Michud *et al.*, 2016b). However, if cellulose has a low DP and sufficient mobility, at a certain point further increase of cellulose concentration starts to decrease the viscosity of the solution due to spontaneous arrangement of polymer chains, leading to inferior spinning (Kim *et al.*, 2005). Therefore, cellulose solutions are commonly prepared with a cellulose concentration of 11-15% for the production of Ioncell fibres (Ma *et al.*, 2023).

Another crucial factor affecting the properties of man-made cellulosic fibres is the cellulose content in the starting material. Using pulp with high cellulose content leads to regenerated fibres with superior mechanical strength (Ma *et al.*, 2018b). Therefore, non-cellulosic impurities, such as hemicellulose and lignin, should be removed from a raw material before its use in producing MMCFs. This allows the solvent to penetrate the cellulose more effectively leading to sufficient dissolution (Ma *et al.*, 2018a). Lignin and hemicelluloses also have a lower DP and cannot orient themselves along the fibre axis as cellulose does, negatively impacting the properties of the resulting fibres (Ma *et al.*, 2018b). This effect has been demonstrated by Moriam *et al.* (2021), who showed that using a high purity pulp with a low amount of non-cellulosic impurities resulted in Ioncell fibres with increased strength and toughness. The improved cellulose content in combination with high DP and a longer cylindrical capillary geometry of the spinneret could produce fibres with a strength of 61.5 cN/tex (925 MPa) and a toughness of 83.3 MPa. The presence of non-cellulosic components is a distinct issue when using unconventional raw materials like recycled materials and non-wood pulps

since they can contain more impurities. However, these types of materials are often much cheaper than materials with higher purity and may be produced with a smaller environmental footprint. Thus, a reduction in fibre properties due to non-cellulosic impurities is sometimes acceptable if it is compensated by reduced costs and additional environmental benefits.

For some properties of cellulose, a balance must be found to optimize the properties of the regenerated fibres while simultaneously allowing sufficient dissolution. For instance, a high crystallinity affects dissolution negatively but some mechanical and physical properties of regenerated fibres positively. To achieve dissolution, a solvent must fully diffuse into the structure of a fibre (Klemm *et al*, 1998). This happens first in the amorphous domains due to their high level of disorder, and then slowly in the crystalline domains of the fibre (Jiang *et al*, 2012; Kang *et al*., 2016). Therefore, the lower the crystallinity index of a fibre is, the faster dissolution can occur (Ghasemi *et al*., 2018). Dissolution can also be enhanced by decreasing fibre diameter, because solvent molecules can saturate thinner fibres faster, allowing more solvent molecules to be accessible (Ghasemi *et al*., 2018). In contrast, Kreze and Malej (2003) showed that a higher crystallinity in man-made cellulosic fibres can affect the mechanical and sorption properties in a positive way. When comparing Lyocell, viscose, and modal fibres, they found that Lyocell had the highest tenacity and modulus out of the fibres, and its mechanical properties were least affected by wetting. Lyocell fibres were able to keep over 90% of their tensile strength when wet, while viscose fibres retained around 60% and modal fibres only 50%. According to the study, this is because Lyocell fibres have the highest crystallinity index and orientation factor. However, other more recent studies such as the one by Javed *et al*. (2014) have shown that modal fibres retain about 60% of their tenacity when wet and viscose fibres retain only 50%. The study by Kreze and Malej (2003) also showed that water adsorption of Lyocell fibres is in between viscose and modal fibres, with viscose having the highest water sorption. Water cannot penetrate the crystalline areas of cellulose (Müller *et al*., 2000), but water

absorption can happen within the disordered regions and between cellulose fibrils or crystals (Aulin *et al.*, 2009). Thus, the amount of water absorbed must be affected by the crystallinity index of cellulose, as well as the size and shape of the gaps between the fibrils or crystals. In the study by Kreze and Malej, Lyocell fibres were shown to have similar voids in between fibres as viscose, but due to its lower crystallinity index, viscose retains more water. Although modal is less crystalline than Lyocell, the voids in between the fibres are smaller in modal, leading to a lower water retention value. A smaller length and greater orientation of the voids in between regenerated cellulosic fibres have also been suggested as causes for an increase in mechanical properties (Jiang *et al.*, 2011).

2.3.3 Straw Pulp for Man-Made Cellulosic Fibres

The chemical composition of straw differs from wood pulp, which is a possible factor affecting the properties of regenerated cellulosic fibres made from straw. In general, wheat straw contains 11-26% lignin, 32-45% cellulose, and 20-45% hemicellulose (Zhang *et al.*, 2022). However, these values vary on the source and species of the straw as well as soil type, fertilizer, and other cultivation conditions (Zhang *et al.*, 2022). Typically, wood contains more lignin than straw (Zhang *et al.*, 2022). Wheat straw also contains some inorganic impurities, mainly silicon (Si), and trace amounts of minerals, such as potassium (K) and calcium (Ca) (Biricik et al., 1999), which can lead to issues with chemical recovery and damage to processing equipment (De Assis *et al.*, 2018). According to Hallac and Ragauskas (2011) the native cellulose found in wheat straw has a DP of 2660, and it can decrease to a level of 2480-1421 upon pulping, depending on the pulping method used. The average length of cellulose fibres in wheat straw fibres is 1.18 mm, and the width is 13.6 μm (Singh *et al.*, 2011). In contrast, wood derived cellulose fibres have an average length of 1–3 mm and width of 10-50 μm (Chinga-Carrasco, 2011).

Straw has been previously used to produce man-made cellulosic fibres, though the research is limited. Chen *et al.* (2015) studied the use of wheat

straw as a raw material for MMCFs using a wet spinning method and five different non-derivatizing solvents. The results of the study showed that all five solvents could successfully dissolve the straw pulp and be used for creating regenerated fibres. However, the choice of solvent affected the viscosity, cross-section, and surface morphology of the resulting fibres. The tensile strength of the fibres was between 11.5 and 15.3 cN/tex, which is not close to the values of Ioncell fibres produced from high purity wood pulp (61.5 cN/tex) (Moriam *et al.* 2021) or commercially available regenerated cellulosic fibres such as viscose (21.5 cN/tex) or Lyocell (42.9 cN/tex) (Jiang *et al.*, 2011). The authors acknowledge that the mechanical properties of the MMCFS made from straw pulp could be improved by optimising spinning conditions to mimic those of fibres with superior quality, as well as increasing the cellulose concentration. Thus, by using the Ioncell process, fibres with better performance could potentially be obtained. Furthermore, Chen *et al.* (2015) found that the strength of the fibres produced from straw pulp had a direct correlation to the molecular weight of the straw. This is in accordance with the behaviour of regenerated fibres produced from wood pulp and is another factor that could be used to enhance the performance of straw pulp based MMCFs.

Lim *et al.* (2001) studied the use of rice straw in the development of regenerated cellulose fibres using a wet spinning method and NMMO as a solvent. The resulting fibres had a tensile strength of 18.6 – 26.6 cN/tex (280-400 MPa). The results are comparable to the values of viscose (21.5 cN/tex), but significantly lower than the highest tenacities possible with the Ioncell process (61.5 cN/tex). Nonetheless, this indicates that it is possible to use straw to produce man-made cellulosic fibres with tenacities in the range of commercially available MMCFs.

Ionic liquids have also been used to produce man-made cellulosic fibres from straw pulp. Protz *et al.* (2021) demonstrated the successful dissolution of wheat straw using dry jet-wet spinning and the ionic liquid 1-butyl-3-methylimidazolium cyclohexyl carboxylate ([BMIM][Chc]). In the study,

bicomponent cellulose-lignin fibres were produced. The solutions prepared from wheat straw had an acceptable spinning stability and were able to reach a draw ratio of 5.7. The solvent [BMIM][Chc] was able to dissolve wheat straw, but the dissolution time was three times as long as for dissolving pulp of the same concentration, and only 68% of the wheat straw was dissolved, while the value was 95% for dissolving pulp. The fibres using wheat straw as a raw material reached a tenacity of 20.5 cN/tex, which is comparable with viscose fibres (21.5 cN/tex), but twice as low as fibres produced from regular dissolving pulp in the same study (40.4 cN/tex). The fibres had a rather low elongation at break (3%) but an acceptable modulus (1200-1400 cN/tex). This study shows that straw pulp can successfully be used to produce regenerated fibres using ionic liquids and the dry-jet wet spinning process, but conditions such as dissolution time may need to be adjusted in comparison to methods used for dissolving wood-derived pulp. The mechanical properties still have potential for improvement by applying the Ioncell process and optimizing the spinning conditions and composition of the starting material.

2.3.4 Recycled Nonwovens for Man-made Cellulosic Fibres

Recycling with the Ioncell process has been studied for multiple different types of textiles and materials (Ma *et al.*, 2016; Ma *et al.*, 2018a; Asaadi *et al.*, 2016; Sederholm 2022, Haslinger *et al.*, 2019a). These studies have found the Ioncell process to be a promising technology for recycling various cellulosic materials including cotton waste, man-made cellulosic fibres, wastepaper, and newsprint. In general, the DP of cellulose molecules in textile fibres decreases upon long-time use and laundering due to mechanical and chemical degradation (Palme *et al.*, 2014). As described earlier, a decrease in DP correlates with diminished mechanical properties, as well as a lower intrinsic viscosity, which directly affects spinnability. Therefore, fibres made from recycled textiles are often weaker than ones made from virgin material and the spinning can be more difficult. As nonwovens are often used in applications where high strength is not needed, the decrease in

strength due to recycling might not be detrimental. However, the binders and other additives found in nonwovens can pose an issue, since in the way of other impurities, they can hinder the dissolution of cellulose and interrupt the interactions of cellulose molecules, making the resulting fibres weaker (Ma *et al.*, 2023).

A study by Wedin *et al.* (2019) showed that used textiles made from man-made cellulosic fibres had a significantly lower DP and an inferior polydispersity index (PDI) than used cotton fabrics. This is partly because cotton has a higher starting DP than MMCFs. The study concluded that cotton fabrics are therefore more suitable for recycling with Lyocell-type processes than MMCFs. Furthermore, the studies conducted by Sederholm (2022) demonstrated that Ioncell fibres display decreased tenacity and elongation after closed-loop recycling with the Ioncell process due to a decrease in DP and crystallinity. However, the recycled fibres still exhibited superior tenacity in comparison to cotton, Lyocell, and viscose fibres as well as higher elongation than cotton. Therefore, fibres produced with the Ioncell process from recycled MMCFs may not be suitable for infinite recycling on their own but could possibly be recycled infinitely with additions of virgin wood pulp or other materials like high DP cotton waste to compensate for the reduction of DP in the substrate. Furthermore, recycled MMFCs are suitable for production of new textiles, especially for applications that do not require very high strength.

Furthermore, Wedin *et al.* (2019) found that textiles from the home consumer sector were more suitable for recycling than ones from the service sector. This is because industrial scale laundering causes a more significant decrease in the DP of cellulosic fibres than domestic laundering, which decreases the strength of resulting regenerated fibres. Many nonwovens are single-use products or products not designed to be washed regularly. Thus, the DP of cellulose fibres in nonwovens might not be affected as significantly by repeated laundering, making recycling easier.

3 Research Materials and Methods

3.1 Materials

Standard dopes were made from Enocell 5 provided by Stora Enso. The properties of the material are shown in Table 1.

Table 1. Properties of the Enocell pulp provided by Stora Enso as informed by the manufacturer.

Property	Value
Cellulose content	92.3%
Hemicellulose content	7.3%
Lignin content	0.4%
Intrinsic viscosity	434 ml/g
M_n	50.5 kDa
M_w	141.5 kDa

Biolace® 100% Lyocell nonwoven was provided by Suominen. The properties of the material can be seen in Table 2.

Table 2. Physical properties of the nonwoven fabric provided by Suominen as informed by the manufacturer.

Physical Property	Typical Value	Test Method
Basis weight	60 g/m²	NWSP 130.1
Thickness	0.65 mm	NWSP 120.6
Tensile strength CD	110 N/5cm	NWSP 110.4
Tensile strength MD	35 N/5cm	NWSP 110.4
Elongation at break CD	15%	NWSP 110.4
Elongation at break MD	90%	NWSP 110.4

Straw pulp was provided by Fortum.

3.2 Fibre and Yarn Production

3.2.1 Ionic Liquid Preparation

The ionic liquid was prepared by adding 1,5-diazabicyclo[4.3.0]non-5-ene (DBN) into a 6 l reactor kept at 70 °C. An equimolar amount of acetic acid was added to the reactor at a rate of 6 ml/min with constant stirring. After all acetic acid had been added, the mixture was stirred for a further 30 minutes. The ionic liquid was then collected into glass bottles and stored at room temperature with the caps sealed with parafilm.

3.2.2 Preparation of Cellulosic Material

The straw pulp was dried in an oven overnight to obtain a dry matter content (DMC) of at least 94%. The DMC was measured using a Precisa moisture Analyzer XM 60.

The nonwoven fabric and straw pulp were ground using a Wiley M02 mill manufactured by the Arthur H. Thomas company. The Enocell pulp was ground using a Fritsch Cutting mill. The ground nonwoven was sieved using an Allgaier sieve.

3.2.3 Removal of Additives

After preparation of some dopes of inferior quality using untreated ground nonwoven fabric as a raw material, it was determined that some kind of additive or additives were present in the fabrics and were disrupting the dissolution process. Therefore, the unidentified substance or substances should be removed before dissolution. The substances to be removed were suspected to be some type of binder and a titanium dioxide mattifying agent. Various methods were tested for the removal of the substances, and the most promising ones were studied further.

Small samples of 1 cm x 1 cm were taken from the nonwoven fabric and placed in 100 ml bottles with different solvents to assess whether a binder would be

dissolved. The solvents tested were NaOH (3%), NaOH (5%), acetone, propanol, and hexane. The samples were stirred in the solvents for up to two hours and examined at certain time intervals. Since the chemical composition of the binder was unknown, the solvents were chosen based on suggestions described in the literature and with the aim to test a variety of different types of solvents. It is likely that the binder consists of polyacrylates or ethyl vinyl acetate since they are commonly used as binders in nonwovens (EDANA). The patent by Mitchell (1974) describes the removal of polymeric binders from nonwovens by boiling the fabrics in sodium hydroxide solutions with concentrations between 1% and 5% for periods of time between 15 minutes and two hours. The method is based on the idea that the ester groups of a polymeric binder with an ester constituent will be hydrolysed, after which the sodium ions of the NaOH will attach onto the ester groups of the remaining polymer molecule and thus form a charge on the polymer (Figure 11). This makes the polymer molecules soluble in water, removing them from the cellulose substrate into the NaOH solution. A too high NaOH concentration was to be avoided to decrease the need for chemicals as well as avoid damaging the cellulose. NaOH is known to be able to decrease the DP of textile fibres such as viscose at high temperatures (Mitchell, 1974) and dissolve cellulose and lower its crystallinity index at concentrations above 6% at low temperatures (Zhou & Zhang, 2000; Iolevich, 2001; Qi *et al.*, 2011). Acetone and propanol have also been mentioned as possible solvents for nonwoven binders (Brody & Gillespie, 2005; Matsumoto *et al.*, 1974). Finally, hexane was used to test the effects of a nonpolar solvent.

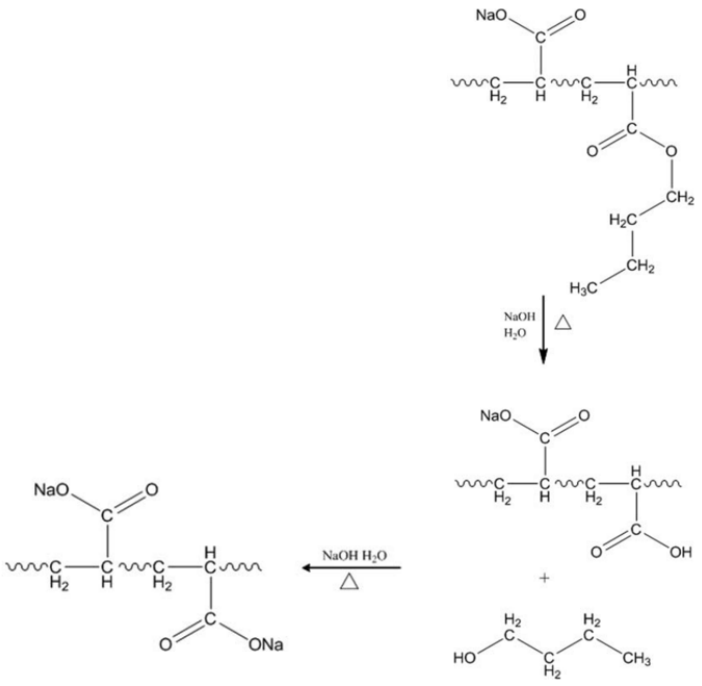

Figure 11. An example reaction showing the interactions between NaOH, water, and a polyacrylate molecule. In the first step, the water molecule hydrolyses the polyacrylate at the ester group, creating two fragments: one with an alcohol group and one with a carboxylic acid group. The Na^+-ion is then attached to the carboxylic acid group of the original polyacrylate, creating a sodium polyacrylate, which is water soluble. (Modified from Xu et al., 2015)

Based on the results of the initial tests, NaOH (3%) and NaOH (5%) were chosen for further testing. The test was repeated for both solvents with the addition of heating the solvents to 40 ˚C before adding the nonwoven fabric and keeping the heat at a range of 37 ˚C - 45 ˚C during stirring. The stirring and heating were continued for about 30 minutes. The four samples dissolved in NaOH (3%) and NaOH (5%) both unheated and heated were collected, washed with deionised water, and dried. Their ability to dissolve in ionic liquid after the NaOH treatment was tested by placing a few fibres of each sample onto microscope glass slides and placing a few drops of ([DBNH][OAc]) onto the fibres. The samples were examined using a Zeiss Axio LTS420 light transmission microscope under polarized light and a 5x magnification while heating at 60 ˚C.

For testing the effects of the NaOH treatment on dope production, spinnability, and performance of the resulting fibres, larger batches of NaOH-treated nonwoven were produced. One larger batch of NaOH treated nonwoven fabric was prepared using 12 g of ground nonwoven fabric and 2 l of 4% NaOH solution heated at 40 ˚C for 50 minutes. The ground fabric was collected, repeatedly washed with deionised water, and dried in the oven at 60 ˚C overnight. All further batches were prepared with an additional step of sieving the ground nonwoven before the NaOH-treatment. The aim of the sieving was to remove the visible round particles from the material, that were suspected to be another possible cause for inadequate dissolution of the ground nonwoven in ionic liquid. After sieving, one large batch of NaOH-treated nonwoven was prepared in the same manner as described above. Another two batches were prepared from the sieved nonwoven, one with a longer immersion time in the 4% NaOH-solution (90 minutes), and one with increased NaOH-concentration (6%) and temperature (60 ℃).

The suspected TiO_2 was attempted to be removed by treating the ground nonwoven with sulfuric acid (H_2SO_4). Hot H_2SO_4 has been shown to be able to dissolve titanium dioxide by forming sulphate salts $Ti(SO_4)_2$ and $TiOSO_4$ (Dubenko *et al.*, 2020; EU Directive 95/45/EC, 1995). However, a mild concentration must be used in the removal of TiO_2 from the cellulosic fabric to avoid cellulose hydrolysis (Xing *et al.*, 2018). The treatment was performed on ground nonwoven that had first been sieved and treated with 4% NaOH at 40 ˚C for 50 minutes. This initial step is necessary to first remove the binder from the fabric, loosening the fibres and fabric structure and thus allowing the TiO_2 particles to be released from the fabric. 800 ml of 0.01 M H_2SO_4 was heated to 40 ˚C in a 1 l beaker with magnetic stirring, after which around 5 g of the nonwoven was added. The heating and stirring were kept on for 40 minutes. The treated nonwoven was filtered from the acid and neutralised by immersing the nonwoven in deionised water and adding 4% NaOH-solution until the pH reached 8. The nonwoven was stirred in the alkaline solution for 10 minutes and the neutralisation process was repeated.

Finally, the material was washed repeatedly with deionised water and dried overnight in an oven.

The effects of using a more concentrated H_2SO_4 at higher temperatures was also tested. 2 g of ground and sieved nonwoven first treated with 4% NaOH and 0.01 M H_2SO_4 was stirred in 200 ml of 0.05 M H_2SO_4 heated to 60 ˚C for 10 minutes. The neutralization and washing steps were repeated as previously stated. The effects of the treatment were tested by starting the dissolution process of the treated nonwoven in ionic liquid as described in dope preparation to see if the white colour of the TiO_2 was still present. The dissolution attempt was terminated as soon as sufficient results of the colour could be observed and the dope was discarded. Additionally, the intrinsic viscosity was measured as described in the section 3.3.2 “Intrinsic Viscosity”, to ensure that the harsher H_2SO_4-treatment had not significantly damaged the cellulose and the viscosity had not been drastically decreased. Based on the results of these experiments, the treatment using 0.05 M H_2SO_4 heated to 60 ˚C with a treatment time of 10 minutes was chosen as the preferred treatment, and further experiments were conducted using these parameters.

For production of larger dopes for a larger fibre spinning unit, larger batches of pretreated nonwoven had to be prepared. These treatments were performed in a 5 l reactor with 150 g of ground and sieved nonwoven. 4% NaOH was first heated to 40 ˚C, after which the nonwoven was added in small increments. After all nonwoven pulp had been added, the mixture was stirred at ~350 rpm for 50 minutes. The contents of the reactor were emptied, and the nonwoven was collected within a filtration cloth. The excess liquid was removed by centrifuging, and the nonwoven was beaten with a Metos Karhu 20 mixer to separate larger clumps. The following steps were washing with deionised water, reacting with 0.05 M H_2SO_4, washing twice in deionised water set to pH 8 with NaOH, and finally washing with deionised water. The nonwoven was mixed for 10 minutes during all steps and the temperature was kept at 60 ˚C. Filtration, centrifugation and beating were repeated in between each step. Finally, the nonwoven was dried overnight in

an oven kept at 50 ˚C. The described procedure was repeated without the steps involving H_2SO_4 and altering the pH, to obtain a large batch of ground nonwoven treated only with 4% NaOH.

3.2.4 Dope Preparation

The dopes were prepared following the methods of Sixta *et al.* (2015) and Hauru *et al.* (2014). First, the IL was melted in a water bath set to 75 ˚C. The IL was then poured into a kneading device, after which the cellulosic material was added in the required amount to reach the desired cellulose concentration (10-13%). The components were manually mixed and then left to stir in the kneader at a speed of 30 rpm, temperature of 80 or 85 ˚C, and pressure of 50 mbar. The duration of mixing was varied depending on how long the specific material took to dissolve. The dissolution was monitored visually by opening the kneading device at appropriate intervals and checking to see if there were undissolved particles visible. The opening of the device was also used to stir the mixture manually and push it down from the sides of the device to ensure even mixing. After sufficient dissolution, a small sample was taken for microscopy analysis, and the rest of the mixture was transferred to a hydraulic press for filtration. The device was heated up to 90 ˚C and the dope was pressed through a filter of 6 µm in the case of the dopes prepared from standard Enocell and straw pulp, and 30 µm in the case of recycled nonwovens. The dope prepared from straw pulp at 13% cellulose concentration was filtered through a 30 µm filter due to a shortage of the 6 µm filters. Another small microscopy sample was taken after filtration, as well as a sample for rheology measurements. The rest of the dope was wrapped in plastic film and parafilm and stored at 5 ˚C. Some of the dopes were prepared using a small version of the equipment, and some on a larger unit, depending on whether the fibre spinning would be done on a monofilament or multifilament unit. The details of the preparation of each dope are summarized in Table 3.

Table 3. Kneading parameters for prepared dopes: starting material, dry matter content DMC (%), size of unit used, cellulose concentration c (%), mass of ionic liquid m_{IL} (g) and dry cellulose $m_{cellulose}$ (g), and mixing time t (min).

Starting Material	DMC (%)	Unit	c (%)	m_{IL} (g)	$m_{cellulose}$ (g)	t (min)
Enocell 5	94.39	Small	13	29.9	4.5	120
Nonwoven	94.45	Small	10	23.8	2.6	120
Nonwoven	94.45	Small	13	31.4	4.7	200
Nonwoven	94.45	Small	10	35.3	3.9	120
Nonwoven, not sieved, 4% NaOH-treated 40 °C 50 min	98.8	Small	13	27.5	4.1	160
Nonwoven, 4% NaOH-treated 40°C 50 min	99.03	Small	13	25.3	3.8	200
Nonwoven, 4% NaOH-treated 40°C 90 min	98.71	Small	13	26.1	3.9	170
Nonwoven, 6% NaOH-treated 60 °C 60 min	98.82	Small	13	27.4	4.1	100

Nonwoven, 4% NaOH-treated 40°C 50 min & H2SO4-treated 0.01 M 10 min	98.04	Small	13	14.9	2.2	190
Nonwoven, 4% NaOH-treated 40°C 50 min & H2SO4 0.01 M 10 min	98.83	Small	13	26.1	3.9	100
Nonwoven, 4% NaOH-treated 40°C 50 min & H2SO4 0.05 M 10 min	95.7	Big	12	1262	172.1	150
Straw pulp	95.7	Big	12	1262	172.1	150
Straw pulp	93.42	Big	13	1309	195.7	140
Nonwoven, 4% NaOH-treated 40°C 50 min	94.38	Big	12	1304	194.8	180

3.2.5 Fibre Spinning

The dopes were spun into fibres following the dry jet-wet spinning method of the Ioncell process described by Sixta *et al.* (2015) and Hauru *et al.* (2014). Some of the dopes were spun on a monofilament unit (Ks15) and some on a multifilament unit (KS80), both produced by Fourné Maschinenbau GmbH, but the principles are the same for both. Before spinning with the small unit, the dope was placed inside a spinning cylinder, which was then placed in an oven set to 75 °C for 30 minutes to melt the dope. For spinning with the large

unit, the dopes were melted by placing heating jackets around the cylinder and heating until the dope reached about 75 ˚C. The cylinder was then attached to the spinning device, temperature and pressure sensors were attached and a heating jacket was added also for the smaller unit. The cylinder was then set to heat at the appropriate spinning temperature depending on the dope used. The dope was then extruded from the spinneret into a coagulation bath of deionised water through an air gap of approximately 1 cm. The temperature of the coagulation bath was 5 ˚C for the monofilament unit and 9-12 ˚C for the multifilament unit. The filaments were guided from the coagulation bath onto a godet pair via a roller. The extrusion speed and speed of the godet pair were adjusted to obtain a desired draw ratio (DR). The DR was calculated using formula (1),

$$DR = \frac{V_{\text{godet}}}{V_{\text{extrusion}}} \quad (1)$$

where DR is the draw ratio, V_{godet} is the speed of the godets (m/min), and $V_{\text{extrusion}}$ is the speed of extrusion (m/min).

The fibres were collected at different draw ratios and cut into 10 cm pieces for fibre testing, 1 cm pieces for production of nonwoven fabric, or 4 cm to produce yarn. Fibres from the small spinning unit were covered in 80 ˚C deionised water and kept at least overnight to remove excess ionic liquid, and then dried at room temperature overnight. Fibres from the big spinning unit were kept in deionised water overnight, then washed with deionised water at up to 67 ℃ for two hours and dried at room temperature for at least one day.

3.2.6 Production of Nonwovens and Textiles

Both fibre types produced from recycled nonwovens with the multifilament unit and fibres produced from straw pulp at 12% cellulose concentration were used for production of new airlaid nonwovens by VTT Technical Research Centre of Finland. The parameters used in the production can be found in the Appendix in Table 17. All fibres were produced at DR 8 and cut to 1 cm length.

The fibres were first coated with spin-finishing containing Afilan CVS and Leomin PN at an 80:20 ratio at a 1:20 liquor ratio to dry fibres. Deionised water was first heated to 50 °C, after which Leomin PN was added and stirred until dissolved, followed by the addition of Afilan CVS and the fibres. This was stirred for five minutes, after which the fibres were removed, and the liquid was pressed out to obtain a 300% pick-up of liquid. The fibres were then set out to dry in room temperature for two days.

Some of the fibres produced from recycled nonwovens pre-treated with NaOH (4%, 40 °C, 50 minutes) and H_2SO_4 (0.05 M, 60 °C, 10 minutes) were also used to produce yarn that was further used for producing a textile sample. Staple fibres of 4 cm spun at a draw ratio of 11 were used. The fibres were first coated with spin-finishing as described above, and conditioned overnight in a room with 65% humidity and at 20 °C. The fibres were then loosened and separated from each other using a Mesdan s.p.a. 281c Trash Analyzer that also removed larger impurities. Next, the fibres were carded in batches of roughly 30 g with a Mesdan s.p.a. Laboratory Carding Machine 337A. Each carded batch was folded into slivers and fed into a Mesdan s.p.a. Stiro Roving Lab 3371 for drawing. Two of the drawn slivers were then combined and drawn again together, and the final sliver was folded from end to end and drawn again. These pre yarns were spun into yarn with a SER.MA. TES laboratory yarn spinning device to a target linear density of 25 tex and coned with a Simet SE.2 spooling machine. Finally, the yarn was used to produce a small sample of knitted fabric using a Mesdan Lab Knitter 294E.

3.3 Characterization

3.3.1 Dope Rheology and Dissolution

Rheology measurements and polarized microscopy were used to evaluate how well the dissolution of cellulose occurred in the dope preparation and how suitable the dopes were for spinning. The rheology measurements were conducted using an Anton Paar MCR302 rheometer with a plate diameter of

25 mm and 1 mm gap size. A frequency sweep test was applied to each sample at 5 ˚C temperature intervals starting from 50 ˚C up to 90 ˚C.

The dopes were examined with a Zeiss Axio LTS420 light transmission microscope under polarized light and a 5x magnification. One sample was collected before filtration and another one after. If the samples were not examined right after preparation, they were refrigerated and then heated up to 60 ˚C using the heating stage of the microscope. The samples were examined for any fibres or particles that might have been left undissolved during the dope preparation.

3.3.2 Intrinsic Viscosity

The intrinsic viscosities of the starting materials and produced fibres were measured following the SCAN-CM 15:99 standard procedure. The measurements were also completed on the nonwoven fabric after various pre-treatments to see the treatment's effects on viscosity. 200-250 mg (dry mass) of each sample was measured into a plastic bottle along with three small copper rods and 25 ml of deionised water. The bottles were shaken with a VWR Standard Analog Shaker for 30-60 minutes. Then, 25 ml of cupri-ethylenediamine-solution (CED) was added to each bottle, and they were shaken for another 30 minutes. The nonwoven did not appear to be fully dissolved after this time, so another 15 minutes was added to the shaking time. Next, the samples were run through a capillary that was kept at a standard temperature with a water bath of 25 ˚C. The time it took for the sample to travel between two fixed points in the capillary was measured. This was used for calculating the relative viscosity using formula 2,

$$\eta_{\mathrm{rel}} = h \cdot t \qquad (2)$$

where η_{rel} is the relative viscosity, h is the viscometer constant, and t is the time it takes for the sample to travel between two points (s).

The relative viscosity was then used to determine the corresponding limiting viscosity from a standard table. This could then be used to calculate the intrinsic viscosity with formula 3,

$$\eta = \frac{\eta[c]}{c} \qquad (3)$$

where η is the intrinsic viscosity (ml/g), $\eta[c]$ is the limiting viscosity, and c is the cellulose concentration (g/ml).

Two samples were prepared per material and two to three measurements were recorded per sample. Samples prepared from nonwovens treated with sulfuric acid had only one sample. The average of these measurements was taken to obtain the average intrinsic viscosity of each material. This value was also used to calculate the DP of the material using formula 4,

$$\mathrm{DP} = \eta \cdot 2.381 \qquad (4)$$

where DP is degree of polymerization and η is intrinsic viscosity.

3.3.3 Molecular Weight Distribution

Gel permeation chromatography (GPC) was used to analyse the molecular weight distribution of the starting materials. The samples were prepared for GPC following the procedures described by Sixta *et al.* (2012). 50 mg of each sample was measured into a 6 ml polypropylene tube with a 20 µm PE-frit filter. 4 ml of deionised water was added to each sample, and they were left covered overnight to activate the cellulose. The following day, the water was removed using a vacuum suction. To remove any leftover water, 2 ml of acetone was added to the samples and immediately removed. Another 4 ml of acetone was added to each sample and left for about five hours. The tubes were then emptied by vacuum and filled with 4 ml of dimethylacetamide (DMAc) and left to activate for three days. Next, the tubes were emptied again by vacuum and the residues were transferred to glass bottles. 5 ml of LiCl/DMAc was added to each bottle to dissolve the cellulose overnight with magnetic stirring. The following day the samples were diluted by combining

0.5 ml of each solution with 4.5 ml of pure DMAc, giving a sample concentration of 1 mg/ml. A small amount of each diluted sample was then transferred into a vial through a 0.2 μm syringe filter. A duplicate was produced for each vial. The samples were then analysed using a Dionex Ultimate 3000 HPLC by Thermo Scientific.

3.3.4 Carbohydrate and Lignin Content

Monosaccharides and lignin in the starting materials were analysed and quantified according to the standard NREL/TP-510-42618 (Sluiter *et al.*, 2008). The measurements were conducted in duplicate. 300 mg of each sample was weighed into a 250 ml bottle and 3 ml of 72% sulfuric acid was added. The material was mixed with the acid with a glass rod, and the bottles were placed along with the rods into a water bath of 30˚C for one hour. The samples were occasionally stirred manually to aid dissolution. Next, 84 ml of deionised water was added to each bottle while simultaneously using the water to wash all remaining sample from the glass stirring rods into the bottles. A sugar recovery standard (SRS) was prepared to determine the amount of sugars lost in the hydrolysis process and used to correct the results of the samples. The SRS was prepared in a way that resembles the amount of sugars found in the tested samples, by making 1000 ml of solution containing 1000 mg glucose, 500 mg xylose, 200 mg arabinose, 200 mg galactose, 200 mg mannose, and 100 mg rhamnose in deionised water. The samples and SRS were then placed in a Systec DE 23 autoclave for two hours on the "hydrolysis" setting and then cooled down for one hour.

The acid insoluble lignin was determined by vacuum filtering the hydrolysed samples through previously weighed VitraPOR Borosilicate 3.3 crucibles and washing with deionised water. A small sample of each filtrate was collected for determining acid soluble lignin and carbohydrates. The crucibles and residues were dried in an oven at 105 ℃ for two hours, then cooled down for at least one hour in a desiccator, after which their weights were recorded. The

amount of acid insoluble lignin (AIL) was calculated assuming that all residue is lignin, using formula 5,

$$AIL = \frac{m_{\text{crucible+AIL}} - m_{\text{crucible}}}{ODM_{\text{sample}}} \cdot 100\% \qquad (5)$$

in which AIL is the percentage of acid insoluble lignin (%), $m_{\text{crucible+AIL}}$ is the combined mass of the crucible and AIL after drying and cooling (g), m_{crucible} is the mass of the crucible (g), and ODM_{sample} is the oven dry mass of the sample in the beginning (g).

The acid soluble lignin was determined using a Shimadzu UV-2550 UV-visible spectrophotometer. A wavelength of 205 was used for analysing the samples. The previously collected filtrates were diluted with deionised water to appropriate concentrations in order to obtain an absorbance between 0,7-1,0 at wavelength 205. All samples were analysed in duplicate. The amount of acid soluble lignin (ASL) was calculated with formula 6

$$ASL = \frac{UV_{\text{abs}} \cdot V_{\text{solution}} \cdot Dilution}{\varepsilon \cdot ODM_{\text{sample}} \cdot l} \qquad (6)$$

, in which ASL is the percentage of acid soluble lignin (%), UV_{abs} is the average absorbance of the samples measured at appropriate wavelength , V_{solution} is the volume of the solution before hydrolysis (ml), $Dilution$ is the dilution factor used for preparing the samples for UV spectrophotometry, ε is the absorptivity of biomass at a specific wavelength (110 was used), ODM_{sample} is the oven dry mass of the sample in the beginning (g), and l is the pathlength of the UV-vis cell (cm).

Due to the nature of the measurements used to determine lignin content, lignin cannot be distinguished from other non-saccharide components. Therefore, the results do not necessarily represent an accurate lignin content, but could include other compounds as well, especially in the case of the recycled nonwoven since the fabric contains multiple additives. Thus, the results are presented as non-saccharides and not as lignin.

The carbohydrate content of each sample was determined with high performance liquid chromatography (HPLC). The filtrates of each sample were diluted with deionised water to obtain a ratio of 1:100 for Enocell and straw pulp, and 1:50 for the recycled nonwoven and SRS. Later, due to inferior results, another sample was prepared of the nonwoven sample with a ratio of 1:100 as well. A small amount of the diluted samples was transferred to autosampler vials through a 0.2 µm filter. Two vials were filled for each sample. The HPLC results were used to calculate the amounts of different monosaccharides in each sample utilising Janson calculations (Janson, 1970).

3.3.5 Fourier Transform Infrared Spectroscopy (FTIR)

The chemical composition of the starting materials and the produced fibres were characterized using Fourier Transform Infrared Spectroscopy (FTIR). The measurements were conducted using a Spectrum Two FTIR Spectrometer by PerkinElmer from wavenumber 400 to 4000 cm^{-1}.

3.3.6 Ash content

The ash content of the starting materials and produced fibres were measured by placing about 200-400 mg of each sample in a Selecta Horn muffle furnace at 575 ˚C for four hours. During the heating process, inorganic minerals are not destroyed, while water and volatile compounds found in biomass are vaporized, and organic substances are burned (Liu, 2019). Thus, the residue left after heating represents the amount of inorganic minerals present in a sample (Liu, 2019). The masses of the samples were measured before and after heating, and this information was used to calculate the ash content using formula 7,

$$\mathrm{AC} = \frac{m_\mathrm{f}}{m_\mathrm{i}} \cdot 100\% \qquad (7)$$

, where AC stands for ash content, m_f is the final weight of the sample after heating, and m_i is the initial dry weight of the sample before heating.

3.3.7 Elemental Analysis

The elemental composition of each starting material and the produced fibres were analysed with a Thermo Scientific Thermo Flash Smart CHNSO Elemental Analyser. The percentage of carbon, nitrogen, and hydrogen were evaluated, and the samples were analysed in triplicate.

3.3.8 Simultaneous Thermal Analysis (STA)

The amount of inorganic matter and thermal behaviour of the starting materials and produced fibres were evaluated with thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC). The measurements were conducted with a Netzsch STA 449 F3 Jupiter and QMS 403 Aëlos Quadro thermal analyzer. Around 5 mg of each sample was placed in a crucible into the device that was heated from 40 ˚C to 900 ˚C with a heating rate of 20 K/min. A helium atmosphere was used with a gas flow of 70 ml/min.

3.3.9 Fibre & Yarn Properties

The performance of the different produced regenerated fibres was evaluated using a Textechno Herbert Stein Favigraph. The tenacity, elongation at break, and linear density of 20 fibres were measured for each sample in both dry and wet conditions. The fibres were conditioned overnight at relative air humidity of 65% and temperature of 20 ˚C prior to measurement. The birefringence of the fibres was then tested by measuring the linear density of three fibres per sample at three different locations of the single fibre. The fibres were then attached to microscope slides keeping them taut and straight and examining them using a Zeiss Axio Scope A1 transmitted light polarization microscope with a tilting compensator. The tilting angles of the fibres were recorded from three different locations on each fibre and used for calculating the total orientation of the fibres.

The properties of the produced yarn were measured using a MTS 400 tensile and compression testing machine after conditioning the yarn at a humidity

of 65% and temperature of 20 °C overnight. 30 samples of yarn were measured and used for calculating average values for tenacity and elongation at break. A load cell of 50 N was used with a gauge length of 250 mm.

3.3.10 Scanning Electron Microscopy (SEM)

The cross-section and surface of the produced fibres was examined using a Zeiss Sigma VP scanning electron microscope (SEM). For preparation for the SEM, a small bundle of the fibres was frozen in liquid nitrogen and cut in half with scissors. The pieces were allowed to defreeze, after which they were attached to tilted SEM sample holders with double sided carbon tape. The samples were coated with an 80 Au/20 Pd mixture using a sputter coater (Quorum Technologies, Q 150 R) to impart electric conductivity to their surface. The obtained images were edited with Adobe Photoshop Express Photo Editor to enhance the image quality.

3.3.11 Optical Properties

The colour and brightness of the starting materials and produced fibres were measured using a GretagMacbeth Spectroscan 16130-818. A bundle of the examined material was held against a white background and scanned multiple times at different locations until a sufficiently small standard deviation was obtained. Values with a significant divergence from the average were discarded.

4 Results

4.1 Dissolution and Spinnability

The recycled nonwoven fabric took more time to dissolve in ionic liquid at the same cellulose concentration than the standard Enocell pulp. The appearance of the dopes produced from untreated, ground nonwovens was also different. Standard dopes are generally translucent, while the recycled nonwoven dopes were opaque (Figure 12). This indicates the presence of solid particles in the dope, possibly from undissolved cellulose or additives found in the original fabric. This could be partly due to some white, round particles that were visible in the dopes. The microscope images of the dopes prepared from recycled nonwovens (Figure 13) showed some undissolved fibres after filtration, but not a significant amount. Therefore, it was concluded that the nonwoven fabrics likely contained something other than cellulose that was interfering with the dope preparation.

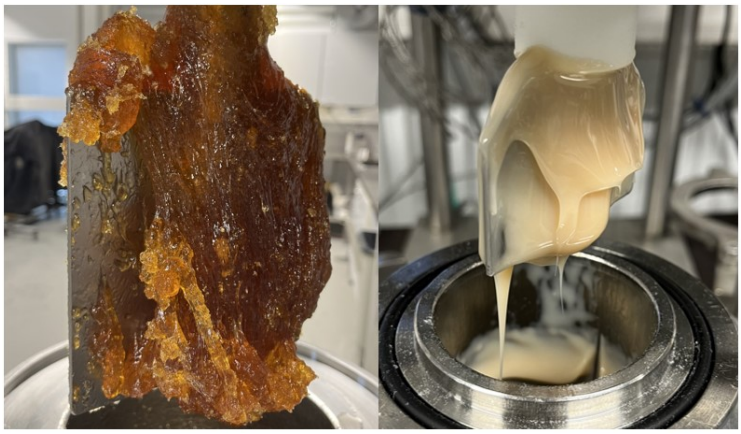

Figure 12. Typical cellulose-ionic liquid dope (left) and dope produced from recycled nonwovens (right).

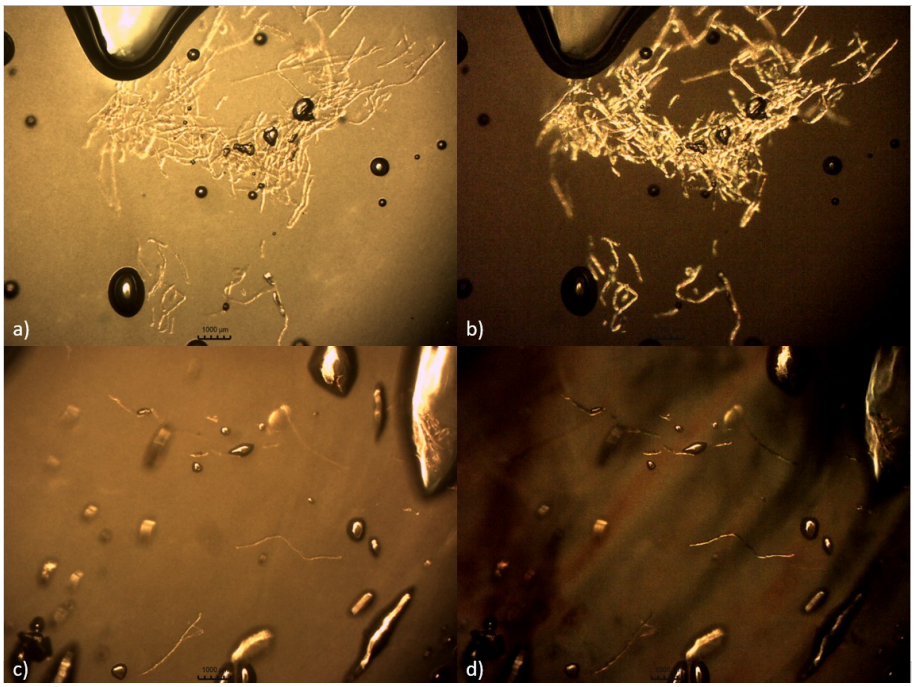

Figure 13. Microscope images of the dope produced from ground, untreated nonwoven at 13 % cellulose concentration before filtration without cross-polarization (a) and with cross-polarization (b) as well as after filtration without cross-polarization (c) and with cross- polarization (d).

Common additives found in nonwovens that could have a disrupting effect in the recycling process are binders and TiO_2. Binders are adhesive materials that are added to nonwovens to attach fibres to each other and hold them together (Chapman *et al.*, 2022). This could mean the cellulose fibres are too strongly attached to each other to easily dissolve in the IL, and the binding chemicals could interfere with the interactions of the cellulose molecules. Titanium dioxide (TiO_2) in turn is commonly used in the textile industry and added to nonwovens as a whitening and mattifying agent as well as to improve stability under ultraviolet light and add antibacterial properties (Gao *et al.*, 2020; Karbownik *et al.*, 2009; Metanawin & Metanawin, 2022; Verleysen *et al.*, 2022). Thus, a TiO_2 pigment could be the source of the opaque, milky appearance of the dopes prepared from recycled nonwovens. Just as the binder, the presence of TiO_2 could also interfere with the interactions between cellulose molecules, creating weaker fibres. However, if the produced fibres are to be used for production of new nonwovens, TiO_2 would possibly be added back in, making its removal for recycling purposes

redundant. Therefore, if spinning at a high enough draw ratio is possible and the fibre properties prove to be sufficient, it would be more economical and ecological to keep the TiO_2 in the material throughout the recycling process.

A variety of pretreatments were performed on the nonwoven fabric to remove the possible additives, and the most promising ones were used for preparation of dopes and fibre spinning. The process of determining the most optimal method for removing the binder included testing the effects of multiple solvents, the results of which are described in the upcoming section 4.2. Removal of Binder. The current section will in turn describe the results of the dope preparation and fibre spinning after the most promising treatments. The success of the removal of TiO_2 is measured by the effects on the appearance of dopes and fibre spinning, which is why the removal of this additive is discussed primarily in the current section.

The spinnability of a certain dope can be evaluated by the maximum draw ratio reached in the spinning and how long the fibres could be collected at a certain draw ratio. Ma *et al.* (2018b) have defined spinnability of Ioncell fibres in the following way: DR<2: non-spinnable, DR 2-8: poor, DR 8-14: good, DR>14: excellent. However, this categorization is not directly applicable to all kinds of dopes, since the raw material used, cellulose concentration of the dope, additives, and desired application will have an effect on which DRs are considered good. In the case of nonwovens, thicker fibres are often used and thus DR 8 is a suitable option. The values describing the spinnabilities of each dope are summarized in Table 4.

Table 4. Description of spinning results for each dope, including cellulose concentration in the dope c (%), pre-treatments performed on starting material, maximum draw ratio reached during spinning, maximum collection time at max DR (min), ideal temperature (°C), and size of the spinning unit. If the section Max Draw Ratio *is marked with "-", no collection of fibres was possible. The maximum draw ratio for fibres produced with the multifilament unit are presented as the maximum DR that allowed continuous spinning, unless stated otherwise.*

Cellulose wt-%	Raw Material	Max Draw Ratio	Collection Time (min)	Ideal Temperature (°C)	Spinning unit
10%	Nonwoven (NW)	-	-	-	Small
13%	Nonwoven (NW)	3	1,5	66	Small
13%	NW, not sieved, NaOH 4%, 40 °C, 50 min	7	1,5	63	Small
13%	NW, NaOH 4%, 40 °C, 50 min	8	1	67	Small
13%	NW, NaOH 4%, 40 °C, 90 min	-	-	-	Small
13%	NW, NaOH 6%, 60 °C, 60 min	-	-	-	Small
13%	NW, NaOH 4%, 40 °C, 50 min & H_2SO_4 0.01 M	9	1	65	Small
13%	NW, NaOH 4%, 40 °C, 50 min & H_2SO_4 0.05 M	11	Continuous	63	Big
13%	NW, NaOH 4%, 40 °C, 50 min	8	Continuous	66	Big
12%	Straw pulp	8	5	Not determined	Big
13%	Straw pulp	8	Continuous	87	Big

Two batches of dopes with 10% concentration were made, the first of which had issues in the filtration stage leading to improper filtration and loss of

material. The second batch of 10% concentration dope was used for fibre spinning, but the filaments were too weak for collection although they were able to reach the godet couple. Therefore, all further dopes prepared from recycled nonwoven were produced with a cellulose concentration of 13%. Increasing the cellulose concentration to 13% made the dope spinnable, but the maximum DR was still only 3 with a short collection time.

The dopes prepared from nonwoven fabric after various NaOH- and H_2SO_4-treatments were generally dissolved faster than the dopes produced from untreated recycled nonwoven. This could be due to the removal of the binder, which would make the fibres of the nonwoven looser and allow the IL to penetrate the structure more easily. However, the colour of the dopes was still an opaque light orange, indicating that neither treatment was able to remove all solid matter that is insoluble in the ionic liquid. Nonetheless, the effects of the treatments can be seen in the improvement of the spinnability of the dopes. As seen in Table 4, the maximum draw ratio increased from 3 to 7 after treatment with 4% NaOH at 40 °C for 50 minutes and further to 8 after adding a sieving step to the process. Based on these results, larger particles were sieved from the ground nonwoven before preparation of all further dopes. Harsher NaOH-treatments with a longer reaction time or higher NaOH-concentration and temperature appeared to make the dopes non-spinnable.

The potentially present TiO_2 pigment was attempted to be removed by a H_2SO_4-treatment after initial treatment with NaOH. Although the appearance of the dope did not change after treatment with 0.01 M H_2SO_4, the fibre spinning was improved and a DR of 9 was reached for about 30 seconds and spinning with DR 8 was possible for a longer time of seven minutes. This indicates that the TiO_2 was likely partially removed by the treatment but not completely. Therefore, a harsher version of the treatment was performed with 0.05 M H_2SO_4 at 60 °C for 10 minutes, but this treatment was still not able to change the appearance of the dope. However, since the intrinsic viscosity of the nonwoven treated with 0.05 M H_2SO_4 showed no

signs of significant cellulose degradation, and the harsher treatment is more likely to remove a larger portion of TiO_2, this treatment was selected for further examination.

Two different formulations of dopes made from recycled nonwovens were prepared for spinning with a larger unit. One was similar to the dope that performed the best in the trials with the small spinning unit: ground, sieved nonwoven treated with 4% NaOH for 50 minutes and 0.05 M H_2SO_4 for 10 minutes. As seen in Table 4, the use of the large unit allowed the increase of the maximum DR up to 11 with continuous collection, when spinning the NaOH- and H_2SO_4-treated nonwoven substrate. The improvement is largely due to the nature of the larger spinning unit with a spinneret containing 400 holes in contrast to the single one in the small spinning unit, as some filaments can break during spinning while collection can still be continued. Additionally, the H_2SO_4-treatment was performed with a higher concentration than for the small spinning, which could have led to a more thorough removal of TiO_2 and thus improve the spinnability.

The other dope was prepared from ground, sieved nonwoven treated only with 4% NaOH for 50 minutes but with no H_2SO_4-treatment. Again, the spinning was improved in comparison to spinning the same formulation using a monofilament unit. A draw ratio of 11 was reached for a short period of time, and stable spinning was possible with DR 8. However, many of the filaments broke during their initial stretching as the godet speed was increased to obtain DR 8. Therefore, the yield was not very high, but it is possible that this could be improved by altering the spinning temperature. Since DR 8 is ideal for production of nonwovens, these results show that nonwovens can be recycled with the Ioncell process without removing TiO_2 for production of new nonwovens. The removal of TiO_2 with H_2SO_4 can however be added if thinner fibres are required for a certain application or if the fibres are used for production of knitted or woven textiles.

According to the categorization by Ma *et al.* (2018b), the draw ratios reached for Ioncell fibres from recycled nonwovens could be classified as good, but they cannot reach excellent values. Similarly, previous studies using recycled materials for producing Ioncell fibres have shown a varying decrease in draw ratio, with Haslinger *et al.* (2019a) reaching DR 8.8 using a recycled cotton polyester blend, Haslinger *et al.* (2019b) reaching DR 12 with dyed cotton waste, Selinger (2022) reaching DR 8-11 using recycled banknotes, Airaksinen (2023) reaching DR 14.3 for a short period and DR 8 for continuous spinning with recycled viscose fibres, and Sederholm (2022) reaching DR 11-14 with recycled Ioncell fibres derived from Enocell pulp. However, very high draw ratios have also been achieved with recycled raw materials, as demonstrated by Ma *et al.* (2016) by reaching DR 19.8 using cardboard as raw material and Ma *et al.* (2018a) reaching DR 18 with recycled newsprint. Due to the additives present in the nonwoven used in the current study, it is expected that the maximum draw ratio is lower than for standard Ioncell fibres and Ioncell fibres produced from recycled materials with higher purity.

Straw pulp was used as a raw material to produce two different dopes. One had a cellulose concentration of 12%, that is lower than is typically used, in order to compensate for the high intrinsic viscosity of the straw pulp. Another dope was produced at 13% cellulose concentration based on previously obtained test results. The dope with 12% concentration was able to reach both DR 11 and DR 8 only for short times until all filaments broke. However, this spinning trial suffered from technical issues. The dope was leaking from an untight spinning cylinder, affecting both the pressure and extrusion velocity of the dope and, thus, interfering with the spinning process. Furthermore, based on the rheology data and the appearance of small air bubbles in the filaments in the air gap, the temperature was too low for ideal spinning. Increasing the cellulose concentration and using a higher temperature during spinning allowed continuous spinning at DR 8, but adequate spinning was still not reached at DR 11. Nonetheless, because nonwovens require slightly

thicker fibres than other textiles, DR 8 is acceptable. Moreover, this is a distinct improvement to previous reports on using dry-jet wet spinning to produce fibres from straw. Protz *et al.* (2021) report a maximum draw ratio of 5.7 for solutions spun from wheat straw using the ionic liquid [BMIM][Chc] as a solvent.

The spinnability of a dope is strongly dependent on its visco-elastic properties. To allow extrusion through the spinneret holes, a cellulose solution must display suitable fluidity, and an appropriate visco-elasticity is required for the filaments to be able to withstand stretching in the air gap without breakage (Ma *et al.*, 2018b). The rheological properties of the dopes with best spinning performance are demonstrated in Figures 14-17 and Table 5. As seen in Figure 14, the viscosities of the dopes produced from recycled nonwovens are lower than the standard Enocell dope. However, the shapes of the curves are similar. As seen in Figure 15, the curves of dopes produced from straw pulp are more linear than the curve of the standard dope. As expected, the dope produced with 13% cellulose concentration has higher complex viscosity than the dope produced with 12% concentration.

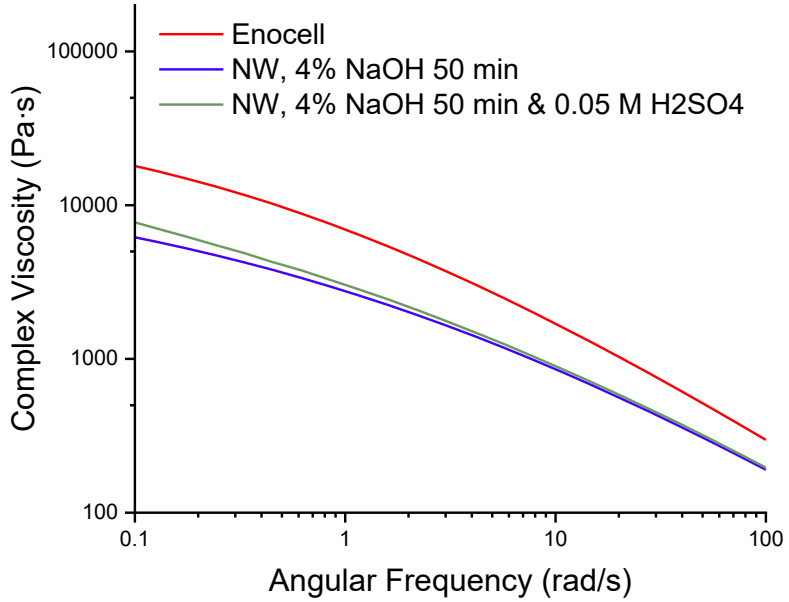

Figure 14. Double logarithmic illustration of the complex viscosity (Pa·s) vs angular frequency (rad/s) of dopes produced from standard Enocell pulp and recycled nonwovens at 65 ˚C.

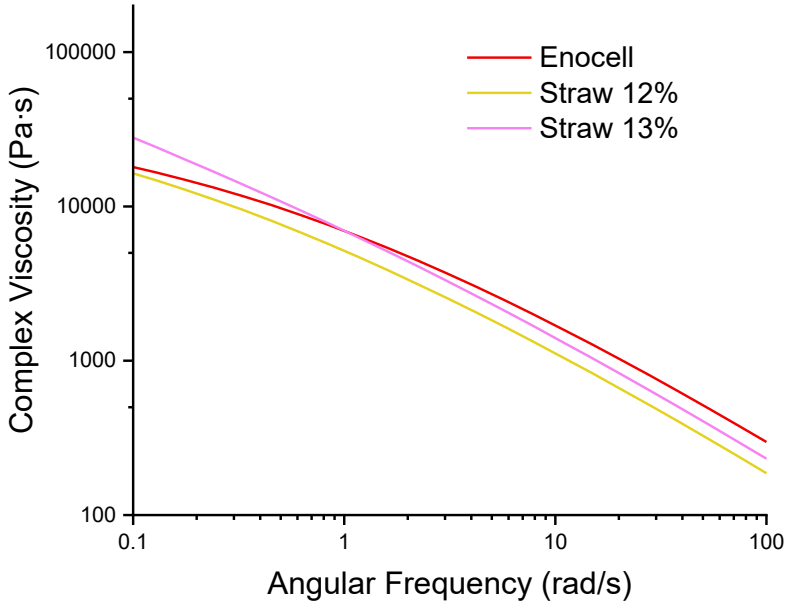

Figure 15. Double logarithmic illustration of the complex viscosity (Pa·s) vs angular frequency (rad/s) of dopes produced from standard Enocell pulp (at 65 ˚C) and straw pulp (at 80 ˚C for 12% and 85 ˚C for 13%).

Table 5 shows the zero-shear viscosity, and angular frequency and modulus at the cross-over point of the samples, while Figures 16-17 demonstrate the loss and storage moduli in relation to angular frequency. The zero shear viscosities are calculated with the Cross model from the complex viscosity vs angular frequency curves by extrapolation. The model is only applicable if the curve follows a certain shape, with a plateau at low values of angular frequency. The curve for the dope made from straw pulp at 13% cellulose concentration seems to follow a power-law dependence, meaning the extrapolation with a Cross-model is not applicable for this sample.

Ideal spinning behaviour is generally achieved when the cross-over point (COP) between the loss modulus and storage modulus occurs at around 1 rad/s or slightly higher, the dynamic modulus at the cross-over point is around 4000 Pa, and the zero-shear viscosity is between 25 000 and 35 000 Pa.s. (Sixta *et al.*, 2015). As seen in Table 5, the standard dope produced from Enocell pulp corresponds best with these values. The dopes produced from recycled nonwovens have a decreased zero shear viscosity and dynamic modulus, while their cross-over points occur at higher angular frequencies.

The dope produced from straw pulp with 13% cellulose concentration shows no cross-over point at ideal spinning temperature, while the dope with 12% cellulose concentration shows a COP far below 1 rad/s and with a relatively low modulus at the COP. Neither of these dopes have ideal rheological behaviour, although both dopes were spinnable and could reach DR 8. Further optimisation of the cellulose concentration could be applied to enhance the spinnability of straw pulp.

Table 5. Zero shear viscosity η_0 (Pa·s), angular frequency at the cross-over point ω (rad/s), and loss modulus and storage modulus at the cross-over point G'=G" (Pa) measured at the appropriate spinning temperature (°C) of each sample.

Sample	Temperature (°C)	η_0 (Pa·s)	ω (rad/s)	G'=G" (Pa)
Enocell	65	31 300	0.83	4500
NW, 4% NaOH 50 min	65	20 000	2.74	3400
NW, 4% NaOH 50 min & 0.05M H2SO4	65	12 300	1.70	2800
Straw 12%	80	37 000	0.32	2200
Straw 13%	85	na	No COP	No COP

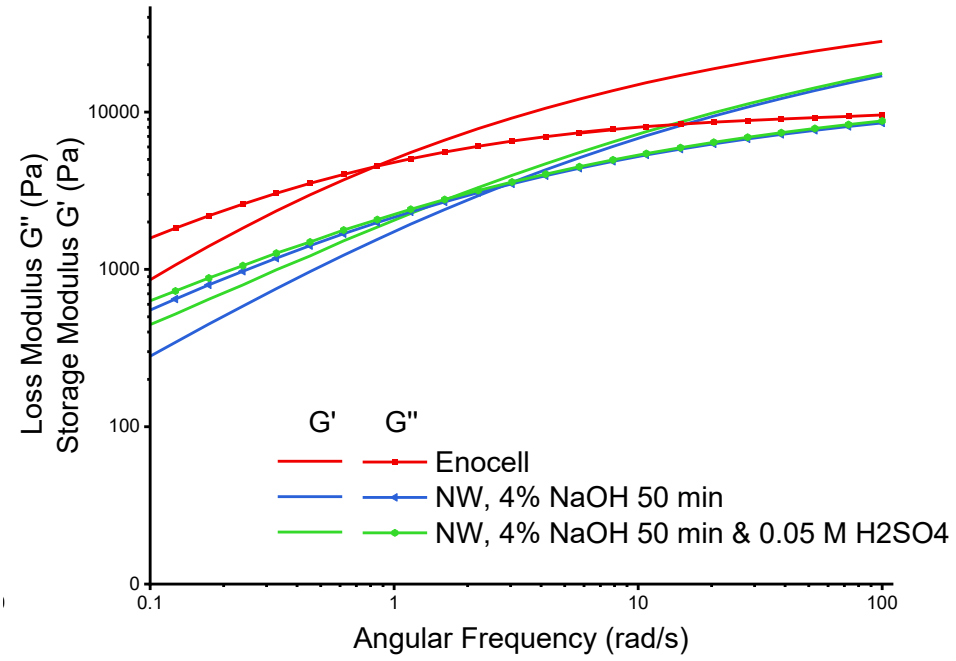

Figure 16. Loss modulus G" (Pa) and storage modulus G' (Pa) vs angular frequency (rad/s) of dopes made from standard Enocell and recycled nonwovens that were produced with a large unit measured at 65 °C.

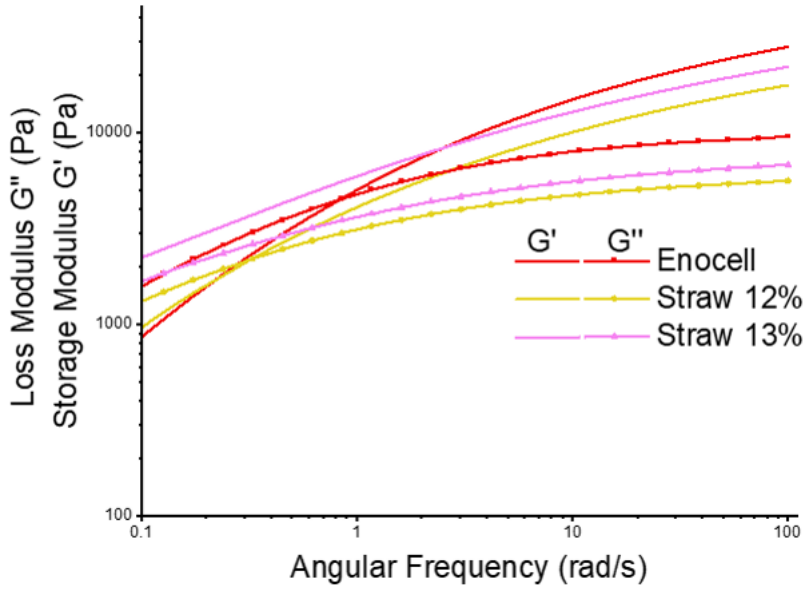

Figure 17. Loss modulus G" (Pa) and storage modulus G' (Pa) vs angular frequency (rad/s) of dopes made from straw pulp at 80 ˚C (Straw 12 %) and 85 ˚C (Straw 13 %).

The rheology data of dopes with non-ideal spinning is summarized in the Appendix in Table 15. The supplementary data in Table 15 supports the findings that dopes produced with a 10% cellulose concentration were not spinnable due to one of the dopes not having a cross-over point at all and one having a distinctly low zero shear viscosity and cross-over point at high angular frequency. The supplementary data also shows that the NaOH-treatments slightly decrease the viscosity of dopes due to cellulose degradation, and the viscosity is further reduced by sieving the ground nonwoven. The dope produced from nonwoven treated with the higher concentration NaOH shows a distinctly lower zero-shear viscosity than the other nonwoven dopes, which is likely why this treatment made the dope non-spinnable.

4.2 Removal of Binder

The initial tests examining the dissolution of the unknown binder in various solvents showed that the nonwoven did not react with acetone, propanol, or

hexane, as can be seen in Figure 18. However, the fibres of the textile samples submerged in NaOH started to loosen and untangle from each other, indicating that the binder was dissolving in NaOH and getting removed from the fabric. The sample in 5% NaOH showed signs of binder dissolution almost immediately, and the fibres kept loosening with time. 3% NaOH appeared to dissolve the binder as well, but slower than at 5% concentration. The samples tested with heating to 40 ˚C showed less of a difference between 3% NaOH and 5%, with 5% concentration appearing to dissolve the binder slower when heated than at room temperature and 3% concentration appearing to have a slightly faster dissolution. The stirring may not have been as effective during treatment with heating, because the magnetic stirrer was smaller than during treatment at room temperature, which could be a reason why the 5% NaOH-treatment was not as effective when heated. The difference in dissolution of binder by different NaOH concentrations, time intervals, and heating are compared in Figure 19.

Figure 18. a) Nonwoven samples before treatment. b) Nonwoven samples submerged in propanol (left) and hexanol (right) for 30 minutes. c) Nonwoven fabric sample submerged in acetone for 30 minutes.

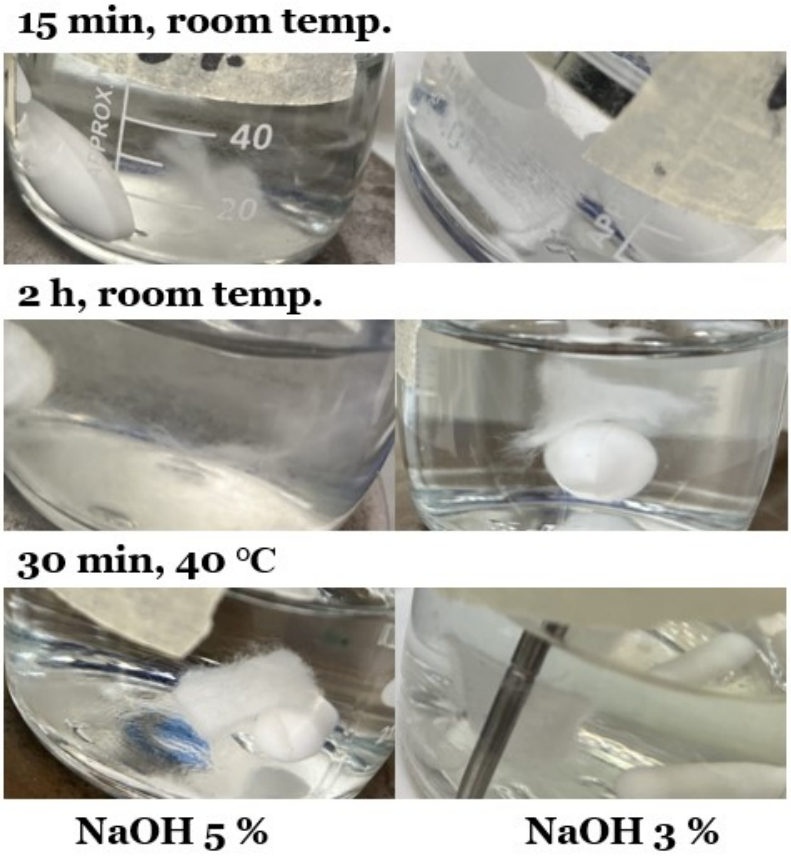

Figure 19. Tests for the removal of binder from nonwoven fabric samples in NaOH at a concentration of 5% (left) and 3% (right) after 15 minutes and 2 hours of stirring at room temperature and after 30 minutes of stirring at 40 °C.

The cellulose dissolution tests performed on the NaOH-treated fibres showed that the treatment did not make a significant difference on the ability of the ionic liquid ([DBNH] [OAc]) to dissolve the cellulosic fibres in the nonwoven fabrics. The untreated fibres and fibres treated with 3% NaOH took about seven minutes to be dissolved by the ionic liquid. However, after moving the microscope slides, there were still some individual undissolved fibres left at a different part of the examined sample for all samples. In contrast, the fibres treated with 5% NaOH solution took only six minutes if the solution had been heated but an unusually long 15 minutes if the solution had been at room temperature. The dissolution test was repeated twice more for the fibres treated with room temperature 5% NaOH, one having a dissolution time of

10 minutes, and one over 15 minutes. This could be due to NaOH-residue being left on the samples due to poor washing.

4.3 Fibre Properties

The properties of the different regenerated fibres produced in this study are summarized in Table 6 along with corresponding literature values for other regenerated cellulose fibres and synthetic fibres used in nonwovens for comparison. The properties correspond reasonably well with the results of the spinning stage, with improved spinnability generally leading to increased tenacity of the fibres. However, although using 13% cellulose concentration improved spinning with straw pulp, the tenacities show a slight decrease. Supplementary data on the properties of produced fibres that did not perform as well is found in the Appendix as Table 16.

The average tenacity and elongation at break were 39.9 cN/tex and 9.0% for the fibres produced from recycled nonwovens after pre-treatment with 4% NaOH and 0.05 M H_2SO_4 at draw ratio 8. The corresponding values for fibres produced after only the NaOH pre-treatment were 38.2 cN/tex and 8.1 %. This shows that although the partial removal of TiO_2 with H_2SO_4 improves spinnability, it does not have a significant effect on the fibre properties. Both fibre types also show a significant increase in tenacity and elongation in comparison to the untreated starting material. The highest average tenacity and elongation at break measured for the standard Ioncell fibres produced in this study were 46.0 cN/tex and 8.0%. This shows that using recycled nonwoven fabric as a raw material for new regenerated cellulosic fibres leads to a small, but not significant, decrease in fibre strength and no change or small increase in elongation in comparison to standard wood pulp. However, Ioncell fibres can reach tenacity values up to 61.5 cN/tex with high purity wood pulp as raw material (Moriam *et al.*, 2021), which is significantly higher than the values obtained for standard Ioncell fibres prepared in this study. This is partly due to the higher purity of the starting material used by Moriam *et al.* as well as the higher draw ratio and the use of a multifilament spinning

unit in comparison to the monofilament unit used to produce the standard fibres in the current study. Furthermore, the properties of the standard Ioncell fibres produced in this study were better at DR 5 than DR 8, which goes against expected outcome. Therefore, something has most likely happened during the spinning process that interfered with the stretching and thus orientation of the fibres, leading to inferior fibre properties than what is typical for standard Ioncell fibres.

The fibres produced from straw pulp had somewhat higher tenacities than the fibres produced from recycled nonwovens reaching closer to the values of standard Ioncell fibres. The highest average tenacity of fibres produced from straw pulp was 45.0 cN/tex, which was achieved by using a 12% cellulose concentration and draw ratio 8. However, the spinning was more stable when using a cellulose concentration of 13%, and the average tenacity for these fibres at DR 8 was 43.8 cN/tex. The average elongation of the fibres was 8.0% for the former and 8.4% for the latter. Since there is not a significant difference between the performance of the fibres depending on whether a cellulose concentration of 12% or 13% is used, the one with superior spinning is the one with more commercial value as its production would be easier to scale up.

Table 6. Average linear density (dtex), elongation at break (%), and tenacity (cN/tex) of produced fibres at most significant draw ratios with standard deviation. Literature values for elongation and tenacity of other common regenerated cellulosic fibres and synthetic fibres used in nonwovens. Values are shown for the produced fibres in dry and wet conditions.

Fibre type	Condition	Linear density (dtex)	Elongation at break (%)	Tenacity (cN/tex)	Source
Enocell 5, DR 5	dry	2.9±0.5	8.0±1.3	46.0±4.4	
	wet	2.7±0.6	9.7±1.8	40.3±7.5	
Enocell 5, DR 8	dry	1.5±0.3	8.1±1.3	43.6±7.2	
	wet	1.3±0.3	9.9±1.7	39.4±6.4	
Nonwoven untreated, DR3	dry	4.1±0.5	7.1±1.7	26.4±4.1	
	wet	4.1±0.7	9.4±2.9	24.1±4.0	

Nonwoven, 4% NaOH, large unit DR5	dry	2.6±0.4	9.0±2.0	37.0±3.7	
	wet	2.6±0.4	10.4±1.9	27.3±4.1	
Nonwoven, 4% NaOH, large unit DR8	dry	1.6±0.2	8.1±1.7	38.2±4.2	
	wet	1.6±0.2	10.0±1.3	35.6±4.7	
Nonwoven, 4% NaOH, large unit DR11	dry	1.9±0.3	8.7±1.8	37.5±5.1	
	wet	1.9±0.3	10.8±1.7	33.5±4.5	
Nonwoven, 4% NaOH + H_2SO_4 0.05 M, large unit DR5	dry	2.8±0.3	10.3±1.7	34.4±3.4	
	wet	2.5±0.2	12.0±1.0	32.4±3.7	
Nonwoven, 4% NaOH + H_2SO_4 0.05 M, large unit DR8	dry	1.7±0.2	9.0±0.7	39.9±4.2	
	wet	1.6±0.1	10.9±1.4	35.9±3.8	
Nonwoven, 4% NaOH + H_2SO_4 0.05 M, large unit DR11	dry	1.6±0.4	8.8±2.2	37.6±3.5	
	wet	1.7±0.3	11.6±1.7	35.5±4.0	
Straw 12% DR 5	dry	2.4±0.4	8.9±1.9	42.6±4.1	
	wet	2.0±0.3	10.7±1.6	40.5±4.6	
Straw 12% DR 8	dry	1.5±0.2	8.0±1.9	45.0±5.3	
	wet	1.4±0.2	11.9±2.2	42.9±3.9	
Straw 12% DR 11	dry	1.1±0.2	8.3±2.0	44.3±7.3	
	wet	1.1±0.1	11.5±1.8	44.6±5.1	
Straw 13% DR 5	dry	2.8±0.5	8.9±1.9	38.9±4.4	
	wet	2.8±0.5	11.2±1.9	36.1±6.0	
Straw 13% DR 8	dry	2.0±0.3	8.4±1.8	43.8±5.0	
	wet	2.0±0.3	10.8±1.9	39.2±3.8	
Straw 13% DR 11	dry	1.34±0.2	8.5±2.0	43.3±3.9	
	wet	1.2±0.2	10.1±1.5	40.1±4.4	
Viscose	dry	-	21±4	20.4±2.7	Sayyed *et al.*, 2019
	wet	-	25.5±4.5	11.1±2.2	
Cellulose acetate	dry	-	30±5	11.5±0.9	Sayyed *et al.*, 2019
	wet	-	40±5	6.2±0.4	
Cuproammonium	dry	-	12.5±0.5	19.5±4.4	Sayyed *et al.*, 2019
	wet	-	26.5±0.5	14.6±0.4	
Lyocell (NMMO)	dry	-	13.5±2.5	44.2±4.4	

	wet	-	17±1	35.0±0.4	Sayyed *et al.*, 2019
Nylon 6.6 (staple fibre)		-	43	37	Hearle & Morton, 2008
Polyester (staple fibre)		-	37	47	Hearle & Morton, 2008
Polypropylene		-	40	52.5±17.5	Cordin & Bechtold (2018); Li (2020)
Polyethylene (high-density)		-	10	34	Hearle & Morton, 2008

Both the fibres produced from recycled nonwovens and from straw pulp have a higher tenacity than most other commercially available regenerated cellulosic fibres like viscose. Only NMMO-based Lyocell fibres slightly exceed the values of the fibres produced form nonwovens and are roughly equal to the fibres made from straw pulp. Since the nonwoven is originally produced from Lyocell fibres, this shows that the recycling process has been relatively successful although there is a slight decrease in both tenacity and elongation. This is highlighted in Figure 20, that shows the tenacities in relation to elongation at break of the fibres produced from recycled nonwovens with comparison to the standard Ioncell fibres and Lyocell fibres. The corresponding values measured for the fibres produced from straw pulp are shown in Figure 21 with comparison to standard Ioncell fibres. As seen in Table 6, the fibres produced in this study also have higher tenacity than some synthetic fibres such as polyethylene and nylon, while polyester and polypropylene are stronger. Synthetic fibres also tend to have significantly higher elongation. Nonetheless, many of the applications that nonwoven textiles are used for, such as hygiene products, wipes, and household textiles, do not require very high strength or elongation. Therefore, the generally

lower elongation of Ioncell fibres and the slight decrease in fibre strength caused by recycling or using straw as raw material are not necessarily critical issues.

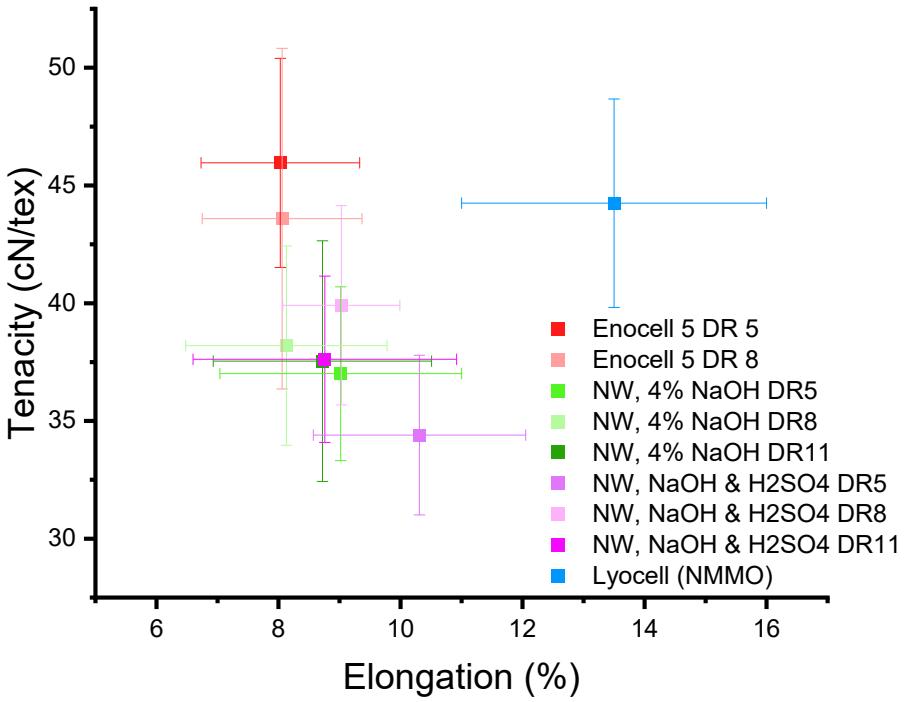

Figure 20. Tenacity (cN/tex) vs elongation at break (%) in dry conditions of standard Ioncell fibres, Ioncell fibres from recycled nonwovens, and Lyocell fibres (value for Lyocell from Sayyed et al. (2019).

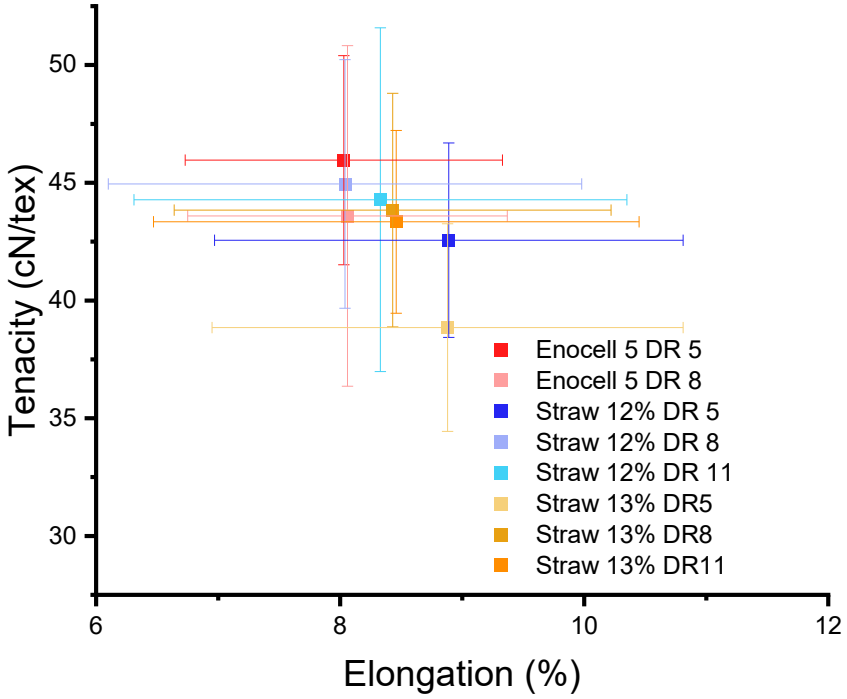

Figure 21. Tenacity (cN/tex) vs elongation at break (%) in dry conditions of standard Ioncell fibres and Ioncell fibres from straw pulp.

The tenacities of fibres produced in previous studies on using recycled materials in the Ioncell process have shown both higher and lower values than the ones in the current study (39.9 cN/tex). Selinger (2022) achieved tenacities of 29.2 cN/tex at DR 7 when spinning fibres from recycled banknotes with a multifilament unit, and 37.3 cN/tex at DR 11 using a monofilament unit. Airaksinen (2023) achieved continuous spinning from recycled viscose fibres at maximum DR 8 and obtained fibres with a tenacity of only 33.45 cN/tex. The starting material used by Selinger had a high amount of impurities, while the starting material of Airaksinen had an exceptionally low molecular weight and viscosity, leading to the inferior spinnability and fibre tenacity in comparison to the current study. When higher draw ratios were reached than in the current study, fibre tenacities were also higher. Ma *et al.* (2016a) produced fibres from recycled newsprint and achieved 43.89 cN/tex at DR 18 and 40.95 cN/tex at DR 12, and Ma *et al.* (2016) achieved a maximum of 51.38 cN/tex at DR 18 using recycled cardboard. In contrast, there are also cases in which inferior or similar spinnability as in the current study led to stronger fibres. Haslinger *et al.*

(2019a; 2019b) respectively reached tenacities of 48 cN/tex from recycled cotton polyester waste while only achieving DR 8.8 and 59.8 cN/tex from dyed cotton waste at DR 12. The high tenacities are likely due to the use of cotton as starting material in the studies by Haslinger *et al.* since it has a high molecular weight and is thus expected to produce stronger regenerated fibres. The elongation at break did not have significant differences between studies, with the exception of the fibres produced from dyed cotton waste by Haslinger *et al.* (2019b) that reached elongation of up to 14.3%. Overall, the fibres produced from recycled nonwovens in the current study performed well considering the draw ratios that were possible to reach and the nature of the starting material in comparison to previous attempts at recycling materials with the Ioncell process.

The fibres produced from straw pulp performed very well in comparison to previous published studies examining the use of straw for production of regenerated fibres. Previous studies have reached tenacities of 15.3 cN/tex, (Chen *et al.*, 2015), 20.5 cN/tex (Protz *et al.*, 2021), and 26.6 cN/tex (Lim *et al.*, 2001). The elongation at break was 4.12% for the fibres produced by Chen *et al.* (2015) and 3.2% for the fibres produced by Protz *et al.* (2021). These values are significantly lower than the highest values achieved in the current study (tenacity 45.0 cN/tex and elongation at break 8.9%). One reason for the inferior performance of the previously studied fibres is the use of lower cellulose concentrations during dope preparation (4% for Chen *et al.* and 10% for Protz *et al.*) compared with the 12% and 13% concentrations used in the current study. Chen *et al.* also used wet spinning that generally produces weaker fibres than dry-jet wet spinning. This is because in dry-jet wet spinning, the filaments pass through an air gap before being immersed in the coagulation bath allowing the filaments to stretch and align along the fibre axis, cool down slightly, and relax stresses accumulated within the spinneret (Bajaj *et al.*, 2005). In addition to the experimental methods used, the properties of the various straw materials used likely had an effect on the

performance of the fibres, which is discussed in more detail in the upcoming sections analysing the characterisation results of the current study.

Figures 22 and 23 show the tenacity vs elongation curves in both dry and wet conditions for the fibres produced from recycled nonwovens and straw pulp respectively. MMCFs often exhibit a significant decrease in tenacity in wet conditions, as is seen in Table 6. During tensile tests performed in wet conditions, cellulose fibres absorb water by forming hydrogen bonds with the accessible OH-groups that are mainly present in amorphous regions (Asaadi *et al.*, 2018). This makes the fibres swell and disturbs the hydrogen bond network, resulting in decreased tensile strength (Asaadi *et al.*, 2018). This issue is also present in but substantially limited by the Ioncell process, due to the high level of orientation that is achieved in fibres by dry-jet wet spinning (Asaadi *et al.*, 2018). The wet-to-dry tenacity ratios of Ioncell fibres have been reported to be in the range 0.6-1.0 for Ioncell fibres (Asaadi *et al.*, 2018). The same can be observed in the fibres produced from recycled nonwovens as well as straw pulp, that exhibit ratios in the range 0.64-0.99 and 0.84-1.00 respectively. As many applications of nonwovens, such as hygiene products and medical textiles, come in contact with liquids, sufficient wet strength of fibres is desirable.

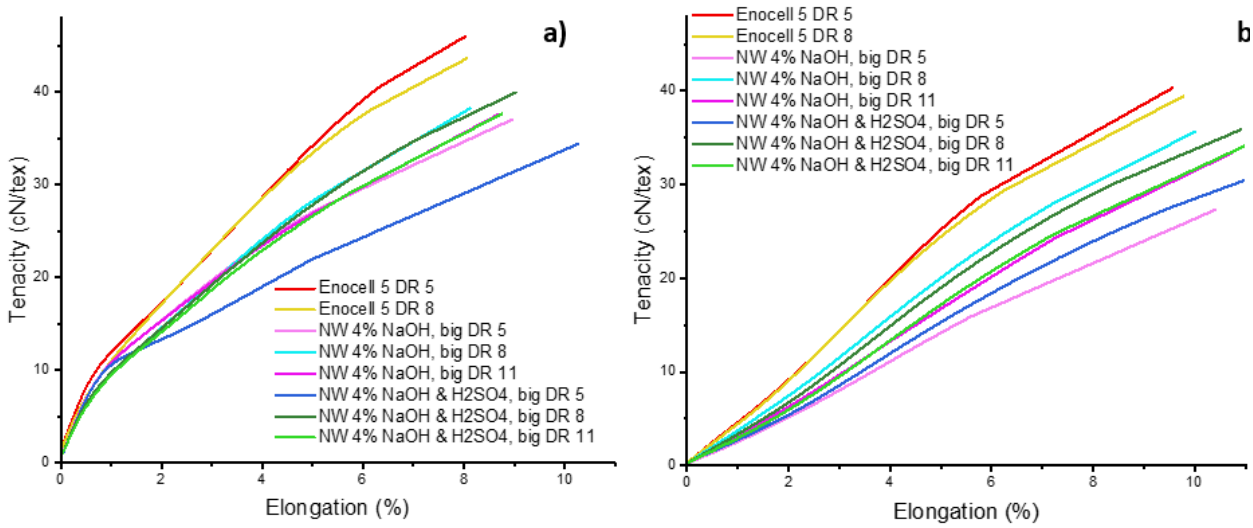

Figure 22. Tenacity vs Elongation measured in dry conditions a) and wet conditions b) for fibres produced from recycled nonwovens and Enocell.

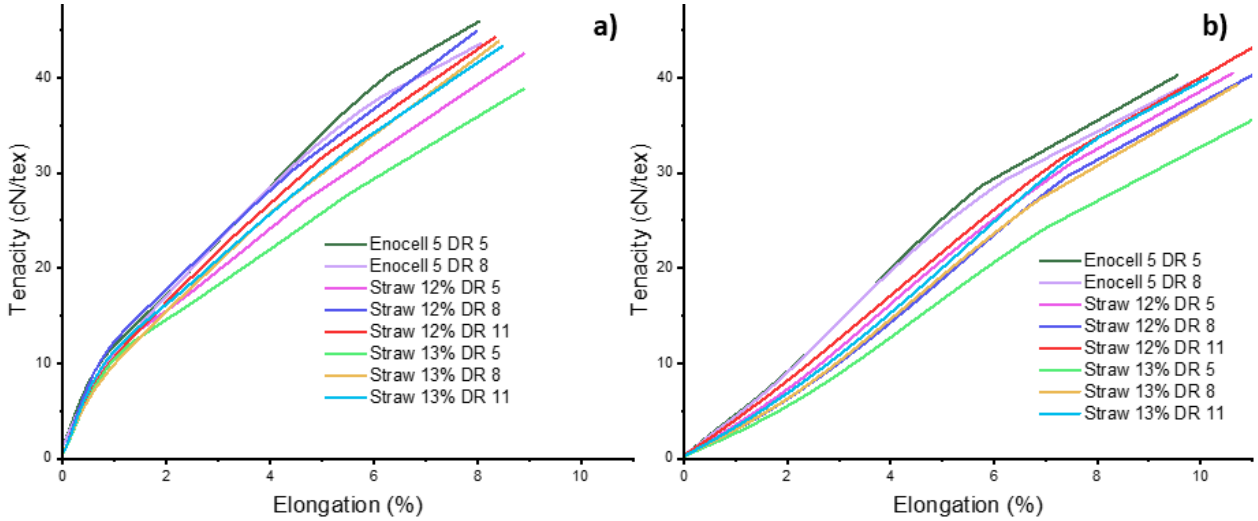

Figure 23. Tenacity vs Elongation measured in dry conditions a) and wet conditions b) for fibres produced from straw pulp and Enocell.

The birefringence and total orientation of the best produced regenerated fibres at most significant draw ratios are presented in Table 7. In a previous study, it was shown that Ioncell fibres with DR 5 have a total orientation of 0.72 and DR 7 have an orientation of 0.67 (Asaadi *et al.*, 2018). Most of the measured results correspond reasonably well with these values but some are slightly lower. There are some exceptions that showed very low total orientations under 0.6, indicating that something has interfered with the ability of the filaments to align and orient themselves properly during the spinning process. This often occurs at lower draw ratios because the fibres are not being stretched enough to allow correct alignment of the cellulose molecules. Therefore, many of the fibres produced in this study show low total orientations at DR 5.

Generally, a higher draw ratio corresponds to a higher orientation and higher tenacity because the drawing process aligns polymer molecules along the chain axis, enhancing orientational strength (Sengupta, 1997). This increases the extent of intermolecular hydrogen bonding resulting in a more uniform structure and thus higher strength (Michud *et al.*, 2016b). However, this is only accurate up to a certain point, after which very high draw ratios induce

molecular chain slippage, that can decrease the orientation and strength of the fibres (Mortimer, Peguy & Ball, 1996; Cicero & Dorgan, 2001). The draw ratios achieved in this study are not very high, but for some of the samples, a higher draw ratio still led to a lower total orientation. This occurs especially in the case of fibres spun at draw ratio 11. For the most part, a decreased total orientation led to a decrease in fibre tenacity.

Table 7. Birefringence (Δn) and total orientation (f_t) of produced fibres at most significant draw ratios

Fibre type	Δn	f_t
Enocell 5, DR 5	0.0446±0.0100	0.719±0.0054
Enocell 5, DR 8	0.0333±0.0033	0.537±0.0007
Nonwoven, 4% NaOH, large unit DR5	0.0212±0.0090	0.342±0.145
Nonwoven, 4% NaOH, large unit DR8	0.0463±0.0064	0.746±0.103
Nonwoven, 4% NaOH, large unit DR11	0.0438±0.0013	0.706±0.020
Nonwoven, 4% NaOH + 0.05 M H_2SO_4, large unit DR5	0.0128±0.0115	0.206±0.186
Nonwoven, 4% NaOH + 0.05 M H_2SO_4, large unit DR8	0.0444±0.0066	0.716±0.106
Nonwoven, 4% NaOH + 0.05 M H_2SO_4, large unit DR11	0.0467±0.0075	0.754±0.122
Straw 12%, DR5	0.0303±0.0139	0.489±0.224
Straw 12%, DR8	0.0458±0.0023	0.739±0.038
Straw 12%, DR11	0.0453±0.0107	0.731±0.172
Straw 13%, DR5	0.0213±0.0102	0.344±0.165
Straw 13%, DR8	0.0462±0.006	0.745±0.097
Straw 13%, DR11	0.0451±0.0056	0.728±0.090

The birefringence of a perfectly aligned fibre has been reported as 0.057 (Sixta *et al.* 2015). Ioncell fibres produced from Enocell pulp at DR 12 have previously been reported to have a birefringence of 0.0453 (Michud *et al.*, 2016a), while the birefringence of Lyocell has been reported as 0.04263 and viscose as 0.0265 (Drambei *et al.*, 2009). Compared to these values, the birefringence of the fibres produced in the current study show reasonably high values with the fibres spun at DR 8 and above being in the range 0.0444-0.0467.

Figure 24 shows the cross-sections of the fibres produced from recycled nonwovens obtained by SEM. All of the fibres were produced with the monofilament spinning unit at DR 5, except the untreated nonwoven (b) that only reached DR 3. The cross-sections of the fibres look similar and show no clear defects. The cross-sections are similar to the ones obtained by Sederholm (2022) for standard Ioncell fibres.

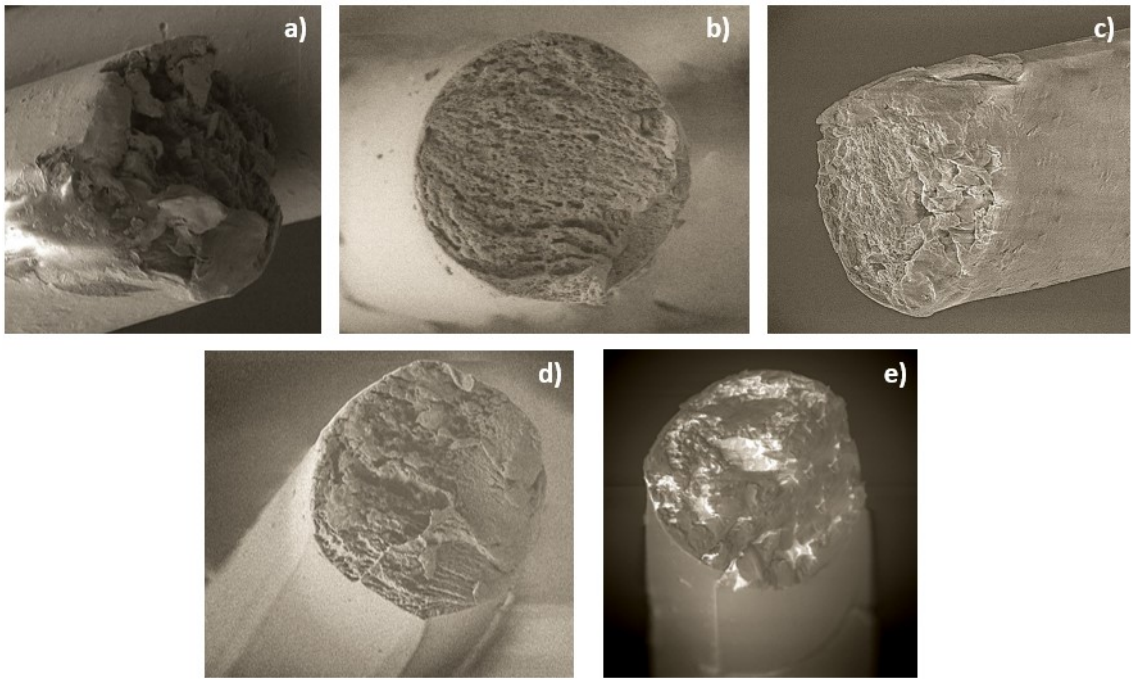

Figure 24. SEM images of regenerated fibres produced from Enocell a), untreated nonwoven b), nonwoven treated with 4% NaOH at 40 °C for 50 minutes c), nonwoven sieved and treated with 4% NaOH at 40 °C for 50 minutes d), and nonwoven sieved and treated with 4% NaOH at 40 °C for

50 minutes and 0.05 M H_2SO_4 at 60 °C for 10 minutes e). Images edited with Adobe Photoshop Express Photo Editor.

4.4 Polymer Properties

The results of the GPC measurements are summarized in Table 8. The molecular weight distributions of the produced fibres in relation to their starting materials are shown in Figure 25 and 26 for recycled nonwovens and straw respectively. The molecular weights measured for Enocell are similar to the results of Sederholm (2022), but the molecular weight distribution (MWD) is wider. The nonwoven has a slightly lower molecular weight than Enocell, while straw pulp has a distinctly higher molecular weight than the other two. This is likely a contributing reason to why the fibres produced from straw were stronger than the ones produced from recycled nonwovens, since a higher molecular weight has been shown to increase fibre tenacity (Bredereck and Hermanutz, 2005; Ma *et al.,* 2023).

The molecular weight and polydispersity index of all fibres produced from nonwovens or straw pulp are lower than their respective starting materials, indicating cellulose degradation during processing into fibres. This can be seen in Figures 25-26 as the curves becoming narrower and taller and shifting towards a lower molecular weight. Furthermore, the amount of both short and long polymer chains was smaller for all produced fibres than for their starting materials. A similar effect was observed by Sederholm (2022) with fibres produced from Enocell pulp with the Ioncell process, with the exception of the low molecular weight fraction, that increased after processing into fibres. The fibres produced from wheat straw by Chen *et al.* (2015) using wet spinning show a distinctly smaller decrease in molecular weight in relation to the starting material used than the fibres produced from straw pulp in the current study. Chen *et al.* also used a starting material with a significantly higher molecular weight to begin with. Their starting material had the molecular weights M_w 1046.9 kDa and M_n 185.85 kDa, while their strongest fibres had the values M_w 784.36 kDa and M_n 165.59 kDa. However, these fibres had a significantly lower tenacity and elongation at break than

the fibres produced in the current study. This indicates that although molecular weight is a significant factor for strength and flexibility of regenerated fibres, other aspects such as spinning method can have a greater influence.

Table 8. Number average molecular weight M_n (kDa), weight average molecular weight M_w (kDa), polydispersity index M_w/M_n, low molecular weight fraction DP<100 (%), and high molecular weight fraction DP>2000 (%) of each starting material, nonwoven after various pre-treatments, and regenerated fibres.

Sample	M_n (kDa)	M_w (kDa)	M_w/M_n	DP < 100 (%)	DP > 2000 (%)
Enocell	55.0±7.7	141.0±2.2	2.6±0.3	5.3±1.5	6.4±2.7
Nonwoven	58.8±2.6	131.2±1.5	2.2±0.1	2.8±0.7	7.9±0.1
Straw pulp	182.1±1.3	381.1±12.2	2.1±0.1	0.0±0.0	44.7±1.4
NW, 4% NaOH 50 min	43.5±4.3	114.2±2.1	2.7±0.2	7.6±1.8	6.6±0.1
NW, 4% NaOH 90 min	41.4±7.2	96.1±23.2	2.3±0.2	8.5±2.7	13.4±6.1
NW, 6% NaOH 60 min	47.9±0.9	107.3±1.3	2.3±0.0	5.3±0.7	5.5±0.1
NW 4% NaOH & 0.05M H_2SO_4	49.6±1.1	155.3±51.2	2.1±0.0	3.8±0.4	5.0±0.1
Fibres, NW 4% NaOH	45.1±0.9	86.1±1.3	1.9±0.0	4.9±0.3	2.0±0.1
Fibres, NW 4% NaOH & H_2SO_4	46.9±0.7	92.2±0.5	2.0±0.0	4.0±0.4	2.5±0.6
Fibres, Straw 12%	120.4±1.5	216.3±1.5	1.8±0.0	0.0±0.0	20.4±0.0
Fibres, Straw 13%	124.2±7.9	218.3±8.5	1.8±0.0	0.0±0.0	20.5±0.8

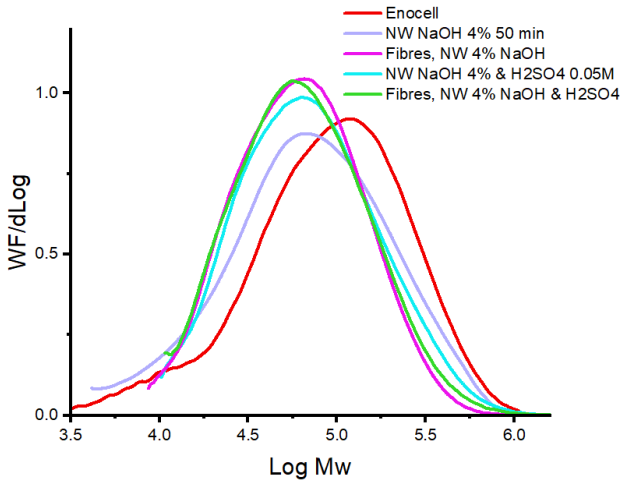

Figure 25. Molecular weight distribution of Enocell, the best fibres produced from recycled nonwovens and their corresponding starting materials.

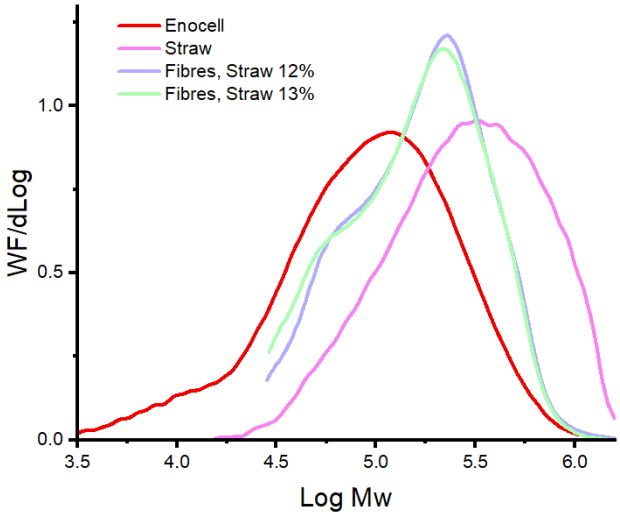

Figure 26. Molecular weight distribution of Enocell, straw pulp and fibres produced from straw pulp.

Figure 27 shows the effects of the pre-treatments on the molecular weight distribution of the nonwoven. The NaOH pre-treatments performed on the nonwoven appear to decrease the molecular weight and slightly broaden the molecular weight distribution. The NaOH-treatments also increase the percentage of shorter polymer chains and for the most part decrease the percentage of long polymer chains. Some of the NaOH-treated nonwovens show a distinctly higher standard deviation in weight average molecular weight than the other examined materials, indicating that the treatments may affect the cellulose molecules unevenly, with some parts experiencing more cellulose degradation than others. The harsher NaOH-treatments appear to have slightly lower molecular weight than the treatment with 4% NaOH for 50 minutes, which was the only one of the treatments that provided spinnable dopes. In contrast, the H_2SO_4-treatment performed after the NaOH-treatment does not appear to have a further effect on the average molecular weight of the nonwoven, but it makes the polydispersity index narrower.

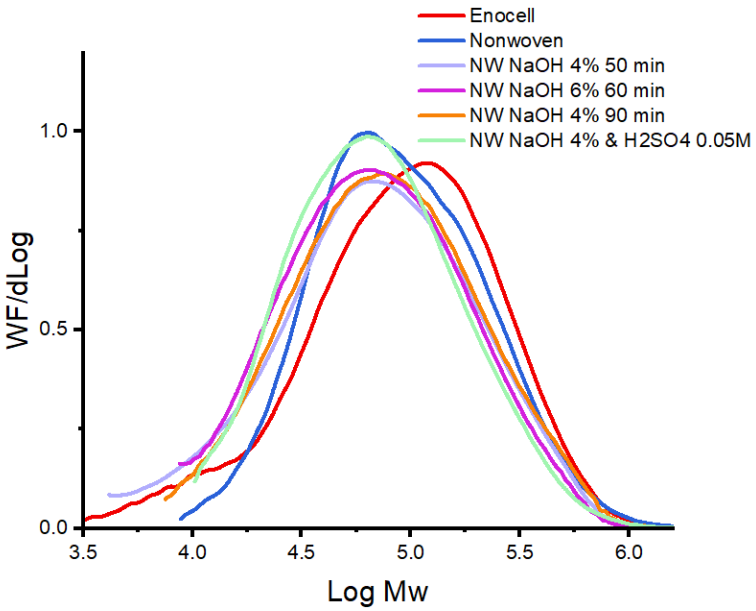

Figure 27. Molecular weight distribution of Enocell, untreated nonwoven, and nonwoven after various pre-treatments.

The intrinsic viscosities of each starting material and each produced fibre type are shown in Figure 28. The results correspond with the trends in measured molecular weights with a lower molecular weight leading to lower intrinsic viscosity. It can be seen that the ground nonwoven has a lower viscosity than the standard Enocell pulp, while straw pulp has a significantly higher viscosity. Neither of these are within the optimal range of viscosity for fibre spinning dopes with a cellulose concentration of 13%, which is 360-500 ml/g (Ma *et al.*, 2023), but the nonwoven is very close, making it sufficiently suitable. The high viscosity of the straw pulp means that a lower cellulose concentration in the dope may be necessary, which is why a lower cellulose concentration of 12% was tested. However, this was not apparent in the fibre spinning, that showed that the dope with 13% cellulose concentration performed better than 12%. The results also show that intrinsic viscosity of the nonwoven is slightly decreased by the NaOH-treatments, with the treatment using higher concentration (6%) having the most noticeable effect. The H_2SO_4-treatment also led to a decrease in intrinsic viscosity when performed in a small batch using both 0.01 M and 0.05 M H_2SO_4, but a larger batch using only 0.05 M concentration appeared

to have a slightly higher intrinsic viscosity. This is likely due to circumstantial variance in the values, since only one sample was measured, and does not indicate that the treatment increases viscosity, but likely has no clear effect on it. This would be in line with the GPC results, that showed no distinct difference in molecular weight after H_2SO_4 treatment.

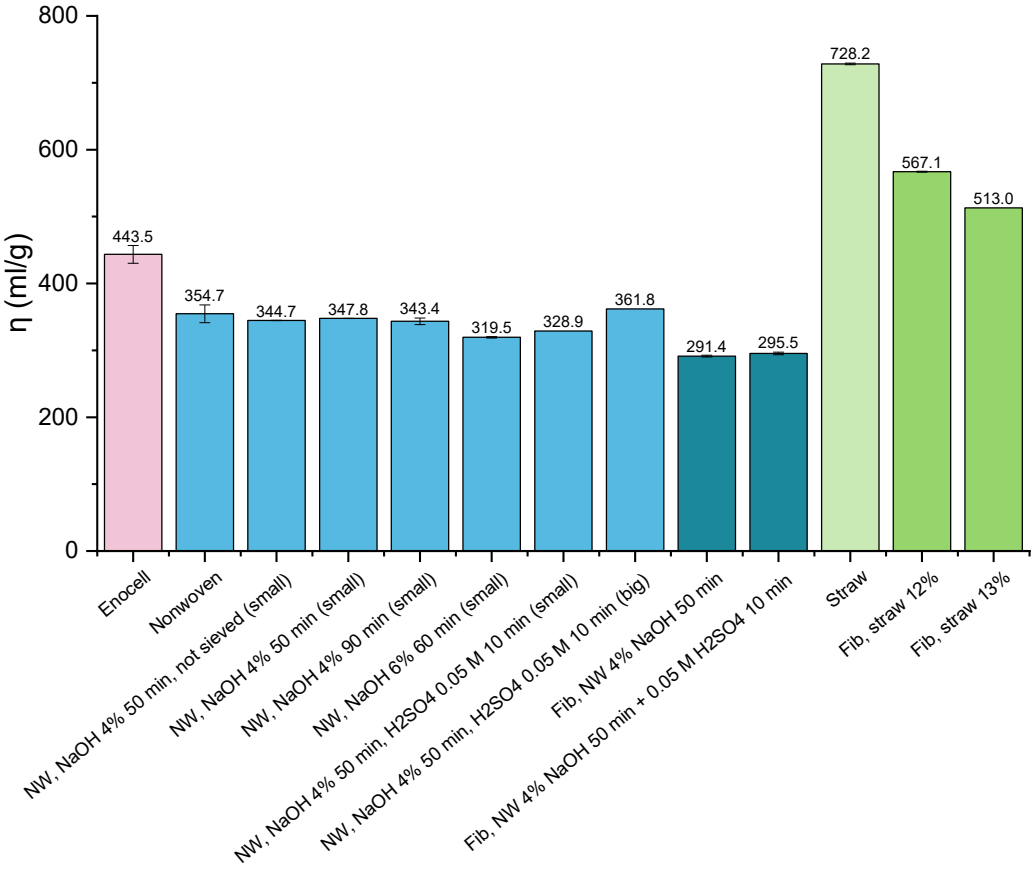

Figure 28. Intrinsic viscosities of Enocell (pink), nonwoven before and after various pre-treatments (light blue), regenerated fibres produced from nonwovens (dark blue), straw pulp (light green), and regenerated fibres produced from straw (dark green).

Intrinsic viscosity is decreased during processing of the starting materials into fibres when using either nonwoven or straw pulp as raw material. This corresponds to the results demonstrated by Sederholm (2022) when studying the closed loop recycling of Ioncell fibres. Intrinsic viscosity decreased after pulp was processed into regenerated fibres and decreased again after recycling them into new fibres, which was expressed as a reason

for why the Ioncell process cannot be used for fully closed-loop recycling. Therefore, it is likely that the same applies to the materials used in the current study. Since the intrinsic viscosity of the nonwoven was low to begin with, it may not be suitable for recycling multiple times with the Ioncell process, although it can be recycled at least once. In contrast, since the intrinsic viscosity of the straw pulp was so high, the produced fibres could possibly be recycled further with the Ioncell process.

4.5 Chemical Composition

The FTIR spectra of all starting materials and produced fibres are presented in Figure 29. The spectra for the different starting materials and fibres are very similar, the main difference being in the intensity of some of the peaks. In general, the peaks are highest for straw pulp, and lowest for NaOH-treated nonwovens. All materials exhibit a peak in the range of 3300-3330 cm^{-1}, which represents intermolecular OH-bonds (Hinterstoisser & Salmén, 2000). A smaller peak occurs in all starting material samples in the range of 2835-2870 cm^{-1}, which correlates to CH_2 stretching (Abidi *et al.*, 2014). A group of small peaks can be seen in the range 1315-1420 cm^{-1}, representing CH_2 rocking and CH bending (Abidi *et al.*, 2014). A stronger peak occurs between 990 and 1015 cm^{-1}, with straw pulp exhibiting the strongest peak. This peak represents CO stretching (Abidi *et al.*, 2014). Very small peaks at about 890 cm^{-1} represent β-linkages of cellulose (Abidi *et al.*, 2014). Overall, the spectra of the starting materials shows that they are very close in structure to regular cellulose (Hivechi *et al.*, 2020), indicating that any possible impurities or additives in the starting materials are only present in small quantities on the material surface. The various pre-treatments performed on the nonwoven fabric do not change the FTIR peaks, but the peaks become smaller. This further shows that any additives present in the fabric are not present in significant amounts on the material surface. Furthermore, Figure 29 shows that the chemical compositions of the starting materials are not significantly changed by the dissolution and fibre spinning

process but the peaks generally become smaller. Similar FTIR spectra were obtained by Sederholm (2022) and Selinger (2022).

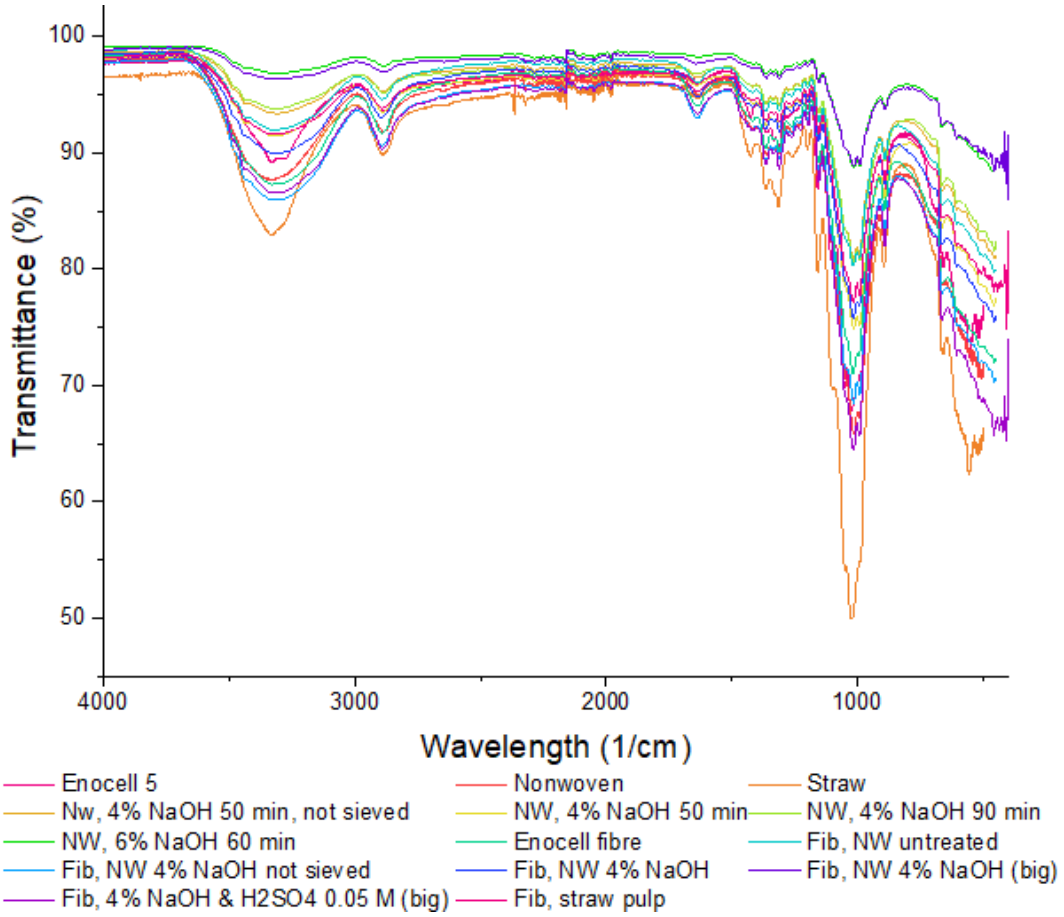

Figure 29. FTIR graphs for all starting materials, nonwoven after different pre-treatments, and produced fibres.

The ash content or amount of inorganic matter in all starting materials, both treated and untreated, as well as the resulting fibres can be seen in Table 9. Compared to the standard Enocell pulp, both the nonwoven and straw pulp have a significantly higher ash content. The higher ash content in the nonwoven originates from various additives, such as TiO_2 and possibly other substances used as binder or finishing. Straw in turn contains more inorganic matter than wood, because as an annual plant it obtains higher amounts of substances like silica, potassium, and calcium from the earth (Biricik et al., 1999). This is likely a contributing factor to the inferior spinnability and mechanical properties of regenerated fibres produced from these materials

in comparison to standard Ioncell fibres. However, the ash content alone cannot explain this behaviour, since previous studies on the Ioncell process have shown that using recycled feedstocks with considerably higher ash contents can lead to both worse and better spinnability than in the current study. Selinger (2022) used bleached, recycled banknotes with an ash content of 5.46% and achieved maximum DR 8 with a multifilament spinning unit, while Ma *et al.* (2018a) used recycled newsprint treated with alkaline polyol pulping with an ash content of 1.5% and reached DR 18. The study by Protz *et al.* (2021) used straw with an ash content of 4.3% for dry-jet wet spinning fibres with a DR of 5.7. Since straw pulp used in the current study had significantly fewer inorganic impurities, this is likely a contributing factor as to why a higher draw ratio of 8 could be achieved in the current study. Moreover, the straw used by Lim *et al.* (2001) to produce regenerated fibres had an ash content of 2.1% and lead to fibres that did not perform as well as the ones in the current study, further indicating that a higher amount of inorganic purities in a starting material could contribute to inferior regenerated fibres.

Table 9. Ash content in the starting materials before and after possible treatments, and the regenerated fibres.

Material	Ash content (%)
Enocell	0.17 ± 0.10
Straw	0.93 ± 0.45
Nonwoven	0.98 ± 0.29
NW 4% NaOH 50 min	1.54 ± 1.10
NW 4% NaOH 90 min	0.67 ± 0.10
NW 6% NaOH	0.43 ± 0.05
NW H_2SO_4 0.01M	0.98 ± 0.16
NW H_2SO_4 0.05M	0.52 ± 0.03
NW fibres NaOH	0.68 ± 0.02
NW fibres NaOH + H_2SO_4	0.62 ± 0.49
Straw fibres 12%	0.69 ± 0.16
Straw fibres 13%	0.20 ± 0.14

The ash content of the nonwoven can be decreased by both NaOH and H_2SO_4 pre-treatments, with harsher treatments removing more inorganic matter. The higher ash content measured after treatment with 4% NaOH for 50

minutes is likely higher than the untreated nonwoven due to some traces of sodium from the treatment still being left on the fibres indicating insufficient washing. In contrast, the ash content decreases by about 34% in comparison to the untreated nonwoven, when the treatment time is increased to 90 minutes and about 43% if the concentration is increased to 6%. This indicates that a harsher NaOH treatment is able to remove more impurities from the nonwoven. However, the dopes produced from nonwoven treated with the more intense NaOH-treatments were non-spinnable, meaning that NaOH cannot remove all impurities from the nonwoven without negatively affecting the spinnability. There appears to be no direct correlation with the amount of inorganic matter present in the regenerated fibres in comparison to the starting materials. Fibres produced from nonwovens treated with NaOH and H_2SO_4 show a slight increase in ash content after processing into fibres, while fibres produced from straw pulp have a reduced ash content. The decrease is more significant when using 13% cellulose content than when using a 12% content, although there should be no considerable difference between the two.

The results of the elemental analysis can be seen in Table 10. The amount of carbon and hydrogen is similar in all starting materials, but the nonwoven showed less nitrogen than the measurement was able to detect. This result seems reasonable since the nonwoven consists of Lyocell fibres and Su *et al.* (2021) also observed no detectable amounts of nitrogen in the elemental analysis of Lyocell fibres. Since the nonwoven contains additives, the relative nitrogen content is also lowered due the increased presence of other elements. All produced fibres show an increase in nitrogen content in comparison to their respective starting materials. This is because there is nitrogen in the ionic liquid used for dissolving the cellulose and some traces are left in the fibres after coagulation and washing. The higher value of nitrogen in the fibres produced from nonwoven treated with NaOH and H_2SO_4 indicate poorer washing of the fibres. The carbon and hydrogen contents remain almost unchanged throughout the fibre production process.

Table 10. Elemental analysis results, showing the nitrogen, carbon and hydrogen contents (%) of each starting material, treated nonwovens, and produced fibres. LOQ refers to limit of quantification.

Material	Nitrogen (%)	Carbon (%)	Hydrogen (%)
Enocell	0.01±00	42.05±0.12	6.19±00
Straw pulp	0.01±00	43.23±0.01	6.29±0.05
Nonwoven	<LOQ (=0.01%)	41.71±0.02	6.29±0.02
Nonwoven, 4% NaOH	<LOQ (=0.01%)	41.20±0.04	6.23±0.01
Nonwoven 4% NaOH & 0.05M H_2SO_4	<LOQ (=0.01%)	41.58±0.01	6.25±0.08
Fibres, NW 4% NaOH	0.04±0.01	41.58±0.04	6.23±0.02
Fibres, NW 4% NaOH & 0.05M H_2SO_4	0.05±0.07	42.80±0.63	6.29±0.08
Fibres, Straw pulp	0.03±0.01	42.92±0.11	6.30±0.02

Figure 30 shows the thermogravimetric (TG) data of the untreated and treated starting materials as well as produced fibres, and the corresponding differential thermogravimetric (DTG) data. These represent the relative mass loss in the sample during heating and the rate of sample degradation respectively. The onset temperatures, indicating the beginning of sample degradation, and the DTG peaks, corresponding to the temperature at which the highest degradation rate occurs, are summarised in Table 11. Table 11 also shows the total mass loss after the main degradation has occurred accounting for any slight mass loss occurring in the beginning due to moisture, as well as the total mass in percentages that is left once the samples have been heated to 600 °C. The onset temperatures and DTG peaks are similar for most samples. In contrast, as seen in Figure 30, the extent of degradation differs between samples. The mass reduction is greater for straw pulp than nonwoven, which is to be expected due to the additives present in the nonwoven. The boiling point of TiO_2 is 2972°C (Sungur, 2021), meaning that it is not degraded during the measurement and is left in the nonwoven samples. In accordance with the ash contents of the samples, the amount of residue left in the nonwoven samples after thermal analysis decreases after treatment with 0.0 5M H_2SO_4 and slightly increases from this after processing into fibres. This indicates that the H_2SO_4-treatment is

successfully removing non-degrading components from the nonwoven. In contrast, the NaOH-treatment appears to increase the amount of non-degrading matter. The same phenomenon was observed in the ash content measurements and is likely a result of poor washing of the NaOH from the nonwoven. This is supported by the fact that the fibres produced from nonwovens treated with NaOH show an increased mass loss in comparison to untreated nonwoven, indicating that the NaOH-treatment is not permanently adding non-degradable matter to the material. Notably, the fibres produced from straw pulp show a decreased mass loss in comparison to their starting material, which was not observed in the ash content measurements. Overall, similar TG and DTG curves were obtained by Sederholm (2022) for standard Ioncell fibres with onset temperatures slightly lower than the ones in the current study (312-322 °C).

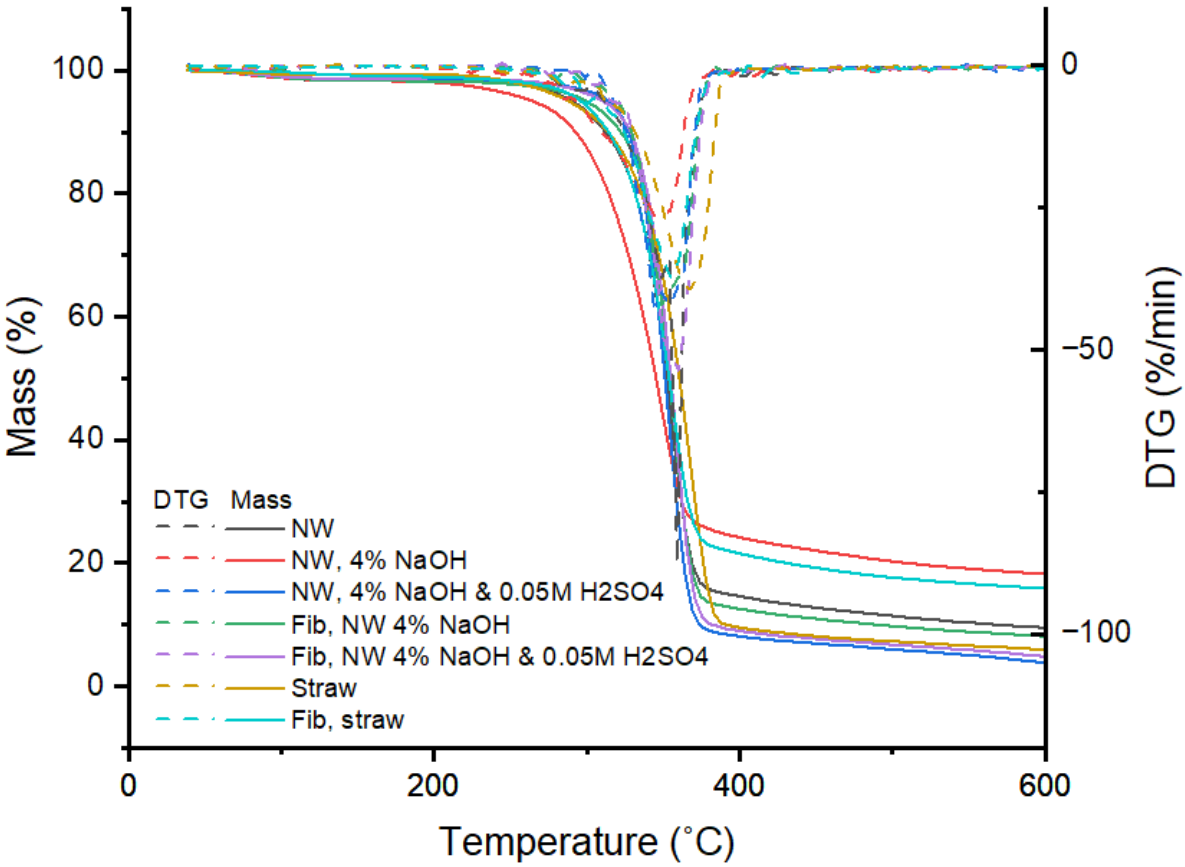

Figure 30. Sample mass (%) (left) and degradation rate (%/min) (right) vs temperature (°C) for starting materials, nonwoven after best pre-treatments, and produced fibres.

Table 11. Onset temperature (°C), temperature at DTG peak (°C), mass loss after main degradation (%), and mass left after heating to 600 ˚C (%).

Sample	Onset Temperature (˚C)	DTG Peak (˚C)	Mass Loss After Main Degradation (%)	Mass Left at 600 ˚C (%)
Nonwoven	344.8	350.1	82.8	9.5
Nonwoven, 4% NaOH	313.8	345.1	72.7	18.3
Nonwoven 4% NaOH & 0.05M H_2SO_4	342.5	347.7	89.4	3.9
Fibres, NW 4% NaOH	348.1	353.1	86.4	8.1
Fibres, NW 4% NaOH & 0.05M H_2SO_4	342.9	360.1	88.8	4.9
Straw Pulp	341.1	364.0	88.9	6.0
Fibres, Straw pulp	330.6	352.0	75.5	15.9

Figure 31 shows the normalized results of the DSC measurements, representing the amount of energy absorbed or released during the thermal analysis of the materials. The nonwoven samples have a sharp drop in energy in the range 285-305 °C, excluding the nonwoven samples that have been treated with H_2SO_4. This could mean that the H_2SO_4-treatment has removed some kind of additive from the nonwoven that is degrading at this temperature. The curve for NaOH and H_2SO_4-treated nonwoven also exhibits a sharp peak at 318 °C, while the other materials have a rounded peak in the same range.

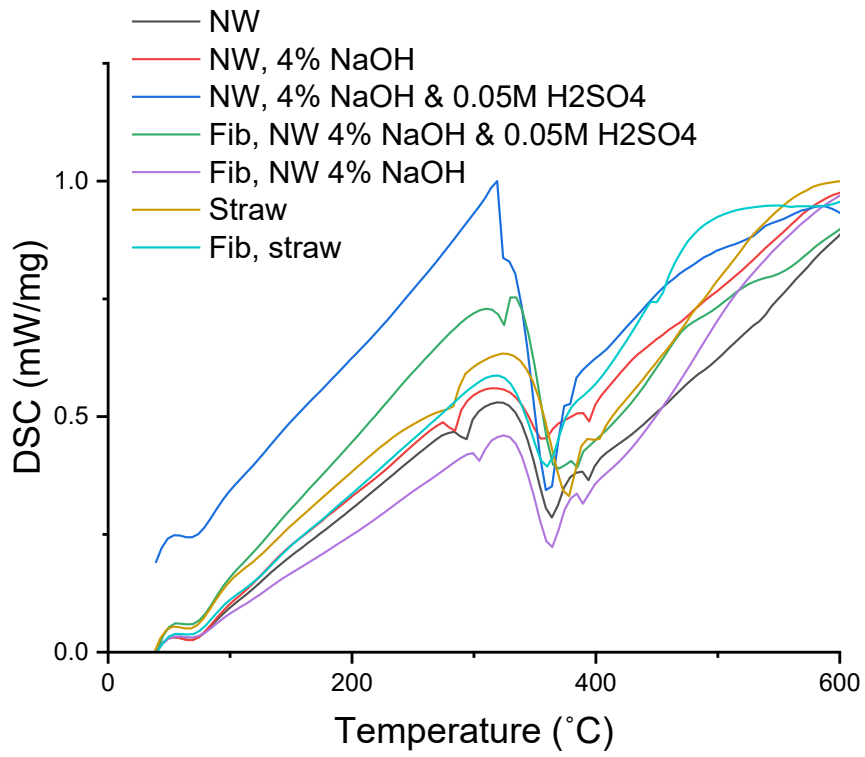

Figure 31. DSC (mW/mg) vs Temperature (°C) of starting materials, nonwoven after best pre-treatments, and produced fibres using normalized data.

Table 12 shows the lignocellulosic composition of the starting materials and produced fibres. As lignin cannot be distinguished from other non-saccharides in the current study, the percentage of the samples that are not cellulose or hemicellulos is referred to as non-saccharides. The results measured for Enocell pulp correspond well with the same values measured by Sederholm (2022). The straw pulp has distinctly less cellulose and more hemicellulose than the standard wood pulp as well as significantly more non-saccharides. This is a contributing factor for the poorer spinnability and fibre properties obtained with straw pulp in comparison to Enocell pulp, as a higher cellulose content and less impurities correlate with improved performance (Ma *et al.*, 2023). The straw pulp showed a lower non-saccharide content than the straw used by Protz *et al.* (2021) that had a lignin content of 19.4% and a significantly higher non-saccharide content than presented by Lim *et al* (2001) who used rice straw with a lignin content of

1.4%. The differences in non-saccharide content come from the processing methods used to treat the straw. Protz *et al.* used untreated straw, while Lim *et al.* treated their straw with NaOH to remove non-cellulosic impurities. The higher lignin content in the straw used by Protz *et al.* could be a reason why their fibres did not perform as well as the fibres produced in the current study or by Lim *et al.* However, the fibres produced in the current study were also superior to the ones produced by Lim *et al.* despite the higher non-saccharide content, indicating that other factors can have an even greater effect.

The nonwoven also shows a lower cellulose content and higher non-saccharide content than Enocell pulp. This is likely due to the additives found in the nonwoven, that increase the non-saccharides content and decrease the relative amount of other components in the fabric. The fibres produced from nonwovens show an increase in cellulose content and decrease in hemicellulose and non-saccharides content. Since the additives in the nonwoven are removed during pre-treatments before fibre spinning, it is reasonable that the amount of non-saccharides decreases and the relative amount of cellulose would thus increase. The fibres produced after NaOH and H_2SO_4 treatment show a greater decrease of non-saccharides and increase of cellulose than the ones produced from only NaOH-treated nonwoven, indicating that the acid treatment has successfully removed more additives from the nonwoven. The fibres produced from straw pulp also show an increase in cellulose content due to the relative decrease of non-saccharides.

Table 12. Cellulose, hemicellulose and non-saccharides contents (%) of starting materials and produced fibres.

Material	Cellulose (%)	Hemi-cellulose (%)	Non-saccharides (%)
Enocell	94.9	4.6	0.5
Straw	78.2	9.3	12.5
Nonwoven	90.2	2.9	6.9
Fibres, NW 4% NaOH	90.9	2.4	6.7
Fibres, NW 4% NaOH & H_2SO_4	95.6	1.4	3.0
Fibres, Straw	84.9	8.1	7.0

4.6 Optical Properties

The CIELab colour space values of the starting materials and produced fibres are shown in Table 13. The CIELab colour space refers to a system developed by the Commission Internationale de l'Eclairage (CIE), that attributes a certain colour with a numerical value (Westland, 2012). According to the system, each colour can be expressed as three coordinates: L, indicating lightness, and a and b, both referring to chroma and hue that together form colour (Berns, 2014). In general, the a axis indicates a balance between red and green, while b refers to a balance between yellow and blue (Berns, 2014). Positive values on the axes refer to more red and yellow shades, while negative values refer to more green and blue shades. Table 13 shows that Enocell is the lightest of the starting materials and straw pulp is the least light and more yellow than the other two. As expected, the produced fibres are less light than their starting material, with the exception of the fibres produced from straw pulp. Nonetheless, these fibres are more yellow and red than the straw pulp. Fibres produced from nonwovens are also somewhat more yellow than their starting material. The fibres treated with 4% NaOH and spun with a multifilament unit appear redder than the other fibres, while all other materials have a slight green hue. This could mean there is some residue of ionic liquid present in these fibres, since the IL used is orange in colour, but this cannot be said for certain since the differences in the values are relatively small in contrast to the full scale of values.

Table 13. CIELab values for all starting materials and most successful regenerated fibres.

Material	L	A	b
Enocell	85.32±1.13	-0.99±0.08	2.77±0.28
Enocell fibres	82.70±0.45	-1.25±0.48	2.85±1.00
Nonwoven	83.75±2.15	-0.44±0.05	1.32±0.14
Nonwoven fibres, untreated	81.49±0.99	-0.56±0.22	11.09±1.17
Nonwoven fibres, 4% NaOH	82.14±1.33	4.98±0.68	12.40±1.62
Nonwoven fibres, 4% NaOH & 0.05 M H_2SO_4	82.73±1.52	-2.08±0.14	6.23±0.64
Straw pulp	71.32±1.73	-1.45±0.03	5.02±0.19
Straw fibres	74.59±1.00	-0.87±0.85	10.25±2.13

4.7 Nonwoven and Textile Samples

The fibres spun at DR 8 with the large spinning unit were used to produce samples of airlaid nonwovens at VTT Technical Research Centre of Finland. All fibres could successfully be used to produce nonwovens, although the fibres were distributed somewhat unevenly. The process was easier for the fibres produced form recycled nonwovens, while the straw fibres had a longer airlaying time. The nonwoven sheets are depicted in Figure 32.

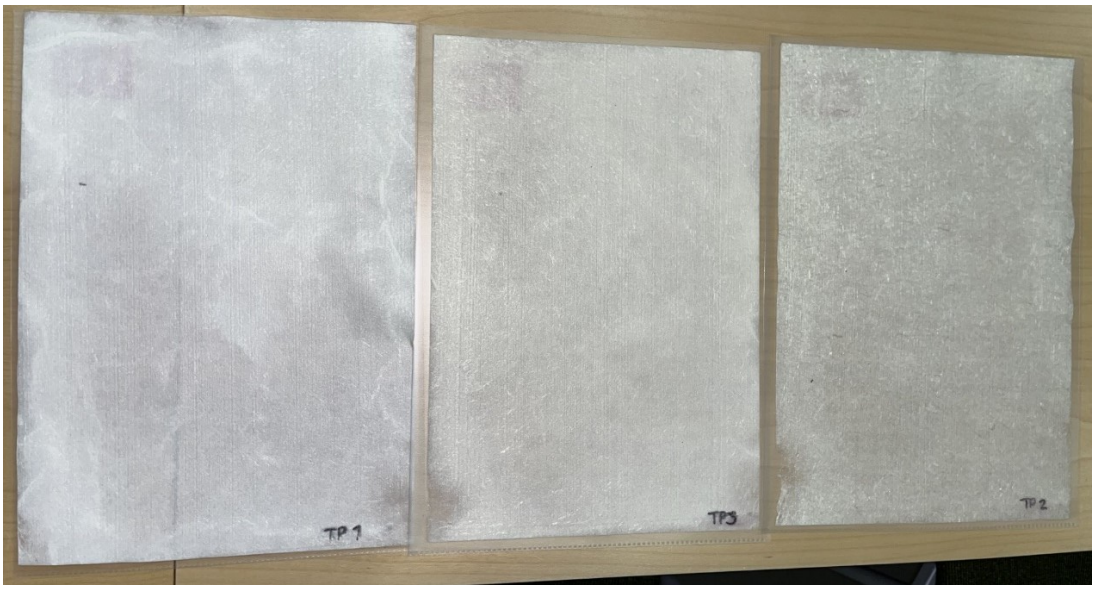

Figure 32. Nonwoven sheets produced by VTT from recycled nonwoven treated with 4% NaOH and 0.05 M H_2SO_4, recycled nonwoven treated with 4% NaOH, and straw pulp in this order.

The fibres prepared from recycled nonwoven treated with both NaOH and H_2SO_4 at DR 11 were also used to produce yarn and a sample of knitted fabric (Figure 33). Table 14 shows the properties of the produced yarn produced. The tenacity and elongation at break are slightly lower than reported by Sederholm (2022) for yarn produced from recycled Ioncell fibres (22.68 cN/tex; 7.71%), but slightly higher than achieved by Airaksinen (2023) from yarn produced from Ioncell fibres from recycled viscose (17.66 cN/tex; 5.81%). It should be noted that in addition to the properties of the used fibres, the properties of yarn depend also on factors such as twist of the yarn. Therefore, yarn properties are not fully comparable between different fibres, and the properties of the yarn produced in this study could be improved by optimising the yarn production process.

Table 14. Linear density (tex), elongation at break (%), and tenacity (cN/tex) of the yarn produced from recycled nonwovens.

Linear Density (tex)	Elongation at Break (%)	Tenacity (cN/tex)
27.9±1.2	6.1±1.0	19.2±4.7

Figure 33. Upcycling process from original nonwoven, to produced regenerated fibres, to yarns, to knitted fabric.

5 Future Research

In order to advance the research conducted in this thesis further, some additional analyses could be performed. The elemental composition of the starting materials and produced fibres could be analysed with Inductively coupled plasma optical emission spectroscopy (ICP-OES). This would be especially useful for the recycled nonwovens, because it would allow the determination of Ti-content in the material before and after the pre-treatments to quantify how much of the TiO_2 could be removed by the treatments. Furthermore, X-ray diffraction could be used to analyse whether the crystalline structure of the produced fibres is affected by the non-cellulosic impurities in both starting materials.

Future research could also focus on broadening the scope of this study by testing how different nonwovens would behave when recycled with the Ioncell process. Since the additives in other nonwovens can be different from the ones in this study, the pre-treatments may need to be adjusted for different substrates.

6 Conclusions

This study evaluated the production of man-made cellulosic fibres for production of nonwovens from cellulose sources alternative to wood. Regenerated cellulosic fibres were successfully produced with the Ioncell process using recycled nonwovens and straw pulp as raw material. These fibres were successfully used to produce nonwovens, and the fibres produced from recycled nonwovens were also upcycled by spinning them into yarn and producing a knitted fabric sample. The properties of the produced fibres were measured and the changes in chemical composition and polymer properties of the materials were evaluated throughout the process.

The recycling of nonwoven fabric with the Ioncell process was tested and optimised with a monofilament dry-jet wet spinning unit. The initial spinnability of the untreated nonwoven was poor due to the presence of unknown additives in the fabric. One of the additives was identified as a binder, as they are commonly used in nonwovens for attaching fibres together. TiO_2, a pigment commonly added to nonwovens as a mattifying and whitening agent, was suspected to be another additive in the nonwoven due to the opaque light orange colour of the cellulose-ionic liquid solutions. Various pre-treatments were tested on the nonwoven for the removal of these additives to thus improve the spinnability of the material. The most successful pre-treatment for binder removal proved to be immersing the ground nonwoven in 4% NaOH for 50 minutes with heating at 40 °C. The nonwoven could then be further treated with 0.05 M H_2SO_4 for 10 minutes with heating at 60 °C for removal of the TiO_2. The NaOH-treatment significantly improved spinnability with the monofilament unit and the H_2SO_4-treatment showed a further but less significant improvement. However, the colour of the solution remained the same, indicating that some but not all TiO_2 had been removed.

A multifilament unit was further used to produce fibres treated with only the NaOH and with both NaOH and H_2SO_4. Continuous spinning was reached at

draw ratio 8 with the former and draw ratio 11 with the latter. DR 11 produces thinner fibres with a lower linear density than DR 8, but thicker fibres are acceptable for nonwovens production allowing the use of fibres spun at DR 8. Furthermore, keeping TiO_2 in the fibres could be an advantage since this eliminates the need to add the pigment back in at a later stage. Therefore, it can be concluded that using only a NaOH-treatment is a viable option to produce fibres for nonwovens, while the addition of a H_2SO_4-treatment is an option for production of yarn and textiles, or if thinner fibres are needed. Further testing would be needed to evaluate the optimal conditions of the pre-treatments more specifically.

Straw pulp was spun with a multifilament dry-jet wet spinning unit using 12% and 13% cellulose concentrations. Stable spinning could not be reached with 12% concentration, but it was reached at DR 8 with 13% concentration. However, when spinning with 12% cellulose concentration, the spinning temperature was too low, and the dope was leaking from an untight spinning cylinder. These factors likely affected the outcome of the spinning trials, meaning a definitive conclusion cannot be made on which cellulose concentration is superior. Since only two spinning trials were performed, a more comprehensive study of the exact spinning conditions may further improve the spinnability and properties of the produced fibres.

The tenacities of the produced fibres were slightly lower than for standard Ioncell fibres and their elongations were similar or slightly improved. The decrease in tenacity is due to an elevated amount of inorganic impurities in comparison to standard wood pulp in the case of the nonwoven, and a mix of organic and inorganic impurities in the straw pulp. The fibres produced from straw pulp are stronger than the fibres produced from recycled nonwovens, because the molecular weight of the straw pulp was significantly higher. The tenacity and elongation of the fibres produced from straw pulp were also higher than in previous studies on producing regenerated fibres from straw, due to the use of a starting material with higher purity than in the other studies and the use of dry-jet wet spinning and the Ioncell process. The fibres

produced from nonwovens treated with both NaOH and H_2SO_4 had a slightly higher tenacity than the fibres produced from only NaOH-treated nonwoven, but the difference was only marginal. The tenacities of the recycled fibres were also respectable in comparison to fibres produced from recycled materials with the Ioncell process in previous studies.

Both straw pulp and recycled nonwoven produced fibres with higher tenacities than most commercially available man-made cellulosic fibres, with only NMMO-based Lyocell reaching similar values to the fibres produced from straw pulp. As Lyocell is the starting material of the nonwovens, this shows that the recycling process of the nonwovens has been successful with only a slight decrease in fibre properties. The fibres produced from either nonwovens or straw pulp also exhibited an improved wet-to-dry tenacity ratio in comparison to other MMCFs due to the increased fibre orientation achieved by dry-jet wet spinning. The fibres were also stronger than some synthetic fibres commonly used in nonwovens, such as polyethylene and nylon, while polyester and polypropylene fibres are stronger. The produced Ioncell fibres generally had lower elongation than other commercially available fibres due to the nature of the Ioncell process. Nonetheless, the performance of the fibres can be seen as respectable in relation to other commercially available fibres and can thus be concluded as suitable for production of nonwovens.

Overall, the results of this thesis show that both straw pulp and recycled nonwovens can be used to produce regenerated fibres with the Ioncell process. The produced fibres have competitive properties in relation to commercially available fibres and are suitable for production of nonwovens, as demonstrated by the preparation of airlaid nonwoven samples. Recycled nonwoven fabric could also be upcycled into yarn and knitted textiles. Further research is needed to evaluate the yield of the recycling of the nonwoven, the suitability of the pre-treatments on other types of nonwovens, and the effects of using a different ionic liquid that would be recyclable.

7 Project Partners

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9 Appendix

Table 15. Zero shear viscosity η_0 (Pa·s), angular frequency at the cross-over point Ω (rad/s), and loss modulus and storage modulus at the cross-over point G'=G" (Pa) measured at the appropriate spinning temperature (°C) of samples that did not have the best spinning behaviour.

Sample	Temperature (°C)	η_0 (Pa·s)	Ω (rad/s)	G'=G" (Pa)
NW, 10% cellulose #1, not sieved	65	2400	6.56	2100
NW, 10% cellulose #2, not sieved	65	Na	No COP	No COP
NW, 13% cellulose, not sieved	65	26 800	0.89	2600
NW, not sieved, 4% NaOH 50 min	65	16 800	1.55	3700
NW, 6% NaOH 60 min	65	9300	2.65	4200

Table 16. Average linear density (dtex), elongation at break (%), and tenacity (cN/tex) of rejected fibres with standard deviation. Values are shown in dry and wet conditions.

Fibre type	Condition	Linear density (dtex)	Elongation (%)	Tenacity (cN/tex)
Nonwoven, not sieved 4% NaOH, DR5	dry	2.7±0.5	6.8±1.7	29.3±7.3
	wet	2.8±0.6	10.0±2.2	27.4±7.0
Nonwoven, not sieved 4% NaOH, DR7	dry	2.1±0.6	5.9±1.3	30.4±6.9
	wet	2.2±0.5	7.7±1.5	25.6±6.0
Nonwoven, 4% NaOH, DR5	dry	2.6±0.2	10.4±1.3	36.1±2.2
	wet	2.5±0.3	13.9±2.0	35.7±2.6
Nonwoven, 4% NaOH, DR8	dry	1.8±0.3	8.9±1.1	40.8±4.7
	wet	1.7±0.2	11.9±1.8	35.9±3.8
Nonwoven, 4% NaOH + H_2SO_4 0.01 M, DR5	dry	3.3±0.7	8.5±1.7	31.8±4.1
	wet	3.0±0.5	12.8±2.7	28.8±5.6
Nonwoven, 4% NaOH + H_2SO_4 0.01 M, DR8	dry	2.4±0.4	7.0±1.6	28.9±4.5
	wet	2.4±0.4	9.5±2.8	26.6±4.9
Nonwoven, 4% NaOH + H_2SO_4 0.01 M, DR9	dry	3.1±0.7	6.1±1.1	25.8±5.4
	wet	3.4±0.6	8.4±2.1	22.9±4.6

Table 17. Parameters used in production of nonwovens at VTT.

Mass of fibres (g)	Mass of fibres on forming wire (g)	Temperature (°C)	Relative humidity (%)	Rolling before hydroentanglement (once back and forth)	Binding	Drying temperature (°C)
5.42	5.36	21.0	45.0	Yes	Hydroentanglement: pre-wetting 1x10 bar + 1x20 bar + 1x40 bar + 1x60 bar (both sides of the sheet)	105
5.19	5.07	21.0	43.3			
5.26	5.19	21.5	43.5			

Fibres	Target grammage (g/m^2)	Fibre laying method	Opening of fibres	Make-up air (mm)	Rotation speed of the drum (rpm)	Airlaying time (min)
Nonwoven, NaOH (4% 50 min) & H_2SO_4 (0.05M 10min)	80	Airlaying	Blender and compressed air	30	60	3
Nonwoven, NaOH (4% 50 min)						3
Straw pulp (12% cellulose)						12