JMBFS

Journal of Microbiology, Biotechnology and Food Sciences

International peer-reviewed scientific online journal

Published by Faculty of Biotechnology and Food Sciences

BIONANOTECHNOLOGIES: SYNTHESIS OF METALS' NANOPARTICLES WITH USING PLANTS AND THEIR APPLICATIONS IN THE FOOD INDUSTRY: A REVIEW

Svitlana Tsekhmistrenko[1], *Vladimir Bityutskyy*[1], *Oksana Tsekhmistrenko*[1], *Serhii Merzlo* [1], *Natalia Tymoshok*[2], *Alexander Melnichenko*[1], *Svitlana Polishcuk*[1], *Alexander Demchenko*[2], *Igor Yakymenko*[3]

Address(es):
[1]Bila Tserkva National Agrarian University, Department of Biotechnology, 8/1, Soborna Sq., 09117 Bila Tserkva, Ukraine, +38(068) 034 48 48.
[2]Zabolotny Institute of Microbiology and Virology of NAS of Ukraine, Kyiv.
[3]National University of Food Technologies, Kyiv, Ukraine.

*Corresponding author: Svetlana.tsehmistrenko@gmail.com

https://doi.org/10.15414/jmbfs.1513

ARTICLE INFO

Received 3. 4. 2019
Revised 17. 5. 2019
Accepted 28. 5. 2021
Published 1. 6. 2021

Regular article

OPEN ACCESS

ABSTRACT

The basic principles of bionanotechnology and modern "green" chemistry are considered. The methods of synthesizing nanoparticles such as "top-down" and "bottom-up" are analyzed. The advantages of "green" synthesis of nanoparticles from plant raw materials with unique properties and a wide range of applications are presented. There are systematized factors that influence the biosynthesis of nanoparticles: the nature of the plant extract, the pH of the reaction mixture, the temperature of the incubation, the reaction time, the concentration and the electrochemical potential of the metal ion. The prospect of using nanotechnological approaches for the whole chain of agricultural production, as well as in the food industry, which will promote the creation of new nanomaterials, is shown. Nanotechnologies are successfully used to ensure the quality of food, enrichment of food products with minerals, vitamins, antioxidants, improvement of organoleptic properties, prolongation of life and antimicrobial packaging of food products. The focus is on the biosafety of nanomaterials whose properties are multifaceted, ambiguous and require a comprehensive, safe, responsible and scientifically sound approach.

Keywords: bionanotechnologies, "green" synthesis, metals' nanoparticles, food industry, biosecurity of nanomaterials

INTRODUCTION

Over the past decades there has been a rapid increase in the number of nanotechnology research. Nanotechnology is an interdisciplinary science that deals with the synthesis of nanomaterials smaller than 100 nm (**Zhiharev and Ljashenko, 2007**). Bionatechnology combines biological principles with physical and chemical approaches to obtain nanosized functional particles. New unique properties, including catalytic, electrical, optical, mechanical and electromagnetic properties, have contributed to the production of nanoproducts that have been used in several sectors, such as electronics, medical diagnostics, therapy, agriculture and the food industry. There are various chemical and physical methods used to synthesize nanoparticles. These methods require special expensive equipment and the use of toxic reagents that are not acceptable for large-scale synthesis (**Prasanna *et al.*, 2019**). Biosynthetic or "green" methods play an extremely important role in nanotechnology, since they are simple, expressive, cost-effective and environmentally friendly (**Herrmann *et al.*, 2007**). The introduction of the concepts of "green chemistry" and "nanotechnologies" is one of the revolutionary events in science, which has contributed to the research on environmental safety and the reduction of the size of objects. The unification of these two areas paved the way for a new green and nanoscale oriented science called "green nanotechnology" or bionahnotechnology (**Das *et al.*, 2017; Thakur *et al.*, 2018**). The twelve "principles of green chemistry" are actively suggesting looking for green options for nanoproducts (**Das *et al.*, 2017**). The principles of green chemistry are a philosophy that is applied in all areas of chemistry, and not just in one chemical discipline, and is aimed at preventing contamination at the molecular level. These principles provide for the application of innovative scientific decisions, which leads to a reduction in the formation of hazardous substances, since it prevents the formation of contaminants. It reduces the negative impact of chemical products and processes on human health and the environment, reduces and even in some cases eliminates the danger from already existing products and processes (**Anastas and Eghbali, 2010**).
Metals' nanoparticles are one important and widely studied group of materials that show a great deal of diversity and many different uses. The ecological purity of the production of metals' nanoparticles, which application grows at a significant pace, is an urgent problem of the present. The methods of "green" synthesis are environmentally friendly, since organisms have been adapted to survive in environments that contain high levels of metals, developing mechanisms to cope with them. These mechanisms may include the change in the chemical nature of the toxic metal, so that it no longer causes toxicity. That leads to the formation of nanoparticles of the corresponding metal. Thus, the formation of nanoparticles is a "by-product" of the resistance mechanism against a particular metal, and this can be used as an alternative way of obtaining them (**Pantidos and Horsfall, 2014**).
Nanocomposites are currently being introduced into commercial products at a faster pace than the development of knowledge and regulations to reduce the potential health and environmental impacts associated with their production, use and disposal (**Thorley and Tetley 2013**). Since nanomaterials find new applications on a daily basis, their potential toxic effects should be monitored (**Jain et al 2016**). It is necessary to study the influence of nanoparticles on the human body and animals due to their characteristics, namely the method of preparation, size, form, reactivity and other characteristics (**Spivak et al 2013; Navya and Daima 2016**). After all, it is often unknown how the new materials what are being developed and that appear on commercial markets, after their entry into the environment, will behave. It is important to provide regulatory means to avoid potential risks associated with the continued development and use of nanomaterials, as well as the future development of new materials.

Methods of synthesizing of metals' nanoparticles

Methods of synthesizing of nanoparticles (NP), including metals nanoparticles (NPm), are improved, new ones are appearing. All methods are aimed at obtaining stable systems, since NPs are characterized by high surface energy and are prone to the formation of large aggregates. Morphology (size and shapes) and properties of nanoparticles depend on the method of synthesis and conduct conditions (**Kharissova *et al.*, 2013**).
During the last decade, innovative approaches to the synthesis of various nanomaterials such as nanoparticles of metals, quantum dots, carbon nanotubes (fullerenes, graphene, nanotubes) and their composites have appeared in the field

of nanotechnology (**Kim *et al.*, 2007; Laurent *et al.*, 2008; Oskam, 2006; Prasanna *et al.*, 2019; Su *et al.*, 2014**). To obtain nanomaterials of the required size, shape and functionality in the existing literature, two different principles of synthesis have been investigated. There are "top-down" and "bottom-up" methods (Fig. 1). Top-down synthesis methods are physical methods of grinding, dispersing volumetric macroforms of metals to nanoparticles using high temperatures, pressure and vacuum (**Guozhong, 2004**). These methods require complex equipment, are expensive and do not allow adjustment of the size, geometry and stability of nanoparticles of metals (NPm). "Bottom-up" methods are chemical and physico-chemical based on the restoration of metal cations to neutral metal atoms, which unilaterally aggregate to nanosized clusters - NPm (**Ge *et al.*, 2013**). Currently it is important to transfer of metal ions to zero-valence atoms, and then maintaining the bottom-up synthesis approach up from the bottom (**Das *et al.*, 2017**). In some cases, depending on the "bottom-up" methods of synthesis and their conditions, it is possible to achieve a state close to monodisperse and its high stability. In some cases, depending on the methods of synthesis "bottom-up", its conditions, it can achieve a state close to monodisperse and its high stability.

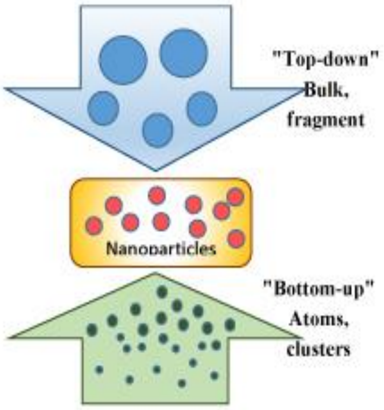

Figure 1 "Top-down" and "bottom-up" synthesis of nanoparticles

Today, the use of nanotechnology advances allows for the production of many polyfunctional materials, but they have shortcomings, such as the use of hazardous chemicals such as sodium borohydride ($NaBH_4$) and trinatrium citrate ($Na_3C_6H_5O_7$), complex and consumable methods and the lack of scalability of synthesis processes (**Das *et al.*, 2017**) To evaluate the characteristics of the NPs, their dimensions, size distribution (polydispersity), particle geometry, physical properties (optical, electrical, magnetic, thermal conductivity, etc.) (**Jamdagni et al., 2018; Sumitha *et al.*, 2019**) are taken into account.

The study of synthesized nanoparticles characteristics is carried out using UV-Vis spectroscopy, infrared Fourier spectroscopy and fluorescence spectroscopy, transmission and scanning electron microscopy, X-ray diffraction, cyclic voltammetry (**Thomas *et al.*, 2019).**

However, despite the widespread use of traditional nanoparticle technology, they tend to be expensive, labor-intensive, associated with the risk and potential danger to the environment and living organisms (**Santhoshkumar *et al.*, 2017**). Thus, there is a clear need for alternative economically feasible and at the same time safe and environmentally friendly methods of nanoparticles production. In order to eliminate the disadvantages of physical and chemical methods, recently bionahnotechnologies ("green" nanotechnologies) are intensively developing (**Chauhan *et al.*, 2019; Khan *et al.*, 2017**). "Green" synthesis is an environmentally friendly alternative to traditional synthesis methods and aims at avoiding or minimizing toxic components what are used or produced by physical and chemical methods and are able to compete successfully with them in terms of speed, controllability, bioconversion and reduced cost of the final product (**Kharissova *et al.*, 2013; Pal *et al.*, 2019; Iravani, 2011**).

The advantage of nanostructures synthesized by the "green" approach lies in the fact that bio-objects are used as biofacies contain a large variety of molecules. At the same time they are restoring and stabilizing the surface of synthesized NPs, thus forming coating layers that additionally provide stability and biocompatibility with "green" NPs (**Khatami *et al.*, 2018**). Although one of the problems encountered by most of the physically and chemically synthesized NPs is aggregation. Biomolecules what cover the surface of the "green" synthesized NPs help to avoid aggregation through the existing coating layer (**Khan *et al.*, 2017; Ahmad *et al.*, 2015; Pal *et al.*, 2019**).

There is a large variety of plants in nature, what causes a wide selection of reducing agents able to serve as a material for the synthesis of metals' nanoparticles (**Pal *et al.*, 2019**). Multicellular and single-celled organisms, due to their ability to accumulate metals, are used for the synthesis of nanoparticles (**Sardar and Mazumder, 2019; Singh *et al.*, 2019**). During the last decade, it has been shown that diverse biological systems, including plants and algae (**Das *et al.*, 2017; Iravani *et al.*, 2011; Roseline *et al.*, 2019**), bacteria (**Shahverdi *et al.*, 2007; Pantidos and Horsfall, 2014**), yeast (**Lin *et al.*, 2005**), mushrooms (**Narayanan and Saktiivel, 2011b; Chhipa, 2019**) and viruses (**Shenton *et al.*, 1999; Lee *et al.*, 2002**) can convert ions of inorganic metals into metallic nanoparticles due to the recovery process carried out by proteins, enzymes, and metabolites contained in these organisms.

The resulting particles are very promising for biological applications due to their biocompatible nature. Microorganisms in the presence of high metal concentrations have developed specific evolutionary adaptation mechanisms to reduce their toxicity. These mechanisms include the change in the chemical nature of the metal, what leads to the formation of nanoparticles as "by-product" of the adaptive protection mechanism. Such "green" synthesis can be used as an alternative to traditional technologies for the generation of first-generation nanoparticles based on physical and chemical methods (**Pantidos and Horsfall, 2014**).

It is significant that the production of nanoparticles using plants described in this review has several advantages. There are the availability of cheap material, low toxicity, simplicity, short production times, safety, the ability to regulate the required volume of production, and the suitability for large-scale production among those advantages (**Sanjay, 2019**). For in vitro biosynthesis, the use of redoxactive components of secondary metabolites of plants is used. In this case, the initiation of the origin of nanoparticles is due to the restoration of metal ions due to reduction compounds / phytochemicals present in tissue extracts. The nuclei are further enhanced by adsorption of metal atoms after recovery, which results in the formation of metals nanoparticles (**Rajeshkumar and Bharath, 2017**). The components of living cells, such as carbohydrates, fats, proteins, enzymes, flavonoids, terpenoids, polyphenols and alkaloids, can associate metal ions with nanoparticles (**Rautela *et al.*, 2019**).

The biosynthesis of nanoparticles can be extracellular and intracellular (**Iravani, 2014; Santhoshkumar *et al.*, 2017**).

Regardless of the nature of the reducing agent, the synthesis of NPs occurs in a series of sequential reactions and interactions, resulting in a balance in the polydisperse system. The following main phases of the "green" synthesis of NPs are singled out: activation (recovery of metal ions (Mn^{n+}) and formation of zero-valent atoms (Me^0); nucleation of neutral metal atoms with the formation of proton-particles; increasing (aggregation) of smaller particles formed at the nucleation stage and aggregation into larger agglomerates (**Makarov *et al.*, 2014**).

Intracellular methods for the synthesis of nanoparticles include the cultivation of plants in rich metal organic media (**Gardea-Torresdey *et al.*, 2002**), in rich soil soils (**Haverkamp *et al.*, 2007**), as well as in metal-enriched hydroponic solutions (**Harris and Bali, 2008**). The biosorption of nanoparticles of metals is restored on the periplasmic membrane. Recovery of Pd^{2+} to Pd^0 in the periplasmic membrane was shown (**Yong *et al.*, 2002**). An enzyme hydrogenase is located on the membrane, what serves as a nucleation site and contributes to the initial growth of Pd nanoparticles, possibly by transporting electrons to recover Pd (II).

Non-cellular methods include the synthesis of nanoparticles using an extract of various biological tissues that are prepared in various ways (**Pal *et al.*, 2019**).

In the extracellular process, the ions are restored with the help of proteins, enzymes and organic molecules in the medium or components of the cell wall. Many organisms have the ability to use the mechanisms of metal recovery, which are synchronously related to the oxidation of enzymes, especially reductase (**Deplanche *et al.*, 2010**). This leads to the formation of stable and inert metal nanoparticles, which can then be safely removed from the contaminated sample. Extracellular synthesis is more cost-effective than intracellular, because of its lower cost, more simple extraction technology and higher efficacy (**Deplanche *et al.*, 2010; Fang *et al.*, 2019**).

Methods of "green" chemistry have been actively developed in recent years as an alternative, effective, cheap and environmentally safe method for obtaining polyfunctional nanoparticles with given properties. The priority in green synthesis is the use of non-toxic plants with medicinal properties, which is important for their further use in biology, medicine and food industry.

The overall progress achieved in bionahnotechnology is remarkable, and most importantly, its environmental action has made the "green" synthesis a more generalized and attractive alternative to traditional methods for the synthesis of nanoparticles (**Das *et al.*, 2017**).

Factors influencing the "green" synthesis of metals' nanoparticles in plants

Synthesis of metals' nanoparticles with the help of plants is environmentally friendly, inaccurate and not expensive and refers to the "green" synthesis. Plants are a good raw material for the "green" synthesis of nanoparticles that have a wide range of applications in various industries (**Kumar *et al.*, 2019a; Pal *et al.*, 2019**).

The process of restoring metals' ions to form nanoparticles depends on a large number of factors (**Kumar *et al.*, 2019a**). The nature of the plant extract, the pH of the reaction mixture, the incubation temperature, the reaction time, the concentration, and the electrochemical potential of the metal ion are influenced by the nature of the nanoparticles (Fig. 2) (**Raveendran *et al.*, 2003; Haverkamp and Marshall, 2009; Selvakannan *et al.*, 2004; Willett *et al.*, 2005; Yadi *et al.*, 2018**). The relationship between the applied parameters, in particular the concentration of the plant extract, the concentration of metal ions, the reaction time and temperature with the shape and size of the nanoparticles of the metals obtained (**Mishra *et al.*, 2014**), have been revealed.

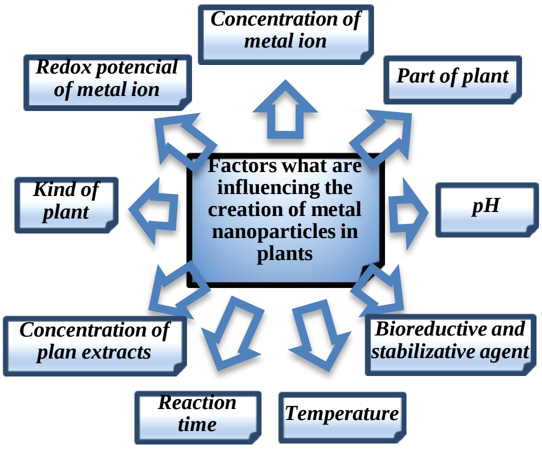

Figure 2 Factors influencing the "green" synthesis of nanoparticles of metals from plant raw materials

The content of active biomolecules, their combination and concentrations are due to the nature of the plant extract. The size and shape of the nanoparticles play a decisive role in determining the overall biocompatibility (**Alkilany and Murphy, 2010**). For their biomedical applications, nanoparticles with a size up to 100 nm are the most suitable. It was found that Ag nanoparticles could be obtained from different plants and their parts, while the sizes of nanoparticles vary. Thus, Ag particles with size 90 nm were obtained from the leaves of *Digitaria radicosa* (**Naraginti *et al.*, 2016**), 11-100 nm from *Elephantopus scaber* (**Kharat and Mendhulkar, 2016**), - 20-80 nm from *Butea monosperma* (**Patra *et al.*, 2015**), 25 nm with *Thymus serpyllum* (**Erci and Torlak, 2019**). The size of the silver particles obtained from the *Diospyros paniculata* root is 14-28 nm (**Rao *et al.*, 2016**), *Acorous calamus* particles are of an average size of 31 nm (**Naraginti *et al.*, 2016**), from Rhizome *Curculigo orchioides* - 15-18 nm (**Kayalvizhi *et al.*, 2016**), from fruits of *Emblica officinalis* 10-70 nm (**Ramesh *et al.*, 2015**), from *Tectona grandis* seeds - 10-30 nm (**Rautela *et al* ., 2019**), from *Durio zibethinus* peel - 20-60 nm (**Sumitha *et al.*, 2019**), from saffron waste (*Crocus sativus L.*) 12-20 nm (**Bagherzade *et al.*, 2017**).

Gold nanoparticles can be of different shapes - spherical, cylindrical, cubic, triangular, and different sizes, depending on the nature of the raw material (**Santhoshkumar *et al.*, 2017**). Thus, from *Carica papaya* leaves nanoparticles of size ranging from 15-28 nm were obtained (**Muthukumar *et al.*, 2016**), from *Hibiscus sabdariffa* - 10-60 nm (**Mishra *et al.*, 2016**), from *Hygrophila spinosa* - 50-80 nm (**Koperuncholan *et al.*, 2015**). Gold nanoparticles from *Ocimum sanctum* were characterized by different sizes (1-50, 10-300, 50-300,> 200) (**Lee *et al.*, 2016**). An inexpensive, fast and environmentally friendly approach to the synthesis of AuNPs using *A. Rosea* leaf extract is proposed. *A. Rosea* leaves extract can synthesize triangular, pentagonal, hexagonal and spherical AuNPs (**Khoshnamvand *et al.*, 2019**). Gold nanoparticles derived from banana extract using banana pith extract had antibacterial activity against bacteria as gram-positive (*Bacillus subtilis*) and gram-negative (*E.coli, Pseudomonas aeruginosa*) (**Nayak *et al.*, 2018**).

The formation of nanoparticles is influenced by the presence of compounds involving with bioreduction and stabilization (**Dauthal and Mukhopadhyay 2016; Ahmed *et al.*, 2014; Kumar *et al.*, 2019a; Khedri *et al.*, 2018**). Various photochemical compounds in plant extracts that are involved in the bioreduction of metals' ions act through their synergistic activity. Various photochemical compounds in plant extracts that are involved in the bioreduction of metals' ions act through their synergistic activity (**Shankar *et al.*, 2004**). Catechins, Teaflavins and Arabugins are required for the formation of gold nanoparticles from *Camellia sinensis* (**Nune *et al.*, 2009; Alshatwi *et al.*, 2015**). Similarly, the separation and purification of azadiachratine tetraenotriterpenoide confirmed the role of *Azadirachta indica* like regenerative and stabilized agent of Au and Ag nanoparticles in GS (**Shukla *et al.*, 2012**). *Jatropha curcas*, curcain enzyme and the cyclic octapeptide (Curcacycline A and B) play a role in the bioreduction and stabilization of AgNPs synthesized by latex (**Bar *et al.*, 2009**). According to the "recognition-restriction-limited nucleation and growth" model, in the case of the synthesis of AgNPs, silver-ions were captured on the surface of the protein through electrostatic interaction. (**Li *et al.*, 2007**). Other researchers deny the involvement of enzymes in the recovery of metal ions, since the herbal extract was heated (**Narayanan and Saktiivel 2011a; Song *et al.*, 2010**). According to the "recognition-restriction-limited nucleation and growth" model, in the case of the synthesis of AgNPs, silver-ions were captured on the surface of the protein through electrostatic interaction. (**Li et *al.*, 2007**). Other researchers deny the involvement of enzymes in the recovery of metal ions, since the herbal extract was heated (**Narayanan and Saktiivel 2011a; Song *et al.*, 2010**).

Polyphenol is used for green synthesis of Pt nanoparticles (**Alshatwi *et al.*, 2015**), and epicatechin and quercetin-glucuronide were used for Fe_2O_3NPs (**Wang *et al.*, 2014**). Common phytochemical components, such as phenols, alkaloids, terpenoids and some pigments are responsible for the "green" synthesis of various low-molecular metals (**Das *et al.*, 2010; Nasrollahzadeh *et al.*, 2014; Sangeetha *et al.*, 2011**). Different phenolic acids such as caffeic (**Aromal and Philip, 2012**), ellagic acid (**Edison and Sethuraman, 2012**), gallic acid (**Huang *et al.*, 2010**), and protocatechuic acid (**Kumar *et al.*, 2012**), as well as alkaloids are acting as bioreducting agents for the synthesis of nanoparticles of metals. It is believed that bioreduction of silver and gold ions occurs through the hydroxyl groups of flavonoids (**Ghoreishi *et al.*, 2011**) and terpenoids (**Shankar *et al.*, 2003**), oxidized to carbonyl groups and for the use of flavonoid glycoside apine (**Kasthuri *et al.*, 2009**).

The pH of the plant extract is very important for the formation of nanoparticles (**Armendariz *et al.*, 2004; Ghodake *et al.*, 2010; Sathishkumar *et al.*, 2010**). The change in pH affects the charge of natural extract phytoreagents and their ability to bind and recover metal cations during the synthesis of nanoparticles. These changes contribute to the modification of the shape, size and release of nanoparticles. In the extract of *Avena sativa* at pH 3-4, golden nanoparticles of small size were formed, while at pH 2, larger aggregated particles were observed. Perhaps this is due to the fact that at pH 3-4 the bigger number of functional groups becomes accessibler than at pH 2 (**Sathishkumar *et al.*, 2010**). On the contrary, using pear fruit extracts has shown that hexagonal and triangular gold nanoplates are formed in alkaline media, while in the acidic medium nanoparticles are not formed (**Ghodake *et al.*, 2010**). It is found that significantly more silver nanoparticles from *Curcuma longa* tubers are synthesized at alkaline pH values (**Sathishkumar *et al.*, 2010**). The work (**Vennila *et al.*, 2018**) found that the optimum conditions for the synthesis of silver nanoparticles is the medium of aqueous extract of *Spermacoce hispida* leaves at pH 8 and 40 °C. AgNPs have been found to have a concentration-dependent antioxidant activity. Experimental results show that if adjusting the size of the synthesized LF Al_2O_3 from *Prunus × yedoensis* leaf extract and optimizing its pH, higher catalytic and antibacterial activity is manifested (**Manikandan *et al.*, 2019**).

Another important factor influencing the formation of nanoparticles in plant extracts is the temperature (**Lin *et al.*, 2010; Lukman *et al.*, 2011; Das *et al.*, 2011; Eghbali-Arani *et al.*, 2018; Kumar V. *et al.*, 2019**). In general, the temperature increasing helps the reaction rate and the effectiveness of nanoparticle synthesis increasing. In alfalfa (*M. sativa*), triangular silver nanoparticles are formed only at temperatures above 30 °C (**Lukman *et al.*, 2011**). The increase in reaction temperature is accompanied by an increase in the efficiency of the recovery of Ag ions in the Lemon verbena extracts (**Cruz *et al.*, 2010**). Moreover, at high temperatures, particles are formed much more often than at room temperature. Temperature can also affects the structural form of synthesized nanoparticles. At room temperature, in Cassia fistula plants, mostly silver nanowires are formed, while at temperatures above 60 °C the bulk of the spherical nanoparticles is formed (**Lin *et al.*, 2010**). In this case, it is believed that higher temperatures can changes the interaction of phytoreagents with the surface of nanoparticles, thereby inhibiting the inclusion of adjacent nanoparticles into the structure of the nanoscale. In addition, higher temperatures in some cases may contribute to the nucleation process to the detriment of the secondary recovery process and the subsequent condensation of the metal on the surface of nanoparticles forming. This phenomenon explains the formation of spherical gold nanoparticles at 80 °C in alcohol extracts *Nyctanthes arbor-tristis* (night jasmine), in contrast to nanoparticles of various shapes formed at room temperature (**Das *et al.*, 2011**). The rate of synthesis of AgNPs increased significantly during 2 minutes from the start of $AgNO_3$ incubation with *Laminaria japonica* algae extract and temperature rise from 90 °C to 120 °C. At room temperature, it takes 48-72 hours for completing (**Kim *et al.*, 2018**). The silver nanoparticles obtained from the leaf extract of *Platycodon granddiflorum* at 50 °C had an average size of 21 nm while a temperature of 30 °C was 19 nm. In general, low frequency synthesized at 50 °C had a good shape and structure with high stability (-5.23) (**Anbu *et al.*, 2019**).

In determining the shape, size, and speed of the nanoparticle recovery process, the concentration of plant extracts plays an important role (**Kumar *et al.*, 2019b**). When used for the synthesis of AgNPs and AuNPs of fruit extract (*Tanacetum vulgare*) at concentrations of 0.5 ml, 1.0 ml, 1.8 ml, 2.8 ml, 3.8 ml and 4.8 ml, a decrease in the size of the particles was observed with an increase concentration of extract (**Dubey *et al.*, 2010**). Similar results were obtained for the synthesis of AgNPs and AuNPs from the leaf extract of *Chenopodium album* (**Dwivedi and Gopal, 2010**). The average size of AgNPs obtained from *C. amboinicus* (**Narayanan and Sakthivel, 2011a**) and *Carica papaya* peel extract (**Balavijayalakshmi and Ramalakshmi, 2017**) decreased with increasing extracts concentration. The shape, size and yield of nanoparticles in green synthesis depend on the time of incubation / reaction. During the AgNPs synthesis using the extract of *Capsicum annuum L.* at 5 h incubation time, the nanoparticles were spherical and their size was 10±2 nm. In the case of increasing the incubation time to 9 and 13 hours, the size of the nanoparticles increased to 25±3 nm and 40±5 nm, respectively (**Li *et al.*, 2007**).

Concentration of metals' is also influencing the synthesis of nanoparticles (**Kumar *et al.*, 2019a**). **Dubey *et al.*,** (2010) synthesized AgNPs and AuNPs using fruit musk extract at different concentrations of metals' ions (1-3 mM). Increasing in the concentration of silver ions led to an increase in the absorption peak in the case of AgNP, but in the case of AuNPs, the absorption peak increased at 1 mM and 2 mM gold ion concentrations, while peak absorption decreased with 3 mM gold ions. At a higher concentration of metal ions, AgNPs were larger. In addition, they observed that an increase in the reaction time resulted in an increase in the acute absorption peak for both AgNP and AuNP.

The efficiency of the synthesis of metals' nanoparticles also depends on the ion electrochemical potential (**Haverkamp R. and Marshall, 2009; Sims *et al.*, 2017**). Thus, the ability of a plant extract to effectively regenerate metal ions can be significantly higher in the case of ions with a large positive electrochemical potential (eg, Ag^+) than in low-electrochemical potential ions such as $[Ag(S_2O_3)_2]^{3-}$)] (**Haverkamp and Marshall, 2009**). AuNPs superficial charge, calculated on the zeta potential basis, facilitates their physical and chemical stability and further involvement in metabolism and bioaccumulation (**Sengani *et al.*, 2017**). The toxicity level of AuNPs strongly depends on the charge of surface particles, so positively charged gold nanoparticles cause cell death at lower concentrations, while neutrally charged particles detect cell death at much higher concentrations (**He *et al.*, 2017**).

The proteins that make up the herbal extract can significantly influence the formation of nanoparticles (**Saravanakumar *et al.*, 2018**). Recently, the "green" synthesis of metals' nanoparticles uses approach combining the use of plant extracts with the addition of biomaterial, peptides and proteins, the amino acid sequence and structure of those amino acids optimized for the effective production of nanoparticles. The best ability to recover metal ions belongs to tryptophan, tyrosine, arginine and lysine (**Selvakannan *et al.*, 2004**). ZnO nanoparticles of 30 to 50 nm containing functional molecules such as carboxylic acids, amines and nitro groups obtained from the *Cassia alata* plant. They had anti-dermatophytic potential against *Trichophyton mentagrophyte, Trichophyton rubrum, Epidermophyton floccosum, Microsporum audouinii* and *Microsporum canis* (**Sujatha *et al.*, 2018**).

Nanomaterials in the food industry

In modern conditions, nanotechnologies are increasingly becoming widespread, especially in the food industry (**Patel *et al.*, 2018; Hamad *et al.*, 2018; Sonawane *et al.*, 2018**). Nanotechnologies have the potential providing healthier, safer and better food, as well as improved packaging of food products. The application of nanotechnologies in the food industry will contribute to the creation of new materials through a range of their unique physical and chemical properties, useful for a number of other industries (**Dasarahally-Huligowda *et al.*, 2019**). It is promising to apply nanotechnological approaches to the entire chain of agricultural production, from the field to consumers (**Bityutskyy *et al.*, 2017; Fortunati *et al.*, 2019**). This involves the creation and use of organic and inorganic materials in a nanoscale, with individual physical, chemical and biological properties. It had been established that substances in the form of nanoparticles, in comparison with molecular and macroscopic forms, have for the most part different physical and chemical properties (**Jafarizadeh-Malmiri *et al.*, 2019c**).

The field of nanotechnology in the food industry is rapidly developing and has various applications in the food analysis (**Nurfatihah and Siddiquee, 2019**), the application of nanosensors (**Ragavan and Neethirajan, 2019; Hernández-Muñoz *et al.*, 2019**), tracking devices, packaging (**Jafarizadeh-Malmiri *et al.*, 2019a; Hernández-Muñoz *et al.*, 2019**) etc. Nanotechnologies at various levels in the food industry, including food packaging, food processing, and various conservation methods aimed at increasing the shelf life of the food (**Hamad *et al.*, 2018**). Nanotechnologies are successfully used to ensure the quality of food and food safety (detection of pathogenic microorganisms or toxic metabolites), enrichment of food products (minerals, vitamins, antioxidants and essential oils), improvement of organoleptic properties (increase of taste or color), extension of shelf life and antimicrobial food packaging (**Chen *et al.*, 2017; Duncan, 2011; Hamad *et al.*, 2018**). Nanotechnologies for intelligent packaging developing are focusing mainly on protecting the product from oxygen, moisture and maintaining freshness (**Cabedo *et al.*, 2018; Oh *et al.*, 2019**). Functional packaging should have increased mechanical strength, barrier properties, flexibility and stability, be capable of biological decomposition, low-yielding and environmentally friendly (**Kuswandi and Moradi, 2019**).

Commonly accepted methods for detecting pathogens transmitting by food or their toxic metabolites are time-consuming and expensive compared to nanotechnology methods that are faster, more accurate, and more cost-effective (**Bata-Vidács *et al.*, 2013; Choleva *et al.*, 2018; Liao *et al.*, 2018**).

Nanomaterials are generally classified as: nanoparticles, nanomolecules, nanoemulsions, nanolaminates, nanocapsules, nanofibers, nanotubes etc. (Fig. 3), which can be synthesized by many methods and have many applications in the food industry (**Liu *et al.*, 2018**). Nanomaterials can be used as nutritional supplements (nanoinsiders) and as food packaging (nanooutsiders) (**Ranjan *et al.*, 2014**).

Biopolymer nanoparticles can be formed using biopolymers of food quality, namely, proteins or polysaccharides by self-concentration or aggregation. Polylactoglicolic acid is used to encapsulate and deliver medications, trace elements and vitamins (**Ravichandran, 2010**).

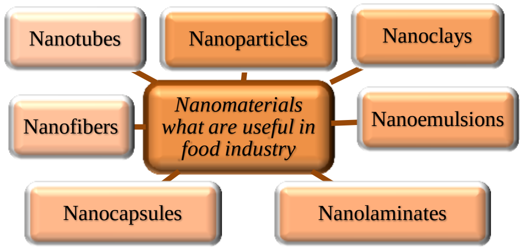

Figure 3 Nanomaterial's what are useful in food industry

Nanoemulsions with droplets diameter less than 100-500 nm in diameter can be included in functional food ingredients (**Ranjan *et al.*, 2014; Artiga-Artigas *et al.*, 2019**). Substances with common nanosilver and nanoclay structure can act as a barrier to gas and moisture (**Akbari *et al.*, 2006; Cabedo *et al.*, 2018**). These nanocomposites are potentially used in packaging for cheese, confectionery, processed meat and cereal products (**Cagri *et al.*, 2004; King *et al.*, 2018**). An additional application includes an extrusion coating for fruit juices, liquid dairy products, as well as beer and carbonated drinks (**Akbari *et al.*, 2006; Jafarizadeh-Malmiri *et al.*, 2019b**).

Nanolaminates consist of two or more layers of nanosized material and physically or chemically bonded to each other. They may contain edible nanolaminates constructed of polysaccharides, proteins and lipids (**Ravichandran, 2010**). They can be built in the form of food coats and films for a variety of food products such as vegetables, fruits, meat, candy, chocolate, bakery products. Such a coating or film serves as a barrier to moisture, gas and lipids. In addition, nanolaminates can be carriers of functional components, namely flavors, dyes, antimicrobial compounds, antioxidants, and improve the texture properties of various foods (**Ranjan *et al.*, 2014**).

Nanocapsules are made of lipids or natural polymers and they are widely used in foods (**Ponce *et al.*, 2018**). The formation of nanocapsule involves the inclusion of bioactive compounds in small particles. Encapsulation using nanoemulsions is a powerful technique for protecting food ingredients, including vitamins, lipids, antioxidants and antimicrobial agents. (**Kumar and Sarkar, 2018**). Immobilization of the enzyme on the appropriate carriers increases their functional efficiency and reproducibility while reducing the complexity and reducing the contamination. They are useful in baking, making dairy products, jams, jellies, processing beverages (beer, wine, juices) (**Sneha *et al.*, 2019**). Rosemary extract nanocapsule with natural antimicrobial polysaccharide chitosan and γ-polyglutamic acid (γ-PGA) is used to improve antimicrobial activity (**Lee *et al.*, 2019**).

Nanotubes could be made of several globular proteins and be useful to enhance the processes of immobilization of enzymes (**Singh, 2016**).

The complexity of preserving food is due to the largely inappropriate use of antibiotics. The use of nanomaterials with antimicrobial activity is a new protection from many drug-resistant infectious organisms (**Bata-Vidács *et al.*, 2013**). Instead of interfering with a certain biochemical process, as conventional antibiotics do, nanoparticles are likely to inhibit several processes in microorganism cells in a less specific manner. Nanoscale materials that possess antimicrobial properties include nanoparticles based on silver oxide (Ag_2O) (**Allahverdiyev *et al.*, 2011**), Titanium dioxide (TiO_2) (**Chawengkijwanich and Hayata 2008**), copper and copper oxide (CuO) (**Chauhan *et al.*, 2019**), zinc oxide (ZnO) (**Paul *et al.*, 2019**), cerium dioxide (**Tsekhmistrenko *et al.*, 2018a**). Among them, silver nanoparticles (AgNPs) are the most powerful anti-microbial agents of a wide spectrum of action. Conventional antibiotics eradicate only 5-6 pathogens, while silver molecules can destroy more than 650 pathogens in 6 minutes of contact (**Han and Li, 2008**). This indicates the possibility of their use as a preservative and packaging material due to their safe condition and low cost (**Akbar *et al.*, 2019**).

Polypropylene films coated with TiO_2 nanoparticles exhibited photoconductive biocidal properties against nine food bacteria and yeasts (**Cerrada *et al.*, 2008**), which were able to inhibit the growth of *E coli* in freshly cut salad (**Chawengkijwanich and Hayata 2008**). Silver nanoparticles (AgNPs) exhibited greater killing power than vancomycin (**Gu *et al.*, 2003**).

Enzyme-like activity of metals' nanoparticles allow them to be widely used in various industries (**Cormode *et al.*, 2018; Li *et al.*, 2018; Wang *et al.*, 2016; Tsekhmistrenko *et al.*, 2018b**). The activity of peroxidase and oxidase are exhibited by iridium nanoparticles (**Cui *et al.*, 2017**), rhodium (**Choleva *et al.*, 2018**), ruthenium (**Cao *et al.*, 2017**), cerium oxide (**Estevez *et al.*, 2017**), manganese oxide (**Yan *et al.*, 2017**), cobalt compounds (**Jia *et al.*, 2016; Wang *et al.*, 2017; Zhao *et al.*, 2018**), $NiCo_2O_4$ (**Song *et al.*, 2018**).

Nanotechnology applications in the food sector are expected to bring a number of benefits, including new flavors and product texture, less use of fats, and improved nutrient uptake (**Ponce *et al.*, 2018**). The nanosensory devices will allow rapid, selective, responsive, cost-effective and, in some cases, built-in, online, and real-world detection of a wide range of compounds, even in compound matrices, and may allow the development of new strategies for detecting allergens (**Gómez-Arribas *et al.*, 2018**).

Packages with nanocomposites of metals have a number of advantages, including reduction of the use of preservatives, higher rate of reactions for suppressing microbial growth, extending the shelf life of food products (**Hoseinnejad *et al.*, 2018**). As biosensors for assessing the quality and safety of food products, nanoparticles are used (**Ragavan and Neethirajan, 2019; Jafarizadeh-Malmiri *et al.*, 2019c**).

The most promising is the use of nanoparticles obtained by the "green" synthesis method (**Dwivedy *et al.*, 2019**). However, the use of nanoparticles in all industries should be clearly controlled, as there is a report on the toxicity of individual nanomaterials (**Jain *et al.*, 2018; Das *et al.*, 2019**).

Risks of using metals' nanoparticles

The widespread use of metals' nanoparticles and their oxides is not possible without assessing their potential impact on end users (**Kaphle *et al.*, 2018**). The issues of biosafety of nanomaterials are multifaceted and ambiguous, requiring an integrated, safe, responsible and scientifically grounded approach (**Communication, 2008**). There are currently not enough reports on the toxicodynamics and toxicokinetics of nanoparticles in humans and animals and their impact on the environment (**Kaphle *et al.*, 2018**). To this end, it is necessary to systematize information on the interrelationships between the toxicity of nanoparticles and their composition, concentration, size, form, reactivity, etc. (**Navya and Daima 2016**). It is important to investigate the molecular mechanisms of the influence of nanoparticles on the body, organs, tissues, cells and mechanisms of development of remote toxic effects, as well as ways to eliminate or weaken their undesirable effects. The pollution of nanoparticles, and, consequently, the change in their properties is reported not only in the environment but also in the food production chain, which has an impact on humans almost inevitable (**Mattarozzi *et al.*, 2019**). Nanotechnologies improve our lives, especially in nutrition. They could provide potential problems which could not be ignored (**Ummi and Siddiquee, 2019**). Such studies are possible in case of use of key system characteristics of biological systems in vivo and in vitro (physiological, biochemical, immunological, genetic, cytological, etc.) that are susceptible to toxic effects (**Roman'ko M. E., 2017; Prajitha *et al.*, 2019**). The toxic effects of nanoparticles may be due to the fact that they are capable of penetrating the cell bypassing the respiratory, skin, gastrointestinal, hematoencephalic, placental and other barriers and selectively accumulate in cells and sub-cell structures (**Colon *et al.*, 2009; Fischer and Chan 2007; Ranjan *et al.*, 2018**).

The food industry intensively uses silver nanoparticles as sensors, nutritional supplements, and packaging components. However, they present a potential toxicity risk for the consumer, the target of which is the intestine and liver. Silver nanoparticles can produce free radicals and cause oxidative stress in cells, causing oxidative damage. Oxidative stress mediates toxicity, causing inflammatory reactions and death from necrosis or apoptosis (**Gaillet and Rouanet, 2015**).

It is necessary to balance the efficiency and toxicity of nanomaterials before using such materials (**Thorley and Tetley 2013**). Nanomaterials what are intended for use in foods and for therapeutic purposes should be thoroughly investigated *in silico, in vitro* and *in vivo* toxicity analysis (**Jain *et al.*, 2016**). For these innovative systems, additional research needs to achieve minimal side effects and a wide range of applications.

Conclusions and perspectives of further research

Among a number of methods used to synthesize nanoparticles of metals and metal oxides, a "green" synthesis based on plants deserves a great deal of attention, due to its environmental friendliness. There is a large array of reports about the possibility of synthesizing nanoparticles from different parts of plants, including leaves, stems, bark, flowers, roots, peas, fruits and seeds. The size and shape of nanoparticles are controlling by physical and chemical parameters. The peculiarity of the "green" synthesis of nanoparticles is the difficulty of explaining the mechanisms of synthesis, prediction and identification of specific bioreducting and stabilizing molecules. The size, shape and properties of nanoparticles obtained by nanotechnological methods depend on the type and part of plants, temperature, reaction of the medium, concentration of metal ions and other parameters. Studying features of metals nanoparticles synthesis from plant raw materials and their specific properties will provide wide application in biology, medicine, agriculture, and in the food industry. All this together will contribute to improving the health of people, the duration and quality of life.

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