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Banana Peel Waste as a Source of Nanocellulose Materials - Recent Progress, Challenges and Future Prospects

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Abstract: *This review highlights the recent progress in nanocellulose (NC) extraction from banana peel waste. Recent studies have successfully extracted nanocellulose in the forms of cellulose nanocrystalline (CNC), cellulose nanofibers (CNF), and bacterial nanocellulose (BNC). Acid hydrolysis using H_2SO_4 has emerged for extracting CNC, while a combination of chemical and mechanical fibrillation has been most utilized for CNF extraction, and bacterial fermentation for BNC. The critical challenge for the industrial production and commercialization of NC is to produce low-cost material, reduce production costs and energy consumption, and implement environmentally friendly manufacturing processes. Future studies should explore other efficient methods, such as TEMPO-mediated oxidation. Surface modification shows promise in enhancing the hydrophobicity of banana peel NC, potentially unlocking applications in diverse industries. However, to ensure sustainability, it is crucial to consider the three pillars of sustainability. Therefore, future research should include process optimization to improve efficiency and eliminate unnecessary actions in the process.*

Keywords: Nanocellulose, banana peel, circular economy, acid hydrolysis, sustainability

1. INTRODUCTION

According to Statista [1], in 2021, the volume of bananas produced worldwide reached approximately 125 million tons (MT) and this has been generally increasing since 2010. In 2021, Asia was the leading region from banana production with approximately 68 MT, next to Africa, South America, and Central America with the corresponding volume produced of 23 MT, 19 MT, and 10 MT, respectively [2]. In the Philippines, the volume of banana produced in year 2021 was amounted to approximately 9.01 MMT reflecting a slight decrease from the previous year (9.1MMT) [3]. Therefore, in that year, the volume of BP produced can be estimated to be around 3.15-4.50 MMT as the weight of the accounts 35-50% of the total weight of the fruit [4]. According to the Agricultural Outlook 2021-2030 report, the increase in banana production, anticipated to grow by 1.4% annually and reach 138 million tons by 2030, will result in a corresponding rise in the volume of banana peel waste [5]

The current approach for banana peel disposal is landfilling. This is not a sustainable approach as the action requires additional effort and cost for the site disposal, labor, and transportation. Additionally, the presence of this waste in the landfill can pose major environmental problems, such as soil and water pollution, eutrophication, the greenhouse effect, global warming, and other health concerns when not properly managed due to their fermentability and high biodegradability [6]. Therefore, utilization of these agricultural waste is of high significance. Recent efforts were made to explore the potential of banana peel waste as feedstock for ethanol production [7], a novel material for wastewater treatment [8], for cellulose and NC production [9], for food production and other industrial applications.

Among these, banana peel as a source of NC has emerged and is gaining popularity in the market due to the unique properties of this biomaterial. NC is a naturally occurring nanomaterial having a size of 100 nm in any of its dimensions [10], that can be derived from the cell walls of plants [11] including banana wastes and by-products such as banana peel. According to Pereira et al. [12], the cell wall of banana peel is made up of polysaccharide fractions, represented by cellulose (18–59%), hemicellulose (17–20%), and pectin (10–20%), suggesting that banana peel is a promising source of NC. For this reason, numerous research studies have emerged aiming to explore the potential of banana peel NC in packaging applications [13-14], water filtration [15-16], nanopaper [17], biomedical and biosensors [9], and as a reinforcing agents for diverse applications[18-22]. Given the promising potential of banana peel as a raw material for NC production and its sustainability for diverse applications, it is crucial to track recent progress and challenges of this technology. This will help identify possible research areas to further improve the technology. While some reviews on the potential of BP as a source of phenolic compounds [23], biosorbents [24], bioactive ingredients [25], and other value-added materials. Also, a review on banana biomass waste as a resource for NC exists, it primarily focuses on applications in the food industry, such as food packaging and stabilizers [26], there has been no specific review on the recent progress of BP as a resource of NC. As a response, this mini review aims to explore recent advancements, challenges, and research opportunities in banana peel NC production and commercialization.

2. COMPOSITION OF BANANA PEELS

As shown in Fig. 1, the components of the banana fruit include the neck, fingertip, peel, and pulp. The neck of a banana fruit is the section that connects the banana to the crown, serving as the attachment point to the main stem,

while the fingertip is the small, rounded end of the banana, opposite the neck, marking the termination of the fruit. The peel serves as the outer covering of the banana fruit [27], while the flesh, or pulp, is the edible inner part. According to Hikal et al. [27], the average weight of a banana fruit is 125 grams, consisting of approximately 75% water and 25% dry matter in which peel accounts for 35%-50% of the total mass fruit [4].



Fig. 1. Component of Banana fruit. Figure modified from [28].

In Table 1, the chemical composition and the lignocellulosic fraction in dry matter of banana peel is presented. The three main polysaccharides, illustrated in Fig. 2, present in the cell wall of banana peel are lignin (18.4%), hemicellulose (17-20%), and cellulose (18-59).

Table 1. Chemical and lignocellulosic fraction of banana peels

Chemical Fractions (%)		Reference
Crude protein	5.5-7.87	[26]
Lipid	2.24-11.6	
Ash	9-11	
Carbohydrate	59.51-76.58	
Dietary fiber	47-53	Reference
Lignocellulosic fractions (%)		
Lignin	18.4	
Hemicellulose	17-20	
Cellulose	18-59	[12]
Pectin	10-20	

However, the composition and content of these three components vary depending on the species, types, and sources of lignocellulosic biomass [11]. In plant cell walls, lignin acts as the adhesive, binding cellulose, and hemicellulose complexes together [29]. Hemicellulose, a

heteropolymer, consists of short, linear, and branched chains of various monomers such as pentoses and hexoses, adhering to cellulose fibrils via hydrogen bonds and Van der Waals interactions [11].

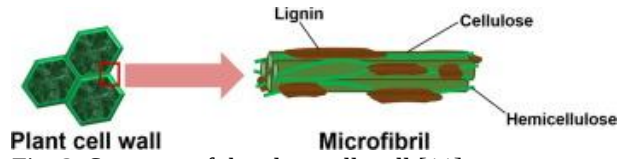


Fig. 2. Structure of the plant cell wall [11].

Cellulose, the primary component in lignocellulosic biomass, is predominantly found in the plant cell wall and serves as the main source for NC extraction. To extract the cellulose from the cell wall of the plant, the main goal is to remove the lignin and hemicellulose as they give a shield effect to the cellulose component.

3. APPLICATIONS OF NANOCELLULOSE

With the growing concerns in environmental sustainability, alongside challenges such as resource depletion and pollution, NC presents itself as a solution offering renewable and biodegradable properties; therefore, potentially replacing the conventional petroleum-based materials [30-31]. It can be used as filler to develop a biodegradable material that could replace a single used plastic items [32]. NC are sustainable biomaterials derived from nature that include both cellulosic properties and the key features of nanomaterials, including a large surface area, adaptable reactive sites or functions, and stability in the scaffolding to support inorganic nanoparticles [33]. Due to these unique properties of NC, it gained wide attention in packaging industry, food industry, paper industry, textile industry, automotive industry, construction industry, electronics and electrical industry, cosmetics industry, biomedical applications, and other industries as shown in Figure 3.

In fact, the worldwide NC market was worth USD 351.5 million in 2022, and it is expected to increase at a compound annual growth rate (CAGR) of 20.1% from 2023 until 2030 [34].

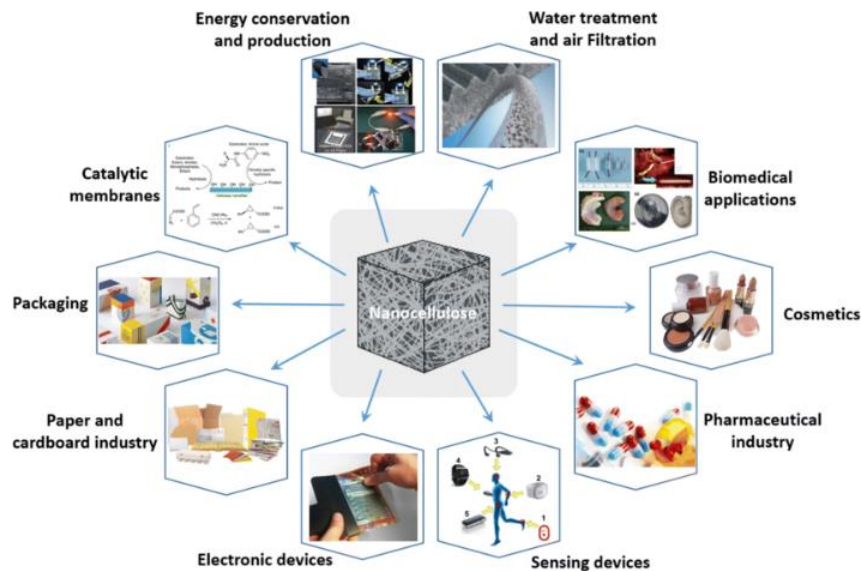


Fig. 3. State of the art applications for new nanocellulose based materials [35].

In terms of NC extracted from banana peel wastes, it was found that this nanomaterial has the potential for packaging applications [13-14], water filtration systems [15-16], nanopaper [17], stabilizer [36], biomedical and biosensors [9], bioplastic for vehicular use [37], and reinforcing agent [19, 20, 22, 36].

4. RECENT PROGRESS IN BANANA PEEL NANOCELLULOSE PRODUCTION

In recent years, banana peel has been explored for its potential in NC extraction such as cellulose

nanocrystalline (CNC), cellulose nanofibers (CNF), and bacterial nanocellulose (BNC) [38], as summarized in Table 2. These three main types of NC vary according to their extraction method and characteristics. CNC are rod-like nanostructures with high crystallinity which can be extracted using chemical synthesis, CNF are long, flexible fibers with both crystalline and amorphous regions and are usually obtained using mechanical processes and chemical treatment, while BNC is produced by bacteria and enzymes and consists of highly pure, entangled fibers.

Table 2. Recent studies on banana peel wastes as a resource of nanocellulose

NC	Isolation Method	Applications	Highlights	References
CNC	One-pot oxidation-hydrolysis system	Cellulose-based polymers	Average size of 362.2-171.6 nm (unripe peels), and 701.6-78.6 nm (ripe peels).	[39]
CNC	Acid hydrolysis	Bioplastic for vehicle use	Particle size is 20 nm	[37]
CNC	One-Pot Microwave and Mild Oxidative Hydrolysis System	Reinforcing agents	High I_C of 85% was obtained at 6% H_2SO_4 , average size of 20 nm, ZP of -42.9 mV, yield is 28.1%.	[18]
CNF	High intensity ultrasonication combined with chemical treatment	Reinforcement agent (nano composites)	Diameter is 20-35 nm, Length is 263-589 nm, Aspect ratio is 13.19-16.83, I_C is 30.5-63.64%	[40]
CNF	High intensity ultrasonication combined with chemical treatment	Nanopaper	Average diameter is 10.3-43.8 nm, I_C is 37.16-68.4%, ζ -potential is -2.6 to 5.4 mV	[17]
CNF	Alkaline treatment, bleaching, and acid hydrolysis	Polymer composites	Yield is 5.1% (db), ζ -potential is 16.1 to -44.1 mV, diameter of 10.9-22.6 nm, length of 335.1-454.9 nm, aspect ratio of 15.1-42.7 nm	[41]
CNF	Microwave digestion method and ball milling assisted ultra-sonication, acid hydrolysis	Cellulose-based polymers	Yield is $76.58 \pm 1.97\%$ (w/w) based on micro cellulose, diameter of 73 nm, average PS based on DLS is 1285 nm, I_C is 77.2%.	[42]
CNF	Alkaline treatment followed by enzymatic treatment with xylanase	Biodegradable materials	I_C of 66.2%, diameter 3.7 nm, ζ -potential of -29.1 mV	[43]
CNF	Chemical and enzymatic treatment	Reinforcing agents	Average diameter of 10.9 nm (CT), and 7.6 nm (ET), length of 454.9 nm (CT), and 2889.7 nm (ET), CI is 58.6% (ET) and 49.2% (ET).	[31]
CNF	Alkaline treatment, bleaching, acid hydrolysis,	Reinforcing agents	Average diameter of 3.72 nm, ζ -potential values ranging from -37.60 to -67.37 mV, CI of 63.1-66.4%. Regarding	[21]

NC	Isolation Method	Applications	Highlights	References
	high pressure homogenization		cytotoxicity, low CNF concentrations (50–500 µg/mL) did not cause cell death, but CNFs at concentrations above 1000 µg/mL significantly decreased cell viability	
CNF	Alkali treatment, bleaching, acid hydrolysis	Biomedical and biosensors	The optimal condition is 200 mL of sulfuric acid 55% v/v at 50 °C for 60 min with the yield of 29.9%. PS is 209 nm and ζ-potential are -43 mV, I _C is 64.12%.	[9]
CNF	Alkaline treatment, enzymatic hydrolysis using enzyme xylanase, ultrasound, and high-pressure homogenizer	Stabilizer	Length of 1492-3493 nm, diameter of 3.3-3.5 nm, aspect ratio of 450-972 nm, ζ-potential of -24.3 to -57.9 mV, I _C of 51-78%.	[36]
CNF	Alkali treatment, bleaching, acid hydrolysis	Reinforcing agents	I _C of 53.48-56.50%, best sample was noted in 5% H ₂ SO ₄ concentration.	[20]
CNF	One-Pot Nitro-Oxidation method	Reinforcing agents	Diameter of 20-50 nm, and CI of 21-63%.	[22]
CNF	Chemical and mechanical treatment	Packaging	Addition of CNF reduce film's water solubility and water vapor permeability, improved mechanical, and thermal properties.	[13]
BNC	Bacterial fermentation using <i>Gluconacetobacter xylinus</i>	Water filtration	PS is 30-50 nm, I _C is 86.94% and cellulose type I, ζ-potential is -11.39 mV	[15]
BNC	Bacterial fermentation using <i>Gluconacetobacter xylinus</i>	Water filtration	Diameter sizes between 30-50 nm, bacterial growth pH range of 3.93–4.26.	[16]
BNC	Bacterial fermentation using <i>K. xylinus</i> IITRDKH-20 strain	Active packaging	Yield is 9.24 ± 0.31 g/L (static intermittent fed-batch), and ~5.34 ± 1.03 g/L (static batch)	[14]
BNC	Wet retting method	Reinforcing agents	Maximum thermal decomposition temperature is 405°C, I _C of 15.75-27.55%.	[19]

CNC-Cellulose nanocrystalline; **CNF**-Cellulose nanofibers; **BNC**-Bacterial Nanocellulose; **PS**-Particle size; **I_C**-Crystallinity index; **DLS**-Dynamic light scattering; **CT**-Chemical treatment; **ET**-Enzymatic treatment.

Acid hydrolysis is a primary method for extracting CNC from banana peel, utilizing strong acids like sulfuric acid, hydrochloric acid, nitric acid, phosphoric acid, and hydrobromic acid [26] to hydrolyze the disordered regions in the cellulose fibrils of banana peel. Sulfuric acid is the most commonly used acid because it effectively isolates nanocrystalline cellulose and disperses it into a stable colloidal system through the esterification of hydroxyl groups by sulfate ions [11]. Aside from this method, Mohd Jamil et al. employed a one-pot microwave and oxidative hydrolysis system for CNC isolation from BP [18]. This technique is a combination of microwave pre-treatment and mild oxidation hydrolysis using hydrogen peroxide (H₂O₂), and sulfuric acid (H₂SO₄). The extracted CNC exhibits a promising characteristic such as high crystallinity index (I_C) of ~85% and average size of 20 nm being obtained at 6% H₂SO₄. A high I_C (>80%) of NC is a good manifestation that the amorphous regions in cellulose microfibrils have been effectively removed. NC with higher crystallinity, produced via a one-pot isolation method, generally offers superior tensile strength, reinforcement, and thermal stability, making it ideal for high-strength nanocomposite materials.

The isolation of CNF from BP is usually performed using the combination of chemical and mechanical treatment. For example, a chemical treatment paired with a high-intensity ultrasonication was employed by Khawas et al.

[17], in which the produced CNF is in nanosized range (10.3-43.8 nm), with an I_C of 37.16-68.4%. However, the obtained ζ-potential is -2.6 to 5.4 mV which suggest that the particle has low charge and would clump together due to a lack of charges to repel each other. NC is considered stable when its ζ-potential exceeds 30 mV, while absolute values below 15 mV are associated with particle agglomeration [44]. Tibolla et al. [21] were able to isolate CNF with a stable ζ-potential in the range of -37.60 to -67.37 mV using chemical treatment and high-pressure homogenization technique. Costa et al. used the same isolation procedure, resulting in extracted CNF with a stable ζ-potential ranging from -24.3 to -57.9 mV [36]. A NC with stable ζ-potential suggests better electrostatic repulsion between charged particles, leading to improved dispersion, which is a desirable property of a nanofiller [45]. Another method, called the one-pot nitro-oxidation method, was utilized by Van Nguyen and Ngan Nguyen for CNF isolation from banana peel [22]. The diameter of the extracted CNF was in the nanoscale range (20-50 nm), and it exhibited I_C of approximately 21-63%.

Meanwhile, the BNC isolation method from BP is facilitated by certain types of bacteria such as *Gluconacetobacter xylinus*, the most efficient producer of BNC [26]. This approach was utilized by Sijabat et al., where extracted BNC falls in nano range, that is 30-50 nm, with less stable ζ-potential of around -11.39 mV [15]. Naeem et al. employed the wet retting method for

successful extraction of BNC from BP [19]. In this method, ~7 g of dried banana peel fibers was added to a Hestrin and Schramm culture medium, autoclaved, inoculated with *Gluconobacter* strain, and fermented at 30 °C for 3 days under agitation, leading to the formation of BNC coating the fibers, which were then treated with NaOH, washed, and dried to produce self-assembled BNC. The obtained BNC exhibited a thermal decomposition temperature of 405 °C, and a relatively low I_C (15.75-27.55%) suggesting the presence of amorphous regions in the cellulose microfibrils.

5. CHALLENGES

Despite its excellent properties and versatility, NC has not yet been fully commercialized due to its high production costs, significant energy consumption, and concerns about environmental and human health safety [46]. While the raw material, BP, is generally regarded as waste in this context, it is important to note that other materials used for chemical treatment and acid hydrolysis can be quite costly. Therefore, a critical challenge for the industrial production and commercialization of NC is to achieve the production of low-cost material, reduce production costs, minimize energy consumption, and implement environmentally friendly manufacturing processes.

Acid hydrolysis has several drawbacks, including severe equipment corrosion, significant cellulose degradation, substantial environmental pollution, and high water consumption [47], posing another challenge in the economics and environmental impact of this technology. While it is possible to recover the acid in this method, the minimum product selling price of NC produced in this approach is higher than no acid recovery [48]. Another isolation technique that does not require extensive acids is the mechanical process in which high shear force is applied to cleave cellulose fibers along the longitudinal axis, producing nano fibrillated cellulose; common methods include high-pressure homogenization, ultrasonication, and ball milling [11]. Although it temporarily addresses the problems encountered in acid hydrolysis, this is an energy-intensive approach. Additionally, other issues include the overheating of raw materials, low recovery and uniformity of NC, and a reduction in the crystallinity of CNF [49]. Also, enzymatic hydrolysis is a time-consuming approach as it requires long reaction time.

In addition to the drawbacks encountered during NC production, another significant challenge preventing NC from reaching its full market potential is the lack of process-adapted measuring tools [50]. These tools need to characterize NC quickly and reliably to meet industrial demands cost-effectively.

6. FUTURE DIRECTIONS

6.1. Isolation Method

While previous studies have successfully extracted NC from BP wastes, there are several isolation techniques that have not yet been utilized or investigated for their effects on NC characteristics. For instance, the TEMPO-mediated oxidation has not yet been employed for CNF extraction from BP. This technique is a promising and efficient method to transform plant cellulose into

cellulose nanofibrils, requiring minimal mechanical force to break down the structure of the oxidized products and isolate the cellulose nanofibrils [51]. The effects of other isolation techniques, such as deep eutectic solvents, ionic liquid treatment, micro fluidization, supercritical fluid processing, plasma, and steam explosion on the properties of banana peel NC have not yet been investigated.

6.2. Surface Modification

NC in the form of CNC, CNF and BNC, exhibits strong hydrophilicity due to the abundance of hydroxyl groups [52], as illustrated in Fig. 4. They also contain anionic units, such as sulfate or phosphate esters, introduced during hydrolysis, which significantly enhance their hydrophilic nature [53]. The inherent hydrophilicity of NC complicates its dispersion in hydrophobic polymers, often resulting in aggregated structures that undermine its effectiveness as a reinforcing filler in conventional plastics [53]. The agglomeration of NC particles presents a significant challenge in achieving optimal physico-mechanical properties in composite materials, thereby creating new opportunities for research on surface modification. Through surface modification approach, the hydrophilic nature of cellulose is enhanced, leading to improved compatibility with a wide range of non-polar and polar matrices.

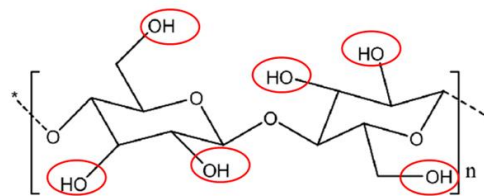


Fig. 4. Chemical structure of nanocellulose. Red circle represents the hydroxyl groups in nanocellulose structures.

In recent years, numerous studies on the surface modification of NC have emerged to enhance its effectiveness as a drug delivery agent [54], to improve the adsorption capacity to contaminants [55] and heavy metals [56], to develop a biocompatible, stable, and hydrophilic substrate for MRI [57], and as a smart and sustainable bio-materials [58]. Surface modification of NC involves several methods, such as chemical approaches including acetylation, esterification, and oxidation of surface hydroxyl groups [55], silylation, amidation, carbamation, sulfonation, and phosphorylation [53]. This provides opportunities for future studies to explore the potential of surface modification of NC isolated from banana peel wastes. This will widen the applications of NC from banana peel in diverse applications or industries.

6.3. Sustainability

The increasing focus on extracting NC from a range of agricultural and industrial wastes is predominantly driven by the ongoing quest for sustainability. While the concept of utilizing banana peel as a resource for new, high-value, and promising nanomaterials sounds appealing, it does not guarantee sustainable effects. A sustainable process or material must adhere to the three pillars of the sustainability triangle [59], as illustrated in Fig. 5. A widely accepted definition of 'sustainability' involves three interconnected 'pillars' that encompass

economic, social, and environmental (or ecological) factors or goals [60].

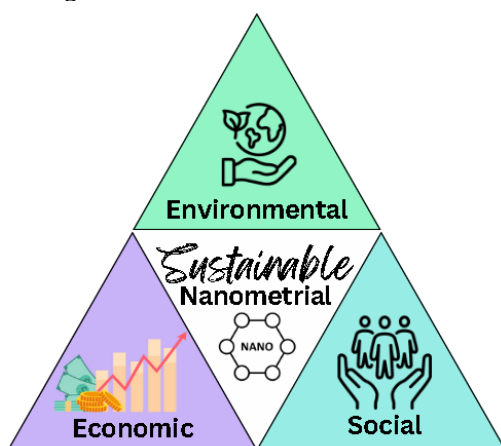


Fig. 5. Sustainable triangle of a nanomaterials.

Future studies must consider assessing the economic feasibility of this technology by employing a techno-economic analysis (TEA). As NC production technologies advance, it is crucial to assess their economic feasibility, technological maturity, and commercialization potential through systematic TEA [61]. TEA is a thorough methodology utilized to evaluate the economic viability and sustainability of new technologies. Aside from this, environmental sustainability must also be considered, hence, future studies must investigate the impact of NC and its production processes on the environmental aspects. This can be carried out using a thorough life-cycle assessment (LCA). An LCA is a method used to evaluate a product's environmental impact across its entire life cycle, from the extraction of natural resources to its disposal [62]. It allows us to quantify the waste, emissions, and resources consumed throughout the process [63], from the production of raw banana peels to the extraction of NC. Finally, future study must also assess the social sustainability of this technology. As noted by Woodcraft, social sustainability involves creating sustainable, thriving communities that promote well-being by understanding and addressing the needs of people in their living and working environments [64]. Future studies on NC extraction from banana peel wastes should explore the social sustainability aspects, such as the potential for job creation in local communities and the improvement of waste management practices. Additionally, research could assess the social acceptance and benefits of using agricultural waste for high-value products, promoting community engagement and sustainable development. Also, it is essential to assess the potential health risk posed by this technology to society. A good reference of this is the study of Stoudmann et al., [65] on the environmental risk assessment of NC, and their review on human hazard potential of NC [66], and risk assessment of NC [67].

Along with these three pillars of sustainability, it is essential to establish an optimum process for NC extraction through process optimization. Process optimization is a way to eliminate unnecessary costs by analyzing and redesigning existing processes to remove inefficiencies. By eliminating the inefficiencies through the process of extraction, it enables us to minimize the production and labor cost as well as the volume of CO₂

emissions. One statistical approach that is particularly useful for this approach is the response surface methodology (RSM) [68]. This technique was first introduced by Box and Wilson and is a collection of mathematical and statistical techniques in which the purpose is to analyze, by empirical model, the problems being encountered. RSM includes the optimization of the process for the settings of factorial variables, in such a way the responses reach the desired maximum, minimum or ranged value [69]. With this approach, ideal conditions may be quickly reached at a minimal cost and without requiring significant experimental testing. Additionally, the response surface representation makes it easy to understand how factors relate to one or more response variables [70]. In recent years, process optimizations for NC extraction from various agricultural wastes have been conducted, focusing on factors such as acid concentration, reaction time and temperature, loading rate, homogenization rate, and cellulose-acid concentration [71-73].

7. CONCLUSIONS

This mini review has highlighted the recent progress on NC derived from banana peel waste. Recent studies were conducted and successfully extracted NC in the form of CNC, CNF, and BNC. Acid hydrolysis using H₂SO₄ was the most utilized method for extracting CNC, while a combination of chemical and mechanical fibrillation was the most utilized method for CNF extraction, while bacterial fermentation for BNC. Future studies may consider some other efficient and effective methods such as TEMPO facilitated oxidation that has been employed in diverse agricultural wastes but not in banana peel to be exact. Surface modification also presents promise in terms of enhancing the hydrophobicity of banana peel NC and potentially unlocking its applications in diverse industries. Furthermore, utilization of this discarded waste not for synthesis of such high-value material does not warrant sustainability not unless three pillars of sustainability are considered. This suggests that in future studies, process optimization, techno-economic analysis, life cycle assessment and social impact investigation should be considered. In brief, banana peel wastes as a resource of NC presents promise considering the unique characteristics of this nanomaterial and the volume of this waste generated.

8. ACKNOWLEDGEMENT

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