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A Progress on Nanocellulose as Binders for Loose Natural Fibres

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Abstract: Nanocellulose can be obtained by two ways: extracting from wood to be nanofibrillated cellulose (NFC) and synthesised from bacterial cellulose (BC). Typically, the use of nanocellulose is for coating of fibres or reinforcement of polymer to make nanocomposites. This nanocellulose had been developed to be binders for natural fibre non-wovens. This binder has function to hold the loose fibres together so that the mechanical properties are improved. This article reviews the use of nanocellulose as binder for improving the mechanical properties of nonwoven natural fibre preforms. The manufacturing process, the comparison to conventional polymer binders and the potential of this in the future are also discussed.

Keywords: nanocellulose, bacterial cellulose, nano-fibrillated cellulose, binder, non-woven, natural fibre

1. Introduction

The growing demand of greener products shows a significant increase due to the global problems such as global waste, high oil price and environmental of landfill sites ¹⁻³. There are many efforts having been conducted in the area of polymers by adding natural fibres as the reinforcement to manufacture environmentally-friendly composite materials. Natural fibres are likely to be promising candidate for polymers as the replacement of glass fibres to produce greener composites due to its high specific stiffness and strength, wide availability and also renewability ⁴⁻⁷. Though natural fibres have been used in some applications such as automotive ^{8,9}, indoor element in housing ¹⁰ and interior panelling for train ¹⁰, this fibre still has drawbacks including incompatibility with hydrophobic polymers ¹¹⁻¹³, low thermal stability ¹⁴⁻¹⁸ and inherent variability in dimensions and mechanical properties ^{9,19-21}. A little effort can only be done for solving the latter problem. However there has been considerable research effort conducted to tackle with hydrophilic properties of natural fibres by surface modification to enhance fibre-matrix interface ²²⁻²⁶. Two of popular treatments (despite having been old treatments) for natural fibres is alkaline treatment ²⁷⁻³¹ and silane treatment ³²⁻³⁵.

Another effort that can be done to enhance the properties of natural fibres is by forming non-woven preforms from natural fibres ³⁶⁻³⁹. Processing natural fibres into non-wovens (or called preforms) can be done by braiding the yarns using needle-punching equipment

⁴⁰⁻⁴⁴. Non-wovens have good mechanical entanglement from cellulosic fibres and even can be utilised without matrix or resin. However, in order to get better bonding among the fibres, natural fibres non-wovens are commonly combined with the binders made of polymers (thermosets and thermoplastics)^{45,46}. In other words, the use of binder actually has the same role as surface treatment for natural fibres which may increase not only the bonding between fibres and matrix during impregnation with polymer but also the entanglement among fibres. However, the use of conventional polymers evokes some problems including the incompatibility with natural fibres since polymers usually have hydrophobic properties.

In order to get green non-woven preforms, another alternative material can be used as binder. Nanocellulose which is recently investigated for coating natural fibres to enhance the fibre-matrix interface properties ⁴⁷⁻⁵⁰ has been developed to bind loose natural fibres to replace the role of conventional polymers as binders. There have been works focusing on this with utilising the hydrophilic behaviour of natural fibre to absorb the water in nanocellulose dispersion, so the nanocellulose is allowed to spread along the fibres ⁵¹ and will improve the bonding between the fibres in non-woven preforms which enhance the mechanical performance as well.

This review focuses on the recent progress in the development of nanocellulose as binders for upgrading the properties of natural fibre non-wovens. In section 2, the types of nanocellulose based on the sources and how it can

be obtained are revealed. In section 3, the effective methods to manufacture natural fibre non-wovens with the addition of nanocellulose binders are discussed. The comparison of nanocellulose and polymers as binders is also presented as well as the conclusion at last.

2. Nanocellulose

There are two ways of producing nanocellulose, bottom-up by biosynthesis of bacteria and top-down by disintegration of plant materials ^{52,53}. In terms of biosynthesis of bacteria, nanocellulose resulted is called bacterial cellulose (BC) which can be produced by two methods ⁵³. The first method is static culture resulting white BC pellicle at the air-liquid from the accumulation of a thick which has structure like leather ⁵³. The second method is stirring culture which can form suspended fibre or irregular pellets based on synthesis ⁵⁴. The latter method is known as the first method to produce BC introduced by Brown ¹. According to Lee et al. ⁵², BC has lateral dimension of 25-86 nm and is highly crystalline

nano-sized cellulose (see figure 1). It has degree of crystallinity of up to 90% without the impurities such as hemicellulose and lignin ⁵⁵⁻⁵⁸. Moreover, BC has young's modulus of up to 114 GPs for a single fibre ^{59,60}. These properties have attracted many researcher to investigate its potential, especially in the area of tissue engineering ^{55,61}, biomedical engineering ⁶² and advanced composites ⁶³. BC can be used in various biomedical applications such as implants and scaffolds, drug delivery, and wound dressing materials ⁶⁴.

Regarding nanocellulose produced using disintegration of plant materials, it is usually termed nano-fibrillated cellulose (NFC). There are techniques to extract NFC from plants. One of techniques is by using ultrasound to disintegrate larger bundles of natural fibres into small fibrils ^{52,65-69}. Another technique is the use of high-pressure homogeniser investigated by Turbak et al. ⁷⁰ to decrease the size of wood fibres into the nanometre scale. Generally, NFC has similar diameter to BC at about 100 nm. However, it has lower critical surface energy (42 mN/m) than BC (57 mN/m) due to its impurities which are

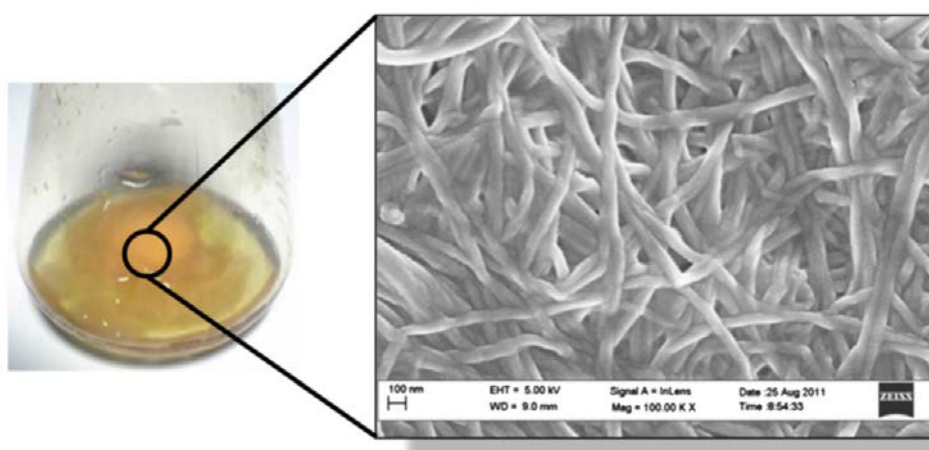


Fig. 1: Gel-like pellicle of BC and the appearance of pellicle under Scanning Electron Microscope (SEM) ⁷¹ with permission of ACS Publication.

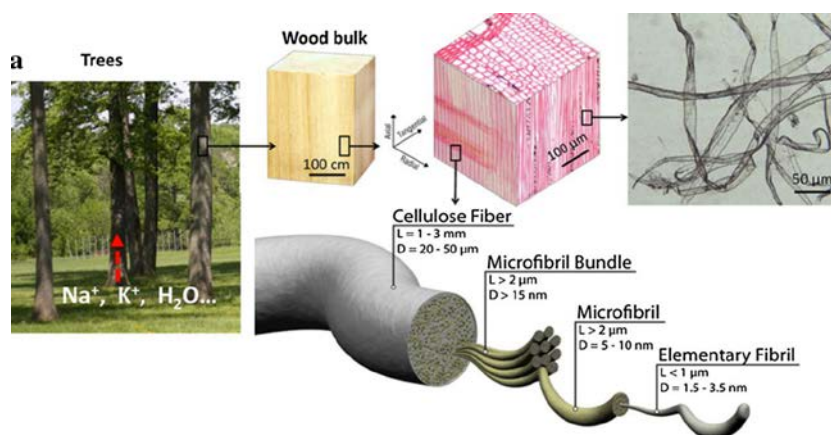


Fig. 2: Wood fibre structure ², with permission of ACS.

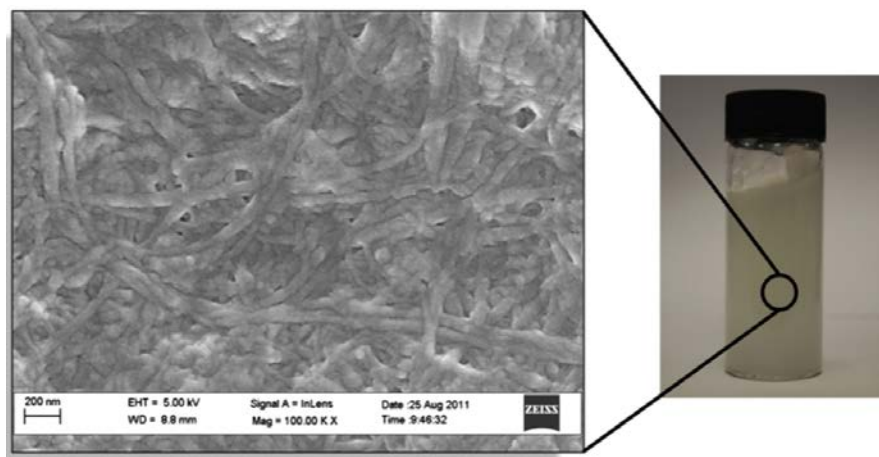


Fig. 3: NFC suspension ⁷¹⁾ with permission of ACS.

affected by the presence of lignin and hemicellulose ^{72,73)}. Figure 2 shows the hierarchical structure of wood fibres followed by NFC suspension in water investigated using SEM (figure 3). NFC is also good as electrical insulation proven by Wang et al, ⁷⁴⁾. Due to good mechanical properties and electrical properties, the potential application of NFC is not only in the structural applications but also collapsible electronic devices ⁷⁴⁾.

3. Natural Fibre Non-wovens with Nanocellulose Binders

The use of BC as binders was developed by Lee et al. ^{75,76)} using simple papermaking process to produce non-woven sisal fibre preforms. The manufacturing of fibre preforms was conducted by soaking sisal fibres in BC dispersion overnight, followed by dewatering, cold compression and hot compression at 120°C for 4h to consolidate hornified nanocellulose network to form BC-sisal fibre preforms. The results show that the addition of 10 wt.% BC as binders increases tensile strength to $13.1 \pm$

2.1 kNm^{-1} and decreases porosity to 61% as shown in table 1. Tensile index presented in table 1 is the ratio of tensile strength (N/m) and grammage (m^2/g) in which grammage is the areal density of non-wovens. Grammage usually uses to measure the density of paper. In addition, as shown in figure 4 that without the addition of BC binders, the natural fibres are loose and cannot be used to withstand load.

Fortea-Verdejo et al. ⁷⁷⁾ followed the process above to produce BC-flax fibre preforms and also used another nanocellulose type, nano-fibrillated cellulose (NFC) which is produced from wood via top-down approach ⁵²⁾, and pulp to bind loose flax fibres. Two different filtration methods were also compared in this work: single filtration method (used by Lee et al. ^{75,76)} as well) and a layer-by-layer filtration technique. The difference of single-step and a layer-by-layer filtration method is shown in figure 5. The results show that nanocellulose (both BC and NFC) totally surpassed pulp as binder for flax nonwovens since pulp has lower surface area which leads to lower contact area between pulp and flax fibres.

Table 1. Properties of natural fibres using 10 wt.% of nanocellulose or other materials as binders

Fibres	Binder	Tensile strength (kNm^{-1})	Tensile strength (MPa)	Tensile index (N m g^{-1})	Flexural strength (MPa)	Flexural modulus (GPa)	Porosity (%)
Sisal ^{75,76)}	BC	13.1 ± 2.1	-	-	-	-	61 ± 3
Flax ⁷⁷⁾	BC	6.5 ± 1.5	4.4 ± 0.9	7.5 ± 1.9	9.0 ± 1.7	0.8 ± 0.3	63.1 ± 4.2
Flax ⁷⁷⁾	NFC	6.3 ± 0.8	5.0 ± 0.7	7.4 ± 1.2	7.0 ± 1.8	515 ± 150	57.7 ± 2.9
Flax ^{77)*}	BC	17.6 ± 2.5	12.7 ± 1.1	19.6 ± 2.3	14.6 ± 4.1	1501 ± 550	57.1 ± 2.2
Flax ⁷⁷⁾⁽²²⁾	PVA	1.8	-	4.6	-	-	83 ± 3
Flax ⁷⁸⁾	PP	5.4	-	14	-	-	-

*manufactured by a layer-by-layer filtration method; PVA: Polyvinylalcohol



Fig. 4. Sisal fibres without the addition of BC (top) and sisal fibres with the addition of BC (bottom) with permission of JoVE ⁷⁶.

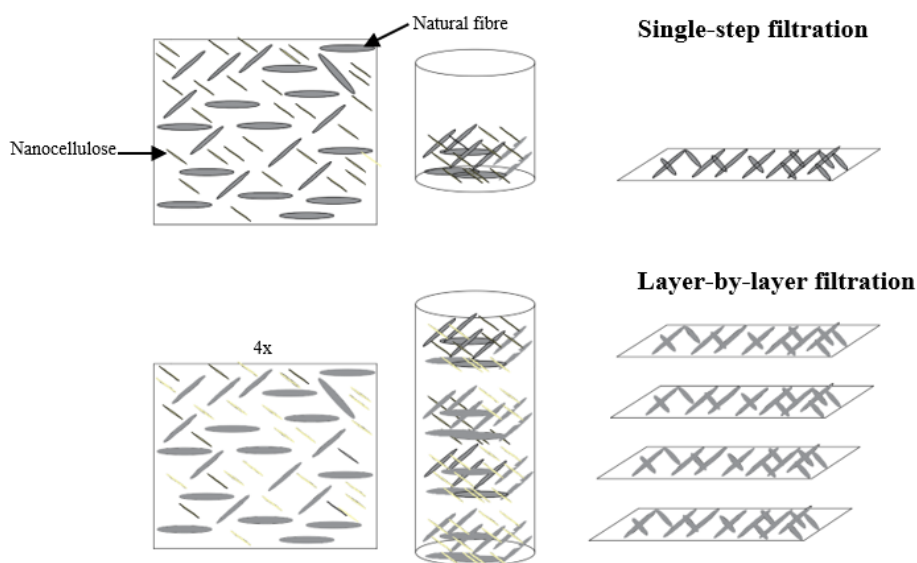


Fig. 5. Single-step and layer-by-layer filtration methods to produce natural fibre non-wovens with the addition of nanocellulose.

It can be seen in table 1 that BC-flax fibre preforms manufactured by a layer-by-layer filtration method had higher mechanical properties since the distribution of fibrous nanocellulose network throughout the non-woven preforms is more uniform ^{77,79}. As shown in table 1, BC-sisal fibre preforms possesses higher mechanical properties than BC-flax fibre preforms having the same filtration method. However, once it is compared to BC-flax fibre preform with a layer-by-layer filtration method,

mechanical properties of BC-sisal preform were lower. This means that the mechanical properties of the nonwovens are not strongly dependent on the types of natural fibres but on the nanocellulose network because single-step filtration method has non-uniform nanocellulose network while layer-by-layer filtration method has more complex step and more thorough to ensure homogenous nanocellulose network which results better mechanical properties ⁷⁷. Furthermore, the

properties of all-natural nonwovens using nanocellulose as binders outperformed flax nonwovens using PVA binder which means that the addition of nanocellulose is more effective (table 1).

4. Concluding Remarks

The use of nanocellulose has been developed not only to coat natural fibres but also to be binders for the natural fibre nonwovens. This is agreed that the use of nanocellulose (from any source) can upgrade the properties of natural fibre preforms. A layer-by-layer filtration method is capable of enhancing the properties of the nonwovens to the next level. It is also remarked that the properties of natural fibres preform are highly influenced by the nanocellulose network structure, no matter the types of natural fibres used. The addition of nanocellulose as binder is also better than of conventional polymer (PVA) as binder. Using those method for binding natural fibre, nanocellulose does not only act as binder but also as nano-reinforcement which will enhance the mechanical performance of composites while impregnated into the resin. As suggestion, those methods can be used in other fibres (not only restricted to natural fibre) as long as it has hydrophilic nature which can absorb water in nanocellulose dispersion which makes nanocellulose spread along the fibres when fibres are soaked in nanocellulose dispersion. The layer of nanocellulose coating will be formed when the fibres are dried. The other potential fibres that can be used with the combination of nanocellulose binders are wood flour and recycled paper.

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