Composites Part B 172 (2019) 16–25

Contents lists available at ScienceDirect

Composites Part B

journal homepage: www.elsevier.com/locate/compositesb

Eco-friendly polymer composites for green packaging: Future vision and challenges

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ARTICLE INFO

Keywords:
Biodegradable biopolymer
Green packaging
Intelligent food packaging
Coffee grounds
Date stones
Expanded organoclay

ABSTRACT

At present, renewable and biodegradable biocomposites materials have drawn much attention as promising green materials in different domains of application such as intelligent food packaging, biomedical and drug delivery, bio-membranes, automotive, as well as in industrial composting applications. The current review deals with the advances in preparation methods and technical applications of these biocomposites. Different biomass materials obtained from renewable resources such as coffee grounds (CG), nanocellulose and date stones are developed to be used as smart reinforcing agents in biodegradable biopolymers for improving their overall properties. Conversely, some drawbacks are associated with the use of lignocellulosic materials as reinforcing agents, especially their high humidity absorption, poor wettability, and incompatibility with most biopolymers. Thus, novel processing techniques and different aspects are proposed in this review to produce high performance lignocellulosic reinforced materials with better properties. Facial and green modification of organoclay (OC) by antibacterial natural rosin and stearic acid to obtain toxicity-free expanded OC is also discussed. Green modification using OC can also be used as compatibilizing and reinforcing material for different incompatible biopolymers such as chitosan, carboxy methyl cellulose (CMC) and polylactic acid (PLA). Ultimately, the future vision on the challenges and the environmental issues towards CO_2 emission which is associated to the risk assessment of these bionanomaterials are also discussed.

1. Introduction

Green packaging based on biodegradable composite materials has currently gained great attention in many disciplines because of unique properties when compared to classical petrochemical-based plastics [1–3]). In addition to their 100% biodegradability and complete decay to carbon dioxide, water and humus. These features can open opportunities for their uses in a wide range of applications such as intelligent nano-food packaging [4–6], bio-membranes for waste water, drug delivery and composting purposes. Thus, the basic function for package material is to achieve the benefits of improved food quality and safety with prolonging its shelf life [[2,7].

Green packaging materials can be biocomposite materials, bio-nanocomposites, or nano-papers [8].Thus, biocomposites could be designated as materials made from biodegradable and renewable particles such as cellulose fibers, coffee grounds [9,10], date stones, etc [11]. On the other hand, nanocomposites that are derived from biopolymers (i.e. PLA, PBAT, PHA) with synthetic or inorganic nano-fillers (i.e. carbon nanotubes, hybrid nanoclay fall under bio-nanocomposites) [12]. The prefix “Bio” in biocomposites or bio-nanocomposites means they are naturally biodegradable by microorganisms.

The resulting organic by products tend to be safe not only for the environment, but also for improving people’s lives. Additionally, a term “Green” has become common and developed not just as a logo or label, but generally refers to composite materials, products, and technologies that have less environmental impact, as well as on the human life because of their safety regulations [13,14]. As a result, composite materials that contain biodegradable and renewable components are considered to be toxicity-free. Consequently, green packaging materials play a vital role to preserve and to protect the food from contaminates and the rot caused by microbes, in order to extend its shelf life. Moreover, it has potential applications in many aspects of food chain and beverages including storage during their transportation, safety

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https://doi.org/10.1016/j.compositesb.2019.05.048
Received 26 October 2018; Received in revised form 28 March 2019; Accepted 5 May 2019
Available online 7 May 2019

regulations, quality monitoring, and food processing [15,16]. To fulfill these advantages, the green packaging biomaterial should be reinforced by nanomaterial or antimicrobial nanoparticles to enhance biopolymeric properties of packaging materials and to meet the global market needs. For example, mechanical properties, antimicrobial activities, barrier properties, and thermal behaviors, as well as energy harvesting and storage [17–21] that can be improved when these kinds of nano-scaled materials are used.

Several studies have been conducted on nano-scales used in food packaging. The European Institute for Health and Consumer Protection estimated that the use of nanostructures in the nano-food packaging market is expected to reach $20 billion by 2020 [22]. Thus, the enhancement in mechanical, antimicrobial and barrier properties could be achieved when biopolymers are compounded with nanomaterials that are produced from plants and animals as reported in literature [23,24]. [25] reported that polymer chain length of pectin in fruit was approximately 100–400 nm, while in another work, the dimensions of casein micelles extracted from the milk were about 300–400 nm. On the other hand, nonclay derivatives as synthetic nanoscale materials, like montmorillonite and layered double hydroxide (LDH) are more efficient and compact regarding to their availability, low cost, and biocompatibility [26–28].

Traditionally, the modification of nanoclay by organomodifier is crucial not only for increasing the d-spacing distance between clay layers, but also for enhancing the filler-polymer homogeneity. This modification is often performed by using long chain of alkylammonium salts and its d-spacing depends mainly on the amount of surfactant (Moustafa et al., 2017e; [29–31]. When, d-spacing of OC is less than 2 nm, this means that all surfactant molecules are chemically interlocked with negative clay layers and give intercalated form. Nevertheless, to obtain a highly exfoliated structure of individual clay platelets in the polymer matrix with expected better properties, the d-spacing distance shall be 3 nm or more relying on the amount of surfactant added [3,32,33].

This can be achieved when part of the organomodifier molecules is chemically counter balanced with clay layers, whereas the other part of the surfactant molecules is physically absorbed into the clay galleries that are equivalent with Br^- anions. Nevertheless, the presence of this type of anion in the galleries can limit its use in food packaging applications because of Br^- toxicity. For this reason, our review article offers an overview of the classifications and modifications of nanomaterials by toxicity-free components and gives an overview of the different sources of these materials, from natural to synthetic, and their uses towards the nano-food packaging. Moreover, the biocompatibility between smart antimicrobial nanomaterial and biopolymer matrix to obtain green packaging that can highly protected the food from unfavorable conditions such as water vapor, oxygen, flavor, and microorganisms.

2. Classification of green reinforcements

The compounding of bio-reinforcing agents such as nanocellulose into polymers has been examined as a way to enhance their properties.

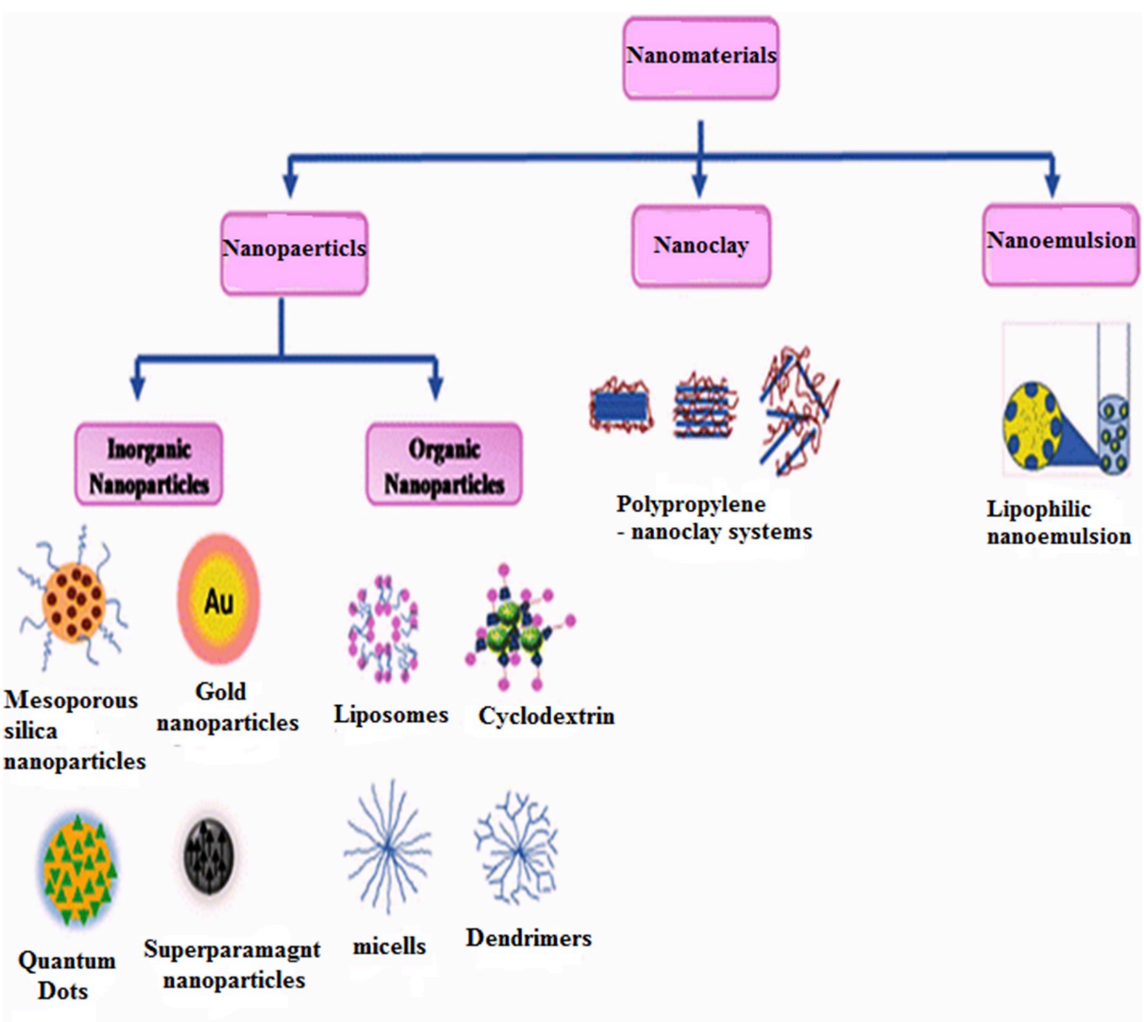

Fig. 1. Classification of nanomaterials based on their categories [41].

Many studies have been focused on the use of bioreinforcing materials derived from renewable resources as an alternative to inorganic reinforcing agents for obtaining full green materials [34]. Based on the previous studies, nanomaterials or green nano-reinforcements can be classified according to their categories [35], their dimensions [36–39], or their origin [40]. In the current review, we focus on the classification of these kinds of materials as natural or synthetic based on their categories and their origin. Fig. 1 shows that nanomaterials are basically classified into three categories, named nanoparticles, nanoclay laminates, and nanoemulsions [41].

2.1. Nanoparticles

From the figure, nanoparticles can be divided into organic and inorganic nanoparticles based on resource of material made.

2.1.1. Organic nanoparticles

They are synthesized by using natural and artificial organic molecules such as protein clusters, liposomes, micelles, and dendrimers. These nanoparticles are considered to be biodegradable, non-toxic materials, and in some particles like liposomes and micelles have a hollow core and known as nanocapsules which are sensitive to thermal and electromagnetic radiation related to light and heat [42]. These unique properties can open the door to use them in wide potential applications such as drug delivery and intelligent nanosensors [43,44].

2.1.2. Inorganic nanoparticles

They are composed of metals and metal oxides based nanoparticles but they do not contain carbon particles. They are usually shaped by precipitation of inorganic salts, which are linked with molecules by covalent or metallic bonds. Among them, nanogold particles (Au) which are typically used in anticancer therapy [16,45] and mesoporous silica nanoparticles. While metal oxide nanoparticles such as iron oxide (Fe_2O_3), aluminum oxide (Al_2O_3), zinc oxide (ZnO), and titanium oxide (Ti_2O_3) are mainly synthesized to increase their reactivity and efficiency.

2.2. Nanoemulsions

Nanoemulsions are formed between biphasic dispersion of two immiscible liquids, either oil in water or water in oil using a suitable surfactant [46]. They have high solubilization degree and greater kinetic stability than simple micelles or coarse emulsions. They are used in several eco-friendly applications such as cosmetics, pesticide delivery industry [47], parenteral delivery, and oral delivery [48].

2.3. Nanoclays

The most promising nanoscale reinforcing material for barrier behavior is nanoclay platelets because it possesses surface area more than 750 m^2/g and thickness of ~1 nm with lateral dimensions ranging from 100 nm to 500 nm [49], as well as high thermal stability [50]. Aggregation of clays leads to tactoid structure in the polymer matrix (microstructure forms) and reduces barrier efficiencies. Organic treatment of nanoclay by organomodifier, however, is crucial for polymer nanocomposites to obtain high hydrophobic material with increment of d-spacing distance of clay layers.

Thus, polymer/layered silicates could be comprised of three different kinds, namely, i) *flocculated or tactoid microcomposites*, in which the polymer chains cannot penetrate the clay layers and remaining stacked (i.e. poor dispersion because there is no expanding in inter layer spaces of clay), as shown in Fig. 2, ii) *intercalated nanocomposites*, where the polymer chains can penetrate and interlock between the layers and but are still preserved, iii) *exfoliated nanocomposites*, for which the individual platelets are completely separated in the polymer matrix with better nanodispersion [52,53]. This new family of composite materials, especially the latter, shows remarkable enhancements of nanocomposite properties such as mechanical and barrier properties at very small amount (i.e. < 5 wt%), when compared with the neat polymers or classical micro- and macro-composite materials. These are promising materials to be used in nano-food packaging applications.

In our previous studies [26,27], facial and green modification of organoclay (OC) that has d-spacing distance less than 2 nm by antimicrobial natural rosin and stearic acid have been achieved to obtain expanded OC (ROC and EOC, respectively) with d-spacing distance of ~4 nm by using melt blending process, (see Fig. 3), instead of in situ or other dissolution methods that may use carcinogenic chloroform or other organic solvents. In this method, the treated clay and gum rosin were initially mixed together using ceramic mortar with mass ratio (1:1). The mixture was then introduced in an oven at 120 °C for 1 h, where the blend was mixed each 20 min and this step was repeated three times to ensure that the melted rosin intercalated well into clay galleries. Afterwards, it was cooled and grinded into a powder form to be examined by x-ray diffraction. Consequently, this material can be used as reinforcing and compatibilizing agent for two incompatible polymers such as polylactic acid (PLA) and poly (butylene adipate-co-terephthalate) (PBAT) (see Fig. 3).

On the other hand, when OC is incorporated with starch that is derived from renewable and sustainable resources to produce biodegradable Starch/OC bionanocomposites (under Trademark name: Plantic® Plastic Tray). It was found that their mechanical, rheological,

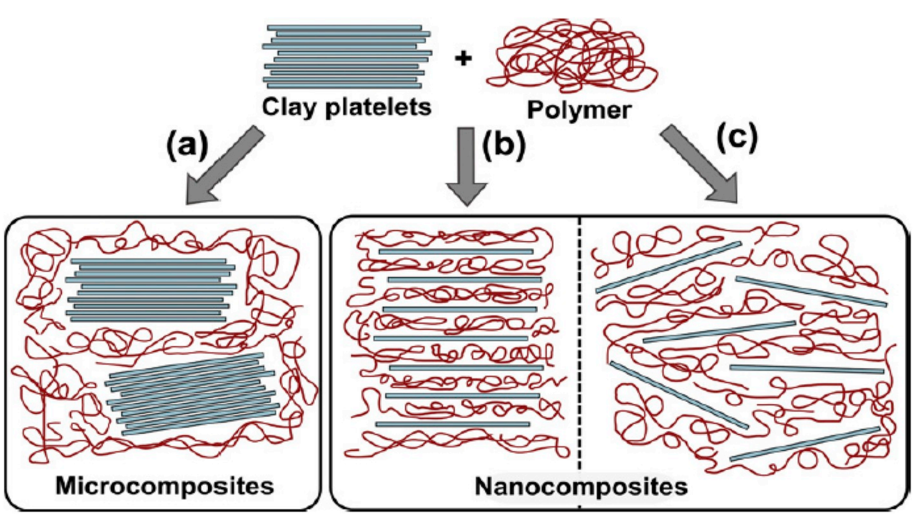

Fig. 2. Flocculated (a), intercalated (b), and exfoliated (c) polymer/layered silicate nanocomposites [51].

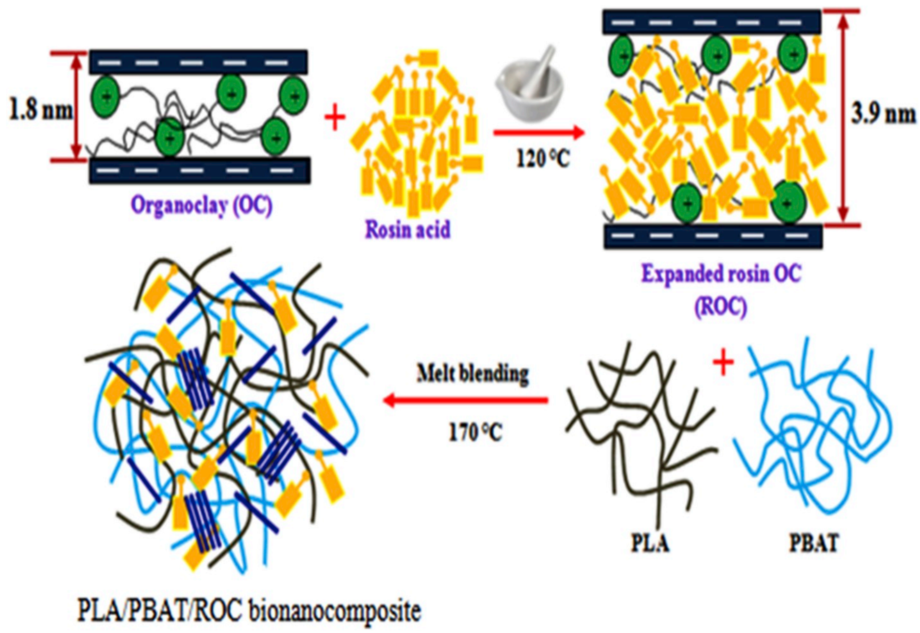

Fig. 3. Schematic representation of green modification of OC by antimicrobial natural rosin and its blending with PLA/PBAT [26]. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

and moisture uptake properties are improved and they were applied in Cadbury®, Dairy Milk, and Mark & Spencer Swiss Chocolate products [5, 54]). Furthermore, the nanostarch-clay has also been used for McDonald's hamburger clamshells in the United States, as an alternative for traditional adhesive [55]. In the same trend, Nylon 6/OC nanocomposites are commercially used in high oxygen barrier packaging for beer and flavored alcoholic beverage [56].

Whereas, nanosilver is applied to nano-food packaging and containers to enhance its antimicrobial behaviors. Huge number of antibacterial nano-packaging products using nano silver and other nanoparticles such as nano-zinc oxide and nano-titanium oxide are readily available on the global market. However, the main drawbacks of these materials are in food-contact (i.e. food contact nanoparticles) that migrate, or that might be predicated to migrate into food, thereby becoming a component of the food, causing a carcinogenicity [5,20,57].

Food and Agriculture Organization (FAO) and World Health Organization (WHO) in 2012 organized a conference entitled "Nanotechnologies in the Food and Agriculture Sectors: Potential Food Safety Implications". They all have reported that the recent activities in food safety management and risk assessment of nanomaterials in the food and agriculture areas shall be taken into account at the national and international level. For this reason, our research focused mainly on lignocellulosic materials derived from natural agriculture resources that currently play a vital role in supporting the nano-food science and packaging, because of their abundantly and nontoxic agro-industrial byproducts.

3. Lignocellulosic materials

Lignocellulosic biomass materials or natural fibers have high impact on several fields of science including engineering and biology agriculture and nano-food sciences because of their availability, low price, and environmentally friendless. They are, moreover, used to designate many types of fibers which are maturely produced from plants, minerals, and animals [58]. They are mainly comprised of three components, namely 1) cellulose (~44%), 2) hemicellulose (~30%), 3) lignin (~26%) (See Fig. 4). Consequently, coffee grounds byproducts are considered as lignocellulosic materials [9,10]). Thus, coffee grounds (CG) have attracted much attention of scientists as "green materials" with many

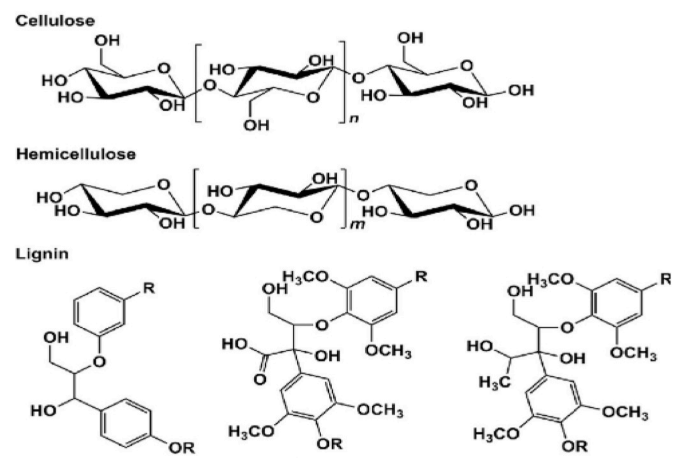

Fig. 4. Chemical block structures of cellulose, hemicellulose, and lignin subunits in lignocellulosic material [59].

potential uses, such as biofuels [60–62], bioenergy [63,64]), or polysaccharides productions (i.e. including starch, chitin, cellulose and chitosan) [65,66].

Currently, they can be used as reinforcing material in biopolymers to produce sustainable green composites with affordable cost [10,20]. However, their hydrophilicity is considered to be a basic barrier to their compounding in polymer matrices due to the poor compatibility between hydrophilic CG phases and hydrophobic polymers, thereby leading to restricted usage as bioreinforcement. Numerous studies have recently been conducted on improving hydrophilicity of CG for material applications [58,67].Among these studies, some focused on compatibilizer or coupling agent addition to the polymer matrix [11,68,69], others on chemical treatment of CG by antimicrobial rosin [[70]] [10]. developed a novel method by using torrefaction process for CG, which is a mild thermal treatment (230-310 °C) under inert gas, with the aim of producing a hydrophobic material, which can achieve their compatibility in the polymer matrix without requiring a compatibilizer.

In latest years, the nanocellulosic materials have attracted the attention of researchers for taking full advantage of the mechanical and barrier properties of packaging materials. Cellulose microfibrils consist of crystalline and amorphous areas that are arbitrarily dispersed along their length. In the former crystalline areas, the cellulose chains are closely packed, while the latter amorphous areas are supplementary vulnerable to chemical or enzymatic attack. The mechanical treatment of cellulose fibers under grinding or homogenization allows the fibrillation of native cellulose fibers to different degrees depending on the intensity of the processing, resulting in microfibrillated cellulose (MFC) or nanofibrillated cellulose (NFC). The nanocrystalline cellulose (NCC) or cellulose nanowhiskers (CNW), is pure cellulose in crystalline form with nanoscale dimensions, that could be managed from different sources of biomass, in moderate conditions of acid or enzyme hydrolysis, to produce powder, liquid or gel forms by the elimination of these amorphous regions.

The resulting NCC has a rigid rod-shaped structure, 1–100 nm in diameter and tens to hundreds of nanometers in length [71]. This results in one of the strongest and stiffest natural materials existing, it displays amazing properties: great tensile strength (7500 MPa); high stiffness (Young's modulus of 100–140 GPa); high aspect ratio (70); large surface area (150–250 m^2/g); and enhanced electrical and optical properties (Revol et al., 1998). This green material has been encouraged for expanded applications, for example, a possible nanofiller for the preparation of industrial composites. Utilization of cellulosic nanofibers in packaging applications will reduce the costs of packed products because of their extensive obtainability and low cost.

The effect of the adding of cellulose nanocrystals on the barrier properties and on the migration performance of poly(lactic acid), PLA-based bionanocomposites were studied in view of the promising participation in food packaging purposes [72]. Besides, the efficiency of cellulose nanocrystal extraction from Phormium tenax leaf natural fibres by acid hydrolysis [72], whereas the possibility to use cellulose nanocrystals extracted from okra "bahmia" bast fibres as reinforcement phase in PVA biodegradable matrix have been reported by Fortunati et al., [72]. Also, Fortunati et al., [73] successfully prepared PVA bio-nanocomposites reinforced with cellulose nanocrystals (CNC) extracted from commercial microcrystalline cellulose (MCC) also use the MCC extracted from two types of natural fibres, Phormium tenax and Flax of the Belinka diversity, were fashioned using solvent casting in water.

Morphological, thermal, mechanical and transparency properties were examined although the individual efficacy of the extraction procedure of CNC from the three sources was assessed. The influence of different CNC types and content on PVA properties and water absorption capacity were also estimated. Natural fibres have higher levels of extraction effectiveness when compared with MCC hydrolysis yield. Thermal analysis evidenced that CNC endorses the crystallization of PVA matrix, whereas enhancing its plastic response. It was illustrated that all PVA/CNC composite systems stay transparent owing to CNC dispersion at the nanoscale, due to saturation after the first 18–24 h of water absorption.

3.1. *Torrefaction process for biomaterials*

The torreffaction of lignocellulosic material plays an important role for enhancing the biomass properties in terms of higher calorific value, hydrophobicity, long-term storage, grindability, and handling [26,74]. The basic use of biomass is to produce bioenergy can be realized by different technologies such as thermochemical (combustion, gasification), chemical processes (esterification), or biological by fermentation [75].The direct combustion of biomass is most commonly used because the treatment includes heating of the biomass material at moderate temperatures from 230 °C to 310 °C under inert atmosphere as in short term. The mass loss of the material during the torrefaction step relies on the torrefaction conditions such as heating rate, final temperature, and holding time.

Additionally, the reduction of O/C and H/C ratios can candidate the torrefied CG to use in the production of syngas [76–78]. It can be concluded that torrefaction is a promising operation to improve not only the performance of biomass and wastes for green energy applications, but also a quick treatment for agro-industrial wastes to be used as bio-renforcing agents in the polymer matrix without requiring a compatibilizer. Based on our previous work [27]), when adding 10 wt% torrefied CG (treated at 250 or 270 °C) in PBAT matrix, the tensile strength value obviously increased to ~17.6 MPa and 18.2 MPa, respectively, compared to virgin PBAT or PBAT composites based on untreated CG (Fig. 5). This perspective technique can open many opportunities to use agro-by-products in safe food packaging applications, thereby leading to reduce the environmental hazards resulting from their burning.

4. Biopolymers based lignocelluloseic materials

Non-biodegradable polymers have caused serious environmental issues because of their inappropriate discard. Recently, biodegradable and renewable biopolymers become the first request and the area of great interest for researchers, because they are 100% biodegradable polymers and have UV light protection (Fig. 6). Besides, these materials have emerged as potential candidates for unique applications such as smart food packaging [81,82], healthcare [83], biomedical purposes [84], and their wastes can be used as industrial composting [85].

Biopolymers are broadly classified into two types based on their origin, i) *natural* biopolymers such as starch, chitosan, cellulose, agar, soy proteins, corn zein, and so on, ii) synthetic biopolymers such as poly vinyl alcohol (PVA), polylactic acid (PLA), polybutylene adapate-co-terephthalate (PBAT), and so on. Nevertheless, these biopolymers have generally poor mechanical, lower heat distortion temperature and barrier properties, beside their high cost; thereby these obstacles limit their uses in engineering and industrial sectors [86].To restrict these drawbacks, a reinforcing material shall be added to enhance the biopolymer properties to meet not only desired applications, but also reduce the final cost of the product. Montmorillonite (MMT) and Layered double hydroxide (LDH) are often used as reinforcing material in polymer matrices and played a potent role to improve their properties such as thermal, barrier, mechanical, and flame retardant properties.

[27] reported that a significant improvement in thermo-mechanical and barrier properties of the PBE/PBAT/EOC bionanocomposites were observed at a small amount of expanded OC (typically < 5 wt %). Xie and co-workers [87] have fabricated PBAT/OLDH films for food packaging, where these films exhibited excellent water barrier properties as compared to unfilled PBAT film. Regarding food contact materials, some regulations and limitations for using these kinds of nanomaterials in the edible food packaging shall be taken into consideration. For this reason, lignocellulosic nanofibers such as cellulose nanocrystals (CNC) or cellulose nanofibers (CNF) (e.g. Kenaf, sisal, and jute) have become appealing materials for green food packaging and energy storage applications. This is because they are the second most abundant biomass materials in earth. The main advantages of biodegradable nanofillers extracted from renewable sources in comparison with inorganic fillers are their reinforcing capability, low energy consumption, low density, and high mechanical properties. Furthermore, cellulose monomer has six hydroxyl groups that are bonded together by hydrogen bonding which plays a superior role in crystalline packing and in governing important physical properties of this highly cohesive material [88].

All these features can open the door to use these materials in many promising applications. Thus, they can incorporate with biopolymer to produce novel, high efficient and eco-friendly composites with excellent processability and properties. Consequently, numerous efforts have been done to prolong the shelf life of food packaging material and overcome the environmental issue of handling package wastes leading to a great interest in biodegradable composites [73,89]. For instance, chitosan/CNC/ZnO films as packaging material are made to improve of

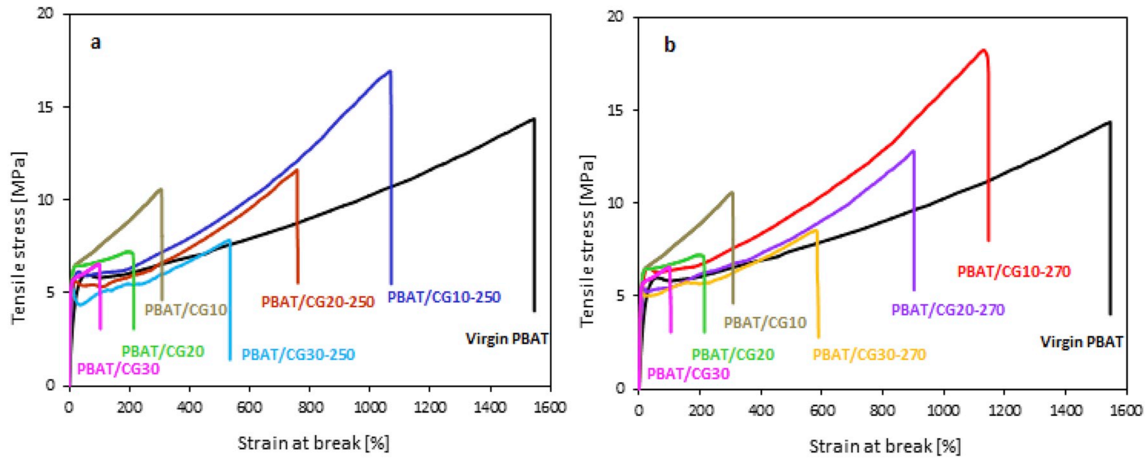

Fig. 5. Stress-strain curves for virgin PBAT and its filled composites with different ratios of untreated CG and torrefied CG at 250 °C (a) and at 270 °C (b) [79]).

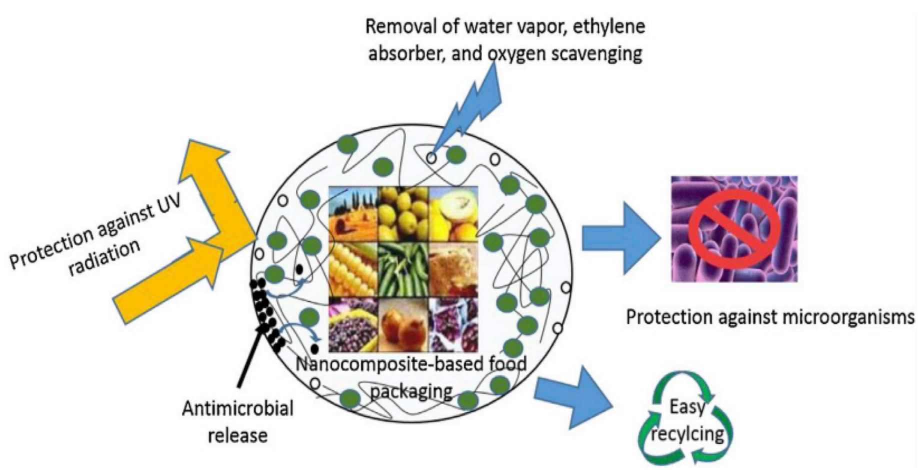

Fig. 6. Functionality of polymer nanocomposites-based food packaging offers considerable advantages in protecting food (e.g. UV light and microbes) [80].

Egyptian soft white cheese shelf-life time [90,91]. In the same senses, chitosan/whey protein as coating film for cheese and fresh dairy products to keep their flavor constant with long storage time [92].

Moreover, the surface of CNC is grafted by natural rosin as a green process to obtain antimicrobial packaging material [70]. Whereas, chitosan/coffee grounds (CG) composites are used as adsorbent to remove the pharmaceutical contaminants from wastewater [93]. Correspondingly, other thermoplastic biopolymers such as PLA/PBAT blend are incorporated with expanded OC by antimicrobial natural rosin to produce high quality bionanocomposites for nano-food industry [94]) [95]. developed a "lamellae barrier walls" from PLA/TMC/graphene nanocomposite to obtain high gas barrier nanocomposites for use in superior barrier applications such as beverage bottles, food packaging and fuel tanks. Thus, it can be summarized that biopolymers based lignocellulosic material have made a major breakthrough in the food science regarding toxicity-free material and migration less properties.

5. Green packaging materials

The basic role of packaging material is to protect food goods or packaged artifacts from outside impacts and damage, and to provide consumers with the right constituents and nutritional needs [96]. It can be also explained as a coordinator regime of preparing goods for safe, efficient and effective handling, transport, distribution, storage, retailing, consumption and recovery, reuse or disposal combined with maximizing consumer value [97,98]. As it is known, food packaging materials were classified on basis of their category into: glass, metal, papers, and plastics.

In this review, we focus only on the plastics as package material. But there are two types of plastics, namely thermosets and thermoplastics. The former are polymers alone or blended with other materials that set irreversibly when heated and cannot be recycled or re-molded. Because they are rigid and durable, they tend to be used primarily in automobiles and construction engineering applications such as adhesives, coatings, otherwise in kitchen utensils, not in food packaging purposes. The later, are polymers that are flexible and soften when exposed to heat and return to their original conditions at ambient temperature. Because thermoplastics can be easily shaped and molded into various products such as plastic films, bottles, and jugs they are ideal for food packaging. Furthermore, substantially all thermoplastics are reusable when melted to produce new products, although the separation poses some practical limitations for certain products as they will always have inferior properties compared to the non-recycled original product. Packaging materials that are fabricated from plastics have a large range of barrier behaviors, but they are generally more permeable than metal or glass packages.

In order to enhance the permeability properties of plastics especially as smart packaging materials, eco-friendly or green reinforcement materials from micro-to nano-scales shall be added to them. Biopolymers and bioreinforcing agents have been promising substitutes to be exploited and established into eco-friendly food packaging materials attributable to its biodegradability [99]. Several efforts have already been done in the field of green packages not only to be versatile enough to withstand handling process, high quality and safe, as well as sufficient for barrier properties to water and gases, but also to comply with international legislations and regulations.

Today's food green package material is often combining several components to exploit each material's functional or aesthetic properties. In this case, most food organizations are recommended to use eco-friendly and green additives for biopolymers to save the consumers from the risks of non-biodegradable or conventional packages. This drives us to talk about intelligent or active packaging (i.e. functional packaging) and antimicrobial packages.

6. Intelligent packaging

Intelligent packaging based on green materials can be defined as packaging that includes an external or internal indicator to provide information about aspects of the history of the package and/or the quality of the foodstuffs [100]. Intelligent packaging is an extension of the communication function of traditional packaging and communicates information to the consumer, and provides by barcode labels, biosensors, time temperature indicators, and gas indicators to be able to sense, detect, or record external or internal changes in the product's zone [101]. The main advantage of intelligent packaging is nowadays its design that is leading the way of packaging innovations because it has the time-temperature indicator [7]. This indicator is useful because it can inform the consumer when the foods have been abused. This means, for example, a food is subjected to a higher temperature more than that recommended for storage or preparation; the quality of the food can deteriorate much quicker. The time-temperature indicator can also be placed on shipping packages or containers as a small self-adhesive label, and an irreversible change, as a color change, will result when the indicator experiences bad conditions. This type of indicator is mostly useful with chilled or frozen foods, since the cold storage during distribution and transportation is important for food quality and safety [102]. In some cases, the inks can be used as smart packaging solution in the packaging material when their colour changes at high temperature and gives a message to the consumer about the time to consume the food, thanks to packaging innovation and technology.

6.1. Antibacterial food packaging

Despite these benefits, there is still relatively narrow insight regarding food decay and rot during handling process and transportation between countries. Sterilizing food process is not sufficient to extend shelf life and maintains food in safety scale. For this reason, antimicrobial materials added to bioplastics that used in the manufacturing of food packaging material to fulfill and improve its antimicrobial property. Consequently, polymer composite based antimicrobial packaging has to be able to prevent the growth rate of microbes and fungi on the package surface, thereby leading to prolong food shelf life and keep food more safety at severe conditions such as storage and handling conditions.

The most commonly used antimicrobial materials in polymer biocomposites involve metal/metal oxides, organoclay, natural biomaterials such as rosin, synthetic antimicrobial agents, and enzymes [103–105]. The incorporation of metal/metal oxides such as nanosilver and nanozinc nanoparticles are the most remarkable studied in the literature and offered strong antimicrobial activity against many bacteria (either Gram-positive or Gram-negative bacteria) and fungi [106–109]. For instance, when polyamide-6 was charged with 2 wt% of Ag-nanoparticles and applied against *E*-coli bacteria, the results showed PA6/Ag-NPs has strong efficacy on the bacteria and can used as antimicrobial material for long-term applications [110].Despite these advantages, there is a critical problem in food packaging that is the migration of nanoparticles from package material to food because of their miniscule dimensions [111].

Accordingly, the main concern on the application of nanoparticles in the food contact packages is regarding the indirect exposure because of the potential migration of nano-scales to food. As a result, FAO and WHO requested that the materials used in food packaging shall be eco-friendly safe, including antimicrobial materials to maintain the food in safe and without causing any impacts on human health [6,113].

7. Biodegradability of biopolymers

Biodegradable food packaging is becoming the first demand and the area of much interest for researchers, because it is 100% biodegradable material when compared with classical petroleum-based polymers. It can be decomposed upon disposal in bioactive medium by microorganisms like bacteria, fungi, algae, or by marine water to H_2O, CO_2, and humus. Biodegradability in biopolymers can be achieved by aerobic or an-aerobic fermentation in the presence of microorganisms which secrete extracellular enzymes to fragment the polymer chains in the packaging material to small molecular weight decay products (Fig. 7). One of our previous studies [27] reported that the biodegradability of PBE/PBAT bionanocomposites was performed in fresh water from the River Nile, Egypt, and marine water within 20 days. The results showed that a high biodegradability rate was noticed for freshwater when compared to marine water. The reason might be due to the microbial effect by microorganisms and algae in freshwater that could attack the polymeric chains and cause polymer chain cleavage [27]; Moustafa et al., 2017e).

8. Future vision and conclusion

A bright vision future might be anticipated for green packaging and intelligent food packaging. The use of intelligent or smart packaging will probably become more popular as more technologies make their way to the global market, innovative packaging in smart regimes will become more common. Eco-friendly polymer composites based on lignocellulosic biomass offer considerable new innovations and exciting opportunities for the safety and quality of the foods. The incorporation of few amounts of biodegradable materials into polymer matrix produces a high performance biocomposite that can be used as biosensors, time temperature indicators, or gas indicators. This enables intelligent food technologies to sense, detect, or record internal or external changes in the product's area, thanks to these technologies. Some experts believe that the next stage of technology in food packaging will include nanomaterials that will allow novel biocomposites such as new antimicrobials and gas scavengers to be involved in packaging films.

Regarding food degradation by microorganisms, antimicrobial natural smart food packaging is highly recommended by food organizations

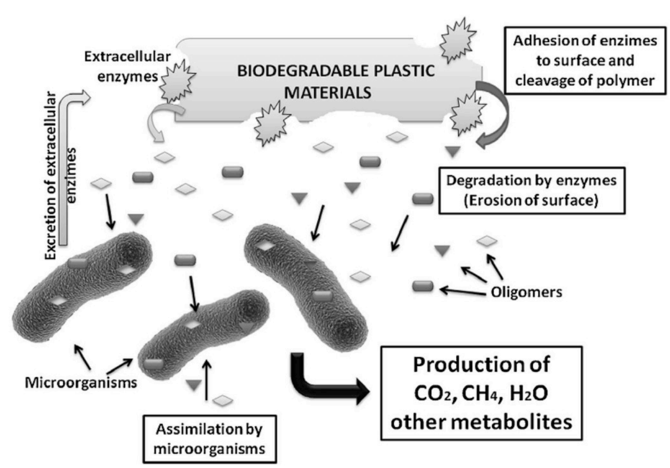

Fig. 7. Biodegradability mechanism in biopolymers [112].

nowadays. The major reason is due to the migration of classical antimicrobial agents or metal oxides from the package material to foods. Consequently, some major parameters for future vision and expectations have been considered:1) price or cost effectiveness of food package material manufacturing as an issue which needs to be dealt with in advance, as expensive equipment are often involved, 2) eco-friendly additives and agents are recommended for green package material, 3) EU regulations and legislations for food package material must be applied and activated not only in the industrial world, but also in the developing countries to meet high level of food safety and transparency to consumers.

In conclusion, green package material is characterized by high tensile strength and stiffness, inexpensive, low density and produce low CO_2 emission. Besides, it is non-toxic as food contact material and 100% naturally biodegradable. As a result, the torrefaction process is highly recommended in green packaging. All these features can open the door to use in a variety of disciplines. Nowadays, the scientists dedicate their efforts on the use of biocomposites in automotive manufacturing because they are low density and produce low CO_2 emission on burning. On the other hand, when most vehicle components are assembled from low density biocomposites this leads to reduce CO_2, save fuel, and cost. Thus, the relation between vehicle weight and CO_2 emission is going to be a potential trend by 2020. There is still narrow insight regarding innovation, development, fabrication, and manufacturing of food package which includes storage, shelf life, and design that has an effect on the food quality and their export because it fails to abide to EU legislations.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.compositesb.2019.05.048.

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