REVIEW PAPER

Advancements in eco-friendly food packaging through nanocomposites: a review

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Abstract

Presently, the food industry continues to be major contributor of plastic waste across the globe. The food industry is in great demand of sustainable food packaging material due to increase environmental and health consciousness. Biobased polymers which are degradable promise to be a key solution. Use of nanotechnological advancements in biopolymers provide a wide range of additional benefit and makes them a suitable packaging material. The benefits include barrier to oxygen permeability, moisture permeability, crystalline structure, other barrier properties, morphology, thermal stability, optical properties, mechanical properties, improving shelf life of the packaged food and antimicrobial characteristics. Polymeric materials (matrix) have been infused with nanofillers such as silicates, carbon, cellulose and starch nanoparticles. This review aims to focus on new and effectual polymeric materials for food packaging that are technologically advanced with nanotechnology as a solution to the challenges faced by food industry with regard to the product safety and materials performance.

Keywords Nanoparticles · Biopolymers · Nanofillers · Properties · Eco-friendly · Food packaging · Nanocomposites

Introduction

The increased concern of plastic wastes across the globe is simultaneously increasing the demand for eco-friendly materials. Food packaging is one of the major contributors of the plastic waste. Hence the need for biodegradable polymers has increased. Although biodegradable polymers can be utilized in food packaging, to make it a suitable replacement for plastics enhancement of properties is needed

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where nanotechnology plays a major role in improving the properties of eco-friendly polymers. It is a well-known fact is that the food packaging material must protect what it sells and sell what it protects. A food packaging material is highly essential to have suitable properties to ensure that it preserves the quality of the food and also does not produce any effect that may detoriate the packaged food for a suitable period of time. In recent times, nanotechnology is analyzed widely for its applications in the food packaging which is being widely studied to improve production processes in order to achieve products with better characteristics, enhanced quality as well as functionalities in food protection and food storage [1, 2].

Nanotechnology enables the creation of novel properties and phenomena through nanometre scale dimensions and large surface-to-volume ratio [2]. These characteristics prove optimal for applications involve food composite materials, controlled and immediate release of substances in active and functional packaging technologies and energy storage in intelligent packing [2]. Nanotechnology contributes to achieve high quality of food products or food components. Every requirement of food safety such as controlling microbial growth and delaying oxidation is being addressed with the help of nanotechnology in the food packaging aspects [3]. Along with monitoring the conditions of food during transport and storage, nanotechnology provides new food packaging materials with improvement, in terms of both performance and properties such as mechanical properties, barrier properties and antimicrobial properties.

Biopolymer nanocomposites incorporated with nanosized particles are a novel class of composite materials [4]. Biopolymer nanocomposites consist of biopolymer matrices and fillers with at least one dimension within 1–100 nm. Certain traditional composites will not be able to go on par with the polymers containing nanosized particles as they exhibit unique combinations of properties [4]. The properties of the entire polymer matrix are slightly changed by novel fillers along with inculcation of new functionality because of their chemical composition and nanosized components [5]. Recently, the application of biopolymeric nanoparticles, viz. silica, cellulose and starch nanoparticles in food packaging, was found to be of great importance. Its wide range of benefits, such as improvements in barrier, mechanical and antimicrobial properties and performances, is the major applications of biopolymeric nanoparticles in food packaging.

Recent researches are addressed to the setup of new food packaging materials, in which biopolymer nanocomposites incorporate nanoparticles [5]. Silica nanoparticles and cellulose nanomaterials have been employed in various branches of food and nutrition sectors, and however, these particles have not been used as an ingredient of food products. This report states the application of nanosized particles of silica, silica derivatives and cellulose nanomaterials in food packaging. In addition, starch is one of the versatile source of nanoparticles which is also found to improve the properties of the matrix when incorporated as the nanofiller by preserving the quality of the food for a longer duration of time and the interesting fact is that the starch nanoparticles incorporation catalyzes the rate of biodegradation of the polymers making it eco-friendly which is the need of the hour [6].

All of the above-mentioned applications of the nanosized particles result in products with better quality, characteristics, or functionalities in food industries

in comparison with employing of larger particles with same composition. However, a number of important issues are to be taken in consideration for the application of nanotechnology in food packaging, viz. safety concerns, due to possible migration of nanoparticles from packing materials into foods and their eventual toxicological effects [3]. On the other hand, there is always a possibility of nano-material migration from packaging (or) containers to food stuff states previous studies [3], 4. Materials that are less than 300 nm or less in diameter can be taken up by individual cells [7] and nanomaterials less than 70 nm can even be taken up by a cell's nuclei where they can cause major damage [8, 9].

Considering all the safety measures and optimizing the conditions of the nano-particles usage can provide a better future in the food packaging sector by its association with nanotechnology. There are reports where many chemically synthesized nanoparticles were utilized for the fabrication food packaging material but because of the toxicity and non-biocompatibility these nanofillers should be replaced with biogenic biopolymeric nanoparticles such as starch, silica, cellulose, chitosan and polyhydroxyalkanotes (PHAs). Therefore, the main focus of the review is to review and highlight the potential biocompatible, thermomechanical and biodegradable biopolymeric nanoparticles which can be utilized for the fabrication of food packaging bionanocomposite films with enhanced properties and can be used as a sustainable solution to the challenges faced by food industry. Some of potential biopolymeric nanoparticles are starch, silicate, cellulose and PHAs which can really do wonders in the food packaging industry due there versatility.

Starch nanoparticles in food packaging applications

Starch is a renewable carbohydrate-based material, and it is an abundant biopolymer found in nature synthesized in plant amyloplast to serve as an energy reserve for the plant. Starch molecules are the polymers of the glucose as its units. The constituents of starch are the amylose and the amylopectin which are linear molecule linked by y-(1–4)-d-glycoside bonds and branched molecules linked by -(1–6) linkages, respectively [10]. The relative amount of the constituent amylose and amylopectin present in the starch depends on the source from which it is collected. The ratio of these two constituents plays a major role in determining the property of the starch [11]. The starch that is used for these industrial applications can either be used as a "native starch" as it is extracted from the plants or it can be used in a modified way as "modified starch" by making the starch undergo certain chemical modifications to attain a specific property as required for the usage. In nature, starch is widely present as a stored energy source for plants. The main source of starch maize, cassava, potato and wheat [12] and is been a topic of interest in the recent days for its food packaging applications either as polymeric source or for its nanotechnological applications due to its diversified presence as it is the second most abundant biomass in the nature as well serving as an affordable source to produce an eco-friendly food package.
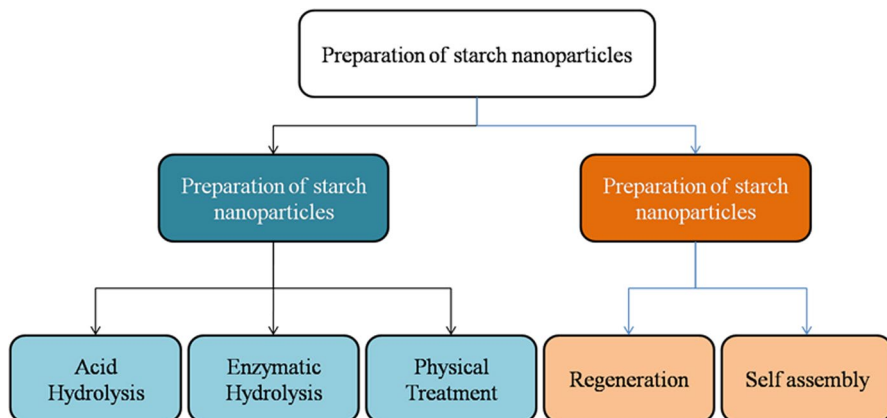
**Fig. 1** Representation of the classification of methodology for the preparation of starch nanoparticles
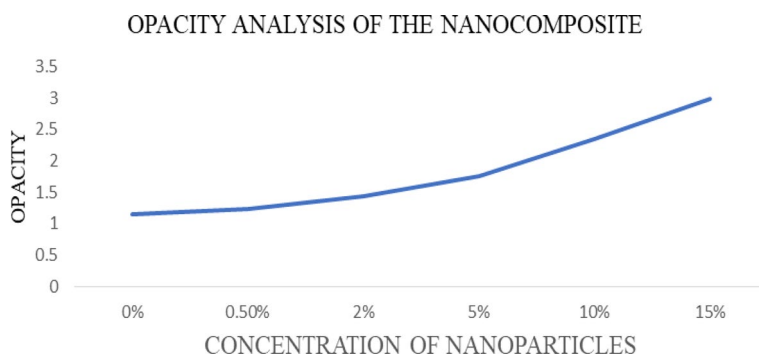
Fig. 2 The concentration of nanoparticles, expressed as a percentage, and the opacity, expressed in absorbance units per unit thickness, were obtained from data reproduced from [21]. The experiment was conducted using a UV–Vis spectrophotometer at a wavelength of 600 nm, with air used as a reference

Synthesis of starch nanoparticles

Acid hydrolysis

Acid Hydrolysis is the widely used method for the preparation of starch nanocrystals due to the simplicity in its procedure (Figs. 1, 2, 3, 4, 5 and 6).

The acid hydrolysis occurs in two steps which include an initial fast hydrolysis followed by a slow hydrolysis. The initial stage is the hydrolysis of the amorphous part of the starch which occurs in a rapid way whereas the slow stage is due to the erosion of the crystalline region of the starch [12]. However, some authors suggest a three stage of hydrolysis which are categorized as the fast stage, slow stage and the very slow stage. Usually crystalline regions are comparatively more resistant to
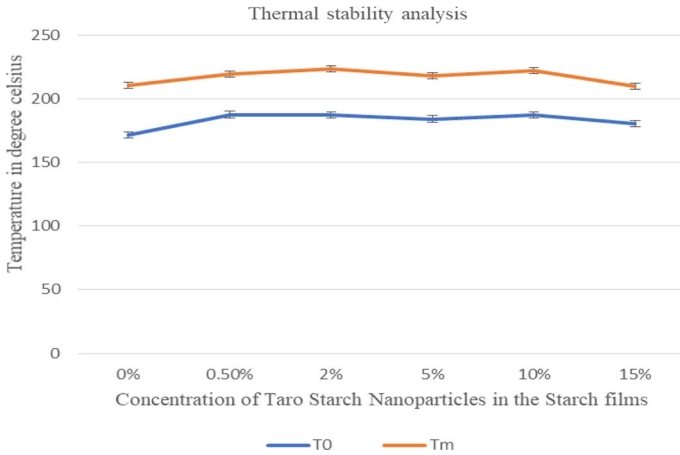
Fig. 3 The figure shows the concentration of nanoparticles expressed as a percentage, and the onset and melting temperatures expressed in degrees Celsius, based on data from [21]
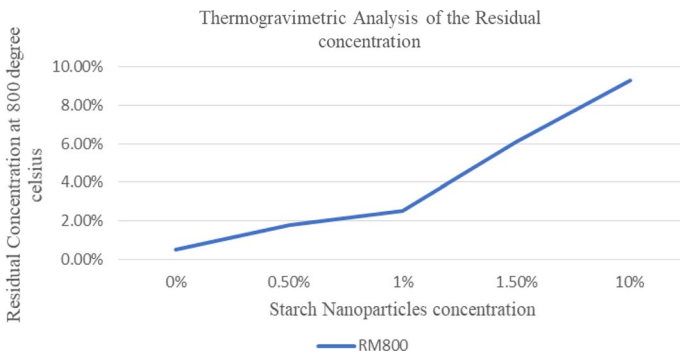

Fig. 4 The concentration of nanoparticles is expressed in percentage and the residual mass at 800 degrees Celsius is expressed in percentage, reproduced from thermogravimetric analysis data [26]

acid hydrolysis than the amorphous regions and hence it is usually isolated by a mild hydrolysis using the acids such as Sulfuric or Hydrochloric acid.

Enzymatic hydrolysis

The enzymatic hydrolysis is a method that is usually used along with other methods of preparation for the attainment of maximal efficiency. The enzymes that are used for the hydrolysis are divided into two sub categories which include the amylases and the transferases. The amylases are utilized for the purpose of cleaving the glucosidic bonds, the amylase enzymes are further sub-divided into three groups as; endo amylase that cleave the internal bond, the exo amylase that cleaves the bonds in the non-reducing end and the debranching enzymes to cleave the branches to create a
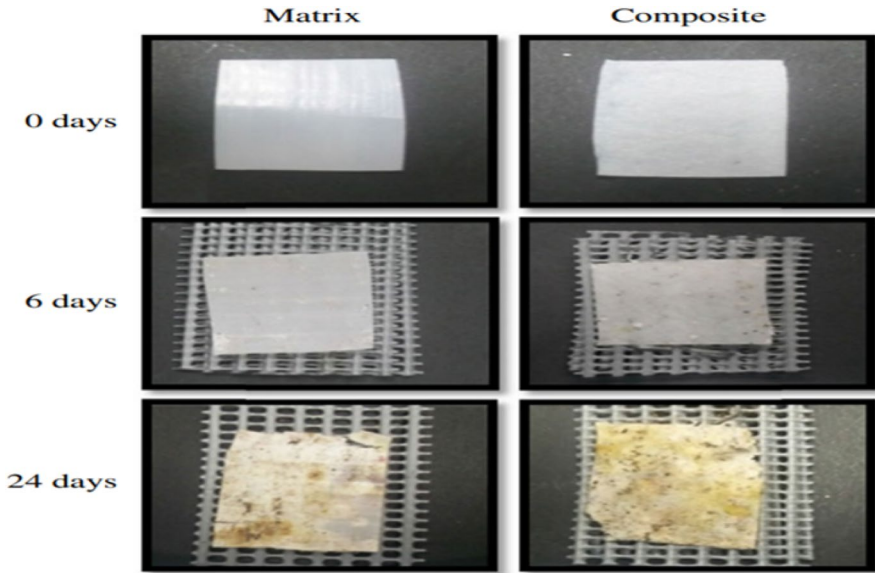
Fig. 5 Experimental result of the biodegradability test of control and nanocomposite. Adapted from "Influence of incorporation of starch nanoparticles in PBAT/TPS composite films" by Paula González et al. [27]

linear polymer of starch [13]. The enzymatic hydrolysis also follows a similar two stage process such as in the acid hydrolysis.

High pressure homogenization

High pressure homogenization is carried out with a microfluidizer by manipulation of the liquid to move through the microfabricated channels of the homogenizer. The homogenizer tends to break the hydrogen bonds inside the particles by the mechanical shear forces created in the homogenizer. This methodology is a simple and highly adopted versatile method that is widely used in the industries but due to lower quantity that can be processed the yield is less. As an example from an experimental data it can be seen that when 5% of the starch slurry was passed through a homogenizer at 207 MPa pressure for 20 times the particle size of the starch was reduced from 3–6 μm to 10–20 nm [14].

Ultrasonication

Ultra-sonication is the process of utilizing the ultrasound waves, to which the particles are subjected and broken down into smaller sized ones. Ultrasound waves have a frequency above the normal human hearing range. The ultrasound waves are generated by the piezoelectric or magneto strictive transducers that create high energy vibrations which are then amplified using the transducers and transferred to a probe which is in direct contact with the starch sample [14]. An experimental data shows
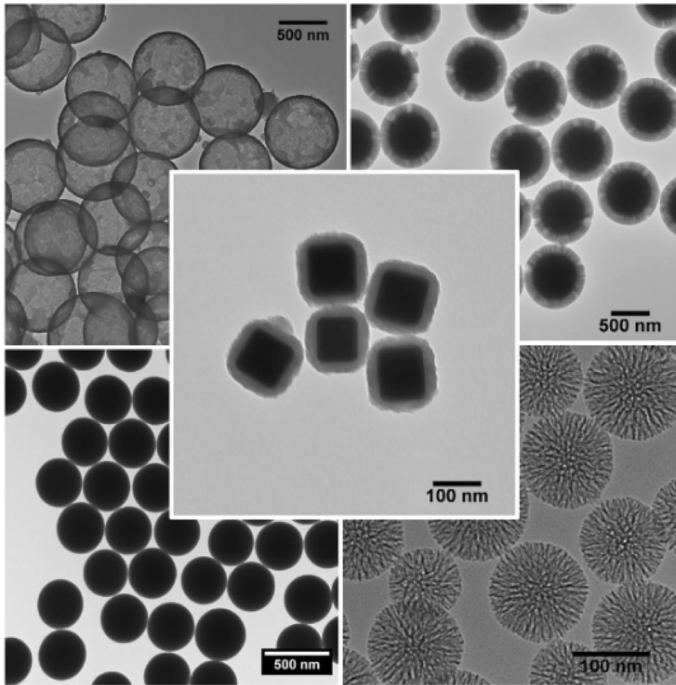
Fig. 6 These images feature 500-nm hollow silica nanoparticles (upper left), 500-nm solid silica nanoparticles with mesoporous shells (upper right), 300-nm solid silica nanospheres (lower left), 100-nm mesoporous silica nanoparticles with hexagonal pores (MCM-41; lower right) and silver nanotubes coated with mesoporous silica shells (center) [31, 32]

that 100 ml of waxy maize suspension with 1.5% of solid content was taken and sonicated at 80% power output for a time period of more than 75 min which resulted in a reduced particle size from micrometers to 100 to 200 nm after 75 min [15].

Gamma radiation

Gamma radiation is a rapid technique which can fragment the molecules by cleaving the glycosidic linkages. Gamma irradiation results in the release of the free radicals which causes the cleavage of bonds. An experimental data suggest that the gamma irradiation resulted in the formation of nanoparticles of approximate size of 20 to 30 nm with an input dosage of 20 kGy waxy maize. The disadvantage of this method is that the starch nanoparticles obtained from gamma irradiation method is more susceptible to thermal degradation due to a larger number of hydroxyl groups in the surface of the particles; however, there is an alternative theory that this susceptibility to thermal degradation is not because of the gamma irradiation instead it is caused due to the pre-heat treatment prior to the irradiation to obtain a stable dispersion [14].

Regeneration strategy

Regeneration is a bottom up approach to prepare the starch nanoparticles, it is usually utilized to produce the amorphous starch nanoparticles. Co-crystallization or complex formation with other components is one of the methodology utilized in regeneration procedure. In this method the starch particles are initially dissolved in a aqueous solvent such as DMSO and then heated for a particular time period of about 1 h followed by which it is stirred for a period of 24 h and it is subjected to a membrane filter such as PTFE of suitable size in nanoscale into a solvent such as n-butanol. The precipitate can be collected by centrifugation and then the selective crystalline components are isolated by the hydrolysis procedure to remove the amorphous part of the nanoparticles [12]. Since most of the nanoparticles formed involves amorphous component, the yield by this procedure is low than compared with other methods.

Self-assembly

A short glucan chain self-assembly is an effective method that is utilized to prepare starch nanoparticle that are thermostable by glucan chain self-assembly at a temperature of 50 degree Celsius. An experimental data show that 15 g of waxy maize dispersed in a citric acid and disodium hydrogen phosphate buffer was gelatinized by heating, stirring, incubating and then by the addition of ethanol as a solvent in dropwise manner, which resulted in a short glucan chain powder by freeze drying, and the short glucan chain powder was then dispersed in deionized water and autoclaved to attain the complete gelatinization. The aliquot solution of the gelatinized component was passed through a nylon membrane of 0.25 μm. This method resulted in the yield of starch nanoparticles ranging between the Size of 30 nm to 40 nm [16]. This method was shown to enhance the relative crystallinity of starch nanoparticles which can be suitable for preparation of nanocomposite films.

Enhancement of packaging properties with starch-based nanocomposites

The nanocomposites are the material which is composed of minimum two components of which one component has a size reduced in the nanoscale and enhance the property of the packaging material.

Composites are the materials that are composed of two basic components:

- Matrix- The matrix plays the role of transmitting the load to the fillers as well as to protect and support the fillers.
- Fillers- The components that are reinforced in the matrix to enhance the properties of the matrix.

The matrix that is widely used is the synthetic polymers which have a versatile applications but it is leading to a greater environmental issue, since most of the

synthetic polymers are non-biodegradable. The solution for this issue is the eco-friendly polymers such as starch, pullulan, polylactide and soybean protein. By incorporating starch nanoparticles into these polymers, it enhances its properties. The nanocomposites are superior to the conventional composites in a way that it can enhance with added benefits of thermal stability, barrier properties, increased water vapor permeability, opacity, thickness etc. [14]. All these enhanced properties are suitable for making the food packaging with added benefits, thus making the food storage and transport in a possible effective way by improving the shelf life of food and preserving the quality of the food.

Barrier property

One of the vital factors to maintain the quality of food is the barrier property which includes barrier from gases, aroma, light and water vapor. This is an important requirement that has to be met by the food packaging system to ensure the quality of the food that is being packaged [17]. Most of the studies suggest that the barrier property of starch nanoparticles-based nanocomposite had an increased barrier effect to oxygen and water vapor than the other alternatives. Some studies suggest that similar to that of the nanoclays the Starch Nanoparticles are creating a tortuous pathway by creating compactness in the polymeric structure by the strong interaction of the nanoparticles and the polymer for the travel of oxygen and water vapor thereby serving as an effective barrier [6]. Some researchers have mentioned that the increase in the concentration of the fillers created a proportional decrease in the water vapor permeability while some other research works suggested that the conventional composites and the nanocomposites with 20% nanocrystals exhibited no difference in the barrier properties but an increase in the percentage to 30 to 40% resulted in a decrease in the water vapor permeability, this kind of observation can be explained as the minimum required concentration to produce a tortuous pathway for the water vapor and oxygen. However, this was contradicted in a case where 2.5% starch nanoparticles in a waxy maize-based polymer decreased the barrier property to about 40% which shows that not just the concentration is a factor but also the way starch nanoparticles are dispersed in the matrix plays a major role [12].

A detailed analysis demonstrated that different polymer matrices needed different levels of filler content to serve as a barrier for the oxygen and water vapor (Tables 1 and 2), in the case of pullalan matrix a concentration above 20% is needed [18].

Table 1 Water vapor permeability analysis of the polymer matrices with nanofillers

S.No	Type of polymer matrix	Optimal concentration to enhance barrier vapor permeability	References
1	Waxy maize starch-based polymer	2.5%	[12]
2	Pullalan matrix	Above 20%	[18]
3	Carboxtmethyl chitosan polymer	Significant increase up to 20%	[19]
4	Natural rubber	Significant increase up to 10%	[20]
5	Corn starch	Significant increase from 10 to 40%	[21]

Table 2 Mechanical property analysis of the matrices after nanofillers are added

S.No	Matrix	Nanofillers	Nanoparticle concentration	Tensile strength	Nanoparticle concentration	Elongation at break	Reference
1	Corn starch-based polymer	Taro starch nanoparticles	10%	Increase, value -2.8 MPa	ND	Decrease	[21]
2	Polyvinyl alcohol film	Pea starch nanoparticles	5%	13.5% increase	20%	7.14% decrease	[23]
3	Starch film	Potato starch nanoparticles	6%	41.33% increase	12%	34% decrease	[25]

In the case of carboxymethyl chitosan, there was a significant decrease up to a concentration of 20% after which the levels remained constant [19] this was suggested to be due to the formation of aggregates above the marked level, the barrier properties osf starch nanocrystals obtained by acid hydrolysis of waxy starch was better when prepared with a nanocomposite film made of natural rubber, this finding was specially attributed to the starch nanocrystals which are like platelets with a thickness of 6–8 nm, a length of 40–60 nm and a width of 15–30 nm tend to block the migration of the oxygen molecules through the nanocomposites, it showed a continuous reduction in permeability with increase in the concentration with a marked level of 10% concentration of the nanofiller [20]. Taro starch nanoparticles that were obtained by the enzymatic hydrolysis with pullulanase and recrystallization process was used in the preparation of the nanocomposite by reinforcing in the corn starch films, the research showed that the water vapor permeability of the nanocomposite film decreased significantly with an increase in the nanofiller concentration, the report was made that with a concentration of about 10% there was an increase in the water vapor permeability, whereas when the concentration increased above 30% to 40%, the water vapor permeability began to decrease which was suggested to be due to the formation of aggregates [21].

Hence it can be seen that it is universally accepted fact that starch nanocrystals has a good range of barrier properties; however, the concentration of the nanofiller used with respect to the polymers in which it is reinforced as well as the dispersion of the nanofillers in the polymer matrix serves as a major factor that has to be considered. When these factors are maintained in an optimal levels the starch nanocrystals can be used as an excellent nanofiller agent for the Food packaging polymers by serving as a barrier for the oxygen and water vapor, which ensures the quality and shelf life of the food product during transport and storage.

Opacity

Opacity is an important criterion and provides an advantage in the packaging of the light-sensitive and UV-sensitive food products such as dairy products, meat and meat-based products, wine and vegetable oils which are all protein and fat rich sources. Photo-oxidation is a major reason for the detoriation of some food products and sometimes it even leads to the formation of the toxic compounds in the food product as well as decrease in the nutritional value. The presence of oxygen and light can lead to the photosensitization reaction which causes the liberation of the reactive oxygen species (ROS) as radicals of oxygen such as superoxide anion and hydroperoxy radical, as well as non-radical derivatives such as ozone and singlet oxygen leading to undesirable chemical changes in the food products that are packed. Studies have shown that the photo-oxidation reactions where decreased when light transmission was lowered by the packaging material [22]. Engineered Nanomaterial can help in this intelligent packaging by serving as a light-sensitive packaging material for the food products.

A research work demonstrated that the corn starch films prepared by incorporating taro starch nanoparticles as nanofillers was measured for its light absorption in a wavelength of 600 nm by using the UV spectrophotometer and it was reported that

the increment of concentration of taro starch nanoparticles from 2 to 15% showed an increase in the opacity and the highest opacity was seen in 15% concentration which can be due to the nanoparticles getting embedded in the interspaces leading to less transmittance of the light, it was also seen that vacuum freeze-dried nanofillers showed a further increase in the opacity which could be due to the agglomeration of the nanoparticles [21].

The nanocomposite prepared by blending pea starch nanoparticles with polyvinyl alcohol demonstrated a decrease in the light transmittance. The light transmittance study was made in the films where the nanoparticle concentration was increased from 5 to 25% which resulted in a decreased transmittance of light through the nanocomposite [23]. Hence, the opacity of the nanocomposite increased with an increase in the concentration of the starch nanoparticles, which provides a suitable evidence for the utilization of starch nanoparticles in the packaging of the light-sensitive food products.

Thermal property

Thermal insulation provided by the packaging material is an important property that is needed by any food packaging material. High-value food products are usually transported by the cold chain transportation; the food products such as meat and meat-based products are usually to be maintained in 1 to 2 degree Celsius; however, a variation in temperature between 2 and 7 degree Celsius can cause negative impact and detoriation of the food during the transportation; and this may decrease the shelf life of the food product. A suitable packaging material must have the ability to provide a thermal buffering and maintain the food product at an appropriate temperature [24]. Studies show that the incorporation of starch nanoparticles in the starch-based films exhibited an increase in the thermal stability as well as the melting temperature (Tm) than compared with the conventional films without starch nanoparticles [21]. Suisui Jiang reported that an improvement in the thermal stability can be attributed to the better interaction between the nanoparticles and the film which resulted in a decreased onset temperature as well as a decreased melting temperature even with about 12% nanoparticles concentration [25]. Ana Paula et al. demonstrated that the Inclusion of starch nanoparticles in the starch films exhibited an increase in the thermal stability to about 15% when compared with the other starch films [26].

The thermogravimetric analysis demonstrated that at 800 degree Celsius, the residual mass increased proportionally with the starch nanoparticles concentration in the films and maximal residual mass was obtained with the acetylated starch nanoparticles. Paula González Seligra et al. stated that poly (butylene adipate-co-terephthalate) (PBAT) is a polymer that is biodegradable which was blended with thermoplastic starch and the Cassava starch nanoparticles was incorporated, the differential thermal analysis suggest that although the incorporation of nanoparticles did not affect the degradation of the PBAT polymer, it still was capable of increasing the degradation temperature due to the strong interaction of nanoparticles with the amylase [27]. Hence it can be understood that the incorporation of starch nanoparticles in the suitable polymer matrix, with an optimal concentration that can provide

the desired effect will enhance suitable thermal stability for the food products which ensure the safe transport and storage food products.

Biodegradability

The major purpose of utilizing starch in the food packaging sector is due to increasing environmental problem of non-biodegradability as there is a huge accumulation of plastics and other non-biodegradable polymers. One of the fastest growing industry across the globe is the food industry because of the tremendous growth in the human population. Along with the growth in the population, there is a simultaneous increase in the food packaging waste. This waste generated does not just include the final food package but it begins from the package of raw material, the waste from the food industry, waste from the retail sector and finally the waste from the household or the customer. This has led to need for the development for the biodegradable products [28]. Starch in general is a biodegradable polymer, and the main discussion is whether the incorporation of the starch in this polymer can increase the biodegradability.

Some studies suggest that the incorporation of the starch nanoparticles created an impact in the biodegradability of the polymer by catalyzing the process by the diffusion of water when it is in the soil causing it to swell and increase the biodegradability [6]. An experimental data suggested that the nanocomposite made of poly (butylene adipate-co-terephthalate PBAT)/TPS (thermoplastic starch) incorporated with cassava starch nanoparticles was cut into pieces and placed in a vegetable compost that was used as a soil and the samples were buried at 4 cm depth. The results suggested that the degradability rate increased when compared with the matrix. It was reported that the nanoparticles inclusion increased the moisture content which was found by the evaluation of the hydrophobicity of the film through contact angle study, and this led to higher rate of microbial attack and thereby faster degradability [27]. Therefore, the starch nanoparticle incorporation as nanofillers in the biodegradable polymers increased the rate of degradability which makes it a promising material in the recent trend and need of eco-friendly packaging materials.

Mechanical properties

The main reason for utilizing plastics for food packaging, although it is non-biodegradable, is due to its strong mechanical properties; if the world finds an alternate solution for replacing these mechanical properties with a biodegradable material, then we can expect a welcome of that product [30].

Though the natural polymers are comparatively weak than the plastics, in this section we will discuss on how this mechanical properties of the natural polymers can be enhanced by the incorporation of the starch nanoparticles as the nanofillers. The tensile strength, elongation of the material at break which is the point at which the film breaks, reflects the flexibility and the ability to stretch, and the reinforcement effects are the major factors and determinants of the mechanical strength of a packaging material.

Many research works report that the starch nanoparticles are beneficial in improving the mechanical properties of the natural polymer matrices; this is mainly because the nanometric dimension of a component such as starch can result in a strong interaction with the different polymer matrices that we prefer; this happens with the nanoparticles occupy the intra- or the intermolecular spaces in the polymer which attributes the polymer by increasing its density; and also the nanoparticles increase the specific surface area which can create a better interfacial interaction between polymer and the nanoparticles leading to increased strength of the nanocomposite [6], in the case of natural rubber matrix incorporated with waxy maize starch nanoparticles [20].

He le ne Angellier et al. reported that the usage of 20% of the nanoparticles reinforced effectively in the matrix without decreasing the elongation at the break along with an improvement in the relaxed modulus to about 10, 75 and 200 times higher in 10%, 20% and 30% concentration. The nanocomposite films prepared with the blend of polyvinyl alcohol with the pea starch nanocrystals exhibited an increased tensile strength and elongation at break than that of the polyvinyl alcohol films. The tensile strength increased up to a concentration less than 10%; however, it decreased above that concentration. This can be attributed to the smaller size and homogenous dispersion of the nanoparticles which forms stronger interactions in the film [23].

The corn starch-based films when incorporated with the taro starch nanoparticles exhibited an increased tensile strength when compared with the conventional film until the concentration of the nanofillers was gradually raised to 10% after which the tensile strength began to decrease when the concentration reached 15% which is similar to the previous case; however, it can still be seen that the tensile strength was higher than the control films. This was explained as the formation of aggregates at increased concentrations and led to decreased surface interactions with the matrix and created a disordered structure which has resulted in the decreased tensile strength. This case is inversely proportional in terms of the elongation at the break since the Eb value decreased as the quantity of rigid fillers increased. Hence, it was found that the maximum tensile strength (TS) value was reached at a lower Eb value [21].

Hence, we can infer that the mechanical properties of the food packaging polymers can be significantly improved with the incorporation of the starch nanoparticles but the factors such as concentration and homogenous dispersion plays a major role in obtaining this advantage by the utilization of starch nanoparticles in the polymer matrices as the nanofillers.

Applications of starch-based nanocomposites in food packaging

Starch is widely used as food packaging application for a longer time and always been a preferred choice of material due to its environmental compatibility, availability and reduced cost. In order to compensate the downside of the starch-based composites, wide range of inorganic material and synthetic polymers was added to the starch nanocomposites. Starch/clay composite is widely researched across the world for its ability to provide significant increase in the mechanical strength

and barrier properties in food packaging and bottling. Addition of MMT with starch reduced water consumption by the starch films providing both stability and tensile strength for starch-based films [94].

The polymers like polyvinyl chloride (PVC), polyethylene terephthalate (PET) pose a risk to environment. Hence development of synthetic biobased polymers is of in the recent trends, by combining the synthetic polymer with starch will improve the overall sustainability and functionality of the packaging material. Polymer composites such as carboxymethyl cellulose (CMC)/starch possess low mechanical strength compared to other toxic polymers; in contrast, PVA performs better than CMC exhibiting non-toxicity and biodegradability and assist starch processability with common industrial equipment [95].

Starch-based composite made from metal oxides such as titanium dioxide (TiO_2) and zinc oxide (ZnO) produced a better results in preservation of the fresh fruits in contrast to other films without incorporation of nanoparticles with better biodegradation capabilities [96]. It is reported that development of pH-sensitive starch/PVA composite films with ZnO and phytochemicals, and these packaging materials will offer a better safety, shelf life and transportation of food items enhancing water barrier, UV barrier properties from the packaging system on reaching the consumer end [97].

Silica-based nanocomposites

Silica nanoparticles will consistently possess a noticeable and a concentrated consideration of academic local area due to their simple planning and their wide use in different modern applications like food innovation, food bundling, wholesome area, catalysis, colors, drug store and mugginess sensors. Size and circulation of these particles decide the nature of a portion of these items.

Silica nanoparticles can be arranged into two principle draws near: top–down and base–up. Decrease in the element of unique size by using extraordinary size decrease methods (Physical methodology) is called as top–down measure though base–up measure includes a typical course used to deliver silica nanoparticles from nuclear or sub-atomic scale. A portion of the strategies that are broadly used to amalgamation silica nanoparticles are the solgel measure, turn-around microemulsion and fire combination. Among these strategies', solgel measure is broadly used to create unadulterated silica particles because of its capacity to control the molecule size, size appropriation and morphology through methodical checking of response boundaries. The surfaces of these nanoparticles are effortlessly changed because of its biocompatibility, warm constants, minimal expense and low poisonousness.

Silica nanoparticles are ordinarily round with profoundly uniform size and shape, yet they can likewise be created in wide range of shapes and estimates or applied as surface coatings to different kinds of nanomaterials. The size, porosity, crystallinity and state of silica nanoparticles can be definitively designed considering silica–based nanomaterials to be streamlined for the assortment of utilizations.

Synthesis of silica nanoparticles

Silica nanoparticles are generally produced by the solgel process, a chemical process that occurs under acidic or alkaline conditions at ambient temperature. The process starts with a precursor, which in aqueous solution can be a silane such as tetraethylorthosilicate (TEOS) Si (OEt)$_4$ or tetramethylorthosilicate (TMOS) Si(OMe)$_4$. The reaction occurs by hydrolysis and condensation in the presence of acid or base catalysts such as HCl or NH$_3$. During the reaction, crystalline nanostructures are formed. The speed of the hydrolysis and condensation steps is fast or slow depending on the choice of acid or base catalyst. In the acid-catalyzed reaction, many small silica particles are formed, which tend to form a gel-like structure, since the rate of hydrolysis is faster than the rate of condensation.

Similarly, in the base-catalyzed reaction, large silica nanoparticles are produced by sweat faster than hydrolysis, yielding solid spheres. Silica results are non-porous and tend to form gel fibers or solid spheres when we only use these products due to acid catalysis or catalyzed reactions. The synthesis of silica nanoparticles using the solgel method seems to be both simple and difficult, and the process can be quite beneficial to handle. The formation of MSNs is done by adding various surfactants to the solution, and the choice of surfactants used is considered an important factor in determining the mesopore structure [33]. MSNs with defined porosity can be easily produced by varying the amount and type of surfactants and adding blowing agents. However, most surfactants are cytotoxic and complete removal of surfactant is essential for the use of MSNs in food packaging. This is usually done by two main methods, calcination and solvent extraction.

Calcination is a thermal process involving the heating of synthesized MSNs to temperatures as high as 800 °C. The surfactants present in the MSN solution are subjected to extremely high temperatures. The drawbacks of calcination are the high temperature, energy prerequisite and the surface alterations that happen. Si–OH bonds on the outer layer of the MSNs respond together to shape Si–O–Si bonds at hight temperatures. The surface is choked and pore size is changed making the molecule more hydrophobic. Surfactant removal from MSNs can also be achieved using solvent removal, depending on the type of surfactant and the acidic or basic reaction. For example, ammonium nitrate can be used as a solvent to remove nanoparticles. Regardless of the method used, complete removal of surfactant must be ensured before MSN is used.

Barrier and thermal properties

The improvement in the obstruction properties of PNCs is all around explained in the composing in regard to the extended convolution with the extension of fillers [35]. The tangled pathway made by the nanofillers changes the scattering speed of the molecules, consequently achieving further developed limit properties. In any case, to achieve the best improvement, the fillers ought to be reliably flowed all through the polymer system, which is consistently difficult to achieve.

Another possible part is the polymer–nanoparticle participation, which can in like manner sway the obstruction properties by immobilizing the polymer strands. Polymer/clay and silicate nanocomposites clays and other silicate materials are truly consistent, clearly non-toxic in nature, and are immediately open at low expenses, which make them outstandingly engaging as fillers. This is the reason nanoclay-based PNCs have been perused generally for food contact applications all through the latest twenty years. Clays, for example, montmorillonite, kaolinite, hectrite and saponite, have been for the most part read for PNC applications [36].

Ordinarily, two essential courses are used to gauge nanocomposites: break down compounding and in situ polymerization [37]. Totally shed nanocomposite systems of polar polymers, for instance, polyamide with nanoclays, have been constantly uncovered [38, 39]; regardless, in nonpolar polymer cross sections, for instance, polyolefins, even intercalated morphologies are difficult to get. Two procedures are generally used to work on the correspondence between the hydrophobic polymer and the aluminosilicate surface of the nanoclays: the modification of clays with alkyl quaternary ammonium particles and the joining of polyolefins with, for example, maleic anhydride used as a compatibilizer [40–42]. The mass properties of the PNC materials depend upon the dissipating of the nanoparticles and their morphology. Monodispersed stripped systems show better impediment properties appeared differently in relation to intercalated and tactoid structures.

Gorrasi et al. found that while tactoid and intercalated montmorillonite/polycaprolactone composite developments showed unimportant improvement in water seethe impediment properties, totally shed plans further develop the obstacle properties by pretty much two critical degrees [43]. Modifiers are moreover used to achieve shedding; for example, dissipated montmorillonite particles have a mean interlayer distance (d-scattering) of only 3 nm, appeared differently in relation to the 8 nm mean interlayer separation of montmorillonite particles functionalized with octadecylamine [44]. Osman et al. have considered the effect of changing montmorillonite clays with quaternary ammonium modifiers bearing perhaps one, two, three, or four long alkyl (octadecyl) chains on the oxygen obstacle properties. Greater d-partitioning in the soil platelets was gotten with modifiers that have a couple of long alkyl chains.

A contrary association between the d-partitioning and the gas vulnerability has been showed up by the designer in changed montmorillonite/polyethylene (PE) nanocomposites [45]. Clays can be participated in different polymers to work on their gas and water smoulder deterrent properties. Regardless, the Incites related to achieving a uniform transport of nanoclays in the polymer framework and complete shedding achieves a confined impact. EVOH/montmorillonite composites have been represented with basically further developed limit properties [46]. Jung et al. have declared a 32% decrease in the oxygen vulnerability with polypropylene (PP) solidifying clay and void glass microspheres [47]. Wang et al. uncovered a lightweight and strong microcellular mixture framed PP/powder nanocomposite [40].

The unbending nature and the Gardner influence strength of the composite improved by 226% and 166%, separately; regardless, the effect on the prevention properties has not been represented. Jacquelot et al. uncovered a gigantic

improvement in gas limit properties (for all intents and purposes 100% for CO2 and 60% for O2) of a PE-based nanocomposite with oxidized paraffins as compatibilizers [48].

Another strategy is in situ polymerization for obtaining polyolefin composites with nanoclays. Nikkhah et al. uncovered a three-cross-over progress in the gas deterrent properties of an in situ-orchestrated PE nanocomposite diverged from virgin PE. Dadbin et al. proposed a single layer PE nanocomposite film that was envisioned to supersede PE multilayer films that are customarily used in the food packaging industry [49]. Thin layer films designed using jet film ejection of the low thickness polyethylene (LDPE)/straight LDPE (LLDPE)/montmorillonite organoclay nanocomposite showed a half decreasing in oxygen permeability at only 3 segments for every hundred (pph) centralization of nanoclay in the blend.

Similarly, in a study by Arunvisut et al. low-density polyethylene (LDPE)/clay nanocomposites showed a 24% reduction in oxygen compared to pure LDPE. Polyethylene grafted maleic anhydride (PEMA) was used as a match to improve the bond between the two materials. The clay component is treated with an alkylammonium surfactant to improve its distribution in the polymer matrix before gently mixing. The findings show that the incorporation of clay nanoparticles and the use of adapters can improve the performance of LDPE for a variety of applications [50].

Mechanical properties

Different models can be found in the coherent composing where nanoclays have been applied to further develop the limit properties of biobased and biodegradable materials. In an actually conveyed assessment by Risyon and partners, the effect of intertwining different groupings of halloysite nanotubes (HNTs) into PLA was explored for a normal improvement in mechanical, warm and block properties. An assembly of 3 wt% HNTs was perceived as the ideal, achieving abatement in oxygen transmission rate from $3.0 \times 10-13$ cm^3 cm/cm^2 s Pa for an ideal PLA film to $2.0 \times 10-13$ cm^3 cm/cm^2 s Pa.

The water smolder permeability decreased from 1.4 g m/m^2 for impeccable PLA to 1.1 g m/m^2 for PLA with 3.0 wt% HNTs. The further developed block properties were credited to the incredible dissipating of HNTs in the polymer lattice and the improvement of stable hydrogen associations between the HNT particles and the PLA, achieving a tangled pathway. Extended hydrogen holding in the polymer grid also achieved worked on mechanical properties and security from distortion, which was reflected in the high inflexibility and Young's modulus (E), disregarding the way that there was a reduced extending at break [51].

In general, all of the mechanical characteristics (impact resistance, Young's modulus, flexural modulus and thermal distortion temperature) appear to be significantly improved in exfoliated nanocomposites filled with layered silicates of high aspect ratio. The construction of a linked, three-dimensional network of long silicate layers [62]. The integration of a polymer matrix through the layered silicate structure takes place in a crystallographically normal way. Several molecular chains of the polymer are often used to create intercalated nanocomposites. Sometimes the interaction of

the silicate layers' hydroxylated edge-edge results in the flocculation of the silicate layers.

The outcome of substantial penetration of the polymer into the silicate layers with the spacing between layers enlarged to 10 nm or more is the exfoliated nanocomposites, which are made up of individual nm thick layers floating in a polymer matrix [63].Phyllosilicate clay that is layered and made up of highly anisotropic platelets and thin water layers. The platelets' average lateral diameters range from a few tens of nm to several m, and their average thickness is around 1 nm. A layer of silicon oxide tetrahedra is sandwiched between two layers of aluminum or magnesium hydroxide octahedra in each platelet. A 2:1 layered phyllosilicate, montmorillonite is composed of two layers of tetrahedral silica sheets filled with an octahedral film of aluminum in the middle.

This kind of clay has a surface charge that is somewhat negative, which is crucial for establishing interlayer space. Exchangeable cations (Na+ and Ca2+) balance out the imbalance of the surface negative charge. Weak electrostatic forces hold parallel layers together [64]. Biswas 2019 reported that the silver/silica composite films inhibits bacteria population on contact and has the potential to be implemented in the antimicrobial food packaging applications [65]. It is reported that MCM-41 and SBA-15 possessed high stability in aqueous phase and capability of controlled release of antioxidants from the active food packaging compared to other packaging materials [66].

Applications of silica-based nanocomposites in food packaging

It has been observed that silicate-based composites, which are frequently employed as fillers, have better biophysical qualities than the original polymer at lower concentrations. To preserve the packaging film's structural stability and barrier qualities, physical strength is needed. The physical and blocking characteristics of materials can be improved with the use of nanoparticles. It is believed that nanoparticles that are involved in water vapor transport, oxygen permeability and the toughness of polymer membranes play a significant influence in preventing and minimizing food deterioration. These advancements maintain food quality while extending the shelf life of food. Some gas molecules permeate into the following polymer during gas permeation; these molecules are primarily bonded to the polymer's surface [67]. Synthesis of biodegradable hybrid coating material using the solgel process involves blending an adequate amount of nanosized silica, an inorganic substance, with polylactic acid, an organic component, to produce a biodegradable organic component. The moisture and gas blocking properties of the PLA/SiO_2 composite material have been enhanced. It is a coating film that may be utilized in food packaging. The blocking qualities of polymer materials are defined in terms of diffusivity, which is determined by the diffusion rate of the airframe as well as the proportion of gas solubility of the polymer matrix [68, 69]. Curcumin improves the functional characteristics of pure chitosan films embedded with mesoporous silica nanoparticles (SBA15). The introduction of SBA15 nanofillers improves the physical characteristics of biocomposite films, as demonstrated in the FTIR analysis of biocomposite

films [70]. SiO$_2$ is employed as a reinforcing and surface enhancing ingredient in practically every polymer composite. Many precursors are required for the synthesis of silica nanoparticles, including methyl triethoxysilane (MTEOS), tetraethoxysilane (TEOS) and phenyl triethoxysilane (PTEOS).

There is an increasing need in the packaging sector for packaging solutions that promote sustainability by focusing on circular economy, which focuses on recyclability of packaging materials. The goal is to reduce traditional plastic consumption while increasing recycling efficiency by preserving the necessary barrier and physical qualities. Packaging materials built of silica-based nanocomposites can provide the desired properties and may be able to replace complicated multilayered polymer structures in this arena [69] (Table 3).

Cellulose-based nanocomposites

Nanocellulose is term insinuating nanocoordinated cellulose. This may be either cellulose nanocrystal (CNC or NCC), cellulose nanofibers (CNF) moreover called nanofibrillated cellulose (NFC), or bacterial nanocellulose, which suggests nanocoordinated cellulose made by microorganisms [52]. CNF is a material made out of nanosized cellulose fibrils with a high point extent (length to width extent). Typical fibril widths are 5–20 nm with a wide extent of lengths, consistently a couple of micrometers.

It is pseudo-plastic and shows thixotropic, the property of explicit gels or fluids that are thick (gooey) under average conditions, but become less gooey when shaken or agitated. Exactly when the shearing powers are killed, the gel recuperates a ton of its extraordinary state. The fibrils are detached from any cellulose containing source including wood-based fibers (squash strands) through high pressure, high temperature and rapid influence homogenization or microfluidization [53–55]. Nanocellulose can moreover be synthesized from strands by hydrolysis, prompting astoundingly clear and rigid nanoparticles which are more restricted (100 s to 1000 nm) than the cellulose nanofibrils (CNF) gained through homogenization, microfluiodization

Table 3 Biodegradable polymer (with silica nanoparticle) and their properties [69]

S. No	Biodegradable polymer (with silica nanoparticle)	Properties
1	Corn starch	The water resistance of the silica-nanocomposite increases
2	Potato starch	Mechanical properties of the nanocomposite increased
3	Poly (lactic acid)	Tensile strength reduced
4	Polycaprolactone (PCL)	Antibacterial activity was observed
5	Polyhydroxyalkanoates (PHA)	Higher Young's modulus
6	Chitosan	Better mechanical and thermal properties
7	Galactomannans	Tensile strength reduced
8	Gelatin	Barrier properties like mechanical strength improved

or pulverizing courses. The ensuing material is known as cellulose nanocrystal (CNC) [71]. Nanochitin is comparative in its nanostructure to nanocellulose.

Synthesis of cellulose nanoparticles

Nanocellulose, which is moreover called cellulose nanofibers (CNF), microfibrillated cellulose (MFC) or cellulose nanocrystal (CNC), can be set up from any cellulose source material, yet wood pulp is consistently used. The nanocellulose fibrils may be restricted from the wood-based fibers using mechanical procedures which open the crush to high shear powers, tearing the greater wood-strands isolated into nanofibers. Consequently, high-pressure homogenizers, processors or microfluidizers can be used. The homogenizers are used to delaminate the cell dividers of the fibers and free the nanosized fibrils. This association consumes incredibly a great deal of energy and characteristics in excess of 30 MWh/ton are ordinary. To resolve this issue, to a great extent enzymatic/mechanical pre-prescriptions [73] and show of charged social affairs for example through carboxymethylation [74] or TEMPO-mediated oxidation are used. [74] These pre-prescriptions can decrease energy use under 1 MWh/ton. "Nitro-oxidation" has been made to prepare carboxy cellulose nanofibers directly from unrefined plant biomass. Inferable from less getting ready strides to remove nanocellulose, the nitro-oxidation method has been found to be a smart, less falsely orchestrated and useful procedure to eliminate carboxy cellulose nanofibers [75].

Functionalized nanofibers using nitro-oxidation have been found to be a brilliant substrate to dispense with considerable metal molecule toxins like lead, cadmium and uranium. Cellulose nanowhiskers are rodlike uncommonly glass like particles (relative crystallinity record above 75%) with a rectangular cross region. They are outlined by the destructive hydrolysis of nearby cellulose strands for the most part using sulfuric or hydrochloric destructive. Shapeless sections of nearby cellulose are hydrolyzed and after careful arranging, clear portions can be recuperated from the destructive plan by centrifugation and washing. Their estimations depend upon the nearby cellulose source material, and hydrolysis time and temperature. Roundabout molded carboxycellulose nanoparticles orchestrated by nitric destructive phosphoric destructive treatment are consistent in dispersing in its non-ionic construction.

Barrier properties

In semi-clear polymers, the glasslike areas are seen as gas impermeable. As a result of commonly high crystallinity, in mix with the limit of the nanofibers to shape a thick association held together by strong between fibrillar bonds (high solid energy thickness), it has been recommended that nanocellulose may go probably as an impediment material. But the amount of nitty gritty oxygen permeability regards are limited, reports attribute high oxygen limit properties to nanocellulose films. One assessment uncovered an oxygen permeability of 0.0006 $(cm^3 \ \mu m)/(m^2 \ day \ kPa)$ for a ca. 5-μm thin nanocellulose film at 23 °C and 0% RH.

In an associated report, a more than 700-overlay decrease in oxygen permeability of a polylactide (PLA) film when a nanocellulose layer was added to the PLA surface was represented. The effect of nanocellulose film thickness and porosity in video structure oxygen vulnerability has been researched. A couple of makers have uncovered immense porosity in nanocellulose films, which is apparently in consistent irregularity with high oxygen limit properties, while Aulin et al. assessed a nanocellulose film thickness close to thickness of glasslike (cellulose Iß valuable stone development, 1.63 g/cm^3) exhibiting a thick film with a porosity close to nothing.

Changing the surface handiness of the cellulose nanoparticle can in like manner impact the vulnerability of nanocellulose films. Films involved conversely charged cellulose nanowhiskers could suitably reduce immersion of unfavorably charged particles, while leaving fair particles all things considered, unaffected. Distinctly charged particles were found to accumulate in the film. Multi-parametric surface plasmon resonance is one of the techniques to consider limit properties of ordinary, changed or covered nanocellulose. The different antifouling, soddenness, dissolvable, antimicrobial limit specifying quality can be assessed on the nanoscale. The adsorption energy similarly as the degree of developing can be assessed ceaselessly and mark free.

Mechanical properties

Glasslike cellulose has a solidness around 140–220 GPa, basically indistinguishable with that of Kevlar and better than that of glass fiber, the two of which are used monetarily to develop plastics. Motion pictures delivered utilizing nanocellulose have high strength (in excess of 200 MPa), high immovability (around 20 GPa) but non-appearance of high strain (12%). Its fortitude/weight extent is on different occasions that of solidified steel. Strands delivered utilizing nanocellulose have high strength (up to 1.57 GPa) and immovability (up to 86 GPa).

Among many different synthetic polymers, cellulose and its derivatives are often employed. Cellulose is said to act as a carrier and stabilizer in addition to giving the film other special qualities. Synthetic polymers, however, have a low thermal stability and a high degree of brittleness, which is one of their main disadvantages. Their adoption is limited because these restrictions render them unstable during heat procedures like melting [29]. The two most often utilized chemicals created from renewable resources are poly-3-hydroxybutyrate (PHB) and polylactic acid (PLA).

These substances are weakly thermally stable, brittle and fragile by nature. In case of natural polymer blending, cellulose is the most stable natural polymer and blending it with other natural polymer with same properties will increase the total stability of the cellulose composites but the major disadvantage is that these composites will have less mechanical strength, low thermal stability and barrier properties.

Incorporation, encapsulation, blending might be the more effective way in attainment of the cellulose composites films. Cellulose composites are also done with presence of proteins, these proteins increase the sensitive of the film but at the same time these reduces the mechanical strength of the films. The most commonly used protein is gelatin, allowing themselves to reinforce different polymers like cellulose.

The tensile strength and solubility greatly depends on the ratio of materials used, finding the perfect ratio might increase the overall quality of these composite films made of cellulose.

The metal composites are known for its antimicrobial qualities compared to other composites and are widely used in food packaging [76]. It has been demonstrated that cellulose containing methylcylopropene is a significant and effective treatment to stop the synthesis of ethylene. The entry of microbes is another way that food can deteriorate. Microbial deterioration is mostly to blame for fruit damage. Different antimicrobial agents are actively incorporated into cellulose to enable continuous release in the matrix following activation [77].

Application of cellulose nanocomposites in food packaging

Food shelf life is crucial because food products may require a few days or even many months before they are delivered to clients [78]. Some types of efficient packaging are required to better preserve food and enhance product shelf life. Ezati et al. created CNF composite films combining glucose-derived carbon dots (GCD) and N-functionalized GCD (NGCD) [79]. The results demonstrated that GCD and NGCD could effectively filter UV light while increasing the membrane's water vapor permeability without compromising its mechanical qualities. At the same time, the composite films were oxidation resistant, with a free radical scavenging rate of 99% ABTS and 80–85% DPPH. When these films were applied to citrus or strawberries, they significantly inhibited fungus development and extended shelf life by 2–10 days. Furthermore, they minimized the weight loss caused by transit and storage, preserving food freshness and enhancing economic advantages [80]. A novel technique, coaxial 3D printing, was used to create cellulose nanofiber (CNF)-based labels with dual functions of fruit freshness preservation and visual monitoring. This fibrous casing is made from a CNF-based ink containing blueberry anthocyanins and offers excellent moldability and print fidelity, as well as a visual pH-sensitive response to monitor freshness. Chitosan containing 1-methylcyclopropene (1-MCP) was injected into the hollow microchannels of the fibers and 1-MCP was captured by the electrostatic interaction between chitosan and CNF. The 3D-printed label extended the shelf life of lychee by 6 days while clearly demonstrating the difference in freshness as demonstrated by headspace gas chromatography and ion mobility spectroscopy [80].

Polylactic acid-based nanocomposites

Polyactic acid (PLA) is the biopolyester that is most commonly utilized in commercial food packaging. Despite this, the biopolymer has severe flaws in its qualities that PET does not, including high stiffness, poor thermal resistance, and comparatively poor gas and vapor barrier properties [81]. Due to its outstanding transparency, which is on par with its petroleum-based counterparts, and its decent water resistance, PLA, which is presently manufactured commercially at a rate of more than 150,000 tonnes per year, is of special interest for use in food packaging. For

example, PLA has a lot lower water permeability than proteins and polysaccharides, but it is still higher than PET and traditional polyolefins. The material has a tendency to be quite stiff, and its thermal resistance—measured by the heat deflection temperature (HDT)—is substantially lower than PET's [82]. While there are some more or less effective solutions for mechanical qualities, thermal and barrier properties are more difficult to address. Enhancing this material's thermal and barrier capabilities while retaining its naturally advantageous qualities, like transparency and biodegradability, is consequently of tremendous industrial interest [83].

Due to the synergistic effect of PLA's biodegradable nature and the barrier-induced effect caused by the dispersion of nanoparticles, PLA reinforced with nanoclay is a viable substitute for the use of standard oil-derivative polymers [84].

Synthesis of PLA nanocomposites

Nearly all agricultural raw resources, including sugar or corn, can be used to produce PLA. But organic waste from agricultural operations can also be put to use. By means of a bacterial fermentation procedure, these fundamental components will be converted into lactic acid, the essential chemical required for PLA manufacturing. By combining the processes of oligomerization and cyclization, this lactic acid can subsequently be converted into lactide, the cyclic dimer of lactic acid. After purification of lactide, the PLA is produced from purified lactide through the process of polymerization [85]. The process of creating PLA-based nanocomposites with a nanocellulosic substrate, such as nanofibrillated cellulose (NFCs), by extruding an aqueous suspension of NFCs into the PLA melt [86].

Barrier and mechanical properties

Even with a low degree of filler loading (5 wt%), the mechanical characteristics of diverse biopolymers have significantly improved thanks to the formation of nanocomposite with organoclays. It has commonly been noticed that the filler content has a significant influence on the mechanical properties of polymer/clay nanocomposites. Here, we'll look at a few cases that have been published in the last few years. The tensile characteristics of starch/MMT nanocomposites synthesized with different filler concentrations of 0 to 11 wt% to the starch were evaluated by Huang and Yu. Tensile strain dropped monotonically with increasing filler loading, with the exception of 8% loading, while tensile strength and Young's modulus increased up to 8% before levelling off [87]. Because of the high stiffness and aspect ratio of nanoclay and the positive interfacial interaction between the polymer matrix and the dispersed nanoclay, the mechanical characteristics of polymer nanocomposites have improved. Excellent barriers against gases (such O_2 and CO_2) and water vapor exist in polymer nanocomposites. Studies have revealed that the kind of clay (i.e., compatibility between clay and polymer matrix), aspect ratio of clay platelets and structure of the nanocomposites all play a significant role in this reduction in gas permeability. In general, polymer nanocomposites with fully exfoliated clay minerals and high aspect ratios would yield the best gas barrier capabilities [88].

Thermal stability

According to Hablot et al. [89], the quaternary ammonium compounds may catalyze the heat breakdown of PLA, as evidenced by a sharp drop in PLA molecular weight. These findings suggest that the biodegradable characteristics of the bio-nanocomposite materials can be tuned by carefully selecting the organoclay fillers. Before making any conclusions, it is necessary to take into account the thermal stability of the organic component of the organoclay, such as quaternary ammonium salts, which are utilized up to 40% for surface modification of nanoclay.

The thermal stability of the ammonium salts is highly limited at temperatures of commercial scale processing employing extrusion or injection moulding necessary for most polymers since most of the structures trend to Hofmann elimination resulting in volatile olefins with amines [90]. Ammonium salts degrade thermally at temperatures as low as 180 °C, and catalytically active spots on the aluminosilicate layer further slow this process down [91]. The amino propyl groups plus their charge interactions are responsible for the AMP clay's ability to hinder microbial development by breaking membrane integrity and allowing vital substances to leak inside the cells.

Applications in the food sector have been suggested for quaternary phosphonium salt functionalized few-layered graphite due to its excellent thermal stability and protracted antibacterial action against *E. coli* and *S. aureus* [92].

Biodegradability

Biodegradation for biodegradable polymers can take the form of fragmentation, a loss of mechanical qualities, or occasionally, degradation brought on by the activity of microorganisms like bacteria, fungi and algae. A complex process called biodegradation of polymers can take place through oxidation and hydrolysis catalyzed by enzymes [93]. Tetto et al. examined the biodegradability of nanocomposites made of PCL for the first time and found that they degraded more readily than pure PCL.

They clarified that the catalytic function of the organoclay during the biodegradation process may be responsible for the increased biodegradability of PCL in clay-based nanocomposites [94]. A series of biodegradation studies with PLA nanocomposite produced from PLA and organoclay were carried out by Sinha Ray et al. [95].

They first examined the biodegradability of PLA and PLA/organoclay nanocomposite films using a soil compost test at 58 °C with both PLA and PLA/organoclay nanocomposite films made by melt blending. They then showed images of the recovered film samples with composting time. They discovered no discernible difference in the biodegradability of plain PLA and PLA nanocomposite until just a month of composting, but within two months, they noticed a noticeably higher disintegration of the PLA nanocomposite, which was totally decomposed by compost. They also tracked the changes in molecular weight (Mw) and residual weight (Rw) in PLA and PLA nanocomposite films over time, finding that for a month, the Mw and Rw losses were essentially the same for both types of films [96].

The PLA nanocomposite films saw a dramatic reduction in residual weight loss after one month, which was consistent with the outcome shown on camera. Later, they used a respirometric test to determine the PLA nanocomposite films' biodegradability by detecting CO_2 evolution during biodegradation [97].

The bionanocomposite packing materials should be kept stable and durable enough to fulfil their packaging duties for the duration of their practical shelf life. It's interesting to note that, depending on processing and environmental factors, nanoparticles have two opposing impacts on polymer nanocomposites: degradation and stability [98]. By altering the amount of solvents employed or by incorporating various forms of organocays modified surfaces with various types of surfactants, hybrid bio-nanocomposites, such as layered silicates contained in a polymeric matrix, increase stability. Depending on the intended usage, the packaging sector may be able to take advantage of such novel features of nanoparticles.

CNTs/natural montmorillonite PLA-based nanocomposites provide proof of the synergistic interaction between various nanofillers. In the study, natural montmorillonite (Cloisite Na+) and hydrid nanoclays with CNTs generated using a CVD technique were introduced into the PLA matrix. The eventual conclusion was that the PLA nanocomposites had both Cloisite Na+ sorption capacity and CNT electrical conductivity [99].

The consumer demand for more environmentally friendly products, the exploitation of biobased feedstock, consideration of recycling options, an increase in price and restrictions on the use of polymers with high "carbon footprints" of petrochemical origin, particularly in such applications as packaging, automotive, electrical and electronic industries, are all contributing factors to the extraordinary rapid growth of biobased polymers at the moment. In this regard, PLA stands in as the most well-liked and promising "green" eco-friendly material, with the best potential for growth in both immediate and long-term uses [100].

Application of PLA nanocomposites in food application

Incorporating inorganic nanoparticles (NPs) significantly lowers permeability and imparts antibacterial characteristics to PLA, although the use of metal and inorganic NPs raises health issues in food packaging [101]. TiO_2, CuO, ZnO, MgO and Fe3O4 are notable NPs that show potential against infections. PLA-based NPs, such as sandwich-architected PLA-graphene films, improve water vapor and oxygen barrier properties, hence prolonging the shelf life of edible oil and potato chips [102]. PLA is strengthened by unmodified montmorillonite clay, which protects processed meats and reduces lipid oxidation [75]. Active PLA-NP packaging increases the safety of perishable products; ZnO-reinforced PLA improves fresh-cut apple quality and shelf life [76], while TiO_2 and Ag-containing PLA matrix prolong mango freshness [77]. Microbial growth is inhibited by a PLA/ZnO layer in fish preservation [78]. PLA composites (PLA/EO/Ag-Cu) protect fresh chicken against contamination. Cu NPs in PLA packaging improve dairy products without altering their taste [80]. Antioxidant NPs, such as SeNPs, reduce oxidation in nuts [83]. PLA NC migration is

investigated, and results demonstrate low migration for Cu, Zn, Al and Ca, as well as PLA/TiO$_2$ and Ag NPs migration within limits [84].

Nanomaterials strengthen environmentally friendly PLA, solving the environmental difficulties associated with plastics. PLA's processability lends itself to food packaging with a variety of molding processes. Reinforced PLA compensates for flaws by improving characteristics while somewhat delaying biodegradation. Despite the abundance of research on PLA nanocomposites, real-world food packaging applications are restricted. More research is needed to evaluate PLA nanocomposites containing inorganic NPs for food safety, with the potential to replace traditional polymers in direct food contact.

Chitosan nanocomposites

Chitosan is unique and naturally available biopolymer and can be synthesized by deacetylation of chitin [81]. It is a step behind cellulose as most available natural polysaccharide and industrial production of chitosan largely depends on the terrestrial arthropods, marine crustaceans, Mollusca and fungal microorganisms as the main source of chitosan. Chitosan is co-polymers containing β-(1–4) glycosidic bonds between altered N-acetyl-D-glucosamine and D-glucosamine.

Chitosan attracts attention as numerous food industries adopt it as a natural preservative and packaging material since it demonstrates both antioxidant and antibacterial action against a wide range of microorganisms, including pathogenic and spoilage microorganisms [82]. The antimicrobial activity of chitosan is not fully understood yet but three models were proposed in regard to its interaction with microbes exhibiting the antimicrobial property. Firstly, the interaction between the positively charged chitosan against negatively charged microbial cell wall results in electrostatic protonation of NH$^+_3$and negative residues with influencing Ca^{2+} by its negative potential that in turn induces osmotic imbalance and hydrolyze peptidoglycan layer.

The second mechanism involves chitosan binding to the DNA of microorganism and interrupting protein synthesis by inhibiting m-RNA. Third mechanism involves chelation of metals and suppression of essential nutrient required for microbial growth [83].Chitosan has the distinguishing benefit over other biobased packaging materials given that it can be incorporated with vitamins and minerals that contain antimicrobial properties [81].

Synthesis of chitosan nanocomposites

Chitosan composites are widely researched based on their quality and compatibility. There are different types of composites among chitosan/biopolymer-based films including integration of various naturally obtained biopolymer with chitosan to increase its stability and overall quality. Chitosan/polysaccharide-based films involve combining the large carbohydrates with chitosan. These films possess a great antimicrobial and antioxidant properties and promising active food packaging.

Polysaccharides like cellulose and starch blends with chitosan well enhancing the general film forming capacity of the chitosan with promoting other factors such as optical property and gas barrier properties. Proteins with special groups can also be used to develop a chitosan composite that promotes its application in food packaging industries. Proteins like caseinate and collagen provide high thermal stability and good adhesion properties to the composites. Extracts obtained from beeswax are used to develop films by introducing them with chitosan, and these chitosan/extract composites inhibit pathogenic microorganisms. Chitosan/synthetic polymer composites contain synthetic polymers like PVA and PLA compared to biopolymer composites, and synthetic polymer possesses high mechanical strength and thermal properties.

Chitosan/metal ion films are known for its antimicrobial property as metal ions like silver possess great antimicrobial properties and also non-toxic in nature. Another metal ion like ZnO enhances both physiochemical and biological properties of chitosan films [84]. It is reported that the two primary issues with bread and bakery products are fungal growth and staling causes significant economic losses every year around the globe.

In order to extend the physicochemical and microbiological shelf life of sliced wheat bread, novel active nanocomposite films and coatings based on CMC-CH-OL-ZnO NPs (0–2%) were created and ZnO/chitosan composites were used in raw meat packaging due to its capability in controlling the spoilage by inhibition of bacterial growth [85, 86].

Chitin contains high level of acetylated groups resulting in their poor solubility in the aqueous solutions and can be increased by reduction of acetyl group partially. There are two ways of synthesis: Chemical method of chitosan production is commonly used by the industries. Raw material from crustaceans, mollusks and fungal cell wall is reduced to form chitin by reducing calcium, protein and decolorizing. In order to remove the acetyl group present in the chitin, it is treated with 40- 50% of NaOH. Reduction of acetyl groups directly corresponds to the concentration of the alkali concentration, reaction time and temperature.

The chemical degradation process is efficient with great yield but pose a risk of environmental pollution compared to enzymatic method that uses chitin deacetylase enzyme to degrade chitin to chitosan. This method is eco-friendly but built around complex processes of selecting and cultivating enzyme-producing bacterial strains in correspondence to yield. Selecting strain with great potential is mandatory in large-scale production of chitosan [87].

Mechanical properties

Mechanical properties include tensile strength, elongation rate and puncture force of the composite films. Different chitosan composite exhibits broad range of mechanical properties. It is reported that the pure chitosan films with a concentration of 2% possessed different tensile strength at different deacetylation level. 80% and 78% DD of chitosan had a tensile strength of 59.4 and 55.6 MPa, respectively, indicating that the degree of deacetylation of chitosan plays a major role in altering the mechanical

properties [88]. Chitosan/biopolymer composites mechanical property increased significantly based to stable biopolymer incorporated. Chitosan and very stable cellulose were combined to produce a material with a tensile strength of 75.0 MPa with an elongation percentage of 17% that is greater than the pure chitosan films [89].

In contrast, the chitosan/protein composites exhibited very low tensile strength compared to other composite but with high elongation percentage. Chitosan/quinoa composite displayed a 177.8% of elongation with very low tensile strength of 2.7 MPa [90]. On other hand, chitosan with plasticizers exhibited balanced tensile strength and percentage of elongation, and chitosan/glycerol composite possessed a tensile strength of 46.4 MPa with an elongation of 35.9% exhibiting its potential as food packaging material [91].

Barrier properties

Barrier properties of chitosan composites are essential in determining its viability in food packaging industries as they contribute in estimating and predicting the shelf life of the product. Among the parameters, water permeability and oxygen permeability as they prevent deteriorating reactions and dehydration. Chitosan possesses low water and oxygen permeability like other polysaccharides, and pure chitosan films (1%) exhibited a water permeability of 8.07×10^{-13} g/m s Pa and with an oxygen permeability of 6.65 cm^{3}μm/m2 day kPa.

Chitosan composites with plasticizers like glycerol exhibited 8.8×10^{-10} g/m s Pa of water permeability and 37.4 cm^{3}μm/m2 day kPa of oxygen permeability that is comparatively low than polyethylene oxide that pose a water permeability of 10.6 g/m s Pa. Therefore, figuring out the right composite material that balance out the properties is of absolute necessity for efficient functioning of the chitosan composites films as a food packaging material [92, 93].

Application of chitosan-based nanocomposites in food packaging

The use of nanocomposites to improve chitosan-based films has gained popularity. Nanoparticles enhance physical, mechanical and antibacterial qualities while influencing biodegradability. Various nanoparticles, such as cellulose nanocrystals, nanosilver, nanosilica, graphene oxide and zinc oxide, improve the functioning of chitosan [88–92]. Chlorogenic acid incorporation onto halloysite nanotubes enhanced heat stability while reducing mechanical characteristics. Antimicrobial and mechanical properties of zinc oxide nanoparticles (ZnONPs) have been demonstrated in chitosan and ZnONP-based nanocomposites [93–96]. Gallic acid-loaded nanocomposites have potent antibacterial and antioxidant properties. Silver and magnesium oxide nanoparticles improve the antibacterial activity and thermal stability of chitosan-based films. Silver nanoparticles increase meat packing shelf life, while silver-loaded nanotitania strengthens composite connections. The use of natural polymer-based nanoparticles such as cellulose nanocrystals improves the characteristics of chitosan films while preserving their biodegradability [97]. This environmentally friendly approach shows potential in meat packaging, improving

mechanical, thermal and antimicrobial features while extending shelf life. Electrospinning is a novel fiber preparation technology that is being used in culinary, biomedical and packaging applications. Arietta et al. [98] fabricated mechanical characteristics and biodegradable end-of-life packaging by combining chitosan with PLA and PHB. Toloue et al. [99] employed electrospinning to improve bioscaffold characteristics in bone tissue engineering by adding alumina. Chitosan nanoparticles embedded in PLA fiber membranes revealed improved antibacterial efficacy [100].

Polyhydroxyalkanoates

Polyhydroxyalkanoates (PHAs), a type of bioplastic, are thought to be the best choice for usage in food packaging. Nonlinear optical activity, biodegradability, hydrophobicity, piezoelectricity, non-toxicity, thermoplasticity, as well as impermeability to gases and/or water are among the PHAs' intriguing properties [101]. PHAs are the 3-, 4-, 5- and 6-hydroxyalkanoic acid polyesters (HA). In bacterial cells, Beijerinck first noticed them as intracellular granules in 1888 [82]. French scientist Lemoigne identified it as a homopolyester containing 3-hydroxybutyric acids shortly after that, in 1926. (3-HBs). In order to store PHAs utilising renewable resources, the archaebacteria, gram-negative, gram-positive bacteria and cyanobacteria have been identified.

Depending on how many carbon atoms are in the side chain, there are three primary types of PHAs. Three to five carbon atoms (C3-C5) make up the short-chain-length vPHAs (scl-PHAs), such as poly-3-hydroxybutyrate (PHB) and poly-3-hydroxybutyrate-co-3-hydroxyvalerate (PHBV). The medium-chain-length PHAs (mcl-PHAs), such as poly-3-hydroxyhexanoate (PHHx) and poly-3-hydroxyoctanoate, each contain 6—14 carbon (C6-C14) atoms (PHO). The PHAs with long chains (lcl-PHAs) include more than 14 carbon atoms [102].

In contrast to pure PHB, the nanocomposite that was created had an intercalated morphology, a greater storage modulus (more than 40%), and it nevertheless preserved biodegradability. Using the solvent intercalation process, Lim et al. investigated the effects of different C25A clay concentrations (3, 6 and 9 wt%) on the thermal characteristics of PHB/C25A nanocomposites. The planned nanocomposite's intercalated structure was revealed by XRD investigation [103].

Synthesis of PHAs

PHAs are produced by microorganisms using eight different metabolic processes. *Cupriavidus necator* uses the PHA biosynthetic pathway I, which has received the most research attention. Two molecules of acetyl CoA-(acetyl coenzyme A), which are produced from the tricarboxylic acid (TCA) cycle, are condensed into acetoacetyl coenzyme A by the enzyme b-ketothiolase (encoded by phbA).

The conversion of acetoacetyl CoA into 3-hydroxybutyryl-CoA (3HB-CoA) monomer is then carried out by the NADPH-dependent acetoacetyl-CoA dehydrogenase, which is encoded by phbB. Finally, the polymerization of 3HB-CoA

Fig. 7 Flow diagram of preparation of silica nanoparticles using calcination process [34]
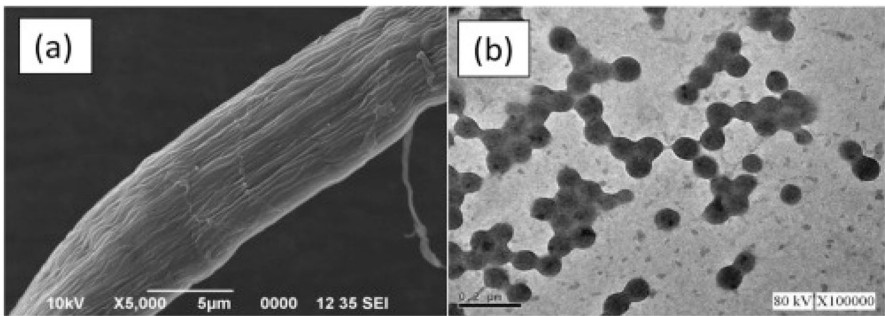

Fig. 8 **a** SEM image of cellulose fibers facial cotton and **b** TEM micrograph of cellulose nanoparticles prepared from 0.01 w/v% of cellulose solution [56]

monomers into PHB homopolymer is carried out by the PHB synthases, which are encoded by phbC.

According to reports, *Pseudomonas oleovorans* and *Pseudomonas aeruginosa* create mcl-PHAs, including PHBHHX, from alkanes, alkanols, or alkanoic acids obtained via the pathway II's b-oxidation of fatty acids. Malonyl-CoA-ACP transacylase and 3-hydroxyacyl-ACP (acyl carrier protein) transferase (PhaG) are responsible for the PHA synthesis in biosynthetic pathway III (FabD).

In Rhizobium (Cicer) sp. strain CC 1192, pathway IV entails the conversion of acetoacetyl CoA into PHB by NADH-dependent acetoacetyl-CoA reductase (Figs. 7 and 8).

The 4-hydroxybutyrate (4-HB)-containing PHAs are produced from 4-hydroxybutyryl-CoA via the pathway V, which was first identified in Clostridium kluyveri (Fig. 9). This pathway is mediated by 4-hydroxybutyrate dehydrogenase (4hbD), succinic semialdehyde dehydrogenase (SucD) and 4-hydroxybutyrate-CoA: CoA transferase (OrfZ) [102–105].
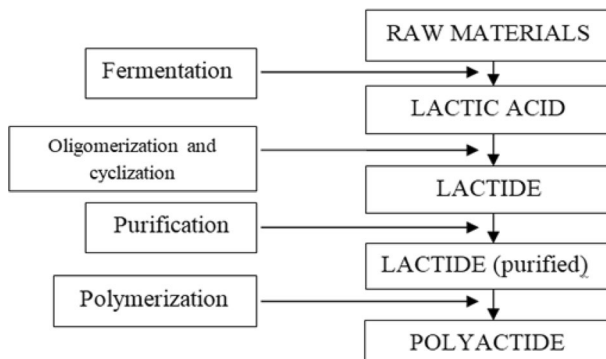
Fig. 9 Synthesis of Polyactide (PLA)

Barrier, mechanical and thermal properties

While the Young's modulus values fluctuated slightly around a value of around 1100 MPa for PHB/poly(vinyl acetate) PVAc/cellulose nanocrystals (CNC) nanocomposites, the tensile strength increased by about 20% with the addition of nanocellulose. It should be highlighted that plain PHB (after annealing treatment) showed a higher elastic modulus value than both plain poly-(hydroxybutyrate-co-hydroxyvalerate)PHBV and PHB/PVAc 80/20 blend.A different pattern of behaviour was seen for PHBV/PVAc/CNC systems (Fig. 8b): the elastic modulus of the composite materials (about 1300 MPa) was higher than that of the original PHBV (820 MPa) as well as of PHBV/PVAc 80/20 blend (1220 MPa), while the tensile strength increased with the CNC content from around 21 MPa for pure PHBV (as well as PHBV/PVAc blend) up to 32 MPa for the composite [109].

Biodegradability

PHA can be biodegraded by bacteria that make it either inside the cell or outside the cell by microbes that do not manufacture PHA but can still utilise it as a carbon and energy source. However, in both situations, specialized enzymes, internal or extra-cellular PHA depolymerases, catalyze the hydrolysis. The large molecular weight of PHA chains limits cell wall penetration by non-producing PHA bacteria. In this manner, PHA chain scission in water soluble monomers and oligomers is essential to their uptake by cells and subsequent cytoplasmic metabolization [106].

Maiti et al. also assessed the biodegradation rates of PHB/MMT as well as PHB/MAE nanocomposites at ambient and 60 °C (2007). The findings showed that all samples' biodegradation rates decreased by 2–3 times at 60 °C. Two things were blamed for the decline in the biodegradation rate: a drop in the number of microorganisms present at 60 °C and/or a rise in crystallinity.

This is due to the fact that polymer chains are more mobile in the amorphous phase at higher temperatures than the glass transition temperature of the matrix

(*16 °C), which increases the likelihood that they will organize into a crystalline structure [106]. Despite the decreased size of the PHBV spherulites in the PHBV/ MMT nanocomposites, Wang et al. demonstrated a reduction in the biodegradation rate of PHBV/MMT nanocomposites in relation to an increase in MMT quantity. According to the authors, the MMT serves as a physical barrier to keep the polymer chains safe and prevent water from diffusing through the sample. Therefore, the lesser availability of water for hydrolysis reactions is the cause of the lower biodegradability of PHBV/MMT compared to pure PHBV [108].

Application of PHAs in food packaging

The PHB-based cups performed on par with HDPE in terms of competency while also outperforming HDPE when storing samples in the light. In a different study, PHB was determined to be suitable for preserving fat-rich items including mayonnaise, margarine and cream cheese based on physical, mechanical, sensory and dimensional assessments [106]. PHA and PHB were used for packaging of sour cream.

The effects of pasteurization on the effectiveness of packaging of PE, PP, PLA and PHB films and concluded that sterilized PHB films could be utilized to package meat salad. When PHB qualities were exposed to gamma radiation at the levels necessary to sterilize food or packaging materials, no significant loss in PHB properties was observed.

To determine the best material for packaging organic tomatoes, Kantola and Helén were the first to compare the performance of biopol-coated paperboard trays and Mater-Bi (Novamont, Italy) starch-based film. They discovered that the tomatoes in the PHB-coated paperboard trays were just as fresh as those in the Mater-Bi starch-based film [107–109].

Conclusion

Nanotechnology has the potential to revolutionize various industries, including food technology. The development of nanocomposites, which are materials composed of nanoparticles dispersed within a matrix, offers exciting possibilities for creating eco-friendly and efficient solutions, particularly in the realm of food packaging. By incorporating eco-friendly nanofillers such as silica, starch and cellulose into biodegradable polymers, researchers can enhance the properties of the resulting nanocomposites.

These fillers, which are readily available and cost-effective, can improve the mechanical strength, barrier properties and thermal stability of the packaging materials. They can also provide additional functionalities, such as antimicrobial properties, oxygen scavenging capabilities and UV resistance, which can extend the shelf life of packaged food products. The use of nanotechnology in food packaging can address some of the challenges associated with traditional packaging materials. For instance, nanocomposites can create a barrier that prevents oxygen, moisture and

other contaminants from reaching the food, thereby reducing spoilage and enhancing food safety.

Additionally, the incorporation of nanomaterials can enable the development of intelligent packaging systems that can monitor and indicate the freshness or quality of the food inside. Furthermore, the use of biodegradable polymers in combination with eco-friendly nanofillers contributes to environmental sustainability. As these materials break down more readily in natural environments, they reduce the accumulation of non-biodegradable waste. This aligns with the broader goal of reducing the environmental impact of packaging materials and promoting a more sustainable future.

However, it is important to note that the field of nanotechnology in food packaging is still evolving, and there are considerations regarding the safety and regulatory aspects of using nanomaterials in direct contact with food. The potential release of nanoparticles and their effects on human health and the environment require careful evaluation and assessment.

In conclusion, the integration of nanotechnology in food technology, particularly in the development of eco-friendly nanocomposites for food packaging, holds significant promise for a better future. By leveraging the unique properties of nanomaterials, researchers can create innovative and sustainable solutions that enhance food safety, extend shelf life and reduce environmental impact.

Declarations

Conflict of interest The authors certify that they have NO affiliations with or involvement in any organization or entity with any financial interest in the subject matter or materials discussed in this manuscript.

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