

## NANO-FIBRILLATED CELLULOSE VS BACTERIAL CELLULOSE: REINFORCING ABILITY OF NANOCELLULOSE OBTAINED TOP- DOWN OR BOTTOM-UP

K.-Y. Lee<sup>1</sup>, T. Tammelin<sup>2</sup>, H. Kiiskinen<sup>3</sup>, J. Samela<sup>3</sup>, K. Schluffer<sup>4</sup>, A. Bismarck<sup>1\*</sup>

<sup>1</sup>Polymer and Composite Engineering (PaCE) group, Department of Chemical Engineering, Imperial College London, South Kensington Campus, SW7 2AZ

<sup>2</sup>VTT Technical Research Centre of Finland, P.O. Box 1000, FIN-02044, Finland

<sup>3</sup>VTT Technical Research Centre of Finland, P.O. Box 1603, FIN-40101, Jyväskylä, Finland

<sup>4</sup>Forschungszentrum Medical Technology and Biotechnology GmbH, D-99947 Bad Langensalza, Germany

\*a.bismarck@imperial.ac.uk

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### Abstract

*This work investigates the difference between nanofibrillated cellulose (NFC) and bacterial cellulose (BC). BC possesses higher critical surface tension of  $57 \text{ mN m}^{-1}$  compared to NFC ( $41 \text{ mN m}^{-1}$ ). The thermal degradation temperature of BC was also found to be higher than that of NFC, in both nitrogen and air atmosphere. These results are in good agreement with the higher crystallinity of BC based on area under the XRD spectra, measured to be 71% as compared to NFC of 41%. Both types of nanocellulose papers were found to possess similar tensile moduli and strengths of 12 GPa and 100 MPa, respectively. Nanocomposites were manufactured by infusing the nanocellulose paper with an epoxy resin using vacuum assisted resin infusion. The cellulose reinforced epoxy nanocomposites were found to possess a stiffness and strength of approximately  $\sim 8 \text{ GPa}$  and  $\sim 100 \text{ MPa}$  at an equivalent fibre volume fraction of 60 vol.-%. In terms of the reinforcing ability of NFC and BC in a polymer matrix, no significant difference between NFC and BC was observed.*

### 1 Introduction

Cellulose is used in the paper [1], pharmaceutical and cosmetic industry [2, 3], and more recently it is explored as reinforcement for polymers [4, 5]. Numerous products are also derived from cellulose; technical textile fibres, such as viscose and Lyocell [6] and thermoplastic polymers, such as cellulose acetate. Currently, much research activity and attention focused on the isolation and production of nano-scale cellulose fibres. For comprehensive reviews on the production and application of nanocellulose, the readers are referred to publications by Klemm et al. [7] and Siró et al. [8]. Interests in nanocellulose come from the fact that nano-scale cellulose combines the physical and chemical properties of cellulose, such as hydrophilicity and the ability to be chemically modified by a broad range of reactions, with the features of nanomaterials – high specific surface area and aspect ratio.

Nanocellulose can be obtained by two approaches: top-down and bottom-up. The top-down approach involves the disintegration of (ligno)cellulose biomass, such as wood fibres into nanofibres. This technique was first reported by Herrick et al. [9] and Turbak et al. [10], whereby wood pulp was fed through a high-pressure homogeniser to reduce the size of the fibres down to the nano-scale. A more recent method of producing nanocellulose from plant-based cellulosic fibres involves using of grinders [11], whereby wood pulp is passed through the slit between a static and rotating grindstone. This high shear fibrillation process converts micrometre-sized cellulose into nanocellulose. Herein, we term these plant derived nano-scale cellulose nanofibrillated cellulose (NFC). Nanocellulose produced via the bottom-up approach utilises the fermentation of low molecular weight sugar using cellulose-producing bacteria, such as from the *Acetobacter* species, to produce nanocellulose [2, 12-14]. These nanocellulose, herein termed bacterial cellulose (BC), is pure cellulose without the presence of pectin or lignin [15]. The cellulose is excreted by the bacteria into the aqueous culture medium as nanofibres, with a diameter ranging from 25 – 100 nm [13, 15]. These nanofibres makes up the pellicles in the culture medium [13].

Utilising nanocellulose as filler in polymer matrices was first reported by Favier et al. [16] The authors reinforced latex (styrene-butyl acrylate copolymer) with cellulose nanowhiskers derived from tunicin to produce polymer nanocomposites. Since then, studies on utilising nanocellulose as filler in polymer matrices have increased significantly over the years. The major driver for this is the potential of exploiting the high stiffness of cellulose crystals. X-ray diffraction and numerical simulations estimated the stiffness of a cellulose crystal to be approximately 100-160 GPa [17, 18], which is highly desirable as reinforcing filler for polymer matrices. However, it is not clear what is the true crystal modulus of cellulose nor its maximum attainable stiffness as reinforcing filler [19]. Nonetheless, nanocellulose has been shown to improve the mechanical performance of the resulting nanocomposites. Yano et al. [20] have obtained a tensile modulus and strength of up to 21 GPa and 325 MPa, respectively, for a BC sheet reinforced epoxy resin with a nanocellulose loading of 70 wt.-%. However, the authors failed to explain why the tensile strength of the nanocomposites was 25% higher than that of the reinforcing BC sheets. Nevertheless, this study showed that high strength nanocomposites can be produced using BC sheets. Laminated BC-poly lactide nanocomposites at a loading fraction of 18 vol.-% have also been studied [21]. The tensile modulus doubled and the strength tripled when compared to neat poly lactide. NFC has also shown to improve the mechanical properties of the nanocomposites. Henriksson et al. [22] produced NFC sheet reinforced hexamethoxymethyl melamine-based nanocomposites; at a volume fraction of 55 vol.-%, a Young's modulus of 9.7 GPa and tensile strength of 108 MPa was achieved. The neat polymer possesses a Young's modulus and tensile strength of only 3.6 MPa and 6.1 MPa, respectively.

It is evident that BC and NFC have the ability to act as reinforcement for the production of high strength and stiffness materials. However, there are currently no systematic studies reporting the surface and bulk properties of NFC and BC, and the reinforcing capability of these two types of nanocellulose for composite materials. With nanocellulose gaining significant research interest and wide availability<sup>†</sup>, it is important to quantify the differences, if any, between NFC and BC. Therefore, the aim of this work is to elucidate the differences in terms of their wettability,  $\zeta$ -potential, crystallinity, thermal degradation behaviour and

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<sup>†</sup> NFC can be obtained from wood pulp via grinder or high-pressure homogenizer. fzmb GmbH is currently producing 30 tonnes per annum of BC.

mechanical properties of NFC and BC. This article also demonstrates the large scale manufacturing of nanocomposites using commercially available vacuum assisted resin infusion, which is intrinsically scalable and discusses the reinforcing ability of NFC and BC in nanocomposites applications.

## 2 Materials and methods

Ultra low viscosity epoxy resin (PRIME 20ULV, Gurit Ltd, Isle of Wight & Hamble, UK) was used as the matrix for the nanocomposites. BC was kindly supplied by fzm GmbH (Bad Langensalza, Germany) in wet pellicle form containing 94 wt.-% water. NFC was produced by grinding of never-dried bleached birch Kraft pulp (*Betula pendula*). The grinding of birch pulp was conducted using a Masuko Mass Colloider (Masuko Sangyo Co., Kawaguchi, Japan). The pulp was passed through the grinder seven times and the final consistency of the aqueous gel-like NFC was approximately 2 wt.-%.

### 2.1 Large scale manufacturing of BC and NFC sheets.

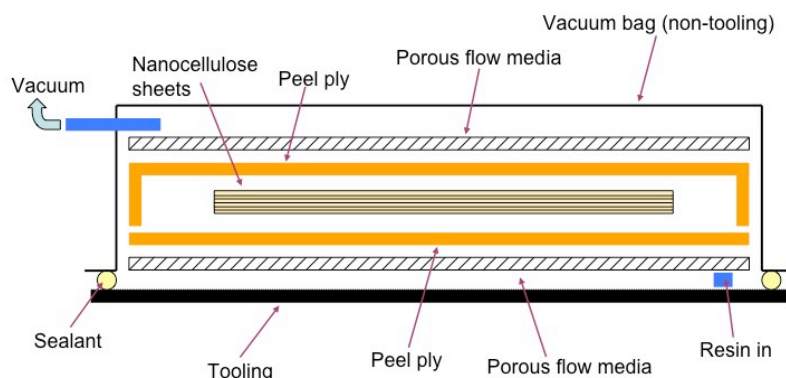
BC and NFC papers with a grammage of 60 g m<sup>-2</sup> were produced using a home-made vacuum assisted sheet former equipment designed by VTT in cooperation with Metso Paper. Firstly, the BC pellicles were cut into small pieces and blended for 2 min using a blender (Breville BL18 glass jug blender, Pulse Home Products Ltd, Oldham, UK) at a consistency of 0.1 wt.-%. The NFC suspension was adjusted to 0.4 wt.-% from 2 wt.-% consistency and blended for 2 min using the blender to produce a homogenous dispersion of nanocellulose in water. These nanocellulose suspensions were then vacuum filtered onto a filter. The filter cake was then wet pressed twice under a weight of 10 kg between bloating papers for 10 s. The partially dried nanocellulose sheets from the wet pressing steps were then sandwiched between bloating papers and metal plates under a weight of 10 kg during the drying process in the oven held at 55°C for at least 48 h to dry. This was to prevent the nanocellulose sheets from shrinking during the drying process. Shrinkage of cellulose sheets will induce flexibility in the fibre network and decrease the load bearing capability of the resulting sheets [23].

### 2.2 Manufacturing of BC or NFC paper reinforced nanocomposites.

The nanocellulose paper reinforced epoxy was produced using vacuum assisted resin infusion (VARI). A schematic diagram of the VARI lay-up is shown in figure 1. A polyester porous flow medium (15087B, Newbury Engineer Textile, Berkshire, UK) was placed on top of the tooling side (a 460 mm x 920 mm heating plate with a temperature control unit), which consisted of a layer of polyester film (Melinex PW 122-50-RL, PSG group, London UK). 11 nanocellulose sheets were laid up and sandwiched between two PTFE coated glass release fabrics (FF03PM, Aerovac, West Yorkshire, UK) and placed on top of the polyester porous flow medium. Another polyester porous flow medium was then placed on top of the release fabric. The whole setup was covered with a vacuum bagging film (Capran 519 heat stabilised Nylon 6 blown tubular film, Aerovac, West Yorkshire, UK) and sealed using a vacuum sealant tape (SM5127, Aerovac, West Yorkshire, UK).

Prior to the infusion, the epoxy and hardener were mixed thoroughly at a ratio of 100:19 by weight and degassed at room temperature under a reduced pressure of 75 mmHg for 5 min. This ensures all air bubbles trapped during the mixing process were removed. The infusion process starts with an air removal step, whereby a vacuum was applied to the system via the

tubing on the non-tooling side with the resin inlet tubing sealed off. When the maximum vacuum was reached ( $\sim 15$  mmHg), the VARI setup was left under this vacuum for 2 h to ensure that there was no leakage in the set up by constantly monitoring the pressure in the vacuum bag. Once the system was determined to be leakage-free, the liquid resin was fed at room temperature from the bottom of the polyester porous flow medium on the tooling side through the nanocellulose sheets and exit via the tubing on the non-tooling side. The inlet and outlet of the system were sealed off again when the resin fully impregnated the nanocellulose sheets. The resin was left to cure at room temperature for 24 h, followed by a post-curing step conducted at 50°C for 16 h.



**Figure 1.** Schematic diagram of the VARI process.

### 2.3 Scanning electron microscopy (SEM)

SEM was used to study the morphology of NFC and BC nanofibres. It was performed using a high-resolution field emission gun scanning electron microscope (LEO Gemini 1525 FEG-SEM, Oberkochen). The accelerating voltage used was 5 kV. Prior to SEM, the nanocellulose was solution cast from water onto carbon tabs stuck on the SEM stubs, air dried and Cr coated (K550 sputter coater, Emitech Ltd, Ashford, Kent, UK) for 1 min at 75 mA.

### 2.4 Tensile properties of the nanocellulose sheets and nanocomposites

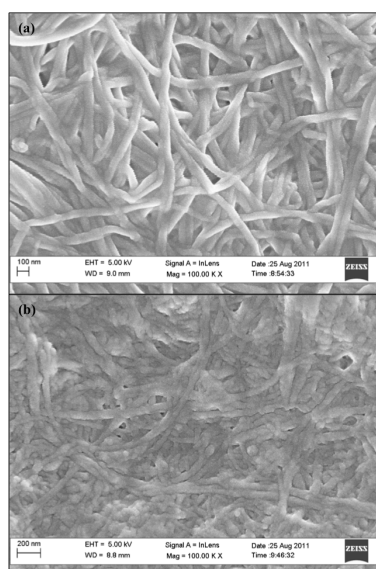
Nanocellulose sheets were cut into dog bone shape specimens using a Zwick cutter. The test specimen possesses an overall length of 35 mm and the narrowest part of the specimen is 2 mm. Prior to the test, the specimens were secured onto testing cards using a two-part cold curing epoxy resin (Araldite 2011, Huntsman Advanced Materials, Cambridge, UK). This was to prevent the clamp of the tensile testing equipment from damaging the test specimens. Tensile test was conducted using a TST350 tensile tester (Linkam Scientific Instruments, Surrey, UK). The load cell and crosshead speed used were 200 N and 1 mm min<sup>-1</sup>, respectively. The sample thickness was determined using a digital micrometre. A total of 5 specimens were tested for each type of nanocellulose. The machine compliance was determined to be  $7.19 \times 10^{-3}$  mm N<sup>-1</sup>.

The nanocomposites were tested in tension using an Instron universal material testing equipment (Instron 4505, Instron Corporation, MA, USA). The tensile test was conducted in accordance to ASTM D3039-00 using a load cell of 10 kN. The test specimens possessed dimensions of 100 × 10 × 1 mm, with a gauge length of 40 mm. Prior to the test, woven glass fibre reinforced polyester end tabs with 1.6 mm thickness were glued onto the samples using a two-part cold curing epoxy resin (Araldite 2011, Huntsman Advanced Materials, Cambridge, UK). The distance between the end tabs were 60 mm. Strain gauges (FLA-2-11, Techni

Measure, Studley, UK) were glued onto the middle portion of the test specimen using cyanoacrylate glue (EVERBUILD Building Products Ltd, Leeds, UK). Specimens were tested until failure at a crosshead speed of 1 mm min<sup>-1</sup>. A total of 5 specimens were tested for each type of nanocomposites.

### 3 Results and discussion

#### 3.1 Morphology of NFC and BC



**Figure 2.** Scanning electron micrographs of (a) bacterial cellulose and (b) nanofibrillated cellulose

The morphology of the two different forms of nanocellulose (solution cast from 0.05 wt.-% concentration) was studied using scanning electron microscopy (SEM) (see figure 2). Both types of nanocellulose possess a fibrous structure with dimensions of approximately 50 nm in diameter and several micrometres in length. It can also be seen that the diameter of the nanofibres are very uniform. This is not surprising for BC as its production is well-controlled by the biosynthesis of cellulose producing bacteria, whereby the nanocellulose is spun out by bacteria and assembled into the ribbon-shaped nanofibres.[24] The production of NFC, on the other hand, was carried out by grinding birch Kraft pulp using Masuko Mass Colloider. The pulp was passed through the grinder several times to ensure a uniform fibre diameter [25]. NFC contains approximately 25% of amorphous xylan, which has an ability to enhance the fibrillation procedure. In addition to this, both the NFC and BC sheets are translucent. The NFC sheet is more transparent compared to BC sheets. This could be due to the better disintegration and distribution of individual nanofibres within the sheet structure. The anionic charges on the NFC surfaces prevented the agglomeration of NFC in the suspension and thus, more evenly distributed in the sheets. In addition to this, the slight difference in thickness may affect the transparency of the sheets [26]. BC and NFC sheets made from suspensions (grammage of 60 m<sup>2</sup> g<sup>-1</sup>) were 79 μm and 64 μm thick, respectively.

#### 3.2 Tensile properties of sheets and nanocomposites

Both types of nanocellulose sheets possess a stiffness and strength of approximately 12 GPa and 110 MPa (see table 2), respectively. These values are consistent with the mechanical properties of nanocellulose sheets studied by Henriksson et al. [22] and Iwamoto et al. [27] At first glance, it seems surprising that BC sheets, which have a much higher degree of

crystallinity, possess almost the same tensile properties as NFC. However, BC paper is more porous compared to NFC paper. The porosity of BC paper was found to be 52% compared to NFC paper of only 38%. The oxygen transmission rates through the sheet structures suggest the same feature. The sheets prepared using NFC possess lower oxygen transmission rates indicating more densely packed fibrillar structure compared to the BC sheets. It is postulated that the more crystalline nature of BC led to a decrease in the packing efficiency of the nanofibres. Nevertheless, it is quite remarkable that BC papers with such a high porosity possess good mechanical properties. In addition to the difference in porosity of the papers, the presence of hemicellulose also affects the mechanical properties of NFC papers. Hemicellulose aids the nanofibres to bonding together forming a composite [27, 28]. As a result, NFC with lower crystallinity and porosity possesses a similar strength and stiffness to BC (higher crystallinity and porosity). BC, on the other hand, has a higher strain-to-failure at 7.5% compared to NFC of only 4.2%. The lower strain-to-failure of NFC papers could be due to fewer physical crosslink points between the nanofibres, which allows for the realignment of the fibres during tensile loading. In addition to this, the presence of hemicellulose could also lead to the reduction in the strain-to-failure of NFC sheets. Although hemicelluloses can improve the bonding within the nanofibres, which is postulated to be the results of film forming ability and natural affinity towards cellulose, the distinctive feature of the hemicellulose films is their brittleness [29]. The tensile work of fracture ( $W_A$ , calculated from the area under the stress-strain curve) was also smaller for NFC papers compared to BC papers. This could be due to the higher strain-to-failure of BC papers, which allows for more energy to be absorbed during tensile loading.

**Table 1.** Mechanical properties and the properties of NFC and BC sheets.  $E_{\text{sheet}}$ ,  $\sigma_{\text{sheet}}$ ,  $\epsilon_{\text{sheet}}$  and  $W_A$  indicate Young's modulus, tensile strength, elongation at break, true density of the sheet and work of fracture, respectively.

Types of nanocellulose	$E_{\text{sheet}}$ (GPa)	$\sigma_{\text{sheet}}$ (MPa)	$\epsilon_{\text{sheet}}$ (%)	grammage ( $\text{g m}^{-2}$ )	$W_A$ ( $\text{MJ m}^{-3}$ )
NFC	$12.8 \pm 1.4$	$103 \pm 13$	$4.2 \pm 0.8$	$59.5 \pm 0.4$	$3.8 \pm 0.7$
BC	$12.0 \pm 1.1$	$123 \pm 7$	$7.5 \pm 0.6$	$57.2 \pm 3.2$	$5.8 \pm 0.6$

A stack of 11 sheets of NFC and BC paper were resin infused with an ultra low viscosity epoxy resin. This resin was chosen to ensure that the liquid will impregnate the nanocellulose papers. The surface tension of the liquid epoxy resin (with hardener) was measured to be  $32.3 \pm 0.1 \text{ mN m}^{-1}$ . This liquid resin should fully wet both BC and NFC papers as the critical surface tension of the papers was higher than the surface tension of the resin. The manufactured nanocomposites possess different fibre volume fractions ( $v_f$ ). In order to be able to compare between the nanocomposites, the Young's moduli of the nanocomposites were normalised to an equivalent fibre volume fraction of 60 vol.-% (see table 3). Both NFC and BC exhibited excellent reinforcing ability when used in nanocomposites as paper form due to the presence of cellulose network in the reinforcing paper. The Young's modulus of the nanocomposites increased to 8 GPa compared to that of the neat resin of only 3 GPa. This increase is inline with the values calculated using the "rules-of-mixtures" for composites, indicating that the maximum possible Young's modulus of the material was achieved. There is a slight difference in the reinforcing ability between NFC and BC when the tensile strength ( $\sigma$ ) was compared. It seems that BC reinforced nanocomposites possesses higher  $\sigma$  compared to NFC reinforced nanocomposites, even when the former possesses slightly lower  $v_f$ . We attribute this to the higher surface energy of BC, which promotes better adhesion between the matrix and the reinforcing fibre

**Table 2.** Fibre volume fraction ( $v_f$ ) and the mechanical properties of the nanocomposites.  $E$ ,  $E_{\text{norm}}$ ,  $\sigma$  and  $\epsilon$  indicate the Young's modulus, normalised Young's modulus to 60 vol.-% and tensile strength of the nanocomposites, respectively.

Types of nanocellulose	$v_f$ (vol.-%)	$E$ (GPa)	$E_{\text{norm}} = (E / v_f) \times 0.6$ (GPa)	$\sigma$ (MPa)
Neat resin <sup>&amp;</sup>	0	3.0	-	71
NFC nanocomposites	$58 \pm 1$	$8.5 \pm 0.2$	$8.8 \pm 0.2$	$96 \pm 1$
BC nanocomposites	$49 \pm 2$	$7.1 \pm 0.1$	$8.7 \pm 0.2$	$102 \pm 1$

#### 4 Conclusions

Nanocellulose obtained top-down (NFC) or bottom-up (BC) was studied and compared in this work. SEM showed that both types of nanocellulose possess a fibrous structure of approximately 50 nm in diameter and several micrometre in length. Both the NFC and BC papers were found to possess similar tensile properties; a Young's modulus of ~12 GPa and tensile strength of ~110 MPa. When used as reinforcement in an epoxy matrix, the nanocomposites were found to possess a high stiffness and strength of approximately ~8 GPa and ~100 MPa, respectively at an equivalent fibre volume fraction of 60 vol.-%. However, no significant difference was observed between the reinforcing ability of NFC and BC in terms of the stiffness of the nanocomposites. The nanocomposites reinforced with BC papers, however, showed slightly higher tensile strength compared to NFC papers by approximately 6%. Nonetheless, the difference between the tensile strength of the nanocomposites reinforced by NFC and BC is not very significant. This implies that both NFC and BC will serve as excellent reinforcing material for the production of nanocomposites.

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