

An overview of the application of nanotechnology in food packaging: benefits and challenges

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ABSTRACT

Nanotechnology in food industry is applied in food production, processing, nutrition and packaging. Its application in food packaging offers smart, intelligent, light weight packaging, reduces food wastage by extending shelf life, enhances food safety, preserves the food color and flavor, prevents food borne diseases, enhances intake of healthy food by reducing the amount of sugar and salt added for food preservation. Addition of nanomaterials to food packaging enhances their durability, temperature resistance, flame resistance, barrier properties, optical properties, processability and recycling properties. This review report the application of nanotechnology in food industry and the focus will be on nanomaterials used in food packaging, their benefits and challenges.

Keywords: *nanopolymers, food pathogens, biomaterials, nanotechnology, food packaging, food preservation.*

INTRODUCTION

The shelf life and safety of food products is influenced by the processing technique and the nature of food package used. Food packaging is very important and protects food products from bacteria, compression, oxygen, water vapor, dust, agglomeration, temperature and makes handling convenient (Bix *et al.* 2003). It extends shelf life, provides important information on the usage and disposal of the food product and package (Bix *et al.* 2003, internet). Food-borne disease is a major safety concern in the food industry and some of the bacterial enteric pathogens that contribute to food related death are Salmonellosis (31%), listeriosis (28%), *Campylobacter* (5%), *Escherichia coli* (4.3%) and *Staphylococcus aureus* (0.8%) (Aymerich *et al.* 2006; Mead *et al.* 1999). Micro-organisms which causes food spoilage contribute to food wastage and huge economic losses. Chemical reactions also occur in food production resulting in reduced shelf life of perishable foods and this in turn affects the food quality (Coma, 2013). For instance, fat deterioration is due to reactions such as: auto-oxidative degradation of

lipids during storage and the oxidation process as a result of the presence of metals in fats (Coma, 2013). To inactivate the catalysis effect of these metals in the fats, sequestering agent *e.g.* citric acid is usually added.

Food contamination that is caused by oxidative processes, pathogen or spoilage micro-organisms usually result in food related death, diseases and wastage. And because of the aforementioned effects, there is a pressing need to develop food packaging that can extend shelf life, improve safety and also maintain the quality of the food. The types of food packaging materials used can be classified as: flexible and rigid plastics, bottles, paper and boards, ceramic, wood, metals, nanomaterials *etc.* Food contact with the packaging materials used is not expected to release chemicals into the food causing harm to human health. To determine the type of packaging material used for food products, the chemical composition of the packaging material and the levels at which these compounds can partition into

food products must be clearly studied and understood (Muncke, 2012).

Nanotechnology is focused on the manipulation of atomic, molecular and supramolecular scale of a matter to a size of 1–100 nm (Drexler, 1986). It is a very useful technology that has revolutionize various industrial sectors e.g. medicine, energy, information technology, environmental science, medicine, homeland security, food safety and transportation. Nano-based materials exhibit unique qualities such as larger surface area, improved optical, reactive and catalytic properties (Kahn, 2006). Most of the materials used for food packaging are non-biodegradable resulting in a serious global environmental problem. New materials that are bio-based, biodegradable and edible have been

developed for food packaging to extend shelf life and improve quality of food (Tharanathan 2003; Azeredo 2009). However, these bio-based materials exhibit serious limitations such as: they are expensive, brittle, have poor gas and moisture barrier. The application of nanotechnology to develop food packaging opens new possibilities such as: smart and intelligent packing, hygienic food packaging, light weight and strong packaging, extend shelf life of food products, improves and maintain food flavor and color, improves nutrient uptake, and is cost effective. Although nanotechnology offers huge advantages in food packaging, it also poses some risk to human health. This review will be focused on nanomaterials currently used in food packaging, their benefits and challenges.

NANOTECHNOLOGY APPLICATIONS

There are two approaches for preparing nanomaterials namely: bottom up and top down method. In bottom up method, atoms and molecules are assembled in nanostructures whereas in the top down method, the bulk materials sizes are reduced using nanolithography

or milling (Azeredo 2009, Foster and Konrad 2003). The top down method is used for commercial scale production. Nanotechnology offers several advantages based on it applications as listed in Table 1.

Table 1. Applications of nanotechnology (adapted from www.nano.gov and www.nanopro.biz)

Sector	Applications	Advantages
Aviation industry	Nanocomposites in advanced sensors and faster electronics	Data processing with reduced emission of carbon dioxide, lighter materials, less fuel consumption, reduced cost, improved functionality of materials, minimised risk and production of flexible and new systems
Agriculture sector	Biosensors	Detection of pathogens, removal of contaminants, high crop yields, reduction in the use of pesticides and improved water management
Automotive industry	Lubricant / hydraulic additives, nanoparticles in catalytic converters, fuel cells and hydrogen storage	Reduction of carbon dioxide, lighter materials and less fuel consumption.
Chemical industry	Fuel cells and catalysts	Reduction of waste and carbon dioxide
Energy generating industry It is used in	Thermal insulation and energy storage devices, DC-DC power converters, fuel cells, nanocomposites for high temperature applications for energy harvesting and storage.	Use of less energy and low production of carbon dioxide
Cosmetics industry	Sunscreens, beauty care products	It provides UV protection, enhances delivery of medicated skin products.
Creative Industry	Advanced display systems	Changing effects and advanced display systems
Security industry	Security body armour, chemical and biological sensors	For better detection and surveillance techniques
Electronics	For advanced display technologies	To provide faster, smaller and enhanced hand held

Sector	Applications	Advantages
	with conductive nanomaterials, quantum computing, data storage, printable and flexible electronics, magnetic nanoparticles for data storage	devices.
Environmental management		For air and water filtration, for removal of virus cells from water, waste and water treatment, hazardous materials disposal, in-building environmental systems, remediation and de-icing
In Medicine	Wound dressings, drug delivery devices, biological imaging for medical diagnostics and fluorescent biological labels	Better diagnostics and improved drug delivery
In household products	Water and air purification systems, deodorizers and anti-misting systems.	
Oil and Gas industries	For safety monitoring, improved oil recovery and well management	
Textiles Industry	Textile materials that are stain-resistant, self-cleaning and anti-bacterial coatings, wearable textiles with solar cells and sensors.	
Food Industry packing	Packaging materials, biosensors, food encapsulation for improved delivery	Packaging materials with improved barrier properties and heat-resistance, anti-microbial and anti-fungal packaging, smart sensing, biodegradable packaging, environmentally friendly for tracking, quality monitoring and anti-counterfeiting, to provide enhanced information on product.

Nanotechnology application in food industry

The applications of nanotechnology in food industry are still very new in most countries and are being investigated in food packaging. In 2006, the global market value for nano-enabled food and food packaging materials was estimated at around US \$4 million and this amount was expected to rise to US \$20 billion by 2010 (Chaudhry and Castle, 2011; Helmut Kaiser Consultancy, 2004) and to US \$6 billion by 2012 (Chaudhry and Castle, 2011; Cientifica, 2006). The market value is expected to rise to US \$15 billion by 2020 (<http://persistencemarketresearch.com/>, accessed 25th June 2015). Food packaging applications is estimated to make up the largest share of the current and short-term predicted market for nano-based products in the food sector such as: active and smart packaging, health-foods and functional food products (Chaudhry and Castle, 2011; Cientifica, 2006). The application of nanotechnology in food industry can be divided into four broad areas (Figure 1): food production, processing, nutrition and packaging (Duncan 2011).

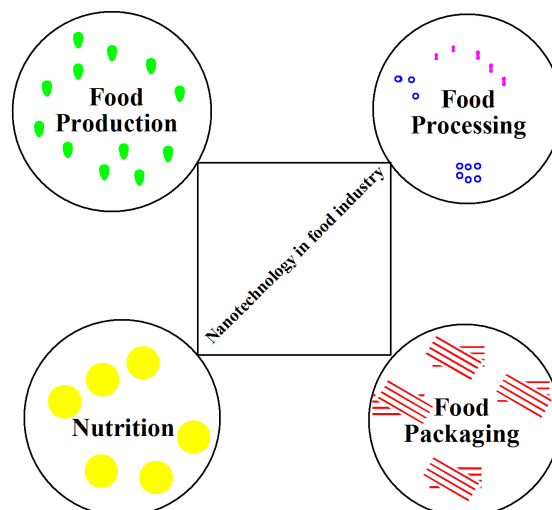


Figure 1. Nanotechnology application in food production

The application of nanotechnology in food production (Figure 2) include (Sekhon, 2014): nano-based formulations of pesticides for controlled release, fertilizers for crop improvement, biocides, veterinary medicines with enhanced therapeutic effects; improved nano-based nutritious animal feeds; nanosensors and nanobiosensors in crop protection and the identi-

fication of diseases and residues of agrochemicals; nanodevices for the genetic manipulation of plants and postharvest management. Other applications include nanoparticle-mediated gene or DNA transfer in plants for the development of insect-resistant varieties (Sekhon, 2014).

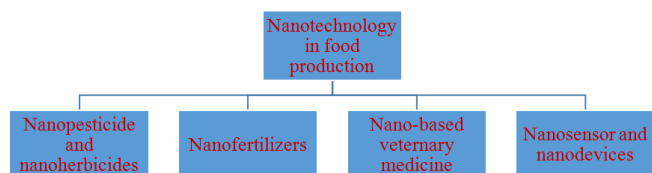


Figure 2. An illustration of the application of nanotechnology in food industry

There are research reports on nanoformulations of water soluble pesticides which improve water solubility. Microencapsulation of these formulations have enhanced their hydrophobic and dispersion in aqueous media, penetration in the plant and controlled the release of active compounds. Boehm *et al.*, reported the encapsulation of ethiprole, an insecticide onto nanospheres with enhanced penetration in plant when compared to the classical suspension of the insecticide (Boehm *et al.* 2003). In another research report, Adak *et al.* 2012 a & b, imidacloprid was encapsulated onto polyethylene glycol and various aliphatic diacids. They were found to be effective against selected pests of soybean *e.g.* stem fly, *Melanagromyza sojae* Zehntmer and white fly, *Bemisia tabaci* Gennadius. The encapsulated imidacloprid formulations exhibited better pest control than its commercial formulations. There are several other reports on the application of nanoformulations of pesticides with improved pest management and targeted killing of insects (Scrinis and Lyons, 2007). There are also several reports of nanoformulations of herbicides (Sarijo *et al.* 2010; Hussein *et al.* 2005; Hussein *et al.* 2007; Hussein *et al.* 2010, Hussein *et al.* 2012). Nanoformulation of sulfur nanoparticles with antifungal activity have been reported to be effective against phytopathogens responsible for early blight of tomatoes and apple scab disease (Rao and Paria, 2013).

Nanofertilizers exhibits ultrahigh absorption, increased crop production, enhanced photosynthesis and significant expansion in the

leaves' surface area. Their use has resulted in an increased efficiency of selected elements, reduced soil toxicity and frequency of application of fertilizers (Sekhon, 2014). In the encapsulation of nutrients onto nanomaterials, it is performed by coating with a thin protective film, as emulsions or nanoparticles (DeRosa *et al.*, 2010).

The applications of nanotechnology in food production also involves nanobiosensors that are used to detect fertilizers, herbicide, pesticide, insecticide, pathogens, moisture, soil pH for sustainable agriculture and improved crop production (Rai, Acharya and Dey, 2012). Nanosensors are also able to detect soil nutrient content, crop pathogens and plant viruses (Jones 2014; Brock 2011; Otles and Yalcin 2010). Nanotechnology helps scientists to understand plant roots adaptability to their environment (McLamore, 2010).

Nanotechnology in food processing (Nano-based food (nano inside))

Nanotechnology in food processing involves nano-based food. It includes the addition of nano-scale materials to food products and nanoencapsulation techniques for delivery of food components such as vitamins *etc.* (Nichols-Richardson, 2007). The nano-sized ingredients bioavailability can be increased resulting in reduced concentrations in food products (Weiss, Paul and Julian, 2006; Swarnali Das Paul and Divya Dewangan, 2014). Nano-food fortification offers improved appearance, taste, reduced fat, salt and sugar content and increases nutritional value of food products. Nano-based food (nano inside) involves the use of technique known as nanoencapsulation. It protects bioactive compounds, such as: carbohydrate, vitamins, antioxidants, proteins and lipids resulting in functional foods with improved functionalities and stability.

Nanoencapsulation of food products

The main purpose for encapsulation of food products is due to several advantages that it offers, such as (Zuidam and Shimoni, 2010): to protect the encapsulated products from environmental factors such as light, oxygen, and heat; to retain the properties of the products; to improve the stability of the product during

processing; for control release of the active ingredients; off taste masking; for immobility of active agents in food processing systems; for creation of visible and textural effects and for adjustable properties of active ingredients. Different techniques have been developed for encapsulation of food products (Figure 3). In general, these techniques may be divided into three main groups (Yáñez *et al.* 2003; Quintanilla-Carvajal *et al.* 2010): (a) physical processes such as spray drying–coating, spray drying and extrusion *etc.*; (b) physicochemical processes such as coacervation and entrapment into liposomes; (c) chemical processes such as interfacial polymerization and molecular inclusion. In the selection of the most suitable encapsulation process for selected food products, certain factors are usually taken into consideration such as: the required size of the particles, the physicochemical properties of the core material, the nature of the food product to be encapsulated, cost and the desired release mechanism (Yáñez *et al.*, 2003; Quintanilla-Carvajal *et al.*, 2010).

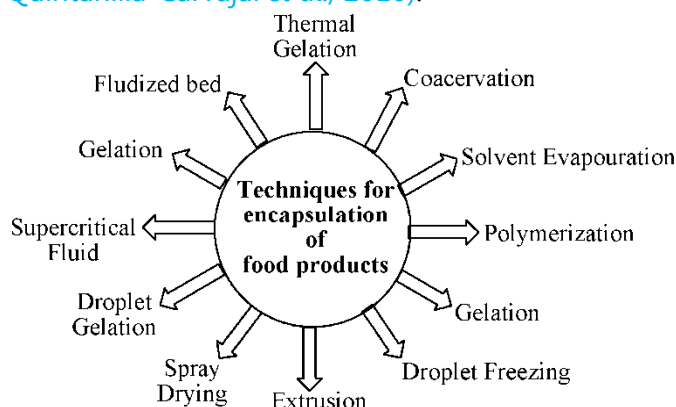


Figure 3. Techniques for encapsulation of food products

Nanoliposomes

Nanoliposomes are colloidal structures composed of phospholipids. They are closed, continuous, vesicular structures composed of phospholipid bilayers in an aqueous environment (Mozafari and Mortazavi, 2005). They can be prepared from natural sources, such as egg, soy, or milk *etc.* (Thompson *et al.*, 2007; Mozafari *et al.*, 2008). They are presently being investigated and developed as nanocarriers systems for the protection and delivery of bioactive agents in food, pharmaceutical and cosmetics industries (Mozafari *et al.*, 2008a). The mechanism for their formation is

by hydrophilic–hydrophobic interaction between phospholipids and water molecules (Mozafari *et al.* 2008b). Liposomes and nanoliposomes have been used for encapsulation of selected food products: flavoring agents such as oils, spices, seasoning and sweeteners; essential oils, amino acids, vitamins and minerals; leavening agents; antioxidants, antimicrobials, preservatives and omega 3 (Gibbs *et al.*, 1999; Mozafari *et al.*, 2006; Wilkinson and Kilcawley 2005; Mozafari *et al.* 2008b). They have been used for enzymes encapsulation for cheese processing and they protect casein from premature hydrolysis during cheese production, accelerate ripening of cheese, they partition well in the curd, increase shelf life of cheese products and fortify cheese products with vitamins and minerals (Mohammadi *et al.*, 2015; Payne *et al.*, 1986). Nanoliposome have also been used for encapsulation of antioxidants. Antioxidants protect the nutritive quality of the food and also protect the body against certain diseases (Mozafari *et al.* 2006 and 2008b). Antioxidants possess limited ability to cross cell membranes and are removed from the cells rapidly, resulting in low bioavailability (Mozafari *et al.*, 2006 and 2008b). Encapsulation of antioxidants onto nanoliposomes enhances their bioavailability.

Nanocapsules

Nanocapsules are nano-scale shells and consist of a polymer-based membrane and inner liquid core (Figure 4). The membrane acts as protective barrier and preserves the functionality and bioavailability of food additives (Gray, 2010). They are used for drug delivery, food enhancement and nutraceuticals *etc.* In food industries, they offer several benefits: protection of food products from environmental factors, for controlled release mechanisms, for changing food textures, flavorings, colorings and extend shelf-life (Ezhilarasi *et al.* 2012). Nanocapsules are also used for encapsulation of nutraceuticals (*i.e.* substances that are placed in food to enhance nutrition) for enhanced bioavailability. Lertsutthiwong and Rojsitthisak in 2011 reported the encapsulation of turmeric using chitosan-alginate based nanocapsules. Yi *et al.*, 2014, reported nanocapsules of jasmine essential oil via gelatin/gum arabic based complex coacervation.

The capsules possessed a good heat-resistance capability against humid heat (80 °C).

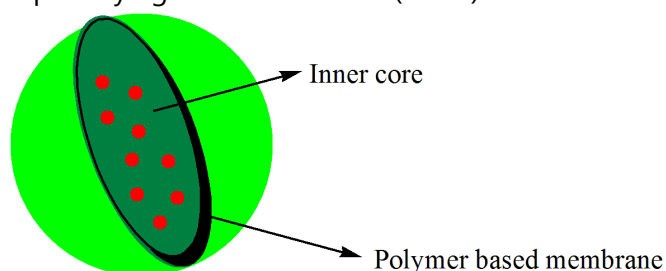


Figure 4. Schematic representation of a nanocapsule

Nanoemulsions

Nanoemulsions are nanocarriers that are used for the encapsulation of lipophilic compounds such as: nutraceuticals, antimicrobial agents, drugs, antioxidants and flavors (Silva *et al.*, 2012; Sanguansri and Augustin, 2006; Weiss *et al.*, 2008; Wissing *et al.*, 2004). Application of nanoemulsions in food products results in the use of less fat thereby offering a healthier option to the consumer. They are used in low fat nanostructured mayonnaise, spreads and ice creams (Chaudhry *et al.*, 2008; Cushen *et al.*, 2012). The use of nanoemulsion in ice cream has resulted in fat reduction from 8-16% to 1% (Hall, 2005; Cushen *et al.*, 2012). Encapsulation of lipophilic compounds to nanoemulsions have increased bioactivity, desirability and palatability of the compounds (McClements *et al.*, 2007). They enhance the bioavailability of the encapsulated food products due to high surface-to-volume ratio and small particle size (Acosta, 2009).

Nanomicelles

Nanomicelle is an aggregate composed of amphiphilic block copolymers, which aggregate into a core shell structure by hydrogen bond, electrostatic interaction and van der Waals force (Ai *et al.*, 2014). Micelles are spherical particles (5 to 100 nm) and they are used to encapsulate water-insoluble compounds such as: vitamins, lipids, antimicrobials and antioxidants. They have been used to encapsulate various food products such as: lycopene, lutein, omega-3 fatty acids. They have been used for encapsulation of alpha-tocopherol in fish oil and essential oils in carbonated beverages (Chen *et al.*, 2006; El and Simsek, 2011). AQUANOVA, a company in Germany produces nanomicelle-based carrier system with a diameter of 30 nm. They are used in

food and beverage products. This company has also incorporated curcumin into nanomicelles for enhanced absorption and bioavailability. NutraLease Ltd. Israel uses nanomicelles of diameter 30 nm to encapsulate various vitamin D and E, lycopene, phytosterol, lutein etc. for enhanced solubility and bioavailability. Shemen Industry has used nanotechnology (nano-sized self-assembled structured liquid) to produce canola active oil containing minute compressed micelles known as nanodrops (ElAmin 2006). NanoCluster™ delivery system for food products are prepared by RBC Life Sciences® Inc. USA. Some of the products are nanoceutical slim shake chocolates (ElAmin, 2006).

Nanofibres

Nanofibers are used as carrier systems for the delivery of antimicrobial agents, enzymes, drugs, colors, flavors, antioxidants and other functional compounds (Rezaei *et al.*, 2015; Shen *et al.*, 2011). They have diameters less than 10 nm, high surface area and offer high encapsulation efficiency. They can maintain the quality of food products (Fernandez *et al.*, 2009; Rezaei *et al.*, 2015). The delivery rate of nanofibres can be enhanced by increasing their surface area. They also act as delivery systems for microemulsions containing solubilized lipophilic functional compounds (Kriegel *et al.*, 2009). Some of these microemulsions containing solubilized lipophilic functional compounds are often susceptible to heat, light and oxygen. Nanofibres are useful in the stabilization of these compounds (Kayaci and Uyar, 2012). They are used for nutraceutical delivery in food formulations (Rezaei *et al.*, 2015).

Nanotubes and nanoparticles

Carbon nanoparticles are amorphous in nature, spherical with sizes in the range of 4–30 nm and useful for biological applications. Nanotubes have been found to be effective in destroying the cell wall of *E. coli* (Kang *et al.*, 2007). The application of nanotube has significant potential for use in food systems. Nanotube membranes have been employed for analytical purposes such as sensors for molecular identification of enzymes, antibodies, selected proteins and DNA (García *et al.*, 2010; Rouhi, 2002). However, nanotube membranes have very limited control of their structure and chemical affinity

thereby limiting their use in food industry (García *et al.*, 2010). Functionalizing nanotubes have resulted in nanotube membranes that can effectively separate molecules based on their molecular size and shape and their chemical affinity. Lee and Martin (2002) developed membranes containing mono-dispersed gold nanotubes. The interior of the nanotubes was hydrophobic and the nanotube membranes were able to transport neutral hydrophobic molecules. These technology has a potential to be used in future for the separation of food biomolecules with functional value (e.g. vitamin, peptides, proteins or minerals), which can be used for food fortification or the manufacturing of dietary supplements (García *et al.*, 2010, Moraru *et al.*, 2003).

Nanometer-sized particles have been prepared from food-based biopolymers such as proteins or polysaccharides (Gupta and Gupta, 2005; Ravichandran, 2010). Nanoparticles can be used to encapsulate and release functional ingredients to target cell/tissue. Most biopolymer based nanoparticles are prepared from polylactic acid. They have been used to encapsulate and deliver proteins, vaccines and drugs (Ravichandran, 2010; Riley *et al.*, 1999). Nanoparticles are added to foods to improve flow properties, color and stability during processing and to increase shelf life. Some examples of nanoparticle added to food are: aluminosilicate materials which are used as

anticaking agents in granular or powdered processed food products; anatase titanium dioxide used as food whitener and brightener additive and it is used in confectionery, some cheeses and sauces (Ashwood *et al.*, 2007; Powell *et al.*, 2000; Alfadul and Elneshwy, 2010).

Nanospheres

Nanospheres have size range of 50-1000 nm and are investigated for their potential application in food industry. Salvona Technologies developed a delivery system, MultiSal™ for delivery of multiple active ingredients e.g. water-soluble and fat-soluble ingredients (Shefer and Shefer, 2003). This system is a nanosphere containing food-approved hydrophobic materials encapsulated in moisture-sensitive or pH-sensitive bioadhesive microspheres. When the microsphere comes in contact with moisture, such as saliva, it dissolves and releases the encapsulated bioactive agents. They are easy to handle; stable, enhancing shelf life of food products, protects against oxidation, retains volatile ingredients; reduces the loss of flavor; masks taste; exhibits moisture-triggered controlled release mechanism thereby providing a high impact flavor burst; exhibits pH-triggered controlled release mechanism; exhibits heat-triggered release mechanism to release active ingredients at specific temperature; exhibits consecutive multiple release mechanism of active ingredients and enhances bioavailability of biologically active agents (Shefer, 2003; 2011).

NANOTECHNOLOGY IN FOOD PACKAGING

Packaging is used to maintain the quality and extend the shelf life of food products. Nanomaterials have been incorporated into polymers for reinforcement resulting in improved food packaging materials. Nanomaterials with enhanced mechanical and thermal properties have been developed for food packaging to protect food products from exterior mechanical, thermal, chemical or microbiological effects (Wesley *et al.*, 2014). The commonly reported type of nano-based food packaging is the nanocomposites.

Nanocomposites

Nanocomposites possess improved mechanical strength; reduced weight; increased heat resistance and improved barrier against

oxygen, carbon dioxide, ultraviolet radiation, moisture and volatiles (Sekhon, 2010). Due to the aforementioned properties, they are used for food packaging.

Polymer-clay nanocomposites

Incorporation of clay nanoparticles onto polymers to form polymer-clay nanocomposites are among the first nano-based food packaging. The most studied type of clay is montmorillonite (MMT). MMT is a hydrated alumina-silicate layered clay made up of edge-shared octahedral sheet of aluminum hydroxide between two silica tetrahedral layers (Weiss *et al.*, 2006). It is used as a nanocomponent to enhance the mechanical properties. It has a moderate negative surface charge useful for equilibrium layer spacing.

Composites that are prepared from the interaction between clays and polymers are (Ray *et al.*, 2006): in the non-intercalated nanocomposites, the polymer cannot intercalate between the silicate sheets; in the intercalated nanocomposites the separation of clay layers expand the interlayer spacing and in the exfoliated nanocomposites there is separation of clay platelets into random arrangements (Ray *et al.*, 2006). Exfoliated polymer-clay nanocomposites are referred to as the ideal nanocomposites, however, they are difficult to obtain (Ray *et al.*, 2006). They are prepared generally by three methods namely (Ray *et al.*, 2006): melt processing, solution method and *in-situ* interlamellar polymerization techniques (Figure 5).

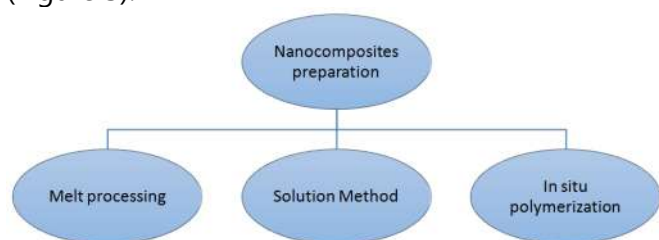


Figure 5. A schematic representation of the general methods for preparation of nanocomposites

In the melt processing technique, the clay filler is incorporated into the molten state of the polymer to form the nanocomposite material under a solvent-free condition (Ray *et al.*, 2006). In the solution method, the organoclay is swollen in a solvent followed by the addition of solution of polymer resulting in polymer molecules crawling between the silicate layers of the filler (Ray *et al.*, 2006). The solvent is evaporated resulting in intercalated/exfoliated nanocomposites (Ray *et al.*, 2006). The *in-situ*/interlamellar polymerization involves swelling of the layered silicates by absorption of a liquid monomer (Ray *et al.*, 2006). The monomer migrates into the galleries of the layered silicate resulting in polymerization within the intercalated sheets. Polymerization is initiated using either heat or radiation, diffusion of a suitable initiator, or by an organic initiator (Ray *et al.*, 2006). Polymer-clay nanocomposites possess several advantages such as thermal stability, barrier protection, mechanical strength and improved flammability properties. They exhibit

barrier properties against oxygen, carbon dioxide, ultraviolet, moisture and volatiles. Polymers used in food packaging have inherent permeability to gases and vapors thereby limiting their application in food packaging. Incorporating clay onto polymers has been reported to improve its barrier protection. The barrier properties of polymer-clay composites are attributed to the individual clay platelets which act as a maze or tortuous path thereby hindering the permeability of gas and vapor molecules into the polymer matrix ((Ray *et al.*, 2006). Akkapeddi *et al.*, (2003) reported Nylon-6 with good oxygen barrier properties which were enhanced by the addition of nano-scale silicate platelets via an *in-situ* polymerization process (Ray *et al.*, 2006). Sorrentino *et al.* (2006) developed a geometric model that can predict the effective diffusivity through polymer-clay composites as a function of clay sheet orientation, volume fraction, polymer-clay interaction and aspect ratio (Ray *et al.*, 2006). The type of organoclay used influenced the moisture absorption and diffusion (Ray *et al.*, 2006). Kim *et al.* (2005) reported reduced moisture permeability of octadecylamine-modified MMT when compared to quaternary alkylamine-modified MMT. This observation was attributed to larger interlayer distance in octadecylamine-modified MMT (Kim *et al.*, 2005, Ray *et al.*, 2006). Jang, Rawson, and Grunlan 2008, prepared a multilayer film from anionic sodium MMT and cationic polyacrylamide on a PET substrate. The films were potentially microwaveable with a good optical transparency and were found to be an alternative for aluminum foil in food packaging.

Some polymer-clay nanocomposites that are presently used for food packaging include (Chaudhry *et al.*, 2008): Imperm® resin produced by Nanocor® Inc. which is used to prepare multi-layer PET bottles and sheets used for food and beverage packaging. It reduces the loss of CO₂ from the drink keeping the beverages fresher thus, extending shelf-life; Aegis® OXCE high nylon barrier resin is manufactured by Honeywell inc. and it is used to prepare nanocomposite film used in co-injection PET bottle applications for packaging beer, fruit juice and soft drinks for retaining CO₂; Durethan® KU2-2601 manufactured by Bayer AG is a film with increased barrier

properties, used in paperboard juice containers (Chaudhry *et al.*, 2008; Joseph and Morrison, 2006).

Application of bio-based natural fibers for the preparation of nanocomposites for food packaging

Bio-based natural fibers are renewable biomaterials and are classified as either plant based or animal based. They are used as reinforcements to prepare light weight, low cost, eco-friendly, hygienic, naturally degradable and carbon dioxide neutral composites (Mahdavi *et al.*, 2010; Majeed *et al.*, 2013). They are biodegradable and do not cause any environmental hazards. However, they are not very attractive for commercial application because of their poor compatibility with hydrophobic polymer matrices, high moisture uptake and their variability in structure (Majeed *et al.*, 2013). The addition of natural fibers to polymer matrices have been found to improve the biodegradation of the composite materials (Majeed *et al.*, 2013; Kim *et al.*, 2006; Contat-Rodrigo and Greus, 2002; Tserki *et al.*, 2006). The three main components that are found in natural fibers are: cellulose, hemicellulose and lignin and the percentage of these components vary in different natural fibers. Other components that are found in natural fibers are waxes, pectin, inorganic salts and nitrogenous salts (Majeed *et al.*, 2013). The mechanical properties of natural fibers are influenced greatly by the cellulose component (Mwaikambo and Ansell, 2006). One of their main component, lignin regulates the transport of liquid in the plant and is composed of phenylpropane derivatives (Majeed *et al.*, 2013).

Polymer-cellulose nanocomposites

Cellulose is a natural polymer and is a plant based bio-based fiber. It is widely available, requires low energy in manufacturing, easily recyclable, environmentally friendly and affordable (Azeredo, 2009). Due to the aforementioned properties, it is a nanomaterials used for the preparation of affordable, lightweight and high-strength nanocomposites (Azeredo 2009; Helbert, Cavaille and Dufresne, 1996; Podsiadlo *et al.*, 2005). Two types of nanoreinforcements can be obtained from cellulose namely: microfibrils and whiskers (Azeredo 2009; Azizi Samir *et al.*, 2005). The

microfibrils have diameters of 2–20 nm and their length are in micrometer range and this is dependent on their source (Azizi Samir *et al.*, 2005). The whiskers have a diameter of 8–20 nm and lengths ranging from 500 nm to 1–2 μm (Azizi Samir *et al.*, 2005). The properties of polymer-cellulose nanocomposites have been reported to be strongly related to the geometric and mechanical percolation effects, dimensions and consequent aspect ratio of the fibers (Dubief *et al.*, 1999; Hubbe, Rojas, Lucia and Sain, 2008; Azeredo, 2009). The aspect ratios are dependent on the origin of the cellulose used and whisker preparation conditions (Azeredo 2009; Azizi Samir *et al.*, 2005). There are several reports on the effects of the addition of cellulose to matrix of synthetic polymers. Helbert, Cavaille and Dufresne, in 1996 reported poly(styrene-co-butyl acrylate) latex film containing 30 wt.% of straw cellulose whiskers. The modulus was more than a thousand times higher than that of the bulk matrix without the straw cellulose whiskers. The increased modulus was attributed to the stiffness of the whiskers, the formation of a fibril network within the polymer matrix and the linkage of the cellulose fibers by hydrogen bonds. Other researchers have also reported the effects of cellulose fibers in improving strength and modulus of polymers at temperatures above the glass transition temperature (T_g) of the polymer matrix (Dufresne *et al.*, 2000; Dufresne and Vignon, 1998). However, hindered elongation due to the addition of fibers to polymer matrix was also reported by some researchers (Freire *et al.*, 2008; Kim *et al.*, 2009; Wang and Sain, 2007). Other researchers reported that polymer elongation was improved by the addition of fibers (Pettersson and Oksman, 2006b; Zimmermann *et al.*, 2004; Azeredo, 2009; Wu *et al.*, 2007). The elongation of polyurethane was improved by cellulose nanofibrils but decreased by conventional micro-scale cellulose filler. This observation suggested that the degree of matrix-cellulose interactions influences the polymer elongation (Azeredo, 2009). Jordan *et al.*, 2005 reported that the addition of nanoreinforcements that exhibit poor interaction with the polymer matrices results in a decrease in the elongation

and strength of the material; however, modulus is dependent on these types of interactions.

The effects of cellulose nanoreinforcements on performance of starch have been investigated by some researchers. Starch-based materials enhance the biodegradability of plastics. However, starch are very brittle and as such, plasticizers are used to improve its flexibility. However, the addition of plasticizers results in a decrease in the thermomechanical properties of starch. Lima and Borsali, 2004 reported that the addition of whiskers to starch improves their thermomechanical properties, reduces water sensitivity, and maintains their biodegradability. An increased in Tg effects due to the addition of cellulose fibers to starch was also reported and this effect is as a result of the formation of a cellulose microfibrils network within the matrix, resulting from hydrogen bonds which can be formed during the evaporation step (Angles and Dufresne 2001; Angles & Dufresne, 2000).

Polymer films containing cellulose have been found to exhibit improved moisture barrier (Paralakar, Simonsen, & Lombardi, 2008; Sanchez-Garcia, Gimenez, & Lagaron, 2008). The presence of cellulose is believed to increase the tortuosity in the materials resulting in a slow diffusion process and, hence, low permeability (Sanchez-Garcia *et al.*, 2008). Nano-sized cellulose fibrils have been reported to improve thermal properties of polymers (Helbert *et al.*, 1996; Oksman *et al.*, 2006).

Starch-based nanocomposites

There are several reports on starch-based nanocomposites. Starch is biodegradable, available and affordable (Tang *et al.*, 2012). De Carvalho *et al.*, 2001 reported the preparation and characterization of starch-kaolin composites by melt intercalation techniques. There are also other research reports on the preparation of starch/clay nanocomposites by melt intercalation (Tang *et al.*, 2012; Park *et al.*, 2002; 2003; Wilhelm *et al.*, 2003; Avella *et al.*, 2005; Chen and Evans, 2005; Huang *et al.*, 2005; Pandey and Singh, 2005; Chiou *et al.*, 2007). Some of these nanocomposites exhibited an increased tensile strength and a decrease in water vapor transmission rate (Park *et al.*, 2002; 2003; Wilhelm *et al.*, 2003; Huang *et al.* 2005). Chiou *et*

al., 2007 reported an improvement of thermal stability and water absorbance of wheat starch/MMT nanocomposites. In some other reports, starch-polymer based nanocomposites were found to be potential food packaging. Biodegradable polyester blends were used for the preparation of starch/polyester/clay nanocomposites (McGlashan and Halley, 2003; Kalambur and Rizvi, 2004; 2005). Different methods were used, such as: melt extrusion method (McGlashan and Halley, 2003); reactive extrusion method (Kalambur and Rizvi, 2005).

Soy protein based nanocomposites

Soy proteins are plant based biopolymers and they exhibit good biodegradability (Tang *et al.*, 2012). They are very brittle and in their application for the preparation of nanocomposites, plasticizers are added to overcome the brittleness. The addition of plasticizers decreases the tensile strength of the nanocomposite (Tang *et al.*, 2012). The tensile strength can be enhanced by the addition of reinforcing materials such as nanoparticles. There are some research reports on soy protein/clay nanocomposites (Yu *et al.*, 2006, 2007; Chen and Zhang, 2006). The electrostatic attraction and hydrogen bonding on the interfaces of soy protein and clay resulted in good dispersion of clay in the protein matrix. The strong interaction between soy protein and clay improved the mechanical strength and thermal stability of the nanocomposites.

Gelatin based nanocomposites

Gelatin is an animal based biopolymer. It is obtained by hydrolytic cleavage of collagen chains (Tang *et al.*, 2012). It has poor mechanical properties. Zheng *et al.*, 2002 reported the preparation of gelatin/clay nanocomposites. The nanocomposites tensile strength and Young's modulus improved and it varied with the content of clay and the pH of gelatin matrix. Rouhi *et al.* reported the preparation of fish gelatin-based nanocomposites using ZnO nanorods as fillers (Rouhi *et al.*, 2013). These bio-nanocomposites had great potential for applications in food packaging materials. Kavosi *et al.*, reported gelatin composite films prepared containing multi-walled carbon nanotubes as nanofiller. Incorporation of MWCNT decreased the water solubility, water

swelling, water uptake, and water vapour permeability of the composites. The composites showed significant antibacterial activities against both gram-positive and gram-negative bacteria suggesting that they are potential food packaging materials (Kavoosi *et al.*, 2014). Nagarajan *et al.*, reported nanocomposites films prepared from skin gelatin incorporated with hydrophilic and hydrophobic montmorillonite nanoclays for food packaging (Nagarajan *et al.*, 2014). There are other research reports on gelatin based nanocomposites (Bae *et al.*, 2009; Shakila *et al.*, 2012; Farahnaky *et al.*, 2014; Jongjareonrak *et al.*, 2008).

Gluten Based Nanocomposites

Gluten is a protein composite found in wheat. It is used for food packaging applications because of its high gas barrier properties and hydrophobic nature (Tag *et al.*, 2012). Zhang *et al.*, 2007 reported the preparation of wheat-gluten based nanocomposites by dispersing Cloisite-30B nanoclay particles into plasticized wheat gluten systems under thermal conditions. Tunc *et al.*, 2007, reported the preparation of wheat gluten/montmorillonite nanocomposite films by casting. The presence of montmorillonite resulted in the decrease of water sensitivity and improved tensile properties of the nanocomposites. Olabarrieta *et al.*, 2006 also reported wheat gluten based nanocomposites. In their study, two types of nanoclay, Cloisite Na⁺ montmorillonite and Cloisite 10A (quaternary ammonium modified montmorillonite) were used. The prepared nanocomposites displayed decreased water vapor permeability. El-Wakil *et al.*, 2015 developed bio-nanocomposites by casting/evaporation of wheat gluten (WG), cellulose nanocrystals, and TiO₂ nanoparticles. They exhibited antimicrobial activity against *Saccharomyces cerevisiae*, gram-negative bacteria *Escherichia coli* and gram-positive bacteria *Staphylococcus aureus*. There are several other reports on gluten based nanocomposites for food packaging (Rafieian *et al.*, 2014; Song and Zheng, 2008).

Pectin based nanocomposites

Pectins are biopolymers. They are non-toxic and renewable materials. They have excellent film forming capability and are used as edible films in bioactive packaging. Pectin based films have poor mechanical properties, low thermal stability and

poor moisture barrier (Gorrasi *et al.*, 2012). They are enhanced by the addition of nanoreinforcements. Moreira *et al.*, 2013, prepared nanocomposites containing high-methoxyl and low-methoxyl pectins by casting method. The nanocomposites displayed improved mechanical and thermal properties than the control nanocomposites. Mg(OH)₂ nanoplates added improved the properties of the low methoxyl pectin. The pectin-Mg(OH)₂ nanocomposites released magnesium hydroxide by contact, demonstrating their potential for magnesium supplementation in bioactive packaging. Mangiacapra *et al.*, 2006 reported the preparation of natural pectins and MMT nanocomposites with improved mechanical and barrier properties. Other researchers have reported pectin based nanocomposites (Alves *et al.*, 2011; Moalemiyan *et al.*, 2012).

Carbon nanotube based nanocomposites

Carbon nanotubes (CNTs) are classified as single-walled carbon nanotubes (SWNTs), double-walled carbon nanotubes (DWNTs) and multi-walled carbon nanotubes (MWCNTs) (Lijima and Ichichashi, 1993; Zhou *et al.*, 2004). They exhibit high tensile strength, excellent mechanical properties, can be used as polymers reinforcing agents and as a filler for polymer nanocomposites (Sahoo *et al.*, 2010; Lau and Hui, 2002). Asgari *et al.*, 2014 prepared nanocomposite low-density polyethylene films with carbon nanotube base by solution casting. Their potential for packaging of fresh Mazafati dates were studied at ambient temperature. The nanocomposites films increased the shelf life of Mazafati dates. Kim, Han, and Hong 2008 reported CNTs based nanocomposites prepared from modified CNTs for enhanced intermolecular interactions with poly(ethylene-2,6-naphthalene) matrix. The addition of CNTs improved the thermal stability, tensile strength and modulus of poly(ethylene-2,6-naphthalene). Kavoosi *et al.*, 2014 prepared nanocomposite films from gelatin solutions using multi-walled carbon nanotubes as nanofiller. Incorporation of MWCNT resulted in a decreased water solubility, water swelling, water uptake and water vapor permeability. The nanocomposites also exhibited increased tensile strength, decreased elongation at break and significant antibacterial activities against

both gram-positive and gram-negative bacteria. There are other reports on the preparation of CNTs based nanocomposites from synthetic polymers with enhanced tensile strength improved by the addition of CNTs, such as polyvinyl alcohol (Bin, Mine, Koganemaru, Jiang, & Matsuo, 2006; Chen, Tao, Xue, & Cheng, 2005), polypropylene (Lopez Manchado, Valentini, Biagotti, & Kenny, 2005), polyamide (Zeng *et al.*, 2006) and PLA (Brody, 2006).

Chitosan based nanocomposites

Rhim *et al.*, prepared chitosan-based nanocomposite films by solvent-casting method by incorporation of selected nanoparticles such as: unmodified montmorillonite (Na-MMT), an organically modified montmorillonite (Cloisite 30B), a nano-silver, and a Ag-zeolite (Ag-Ion). The nanocomposite films with the highest intercalation were the Na-MMT-incorporated films. The nanocomposite films showed improved mechanical and barrier properties with decreased water vapor permeability (Rhim *et al.*, 2006). Rhim *et al.* 2013 also reported strong antimicrobial activity of chitosan-based nanocomposite against a range of microorganisms, especially against Gram-positive bacteria. The antimicrobial activity was attributed to the quaternary ammonium group in the organoclay and the antimicrobial activity of chitosan. There are several other reports on chitosan based nanocomposites (Kasirga *et al.*, 2012; Wang *et al.*, 2006; Kaur *et al.*, 2013; Ali *et al.*, 2010; Priya *et al.*, 2014; Tripathi *et al.*, 2011; Haerudin *et al.*, 2010).

Edible nanocomposites

Edible nanocomposite films and coatings are thin, continuous layers of edible material which are used as a coating or as a film placed between food components to provide a barrier to mass transfer (Balasubramaniam, Chinnan, Mallikarjunan, & Philips, 1997; Guilbert *et al.*, 1997). Edible films thickness is less than 254 μm and sheets thickness is greater than 254 μm (Janjarasskul and Krocht, 2010). Their preparation and applications for food differs. Edible coatings are applied directly on food products by addition of a liquid film forming solution or by molten compounds. Edible films are prepared and then applied to foods. They are prepared by casting or by traditional plastic

processing techniques, such as extrusion (Baldwin, 1994; Park *et al.*, 1996). The components of edible films and coatings can be classified as water-soluble polysaccharides and lipids. The polysaccharides include cellulose derivatives, alginates, pectins, starches, chitosan etc. (Ayranci and Tunc, 1997; El Ghaouth *et al.*, 1991). However, how easily these substances can form films of good integrity differ greatly (Park, Chinnan and Shewfelt, 1994). The water-soluble polysaccharides provide thickening quality, adhesiveness, mouth-feel, hardness, viscosity, crispness, compactness and gel-forming ability (Whistler & Daniel, 1990).

Lipid compounds such as waxes, acylglycerols, fatty acids, animal and vegetable fats are used to make edible films and coatings. Lipid films exhibit excellent moisture barrier properties. Waxes are used to coat fruits and vegetables so as to reduce respiration and moisture loss (Avena-Bustillos, *et al.* 1994a; Avena-Bustillos, *et al.* 1994b). They also extend shelf life of fruits (Drake, Fellman and Nelson, 1987). Some of them are used with a supporting matrix due to their fragile nature e.g. fatty acids and fatty alcohols. Composite films can be prepared from a combination of lipid and water soluble polysaccharides (Greener and Fennema, 1989; Kamper and Fennema, 1984). Azeredo *et al.*, 2009 reported the preparation of nanocomposite edible from cellulose nanofibers in different concentrations to mango puree based edible films. The cellulose fibers contributed to the increased in tensile strength and improved water vapor barrier of the films. Edible coatings and films are used to incorporate food additives for enhanced color, flavor and texture, hinder microbial growth and long-term storage of food product (Siragusa and Dickson, 1992).

Nanolaminates are edible food coating and are consisted of two or more layers of material of 1–100 nm per layer (Tarver, 2006). They are used in food-industry. They are prepared from polysaccharides, proteins and lipids. Those prepared from lipids are used to protect food from moisture, however, they exhibit limited resistance to gases with poor mechanical strength (Park, 1999; Tarver, 2006). Presently, food products are coated with nanolaminates by dipping them into a series of solutions that would adsorb the food

product surface or by spraying substances onto the food surface (McClements *et al.*, 2005). The degree of adsorption depends on the nature of the food product surface and the adsorbing substance. Different adsorbing substances form different layers of a nanolaminate and they include selected functional agents such as: antimicrobials, anti-browning agents, antioxidants, enzymes, flavors and colors. Edible coatings and films are used on a wide variety of foods, fruits, vegetables, meats, chocolate, candies, bakery products and French fries (Morillon *et al.*, 2002, Cagri *et al.*, 2004; Ozimek *et al.*, 2010).

Antimicrobial nanocomposites

Antibacterial agents are useful in various industrial sectors such as: environmental, synthetic textiles, packaging, healthcare, medical care, food *etc.* They are classified as organic and inorganic (Emamifar, 2011 a&b). Organic antibacterial materials are less stable at high temperature and pressure. The inorganic antibacterial agents are more stable particularly at high temperature and pressure (Emamifar, 2011). The introduction of antimicrobial agent onto food packaging materials reduces the growth rate of microorganisms resulting in extended shelf life, preserving food product quality and safety (Emamifar, 2011 a&b). There are different forms of antimicrobial food packaging such as: sachet pads that contain volatile antimicrobial agents are added into packages; coating or adsorbing antimicrobials onto polymer surfaces; immobilization of antimicrobials to polymers by ion or covalent linkages, incorporating volatile or non-volatile antimicrobial agents directly into polymers and the application of polymers that are antimicrobial (Emamifar, 2011; Appendini & Hotchkiss, 2002).

The direct addition of antimicrobials to food packaging can result in the antimicrobial leaching into the food product resulting in a cross reaction with food components (Emamifar, 2011). There are several reports on the modification of polymeric matrices with antibacterial agents to prevent growth or reduce adhesion of detrimental microorganisms.

Emamifar *et al.*, 2010 prepared antimicrobial nanocomposite films containing silver and zinc oxide. The nanocomposite films were evaluated and were found to preserve and extend the shelf

life of orange juice. Emamifar *et al.*, 2011a, continued their studied using the silver and zinc oxide nanocomposite packaging orange juice inoculated with *Lactobacillus plantarum*. The microbial population was less in the juice packaged in silver and zinc oxide nanocomposite when compared to those packaged without silver and zinc oxide. Li *et al.*, 2009 prepared nanocomposite polyethylene film with Ag nanoparticles and they retarded the senescence of jujube, a Chinese fruit. An *et al.*, 2008 reported a coating containing Ag nanoparticles for packaging asparagus. The coating decreased microbial growth and increased the shelf life of asparagus. Silver based nanocomposites were also reported to prolong shelf life of fruits and vegetables (Hu and Fu, 2003). Damm, Munstedt, and Rosch 2008, compared the efficacy of polyamide silvernano and microcomposites. The nanocomposites with low content of silver displayed increased efficacy against *Escherichia coli* than the microcomposites with a higher content of silver. Damm, Munstedt, and Rosch 2007 also reported that polyamide silver nanocomposite was effective against *E. coli* after immersion in water for 100 days. The mode of actions of silver nanoparticles antimicrobial activities have been proposed to be under some research: adhesion to cell surface resulting in degrading lipopolysaccharides, forming pits in the membranes and increased permeability of the nanoparticles into bacterial cell (Azeredo, 2009; Sondi and Salopek-Sondi, 2004); by damaging DNA (Li *et al.*, 2008) and by releasing antimicrobial Ag⁺ ions by Ag nanoparticles dissolution (Morones *et al.*, 2005).

Titanium dioxide have been used to inactivate several food pathogenic bacteria (Kim, Kim, Cho and Cho, 2003; Robertson, Robertson and Lawton, 2005). Chawengkijwanich and Hayata, 2008 prepared titanium dioxide powder-coated packaging film and it reduced *E. coli* contamination on food surfaces. Some researchers prepared nanocomposites from a combination of titanium oxide nanoparticle, silver nanoparticles and polyvinylchloride. These nanocomposites exhibited excellent antibacterial activity (Cheng, Li, Pavlinek, Saha and Wang 2006).

Poly(lactic acid) (PLA) nanocomposites

PLA is a biocompatible, biodegradable material with good mechanical and optical properties. The monomer of PLA is lactic acid which is obtained from the fermentation of carbohydrate feedstock. It has a high permeability for gases such as oxygen and water vapor which makes them useful for food packaging. Svagan *et al.* 2012 reported improved PLA for oxygen and water vapor sensitive food packaging. There are also other reports on the preparation of PLA nanocomposites in which reinforcement such as nanoclay and silicates are used to improve the mechanical and oxygen barrier properties (Cabedo *et al.*, 2006; Sinclair, 1996; Thellen *et al.*, 2005; Fukushima *et al.*, 2009 and 2012; Di *et al.*, 2005; Pluta *et al.*, 2002).

Silicate nanocomposites

Addition of silica nanoparticles to polymer matrices have been reported to improve the mechanical and barrier properties. Wu, Zhang, Rong, and Friedrich 2002 reported the preparation of nanocomposites by the addition of silicate nanoparticles to polypropylene. The nanocomposites exhibited improved tensile properties. Vladimirov *et al.*, 2006 incorporated silicate nanoparticles to isotactic polypropylene (iPP) matrix and maleic anhydride grafted polypropylene (PP-g-MA) was used as a compatibilizer. The nanocomposites were stiffer which improved the oxygen barrier. In another report, nanocomposites were prepared by radical co-polymerization of vinyl silica nanoparticles and vinyl acetate. The nanocomposites displayed improved thermal and mechanical properties. (Jia, Li, Cheng, Zhang, and Zhang 2007). Tang, Zou, Xiong, and Tang, 2008, prepared silicates nanocomposite biodegradable films that exhibited improved tensile strength and water resistance.

Intelligent packaging

Due to an increasingly complex society there is a pressing need for the integration of technologies into food packaging to guarantee food safety, quality and traceability (Vanderroost *et al.*, 2014). The aim of integrating these technologies is to reduce waste by efficient use of resources which process optimization, recycle and reuse. Intelligent packaging monitors changes in

the food product or the environment (Vanderroost *et al.* 2014). The three major technologies that exist in intelligent packaging are (Vanderroost *et al.* 2014; Kerry *et al.* 2006): biosensors, indicators and radio frequency identification (RFID) systems. The difference in these three technologies is based on the physical composition, amount and type of data that they can capture and how the data is captured and distributed (Vanderroost *et al.*, 2014; Heising *et al.*, 2014).

Radio Frequency Identification System

Radio frequency identification (RFID) system uses radio waves to track product. It uses tags (data carriers), readers (receivers) and computer systems (software, hardware, networking and database) (Brody *et al.*, 2008; Kumar *et al.*, 2009).

The tags are composed of an integrated circuit, a tag antenna and a battery for passive tag (Brody *et al.*, 2008; Kumar *et al.*, 2009). The integrated circuit contains a non-volatile memory microchip for data storage, an AC/DC converter, encode/decode modulators, a logic control and antenna connectors (Brody *et al.*, 2008; Kumar *et al.*, 2009). This system is very flexible because of the nature of data transfer between the tag and the reader thus, are ideal for food packaging. Data stored in tags is activated by the readers when it enters the electromagnetic zone of a reader. The data is transmitted to a reader for decoding and the decoded data is transferred to a computer system for processing (Brody *et al.*, 2008). However the use of RFID is limited because the tags and the infrastructure required for RFID systems is too expensive for use on individual primary packages (Brody *et al.*, 2008).

The use of RFID in the food industry is currently focused on tracking and identification of food products. Retail chains such as Wal-Mart and Home Depot have used this technology for distribution (Joseph and Morrison, 2006). In 2003, the top suppliers of Wal-Mart were required to use RFID tags. U.S. Department of Defense and other retailers such as Albertsons, Target, Tesco and Marks and Spencer are presently using RFID technology. This technology provides security and safety for food companies through tracking the origin of supplies. According to the FDA, 1307 recalls of processed foods occurred between 1999

and 2003 (Brody *et al.*, 2008). Application of RFID technology can avoid such recalls in future. All stages of the supply chain *i.e.* from farming to consumption are very critical to food recall problems (Stauffer, 2005).

The RFID system is also limited by the shielding effect of metal. The data on the tags placed on metal packages cannot be read correctly. As a result of the aforementioned limitation, high frequency RFID tags are used on metal packaging such as beer barrels. Another limitation of RFID system is that water molecules can absorb microwave signals, resulting in signal loss or interference during data acquisition from microwave RFID tags (Brody *et al.*, 2008). Many food products contain high water content limiting the use of RFID system. To overcome RFID system interference with water molecules, ice cream manufacturers place tags over an air gap in the container (Brody *et al.* 2008). Unilever (London, U.K.) uses RFID technology to handle and track products in the warehouse (Kumar *et al.*, 2009). United biscuits (Hayes, U.K.) uses RFID technology to control the movement of raw materials, in weighing, mixing and baking processes involved in the preparation of biscuits and cakes (Angeles, 2005; Kumar *et al.*, 2009). Cheese manufacturers use RFID to trace cheese along the supply chain with great precision (Regattieri *et al.*, 2007; Kumar *et al.*, 2009).

Challenges in the implementation of RFID Technology

The technical challenges in implementing RFID technology are that it is unreliable in a retail environment. Reading of ultra-high frequency tags near a human body is difficult because of interference from the water content of human body (Kumar *et al.*, 2009; Roberts, 2006). Tags on products with large amount of liquid or metals cannot be read easily because liquids absorb signals while metals reflect the signals (Kumar *et al.*, 2009). The reading range for low frequency RFID systems is about 1 m while the ultra-high frequency RFID systems operating range is from 3 up to 4 m. Developing RFID readers with wide operating ranges will result in wide application of the technology (Kumar *et al.*, 2009; Want, 2004). The reader cannot communicate accurately with a tag, which is oriented perpendicular to the antenna

of the reader. And because of this, some of them are invisible to the reader (Kumar *et al.*, 2009). Most RFID readers do not operate properly in the presence of another reader. There are differences in frequencies allocated for RFID applications because it is managed by the regulations of different countries. This implies that a tag operating at a certain frequency in one country may not be readable in another country, which uses the same frequency for a different purpose (Kumar *et al.*, 2009). Cost is another major limitation of RFID technology. The tags are expensive making it would be uneconomical to incorporate tags into every retail item. (Kumar *et al.*, 2009; Roberts, 2006). Recycling issues is also a factor with RFID. Adhesives, computer chips, copper from the antenna, and conductive inks of an RFID tag can contaminate polyethylene terephthalate and high density polyethylene during the recycling process (Kumar *et al.*, 2009; Foley, 2006). Contamination can be minimized by removing the recycling tag (Arnold, 2005; Kumar *et al.*, 2009). Consumer acceptance also limits the application of this technology because it has the ability to track their movement and buying habits. An unauthorized user can scan tags and gain access to private information such as details of shipments and inventory (Weinstein, 2005; Kumar *et al.*, 2009).

Nanobiosensor

A biosensor is an analytical system that is very sensitive and it is composed of biological based materials (Reddy and Ratna, 2013). Some of the biological based materials used in the construction of biosensors are DNA, enzymes, antibodies, tissues, microorganisms and receptors (Reddy and Ratna, 2013). The system uses a sensing device that converts the biological response into an electrical signal that is amplified, and quantified by a processor (Reddy and Ratna, 2013). There are different types of biosensors namely: Immunosensors, acoustic, electrochemical, potentiometric, optical, enzyme based, electrode based, nucleic acid-based, whole cell-based, tissue/whole organism-based, amperometric and calorimetric biosensors (Otlés and Yalcin, 2012). Monitoring the quality of food products throughout the processing is a major concern in food industry. Consumers select foodstuff based

on several factors such as: color, aroma, sensory perception, texture and shape *etc.* (Otles and Yalcin, 2012). These factors are influenced by fat and moisture content, protein, carbohydrate levels *etc.* (Otles and Yalcin, 2012). An accurate technique for the assessment of food product quality is a pressing need. Biosensors are potentially user-friendly, portable, fast, cheap and accurate system for monitoring quality of food product (Rana *et al.*, 2010). It can be integrated into food packaging and it is able to detect pathogens, certain chemical compounds and toxins in food thereby, provide the real time status of food product freshness (Liao, Chen and Subramanian, 2005; Azeredo, 2009). Nanosensors in food packages can be divided into different types based on their function. Some of these functions are detection of atmospheric impacts, microorganisms and chemical detection inside the package (Iles *et al.*, 2011). There are reports on commercialized nanosensors which are used to detect pathogens, product tampering, food spoilage, chemical contaminants, monitor products through the processing chain (Nachay, 2007).

Carbon nanotube based sensors

CNTs based sensors are rapid with high-throughput detection, cost effective, consume less energy, recyclable and the use of labels becomes unnecessary (Azeredo, 2009). Some researchers reported gas sensors that can be integrated into food packaging that are affordable to gauge freshness of food products. They were prepared by spraying carbon nanotubes onto flexible plastic sheets. These sensors exhibited rapid detection and response to changes in the concentrations of gases: ammonia, carbon dioxide and nitrogen oxide. They consume less energy and operate at room temperature (Abdelhalim *et al.*, 2013; Abdellah *et al.*, 2013a; Abdellah *et al.*, 2013b). In a report by Nguyen *et al.*, 2013, fabricated CNT-based structures were able to detect CO₂. The CNT-based gas sensors were found to be a potential gas sensor for affordable determination of food product freshness and quality (Nguyen *et al.*, 2013). In another research report, sensors were prepared from carbon nanotubes functionalized with selected cobalt porphyrin complexes (Liu *et al.*, 2015). The sensors had a detection limit below

0.5 ppm for ammonia and were able to differentiate ammonia from other chemical vapors containing various functional groups. The sensors were tested on putrescine and cadaverine simulating meat deterioration. Total volatile basic nitrogen was also measured using the sensors on samples of cod, salmon, chicken and pork. The results coincided with the expected deterioration for these foods (Liu *et al.*, 2015).

Enzyme based biosensors

Enzyme based biosensors are analytical systems that are composed of a biological sensing element closely connected to a transducer system (Velasco-García and Mottram, 2003; Cock *et al.*, 2009). The biological sensors include enzymes, cellular organelles, complete cells animal or vegetal tissue and are used to detect any of the substrates that participate in the reaction by the disappearance of a known substrate distinct from the substrate that is being sought for or by the appearance of a known product (Davis *et al.*, 1995; Mello and Kubota, 2002). The biological sensors can be reused (Gajovic *et al.*, 2000; Cock *et al.*, 2009). Enzymes are employed as biological recognition elements because of their rapid response, high selectivity, ability to regenerate, commercial availability isolation and purification (Luong *et al.*, 2008; Cock *et al.*, 2009; Hall, 2002). Some enzymes used in biosensors are oxidoreductase, glucose oxidase, horseradish peroxidase and alkaline phosphatase (Rogers and Mascini, 1998; Cock *et al.*, 2009). They are stable in catalyzing reactions of oxide reduction (Mello and Kubota, 2002). However, they exhibit some limitations, such as: their detection limits are either satisfactory or excessive; they are unstable over a long period of time and usually require immobilizing them; their purification is difficult and expensive (Tothill, 2001; D'Orazio, 2003, Mello and Kubota, 2002). In enzymatic biosensors the enzymes are immobilized (Davis *et al.*, 1995). There are several reports on the application of enzyme based biosensors. Lange and Wiltman *et al.*, 2002, reported the application of enzyme sensor to analyze meat, fish, beer, sauerkraut, dairy products, wine and fermented foods. The data obtained was compared with the conventional Liquid Chromatography and a mean correlation

coefficient of 0.854 was found (Lange and Wiertman *et al.*, 2002). Baeumner *et al.*, reported the detection of food product freshness based on L-lactate detection in tomato paste and infant food (Baeumner *et al.*, 2003). Baeumner *et al.* also reported graphite-Teflon-tyrosinase composite biosensor developed for the quantification of benzoic acid in soda drinks and mayonnaise (Baeumner *et al.*, 2003). They are used for the analysis of aspartame with carboxyl esterase, alcohol oxidase, carboxypeptidase, L-aspartase, peptidase, aspartate aminotransferase, glutamate oxidase and α -chymotrypsin (Odaci *et al.*, 2004). They are also used for the analysis of sorbitol with sorbitol dehydrogenase and nicotinamide adenine dinucleotide (NAD⁺) (Saidman *et al.*, 2000); analysis of benzoic acid with tyrosine (Morales *et al.*, 2002); analysis of sulphites with sulphite oxidase; to detect sorbitol which also interact with artificial edulcorants such as xylitol; they are used for the determination of benzoic acid by the presence of other antioxidants such as butyl hydroxyanisole (BHA) and propyl gallate (Patel, 2002). Smyth *et al.*, (1999), determined ethanol accumulation in lettuce, cauliflower, broccoli and cabbage processed and packed in a modified atmosphere. The response from the biosensor was very similar to the results observed from gas chromatography. There are also reports on the application of enzyme based sensors for the determination of ethanol formation during apple storage in a controlled atmosphere, the development of putrefaction in tubercles like potatoes which is associated with quality loss. It has been used to determine the content of some organic acids and sugars as indicators of fruit and vegetable maturity (Ángeles and Cañizares, 2004). They are used in wine industry to detect disagreeable flavors and aromas which are responsible for huge loss in the industry (Moore *et al.*, 2003). They can detect freshness of fish (Volpe and Mascini, 1996). They can be used to distinguish milk that has been submitted to ultra-high temperature treatment and milk sterilized in the container (Cock *et al.*, 2009).

Nucleic acid based biosensors

In a nucleic acid based biosensor, DNA or RNA target is detected through the hybridization reaction

between DNA or RNA sensing elements. DNA and RNA possess the ability to fold into distinct three-dimensional structures and can form receptors and bind targeted molecules with high specificity and affinity. It has been reported that RNA and DNA aptamers can interact with a diverse assortment such as ions, small molecules, peptides, single proteins, organelles, viruses and even whole cells. (Wilson and Szostak 1999; Mok and Li 2008). DNA-based biosensors have been used to detect *E. coli* in public beach water (Sun *et al.*, 2006). *Salmonella* spp. was also detected using DNA streptavidin modified magnetic beads and electrochemical detection (Lermo *et al.*, 2007). *L. monocytogenes* was detected using a magneto electrochemical luminescence PCR detection platform (Pöhlmann *et al.*, 2009). *E. coli*, *Bacillus subtilis*, *B. atrophaeus* and *L. innocua* in meat juices have been detected using nucleic acid biosensor (Pöhlmann *et al.*, 2009). Label free detection was performed using synthesized target DNA and real DNA samples from *S. choleraesuis* in dairy food (Berdat *et al.*, 2008). Applications in food safety control have been reviewed (Amaya-Gonzalez *et al.*, 2013). *E. coli* was detected in milk and apple juice with detection level of 6 CFU/mL and 25 CFU/mL, respectively (Zelada-Guillen *et al.*, 2010). *Vibrio cholera* was detected at 0.85 ng/ μ L genomic DNA (Adley, 2014).

Nanowheels

Some researchers developed molecules shaped like a tiny wagon wheel with a diameter of 7 nm (Wiley & Sons, 2007). They were prepared using two-dimensional particles such as inorganic alumina platelets and used as fillers for plastics. They impart excellent mechanical properties to these materials. Nanocomposites made of alumina platelets and polymers have been reported to be extraordinarily rigid, strong, and thermally stable. The addition of nanoscopic platelets to plastics can improve the liquid and gas barrier properties of plastics. The use of nano platelets would also make plastic-based food packaging affordable and environmentally friendly (Wiley & Sons 2007).

BENEFITS OF NANOTECHNOLOGY IN FOOD INDUSTRY

Application of nanotechnology in food industry offers several benefits such as (Chaudhry

et al., 2008, 2010; Chaudhry and Castle, 2011): efficient food production requiring the use of less

agrochemicals, resulting in reduced environmental hazard and introduction of harmful chemicals into food; hygienic food processing techniques, resulting in better food with excellent safety, quality and free from food borne illnesses; improved tastes, flavors and mouth feels of food products; extended shelf life of food products, thereby reducing food wastage and a more dependable food supply; innovative lightweight, stronger, functional packaging contributing to reduction in cost of transportation and packaging materials; it ensures food authenticity, safety, and traceability by using smart labels. The effects of

nanopackaging on food products is dependent on the active ingredient of the nanomaterial, the polymer matrix onto which the nanomaterial is incorporated. Some nanomaterials have reported to induce cell death in eukaryotic cells (Long *et al.*, 2006; Nel, Xia, Madler and Li, 2006) and exhibit cytotoxic effects by inhibiting growth in prokaryotic cells (Brayner *et al.*, 2006; Thill *et al.*, 2006). These findings are very useful in food industry for control of food spoilage microorganisms and pathogens.

CHALLENGES OF NANOTECHNOLOGY IN FOOD INDUSTRY

There are many benefits of these technologies, however, there are concerns over the potential negative effects. The decrease in particle dimension results in increase of surface area which in turn increases solubility, absorption and biokinetics. Chen and Mikecz (2005), reported an *in vitro* study on human epithelial cell cultures using fluorescence-labeled SiO₂ nanoparticles. Particles that had sizes smaller than 70 nm entered cell nuclei. The use of nanomaterials in the food industry can result in these nanomaterials having access to tissues in the human body and thereby resulting in accumulation of toxic contaminants (Cushen *et al.*, 2012; Chau *et al.*, 2007). Nanoparticles possess the possibility to form compounds with selected food material, or remain in a free state while in the alimentary canal. However, the effect of this on absorption is unknown (Cushen, 2012). There are knowledge gaps in the understanding of the properties and effects of nanomaterial used in food applications which makes it difficult to assess the risk it poses to consumers. Some of the knowledge gaps that need more research include (Chaudhry and Castle, 2011): a clear definition of nanomaterials and nanotechnologies; valid techniques for detection and characterization of nanomaterials in food products; toxicology studies of nanomaterials used in food products to confirm its safety; studies on the adsorption, distribution, metabolism and elimination profiles of nanomaterial; long term health consequences of ingestion of nanoparticles

from food products; studies on the potential risks of functionalized nanobiomaterials; establishment of international research collaborations and networks that can address different aspects of the existing and new nanotechnology food sectors; development of clear and consistent guidelines for risk assessment of nano-food products; establishment of a global body to ensure quality and that products are not just labeled for commercial gain; promotion of industry best practices and self-regulation in the use of nanotechnologies and a harmonized regulatory system at the global level for evaluation of nano-food products and labeling of nano-food products to inform the consumer. According to Chaudhry *et al.*, (2008), Chaudhry and Castle (2011), the major concerns in the application of nanomaterials are: food products containing indigestible, insoluble and biopersistent nano-additives such as metals or metal oxides, functionalized nanomaterials; adsorption, distribution, metabolism, elimination profiles and toxicological properties of nanoparticles are not yet fully known at present; there is a risk of migration of nanopesticides and nano-sized veterinary medicines into the food products during processing and incorporation of nanomaterials to food packaging poses a risk to consumers because nanomaterial may migrate into food products. Modeling studies reported that nanomaterials can migrate from the package while others reported no migration of the nanomaterials. (Chaudhry and Castle, 2011; Avella *et al.*, 2005;

Bradley, Castle and Chaudhry, 2010). Simon, Chaudhry and Bakoš (2008), modelling reports suggested that detectable migration of nanomaterials from packaging to food will only take place where very small nanoparticles in the lower nm range have been incorporated into a polymer matrix that has a low dynamic viscosity. In such polymer matrix, the particles are not bound to the matrix. However, more research is needed to determine the migration patterns in other polymer-nanomaterial composites.

Nanoadditives in food can undergo various transformations in the food and GI system as a result of several reactions such as: reaction with stomach acid and enzymes, agglomeration, aggregation and binding with food components (Chaudhry and Castle, 2011). These transformations can result in nanomaterials losing their nano attributes (Chaudhry and Castle, 2011). There is very little report on the effects of these transformations on nano-based food safety (Chaudhry and Castle, 2011). Other areas of concern are food products containing natural processed food nanostructures that are not biopersistent but are digested in the gastrointestinal tract (Chaudhry and Castle, 2011); food products containing encapsulated food additives in nano-sized carriers which are biopersistent but carry the encapsulated substances across the gastrointestinal tract. This can result in increased bioavailability of food additives: vitamins and minerals which may not be beneficial for the consumers health. This can also result in a high uptake of food colors or preservatives by the body above the acceptable daily intake (Chaudhry and Castle, 2011).

Regulation of the application of nanotechnology in food industry

There is a need for regulatory bodies around the world to establish rules and guidance on the use of nanomaterials in food products. Validated techniques to detect and characterize nanomaterials in complex food matrices are currently not available (Chaudhry and Castle, 2011). Some organizations are already involved in nanotechnology research, regulations and guidelines. The Food and Drug Administration (FDA) has provided its perspective on nanotechnology on its Web site

(<http://www.fda.gov/nanotechnology/>). FDA regulates products and not technologies. It has limited regulatory authority over certain categories of products and may have limited authority over the use of nanotechnology related to some food products (Wesley *et al.*, 2014).

Regulation of Nanomaterials in the United States

In 2006, FDA initiated the Nanotechnology Task Force to address approaches for FDA-regulated products that contain nanomaterials. The Task Force recommendations were focused on engineered nano-scale materials (ENMs) and not on natural or incidental nanomaterials (Clark and Baughan, 2012). Their reports discouraged the adoption of a precise definition of nano-scale materials and recommended a case-by-case assessment of ENMs. FDA followed the recommendation and did not define ENMs in the draft guidance that the Agency published in June 2011. The guidance indicates FDA's intention to make a distinction between products that have been deliberately manipulated to control particle size for purposes of achieving desired properties, and those products that contain incidental or background levels of nanomaterials or products that contain materials having a size that naturally falls within the nano-scale range (Clark and Baughan, 2012).

In June 2011, FDA agency published a draft guidance and two important aspects that are to be considered when evaluating new products, these are (Clark and Baughan, 2012): whether an engineered material or end product has at least one dimension in the nano-scale range (approximately 1 nm to 100 nm); or whether an engineered material or end product exhibits properties or phenomena, including physical or chemical properties or biological effects, that are attributable to its dimensions, even if these dimensions fall outside the nano-scale range, up to one micrometer.

Regulation of Nanomaterials in the European Union

In February 2007, European Food Safety Authority Regulatory agency formed a panel to conduct a risk assessment of nanoparticles in food and food packaging. Denmark's National Food Institute is working on a project to gather toxicology information on nanoparticles and the UK Food Safety Authority has put together a

report on potential areas for future regulation regarding the use of nanotechnology in foods (Nanotechnology in food industry, internet). In some countries such as Australia and New Zealand, new food products that are manufactured using nanotechnology are made to undergo comprehensive scientific safety assessment before they are supplied. Food Standards Australia and New Zealand (FSANZ) have also adopted strategies that continually review the potential risks associated with nanotechnologies in foods. These strategies include (Nanotechnology and Food, 2014): amending/updating the FSANZ application handbook to support new food regulations and ensure applicants provide all the necessary information to help FSANZ conduct a risk assessment; advising the food industry about the amendments to the application handbook involving nanotechnology and asking industry for information about proposed nanotechnology applications; engaging with other national and international regulatory agencies, industry and the public to outline FSANZ's regulatory responses.

In EU, a comprehensive science-based definition of nanomaterial is currently developed. The definition is applicable to engineered nano-scale materials and substances that naturally contain one or more external dimensions that are

less than 100 nm in size. Several legislation and technical guidance have been adopted on the use of nanomaterials.

Regulation of nanomaterials in Canada

Canada does not have any explicit legislation on the use of nanomaterials in food packaging or food products. However, the potential risks and benefits of nanomaterials in food packaging are evaluated within existing legislative and regulatory frameworks (Clark and Baughan, 2012). Health Canada published a policy statement on the October 6, 2011. The policy statement defines a nanomaterial as any manufactured substance or product and any component material, ingredient, device, or structure that is (Clark and Baughan, 2012):

- At or within the nano-scale (1 to 100 nm) in at least one external dimension, or has internal or surface structure at the nano-scale, or;
- Smaller or larger than the nano-scale in all dimensions and exhibits one or more nano-scale properties that are attributable to size and their effects, and are distinguishable from the chemical or physical properties of individual atoms and molecules, and bulk material.

CONCLUSIONS

Nanotechnology offers many potential benefits to the food industry which include food production, food processing and food packaging.

Some of the potentials includes nano-based packaging materials that have excellent barrier to oxygen and water vapor, addition of nanoparticles to food packaging that are potent antimicrobial agents that can kill food borne pathogens, nanosensors which are able to detect gases, microbes or chemical contaminants in complex food matrices and nanoencapsulation that help to fortify food products with essential nutrients.

Some of the other benefits of nanotechnology in food industry are improved tastes and textures of food products, reduction in the use of fat, salt and food additives, enhanced absorption of nutrients and supplements,

preservation of quality and freshness, better traceability of food products and extension of shelf life.

It is envisaged that nanotechnology-based food products availability will increase to consumers worldwide in the next few years. It is estimated that several companies worldwide are conducting research and development into the use of nanotechnology in food packaging, delivering food and nutritional supplements.

However, despite the great benefits that nanotechnology offers there is still a pressing need to study the properties, behavior, effects, toxicological safety, long term impact on human health, adsorption, distribution, metabolism and elimination profiles of nanomaterials.

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Conflicts of Interest

The authors declare no conflict of interest.

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