

NANO-CELLULOSE REINFORCED POLYMERS DERIVED FROM BANANA TREE AND OTHER FIBRE SOURCES

P.Hornsby^{1*}, M. Mohamad¹, C.McMullan¹
S.Sharma² and E.Carmichael²

¹ *School of Mechanical and Aerospace Engineering, Queen's University Belfast, UK*

² *Applied Science Division, Agri-Food and Bioscience Institute, Newforge Lane, Belfast, UK*

* *peter.hornsby@qub.ac.uk*

Keywords: Cellulose nanofibres; Polymer reinforcements; Microfibrillation

Abstract

Consideration is given to the formation of cellulose microfibrils from different fibre feedstocks using combinations of chemical and mechanical treatments, based on microfibrillation. Examples will be presented showing the influence of preparation method on the structure of the fibres and their effect on the physical properties of selected polymers, including polyethylene, poly(vinyl alcohol) and polyamide 6.

1.Introduction

Whilst reinforcement of polymers using conventional natural fibres is well established commercially, in particular for use in the automotive industry, there is increasing interest in the use of cellulose nano-fibres derived from a variety of natural sources, which due to their size, potentially have a much higher reinforcing capability than the currently used fibre feedstocks. This paper will highlight preparation methods for nano-cellulose reinforcements derived from banana tree fibres, flax and grass, based on microfibrillation and will exemplify the benefits and challenges of using them as additives in hydrophilic and hydrophobic polymers.

2. Experimental procedures

Nanocellulose fibres from different sources (flax, banana tree and grass) have been prepared using a high pressure microfluidizer. To facilitate the ease of structural breakdown in this process, different chemical pretreatments were applied, including acid hydrolysis, mercerization and TEMPO. Products made by these methods were characterised by DLS, FTIR, XRD, TEM, SEM and TGA. Differences in particle size, cellulose crystalline content and thermal stability were then compared.

Polymer composites containing these nano-cellulose fibres were prepared by solution casting and melt lamination techniques with nano-cellulose of up to 5wt%. Polyvinyl alcohol (PVA) composites were made by mixing aqueous solutions of the polymer with nanofibre suspensions then evaporating the resulting mixture. A similar solvent approach was adopted using polyamide 6 (PA-6) dissolved in formic acid. Laminate composites of high density polyethylene (HDPE) were made by first preparing nanocellulose paper, by evaporation of aqueous suspensions, then melt compression within preformed sheets of the polymer of known thickness.

Mechanical tests were undertaken on these composite samples in both tensile and dynamic testing modes.

Further details of nano-cellulose preparation, composite preparation, testing and characterization procedures can be found in [1-3].

3. Results and discussion

By way of example, Figure 1 shows the high aspect ratio achievable from flax fibres exposed to microfluidisation.

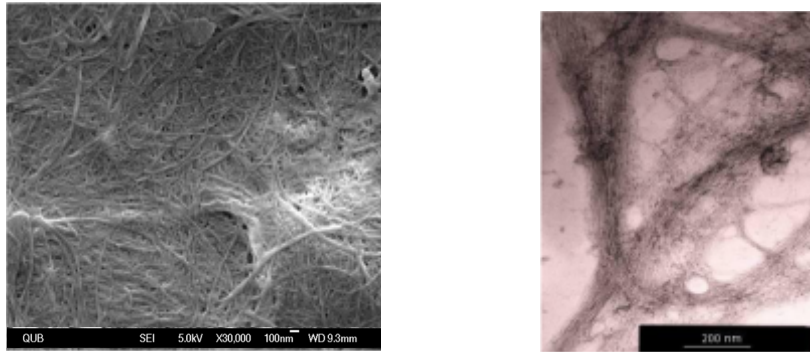


Figure 1 (left) SEM micrograph after 80 passes using silane pretreatment (right) TEM micrograph of flax nanofibres after 40 passes using high pressure (1800 bar)

As shown in Figure 2, multiple passes of flax through a microfluidiser causes the degree of cellulose crystallinity to increase as non-cellulosic components are removed. Figure 3 compares dynamic light scattering results for cellulose nanofibres derived from banana fibres demonstrating the strong influence of pre-treatment route prior to microfluidisation.

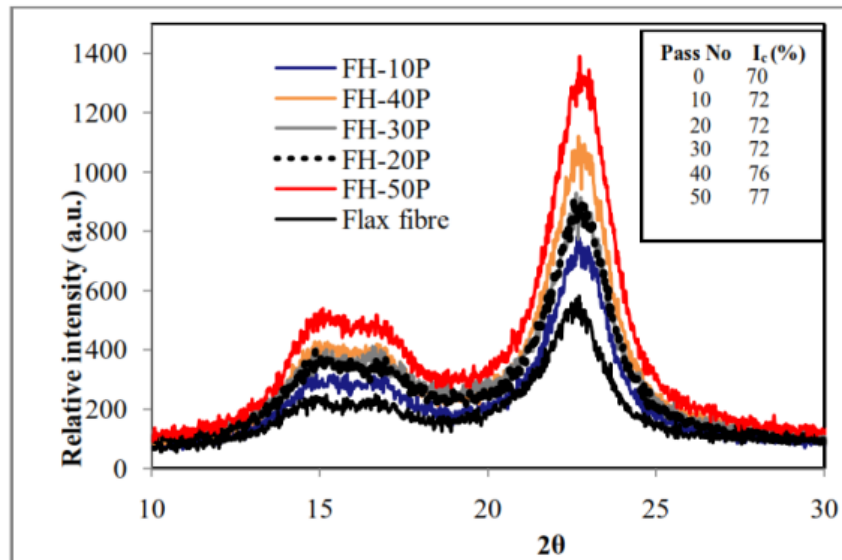


Figure 2. XRD patterns showing effect of multiple passes through microfluidiser on degree of crystallinity of cellulose nanofibrils

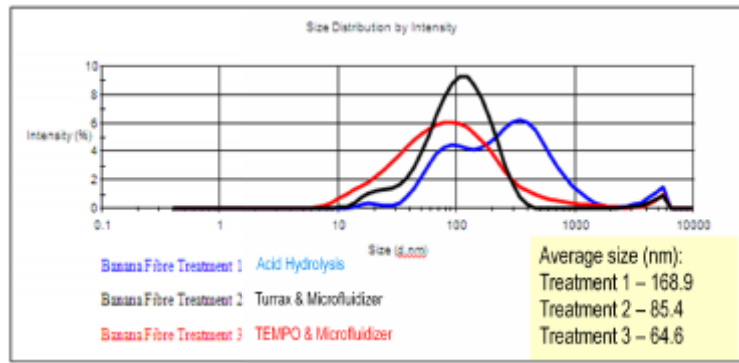


Figure 3. Effect pre-treatment and microfluidisation procedure on the particle size distribution of banana nano-fibres.

Reinforcement with nanocellulose fibres has been studied in a number of polymers, including polyethylene (HDPE), polyvinyl alcohol (PVA) and polyamide 6 (PA-6), using solution casting and melt lamination procedures. For example, Figure 4 shows solution cast samples of PVA/banana nano-cellulose fibres indicating how the colour and clarity of the films is influenced by the preparation method.

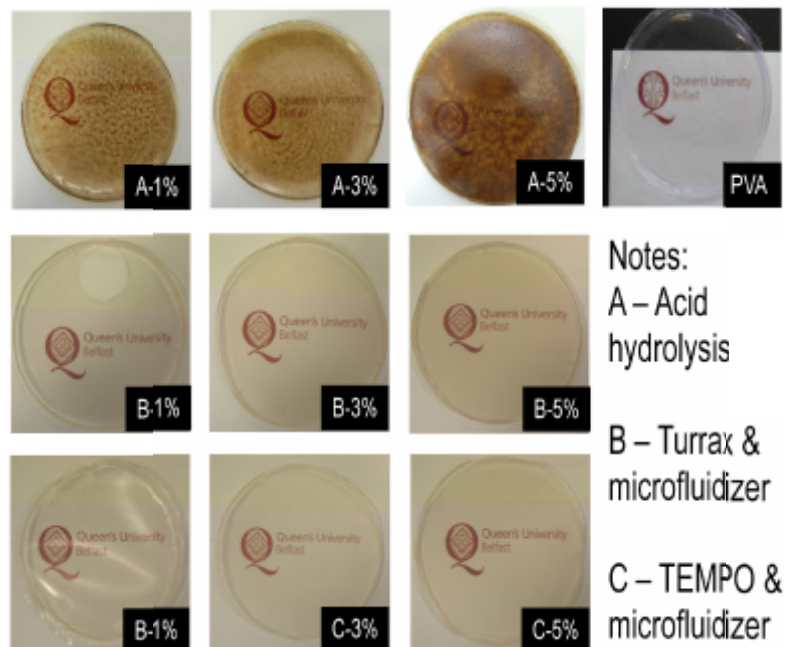


Figure 4. Solution cast PVA/banana cellulose nanofibres showing the effect of weight% addition level and preparation method on colour and clarity. .

Figure 5 demonstrates the reinforcing efficiency achievable using cellulose nanofibres derived from banana fibres using the different methods of pre-treating the fibres prior to mechanical shear (routes A-C). Tensile modulus is increased by 300% with 5wt% addition of nanofibres to PVA, whereas the enhancement of tensile strength is limited to 30%.

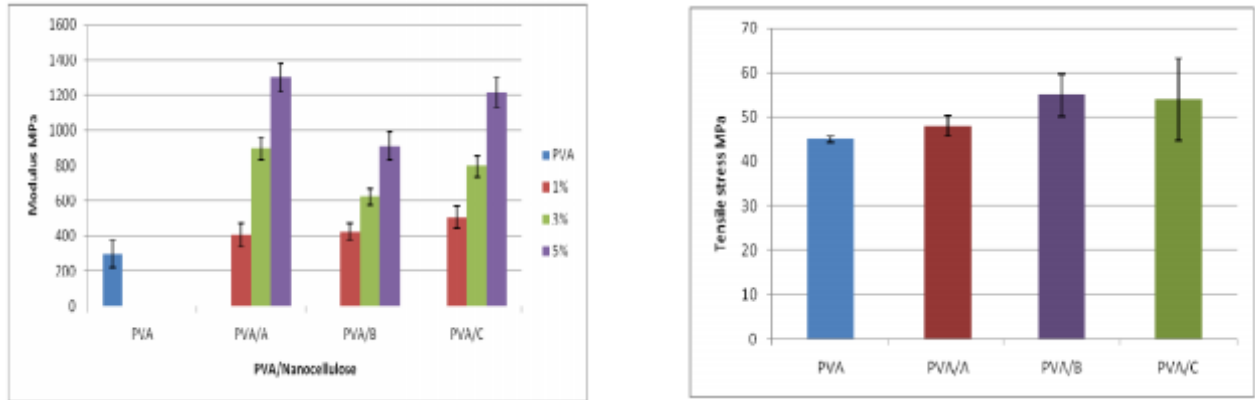


Figure 5. Tensile properties of solution cast PVA/banana cellulose nanofibre composites.

Laminated composites were prepared by compression moulding of multiple layers of maleic anhydride functionalized polyethylene and nano-cellulose paper derived from microfluidised suspensions of grass fibres. As seen in Figure 6, there is evidence of interfacial bonding between the modified polyethylene and nanocellulose paper, thought to result from ester formation. By comparison, composites made using non-functionalised polyethylene delaminated under testing with no evidence for interfacial bonding (Figure 7).

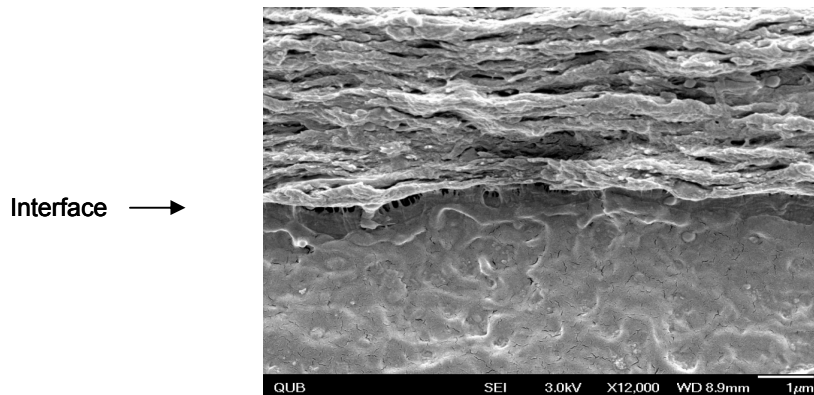


Figure 6. Interface region between maleic anhydride modified polyethylene and nanocellulose paper laminate.

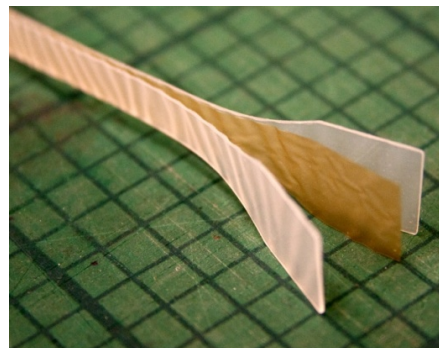


Figure 7. Delamination at nano-cellulose /polymer interface using unfunctionalised polyethylene

Through multiple laminations using nano-cellulose paper 118% increase in modulus was obtained with only 5.3wt% nanocellulose addition, relative to unreinforced polyethylene (Figure 8).

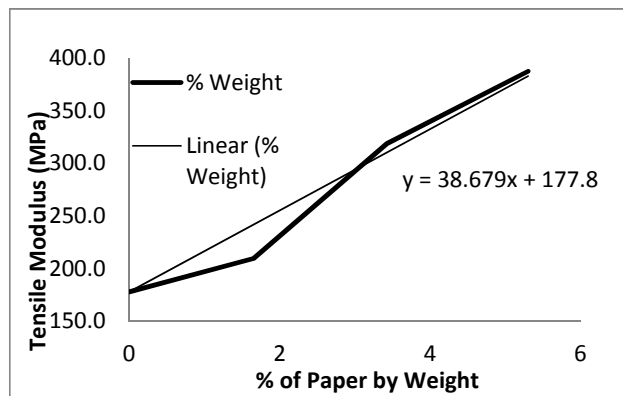


Figure 8. Effect of nanocellulose addition on the tensile modulus of anhydride modified polyethylene laminates.

Current work is focusing on composite preparation procedures using traditional melt compounding with polyethylene. This will involve the use chemical modification procedures on the fibres to prevent nanofibre agglomeration and to aid dispersion, together with the effects of uniaxial and biaxial deformation on structure and properties of the composites.

4. Conclusions

Nano-cellulose fibres have been prepared from a variety of feedstocks, including grass, flax and banana tree fibres using combinations of chemical and mechanical treatments, involving high pressure microfluidisation. The fibre aspect ratio, degree of crystallinity and thermal stability, critically depend on the preparation procedure employed. Polymer composites made from these fibres, by solution casting and melt lamination methods, demonstrate a high level of mechanical enhancement is possible with very low addition levels of reinforcement, typically around 5wt%.

5. References

- [1] Hornsby, P.R. and Qua, E. Preparation and characterization of polyvinyl alcohol nanocomposites made from cellulose, *Journal of Applied Polymer Science*, **113**, pp. 2238-2247, (2009).
- [2] Hornsby, P.R. and Qua, E. Preparation and characterisation of nanocellulose reinforced polyamide-6, *Plastics, Rubber and Composites*, **40**, pp. 300-306, (2011).
- [3] Sharma, H.H.S, Carmichael, E., Muhamad, M., McCall, D., Andrews, F., McRoberts, C., and Hornsby, P.R. Biorefining of perennial ryegrass for the production of nano-fibrillated cellulose, RSC Advances, In Press (2012).