2016-09-13

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Springer

https://doi.org/10.1007/s13197-016-2325-6

J Food Sci Technol (September 2016) 53(9):3395–3407
DOI 10.1007/s13197-016-2325-6

REVIEW

Transformation of food packaging from passive to innovative via nanotechnology: concepts and critiques

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Revised: 9 August 2016 / Accepted: 25 August 2016 / Published online: 13 September 2016

Abstract In recent decades, there is a global advancement in manufacturing industry due to increased applications of nanotechnology. Food industry also has been tremendously changing from passive packaging to innovative packaging, to cope with global trends, technological advancements, and consumer preferences. Active research is taking place in food industry and other scientific fields to develop innovative packages including smart, intelligent and active food packaging for more effective and efficient packaging materials with balanced environmental issues. However, in food industry the features behind smart packaging are narrowly defined to be distinguished from intelligent packaging as in other scientific fields, where smart materials are under critical investigations. This review presents some scientific concepts and features pertaining innovative food packaging. The review opens new research window in innovative food packaging to cover the existing disparities for further precise research and development of food packaging industry.

Keywords Active packaging · Food Nanotechnology · Innovative packaging · Intelligent packaging · Nanomaterials · Smart packaging

Introduction

In recent decades, there is a global advancement in manufacturing industry due to increased application of nanotechnology. Food industry also has tremendous availed this technological advancement by reverting its food processing, packaging and storage methods. Passive food packaging is gradually changing to innovative packaging systems; namely smart packaging, intelligent packaging and active packaging (Ampatzidis et al. 2008; Meng et al. 2015; Puligundla et al. 2012; Ranjan et al. 2014). Innovative technology is underway and active research is taking place to produce more effective and efficient food packaging and processing techniques for improved food quality and safety (Brizio and Prentice 2015; Mahieu et al. 2015). In addition, the food industry is pioneering the applications of hybridized packaging materials such as nanocomposites with more desired packaging features through nanotechnology. Nanocomposites exhibit improved mechanical, electronic, optical, electrical and barrier properties and can be integrated with communication devices including radio frequency identification (RFID) devices (Feng et al. 2015), nanosensors, biosensors (Ranjan et al. 2014), barcodes and electronic noses (Sarapulova et al. 2015). Innovative packaging focuses on one or more of the following pillars: improving the quality, safety and shelf-life extension of foods (Realini and Marcos 2014), production of less expensive packaging materials (Wu et al. 2015a), reduced processing labor, produce convenience food products, reduced use of preservatives in food formulations and monitoring food quality in supply chain (Bastarrachea et al. 2011; De Jong et al. 2005). Then, innovative food packaging systems would interact with human demand as food producers and handlers, food processors and packers, distributors, business-persons, consumers to policy-makers, in

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order to address the issues pertaining food processing, postharvest losses and food security (Koskela et al. 2015; Rukchon et al. 2014).

Nanopackaging materials integrated with communication and detective devices including RFID, barcode tags and nanosensors as for food bonafide (Neethirajan and Jayas 2011; Wanihsuksombat et al. 2010) is expected to create new era of guaranteed food safety, quality and new functionalities of food materials (Chellaram et al. 2014; Dobrucka and Cierpiszewski 2014; Puligundla et al. 2012). In smart packaging, the self-healing and self-cleaning are predominant models. Self-healing is attributed by dynamic swelling and electrostatic repairing of the polyelectrolyte multilayers in the vicinity of the fracture, toughness, stresses, corrosion, tears, barrier properties and even molecular weight fractures (Sharon 2012; Wu et al. 2008; Zhu et al. 2015). For self-cleaning materials, superhydrophobicity and reduced contact angle as a result of minimized surface tension and energy, which creates the free contaminant surface of substrate (Liu et al. 2015c; Martines et al. 2005; Ye et al. 2015). Active packaging such as antimicrobial packaging encapsulates the preservatives in polymer matrix which leads to reduced amount of preservatives and their controlled released in programmed fashion onto the packed foods (Acosta 2009; Mahieu et al. 2015). This allows effective food preservation and prolonged shelf life of foods by retarding microbial growth and extending their lag phase leading to improved safety and quality of food products (Biji et al. 2015; De Jong et al. 2005). Therefore, the practice is significant for preventing the post-processing contamination and spoilage of foods (Bastarrachea et al. 2011; Imran et al. 2010; Koskela et al. 2015; Wen et al. 2016).

Currently, nanotechnology has garnered the utilization of bioplastic polymers over conventional petrochemical plastics due to their promising packaging properties such as biodegradability, biocompatibility and sustainability (Mahieu et al. 2015). Despite the technical limitations of bioplastic polymers for packaging including low processability, excessive brittleness, high cost, low heat deflection temperatures, poor barrier to oxygen and moisture and hydrophilicity, nanotechnological strategies have been deployed to improve their performances (Tang et al. 2011). The strategies such as polymer blending, nanocomposites, cross-linking and other modifications techniques have used to tailor their optical, mechanical and chemical properties for packaging. In this review, the science, concepts and features behind innovative food packaging with respect to advancement in nanotechnology are addressed. However, the concepts and features of smart packaging have been implicitly postulated for the first time in food industry. These features engineer new research opportunity in food packaging, packaging forming engineering and addresses the existing knowledge disparities in food production industry.

Nanotechnology and food industry

Nanotechnology is defined as the application of scientific knowledge to measure, create, pattern, manipulate, utilize or incorporate materials and components at the nanoscale (de Azeredo 2009). Nanotechnology focuses on designing, synthesizing, characterizing and applying of materials, patterned devices and the systems with functionalized organizations in at least one dimension on the nanoscales (Duncan 2011). The nanomaterials have unique physical and chemical properties when compared to that of macroscale materials of the same chemical compositions (Duncan 2011). Nanotechnology is described by scientific community as the revolutionary super-cutting-edge field (Chellaram et al. 2014; Imran et al. 2010; Rashidi and Khosravi-Darani 2011) that intends to revolutionize future manufacturing industry including chemical processing, mechanical industry, electronic technology, sensors technology, and from pharmaceutical industry to agricultural and food supply chain (Bumbudsanpharoke and Ko 2015; Dasgupta et al. 2015; Neethirajan and Jayas 2011; Sarapulova et al. 2015).

To date, nanotechnology is the cornerstone of research in innovative packaging technology; smart packaging, intelligent packaging and active packaging (Dasgupta et al. 2015). Taking the advantages of nanotechnology, there is active research in food storage, food processing and packaging, food safety and quality monitoring systems (Ranjan et al. 2014), tracking, tracing, brand protection and actives/nutrients delivery systems (Fig. 1) (Bumbudsanpharoke and Ko 2015; Cushen et al. 2012; Feng et al. 2015; Rashidi and Khosravi-Darani 2011). Despite this research,

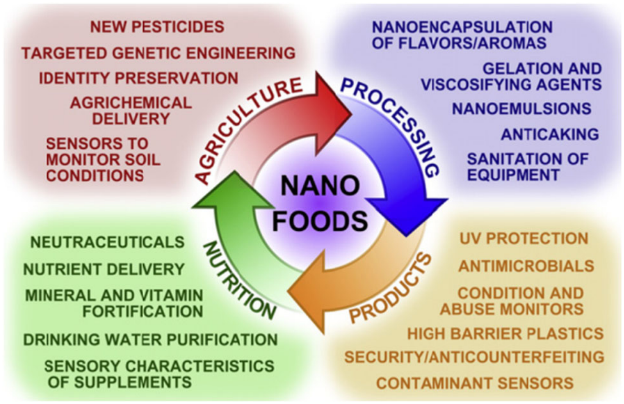

Fig. 1 Prospective uses of nanotechnology in food production and agricultural sector. The nano-based food products are discussed in this article (Duncan 2011)

some nanotechnology related products are in market globally. According to Vance et al. (2015), nanotechnology related products are more than 1827 in the market and are produced by 622 companies from 32 countries (Huang et al. 2015). Food and beverage nanoproducts in the market account for about 6.5 % of total products. Many of these products are nanoclay and silver-based related products predominantly for manufacturing food packaging and food containers from South Korea, China, USA and Japan (Bumbudsanpharoke and Ko 2015; Cushen et al. 2012). The current market trends of nano-food products is about US$ 1 billion (Acosta 2009; Neethirajan and Jayas 2011), with growth potential over US$ 20 billion in the next decade at annual growth rate of 11.6 % (Blasco and Picó 2011). Most of these products are related to coating technology of packages, and nanoencapsulation of nutritious and health-promoting related products and beverages (Ziani et al. 2012).

The higher projection of food nanotechnology investment (Blasco and Picó 2011; Bumbudsanpharoke and Ko 2015; Cushen et al. 2012) is assisted with revolution in the field of information technology, photonics and nanophotonics, electronics, computing, data storage, advance materials sciences, understanding of polymers, and advanced characterization of materials at precise nanoscales and shapes (Ampatzidis et al. 2008; Cushen et al. 2012; Sarapulova et al. 2015). Advanced analytical and imaging technology including laser and scanning forces, digital optics, transmission and scanning electron microscopes (TEM, SEM) have influenced modern understanding and probing of materials based on their particle size, morphology, geometric structures and interactions of materials with their environments (Rashidi and Khosravi-Darani 2011). To date, the research is paving for comprehensive understanding of polymer nanomaterials for enhanced smart packaging, intelligent packaging and active packaging phenomena in more complex forms. The knowledge enables for integrated packages with additives such as antimicrobial agents, nutrients, probiotics (Ziani et al. 2012), moisture and oxygen barriers, damage repairing (Imran et al. 2010; Neethirajan and Jayas 2011), self-cleaning agents (Youngblood and Sottos 2008) and information provisional devices (Bastarrachea et al. 2011; de Azeredo 2009).

Polymeric nanocomposites and packaging

Synthetic and natural polymers are widely used in food industry and other life science fields such as biomedical, pharmaceutical and agriculture sector due to their particular desirable properties. However, the polymers used in food packaging do not singly meet the packaging performance requirements. Therefore, many techniques include blending, nanocomposites, chemical modification, plasticization and compatibilization have been used to achieve intermediate or even superior properties from the hybrid materials for improved performance (Tang et al. 2011). Improving performance targets on mechanical properties and impact resistance, increasing barrier properties, biodegradation rate, cost reduction and thermostability of packaging materials. The prominent strategies used to produce hybrid materials for food packaging include blending of polymers and polymer nanocomposites. Nanocomposites are composites or hybrid materials that comprise a dispersion of nanometer size particles in a polymer matrix (Tang et al. 2011). Most used nanomaterials for this purpose are carbon nanotubes (CNTs), clay platelets (montmorollites/nanoclays) and bionanocomposites (de Azeredo 2009; Yildirim et al. 2015).

Notable improvements for polymer nanocomposites include biodegradation rate, barrier properties, mechanical strength, heat resistance, flexibility, compatibility and even flame retardation. The final properties depend on nanocomposites dispersion and their interactions at interface (Ghanbarzadeh et al. 2015; Rhim et al. 2013). The nanocomposites tend to immobilize into a layered inorganic matrix that organizes the species into two-dimensional arrays resulting in novel functions of the individual polymer matrix. These have been observed to affect some quality parameters like tensile strength, water vapor permeability (WVP) and moisture of the hybrid composites. Even low content of nanoparticles influences the homogenous dispersions of nanoclays within the polymeric nanocomposites leading to relatively compacted and smooth surface of polymeric films and with improved thermal stability. However, when nanoclays contents reach their saturation point, negative effects such as poor surface and optical transparency due to particle aggregations, have been reported (Li et al. 2015).

Innovative food packaging systems

The application of nanotechnology for innovative food packaging has been through bioinspiration and copying from the nature, using polymers (Hager et al. 2015; Zhu et al. 2015). As well known, most of the important components of living cells are made up of polymers including carbohydrates, proteins and nucleic acids as well as fatty acids or lipids are involved in regulations and control the biological processes (Sharon 2012). Therefore, the interest in novel food packaging is to develop intelligent, active and smart packaging systems by mimicking the biological processes, which would safeguard the integrity of packages and foods in food chain systems. Despite this, countless of publications advocate synonymously three terms in innovative packaging: smart systems to active packaging systems and intelligent systems to smart packaging systems and vice versa (Biji et al. 2015; Chellaram et al. 2014; Dobrucka and Cierpiszewski 2014; Puligundla et al. 2012; Ranjan et al. 2014). In other scientific fields, the term “smart materials or polymer” is well defined based on its fundamental sciences and underlying features (Fig. 2). In general, the features for smart packaging are a leading conundrum in classifications and fewer research efforts is focused in food production industry (Wu et al. 2016; Zhu et al. 2015).

Intelligent food packaging

Intelligent packaging system is a system which monitors the conditions and quality of food products in supply chain, especially during distribution and storage and provides the status of the food to the end-user or consumer (Biji et al. 2015). Intelligent packaging system could be applied for detecting, sensing and reacting to the dynamically changing signals of the products environment in the range of parts per trillion (Neethirajan and Jayas 2011; Realini and Marcos 2014; Sharon 2012). Figure 2 shows that intelligent systems can be categorized as (i) data carries including barcode, RFID (Feng et al. 2015), electronic article surveillance (EAS), and digital watermark, (ii) quality indicators such as freshness indicator (Shukla et al. 2015) and time–temperature indicator (Pereira et al. 2015; Wanihsuksombat et al. 2010), (iii) sensors (Biji et al. 2015) and (iv) others are organic light emitting diodes (OLED) and hologram (Dasgupta et al. 2015; Sarapulova et al. 2015). In food industry, most reported systems are in order of (ii), (iii) and (i). These systems can be developed with the perspectives of managing food safety for compromised products, to smoothen judgments and decision-making processes, supplement to food safety and quality management systems, endowed with biosecurity information and alerts to end-users on any possible problem (Wanihsuksombat et al. 2010). The revolution in food industry and especially the capability in traceability and tracking of food products are anticipated to safeguard the public health and food safety globally (Koskela et al. 2015). This can be done in connection to changes in temperature (Wanihsuksombat et al. 2010), relative humidity (RH), pH and volatile compounds (Qin et al. 2012; Rukchon et al. 2014) to qualify the product authenticity in food supply system (Neethirajan and Jayas 2011).

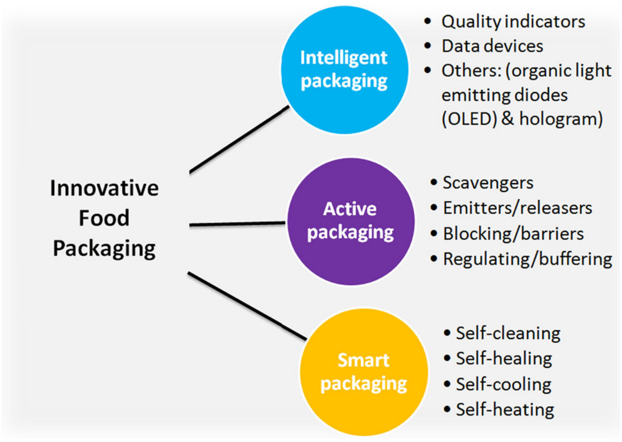

Fig. 2 Classification and features of innovative food packaging. Smart packaging phenomena is based on the background features of the polymer, intelligent packaging systems provides information to use on the packed food integrity and active packaging systems surveillance the packed food integrity

Integration of nano-based communication devices such as RFID tags and barcode with wireless sensors in packaging materials could be the transformative force of packaging technology and food supply chain (Biji et al. 2015; Koskela et al. 2015; Sarapulova et al. 2015). These devices can be used to provide assurances tamper proof (Realini and Marcos 2014), product authenticity, detecting, sensing, anti-theft, anti-counterfeiting (Dobrucka and Cierpiszewski 2014) and product traceability (Dasgupta et al. 2015; Feng et al. 2015; Neethirajan and Jayas 2011; Puligundla et al. 2012). Feng et al. (2015) assembled a chipless and wireless RFID sensor tag comprising two planar inductor-capacitor resonators to monitor the RH and identify the short-range items via inductive-coupling for paper packaging (Koskela et al. 2015). The constructed RFID tangs showed excellent sensitivity to RH at the ranges of 20 and 70 %. Feng et al. proposed for integrating the developed system to cheaper RFID sensors on conventional packaging materials as to make them intelligent.

Luminescent films of ZnO nanoparticles embedded on polyvinylpyrrolidone (PVP) exhibited significant sensing capacity for the status of various food substrates by varying their intensity of luminescence (Sarapulova et al. 2015). The materials were potential for developing nanophotonic devices including screen materials, flexography coatings, pad and inkjet printing materials that would ensure the functionality of novel packaging. RFID tags comprising RFID reader and Differential Global Positioning System (DGPS) were developed for short-range mapping of the hand-harvested fruits in the orchard field (Ampatzidis et al. 2008). The passive RFID tags mounted to the trees and fruit harvesting bins and the readers with DGPS and without were attached on the orchard tractor. The accuracy and reliability of GPS signals of the developed system were interfered with presence of tree canopies in the field, reading a missing ratio of 0.32 % for the detection of the bins. However, there was 100 % detection when the RFID tags were attached to suitable tree branches. In the second method, the absence of GPS hindered its successfulness in trials.

The food quality or freshness indicators play a crucial important in food industry. Food indicators are chemical substances that can indicate the presence, absence and/or concentrations of other chemical substances in a particular composition. They also refer to chemical substances which indicate the degree of interactions of two or more substances by means of a particular standardized characteristics such as changes in color of the substrates (Biji et al. 2015). Freshness indicator provides the information of the food product quality from characteristic changes from chemical effects or the growth of spoilage microorganisms in packed food products. The metabolites evolved from microbial spoiled food products can react with the indicators integrated onto packaging materials, hence provides visual indications about the quality of the food products. Some common indictors used in food industry are gas sensing dyes (Lee et al. 2015; Puligundla et al. 2012), and indicators based on time–temperature (Pereira et al. 2015; Wanihsuksombat et al. 2010), microbial spoilage, microbial growth, physical shock, pH, and food freshness (Rukchon et al. 2014; Sarapulova et al. 2015). Many volatile chemical metabolites produced from microbial activities including diacetyl, amines, carbon dioxide (Meng et al. 2015; Nopwinyuwong et al. 2010), ammonia gas and hydrogen sulfide gas evolved during the action of food aging, especially in meat and meat products can be easily monitored by freshness intelligent devices (Koskela et al. 2015; Shukla et al. 2015).

Real-time colorimetric sensor was integrated with bromophenol blue indicators for monitoring the quality of refrigerated buffalo meat at 4 ± 1 °C with response of total volatile basic nitrogen released to the package headspace (Shukla et al. 2015). The released gas indicated excellent correlation with microbial load of the refrigerated meat. The deteriorating quality of the refrigerated meat products changed the color of indicators from yellow to blue (Lee et al. 2015; Shukla et al. 2015). The chemical barcodes of cellulose integrated with pH-dye indicators were developed to monitor the quality of the skinless chicken breast with respect to microbial spoilage (Meng et al. 2015; Rukchon et al. 2014). The indicators, one made of methyl-red and bromothymol blue and the other with phenol red and bromocresol green exhibited better correlation between them in microbial growth patterns and the level of carbon dioxide evolved from the packaging. The indicators for headspace concentrations of carbon dioxide in packed kimchi during storage was developed by incorporating Coomassie Brilliant Blue dye (BB) into chitosan biopolymer (Lee et al. 2015; Meng et al. 2015). The correlation analysis of carbon dioxide gas based on transparent of chitosan films showed potential visual indicator for monitoring the quality of kimchi (Nopwinyuwong et al. 2010).

Moreover, the chemical sensors integrated with microarrays for characterization and identification of various types of Chinese liquors sourced from different regions in China exhibited various reaction patterns based on the locality of a particular liquor (Nopwinyuwong et al. 2010; Qin et al. 2012). Additionally, the real-time time–temperature indicator (TTI) were synthesized from chitosan and polyvinyl alcohol (PVA) polymers doped with thocyanins extracts from *Brassica oleracea var. capitata* (Red Cabbage) (Pereira et al. 2015). The TTI was developed for indirectly testing the quality of packed foods through detection of pH changes when subjected to improper storage temperatures. The indicators showed color changes of the pasteurized milk at the time of their chemical deterioration as evidence of quality to consumers. This demonstrates how the development of freshness indicators are useful tools in food industry for providing authentication and to avoid ailing food products for consumptions (Brizio and Prentice 2015; Wanihsuksombat et al. 2010).

The sensor is another subject in intelligent packaging which is used for detecting, locating, recording, quantifying energy and transmitting the information pertaining the detected or measured physical or chemical property (Biji et al. 2015). Sensors have (bio)receptor for recognizing the target analyte and transducer for conveying the electronic signals into measurable responses. The biosensors can modulate the external signals like light, pH, temperature, mechanical force, electric field, metabolites or solvent composition by utilizing the hydrophilicity and hydrophobic states of materials (Meng and Jinlian 2010; Wan et al. 2015; Wu et al. 2015b). There are various types of sensors such as nanosensors, gas sensors, printed electronics, CNTs-based sensors, fluorescence and luminescence biosensors, quartz crystal microbalance biosensors, surface plasmon resonance biosensors, electrical biosensors, and field effect transistor biosensors, electronic nose and chemical sensors (Koskela et al. 2015; Sarapulova et al. 2015). Quantum dots and graphene were used to construct a chip-integrated triple-dimensional sensor with electrochemical, fluorescence and mass-sensitivity elements to detect oil samples (Liu et al. 2015a). The sensor was able to discriminate eight different oil samples in laboratory and exhibited high analytical accuracy by 92.5 % for unknown samples compared to analysis done by high performance liquid chromatography (HPLC). Koskela et al. (2015) developed sensor tags embedded on copper acetate for real-time monitoring of microbial spoilage of raw broiler meat based on the evolution of hydrogen sulfide (H_2S), a product of microbiological metabolism. The sensor was effective to monitor the H_2S contents associated with bacterial and

Enterobacteriaceae metabolites in the modified packaged broiler meat at 6 °C.

Active food packaging

Active packaging systems are systems where by the products, packaging materials and the environments are interacting for performing the dynamic roles to enable extended shelf life, quality and safety or organoleptic properties of food products (Biji et al. 2015; Dobrucka and Cierpiszewski 2014). Active food packaging systems can be categorized as (i) scavengers including carbon dioxide, oxygen and ethylene scavengers (Mahieu et al. 2015), (ii) blocking (such as ethylene blocking), (iii) releasing (including antimicrobial packaging, antioxidant, insect repellent and flavor releasing) and (iv) regulating/buffering (such as moisture regulating/buffering). Active packaging system may incorporate active substance that could release active ingredients into food matrix (De Jong et al. 2005) or prevent infringement of unwanted substances from the environment onto packaging (Mahajan et al. 2008) (Fig. 2). The prominent forms of active food packaging are based on (i) antimicrobial agents (Gemili et al. 2010; Wen et al. 2016), (ii) scavenging systems for oxygen using ferrous oxide, zeolite, palladium, cobalt, photosensitive dyes, glucose oxidase, activated carbon, ethanol oxidase, ascorbic acid, ascorbate salts or catechol (Joven et al. 2015; Mahieu et al. 2015) and (iii) moisture absorbers using sachets, pads, sheets or blankets and therapeutic probiotics food (Dobrucka and Cierpiszewski 2014; Mahajan et al. 2008; Yildirim et al. 2015).

Antimicrobial packaging systems are integrated with antimicrobial agent for the purpose of preventing microbial growth and intrusions in food products and extending its shelf life. It aims on safety assurance, quality maintenance and extending the shelf life of the food products by killing or preventing the growth and entrance of spoilage and pathogenic microorganisms (Alkan and Yemenicioğlu 2016). Broadly antimicrobial substances including enzymes, organic salts and their acids, macromolecules (chitosan), bacteriocins, natural extracts, essential oils (EOs), fungicides, metallic salts and nanomaterials, are common antimicrobial susbstances used in food packaging industry. The natural antimicrobial agents of plant and animal origin including bacteriocins, enzymes, phenolic extracts and EOs have been of interest due to their safety and regulatory issues (de Oliveira et al. 2013; Wen et al. 2016). Example, Alkan and Yemenicioğlu 2016 developed antimicrobial zein film of natural phenolic compounds including phenolic acids of gallic, vanillic acid and their extracts from oregano, clove, walnut shell and artichoke stem. In addition, the EOs of cinnamic acids, thymol, carvacrol, eugenol and citral were used. The developed active films significantly inhibited the growth of *Xanthomonas vesicatoria, Erwinia carotovora*, *E. amylovora* and *Pseudomonas syringae*. Moreover, the concentration range between 1 and 4 mg/cm^2 phenolic acids, 1 and 4 mg/cm^2 EOs and 4 and 8 mg/cm^2 clove extract inhibited the growth of most of the pathogens. Galic acid produced the most potent films against *E. amylovora* and *P. syringae*, vanillic acid against *E. carotovora*, and cinnamic acid, thymol or carvacrol against *X. vesicatoria*.

Antimicrobial nanofibrous films of PVA-b-cyclodextrin blended with cinnamon EOs/exhibited excellent inhibition to *Staphylococcus aureus* and *Escherichia coli* growth in a media (Mahieu et al. 2015; Wen et al. 2016). The in vivo test of active films showed extended shelf life of strawberry. Similarly, de Oliveira et al. observed effective prevention of *Salmonella enteritidis* in refrigerated ground beef at ≈4 °C incorporated with oregano and lemongrass EOs for 6 days in sterile plastic bags (de Oliveira et al. 2013). The microbial efficacy tests at various concentrations of EOs, 3.9, 7.8 and 15.6 µl/g, on the processed meat were effective against the microbial population at the refrigerated temperature. In addition, ascorbic and iron powders or copper chloride as a catalyst for extruded oxygen scavenging thermoplastic starch films exhibited reduction in oxygen transmission rate of films from 20.9 to 1 % for 15 days at 80 % RH (Mahieu et al. 2015). Similarly, the oxygen scavenging polyethylene terephthalate (PET) films, PET-aluminum oxide coatings, polylactic acid films and oriented polypropylene (o-PP) films were deposited with palladium layers through vacuum deposition on silicon oxide ($SiO\chi$) layer for modified atmosphere packaging (Yildirim et al. 2015). The coating of the films significantly reduced the oxygen transmission rate through the film layers up to 33-fold. Moreover, the films of low-density polyethylene (LDPE) blended with activated carbon and sodium erythorbate were able to absorb 80 % of oxygen concentration from the package headspace, equivalent to absorbing 3.57 mg of oxygen from the headspace for 11 storage days (Joven et al. 2015).

Smart food packaging

Smart packaging can be defined as the packaging system that undergoes automatic and autonomous micro/nanostructures modulations as the course of dynamic changes in its environments. Generally, smart materials have capacity to control their interfacial properties based on ionic channels, bioinspired surface motions, wettability, adsorption and adhesion (Zhu et al. 2015). To date, the fundamental science of smart materials investigates self-cleaning, self-healing also called shape memory polymers (SMPs), self-cooling and self-heating materials, while self-healing and self-cleaning are predominant (Bleay et al. 2001; Wu et al.

2008, 2016). In food industry, smart packaging is interchangeably used with intelligent food packaging and/or active food packaging and features of self-healing, tear and wear of packaging, however, lack their principles and accomplishment models (Chellaram et al. 2014; Imran et al. 2010; Neethirajan and Jayas 2011; Wu et al. 2015b).

Self-healing polymer materials

Self-healing polymers or SMPs have the capacity to recover and remedy the damaged material to its original shape or structural features as the function of the exerted external stimuli (Pei et al. 2016). Figure 3 shows the reversible equilibrium in polymer materials to attain self-healing by lowering their molecular mass and viscosity in which the defects can be easily mended in the re-polymerization processes (Blaiszik et al. 2010; Meng and Jinlian 2010). Self-healing biomaterials are usually brittle (Imran et al. 2010) or amorphous in nature which allows for reversible equilibrium formed by covalent bonds, non-covalent, reversible bonds, covalently cross-linked by physical networks of crystalline domains, ionic aggregates or multiple hydrogen bonds (Hager et al. 2015; Youngblood and Sottos 2008). Some of the polymers exhibiting the damage or temporary shape recovery and memorization of their original shapes include poly(ether–ether-ketone), crosslinked polyethylene, polyester, polyurethane (PU), epoxy resin and polynorbornene (Haghayegh et al. 2015; Wu et al. 2016; 2015b). Sucessful and efficient self-healing processes rely on the quiescent of materials, stability of healing agents, promptly deliverance of healing agents, reactivity of healing agent to polymer matrix, volume shrinkage during polymerization processes, degree of crosslinking of polymer and healing agent, process repetitiveness and activator or catalyst within the materials (Youngblood and Sottos 2008).

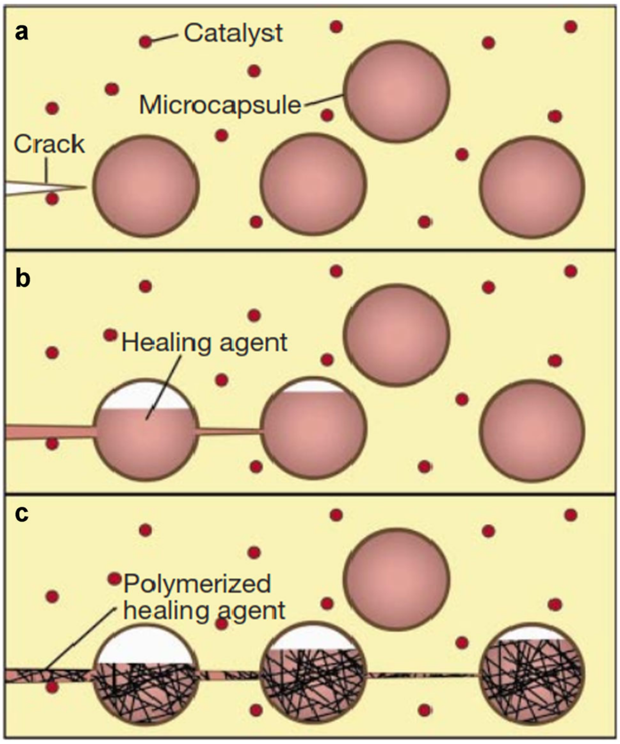

Fig. 3 The activated autonomic healing scenario presenting the healing agent and catalyst in micro-embedded onto the composite matrix. **a** Crack formed in the composite material due to damages; **b** the damage's signals rupture the embedded microcapsule and release the healing agents by capillarity for recovery; **c** the crack of composite matrix is closed at the surface after catalyzation of healing agent through polymerization process (White et al. 2001)

Self-healing phenomenon has been broadly categorized into two groups namely: extrinsic which is based on capsule healing and vascular healing systems, and the intrinsic healing polymers (Blaiszik et al. 2010; Haghayegh et al. 2015; Zhu et al. 2015). The intrinsic self-healing works on the principles of chemical, physical and the intra/intermolecular interactions in the polymer matrix, which are autonomic at the microscale, nanoscale or molecular levels (Blaiszik et al. 2010; White et al. 2001; Youngblood and Sottos 2008; Zhu et al. 2015). According to Blaiszik et al. (2010), when the additives like catalyst are incorporated within the materials, the process of self-healing depends on external activation at interface including moisture, heating, light, chemical constituents, water/solvent or magnetic fields (Ma et al. 2016; Wu et al. 2016; 2015b) (Fig. 3). Activation of healing process by heating and photo-irradiation are common, where the glass transition (Tg) of materials (Bleay et al. 2001; Wu et al. 2008) acts as bioswitch to trigger the recovery process by damage (Wan et al. 2015). When the polymeric materials are heated to a temperature above their Tg, they recover their original shapes by releasing their internal stored stresses in crosslinking structures that can stretch the polymer chains from irreversible chain mobility during deformation (Meng and Jinlian 2010; Youngblood and Sottos 2008).

The extrinsic systems use the pre-incorporated healing agents in the polymer resins and involve no molecular modifications. The healing agents in the liquid forms are micro/nano-encapsulated within the shell containers as microcapsules and pipelines before incorporation in the polymer matrix (Fig. 4), by which the encapsulated healing agent flow by capillarity to the area of damage and cures (Bleay et al. 2001; Haghayegh et al. 2015; White et al. 2001; Zhu et al. 2015). Some of the contained monomers that can be used for fracture curing include: methyl methacrylate, cynoacrylate, azobenzene (Wu et al. 2016), acrylic and thiol-ene monomers and poly(ethylene-co-butylene) (White et al. 2014). Wu et al. (2016) investigated the shape-memory features of substrates formed from polycaprolactone-PU blended with azobenzene and

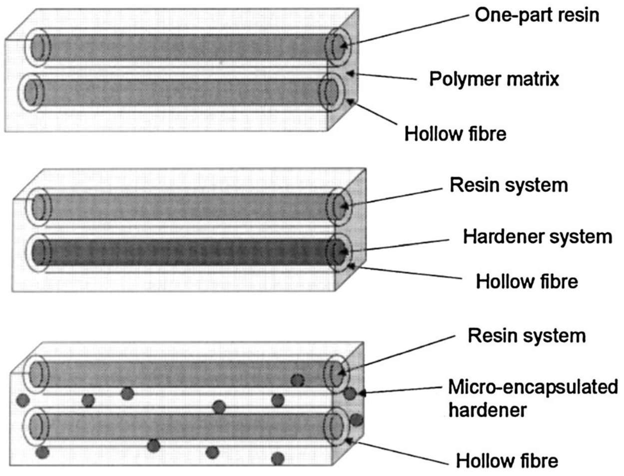

Fig. 4 Schematic presentation of non-activated smart repair concept for polymer matrix. When the containers are damaged by the external impacts the polymer resin flows in the liquid form to the damaged area and cures the damage (Bleay et al. 2001)

observed that PUs was with hard segments in the side-chains with low stability compared PUs with azobenzene in their main chains for effective self-healing process. In other case, the shape memory epoxy resin was constructed using 9,9-Bis [4-(2-hydroxyethoxy)phenyl] fluorine (BHEPF) and dimethyl sulfoxide (DMSO) and cured in presence of 4,4́-diaminodiphenylsulfone (DDS) was conducted in conventional oven for 2 h and for 4 h in a convection oven (Wu et al. 2015b). Heating of the resins at 150 and 180 °C for 2 and 4 h confirmed that the shape memory properties of the resins relied on the ratios of storage modulus, fold-deploy and tensile-shrinkage tests, respecitively. The ratio exhibited between glassy and rubbery states was 400 for storage modulus tests and 99 % was observed as shape fixed ratio for fold-deploy shape memory test, with 100 % shape recovery ratio within 30 s at 185 °C. At the same temperature by the tensile-shrinkage test, 25 % of strain was observed with the recovery stress of 2.5 MPa and 99 % ratio for shape recovery and shape fixed after six test cycles.

Self-cleaning polymer materials

The phenomena of self-cleaning effects are based on the inherent washing force properties of the superhydrophobical solid surfaces, which allow for ease rolling-off of water droplets with debris (Ye et al. 2015) and other particulates by controlling wetting properties (Liu et al. 2015c; Youngblood and Sottos 2008). The rolling of debris reduces the contact point and surface tension of water stimulated by surface energy through condensed water vapors, enabling capture and localization of debris to produce contaminant free surfaces (Liu et al. 2015c). This hydrodynamic shear stress mechanism is controlled in a gas flow or liquid stream that can be accomplished through three removal mechanisms including floating, lifting and then aggregating processes after overcoming the attractive forces of capillary and van der Waals interactions (Ye et al. 2015). The phenomenon mimics the natural mechanisms of self-cleaning effects of lotus leaf (Fig. 5a), water strider's leg (Fig. 5b) and other living things with unique micro/nanoscale structures on their surfaces (Liu et al. 2015b). Self-cleaning effects of superhydrophobic surface are promoted by the large contact angle and small hysteresis (Liu et al. 2015c) and governed by three-phase contact lines of the drop-in planes of liquid–gas interface, solid–gas interface and solid–liquid interface. The phenomenon is described according to Young's and Wenzel equation (the later not explained here) (Martines et al. 2005; Ye et al. 2015):

$$Cos\,\theta = (\gamma_{SV} - \gamma_{SI})/\gamma_{IV} \qquad (1)$$

where γ_{SV} is solid–gas interface energy, γ_{SI} is liquid–gas interface energy, γ_{IV} is solid–liquid interface energy, and θ is contact angle created between water droplet and solid substrate.

The superhydrophobicity of surfaces (angle >150° with water) prevent the wetting properties of substrate due to the reduced contact point and low surface tension of water (Kumar et al. 2015; Ye et al. 2015). Common materials in construction of bioinspired self-cleaning packages include titania and silicon nanoparticles, CNTs and graphite (Liu et al. 2015a, c; Wu et al. 2015a; Yildirim et al. 2015). The self-cleaning copolymer brushes of N-isopropylacrylamide (NIPAAm) and poly(ethylene glycol) methacrylate (PEGMA) embedded on polypropylene (PP) surface increased oil contact angle higher than 141° and lowered the oil adhesive forces below 20 µN (Ye et al. 2015). The self-cleaning tests showed only 0.2 wt% of oil residues remained on the modified surfaces of P(NIPAAm-5PEGMA) (Wan et al. 2015). Similarly, the PET film coated by titania-silica nanocomposite realized increase in hydrophobicity and decrease of the water contact angle in the modified films surface from 82.7° to 18.7° (Kumar et al. 2015; Wu et al. 2015a). Liu et al. (2015b) developed the self-cleaning surface of glass substrate, (Heptadecafluoro-1,1,2,2-tetrahydrodecyl)trimethoxysilane and exhibited superhydrophobicity features with rough surface, hill-like morphological surface and wrinkled surface resembling the microstructural features of lotus leaf. The contact angle of the glass surface increased to 169° with tilt angle lower than 5°. The potential self-cleaning behavior was attributed by the dust-repellent properties of the surface, stable plastron and water-jet impact resistances.

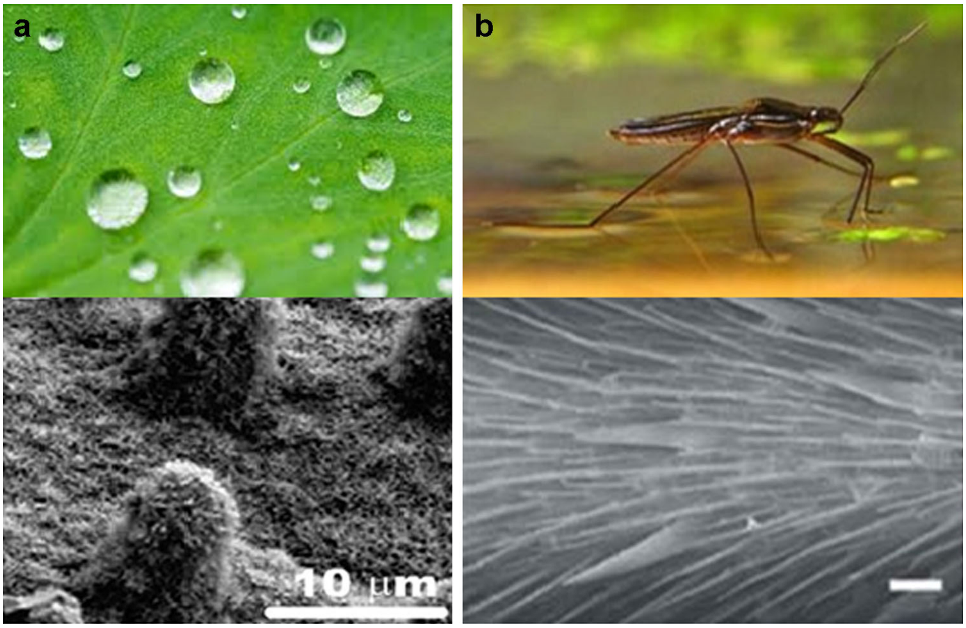

Fig. 5 **a** Self -cleaning of lotus leaves; **b** dewetting of water strider's legs in water. The lower parts show the SEM micrographs of natural superhydrophobic microstructures of lotus leaf and water strider leg microstructures [collected from internet sources and others adapted from (Youngblood and Sottos 2008)]

Distinctive features in novel food packaging systems

Innovative packaging develops the accurate, cost-effective (Rashidi and Khosravi-Darani 2011; Wu et al. 2015a), fast, consistent and noninvasive techniques for evaluating the real-time quality and safety of food products (Bastarrachea et al. 2011; Duncan 2011; Nopwinyuwong et al. 2010; Puligundla et al. 2012; Ranjan et al. 2014; Realini and Marcos 2014). Smart and intelligent packaging consists of overlapping features on the use of stimuli, as the course of action. However, smart packaging deals with the geometrical and architectural integrity of packages. It includes the invisible resilient features enabling the packages to modulate and recover their original shapes and sizes after the release of internal stress due to signal activations (Imran et al. 2010; Meng and Jinlian 2010; Neethirajan and Jayas 2011). Smart packaging involves in perambulation of the integrity of micro/nanostructures in the package as well as the quality of the package itself (Youngblood and Sottos 2008). In general, the smart packaging system reviews the sustainability of both intelligent and active packaging internal features (Meng and Jinlian 2010). For example, the smartness of packaging ensures the recovery or memorizes the *status quo* of packaging after the dynamic depolymerization and repolymerization and then hydrophilicity to hydrophobicity in intelligent packaging (Fig. 6).

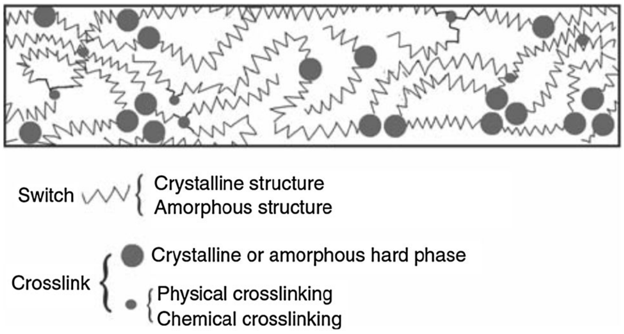

Fig. 6 The molecular mechanism of thermal-active smart memory effects. The transition and melting temperatures of constituting polymers act as bioswitch or recovery point of materials. Simply, the rubbery states of the material at high temperatures allow to release the internal stress at lower temperatures where recovery to original shape occurs (Meng and Jinlian 2010)

Intelligent packaging features deal with communication function to processors, distributors, retailers and consumers to facilitate decision-making (Neethirajan and Jayas 2011; Nopwinyuwong et al. 2010) and to enhance the quality and safety of food products. Intelligent packaging can also contribute to onsite quality and safety management in food processing and production systems and help to establish intervention strategies to reduce or eliminate the associated hazards (Biji et al. 2015). American Heritage Dictionary defines intelligent as act of showing rationally and sound judgments or with some storage and processing capacity of data. Sound decision-making is a main prerequisite of intelligent food packaging for effective communication and sharing information among the parties. They target the regulatory standards and the integrity of the packed food products. Many of their features are based on fundamental bioanalysis of biological substances and food matrix, which deliver signals to the target user in form of indicators.

Provisionally, active food packaging surveillances the quality and safety of packed food products from physical, microbial and biochemical deterioration agents include exclusions of moisture, oxygen and microorganism. Active food packaging can be adopted easily in a wide range of environments to reduce the postharvest losses and hence improving the food security, especially in developing countries (Koskela et al. 2015; Neethirajan and Jayas 2011). In this case, active food packaging can be easily integrated with smart and intelligent packaging features, which presents the multipurpose packaging systems. For example metallic, metallic oxides and their nanoparticles can be diversely applied in antimicrobial applications, barrier functions, detecting systems (Imran et al. 2010; Yildirim et al. 2015), photoreactive substances (Rashidi and Khosravi-Darani 2011) and self-cleaning materials when coated on surface of various substrates (Liu et al. 2015a, b; Wu et al. 2015a). These include nanomaterials from iron oxides, gold, alumina, silver, silica, zinc oxide, CNTs, graphites (Ranjan et al. 2014) and titania (TiO_2). From the features describe in all sections above, it is obvious to note that some features described as “smart” in food packaging may mislead the future explorations of innovative packaging in food industry. Developing the smart packaging system based on the presented phenomena have various added advantages over the previous reported from utility and time serving to durability and reliability perspectives (Zhu et al. 2015).

Safety and legislation issues

Every new technology comes with new unknowns to society. Nanotechnology applications in food industry is a pivotal to evolution of innovative packaging techniques; however, comprehensive understanding of the likelihood of interactions between nanomaterials used in packaging and the food matrix is critically needed. Addressing the safety and legislation prospects of nanotechnology, three scientific domains towards established food safety and legislation systems for nanomaterials have been considered.

Safety concerns

The quality of packages and packaging materials relies on the scientific assurances of the possible interactions between packages and food matrix as well as the possible adverse repercussions of engineered nanomaterials to food, package, consumers or all. Poor packaging may cause serious deterioration and recontamination of food product. It can lead to serious alterations of product appearance, organoleptic and chemical modifications of formulations, decrease product acceptability, reduction in nutrients bioavailability and bioaccessibility, enhance oxidation, increase waste, cause safety calamity and result in lowering overall product quality (Biji et al. 2015; Yildirim et al. 2015). The most pending issue with current development of food nanotechnology is clear and comprehensive understanding of the fate and toxicity of nanomaterials in food products and their ultimate environmental impacts and human health side effects (Bumbudsanpharoke and Ko 2015; de Azeredo 2009). In addition, the interactions between nanomaterials and biological systems are not yet comprehensively elucidated and understood. It is claimed that, nanomaterials have capability of penetrating in biological systems and have linked to poor clearance from the body depending on their size and shape (Bumbudsanpharoke and Ko 2015; de Azeredo 2009).

Nanomaterials analysis

Identification, characterization, quantification and ultimate validation of nanomaterials in food matrices are quite complex phenomena in analytical sciences. Acquiring comprehensive scientific evidences of nanomaterials through risk assessment and characterization is a fundamental task for development of proportionate verification frameworks. Therefore, the lack of reliable analytical techniques and thereafter data lead to inadequate safety laws, testing protocols and workplace health procedures for nanofoods, nanofoods packaging and nanobased chemicals (Neethirajan and Jayas 2011). The measurement of engineered nanomaterials and especially metallic nanoparticles which exhibit higher dependency in thermodynamic conditions in food matrix imposes difficulties in standardizing their validation protocols (Huang et al. 2015). In addition, the nanomaterials present in food matrix might be at low concentration and commonly nanomaterials analysis deploys various detection techniques in order to gather the reliable information for risk assessment (Blasco and Picó 2011; Kuorwel et al. 2015). Some of the sensitive analytical approaches which are commonly used in analysis of nanomaterials can be subdivided into three groups, (i) imaging techniques like atomic force microscope, SEM and TEM, (ii) spectroscopic techniques including X-ray diffraction and small/wide-angle X-ray scattering, dynamic light scattering (DLS) and (iv) quantitative analytical techniques (Blasco and Picó 2011; Dasgupta et al. 2015; Huang et al. 2015; Kuorwel et al. 2015).

Legislation issues

Despite nanoscience and nanotechnology applications receive higher research priority in food production and agricultural sector worldwide, no safety assessments or existing regulatory frameworks either in developed or developing countries (Amenta et al. 2015; Bumbudsanpharoke and Ko 2015; Vance et al. 2015). The nano-related food products are regulated horizontally based on existing chemicals, cosmetics and food laws in various countries (Cushen et al. 2012). Generally, many parts of the world implement precautionary principles to regulate the nano-based products, as scientific evidences required by regulatory bodies to establish safety guidelines are insufficient, inconclusive, or uncertain. Therefore, there are global intentions by regulatory institutions, academia and industries to gathering required information and standardizing for the fate of proceeding to establishment of required legal frameworks for testing nanomaterials in nano-based food products. Although the current information shows that the amount of nanomaterials migrating from packaging materials into food matrix is negligible (Bumbudsanpharoke and Ko 2015). However, the effect and reactions of these migrated nanomaterials to the environment and biological systems remain imprecise. The Food and Drug Administration (FDA), United States, has listed some nanomaterials as Generally Recognized As Safe (GRAS), allowed as food contact materials comprising of nanoclays or montmorollites, alumina, zinc oxides and carbon blacks. These materials do not need the competent authorities to authorize them in the market. The scenario is different with European regulatory frameworks where only titanium nitride, carbon blacks and silica can be used in plastic food packaging. The application of nanoclay, silver materials, aluminum nanomaterials and zinc oxides in food packaging is not authorized in the European market (Bumbudsanpharoke and Ko 2015).

Conclusion: future trends in packaging

The innovative food packaging based in nanotechnology covers small global segment to assume all privileges in food industry. Food nanotechnology industry is at its infancy stage, however, the annual research and investments in the sector is steadily growing both in academia and industrial institutions. There are technical and functional hindrances to development of nanomaterials for food packaging such as safety regulations, standardizations and trained workforces in different geographical locations. Currently there is a void knowledge regarding the migrations of nanomaterials from packaging onto food matrix, possible interactions which can occur between nanomaterials and biological systems and the concentrations of nanomaterials with ultimate impacts to human health and the environment at large. More knowledge is needed on toxicity characteristics of nanomaterials and their physicochemical properties such as surface charges, particle size, geometrical structure and shapes regarding various modes of exposure to human beings and environment for appropriate and reliable techniques for their identifications, characterizations and quantifications in complex food matrices as well as their disposal strategies.

However, innovative food packaging is the future tool to revolutionize the global agricultural production and food chain systems. Innovative food packaging has many features desirable in food packaging industry from mere quality control of food products to the biosecurity and safety applications. At large, the kind of packages is of paramount in this era of global food insecurity, high post harvest losses, global population increase, climate change, and increased competition of water for agriculture and domestic use. Developing countries should more embrace in the research and future adaptation of innovative packaging technologies that will help in reduction of the food loss in food supply chains, especially during storage, processing, packaging, and transport and distribution.

Acknowledgement The authors appreciate the financial support from the Government of Tanzania through the Commission for Sciences and Technology (COSTECH) and Centre for Science & Technology of Non-Aligned and Other Developing Countries (NAM S&T Centre), Government of India, through Research Training Fellowship for Developing Country Scientists (RTF-DCS) 2014/2015. N.M thanks the management of the NM-AIST for permission granted to attend the training and the Central Institute of Post-Harvest Engineering and Technology (CIPHET), Ludhiana, Punjab, India for hosting the research work.

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