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A Review on Synthesis of Plant-Mediated Metal Nanoparticles for Fabric Coating

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Biosynthesis, or also known as green synthesis method, has received an enormous attention from the scientific community over the physical and chemical synthesis methods for the production of metal or metal oxide nanoparticles (NPs). Physical and chemical syntheses are typically expensive and generate a large number of hazardous by-products which can cause toxicity to either humans or the environment. Thus, biosynthesis is an alternative for production of NPs. Biosynthesis involves the use of plant extracts and natural bio-resources such as microorganisms and enzymes. It has been demonstrated that the biosynthesis method is suitable for large-scale production since it is simple, cheap, easy, fast, non-toxic, and environmentally friendly. This review mainly focuses on the synthesis of plant-mediated metal NPs for fabric coating. Importantly, the characteristics of the NPs and their applications after fabrication onto the fabrics are also highlighted. The morphology and size of the NPs are influenced by the concentration of plant extracts, control of metal salt concentration, temperature, and pH. This review also summarizes the advantages, limitations, and future challenges of biosynthesis towards the production of fabric coatings. There are several advantages in utilizing green substances, including moderate operation conditions with a minimal use of toxic chemicals and low energy consumption.

Key words: Biosynthesis; Green synthesis; nanoparticles; fabric coating

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Nowadays, the synthesis, characterization, and application of nanomaterials have been gaining attention due to their unique properties, such as extremely small size, high surface-area-to-volume ratio, surface modifiability, great biocompatibility, and excellent magnetic properties. Nanoparticles (NPs) have become significant in many fields such as the textile industry, health care, agriculture, energy, sports, etc. [1]. NPs can be obtained through chemical, physical or biological synthesis processes. Chemical and physical syntheses of NPs are costly and involve the use of toxic and hazardous chemicals which may harm the environment and biological life cycles. Also, the use of toxic chemicals may limit its applications, specifically in biomedical fields [2]. Thus, the development of biological synthesis approaches which are economical, environmentally friendly, and free from the hazardous by-products has gained interest in this contemporary world.

The biosynthesis process of NPs involves the use of various natural resources such as microorganisms, algae, plants, plant tissues, fruits, and plant extracts [3]. The unicellular and multicultural microorganisms can produce intracellular and extracellular inorganic NPs [1]. However, plant extracts have the edge over microorganisms as synthesizing agents because they occur extracellularly and are considered as a straightforward synthesis approach [4-5]. Besides, plant extracts may act both as stabilizing and reducing agents in the synthesis of NPs [6]. The source of plant extracts may affect the morphology of the synthesized NPs to a great extent due to the different concentrations of biochemical reducing agent contents in different plant extracts. The composition of biochemical reducing agents differs seasonally and regionally in most plant extracts due to the nutrient uptake of the plants. Thus, this will lead to a variation of NPs in every batch and directly affects the application and performance of the end products.

In this review, we focus on the plants used in NP biosynthesis for fabric coating and its applications. Such applications include antibacterial, antioxidant, hydrophobic property, ultraviolet (UV) protection, and also as a dye for fabric. We also highlight the morphologies and characteristics of NPs coated on fabric. The advantages, limitations, and future challenges of the biosynthesis method towards the production of fabric coatings are also discussed. The main aim of this literature is to systematically describe the general biosynthesis procedures and the application of green-mediated metal NPs on fabric. Also, it will give benefits to researchers involved in this exciting field, while serving as a useful guide for readers with a general interest in this topic.

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1. Overview of Biosynthesis Process

Plants contain biomolecules such as proteins, carbohydrates, and coenzymes, which are excellent sources for synthesizing metal or metal oxide NPs due to their potential in reduction of metal salts into NPs. Besides, plants also have the capability to reduce metal ions in a shorter time than bacteria and fungi [7]. In addition, the NPs synthesized by plant extracts can progressively and selectively adsorb biomolecules, resulting in interaction with the biological system by forming a corona which enhances the NPs' efficiency [8]. The plant-mediated metal NPs also provide additional benefits such as antimicrobial properties to the NPs, which could directly enhance the NPs' performance and efficiency on fabric [9-11]. This is due to the presence of functional groups such as phenolics, saponin, anthraquinones, and flavonoids in plant extracts.

The composition of phytochemicals in plant extracts is also varied depending on the source of the plants, which leads to the variation of NPs' characteristics. The main components of phytochemicals present in plants are terpenoids, ketones, sugar, carboxylic acid, aldehydes, and amides [12]. Flavonoids, terpenoids, amino acids (e.g., cysteine, lysine, methionine and arginine), and alcoholic compounds (e.g., hydroquinone, plastohydroquinone, quinol) have the ability to enhance the reduction of metal ions [13-16]. Meanwhile, sugars such as glucose and fructose are responsible for the formation of metallic NPs. According to Panigrahi *et al*. [17], glucose has a stronger capability to reduce metallic NPs compared to fructose. The strong metal-support interaction could reduce the size and shape of NPs. Functional groups like ether (–C–O–C–), alcohol (–C–O–), alkene (–C=C–), and carbonyl function (–C=O) such as ketones, aldehydes, carboxylic acids, and amides in plants are known to be the capping ligands for NPs to prevent further growth and agglomeration of NPs for its stabilization purpose [18].

Various parts of plants, including leaves, roots, stems, rinds, fruits, and seeds can be used for synthesizing metal NPs [7]. Synthesizing of NPs from plants starts with the preparation of plant extracts. An extraction method of a plant plays a crucial role in the success of the biosynthesis process as it will affect the physicochemical properties of extracts and indirectly affect the NPs' performance [19]. Typically, the extraction process of a plant involves six steps, namely collection and authentication of plant material and drying, size reduction, extraction, filtration, concentration and drying, and reconstitution [20]. There are various methods used for plant extraction such as infusion, maceration, percolation, soxhlet extraction, decoction, microwave-assisted extraction, ultrasound-assisted extraction, supercritical fluid extraction, and accelerated solvent extraction [21]. According to Gupta *et al*. [22], the quality of extracts is also influenced by the part of the plant used, solvent used, and ratio of plant to solvent.

In the biosynthesis process, the plant extract is added into metal precursor solutions as bio-reduction and stabilizing agents to facilitate NPs' syntheses [23-24]. The metal salt will be reduced and changes in the color of the mixture solution are monitored. Medium and large particles are removed by centrifugation and washed thoroughly with water. According to Singh *et al.* [25], good mono-dispersity of NPs such as narrow size distribution can be achieved by controlling the synthesizing parameters. The rate of NP formation, yield, and stability are controlled by the phytochemical contents of the plant extract, phytochemical concentrations, metal salt concentration, temperature, and pH [26].

Generally, there are three steps involved in the bio-reduction of metal NPs using plant extracts. The first step is the activation step. In this step, the reduction and nucleation of metal ions occur and lead to the formation of new structures through self-organization (nucleation) of reduced metal atoms. Then, in the growth phase, small-sized NPs join with each other to form larger particles, together with increasing thermodynamic stability of the NPs. The bio-reduction of metal NPs is completed with the termination phase, where the shape of NPs is formed [9], and the reduction of metal ions by active metabolites takes place. The stability of the NPs is depending on the shape of the NPs formed. The NPs are centrifuged and washed to remove impurities and separated using suitable separation methods before used [27].

2. Overview of Fabric Functionalities

The new development of fabrics through nanotechnology has created new fabric materials by producing dynamic, innovative, and interactive products which are able to offer protection, performance, and comfort. It grows rapidly and many developments of new products and applications which have significant added values in terms of functionality have been produced. Traditionally, fabrics serve the purposes of decoration, comfort, and safety. The production of nano finishing-coated fabrics using plant-mediated metal NPs has created complex, multifunctional, and even "intelligent" protective fabrics without the change of fabrics' basic functions. Such functions produced from the development of nanotechnology include antimicrobial, antioxidant, UV resistance, and hydrophobicity of protective fabrics [28-39]. Besides, pigments contained in plants also can act as dye or colorant for fabrics [40-42].

The demand for antimicrobial fabrics has become popular due to the increase in health awareness and healthy lifestyles. Antimicrobial compounds extracted from plants and herbs can be used for the production of protective fabrics to combat the growth of microbial on fabrics [43]. These natural compounds have antimicrobial effects as well as being safe, non-toxic to the skin, environmentally friendly, and ease of availability [44]. Besides, they do not exhibit the side effects associated with synthetic chemicals. The natural defensive amino acids and peptides can inhibit various microorganisms, including both gram-negative and gram-positive bacteria [45]. There are about 12 to 50 amino acids and they are classified based on size, predominant acid structure, or conformational structure. Lysine and arginine are responsible for amphipathic structures that interact with microbial membranes and their positive charges. The mechanism of antimicrobial peptides (AMPs) involves the synthesis of macromolecules like ribonucleic acid (RNA) and deoxyribonucleic acid (DNA), which affect several internal processes and actively respiring cells, which result in the loss of adenosine triphosphate (ATP) [46].

Antioxidants are the substances used to inhibit oxidation which produces free radicals that cause damage to cells [47]. Tocopherols, carotenoids, luteolin, glutathione, vitamin E, and vitamin C are some of the examples of antioxidant substances [41, 48-51]. Antioxidant fabrics are high-performance fabric materials with active substances used to improve skin health [52]. However, it is not considered as a cosmetic product, even though it is able to give benefits to the skin. The fabrics help in reducing the signs of aging, improving firmness and elasticity of the skin, rejuvenating and promoting wellness, and would also give a pleasant feeling [47, 53]. The cosmetic substance attached to the fabric will reach the specific target when in contact with the human body by releasing the active ingredients, which results in improving or repairing the skin structure [50].

Ozone depletion causes the increase of UV radiation that reaches the Earth’s surface. It gives negative effects on the skin such as skin cancer, skin aging, and sunburn. Also, prolonged exposure to UV radiation can cause damage to the immune system and eyes [54]. UV protection fabrics can protect the skin against harmful effects of the sun. However, the effect depends on a few factors such as dyeing and finishing, nature of fibers, and fabric construction factors. A transparent layer of UV absorbent materials produced from nanostructures and natural materials on the surface of the fabric may improve the UV protection factor (UPF) and sun protection factor (SPF). Plant extracts have been used to develop UV protection fabrics due to the presence of phytochemical substances such as alkaloids, flavonoids, carotenoids, and isothiocyanates [55]. In plants, these substances act as a mechanism to protect plants from UV radiation. Besides, the use of plant extracts for the production of UV protection fabrics can prevent pollution by avoiding the use of hazardous chemicals and indirectly promotes eco-friendly fabrics [56].

Hydrophobic or also known as water-repellent fabrics have an extremely low absorption of water with high stability. The hydrophobic fabrics have become popular recently as it can enhance the self-cleaning characteristic of the fabrics [57]. It is different from waterproof fabrics as it does not block air to pass through the fabrics. In other words, the fabrics are breathable [58]. The hydrophobic characteristic of the fabrics can be imparted through coating methods. The active components contained in plant extracts such as fatty acids, phenolic compounds, terpenoids (sesquiterpenoids, diterpenoids), and coumarins act as a capping agent for NPs synthesis [59-61]. These compounds can increase the ability of NPs to form colloidal particles by utilizing a few interactions such as hydrogen bonding, Van der Waals forces, aromatic pi stacking force, and electrostatic attractions [62]. The hydrophobic fabrics generally have other functional characteristics such as antibacterial property, flexibility, strength, soft, and lightweight [63]. Hydrophobic fabric coatings have been applied either for general or industrial wear such as clothing, aprons, lab coats, and uniforms as it can ensure the material to be safer or remain stain-free as it does not absorb any liquid [64].

The application of natural dyes from plant extracts has gained interest due to the worldwide concern over the use of eco-friendly and biodegradable materials. Besides, the color of dyes is permanent on fabrics and it is resistant to sunlight and can withstand many washing cycles [65]. The dyeing process of fabrics with natural dyes involves the use of less water, salt, and heavy metals, which is economical, eco-friendly, and robust than synthetic dyes [66]. Dyeing can be done either in acidic, alkaline or neutral conditions depending on the types of fabrics, source of dye, and metal precursor used [67]. It works best with natural fibers like cotton, viscose, bamboo, linen. and silk. Dyeing fabrics with plant extracts can give other functional properties to the fabrics, such as antimicrobial and antioxidant [40-42].

3. Applications and Characteristics of Biosynthesized Nanoparticles

Numerous researches have been conducted on the use of plant extracts as bio-reducing and stabilization agents for the production of metal NPs [28-42]. The researches mainly involved the development of finishing fabrics for antimicrobial protection. Besides, multifunctional fabrics with a combination of antimicrobial property with one or more fabric protection properties such as hydrophobicity, antioxidant, UV protection and CV dye degradation, and also the use of plant extracts as dyes have been fabricated.

In a work by Shastri *et al*. [28], *Azadirachta indica* leaf extract was used to synthesize silver NPs. *A. indica* leaf extract was obtained by boiling 2 g of leaves in 100 ml of distilled water. The addition of 5 ml of *A. indica* extract into 45 ml of 0.05 M silver nitrate ($AgNO_3$) solution resulted in the change of color from colorless to yellowish-brown, which indicated the formation of Ag NPs. The presence of flavonoids and terpenoids in *A. indica* was responsible for stabilizing metal ions through interactions with carbonyl groups or pi-electrons. Spherical NPs with the size of ~5-50 nm were observed (Figure 1(a)). A pale thin layer of the capping organic material from *A. indica* leaf extract was spotted surrounding the *A. indica*-Ag NPs. The *A. indica*-Ag NPs were coated on cotton and nylon fabrics and both fabrics were efficient against bacteria that cause foot odor (*Klebsiella pnuemoniae*, *Pseudomonas aeruginosa*, *Staphylococcus aureus*, and *Candida albicans*). The attachment of silver NPs to the surface of a bacterial cell may affect its respiration and results in the death of the cell. However, the nylon fabric had greater antimicrobial activity, as exhibited by the larger inhibition zone than the cotton fabric. The antimicrobial properties for both *A. indica*-Ag NPs-coated nylon and cotton fabrics were efficient about 50% to 80% and 30% to 60%, respectively, even after five times of washing.

Nalini and Vijayaraghavan [29] used aloe vera gel for synthesizing silver and gold NPs using silver nitrate ($AgNO_3$) and hydrogen tetrachloroaurate ($HAuCl_4$). The syntheses were conducted at 32 ± 2 °C and pH 6.9 ± 0.2. Then, the NPs were impregnated on a cotton fabric by manual padding, followed by air-drying and curing.

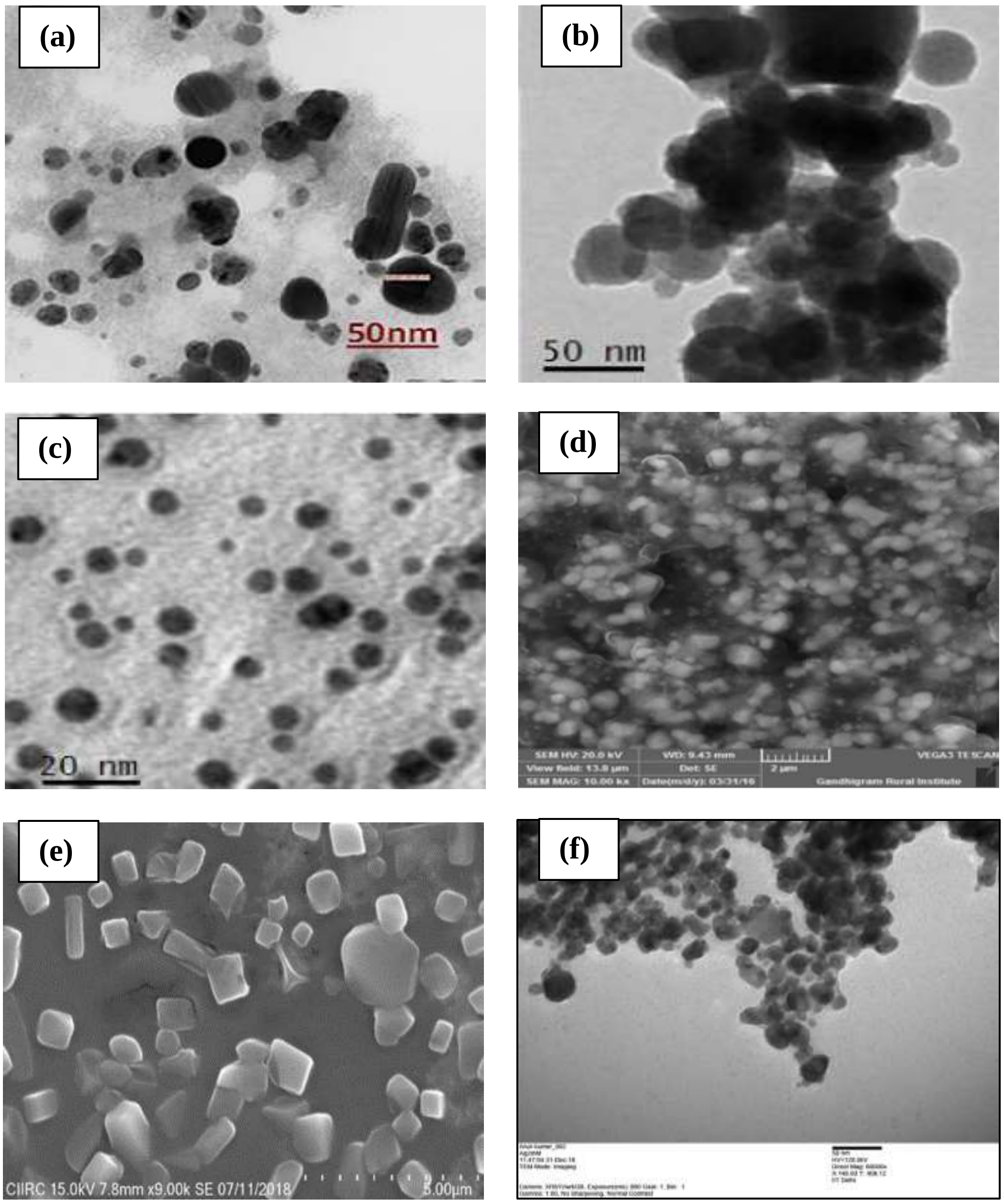

Figure 1. TEM images of NPs: (a) *A. indica*-Ag NPs [28]; (b) Ag-aloe vera NPs [29]; (c) Au-aloe vera NPs [29]; (d) *A. indica*-Ag NPs [33]; (e) *C. papaya*-CuO NPs [37]; (f) Chitosan-Ag NPs [41].

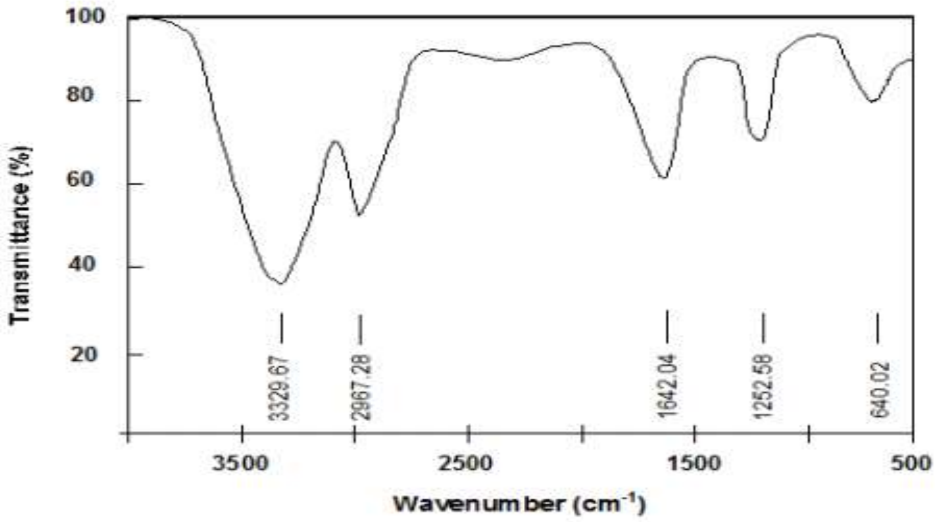

Figure 2. FTIR spectrum of Ag NPs synthesized from aloe vera gel extract [29].

The change of color from colorless to pale yellow and then to deep red-brown confirmed the reduction of Ag^{3+} to Ag. Meanwhile, for Au NPs, the change in color from light yellow to red and deep blood red confirmed the reduction of Au^{3+} to Au. Based on FTIR analyses (Figure 2), functional groups like amine (C–N), ether (C–O–C), amide linkages, and carboxylate (COO–) present in aloe vera acted as capping agents, resulting in the reduction of ions [68], while alcohol and phenol acted as reducing agents. The Ag and Au-aloe vera NPs coated on the cotton fabric were efficient against *Escherichia coli* and *S. aureus* due to the large surface area and high surface energy of nanomaterials which caused the immobilization and high durability of the fabric. Besides, the interaction of bacteria cell walls and sulphur proteins of the impregnant fabric inhibited the bacteria's metabolism, resulting in bactericidal effect. Poly-dispersed spherical NPs with sizes ranging between 12 – 40 nm for Ag-aloe vera NPs (Figure 1(b)) and less than 15 nm for Au-aloe vera NPs (Figure 1(c)) were observed.

Vankar and Shukla [30] produced antimicrobial cotton and silk fabrics using lemon (*Citrus limon*) leaf extract as reducing and capping agents for silver NPs. The extract was prepared by air-drying the leaves, then soaking in boiled distilled water before filtering. The biosynthesis process was performed with the addition of lemon leaf extract to $AgNO_3$ solution. The reduction of Ag^+ ion to Ag occurred after 1 h with the color change of the solution from clear to yellowish-brown. The Ag NPs were coated onto cotton and silk fabrics through the dip and dry process. Both types of fabrics had different colors after coating with Ag NPs. The coated cotton fabric was grayish-brown, while the silk fabric was greenish-brown in colour. However, the colors changed to white after washing but it did not affect the antimicrobial activity of the coated fabrics. Both coated fabrics showed antifungal activities towards *Fusarium oxysporum* and *Alternaria brassicicola* even after five times of washing due to the synergistic effect of silver and essential oil contents of lemon leaves. The morphology of the Ag NPs was observed in various shapes and the size was more than 100 nm.

The antimicrobial textile developed by Jha and Prasad [31] involved the biosynthesis of silver nanoparticles (Ag NPs) with *Clerodendron infortunatum* leaf extract. *C. infortunatum* leaves were alcoholic extracted by boiling 25 g of dried cut leaf with 100 ml of 50% ethanol until the change of color from clear transparent to light yellow-green. For the reduction of silver ions, 5 ml of *C. infortunatum* leaf extract was added into 25 ml of 0.25 M $AgNO_3$ solution with constant stirring at 50 – 60°C. The change of color from deep straw to yellowish-brown and finally black occurred immediately after the addition of the extract, indicating the formation of silver NPs due to excitation of surface plasmon resonance in the range of visible region at 414 nm using UV-Visible Spectroscopy (Figure 3). The NP solution was impregnant onto a cotton fabric through the pad-dry-cure method. Spherical shaped NPs with sizes ranged between 2 – 6 nm were observed. The Ag NPs treated cotton was effective against *S. aureus*. Besides, the treated fabric also had good stability as no aggregation in the solution was observed due to encapsulation by proteins present in the leaf parenchyma. The phytochemicals composition of *C. infortunatum* such as alkaloids, steroids, terpenoids, phenolics, flavonoids, tannins, and saponins could inhibit the microbes present on the surface of the textile and also give a hydrophobic property of the textile material through π-stacking and van der Waals interactions.

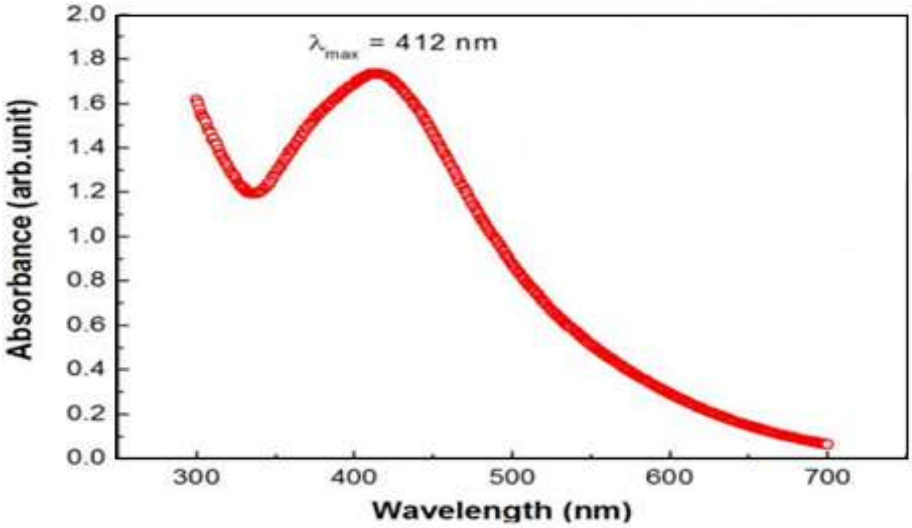

Figure 3. UV-vis spectrum of Ag NPs synthesized with *C. infortunatum* leaf extract [31].

Zingiber officinale or ginger was used to synthesize iron NPs coated onto cotton fabric [32]. The ginger extract was prepared by mixing dried ginger powder in isopropyl alcohol at 80°C for 1 h and the mixture was filtered to collect the supernatant [69]. The green synthesis of the iron NPs (Fe NPs) was conducted by adding the ginger extract to iron(III) chloride ($FeCl_3$) at room temperature. The color change of the solution to black-brown indicated the reduction of Fe^{3+} ions due to the presence of aldehyde and polyphenol groups in the leaf extract [70]. The Fe NP solution was dried to form fine black powder [71-72]. The Fe NPs powder was dissolved in methanol using an ultra sonicator and coated onto a cotton fabric using dip-coating process. The Fe NPs had an average size of 56.2 nm with crystal plane shapes and had antimicrobial properties against gram-positive (*Bacillus subtilis* and *S. aureus*) and gram-negative (*E. coli* and *K. pneumoniae*) bacteria due to the presence of gingerol.

Kumar [33] developed an antimicrobial fabric by synthesizing silver NPs ($AgNO_3$) with *Acalypha indica* leaf extract. The formation of *A. indica*-Ag NPs was indicated by the change of the color of the solution from clear to yellowish-brown. The *A. indica*-Ag NP solution was impregnant over a cotton fabric through the dipping process and dried at 60°C. The morphology of rod shaped *A. indica*-Ag NPs with the size of 100 nm was observed (Figure 1(d)). The coated cotton fabric was efficient against both gram-positive and gram-negative pathogenic microorganisms such as *B. cereus*, *K. pneumonia*, *P. aeruginosa*, *C. albicans*, and *C. glabrata.*

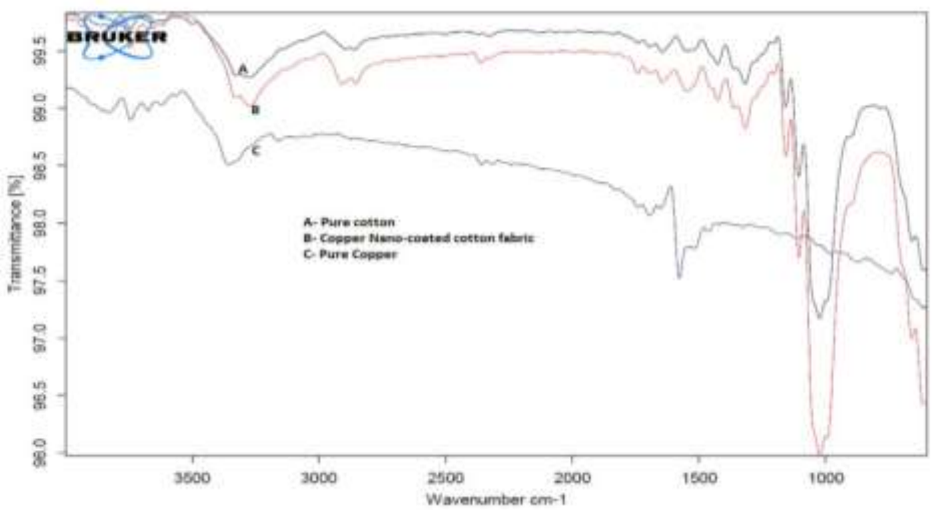

Figure 4. ATR-FTIR spectrum of pure cotton, green copper NPs coated cotton fabric, and pure copper nitrate [34].

The green synthesis of copper NPs with *Tinospora cardifolia* leaf extract was conducted by Sharmaa *et al*. [34]. *Tinospora cardifolia* extract was prepared aqueously by heating 25 g of dried leaves with 50 ml of water till the color of the solution changed from colorless to brown. The copper NPs were prepared by a one-step synthesis process. 2 ml of leaf extract was added into 10 mL of 0.25 M copper nitrate ($Cu(NO_3)_2$) solution at 87°C. The reduction of copper ions was confirmed with the color change of the solution from sky blue to dark green. The presence of alkaloids and steroids (OH), fatty acid (C–O), carboxylic (O–H), ethers (C–O–C), and esters (C–O) were confirmed through FTIR analysis (Figure 4). A cotton fabric was coated with the Cu NPs by dipping process before being washed and dried at normal temperature. The NPs were spherical in shape with the range of particle size of 63.3 – 143 nm. Also, the NPs had a smooth shape, which no cracks or pinholes visualized. The Cu NPs coated fabric was more efficient against gram-positive bacteria (*S. aureus*) than gram-negative bacteria (*E. coli*) because of the complexity in the structure of gram-negative bacteria. It had antimicrobial properties even after 50-cycle washing due to the capping of the Cu NPs with *T. cardifolia.*

The study of the antimicrobial activity of the silver nanocoated cotton fabric to produce good dressing and clothing material was conducted using an aqueous extract of *Allium cepa L* [35]. The aqueous extract of *A. cepa L* was prepared by boiling it with distilled water until a pinkish solution was formed. The synthesis of silver NPs was conducted through a one-step synthesis process. A sudden color change of the solution was observed after the addition of *A. cepa L* extract into silver nitrate solution due to the change in pH from 7.4 to 12.5. The brownish color of the solution was formed after a complete reaction between silver nitrate and *A. cepa L* extract. The cotton fabric was coated with the silver NPs by dipping, washing, and drying processes. *Allium cepa L.* acted as a good reducing and capping agent which produced stable silver NPs. The particle size ranged from 36 to 98 nm with a spherical shape. The silver NP-coated fabric was effective against both gram-positive (*S. aureus*) and gram-negative (*E. coli*) bacteria. However, it was more effective against gram-positive bacteria than gram-positive bacteria due to the cell wall structure of the bacteria strains [73]. The silver NP-coated fabric was economical and has good resistance to washing even after 50 laundry cycles.

A study conducted by Balashanmugam and Kalaichelvan [36] used *Cassia roxburghii DC.* aqueous extract to synthesize silver NPs. The extract was added into silver nitrate solution and kept in a dark place to minimize the photoactivation of silver nitrate. The change in color from pale yellow to dark brown confirmed the conversion of Ag^+ ions to Ag NPs due to the reduction by the capping agent of the plant extract. The cotton fabric impregnated with Ag NPs had high antimicrobial properties against both gram-positive bacteria (*B. subtilis, S. aureus, Micrococcus luteus*) and gram-negative bacteria (*P. aeruginosa, E. coli, E. aerogenes*). The biosynthesis of Ag NPs with *C. roxburghii* exhibited high stability with spherical shapes and sizes of 10 to 30 nm.

Research conducted by Turakhia [37] had shown the antimicrobial and hydrophobic properties of cotton fabrics. *Carica papaya* leaves showed the highest reduction capability against copper sulphate ($CuSO_4$) when compared to other parts of the plant, such as seeds and fruits. The green synthesis of copper oxide (CuO) NPs was carried out by adding 0.01 M $CuSO_4$ solution to the *Carica papaya* leaf extract (volume ratio 95:5) for the reduction process to occur.

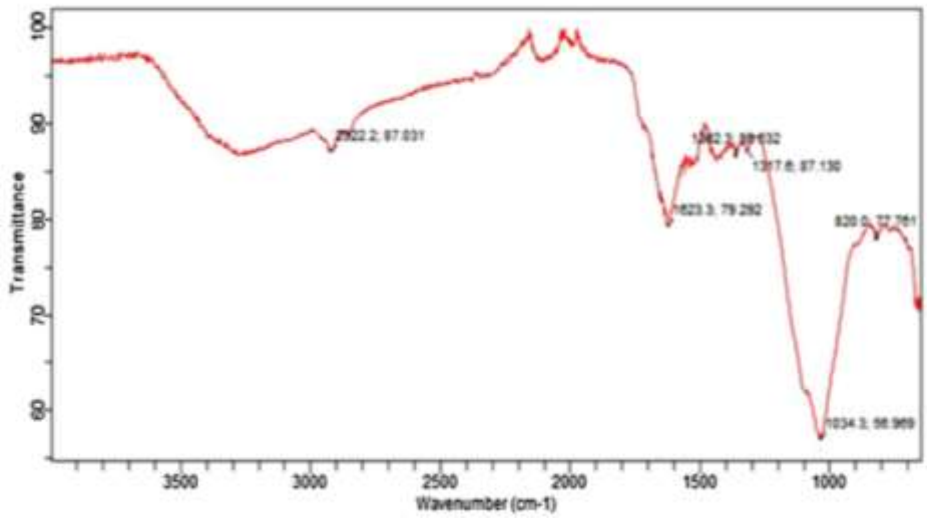

Figure 5. FTIR spectrum of CuO NPs synthesized with *Carica papaya* leaf extract [37].

The color of the colloidal suspension formed was turned green from blue with a brownish-black precipitate. Results from FTIR analysis (Figure 5) had confirmed the presence of carbonyl groups (C=O), phenol, and alcohol groups (OH, H–OH), which has a similar pattern to the synthesized copper oxide using gum karaya. The cotton fabric coated with CuO NPs had an excellent antimicrobial efficiency against the gram-negative bacteria *E. coli* even after 30 cycles of washing. The combination of metal ions with phytochemicals and phenols from the plant extract increased the antimicrobial activity. Besides, the CuO NPs coated fabric also exhibited higher tensile strength and hydrophobicity. Evenly distributed square and rectangle shaped NPs with sizes less than 100 nm were observed on the cotton fabric (Figure 1(e)).

Ruellia tuberosa aqueous extract was used as a reducing agent for the synthesis of CuO NPs [38]. Copper sulphate ($CuSO_4$) was used as a precursor and the biosynthesis of CuO NPs was carried out by the addition of *R. tuberosa* aqueous extract into $CuSO_4$ solution. The color of the solution gradually changed from yellowish green to brownish black after 5 – 7 h of continuous stirring, which indicated the reduction of Cu ions had occurred. The morphology of rod shaped NPs with poly-dispersed form and size ranging from 20 – 100 nm was observed. The cotton fabric treated with CuO NPs showed antimicrobial activities against both gram-positive (*S. aureus*) and gram-negative (*E. coli, K. pneumonia*) bacteria. Besides, the treated fabric also had an efficient photocatalytic degradation of crystal violet (CV) dye under direct sunlight.

The UV protection fabric produced by Ganesan and Prabu [39] involved the use of chloroauric acid as a precursor and *Acorus calamus rhizome* aqueous extract as a reducing agent. The synthesis of Au NPs was conducted by mixing the extract, precursor solution, and pH buffer solution at room temperature until the change of color was observed. The color change of the mixture from light to dark brown confirmed the reduction of Au^{3+} to Au. The pad-dry-cure method was used to coat a cotton fabric. The Au NPs formed higher distribution spherical ball morphology with a size less than 100 nm. The Au NPs coated cotton fabric had higher UV protection efficiency compared to the uncoated cotton fabric. The ultraviolet protection factor (UPF) value for the Au NPs coated cotton fabric was 31.9, while the UPF value for the uncoated cotton fabric was 3.9. The deposition of Au NPs in the spaces between the yarns in cotton fabrics prevents the penetration of UV radiation through the fabrics [74-75]. Besides, the Au NPs coated cotton fabric also had antimicrobial properties against both gram-positive (*S. aureus*) and gram-negative (*E. coli*) bacteria.

Yu *et al*. [40] developed a multifunctional cotton fabric using silver NPs and black rice aqueous extract as reducing agent and dye. Anthocyanins contained in black rice aqueous extract were used as pigments to dye a cotton fabric. The main component of anthocyanins is cyanidin-3-glucoside [76]. The black rice aqueous extract was prepared by soaking black rice dust in deionized water overnight for anthocyanins to be obtained. The multifunctional cotton fabric was produced by pre-treating the cotton fabric in the $AgNO_3$ solution through the padding process. Then, the pre-treated fabric was dyed with black rice aqueous extract and in situ synthesis of Ag NPs, which was done at 60°C to maintain the stability of the structure of anthocyanins. The pH value during the synthesis process plays an important role as it greatly influences the appearance and strength of the color. The coated cotton fabric was in yellow color at pH 10, while at pH between 3 – 7 the colour of the coated fabric was pale yellow. Meanwhile, higher pH values give better washing durability compared to low pH values. The coated Ag NPs cotton fabric also had UV protection and antimicrobial properties against *S. aureus* and *E. coli*. The Ag NPs were evenly distributed on the cotton fabric with an average size range of 50 to 220 nm.

A simple economical green fabrication procedure was developed by Shahid-ul-Islam *et al*. [41] to produce colorful chitosan-modified cotton fabrics using silver NPs with the presence of peanut waste shell extract. Fresh peanut shells were cleaned, air dried, and ground. The powdered peanut shells were boiled and filtered. A chitosan solution was prepared by mixing the solution with 2% acetic acid at 60°C. A cotton fabric was immersed into chitosan solution and padded, cured, and dried. Meanwhile, $AgNO_3$ solution was prepared by using distilled water. The chitosan-cotton fabric was immersed in the $AgNO_3$ solution. The prepared peanut shell extract was added to the solution and heated. The reduction of Ag^+ to Ag occurred due to the presence of polyphenolic moieties in the peanut shell extract, resulting in the color change of the solution [77-78]. The chitosan-Ag NPs coated cotton fabric was yellowish in colour. The increase of $AgNO_3$ and peanut shell extract concentrations changed the color of the fabric from pale yellow to deep brownish yellow. The fastness properties, which are washing and rubbing, of the coated fabric were within the acceptable range between 3 to 4.5. Besides, the coated fabric had an antioxidant activity due to the presence of polyphenolic hydroxyl groups, which have potent radical scavenging activity. Also, it was effective against *E. coli* and *S. aureus*. Spherical shaped and dispersed Ag NPs with sizes less than 60 nm were observed (Figure 1 (f)).

Shahid-ul-Islam *et al.* [42] used silver NPs, chitosan, and pineapple crown extract biomolecules to fabricate a coloring, antimicrobial, and antioxidant linen fabric. Chitosan and pineapple crown extract acted as a bio-reducing and stabilization agent, while $AgNO_3$ was used as a precursor. The pineapple crown was extracted using methanol/water as the solvent. In this one-step synthesis process, $AgNO_3$ solution was added into the chitosan solution, followed by the immersion of the linen fabric into the colloidal mixed solution. Then, the pineapple crown extract was added, which resulted in the color change of the mixture to reddish brown. The linen fabric treated with chitosan-Ag NPs was deep yellow in color. It exhibited strong antibacterial activities against *E. coli* and *S. aureus* due to the presence of chitosan polysaccharide and Ag NPs, which have ionic interactions with the bacterial cell wall, causing leakage of the intracellular matrix, and finally resulting in cell death [79]. The presence of antioxidants like chitosan, pineapple extract biomolecules, and silver ions on the linen fabric enhanced the antioxidant activity through synergism, chelation, and H-donation mechanisms [80]. The morphology of roughly spherical shaped chitosan-Ag NPs with sizes about 10 – 60 nm of was observed.

4. Factors Affecting the Morphology and Size of Nanoparticles

The morphology and size of NPs play an important role as they can influence the NPs performance. According to Salem *et al*. [81], smaller NPs are more efficient than larger NPs because they can penetrate deep into the fabric and tightly adhere to the textile fibers. The smaller NPs also can easily enter the porous fibers through osmotic pressure [82]. There are a few factors which control the morphology and size of NPs, such as temperature, metal salts and plant extract concentrations, and pH.

Low synthesis temperatures produce small sized NPs with less agglomeration particles. According to Rachael and Rajiv [83], large surface area and tendency to stick result in aggregation of NPs. An increase of synthesis temperature results in larger sized NPs, thus enhances the agglomeration of the NPs and increases the frequency of collisions among nucleating atoms, resulting in an exchange of energy. The exchange of energy makes nucleated atoms more active and directly increases the surface mobility [84]. Research conducted by Basri *et al*. [85] have shown the different morphologies and sizes of ZnO NPs when synthesized at different temperatures (Figure 6). Flower-rod shaped ZnO NPs with diameter sizes between 73 – 123 nm were observed at 60°C, while ZnO NPs synthesized at 28°C exhibited a mixture of spherical and rod shaped NPs with diameter sizes between 8 – 46 nm.

The concentration of metal salt and plant extract also influences the size and morphology of NPs. Under sufficient metal salt and plant extract, the size of NPs could increase with the increase of temperature. Insufficiency of metal salt and excess of plant extract result in competition of nucleation and growth for metal salt ions. The lack of metal salt could decrease the size of NPs due to an explosive nucleation [86]. Tavakoli *et al*. [87] has demonstrated that metal salt concentration and aging time influence the morphology of CuO NPs. Spherical shaped NPs with the size of 9 nm were observed with 1 M copper chloride ($CuCl_2$) solution. 6 mM $CuCl_2$ solution resulted in platelet NPs with sharp edges and rod shape and size of 23 nm. The change of morphology of NPs from platelet with sharp edges to spherical and rod shapes was reported with increasing of aging time from 0 to 72 h. This is due to the reduction of surface energy for thermodynamically stabilization purposed [88].

The pH value of plant extracts may control the morphology of the NPs, where the shifting from neutral to basic conditions leads to destabilizing effect of reducing agent component, which results in irregular spherical shape of NPs. Bigger sized NPs can be produced by increasing the pH value. This may boost the homogenous nucleation and intensify the reaction rate of bio-reduction [86]. According to Aziz and Jassim [87], only spherical shaped NPs can be produced with increase of pH value due to the presence of reducing and stabilization agents in plant extracts. Besides, the phytochemical components of plants such as polyphenolic compounds, gallic acids and derivatives are not stable at high pH and result in the formation of individual spherical NPs [88].

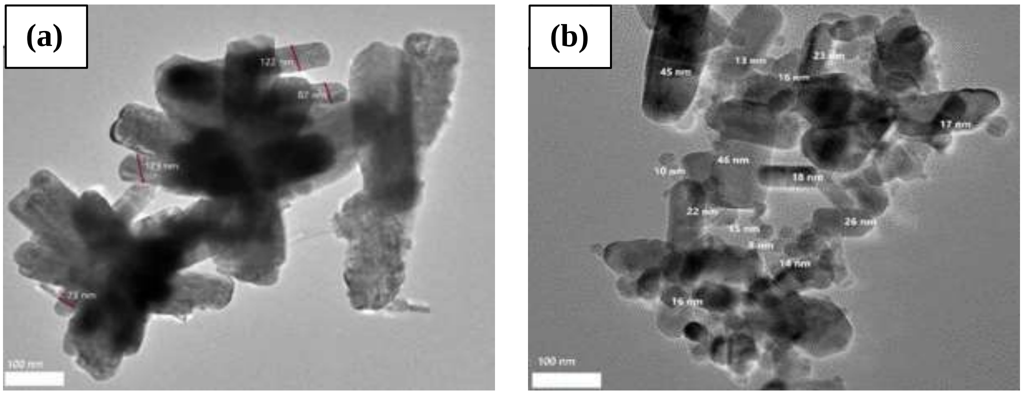

Figure 6. The transmission electron microscopy (TEM) images of ZnO NPs: (a) synthesized at 60°C; (b) synthesized at 28°C (room temperature) [85].

5. Advantages, Limitations and Future Challenges

The "greener" environmental process in nanotechnology is gaining popularity recently due to the awareness of worldwide problems associated with environmental contamination issues. Thus, the application of plant-mediated metal NPs for fabrication has become an alternative in the field of nanotechnology. The development of plant-mediated metal NPs through biosynthesis could reduce the use of chemicals, which will directly reduce the environmental impact. In addition, biosynthesis is energy efficient as the process does not involve the use of much equipment like chemical and physical synthesis processes. Consequently, this will cut the operational cost, and in the long run would benefit the investors in terms of the overall cost of production. Besides, unlike physical and chemical synthesis methods, biosynthesis method of plants is rapid or not time-consuming due to its straightforward synthesis process. The antimicrobial properties of plants could create multifunctional textiles with a combination of other protection properties such as hydrophobicity, antioxidant, and UV protection. In addition, pigments from plants such as carotenoids, anthocyanins and flavonoids can also act as dyes, which give an antioxidant and UV protection properties to the fabric.

However, there are some limitations in merging the plant-mediated metal NPs for fabric into industrialization. The variation of biomolecule contents of plant extracts due to different seasonal and geographical locations will lead to the variation of NPs in every batch. Thus, it can be a major hump for the process and product optimization. In addition, the unawareness of the product performance due to the variation of NPs produced will also give a negative impact on the nanotechnology fabrication industry. Focusing on obtaining the stable NPs by varying the physicochemical parameters of plant substrates is important in order to successfully commercialize the product. Another obstacle that might be faced is the lack of expertise in the research and development (R&D) stage, resulting in poor quality and no assortment of the end product. Besides, it also will lead to the expensive price of the product due to high demand, but less product produced.

CONCLUSION

The synthesis of plant-mediated metal NPs has been discussed to be economical, energy and cost effective, and environmentally friendly over physical and chemical synthesis methods. Owing to the richness of plant biodiversity, their potential as reducing and stabilization agents for NPs synthesis still needs to be explored to enhance its applications in the textile industry. However, there is no doubt about the potential and applicability for NP synthesis to evolve in nanotechnology fabrication industry. Also, fulfilling the demands of consumers in terms of safety, health and comfortability and awareness of the environment have to be considered in producing the fabric. It is hope that with recent progress and ongoing efforts on improving NP synthesis, the implementation of plant-mediated metal synthesis for fabric in pilot plants and its product commercialization will take place in the near future.

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