REVIEW PAPER

Poly(lactic acid)/nanocellulose biocomposites for sustainable food packaging

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Abstract Considerable interest in food packaging research has been prompted by the rising environmental impact of waste, customer awareness for readily accessible foods with extended shelf life, and ecological consciousness about the scarcity of natural resources. The utilization of plastic-based food packaging has resulted in a significant environmental concern due to the accumulation of substantial quantities of plastic in the surrounding areas. Research efforts are being motivated by ecological and health considerations in the pursuit of developing biodegradable films. Besides, poly(lactic acid) (PLA) has been suggested as a possible substitute for petroleum-based polymers because of its sustainability, simple accessibility, and biodegradability. PLA is a biodegradable plastic made from sugar beet or maize starch that may be fermented by bacteria to generate material with desirable qualities like transparency and rigidity. However, there is a need for further improvement in certain properties of PLA, such as flexibility, toughness, permeability, and thermal properties. One potential approach for enhancing these attributes involves the integration of nano-reinforcement. The utilisation of nanocellulose derived from biomass has garnered significant interest in recent times owing to its renewable nature, ability to biodegrade, impressive mechanical strength, low density, and considerable economic worth. In this study, we present a comprehensive overview of the most up-to-date methods for synthesising nanocellulose and its use as a filler material in the manufacture of PLA nanocomposites for food packaging. In addition, this study examines

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the emerging challenges and potential advantages associated with the utilization of PLA biocomposites incorporated with nanocellulose in the food packaging sector.

Introduction

Polymers are an essential component of modern life because of the desirable features they possess, such as stability, resilience, and the ease with which they may be produced (Chen and Patel 2012). Plastics have several uses and can be found in a wide number of industries, including packaging, building and construction, automotive applications, electrical and electronics applications, aerospace applications, and corrosion prevention and control (Silvestre et al. 2011). At the moment, more than 40% of the entire quantity of plastics consumed across the globe is utilised in the packaging industry (Surendren et al. 2022). The global flexible packaging market share by applications is shown in Fig. 1a. The key functions of packaging can be generally categorized into two main groups: primary functions and secondary functions. The primary functions of packaging encompass various aspects, including protection, storage, loading,

and transportation of the product. To fulfil these functions effectively, the packaging must possess qualities such as strength, non-porosity, and the ability to resist external conditions that may be encountered in the storage or transportation surroundings (Petersen et al. 1999). Secondary purposes, such as those related to encouraging sales of the product, may require the packaging to have a good physical appearance (glossy) or to have good transparency in order to draw the attention of customers. It may be necessary to include information on the packaging, such as the product's contents, the nutrition content (of food products), and directions for use; this will require that the material used for the packaging be printable. In addition, the recyclability of polymers can be a significant factor in evaluating whether or not they are suitable for use as a packaging material, particularly for high-volume applications (Lange and Wyser 2003; Chen et al. 2020a, b).

The determination of packaging material properties is influenced by the physical and chemical attributes of the product, as well as the external conditions encountered during storage and transportation. Plastics are highly desirable materials for packaging applications due to their versatile range of properties that can be customised to meet specific product requirements (Lau and Wong 2000; Subramanian 2000). Polyethylene (PE), polypropylene (PP), polyethylene terephthalate (PET), polyvinyl
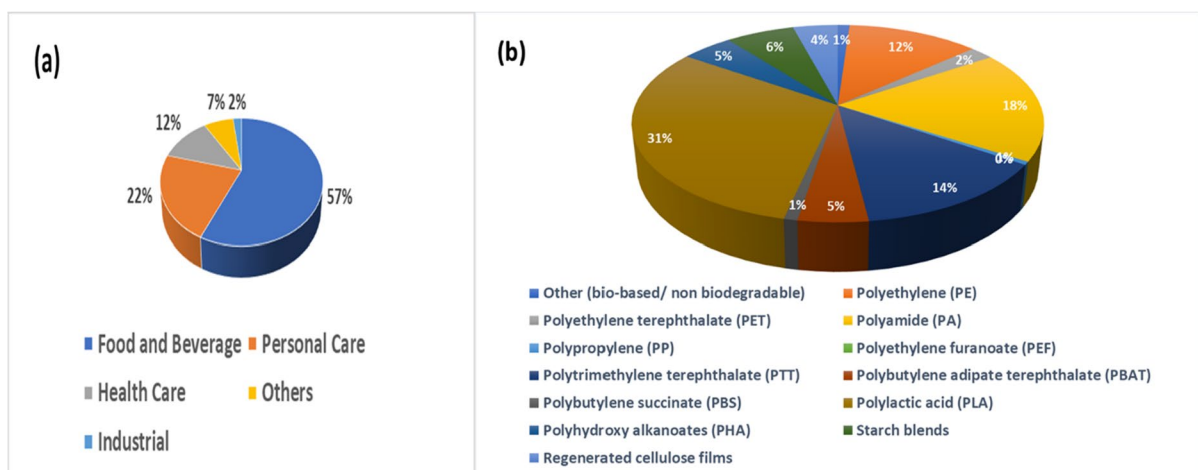

Fig. 1 **a** The global flexible packaging market share by applications (https://www.gminsights.com/industry-analysis/flexible-packaging-market) [EUBIO_Admin Market n.d., Flexible Packing Market

Size | Industry Analysis (2024a)–2032 n.d.,] **b** Global production capacities of bioplastics in 2023 (https://www.european-bioplastics.org/market/)

chloride (PVC), and polystyrene (PS) are widely recognised as the predominant types of packaging plastics (Siracusa and Blanco 2020, Guillard et al. 2018). These materials collectively constitute over 90% of the overall volume of plastics employed in packaging applications. The production of plastics commonly relies on raw materials derived from petroleum products obtained through various refining processes (Khoo et al. 2010). The majority of conventional plastics, which are derived from fossil fuels, are known to be non-biodegradable, leading to potential environmental pollution. The growing awareness and concern surrounding environmental issues have sparked a surge in research on bio-based sustainable plastic packaging materials in recent years (Peelman et al. 2013; Jabeen et al. 2015; Shlush and Davidovich-Pinhas 2022). According to the American Society for Testing and Materials, biodegradable polymers refer to polymers that possess the ability to undergo degradation through the action of naturally occurring microorganisms in the environment, including bacteria, fungi, and algae. This degradation process leads to the formation of carbon dioxide, methane, water, and inorganic constituents. These polymers have the potential to be diverted from surface disposal landfills and instead used in the production of compost (Lenz 1993).

Biopolymers are polymers derived from natural materials, either chemically produced from biological substances or completely synthesised by living organisms. (Smith et al. 2016). Biopolymers are commonly categorized into five distinct groups based on their origin (Phiri et al. 2023; Jacob et al. 2020; Scarfato et al. 2015).

Class I: This category includes biopolymers made from naturally occurring substances. These renewable resources include (i) biomass from agricultural sources, (ii) polysaccharide materials derived from animals, and (iii) protein-based materials derived from either animals or plants. Starch and cellulose are the two main biopolymers that are produced using agricultural resources. Chitin and chitosan are two examples of biopolymers generated from animal-sourced polysaccharide material. Wheat, zein, pea, soy, and canola (plant origin) and casein, gelatin, keratin, and whey (animal origin) are biopolymers made from protein-based resources.

Class II: This group includes biopolymers that come from microbial sources. Polyhydroxy alkanoate (PHA) is among the biopolymers derived from microbial sources.

Class III: This category encompasses biopolymers that are derived from monomers obtained through renewable resources or microbial fermentation. An example of such a biopolymer is Poly (lactic acid) (PLA).

Class IV: In this category, biopolymer monomers are derived from petrochemical sources. Polycaprolactone (PCL) and Poly(butylene adipate-coterephthalate) (PBAT) are examples of class IV biopolymers in which both monomer and biopolymer are produced by conventional methods.

Class V: This is a particularly novel category in which biopolymers are derived from a combination of renewable and petrochemical monomers. Class V biopolymers include poly (trimethylene terephthalate) (PTT), which can be produced from terephthalic acid (petrochemical origin) and 1,3-propanediol (biological origin).

Polylactic acid (PLA), also referred to as polylactide, is a highly promising polymer within the realm of biodegradable materials (Mukherjee and Kao 2011). It holds a significant position, constituting approximately 31% of the overall bioplastic production, as illustrated in Fig. 1b.

PLA is a type of linear aliphatic thermoplastic polyester that is sourced entirely from renewable resources. These resources include sugar, corn, potatoes, cane, beet, and other similar sources (Jamshidian et al. 2010; Inkinen et al. 2011). PLA has gained significant commercial traction and is widely employed in the realm of food packaging. It finds extensive application in packaging perishable or time-sensitive food products, serving as containers, drinking cups, salad cups, overwrap and lamination films, as well as blister packages. Nevertheless, the utilisation of these materials in food packaging has been limited due to their low thermal stability and inadequate barrier properties (Auras et al. 2004; Ahmed and Varshney 2011. The integration of nanomaterials has been proposed as a potential solution to mitigate these issues and enhance the potential of PLA-based packaging systems (Mulla et al. 2021).

Nanocellulose has been suggested as a potential load-bearing component for the development of

cost-effective biomaterials. This is attributed to its advantageous characteristics, including a high aspect ratio, favourable mechanical properties, and complete degradability and renewability. When comparing nanocellulose to other inorganic reinforcing fillers for biopolymers, several additional benefits can be observed. These advantages include the broad availability of sources for nanocellulose, its low energy consumption during production, the ease of recycling through combustion, and its relatively easy processability due to its non-abrasive nature. These characteristics enable nanocellulose to achieve high filling levels in biopolymers, resulting in substantial reductions in costs (Kim et al. 2015). By 2029, the market for nanocellulose is expected to have grown from its 2022 valuation of 0.40 billion to 0.49 billion, with a compound annual growth rate (CAGR) of 21.9%. Figure 2a shows the amount of nanocellulose produced worldwide. Figure 2b also displays the number of publications produced on PLA/cellulose nanocomposites over the last ten years (based on Google Scholar).

The objective of this review is to provide a comprehensive overview of the benefits associated with the incorporation of nanocellulose into a PLA matrix for the purpose of creating bionanocomposites suitable for biodegradable food packaging applications. This paper commences by providing a thorough review of the properties of PLA-based food packaging material, with a specific emphasis on its performance. This article aims to explore the production and extraction methods of nanocellulose derived from agricultural wastes. This article provides a comprehensive summary of the properties of PLA-NC bionanocomposites as food packaging materials. These properties include morphology, rheology, mechanical strength, thermal stability, barrier properties, antioxidant activity, antibacterial properties, and biodegradability. Finally, this article provides a concise overview of the safety considerations related to the development of PLA-NC bionanocomposites for their application in food packaging.

Poly(Lactic Acid) (PLA)

In recent years, Poly(lactic acid) (PLA) has emerged as one of the biopolymers that has garnered the most interest due to its economic and commercial feasibility throughout the processing stage. PLA is a member of the family of aliphatic polyesters that are constructed from alpha-hydroxyacids (Anderson and Shive 1997). Other members of this family include polyglycolic acid and polymandelic acid. PLA polymerization may be achieved by either microbial fermentation of renewable resources, such as rice, wheat, maize, cane, potatoes, beets, etc., or chemical polymerization. PLA can be industrially produced using either lactic acid (LA) polymerization or lactide ring-opening polymerization (ROP). As depicted in
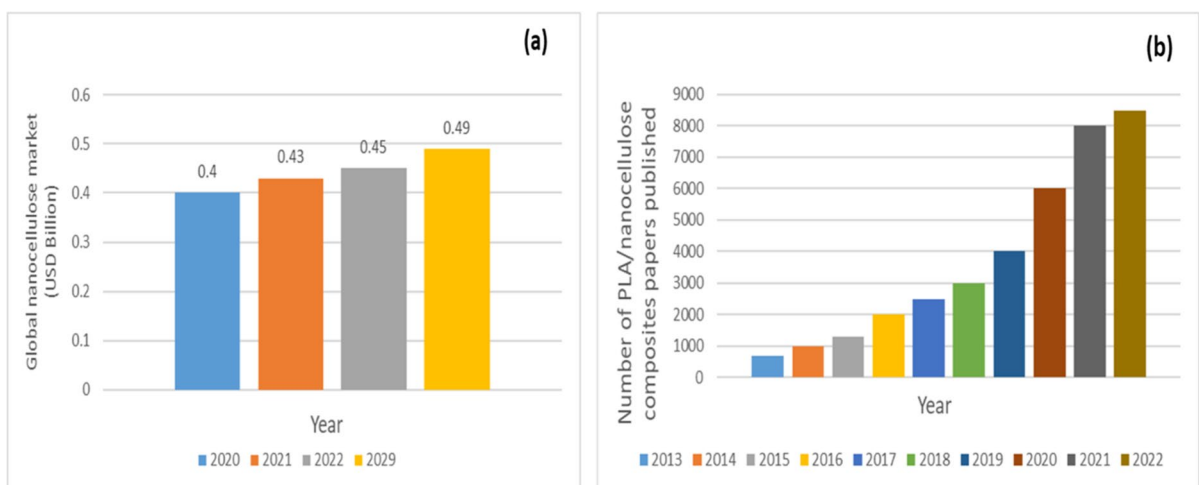

Fig. 2 a Global nanocellulose market (https://www.futuremarketinsights.com/reports/nanocellulose-market) [Nanocellulose Market n.d.,] **b** Number of publications produced on PLA/cellulose nanocomposites over the last ten years (based on google scholar)
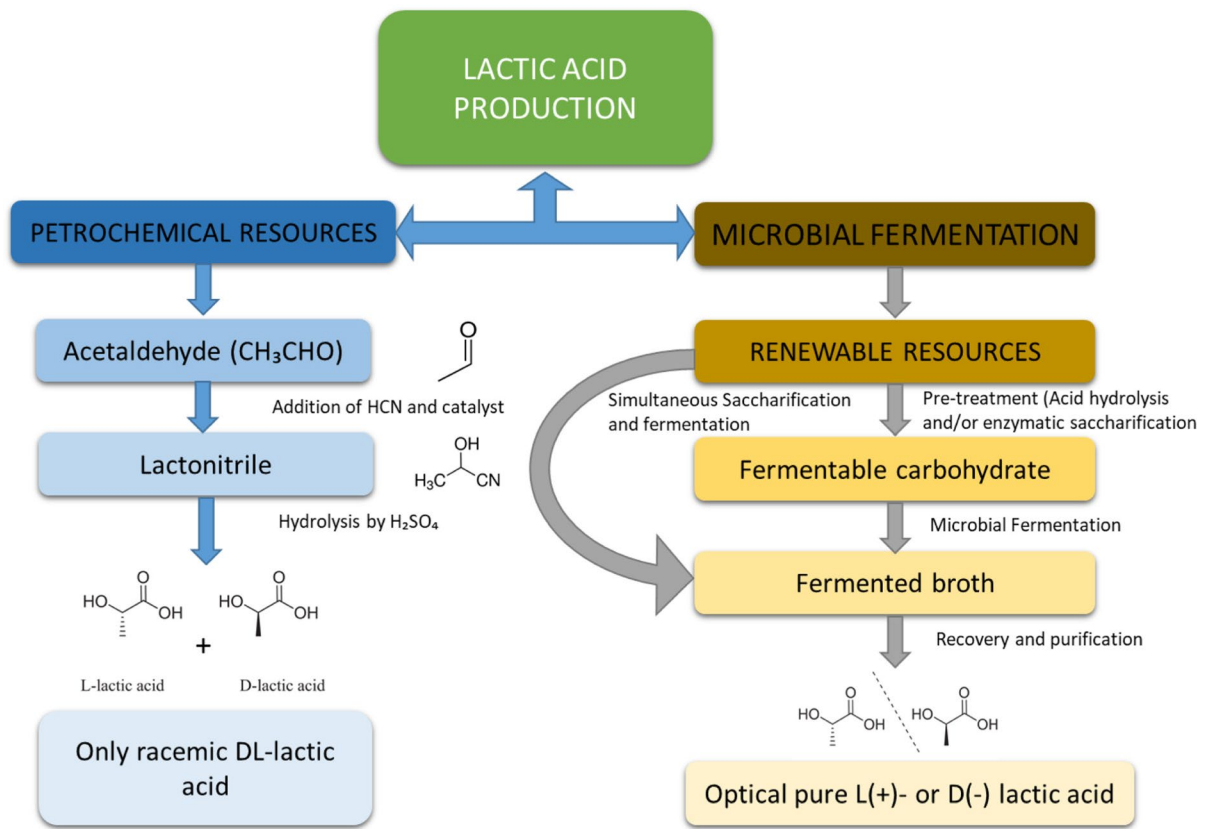
Fig. 3 Overview of lactic acid production through chemical synthesis and microbial fermentation (Ahmad et al. 2020)

Fig. 3, the lactides used to make PLA are generated by the microbiological fermentation of agricultural wastes, making it a more environmentally friendly alternative to items obtained from petrochemical resources (Mulla et al. 2021; Madhavan et al. 2010).

Table 1 displays the lactic acid yield from various bacteria and sources. Since it was discovered that PLA's performance was superior to that of synthetic plastic materials in many situations, its usage as a green food packaging material is on the rise.

PLA merits and demerits

Packaging materials enhance the product's shelf-life by protecting it from potential degradation caused by physicochemical or biological factors. Additionally, these materials play a crucial role in maintaining or even enhancing the overall quality and safety of the product throughout its storage and handling processes. Furthermore, it is imperative to consider the

biodegradability of materials once they have reached the end of their useful life, as this can help mitigate potential environmental waste disposal challenges. Hence, the rising need for the utilization of biodegradable polymers derived from renewable sources as a substitute for petroleum-based polymers in packaging materials has been observed, owing to their potential for reducing environmental pollution (Siracusa et al. 2008; Taib et al. 2023). PLA is a commonly used biopolymer in industrial applications. The use of PLA in packaging offers numerous advantages compared to conventional petroleum-based polymers:

(a) Transparency is high compared to poly(ethylene terephthalate) (PET) and poly(styrene) (PS), with a transmission of visible light between 540 and 560 nm (Armentano et al. 2013).

(b) Decomposition of PLA in a biological context, such as in presence of soil or compost occurs in two stages: hydrolytic degradation and enzymatic

Table 1 The lactic acid yield from various bacteria and sources

Substrate	Microorganism	Lactic acid yield	References
Wheat starch	Lactococcus lactis ssp. lactis ATCC 19435	0.77 to 1 g/g substrate	(Hofvendahl et al. 1999)
Whole wheat	L. lactis and Lactobacillus delbrueckii	0.93 to 0.95 g/g substrate	(Hofvendahl and Hahn-Hagerdal 1997)
Barley	Lactobacillus casei NRRL B-441	0.87 to 0.98 g/g substrate	(Linko and Javanainen 1996)
Cassava bagasse	L. delbrueckii NCIM 2025, L. casei	0.90 to 0.98 g/g substrate	(Rojan et al. 2005)
Potato starch	Rhizopus oryzae, R. arrhizus	0.87 to 0.97 g/g substrate	(Huang et al. 2005)
Corn starch	L. amylovorus NRRL B-4542	0.935 g/g substrate	(Nagarjun et al. 2005)
Wheat and rice bran	Lactobacillus sp	129 g/ l	(Yun et al. 2004)
Agro industrial starchy waste	Enterococcus faecalis RKY1	~0.93 g/g substrate	(Oh et al. 2005)
Cellulose	L. bulgaricus NRRL B-548 N	80 g/l	(Venkatesh 1997)
Corncob	Rhizopus sp. MK-96–1196	90 g/l	(Miura et al. 2004)
Wood	Rhizopus oryzae NRRL 395 N	0.85 g/g substrate	(Woiciechowski et al. 1999)
Waste paper	Rhizopus oryzae N	0.8 g/g substrate	(Park 2004)
Wood hydrolyzate	Enterococcus faecalis RKY1	~0.9 g/g	(Wee et al. 2004)
Mixed organic wastes (Banana peels and food waste materials)	Enterococcus durans BP130	6.7%	(Hassan et al. 2019)
Wheat straw hemicellulose hydrolysate	Lactobacillus pentosus and Lactobacillus brevis	95%	(Garde et al. 2002)
peanut meal	Bacillus sp. WL-S20	225 g/l	(Meng et al. 2012)
cassava bagasse	mixed culture of Bacillus coagulans and lactobacillus rhamnosus	0.88 g/g	(Chen et al. 2020a, b)
vitamin-supplemented soybean hydrolysate	Lactobacillus rhamnosus ATCC 10863	125 g/l lactic acid from 150 g/l glucose	(Kwon et al. 2000)
Rice straw	Lactobacillus lactis 2369	82.2 gL^{-1}	(Mottaghi et al. 2022)

degradation. The hydrolysis process begins with water penetration into the polymer and progresses to random nonenzymatic chain scissions of the ester group. The breaking of ester bonds leads to a decrease in molecular weight and the generation of oligomers and lactic acid, which may be digested by microorganisms such as fungus and bacteria (Kara and Molnár 2023; Arena et al. 2011). A molecular weight of 10,000–20,000 g/mol seems to be the crucial upper limit for bacteria to metabolize and subsequently convert to carbon dioxide, water, and humus. While the first hydrolytic reaction may occur in the bulk polymer through water diffusion, the second step, mediated by bacteria, happens exclusively at the surface (Yu et al. 2023; Kale et al. 2007). When lactic acid is transformed into PLA, significant alterations in both the chemical and biological breakdown processes take place. PLA is not as easily broken down and consumed by living organisms

as lactic acid itself. The biodegradability of PLA is influenced by molecular weight, chemical bonding, stereochemistry, crystallization, water uptake, acidity, etc. (Naser et al. 2021). Degradation of PLA happens in environments with high temperatures (above 55 °C) and humidity, where specific microorganisms with enzymes like carboxylesterases, lipases, cutinases, and proteases are present (Ahsan et al. 2023).

(c) PLA has been extensively studied for its biocompatibility and ability to degrade into non-toxic components. Notably, the Food and Drug Administration (FDA) has granted approval for the implantation of PLA in the human body (Zhang et al. 2006). The incorporation of PLA in various applications, such as medical implants and drug-delivery systems, has been explored due to its ability to undergo scission in the body. This process results in the formation of monomeric units of lactic acid, which serves as a natural interme-

diate in carbohydrate metabolism. The hydrolysis of PLA has been observed to occur in vivo, and interestingly, it does not appear to rely on the presence of enzymes for catalysis (Schwach and Vert 1999). The degradation rate of polymers is influenced by various factors, including the size and shape of the polymer, the ratio of isomers present, the hydrolysis temperature, the presence of low-molecular weight impurities, and the concentration of catalysts. According to the existing data, the duration of degradation has been observed to range from 1 to 2 years. The degradation of PLA results in the production of lactic acid, a compound that is naturally generated during metabolic processes (Chen et al. 2007; Gupta et al. 2007; Papageorgiou et al. 2010).

(d) Processability: The primary conversion methodologies employed for PLA revolve around melt processing techniques. According to research, it has been observed that commercial grades of PLA can be effectively processed using a conventional twin-screw extruder. The melt viscosities of high molecular weight PLA have been found to range from 500 to 1000 Pa.s at shear rates of 10 to 50 s^{-1}. These findings suggest that PLA can be successfully utilized in various industrial applications using established processing techniques (Auras et al. 2004). Nature Works offers a range of PLA grades that are customized for different processing methods and applications, including extrusion, thermoforming, injection stretch blow molding, film production, fiber production, and foam production (Avinc and Khoddami 2009). Nevertheless, the primary limitation for processing in a molten state is the thermal degradation of the material. The thermal degradation of PLA is commonly associated with various chemical processes, including hydrolysis, depolymerization, oxidative random-chain scission, as well as inter and intramolecular transesterification. These processes lead to the production of lactide monomers and oligomers. The early reduction in molecular weight is significantly influenced by the presence of residual monomer and traces of water. Hence, the proper drying of PLA pellets prior to processing is considered to be of significant importance in various research studies (Najafi et al. 2012; Murariu and Dubois 2016).

The aforementioned characteristics provide PLA a viable and environmentally friendly substitute for petrochemical-based synthetic polymers in the context of packaging materials. Nevertheless, commercial PLA also possesses certain limitations that hinder its current application in the field of food packaging (Muller et al. 2017; Rasal et al. 2010; Krishnan et al. 2016).

- One such drawback is its inherent brittleness, which restricts its suitability for flexible films, sheets, or injected parts requiring high impact strength. Consequently, this characteristic also limits the range of processes that can be employed, primarily focusing on blown-film and thermoforming techniques.
- The thermomechanical resistance of the material is limited by its poor crystallization behavior.
- The unsuitability of PLA for hot filling liquid food packaging can be attributed to its high hydrolysis rate and low thermal resistance.
- The low gas barrier properties of the material make it susceptible to reactions or damage from O_2, CO_2, or H_2O, which can affect the quality of the food.

Numerous research endeavors have been focused on addressing these limitations through various approaches, including the incorporation of natural additives and plasticizers, copolymerization, and polymer blending. The enhancement of thermomechanical and barrier properties of PLA through the integration of nano-fillers into its matrix has been documented in previous studies (Marra et al. 2016; Swetha et al. 2023; Yang et al. 2020; Huang et al. 2022). Therefore, the nanocomposites based on PLA show great potential as biodegradable materials suitable for use in the packaging sector.

Nano fillers used for food packaging applications

Bio-nanocomposites typically include a polymer matrix integrated with a nanofiller. Fillers may be classified into two types: organic and inorganic. However, the efficiency of nanofillers is contingent upon many aspects. This encompasses many geometrical features, such as size, form, and aspect ratio, as well as mechanical parameters, such as ultimate tensile strength, elastic modulus, and degree of dispersion

etc. (Lopez De Dicastillo et al. 2020). In addition to its role in enhancing structural integrity, nanomaterials may also fulfil several additional functions, such as incorporating active or intelligent attributes, such as antimicrobial capabilities and biosensing capabilities (Priyadarshi et al. 2022). Figure 4 provides a comprehensive list of the nanofillers often used in food packaging applications.

Nanofillers used in food packaging may be broadly categorized according to the specific raw materials employed, including both organic sources (such as cellulose whiskers, lignocellulosic materials, chitin whiskers, and starch nanocrystals) and inorganic nanofillers. Inorganic nanofillers include a range of materials, including metallic nanoparticles such as gold (Au) and silver (Ag), metal oxides including magnesium oxide (MgO), titanium dioxide (TiO_2), copper oxide (CuO), and iron oxide (Fe_3O_4), as well as carbon nanotubes and layered silicates such as montmorillonite (MMT) clays, hectorite, and Saporite (Olayil et al. 2022; Wu et al. 2022).

However, it has been observed that a significant portion of these nanofillers lack biocompatibility and/or biodegradability properties. Researchers have focused on PLA-based "green composites" or "biocomposites" made with biodegradable, reusable, and nontoxic nanofillers (Zubair and Ullah 2020). Cellulose, due to its abundant presence in nature, non-toxic properties, and biocompatibility, has gained recognition as a potential biofiller candidate for the development of PLA composites.

These composites have various applications, including food packaging and biomedical purposes (Nazrin et al. 2020). Cellulose-based nanoparticles have been extensively studied and have shown promise in various applications. These nanoparticles possess supramolecular structures, which are organized on a molecular level and contribute to their unique properties. The diameters of these nanoparticles typically range from a few to several tens of nanometers, allowing for precise control over their size and morphology. This level of specificity in their structural characteristics enables tailored functionality and enhanced performance in a wide range of fields (Ahankari et al. 2021; Silva et al. 2020). The classification of cellulose materials can be based on their types and dimensions, which include cellulose nanocrystals (CNCs), cellulose nanofibers (CNFs), and bacterial cellulose (BC). The acquisition of these nanoparticles can be achieved using diverse methodologies from a range of cellulose sources. In addition to their recognized environmental advantages, cellulose nanoparticles exhibit notable characteristics such as a high aspect ratio, substantial surface area, low specific density, and superior strength and modulus when compared to conventional fibers. In recent years, there has been a significant increase in scientific interest surrounding the development of PLA/nanocellulose bionanocomposites, following the commercialization of nanocellulose products (Poulose et al. 2022; Saallah et al. 2020; Kumar et al. 2021).

Fig. 4 List of nanofillers used in the packaging applications (Olayil et al. 2022)
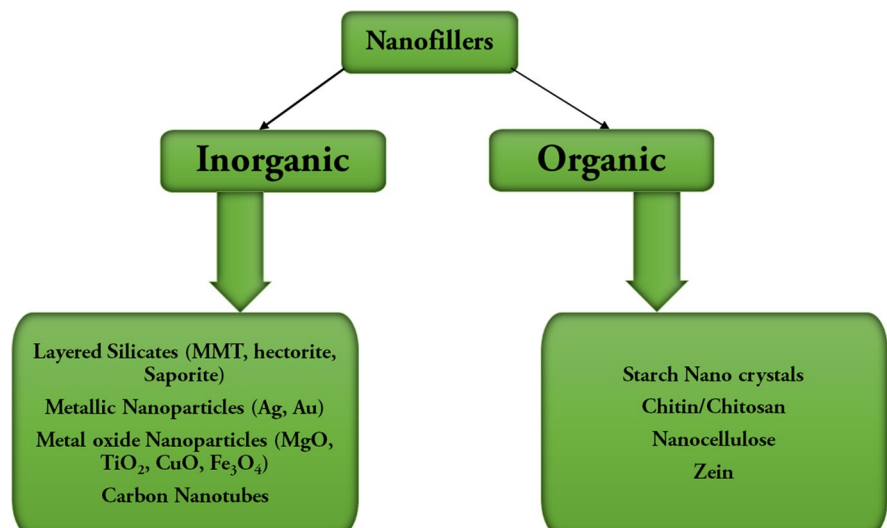
Nanocellulose: history and features

Lignocellulose biomass

Lignocellulosic biomass encompasses a diverse range of natural organic materials, primarily derived from plants or plant-based sources. This category represents the largest quantity of sustainable carbon materials and holds significant potential as a feedstock for the sustainable production of biochemicals, bioethanol, and biofuels (Lee et al. 2014). Lignocellulosic biomass is a well-known and widely studied source of natural fiber. Its potential as a substitute for petroleum-based polymers has been extensively investigated, primarily due to its remarkable environmentally friendly properties. In addition, it is worth noting that biomass waste, including agricultural residues and forest remnants, holds significant promise for repurposing as a source of fuel or raw material for the creation of valuable products. Importantly, this potential does not interfere with the availability of resources for human and animal consumption (Pradhan et al. 2022; Phanthong et al. 2018).

The cellular composition of lignocellulosic biomass mostly comprises three distinct polymers, namely lignin, hemicellulose, and cellulose. Nevertheless, the composition and quantity of these three components exhibit significant variation as a result of the diversity in species, kinds, and sources of lignocellulosic biomass (Nanda et al. 2015). Lignin comprises about 10–25% of the total weight of dry lignocellulosic biomass. Lignin functions as a binding agent that links cellulose and hemicellulose complexes inside and around plant cell walls. Lignin, by its binding function, imparts rigidity, compressive strength, resistance to degradation, and impermeability to water in the cell walls of plants. Hemicellulose comprises about 20–35% of lignocellulosic biomass (Yoo et al. 2020). Hemicellulose is a heteropolymer consisting of many types of monomers, including pentoses and hexoses, arranged in short, linear, and branching chains. Xylans and glucomannans are recognized as prevalent forms of hemicellulose. Hardwood is known for its plentiful composition of xylans, while softwood mostly contains glucomannans. Hemicellulose exhibits adhesion to cellulose fibrils through hydrogen bonding and Van der Waals interactions (Luo et al. 2019).

Cellulose is the major constituent of lignocellulosic biomass, mostly found inside the plant cell wall, accounting for around 35–50% (Bajpai 2016; Demirbas 2005). Cellulose is a linear polymer made up of D-glucopyranose units connected by β-1,4-glycosidic linkages. It is often biosynthesized into cellulose I, a metastable form made up of thin, rod-like crystalline microfibrils. These microfibrils consist of a combination of two crystalline allomorphs, known as cellulose Iα and cellulose Iβ, in certain compositional ratios. Cellulose I may undergo an irreversible transformation into a stable crystalline structure known as cellulose II via two separate processes: regeneration and mercerization. During the allomorphic change from cellulose I to cellulose II, the molecular chains connecting neighbouring cellulose microfibrils are thoroughly mixed and reorganized to generate an antiparallel chain structure. When ammonia-cellulose I or ammonia-cellulose II (a complex of ammonia and cellulose) decomposes, it produces Cellulose III, a frequently occurring form of cellulose. It is referred to as III_I when the chains are parallel and III_{II} when the chains are antiparallel. These allomorphs vary in terms of their relative chain direction, chain stacking, and unit-cell properties. They also differ in the orientation of hydroxy- and hydroxymethyl groups, as well as the corresponding hydrogen-bonding networks (Chen et al. 2016; Wu et al. 2020; Nishiyama et al. 2002; Wada et al. 2004).

Cellulose I exhibits a *tg (trans-gauche)* conformation at C6, enabling the positioning of O6 adjacent to the glycosidic linkage. It can form a hydrogen bond with the oxygen atom (O2) of the previous glucosyl unit in the same chain and can also donate a hydrogen bond to the oxygen atom (O3) of the adjacent chain. The O3 atom acts as the donor for a hydrogen bond with the oxygen ring on the preceding residue in the same chain (Nishiyama 2009; Jarvis 2022). There is some disorder in the O6 and O2 hydrogen bonding, and some hydrogen bonds have two acceptor oxygens present at once. Cellulose II exhibits antiparallel chains, resulting in a *gt (gauche-trans)* conformation for C6 (Wohlert et al. 2022; Jarvis 2022). Consequently, the distance between O6 and O2 is too great to facilitate hydrogen bonding. The hydrogen bond O3'H–O5 is a common structural motif found in dry cellulose and hemicelluloses, which have a flat-ribbon 2_1 helical conformation. The three-dimensional hydrogen-bonding network in cellulose III_I is more

cooperative than the two-dimensional cellulose Iα and Iβ due to its ability to span multiple sheets (Lindman et al. 2010; Habibi et al. 2010; Wohlert et al. 2022, Chen et al.2016; Jarvis 2022; Wada et al. 2004; Langan et al. 2001; Ford et al. 2005).

Nanocellulose

Cellulose fiber is characterized by its exceptional physical and mechanical qualities, which may be attributed to the presence of many hydroxy groups and an effective hydrogen bonding network (Wang et al. 2016). Nevertheless, it has been shown that cellulose fibrils typically form aggregates with both a crystalline structure, characterized by high order, and an amorphous structure, lacking a distinct order. In the crystalline regions, the chain molecules exhibit a highly organized arrangement, hence facilitating enhanced rigidity and robustness of cellulose. The amorphous components of the bulk material contribute to its overall flexibility. Cellulose fibrils, found within the composite structure of ordered and disordered areas, often exhibit diameters ranging from 3 to 100 nm and lengths between 1 and 4 μ m in the case of common lignocellulosic biomass (Bangar and Whiteside 2021; Isogai 2021).

Nanocellulose is extracted from numerous sources of cellulose. The main area of interest is in the dimensions of nanocellulose fibers, which typically exhibit a diameter of less than 100 nm and a length of several micrometers (Isogai 2021). Nanocellulose has remarkable strength properties, in addition to being lightweight and having a low density of around 1.6 g/ cm^3. Furthermore, it is biodegradable in nature. In particular, the material exhibits a very high rigidity, with an elastic modulus of 220 GPa, surpassing that of Kevlar fiber. Furthermore, it is worth noting that nanocellulose has a remarkable tensile strength of up to 10 GPa, surpassing that of cast iron (Mondal 2017). Additionally, its strength-to-weight ratio is eight times larger than that of stainless steel. Furthermore, nanocellulose exhibits transparency and has a high density of hydroxy groups on its surface, making it highly reactive. These hydroxy groups may be modified to impart a wide range of surface characteristics (Dufresne 2019).

Nanocellulose can be classified into three primary categories: cellulose nanocrystals (CNCs), cellulose nano fibers (CNFs), and bacterial nanocellulose

(BNC). The various types under consideration exhibit similarities in terms of their chemical composition. However, they display differences in morphology, particle size, crystallinity, and certain properties. These variations arise from disparities in their sources and the methods employed for extraction (Qi et al. 2023; Randhawa et al. 2022; Dhali et al. 2021).

Cellulose nanocrystals (CNCs)

CNCs are produced through acid hydrolysis or heat-controlled methods using sulfuric acid. This process prevents aggregation in aqueous suspensions due to electrostatic repulsion between cellulose particles. Acid treatment removes noncellulose components and most amorphous cellulose parts, producing pure CNCs. Almost 100% of CNCs have crystalline regions (around 54%–88% high crystallinity) with a rod-like shape or whisker shape, with diameters ranging from 2 to 20 nm and lengths ranging from 100 to 500 nm (Akatan et al. 2022; Lavoine et al. 2012). Figure 5 depicts the schematic representation of cellulose nanocrystal, which is obtained by the process of acid hydrolysis from cellulose fibrils. The hydrolysis and subsequent removal of the amorphous components is achieved by the use of acid, while the crystalline components remain unaffected. The extraction technique used in this study leads to the inference that cellulose nanocrystal has a high degree of crystallinity and possesses a morphology that includes short rod-like structures (Dufresne 2019; Grishkewich et al. 2017).

Cellulose nanofibers (CNFs)

Cellulose nanofibers, also referred to as cellulose nanofibril or nanofibrillar cellulose, is a type of nanocellulose that is characterized by its elongated structure, flexibility, and intricate entanglement. This unique material can be obtained through mechanical extraction techniques from cellulose fibers. The observed structures exhibit elongated fibril morphology, characterized by diameters ranging from 1 to 100 nm and lengths spanning from 500 to 2000 nm. The composition of the substance under consideration includes a cellulose content of 100%, comprising both crystalline and amorphous regions (Nagarajan et al. 2021; Li et al. 2021). Figure 6 illustrates the schematic of CNFs, which can be separated from
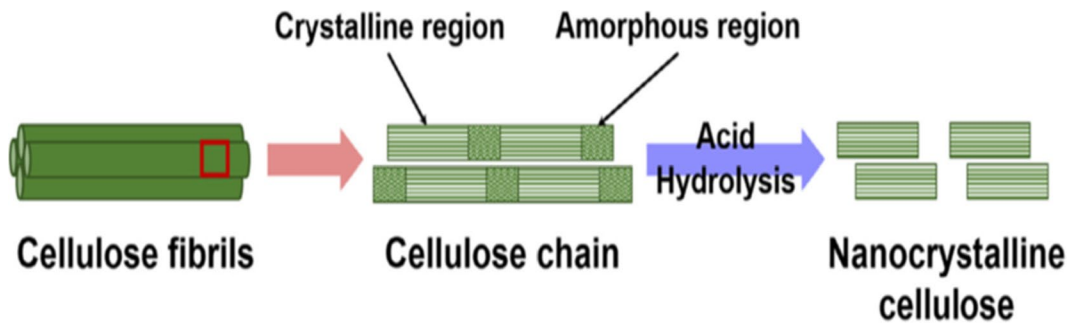
Fig. 5 Schematic representation of the extraction of cellulose nanocrystal from cellulose fibrils via acid hydrolysis (Phanthong et al. 2018)
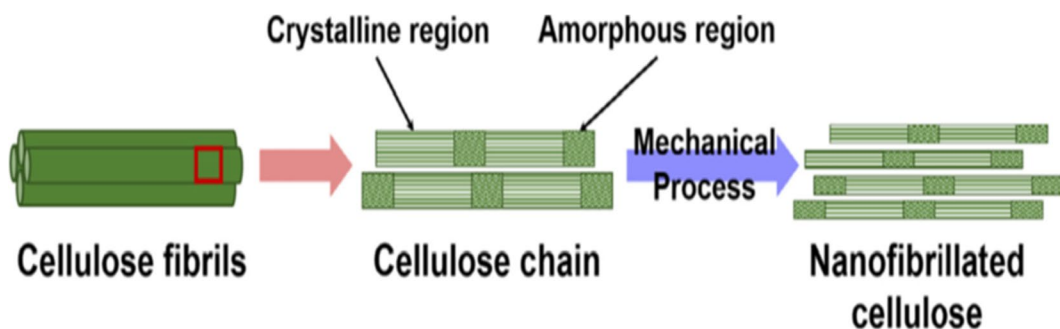

Fig. 6 Schematic representation of extraction of CNFs from cellulose fibrils via mechanical cleavage (Phanthong et al. 2018)

cellulose chains by forcing fibrils to split along their longitudinal axis using a mechanical technique. When compared to CNCs, CNFs is longer, has a higher surface area, length to diameter ratio, and a higher widespread hydroxyl group content, making it more readily modifiable on the surface. (Abitbol et al. 2016; Prakash Menon et al. 2017; Phanthong et al. 2018).

Bacterial nanocellulose (BNC)

There exists an alternative kind of nanocellulose that distinguishes itself from CNCs and CNFs. CNCs and CNFs can be derived from lignocellulosic biomass through a top-down process. In contrast, bacterial nanocellulose is synthesised through a bottom-up process, wherein bacteria, primarily *Gluconacetobacter Xylinus*, construct it by accumulating low molecular weight sugars over a period ranging from a few days to two weeks. Unlike cellulose derived from plants, bacterial cellulose does

not contain any additional compounds found in plant pulp or derived from animals, hence different chemical procedures are not required to obtain pure cellulose. But bacteria are present in the untreated bacterial cellulose membrane. It is necessary to eliminate these cell remnants as well as additional contaminants such salts, organic acids, and residual sugars from the culture media. While several techniques are utilized, including centrifugation, filtering, chemical extraction, and washing, the most popular procedure is washing bacterial cellulose in a hot NaOH solution (Wan et al. 2019). Bacterial nanocellulose has identical chemical compositions to two other types of nanocelluloses. The structure consists of twisted ribbons characterized by typical diameters ranging from 20 to 100 nm and micrometer-scale lengths, resulting in a significant surface area per unit (De Oliveira Barud et al. 2020). Figure 7 displays a visual representation of the comparative microscopic images of CNCs, CNFs, and BNC.
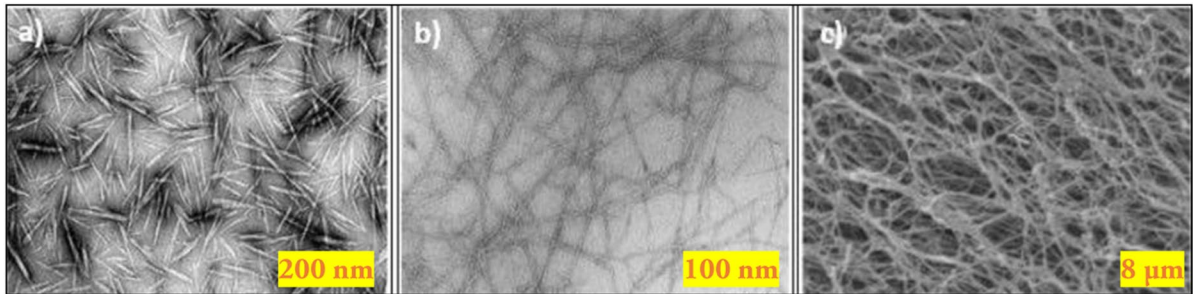
Fig. 7 Comparison microscopic image of **a** cellulose nanocrystals (CNCs), **b** cellulose nanofibers (CNFs), **c** bacterial nanocellulose (BNC) (Nazrin et al. 2020)

Nanocellulose extraction from lignocellulosic biomass

The exploration of nanocellulose extraction from lignocellulosic biomass is very appealing owing to the remarkable characteristics of nanocellulose and its potential for future uses. This is especially important for the extraction process from agricultural residue. In most cases, the pre-treatment process is used to eliminate non-cellulosic constituents, including lignin, hemicellulose, and other substances (Yu et al. 2021). Various extraction procedures are used to extract nanocellulose from cellulose fibrils and are the following:

Biomass pre-treatment

In recent years, there is a growing interest in using agricultural wastes as a viable resource for the production of nanocellulose. Not only is nanocellulose readily accessible in nature, but the use of agricultural leftovers may enhance the conversion of non-valuable wastes into very profitable nanocellulose products (Gupta and Shukla 2020). Furthermore, the effective use of agricultural waste is beneficial for the environment. As previously mentioned, lignocellulosic biomass is composed of cellulose along with non-cellulosic components like lignin, hemicellulose, and several other chemicals. The pre-treatment of biomass is a crucial stage in the process of eliminating non-cellulosic constituents and isolating cellulosic materials for subsequent nanocellulose extraction (Lunardi et al. 2021).

There are two commonly used procedures for biomass pre-treatment, namely acid chlorite treatment

and alkaline treatment (Phanthong et al. 2018). The acid-chlorite treatment also referred to as the delignification process or bleaching process, is extensively used in pulp businesses. The removal of lignin and other components from lignocellulosic biomass may be achieved using a process involving the mixing of distilled water, sodium chlorite, and acetic acid. This process entails stirring the biomass at temperatures at 70 to 80 ^{0}C for 4 to 12 h. Acetic acid and sodium chlorite are introduced into the mixes periodically, specifically at hourly intervals, in order to regulate the pH level. Subsequently, the mixture is subjected to continuous stirring for one night, after which it is subjected to a series of washes with distilled water until a state of neutral pH is achieved. The solid products that were produced were collected and subjected to a drying process in an oven at a temperature of 50 ^{0}C. This resulting material is referred to as holocellulose, mostly consisting of hemicellulose and cellulose present inside the fibers. The presence of a white-colored holocellulose fiber signifies the effective elimination of lignin and other contaminants (Phanthong et al. 2015; Hubbell and Ragauskas 2010).

The alkaline treatment involves the use of alkaline substances to eliminate the amorphous polymer of hemicellulose and residual lignin. Sodium hydroxide (4 to 20 wt.%) is commonly employed as the alkaline agent in various applications. In these processes, it is typically mixed with holocellulose and subjected to stirring for 1–5 h. The solid products obtained from the experiment were subjected to a washing process using distilled water until a neutral pH was achieved. Subsequently, the washed solids were dried in an oven set at a temperature of 50 ^{0}C. The fiber products obtained from this treatment primarily consist of cellulose, with

the removal of other non-cellulosic materials (Sharma et al. 2021; Alemdar and Sain 2008).

Extraction of nanocellulose

Numerous methods have been devised for the isolation of nanocellulose from cellulosic substrates. The use of various extraction techniques led to variations in the characteristics and attributes of the acquired nanocellulose. This section categorizes the primary extraction methods into three techniques: chemical treatment, enzymatic hydrolysis, and mechanical processes.

Acid hydrolysis

Acid hydrolysis is a primary method used in the isolation of nanocellulose from cellulosic substances. The susceptibility of cellulose chains to acid hydrolysis is attributed to the presence of both ordered and disordered areas within the chains. Acid hydrolysis preferentially targets the disordered regions, leaving the ordered sections intact. Sulfuric acid is the primary acid used for acid hydrolysis, as shown by previous studies (Akatan et al. 2022; Bondeson et al. 2006; Bacha 2022). The process of hydrolyzing cellulose pulp with sulfuric acid is heterogeneous, since the acid permeates the pulp fiber and breaks the glycosidic linkages inside the cellulose polymer. Hydrolysis may also happen on the crystalline areas depending on reaction timings, and some of the hydroxyl groups on the crystalline surface will change into sulfate groups (e.g., cellulose-OH to cellulose-$OSO_3^-H^+$). The characteristics of nanocellulose produced are primarily influenced by key regulating parameters, including reaction time, temperature, and acid concentration (Borjesson and Westman 2015; Peng et al. 2011). One major disadvantage of acid hydrolysis is the production of acid wastewater during the washing procedure required to neutralize the pH value of the nanocellulose solution. Typically, the washing procedure is the addition of cold water, followed by centrifugation until a neutral pH level is attained. An alternative approach to washing the acquired substances involves the use of alkaline agents, such as sodium hydroxide, to achieve pH neutralization (Johar et al. 2012).

TEMPO (2,2,6,6-Tetramethylpiperidine-1-oxyl radical) oxidation

The use of TEMPO as a catalyst, in conjunction with hypochlorite as the major oxidant, enables the oxidation of the hydroxy group present in cellulose to form carboxylates. Consequently, the CNFs acquired have an average diameter of roughly 3–4 nm and a length of a few microns, with a surface composed of carboxylic acid groups (Levanic et al. 2020). The TEMPO-mediated oxidation process is achieved by the use of TEMPO/NaBr/NaClO in an aqueous solution with an elevated pH level. This method involves dissolving TEMPO and NaBr in water, followed by the initiation of oxidation with the addition of NaClO. Additionally, an alternate oxidation system known as TEMPO/NaClO/$NaClO_2$ may also be used in this procedure, particularly in neutral or slightly acidic conditions (Isogai et al. 2011). TEMPO-oxidized cellulose nanofibers consistently exhibit a consistent width (3 to 4 nm), with a high aspect ratio. These nanofibers have the potential to be used in several applications, such as transparent and flexible displays, gas-barrier films for packaging, and nanofiber fillers in composite materials (Fukuzumi et al. 2009).

Enzymatic hydrolysis

It is a biological process that uses enzymes to digest/modify cellulosic fibers into nanocellulose. Microbial processing is used to facilitate the degradation of hemicellulose and lignin, using enzymes such as cellobiohydrolase and endoglucanase (Winuprasith and Suphantharika 2015). Cellobiohydrolase has enzymatic activity towards crystalline cellulose, while endoglucanase demonstrates enzymatic activity towards amorphous cellulose. The use of enzymatic processes for cellulose production has been proposed as an environmentally sustainable approach. Numerous studies have extensively demonstrated the phenomenon of synergistic interactions between enzymes in relation to the process of surface modification of cellulosic biomass. According to reports, enzymatic hydrolysis in conjunction with homogenization techniques on softwood wood have been found to produce nanocellulose with a higher aspect ratio compared to acidic treatment (Phanthong et al. 2018). Enzymatic hydrolysis involves the catalytic activity of enzymes, which is facilitated by hydrogen bonding

between cellulose fibers. Furthermore, the hydrolysis of hemicellulose has significant importance for many reasons. Firstly, it serves as a preventive measure against cellulosic hydrolysis. Secondly, it facilitates the production of monosaccharides that may be used for the process of fermentation, ultimately leading to the production of bioethanol (Wahlstrom and Suurnakki 2015). The effective hydrolysis of cellulosic aggregates necessitates the association of cellulases and hemicellulases. In general, the enzymatic biological treatment may be conducted under modest working conditions; nevertheless, it requires a prolonged duration of action. In order to address this issue, the process of enzymatic hydrolysis is consistently integrated with complementary methodologies (Moniruzzaman and Ono 2013). The study conducted by Moniruzzaman and Ono (2013) focused on investigating the process of separating cellulose fibers from wood chips. This was achieved by a pre-treatment method including the use of an ionic liquid, which aimed to enhance the accessible surface area of the cellulose fibers. Subsequently, enzymatic hydrolysis was carried out using laccase. The nanocellulose that was synthesized exhibited a greater degree of crystallinity and thermal stability in comparison to the original wood fibers (Moniruzzaman and Ono 2013).

Mechanical process

The mechanical process involves extracting cellulose fibrils with significant shear force, causing cellulose fiber cleavage along their longitudinal axis, resulting in the formation of CNFs. The mechanical techniques that are often used include high pressure homogenization, ultrasonication, and ball milling processes (Abdul et al. 2012). The process of high-pressure homogenization (HPH) involves the passage of cellulose slurry into a vessel with both high pressure and high velocity. The generation of impact force and shear force inside a fluid medium facilitates the cleavage of cellulose microfibrils, resulting in a reduction of their diameter to the nanometer scale (Abdul et al. 2014). Ultrasonication is a technique used to defibrillate cellulose fibers by using the hydrodynamic forces generated by ultrasound. This process generates mechanical oscillating power, which leads to the creation, enlargement, and collapse of microscopic gas bubbles as the liquid molecules acquire ultrasonic energy (Dufresne 2019). Ball milling is another

mechanical method that has been widely employed for the defibrillation of cellulose fibers (Barakat et al. 2014). It significantly affects the structure, morphology, crystallinity, and thermal stability of the materials and is used to grind native cellulose fibers, producing amorphous cellulose. This cellulose exhibits reductions in crystallinity, size, morphology, and alterations in crystalline lattice structure. The primary elements of a ball milling machine are milling balls, also known as grinding media, housed in a milling jar or milling beaker. Various types of ball millers are widely used in industrial and laboratory settings, such as the planetary ball mill, mixer ball mill, and vibration ball mill. The planetary ball mill is a mechanical process that involves the collision of balls and friction between the balls and the wall of a rotating milling jar. The characteristics of ball-milled products are influenced by factors such as the quantity and size of balls, the speed of milling, the state of milling (dry or wet), the weight ratio between balls and materials, and the duration of milling (Phanthong et al. 2018; Piras et al.2019). The list of various biomass sources of nanocellulose, their isolation procedures, and their dimensions are shown in Table 2.

PLA/ nanocellulose composite films for food packaging

PLA is becoming more prevalent as a packaging polymer for a wide range of fresh food items, including fruits and vegetables. It is often used in the form of films, containers, and coatings. Despite considerable research endeavors, the thermal, mechanical, and barrier characteristics of PLA remain inadequate for some rigorous applications in the field of food packaging (Marano et al. 2022; Farah et al. 2016). Nanocellulose, when incorporated into PLA biocomposites, significantly improves tensile strength and elastic modulus. Its compact network formation in composites is advantageous due to robust hydrogen bonding, which significantly impedes molecule passage, making it a valuable filler for barrier applications (Vatansever et al. 2019). Nevertheless, the use of nanocellulose in hydrophobic polymers, such as PLA, is hindered by its high polarity and inadequate compatibility, resulting in diminished moisture resistance and suboptimal mechanical characteristics. Various surface modification techniques, including monomer

Table 2 List of various biomass sources of nanocellulose, their isolation procedures, and their dimensions

Biomass Source	Isolation Procedure	Shape of nanoparticles	Dimension	References
Paraguayan Residual Agro-Industrial Biomass	Acid hydrolysis	Whisker-like	Diameter of 230 ± 42 nm and a height of 12 ± 2 nm	(Velazquez et al. 2022)
Miscanthus (*M. sacchariflorus* and *M. sinensis*) and Sorghum (*S. saccharatum* and *S. bicolor*)	Acid Hydrolysis	Spherical	Diameter of 27 to 54 nm	(Babicka et al. 2022)
Husks of short, medium and long rice grains	Acid hydrolysis	Nanowhiskers	Diameters of 11.7–28.9 nm	(Rashid and Dutta 2020)
Pistachio shell	Acid hydrolysis	Spherical	Diameter of 68.8 ± 20.7 nm	(Kasiri and Fathi 2018)
Vietnamese agricultural wastes (nypa fruticans trunk, coconut husk fiber, and rice husk)	Acid hydrolysis	Nanorod	Lengths of 200–500 nm and widths of 10–15 nm	(Nang An et al. 2020)
Brazilian satintail (*Imperata Brasiliensis*) plant	Acid hydrolysis	Nanorod	Diameter of 10 to 60 nm and length of 150 to 250 nm	(Benini et al. 2018)
Wheat straw and soy hulls	Mechanical process	Nanofibre	Diameters of 10–80 nm and lengths of a few thousand nanometers	(Alemdar and Sain 2008)
Tomato peels	Acid hydrolysis	Nanorod	Diameter 7.2 nm and length almost 135 nm	(Jiang and Hsieh 2015)
Pine cones	Mechanical process	Nanofibre	Diameter almost 15 nm	(Rambabu et al. 2016)

grafting, silylation, surfactant treatment, acetylation, and esterification, have been used to reduce the hydrophilicity of nanocellulose in order to enhance its dispersion in PLA (Song et al. 2014; Raquez et al. 2012; Fortunati et al. 2012; Jonoobi et al. 2012; Lee et al. 2009).

Properties of PLA/nanocellulose composites

Morphological properties

Atomic force microscopy (AFM) and Scanning electron microscopy (SEM) are widely used techniques for visualizing the morphology and microstructure of materials (Falsafi et al. 2020). SEM works by scanning the surface of a sample with a beam of electrons with a relatively low energy. Photons and electrons are emitted from or near the sample surface as the beam approaches and penetrates the material (Akhtar et al. 2018).

Frone et al. (2013) reported on morphological investigations pertaining to the fabrication of bionanocomposites composed of PLA/CNC. The morphology of both pure PLA and nanocomposites was assessed using a novel and advanced AFM method

known as Peak Force QNM (Quantitative Mechanical Property Mapping at the Nanoscale). This approach effectively highlighted the nanoscale features by elastic modulus mapping. Figure 8a-b show typical topography views of PLA composites including cellulose nanofibers, both with and without silane treatment. The analysis of these data revealed a more pronounced dispersion in the PLA composite with silane-treated CNFs. The distribution of CNFs on the surface of the PLA/CNFs composite was found to be more uniform, with fewer instances of fiber agglomeration seen in Fig. 8b in comparison to Fig. 8a. The majority of CNF agglomerates were situated towards the surface of the material, in contrast to CNFs, which were found deeper into the polymer. This observation implies that the PLA/CNFs exhibited a more favourable interface between the cellulose fibers and the matrix (Frone et al. 2013).

Herrera et al. (2015) reported the use of SEM analysis in their study on the fabrication of PLA/CNF bionanocomposites. The brittleness of the fracture surface of neat PLA was evident, as seen in Fig. 9a. This was due to the consequence of the inherent fragility of PLA. It was anticipated that the incorporation of glycerol triacetate (GTA) as a plasticizer
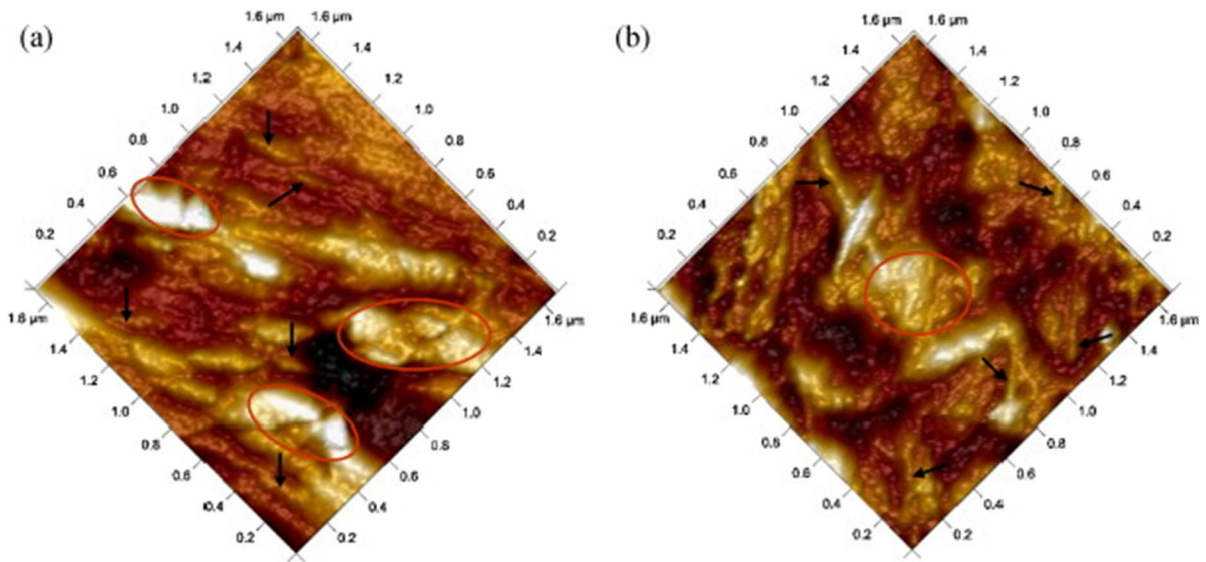
Fig. 8 AFM micrographs of **a** untreated and **b** silane treated CNF reinforced PLA composites at a scan size of 1.7 μm × 1.7 μm (Frone et al. 2013)
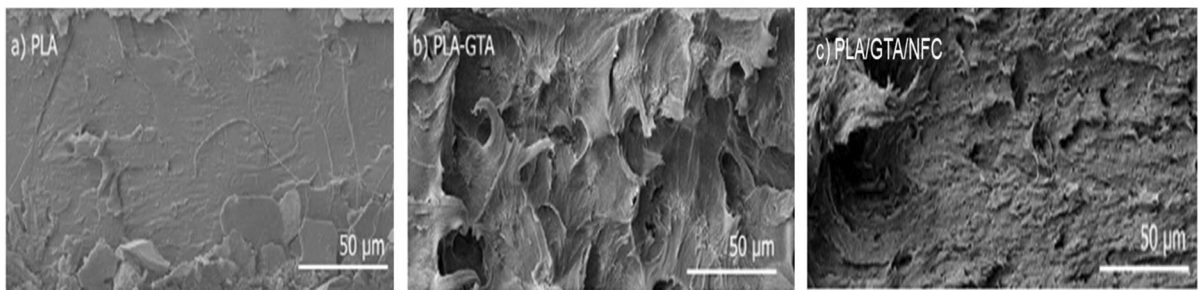

Fig. 9 Fractured surfaces morphology of **a** PLA, **b** PLA/GTA, and **c** PLA/GTA/CNF films (Herrera et al. 2015)

would result in improved dispersion and distribution of CNFs inside the matrix. The micrographs of PLA/GTA and PLA/GTA/ CNF bionanocomposites are shown in Fig. 9b and c, respectively. The fracture surface of the PLA/CNF bionanocomposites with GTA plasticizer in Fig. 9c exhibited a ductile fracture, as seen by the significant presence of crazes. The formation of crazes was subject to several influencing variables, including the level of crystallinity and the size of spherulites. The inclusion of CNFs in bionanocomposites resulted in a decrease in the available area for spherulite formation, which led to a decrease in the brittleness of the materials (Herrera et al. 2015). Fractured surfaces of PLA and PLA/CNC composites were studied by Fortunati et al. (2015). The neat PLA

film sample had a rather smooth fractured surface. In contrast, the unmodified CNC-based nanocomposites exhibited a rougher and fractured surface as can be seen in Fig. 10. This roughness indicates a greater brittleness in the CNC-based formulations compared to the pristine PLA film. Furthermore, the fractured surface of PLA/3CNC exhibits increased roughness as a result of the greater concentration of CNC in comparison to PLA/1CNC. Distinctive behavior was seen in formulations including surfactant modified CNCs, whereby the presence of the surfactant induced a porous structure. This phenomenon was more pronounced in the composite material composed of 3 wt% surfactant modified CNCs (3 s-CNC). The fracture surface of PLA/1 s-CNC exhibited

Fig. 10 FESEM investigation of fracture surfaces of PLA and PLA based nanocomposites containing 1%wt and 3%wt of unmodified (CNCs) and surfactant modified (s-CNCs) cellulose nanocrystals (Fortunati et al. 2015)
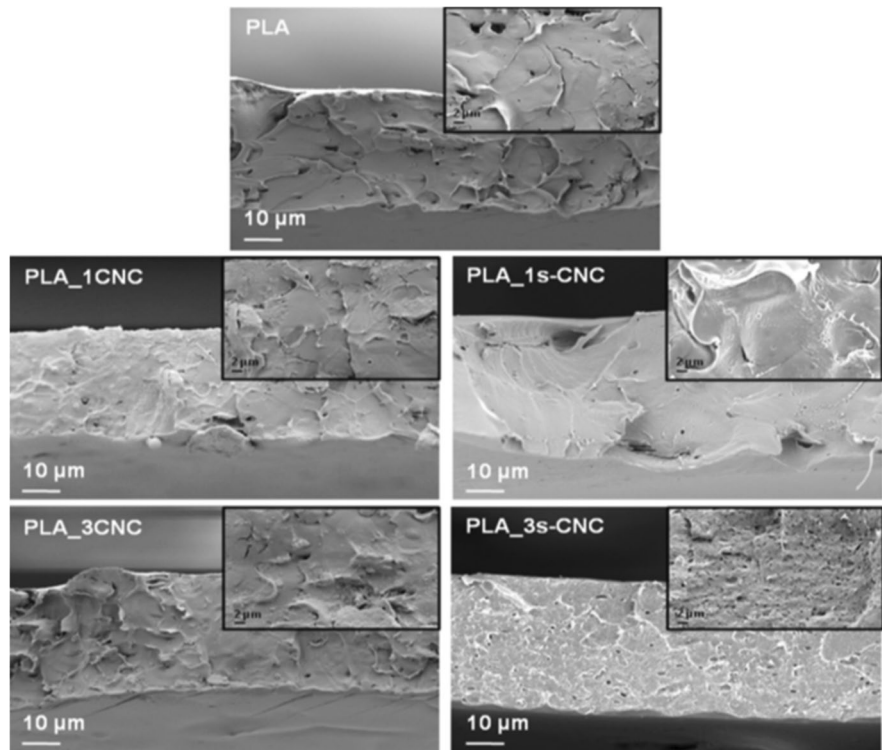
compelling evidence of the homogeneous dispersion of modified cellulose nanocrystals and plastic deformation (Fortunati et al. 2015).

The influence of several thin-film production procedures, in conjunction with varied concentrations of CNCs ranging from 1 to 5 wt%, on the characteristics of PLA films was reported by Shojaeiarani et al. (2020). The nanocomposite thin films were fabricated using solvent casting and spin-coating methods. SEM analysis demonstrated that the spin-coated films exhibited a distinct difference compared to the solvent cast samples. Specifically, the spin-coated films had a smaller size and a more uniform distribution of CNCs inside the PLA matrix (Shojaeiarani et al. 2020). This observation was especially evident when the CNCs concentration was low, at 1 wt% and 3 wt%. The primary factors influencing the prevention of microsized agglomeration development in spin-coated films are a rapid solvent evaporation rate and the synergistic effects of centrifugal force and surface tension during the spin-coating procedure. In a similar manner, nanocomposites of PLA/CNCs with CNC concentrations of 1, 3, and 5 wt% were fabricated using two methods: (a) solution casting and (b) dilution of

a solution-casted masterbatch of PLA/CNCs by melt mixing in a twin-screw extruder. This study examined the influence of the preparation procedure on the quality of CNC dispersion and, consequently, on the properties of the nanocomposites. The findings from the morphological investigations indicated that the solution casting method resulted in a more effective dispersion of CNCs within PLA. Conversely, the melt processing of nanocomposites led to the formation of CNC agglomerates, potentially due to the presence of highly robust hydrogen bonding among CNC nanoparticles (Arslan et al. 2020).

Rheological properties

The rheological characteristics of composite materials are of great importance in enhancing the processability of composites, particularly in processing operations that entail high shear rates, such as extrusion and injection molding (Ching et al. 2016). CNCs have been documented as effective rheological modifiers for various polymers due to their capacity to embed themselves between polymer chains, hence imparting notable stability to the polymer network.

The evaluation of nanocellulose crystal dispersion within a polymer matrix can be assessed by analyzing the storage modulus (G'), loss modulus (G"), and complex viscosity (η*). These parameters serve as indicators to determine the extent of dispersion achieved. The observation of higher G' and G" values suggests a uniform dispersion of CNCs within a polymer matrix, whereas the decrease in G' and G" can be explained by the agglomeration of nanocrystals within the polymer matrix (Ching et al. 2016; Mariano et al. 2016). The complex viscosity (η*) of the system can be modulated by the presence of additional particles, as elucidated by the Einstein-Batchelor theory (Moustafa et al. 2019). When assessing material processing behaviors, the melt flow index (MFI), which measures the ease with which a thermoplastic polymer can flow at a given temperature and pressure, is essential. MFI is necessary for PLA to ensure that it may be used in a variety of applications, including food packaging, which is also impacted by adding nanocellulose, while also improving the material's thermal and physical characteristics. While studying the influence of added CNCs (1.5%) on PLA, Bajwa et al. (2021) observed a decrease in MFI from 6.75to 5.31 g/10 min, which was attributed to the flow burden of CNCs within the matrix (Bajwa et al. 2021).

The identification of the rheological percolation threshold can be achieved by determining the concentration at which the network formation of cellulose nanoparticles occurs. At these concentrations,

it is expected that the long-range relaxation of polymer chains would be restricted, leading to a notable rise in both viscosity and moduli at lower frequencies (Vatansever et al. 2019). In a study conducted by Musa Kamal and co-workers, they identified several rheological features that serve as indicators of the initiation of solid-network formation. These features include: (a) a sudden reduction in the linear viscoelastic region, (b) a higher value of G' compared to G" at low frequencies, (c) an increase in complex viscosity at low frequencies, and (d) the presence of an infinite relaxation time derived from the relaxation time spectra. These findings provide valuable insights into the characteristics associated with the formation of solid networks (Khoshkava and Kamal 2014). Hence, the establishment of the network structure is contingent upon the uniform distribution of nanoparticles. Therefore, the observed rheological behaviors may serve as a potential metric for assessing the quality of nanocellulose dispersion within PLA (Dufresne 2017).

In a separate study, the incorporation of CNCs into PLA was achieved through the use of the Pickering emulsion technique. In the rheological measurements, the storage modulus (G') and complex viscosity (η*) of both neat PLA and PLA/CNC composites were investigated as a function of frequency (ω), as shown in Fig. 11. The rheological characteristics of all samples were found to be stable. The frequency dependence of the storage modulus of neat PLA exhibited characteristics commonly observed in
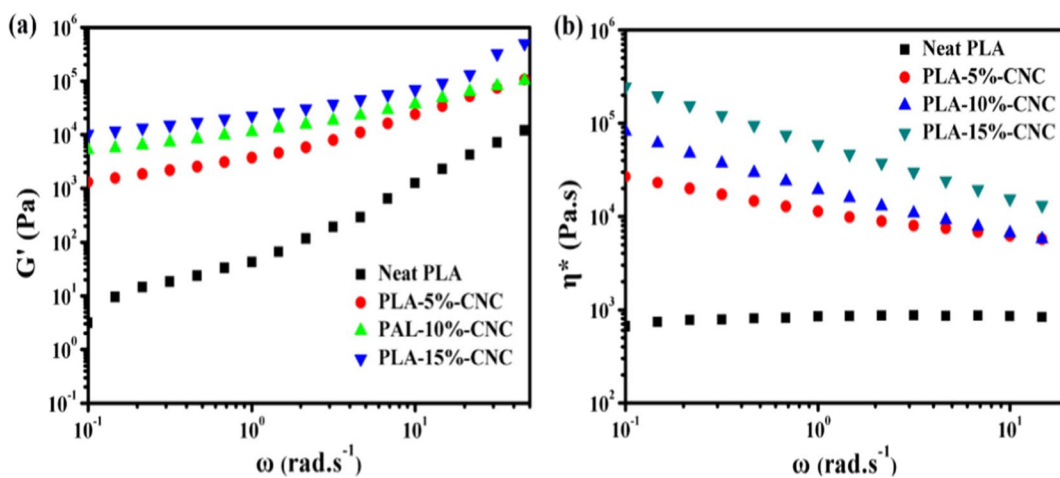

Fig. 11 Values for **a** storage modulus and **b** complex viscosity of neat PLA and the PLA/CNC composites (Zhang et al. 2019)

polymer melts, as illustrated in Fig. 11a. In contrast, it was observed that the storage modulus of the CNC composites exhibited an upward trend and eventually reached a plateau when subjected to low frequencies. The complex viscosity of pure PLA was observed to exhibit a prolonged plateau, as depicted in Fig. 11b. At higher frequencies, a minimal shear-thinning tendency was observed. In the study, it was observed that all composites displayed a clear shear-thinning behavior at low frequencies, without the presence of a plateau region (Zhang et al. 2019). The findings of this study suggest that there is a possibility for the formation of a network between CNCs and the PLA matrix. This network formation appears to facilitate the transition of the material from a fluid-like state to a solid-like behavior, particularly at elevated temperatures (Kamal and Khoshkava 2015). This behavior can be attributed to the effective dispersion of CNC within the PLA matrix. The effective dispersion of nanoparticles in PLA matrices has been identified as the cause for the reported similar rheological behavior seen in other PLA/CNC composites. The study conducted by Bagheriasl et al. (2018) found that all samples of PLA/CNC displayed consistent rheological properties, with variations of less than 2%. In the case of PLA with a 1 wt% concentration of CNC (PLA/1CNC), the values of G" and G' exhibited a significant increase of 1 and 4 orders of magnitude, respectively, when compared to pure PLA. Moreover, when the concentration of CNCs rises, there is an additional enhancement seen, with G" and G' increasing by up to 3 and 6 orders of magnitude, respectively, relative to pure PLA. The nanocomposites exhibited a notable rise in both the loss and storage moduli when the amount of CNC was increased. This effect was especially pronounced at lower frequencies, suggesting a solid-like behavior (Bagheriasl et al. 2018).

In another contribution, PLA nanocomposites with spray-and freeze-dried cellulose nanocrystals (SCNCs and FCNCs) were developed via solution casting using four different solvents. This study examined the influence of several solvent types, namely tetrahydrofuran (THF), chloroform (CHL), dimethylformamide (DMF), and dimethyl sulfoxide (DMSO), on the dispersion characteristics of SCNCs and FCNCs in PLA. The impact of the dielectric constant of solvents on the dispersion quality of CNCs was shown. The use of THF and CHL, which possess low dielectric constants, did not result in a

satisfactory dispersion of CNCs in PLA. Conversely, the application of DMF, specifically DMSO, proved to be more successful in improving the dispersion of CNCs, which was attributed to the much greater dielectric constant shown by DMF and DMSO. In the present context, the rheological percolation threshold values in PLA/SCNC nanocomposites, which were generated utilizing DMF and DMSO, were determined to be around 1.52 and 0.12 wt% CNC, respectively, using an empirical power-law equation. Furthermore, it was noted that the disparities in rheological characteristics between PLA/SCNC and PLA/FCNC nanocomposites were not significant; however, the SCNCs exhibited a somewhat more pronounced enhancement in the rheological properties of nanocomposites made with DMSO compared to FCNCs (Ozdemir and Nofar 2021). The impact of spray-dried lignin-coated cellulose nanocrystals (L-CNCs) filler on the rheological characteristics of PLA composites was investigated by Gupta et al. (2017). The rheological percolation threshold concentration for PLA/L-CNC composites was determined to be 0.66 wt% by power law analysis. The exceptional dispersion of L-CNCs inside the PLA matrix is responsible for the observed low rheological percolation concentration. The incorporation of a mere 0.5 wt% of L-CNCs into the PLA matrix yielded a significant enhancement of around 60% in the storage modulus compared to pure PLA, as determined by dynamic mechanical analysis (Gupta et al. 2017). Vatansever et al. (2020) investigated the effects of varying molecular weights and crystallisability properties of PLA on the dispersion quality and formation of percolation networks in CNCs. The findings indicated that achieving lower CNC percolation concentrations is possible by using a PLA matrix with a lower molecular weight. This was because a lower molecular weight allows for easier interpenetration of shorter chains and CNCs during their dissolution in the solvent. In contrast, it was observed that the concentration of CNC percolation was reduced even further with the utilization of PLA which exhibited higher crystallizability. In order to enhance the quality of CNC dispersion in hydrophobic polymers like PLA, it is important to consider various approaches (Vatansever et al. 2020). These include modifying the surface of CNCs, utilizing surfactants or compatibilizers, and most importantly, selecting an appropriate polymer matrix based on its

molecular structure and configuration. This selection plays a critical role in facilitating more effective dispersion of CNCs within the polymer matrix (Arias et al. 2015).

Mechanical properties

In the investigation of potential applications for bionanocomposites across diverse industries, including biomedical and packaging sectors, the evaluation of tensile properties emerges as a crucial factor to be considered (Ma et al. 2014). Tensile properties encompass various physical characteristics, including tensile strength, elongation at break, and tensile modulus. These properties play an essential role in determining the material's physical capabilities, particularly in critical and extreme conditions (Salmieri et al. 2014).

Numerous investigations have been undertaken to examine the potential impact of nanocellulose on the mechanical properties of composites from a theoretical perspective. In their study, Aitomäki and Oksman employed a micromechanical model known as the Halpin–Tsai model to assess the modulus efficiency (Aitomäki and Oksman 2014).

$$E_1 = E_m \frac{1 + \zeta \eta v_f}{1 - \eta v_f} \tag{1}$$

where, E_m and E_f is the elastic modulus of the matrix and fiber, V_f is the fiber volume fraction and f is the reinforcement efficiency and ROM model to calculate the strength of the composite.

$$\sigma_R = \eta_{Os} \eta_{ls^\sigma f} v_f + (1 - v_f) \sigma_{mf} \tag{2}$$

where, η_{os} is the orientation factor and σ_f is the fiber strength.

The authors used the given data pertaining to the elastic modulus and strength of several nano cellulosic biocomposites. Subsequently, a reinforcing efficiency factor was computed by using the Halpin–Tsai and ROM models. Both models' interpretations concluded that the nanocellulose demonstrated the greatest reinforcing factor. According to the data provided by Ramires and Dufresne, the mechanical properties of nanocomposites are influenced by three primary factors: a) dimension and morphology, b) processing technique, and c) nanostructure of the matrix and matrix/filler interface. The use of nanocellulose with a high aspect ratio and specific surface area, characterized by a rough surface and reduced fiber diameter, has the potential to enhance the adhesion between nanofillers and matrices, hence leading to improved mechanical properties (Ramires and Dufresne 2011). Several investigations on nanocellulose reinforced PLA bionanocomposites have been conducted in recent years, and tensile testing has been employed to characterize their performance.

Zabidi et al. (2022) conducted a study in which they fabricated and evaluated active and pH-sensitive films composed of PLA and CNFs. The PLA and PLA/NFC films were fabricated using the solvent casting method, including 1.5% CNFs. Various concentrations (5%, 10%, and 15%) of essential oils (EO), such as thymol and curry, were included in the films. The anthocyanin powder with a fixed amount of 1% was added to the films to serve as a pH indicator. The incorporation of CNFs resulted in a decrease in the tensile strength of films, while simultaneously enhancing their flexibility. This may be attributed to the plasticizing influence exerted by EOs. The PLA/EO and PLA/ CNFs/EO films including curry exhibited a somewhat superior strength compared to the films containing thymol. The degree of flexibility shown by films was found to be positively correlated with the concentration of EO, irrespective of the specific kind of EO used (Zabidi et al. 2022). In a study conducted by Srisawat et al. (2023), PLA/CNF composites were fabricated using a melt-compounding technique. CNFs was obtained from wood sawdust and utilized in its unmodified form. These CNFs had diameters measuring around 10 nm and lengths spanning several microns. This study examined the impact of polymeric plasticizer selection on the mechanical performance of PLA and PLA/CNF composites. In this study, the performance of poly(butylene adipate) (PBA) as a polymeric plasticizer was evaluated and compared to that of poly(ethylene glycol) (PEG), which is widely used as a plasticizer for PLA. Figure 12a presents stress–strain curves of neat PLA, plasticized PLA samples, and PLA composites incorporating CNFs. The observed PLA material demonstrated brittle failure behavior, characterized by a sudden and severe failure occurring at a relatively low level of strain. The ductility of PLA was significantly enhanced through the incorporation of polymeric plasticizers. The plasticized samples
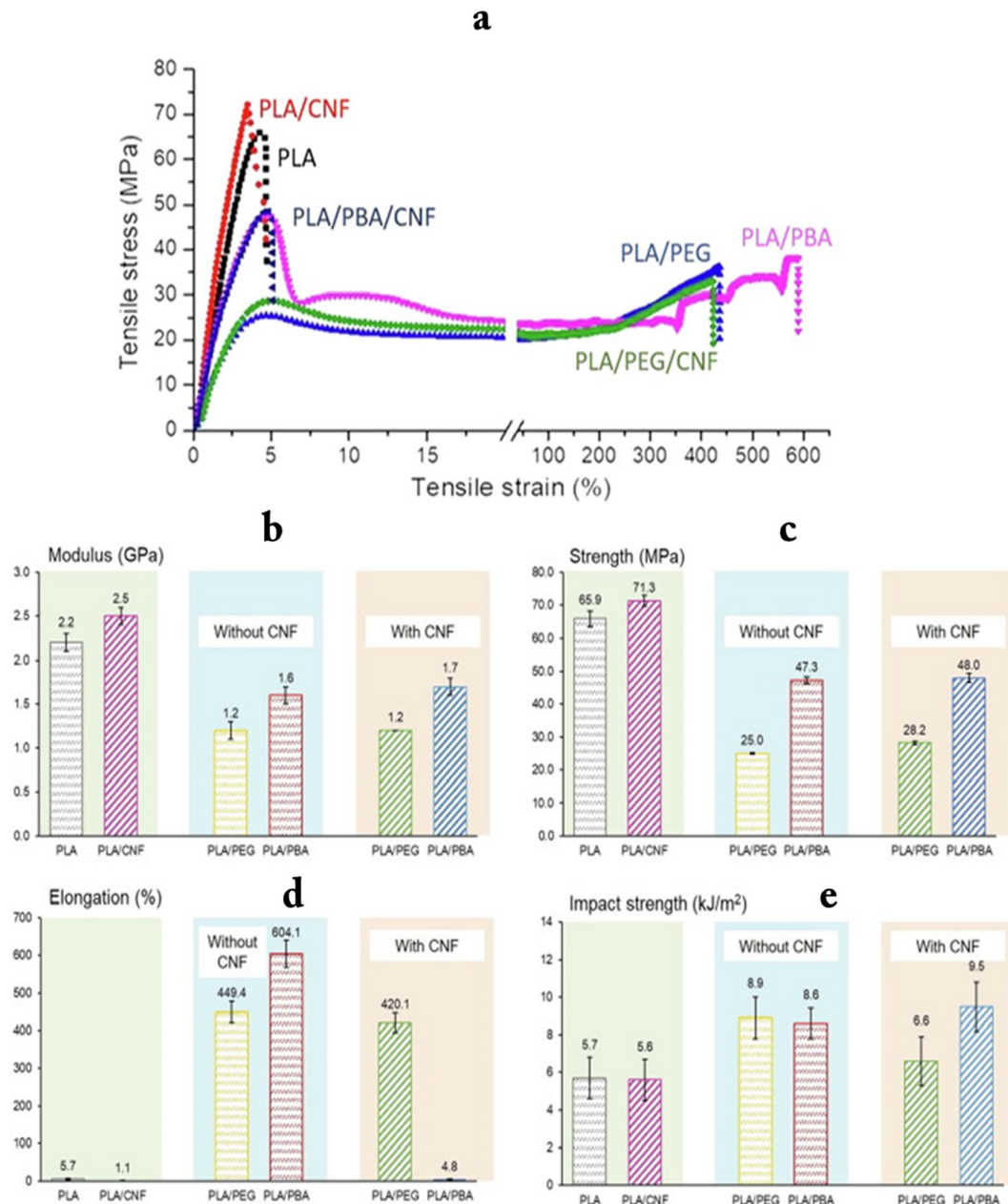
Fig. 12 **a** Stress–strain curves, **b** modulus, **c** tensile strength, **d** elongation at break, and **e** impact strength of PLA, its plasticized samples, and composites (Srisawat et al. 2023)

exhibited a notable plastic deformation response when subjected to tensile drawing, characterized by a significant elongation at break exceeding 400% (Srisawat et al. 2023). The mechanical performance of the composites was found to be influenced differently by PBA and PEG, as evidenced by the

results of the tensile and impact tests depicted in Fig. 12b-e.

In a separate study, the fabrication of PLA/cellulose-nanowhisker (CNWs) nanocomposites was conducted through a two-step process involving twin screw extrusion and subsequent injection molding.

In this study, the CNWs were uniformly dispersed within a plasticizer prior to the melt mixing process. The Young's moduli and strength of the nanocomposites were found to be higher compared to the neat matrix. This can be attributed to the reinforcing effect of the CNWs present in the nanocomposites. In contrast, it was observed that the nanocomposites exhibited a reduced elongation at break. This phenomenon can be attributed to the incorporation of fibers, which potentially restrict the mobility of polymer chains, thereby diminishing the ductility of the polymer. In contrast to the anticipated outcomes, it was observed that the nanocomposite with the lowest content of CNWs exhibited the most superior mechanical properties. The observed outcome could be ascribed to the aggregation of the CNW filler at high levels of loading (Moran et al. 2016). The effective preparation of L-lactide functionalized cellulose nanocrystals incorporating PLA (PLA/CNC-g-PLA) was achieved by the use of electrospinning technology. The enhancement of the mechanical characteristics of the composite fiber membranes was achieved by improving the molecular chain orientation using the electrospinning technique. The tensile strength and elongation at break of the composite fiber film exhibited a significant increase of 2.4 and 8.2 times, respectively, with the incorporation of 5% CNC-g-PLA (Chen et al. 2023).

In another investigation, CNCs were subjected to modifications including methacrylamide, cetyltrimethylammonium bromide, and zinc oxide. These modified CNCs were then used in the process of spray-coating the surface of PLA sheets. The stress at the yielding point of the PLA sheets was around 10 MPa, and the films experienced fracture at a strain of approximately 400%. The yield point, which denotes the movement from elastic deformation—where the material can return to its original shape—to plastic deformation—where irreversible changes occur—is essential for assessing the mechanical properties of PLA biocomposites. Understanding the yield point is critical for PLA biocomposites, including those reinforced with nanocellulose, to assess their mechanical strength and suitability for applications such as sustainable food packaging and automotive parts. Different types of PLA films with coatings showed a significant increase in yield strength (20 to 30 MPa), indicating their enhanced ability to withstand

external forces before irreversible deformation. The Young's modulus of all the coatings showed a notable improvement, with PLA-CC8 (PLA/ cetyltrimethylammonium bromide modified CNCs) and PLA-MC8 (PLA/ methacrylamide modified CNCs) demonstrating enhanced tensile strength. However, it is worth noting that only PLA-CC8 managed to preserve the ductility of the original PLA films. The observed increase in mechanical strength may be attributed to the effective integration of the surface coating with the polymeric matrix, hence serving as a supportive framework. The observed ductility of PLA-CC8 might potentially be due to the favorable interfacial compatibility, which facilitated efficient stress transmission and delayed the occurrence of stretching failure (Huang et al. 2023). In order to mitigate the inherent incompatibility between polar bacterial cellulose (BC) and nonpolar PLA, an acetylation process was used using a non-conventional pathway facilitated by citric acid as the catalyst. The derivatized BC (AcBC) was introduced into the PLA matrix at different filler loadings. The analysis of tensile characteristics indicated that the introduction of pure BC into PLA resulted in a decrease in both stiffness and strength. This may be attributed to the significant aggregation and inadequate dispersion of the BC nanoribbons inside the less polar PLA matrix. Furthermore, the inadequate interfacial adhesion between the composite constituents resulted in a diminished capacity to transmit stress from the matrix to the reinforcement, thereby imposing restrictions on the tensile strength of the composites. On the other hand, the functionalization of BC resulted in a notable enhancement in the dispersion of fillers and the interaction between fillers and the matrix. Consequently, the incorporation of AcBC content led to a significant enhancement in the stiffness of the nanocomposites, exhibiting an improvement of up to 40% compared to the pure PLA. Simultaneously, the tensile strength of the matrix remained unaffected. The enhancement of the mechanical characteristics of nanocomposites has been attributed to the integration of a phase with lower compliance into the polymer matrix (Ferreira et al. 2019). In this particular scenario, a portion of the external stress was assimilated by the more rigid phase, while a portion was dissipated via friction between particles and between particles and the polymer. This was contingent upon the

successful attainment of appropriate dispersion of nanofillers and compatibility between the reinforcement and matrix (Ávila Ramírez et al. 2020).

Thermal properties

The glass transition temperature (T_g), melting temperature (T_m), enthalpy (H), heat capacity (C_p), crystallization process, and spherulite formation of bio-polymers reinforced with nano cellulose have been extensively studied using differential scanning calorimetry (DSC). (Leyva-Porras et al. 2019; Krishnasamy et al. 2019). It has been reported that when a reinforcer acts as a nucleating agent, there is an observed increase in the number of heterogeneous nuclei for crystallization, leading to a higher crystallization temperature of the polymer. Nevertheless, the presence of the reinforcer as an anti-nucleating agent leads to a reduction in the crystallization temperature. CNCs exhibit nucleating properties, and their incorporation into polymers has the potential to increase the melting temperature, crystallization temperature (T_c), and degree of crystallinity of the polymers (Tarani et al. 2021; Ferreira et al. 2017; Khoshkava et al. 2015). The presence of thick crystalline lamellae in the polymer is correlated with an elevated melting temperature, whereas the crystallization of imperfect polymer crystals is linked to a reduced melting temperature (Ferreira et al. 2018).

Clarkson et al. (2020) conducted a study using a solvent-free melt-compounding process to investigate the incorporation of minimal concentrations of CNCs and CNFs into PLA. To disperse the nanoparticles within the PLA matrix, PEG was employed as a dispersing agent. In this study, the effectiveness of CNCs and CNFs as nucleating agents for PLA was investigated, with a focus on their similarities to talc. The Avrami analysis was conducted to determine the crystallization rate, half-time, and Avrami exponent. The results indicated a potential synergistic effect between nanocellulose and PEG, even at low concentrations. The experimental results demonstrated that the crystallization half-time of samples containing nanocellulose was observed to be lower than that of talc at elevated temperatures. This suggested that under specific conditions, the presence of nanocellulose promotes faster crystallization. The analysis of secondary nucleation was conducted to investigate the impact of CNCs on the surface energy of samples.

The findings indicated a decrease in surface energy for samples containing CNCs. This observation provided additional evidence that CNCs can act as an effective nucleation agent, particularly due to the increased mobility of plasticized PLA. These results suggested that even at very low concentrations, CNCs can effectively induce nucleation in the material (Clarkson et al. 2020). Singh et al. (2020) successfully synthesised PLA biocomposites containing up to 30% wt % of cellulosic fibers (CF) modified with poly(ethylene oxide) (PEO) using a corotating twin-screw extruder and an injection moulding machine. The thermal characteristics, in particular the glass transition and cold crystallization temperatures, were greatly affected by the addition of PEO and CF. As a result of PEO's plasticizing impact on the samples, the T_g of PLA/PEO and PLA/CF/PEO biocomposites was found to be lower than that of pure PLA. In addition, a higher degree of crystallinity in the PLA and PLA/PEO samples may be the cause of the T_{cc}'s shift to a lower temperature after the addition of CF to PLA (Singh et al. 2020).

The CNC-PLLA or CNC-PDLA composites were synthesised using the in-situ polymerization of CNCs and L-lactic acid or D-lactic acid. The composites of PLA/CNC-PLLA and PLA/CNC-PDLA exhibited improved crystallization capability and mechanical characteristics. The incorporation of CNCs derived from poly(D-lactic acid) (PDLA) exhibited a more pronounced improvement in the crystallization behaviour and mechanical characteristics of the resulting nanocomposites in comparison to CNCs derived from poly(L-lactic acid) (PLLA). Under identical cooling conditions, the nanocomposites containing 10% CNC-PDLA exhibited a significant increase in crystallinity. Specifically, slow cooling at a rate of 5 ^{0}C min^{-1} resulted in an 86.7% enhancement, while rapid cooling at a rate of 25 ^{0}C min^{-1} led to an astonishing 879% increase in crystallinity compared to pure PLA (Chai et al. 2020). The nucleation processes of CNCs inside the semi-crystalline PLA matrix were investigated in a separate study. Poly(vinyl acetate) (PVAc) chains were successfully grafted onto the surface of CNCs by a highly efficient radical polymerization procedure in an aqueous environment. The findings of this study demonstrated that despite achieving a homogenous dispersion of CNCs by greater PVAc grafting density, the nucleation effect was effectively reduced. This may be attributed to the presence of

abundant PVAc chains at the interface, which led to the dilution of PLA chains (Wu et al. 2023).

Thermogravimetric analysis (TGA) has been widely used as an analytical method for investigating the thermal stability of materials utilized in diverse environmental, pharmaceutical, food, and petrochemical contexts (Yang et al. 2019). TGA is a technique that quantifies changes in the mass of a specimen in relation to variations in temperature and duration. TGA has been used in several research studies using PLA/cellulose composites to assess the thermal stability and oxidative stability of the materials, determine the composition of multi-component systems, predict the product's durability, analyze kinetic decompositions, and measure the moisture and volatile content of the materials (Nurazzi et al. 2022). TGA was employed by Wang et al. (2020) to investigate the effect of incorporation of CNFs as a reinforcing component in PLA. The study revealed that the PLA/CNF composites had a higher first degradation temperature compared to pure PLA, suggesting that the incorporation of cellulose nanofibers led to enhanced thermal stability in the composites. The derivative thermogram (DTG) analysis reveals that PLA exhibited a single decomposition peak at 331 ^{0}C. In contrast, cellulose nanofibers exhibited two temperature peaks. The first peak, which occurred at approximately 40 ^{0}C, was associated with the hydrophilic groups present in the nanofibers. The second peak, observed at around 346 ^{0}C, was attributed to dehydration reactions and the generation of volatile products through chain scission and decomposition. Furthermore, the incorporation of 5.0 wt% of CNF into PLA/CNF composites resulted in a significant increase of 20 ^{0}C in the start degradation temperature and 10 ^{0}C in the maximum degradation temperature. The results suggested that the PLA/CNF composites enhanced the thermal properties in comparison to pure PLA (Wang et al. 2020).

In a separate study, Sucinda et al. (2021) used varying quantities of *Pennisetum purpureum*/Napier cellulose nanowhiskers (NWCs) to enhance the thermal properties of PLA by solvent casting method. The thermal degradation of the sample was seen to occur in two distinct phases, as shown in Fig. 13. During the first phase, it was observed that all film samples exhibited thermal stability within the temperature range of up to 79 ^{0}C. The bionanocomposite film surface experienced weight loss might be
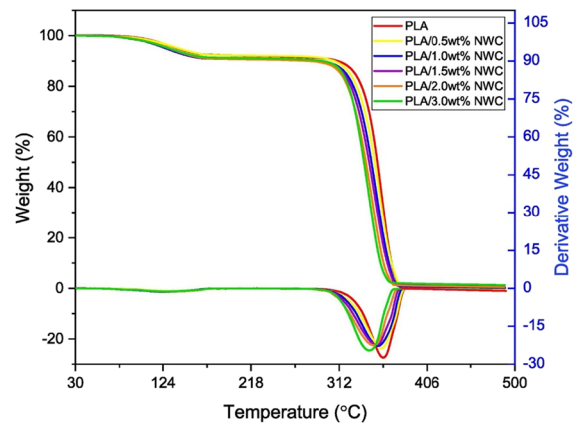

Fig. 13 TGA and DTG curves for PLA/NWC bionanocomposite films (Sucinda et al. 2021)

attributed to the evaporation of bound moisture, as seen in previous studies. During the second stage, it was observed that the weight loss started to increase when the samples contained NWC in the range of 0.5–1.5 wt%. However, as the amount of NWC in the sample increased to 3 wt%, the weight loss exhibited a declining trend. The PLA/0.5% NWC TGA curve exhibited the highest T_{onset} and T_{peak} values, measuring at 101 ^{0}C and 136 ^{0}C, respectively. The results suggested that the PLA/0.5% NWC bionanocomposite film exhibited notable thermal stability within this specific range (Sucinda et al. 2021). In a separate study, it was shown using TGA/DTG analysis that the deterioration of the CNC-based composite occurred at a lower temperature range in comparison to the MCC-based composite. The decrease in cellulose particle size provided a greater surface area-to-volume ratio, which resulted in an increased rate of heat transmission. The increased rate of heat transmission led to a decrease in the observed degradation temperature of the nanocomposites (Bhiogade and Kannan 2021). In order to create the PLA composite films, Rahman et al. (2023) first isolated fibers from the banana rachis, which they then used to synthesize CNCs. It was empirically observed that, up to an optimum level, the thermal stability of the created nanocomposite films increased together with the amount of CNC in the PLA matrix. Maximum thermal stability was achieved with PLA/CNC-20, resulting in about 40.3% residues remaining after 600 ^{0}C; all produced films continued the growing trend except PLA/ CNC-10, which exhibited a reverse tendency and

drop down with the lowest remaining residual mass which was around 2.1% after 600 ^{0}C. Compounding issues, such as CNC agglomeration and poor adherence to the PLA matrix, may have been brought on by this unintended consequence (Rahman et al. 2023).

Barrier properties

The use of cellulose nanoparticles has the potential to enhance the barrier qualities of PLA by establishing a tortuous channel. This characteristic renders the bio-composite very advantageous for various packaging applications (Kalia et al. 2011). The schematic representation in Fig. 14 depicts the tortuous pathway by which water and gas molecules permeate as a result of the incorporation of cellulose nanoparticles. In addition to possessing a solid structure characterized by low permeability, cellulose nanoparticles have the potential to establish a robust polymer-particle association, resulting in the formation of a cohesive network inside the matrix. This network has the ability to impede molecular mobility and thus restrict the diffusion of penetrant substances (Arslan et al. 2021). It is important to note that the shape, orientation, and effective dispersion of nanoparticles inside the polymer matrix significantly impact the barrier characteristics of PLA. The key characteristics that render cellulose nanoparticles suitable for packaging applications are their degree of crystallinity, aspect ratio,

surface polarity, and internal cohesion. The aforementioned features exhibit advantageous characteristics with regard to barrier properties (Ferreira et al. 2018; Helanto et al. 2019). Specifically, the presence of crystallinity enhances water resistance and reduces molecular penetration. However, it is important to note that cellulose, owing to the existence of hydroxy groups, also exhibits poor water resistance and moisture absorption. It is important to acknowledge that the evaluation of barrier qualities in polymers primarily involves the assessment of oxygen transmission rate (OTR), water vapour permeability (WVP), and overall migration tests. These parameters are significant as they have a discernible influence on deteriorative processes (Salmieri et al. 2014). Table 3 displays the mechanical, thermal and barrier properties of recently reported nanocellulose-reinforced PLA-based composite films.

Numerous studies examined the barrier characteristics of nanocomposite systems consisting of PLA and CNCs. The construction of three-phase multilayered materials (TMLs) included the formation of a composite structure by combining a film made from CNFs and CNCs with two sheets of PLA by a heat-pressing technique. Prior to the fabrication of nanocellulosic films, CNFs and CNCs underwent a modification process including the adsorption of a nanoemulsion derived from rosin. The findings of the study indicated that incorporating nanocellulose
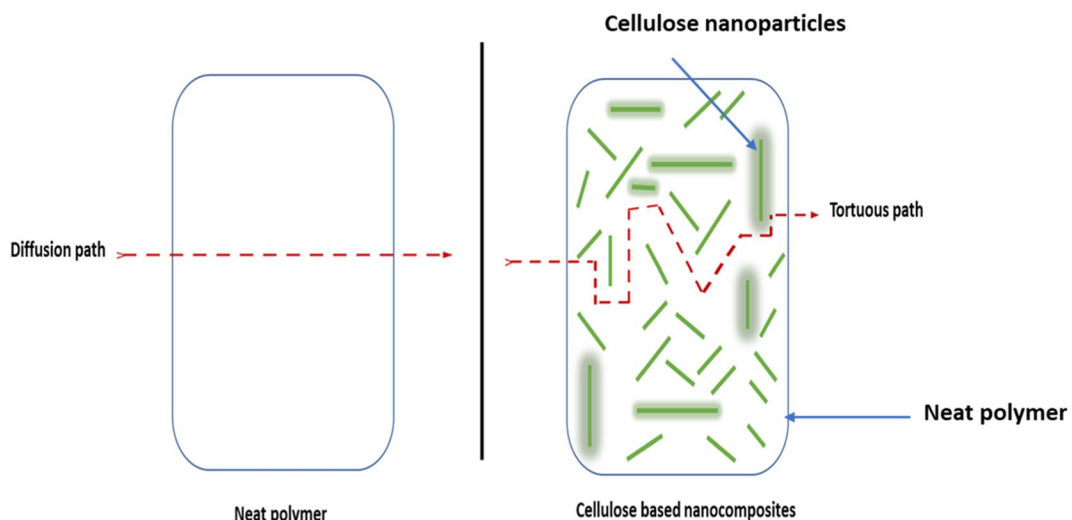

Fig. 14 Schematic representation of the tortuous path for water and gas molecules to diffuse due to addition of nanocellulose (Arslan et al. 2021)

Table 3 Properties of PLA/ nanocellulose composite films

Type of nanocellulose	Modifications	Nanoparticle content wt%	Mechanical properties			Thermal properties		Reduction in OTR (%)	Reduction in WVP (%)	References
			Tensile Strength (MPa)	Tensile Modulus (GPa)	Elongation at break (%)	T_m (°C)	T_g (°C)			
CNC	-	5	77.9	3.9	2.7	-	-	-	-	(Oksman et al. 2006)
CNC	-	3	55	2.61	2.3	147.9	58.7	-	-	(Pracella et al. 2014)
CNC	Silylation	4	14	0.11	250	154.2	30.6	-	-	(Qian and Sheng 2017)
CNC	-	3	60	3.5	2.3	165	62.5	-	-	(Arias et al. 2015)
CNC	Esterification		59.9	4.7	3.6	-	-	-	-	(Shojaeiarani et al. 2018)
CNC	Acetylation	3	62	2.1	4.2	-	-	-	-	(Xu et al. 2016)
CNC	PLA-g-CNC	1	51	2.1	1.9	165.5	52.7	-	-	(Dhar et al. 2016)
CNC	Urethanization	5	74	3.5	-	-	-	-	-	(Gwon et al. 2016)
CNC	Esterification	1	42	2.8	10	-	-	-	-	(Robles et al. 2015)
CNC	-	5	55.5	2.7	9.7	-	-	-	-	(Bondeson and Oksman 2007)
CNC	CNC-ICN	2.5	52	-	5.6	152.2	59	-	-	(Espino-Perez et al. 2013)
CNC	-	5	46.1	4.4	18	149.7	54.1	-	-	(Fortunati et al. 2012)
CNC	Silylation	2	58.6	1.4	8.3	171.8	-	-	-	(Pei et al. 2010)
CNC	-	2	53	4.75	-	-	-	-	-	(Okubo et al. 2009)
CNF	Carboxymethylation Esterification	5	63	3.75	-	-	-	-	-	(Eyholzer et al. 2012)
CNF	Surface coating/Polyethylenimine	-	-	-	-	-	-	65–90	50	(Kiziltas et al. 2016)
CNF	Surface modification/acetylation	5	-	-	-	-	-	64	46	(Feng et al. 2017)
CNF	-	10	57	3.6	-	-	-	-	-	(Quero et al. 2012)
CNF	In situ chemical grafting	5	58.5	2.55	7	-	-	-	-	(Lee et al. 2012)
BNC	Glyoxalization of BC	15	39	3.2	1.4	-	-	-	-	(Martínez-Sanz et al. 2013)
BNC	-	5	67	4.7	1.9	166	57	-	-	(Blaker et al. 2014)
BNC	Polyglycidyl methacrylate-Grafting	3	55	2.8	4	167.4	61.3	7	-	(Ambrosio-Martín et al. 2015)
BNC	Hexanoic and dodecanoic acid	2	141	5.7	17	170.2	58	-	-	(Aulin et al. 2013)
BNC	Lactic acid oligomers grafting	5	46	1.66	2.7	145	53	24	19	(Trifol et al. 2016)
BNC	Compatibizer/EVOH	3	-	-	-	157.4	48.4	-	43	(Martinez-Sanz et al. 2012)

as an intermediate layer between two PLA films led to a notable improvement in the barrier properties against oxygen. The oxygen permeability decreased by approximately 84–96% for nanocellulose alone, and by about 44–50% for nanocellulose combined with rosins as the inner layer (Le Gars et al. 2020). In a study conducted by Vilarinho et al. (2021), it was observed that the incorporation of CNCs and green tea extract (GTE) into PLA resulted in the most significant decrease in oxygen transmission ratio and water vapor permeability, with reductions of 60% and 33% respectively. These findings provided evidence of a synergistic effect resulting from the combined presence of CNCs and GTE in the PLA matrix. The nanocomposite exhibited excellent filler dispersion and a high level of crystallinity, resulting in a highly effective barrier effect (Vilarinho et al. 2021).

According to Xu et al. (2020) adding 3% CNCs decreased the oxygen transmission rate of PLA composite films from 209.9 to 180.8 $cm^3/m^2/day$. Adding compatibilizer, methylene diphenyl diisocyanate (MDI, 4%), further decreased the rate to 109.3 $cm^3/m^2/day$. PLA's water vapor transfer rate decreased with the addition of 3% NCC and 4% MDI, from 44.4 to 28.6 $g/m^2/day$. Due to interactions between the isocyanate groups of MDI and the hydroxy end groups of PLA, the addition of MDI enhanced the hydrophilicity of PLA films. Therefore, it's possible that the inclusion of MDI improved the hydrophilic CNC nanofillers' dispersion in the PLA matrix, enhancing tortuosity and lowering OTR and WVTR (Xu et al. 2020). Based on the report of Liu and Matuana (2019) due to the enhanced crystallinity of the CNCs added to the PLA matrix, the WVP and OP values of the PLA films were dramatically lowered in the case of PLA/CNC extruded cast films, resulting in a tortuosity effect. With an increase in CNC content, WVP and OP decreased practically linearly. The inclusion of 2% CNCs resulted in overall reductions of around 45% in WVP and 49% in OP. When tested at 23 C, the OP values for the neat PLA and PLA-2%CNC cast films did not change when the RH increased from 0 to 75% (Liu and Matuana 2019). The impact of CNC concentration on the oxygen permeability (OP) and water vapor permeability (WVP) of extrusion-blown PLA films was investigated by Karkhanis et al. (2018) at different relative humidities (RH) and temperatures. Both WVP and OP obeyed the Arrhenius equation; however, WVP showed a drop as temperature rose,

whereas OP showed a reverse trend, resulting in positive activation energy (E_p) for OP and a negative E_p for WVP, independent of CNC content. However, introducing CNCs to the PLA matrix raised the E_p for both WVP and OP. As per Fick's law, WVP was unaffected by the RH. Because of the tortuosity effect caused by highly crystalline CNCs in the nanocomposites, which raised PLA's degree of crystallinity, the nanocomposite films had lower WVP (~40%) and OP (~75%) than pristine PLA films. In fact, regardless of the testing circumstances, there was a negative correlation between higher crystallinity and the values of WVP and OP (Karkhanis et al. 2018).

A recent study investigated the application of nanocellulose fibrils containing a significant amount of lignin (NCFHL) in the production of biocomposites using PLA. The water vapor barrier effectiveness of pure PLA was shown to improve with the incorporation of NCFHL content up to 10 wt%. The weight variation tendency experienced a nearly 50% reduction with the introduction of a 10 wt% concentration of NCFHL, in comparison to the pure PLA sample. The inclusion of nanofibrils with high aspect ratios, together with the formation of a dense network by these fibrils, may lead to an increase in tortuosity for water molecules that permeate through the films. Moreover, the composites exhibited enhanced barrier efficacy towards gas molecules due to the high crystallinity of the NCFHL (Nair et al. 2018). In another work, PLA/functionalized cellulose nanocrystal formates (CNCFs) were synthesized using solution casting. Subsequently, the binary films were subjected to a spray coating of silver ammonia aqueous solution in order to produce PLA/CNCF/Ag ternary nanocomposites. The PLA/CNCF/Ag(6) composite showed the most significant decreases in water absorption (71.8%) and water vapour permeability (60.1%). These findings indicated that the incorporation of Ag nanoparticles and CNCF positively influenced the barrier properties of the composite material. The observed outcomes were attributed to the tortuosity of the transport pathway resulting from the heightened crystallinity (Yu et al. 2016).

Optical properties

Transparency is crucial in the industrial manufacture of food packaging materials due to its significance in allowing customers to visually observe the contents

of the package. The ultraviolet (UV) light transmission via packaging is a crucial factor for protecting light-sensitive foods and ensuring the safety of food goods until delivery (Arrieta et al. 2017). PLA is susceptible to oxidative breakdown upon exposure to UV radiation. The presence of oxidation reactions leads to the degradation of the PLA backbone, resulting in a concomitant alteration in the polymer's color. When the structural integrity of the PLA backbone is compromised, UV radiation has the ability to permeate the packaging material and initiate the oxidation process in both edible and non-edible items that are being kept. Oils and cosmetics are illustrative of consumable and non-consumable goods, correspondingly. Hence, it is of utmost significance to prioritize the preservation of stored items from UV radiation by focusing on improving the UV protective capabilities of PLA, along with other necessary attributes necessary for packaging purposes (Narayanan et al. 2017; Chen et al. 2013).

A comparative analysis was conducted to assess the UV-shielding effectiveness of PLA nanocomposites containing lignin-coated cellulose nanocrystals (LCNC) compared to PLA nanocomposites containing lignin nanofillers (LNP). The UV–Vis analysis demonstrated a synergistic impact when integrating either LNP or LCNC nanostructures in relation to their capacity to block UV radiation. The incorporation of low-content (3 wt%) LCNCs into the PLA matrix resulted in the most effective blocking of UV radiation, with a 75.3% reduction in UV-A and a 45.81% reduction in UV-B (Shojaeiarani et al. 2022). Polydopamine (PDA) decorated CNCs were synthesized by Xu et al. (2022). The PDA@CNCs was then incorporated into PLA film using a solution precipitation method followed by hot-pressing. Figure 15 displays the images together with the associated transmittance values of various samples in the wavelength range of 200 to 900 nm, both before and after undergoing UV aging. Following the
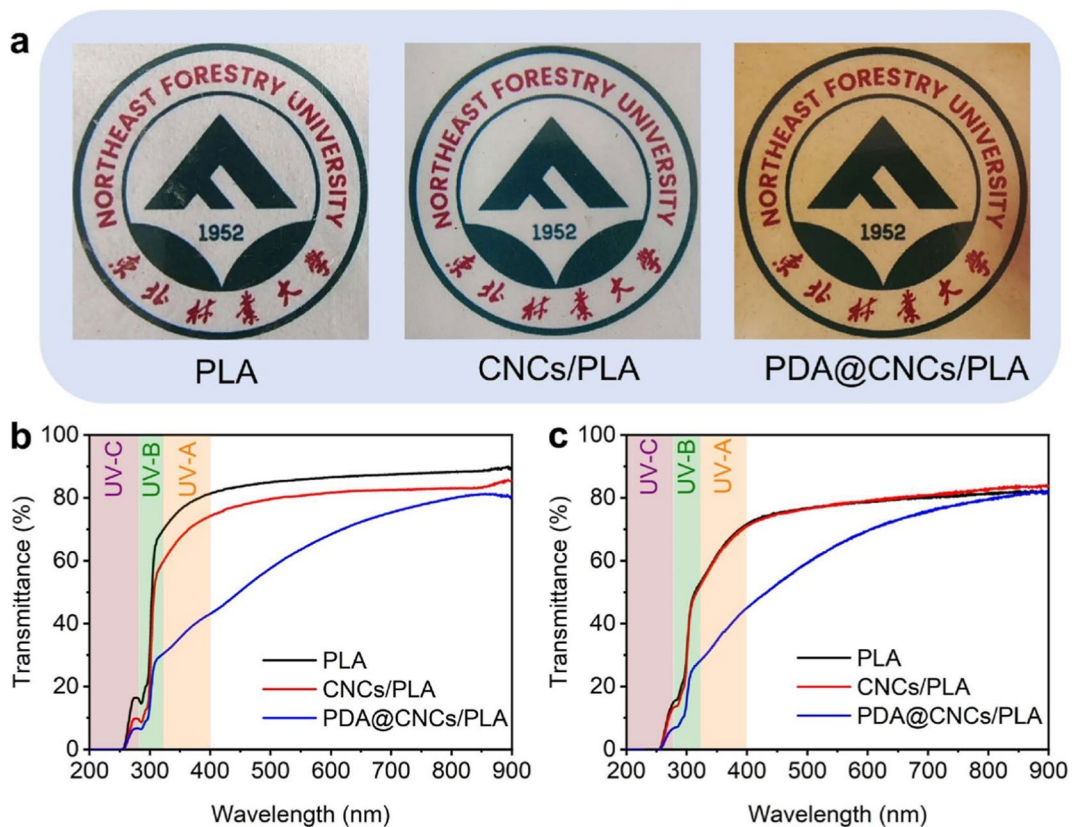

Fig. 15 a The images of PLA, CNCs/PLA, and PDA@CNCs/PLA films b UV spectra of PLA, CNCs/PLA, and PDA@CNCs/PLA films before UV aging, and c UV spectra of PLA, CNCs/PLA, and PDA@CNCs/PLA films after UV aging (Xu et al. 2022)

incorporation of CNCs, there was a marginal reduction seen in the transmittance of the PLA film. At an equivalent degree of addition, the PDA@CNCs/PLA film demonstrated a significant reduction in transmittance while also displaying a discernible UV shielding characteristic. Following exposure to UV aging, the transmittance of all three films exhibited a marginal decline (Fig. 15c), providing direct evidence of the inherent UV shielding capabilities possessed by the materials (Xu et al. 2022).

Jamaluddin et al. (2019) conducted a study to investigate the impact of Acetylated cellulose nanofiber (ACNF) on the optical characteristics of PLA. The PLA film exhibited a remarkable transmittance of 85%, surpassing all other PLA composite films in terms of optical clarity. The addition of cellulose fibers resulted in reduced transmittances of 72% and 70% for PLA/ACNF and PLA/CNF, respectively. Both PLA/ACNF and PLA/CNF composite films exhibited almost identical levels of transmittance. However, PLA/ACNF composite films had a slightly greater transmittance of 2% due to the favorable compatibility of ACNF. The transparency of the samples may also be influenced by their crystallinity (Jamaluddin et al. 2019). Another work described the use of PLA nanocomposite films that included cellulose nanocrystal-zinc oxide (CNC-ZnO) hybrids. The incorporation of a significant proportion (15wt %) of CNC-ZnO hybrids into the PLA matrix resulted in the effective blocking of the highest levels of UV radiation, with a reduction of 85.31% in UV-A and 95.90% in UV-B. The incorporation of CNC-ZnO hybrids into the PLA matrix resulted in synergistic UV-shielding effects, leading to the development of PLA nanocomposite films with superior UV-shielding capabilities in comparison to pure PLA (Wang et al. 2019).

Antibacterial and antioxidant properties

The occurrence of food-borne diseases has generated public consciousness about the significance of preventing microbial contamination in both raw materials and processed food items. The food that has been infected transforms into rubbish, and the act of consuming this contaminated food poses a significant risk to the consumer's health (Velasquez et al. 2021). Many studies have been conducted on active packaging to tackle the challenges related to the growth and

activity of harmful microorganisms in food, aiming to improve food safety. Active food packaging is capable of performing several activities, including acting as an antioxidant, oxygen scavenger, flavor enhancer, moisture absorber, UV barrier, and antibacterial agent. The use of active packaging, which involves the release of active biocidal chemicals into food, has garnered significant interest for its potential to extend the shelf life of food and reduce the occurrence of foodborne germs. The achievement of antimicrobial activity may be realized by the direct incorporation of active biocides into food items or by their application in the vicinity of the food. The primary purpose of antimicrobial packaging is to mitigate and subsequently inhibit the growth of spoiling bacteria (Ribeiro-Santos et al. 2017; Sung et al. 2013).

The acquisition of nanotechnology-based food packaging may be achieved by integrating bio-based nanocellulose with antibacterial and antioxidant functional agents, which can be produced from natural resources or metal nanostructures (Echegoyen and Nerín 2013). The integration of these two nanomaterials yields a sustainable bionanocomposite exhibiting improved antimicrobial and antioxidant properties. This composite material holds promise as an effective packaging solution, safeguarding food products against microbial contamination while simultaneously addressing concerns related to the build-up of agricultural waste and environmental degradation (Fortunati et al. 2014).

Bioactive PLA–CNC–oregano films were fabricated by integrating oregano essential oil (EO) as an antibacterial agent and the films were used as packing material for mixed vegetables. The purpose of this storage was to assess the effectiveness of the films in inhibiting the growth of *Listeria monocytogenes*. The microbiological analysis of mixed vegetables inoculated with *L. monocytogenes* revealed that PLA-CNC-oregano films effectively inhibited bacterial growth in the vegetables by day 14. This demonstrated the significant antimicrobial properties of these films in the specific environment (Salmieri et al. 2014). Khodayari et al. (2019) conducted a study to assess the impact of incorporating CNCs, Tanacetum *balsamita* L. essential oil (TBE), and propolis ethanolic extract (PEE) into PLA composite films. The objective was to determine the effectiveness of these composite films in prolonging the shelf life of vacuum-packed cooked sausages. Based on the

findings obtained from the disc diffusion experiment, it was shown that the presence of TBE in PLA films has a significant impact on their antibacterial properties ($p < 0.05$). Conversely, PLA films including only PEE and/or CNCs did not exhibit any inhibitory effects on bacterial growth. The integration of PEE into other films that also included TBE exhibited synergistic effects on the studied microorganisms. The study revealed that Gram-positive bacteria exhibited greater susceptibility to the active PLA films compared to Gram-negative bacteria. Additionally, it was observed that *B. cereus* showed the highest level of sensitivity to films containing TBE and PEE (Khodayari et al. 2019). The researchers Abdulkhani et al (2017) *fabricated* PLA/CNFs composites that were coated with an ethanolic extract of propolis (EEP). The modified PLA/CNFs films exhibited antibacterial properties against Gram-positive bacteria, even when exposed to very low concentrations of EEP. The inclusion of EEP in the tested films exhibited a significant enhancement in the antibacterial efficacy against Gram-positive bacteria, including *Bacillus anthracis, Staphylococcus aureus, and Salmonella enteric*. However, no discernible impact was seen on Gram-negative bacteria (Abdulkhani et al. 2017).

A few types of metal nanoparticle antibacterial agents have been investigated, such as copper (Cu), gold (Au), zinc (Zn), silver (Ag), and titanium (Ti). The antibacterial activity of silver nanoparticles (AgNPs) was shown to be superior to that of other metal nanoparticle agents (Carbone et al. 2016). The

reduced dimensions of AgNPs result in an increased surface area, facilitating their contact with microbial cells and leading to enhanced antibacterial efficacy when compared to bigger Ag particles. In addition, nanosilver has a wide range of antibacterial capabilities, making it effective against microbes that are resistant to antibiotics or several drugs (De Azeredo 2013). The supposed functional mechanism of AgNPs is as follows: i) AgNPs attach to membrane proteins and the cell wall, leading to the formation of pits on the cell surface and subsequent penetration into the cell. This process causes harm to the bacterial cell structure, leading to cell lysis and subsequent death. ii) The growth-inhibiting ability against microorganisms is due to the denaturation of enzymes and the disruption of the bacteria's respiratory chain. iii) The electrostatic attraction among positively charged nanoparticles and the negatively charged cell membranes of bacteria results in the destruction of the bacterial cytoplasmic membrane. This damage triggers the production of reactive oxygen species (ROS) and oxidative stress. iv) AgNPs modulate the phosphotyrosine profile of bacterial peptides through the inhibition of signal transduction and growth of cells. v) The presence of AgNPs hinders DNA integrity and inhibits cellular replication (Duncan 2011; Gan and Chow 2018).

In a particular work, the researchers produced PLA/functionalized CNFs by a process of solution casting. Subsequently, the binary films obtained were subjected to a spray coating of silver ammonia
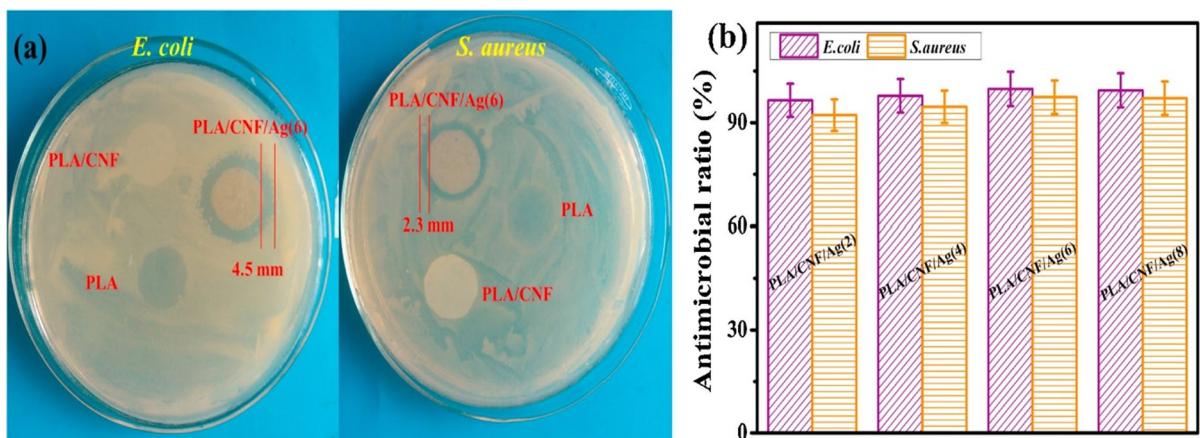

Fig. 16 **a** Antibacterial properties of PLA/CNF/Ag(6) to *E. coli* and *S. aureus* **b** Antimicrobial ratios of ternary PLA/CNF/Ag nanocomposite films at different Ag contents (Yu et al. 2016)

aqueous solution, resulting in the fabrication of PLA/CNF/Ag ternary nanocomposites. The antimicrobial characteristics of pure PLA and PLA nanocomposite films were assessed against *Escherichia coli* (*E. coli*) and *Staphylococcus aureus* (*S. aureus*), as seen in Fig. 16a**.** It is evident that the PLA and PLA/CNF films do not exhibit any inhibition zone against bacteria. However, the PLA/CNF/Ag(6) film demonstrated inhibition zones of 4.5 mm and 2.3 mm against *E. coli* and *S. aureus*, respectively. Figure 16b illustrates the antibacterial efficacy of several nanocomposite films. The results indicated that the PLA/CNF/Ag composite exhibited a higher antimicrobial ratio against both *E. coli* and *S. aureus* compared to the neat PLA and PLA/CNF binary systems. The PLA/CNF/Ag(6) composite exhibited the greatest antibacterial ratios, that is, 99.8% for *E. coli* and 97.4% for *S. aureus*. This notable antimicrobial efficacy may be attributed to the enhanced antimicrobial surface area achieved by the uniform deposition of Ag nanoparticles. A marginal decrease in the antimicrobial efficacy was detected for the PLA/CNF/Ag(8) composite, with reductions of 98.4% for *E. coli* and 96.1% for *S. aureus*. This decrease may be attributed to an increase in the size of aggregated silver nanoparticles, resulting in a weakened antibacterial capacity (Yu et al. 2016).

Oxidation is a prominent degradation mechanism that takes place during the deterioration of food. The definition of an antioxidant pertains to any substance that has the capability to either delay or inhibit the process of oxidation in a given substrate (Papuc et al. 2017; Domínguez et al. 2019). The antioxidant capabilities of CNFs or CNCs in combination with PLA were described by Le Gars et al (2020)*.* Using a heat-pressing technique, two PLA sheets were complexed with a dry CNF- or CNC-based film to create three-phase multi-layered materials (TMLs). Prior to the production of nanocellulosic films, CNFs and CNCs underwent modification through the adsorption of a rosin-based nanoemulsion. The 2, 2-Diphenyl-1-picrylhydrazyl (DPPH) test was also conducted on the multilayered materials TML−CNC−rosin and TML−CNF−rosin. In this experimental study, the inner cellulosic layer of the multilayered materials was directly exposed to the DPPH solution, and attained radical scavenging activity (RSA) values in a plateau of 29% and 43% for the TML−CNC−rosin and TML−CNF−rosin samples, respectively. The

findings of this study demonstrated the efficacy of rosin nanoparticles in enhancing the performance of multilayered materials (Le Gars et al. 2020). Bayer et al. (2023) conducted a study to investigate the impact of cellulose fiber content on the characteristics of PLA composites. Antioxidant composites were produced by immersing cellulose fibers in limonene, a naturally occurring terpene. The composites that were treated with limonene exhibited sustained release and efficacy of antioxidants for a duration of three days. This was confirmed via the use of three distinct assays: DPPH, cupric ion reducing antioxidant capacity (CUPRAC), and free iron ions (Fe^{2+})/ferrozine chelating assays. The biocomposites exhibited short-term antioxidant activity, lasting for 2 h. The levels of antioxidant activity ranged from 50 to 70%, depending on the percentage of cellulose fibers used in the DPPH and CUPRAC tests. However, the antioxidant activity was lower, ranging from 20 to 55%, in the metal chelating assay. The composites exhibited enhanced iron chelating antioxidant activity of 75% after a 5-day period, which may be attributed to the continuous release of limonene. Additionally, the composites had a recorded activity of 90% for the DPPH and CUPRAC assays (Bayer et al. 2023).

Biodegradation properties

The term "biodegradability" refers to the process of material deterioration after interactions with biological components. The issue of biodegradability is of significant importance within the packaging sector that relies on plastic materials. The use of non-biodegradable polymers has a detrimental impact on the environment and climate (Goswami and O'Haire 2016). Researchers are increasingly showing interest in the use of environmentally friendly packaging materials. Hence, the use of polymers with enhanced biodegradability characteristics is a promising alternative for addressing the challenges associated with biodegradation. Modified polymers are specifically engineered to undergo degradation in the absence of oxygen, resulting in the production of beneficial substances like manure. These polymers do not exhibit long-term persistence in the environment. The deterioration of PLA-based materials may occur when exposed to various environmental conditions, leading to undesirable outcomes. The occurrence of irreversible alterations inside a polymer resulting from

degradation processes ultimately leads to the loss of its properties (Haider et al. 2019). PLA has the inherent ability to undergo degradation via the process of ester bond hydrolysis. PLA, when subjected to appropriate environmental circumstances characterized by the presence of moisture, oxygen, and naturally existing microbes, undergoes decomposition into water, carbon dioxide, and a negligible amount of harmless residual substances. This property of PLA represents a significant benefit associated with its use. The breakdown rates of PLA are subject to the effect of many parameters, including the isomer ratio, hydrolysis temperature, pH level, duration of burial, humidity levels, oxygen exposure, as well as the form and size characteristics of the material (Pradhan et al. 2010; Zaaba and Jaafar 2020; Teixeira et al. 2021). PLA degradation may occur via many methods. The mechanisms included in this category consist of hydrolytic, oxidative, thermal, microbiological, enzymatic, chemical, and photodegradative processes. These processes mostly induce both main chain and side chain scissions (Elsawy et al. 2017). Several studies have examined the biodegradability of PLA, but there is a lack of research on the degradation of PLA/nanocellulose nanocomposites. The addition of CNCs can enhance the hydrophilic properties of the polymer, thereby promoting an increase in the biodegradability of the matrix (Galera Manzano et al. 2021).

The study reported on the breakdown behavior of pure PLA and PLA composites containing 3 wt% CNC, both modified and unmodified with non-ionic surfactants (S) at a weight ratio of 1:1 (CNC:S), in garden soil. The study included the evaluation of four distinct non-ionic surfactants, each with a hydrophilic-lipophilic balance (HLB) within the range of 4.3 to 16.7. These surfactants were identified as sorbitan monolaurate (Span 20), sorbitan monooleate (Span 80), polyoxyethylene sorbitan monolaurate (Tween 20), and polyoxyethylene sorbitan monooleate (Tween 80). The findings of the study indicated that the inclusion of surfactants had a positive impact on the biodegradation rate of the composites. Specifically, it was observed that surfactants with lower molecular weights were more effective in enhancing the biodegradation rate of PLA/CNC/S composites. However, when considering surfactants that fall within the same chemical family, it was seen that the PLA/CNC/S composite exhibits the best rate of biodegradation when the surfactants possess a high

HLB and a small spherulite size (Gois et al. 2023). Hegyesi et al. (2019) conducted a study on the enzymatic degradation of PLA and its nanocomposites, which were reinforced with CNCs. The degradation process was catalyzed using lipase from *Candida rugosa* and proteinase K from *Tritirachium album*. The composites were synthesized using the Pickering emulsion method, including nanocellulose at weight percentages of 5, 10, and 15. The experimental findings indicated that the lipase enzyme does not exhibit catalytic activity in the breakdown of PLA, but the proteinase K enzyme has a high level of efficiency in this process. The production of lactic acid during the reaction resulted in a significant fall in the pH of the degradation medium, causing the enzyme to undergo denaturation. In addition to pH, the concentration of ions in the solution also has an impact on the pace of degradation. A lower ionic strength was shown to be more favourable in this regard. The use of CNCs as reinforcement in PLA led to an accelerated degradation rate, resulting in fast disintegration of the samples and the polymer underwent deterioration within a span of three days (Hegyesi et al. 2019).

Accelerated weathering test methods are used to examine the deterioration of polymeric materials after exposure to simulated conditions. The degradation of PLA biocomposites is influenced by the fiber/filler content and can occur at varying rates compared to neat PLA degradation. Polylactic acid (PLA) exhibits a slight polarity that promotes degradation through hydrolysis and photolysis, resulting in faster breakdown when exposed to moisture and UV radiation compared to traditional polymers. The rate of degradation is influenced by the initial molecular weight, sample dimensions, crystallinity, and the inclusion of fillers or reinforcements. PLA films may undergo substantial alterations following brief exposures due to their limited crystallinity. The weight-average molecular weight (Mw) of PLA decreased significantly after 300 h of accelerated weathering. PLA undergoes hydrolytic degradation through chain cleavage, primarily in amorphous regions. The hydrolysis process is self-catalyzed and is influenced by the level of absorbed water. Weathering causes greater damage in amorphous PLA due to the vulnerability of non-compacted polymeric chains, while semi-crystalline regions require more energy to disrupt the chains. (Gonzalez-Lopez et al. 2020). Kaynak and Dogu (2016) investigated the accelerated weathering (both UV and moisture)

behaviour of neat PLA and PLA/MCC biocomposites by comparing their mechanical and thermal properties under increasing accelerated weathering periods. They compared the outdoor usability of PLA and PLA/MCC (PLA reinforced with microcrystalline cellulose (MCC)) (Kaynak and Dogu 2016).

In a separate investigation, the synthesis of nanocomposites comprising PLA and CNCs was conducted. This included dispersing CNCs in an aqueous poly(vinyl acetate) (PVAc) emulsion, which was then subjected to melt extrusion with PLA. The process of functionalizing PVAc by radical grafting of glycidyl methacrylate (GMA) was conducted simultaneously with the polymerization of PVAc. Ammonium cerium (IV) nitrate was used as the initiator for this reaction. The investigation on degradability revealed that the rate of disintegration of PLA/PVAcGMA/CNC in soil was found to be greater compared to that of PLA/PVAc/CNC. This disparity was attributed to the presence of epoxy groups in GMA, which likely facilitated the acceleration of biodegradation by promoting the creation of radicals (Haque et al. 2017). The biodegradation capabilities of transparent PLA nanocomposite films including CNC-zinc oxide (CNC-ZnO) hybrids were evaluated by Wang et al. (2019). This evaluation was conducted under controlled hydrolytic, soil burial, and heat conditions. The study revealed that the weight loss of PLA nanocomposites, after a hydrolytic degradation period of 70 days, exhibited an increase from 9% for pure PLA to 25% when including 15 wt% of CNC-ZnO hybrids. In the context of soil burial testing was shown that PLA exhibited the lowest deterioration rate, resulting in a mere 8% reduction in weight after a duration of 110 days. Conversely, the PLA nanocomposite film containing 15 wt% of CNC-ZnO hybrids showed a
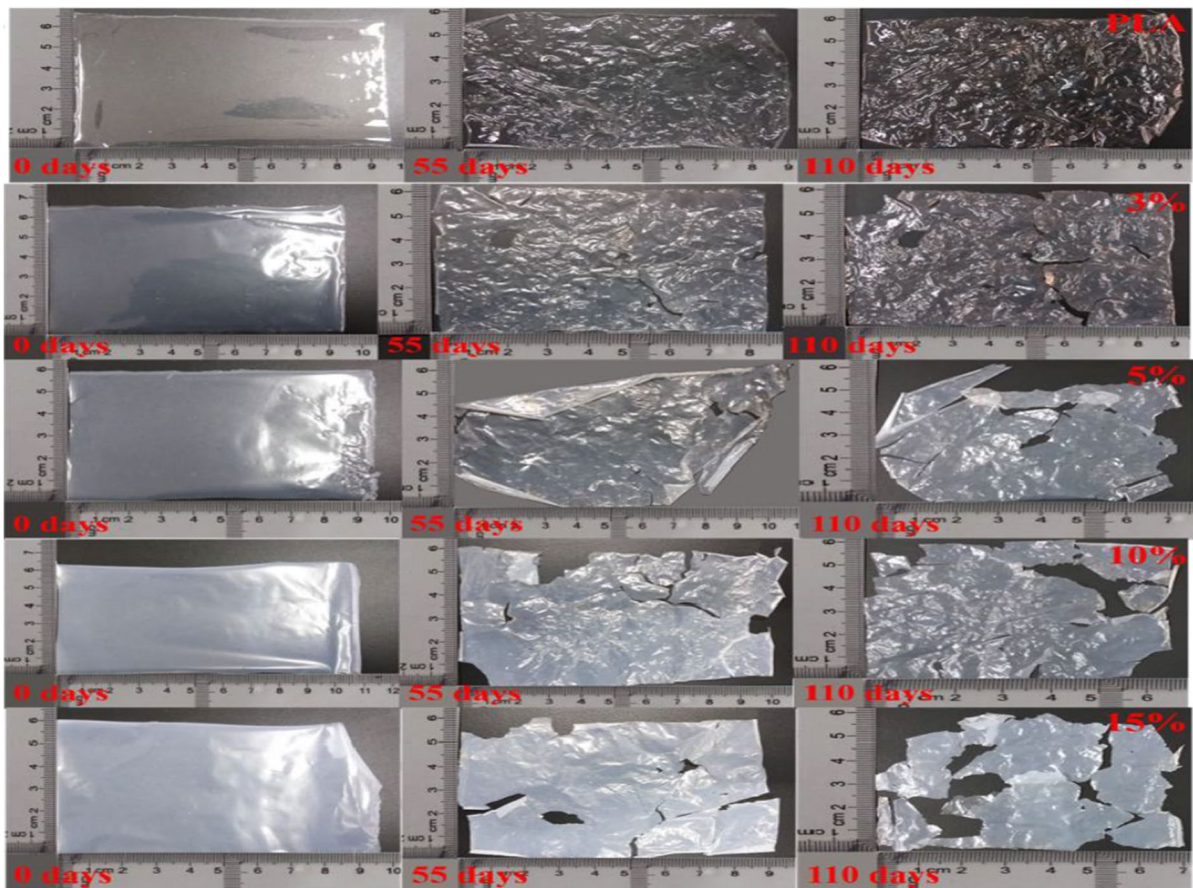

Fig. 17 Images of PLA nanocomposite films before and after soil degradation (Wang et al. 2019)

degradation rate of around 28% (Wang et al. 2019). Figure 17 illustrates the changes in the visual characteristics and size of PLA nanocomposite films during the process of soil degradation.

Safety of nanocellulose bionanocomposites

According to prevailing European regulation, it is essential for all materials that come into contact with food, often referred to as Food Contact Materials (FCMs), to adhere to the standards outlined in the framework Regulation (EC) No 1935/2004. *"Materials shall be manufactured in accordance with good manufacturing practice so that they do not transfer their constituents to food in quantities that could endanger human health; or cause an unacceptable change in the composition of the food; or cause a deterioration in the organoleptic characteristics thereof,"* in accordance with the established regulations. While this regulation does not specifically provide guidelines for nanoparticles used in food contact materials, it is important to note that these criteria are equally applicable to composites based on nanocellulose (Visanko et al. 2015). According to Regulation (EU) No. 10/201 on plastic materials and Regulation (EC) No. 450/2009 on active and intelligent packaging materials, it is stipulated that substances in nanoform should undergo individual evaluation. The processes governing mass transfer and interaction between nanoparticles and their respective host materials and food substances may exhibit variations compared to those often seen at larger particle size scales. Consequently, nanoparticles have the potential to induce diverse amounts of exposure and exhibit hazardous properties. Consequently, the premarket authorizations that are granted based on a risk assessment of a medicine with a conventional particle size do not extend to the utilization of the same substance in its nano-scale dimensions. The utilization of the substance in its nano-scale form is permissible only if explicitly authorised and included in the positive lists specified in the aforementioned regulations (Dainelli 2015; Cirillo et al. 2015).

Cellulose has many uses in the realms of food and pharmaceuticals and is recognised as being "generally regarded as safe" (GRAS). Nevertheless, it should be noted that nanocellulose, although known to increase the quality and safety of food, does not fall under the GRAS category (DeLoid et al. 2019). According to European Regulation-No 10/2011, cellulose and cellulose derivatives have been deemed safe and authorized for use as polymer additives, manufacturing aids (such as cellulose acetate butyrate and hydroxyalkyl cellulose), and starting materials (such as nitrocellulose and lignocellulose) in packaging applications (DeLoid et al. 2016). Nevertheless, it should be noted that nanocellulose is not currently included in the list of recognized substances for use in food contact applications. Numerous epidemiological investigations have provided evidence indicating that nanoscale materials exhibit unique properties and interact with biological systems in a manner distinct from their native equivalents (Silva et al. 2020). The morphology, dimensions, and clustering properties of nanoparticles have the potential to influence the interactions between nanocellulose and biological tissues. The safety assessment of nano-cellulose in food packaging is contingent upon the transfer of nanocellulose into food, since this factor governs its toxicological properties and potential human exposure. Several researches have shown the deleterious impact of nano-cellulose (Serpa et al. 2016). Endes et al. (2016) conducted a comprehensive assessment of several in vitro and in vivo research, as well as ecological models. The researchers performed a study on the possible risks posed by various kinds of nanocellulose to human health and the environment. The authors highlighted many important aspects, such as the capacity of nano cellulose to create reactive oxygen species (ROS) and its potential cytotoxic, genotoxic, and oxidative consequences. The collected data revealed a certain degree of diversity in the acquired results, with some studies reporting little or no toxicity associated with nano-cellulose, while others highlighted the presence of detrimental effects (Endes et al. 2016). Hence, it is recommended that a comprehensive toxicological investigation be conducted prior to the use of nanocellulose in materials intended for food contact. It is important to consider migration behavior, the persistence of nanoforms, and the change of nanomaterials from nano to non-nano form inside food matrices. Moreover, it is imperative that future research prioritize the investigation of nanocellulose safety, particularly in relation to its suitability for food contact applications, since it has not yet received approval for such uses.

Future perspective and conclusion

The packaging sector has a significant interest in biopolymers because to the prevailing worldwide environmental concerns and the challenges associated with the disposal of solid wastes generated by petroleum-based polymers. The integration of nanocellulose into PLA has resulted in the development of novel and environmentally friendly nanocomposites that exhibit improved performance characteristics. Despite the many benefits of PLA, it is important to acknowledge some limitations such as its poor toughness, delayed crystallization kinetics, and low melt strength. However, these downsides may potentially be mitigated with the incorporation of cellulose nanoparticles. Nevertheless, the effective incorporation of nanocellulose into PLA by direct melt mixing remains a significant obstacle due to the inherently hydrophilic nature of nanocellulose and its subsequent dispersion. Numerous investigations have been conducted to assess the efficacy of PLA nanocellulose nanocomposites through the utilization of various production methods, including solution casting and hybrid processes. Additionally, the incorporation of surfactants, compatibilizers, and surface chemical modifications has been explored to enhance the dispersion quality of nanocellulose, as it directly impacts the performance of PLA nanocellulose nanocomposite systems. The findings of these investigations demonstrate that the integration of uniformly distributed nanocellulose has the potential to enhance the crystallinity, crystallization rate, and mechanical characteristics of PLA. The integration of cellulose nanoparticles, in conjunction with the heightened crystallinity of PLA, leads to further improvements in the barrier characteristics against oxygen, water, and other volatile chemicals. The effective advancement of cellulose nanocomposites based on PLA has significant potential for diverse applications, particularly in the domain of packaging, where they might potentially supplant petroleum-derived polymers.

The use of cost-effective modifications and isolation procedures for biopolymer nanocellulose enhances its desirable qualities, hence facilitating its adoption as a commercially viable biomaterial in diverse large-scale applications. Hence, the forthcoming progress that prioritizes the economical and environmentally sustainable methods of extracting and modifying nanocellulose will facilitate the rapid and advantageous development of this remarkable biomaterial for industrial purposes. Nevertheless, the monitoring of nano-cellulose absorption and its subsequent actions within the biological system presents a formidable task, mostly owing to its diminutive size and the absence of viable analytical techniques. In order to tackle these problems, it is imperative that future research endeavors focus on attaining an in-depth understanding of the biological behavior and exposure dosage of nano-cellulose. Moreover, it is essential to do further investigation into nanocellulosic materials in order to address the increasingly limited regulatory, economic, and technological disparities between sustainable and traditional packaging methods used in the food industry.

This review provided a complete examination of the advantages linked to the addition of nanocellulose into PLA matrix for use in biodegradable food packaging. This research begun by conducting a comprehensive examination of the characteristics of food packaging material based on PLA, with a particular focus on its performance. This article provided a concise overview in the synthesis and extraction of nanocellulose, specifically focusing on its derivation from agricultural waste materials. This article provided a thorough overview of the characteristics shown by PLA-NC bionanocomposites when used as materials for food packaging. The aforementioned features include several aspects such as shape, rheology, mechanical strength, thermal stability, barrier properties, antioxidant activity, antibacterial activities, and biodegradability. This article provided a succinct summary of the safety issues associated with the advancement of PLA-NC bionanocomposites for their use in food packaging. We anticipate that this review will encourage research aimed at enhancing the characteristics of PLA/nanocellulose, thereby expanding their industrial applications and promoting the sustainable use of renewable materials.

Author contributions Jissy Jacob: Conceptualization, Investigation, Writing-original draft & editing.

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Hanna J. Maria: Editing.

Laly A. Pothan: Conceptualization, Editing.

Sabu Thomas: Conceptualization, Editing.

Sana Kabdrakhmanova: Editing.

David Laroze: Editing.

Declarations

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