REVIEW PAPER

Biodegradable nanocellulose reinforced biocomposites for food packaging – a narrative review and future perspective

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ABSTRACT

Conventional food packaging materials such as plastic or other petro-based polymers provide the necessary strength and barrier qualities, but they are associated with economic and environmental associated hazards. To have a material that can effectively compete with petroleum-based plastics, the eco-friendly biodegradable biopolymer-reinforced bio-nanocomposites from renewable agricultural biomass can be a promising alternative. Lignocellulosic biomass from agricultural wastes can serve as promising raw materials for nano cellulose extraction. This review focuses on extensive studies done on the pretreatment, and isolation of nanocellulose along with their role as reinforcement agents in various biopolymers such as poly-lactic acid, starch, chitosan, alginate, and carrageenan. The uniqueness of this review is to give readers a thorough overview of the impact of nanocellulose on the mechanical, barrier, and thermal properties of the biopolymeric film. The review will also provide a comprehensive understanding of the supply chain's elements, their opportunity, and constraints for shifting the sustainable production system of cellulose nanocomposites towards a circular economy. Efforts have also been made to combine all the recent studies related to key aspects and engineering of nanocellulose biocomposites. The concept of 3D/4D printing technology of cellulose nanocomposites in conjunction with Shape Memory Polymers (SMP) in the domain of commercial development of smart materials and self- folded packages for food applications has also been discussed.

Keywords: Biodegradability, Food packaging, Nanocellulose, Nanocomposites, 3D/4D printing.

INTRODUCTION

In the modern era, petroleum-based plastic has been used vigorously due to its versatility, convenience, stability, and durability. These plastics, however, disrupt the ecosystem because of their non-biodegradability and environmental impact. Plastic processing being an energy-intensive operation results in significantly higher greenhouse gas emissions (GHGs) that increase the threat of global warming, worsens the quality of the air, and raises several health issues. Apart from air quality issues, plastic waste generation has become a serious issue in the last few decades due to its resistance to decomposition. The United Nations Environment Programme (UNEP) has reported that 7 billion tonnes of plastic waste are generated annually in the whole world and only 10% gets

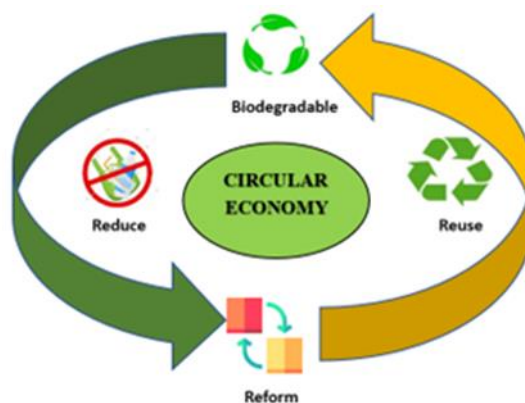* Corresponding Authors Email: *sadhanajadon10@gmail.com*

Fig. 1. Systematic use of ''Circular economy'' to reduce plastic waste and motivate utilization of biodegradable polymers from renewable resources.

recycled. The remaining is mismanaged and ends up in the land, ocean, and river where they persist for long centuries and cause environmental pollution. If it is not improved, then plastic waste of 13.2 billion tons will reach the environment and landfills by 2050[1]. Managing and disposing of plastic trash is a significant challenge to the quality of goods created from recycled plastics and also the recyclability of post-consumer plastics. The concept of a "circular plastic economy" motivates the utilization of renewable resources and minimizes waste production as depicted in Fig. 1. As per this model, eco-friendly bio-based polymers such as PLA (Polylactic acid), bio PBS (Biodegradable Polybutylene succinate), PCL (Polycaprolactone), PHA (Polyhydroxyalkanoates), PBSA (Polybutylene succinate-co-butylene adipate), PMAs (Poly (methyl methacrylate)) and Starch, cellulose can replace consumer plastics such as PS (Polystyrene), PET (Polyethylene terephthalate), PVC (Polyvinyl chloride), PE (polyethylene), PP(Polypropylene) in the food packaging industry [2]. The implementation of this model will help in achieving sustainable goals with the utilization of agro-waste (non-fossil) resources, new recycling techniques, and with less toxic chemicals and solvents. The production of Bio-based films from agro-food waste is natural and remarkably low-cost carbon and 100 % biodegradable. Thus, this model holds considerable advantages in reducing carbon footprints and maintaining environmental sustainability.

Food packaging films should satisfy basic requirements, including legislation, security, and a variety of other requirements, as well as effectiveness. It must be inventive, simple to use, odorless, and good in appearance. The main objective of food packaging film is to protect against microbial contamination, moisture loss, physical damage, and chemical changes. Besides physical protection, a suitable packaging film should be effective in maintaining food stability, quality, and safety during storage. Conventional materials such as plastic, aluminum, tin, and petro-based polymers provide the necessary strength and barrier qualities, but they are associated with economic and environmental associated hazards. To effectively compete with petroleum-based plastics and to have an alternative to them, there has been an increase in interest in using renewable agricultural biomass to manufacture biodegradable bioplastics. Biodegradable plastics could serve as a feasible solution for overburdened landfills while also helping the recycling of non-renewable materials. Biobased polymers are renewable materials that undergo degradation at a faster rate in the natural environment due to different micro-organisms, moisture, and ultraviolet (UV) sunlight without releasing any toxic gases into the environment. Biopolymers such as PLA, PCL, PHA, and PBS are gaining much attention as they are derived from renewable plant-derived material. The modification of the molecule features viz., the weight of the molecule, sequence of the monomer distribution, and crystallinity can regulate the rate at which these biopolymers disintegrate. Urbanek et al. [3] studied the biodegradability of PCL, PBS, and PBSA by antarctic microorganisms at low temperatures. Biodegradation activity was found to be 49.68% for PBSA and 33.7% for PCL, 45.99% for PBSA, and 49.65% for PCL at 20°C in the presence of *Sclerotinia* sp. B11IV and *Fusarium* sp. B3′M

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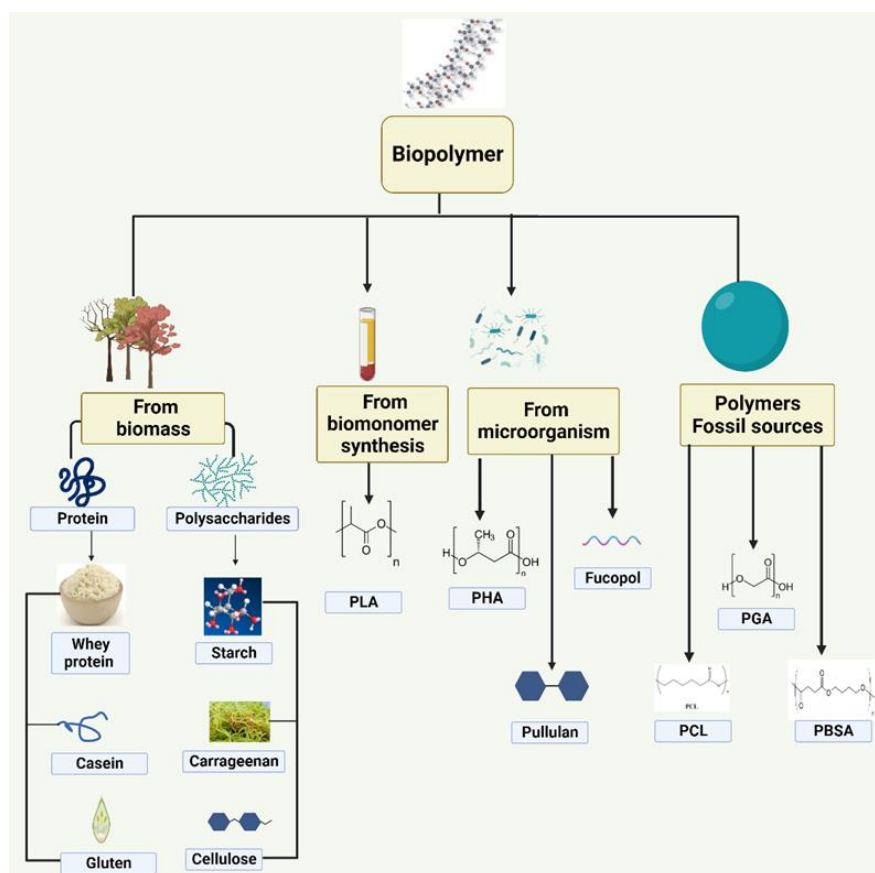*J. Water Environ. Nanotechnol., 8(3):293-319 Summer 2023*

Fig. 2. Various biopolymers derived from different renewable and non-renewable sources. PLA = polylactic acid; PHA = polyhydroxyalkanoates; PCL = polycaprolactone; PGA = polyglutamic acid; PBSA = poly butylene succinate-co-butylene adipate.

strains. Cho et al. [4] evaluated the biodegradation behavior of PHB by marine *Bacillus* sp. JY14 under different environments and saline conditions. The result showed approx. 98% degradation after 14 days.

With the rise of the packaging industry, biopolymer-based packaging is being considered as an alternative to plastic packaging to reduce the volume of waste. Biopolymers get easily broken down through the action of enzymatic action of microbes. Bio-based polymers for food packaging include polysaccharides derived from biomass, such as starch, pectin, cellulose, and protein (casein and gluten), as well as chemically synthesized biopolymers derived from renewable monomers, such as polylactic acid; and polymers derived from microorganisms, like polyhydroxyalkanoates and pullulan [5]. Some of the biopolymers derived from different renewable and non-renewable sources are illustrated in Fig. 2. The polymeric-based films/ coatings from these biopolymers are finding

application in food packaging to protect food quality and shelf life by incorporating antibacterial, antioxidant, natural ingredients, and colors [6-8]. Biopolymers can be utilized to package food products effectively, however, because of their low barrier and mechanical properties, the usage of bio-based materials for food packaging is currently fairly restricted [9-11]. Blending biopolymers with other biodegradable polymers is viewed as an efficient strategy to boost the mechanical and gas barrier qualities because of molecular entanglement, which may be used to change the characteristics of biopolymers and also to minimize overall cost [12]. The purpose of blending could be to improve or modify qualities to a specific application and to maximize a composite performance[13-14].

Nanotechnology has garnered considerable attention for establishing cellulose-based food packaging with better barrier qualities [15]. The introduction of nano-reinforcing agents into biopolymers has turned out to be a suitable
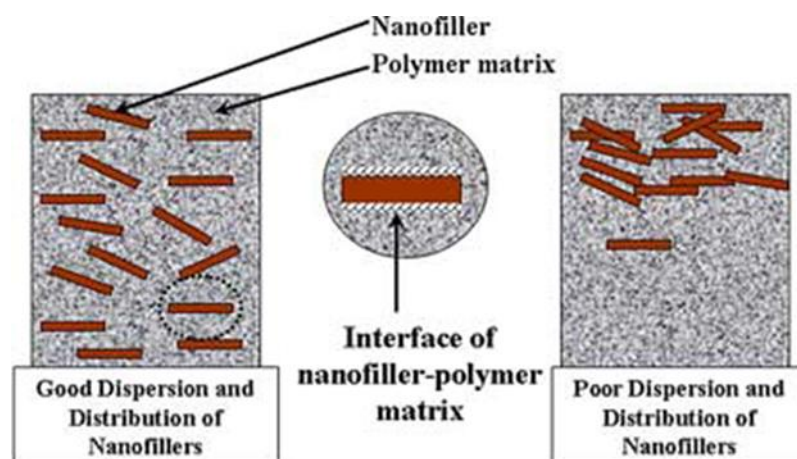
Fig. 3: Nanocellulose – matrix dispersion (Reproduced from: [23])

approach for the enhancement of mechanical strength, thermal decomposition behavior, and water vapor barrier property enabling their usage in commercial applications, as shown in Table 1. Infusion of nanoparticles in biopolymers into bionanocomposites, on the other hand, is a promising technique to improve material properties while ensuring biodegradability [16]. While some bio nanocomposites have been commercialized, many more are still in the research phase. Abral et al. [17] reported that the addition of bacterial cellulose (BC) nanofibers in tapioca starch/chitosan-based films resulted in improved tensile strength thermal resistance, moisture resistance, and water vapor barrier properties. Similar improvement in mechanical and barrier properties by utilizing nanocellulose from varied sources has been reported [18-19]. It was reported by Mulla et al. [20] that the properties of PLA biopolymer got improved by adding nanoparticles as reinforcement agents. Rizal et al. [21] reported improvement in physical, mechanical, and degradation properties of PLA/chitosan-based nanocomposites reinforced with CNF (Cellulose nanofibers) isolated from *Schizostachyum brachycladum* bamboo. Apart from this, thermal properties were also enhanced with CNF addition without affecting mechanical strength. Problems of high-water sensitivity, low barrier, and mechanical properties that were encountered with chitosan-based films got overcome by reinforcement with nanoparticles [12, 22].

The main aspects to consider during nanocomposite production are filler-to-matrix interfacial adhesion, reinforcement loading, and dispersion of nanocellulose filler in the polymer matrix. As shown in Fig. 3, if the bonds between the nanocellulose filler and matrix materials get weakened due to their poor compatibility, it leads to poor dispersion and distribution of nanofillers. This results in reduced nanofiller efficacy and nanocomposite [23]. However, high surface area, nanosize range, good stiffness, aspect ratio, and uniform distribution of nanocellulose inside a polymer matrix alter the mobility of the polymer chain and thus strengthen the characteristics of the resulting nanocomposite. Furthermore, the interactions formed between the polymeric and nanostructures reduce the spaces in the polymeric chains that can engage with water molecules, enhancing the moisture barrier properties of the nanocomposites.

In the present review, a comprehensive outline of the nanocellulose preparations, their interactions with different nanocomposites, and the environmentally friendly applications of nanocellulose-based nanocomposites have been presented. The uniqueness of this review is to provide readers with an in-depth look at the most recent research on nanocellulose reinforcement in polymeric matrices and its impact on the properties of nanocomposites-based food packaging. Finally, the status, perspectives, advantages of cellulose nanocomposites, and application of 3D/4D printing in the creation of environmentally friendly smart food packaging material have also been presented. The information provided in the review will help conduct further research in the

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Table 1: Comparison of nanocellulose-based food packing materials with as starch, pullulan and PVA

Nanocellulose based biopolymer biocomposites	Biopolymer matrix and its properties	Nanocellulose source	Method	Key Findings	Reference
Cellulose Nanofibrils based Starch-Hibiscus-Composite Film	Starch- Higher availability, good compatibility with antimicrobial materials and better gas barrier property	Coffee husks	Solvent-casting method	Presence of TEMPO-oxidized CNF enhanced the mechanical properties and water vapor permeability	[24]
BC nanofiber reinforced Starch/chitosan film	Chitosan-antimicrobial activity, good compatibility with other biopolymers, considerable mechanical strength	Wet pellicle	Solvent-casting method	Incorporation of BC nanofibers in starch/chitosan-based edible coatings improved their moisture resistance and water barrier properties	[17]
Dialdehyde nanocellulose based PVA composite film	PVA- Good film forming properties, high physical properties and excellent transparency	Industrial	Solvent-casting method	Improvement in mechanical, thermal and hydrophobic properties due to the presence of chemical network structure	[25]
NC reinforced Polyvinyl alcohol film		Teff straw	Solvent-casting method	Inclusion of nanocellulose content less than (<7%) improved its mechanical properties and moisture barrier properties and did not affect transparency	[26]
Pullulan/carbon nanotubes-CNF Based Nanobiocomposites	Pullulan-Good tensile strength, better barrier properties but lower elongation at break	Industrial	Solution casting method	Improved tensile strength, water barrier properties, reduced moisture susceptibility and biodegradability	[27]
Pullulan/ bacterial cellulose and ferulic acid-based composite		Industrial	Solution casting method	Incorporation of 2% BC improved mechanical and barrier properties	[28]

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field of nanocellulose biopolymers to address various engineering and environmental problems associated with their bulk production and potential commercial use in food packaging.

CELLULOSE AS A BIOPOLYMER FOR BIODEGRADABLE FILMS

Cellulose is the most abundantly found biopolymer and a major component of agricultural biomass. It is present as a predominant constituent in the cell wall of plants but is also possessed by some bacteria. Cellulose has been considered to be a suitable biopolymer for biodegradable films due to specific properties such as recyclability, renewability, harmlessness, process ability, cost-effectiveness, and mechanical properties [29]. D-glucopyranose monomers constitute the basic units of cellulose polymer chains. Two units of anhydroglucose combine to produce an anhydrocellobiose structure, which is connected by β-(1-4) glycosidic linkages [30]. The poor solubility of cellulose in water is attributed to an extensive network of cellulose molecular chains and strong H- bonding between the OH groups of adjacent cellulose chains. The -OH groups and oxygen atoms of the pyranose ring and the glycosidic bond can be used to create ordered hydrogen bond networks [31]. Hydrogen bond networks are thought to be a principal factor in deciding their solubility, stiffness, crystallinity, and chemical stability. Intermolecular interactions give the polymer chain rigidity, while intramolecular bonds offer it structures [32-33].

Cellulose biocomposites are green and sustainable materials prepared by the incorporation of natural fibers in the polymer matrix to enhance the mechanical properties of the composite [34]. They are considered alternatives to synthetic fibers and can be developed in a similar way to compression molding, resin transfer molding, extrusion, and hand lay-up. Their physical properties can be enhanced by surface modification, coating, chemical treatment, and also by hybridization techniques. By integrating antibacterial compounds in cellulose and its esters, antibacterial activity can be introduced to cellulosic composites to improve food preservation [35-37]. However, nanocellulose-based composites are emerging materials to overcome critical drawbacks such as lower mechanical properties, higher moisture permeability, incompatibility, and poor thermal properties.

Cellulose nanomaterials

Nanocellulose is a nanosized structure and can primarily be divided into two categories:

(i) Cellulose Nanofibers: Nanofibrillated cellulose (NFC), microfibrillated cellulose (MFC), and cellulose nanofiber (CNF) are other acronyms for cellulose nanofibrils, which have a diameter of 5 to 50 nm and a length of up to several micrometers. The attributes of CNF such as size, shape, surface charges, crystallinity, and stiffness are all determined by the raw material type, the isolation method, and the pretreatment processes adopted [38]. It is observed as a semi-flexible network entirely made up of crystalline and amorphous nanofibril agglomerates and individual fibers. It is mechanically produced through the disintegration and defibrillation of cellulose fiber and resulting in nanosized fibers. Pretreatment is required before mechanical treatment to improve nanofiber efficiency and lower energy consumption costs. Unlike cellulose nanocrystals (CNCs), CNF requires fewer chemicals and energy during the pretreatment step, rendering CNF a cost-effective and sustainable reinforcement agent [39]. High aspect ratio, low density, easy-to-modify surface, and web-like structure make the CNF a potential reinforcement agent contributing to the high strength and stiffness of the nanocomposites. About H-bonding connecting each fiber, cellulose fibers can self-associate through interconnections due to their flexible structure, which confers a further reinforcing effect in nanocellulose-reinforced nanocomposite film [31, 40].

(ii) Cellulose nanocrystals: The elongated rod-shaped CNCs, commonly referred to as cellulose whiskers, are made via acid hydrolysis by dissolving amorphous components and possess a much more crystalline region than CNF. The length of CNC ranges from 100–250 nm and the width from 3–50 nm depending upon the source of cellulose and hydrolysis condition. Since cellulose nanoparticles are nanoscale in size, they have a great extent of surface hydroxyl groups that can be modified to give any desired surface functionality. Due to its remarkable qualities, numerous industries including packaging have started producing and commercializing CNC for commercial applications. The amorphous portions of cellulose materials

are swiftly digested, releasing pristine crystalline areas. To isolate CNCs, strong acids such as hydrochloric and sulphuric acids are generally employed, disrupting the glycosidic bond and causing dissolution of amorphous parts. Sulfuric acid is usually used to make CNC because it causes negatively charged sulfate groups to develop on the surface. This may influence the thermal stability of CNCs which can be compensated by sodium hydroxide (NaOH) neutralization [41-43].

NANOCELLULOSE FROM AGRO-INDUSTRIAL WASTE/NATURAL FIBERS

The wide availability of lignocellulosic biomass and the need for promising alternatives to plastic have created opportunities for innovations in the domain of biodegradable packaging. The use of lignocellulosic biomass from agricultural wastes for nanocellulose extraction is a sustainable strategy since it reduces environmental disposal issues while also providing waste management benefits. Lignocellulosic biomass is a great source of cellulose and found complexes with undesired compounds such as lignin and hemicellulose [44]. The available sources of lignocellulosic materials for nanocellulose isolation are sugarcane bagasse, corn, wheat, barley, soybean straw, and rice straw. Farmers burn rice straw in the paddy field after harvesting, which is a significant agro-industrial waste. When used as a fuel source, it produces significant amounts of ash and other toxic materials, which are released into the atmosphere. Alternatively, rice straw is thus an economical, promising, and potentially good source for nanocellulose and biocomposite production [45-47]. Lignocellulosic biomass is processed in two steps to produce nanocellulose. Firstly, pretreatment is required to recover cellulose from complex hetero-matrix structures. This is accomplished by separating lignin and hemicellulosic parts without causing any destruction to cellulose fibril. Secondly, the pretreated cellulose fibrils are subjected to chemical and mechanical treatment to obtain nanocrystals or nanofibers [48].

Natural fibers have garnered a lot of attention in recent years because of their low cost and less pollution from production, thereby minimum health risks and sustainability. The widespread availability of natural fibers from underutilized agricultural biomass is an additional advantage. Natural fibers mostly consist of cellulose, hemicellulose, and lignin, with lower amounts of pectin, extractives, and pigments. Cellulose, being the most prominent constituent bounded by matrix including lignin and hemicellulose. Owing to inter- and intramolecular H-bonding within the hydroxyl groups of the nearby cellulose chains, cellulose is insoluble in water, creating a hydrophobic situation. Hence, the substantial intermolecular hydrogen bonds and Van der Waals force present in the crystalline region of microfibrils of cellulose present a hurdle for hydrolysis and nanocellulose production. In the process of extracting nanocellulose form biomass, the material is subjected to pretreatment. However, before the pretreatment step, biomass is ground to reduce its size and contact surface, thereby increasing the effect of pretreatments so that the reagents can interact with the plant wall and enhance the accessibility of cellulose fiber to chemical attack before nanocellulose isolation. The efficiency of the primary treatment determines the quality of the natural fibers, which can have a significant impact on the qualities of the resulting composites. A newly developing fibers extraction technique known as a retting process involves the separation of fiber bundles from plant stems leading to maximum fiber loosening from woody tissue. It can naturally occur in water bodies or be induced by a mechanical, enzymatic, or chemical process. Among them, microbiological and enzymatic retting is an extensively used procedure for obtaining greater quality extraction from flex, hemp, and jute fibers. Fiber quality is greatly influenced by the disintegration of pectic components which results in fiber separation. The various steps involved in nanocellulose extraction are discussed below.

Pretreatment

The pre-treatment process, being an important initial step is carried out in raw material before nanocellulose production to enhance the efficiency of the extraction process by removing undesired substances like waxes, lignin, hemicellulose, and ashes. Additionally, the pretreatment process aids in the optimum transformation of cellulose fiber into micro/nanofibrils by enhancing fibrillation [49-50]. Pretreatment involves fiber cleaning, extractive removal, acid treatment, alkalization, and bleaching. The sequence of the pretreatment process applied is determined by the fiber type. For instance, since cotton linters contain fewer extractives than sisal leaves, substantial washing may not be necessary in that case [34]. Pretreatment

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breaks down cross-linkages between biomass components by modifying the lignin's structure and partially depolymerizing the hemicellulose. The crystalline features of the pretreated cellulose fibers have been discovered to be severely reduced by the presence of lignin and hemicellulose, which further affects its mechanical properties. Hence, the separation of hemicellulose and lignin is essential to produce cellulose in its purest form, which has higher properties. Pretreatment methods include chemical pretreatment, physical pretreatment, enzymatic pretreatment, oxidation methods, and a combined approach.

Chemical pretreatment

Acid pretreatment: Acid hydrolysis is a suitable strategy since it hydrolyses and releases the amorphous component of the cellulose structure. The sulphuric acid-based acid treatment is a widely used pretreatment that effectively lowers biomass intransigence by hemicellulose elimination, cellulose crystallinity modification, and increment in biomass porosity through lignin redistribution. The duration and temperature of the hydrolysis reaction, as well as the acid content, are crucial factors that affect the morphology and size of the resulting fibers [51]. It's vital to have the ideal acid content because a larger quantity might harm the instrument, and structure and may cause corrosion of the reaction vessel.

Alkaline pretreatment: Sodium hydroxide, calcium hydroxide, and potassium hydroxide are used in alkaline pretreatment to get access to biomass structure, particularly by dissolving a considerable amount of lignin in the alkaline medium [52]. Alkali treatment leads to disruption of the structural linkage between hemicellulose and lignin by saponification of the intermolecular ester bond and removal of the acetyl groups and uronic acid substrates from the hemicellulose during the solvation process [53]. Following the lignin solubilization stage, lignin will no longer serve as a barrier to protect the cellulose as disruption of the lignin structure occurs due breakage of its glycosidic ether bond. This helps to disperse cellulose fibers, which causes cellulose to swell and eventually affect shape, size, thermal characteristics, and mechanical qualities. Thereafter, cellulose become more accessible to subsequent downstream processes, disclosing the structure of cellulose.

Organosolv pretreatment: An emerging pretreatment process called organosolv pretreatment uses organic solvents like methanol and ethanol to simultaneously degrade lignin and hemicellulose from lignocellulosic biomass. Addition of catalysts such as mineral acids (hydrochloric acid, sulfuric acid, phosphoric acid), and bases (lime, sodium hydroxide, ammonia) is advantageous in enhancing the delignification process and also lowering the temperature [54]. Lower boiling point and high volatility of alcohol are preferable due to easy recovery via simple distillation. The solvents may be easily recycled and reused, which is a benefit of this pre-treatment technique. The intermolecular ester bonds between lignin and hemicellulose are broken by the saponification reaction, which is initiated by the OH ion produced by the alcohol solvent.

Ionic liquid pretreatment: Ionic liquids, a green alternative to volatile organic solvents consists of small inorganic anion and large organic cation. The ionic liquid can effectively dissolve biomass or selectively remove lignin and hemicellulose based on the specific combination of anion and cations. A good ionic liquid should be non-toxic, non-volatile, strong chemically stable, low viscous, low/no vapor pressure, and have a high dissolve power due to the tunable nature of cation and anion. Imidazolium-based ($[(C_3N_2)X_n]^+$), ammonium-based $[NX_4]^+$, pyrrolidinium-based ($[C_4N)X_n]^+$, pyridinium based ($[(C_5N)X_n]^+$), phosphonium-based $[PX_4]^+$, sulfonium-based $[SO_3]^+$, and, choline are the different types of Ionic liquids being utilized [55]. Ionic liquids are considered to be beneficial for maximum cellulose dissolution at minimal cost as they consist of aromatic imidazolium or pyridinium cations and OAc^-, $HCOO^-$, $(MeO)_2PO_2^-$ or Cl^- anions.

Physical Pretreatment

Physical treatments include such as grinding, ball milling, two-roll milling, hammer milling, and the use of vibro-energy. The shear forces produced during milling allow for efficient reduction of particle size and cellulose crystallinity. The degree of polymerization and cellulose crystallinity is significantly influenced by the milling time and technique. Vibratory ball milling is the most effective milling technique for increasing biomass digestibility. Even though these techniques are favorable because they act effectively and emit little harmful material, a major challenge that must be addressed for the successful commercialization of nanocellulose is their high energy consumption.

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Enzymatic Pretreatment

Enzyme utilization in biological treatment is the most effective technique since it provides numerous benefits such as process control, environmental friendliness, and selectivity, making it one of the most suited approaches that are most frequently used to overcome financial constraints. Complex lignocellulosic biomass is typically digested using enzymes like cellulases and xylanases. They precisely solubilize the fiber and prevent excessive cellulose chain breakdown into glucose. Subunits of cellulase enzyme complex (endocellulases, exocellulases, β-glucosidases, and cellulose phosphorylases) work synergistically for the effective hydrolytic degradation of β (1→4) linkages. Firstly, endoglucanase produces non-reducing oligosaccharide segments by randomly cleaving internal bonds of the cellulose chains, which favors fibrillation. It is further acted upon by cellobiohydrolase (exoglucanase) releasing cellobiose, and then individual monosaccharide units are produced from non-reducing ends of the cello oligosaccharides by the action of ß-glucosidase [56]. Besides that, phenol oxidase (laccase) (Lac) and peroxidases, such as Lignin peroxidase (LiP), versatile peroxidase (VP), and manganese peroxidase (MnP), are the categories of ligninolytic enzymes that possess a substantial role in the enzymatic degradation of lignin [57].

TEMPO-mediated oxidation

Additionally, TEMPO-oxidation is a common chemical pretreatment method performed under mild conditions via the oxidative process. TEMPO (2, 2, 6, 6-tetramethyl-piperidine-1-oxyl) which is a water-soluble radical carry out its reaction by utilizing sodium bromide (NaBr) as the efficient catalyst and sodium hypochlorite (NaClO) as the main oxidant component. In a relatively aqueous environment, (TEMPO) radicals can oxidize cellulose, converting its hydroxyl groups into a carboxylic acid, before mechanical treatment by considerably avoiding the drawback of spending more energy to do so [58]. A high aspect ratio is possessed by cellulose nanofibers obtained via TEMPO radical oxidation leading to a plethora of possibilities such as imparting effective barrier properties in composite film.

Nanocellulose isolation

The pretreated biomass can be treated differently to produce either cellulose nanocrystals or cellulose nanofibers.

Acid hydrolysis

Acid hydrolysis, a commonly used method for CNC isolation utilizes strong acids, especially sulphuric acid to hydrolyze amorphous parts and to also maintain high crystallinity. The process of esterification between the -OH group on the surface of cellulose and the anionic sulfate ester groups results in a negative charge on the surface of the nanocrystals, which makes them easier to disperse in water. In this process, the acid targets the amorphous areas of the fibers by penetrating the cellulose chains with hydronium ions, which causes the hydrolysis of glycosidic bonds and finally results in the release of crystalline sections [59]. It creates rod-shaped CNC with high crystallinity (54–88%), and the characteristics of the CNC change according to the type, concentration, temperature, and time of the acid [58].

Mechanical treatment

For the production of CNF, mechanical treatments consisting of high-intensity ultra-sonication, high-pressure homogenization, micro-fluidization, micro-grinding, electrospinning, cryo-crushing, ball milling, etc. are employed [60]. Mechanical treatments are effective chemical-free methods that result in fiber delamination and the separation of nanometric fibers. The limitations of this treatment include high energy consumption and equipment obstruction.

Ultrasonication: Ultrasonication is a mechanical method that defibrillates pretreated cellulose fibers using ultrasonic hydrodynamic forces to produce cellulose nanofibers [61] Concentration of cellulose, process time and ultrasonication power are important parameters that influence the ultrasonication process [62]. Ultrasonication can also be successfully combined with a high-pressure homogenization process to enhance the nanofiber isolation rate. Dilamian et al. [63] extracted cellulose nanofibers through combined ultrasonication - high-pressure homogenization from chemically pre-treated cellulose fibers and obtained cellulose nanofibers with 6-20 nm diameter and 177 high aspect ratios.

High-pressure homogenization: This process involves passing cellulose pulp from a nozzle at high pressure, high speed with impact and shear force. The hydrogen bond network of cellulose is damaged through the application of these forces. These

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forces allow the fluid to produce a shearing effect in streams, which in turn leads to the formation of nanosized fibers. Wang et al. [64] applied high hydrostatic pressure (HHP) for the extraction of NC from pre-treated wood and bamboo waste. The obtained nanocellulose had good stability and high crystallinity, which demonstrated its potential for the reduction of environmental pollution.

Microfluidisation: A microfluidizer is made up of an intensifier pump that accelerates the suspension of the fibers through an interaction chamber that generates shear forces against colliding streams to cause fiber defibrillation. It's interesting to note that droplet coalescence caused by repeated cycles (more than two) of micro fluidization or pressure exceeding 124 MPa increases the average particle size of cellulose nanofibers [65]. Microfluidisation is a traditional mechanical technique that shows similarity with high-pressure processing but produces finer particle size distribution and more crystallinity than high-pressure homogenization [66].

Electrospinning: Electrospinning is a facile and efficient method that uses electrostatic forces to produce nanoscale polymer fibers with good structural stability and a large surface area. Surface tension, solution viscosity, voltage, spinneret-collector distance, temperature, dampness, and electrical conductivity, all affecting the development and size of nanofibers [67].

Micro grinding: In the micro-grinding process, fibrillation takes place in which H-bonds are ruptured and the cell wall is disintegrated under the influence of shear forces. This leads to the isolation of nanosized fibers [68]. Excessive energy consumption, an uneven distribution of nanocellulose size, overheating of raw materials, a negative impact on cellulose crystallinity, and grinder clogging are some of the limitations. Clogging, however, can be prevented by adjusting the stone spacing of the micro-grinder [69].

Cryocrushing: Cryocrushing is an alternative mechanical method in which prepared cellulose fibers are dipped in liquid nitrogen, which freezes the water content in the fibers. It is then treated to high-impact crushing with a mortar and pestle while submerged in liquid nitrogen. Ice crystals apply pressure to the cellulose fiber cell wall under high-impact forces, breaking microfibrils off the fiber walls and converting them to nanocellulose. The disadvantages of this technique are its lower recovery rate, high production cost, more

power consumption, and low homogeneity of nanocellulose. This method is rarely utilized for commercial purposes because it is ineffective at producing fine CNFs [70].

Ball milling: The characteristics of CNFs obtained in the ball milling process are determined by the ratio of ball and cellulose mass, grinding time, size of balls, and also by the number of balls present in a container. Maintaining the uniformity of the CNF, as well as high power consumption and generation, are some of the challenges associated with this process. Zhang et al. [71] investigated the considerable effect of ball milling on softwood pulp for CNF preparation. The diameter of cerium-doped zirconia balls in the range of 0.4-0.6mm range was found to be effective for a sufficient yield of CNFs without alkaline treatment. The optimum quantity of CNF was obtained when the mass ratio of ball to cellulose was between 60:1 and 80:1.

NANOCELLULOSE-REINFORCED NANOCOMPOSITES

Nanocellulose – Polylactic acid

Lactic acid serves as the monomer for polylactic acid (PLA), linear thermoplastic polyester. Lactic acid is synthesized from renewable sources via the fermentation pathway. Due to several benefits like processability, biocompatibility, transparency, flexibility, and low energy consumption during manufacturing, polylactic acid is currently the most broadly explored biopolymer in several food contact materials. It is biodegradable that is obtained via biotechnological processes from raw materials such as corn, rice, sugarcane, and potatoes. The renewability of PLA assists in minimizing the environmental effects associated with the widespread use of conventional polymers generated from non-renewable fuel sources [72]. The water vapor permeability and fragility of PLA restrict its performance in diverse applications, despite its intriguing mechanical characteristics. In this case, incorporating nanofiller into the polymeric matrix, such as nanocellulose could have been a feasible alternative. Enhancement in the mechanical, rheological, heat-resistant, crystallization, and biodegradability of PLA composites has been ascribed to the addition of nanocelluloses [73].

The nanocomposite (NCs) synthesis methods have been stated to play a central role in improving the PLA membrane performance (Table 2). The idea underlying reinforcement is that since nanoparticles possess high surface area and high

Table 2. Nanocellulose based PLA bio-nanocomposites

Type & source of nanocellulose	Isolation method of NCs	Load concentration of nano-cellulose (%)	Method of preparation	Improved properties	Reference
CNC (Microcrystalline cellulose powder)	Acid hydrolysis	5	Casting	Thermal stability	[76]
CNF (Linter dissolving pulp)	Ultrafine grinder	5	Twin screw extruder	Tensile strength and Young's modulus	[77]
CNC	Acid hydrolysis	0.5	Casting	Tensile strength, elongation at break	[78]
CNF (alkylation)	Ultrasonication	1.5	Casting	Load dispersions	[79]
BNC (*Komagateibacter xylinus*), CNF (softwood)	Cryogrinding	1.5	Casting	Water vapor transmission rate	[80]
CNF (wheat straw)	Acid hydrolysis	1	Casting	Water vapor transmission rate	[81]
CNF (Carrot pomace)	Thermochemical process	0.005	Casting	Hydrophobicity, Gas barrier property	[82]

strength, this results in a larger contact area and more interaction between the polymeric chains and the nanoparticles, strengthening the properties of nanocomposites [74]. To disseminate the nanocelluloses in PLA, many strategies have been explored to impart uniformity in the dispersion state. Esterification, oxidation/amidation, and silanization are three types of chemical modification that have been implemented to introduce small molecules to substitute hydrophilic hydroxyl groups on cellulose surfaces. Polymer grafting aims to improve interfacial compatibility through proper H –bonding without compromising its biodegradability [75]. Consequently, while developing a high-performance material for food packaging, features such as excellent nanocellulose dispersion and self-association must be taken into account.

Nanocellulose - starch

Starch is a polysaccharide that is widely present in plants. The availability, renewability, biocompatibility, processability, and non-toxicity of starch are among its intriguing qualities. [83]. There are many crops from which starch may be derived, such as potatoes, wheat, rice, maize, peas, bananas, barley, rye, oat, tapioca, etc. Starch properties are determined by the plant source, the amylose/amylopectin ratio, the genotype, the particle size distribution and morphology, and other factors including composition, pH, and chemical changes.

Due to its large hydrogen bonding network, starch has been used to create biobased films with superior oxygen barrier properties, however, its high hydrophilic nature limits its use. The increase in crystalline structure or amylopectin content makes starch films more resistant to moisture and relative humidity of the surrounding. Although it has a high tensile strength, its mechanical qualities are limited by its brittleness and amorphous region. Plasticizers can be acted to counteract the brittleness of starch and thus, improving flexibility and stability [84]. Plasticizer addition lowers the intermolecular force between polymer chains, allowing for modification of mechanical and water vapor barrier properties. Plasticizers boost the polymer chain's flexibility and resistance to deformation [85]. The potential effects of NFCs on the properties of biofilms have been presented in Table 3, which depicts that variations in mechanical qualities rely not only on the origins of the raw materials but also on the production method. To remove unwanted elements like lignin

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Table 3. Starch based polymer, nanocomposites (NC) and their mechanical properties

Starch source	Type & source of nanocellulose	Isolation method of NCs	Load concentration of nanocellulose (%)	Method of preparation	Improved properties	References
Industrial Thermoplastic starch	CNF (henequen)	Chemo-mechanical method	0.4	Casting	Tensile strength, elastic modulus, water vapor barrier properties	[86]
Corn	CNF (Microfibrillated Cellulose)	TEMPO mediated oxidation system	5	Casting	Tensile strength and Young's modulus, moisture susceptibility and transparency	[87]
Sugar palm	CNC (Sugar palm fibers)	Acid hydrolysis	0.05	Casting	Tensile strength, tensile modulus	[88]
Banana peel bran	CNF (unripe plantain bananas)	Chemi-mechanical	5	Casting	Tensile strength and Young's modulus	[89]
Cassava	CNF (Bamboo fibers)	Industrial	1	Casting	Tensile strength, Elongation at break	[90]
Corn	CNF (Kenaf bast fibers)	Mechanical process	10	Casting	Tensile strength and storage modulus	[91]
Potato	CNF (Rice straw)	Chemo-mechanical method	15	Casting	Yield strength, young modulus, humidity absorption resistance	[92]

and hemicellulose and isolate NFCs, chemical-mechanical methods including acid hydrolysis and TEMPO-mediated oxidation system have widely been used.

To enhance the useful aspects of starch in terms of water resistance and mechanical strength, starch has been expansively explored in combination with hydrophobic polymers such as PLA, and polycaprolactone (PCL), and also with reinforcement agents like clay nanoparticles. Mao et al. [93] prepared starch nanofibrillated cellulose (NFC)-reinforced nanocomposites with Polylactic acid (PLA). Rheological properties of PLA-based nanocomposite suspensions enhanced with an increase in the content of NFC. For 3% NCC (Nanocrystalline cellulose)), the PLA/starch composites showed a rise in tensile strength by 14.7%, and Young's modulus by 166.1%. Starch promoted the biodegradable qualities of PCL, while also bringing down the cost.

MMT (montmorillonite) is employed sparingly to create biodegradable starch and polymer blends with dramatically enhanced tensile qualities. In a research work done by Mulla et al. [94] nanocomposites of polycaprolactone/corn starch (PCL/CS) were reinforced with sodium

montmorillonite (Na-MMT) modified by Fatty hydroxamic acid (FHA). It was observed that showed that reinforcement of FHA-MMT into the PCL/CS blend led to increasing tensile strength of the blend, elongation at break, and thermal stability of the polycaprolactone/corn starch (PCL/CS) nanocomposite. The percentage of MMT and the proportion of starch to polycaprolactone/, which also optimized mechanical and thermal properties, determined the type of nanocomposite created. These results indicated that further exploration is needed to completely assess the attributes of starch-based nanocomposites, particularly the surface interactions between the matrix and filler. This is particularly true considering that there are many different kinds of cellulose nanofibers and clays, each with unique properties. Nanocomposite blends, with different glycerol– clay nanoparticles ratios in cassava starch film indicated that the incorporation of clay nanoparticles greatly affected both the barrier properties when compared to original starch blends containing sucrose, inverted sugar, and glycerol [95]. However, complementary approaches should be used to safeguard the integrity and barrier properties when applying them to food packaging. Furthermore, the extensive delamination of clay nanoparticles produced by equipment like the twin screw extruder used in the packaging industry may result in films with higher mechanical and barrier qualities.

Nanocomposites from cellulose microfibrils gelatinized with thermoplastic starch have been reported to reduce moisture sensitivity, increasing mechanical properties [96]. It was found that there is a feasibility to incorporate CNF with sufficient water content in the extrusion of bionanocomposites. The composites were effectively made using a twin-screw extruder. The mechanical properties of nanocomposites made from thermoplasticized starch packed with (CNFs) and produced by twin-screw extrusion of corn starch granules, glycerol as a plasticizer, and oxidized cellulose fibers have been demonstrated to be improved by *in situ* preparation during the extrusion process. Due to the integration of CNFs above 10%, the reinforcing impact was more important in raising the modulus [97].

Nanocellulose-Chitosan

Chitin deacetylation can be used to partially produce the polysaccharide known as chitosan. The advantage of using it is its propensity to degrade naturally, biofunctionality, antimicrobial

qualities, and superb compatibility with other natural polymers. However, the hydrophilic nature and high-water sensibility limit its use in food packaging materials [98]. The preparation process of chitosan-based polymer and NC nanocomposites influence the mechanical properties of such films, as shown in Table 4.

Effective moisture transfer control is a key attribute in the context of food preservation, particularly in high humidity levels conditions. To compensate for this behavior, several strategies have been proposed to control the swelling behavior of chitosan films such as cross-linking with reagents like genipin, formaldehyde, etc. [107]. The need of combining chitosan with additional constituents (proteins or polysaccharides) to make chitosan films more useful and stability has been emphasized [108]. The integration of nano cellulosic material greatly strengthens the young's modulus, heat resistance, and water vapor resistance of chitosan-based matrices, showing strong and uniform nanofiller distribution in the chitosan matrix and facile connection between nanocomposite phases [101].

In a biopolymer matrix, metal oxide nanoparticles are utilized vigorously as a nano-reinforcement agent to increase the mechanical strength, barrier, and antibacterial properties of the film. Nano-ZnO and nano-organo-clay nanoparticles were explored for their reinforcement impact in chitosan matrix for mechanical, thermal, and antibacterial properties of nanocomposite generated via polymer intercalation process [109]. The resultant nanocomposite showed superior mechanical characteristics as a result of the compatibility of the functional groups of chitosan, montmorillonite, and metallic oxides. Combining the effect of chitosan and zinc oxide nanoparticles Np-ZnO provided both bacteriostatic and bactericidal effect aginst *S. aureus* and *E. coli*. ZnO and TiO_2 facilitated the loss of membrane integrity by promoting peroxidation of the phospholipids found in microbial cell membranes. Roy et al., [110] reported the synergistic effect of tannic acid (TA) as a cross-linker and the reinforcing agent of titanium dioxide (TiO_2). Good compatibility between both the components (cross-linker and reinforcement agent) of nanocomposites resulted in satisfactory mechanical strength and thermal properties of the resultant film.

The addition of TiO_2 in chitosan/TA improved the mechanical and thermal properties of developed

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Table 4. Chitosan based polymer, NC nanocomposites and their mechanical properties

Type & source of nanocellulose	Isolation method of Ncs	Nanocellulose concentration (%)	Method of preparation	Improved properties	References
CNF (Carrot)	High-intensity ultrasonication	0.1	Vacuum Filtration	Increased composite Hydrophobicity, Thermal stability and antimicrobial activity	[99]
CNF (*Gluconacetbacter medellinensis* bacterium)	Fermentation	5	Casting	Tensile strength, young modulus,	[100]
CNC (Industrial)	Industrial	0.18	Casting	Thermal properties, antibacterial property	[101]
CNC (Corn cobs)	TEMPO mediated oxidation	14	Casting	Young modulus; water vapor permeability; tensile strength,	[102]
CNC (Microcrystal cellulose)	Acid hydrolysis	15	Casting	Crystallinity, good interfacial interaction	[103]
CNC (Industrial)	Industrial	2	Microfluidization	Elongation; tensile strength	[104]
CNC (Microcrystalline cellulose)	Acid hydrolysis	0.5	Casting	Mechanical properties	[105]
CNC (Ramie)	Acid hydrolysis	4	Casting	Mechanical property and water barrier property	[106]

nanocomposites owing to good compatibility between cross-linker and reinforcement agent. Talebi et al. [111] successfully fabricated a composite reinforced with cellulose nanocrystal (CNC) and cellulose nanofiber (CNF) with different concentrations. It was observed that by the incorporation of 7% CNF and CNC in the chitosan matrix, the tensile strength was increased by 104% and 52%, respectively. CNF presence also resulted

in higher values for tensile modulus and strength in comparison to CNC.

Nanocellulose - Alginate

Two polymers namely, D-mannuronic acid (M) and -L-guluronic acid (G) residues—make up the linear, anionic, water-insoluble polysaccharide known as alginate, which is generated from brown algae. It comprises alternating segments

of mannuronic and glucuronic acid (MG blocks), which directly affect its properties and can also change depending on its source [112]. The most noteworthy property of alginates is their propensity to react with polyvalent metal cations, especially calcium ions. The tridimensional network that results from the cooperative interaction of the M and G blocks due to the ions enables their coordination and packing. The crosslinking arrangement with polyvalent cations has been used to enhance the moisture barrier, mechanical resistance, cohesion, and stiffness of alginate films. The linear structure of alginate and the crosslinking encouraged by the addition of calcium led to the creation of a film, which also improved its barrier and mechanical capabilities. Lavric et al. [113] prepared 25 different biopolymer film combinations using chitosan, alginate, or nanocelluloses either separately or in various combinations. Three types of nanocellulose—CNC, —CNF, and —BNC were examined and evaluated for their morphology (SEM), density, contact angle, surface energy, mechanical strength, and oxygen and water barrier properties. The presence of CNC increased hydrophobicity and decreased moisture absorption. However, a combination of chitosan and alginate with nanocellulose demonstrated improved water vapor barrier property (15-45%) and oxygen permeability (45%) than single biopolymer-based nanocomposites.

Nanocellulose- Carrageenan

Carrageenan, a naturally occurring linear polysaccharide, is obtained from a cell wall of red seaweed belonging to the Rhodophyceae family. The propensity to form strong films and coatings with the highest mechanical strength is distinctive to kappa carrageenan among all other types. Kappa (κ) carrageenan, mainly extracted from red sea moss *Kappaphycus alvarezii*. It forms a rigid and 3D gel in the presence of potassium salts through the association of its linear helical parts. These composites, like others derived from seaweeds, efficiently shield to protect vegetables and fruit against turgor loss, compound oxidation, and ageing processes. If combined with ascorbic acid, they also reduce the number of bacteria [114].

Laponite and carrageenan coatings with silver nanoparticle loading on oxygen plasma-surface modified Polypropylene films made via green synthesis optimized the high strength, oxygen, and water barrier performance significantly [115].

The resultant film was found to be shown high antibacterial properties against common food spoilage bacteria. Roy et al. [116] incorporated melanin (Mel) as a reducing and capping agent and found that combining carrageenan and AgNP increased the film's qualities, including its thermal stability, mechanical, barrier functionalities, and antibacterial activity. In a similar study by Roy and Rhim [117], carrageenan was combined with zinc nanoparticles formed using melanin as a stabilizing agent. The resulting film had a better water vapor barrier, greater thermal stability, enhanced mechanical characteristics, and demonstrated excellent antibacterial activity against Gram-negative foodborne pathogenic bacteria. A broad-spectrum antibacterial activity was demonstrated by copper sulfide nanoparticles (CuS-NPs) added to natural carrageenan with a photothermal impact. Mechanical strength, thermal property, and transparency were among the major aspects that were focused on and demonstrated to be improved. Strong antibacterial uses for CuS-NPs with NIR light irradiation in the food packaging sector have shown a significant amount of potential in an antibacterial test on beef [118].

PROPERTIES OF NANOCOMPOSITES

Depending on how nanoparticles are distributed throughout the polymer matrix, composite films are shown to have improved mechanical and barrier properties.

Mechanical properties

Nanosized cellulose has a high strength, biocompatibility, and aspect ratio, which improves the mechanical properties of nanocellulose-reinforced polymer nanocomposites [119]. Nanocellulose holds a lot of strength as a reinforcement phase towards certain biodegradable polymer matrices. The miscibility of composite components is well recognized to have a considerable effect on their overall qualities. The homogenous and consistent distribution of nanoparticles in the matrix material determines the mechanical performance of composite films. The upsurge in Young's modulus and tensile properties is projected to come at the outlay of elongation and breaking strength [120].

According to Jancy et al. [121], reinforcement cellulose nanoparticles to the PVA matrix sufficiently improved the mechanical performance, resulting in a seven times upsurge in strength

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and a six-fold rise in elongation at break. The crystallinity of CNP is related to higher stiffness, which ultimately culminated in the higher tensile strength of the bio-nanocomposite. The amount of nanocellulose present in the polymer matrix affects the biocomposite film's stiffness and tensile strength. Segregation occurred when the concentration reached a certain level. PVA-chitosan films blended with CNCs were developed by Miri et al. [122], and the results revealed a rise in young's modulus of 105%, tensile strength of 77%, and toughness of 68% with just a 5-weight percent addition of CNCs to a PVA/CS blend. Santos et al. [123] integrated sugar palm CNF into sugar palm starch (SPS), leading to a 100 percent rise in strength and young modulus with merely 1% CNF. It was observed that ultrasonication promoted NC dispersion and resulted in the improvement of film properties.

Casting evaporation in the presence of an aqueous solution is considered as suitable preparation process because of the uniform dispersion of nanoparticles in the polymeric matrix and also maintains surface hydroxyl groups leading to improved mechanical performance. However, the reinforcement of nanocellulose is functional up to a certain limit. Under a certain range, nanocellulose dispersion forms a strong network within the polymer matrix. If the concentration of nanocellulose is exceeded above this limit, segregation may prevail leading to weakening of moisture barrier properties and thermal degradation [124].

Thermal properties

Considering the possible application of nanocomposites in food packaging, thermal stability must be taken into consideration. Due to the existence of H- bonding, cellulose chains are organized in a compact form. The dense crystalline structure of cellulose is considered to be responsible for its temperature resistance, which also helps it to withstand atmospheric forces. The thermal stability of nanocomposites may be caused by variations in nanocellulose supply, polymer matrix, production methods, and drying process [125]. The hydrophilic properties of nanocellulose cause it to absorb moisture, resulting in its constituents breaking down through hydrolysis and eventually losing their thermal stability. Acid hydrolysis in the presence of sulphuric acid incorporates sulfate esters, resulting in a lowering of heat stability of the

nanocomposites [42, 126].

Good dispersion of nanocellulose and strong intermolecular bonding among nanosized cellulose and polymer matrices act as driving forces in strengthening thermal stability. Soy protein isolate nanocomposites containing (CNCs) and CNF reinforcement were produced by Tosi et al. [127]. (CNFs). The comparison between thermal stability showed that CNF incorporation in soy protein nanocomposite provided better thermal stability than CNCs incorporation. This effect was found to be related to the high aspect ratio and homogenous dispersion of CNFs in the protein matrix, whereas in the case of CNC-incorporated nanocomposites, high surface area lowered the thermal stability.

Mandal et al. [128] emphasized that a tough interaction through H-bonding between nanocrystalline cellulose and polyvinyl alcohol polymeric matrix strengthened the thermal stability of the resultant nanocomposite in the crosslinked state. Utilizing different ratios of bagasse-extracted nanocellulose PVA nanocomposites in the linear and crosslinked states were created. The intense association between the nanofiller and the matrix boosted the heat energy requisite for the breakage of cross-linked PVA-CNC nanocomposites, which promoted disintegration susceptibility.

Barrier Properties

Food packaging should prevent the mass movement of water vapor and oxygen, which is related to numerous processes involved in food deterioration in addition to enclosing and protecting the food from physical damage. Water vapor permeability (WVP) and oxygen permeability (OP) are the parameters that show a film's resistance to gas diffusion and moisture penetration, respectively [129]. Crystalline structure, pressure, temperature, hydrophilicity (polysaccharides are naturally hydrophilic), density and thickness, pore size and structure, and water vapor transmission rate (WVTR) are factors that influence a packed polymer/composite film.

Saxena et al. [130] stated the impact of adding CNC on the barrier characteristics and composite formed by casting an aqueous solution comprising CNC, sorbitol, and xylan. Oxygen transmission rate tests demonstrated that the films had much lower oxygen permeability than the control films. The optimal path length for molecules to diffuse determines the shape, homogenous distribution, and interaction of nanocellulose with the polymeric

matrix, all of which have a substantial impact on the gas barrier performance. In addition, a high degree of crystallinity and capacity for robust network formation of nanocellulose, due to the existence of hydroxyl groups, depicts its potential to possess high gas barrier properties [131]. However, this pattern persisted up to a certain limit, or 70 % relative humidity, after which free hydroxyl groups begin absorbing moisture from the environment, showing a major impact of atmospheric moisture conditions. Developing multilayered composite films is another approach to expand the barrier performance of food packaging film. Gars et al. [132] created PLA-based multilayered materials by coating CNFs and CNCs with rosin-based nanoemulsions and sandwiching nanocellulose films between PLA sheets. Oxygen permeability was reduced by 84–96% when nanocellulose was added as an inner layer between two PLA films, significantly enhancing the oxygen barrier.

Surfactants are effective in improving the matrix - nanocellulose interaction and NC dispersion. To strengthen the PLA matrix, Fortunati et al. [133] utilized CNC and surfactant-modified (s-CNC) to prepare nanocomposite. Improved interaction between the cellulose structures and the PLA matrix was observed by s-CNC dispersion in the matrix. The incorporation of 1% and 5% s-CNC showed strong oxygen barrier properties, whereas the addition of 1% of modified CNC led to a 34% reduction in water permeability. By using electron microscopy, the homogenous distribution of s-CNC in the matrix in the presence of surfactants was validated. The quality of the food may be adversely impacted by moisture permeation through the food packing material. The decrease of film cohesion with increasing relative humidity affects the potential of the film to act as a gas barrier. Due to the availability of a hydrogen bond network, significant cohesion energy density was seen when CNCs were blended into a matrix. Nanoparticles caused the free volume to decrease and chain flexibility to decrease. Slavutsky et al. [134] observed that the CNC-created tortuous path dramatically enhanced the water barrier properties of the films. The solubility, contact angle, and water sorption isotherm of the films indicated that reinforcement of CNC reduced the sensitivity to water molecules. Srivastava et al. [135] reported that nanocomposite film formed with PVC-banana pseudostem fiber – nanocellulose displayed good water vapor permeability that was inversely proportional to amount of nanocellulose added.

Biodegradability

Since nanocellulose polymers have inherent bioproperties and are a good substitute for synthetic materials, they are being actively researched for their biodegradability. The biodegradability of cellulose nanoparticles in an aqueous environment was first documented by [136], where it was demonstrated that nanocellulose decomposed at a faster rate than its macroscopic forms owning to its surface area. Fullerenes and carbon nanotubes did not undergo biodegradation, while starch nanoparticles gave similar results to cellulose nanoparticles.

Abraham et al. [137] employed vermicomposting method using worm Eudrilus eugeniae to test biodegradation behavior of the composite formed by reinforcing nanocellulose in natural rubber. It was found that the cross-linked composites deteriorated more slowly than the non-cross-linked elements. Potivara et al. [138] reinforced BNC with natural rubber film and subjected it to a soil burial test to determine its biodegradability. Within 5 to 6 weeks in soil, the composites had entirely broken down. Deepa et al. [139] documented the rate of degradation in cellulose nanofibril-reinforced sodium alginate nanocomposite film into compost within 5 weeks. However, the rate of degradation was influenced by the amount of reinforcement in the sodium alginate matrix. The method of soil burial has been employed to study the biodegradability of PLA/CNF nano-biocomposite [140], nanocellulose blended PVA composite film [141], nanocellulose based starch film [142], nanocellulose reinforced nitrile butadiene rubber (NBR) nanocomposite [143], and nanocrystalline cellulose reinforced Polybutylene adipate terephthalate (PBAT) composites [144]. A summarized list of nanocomposites that have been tested for its biodegradability by soil burial test is given in Table 5.

APPLICATION OF 3D/4D PRINTING FOR FOOD PACKAGING

In the food packaging industry, 3D-printing is becoming an emerging technology to develop bio-based packaging materials. Using eco-friendly and non-toxic materials, 3D printing is a cutting-edge way for creating intelligent packaging, smart devices, and self-indicating, multifunctional components more economically than traditional production techniques. Wang et al. [146] created

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Table 5 Biodegradability of different nanocomposites

Nanocellulose	Matrix	Test period	Findings	References
Nanocrystalline cellulose	Poly (butylene adipate-co-terephthalate (PBAT)	4 months	Degradation rate of PBAT was increased in presence of nanocellulose. Polymer chains were converted into smaller particles but complete degradation was not seen. Based on this finding, PBAT may remain as microscopic form so it requires further degradation.	[144]
CNF	Cellulose	70 days	All cellulose nanocomposites (ACCs) showed high biodegradability, with mass losses of up to 73% after 70 days of soil burial. By changing the matrix phase's structure, it may be possible to alter the biodegradability of ACCs.	[145]
Nanocellulose	Starch	15 days	Biodegradation rate was found to be slow with increasing vibration time because proper dispersion of fiber in matrix by ultrasonicator led to low water absorption which made it less prone to microbial invasion.	[142]
Nanocellulose	PVA	10 weeks	After 10 weeks, reduction in 90% weight was observed in compost than normal soil. NCC modified PVOH found to be degraded more rapidly than pure PVOH.	[141]
Nanocrystalline cellulose	Nitrile butadiene rubber (NBR)	6 weeks	For NBR/Aox-NCC composite films, no significant results were achieved within 2 months instead of to within 6 months, indicating that the degradation process took substantially longer to complete. The NCC has been modified with gallic acid, which is believed to boost the composites' resilience to degradation in soil environments.	[143]

fundamental food materials (cellulose, starch, and protein) based on 2D films through the implementation and utilization of the concept of 4D printing in the food sector. In the presence of water, it was able to change its shape from 2D to 3D. It was discovered to be appropriate for the "flat packaging" approach, which greatly lowers shipping charges and storage requirements. Nida et al. [147] prepared food package casing using sugarcane bagasse (SCB) through extrusion-based 3D printing for the packaging of foods with low moisture content. Zhou et al. [148] developed a biodegradable CMC-based antimicrobial-cushioning dual function for food packaging aerogel via coaxial 3D-printing technology. The aerogel had good cushioning and resilience performance with an average compression resilience rate of >90%, inhibition effect against E. *coli* and *S. aureus*. 3D-printing technology has also been used to prepare antimicrobial composite film by incorporating nano silica-carbon-silver particles in polymeric solution [149]. The nanocellulose-based hydrogels were developed by Lu et al. [150] as an intelligent packaging material to indicate the freshness of meat in the form of a pH-sensitive indicator. Parimita et al. [151] proposed an efficient method to develop a citric acid crosslinked chitosan-based hydrogel which was triggered by the presence of solvent resulting in shape morphism behavior.

Nowadays, 4D printing in conjunction with Shape memory polymers (SMP) has resulted in several breakthroughs in the domain of smart materials in food packaging. Shape memory polymers are intelligent materials that instantly alter shape in response to external environmental stimuli. On exposure to external stimuli, such as heat, light, electrical or magnetic fields, humidity, magnetism, or chemicals, stimulus-responsive polymers demonstrate the capacity to maintain their original morphologies from a programmed temporary shape [152]. Certain polymers, such as thermoplastic polyurethane, polycaprolactone, ethylene-vinyl acetate copolymer, and polylactic acid can be turned into shape memory polymer fibers (SMPFs) with a large aspect ratio and practicability. PLA is presently the most frequently utilized in 4D printing due to its processibility, shape memory properties (SMP), low printing temperature, and nontoxicity [153]. However, further research into the reinforcement of nanomaterials into shape memory polymers (SMPs) and hydrogels is required

to address issues like the robustness of the structure in the designs and the prediction of the bio-printed structures. With the requirement of appropriate ink, there is also a need for an appropriate printer that can help in designing the structures that would possess all the properties of a given traditional packaging materials. Another issue to be addressed is the consumer acceptance and legal framework associated with 3D/4D printing technology of food packaging materials. In order to acquire the confidence of the consumer it is vital to clarify by providing related information regarding the nature of the items and the safety through explanatory label systems.

FUTURE WORK

Despite several encouraging developments on a pilot scale, there are still some difficulties to overcome before nanocellulose composites can be manufactured on a large scale. The commercialization of nanocellulose is hindered by myriads of conventional technologies, such as the release of significant amounts of acid waste water effluent during acid hydrolysis, high energy requirements during mechanical processing, and the protracted reaction times during enzymatic hydrolysis. The motive for exploring nanocellulose includes the development of novel extraction techniques that require a simple route and little to no wastewater. Further research is needed to develop simple, one-step extraction techniques employing mild reaction conditions to address the current challenges because multiple stages are costly and restrict commercial and industrial use. Therefore, the focus of future studies should be on preserving and/or increasing the morphological features of nanocellulose through straightforward and economical processing. The biodegradation process should be properly explored because it is a complex and challenging process to understand when composite materials are exposed to a variety of environmental factors. Further research on this topic is anticipated to monitor the release of harmful substances into the environment and better understand the mechanism of bionanocomposite that contain nanofiller/additives. Recycling cellulose nanocomposite is a viable alternative to degradation because it would not only save time but also prevent the wastage of resources. Additionally, to withstand heat damage, nanocellulose must be coupled with thermoplastic films or sealant layers to resist heat damage since it has not yet been

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rendered heat-sealable.

Even if the safety of bulk cellulose is well established, nanocellulose must be assessed in terms of potential effects, toxicity, and migration before its implementation in food contact packaging materials. Because of the possibility that they could enter cells through their small size and eventually stay in the system. Then, more research must emphasize those areas to ensure that any issues about the safety of nanocellulosic packaging materials are addressed to enable their safe and long-term commercialization. On the other hand, it is important to have a better understanding of legislative and regulatory provisions to deal with a major challenge in the commercialization of emerging food packaging technologies. It is important to conduct more studies on product evaluation, safety concerns, and the suitability of developed packaging solutions for interaction with food. Environmental awareness among customers, which will have a positive and significant impact on their purchasing decisions, can be used to encourage sustainable packaging. A close relationship between industry and academia is necessary to speed up commercial applicability.

There are some issues of 3D/4D print technology also that still need to be addressed. One such issue is that not all the biomaterials can be printed i.e., appropriate ink materials are not available and not all the operations of a standard packaging production can be carried out. Currently, the majority of the available polymeric materials and hydrogels are only responsive to a single stimulus, while bioprinted products are responsive to multi-stimuli exhibiting simple deformation such as curling, folding, and bending. Further studied are required on these aspects to fully utilize the potential of 3D/4D print technology in the realm of food packaging.

CONCLUSION

The biocompatibility, biodegradability, high aspect ratio, transparency, hydrophilicity, and non-toxicity of nanocelluloses are factors that contribute to their significance as potential candidates in nanocomposite food packaging film. The biopolymers are having poor tensile strength and high moisture absorption properties, which prohibit them from being extensively used in food packaging. Incorporating nanoparticles often satisfactorily enhanced the mechanical strength, thermal resistance, and barrier properties of these biopolymers making them ideal for food packaging.

By enhancing the compatibility of hydrophilic nanocellulose with hydrophobic polymers and promoting their dispersion, the modification of the nanoparticles considerably enhances the barrier characteristics. Being new and sustainable

raw materials for the nanotechnology community, nanocellulose (CNC, CNF, and BNC) is a crucial component of the integrating models. Agricultural biomass can be used to extract nanocellulose as a competitive substitute, which can then be transformed into nanofibers or nanocrystals by combining pretreatment with acid/mechanical processes. The influence of nanoparticles on polymeric matrices is shaped by a multitude of variables, including their source, diameter/length ratio, and compatibility with the polymer. Apart from that, a variety of approaches can be implemented to avoid unnecessary self-aggregation and to maintain high mechanical performance, which has been primarily ascribed to the nanoparticles' inescapable coating (to avert self-aggregation during drying) and the intrinsic concealing of surface hydroxyl groups that prevented mechanical permeation through H-bonding. To attain a mass manufacturing application, nevertheless, ongoing technological difficulties and financial problems must be resolved. Further pilot-scale research investigations are expected to develop high-value nanocellulose-based composites that can not only reduce energy use but also address certain long-standing environmental problems, while also increasing commercial uses.

CONFLICT OF INTEREST

The authors hereby declare that there is no conflict of interest.

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