Review Article

Fruit and Vegetable Mediated Green Synthesis of Silver Nanoparticles – A Review

Ritika Desai and Samit Dutta*

College of Food Processing Technology & Bio Energy, Anand Agricultural University, Anand, Gujarat, India

Abstract

The green method for the synthesis of silver nanoparticles is gaining enthusiasm among scientists and researchers. It counters the perils of physico-chemical methods, green methods are fast gaining popularity. The eco-friendly green method is economical, energy-efficient, easily scaled up for vast-scale synthesis of nanoparticles, and does not require harsh conditions for synthesis. Fruit and vegetables mediated synthesis exhibits a large scope for research due to the multitudinous bioactive components present in fruits and vegetables which act as reducing as well as capping and stabilizing agents. The present review briefly discusses the methods, characterization techniques, and various factors affecting the fruit and vegetable mediated green synthesis of AgNPs along with some applications.

Keywords: Green synthesis, AgNPs, fruit and vegetables mediated, nanoparticles

***Correspondence**
Author: Samit Dutta
Email: samit@aau.in

Introduction

The term 'Nanotechnology' refers to the creation, characterization, handling, and utilization of structures at the nanoscale (1-100nm) [1]. Materials in the nanoscale level exhibit improved properties (physical, chemical, magnetic, optical, electrical, etc.) than the material in bulk, by virtue of their distinct characteristics such as the large surface area to volume ratio, size, shape, and morphology [2, 3]. The synthesis of nanoparticles (NPs) is a rising domain of research and is picking up significantly throughout the world. NP synthesis involves two approaches, "Top-down" and "Bottom-up". The former method involves splitting up of suitable bulk materials by various size-reduction techniques while the latter call for tailoring of atoms to new nuclei which grow into a particle of a nanoscale [4].

NPs from noble metals, particularly Ag, Au, Pd, and Pt are the most widely studied nanoparticles today. Silver has been used since ancient times by Greeks, Romans, Persians, and Egyptians to store food products [5]. "Charak Samhita", an ancient Indian Ayurvedic Medicine book also mentions the therapeutic potentials of silver [6]. It is well known for its toxicity against microorganisms. The antimicrobial activity of various metals follows the order [7],

$$Ag > Hg > Cu > Cd > Cr > Pb > Co > Au > Zn > Fe$$

Conventionally the nanoparticles were synthesized using two routes viz. physical and chemical. Chemically, the nanoparticles were synthesized using chemical reduction, sonochemical, electrochemical, microemulsion, etc. while the physical method involved techniques such as pulsed laser ablation, evaporation-condensation, arc discharge, ball milling, and pulse wire discharge [3]. However, these methods necessitate the use of toxic chemicals which are absorbed on the surface and may have detrimental effects on medical applications [8]. These methods have effective yields but have high operational costs and energy needs [9]. To counter the perils caused by the physico-chemical methods, the need for green synthesis arose. The ecofriendly green method is economical, energy-efficient, easily scaled up for vast-scale synthesis of nanoparticles, and furthermore, it does not require high temperature, pressure, and toxic chemicals [10, 11].

With multitudinous advantages and the surging popularity among researchers, the green synthesis of silver nanoparticles is a dynamic region of research today. Green synthesized AgNPs can be achieved by using plant and plant extracts (leaves, stem, fruits, peel, latex, vegetables), microorganisms (bacteria, fungi, yeast), and templates (cell membranes, viruses, DNA) [12]. The present review focuses on fruit and vegetable mediated green synthesis of AgNPs and also summarizes the process of production, various factors affecting the synthesis, characterization techniques of formed AgNPs, and some applications.

Green synthesis process

As discussed earlier, the major advantage of the green method lies in its single-step process. Silver metal ion solution and a biological reducing agent are the perquisites for the green synthesis of silver nanoparticles (AgNPs) [9]. The silver metal ion solution of different concentrations can be produced from various water-soluble salts of silver, such as $AgNO_3$. Concentrations ranging from 0.1mM to 10mM can be used for AgNP synthesis, however, 1mM is preferred by most of the researchers.

Fruits and vegetables are a potential source of biologically active compounds. Various biomolecules such as polysaccharides, vitamins, amides, polyphenolic compounds, alkaloids, organic acids, etc. present in fruits and vegetables can reduce the Ag^+ ion from the metal ion solution to AgNPs thereby acting as biological reducing agents [13]. The process flowchart for the green synthesis of AgNPs mediated by fruits and vegetables is illustrated in **Figure 1**.

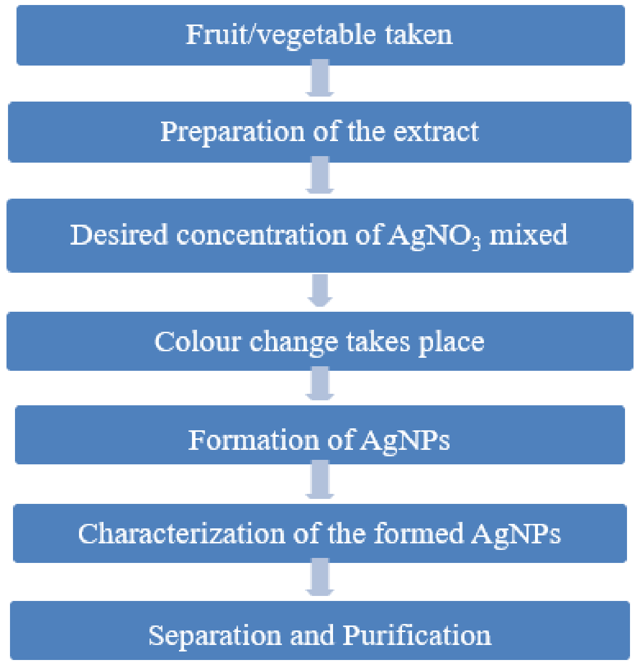

Figure 1 Process flowchart for fruit and vegetable mediated green synthesis of AgNPs

Mechanism

The formation of AgNPs using fruits and vegetables undergoes a two-step process mechanism, namely atom formation and polymerization. Fruits and vegetables are said to be the natural source of bio active compounds. These bioactive compounds act as reducing and stabilizing agents. The bioactive compounds act as electron donors and reduce the Ag^+ ions obtained from the aqueous solution of silver salt to Ag^0. The Ag^0 act as nucleation centers and as catalysts for the reduction of remaining Ag^+ ions present in the bulk of the solution. The NPs formed have high surface energy and thus tends to convert themselves to low surface energy by agglomeration. Gradually, the ions agglomerate to form larger particles having high values of nuclearity.

The phytochemicals present in fruits and vegetables stabilize the growth of formed NPs by acting as capping agents. The capping agents are amphiphillic in nature i.e. having a polar head and a non-polar tail. The polar head of the capping agents reacts with the formed NP and forms a layer around it, thereby preventing further growth of the nanoparticle.

The biomolecules perform a dual role by acting as capping and stabilizing agents along with their reducing properties [14]. Thus, there is no need to add toxic capping and stabilizing agents from outside as in the case with the chemical synthesis. Capping/stabilizing agents prevent the agglomeration of the formed AgNPs and make them stable. Ramesh *et al* used the *Emblica officinalis* fruit extract as stabilizing and reducing agent. Some of the fruit and vegetable extracts used for the synthesis of AgNPs are given in **Table 1**.

Factors affecting the green synthesis of AgNPs

Reaction temperature

As discussed earlier, the green method does not necessitate harsh conditions like temperature and pressure. Synthesis of AgNPs from fruit and vegetable extracts can be easily accomplished at room temperatures and ambient pressure [15, 16]. For red pumpkin extraction, [17] used hot aqueous extract heated to 60°C for 2 hours. [18] in their study found that microwave-mediated synthesis was more rapid and favorable compared to conventional heating. [19] showed that rapid and small-sized AgNPs were synthesized as the temperature was increased from 40 to 80°C.

Table 1 Green synthesis of AgNPs using fruit and vegetable extracts.

S. No.	Fruit / Vegetable extract	Size	Shape	Reference
1	*Amaranthus dubius* C	18 – 21 nm	Spherical	[31]
2	*Emblica officinalis*	15 nm	Spherical	[32]
3	*Malus pumila*	27 – 33 nm	Spherical	[33]
4	*Ficus carica*	20 – 80 nm	Spherical	[34]
5	*Vitis vinifera*	18 – 20 nm	Spherical	[7]
6	*Opuntia ficus-indica* peel	20 - 60 nm	Spherical	[22]
7	*Carica papaya* L.	10 – 50 nm	Cubic and hexagonal	[15]
8	*Dillenia indica*	40 – 100 nm	-	[35]
9	*Solanum tuberosum*	10 nm	Spherical	[16]
10	*Lycopersicon esculentum*	14 – 28 nm	-	[27]
11	*Allium cepa*	5.3 – 10.2 nm	Spherical	[36]
13	*Rubus glaucus*	12 – 15 nm	Spherical	[37]
14	*Capsicum frutescence*	20 – 25 nm	-	[38]
15	*Brassica oleracea var. botrytis* Waste	5 – 50 nm	Spherical	[18]
16	*Sambucus nigra*	20 - 80	Spherical	[39]
17	*Vitis vinifera* pomace	3 – 14 nm	Spherical	[40]
18	*Citrus sinensis*	5 – 50 nm	Spherical	[41]
19	*Tanacetum vulgare*	16 nm	Spherical	[42]
20	*Punica granatum* peels	5 nm	Spherical	[20]
21	*Averrhoa carambola*	12 – 16 nm	Spherical	[21]
22	*Tamarindus indica*	6 – 8 nm	Spherical	[42]
23	*Ipomoea batatas* L.	26 – 37 nm	Spherical	[23]
24	*Raphanus Sativus*	30 – 60 nm	Polygonal	[43]
25	*Cucurbita maxima* L.	67 nm	Spherical	[17]
26	*Manilkara zapota* pomace	23.6–65.9 nm	-	[24]
27	*Citrullus lanatus*	20 – 34 nm	Spherical	[19]
28	*Lyciumbarbarum*	3 – 15	Spherical	[25]
29	*Vitis vinifera* seeds	25 – 35 nm	Polygonal	[44]
30	*Cocos nucifera* coir	23 nm	Spherical	[45]
31	*Ananas comosus*	12 nm	Spherical	[46]
32	*Glycosmis pentaphylla*	17 nm	Spherical	[47]
33	*Solanum melongena* L.	92.4nm	Spherical	[48]

Metal ion concentration

Various concentrations ranging from 0.1mM to 10mM were taken under consideration. The concentration of 1mM was found suitable in the majority of the experiments [15, 20 – 23]. [19] observed a steady increase in the intensity of the absorbance peak with the increase in the concentration of $AgNO_3$ solution from 0.1mM to 1mM. Few authors have mentioned the use of 7mM [24] and 9.41 mM [25] for the efficient synthesis of AgNPs.

pH of the reaction mixture

Another factor responsible for the synthesis of AgNPs is the pH of the reaction mixture. The biomolecules in the extract carry charges. Many authors have reported that the pH alters the charges on the biomolecules, thereby affecting the NP synthesis [26]. The basic medium promotes NP synthesis and produces stable and uniform-shaped nanoparticles [9]. [27] reported a rapid growth rate of NP in an alkaline medium. Further increase in the pH (> 11) is associated with the formation of unstable and clustered NPs [28]. [21] revealed a slow increase in the intensity of plasmon at acidic pH.

Extract contents

Different kinds of fruit and vegetable extracts were used by researchers as reducing agents to prepare AgNPs. The concentration of the extract used varies according to the type and amount of bioactive compounds present in them. [18] and [19] showed that more amount of small-sized AgNPs was synthesized when the volume of extract was increased. This can be attributed to the fact that higher volumes provide more bioactive compounds and hence more reducing agents spur up nucleation [29]. Also, a higher concentration of extract furnishes a sufficient amount of capping agents which hinders the agglomeration of the formed AgNPs and accounts for small-sized stable NPs formation [30].

Other factors such as duration of reaction, and agitation also play a significant role in the synthesis of AgNPs.

Characterization techniques

The physical, chemical and biological properties of formed AgNPs are completely dependent on the shape, size, morphology, and composition of the AgNPs [49, 50]. For desired applications, AgNPs with certain characteristics is preferred. Consequently, the characterization of formed AgNPs is crucial. For characterization of AgNPs on the basis of shape, size, morphology, and polydispersity [51], numerous techniques are employed as described below.

UV-visible spectrophotometer

This technique is useful for the identification and characterization of the shape, size, and size distribution of AgNPs. Based on the size and shape, plasmonic NPs absorb radiations in the visible and near-infrared regions [52]. Thus for the characterization of NPs of 2 – 100nm size, the wavelength of 200 – 800 nm is used [53].

Fourier transforms infrared spectroscopy (FTIR)

In order to identify the organic functional groups attached to the AgNP surface, the FTIR technique is employed [54].

Transmission electron microscopy (TEM) and Scanning electron microscopy (SEM)

For the morphological characterization of the formed AgNPs scanning electron microscopy (SEM) and transmission electron microscopy (TEM) are used. These electron microscopes efficiently provide morphological data in the range of nanometer to micrometer [55]. A transmission microscope has greater resolution than SEM and is utilized for determining the exact shape and size of the synthesized NPs whereas SEM caters to providing elemental information at the micron scale.

Zeta potential measurement

In order to determine the stability of the AgNP synthesized, Zeta potential is used. Greater the value of zeta potential, the greater the stability of the NPs [56].

Dynamic light scattering (DLS)

Presently, dynamic light scattering is one of the most common methods for characterizing the size of AgNPs in aqueous suspension. It also finds application in determining the surface charge and polydispersity index of the prepared nanoparticles [51, 57].

Auger electron spectroscopy (AES) and X-ray photoelectron spectroscopy (XPS)

XPS is sometimes utilized in cases where the determination of NP size by other methods becomes difficult. The presence of coating material on NP surfaces along with their thickness and composition can be easily identified using AES and XPS [56].

Energy dispersive spectroscopy (EDS)

The knowledge of AgNP's elemental composition can be obtained from EDS [58].

Low energy ion scattering (LEIS)

It is an extremely sensitive spectrometry technique that provides information regarding the elemental composition in the outermost layer of the AgNPs. It is a powerful tool for analyzing and establishing a relationship between surface-related phenomena (adhesion, wetting) and surface chemistry [59, 60].

Applications

Silver nanoparticles (AgNPs) have a growing enthusiasm owing to their lower toxicity in humans and thus contribute to diverse applications such as in optics [61], bio labeling [62], catalytic activity [63], nanomedicine [64], chemical sensing, household water filters, cellphones, laptop keyboards, children's toys, contraceptives [7], data storage [65], cell biology [66], agriculture, cosmetics [67], textiles [68], antioxidants and antimicrobial agents [69], the food industry [70] and many more.

Conclusion

The green method for the synthesis of NPs shows that nature has a beautiful way of creating the most efficient tiny functional materials. In summary, it is concluded that sufficient literature and research has been carried out on green synthesis as it is eco-friendly, cost-effective, easy to scale up, safe products, and lesser generation of waste. Moreover, it is less time-consuming and can be carried out under mild environmental conditions. Thus, the present review highlighted the significance of fruit and vegetable mediated synthesis of silver nanoparticles. There is a vast scope for future researchers as great diversity in fruit and vegetable varieties is way to be exploited.

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