



A CRITICAL REVIEW ON ENZYMATIC ASSISTED ISOLATION OF CELLULOSE NANOFIBRE FROM PLANT FIBRES AND ITS FUNCTIONAL APPLICATION IN NANO-BIOCOMPOSITE

¹Ojo, O. T. and ²Olaiya, N. G.

¹ Department of Industrial and Production Engineering,
Federal University of Technology Akure, PMB 704 Akure, Nigeria

² School of Industrial Technology, Universiti Sains Malaysia, 11800 Penang, Malaysia

*Corresponding E-mail: otojo@futa.edu.ng

Oluwole T. O. and Niyi G. O. (2022): A Critical Review on Enzymatic Assisted Isolation of Cellulose Nanofibre from Plant Fibres and its Functional Application in Nano-biocomposite. *Journal of Engineering and Engineering Technology* /16(1), 105-112

Received 09-02-22

Accepted: 17-03-22

Abstract

Pre-treatment technique of biomass for cellulose nanofiber isolation is very significant on its properties. Several techniques have been used for pre-treatment of biomass, however much has not been done on the use of combined pre-treatment for Cellulose Nanofibre (CNF) isolations. The use of enzymes has been attached to the extraction of ethanol from biomass. In this study, a critical review of the application of combine mechanical-enzymes in isolation of CNF was discussed. A parametric study of factors that influence the output of mechanical and enzymatic hydrolysis was done with reference to previous work in this area. The review concluded by assessing the viability of enzymatic hydrolysis in combination with other techniques for successful isolation of cellulose nanofibre

Keyword: Pre-treatment, enzyme, hydrolysis, cellulose nanofibre, biomass

Introduction

The purpose of pre-treatment is directed toward removal of lignin and hemicellulose in the precursor material. The removal makes the fibre less resistance to fibre hydrolysis as these non-cellulosic materials form a cover sheet for cellulosic content in plants. Cellulose prepared from biomass also require these processes to access its cellulosic content (—Kumar et al. (2009). Several techniques have been established for pre-treatments. Pre-treatments can either change the physical and chemical structure of biomass in order to enhance the rate of hydrolysis. Physical pre-treatment is milling, grinding; while chemical pre-treatment involves the use of mild acid, alkaline, organic solvent and oxidising agent. Furthermore, physical chemical process such as stem hydrolysis, hydro thermolysis and wet oxidation can also be used (Bhatnagar and Sain, 2005). Other pre-treatment technique are electrical, ionic liquids and biological (Janardhnan and Sain, 2011). The precursor material determines the most effective of these pre-treatment processes. Often times, the pre-treatment use is determined by the composition of the biomass precursor material. The efficiency of the technique is also depended on the parameters such as concentration, time and so on.

In the isolation of CNF, defibrillation of fibre is needed with mechanical treatment. Mechanical pre-treatment is not economical because it can damage the fibrils and consumes energy. The energy consumed is proportional to the effectiveness of the output from the mechanical treatment, that is, higher yield, higher energy and vice versa (Chinga-Carrasco, 2011). An improvement in the pre-treatments process is the replacement of mechanical treatment with chemical and enzymatic treatment. These pre-treatments consumes lesser energy to as low as 1000 kWh/ton for cellulosic fibres as against 20000 - 30000 kWh/ton in mechanical treatment (Alle *et al.*, 2020). Several industrial preparation of nanocellulose makes use of chemical pretreatment before mechanical isolation using milling machine. However, calculated chemical treatment (such as acid, alkaline, or enzymatic hydrolysis) must be used so as not to damage the fibre. The pre-treatment enables accessibility of cellulose in the fibre (Luo and Wang, 2017).

Most plant precursor fibre is lignin, which prevent access to the cellulose fibre because of its resistance to hydrolysis. Figure 1 presents the schematic diagram of pre-treatment processes and the parameters are summarized in Table 1.

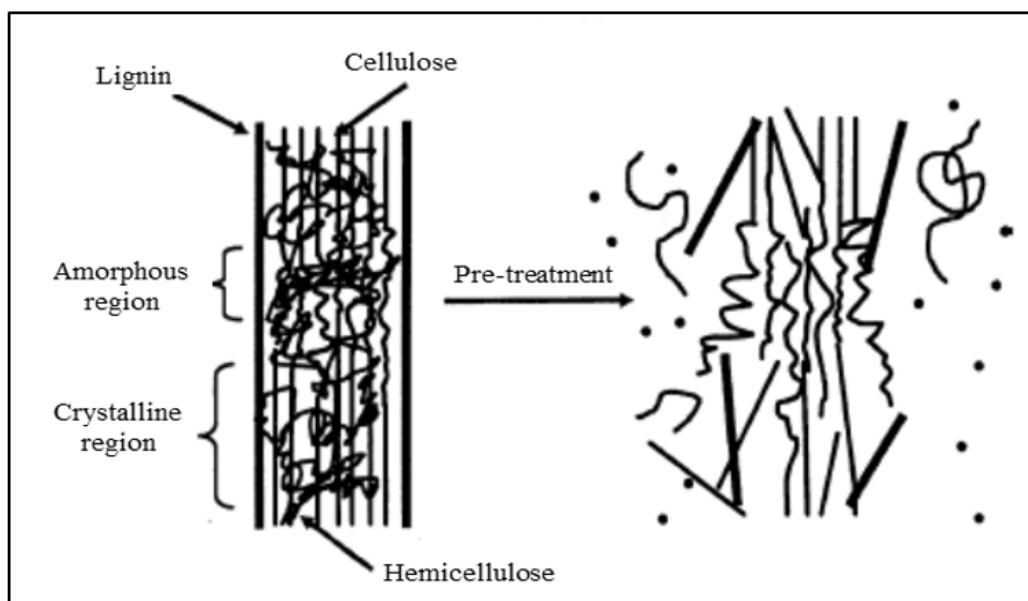


Figure 1: Schematic role of biomass pre-treatment (Kumar and Sekaran, 2014)

Table 1: Various Pre-treatment Process of Lignocellulosic Fibres (–Lavoine et al., 2012)

Pre-treatment	Advantages	Limitations
Biological		
Enzyme-assisted hydrolysis	Degrades lignin and hemicelluloses; low energy requirements	Very low in rate of hydrolysis
Chemical		
Acid hydrolysis	Hydrolyzes hemicellulose to xylose and other sugars; alters lignin structure	High cost; equipment corrosion; formation of toxic substances
Alkaline hydrolysis	Removes hemicelluloses and lignin; increases accessible surface area	Long residence time required; irrecoverable salts formed and incorporated into biomass
Ozonolysis	Reduces lignin content; does not produce toxic residues	Large amount of ozone required; formation of toxic substances
Organosolv	Hydrolyzes lignin and hemicelluloses	Solvents need to be drained from the reactor, evaporated, condensed; high cost
Mechanical		
Grinding	Reduces cellulose crystallinity	Power consumption usually higher than inherent biomass energy
Physicochemical		
CO ₂ explosion	Increases accessible surface area; cost-effective; does not cause formation of inhibitory compounds	Does not modify lignin or hemicelluloses

Pre-treatment techniques of plant fibres for cellulose nanofibre isolation

Chemical Pre-treatment

The first application of chemical treatment before its use in cellulose preparation is in paper making (Abral *et al.*, 2021). Chemical treatment has been proven to be effective for lignin removal; termed delignification. Chemical pre-treatment of lignocellulosic has been traditionally used in pulp and paper industry for delignification of cellulose. It is highly effective with

minimal supervision. It also has an advantage over mechanical treatment because the energy consumed by this technique is minimal and the formation of inhibitor is also insignificant. The Pre-treatment of cellulose precursor material is done using alkaline or bases. The pre-treatment is done to remove the lignin content in the precursor material (Kumar *et al.*, 2009). The alkaline pre-treatment dissolves the hemicellulose, lignin, and pectin from cellulosic fibres. The pre-treatment is performed at room conditions for hours (Zuluaga *et al.*, 2009). The pre-treatment process is in two steps pulping first and then bleaching. Basically,

the objective of pulping is to remove lignin from raw fibres. It is also called delignification, that is the lignin is degraded and becomes soluble which is then washed away with water (Oladele *et al.* 2018)

There are several chemicals for the pre-treatment processes; this includes sodium, calcium, potassium, and ammonium hydroxide. However, sodium hydroxide is often used and it is called alkaline pre-treatment (Ma *et al.*, 2017). The obtained pulp fibre after delignification has been reported with high crystallinity. Secondly, bleaching is employed to remove other impurities in the cellulosic pulp fibres and its dependent of the source of the cellulosic material. Several chemicals have been used for the bleaching process for example chlorine, hydrogen peroxide, ozone or peracetic acid (Thambiraj and Shankaran, 2017). Alkaline pre-treatment is very important in cellulose preparation using enzymatic hydrolysis. The effectiveness of hydrolysis process using enzymes is depended significantly on the pre-treatment. The presence of lignin has been reported to reduce the effectiveness of the enzyme in the hydrolysis of the precursor material to cellulose (Wang *et al.*, 2017). Chang and Holtzapple, (2000) concluded that there is significance correlations between the lignin and enzymatic digestibility. The authors reported that effective delignification resulted in high digestibility by the enzymes. The lignin presence was described to cause barrier for the enzymes to have direct reaction on the cellulosic material. Other impurities such as acetyl group were also reported to reduce the enzymatic action of the raw fibre (—Kumar *et al.*, 2009).

Physical Pre-treatment

Physical pre-treatment are non -mechanical pre-treatment and they are used for breakdown of lignocellulosic materials into fine particles (—Kumar *et al.*, 2009). Examples of physical pre-treatment are steam or autohydrolysis, hydro-thermolysis, and wet oxidation (Tribot *et al.*, 2019). Apart from steam, pre-treatment with liquid water is also done occasionally. When liquid water is used, hydro-thermolysis, aqueous or steam fractionation, aquasolv, and uncatalyzed solvolysis are some of the terms used to describe this form of pre-treatment (Kumar and Singh, 2008). In this method, to keep the water in a liquid state, pre-treatment is carried out at a high temperature and under pressure (). At temperatures of 200-230 °C, the residence time for this process is around 15 minutes. During this process, roughly 40-60% of the total plant fibre is dissolved, including 4-22% cellulose, 35-60% lignin, and all hemicellulose. Since the lignocellulose particles will break apart during the treatment process, no size reduction of biomass is needed prior to hot water pre-treatment. Pérez *et al.* (2007) investigates the effects of temperature (170 and 200 °C), solid

concentration [5% and 10% (w/v)], residence time (0 and 40 minutes), and reactor overpressure (30 bar) on the pre-treatment of wheat straw. Solid compositions and liquid fractions obtained after filtration, as well as the solid fraction's susceptibility to enzymatic hydrolysis using commercial celluloses enzyme, were used to assess the efficacy of pre-treatment. The authors discovered that temperature and time had an effect on pre-treatment, and that increasing both temperature and time increased enzyme hydrolysis yield (Espinosa *et al.*, 2019)

Various treatment systems, including sufficient treatment time, strain, and temperature, have been tried and produced to optimise these processes (Wang *et al.*, 2017). Since the properties of hydrothermolysis can be regulated by temperature, pressure, and treatment time, it's important to pick the right treatment system to monitor the substances' decomposition route (Le Troedec *et al.*, 2008). As previously described, separating lignin from cellulose in a single step is difficult. As a result, a new method was proposed in which alkaline pre-treatments were applied to cellulosic bamboo fibres before high-pressure enzymatic hydrolysis was used to remove lignin and other impurities from the cellulosic bamboo fibres in this study (Zuluaga *et al.*, 2009).

Mechanical Pre-treatment

The most common mechanical pre-treatment methods used in the defibrillation of the CNF include refining, chip-out, homogenisation, microfluidisation, grinding, cryocrushing, ultrasound, steam explosion, electromagnetic or combined approaches (Jawaid and Khalil, 2011). Due to their high energy consumption, mechanical insulation approaches result in a major reduction in output and fibre length. The biomass characteristics and final particle size determine the power and energy consumption for mechanical comminution of lignocellulosic materials. Researchers determined that the energy input for particle size reduction in the range of 3-6 mm is less than 30 kWh per tonne of biomass. In addition to inefficient fibre disintegration, these techniques tend to harm the structure of microfibrils by reducing both the degree of crystallinity and the polymerization of cellulose (kumar *et al.*, 2009).

A method for treating cellulosic fibres using chemical or enzymatic pre-treatment before mechanical isolation is introduced to resolve this issue (Khalil *et al.*, 2012; Chauhan, 2020). A technique that incorporates chemical and mechanical treatment is known as chemo-mechanical therapy (Fatah *et al.*, 2014). According to reports, chemo-mechanical pre-treatment of cellulosic fibres lowered energy consumption from 30,000 kWh/ton to 1000 kWh/ton (Siro *et al.*, 2011). A combination of two or more mechanical isolation processes, on the other hand, produced CNF with

promising properties. Qing *et al.* (2013), discovered that combining refining and microfluidization processes produced standardised CNF with high fibrillation in their study. In another study, Wang and Cheng, (2009) discovered that combining high-intensity ultrasonication with high-pressure homogenization produced effective fibrillation and uniform CNF. In addition, cryocrushing and refining are often used in conjunction with homogenization to improve the performance of the manufacturing process.

Homogenization is the process of changing the particle size of a material to achieve homogeneity. Anything homogeneous is described as having the same composition, structure, or personality. Other words for homogenising include blending, mixing, dispersing, disrupting, boiling, emulsifying, among others. The three major classes are pressure homogenization, ultrasonic pressure homogenization, and mechanical homogenizer (Dharkhar, 2014). Rotor-stator homogenizers and blade form homogenizers are the two types of mechanical homogenizers. The rotor-stator homogenizer disrupts

fibres with hydraulic, mechanical, and cavitation shear. It's also known as high shear mechanical dispersion (HSMD) or high shear homogenization, and it necessitates high-energy sonic and ultrasonic pressure gradients. Cavitation happens when a solid object moves through a rapidly flowing liquid. A rapidly spinning rotor (blade) in a static head or tube (stator) with slots or holes draws fibres up. Fibers are thrown outward by centrifugal force in a pump and exit through slots or holes. As the rotor spins at a high speed, a combination of intense turbulence, scissor-like mechanical shearing, and cavitation occurs within the narrow gap between stator and rotor, rapidly reducing fibre size. To put it in another way, HSMD is a mechanical homogenization strategy that employs two distinct mechanical behaviours. The cutting technique shortens the fibres, while the shear force method frees the microfibrils. The rotor-stator configuration and size, rotor tip speed, initial sample size, medium viscosity, processing time or flow rate, medium volume, and sample concentration all influence the homogenization process' performance (Dharkhar, 2014; Zhang *et al.*, 2020). A schematic diagram of fibre fibrillation using a rotor-stator homogenizer is shown in Figure 2.

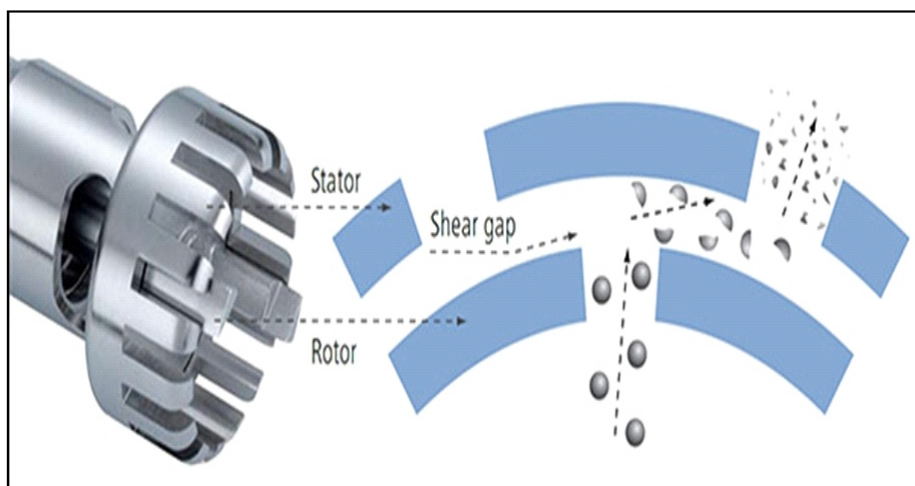


Figure 2. Schematic diagram of fiber fibrillation by Ultra Turrax rotor-stator homogenizer (Cole-Parmer, 2016)

Cellulosic fibre is disintegrated in this process, resulting in smaller fibre dimensions and the liberation of microfibrils. HSMD can generate CNF in small (gram-scale) and large (kilogram-scale) quantities, depending on the equipment, condition, and method used. The Ultra Turrax high-speed dispersing unit is a gram-scale HSMD that is designed for batch-wise operation with a limited inlet volume amount. The kg-scale equipment, on the other hand, was created for industrial use. The Ultra Turrax high-speed dispersing machine consists of a dispersing chamber with a ring generator and a chamber volume. Since viscosity tends to rise during the disintegration process, a pulp with a very low consistency is needed to ensure proper

mixing. Zhao *et al.* (2013) used a simple, environmentally friendly mechanical refining pretreatment with high shear homogenization method to successfully extract CNF with high crystallinity and low thermal stability from dry softwood pulp. A possible novel nanopaper with high optical transparency for advanced applications has been developed. Some researchers have used mechanical disintegration in the fabrication of nanocomposites, especially at the laboratory scale. It acts as a dispersion mechanism, allowing the polymer matrix and nanofibers to disperse more evenly (Hedayati *et al.*, 2011; Sehaqui *et al.*, 2012).

Clogging, on the other hand, is a common problem that

takes time to fix because long fibres clog the system because of small slots or holes that must be disassembled and cleaned. As a result, the fibre size must be decreased before going through the homogenizer to prevent stacking the small slots. Prior to the homogenization process, pre-treatments and a combination of chemical and mechanical treatment are needed. Researchers combined mechanical pre-treatments such as refining (Muhamad *et al*, 2019), cryocrushing (Kam *et al*, 2019), enzyme (Martelli-Tosi *et al*, 2016), and chemical pre-treatments (Alemdar and Sain, 2008) to minimise the size of fission products. To manufacture CNF with diameters ranging from 10-90 nm from kenaf bast fibres, Jonoobi *et al*. (2011) used a combination of pulping and bleaching as a chemical pre-treatment when grinding and cryocrushing as a mechanical pre-treatment before homogenization. They have used a grinding pre-treatment on the kenaf stem and core to produce CNF with widths of 15-80 nm and 20-25 nm, respectively, from the kenaf stem and core.

Application of Cellulose Nanofibre Based Nanocomposites

Previously, composites were created by mixing natural fibre with a petroleum-based polymer. Bio-derived polymer and petroleum-based polymer were used to create green composites. Bio-based material was integrated into a petroleum-based polymer to increase biodegradability. The creation and application of nanocomposites made of nano-sized fibres and microfibrils derived from natural fibre are currently thriving in biocomposites research. Completely green nanocomposites are emerging due to green technology being a serious field of research that is gaining increasing attention from both industries and academic affiliations (Ramesh *et al*, 2017). Green composites are a unique choice for pulp and paper, food processing, automotive, coatings, construction, furniture, medical and pharmacy, cosmetics, and electrical and electronic industries since they are manufactured from renewable agricultural products. Natural fibres and recycled materials have recently gained popularity in these fields due to their low cost, light weight, sustainability, and ability to reduce hazardous waste (Ramesh *et al*, 2017; Sulaiman *et al*, 2018; Sudesh and Iwata, 2008).

Nanofibre is commonly used as a reinforcement in several applications, including hard composites and thin films (Abdul Khalil *et al*, 2020; Eichhorn *et al*, 2010). As a result, it is used in almost every industry, from construction materials to food packaging. A wide range of products are made with CNF and nanocrystalline cellulose (NCC) reinforced polymer matrices. Film and flexible packaging, compostable alternatives to plastic film, high-quality paper and sheets, super-strong pulp, strong tissue, mineral paper, wood fibre composites, and wood panels are among CNF's products (Cherian *et al*, 2011). The properties of

iridescent NCC films include plastics, deposition on glass, cosmetics, architectural industries, inks, protection paper, varnishes and coatings, thermal barriers, ultraviolet reflectance, infrared reflectance, and ultraviolet barriers (Huq *et al*, 2012).

A network of intertwined nanofibrils with a high aspect ratio and random surface nanofibril orientation makes up cellulose nanopaper. It has become a common commodity in nanotechnology applications due to its ability to customise porosity, high durability, and resource renewability (Henrikson *et al*, 2008). Nanopapers are lightweight, transparent, and foldable (Sehaqui *et al*, 2012). The high strength of nano papers is greater fibril strength, high inter-fibril adhesion properties, and uniform distribution defects (Henrikson *et al*, 2008). Nanopapers are an ideal substrate for continuous roll-to-roll processing in the manufacture of electronic devices, and they may take the place of expensive batch glass processing. Nano paper, unlike glass, has a high heat-transfer capability and is transparent due to its tightly packed structure and minimal space between fibres, which prevents light scattering (Sehaqui *et al*, 2012). Furthermore, Yousefi *et al*. (2013) discovered that it outperforms micro paper in barrier properties, tensile strength, and modulus of elasticity. Nano paper has applications in packaging, loudspeaker membranes, battery membranes, and biomedical applications.

Product protection, shelf-life extension, cost-efficiency, customer convenience, and environmental issues are major concerns regarding food packaging. To avoid food degradation, researchers are developing potential materials with suitable mechanical properties and the ability to provide an adequate barrier to oxygen, water vapour, light, microorganisms, and other pollutants (Tang *et al*, 2012). CNF was found to contribute to the impermeability of films when prepared to an acceptable thickness, which is essential in food packaging (Hermawan *et al*, 2019). On the other hand, microfibrillated cellulose (MFC) has emerged as a potential packaging material reinforcement due to its combination of water barriers and mechanical properties (Hermawan *et al*, 2019; Chinga-Carrasco, 2011).

CNF has found applications in biomedical sciences due to its excellent mechanical, porosity, and rheological properties (Abdul Khalil *et al*, 2020). It is used in the development of artificial ligaments and tendons. It's also been used as a reinforcement in developing collagen implantable scaffolds (Khalil *et al*, 2020). Zhang *et al*. (2019) developed biocomposite hydrogels reinforced carboxymethylated CNF to replace nucleus pulposus in intervertebral discs. CNF reinforced polyurethane matrices for implantable applications have also been developed by Cherian *et al*. (2011) used native CNF hydrogel as a scaffold for 3D cell cultures. At the same time, Shimotoyodome *et al*. (2011)

examined the effect of CNF intake on postprandial blood metabolic variables. According to recent biomedical research, CNF can also be used as a filler in thin films for tablet processing (Kolakovic *et al*, 2012).

Several researchers looked at other applications, such as transparent films for coating industries, electronic devices, and excellent barrier properties (Sharma *et al*, 019). MFC, for example, has recently been tested in printing applications and found to increase print quality and ink density when used as a coating agent (Luu *et al*, 2011). The overall performance of nanocomposites is influenced by the properties of nanocellulose as reinforcement material and the interaction between nanocellulose and the polymer matrix (Murphy and Collins, 2018). Mechanical, thermal, and optical properties of CNF nanocomposites are the most expected changes in performance. CNF nanocomposites' proper processing, combined with their high mechanical properties and wide surface area, allows for forming a dense hydrogen bonding network. The formation of hydrogen bonding is one of the most important factors irresponsible for strength in materials intended for the production of scaffold and packaging applications mentioned above (Luu *et al*, 2011).

Conclusion

This study has critically examined different methods of pre-treatment of biomass to obtain functional cellulose nanofibre. These pre-treatment techniques have been shown to be mutually exclusive and effective. Incorporating cellulose nanofibre in biocomposites for various applications is on the increase. The enormous increase in biocomposite properties with the addition of cellulose nanofibre has resulted in increased demand. This has resulted in its high cost, which can be reduced if extracted from waste. Furthermore, the valorisation of biomass waste is of significant importance to combat pollution globally. The techniques reviewed can be applied in the industrial production of cellulose nanofibre from waste. The continuous use of waste in the production of functional material will lead to a long-term reduction in land and marine pollution.

Reference

- Abdul Khalil, H., Adnan, A., Yahya, E. B., Olaiya, N., Safrida, S., Hossain, M., Balakrishnan, V., Gopakumar, D. A., Abdullah, C. and Oyekanmi, A. (2020). A Review on plant cellulose nanofibre-based aerogels for biomedical applications. *Polymers*, 12, 1759.
- Abdal, H., Chairani, M. K., Rizki, M. D., Mahardika, M., Handayani, D., Sugiarti, E., Muslimin, A. N., Sapuan, S. and Ilyas, R. (2021). Characterization of compressed bacterial cellulose nanopaper film after exposure to dry and humid conditions. *Journal of Materials Research and Technology*, 11, 896-904.
- Alemdar, A. and Sain, M. (2008). Isolation and characterization of nanofibers from agricultural residues Wheat straw and soy hulls. *Bioresource technology*, 99, 1664-1671.
- Alle, M., Bandi, R., Lee, S.-H. and Kim, J.-C. (2020). Recent trends in isolation of cellulose nanocrystals and nanofibrils from various forest wood and nonwood products and their application. *Nanomaterials for Agriculture and Forestry Applications*. Elsevier.
- Bhatnagar, A. and Sain, M. (2005). Processing of cellulose nanofiber-reinforced composites. *Journal of Reinforced Plastics and Composites*, 24, 1259-1268.
- Chang, V. S. and Holtzapple, M. T. (2000). Published. Fundamental factors affecting biomass enzymatic reactivity. Twenty-first symposium on biotechnology for fuels and chemicals, 2000. Springer, 5-37.
- Chauhan, P. S. (2020). Lignin nanoparticles: eco-friendly and versatile tool for new era. *Bioresource Technology Reports*, 9, 100374.
- Cherian, B. M., Leão, A. L., De Souza, S. F., Costa, L. M. M., De Olyveira, G. M., Kottaisamy, M., Nagarajan, E. and Thomas, S. (2011). Cellulose nanocomposites with nanofibres isolated from pineapple leaf fibers for medical applications. *Carbohydrate Polymers*, 86, 1790-1798.
- Chinga-Carrasco, G. (2011). Cellulose fibres, nanofibrils and microfibrils: The morphological sequence of MFC components from a plant physiology and fibre technology point of view. *Nanoscale research letters*, 6, 1-7.
- .Dhankhar, P. (2014). Homogenization fundamentals. *IOSR Journal of Engineering*, 4, 8.
- Eichhorn, S. J., Dufresne, A., Aranguren, M., Marcovich, N., Capadona, J., Rowan, S. J., Weder, C., Thielemans, W., Roman, M. and Renneckar, S. (2010): Current international research into cellulose nanofibres and nanocomposites. *Journal of materials science*, 45, 1-33.
- Espinosa, E., Bascón-Villegas, I., Rosal, A., Pérez-Rodríguez, F., Chinga-Carrasco, G. and Rodríguez, A. (2019). PVA/(ligno) nanocellulose biocomposite films. Effect of residual lignin content on structural, mechanical, barrier and antioxidant properties. *International journal of biological macromolecules*, 141, 197-206.
- Fatah, I. Y. A., Khalil, H., Hossain, M., Aziz, A. A., Davoudpour, Y., Dungani, R. and Bhat, A. (2014). Exploration of a chemo-mechanical technique for the isolation of nanofibrillated cellulosic fiber from oil palm empty fruit bunch as a reinforcing agent in composites materials. *Polymers*, 6, 2611-2624.
- Hedayati, M., Salehi, M., Bagheri, R., Panjepour, M.

- and Maghzian, A. (2011). Ball milling preparation and characterization of poly (ether ether ketone)/surface modified silica nanocomposite. *Powder Technology*, 207, 296-303.
- Henriksson, M., Berglund, L. A., Isaksson, P., Lindstrom, T. and Nishino, T. (2008). Cellulose nanopaper structures of high toughness. *Biomacromolecules*, 9, 1579-1585.
- Hermawan, D., Lai, T. K., Jafarzadeh, S., Gopakumar, D. A., Hasan, M., Owolabi, F. T., Aprilia, N. S., Rizal, S. and Khalil, H. A. (2019). Development of seaweed-based bamboo microcrystalline cellulose films intended for sustainable food packaging applications. *BioResources*, 14, 3389-3410.
- Huq, T., Salmieri, S., Khan, A., Khan, R. A., Le Tien, C., Riedl, B., Fraschini, C., Bouchard, J., Uribe-Calderon, J. and Kamal, M. R. (2012). Nanocrystalline cellulose (NCC) reinforced alginate based biodegradable nanocomposite film. *Carbohydrate polymers*, 90, 1757-1763.
- Janardhnan, S. and Sain, M. M. (2011). Targeted disruption of hydroxyl chemistry and crystallinity in natural fibers for the isolation of cellulose nano-fibers via enzymatic treatment. *BioResources*, 6, 1242-1250.
- Jawaid, M. and Khalil, H. A. (2011). Cellulosic/synthetic fibre reinforced polymer hybrid composites: A review. *Carbohydrate polymers*, 86, 1-18.
- Jonoobi, M., Harun, J., Tahir, P. M., Shakeri, A., Saifulazry, S. and Makinejad, M. D. (2011). Physicochemical characterization of pulp and nanofibers from kenaf stem. *Materials letters*, 65, 1098-1100.
- Kam, D., Chasnitsky, M., Nowogrodski, C., Braslavsky, I., Abitbol, T., Magdassi, S. and Shoseyov, O. (2019). Direct Cryo Writing of Aerogels via 3D Printing of Aligned Cellulose Nanocrystals Inspired by the Plant Cell Wall. *Colloids and Interfaces*, 3, 46.
- Khalil, H., Jummaat, F., Yahya, E. B., Olaiya, N., Adnan, A., Abdat, M., Nam, N., Halim, A. S., Kumar, U. and Bairwan, R. (2020). A review on micro-to nanocellulose biopolymer scaffold forming for tissue engineering applications. *Polymers*, 12, 2043.
- Khalil, H. A., Bhat, I., Jawaid, M., Zaidon, A., Hermawan, D. and Hadi, Y. (2012). Bamboo fibre reinforced biocomposites: A review. *Materials & Design*, 42, 353-368.
- Kolakovic, R., Peltonen, L., Laukkanen, A., Hirvonen, J. and Laaksonen, T. (2012). Nanofibrillar cellulose films for controlled drug delivery. *European Journal of Pharmaceutics and Biopharmaceutics*, 82, 308-315.
- Kumar, A. P., Depan, D., Tomer, N. S. and Singh, R. P. (2009). Nanoscale particles for polymer degradation and stabilization trends and future perspectives. *Progress in polymer science*, 34, 479-515.
- Kumar, A. P. and Singh, R. P. (2008). Biocomposites of cellulose reinforced starch: Improvement of properties by photo-induced crosslinking. *Bioresource Technology*, 99, 8803-8809.
- Kumar, K. P. and Sekaran, A. S. J. (2014). Some natural fibers used in polymer composites and their extraction processes: A review. *Journal of Reinforced Plastics and Composites*, 33, 1879-1892.
- Lavoine, N., Desloges, I., Dufresne, A. and Bras, J. (2012). Microfibrillated cellulose barrier properties and applications in cellulosic materials: A review. *Carbohydrate polymers*, 90, 735-764.
- Le Troedec, M., Sedan, D., Peyratout, C., Bonnet, J. P., Smith, A., Guinebreiere, R., Gloaguen, V. and Krausz, P. (2008). Influence of various chemical treatments on the composition and structure of hemp fibres. *Composites Part A: Applied Science and Manufacturing*, 39, 514-522.
- Luo, X. and Wang, X. (2017). Preparation and characterization of nanocellulose fibers from NaOH/Urea pretreatment of oil palm fibers. *BioResources*, 12, 5826-5837.
- Luu, T., Richmond, F., Bilodeau, M. and Bousfield, W. (Year): Published. Nano-fibrillated cellulose as a paper surface treatment for inkjet printing. Nano-fibrillated cellulose as a paper surface treatment for inkjet printing. TAPPI international conference on nanotechnology for renewable materials, Arlington, USA, 2011.
- Ma, L., Zhang, Y. and Wang, S. (2017). Preparation and characterization of acrylonitrile-butadiene-styrene nanocomposites reinforced with cellulose nanocrystal via solution casting method. *Polymer Composites*, 38, E167-E173.
- Martelli-Tosi, M., Torricillas, M. D. S., Martins, M. A., Assis, O. B. G. D. and Tapia-Blácido, D. R. (2016). Using commercial enzymes to produce cellulose nanofibers from soybean straw. *Journal of Nanomaterials*, 2016.
- Muhamad, M., Hornsby, P., Carmichael, E., Zakaria, M., Seok, Y. B., Mohamed, S. and Sharma, S. (2019). Published. Characterisation of cellulose nanofibres derived from chemical and mechanical treatments. MATEC Web of Conferences, 2019. EDP Sciences, 01002.
- Murphy, C. A. and Collins, M. N. (2018). Microcrystalline cellulose reinforced polylactic acid biocomposite filaments for 3D printing. *Polymer Composites*, 39, 1311-1320.
- Oladele, I. O., Isola, B. A., Taiwo, S. A., Ibrahim, I. O. and Masud, A. O. (2018). Development of natural polymer reinforced cement/waste paper pulp composites for structural application. *Leonardo Electronic Journal of Practices and Technologies*, 33, 1-14.

- Pérez, J. A., González, A., Oliva, J. M., Ballesteros, I. and Manzanares, P. (2007). Effect of process variables on liquid hot water pretreatment of wheat straw for bioconversion to fuel-ethanol in a batch reactor. *Journal of Chemical Technology & Biotechnology: International Research in Process, Environmental & Clean Technology*, 82, 929-938.
- Qing, Y., Sabo, R., Zhu, J., Cai, Z. and Wu, Y. (2013). Comparative study of cellulose nanofibrils Disintegrated from different Approaches. In *Proceedings of Advancements in Fiber-Polymer Composites: Wood Fiber, Natural Fibers, and Nanocellulose*, May 6-7, 2013, Milwaukee, WI; 2013; 3 p., 1-3.
- Ramesh, M., Palanikumar, K. and Reddy, K. H. (2017). Plant fibre based bio-composites: Sustainable and renewable green materials. *Renewable and Sustainable Energy Reviews*, 79, 558-584.
- Sehaqui, H., Ezekiel Mushi, N., Morimune, S., Salajkova, M., Nishino, T. and Berglund, L. A. (2012). Cellulose nanofiber orientation in nanopaper and nanocomposites by cold drawing. *ACS applied materials & interfaces*, 4, 1043-1049.
- Sharma, A., Thakur, M., Bhattacharya, M., Mandal, T. and Goswami, S. (2019). Commercial application of cellulose nano-compositesA review. *Biotechnology Reports*, 21, e00316.
- Shimotoyodome, A., Suzuki, J., Kumamoto, Y., Hase, T. and Isogai, A. (2011). Regulation of postprandial blood metabolic variables by TEMPO-oxidized cellulose nanofibers. *Biomacromolecules*, 12, 3812-3818.
- Siró, I., Plackett, D., Hedenqvist, M., Ankerfors, M. and Lindström, T. (2011). Highly transparent films from carboxymethylated microfibrillated cellulose: the effect of multiple homogenization steps on key properties. *Journal of Applied Polymer Science*, 119, 2652-2660.
- Sudesh, K. and Iwata, T. (2008). Sustainability of biobased and biodegradable plastics. *CLEANSoil, Air, Water*, 36, 433-442.
- Sulaiman, M. S., Ramle, S. F. M., Hashim, R., Sulaiman, O., Amini, M. and Geng, B. (2018). The classical mechanics engineered of *Bambusa vulgaris* and *Schizostachyum brachycladum*. *Journal of Tropical Resources and Sustainable Science*, 6, 57-61.
- Tang, X., Kumar, P., Alavi, S. and Sandeep, K. (2012). Recent advances in biopolymers and biopolymer-based nanocomposites for food packaging materials. *Critical reviews in food science and nutrition*, 52, 426-442.
- Thambiraj, S. and Shankaran, D. R. (2017). Preparation and physicochemical characterization of cellulose nanocrystals from industrial waste cotton. *Applied Surface Science*, 412, 405-416.
- Tribot, A., Amer, G., Alio, M. A., De Baynast, H., Delattre, C., Pons, A., Mathias, J.-D., Callois, J.-M., Vial, C. and Michaud, P. (2019). Wood-lignin: Supply, extraction processes and use as bio-based material. *European Polymer Journal*, 112, 228-240.
- Wang, S. and Cheng, Q. (2009). A novel process to isolate fibrils from cellulose fibers by high-intensity ultrasonication, Part 1: Process optimization. *Journal of applied polymer science*, 113, 1270-1275.
- Wang, Z., Yao, Z., Zhou, J. and Zhang, Y. (2017). Reuse of waste cotton cloth for the extraction of cellulose nanocrystals. *Carbohydrate Polymers*, 157, 945-952.
- Yousefi, H., Faezipour, M., Hedjazi, S., Mousavi, M. M., Azusa, Y. and Heidari, A. H. (2013). Comparative study of paper and nanopaper properties prepared from bacterial cellulose nanofibers and fibers/ground cellulose nanofibers of canola straw. *Industrial Crops and Products*, 43, 732-737.
- Zhang, D., Cai, J., Xu, W., Dong, Q., Li, Y., Liu, G. and Wang, Z. (2019). Synthesis, characterization and adsorption property of cellulose nanofiber-based hydrogels. *Journal of Forestry Engineering*, 4, 92-98.
- Zhang, Q., Lei, H., Cai, H., Han, X., Lin, X., Qian, M., Zhao, Y., Huo, E., Villota, E. M. and Mateo, W. (2020). Improvement on the properties of microcrystalline cellulose/polylactic acid composites by using activated biochar. *Journal of Cleaner Production*, 252, 119898.
- Zhao, J., Zhang, W., Zhang, X., Zhang, X., Lu, C. and Deng, Y. (2013). Extraction of cellulose nanofibrils from dry softwood pulp using high shear homogenization. *Carbohydrate Polymers*, 97, 695-702.
- Zuluaga, R., Putaux, J. L., Cruz, J., Vélez, J., Mondragon, I. and Gañán, P. (2009). Cellulose microfibrils from banana rachis: Effect of alkaline treatments on structural and morphological features. *Carbohydrate Polymers*, 76, 51-59.