

**A Review on Green Synthesis of Silver Nanoparticles Conjugated with
Activated Charcoal to form Silver-Charcoal Nanocomposites and its
Applications in Water Filter Membrane**

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ABSTRACT

The increasing interest to carbon and silver nanotechnology and its various applications nowadays caught the attention of many researchers. In this review, the types of plants used for green synthesis of silver nanoparticles and its antimicrobial activity against different types of infectious agents were cited. This challenge boosts the interest of researchers to promote a more eco-friendly way of synthesis of nanoparticles compared to the present ones that can also potentially used in various fields of its application. Occurrence, shape, morphology, and size of silver nanoparticles are also revealed with references. Applications and mechanism of silver nanoparticles in water filtration membranes are also highlighted with suitable references. As charcoal are considered as one of the significant filter types capable of arresting microbial metabolism from odour causing, many articles related to charcoal and its antibacterial mechanisms were reported in this review. Charcoal in water filtration units or in membranes were also described emphasizing the role of activated carbon or charcoal for preventing the growth of bacteria in water or potable water. As nanocomposites are gaining more attention in the membrane filter production industries, in this review, we focussed about the mechanism, synthesis, and application, including their advantages and disadvantages, of charcoal, silver, and silver+ charcoal nanocomposites.

Keywords: Silver, green synthesis, nanoparticles, charcoal, silver+ charcoal nanocomposites

INTRODUCTION

Despite the contemporary improvement of the hygiene in the biomedical (hospitals), education (school/colleges), surrounding environment (air/water), and industry (food/textile/animal husbandry), it is an increasingly important public health issue globally. Today, the world is facing real challenges in meeting increasing demands of drinking water as the available supplies of freshwater are exhausting due to the lack of rain, population, growth, and more stringent health-based regulations (Deshmukha *et al.*, 2019). In particular, the infectious diseases are the major challenges to the human being because of emerging > 300 infectious diseases with a new adaptation. The microbial-based infections are a key cause of the diverse infections because of which > 50% people are dying due to a variety of infections (Jones *et al.*, 2008). The various chemical compounds such as alcohols, quaternary ammonium cation, aldehydes, and oxidizing agents, such as sodium hypochlorite, hydrogen peroxides, iodine, etc., have been used as disinfectant effectively. However, these compounds are suffered from various constraints such as harmfulness, corrosive nature, and bacterial resistance (Rutala and Weber, 2008). Nanoscience is currently widely investigated to be used in water purification. This new interdisciplinary subject is referring to the study of chemical and physical changes at the nanoscale level (1–100 nm). Nanotechnology is currently used for developing new materials and devices, and it is also intensively investigated in water purification. Silver ions and silver-based compounds are the well-known antimicrobial agent for the medicinal importance from the 1000 BCE, and they have been used as an efficient health additive in Chinese and Indian Ayurveda medicine (Deshmukha *et al.*, 2019).

The choice of silver is due to its multiple functions in the medical field. As usual, silver nitrate is used for antimicrobial action a long time, but nowadays, nano-based silver has efficient antimicrobial action because of its physicochemical property in which larger surface to volume ratio resulted into higher surface exposure to the microbes which leads to furnish better antimicrobial activity (Pantic, 2014). In addition, the special properties such as size, shape, and phases play a crucial role of bacterial inactivation or killing of bacteria. These physicochemical properties of the silver nanomaterials and its compounds have foremost applications in the environmental, biomedical, and industrial sectors (Woodward, 1963). Silver nanoparticles (AgNPs) are playing the crucial role in the air/water purifications, in biomedical fields as a therapeutic agent, textile consumer products, as well as wound dressing (Lu *et al.*, 2008). Its bactericidal effects are observed on *Staphylococcus aureus*, *Pseudomonas aeruginosa*, *Escherichia coli*, *Bacillus cereus*, *Listeria innocua*, and

Salmonella choleraesuis due to higher toxic effect to the bacterial cells (Pandey *et al.*, 2014). Therefore, due to its multidomain uses, AgNPs would have the broad spectrum of the biomedical sector for innovative formulation to resist the bacterial growth.

This paper is referring to the use of nanotechnology, specifically the use of AgNPs in water purification purposes. It describes the properties and methods used for obtaining AgNPs and the types of substrates used in general for AgNP filters and it also proposes a new filter structure. As a novel concept, charcoal from natural sources are conjugated with AgNPs to form a metal–carbon composite. These composites are new and not much work has been conducted using this. Hence, the review on nanoparticles and charcoal conjugates would provide knowledge in developing filter materials coated with these composite for effective water treatment process, prevent disease-causing microbial film formation on the membrane, arrest microbial metabolism by penetrating into their cell structures, damage the cell membrane, and inhibit odour-causing metabolisms.

Green synthesis of silver

Owing to the environmental issue, biogenic synthesis of metal and metal oxide NPs is gaining immense attention from the past decades. The reported literature revealed that various plant parts, such as leaf, roots, seed, fruits, and stem etc., have been utilized for the biosynthesis of NPs. The synthesis of NPs is fully dependent on the biomaterials or phytochemicals present in the extract. This section aims to discuss the extract of various plant parts that mediated synthesis of AgNPs and their application as an antimicrobial.

AgNPs were also recently synthesized using several leaf extract of plants such as *Artocarpus altilis*, *Crotalaria retusa*, *Cardiospermum halicacabum*, *Psidium guajava*, *Cassia auriculata*, and *Terminalia chebula*. In 2016, Anandalakshmi *et al.* (2016) reported *Pedaliium murex* leaf extract–mediated AgNPs. The produced NPs were tested against several microbes and displayed highest zone of inhibition (ZOI) of 10.5mm against *Escherichia coli* and *P. aeruginosa* and least activity against *Klebsiella pneumoniae* (8.5 mm).

Recently, Manjamadha *et al.* (2016) have reported ultrasonic–assisted biosynthesis of spherical AgNPs using *Lantana camara* leaf extract. Bactericidal activity of the synthesized AgNPs revealed that it shows excellent antibacterial activity against Gram-positive and Gram-negative bacteria.

Formation of spherical AgNPs using *Maclura pomifera* was achieved in 2017 by Azizian-Shermeh *et al.* (2017). The produced NPs (0.1 mg mL⁻¹ concentration) displayed a

very high ZOI of 23.4 0.1 mm against *Escherichia coli*, which is higher than Ampicillin, a well-known antibiotic drug.

Al-Shmgani *et al.* (2017) prepared AgNPs using *Catharanthus roseus*. The color of the leaf extract changes from yellowish to reddish-brown after adding 2mM AgNO₃ and exposing to heat at 70 °C for 3 minutes indicating the formation of the NPs. AFM displays the crystalline NPs with grains sized 10–88 nm in diameter with mean size of about 49 nm. The authors claimed that synthesized AgNPs enter the cell of microbes that resulted in a disruption of adenosine triphosphate (ATP) production and DNA replication, generation of ROS, and damage the cell structures as earlier observed by Sahayaraj and Rajesh (2011).

Spherical shape AgNPs with diameter in the range 11–47 nm (by TEM analysis) were produced using *Lavandula intermedia*. The AgNPs were found to be most effective against *Escherichia coli* (Elemike *et al.*, 2016).

In another work, a highly crystalline AgNPs were reported to be synthesized from *Canna edulis* (Otari *et al.*, 2017). The NPs showed highest antimicrobial activity against *S. typhimurium* which is closely related to the work carried out by Sumitha *et al.* (2018).

Artemisia vulgaris–mediated AgNPs were reported by Rasheed *et al.* (2017). Antimicrobial test revealed that the AgNPs exhibited significant inhibition activities against tested pathogens with the highest value being recorded against *S. aureus* (18 mm inhibition zone).

Psidium guajava was applied for the production of spherical AgNPs with average dimension of 25 nm. The authors observed that for 100 mg/ml *Psidium guajava*–mediated AgNPs, the ZOI were 18mm and 16 mm against *A. faecalis* and *Escherichia coli*, respectively, whereas ZOI of 13 to 14 mm were recorded at the same concentration against tested Gram-positive bacteria (Wang *et al.*, 2018).

In general, the reduction in the size of the metallic nanoparticles is expected to increase the antibacterial activity because of its significantly large surface area of the smaller nanoparticles. However, the results obtained by Erci *et al.* (2017) using *Thymbra spicata* leaf extract is worth discussing. In their study, higher antibacterial activity of, say, AgNPs-2 (average diameter 70.2 nm) in comparison with AgNPs-1 (average diameter 25.1 nm) was recorded. They reasoned that this could be due to the shape of AgNPs-2, which have triangles, hexagons, spheres, and irregular shapes, whereas AgNPs-1 exhibit mostly spherical formation (Erci *et al.*, 2017).

Silver nanoparticles in water filtrations/membranes

Several researchers have shown that using different methods in preparing silver nanoparticles can lead to very good bacterial activity removal. Material development is enhancing silver nanoparticles deposition on solid materials for the deactivation of microorganisms in water treatment (Mpenyana-Monyatsi *et al.*, 2012). In the case of drinking water treatment, various forms of silver nanoparticles coated on materials/supports were investigated during the years:

Sand impregnated with AgNP is less expensive and effective and can be self-constructed with limited local skills (Mahmood *et al.*, 1993). Zeolites, as cation exchangers in water treatment, have increased in usage due to their availability, low cost, high surface area and sorption capacity, chemical inertness, and low or null toxicity for human (Matsumura *et al.*, 2003). Fiber glass needs only a small amount of Ag for a strong bactericidal response against *Escherichia coli*. The evidence suggests that the adhered Ag nanoparticles to the fiber glass substrate are active against bacteria in a filtration application. Falling desorption rates suggest the adsorption of Ag by the *Escherichia coli* (Nangmenyi *et al.*, 2009). Activated carbon filter was used as support for silver nanoparticles from silver nitrate solutions using glucose as a reducing agent. The batch method showed that with the increase in contact time and the decrease in the bacterial count, complete inhibition was achieved. In the column test, the prepared AgNPs/AC showed a complete bacterial reduction in synthetic water and actual polluted water samples. Data revealed a direct relation between Ag-NPs concentration and contact time toward bactericidal activity against *Escherichia coli* which represented as an indicator for microorganism's pollution, whereas the concentration of AgNPs and contact time increased, bacterial counts of *Escherichia coli* decreased, and inhibition zone increased for all bacterial concentrations used (El-Aassar *et al.*, 2013).

The blotting paper allows microorganisms to meet the biocide, but attachment to the fiber surfaces limits the levels of silver in the effluent water. The primary purification mechanism is not the removal of bacteria from the effluent by filtration but rather the deactivation of bacteria as they percolate through the AgNP paper structure. Consequently, the filter effluent contains dead bacteria. The large pore size in the filter paper allows for reasonably rapid flow by gravity, without the need for pressure or suction (Dankovich and Gray, 2011). Cellulose filter paper was coated with silver nanoparticles using a chemical

reduction method, with sodium borohydride: silver nitrate ratios of 2:1 and 10:1. The efficiency of this cellulose filter in *Escherichia coli* removal ranged from 99% to 100%. Moreover, the performance of the silver nanoparticle-coated cellulose filter paper was evaluated for silver in the effluent and compliance with the drinking water quality standards. Although further research is still necessary, the performance of silver nanoparticle-coated cellulose filter paper and its short and easy preparation method suggests its suitability as an antibacterial water filter during emergency situations (Praveena *et al.*, 2016). Ceramic filters impregnated with silver nanoparticles were investigated in the laboratory and in the field in Guatemala. The addition of silver nanoparticles to the ceramic filters improved the performance for all mass percentages of sawdust relative to filter media without nanoparticle treatment. Filters with higher porosity achieved higher bacteria removal than those with lower porosity. After laboratory testing, ceramic filters were manufactured and distributed to 62 households. Over the course of the study, the average percent reduction in total coliforms and *Escherichia coli* was around 90% and the average effluent concentration of ionic silver was 0.02mg/L (Kallman *et al.*, 2011).

Polyurethane foams can be used for coating silver nanoparticles in diverse forms. This material can be washed, dried, and stored for extended periods without the loss of nanoparticles. The performance of the material as an antibacterial water filter was checked, and no bacterium was detected in the output water when the input water had a bacterial load of 1×10^5 – 1×10^6 CFU/mL. The antibacterial action was also checked online for a flow rate of 0.5 L/min, and no bacterium detected, which suggests that domestic use of this technology is possible. The results are in line with the WHO requirements for drinking water. The experiments suggest the possibility of the use of this material in drinking water purification, air filtration, domestic and industrial air quality management, antibacterial packaging, etc. The chemicals involved in the synthesis of nanoparticles are commonly available, cheap, and nontoxic (Prashant and Pradeep, 2005).

Charcoal against microbes

One of the most widely used nanoparticles for water purification is activated carbon due to its large surface area and high adsorption capacity (Ortiz-Ibarra, *et al.*, 2007). Activated carbon has proven to remove bacteria such as *Pseudomonas aeruginosa* and *Escherichia coli* from fresh and potable water systems (Percival and Walker, 1999). Despite electrostatic repulsion between negatively charged microorganisms and carbon surfaces, microorganisms attach to activated carbon particles through strong Lifshitz van der Waals

forces (Jucker, *et al.*, 1996). Potable water systems are considered low in ionic strength, so electrostatic interactions can offer the possibility of enhancing the efficacy of activated carbon to remove microorganisms from water by positive charge modification of the carbon surfaces. Once there is a charge reversal, the electrostatic attraction between negatively charged microbial cell surfaces and positively modified carbon particles will be strong (Shi *et al.*, 2007). Moreover, modification in the activated carbon particles by coating with a quaternary ammonium compound gives the activated carbon particles bactericidal properties and decreases the possibility of biofilm growth. In addition to the microorganisms charge, the hydrophobicity of the surfaces that come in contact with microbes is important in adhesion (Bos, *et al.*, 1999). Activated carbon (AC) has been widely used for the removal of various organic and inorganic pollutants in wastewater or gaseous media because of its large surface area and high adsorption capacity (Ortiz-Ibarra, *et al.*, 2007). Activated carbon has proven to remove or adsorb large amount of bacteria such as *Pseudomonas aeruginosa* and *Escherichia coli* from fresh and potable water systems (Quinlivan, *et al.*, 2005). More recently, several natural and engineering nanomaterials have also been shown to have strong antimicrobial properties, including chitosan (Qi *et al.*, 2004), silver nanoparticles (Morones *et al.*, 2005), and carbon nanotubes (Lyon *et al.*, 2006),

Charcoal in water filtrations

Water that is used by the public should be wholesome and must be free from diseases producing bacteria, poisonous substances, and excessive amount of minerals and organic matter. To achieve this, a good filtration system is essential for water treatment. When used as a filter material, charcoal traps impurities in water including solvents, pesticides, industrial waste, and other chemicals. Research also shows that charcoal has the potential ability to remove dissolved iron, turbidity, and pathogenic organism from drinking water. Also, the charcoal-based filter resulted in tastier water by enriching water with minerals, such as sodium and potassium. Moreover, charcoal is readily available, affordable, disposable, and inexpensive material. It is relatively cheap and easier to handle. Charcoal as a filter material performs better when it is of good quality. The cost of charcoal depends on its quality. The higher the quality of the charcoal, the higher is its cost (Awwal Musa *et al.*, 2020).

The use of carbon in the form of charcoal has been used since antiquity for many applications. In Hindu documents dating from 450 BC, charcoal filters are mentioned for the treatment of water. Charred wood, bones, and coconut charcoals were used during the 18th and 19th centuries by the sugar industry for decolorizing solutions. Activated carbon is a

material prepared in such a way that it exhibits a high degree of porosity and an extended surface area (Bandyopadhyaya *et al.*, 2008). A typical carbon particle has numerous pores that provide a larger surface area for water treatment. During water filtration through activated carbon, contaminants adhere to the surface of the carbon granules or become trapped in the small pores of the activated carbon. This process is called adsorption. Activated carbon filters are efficient in removing certain organic materials (such as unwanted taste and odors and micropollutants), chlorine, fluorine, or radon, from drinking water or wastewater. However, it is not effective for other contaminants (Cook *et al.* 2003).

Activated carbon filtration is commonly used in centralized treatment plants and at the household level to produce drinking water and in industries to treat effluents. It is also an upcoming treatment applied for the removal of micropollutants both in drinking water production and for the purification of treated wastewater before disposal. There are two basic types of water filters: particulate filters and absorptive/reactive filters. A particulate filter excludes particles by size, and absorptive/reactive filters contain a material (medium) that either adsorb or react with a contaminant in water. The principles of adsorptive activated carbon filtration are the same as those of any other adsorption material. The contaminant is attracted to and held (adsorbed) on the surface of the carbon particles. The characteristics of the carbon material (particle and pore size, surface area, surface chemistry, etc.) influence the efficiency of adsorption. The characteristics of the chemical contaminant are also important. Less water-soluble compounds are more likely to be absorbed into a solid. A second characteristic is an affinity that a given contaminant has with the carbon surface. This affinity depends on the charge and is higher for molecules possessing less charge (Quinlivan and Knappe, 2005). If several compounds are present in the water, strong absorbers will attach to the carbon in greater quantity than those with weak adsorbing ability. Activated carbon filtration is recognized by the water quality association as an acceptable method to maintain certain drinking water contaminants within the limit of the EPA National Drinking Water Standards (Sutherland *et al.*, 2004).

Silver and charcoal composites

Silver nanoparticles also compatible in different nanoparticles that make a new product or composite with more new uses and more efficient inventions. Here in nanotechnology, even a smallest nanoparticle could bring more efficient scientific results in different applications because of the unique nanoproperties that they possess (Shen *et al.*, 2013). Silver and carbon, as one of the common nanoparticles studied for the past years, are

discovered to have wide application in electronics, medicine, water as a disinfectant, air pollution, and chemical sensors (Zhang *et al.*, 2016) and as an antibacterial agent (Saxena and Tripathi, 2010). However, the physical limitation of carbon and silver nanoparticles is still present, such as the oxidation of carbon nanoparticles with oxygen which degrades its structure and hence produce inefficiency to its application, the optical property of silver and carbon nanoparticle, the chemical sensitivity of both silver and carbon, antibacterial property of silver, and bioimaging property of carbon quantum dots and many others; to combat these problems, scientists developed composites called silver–carbon nanoparticles. In line with these, various methods were developed such as adsorption (Luo *et al.*, 2010), adsorption: ultrasonication and adsorption: coating (Walid and Hong, 2013), and spray pyrolysis (Larrude *et al.*, 2014). Carbon nanoparticles have a larger surface area and adsorption capacity compared to silver nanoparticles. This is the reason why the figure below shows that silver nanoparticles adsorb or adhere in the surface of the carbon nanoparticles. In addition, the large surface is carbon that makes it feasible with capping for more stable nanoparticle composites (Shen *et al.*, 2013).

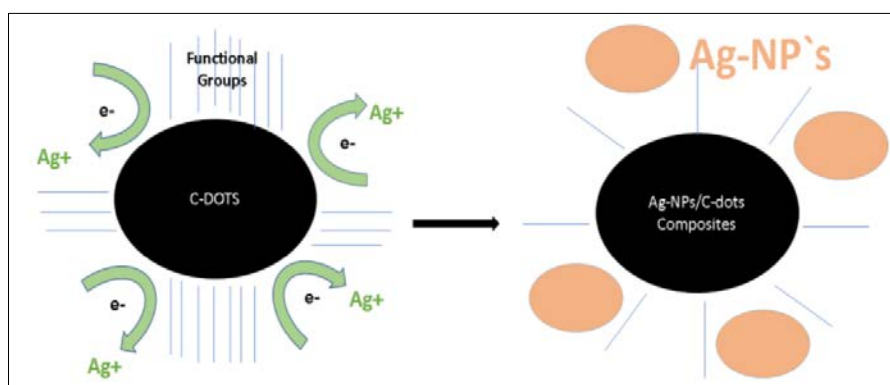


Fig. 1: Schematic representation of silver–carbon nanoparticle composites

Adapted from (Shen *et al.*, 2013). From left to right, the carbon nanoparticle, as dots, undergo adsorption and are heated at 50°C for 5 minutes to produce silver–carbon nanoparticle composites.

Improvements in nanotechnology make eco-friendly and cost-effective solutions to the problem of drinking water contamination problem feasible. Activated charcoal, which is widely used for water purification, can be more effectively used when reinforced with nanoparticles. For antimicrobial water treatment, activated charcoal can be reinforced with silver nanoparticles (SNPs) for best results. Among various methods for the synthesis of nanoparticles, green synthesis mediated by phytoconstituents is preferable, as it is both cost-

effective and environment friendly. Here, we report a green method for synthesizing SNPs from musk melon fruit extract and activated charcoal from coconut shells. The prepared activated charcoal was reinforced with the green-synthesized SNPs and used in microbial water filtration. The efficiency of nanosilver reinforced activated charcoal was higher than virgin-activated charcoal proving improved filtration against microbes after successful reinforcement (Sherly Arputha Kiruba *et al.*, 2015).

Sherly Arputha Kiruba *et al.*, (2013) in another study, green synthesis of biocidal silver-activated charcoal nanocomposite for disinfecting water, revealed significant findings. The researchers thus synthesised and applied the produced silver-activated charcoal (Ag-AC) nanocomposite toward fabrication of antimicrobial water filtration columns. The microbial filtration efficiency of the nanocomposite was found to be higher when compared with the virgin-activated charcoal even with reusage. The antimicrobial property of the silver nanoparticle was tested in fixed bed water filtration columns containing Ag-AC nanocomposites. The use of Ag-AC nanocomposite is advantageous because AC alone cannot be used effectively for microbial filtration and silver nanoparticles alone cannot be used in a column as it needs a substrate to be efficient. Thus, a nanocomposite-filled column was found to be effective against all the four bacteria tested. When compared against normalized values of the control (activated charcoal; AC), the Ag-AC nanocomposite showed highest bactericidal activity against *P. aeruginosa*. The reusability of the Ag-AC column was also checked by passing known colony forming unit (CFU) of bacteria through the column followed by assaying the efficiency of antimicrobial activity. The results indicate that the efficiency of Ag-AC column was maximum against *Escherichia coli* when the column was tested for reusability. With nanocomposite formation, the efficiency of filtration and biocidal activity of AC is enhanced. This is evident from the reusage efficiency of water filtration fixed bed columns (Sherly Arputha Kiruba *et al.*, 2013).

CONCLUSION

In this review, the need for green synthesis of silver nanoparticles using different types of plant extracts and its antimicrobial action against infection causing bacteria were presented. As charcoal has the ability to arrest microbial metabolism which involved odour-causing and infection-causing abilities, the antibacterial action of charcoal was also highlighted. More attention was given to the need of preparation and application of nanocomposites in different fields. Hence, silver nanoparticles after conjugated with charcoal particles were considered as novel nanocomposites. The importance of nanocomposites

containing silver and charcoal was highlighted from different authors' perspectives and presented in this review. More research on this is to be performed using this nanocomposite in our research, and the findings will be published as our future findings. Expected results would be emphasizing its antimicrobial properties against water-borne pathogens and its filtration abilities after coupling with membrane filters.

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