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Review Article

Plant-Mediated Synthesis of Mono- and Bimetallic (Au–Ag) Nanoparticles: Future Prospects for Food Quality and Safety

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The environmental, economic, and operational limits associated with the physical, chemical, and microbiological techniques for the production of nanoparticles (NPs) are the principal obstructions to their rapid commercial applications in various fields including food packaging and sensing to ensure food quality and safety. Over the years, many reports revealed that the nanotechnological (metal-based NPs) application facilitates an alternate, interactive, reliable, as well as simple technology in the food industries and packaging sector. In this review, we summarized the usage of plant extract for the biosynthesis of bimetallic (Au–Ag) and monometallic counterpart NPs. Further, the impact of reaction conditions and identification of reactive phytochemicals with the reaction mechanism of these nanoparticles was reviewed. The recent progress on the applications of Ag, Au, or Au–Ag NPs in food quality analysis and food packaging was comprehensively discussed. The safety aspect of the nanoparticles for food sector use was also briefly stated.

1. Introduction

Food quality and safety are the prime areas of concern among consumers. Food safety is a scientific topic that discusses how food is processed, handled, and stored to protect food from the external environment like foodborne illness. On the other hand, food quality refers to the quality characteristics of food such as appearance, chemistry, physics, flavor, freshness, texture, and micro-organisms. Public health is closely related to food quality and food safety [1–3]. Unsafe food can cause more than 200 diseases (ranging from diarrhea to cancer) by harmful chemical substances or pathogens (bacteria, parasites, prions, viruses). The world health organization (WHO) estimated that about one-tenth (near about 600 million people) of the world's population becomes ill after eating unsafe food, and 4,20,000 die every year [4]. Currently, increasing growth of customer demands for high-quality fresh food and lifestyle variations poses challenges to produce fresh, delicious, and ready-to-eat foods with guaranteed quality and safety and growing preference for new materials in food packaging techniques.

Traditional food preservation methods have an unavoidable and undesirable adverse effect on the healthy properties of food and its quality. In the last 20 years, the use of green-based synthesized nanoparticles (NPs) is one of the best technologies that maintained the quality of the food and at the same time enhanceed safety, prolonged shelf-life, or sensory applications [5, 6]. NPs have a wide range of improved and unique physicochemical properties that set them apart from their bulk counterparts. When material particles are in the nanorange of 1–100 nm in size, they typically exhibit remarkable and even surprising properties. Nanoscaled materials have distinct properties due to their high surface energy, large surface-to-volume ratios, large fraction of surface atoms, huge number of active sites reduced flaws, strong electron transfer abilities, and spatial confinement [7, 8]. Silver (Ag) and Gold

(Au) are the most studied monometallic NPs with a broad range of significant food packaging applications among different noble metals NPs [9, 10]. On the other hand, bimetallic NPs overcome the inadequate properties of monometallic NPs and often display greater catalytic activity than their monometallic counterparts [11, 12].

Wet synthesis methods must be replaced by green chemistry processes that are nontoxic, clean, dependable, biocompatible, benign, and environmentally beneficial [13]. As a result, researchers have redirected their focus to "green" chemistry methods for producing metallic nanoparticles. One such ecologically friendly technology that has received a lot of interest in recent years is biofabrication of metallic nanoparticles using various plant systems. Current research on the biofabrication of metallic nanoparticles using plant extracts has convoyed in a new era of renewable, fast, nontoxic, environmental friendly, and biocompatible nanoparticle fabrication technologies. Plants are the primary photosynthetic autotrophs and producers in the food chain. These are incharge of producing a big amount of biomass in their natural environment [14, 15].

Plants have an incredible ability to convert solar energy to chemical energy. Plants and plant products can thus be used to produce nanoparticles as renewable and sustainable resources. Plants have a diverse range of antioxidant secondary metabolites [16]. Prokaryotic microbial systems exceed plant resources. Microbial systems demand both expensive culture care procedures and downstream processing [17]. Plant resources, as previously said, are a sustainable source of renewable energy, and researchers are interested in fabricating nanoparticles utilizing living plants, plant extracts, or phytochemicals. The ability of plant systems to swallow, collect, use, and recycle various mineral species is at the heart of the plant-mediated synthesis technique for the production of metallic nanoparticles. Plant-mediated synthesis is a fast and low-cost method for manufacturing vast numbers of highly stable nanoparticles [18, 19].

2. Synthesis of Nanoparticles

Several biological, chemical, and physical methods are available for the production of metallic NPs but each method has its own set of advantages and limitations. Some of the solvents, reducing and capping agents, employed in chemical processes have been shown to be unsafe to humans and pose risk to the environment in general. Furthermore, these methods are costly, and formation of toxic by-products may cause environmental pollution [15, 16, 18]. However, physical synthesis methods have limitations such as trained manpower is needed to operate sophisticated instruments and large amounts of energy are required to maintain the high pressure and temperature conditions for synthesis [13, 14]. Therefore, it is necessary to develop a simple, nontoxic, and inexpensive method for the production of monometallic and bimetallic NPs.

2.1. Biosynthesis of Nanoparticles. Among different noble NPs, AgNPs and AuNPs are widely used in fast-growing consumer products which directly come into close contact with the human body, so it is imperative to develop NPs with sustainable and eco-friendly approach that poses no risks to workers and consumers. In recent time, biosynthesis methods have received significant attention with major advantages like being eco-friendly, applicable at room temperature and pressure, and formation of biocompatible by-products; in addition, there are no needs for external reducing and stabilizing agent [10, 20]. Of late, biosynthesis methods for NPs utilizes many biological sources existing in nature such as viruses, bacteria, algae, fungi, and several plant extracts/biomasses. As an alternative to conventional methods, biosynthesis methods in the case of using micro-organisms (like viruses, bacteria, algae, fungi, etc.) have some shortcomings such as adherence of organisms on the surface of NPs may cause infection, inexpensive media requires microbial growth, tedious process of isolation technique, and maintenance of microbial culture [15–17, 21].

2.2. Biosynthesis of Nanoparticles using Biopolymer. Many researchers (both academic- and industry-based) have continued to improve the biosynthesis process of nanoparticles using polymer materials. Based on several aspects such as production, transport, interaction with food, and storage, we need an effective method for biosynthesis of nanoparticles using biodegradable polymers in terms of safety of both consumer and environment [22, 23]. Biopolymers are one kind of polymeric material, which are degraded by naturally occurring organisms (virus, fungi, bacteria, etc.) under suitable conditions of temperature, oxygen, and moisture [24]. Polymers' usage in food packaging material has risen tremendously due to advantages such as strength, stiffness, moisture and oxygen barrier, and flexibility over earlier materials. Polymers are most commonly applied in the food sector for food packaging to keep food fresh under atmospheric conditions; however, it is very important to select the suitable polymer [25, 26]. Biopolymers can be divided mainly into synthetic biodegradable polymers, natural biopolymers, and microbial polyesters. Naturally occurring biopolymers such as agar, alginate, cellulose, chitosan, and starch (starch-polycaprolactone, starch-polylactide, blends of different biopolymers) are from plant carbohydrates and proteins like soy protein, corn zein, casein, collagen, gelatin, wheat gluten, whey protein, and polysaccharides [27–30]. These polymers have unique qualities such as thermal stability, biocompatibility, and flexibility and barrier properties with respect to soil, water, and air. To improve the thermal and mechanical properties of food packaging materials, bio-based nanocomposites are made by embedding metal nanocomposites with biopolymers. In food industries, the use of NPs which is biosynthesized by naturally occurring biopolymers can be safe for human s well as the environment [31, 32].

2.3. Biosynthesis of Nanoparticles using Plant Extract. It is beneficial to employ the plant extracts toward the production of NPs as compared with other biological (micro-organism based) methods due to ecologically sound, less, or no chances of contamination, cost-effectiveness, and very simple laboratory requirement for NPs synthesis. Also, plant-mediated biosynthesis are simple, safe to handle, single-step, unique, faster, improved stability, and suitable for large-scale production [16, 18, 33]. This method can also be utilized to generate metallic nanoparticles on an industrial scale by utilizing tissue culture and downstream process optimization methods. In contrast to traditional synthesis approaches, plant-mediated synthesis procedures primarily use aqueous (water) extract for manufacture and require normal temperature and air pressure, resulting in significant energy savings [34]. All of the criteria for more ecologically friendly synthesis are met by plant-based techniques. Plant-mediated synthesis has become a viable alternative to traditional physical, chemical, and even microbiological techniques as a result of these improvements. In addition, the plant extracts possessing a number of secondary metabolites such as carbohydrates, phenols, flavonoids, alkaloids, proteins, steroids, sugars, tannins, and terpenoids may also impart many functional properties like antioxidant and antifungal activities to the biosynthesized NPs. These secondary metabolites also play a vital role in the reduction and stabilization of NPs. Several plant parts, such as plant leaves, bark, flowers, fruits, roots, seeds, stems, or whole plants are used for the biosynthesis of NPs [15, 35–37]. Some of the reports pertaining to monometallic (Au and Ag) and bimetallic (Au–Ag) NPs biosynthesis mediated by various plant extracts are summarized in Table 1.

The key advantages of plant-mediated synthesis are as follows [15, 16, 18]:

(a) This is a fast process
(b) Use of aqueous solvents
(c) The easy availability of plant
(d) Biocompatible plant extracts (suitable for medicinal use)
(e) This plant extract is suitable for large scale production
(f) Contamination is either negligible or nonexistent (eco-friendly and safe for medicinal use)
(g) This is a simple process that necessitates adequate pressure and temperature (economical)
(h) The combined activity of phytochemicals as a reducing and stabilizing agent (cost-effective)

2.4. Factors Influencing the Plant-Extract-Mediated Synthesis. The optimization process is important in order to customize and control the monodispersity, stability, morphology, large-scale production, and rate of synthesized NPs in plant-extract-mediated synthesis process. Indeed, extracts of plant parts are enriched with phytochemical such as amino acids, carboxylic acids, polyphenols, polysaccharides, proteins, and terpenoids vitamins [21, 56]. The phytochemical present in aqueous extract is believed to serve dual roles successfully participating in the NPs formation (as reducing and capping or stabilizing agents) methods and control the characteristics (morphology and composition) of the biosynthesized NPs. Apart from the characteristics, the stability and quantity of the biosynthesized NPs could be controled not only by phytochemical (reducing and capping or stabilizing agents) but also by varying some other several physicochemical parameters (Table S1) such as metal salt concentrations or proportions, concentration of plant extract, temperature, contact time, and the pH [1, 59, 60]. So, the optimization of operating conditions is very essential in plant-mediated NPs synthesis to accomplish the smaller-sized and large-scale production of NPs along with the reduction of the excess use of precursor materials and laboratory trials. Generally, only one factor at a time was chosen by researchers to investigate the probable optimum level of different parameters as an overall investigation becomes complicated, several variable conditions are there and have interrelationship with them, time-consuming and cumbersome task [61–64]. Monometallic (like Au and Ag) and bimetallic (like Au–Ag) NPs have a distinctive SPR peak depending upon the type of NPs, morphology, and composition of the particle. The SPR peak response of metallic NPs is modulated by changes in the size, shape, and composition of the NPs with respect to one another.

3. Characterization Techniques of Nanoparticles

The diverse applications of NPs primarily depend on their morphology and size. So, it is therefore very important for the chemist to control their properties, that is, the size and morphology of single NP for application in various fields [65]. After synthesis of NPs, characterization of NPs can be performed by several instruments like UV–vis spectroscopy, atomic force microscope (AFM), fourier transform infrared spectroscopy (FTIR), dynamic light scattering (DLS), zeta potentials, mass spectroscopy (MS), transmission electron microscopy (TEM), energy dispersive X-ray spectroscopy (EDX), and other characterization techniques with their functions are shown in Table S2. To understand the various properties of NPs such as morphology, size distribution, composition, surface area, aqueous stability, homogeneity, dispersity, and net charge on the surface are generally determined using these different characterization techniques. The resulting information provides answers to distinguish whether specific NPs can be used as a catalyst or for biological applications, or else to develop their synthesis methods, and in miscellaneous fields [66, 67].

3.1. Bioactive Compounds Involved in Biosynthesis of Nanoparticles. The nature of the active components adsorbed on the NPs' surface has been investigated by various researchers using various techniques such as gas chromatographic–mass spectrometric and FTIR analysis [68, 69]. Plants contain enormous active chemical components belonging to various secondary plant metabolites such as steroids, alkaloid, phenolic acid, enzyme, and terpenoid containing various functional groups (–NH, –CHO, –OH, –COOH, and –COOR). These biomolecules are mainly involved in the biosynthesis of metallic NPs. Previously, various reports suggested that biosynthesis of monometallic (Ag and Au) NPs and their bimetallic (Au–Ag) alloy NPs using plant extract as suitable reducing and stabilizing agents [10, 70, 71].

3.2. Possible Mechanism for Biosynthesis of Nanoparticles using Leaf Extract. Figure 1 displays a possible biosynthetic approach for mono- and bimetallic (Au–Ag) nanoparticles [63, 73, 74]. The biosynthesis of NPs is often categorized into three stages: activation, growth, and termination. Metal ions are reduced when a salt solution is injected into a plant-leaf extract during the activation step, resulting in the formation of metal nuclei, with the plant extract functioning as the

Table 1: Recent progress in green synthesis of mono- (Au and Ag) and bimetallic (Au–Ag) nanoparticles based on a single-plant extract during the last two decades (2000–2020).

Plant	Type of nanoparticles	Size (nm)	Morphology and nature	Applications	References
Azadirachta indica	Au	–	Triangular and hexagonal	Not reported	[38]
	Ag	5–35	Spherical		
	Au–Ag	50–100	Spherical; core–shell		
Volvariella volvacea	Au	20–150	Triangular nanoprisms	Not reported	[39]
	Ag	~15	Spherical		
	Au–Ag	–	Alloy		
Swietenia mahogany JACQ	Au	–	Spherical	Not reported	[40]
	Ag	–	Spherical		
	Au–Ag	–	Spherical; alloy		
Anacardium occidentale	Au	6.5	Spherical	Not reported	[41]
	Ag	5	spherical		
	Au–Ag	8	Spherical; alloy and core–shell		
Brassica oleacea var. capitata	Au	20 ± 3	Spherical and triangular	Not reported	[11]
	Ag	–	–		
	Au–Ag	25–200	Spherical; alloy and core/shell		
Dalbergia sissoo Roxb.	Au	16–25	Spheroids with triangles & hexagons		[42]
	Ag	17–31	Predominantly spherical		
	Au–Ag	–	Predominantly spherical; alloy and core/shell		
Potamogeton pectinatus L.	Au	8.4	Spherical, few nanotriangles	Not reported	[33]
	Ag	50.4	Spherical		
	Au–Ag	6.6	Spherical; alloy		
Piper pedicellatum C. DC	Au	2–40	Triangular, hexagonal, and pentagonal	Not reported	[43]
	Ag	2–30	Spherical		
	Au–Ag	3–45	Spherical, triangular, pentagonal, and hexagonal; alloy		
Plumbago zeylanica	Au	20–30	Nanospheres and nanotriangles	Biofilm inhibition	[44]
	Ag	60	Nanospheres		
	Au–Ag	90	Hexagonal blunt-ended		
Lansium domesticum	Au	20–40	Triangular and hexagonal	Biocompatibility, and antimicrobial activity	[45]
	Ag	10–30	Spherical		
	Au–Ag	150–300	Branched spherical; alloy		
Catharanthus roseus Linn	Au	25–65	Spherical, triangles, hexagonals, and rods	Antibacterial and anticandidal activity	[46]
	Ag	11–26	Spherical		
	Au-Ag	20–25	Spherical; core-shell		
Jasminum sambac	Au	20–50	Spherical	Antimicrobial activity	[47]
	Ag		Spherical		
	Au–Ag		Spherical; alloy		
Rivea hypocrateriformis	Au	20–30	Spherical	Antimicrobial, antioxidant, and anticancer activities	[48]
	Ag		Spherical		
	Au–Ag		Spherical; alloy		
Gloriosa superba	Au	Avg. 20	Triangular and spherical	Antibacterial and antibiofilm activity	[49]
	Ag	Avg. 20	Triangular and spherical		
	Au–Ag	10	Spherical		
Guazuma ulmifolia L.	Au	20–25	Spherical	DNA/protein interactions, photocatalytic, antimicrobial, and anticancer agents	[50]
	Ag	10–15	Spherical		
	Au–Ag	10–20	Spherical; alloy		

(*continued*)

Table 1: Continued.

Plant	Type of nanoparticles	Size (nm)	Morphology and nature	Applications	References
Cannabis sativa	Au	–	Triangular and spherical	Antibacterial and antileishmanial activity	[51]
	Ag	–			
	Au–Ag	–	Triangular and spherical; alloy		
Cetraria islandica (L.) Ach.	Au	6–19	Spherical	Catalytic reduction of 4-NP	[35]
	Ag	6–19	Spherical		
	Au–Ag	6–21	Spherical and polygonal; alloy		
Solidago canadensis	Au	238.2	Spherical, few are triangular, and rod-like shapes	–	[36]
	Ag	180.6	Spherical, few are triangular, and rod-like		
	Au–Ag	186.3	Spherical, few are triangular, and rod-like		
Stigmaphyllon ovatum	Au	80	Triangular	Cytotoxicity	[52]
	Ag	24	–		
	Au–Ag	15	Alloy		
Solidago canadensis	Au	21.3	Spherical	–	[53]
	Ag	32.2	Spherical		
	Au–Ag	25.9	Spherical, few are triangular, and rodlike		
Madhuca longifolia	Au	36–60	Spherical	Wound healing bioefficacy	[54]
	Ag	35–50	Spherical		
	Au–Ag	34–66	Spherical		
Asparagus racemosus	Au	10–50	Spherical	Antibacterial and immunomodulatory potentials	[55]
	Ag	10–50	Spherical		
	Au–Ag	10–50	Spherical; alloy		
Polyalthia longifolia	Au	5–20	Spherical	Catalytic activity for dye (methylene blue, methyl violet, and methyl orange) degradation	[56]
	Ag	5–20	Spherical		
	Au–Ag	5–20	Spherical; alloy		
Moringa oleifera	Au	96 nm	–	Anticancer activities	[57]
	Ag	129 nm	–		
	Au–Ag	11–25 nm	Hexagonal, triangular, and spherical shape		
Pulicaria undulata	Au	–	Irregular or anisotropic	Catalytic activity for the reduction of 4-nitrophenol	[58]
	Ag	–	Irregular or anisotropic		
	Au–Ag	5–12	anisotropic with attached spherical		
Syzygium aromaticum	Au	Avg. 27.12	Hexagons and polyhedral	antioxidant and catalytic reduction of p-NP, methyl orange, and mrthylene blue	[37]
	Ag	Avg. 17.94	FCC type of crystal structure		
	Au–Ag	Avg. 16.04	Hexagonal and polygonal; alloy		

reducing agent. The adjacent metal nuclei consolidate further throughout the growth stage to create the final NPs.

4. Nanoparticles

Nanotechnology has evolved so exponentially in the last decade that we cannot imagine any field without nanoparticle material. In general, nanoparticles are defined as small particles that behave as a whole unit in terms of their characteristics and transport. It can be quantitatively defined as a small object with at least one-dimension size (a single unit small size) and diameter between 1 and 100 nm (nanoscale range) [75]. Nanoparticles exhibit a unique and wide range of improved properties compared with larger particles of bulk material due to the variation in characteristics such as size distribution, ionic state, phase, and morphology of the particles [76]. Their uniqueness arises from their bulk counterpart which is mainly due to a higher surface-to-volume ratio with a reduction in the particle size. These NPs can be metallic, organic, mineral, polymer-based, or a combination

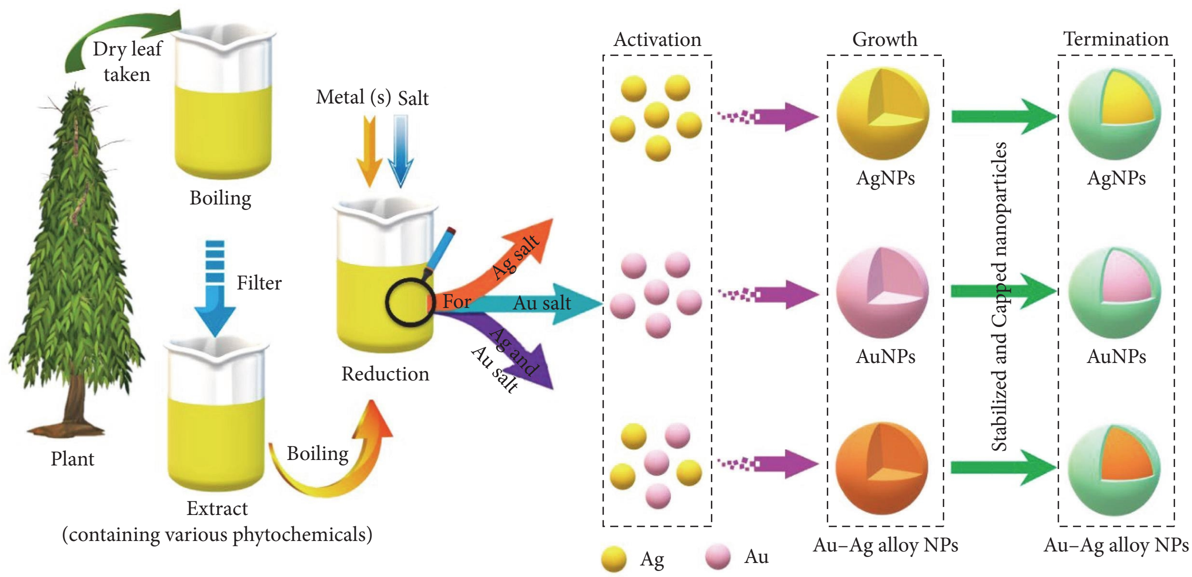

Figure 1: Schematic representation of the possible mechanism for biosynthesis of nanoparticles using leaf extract (source: [72]).

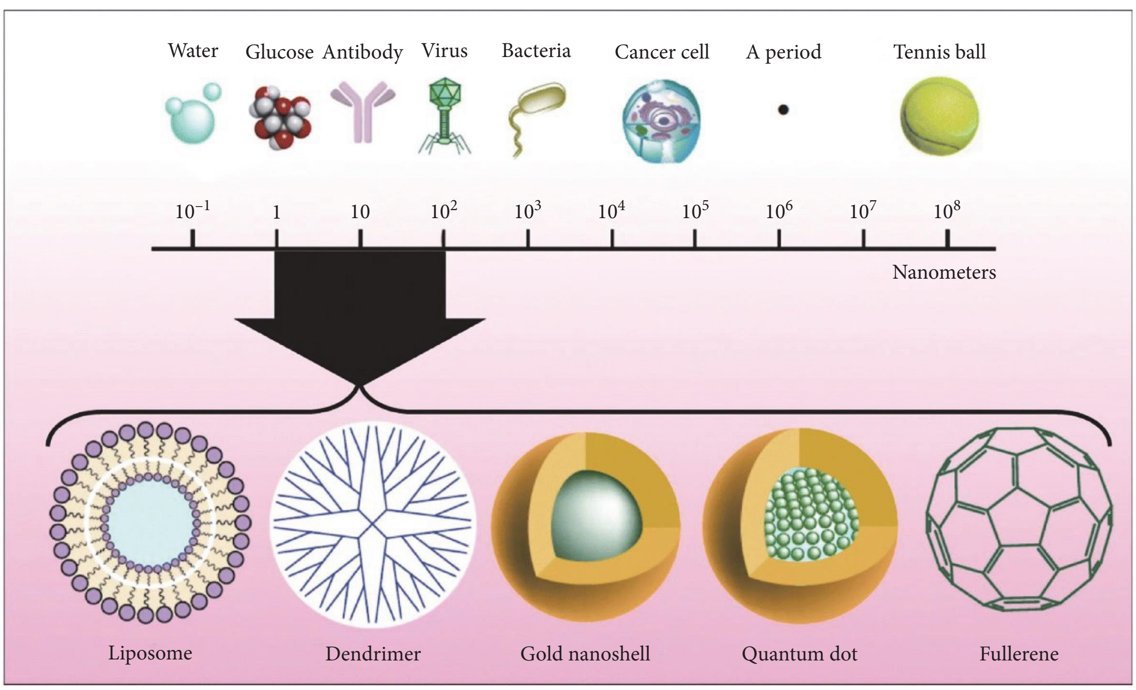

Figure 2: Various size of nanoparticles used in food science (reproduced with permission from [66]).

of two or more materials (nanocomposite). The NPs exist in different shapes viz. rod, spherical, triangular, cubic, hexagonal, star-like or other chain-like structures. One can simply identify the distinct infected targets, and treatment of particular cells or tissues rather than whole-body treatment without side effects is possible with NPs [77]. Over the last few decades, NPs are the most explored research area that improves every aspect of human life, science, and the economy as well as executes the growing needs of society and directly deals with environmental challenges. Among all the NPs, AuNPs and AgNPs are widely applied in cosmetic products, detergents, shoes, shampoos, soaps, and toothpaste, besides their applications in food, pharmaceutical, and medical commodities (Figure 2) [78].

4.1. Metallic Nanoparticles. In the last 20 years, continuous spreading of nanotechnology has been observed by development of various synthesis routes and stabilization methods of metallic NPs. Metallic NPs belong to a type of inorganic NPs (like AuNPs, AgNPs, etc.) that comprise the base composition of pure metal. Specifically, they are divided into monometallic, bimetallic, trimetallic, and polymetallic NPs depending upon the number of base metals. The bimetallic, trimetallic, and polymetallic NPs possess two, three, and more than three metal compositions in the NPs structure while monometallic NPs possess single-metal composition throughout the structure [79–81]. The high reactivity, high stability, paramagnetic behavior, unique optical characteristics, plasmonic properties, and quantum size effect of metallic NPs make them suitable for various potential applications in therapeutic procedures and bioimage diagnostic techniques [82–87]. However, the toxicity risk associated with the usage of metallic NPs in multidisciplinary applications is a major source of concern. So, this must be addressed by producing nontoxic NPs using particular methodologies involving synthesis methods and factors in order to improve quality of life. Figure 1 represents the many nanomaterials that are often employed is provided based on their size in relation to microorganisms or biomolecules that are similar in size to a nanomaterial.

4.1.1. Monometallic Nanoparticles. The monometallic NPs have fascinated scientists for last few decades due to its easy synthesis process, wide range of applications, as well as quite stable even in an adverse environment. As the name suggests, monometallic NPs contain only one metal atom that determines the properties of such type of NPs. Monometallic NPs are classified as metallic, transition, or magnetic NPs depending on the type of metal atom present [88, 89]. AuNPs and AgNPs are the most explored NPs among all monometallic NPs used in nanotechnology field, and they have broad range of significant applications such as in electronic, optical, antimicrobial activity, biomedical sciences, and as catalysts in overcoming environmental pollution [90, 91].

4.1.2. Gold Nanoparticles. The history and even the uses of gold (Au) have been well-known for several thousands of years, and gold is one of the oldest metals that has been discovered by humans. Although there are no definite evidence for the discovery of gold, the earliest use of gold in ornaments comes from the Indus Valley (Mohenjadaro, 3,000 BC), the Sumer civilization (southern Iraq, 3,000 BC), the tomb of King Tutankhamun (Egypt, 1,300 BC), and royal crowns from the Tilliatepe treasure (Scythian, 100 BC). In ancient India, the use of gold started during Vedic (1,000 BC–600 BC) period under the name of "Swarnabhasma" (means, gold ash), as ayurvedic medicine for revitalization and rejuvenation. Gold is the most precious metal across human civilizations for its attractiveness, inherent shiny property, and long-lasting glow for a long time made. It was suggested that gold has always been associated with the gods, eternity, wealth, and the sun (tears of the sun) [92–94]. The bulk-scaled matter of gold has usually been found to be an inactive matter, but nanosize of gold exhibits excellent activities. Among the various monometallic NPs, AuNPs possess some notable novel properties such as being chemically inert, surface plasmon resonance effect, and unique catalytic properties that leads to a key area of nanoresearch [95, 96]. Now a days, AuNPs are of great interest due to their exceptional biocompatibility and exclusive property to conjugate with proteins. AuNPs are expensive but they are widely used in material sciences due to easy production, solubility, low- or no-toxicity effect against human beings, and stability under atmospheric circumstances [97–100]. The effective application properties led to more research in modern science uses of gold NPs. Nanosensors based on AuNPs have been extensively utilized in the detection of metal contaminants (lead, mercury, chromium), pesticides, antibiotics, and dyes to ensure the quality of food products [101]. Apart from nonbiological adulteration, the AuNPs have been found to detect numerous food pathogens in the food. Because of their therapeutic potential, inert and nontoxic nature, and oxidative catalytic properties, AuNPs have piqued the interest of both the medical and food packaging sectors [102]. The colorimetric sensors based on AuNPs are simple, highly sensitive, and cheap and have been widely used in rapid testing and real-time on-site monitoring of food quality and safety [7].

4.1.3. Silver Nanoparticles. Since the ancient times, silver (Ag) has been equally well-known for domestic use, and it is extensively used in food storage, water storage, and wound healing (Greeks, Egyptians, Romans, and other ancient civilizations in 1,000 BC) due to their intrinsic antimicrobial properties and association with the moon due to their white and shining properties. Silver is still used for treating various diseases (respiratory disorders, memory enhancement, neuropsychological disorders) in the form of "RoupyaBhasma" (meaning silver ash) in the Indian ayurvedic and Unani medicine [21, 94]. Silver particles of nanoscale size tend to exhibit diverse physical and chemical properties than their bulk-scaled counterparts, though they are made from the same materials. AgNPs has created a center of concentration over the last few decades due to their antimicrobial properties in the protection of beverages and food for many years. They have enormous industrial applications due to their nontoxicity to human cells at low concentrations but are lethal for the majority of pathogenic bacteria and viruses [14, 103]. These properties make them suitable for a wide range of potential applications such as air sanitizer sprays, coatings of refrigerators, cosmetics, detergents, drug delivery, electronics, food packaging, management of insects in agriculture, medical devices, shampoos, soaps, textiles, toothpastes, vacuum cleaners, washing machines, wet wipes, water purification, and wound dressings [50, 94]. They have enormous industrial applications due to their well-recognized effective antimicrobial properties against pathogens and also it is nontoxic to human cells at low concentrations [104, 105]. Now a days, silver is used more than any other nanoscale materials for manufacturing consumer products. Biosynthesizing AgNPs using plant extracts has gained huge attention in recent decades due to the low cost of synthesis, environment-friendliness, and effective applications as food packaging materials. AgNPs have created a center of attention due to their impressive antimicrobial properties against a wide range of micro-organisms such as bacteria, viruses, yeasts, and fungi [106, 107]. Many studies have reported about the potential application of AgNPs in food packaging science for the protection of bread, beverage, orange juice, fish and meat, and fruits and vegetables. Besides shelf-life improvement of food, AgNPs cause no alteration in food's physical (freshness, color, odor, taste) appearances [106, 108].

4.1.4. Bimetallic Nanoparticles. The invention of hybrid NPs synthesis is a revolutionary stage in the nanoscience and nanotechnology field. As the name suggests, bimetallic NPs can be formed by incorporating two different metal elements in a single particle. Generally, bimetallic NPs can be classified into two types depending upon the mixing patterns between two different metal elements: first, there is a homogeneously mixed Alloy type structure where two different metal elements are mixed in either a statistical distribution pattern or atomically ordered into a common particle. Second, there is a core-shell type structure where one metal (core position) atom is encapsulated by a shell of another metal atom [20, 54, 109]. Bimetallic NPs attract more attention than corresponding monometallic NPs which overcome the limited properties of monometallic NPs. They are important because they usually show enhanced stability, selectivity, and activity compared with monometallic ones. The catalytic, electronic, optical, and thermal properties of the bimetallic NPs can be modified by simply varying the two metals' ratio as well as the geometrical structure of bimetallic NPs. Bimetallic NPs are more significant than monometallic NPs due to the existence of additional degrees of freedom. The new synergistic and bifunctional effects of two metal elements exhibit certain remarkable new properties which enhance their function and application in several different fields [12, 110, 111]. Amongmono- and bimetallic NPs, the bimetallic NPs composites consisting of both Au and Ag have recently become the focus of attraction of researchers as reported in the literature.

4.1.5. Gold–Silver Nanoparticles. Recently, the bimetallic gold–silver (Au–Ag) alloy NPs have drawn most interest than single-metal (monometallic) NPs due to their more effective nature. Apart from the monometallic particles, the use of bimetallic (Au–Ag) alloy NPs could be evidenced from the history when Lydian merchants invented the first coins called "electrum" through the use of (Au–Ag) alloys around 800 BC and also the famous Lycurgus cup (containing 30% Au and 70% Ag) of the fourth century Roman Empire [25]. Bimetallic (Au–Ag) alloy NPs have attracted astonishing attention from researchers for the control of its activity by variations in their molar ratio. In general, bimetallic NPs belong to two categories: alloy NPs (two kinds of metals are homogeneously mixed at atomic level) and core-shell structure NPs (two kinds of metals are heterogeneously mixed at atomic level). Indeed, it is well known that the surface plasmon resonance (SPR) band position and intensity for bimetallic NPs controlled by the ratio of precursor composition or shell thickness. Bimetallic (Au–Ag) alloy NPs exhibit a single SPR with an intermediate position among the SPR band position of monometallic Ag and Au NPs but bimetallic (Au@Ag) core-shell structure NPs show two distinct SPR bands at the position of monometallic Ag and Au NPs [112, 113]. Particularly, in the catalytic reaction process, bimetallic NPs often exhibit higher catalytic activity and selectivity than their monometallic counterparts due to combining advantages (complex structure) of two individual metals [42, 58]. Bimetallic (Au–Ag) alloy NPs has received extensive progress in the field of food safety, biomolecular recognition, biosensing, optical studies, molecular imaging, delivery of drug, medical diagnostics, catalytic studies, etc. The bimetallic Au–Ag NPs provide versatility with respect to their functional properties which is attributed to the combining synergistic effects of two distinct metal atoms [48, 49]. In this regard, it is extremely interesting to take advantage of both (Au and Ag) metals and to produce bimetallic Au–Ag NPs. With the help of nanotechnology, the shelf life of foods can be increased and the extent of food spoilage can be decreased, as finally healthy food can reach the masses and eventually it will improve the health of the people and can aid in reducing the problem of food shortage [114]. Several forms of "nanosystems" such as solid nanoparticles, nanofibers, nanocapsules, nanotubes, nanocomposites, nanosensors, and nanobarcodes are a few of the major nanomaterials that find their use in food processing, packaging, and preservation sectors [115, 116].

5. Applications of Nanoparticles in Food Packaging and Food Quality Analysis

The rapid growth of nanotechnology in recent years has piqued the interest of researchers in a variety of sectors, particularly in food science. Due to increased customer demand for higher food quality without compromising the nutritious element of the foods, nanotechnology has a complete food solution from food manufacturing, processing, and packaging as well as food quality sensing and safety. In particular, nanotechnology has a total food science solution from food manufacturing to processing to packaging [117, 118]. In this context, the use of nanotechnology in the food sector can be summarized mainly in two groups: food quality sensing and food nanostructured ingredients. The former uses as a biosensor or sensing the contaminants (i.e., heavy metals, pesticides, antibiotics, microorganisms, allergenic compounds, food additives, detection of genetically modified foods, and toxins) to evaluate food safety and achieve better food quality. While the latter includes food manufacturing, processing, and packaging by inserting nanomaterials into the packaging structure [119, 120]. In food science, nanomaterials can be employed as food additives, anticaking agents, antimicrobial agents, carriers for smart delivery of nutrients, the durability of packaging materials, fillers for improving mechanical strength, etc. [7, 121]. Indeed, Au and Ag are the most stable and reliable metals that are extensively used due to their lack of reactivity [122, 123]. In this section, our focus is on food quality analysis and packaging applications relevant to food quality and safety monitoring (Figure 3).

5.1. Nanoparticles in Food Packaging. Nanomaterials of various dimensions and sizes offer enormous potential for use in the food manufacturing, processing, packing, and safety of high-quality agrifood. The usage of nanocomposites as an active material for coating and packaging can also be utilized to improve protective packaging. Nanomaterials-based "active" (having antioxidative, antibacterial, and UV absorption properties) and "smart" (having controlled/monitored food conditions) food packaging [126, 127]. It has several advantages over traditional packaging materials, including antimicrobial protection, barrier properties, improved mechanical and thermal strength, as well as

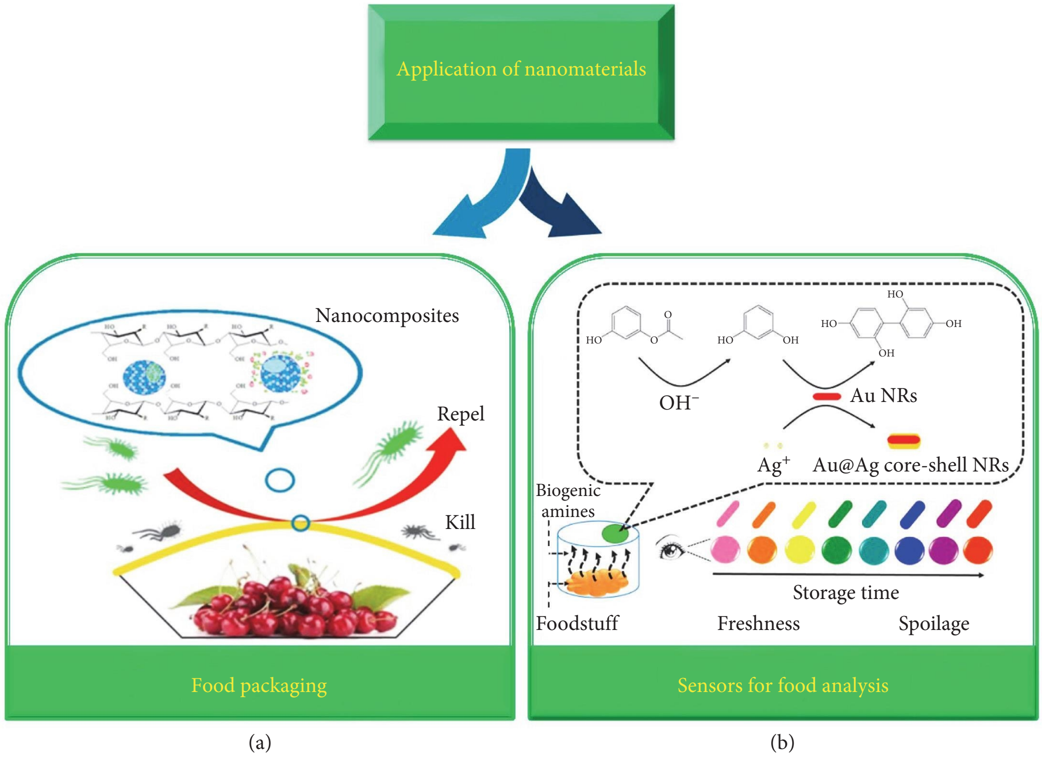

FIGURE 3: Application of nanomaterials in food packaging (a) and nanosensors for food analysis (b). This figure was modified and adopted with permission from [124, 125].

protection from oxygen and other environmental factors. Moreover, it increases the shelf-life of food items by slowing down or preventing deterioration until the product is delivered to the target place [128, 129]. Furthermore, edible nanocoatings on various food components could prevent gas exchange and moisture to keep their original colors, textures, antioxidants, tastes, antibrowning agents, and enzymes activity, as well as increase the shelf-life of produced meals even after the packaging is opened [130, 131]. Many scientists were keen to explore the antimicrobial capabilities of organic materials such as organic acids, essential oils, and bacteriocins, as well as their usage in polymeric materials as antimicrobial packaging. Though, these substances do not fit into the various food processing procedures that demand high pressures and temperatures since they are very sensitive to these environmental conditions [132–135]. Inorganic NPs are capable of providing significant antibacterial activity in low concentrations, mechanical improvement, and also greater stability under harsh circumstances [136, 137]. As a result, there has been a surge of interest in employing these inorganic NPs in food packaging in recent years. Many nanoparticles have been found to exhibit antibacterial properties, including Ag, Au, and its bimetallic Au–Ag metal NPs. The use of Ag, Au, and Au–Ag nanomaterials has shown exciting potential in food packaging of food industry [138, 139], some of them are summarized in Table 2.

5.2. Nanoparticles for Food Quality Analysis. Nantechnology is emerging innovative methods for use in the construction of sensors assuring food safety, extending the shelf life of food, and maintaining food quality. In food science, nanobiosensors or nanosensors are applied for the quantification of existing food constituents, detection of pathogens in processing in food material or plants, and indicating whether the food is fit to consume or not [152, 153]. The functions of nanosensor as an indicator that responds to changes by different environmental reasons such as chemical or microbial contamination, temperature or humidity in storage rooms, pH, or product deterioration [154, 155]. Several nanostructures, including thin films, NPs, nanorods, nanofibers, and nanotubes have been investigated for potential applications as biosensors. Thin film-based optical immunosensors are one of the rapid and highly sensitive detection systems for the determination of cells or micro-organisms [154, 155]. Specific proteins, antibodies, or antigen substances are immobilized on thin nanofilms or sensor chips in these immunosensors that emit signals when target molecules are detected [156]. Nanotechnology can also help to identify heavy metals, pesticides, antibiotics, allergenic compounds, food additives, detection of genetically modified foods, and toxins in the food quality [157]. Among all NPs, AgNPs are one of the most commercially produced NPs due to their antimicrobial activity, whereas AuNPs are extensively studied as a detector/sensor [158, 159]. The recent progress of Ag, Au, and Au–Ag nanomaterials has been briefly discussed in Table 2.

6. Safety Aspects

It is increasingly clear that nanotechnology has a wide range of benefits and has the potential to revolutionize the food industry. The use of nanomaterials in the food industry,

Table 2: Applications of nanomaterials in food packaging and food-quality sensing.

Nanomaterials	Nanocomposites/package materials	Function	Food tested	Application	References
AgNPs	Cellulose films with amino terminated hyperbranched polyamic	Antibacterial, antioxidant	Cherry and tomatoes	Food packaging	[140]
AuNPs	Gelatin	Color change TTI	Cherry and tomatoes	–	[141]
Au/Ag NRs	Agar hydrogel	Color change TTI	Cherry and tomatoes	–	[142]
AgNPs	AgNPs-cellulose	–	–	Antimicrobial food packaging	[143]
AgNPs	AgNPs-polyurethane	*S. aureus* and *E. coli*	Lettuce	Fruit preservation	[144]
AgNPs	AgNPs-pullulan	*L. monocytogenes* and *S. aureus*	Poultry products and meat	Packaging material	[145]
AgNPs	AgNPs-LDPE	Aerobic bacteria	Barberry	Preserves freshness of food during extended storage	[146]
AgNPs	AgNPs-cellulose	Psychotropic bacteria, aerobic bacteria, yeasts, and and molds	Freshly-cut melon	Antimicrobial food packaging	[101]
AgNPs	AgNPs-polyvinyl chloride	–	Wheat bread and red grapes	–	[102]
AuNPs	AuNPs-based alginate plasmonic THI	Irreversible change in color that indicate variation in temperature	Perishable foods	Tunable nanosensor	[7]
AgNPs	AgNPs/GNRs-based electrochemical sensor	Detection of methyl parathion	Fruits and vegetables	Nanosensor	[147]
AuNPs	AuNPs-based glassy electrode	Check of the freshness of food sample	Canned tuna	Nanosensors for xanthine and hypoxanthine	[148]
AuNPs	–	Pathogens (*E. coli* and salmonella spp.)	Cucumber and hamburger extracts	Optical sensor	[149]
AuNPs	AuNPs loaded on MWCNT	Toxins (Bisphenol A)	Soft drinks	Electrochemical sensor	[99]
Au@Ag NRs	–	Hydrogen peroxide (H_2O_2)	Chicken claw	Colorimetric sensor	[150]
Au−Ag alloy NRs	Dual enzyme-induced Au −Ag alloy nanorods	Determination of *Staphylococcus aureus*	Milk	Colorimetric sensor	[151]
Au@Ag NPs	Double strand DNA binding bimetallic Au@Ag NPs	Detection for veterinary antibiotics (kanamycin)	Milk	Optical sensor	[106]
Au@Ag NRs	–	Detection for benzoyl peroxide	Milk	Colorimetric sensor	[107]

NRs, nanorods; MWCNT, multiwalled carbon nanotubes.

farming, cosmetics, and personal healthcare systems could lead to the migration of nanomaterials to the environment and subsequent human exposure through inhalation and skin penetration [160, 161]. The possible routes of exposure could occur that includes: (i) ingestion from intake of nanofood, (ii) leaching of nanomaterials from sensing elements or packaging materials into the food, and (iii) disposal of nanosensors, nanofood, packaging or buried in landfills; further release into the air, water, and soil and reach humans, plants, or wildlife (Figure 4). Ultimately, there is a high chance of risk that nanomaterials residues will end up in the human food chain. Several scientists explored the risks associated with nanomaterials, with particular emphasis on the chance of NPs migrating from packaging material into food and their effect on consumers' body [163–165]. For example, the European Food Safety Authority (EFSA) published a scientific opinion in May 2021 declaring that TiO_2 can no longer be considered safe when used as a safe food additive [166]. However, Garcia et al. [167] reviewed the applications and migration of NPs such as aluminum oxide (AlOx), silicon dioxide/silica (SiO_2), titanium dioxide (TiO_2), and zinc oxide (ZnO) used in food packaging to improve antimicrobial, light-blocking, gas barrier, thermal, and mechanical properties of nanocomposite. Based on existing literature, it was concluded that only a minute number of nanomaterials migrate from food packaging materials into food products and risk of migration could be minimized by applying an extra barrier between the food and the nanocomposite [168–171]. Several researchers studied the toxicity of dietary titanium dioxide (TiO_2; E171) and they raised some concerns about its possible tumor-promoting action [172–174]. Before the use of any nanomaterial in food industries, its toxicity must

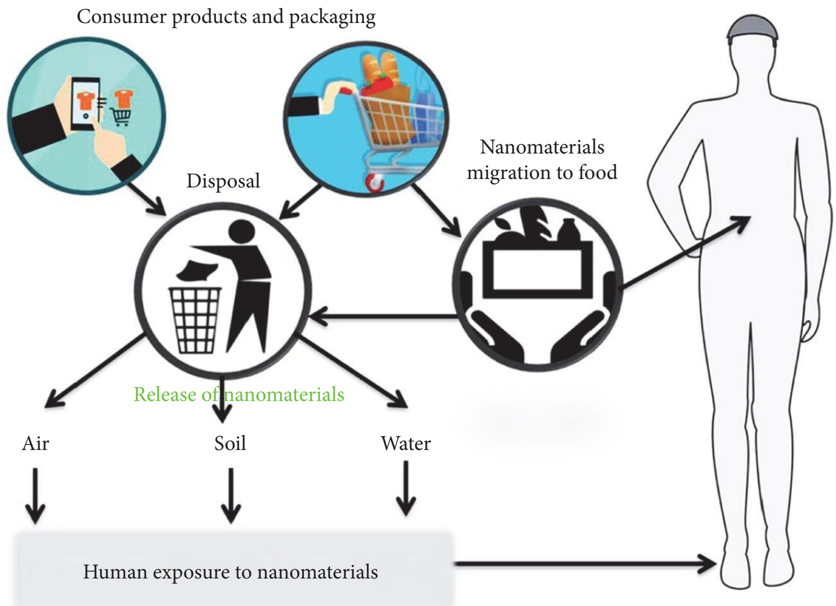

Figure 4: Routes of possible exposure of nanoparticles from agrifood and consumer products into the environment and humans (reproduced from [162] with permission from the Royal Society of Chemistry).

be thoroughly investigated to assure its safety for human health as well as the environment. Nanomaterials may pose a greater risk of bioaccumulation within human organs and tissues due to their small size and also depends on the uptake by the organisms. In addition, nanomaterials' behavior and fate are dependent on their various properties such as concentration of the particles, shape, surface morphology, chemical composition, stability, reactivity, aggregation, phase purity, solubility, surface energy, crystallinity, homogeneity, and bioavailability of nanomaterials in different media [175–177]. Moreover, the properties of nanomaterials are dependent on environmental factors such as temperature, pH, concentration, ionic strength, and composition of natural organic matter that effects their stabilization and aggregation. So, the toxicity should likely be determined on a case-by-case basis as each nanomaterial has a unique feature [178, 179]. Moreover, regulatory agencies must make some standards for commercial products to assure product safety, quality, and health and environmental regulations.

7. Conclusion

The traditional physical, chemical, and microbiological methods for creating NPs are not environment friendly. They have a number of drawbacks, including the need for specifically developed equipment, templates, and extremely high temperatures and pressure. The current review paper provides an environmentally acceptable way to manufacture biogenic nanoparticles from natural plant extracts. Plant parts rich in flavonoids, phenols, steroids, terpenoids, enzymes, and alkaloids, such as leaves, stems, barks, fruit, and flowers, play an important role in reducing and stabilizing metal ions that create metallic NPs. The mechanisms involved in their synthesis, growth, and stabilization involved in biosynthesis of monometallic (AgNPs and AuNPs) and its bimetallic (Au–Ag) NPs are discussed. The phytosynthesized nanoparticles have been extensively used in food packaging and food quality analysis to enhance the shelf-life of foods. The incorporation of bio-based monometallic and bimetallic Ag and Au nanoparticles in packaging materials can help to improve the food shelf life, as well as they can be used biosensors to monitor the real-time food product quality. Although the use of antimicrobial nanoparticles in the food sector is promising still the release of nanoparticles in food and related toxicity is a challenge, which needs to be addressed in near future to ensure complete food safety.

Abbreviations

NPs:	Nanoparticles
NRs:	Nanorods
AgNPs:	Silver nanoparticles
AuNPs:	Gold nanoparticles
Au–Ag:	Gold–silver nanoparticles
MWCNT:	Multiwalled carbon nanotubes
AAS:	Atomic adsorption spectroscopy
AFM:	Atomic force microscope
DLS:	Dynamic light scattering
EDX:	Energy dispersive X-ray spectroscopy
FTIR:	Fourier transform infrared spectroscopy
ICP-MS:	Inductively coupled plasma-MS
MS:	Mass spectroscopy
SEM:	Scanning electron microscope
TEM:	Transmission electron microscopy
HR-TEM:	High-resolution transmission electron microscope
TGA:	Thermogravimetric analysis
UV–vis:	Ultraviolet–visible
XRD:	X-ray diffraction.

Data Availability

Data used in this article are available in the form of tables and figures.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

Supplementary Materials

Table S1: Factors affecting plant extracts mediated biosynthesis of nanoparticles. Table S2: Techniques used for characterization of NPs. (*Supplementary Materials*)

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