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Nano-Cellulosic Fibers from Agricultural Wastes

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Abstract

In recent years, the potential of agricultural wastes has received increasing attention from academia and industry. The aim has been to identify strategies for the conversion of low-value wastes into new materials and other value-added products. Cellulose is a naturally abundant polymer that is readily available in various agricultural wastes. It is a linear polymer consisting of β -D-glucopyranose units (disaccharides) joined by glycosidic β -1,4 bonds. Nanoparticles can be extracted from cellulose fibers using a top-down mechanically or chemically treatment. Cellulose nanomaterials have generated significant interest due to their intrinsic properties such as large surface-to-volume ratios, high tensile strength, stiffness, and flexibility in addition to good dynamic mechanical, electrical, and thermal properties. The use of nanocellulose for reinforcement in matrices improves thermo-mechanical properties, decreases the sensitivity of polymers to water, and preserves biodegradability. The mixing of nanocellulose with polysaccharides improves mechanical properties. Nano-sized cellulose fibers possess unique physical, chemical, and morphological characteristics. Hence, nano-sized cellulose fibers are considered versatile materials for addition to polymers, and application in high gas barriers and packaging materials. Other uses include electronic devices, foods, medicine, cosmetics, and health care. This chapter focuses on the cellulose nanofibers attained from banana, pineapple and corn-based agricultural wastes.

Keywords: fiber, post-harvest, nanocellulose, chemical hydrolysis, enzymatic hydrolysis, agricultural waste

1. Introduction

1.1 Agricultural industry

Agricultural development is one of the most powerful tools used to boost shared prosperity and economic growth as well as to alleviate extreme poverty and feed an estimated population of 9.7 billion people by 2050. In 2018, agriculture accounted for 4% of global gross domestic product (GDP) and over 25% of the GDP of some developing countries. However, current food systems pose threats to human health and the planet since agriculture accounts for 70% of water use, while generating

alarming rates of pollution and wastes. According to the World Bank (2021), one-third of the food produced globally is either lost or wasted [1]. **Table 1** shows the GDP of Malaysia by sector from 2019 to 2021 (at constant 2015 prices).

1.2 Crop production

1.2.1 Banana

Banana, scientifically known as *Musaceae*, originated from India to Papua New Guinea, and parts of Southeast Asia [2, 3]. Banana is one of the most popular fruits worldwide that contains various essential nutrients such as potassium, magnesium, Vitamin B6 and niacin that aid in digestion, healthy heart, and weight loss [4]. The rich taste of banana is used in desserts, parfaits, and baked goods. It can be consumed raw or after cooking via frying, boiling, stewing or few other methods [5]. India is the world largest banana and plantain producer. As of 2019, the production of bananas in India was 30.5 million tonnes, which accounts for 26.02% of the global production. The other top global banana and plantain producers include; China, Indonesia, Brazil and Ecuador accounting for 53.94% of the production. It was estimated that the total global production of bananas in 2019 was 117 million tonnes [6]. The main parts of the banana plant consist of the leaves, fruit bunch, suckers, underground stem and male flowers as depicted in **Figure 1a** while b) shows statistical data of the top ten largest banana producing countries and contributors in 2019.

1.2.2 Pineapple

Pineapple (*Ananas comosus*) is a healthy and delicious tropical fruit that originated in South America, where early European explorers named it after a pinecone. It is rich in various minerals and vitamins especially vitamin C and manganese, which are essential for maintaining a healthy immune system and body metabolism. It can be purchased fresh, canned or frozen for a daily diet [8]. The pineapple plant consists of a crown, slip, fruit, leaf, stem and sucker as shown in **Figure 2a**. **Figure 2b** displays the graphical statistics of the worldwide production of pineapple in 2019. As reflected in **Figure 2b**, Costa Rica is the largest producer of pineapple accounting for 3.33 million metric tonnes in 2019. This is followed by the

	Share (%)		Change (%)	
	2020 ¹	2019	2020 ¹	2021 ²
Services	58.1	6.1	−3.7	7.0
Manufacturing	22.6	3.8	−3.0	7.0
Agriculture	7.4	2.0	−1.2	4.7
Mining	6.9	−2.0	−7.8	4.1
Construction	4.0	0.1	−18.7	13.9
GDP	100	4.3	−4.5	6.5–7.5

¹Estimate.

²Forecast.

Note: Total may not add up due to rounding and exclusion of import duties.

Source: Department of Statistics and Ministry of Finance, Malaysia (2021).

Table 1.
GDP of Malaysia by sector, 2019–2021 (at constant 2015 prices).

Figure 1.
(a) Parts of the banana plant [7]; (b) Top 10 largest producers of banana in 2019 [6].

Figure 2.
(a) Parts of pineapple [9]; (b) top 10 largest pineapple producers worldwide [10].

Philippines, Brazil, and Thailand. The total production of pineapples in 2019 was approximately 28.18 million tonnes [10].

1.2.3 Corn

Corn (*Zea mays* L.) is the third largest agricultural crops globally after wheat and rice. Almost 100% grain corns in Malaysia was imported from Brazil, Argentina and other countries. Corn are important source of foods for both humans and animals. There are plenty types of corn planted across the world, which includes dent corn, flint corn, sweet corn, popcorn, flour corn, pod corn and waxy corn. Corn is processed to prepare food and feed ingredients such as breakfast cereals, corn starch, sweeteners (high fructose corn syrup), cooking oil and lysine. Corn is also utilized in manufacturing industrial products such as ethanol and polylactic acid (PLA). The corn stems can be employed to prepare silage for ruminants [11]. While sweet corn has been traditionally popular fruit in the USA, China, and Brazil

Figure 3.
(a) Parts of corn plant [12]; (b) top 10 largest corn producers worldwide [10].

(Figure 3), although it has recently gained popularity in numerous Asian countries including Malaysia [10]. Corn is typically yellow but comes in various other colors such as red, orange, purple, blue, white and black. Whole corn is packed with fiber and contains vitamins B and C and the elements magnesium and potassium that promote gut health and prevent digestive diseases [13].

2. Agricultural wastes

Presently, organic and agro-industrial residues constitutes a great share in over-all global wastes as depicted in Figure 4a which makes it crucial to effectively manage to exploit it. As biomass feedstocks are abundant, it has high likelihood for future application in biorefinery technologies [14, 16]. It was believed that the biomass wastes in Latin America that has most of the lignocelluloses are mostly acquired from maize, soybean, and sugarcane residues [17].

2.1 Banana waste

Banana residues are not considered among the primary agro-industrial waste to be utilized as biorefinery biomass supply in developing countries. Therefore,

Figure 4.
(a) Composition of global solid waste in 2015 [14]; (b) estimated biomass crop residue flows for Latin America in 2012 [15].

banana waste treatment has become an issue that needs to be resolved [18]. The common banana plant wastes include the pseudostem, floral stalk, banana peels and leaves. Elanthikkal et al. [19] added that banana peel waste is a by-product of processing banana chips and baby foods. The edible part of banana consists of 12 wt% of the whole plant, which indicates the remainder is discarded as agricultural waste thereby causing environmental problems. Chang [20] also mentioned that about 40% of bananas that do not fit standards set by supermarkets are discarded by farmers [2]. The waste produced by a single banana plant can account for nearly 80% of the total plant mass. It is estimated that 220 tonnes of by-products are produced per hectare per year [21], which requires innovative ideas to convert the agricultural wastes into value-added products. **Figure 5a** illustrates a mountain of banana waste being discarded daily as it does not fit the cosmetic look set by the buyers.

2.2 Corn waste

Corn stover is the biggest agricultural waste in the United States (**Figure 6**) due to the high productivity of the farms. Corn stover refers to stalks, leaves and cobs of the remnants of corn harvest. About 100 million dry tonnes of corn stover are collected annually making corn stover not only a renewable and sustainable material but also one of the cheapest and most available residues in the US. Corn cobs serve as a novel source of raw material for the microbial production of fuel alcohols

Figure 5.
(a) Mountains of bananas being discarded daily [20]; (b) waste banana fruit peels [22].

Figure 6.
(a) the process of harvesting corn [23]; (b) corn Stover leftover [24].

and other value-added products [25, 26]. Corn harvest consists of 48% stalk of the total dry mass, 28% leaves, 15% cobs, and 8% husk [27].

2.3 Pineapple waste

The pineapple fruit is typically processed into jam, juice, cordial, vinegar, and food flavoring. Solid or biomass wastes such as core (5–7%), peels (30–35%), a crown (10–15%), and stems are accumulated from the processing or harvesting of the pineapple fruit and plants, as shown in **Figure 7**. In addition, the rough handling of fruits and exposure to extreme environmental conditions during transportation or storage contributes about 55% of product waste [30]. After harvesting, the pineapple waste is burnt or left on the ground, which can cause serious environmental issues. It is estimated that 40–80% of pineapple fruit wastes have high biological oxygen demand (BOD) and chemical oxygen demand (COD) values [31].

2.4 Environmental issues

The poor management of agricultural wastes through practices such as open burning and dumping in landfills causes environmental pollution. For instance, high moisture content of banana waste will eventually produce greenhouse gases if it is not properly managed (e.g. by dumping) thereby resulting in disastrous impacts on the environment. Farmers are known to throw banana tree wastes in rivers, ponds, lakes or in low lying areas where slow degradation occurs thereby generating methane, other gases, and spreading putrescible odors that affect the nearby ecosystem. The farming practice of open fire burning to eliminate banana wastes also contributes to severe air pollution issues [21]. Fruit residues also cause severe environmental problems as it accumulates in the agro-industry.

Fruit wastes will eventually lead to serious environmental problems as it accumulates in the agro-industrial yard with zero significance and commercial value. The high costs of transportation and the limited availability of landfills have resulted in the unscrupulous disposal of wastes into the environment. Additionally, the problems of disposing of by-products have worsened due to legal restrictions. Pineapple wastes have high BOD and COD values, which exacerbates management and disposal [32]. Scientists have focused on the co-digestion of pineapple wastes together with several other fruits, legume, manure and slaughterhouse wastes to decrease the volatile solids by 50–65% [33]. The environmental problems of pineapple wastes are evident in the world's largest producer, Costa Rica and other countries where the juice industry discards 50–65% of the residual biomass [34].

Figure 7.
Pineapple residues from: (a) peels [28]; (b) crown [29].

As for the management of corn wastes, the burning of crop residue eliminates the opportunity to improve organic matter content and potentially leads to substantial loss of nutrients. Nearly all nitrogen and at least 75% of the sulfur in plant residues are lost during combustion. A primary tillage practice designed to chop stalks, deep rip, and establish raised beds in a single operation can save considerable time and fuel expense, compared to several conventional tillage operations. Therefore, it is important to identify and characterize the waste produced to minimize environmental pollution by optimizing waste reduction [35].

3. Cellulose nanofibers (CNF)

3.1 Background

The cell wall of plants contains cellulose, hemicelluloses and lignin. The lignin component accounts for 10–25% by dry weight and acts as a binder between cellulose and hemicelluloses. It is lignin that enhances rigidity, durability, and protects the cell wall thereby acting as a binder. The other two major components of the plant cell wall are cellulose and hemicelluloses that comprise 35–50% and 20–35%, respectively, of the dry weight of lignocellulosic biomass. Cellulose is the linear polysaccharide with repeating units of cellobiose (disaccharide *d*-glucose) units linked by β -1,4 linkage. Hence, there are strong intramolecular or intermolecular hydrogen bonds between adjacent glucose units in similar or different chains through the open hydroxyl groups present in glucose monomer units. The hemicellulose in plant cell walls is mainly composed of xylan and glucomannan, which are pentose and hexose monomers linked by short or branched chains. The compact structures of hydrogen bonding are tightly packed networks in cellulose fibers that provide antibacterial properties, toughness, strength, and water or solvent impermeability to the plant cell wall. Nanocellulose is a fiber or crystal-based form of cellulose measuring a few micrometers in length and diameter < 100 nm and typically extracted from plant, animal, mineral and other natural sources. It is biodegradable, lightweight with a density of about 1.6 gm/cc with 10 GPa of high tensile strength similar to cast iron. It also accommodates reactive hydroxyl groups which are ideal for surface functionalization for various applications. The three types of nanocellulose materials are cellulose nanofibers (CNF), cellulose nanocrystals (CNC) and bacterial nanocellulose (BNC) [36]. **Table 2** shows the chemical composition of lignocellulosic materials derived from various agricultural wastes.

3.2 Methods of cellulose nanofibers extraction

Numerous studies on the use of agricultural waste (biomass) as sources of CNF have been widely discussed over decades. The processes for extracting

Source	Composition (%)				Refs.
Lignocellulosic biomass	Cellulose	Hemicellulose	Lignin	Extracts, pectin and waxes	
Corn cob	28–34	39–47	21–29	5–12	[37]
Pineapple leaf	34–40	21–25	25–29	8–10	[38]
Banana peel	60–65	6–8	5–10		[39]

Table 2.
Chemical composition of lignocellulosic materials from various agricultural wastes.

nanocelluloses (NCs) from biomass are categorized into two main procedures: biomass pre-treatment before NC extraction and method of isolation of NC [36].

3.2.1 Pre-treatment of biomass

The initial step for NC extraction is the pre-treatment of lignocellulosic biomass to eliminate hemicellulose and lignin. The two common methods used for biomass pre-treatment are alkali treatment and acid-chlorite treatment as described below.

3.2.1.1 Alkaline treatment

Alkaline treatment is a process whereby the biomass is treated with an alkali reagent to eliminate non-cellulosic components such as pectins, hemicelluloses and lignin. The filtrate is then washed with water to neutralize it and the obtained solid contains mostly cellulosic components. The common concentration of NaOH used for this treatment is 8–16% and the mixture is heated to 90–160°C for 1–2 h for the removal of hemicelluloses and some parts of lignin. Treatment is usually carried out using a strong base solution such as sodium hydroxide or potassium hydroxide with varying conditions as tabulated in **Table 3**.

Novel approaches such as steam explosion have successfully removed the lignin fraction while producing purer and structurally intact cellulose. Next, the solubilized components are eliminated by rinsing with deionized water and filtered. The conditions applied may vary depending on the fiber source and its constitution [43].

3.2.1.2 Acid-chlorite treatment (bleaching treatment)

The acid-chlorite treatment is the partial or complete removal of lignin from lignocellulosic biomass by combined treatment with sodium chlorite acidified with glacial acetic acid. This process is commonly known as the bleaching or delignification process, which is performed by mixing distilled water, sodium chloride and acetic acid with lignocellulosic biomass at 70–80°C for 4–12 h. The blend is stirred overnight followed by washing with distilled water until neutral (pH = 7) is achieved. The white residual product characterized as holocellulose lignin-free is

Lignocellulosic source	Reagent	Conditions	Refs.
Banana peel	KOH (5% w/v) Bran to KOH ratio (1:20)	27–30°C, 14 h, mechanical stirring	[40]
Banana pseudostem	H ₂ O ₂ (10% v/v), NaOH (1% w/v)	100 rpm, 60 min Autoclave; 110°C, 40 min	[41]
Banana fibers	NaOH 1 M	80°C, 4 h	[19]
Banana fibers	NaOH (2 wt%)	Steam explosion; 20 lb. (12°C /1 h)	[42]
Banana, Pineapple leaf	NaOH (2 wt%)	30°C, 6 h	[43]
Corn stover	NaOH (4 wt%)	150 rpm, 80°C, 2 h 2x treatment	[44]
Cornstalk	NaOH (15 wt%)	55°C, 2.5 h, mechanical stirring	[45]
Pineapple leaf	NaOH (5%) Fiber to solution ratio 1:20 (w/v)	Water bath; room temperature, 1 h	[46]

Table 3.
Alkaline treatment of different lignocellulosic fibers.

Lignocellulosic source	Reagent	Conditions	Refs.
Banana peel	NaClO ₂ (1 %w/v) at pH 5 adjusted with acetic acid	27–30°C, 14 h	[40]
Banana pseudostem	H ₂ O ₂ (10% v/v), NaOH (1% w/v)	100 rpm, 60 min in Autoclave; 110°C, 40 min	[41]
Maize straw	H ₂ O ₂ 2% (v/v) and TAED 0.2% (w/v)/ HNO ₃ 65% (v/v)	48°C, 12 h/120°C, 30 min	[47]
Pineapple leaf	NaOH, acetic acid and 1:3 NaClO solution	25°C, 1 h	[48]
Corn stover	50 g NaCl, 500 ml nanopure water, 50 ml glacial acetic acid	30°C, 24 h, mechanically stirred in a fume hood	[44]
Cornstalk	10 ml ethyl ethanol, 10 wt% NaOH	35°C, 2 h, mechanical stirring	[45]

Table 4.
Bleaching treatment of different lignocellulosic fibers.

then collected and oven-dried. **Table 4** shows the varying conditions and reagents used by other researchers for bleaching treatment.

3.2.2 *Methods of nanocellulose isolation*

In the last decades, various methods to manufacture CNF from regenerated cellulose and natural raw materials have been developed. These methods include electrospinning [49, 50], biosynthesis [51, 52], and mechanical isolation. Some review papers have been published to compare these methods [53–55]. The conventional needle-based electrospinning techniques for producing CNF deteriorates from the use of toxic solvents, very low productivity (spinning rate: 10 ml/h), the use of high voltage [56] and poor crystallinity. Biosynthesis by bacteria yields nanofibers with narrow diameter distributions, high aspect ratios and high crystallinity. However, it has several constraints such as the requirement for strict and high-cost production, poor reproducibility between the bacteria of different generations, and the complex post-treatment purification procedures. According to Sharma [36], the extraction of nanocellulose may be performed via three different methods; acid hydrolysis, enzymatic hydrolysis or mechanical treatment processes.

3.2.2.1 *Acid hydrolysis*

Acid hydrolysis is a common method used to isolate NC. The amorphous region of the cellulose fibrils may be hydrolysed using strong acids such as sulfuric acid via the esterification of hydroxyl groups by the sulphate ions. It will subsequently make the crystalline region of cellulose fibers to produce a stable colloidal dispersion of nanocellulose materials in the remaining reaction mixture. Sulfuric acid is the most used acid for the hydrolysis process. Other researchers have also used several mild acids such as formic, acetic, phosphoric and chloric acids for hydrolysis as summarized in **Table 5**. This reaction depends on three primary reaction parameters; reaction time, temperature and acid concentration, which also influences the properties of the nanocellulose product. The washing procedure is normally conducted using cold water followed by centrifugation or using sodium hydroxide until neutral pH is obtained. For instance, Pelissari et al. [40] mixed 1% v/v H₂SO₄ solution with the insoluble residue at 80°C for 1 h to eliminate the mineral traces and hydrolyse amorphous cellulose, providing the nanofibers product. The insoluble residue was then neutralized with alkaline or acid solution (5% KOH or 10% acetic acid)

depends on the pH followed by centrifugation (10,000 rpm; 5°C; 20 min) until neutral pH was obtained. The final residue was diluted with deionized water and the suspension was stored at 4°C in a sealed bottle.

3.2.2.2 Enzymatic hydrolysis

Enzymatic hydrolysis is a biological treatment process that utilizes enzymes for the digestion or modification of cellulose fibers to obtain pure cellulose. The common enzymes used for this process include; cellulase, endoglucanase, cellobiohydrolase. The mechanism is complex but the enzymatic action is based on breaking or catalytic linking of H-bonds between the cellulose microfibrils. The hydrolysis process is critical to the removal of hemicellulose, which protects cellulose from hydrolysis and the production of monosaccharides from hemicellulose for further fermentation to bioethanol. Cellulases and hemicellulases are structurally related both fundamentally and in relation to their reactant systems that typically function in synergy. Both enzymes are usually required for proficient hydrolysis of assorted lignocellulosic biomasses. The process of enzymatic hydrolysis is carried out under mild condition, but the reaction time required is much longer when compared to the acid hydrolysis process.

3.2.2.3 Mechanical treatment process

Cellulose fibers can be mechanically processed to isolate nanocellulose fibers (NCFs) using various mechanical methods such as ultrasonication, ball milling, and

Lignocellulosic source	Reagent	Conditions	Refs.
Banana peel	H ₂ SO ₄ (1 wt%)	27–30°C, 14 h	[40]
Banana pseudostem	H ₂ O ₂ (10 wt%), NaOH (1 wt%)	100 rpm, 60 min Autoclave; 110°C, 40 min	[41]
Corn cob	H ₂ SO ₄ (64 wt%)	45°C, 90 min	[37]
Cornstalk	30 wt% chloroacetic acid-ethanol solution	75°C, 2.5 h	[45]

Table 5.
Acid hydrolysis treatment of different lignocellulosic fibers.

Lignocellulosic source	Mechanical treatment	Conditions	CNF diameter (nm)	Refs.
Banana peel	High-pressure homogenization	Number of passages: 3, 5 & 7 times 1st stage: 500 bar 2nd stage: 50 bar	11–23	[40]
Banana peel	High-pressure homogenization	Number of passages: 5 times 1st stage: 500 bar 2nd stage: 50 bar	310–508	[58]
Corn stover	High-pressure homogenization	Number of passages: 30 times	5–50	[44]

Table 6.
Multiple mechanical treatments used on various sources of lignocellulosic fibers.

high-pressure homogenization. However, the main limitation of these processes is the high input energy requirements, which require the incorporation of pre-treatment methods to reduce energy consumption. Among the mechanical methods, high-pressure homogenization is also widely applied to isolate CNFs from plants [57]. It is important to note that the diameter of CNFs (which usually varies from around 2 to 20 nm in diameter and up to several microns in length) is largely

Extraction method of NCF	
Sample: Fruit peel (unripe mature green <i>Musa paradisiaca</i>) [40]	Sample: Pseudostem [41]
Oxidation prevention method <ul style="list-style-type: none">Samples soaked in potassium metabisulfite. Solution (1% w/v) for 24 h to prevent browning (oxidation) Drying samples in the oven; 60°C, 24 h	Freeze-drying method <ul style="list-style-type: none">Samples were frozen at -50°C for 12 hFreeze-dried for 48 h
Fine grinding and sieving after drying	
1st alkalinisation treatment <ul style="list-style-type: none">Samples treated with 5% w/v KOH solution (ratio 1:20) under constant stirring at room temperature for 14 h	Alkalinisation and bleaching treatment <ul style="list-style-type: none">Dried samples mixed with deionized water. H₂O₂ (10% v/v) and NaOH (1% w/v) at 100 rpm for 1 hMixture placed in an autoclave at 110°C and 0.5 kg/cm³ for 40 minVacuum filtering and rinsing with deionized water three times to remove remaining traces of NaOH
Neutralization reaction <ul style="list-style-type: none">Insoluble residue neutralized to pH 7 with alkaline (5% KOH) or add solution (10% acetic acid)	Neutralization reaction <ul style="list-style-type: none">Using H₂SO₄ to neutralize the suspension (pH 7)Rinsing with deionized water to remove remnant traces of acid
Bleaching treatment (Delignification) <ul style="list-style-type: none">Using 1% w/v NaClO₂ at (pH 5) adjusted with acetic acidRepeated until sample discolored	Acid hydrolysis treatment <ul style="list-style-type: none">A mixture of deionized water, H₂O₂ and H₂SO₄ (6.25% v/v) with the product suspension was kept under constant stirring at 100 rpm for 1 hThe dispersion was autoclave at 110°C and 0.5 kg/cm² for 40 minVacuum filtering and rinsing with deionized water was carried out three times
Neutralization reaction	
2nd alkalinisation treatment <ul style="list-style-type: none">Similar conditions as the first alkalinisation method Neutralization reaction	
Acid hydrolysis treatment <ul style="list-style-type: none">Insoluble residue mixed with a 1% v/v H₂SO₄ solution at 80°C to eliminate mineral traces and to hydrolyse amorphous cellulose to obtain the required nanofibers	
Neutralization reaction	Neutralization reaction
The final product was diluted with deionized water and centrifuged (10,000 rpm; 5°C; 20 min) and the suspension was stored at 4°C in a sealed container	Drying of product for 48 h at 40°C
Homogenization (mechanical treatment) <ul style="list-style-type: none">1st stage; 500 bar2nd stage; 50 barAliquot suspensions passed through homogenizer 3, 5, or 7 times	

Table 7.
Comparison of CNF extraction method from the banana fruit peel and its pseudostem.

determined by the source. **Table 6** summarizes the various conditions of high-pressure homogenization applied by other researchers.

Tables 7–9 further elaborates the methods used by researchers to obtain NCF from the agricultural wastes of banana, corn and pineapples.

3.3 Applications of CNFs

CNFs possess distinctive properties such as high mechanical strength, tunable surface chemistry, high aspect ratio, crystallinity, barrier properties, non-toxicity and biodegradability. The materials have great potential as a source of green packaging, coatings, fillers and other industrial applications [36]. The application of CNF is described hereafter mainly focusing on the wastes from banana, corn and pineapple.

3.3.1 Nanocomposites

Epoxy-based FRP (Fiber-reinforced Polymer) composites have relatively high thermal stability. Nevertheless, it possesses low crack growth resistance that limits mechanical applications. Studies have confirmed that hybridizing fibers and fillers can significantly enhance the properties of epoxy FRPs [59]. The theory of

Extraction method of NCF	
Sample: Corn stover [44]	Sample: Corn stalk [45]
Sample preparation <ul style="list-style-type: none">A chipper shredder was used to process the stover sample that was field dried. The sample was fed through a mill before grinding with a 6-micron screen, then a 1-micron screen, and finally, a 0.5-micron screen.	Sample preparation <ul style="list-style-type: none">The washed corn stalks were dried in a 60 vacuum drying oven for 6 h, smashed, sifted* through a 100-mesh sieve.
Alkali treatment <ul style="list-style-type: none">The sample was suspended in water at 2%(w/v) and blended using a laboratory blender. The sample was rinsed with water through a 75 µm sieve. It was then resuspended and centrifuged.The supernatant was removed and 500 ml of 4 wt % NaOH was added.The suspension was stirred at 150 rpm at 80°C for 2 h. Then, it was washed thoroughly with nano pure water using a 75 µm sieve. The treatment was repeated.After washing, the sample was centrifuged until a clear supernatant was observed and recovered.The sample was air-dried at room temperature.	Alkali treatment <ul style="list-style-type: none">Corn stalk was weighed and 15 wt% NaOH solution was added and transferred into a 5 ml 3-necked flask.This reaction mixture was placed into a wa bath kept at 55°C for 2.5 h with mechanic stirring.The samples were rinsed few times w deionized water and then dried at 60°C.The weighed sample, 10 mL ethyl alcohol, a 10 wt% NaOH solution were added to a 500 ml 3-necked flask and stirred continuously 35°C for 2 h
Bleaching/ delignification treatment <ul style="list-style-type: none">The delignification solution was prepared using 50 g of NaCl added to 500 ml of nano pure water and 50 ml of glacial acetic acid.The sample was added to the delignification solution and mechanically stirred at 30°C for 24 h in a fume hoodThe sample was air-dried after thorough washing with water.	Acid hydrolysis treatment <ul style="list-style-type: none">12 g of 30 wt% chloroacetic acid-ethanol solutions was added to the reactor at 75°C, 2.5 h.The resulting product was rinsed thorough with deionized water repeatedly and fine dried at 60°C to a constant weight.

Table 8.
Comparison of CNF extraction method from corn Stover and corn stalk.

Extraction method of NCF	
Sample: pineapple peels [34]	Sample: pineapple leaves [46]
Alkalinisation and bleaching treatment <ul style="list-style-type: none">The ground pineapple leaves were incubated in NaOH to remove lignin and hemi cellulose, then bleached in NaClO₂ before incubating in HCl to hydrolyse the cellulose into micro-cellulose.	Sample preparation <ul style="list-style-type: none">The fibers were extracted from pineapple leaves bunches by scraping. Afterwards, the fibers were crushed using the RL-L10 MPL crusher to reduce the length.Sieving process was carried out using an SS304 GMP automatic sieving machine for segregated fibers below 2.00 mm.
Neutralization reaction <ul style="list-style-type: none">After each chemical treatment, the product was thoroughly rinsed with water until a neutral pH was obtained. The solid was recovered by centrifugation at 13000 rpm	Alkali treatment <ul style="list-style-type: none">The short pineapple leaf fiber was soaked in 5 wt % of NaOH solution in a water bath for 1 h at room temperature. The ratio of the fibers and the solution was 1:20 (w/v).The samples were washed and rinsed ten times using distilled water.
Acid hydrolysis <ul style="list-style-type: none">The sample was hydrolysed with H₂SO₄ to attain nanocellulose.Two fractions of particles were separated from the acid solution by centrifugation at 2500 rpmThe solid was incubated with a second H₂SO₄ solution between 30 min and 4 h at 55°CThe product was separated from the liquid viscous phase by centrifugation at 2500 rpm and rinsed few times using deionized water	Composite preparation <ul style="list-style-type: none">All raw materials for the preparation of the composite samples including the treated and untreated fibers, tapioca biopolymer and coupling agents were oven-dried at 80°C for 24 h.

Table 9.
Comparison of CNF extraction method from pineapple peels and leaves.

Hybrid polymer composites	Lignocellulosic source	Refs.
Kenaf/phenolic resin	Silane treated pineapple leaf fiber (PALF)	[60]
Kenaf/Polylactic acid	Cornhusk flour	[65]
Woven Kenaf/unsaturated polyester resin	Banana fibers	[66]
Glass/vinyl ester	Pineapple leaf fiber	[67]

Table 10.
Nanocomposites based on cellulose nanofibers.

hybridization offers great opportunities for applications in automotive, aerospace and construction [60, 61]. The incorporation of nanofillers in FPR improves the physical, mechanical, morphological, thermal and dynamic properties of the composites [62]. Hence, CNF has become a viable alternative when compared to carbon nanofillers [63] due to its biodegradability, high aspect ratio and significant mechanical properties [64]. **Table 10** summarizes the previous research works on the developed Kenaf hybrid thermoplastics, thermoset and biopolymer composites for advanced applications.

3.3.2 Packaging

The synthetic plastic wastes generated from the food packaging industry are disastrous to the environment if left unrecycled. Hence, the development of biodegradable films and coatings has been widely studied recently. These biopolymers

Packaging blend	Lignocellulosic source	Refs.
Polyvinyl alcohol/polyacrylic acid/NCF	Banana pseudostem	[69]
Polyvinyl alcohol/NCF	Banana peel	[73]
Banana starch/Glycerol/NCF	Banana peel	[74]
Potato starch/Glycerol/NCF	Pineapple leaf	[75]

Table 11.
Packaging based on CNFs.

are renewable, abundant, cost-effective and non-toxic. Furthermore, the tensile strength and elongation at break are considered the main mechanical properties required to preserve the integrity of the packed food. Since nanocelluloses possess these properties, they are ideal components of biodegradable food packaging [68]. The popular product features of NCF-based packaging required by researchers include a shelf-life extension [69], heat resistance [53], and intelligent packaging [70]. Other properties include transparent packaging [71], ultra-violet screening packaging, and anti-microbial packaging [72]. **Table 11** shows the various NCF-based packaging blends made from lignocellulosic banana, pineapple and corn residues for packaging by researchers.

4. Comparison of bioplastics with petroleum-based plastics

The plastics industry began in the early 1900s when the first synthetic plastic was created by Leo Hendrik Baekeland in the United States [76]. Since the industry began, annual global plastic production has grown extensively from some 1.5 million metric tons in 1950 to 359 billion metric tons in 2018. The cumulative production of plastic has already exceeded eight billion metric tons worldwide, with further increase is expected in the future. The increase in plastic waste has become a global environmental issue. Although recycling has become more widely practiced in recently, many plastic materials produced over the last 70 years has ended up in landfills, and often ends up in water bodies, contributing to the environmental pollution [77].

Although plastics production uses less than 5% of all gasoline, its renewability is usually a source of concern. Separating various forms of petroleum-based recyclable plastics from other solid wastes is a time-consuming and labor-intensive process, hence, only a limited proportion of plastics are recycled. The failure of petroleum-based plastics to biodegrade is also a point of contention among environmentalists [78].

Table 12 show a collection of LCA literature data [79, 80]; each LCA characterizes and compares the environmental impact of various bioplastics (thermoplastic starch (TPS), polylactic acid (PLA), and polyhydroxyalkanoates (PHA) and traditional plastics (high and low density polyethylene, Nylon 6, polyethylene terephthalate (PET), polystyrene (PS), polyvinyl alcohol (PVOH) and polycaprolactone) with an approach cradle to grave.

Overall, the statistics in **Table 12** demonstrates how the manufacture and utilization of bioplastics is more energy efficient and reduces greenhouse gas pollution as compared to traditional plastics. In contrast, bioplastics has a significant environmental effect due to soil acidification and eutrophication, owing to the use of fertilizers and chemicals in the processing of organic raw materials used in bioplastics manufacturing. However, the inclusion of non-biodegradable

Type of Plastic	Energy Requirement (MJ/Kg)	Global Warming (Kg CO ₂ eq/Kg)
Non-renewable source		
HDPE	80.0	4.84
LDPE	80.6	5.04
Nylon 6	120.0	7.64
PET	77.0	4.93
PS	87.0	5.98
PVOH	102.0	2.70
PCL	83.0	3.10
Renewable source		
TPS	25.4	1.14
TPS +15% PVOH	24.9	1.73
TPS + 60% PCL	52.3	3.60
PLA	57.0	3.84
PHA	57.0	Not Available

Table 12.
Energy requirement and global warming data for each type of plastic.

copolymers in bioplastics necessitates a substantial rise in energy consumption and CO₂ emissions as compared to bioplastics. Indeed, non-biodegradable copolymers are added to mechanical biopolymers to boost their efficiency, lowering the biodegradable capacity of the resulting material. It is vital to note that the above LCA findings were obtained using incineration with energy recovery as the final provision: this option is not especially beneficial to bioplastics due to their low calorific value.

The study of LCA data consistently has confirmed that bioplastics have certain environmental impact indices that are lower than other conventional plastics, although other indices favor the latter; hence, the need to develop an overall environmental impact index that incorporates and accurately weighs all indices [81].

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