Potential of polypropylene nanocomposite reinforced with cellulose nanofiber from oil palm empty fruit bunch as sustainable packaging: A review

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Abstract. Sustainable packaging focuses on the production of packaging that promotes environmental, social, and economic health. The use of thermoplastic such as polypropylene (PP) in packaging has raised concern about environmental impact, so research needed to identify alternative sustainable packaging materials to reduce the environmental impact. Cellulose Nanofiber (CNF) has been considered sustainable packaging material due to its low weight, high strength, high abundance, rigidity, and biodegradability. Therefore, CNF from Oil Palm Empty Fruit Bunch (OPEFB) is the potential additional raw material for developing sustainable packaging. CNF can be used as additional raw material to reinforce the PP matrix, called a polypropylene nanocomposite (PPNC). However, limited research has focused on CNF preparation for PPNC production. Therefore, this review is to explain the potential PPNC reinforced with CNF from OPEFB as sustainable packaging.

1. Introduction

A package is goods or tools used to hold, handle, protect, and present products from producers to consumers. A package is produced according to the type of packaging materials that can be divided into polymers, glass, papers, textile, woods, metals, ceramics, and other types of packaging materials [1]. The packaging materials used in the food and beverage packaging, medical, pharmaceutical packaging, and other application are non-degradable (such as glass, plastics, and metals), and this packaging can increase the environmental impact [2]. This situation caused many requests to increase the exploration and use of eco-friendly packaging. Numerous studies have reported that packaging materials based on biopolymer can reduce the waste from non-biodegradable packaging [3,4].

CNF from natural resources has been recognized as a renewable polymeric material and abundant in nature and as the primary source of sustainable raw materials for sustainable packaging production. CNF has attractive characteristics, such as biodegradability, chemical stability, and biocompatibility. CNF has been utilized as raw materials to produce papers, textiles, and pharmaceuticals [5,6]. In recent years, CNF has attracted researchers’ interest for maximization of the mechanical properties of packaging. The additional CNF in packaging production can minimize the production cost and
minimize the environmental impact [7,8], and then it is can then provide tensile, flexural properties, and superior rigidity [9].

Process design to CNF isolation will ensure the requirement of packaging and product quality such as packaging safety levels, size, ergonomics, height, thickness, stress levels, lifecycle, and cost [10], and define the suitability and limitations of CNF in the development the sustainable packaging [11].

Natural material potential as raw material to CNF isolation is Oil Palm Empty Fruit Bunch (OPEFB) has generated from oil palm extraction process. OPEFB production in Indonesia will continue to increase with increasing the amount of oil palm production. OPEFB has the potential to be developed into products that have added value. Numerous studies have researched CNF isolation from OPEFB (Table 1).

Table 1. Studies researched CNF isolation from OPEFB

| No | Methods                              | Characteristics | References |
|----|--------------------------------------|-----------------|------------|
| 1  | Sulfuric acid hydrolysis             | Size: 1 – 3.5; 53.83 – 58.78 | [12]       |
| 2  | Sodium chlorite hydrolysis           | Size: 4 – 15; 30.77 – 50.38 | [13]       |
| 3  | Acid hydrolysis                      | Size: 1 – 100; 42.52 – 80.42 | [14]       |
| 4  | Sulfuric acid hydrolysis             | Size: 5 – 10; 49.92 | [15]       |
| 5  | Acid hydrolysis                      | Size: 30.717 – 70; 26.65 – 76.15 | [17] |
| 6  | Enzyme hydrolysis from *Trichoderma sp.* | Size: 32.6 – 36.4; 26.65 – 76.15 | [18]       |
| 8  | Sulfuric acid hydrolysis             | Size: 894.25; 31.1 | [19]       |
| 9  | Chemical and Nano grinding treatment | Size: 17.82 | [20]       |

Based on data from Badan Pusat Statistik (BPS) from 2014 to 2017, palm oil production in Indonesia has increased from 29.28 to 34.94 million tons per annum. OPEFB fiber is made up from three complex polymers (cellulose, hemicellulose, and lignin). OPEFB is byproduct that have high cellulose content [20]. Cellulose content in OPEFB around 30 – 40% (w/w) [21], 37.26% (w/w) [22], 29.37% (w/w) [23], 37.3 – 46.5% (w/w) [24], 44.42% (w/w) [18], 69.27% (w/w) [25], 39.13% (w/w) [26], and 36.67% (w/w) [27].

Therefore, CNF from OPEFB is the potential additional raw material to develop sustainable packaging. However, limited research has focused on CNF preparation for PPNC production. Therefore, this review explains the potential PPNC reinforced with CNF from OPEFB as sustainable packaging and defines the suitability and limitations of CNF in developing sustainable packaging.

2. Production of CNF

Nanotechnology is a science focusing on production design, raw material, product characterization, and product application by controlling the size of the product at the nanoscale [28]. It is consisting of three discipline science which are mathematics, chemistry, and physics to processing the raw materials to be a product that has at least one size in nanoscale [29], control a substance at the nanometer (nm) level [30], and the range of particle size from 0.1 to 100 nm [31]. Studies about nanotechnology have been conducted on the CNF isolation from several natural sources and their application to develop added-value products [2,29,32,33]. Natural sources’ selection is pendent on the local fiber availability, chemical components, and economic viability [9]. CNF isolation from natural sources can be conducted in two steps: cellulose fiber pretreatment and CNF isolation (Figure 1) [2,34–36].
2.1. Pretreatments of Cellulose Fibers from OPEFB

Pretreatment of cellulose fibers from OPEFB is carried out after the bleaching process in the CNF isolation. Pretreatment of cellulose fibers consist of 2 types, namely mechanical treatment (include crushing and refining) [37] and chemical treatment include enzymatic hydrolysis [6], fermentation [18], acetylation [34], carboxymethylation [38], TEMPO oxidation [39], and acid hydrolysis [40].

The reasons for performing the pretreatment step of cellulose fibers before CNF isolation from cellulose fibers are [7,36,41]: Efficiency of CNF isolation is increasing, produce highly purified cellulose fibers, removing the lignin and hemicellulose, the hydrophilic surface of cellulose fibers is changed to be hydrophobic, reducing the size of cellulose fibers during isolation process to prevent clogging of CNF instruments, and decrease the energy consumption in the CNF isolation from cellulose fibers.

Many researchers are interested in the production of nanoparticles from cellulose fibers under a top-down condition, which are mechanical treatments, such as crushing, grinding, and high-pressure homogenization, and bottom-up condition, which are biological treatment, such as enzymatic hydrolysis [42,43], and chemical treatment, such as acid hydrolysis [44-46].

A major obstacle in the success of CNF commercialization is the high energy consumption during pretreatment of cellulose fibers. Therefore, researchers have been combining mechanical treatment with a chemical treatment to increase the efficiency of size reduction before homogenization so that it can help to minimize energy consumption [47].

Acid hydrolysis is performed in the presence of acid for depolymerization of cellulose fibers, and the low density of amorphous regions in native cellulose fibers will break up and releasing the individual crystallite cellulose (Figure 2) when subjected to acid treatment [47,48]. Particle size of CNF from OPEFB after acid hydrolysis around 1.0 – 3.5 nm [12], 4 – 15 nm [13], <100 nm [14], 5 – 10 nm [15], and 17.85 nm [20]. The particle size of CNF from OPEFB after enzyme hydrolysis around <70 nm [17] and 32.6 – 36.4 nm [18].

2.2. CNF Isolation

CNF isolation methods from cellulosic fibers using a variety of methods, such as mechanical and chemical methods, including high-pressure homogenization methods [49–52], microfluidization
methods [53,54], micro grinding methods [55,56], high-intensity ultrasonication methods [17,57–59], electrospinning methods [60–62], and steam explosion methods [63–65]. Each technology of the CNF isolation method has advantages and disadvantages (Table 2).

Table 2. The advantages and disadvantages of various CNF isolation methods

| Methods                        | The advantages                                       | The disadvantages                                      | References          |
|--------------------------------|-------------------------------------------------------|--------------------------------------------------------|---------------------|
| High-intensity ultrasonication | • High power output                                   | • Need treatment to dissipate of Generated heat in this method | [5,45,66–67]       |
|                                | • High efficiency of defibrillation                    | • High noise level                                     |                     |
|                                | • High effectiveness of defibrillation                 | • Need pretreatments to release CNF                    |                     |
| High-pressure homogenization   | • High clogging                                       | • Useful to laboratory scale only                      | [66,67]             |
|                                | • The method is quick, effective, and continues        | • High passing time                                    |                     |
|                                | • Can be scaled up from laboratory production to      | • High energy consumption                              |                     |
|                                |   industrial production                               | • Suspension temperature is increasing                 |                     |
| Micro fluidization             | • Less clogging                                       | • Inappropriate for industrial scale                   | [67]                |
|                                | • Uniformity in size reduction                        |                                                        |                     |
|                                | • Need fewer cycle to optimize fibers degradation      |                                                        |                     |
| Micro grinding                 | • Low energy consumption                              | • Difficulty in disk maintenance and replacement       | [66,68]             |
|                                | • Need less cycle to CNF preparation                  | • Reduction in crystallinity of CNF                    |                     |
|                                | • No need to refining pretreatment                    |                                                        |                     |
| Electrospinning                | • Complex hierarchical structures can be obtained by  | • The performance of CNF is not well researched        | [69,70]             |
|                                |   calcination controlling                             |                                                        |                     |
| Steam explosion                | • No recycling and environmental cost                 | • Incomplete destruction of lignin and fibers matrix   | [71–73]             |
|                                | • Low operational cost                                | • Partial hemicellulose solubilization                 |                     |
|                                | • Low energy input                                    | • Creates inhibitors at high temperature               |                     |

Fahma et al. [12] reported that the hydrolysis of 20 g cellulose fibers in 210 mL sulfuric (64 % w/w) under strong agitation at 45°C for 60 minutes is the best condition for CNF isolation from OPEFB. Lisdayana et al. [74] isolate the CNF from OPEFB using an ultrafine grinder under strong agitation (1500 rpm), and then it was ultrasonicated using an ultrasonicator for 30 minutes.

3. Properties of CNF from OPEFB

Property analysis of CNF from OPEFB consists of Morphology and Fractionation analysis, Fourier Transform Infrared Spectroscopy (FTIR) analysis, Degree of Crystallinity analysis. Table 3 shown the properties of CNF from OPEFB from several researches.
Table 3. Properties of CNF from OPEFB from several researches

| Properties                        | Methods                          | Results*                                                                 | Identification of Result                                                                 | References |
|----------------------------------|----------------------------------|--------------------------------------------------------------------------|----------------------------------------------------------------------------------------|------------|
| Morphology and Fractionation of CNF from OPEFB | Atomic Force Microscope (AFM) | The diameter of obtained CNF was 2.05 ± 0.89 nm and can know of the exact length of CNF | [12]                                                                                   |            |
| Transmission Electron Microscope (TEM) | Bruker Tensor 37 spectrometer | The similarity of cellulose fibers and CNF spectrum from OPEFB. This means that their chemical compositions were similar | [12]                                                                                   |            |
| Fourier Transform Infrared Spectroscopy (FTIR) | Nicolet spectrometer | No peaks at 1720 and 1509 cm\(^{-1}\) and low-intensity peak at 1267 cm\(^{-1}\). This means that it was significantly effective in reducing lignin and hemicellulose | [74]                                                                                   |            |
| Degree of Crystallinity          | Wide-Angle X-ray Diffraction (WAXD) | The CNF crystallinity index was 55.09% and then decreased to 54.22% after size reduction of cellulose fibers to CNF using acid hydrolysis | [12]                                                                                   |            |
|                                  | X-ray Diffraction (XRD)          | The CNF crystallinity index was 56.57% and then increased to 64.23% after size reducing of cellulose fibers to cellulose nanofibers using mechanical treatment | [74]                                                                                   |            |

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4. Commercial PP

PP is a thermoplastic polymer and can be produced by polymerizing the propylene molecule. Propylene is a byproduct of the cracking process and from the gasoline refining process. The new process to produce propylene has been obtained by dehydrogenation of propane [75–79]. The crystallinity level of commercial PP is ranging from between 40 and 60% [80]. PP can be categorized
as isotactic PP (PP-it) with melting point ranging from 160 to 166 °C [81], atactic PP (PP-at), and syndiotactic PP (PP-st) with a melting point of 130 °C and crystallinity of 30% [82] (Figure 3).

PP is now the third-largest consumed plastic material after polyethylene (PE) and polyvinyl chloride (PVC). The Primary Advantages of PP are low density, excellent reproduction of mold surface, good sound-deadening properties, versatile (many ways processing), easily colored, easily modified, good water vapor barrier, excellent chemical resistance, and capable of being recycled. Commercial PP has an isotactic index between 85 and 95% with several useful properties such as transparency, high heat distortion temperature, dimension stability, flame resistance, and very suitable for reinforcing, filling, and blending. An isotactic structure leads to a semicrystalline polymer, where the higher isotacticy (isotactic fraction), than greater the semi-crystallinity, softening point, rigidity, e-modulus, and hardness [83]. PP from natural material fibers is one of the most sources to create the polymer composites [84].

Figure 3. Subdividing of polypropylene [85] (Reprinted from ref. 85. Copyright 2020 LibreTexts)

5. Potential Preparation Method to Production of PPNC Reinforced with CNF from OPEFB

5.1. Definition of PPNC

Nanocomposite (NC) typically are particle filled polymers product that was produced from raw materials that have at least one size of the dispersed particles is in the nanoscale (less than 100 nm) [86]. Sandri et al. [87] has defined NC as multi-component materials that comprised multiple different phases in which at least one size of phase is in nanoscale. PP is one of the most widely used packaged materials for industrial packaging because of its good processability, high breakdown strength, and low dissipation [88]. Industrial packaging development from PP based nanomaterials can benefit to properties enhancement of packaging. PP is one of the most important polymers because of its recyclability, design flexibility, low density, hydrophobic, and low production cost, making it a popular preference as a matrix of composites and compatibility when reinforced with polar surfaces, such as cellulose [89]. PPNC can be defined as a product that is formed by infusing nanomaterials into the PP matrix. These PPNC have a great opportunity in industry application because of their remarkable enhancements in material properties compared to the natural polymers.
5.2. Preparation Methods for PPNC Production from CNF

PP is a polymer that potential to be used in industrial applications because of its ease of production, lightweight, and often ductile nature. However, PP has disadvantages, such as low strength and low modulus. Fibers or whiskers can be used as reinforcements materials to the PP matrix to improve PP’s mechanical properties to increase the heat and impact resistance, flame retardancy, mechanical strength, decrease electrical conductivity, and gas permeability to oxygen and water vapor [90]. Numerous studies have researched PPNC production from CNF (Table 4).

Table 4. Research about PPNC production from CNF

| No | Preparation Methods          | Properties                                                                 | References |
|----|------------------------------|---------------------------------------------------------------------------|------------|
| 1  | Six twin-screw elements      | • The elastic modulus of PPNC was increased by the addition of only 1% CNW | [91]       |
| 2  | Solvent casting              | • Good film transparency                                                  | [92]       |
|    |                              | • The slight agglomeration of CNW in the PPNC film                        |            |
|    |                              | • The PPNC tensile strength was increasing 70–80% if compared with neat PP |            |
|    |                              | • The crystallinity was increasing by 50%                                 |            |
|    |                              | • Increased content of CNW because of the higher hydrophilicity, higher thermal degradation temperature, and higher thermal conductivity |            |
| 3  | Twin-screw extrusion         | • The PPCV crystallinity was lower than neat PP                           | [93]       |
|    |                              | • Mechanical, thermal properties above the glass transition temperature of PP |            |
|    |                              | • Water absorption capability was weakened                                 |            |
| 4  | Solvent casting              | • The microscopy results showed a substantial rise in the magnitudes of key rheological parameters of PPNC | [94]       |
|    |                              | • Steady-shear results revealed a strong shear thinning behavior of PPNC  |            |
|    |                              | • PPNC exhibited yield stress.                                             |            |
|    |                              | • Considerable improvement in the modulus of PPNC                         |            |
| 5  | Twin-screw-co-rotating extruder | • The CNF addition increased the tensile modulus (36%), tensile strength (11%), flexural modulus (21%), flexural strength (7%), impact strength (23%) if compared to those of neat PP | [95]       |
| 6  | Solvent casting              | • The crystallization rate of PPNC is faster than neat PP                  | [96]       |
|    |                              | • The activation energy, the equilibrium melting point, and the initial lamellae thickness during isothermal crystallization of PPNC was higher than neat PP |            |
| 7  | Twin-screw extrusion         | • The CNC addition was increasing the tensile strength (1–14%), tensile modulus (15–22%), flexural modulus (13–26%) and decreasing the elongation at break (50–96%), impact strength (10–20%) if compared with neat PP | [97]       |
| 8  | Melt-extrusion               | • The tensile property and the thermal stability of the PPNC with MAPP grafted CNC was higher than that of pristine and TDI grafted CNC systems | [98]       |
|    |                              | • A melt extrusion process with pre dispersion processing exhibited more positive effects on the properties of the PPNC in comparison to the systems without pre dispersion |            |
6. Conclusion

The use of thermoplastic such as PP in packaging has raised concern about environmental impact, so research needed to identify alternative sustainable packaging materials to reduce the environmental impact. PP is one of the most important polymers because of its recyclability, design flexibility, low density, hydrophobic, and low production cost, making it a popular preference as a matrix of composites and compatibility when reinforced with polar surfaces, such as cellulose. CNF has considered a sustainable packaging material due to its low weight, high strength, high abundance, rigidity, and biodegradability. Natural material potential as raw material to CNF isolation is OPEFB has generated from oil palm extraction process. CNF can be used as additional raw material to reinforce the PP matrix, which is called a PPNC. PPNC reinforced with CNF from OPEFB is a potential product as sustainable packaging.

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