

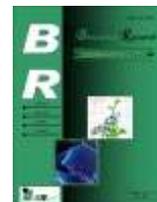


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## Synthesis and industrial applications of bionanomaterials: An update

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Recent trend towards bio-based economy poses great promise to renewable materials like biopolymers as an alternative to petroleum resources. Similarly, reinforcement of biopolymers with nanostructured materials is gaining additional attraction from the researchers in academia and industries, because of the exponential improvement in physical, mechanical, and thermal properties with smaller amounts of nanofillers incorporation. Furthermore, this incorporation can be combined with functionalization routes that may enable new fundamental differences in the properties, biodegradability, biocompatibility, and applications of bionanocomposites compared to pristine ones. This article is intended to review the recent research on bionanocomposites and then compile their most noticeable and novel applications in various industries.

**Keywords:** Biopolymers; Nanofillers; Biomedical applications, Food packaging, Environmental, Textile industry

### INTRODUCTION

The word 'nano' derived from the Greek word 'nanos' meaning 'dwarf' which is equivalent to the billionth of a meter ( $10^{-9}$  m). Based on the number of measurements of the dispersed particles, in the nanometer range, nanomaterials are classified into three classes *viz.* nanotubes, nanoparticles and nanolayers (Saba et al. 2014). In general, for explaining the unique properties of materials along with its applications, nanomaterials have been used since a long time and applied to get novel mechanical, physical and chemical properties of the materials in comparison to same materials without nanoscale features. Polymer nanocomposites are considered as the materials of the 21<sup>st</sup> century (Coiai et al. 2015). Even though the existence of nanocomposites was reported for decades, the first report on the action of these fillers are published by Usuki et al. (1993) and Okada and Usuki (1995). Further research was carried out and the term "nanocomposites" was introduced for the first time in 1994 (Marquis et al.

2011). Now a day, nanomaterials are used in almost all area of human needs including textile, biomedical, health care, food packaging, industrial, electronics, environment, wastewater treatment, agriculture etc. (García-Mayagoitia et al. 2020; Jayakumar et al. 2020; Singh et al. 2020; Velu et al. 2020; Zafar et al. 2016). The important applications of the bionanocomposites in various industries are summarized in table 1. Further, the nanoparticles can be divided into two types that are organic and inorganic nanoparticles. Organic nanoparticles may include carbon nanoparticles whereas inorganic nanoparticles may include semiconductor quantum dots etc. The nanoparticles are basically synthesized by two approaches *viz.* bottom up approach (BUA) and the top down approach (TUA). Biosynthesis of nanoparticles, which arises due to costly physical and chemical processes, fall under the category of BUA, which includes oxidation/reduction reaction (Sahayaraj, 2012).

**Table 1: Prospective applications of bio-nanocomposites in different fields**

| Sr# | Material Composition   | Applications  |
|-----|--|---|
| 01  | Brucite nanoplates-ST, ST-Gelatin- nanorod-rich ZnO, Wheat gluten –lignin NPs, Tilapia skin gelatin- hydrophilic-hydrophobic nanoclays, Soluble soybean polysaccharide-HNC, CNW-PLC, Kefiran-CNC from BIR, ZnO-neem-oil-CS, Potato ST-HNC, PLA lamination on AG-k-carrageenan-Cloisite Na+, AG-cellulose-savory essential oil, Poly(3-hydroxybutyrate-co-3- hydroxyvalerate)-functionalized CNTs, Whey protein isolate-MMT-citric acid | Food contact and food packaging   |
| 02  | CMC/CuO bio-nanocomposite hydrogels, Polyurethane-cellulose nanocrystals, Gold reinforced CS matrix-chloroauric acid, CS-calcium carbonate nanopowder, Chitin-Clay(bentonite) –polyurethane, PLA-hydroxyapatite-carbon nanostructures  | Biomedical applications   |
| 03  | PVA-Pineapple nanofibers-stryphnodendron adstringens bark extract, PLA-natural rubber-organo clay, Poly(Xylitol sebacate)-nanohydroxyapatite, PLA-amorphous magnesium phosphate, CS-halloysite nanotubes, Gelatin-hydroxyapatite graft copolymers  | Tissue engineering  |
| 04  | Epoxidized soybean oil-carboxylic acid functionalized multiwall CNTs   | For electronic appliances   |
| 05  | Wheat gluten –lignin NPs   | Agricultural bags industry  |
| 06  | Starch-based clay (cloisite Na <sup>+</sup> )  | Manufacturing of food containers  |
| 07  | Pyranose oxidase –Pt-MnOx, Pyranose oxidase –Pt-MnOx   | Biosensing with improved sensitivity-Fast response                                    |
| 08  | GOD-graphene, GOD-poly(3-anilineboronic acid)-Pd NPs   | Glucose biosensing  |
| 09  | Pectin-coated chitosan-LDH   | Colon-targeted drug delivery  |
| 10  | Procainamide hydrochloride-MMT-ALG /Procainamide hydrochloride-MMT)-CS   | <i>In vitro</i> drug release  |
| 11  | <i>Penicillium</i> sp-Fe <sub>3</sub> O <sub>4</sub>   | high performance adsorbents for the removal of radionuclides                          |
| 12  | MMT-CS   | Adsorbents of the herbicide clopyralid in aqueous solution and soil-water suspensions |
| 13  | <i>Pseudomonas</i> sp. strain ADP-LDH  | Microbiological applications  |
| 14  | <i>Jatropha Curcas</i> oil based alkyd-epoxy-graphene oxide  | Agricultural, medicinal,  |
| 15  | Soy protein isolate-MMT  | Packaging of high moisture food such as fresh fruits and vegetables                   |
| 16  | Xylan-MMT, Sorbitol-plasticized ST   | Cosmetics formulation   |
| 17  | CS-xanthan gum blend   | Encapsulating bioactive substances such as enzymes                                    |
| 18  | CS–vermiculite   | Foams for cadmium uptake  |
| 19  | Soluble soybean polysaccharide –HNC  | non- Food Packaging industries  |

| Sr# | Material Composition  | Applications  |
|-----|---|---|
| 20  | Poly(aniline-co-pyrrole)-Fe <sub>3</sub> O <sub>4</sub> - alginate acid   | Design of electrochemical biosensor, removal of heavy metal ions, anti-corrosion coating and medicine |
| 21  | CMC-ST-CNC  | Biomaterials, good optical transparency in packaging applications                                     |
| 22  | CMC-LDH   | Textile printing, paper industry, detergents  |
| 23  | RC-zeolite  | Biodegradable packaging, membranes-biomedical areas   |
| 24  | ZnO-AG/ZnO-CMC/ZnO-carrageenan  | Active packaging films to extend shelf- life of food  |
| 25  | ST-clay   | Design of sensors and actuators   |
| 26  | PLC-natural rubber-CNC, Lignin-PLC NPs  | Industrial applications   |
| 27  | ZnO-carboxy methyl chitosan   | Impart antibacterial and UV protection for cotton fabric  |
| 28  | CNW-PLC   | Automotive, medical, applications   |
| 29  | CMC-graphene nano-platelets   | Effective filler in biopolymer based films  |
| 30  | Bacterial NC-pectin   | Prebiotics against drying and gastrointestinal condition  |
| 31  | CS-silver, CMC-LDH, Magnetic graphite –ALG-Ibuprofen NPs  | Drug delivery system  |
| 32  | CS-organoclay   | Hexavalent chromium uptake  |
| 33  | Polysaccharide-sepiolite-polygorskite   | Environmental remediation   |
| 34  | Pea ST-CNW hydrolyzed from pea hull fiber   | Fabricate bionanocomposite films  |
| 35  | Thermoplastic ST-bacterial cellulose nanofibers   | Excellent reinforcement agents for the production of starch-based bionanocomposites                   |
| 36  | Thermoplastic ST-Cellulose nanofibers   | Moisture uptake of thermoplastic starch   |
| 37  | MMT-citric acid-Whey protein-Zein NPs   | Whey protein isolate based food packaging   |
| 38  | Fish gelatin-CS NPs   | Edible films for food packaging applications  |
| 39  | MMT-ST  | Ecofriendly process for the preparation of Ag-NPs in BNCs matrix                                      |
| 40  | Poly(Xylitol sebacate)-nanohydroxyapatite, Mg-HA-TiO <sub>2</sub> -MgO, Gelatin-hydroxyapatite graft copolymers | Bone cement, scaffolds.   |
| 41  | SiO <sub>2</sub> -fluorinated PLA   | Hydrophobic coatings on stone and similar building materials  |
| 42  | Myoglobin-gold-polydopamine-graphene NPs, Graphene-ionic liquid-CS  | third generation biosensors   |
| 43  | Almond shell flour–polypropylene  | Wood plastic composite, fiber industry  |
| 44  | Poly dopamine- Pt NPs   | High performance amperometric immunoassay, bioassay applications                                      |

| Sr# | Material Composition   | Applications   |
|-----|--|--|
| 45  | 1,6-hexanedithi-GOD-Fe <sub>3</sub> O <sub>4</sub> -Au-1,4-benzoquinone                                      | Biocatalysis, biofuel cells, bioaffinity separation  |
| 46  | Multi walled CNTs- monetite  | Cements for orthopedic applications  |
| 47  | NC-polyethyleneglycol  | Wound dressing applications  |
| 48  | PLA-PHB-CNC blends   | Compostable flexible film materials  |
| 49  | Mn/Zn ferrite-CS-NC of apatite NPs   | Diagnosis and treatment of breast cancer   |
| 50  | Sodium fluoride-silicic acid-aluminium chloride-magnesium chloride   | Environmental soil pollution control   |
| 51  | CS-poly(vinyl alcohol)-TiO <sub>2</sub>  | Packaging material for soft white cheese   |
| 52  | AG-CuNP-copper salts -   | Antibacterial activity against both gram-negative and gram-positive food-borne pathogenic bacteria |
| 53  | ST-Gelatin- nanorod-rich ZnO   | Packaging material for pharmaceutical industries   |
| 54  | Waterborne polyesteramide-Organically modified MMT   | Environmental friendly waterborne protective coatings  |
| 55  | Hydroxyapatite-titania   | Biomedical environmentally friendly antimicrobial applications                                     |
| 56  | Pseudomonas oleovorans-octanoic acid-PHO   | Cling films  |
| 57  | Vermiculite -amorphous polyamide   | Electronics packaging  |
| 58  | Senegalia (Acacia) Senegal-iron-silica   | Inhibited bacterial growth   |
| 59  | TiO <sub>2</sub> (rutile)-inulin   | Photocatalytic degradation of methylene blue   |
| 60  | Gluconobacter oxydans-CNTs   | Bioanode for mediated biosensor device   |
| 61  | CS-iron oxide  | Immunosensor for ochratoxin-A  |
| 62  | Bovine hydroxyapatite-diopside powder  | Coatings   |
| 63  | CNTs-GOD   | Potential bionanoelectronic applications, electrochemical detection of glucose                     |
| 64  | CS-Ag  | Bactericide material, food and biomedicine industries  |
| 65  | Poly- L-lactide-grafted CNC-PLA  | Solvent free industrially scalable fabrication process   |
| 66  | Cellulose-Nano SiO <sub>2</sub>  | Fiber manufacturing  |
| 67  | CS-ZnO   | Agricultural products and equipment, other outdoor applications                                    |
| 68  | Poly vinyl alcohol-poly (D,L-lactide-co-glycolide)-bovine serum albumin fluorescein isothiocyanate conjugate | Pharmacological properties appropriate for therapeutic applications                                |
| 69  | Poly(ethylene oxide)-chitin nanofiber networks   | Excellent candidate for reinforced, light weight, renewable materials                              |

| Sr# | Material Composition  | Applications   |
|-----|---|--|
| 70  | Silver nanoparticle and TiO <sub>2</sub> nanoparticles  | Desalination   |
| 71  | TiO <sub>2</sub> nanomaterials with CeO <sub>2</sub>  | Removal of organic contaminants                      |
| 72  | TiO <sub>2</sub> nanocomposites with mesoporous silica  | Elimination of aromatic contaminants from wastewater |
| 73  | Fe <sub>2</sub> O <sub>3</sub> nanoparticles  | Removal of colored humic acids from wastewater       |
| 74  | Fe <sub>2</sub> O <sub>3</sub> and Fe <sub>3</sub> O <sub>4</sub><br>Polymer-grafted Fe <sub>2</sub> O <sub>3</sub>       | Removal of heavy metals and ions                     |
| 75  | TiO <sub>2</sub> nanomaterials and zinc nanoparticles, TiO <sub>2</sub> nanocomposites with multi walled carbon nanotubes | Disinfecting agents                                  |

For the synthesis of bionanoparticles, basically microbes along with plants and animal cells/products have been used. For example, microbes belong to the *Aeromonas*, *Bacillus*, *Verticillium*, *Fusarium*, *Lactobacillus*, *Corynebacterium*, *Desulfovibrio*, *Plectonema*, *Aspergillus*, *Pseudomonas*, *Streptomyces*, *Rhodopseudomonas*, *Rhodopseudomonas capsulata*, *Schizosaccharomyces* etc. have been used for the synthesis of microbes-based bionanoparticles (Sahayaraj, 2012). Moreover, nanomaterials are also synthesized by either plants (*Aloe*, *Avenasativa*, *Azadirachta*, *Capsicum*, *Calotropis gigantea*, *Cinnammum*, *Cymbopogon flexuosus*, *Eucalyptus*, *Jatropha*, *Ipomoea*, *Medicago*, *Mentha*, *piperita*) or its bioactive compounds (geranial, citric acid, azadirachtin) along with bio-waste residues from different food sectors (Xu et al. 2019). Bionanoparticles play significant roles in the field of biology, environment, agriculture and medicine (Sahayaraj, 2012).

Bionanocomposites (BNCs) also known as bio-based nanocomposites are an emerging group of nanostructured hybrid materials. As per the concept of biocomposites, bionanocomposites can be described in two ways. Firstly, bionanocomposites prepared from renewable nanoparticles such as cellulose whiskers and MFC, and petroleum-derived polymers such as PP, PE, and epoxies. Secondly, bionanocomposites prepared from biopolymers such as PLA and PHA, and synthetic or inorganic nanofillers such as carbon nanotubes and nanoclay (Lee and Moon, 2020; Siqueira et al. 2010). The characteristics of the materials such as optical, electrical, mechanical and thermal properties can considerably increase by nanofillers. The features of composite materials

extensively depend on the mixture ratio of organic matrix and the nanofillers (Marquis et al. 2011). In the present decade, there are many new findings related to bionanocomposite formation and its industrial applications. This review contains recent progress in the field of bionanocomposites synthesis and applications.

#### Nanofillers for bionanocomposites

As we know, there are different types of composite materials and their function can be enhanced by a subclass of nanomaterials known as nanofillers. The application of nanofillers for increasing properties of polymers are well reviewed in many literatures (Bhattacharya, 2016). In the beginning, fillers were basically used for reducing the cost of polymeric products; but now it is used in various applications especially for strengthening of polymer's mechanical properties. Based on dimension, nanofillers are classified into three categories viz. one dimensional, two-dimensional and three-dimensional. One-dimensional nanofillers include nanotubes and nanowires; examples of two-dimensional nanofillers are nanoclays and graphene; and three-dimensional nanofillers include spherical and cubical nanoparticles (Lin, et al. 2007). Out of these, nanotubes and graphene that are carbonaceous nanofillers show excellent properties due to their high mechanical strength. In the polymer, nanofillers are incorporated with various mass concentrations such as from 1% to 10%. These nanofillers are included along with traditional fillers and additives, and eventually traditional reinforcement fibres such as glass, carbon or aramid fibers (Marquis et al. 2011). Nanofillers may be organic or inorganic in nature. The examples of inorganic fillers include particles such as silica (SiO<sub>2</sub>), titanium dioxide (TiO<sub>2</sub>),

calcium carbonate (CaCO<sub>3</sub>) etc.; however, organic filler comprises coir nanofillers, carbon black, cellulosic nanofillers etc. (Saba et al. 2014). Different types of nanofillers are described in the following paragraphs.

### Cellulose based nanofillers

In the process of nanocomposites production, some biopolymers including chitin, starch and cellulose can be used as matrix or as reinforcement filler (Feng et al. 2009). Cellulose is the most abundant biopolymer present in nature and found in various resources such as plants and microorganisms, and it is considered as one of the best natural materials for nanofillers to produce nanocomposites (Santos et al. 2016,;). These cellulose based nanofillers have been used in many natural and synthetic polymers and provide better performance of the nanocomposite in relation to their thermal and mechanical properties (Huang et al. 2014; Babaei et al. 2015; Santos and Tavares, 2015). However, there are some challenges in the application of cellulose-based nanofillers derived nanocomposites such as efficient dispersion of particles in the matrix and the compatibility of nanoreinforcement in the matrix. We have to overcome these problems for better performance of cellulose-based nanocomposites (Santos et al. 2016). Cellulose sources include vegetable lignocellulosic materials which are composed of carbohydrate polymers (cellulose and hemicellulose), and lignin (Adel et al. 2011). The important vegetable lignocellulosic materials that can be used to produce cellulose include wood, cotton, sugarcane, flax, wheat straw and others. Out of these, wood is the most significant source of cellulose due to its abundance and many other advantages. Plants are also a significant cellulose source, along with vegetables, due to their abundance in nature and to a preexisting harvesting, processing and handling infrastructure in the pulp and paper, packaging and textile industries (Moon et al. 2011). Bacterial cellulose mostly produced by *Acetobacter* that can utilize glucose, sugar, glycerol or other organic substrates and transform them into pure cellulose (Son et al. 2001). These bacteria can secrete cellulose microfibrils under the specific culture conditions and produce a thick gel known as pellicle (Gatenholm and Klemm, 2010). In the animal kingdom, Tunicates are the only (sea) animals that can synthesize cellulose. These animals have a mantle or tunic, which is composed of an integumentary tissue consisting

of cellulose microfibrils and a protein matrix. This mantle is used as a source of cellulose microfibrils in their mature phase (Moon et al. 2011). A number of algae species also produce cellulose microfibrils within the cell wall, which is composed of xylem and manna found in green algae (*Cladophora*), brown algae (*Phaeophyta*) and red algae (*Rhodophyta*) (Mohammad et al. 2014). Cellulose nanoparticles can be isolated by using several methods such as mechanical treatment (Siro and Plackett, 2010;; Dufresne 2012; De Gruyter et al. 2016; Khawas and Deka, 2016), acid hydrolysis (Santos and Tavares, 2015; Habibi et al. 2010; Yu et al. 2013; Guo et al. 2016), and enzymatic hydrolysis (Siro and Plackett, 2010; Cui et al. 2016; Hassan et al. 2014). However, to obtain a specific particle morphology combination of these approaches may also be used.

The potential and use of cellulose nanoparticles in nanocomposites for various industries have been comprehensively studied (Santos et al, 2016; Mishra et al. 2018). Along with other applications, Hubbe and co-workers mentioned other possible uses of cellulose whisker technology, together with their use in medicine (Hubbe et al. 2008,). Fluorescent cellulose nanocrystals have been produced by transforming the cellulose surface with fluorophores that could be used as markers in nanomedicine (Dong and Roman, 2007). Due to hydrophilic qualities of cellulose, it can also be used for the formation of hydrogels, which are extremely precise materials for medical and pharmaceutical (Hubbe et al. 2008). Some studies have recommended the use of cellulose nanoparticles in the development of support scaffolds for tissue or bone regeneration (Bodin et al. 2007).

### Carbon nanotubes

Since the last two decades, carbon nanotubes have attracted worldwide attention from researchers due to their unusual hollow structures and unique mechanical, electrical and thermal properties. In terms of density, carbon nanotubes are half as dense as aluminum and have 20 times more tensile strengths in comparison to high strength alloys (Bhattacharya, 2016;). The exceptional properties of carbon nanotubes such as high electrical conductivity and large aspect ratio make them outstanding nanofillers to synthesize nanocomposites with multifunctional properties (Tiwari et al. 2016). Other significant properties of carbon nanotubes include extraordinary mechanical properties including

tensile modulus of 1 TPa and tensile strength in the range of 50-150 GPa (Bhattacharya, 2016). The specific properties of carbon nanotubes are influenced by their structural perfection and high aspect ratio, typically ~300–1000 (Coiai et al. 2015). Also, polymer resins integrated with low carbon nanotubes contents demonstrate greater mechanical strength and stiffness (Tjong, 2010). The credit of carbon nanotubes discovery goes to two groups of researchers after their publications (Oberlin et al. 1976; Endo et al. 1976), but the application of carbon nanotubes was started after its rediscovery by Iijima (1991). Carbon nanotubes classified into three categories based on their nanometric diameter (Coiai et al. 2015; Marquis et al. 2011).

1-Single-wall carbon nanotubes (SWCNT): Single-walled carbon nanotubes are made-up of a single graphene sheet, wrapped into cylindrical tubes with diameter ranging from 0.7 to 2 nm and lengths of microns.

2-Double-wall carbon nanotubes (DWCNT): Double-walled carbon nanotubes having diameter between 2 to 4 nm.

3-Multi-wall carbon nanotubes (MWCNT): Multi-walled carbon nanotubes consist of concentric assemblies SWCNTs and are, therefore, characterized by larger average diameters (4 to 150 nm). Depending on the rolling direction (chirality) of the graphene layers, different SWCNTs structures may be generated showing either metallic or semiconducting characteristics.

Carbon nanotubes are manufactured by two possible methods such as catalytic chemical vapor decomposition process at medium temperatures (600-1000 °C) and an electric discharge (arc) process under helium at high temperature (3000 to 4000 °C). In both of these methods, production of carbon nanotubes are not single type but combination of SWCNT, DWCNT and MWCNT, that contains important catalytic residues at their surface (Marquis et al. 2011). The carbon nanotubes composites are basically used as structural materials; however, it is also considered for the potential applications in electromagnetic interference shielding, bipolar plates for fuel cells, chemical sensors, etc. (Tjong, 2010). Some studies reported that carbon nanotubes could promote osteoblasts and neuron proliferation, and found to be effective nano-carriers for several biomolecules such as proteins, DNA and carbohydrates. However, the cytotoxic nature of carbon nanotubes to human dermal cells is also reported by some researchers. Therefore, more research and endorsements are needed

before incorporation of carbon nanotubes nanocomposites into the human body (Tjong, 2010).

### Nanoclays

For the first time polyamide-6-based clay nanocomposites was synthesized by Toyota Research group in early 1990s, and that nanoclays was used as a reinforcement of polymer systems. The study recommended that the nanoclays not only influenced the crystallization process, but also responsible for morphological changes. Based on this study, many scientists and researchers have developed nanocomposites with improved properties by using a variety of clays and polymeric matrices. Nanoclays are considered as universal nanofiller, which belong to one of the large groups of clay minerals. Clay minerals are the basic constituents of clay raw materials and platy structure is the dominant morphology (Nazir et al. 2016). Clay minerals are hydrous silicates and naturally found as platelets with sheet-like structure stacked over one another. Montmorillonite (MMT) is a broadly used clay nanofiller, packed in between two silicate layers of an octahedral sheet of alumina. The diameter of alumino-silicates sheets has dimensions of 100–500 nm and thickness 1–5 nm. Due to these dimensions, the platelets have a high (>50) aspect ratio. In the process of polymer clay nanocomposites formation, either the polymer or the clay needs to be modified. Polymer clay composites can be categorized into three types *viz.* immiscible or conventional composites, intercalated nanocomposites and miscible/exfoliated nanocomposites (Bhattacharya, 2016).

Clay-based nanocomposites enhance physical performances. The most extensively used clay-based nanocomposites are the phyllosilicate that have a shell-shaped crystalline structure along with nanometric thickness (Marquis et al. 2011). Some of the natural and synthetic nanoclays, used as fillers in polymers, are summarized in table 2. Nanoclays have been used in many fields comprising medicine (Ambre et al. 2010; Suresh et al. 2010), pharmacy (Carretero and Pozo, 2010), cosmetics (Carretero and Pozo 2010), catalysis (Garrido-Ramírez et al. 2010; Nagendrappa 2011), food packaging (Majeed et al. 2013) and in environmental protection and remediation (Lee and Tiwari 2012; Ouellet-Plamondon et al. 2012).

**Table 2: Nanoclays identification (extracted from Marquis et al. 2011©).**

| Family                    | Group        | Formula  |
|---------------------------|--------------|--|
| Phyllosilicates           | TO(1:1)      | Kaolinite<br>The reference plate is formed from a tetrahedral plate T and an octahedral plate O. The thickness of the layer is about 0,7 nm. Kaolinite $Al_4Si_4O_{10}(OH)_8$  |
|                           | TOT(2:1)     | Smectite,(Talc, Mica, Mommorillonite), Sepiolite<br>Two tetrahedral plates T in both sides of an octahedral plate O form the reference plate. The thickness of the layer is about 1 nm. The group includes many minerals that are major constituents of clays. |
|                           | TOT:O(2:1:1) | Chlorite, Bentonite, Saponite<br>The reference plate is formed of three plates TOT and another isolated O plate. The thickness of the layer is about 1,4 nm. Chlorite di-tri $Al_2Mg_3Si_4O_{10}(OH)_8$  |
| Polysilicate              | Natural      | Kenyaite, Magadiite, Kanemite, Ilerite, Silhydrite, Zeolite.<br>Magadiite ( $Na_2Si_{14}O_{29}H_2O$ )  |
|                           | Synthetic    | Fluoro Hectorite, Zeolite  |
| Double lamellar hydroxide | Synthetic    | Hydrotalcite<br>Hydrotalcites: $(Mg_6Al_2(OH)_{16})(CO_3^{2-})_4H_2O$  |

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Long et al. (2018) developed an iron oxide-kaolinite nanocomposite ( $\alpha\text{-Fe}_2\text{O}_3\text{-kaolin}_{KAC}$ ) which is able to control hemorrhage. Moreover, the potential of nanoclays are well recognized in textile industry also (Shahidi and Ghoranneviss 2014; Nazir et al. 2016).

### Functional nanofillers

Bionanocomposites are basically made-up of biopolymers and different nanostructured inorganic/organic functional fillers. Some of the most studied functional nanofillers are cellulose nanofibers, hydroxyapatite (HAp), silica nanoparticles, polyhedral oligomeric silsesquioxanes (POSS) etc. (Reddy et al. 2013). Cellulose nanofibers are already described in the previous section of cellulose based nanofillers. Hydroxyapatite is a well-recognized bioactive and biocompatible ceramic present in bones and teeth. These nanofiller have nontoxic, non-inflammatory and non-immunogenic properties (Jevtic et al. 2008). Hydroxyapatite has hexagonal crystal structure with chemical formula  $Ca_{10}(PO_4)_6(OH)_2$ . The functionality of hydroxyapatite depends on its individual characteristics including structure such as size, shape, and crystallinity; its morphology, thermal stability, and solubility; and its synthesis process (Reddy et al. 2013). Silica nanoparticles or nanosilica (silicon dioxide nanoparticles) has capability to be functionalized with various

molecules and polymers along with their thermal stability, porosity, and low toxicity that make them a suitable candidate for biomedical research. It is classified into two categories viz. P-type and S-type depending on their structure. P-type nanoparticles have many nanopores with pore a rate of 0.61 ml/g. However, S-type nanoparticles have smaller surface area compared to P-type nanoparticles. In comparison to the S-type, the P-type has higher ultraviolet reflectivity. Silica nanoparticles are present in the form of a white powder with density and molar mass of 2.4 g/cm<sup>3</sup> and 59.96 g/mol; and the melting and boiling points of 1600°C and 2230°C, respectively. These nanoparticles are well-dispersed in polar solvents such as water and ethanol. There are too many techniques to synthesize silica nanoparticles, and mostly all the techniques employ sol-gel processing at 25 °C under controlled conditions of reactant to solvent ratios. The commonly used techniques for the synthesis of silica nanoparticles are described in detail by Liberman and coworkers (Liberman et al. 2014). Along with the application in nanocomposites as nanofiller, these nanoparticles are also used as an additive for rubber and plastics and in biomedical applications such as drug delivery and theranostics. It is also used in other areas such as electronics, sensor, and catalysis purposes.

Polyhedral oligomeric silsesquioxanes (POSS) are nano-sized (1–3 nm) hybrid materials

with the general formula  $(\text{RSiO}_{1.5})_n$  ( $n = 6, 8, 10 \dots 14$ ), where R may be a hydrogen atom or an organic functional group, e.g., alkyl, alkylene, acrylate, hydroxyl, phenyl or epoxide unit (Ayandele et al. 2012; Kuo and Chang, 2011; Cordes et al. 2010;). It may also be referred to as a silica nanoparticle consisting of a silica cage core and other organic functional groups. Due to the size of POSS nanostructures, they could be considered as the smallest existing silica particles. Based on the reactivity of organic groups, POSS may be classified in three categories such as molecular silica, monofunctional POSS and multifunctional POSS (Kuo and Chang, 2011). In molecular silica, all the organic groups are non-reactive. If there is one reactive organic group, these POSS are referred as monofunctional POSS or Mono POSS. However, if there are more than one reactive organic group, they are called as multifunctional POSS. Different types of POSS containing different R-groups along with their characteristics are already reviewed by Cordes et al. (2010). In comparison to other fillers, POSS molecules are compatible with many polymers because it contains organic constituents on their external surface (Kuo and Chang, 2011). The literature suggested that diffusing POSS nanoparticles into a polymer may increase the strength, modulus, rigidity and reduce the flammability, heat discharge and viscosity of the polymer (Camargo, et al. 2009). These POSS-containing polymer nanocomposites have been extensively studied and commercially used in drug delivery and as a thermoplastic and thermosetting polymers (Kuo and Chang, 2011; Cordes et al. 2010; Liu, 2012; Achilleos and Vamvakaki, 2010; Fina et al. 2010; Keledi et al. 2012).

### Industrial Applications of bionanocomposites

Bionanocomposites, also known as green composites, have been explored for various industrial applications including automobile industry (Agarwal et al. 2009). Cellulosic nanofibers have been commercially used as food and pharmaceutical packaging material including paperboards for dairy based and fruit juice products (Teeri et al. 2007). Green nanocomposites have also been used in critical domains such as tissue engineering and aerospace defense (Qu et al. 2010; Satyanarayana, 2015). Recently, due to their renewable nature and biodegradability biopolymers such as cellulose, starch and chitosan attracted the attention of the scientific community to exploit these materials for various

industrial applications (Manorama et al. 2011). Some of the significant applications of the bionanocomposites are described below in detail.

### Biomedical and pharmaceutical industry

The flexibility and biodegradability of bionanocomposites facilitate these materials to be utilized for biomedical applications. An important characteristic of medical biomaterials is its ability to function appropriately in the human body to produce the desired clinical outcome, without causing adverse effects. Biobased polymers (bionanocomposites) are increasingly being recognized as biocompatible materials for clinical use. For example, plastics and films made from corn-derived PDO have been shown to be non-cytotoxic and non-inflammatory to clinically relevant cell lines. Bionanocomposites with HAp and layered double hydroxides (LDH) have been explored for various biomedical applications. Moreover, soy-derived polymers have been demonstrated to be useful as bone fillers. Bionanocomposites that combine the tissue compatibility of natural polymers and bio-derived polymers along with the physical and chemical properties of nanoreinforcements will find widespread use in clinical medicine. In particular, three emerging areas of medical applications for bionanocomposites are tissue engineering, drug delivery, and gene therapy (Velu et al. 2020).

Bio-nanocomposites offer excellent mechanical properties, biodegradability and biocompatibility which make them the best suitable as ideal green nanocomposites for biomedical applications, such as in vaccination, drug delivery system, tissue engineering and wound dressings (Hitzky et al. 2013). Starch shows remarkable properties including nontoxicity, good biocompatibility, biodegradability, desirable mechanical properties that generates the need to develop starch based bionanocomposites and the latter is great use for biomedical applications (Lane, 2011; Eid, 2011; Gao et al. 2011; Schmitt et al. 2012; Valodkar et al. 2010). In the past, various hydrogels have been prepared using protein-based silver nanocomposites which can be utilized for various biomedical applications (Manjula et al. 2014; Reddy et al. 2013). On similar lines, wheat protein based hydrogel nanocomposites were exploited for antibacterial applications by Jayaramudu et al. (2013). It was also demonstrated that collagen was incorporated in curcumin films to heal dermal wounds (Gopinath et al. 2004). Fama and coworkers studied a combination of starch-based

nanocomposites and multi walled carbon nanotubes (MWCNT) that were successfully utilized for bone-regenerating treatments or as tissue scaffolds (Fama et al. 2011). Another material with excellent antimicrobial properties commonly used in the biomedical field for tissue engineering is hydrogels. A starch-based hydrogel has been synthesized using a gamma radiation polymerization technique (Eid, 2011). Abdel et al. developed nanocomposite hydrogel along with silver nanoparticles, polyacrylamide and starch (Abdel-Halim and Al-Deyab, 2014). Similarly, starch-based films with antibacterial properties were developed using silver nanoparticles for biomedical applications (Yoksan and Chirachanchai, 2010). Silver nanowires were prepared on a waxy starch matrix by Valodkar et al. that depicted excellent bactericidal action against Gram-negative and Gram-positive bacteria (Valodkar et al.2010). Presently, poly lactic acid (PLA) is one of the commonly used polymers used for biomedical applications as it can be synthesized under controlled conditions.

### 3.2. Food packaging industry

Bio-nanocomposites can be utilized as an eco-friendly, cost-effective and renewable film material for food packaging with improved antimicrobial activity. Some of the well-known bionanocomposites used for packaging appliances include cellulose and starch derivatives, poly-(butylene succinate) (PBS), polyhydroxybutyrate (PHB) and polylactic acid (PLA). Renewable resource-based biopolymers such as starch, cellulosic plastics, corn-derived plastics such as PLA, and polyhydroxyalkanoates (PHAs) are some of the most widely used biopolymers to produce nanocomposites for use of food packaging applications. Enhanced barrier properties of the bio-nanocomposites against O<sub>2</sub>, CO<sub>2</sub>, water vapor, and flavor compounds would have a major impact on extending the shelf-life of various fresh and processed foods. In addition, biodegradability of the bio-nanocomposites can be fine-tuned through proper choice of polymer matrix and nanoparticles, which is also a driver for the use of bio-nanocomposites in food packaging. Such property enhancements are generally attained at low nanoclay content (less than 5%) compared to that of conventional fillers (in the range of 10–50%). For these reasons, nanocomposites are far lighter in weight than conventional composite materials, making them competitive with other materials for specific applications such as in food packaging. It will help

to reduce the packaging waste associated with processed food and will support the preservation of packaged foods extending their shelf-life. Moreover, starch-clay bionanocomposites have been investigated for food packaging applications (Cyras et al. 2008). Bio-nanocomposite packaging materials appear to have a very bright future for a wide range of applications in the food and biomedical industries as well as innovative active and intelligent food packaging with bio-functional properties.

### Electronics industry

The advantage of cellulose nanofibers in fabricating transparent and flexible composites found wide applications not only for electronic devices such as displays, solar cells and organic light emitting diodes but also for roll-to-roll fabrication techniques. In roll-to-roll technique, continuous deposition of various functional components is facilitated leading to fabrication of electronic devices. The functional components used in this fabrication include metal wiring, active/gas barrier films. This technology has been widely used for the development of flexible electronic devices. However, the application of this technology for plastic materials (while used as a substrate for active components) suffers due to a high coefficient of thermal expansion (CTE). It is also reported that the addition of bacterial cellulose nanofibers into polymeric resin caused the reduction in CTE. They also suggested that such new composite materials can be used as the potential substrate for roll-to-roll fabrication. A wide range of research has been performed in order to enhance the thermal and tensile properties of petro/bio based polymeric materials for the purpose of roll-to-roll fabrication. As an example, fabrication of poly(lactic acid)/cellulose whiskers nanocomposites and found that the reinforcement of nanostructured cellulose whiskers enhanced their thermal and mechanical properties. They found that the both cellulose whiskers and their nanocomposites were thermally stable up to 220 °C, which indicates their suitability for various device applications.

### Environmental industry

Environmental issues, depletion in petroleum reserves and raising the oil cost in the present years are three critical factors to pay considerable attention to biodegradable polymers from sustainable resources. However, biodegradable polymers show many advantages; there are a few limitations in their functions i.e. low mechanical

strength and weak moisture barrier behavior. Kefiran, a polysaccharide, is extracted from kefir grain as a byproduct of the kefir manufacture process. Kefiran shows anti mutagenic, antimicrobial, mechanical and visual properties. Conversely, the WVP of kefiran is not as excellent as synthetic polymers, thus, it requires few modifications. Nanoparticles are added to biopolymers to provide them additional strength and barrier properties, although there are some problems regarding environmental pollution and toxic nature of nanoparticles. On the contrary, green nanoparticles like nanocellulose are harmless and biodegradable reinforcing substances. Acid hydrolysis is commonly used to produce cellulose nanostructure in which rod-like cellulose nanocrystals are isolated. The aspect ratio of cellulose nanowhiskers reduces to less than two when intensity of hydrolysis is sufficiently high. Such cellulose nanocrystals/spherical cellulose nanocrystals are prepared via beer industrial residues as crude material (Shahabi-Ghahfarrokhi et al. 2015).

In recent years, great interest has been drawn towards naturally occurring nontoxic biopolymers based eco-friendly adsorbents. Adsorbents prepared by alginate (heteropolymer constituted of guluronic acid and mannuronic acidic residues) and its derivatives have been broadly investigated because of their renewability, sustainability and biodegradability. Formerly, a variety of chemically modified alginate have been prepared as an adsorbent for heavy metal ions removal. Biopolymer composites incorporating inorganic nanoparticles have been receiving considerable attention as it improves the performance properties of the individual particles. Therefore, in present paper Au nanoparticles are incorporated owing to their large specific surface area, excellent biocompatibility, easy preparation and huge applications. Au nanoparticles are synthesized via green synthesis using glutathione and oxalic acid due to environmentally benign nature and greater stability of nanoparticles. Glutathione (reduced form have thiol and acetyl groups) acts as a capping and stabilizing agent whereas oxalic acid acts as a reducing agent in the formation of gold nanoparticles. For improving mechanical strength of the bionanocomposite, environment friendly mineral nanofiller, Mica, which is a layered aluminum silicate with reactive groups on its surface, has been reinforced. Furthermore, industrial wastewater usually contains many species of metals. Therefore, adsorption studies for binary system are realistic.

## CONCLUSION

Scientists have been studying bionanomaterials for over centuries. These days, bionanomaterials are playing a very important role in research of engineering and biomedical sciences because of their green and sustainable nature. Additionally, most of these are nontoxic and green reduction agents. Bionanomaterials show numerous businesses and are readily available for real time applications. However, even after the rapid growth of bionanomaterials in several engineering and sciences disciplines over the past few decades, there are still a lot of challenges to design new platforms that can integrate new technologies for the more advanced and substantial commercial output of bionanomaterials. The major challenges for bionanomaterials are their mass production, easy availability, simple refinement methods and their commercial applications without further processing. Researchers are finding potential solutions to these challenges to drive bionanomaterials from the laboratory terminals to the market. Overall, we can conclude that essential efforts are required to develop affordable and innovative technologies to bring these materials in the market for the use of modern society. It is anticipated; using modern technologies derived from nanoscience and biotechnology we might be able to produce more and desired materials for a wide range of real time consumer applications.

## CONFLICT OF INTEREST

The authors declared that present study was performed in absence of any conflict of interest.

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## AUTHOR CONTRIBUTIONS

GY collected literature and wrote the manuscript.

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