

Silver nanoparticles-based composite for dye removal: A comprehensive review

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ABSTRACT

Organic dyes are a severe threat to aquatic life. Most dyes are toxic, non-biodegradable, and likely carcinogenic. Dyes are usually removed from water via adsorption using adsorbents like wood, seaweed, algae, etc. However, traditional treatment methods are ineffective to an extent. Nanoparticles have emerged as excellent materials for dye removal due to their superb surface properties and chemical reactivity. Many different adsorbents are used today; one example is nanocomposites with embedded silver nanoparticles. In nanocomposites, silver nanoparticles embedded on graphene oxide sheets, carbon nanotubes, cellulose, orange peel, biopolymers, etc., are reported. The solid and hydrogel-based metallic and bimetallic silver nanoparticle-based nanocomposites are successful enough to degrade dyes. These nanocomposites had high reusability, stability, high surface area, tunable properties, selectivity, cost-effectiveness, excellent processability, and recyclability. For instance, AgNPS/alginate composites reported high reusability with recyclability up to 25 times. This article sheds light on the use of biopolymer composites with embedded Ag-NPs for effective dye removal. Various preparation methods as well as the kinetics of the reaction are also discussed.

1. Introduction

Water pollution has caused an increased global demand for pure and safe drinking water—moreover, polluted water results in epidemics in many countries (Jaspal and Malviya, 2020). Waterborne diseases are widely spread due to polluted water (Sharma et al., 2020). The presence of organic pollutants causes health issues like carcinogenesis in the long run in humans and animals. It is also noted that water pollution has increased mortality (Sarkar et al., 2017). Even though water pollution occurs through various industrial means, the textile industry is one major factor contributing to a large share of pollutants. Dyes from textile industries used in various products to impart colour prove a significant factor. Hundreds of pigments and dyes are used industrially, and the number is still counting (Sriram et al., 2020; Berradi et al., 2019). The dye effluents have increased soluble solids, pH, colour, salts, metals, BOD (biological oxygen demand), and COD (chemical oxygen demand)

in water bodies (Yaseen and Scholz, 2019). The presence of increased dye amounts poses a threat for biota, as they contain toxic substances like heavy metals and aromatics. For example, the presence of -N=N- bond makes synthetic dyes recalcitrant and carcinogenic; moreover, their complex aromatic structure makes them hardly biodegradable (Homaeigohar, 2020). Therefore, it is obligated to treat these textile effluents before hitting the water resources (Berradi et al., 2019).

Wastewater treatment of textile effluents was carried out using various chemical, physical, and biological treatment processes. Dyes are highly stable under different conditions like light, aerobic digestion, heat, and oxidizing agents. Crini and Lichtfouse (2019) recently compared the techniques and methods used for the treatment of dyes. Various treatment processes like coagulation, reverse osmosis, ion exchange, flocculation, activated carbon adsorption, incineration, ozonation, advanced oxidation, photocatalysis, filtration, biopolymeric hybrid membrane technology, and electrochemical oxidation are discussed.

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However, most of these methods are expensive and also exhibit disadvantages like (i) longer reaction time and bad odour; (ii) formation of sludge which requires other treatment processes; (iii) production of toxic gases which are volatile in case of incineration (Nagajyothi et al., 2020; Mamba et al., 2021). Among these methods adsorption technique is the most sought and among the different adsorbents, activated carbon is widely used. However, activated carbon is ineffective for treating dispersed dyes (Crini and Lichtfouse, 2019).

There has been a rise in the use of many low-cost adsorbents for dye removal from materials like agricultural solid wastes, by-products from industries, and clays (Kanamadi et al., 2006). In recent years, nanomaterials are also used as adsorbents for water treatment, including carbonaceous ones like graphene, carbon nanotubes (CNTs), activated carbon etc.; metallic nanomaterials like ZnO, TiO₂, etc.; bio-nanomaterials like chitosan. Among these, CNTs and activated carbon are the most widely used nano-adsorbents (Vijayakumar et al., 2019). Fig. 1a lists various types of previously published nanomaterials (Chudasama et al., 2016).

Hydrogels are also implemented for the removal of dyes from wastewater. Hydrogels are hydrophilic polymers, they have a higher affinity to retain water, thus increasing their volume (Fig. 1b) (Zhou et al., 2017; Sheikh et al., 2020). The nature and the degree of cross-links determine the coherence of these gels. These gels can be designed into various dimensions like nano and microparticles, slabs, coatings, etc. Hydrogels are similar to natural tissues in their texture and water-retaining capabilities. Polymeric hydrogels have a fantastic ability for reversible swelling under different conditions like ionic concentration, pH, and temperature (Shalla, et al., 2018). This property is very much utilized in the waste water treatment process; i.e., hydrogels are used as adsorbents for dye removal. The presence of a porous, physically well-defined 3-D structure and active functional groups helps them to

capture dyes and metal ions from wastewater.

The functional groups like amine, sulfonic acid, carboxylic acid, and hydroxyl groups in hydrogels acts as complexing agents for removing pollutants from aqueous solutions. The swelling up of the hydrogels (they can swell several times their original size) entraps the dyes onto their polymer network. Furthermore, to improve the adsorption capacity, nanoparticles are entrapped into the hydrogel-like beads, films, or nanocomposites as hybrids; the surface morphology of nanoparticle (NP) loaded copolymer matrix is shown in Fig. 1c (Bhangi, and Ray, 2020). *This review envisions the synthesis and use of silver nanoparticle-based polymeric materials for the successful removal of recalcitrant dyes from the water system, the process is efficient, eco-friendly, and reusable.*

2. Dyes

Dyes are compounds used in various sectors to colour textiles, paper, plastics, rubber, etc. William Henry Perkin first discovered synthetic dyes in 1856 named Mauveine, an organic aniline dye. Dyes are synthetic and natural (from plants, animals, and minerals), but synthetic ones are widely used. They have an aromatic molecular structure due to hydrocarbons, such as benzene, toluene, anthracene, xylene, naphthalene, etc. (Zare et al. 2018; Tara et al., 2019). Dyes also contain auxochromes (-NH₂, -NR₂, -NHR, -Cl, -OH, -COOH) and chromophores (=C=O (carbonyl), -N=N- (azo), C = NH, =C=C=, -CH = N-, NO or N-OH (nitroso), NO-OH or -NO₂ or (nitro) and C=S (sulphur)). Chromophores, the electron receivers impart colour, while auxochromes, the electron donors intensify the colour onto the substrate by improving the adhesion and solubility of colour to the substrate. Most dyes are soluble in water, imparting little colour, while certain dyes, even at lower concentrations, impart darker colours. The chemical structures of some of the important synthetic dyes are shown in Fig. 2 (Tara et al., 2019).

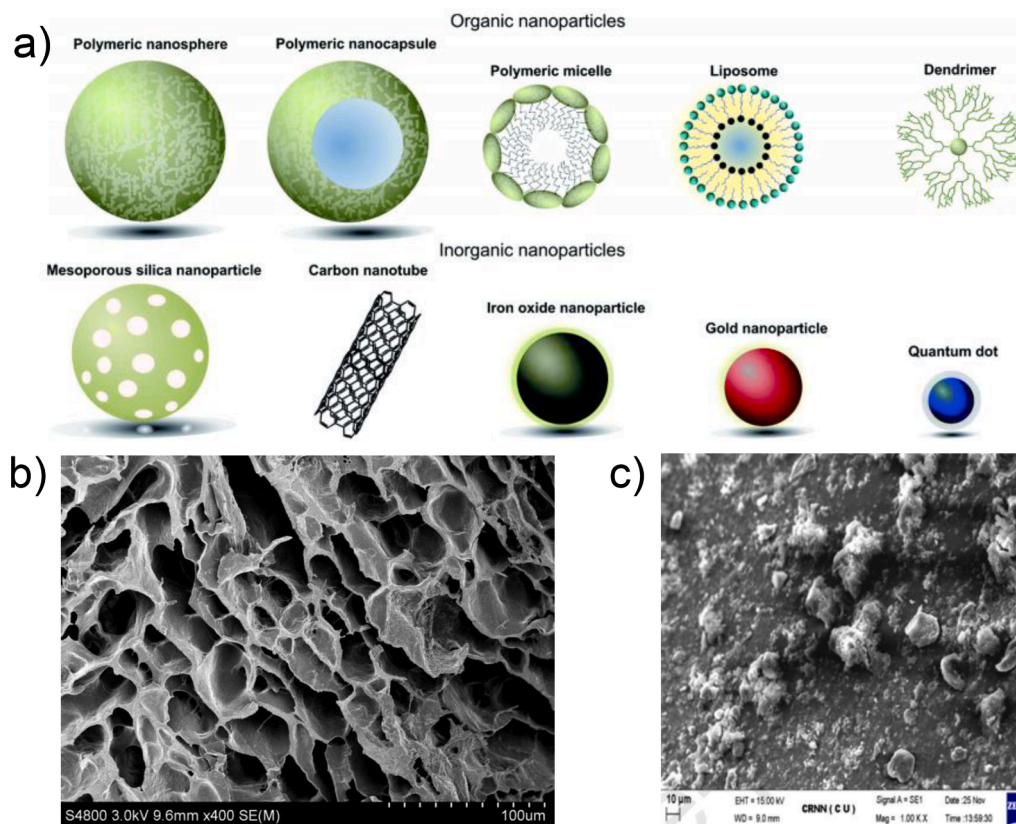


Fig. 1. a), Various types of nanomaterials (Chudasama et al., 2016). b) SEM image of nano-TiO₂/chitosan/poly(N-isopropylacrylamide) composite hydrogel surface. Adapted with permission from Zhou et al. (2017), License Number: 5385750965324. c) SEM image of the G2NP gel surface. Adapted with permission from Bhangi and Ray (2020), License Number: 5385760060385.

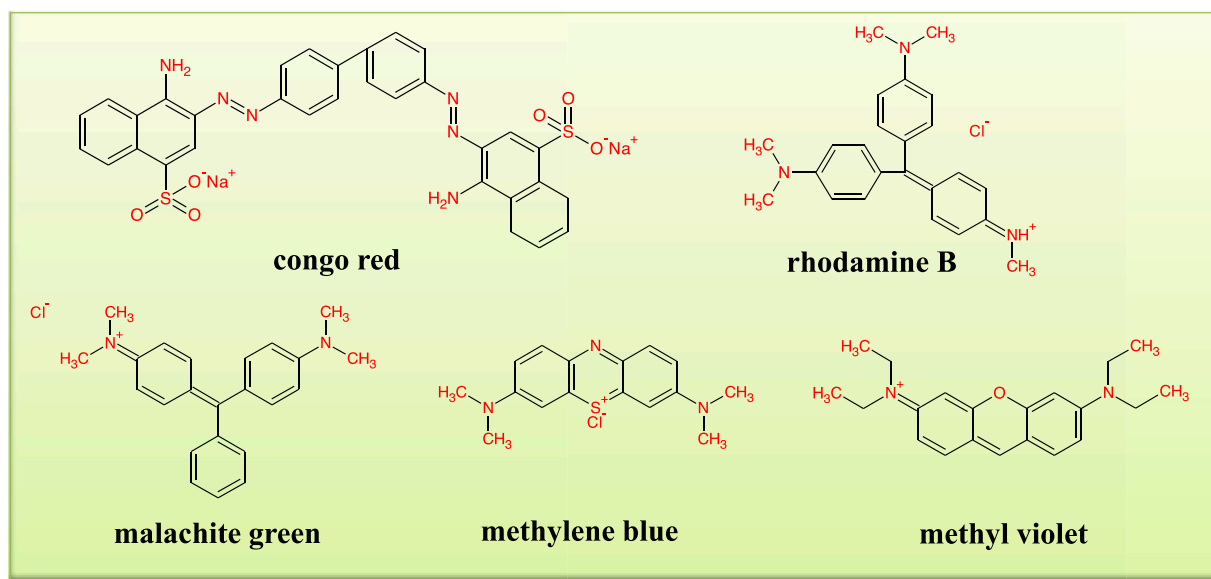


Fig. 2. Chemical structures of some of the synthetic dyes.

3. Classification of dyes

Dyes are vast and diverse and hence classified based on different criteria. They are classified depending on the source, chemical structure, particle charge in solution, and applications.

Based on the source, dyes are classified into natural and synthetic dyes. Natural dyes are obtained from plants (Indigo, Alizarin, Logwood, etc.), animals (Cochineal dye, Tyrian purple, etc.), and minerals (Iron Buff). They could not meet the industrial needs and hence synthetic dyes were developed (Tara et al., 2019). Another grouping of dyes is acidic and basic dyes. Acidic dyes are salts of phenolic, carboxylic, or sulphuric groups. They are soluble in water and are capable of attaching fully or partly to substrates like silk, nylon, wool, and acrylic fibers (Natarajan and Ponnaiah, 2017). Congo Red is the most commonly used acidic dye in various industries. It is a sodium salt of benzidinediazo-bis-1-naphthylamine-4-sulfonic acid and forms strong non-covalent bonds with cellulose fibers. Basic dyes are cationic dyes with azine, monoazoic, and diazoic compounds. The most common basic dye is malachite green, which imparts green colour with a chemical formula of $C_{52}H_{54}N_4O_{12}$. It is usually used to colour leather, jute, wool, paper, etc. It also acts as a food colouring agent. Rhodamine B is also a basic dye with a general formula of $C_{28}H_{31}ClN_2O_3$. It is used to colour cosmetics, paper, and textiles. Another commonly used basic dye is methylene blue, an aromatic molecule with a chemical formula of $C_{16}H_{18}N_3S$. Apart from being used in different industries; they are also used as indicators for oxidation–reduction reactions.

Based on their particle charge dyes are classified as anionic (acid, direct, and reactive dyes), cationic dyes (basic dyes), and non-ionic dyes (disperse dyes) (Zhou et al., 2019). Other classifications of dyes based on their chemical structure into azo dyes, anthraquinone dyes, indigo dyes, xanthene dyes, nitrated and nitrosated dyes, diphenylmethane and triphenylmethane dyes, and phthalocyanine (Berradi et al., 2019; Homaeigohar, 2020; Hasanpour and Hatami, 2020). Dyes are also classified based on their application to the substrate as (i) direct dyes (e. g., Dylon dyes), (ii) basic dyes (e.g., French Silk dyes), (iii) vat dyes, (iv) disperse dyes, (v) pigment dyes, etc. (Zare et al. 2018).

4. The effect of dyes on health and the environment

Industrial effluents that contain dyes are mostly introduced into different water sources like rivers, wades, oceans, and seas. These industrial dyes are mostly non-biodegradable; hence pose a severe risk to

the environment, altering the typical ecological system. Wastewater diffused into the water bodies changes the colour and odour of water, thus making it toxic (Sharma et al., 2018). Prevention of sunlight penetration into the aquatic system is one major factor that leads to more severe issues, the major one being reduced production of photosynthesis by aquatic plants (Hassan and Carr, 2018). Most of the dyes are carcinogens, mainly azo dyes, and can cause diseases like dermatitis, allergies, skin irritation, mutations, and cancer in humans and animals. Different mineral elements like phosphates, nitrites, nitrates, etc., trigger eutrophication in plants, and their long-term presence have bio-accumulation hazards. These dyes also persist as pollutants throughout the food chain, where organisms at the higher trophic levels are highly contaminated than those in lower trophic levels (biomagnification) (Lellis et al., 2019). Moreover, azo dyes that do not bind to the fabrics are released into the water source, which in turn, is used for irrigation, affects the soil microbes, and hinders the germination and growth of plants (Rehman et al., 2018).

Dyes tend to be recalcitrant in aerobic environments, making them non-biodegradable, leading to bio-accumulation. Complex metal dyes, a type used in the textile industry, have higher resistance and shelf life of 2–13 years. When released into the aquatic system, these metal cations accumulate in fish gills due to negative charges, thereby accumulating the fish's other tissues. This reaches humans through the food chain causing many ailments (Khan and Malik, 2018). Chromium in these textile dyes produces oxidative stress, thereby affecting plant growth, especially photosynthesis and CO_2 .

Many dyes reveal mutagenicity. One such example is Azure-B, which can intercalate with the DNA and duplex RNA (Fig. 3) (Haq et al., 2018). It is found that most of the azo and nitro-type dyes exhibit carcinogenicity (Mondal et al., 2018). Crystal violet, widely used for intense colour, causes mitotic poisoning, inducing chromosomal aberrations. This results in tumours in fish, and digestive and respiratory disorders in humans (Mani and Bharagava, 2016). The presence of dyes in water bodies showed great colouration, high levels of BOD, COD, TOC (total organic carbon), TSS (total suspended solids), and high pH levels (Kishor et al., 2021). Synthetic dyes cannot be purified by biodegradation, and hence different combined treatment processes are undertaken.

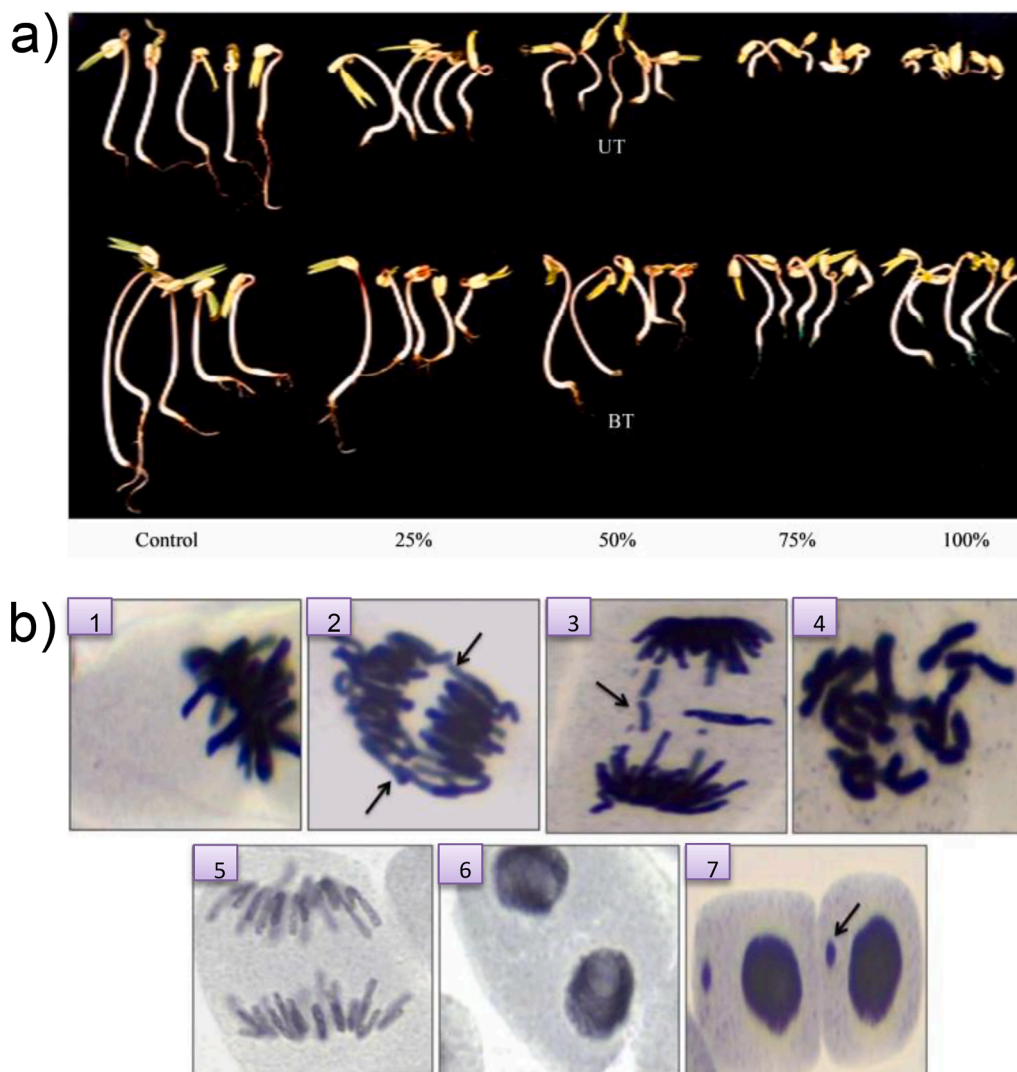


Fig. 3. a) Effect of Azure-B dye on the growth of *V. radiata* species (untreated-UT and bacterial treated-BT), b) Various mutations in the meristematic cells of *A. cepa* caused by Azure-B dye, 1) stickiness of chromosomes, 2) anaphase bridge, 3) fractured chromosomes, 4) c-mitosis, 5) vagrant chromosome, 6) binucleated cell and 7) micronucleated cell. Reproduced with permission from [Haq et al. \(2018\)](#), License Number:5385761256859.

5. Silver nanoparticles (Ag-NPs) based composites for wastewater treatment

5.1. Introduction of Ag-NPs

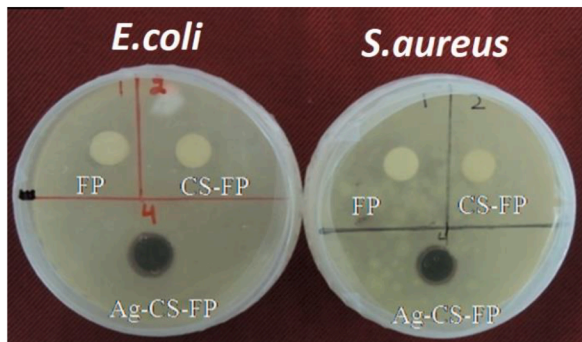
Metal and metal oxide nanoparticles are finding massive applications in different industrial fields in the current era. Wastewater treatment is one such process that exploits the use of nanoparticles as catalysts due to their large surface area ([Ali et al., 2018](#); [Kamal et al., 2020](#)). They are also applied in the field of biomedical applications due to their antibacterial properties ([Fig. 4a-b](#)) ([Haider et al., 2018](#); [Roy et al., 2018](#)). In particular, Ag-NPs are the most widely used ones owing to their unique plasmon, optical, and thermal properties. Ag-NPs can be synthesized in various shapes ([Fig. 4c](#)) ([Srikanth et al., 2016](#)). The advantages of using silver nanoparticles when compared to other counterparts are their cost-effectiveness, stability under ambient conditions, broad absorption spectrum, chemical stability, non-linear optical behaviour, and antimicrobial properties ([Chouhan 2018](#); [Pomal et al., 2021](#)).

Apart from catalyst and biomedical uses, the use of silver nanoparticles in sensors is widely known ([Chouhan 2018](#); [Pomal et al., 2021](#)). When silver nanoparticles are used as catalysts (as they are better photocatalytic materials), there is a need for strong support to

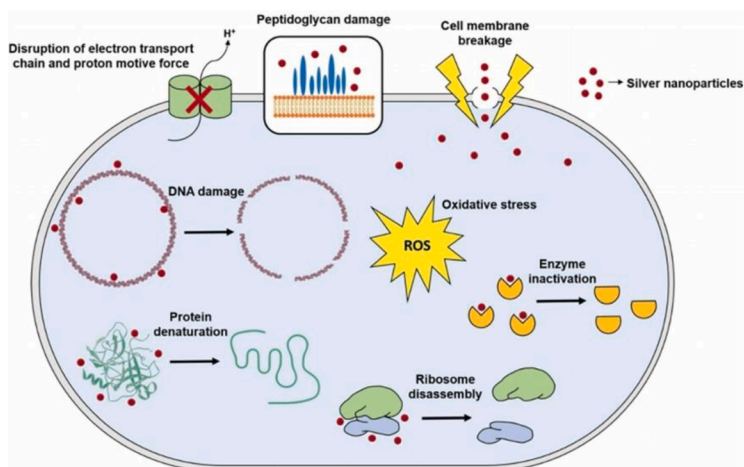
immobilize these catalysts, which is achieved by the use of polymers ([Farooqi et al., 2019](#)). Silver nanoparticles embedded into a polymeric matrix gives the stabilization needed for these otherwise highly active catalysts. Also, the use of nanocomposites enhances the effects of these catalysts ([Fiorati et al., 2020](#)).

Synthesis of Ag-NPs could be attained by many different approaches like physical, chemical, biological, and photochemical methods, and each of these methods had its own set of advantages and disadvantages. Traditional methods like physical methods involved ball milling, sputtering, arc discharge, laser ablation, and evaporation–condensation ([Irvani et al., 2014](#)). The physical method was advantageous as there were no chemicals used, while higher energy consumption, lack of uniformity, and low yield were the disadvantages. Chemical methods involved micro-emulsion, hydrothermal, electrochemical, photochemical, sol–gel, and co-precipitation methods ([Irvani et al., 2014](#)). Chemical methods were advantageous as compared to physical methods due to the high yield. The cons include the use of harmful chemicals in production. To overcome these shortcomings, environmentally friendly methods were used and were termed “green synthesis,” which is cost-effective. Green synthesis of Ag-NPs employed microorganisms, plant tissues, marine organisms, etc. ([Fig. 5](#)). The pros of this method include decreased time, highly stable nanoparticles, and vast biological

a)



b)



c)

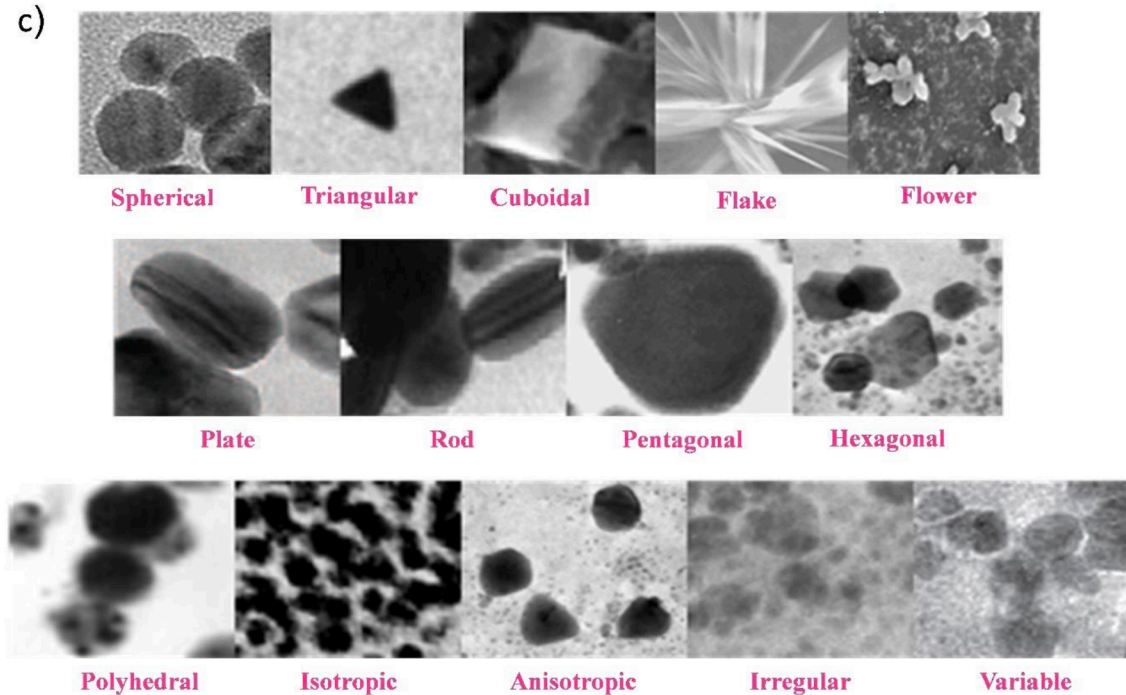


Fig. 4. a) Antimicrobial activity of Ag-CS-FP composite. Reprinted with permission from Haider et al. (2018), License Number: 5385770593088. b) Mechanism of inhibiting microbial growth by silver nanoparticles (Roy et al., 2018). c) Different shapes of Ag-NPs (Srikar et al., 2016).

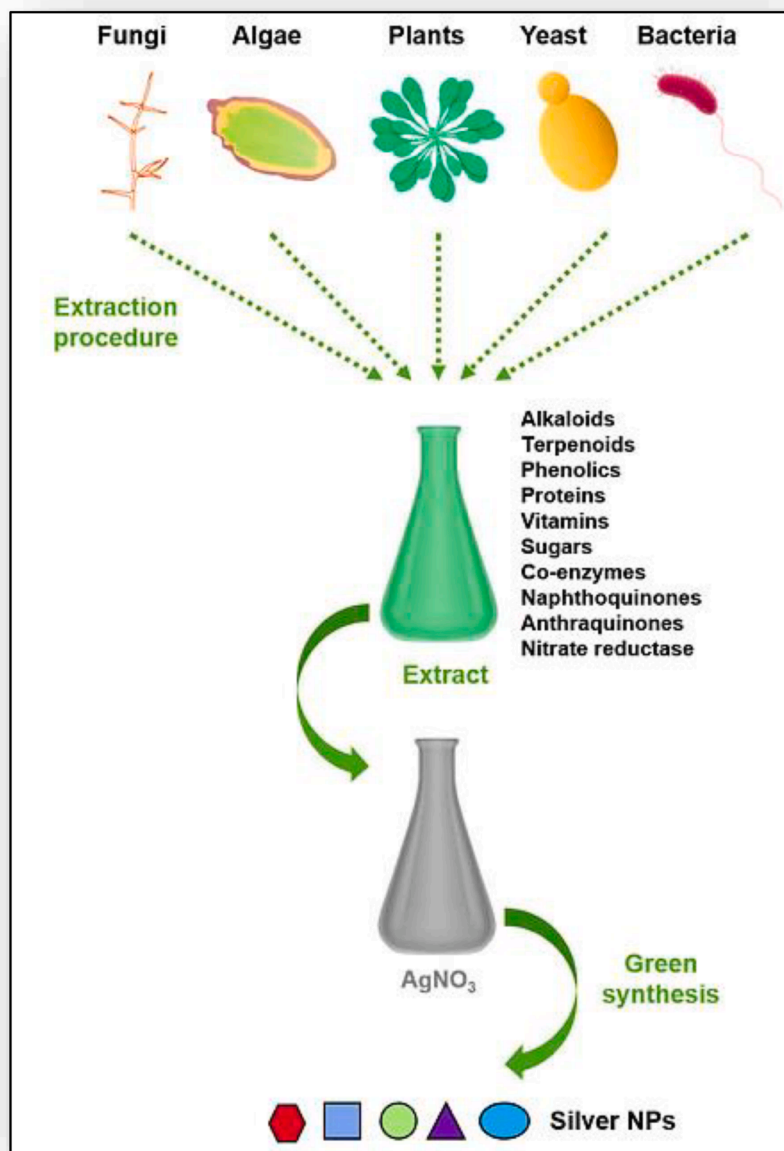


Fig. 5. Green synthesis of silver nanoparticles (Roy et al., 2018). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

resources (Roy et al., 2018). After synthesis, characterization is a most important process which helps to determine the size, purity, shape, morphology, surface charge, etc., which is accomplished by analytical techniques like SEM (scanning electron microscopy), TEM (transmission electron microscopy), AFM (atomic force microscopy), and FTIR (Fourier transform infrared spectroscopy) to name a few (Chouhan 2018).

Ag-NPs are applied in different industries. Their antibacterial activity is used from time immemorial, now employed in household products, cosmetics, consumer products, medical devices, etc. They are also used in the food industry (Kumar et al., 2018), for drug delivery (Mandal 2017), as anti-cancer agents, as antioxidants, in dentistry (Noronha et al., 2017), in diagnosis, (Calderón-Jiménez et al., 2017) in textiles, wound dressings, and wastewater treatment (Calderón-Jiménez et al., 2017; Wu et al., 2018). Polymers are used along with Ag-NPs, which enhance their catalytic ability for wastewater treatment (Saran et al., 2018). An overview of Ag-NP production and applications is shown in

Fig. 6 (León-Silva et al., 2016).

5.2. Ag-NPs based nanocomposites for wastewater treatment

Composites are two or more materials of different constituents combined to form a new material that is superior in its properties. Nanocomposites are composite, were among the multiphase, one phase consists of nanoparticles or nanostructures (Hazra Chowdhury et al., 2019). Engineered nanocomposites exhibit better mechanical properties, electrical conductivity, permeability, and increased chemical resistance. Nanocomposites are classified based on the presence or absence of polymeric material. They are classified as (i) polymer-based nanocomposite and (ii) non-polymer-based nanocomposite or inorganic nanocomposite. Inorganic nanocomposites are further divided into metal-based, and ceramic-based nanocomposites (Hazra Chowdhury et al., 2019). There are two methods through which silver polymer nanocomposites are prepared. Either introducing silver precursors into a

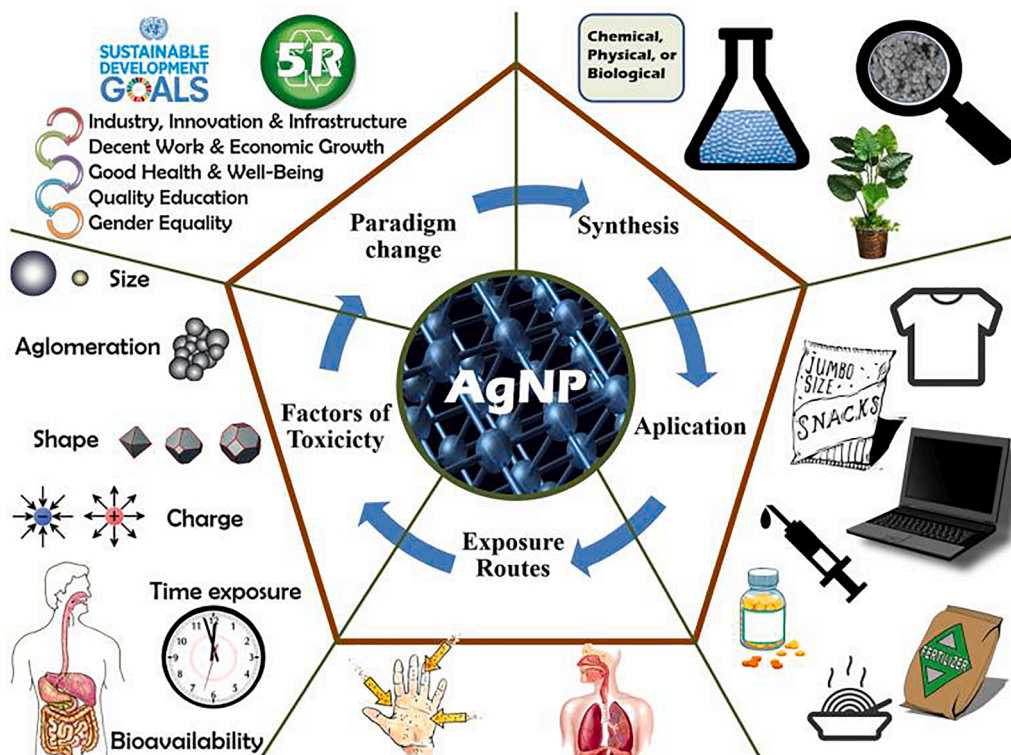


Fig. 6. The overview of AgNP production and applications. Reprinted with permission from León-Silva et al. (2016), License Number: 5,385,780,346,575.

polymer matrix followed by Ag-NPs preparation or by introducing nanofillers directly into the polymer matrix (Karak, 2019).

Nanoparticles are finding profuse use, and among these, silver nanoparticles are widely employed due to their extraordinary chemical, biological, and physical properties (Sadasiyuni et al., 2019). As stated above, Ag-NPs find applications in many fields like wound dressing, medicinal applications, electronics, optics, and water purification (Yaqoob et al., 2020). These already established applications of Ag-NPs made it an ideal candidate for nanocomposite formation. Ag-NPs are also used as a catalyst in reducing organic dyes, but they are easily oxidized and agglomerated due to reduction during catalysis (Liao et al., 2018). This was overcome by supplementing additional support to the nanoparticles. This was when biopolymer composites with embedded Ag-NPs came into action. Many studies are underway using silver nanocomposites to remove toxic dyes and pollutants from the aquatic system. Most researchers focus on synthesizing Ag-NPs using green synthesis rather than physical or chemical methods.

In a study, Kamal et al. (2020) synthesized self-formed Ag-NPs inside a gelatin biopolymer to reduce dyes methyl orange and 4-nitrophenol. Typically, 8 % gelatin hydrogels prepared were immersed in a 10 mM (millimolar) silver nitrate solution, and after three days, there was a colour change, which indicated that Ag-NPs self-formed inside the hydrogel. This prepared hydrogel composite (catalyst) and NaBH_4 (reducing agent) acted upon methyl orange and *N*-nitrophenol with reaction rates of 0.966 min^{-1} and 0.621 min^{-1} , and these silver hydrogels were easily recovered and reusable. In another study, Rani et al. (2020) green synthesized face-centred cubic crystalline structured Ag nanoparticles from kidney beans (*Phaseolus vulgaris*). These Ag nanoparticles can be used as a photocatalyst, catalyst, and antimicrobial agent. As a photocatalyst, it reduced reactive red dye with the kinetics of $1.13 \times 10^{-2} \text{ mmol g}^{-1} \text{ h}^{-1}$, and as a catalyst, it reduced 4-nitrophenol to 4-aminophenol in 15 min (Fig. 7a-c). It is also found that Ag-NPs can be embedded into pectin-based hydrogels. This hydrogel was synthesized using the microwave-assisted method, and later Ag-NPs were embedded into them by immersing the hydrogel in the AgNO_3 solution, and the

colour change denoted the formation of nanocomposite gel. This hydrogel was used as an adsorbent against methylene blue, and adsorption studies revealed an adsorption capacity of 88.72 mg/L and followed pseudo-second-order kinetics (Fig. 7d-f) (Babaladimath and Badalamoole, 2019).

Ag-NPs containing porous hydrogels were prepared with polyacrylic acid-silver and silver ions (PAA-Ag/Ag-NPs) through a self-assembly process and was found to act against three dyes, namely Rhodamine B, Congo red, and methylene blue. The adsorption process followed both pseudo-first and second-order kinetics and the adsorption times for methylene blue was 80 min, and for the other two dyes (Rhodamine B and Congo red) it is found to be 50 min. The formation process of PAA-Ag/Ag-NPs hydrogels is shown in Fig. 8a (Hou et al., 2016) The degradation of methylene blue using silver and magnetite nanoparticles functionalized with poly(ionic) quaternized dialkyl ethanolamine with 2-acrylamido-2-methylpropane sulfonate-co-vinylpyrrolidone was reported (Atta et al., 2020). The presence of a negative charge on the magnetite and silver nanoparticle composite mediates the electrostatic attraction of the methylene blue dye and improves the catalytic activity of both silver and magnetite nanoparticles. Chitosan is also combined with Ag-NPs, which help in pollutant removal. Chitosan and Ag substituted hydroxyapatite nanocomposite beads were produced by embedding chitosan with Ag-hydroxyapatite. The adsorption activity against rhodamine B was determined. The adsorption process showed pseudo-second-order kinetics with an adsorption capacity of 127.61 mg g^{-1} (Li et al., 2018). Some studies also used alginate as polymers. In one such study, Ag nanoparticles prepared by the microwave irradiation method were stabilized using alginate and introduced to three dyes malachite green, methylene blue, and rhodamine B. The adsorption capacity was studied, and it was noted that it followed both pseudo-first and pseudo-second-order kinetics. These nanocomposites had high reusability and were recyclable up to 25 times (Pal and Deb, 2013). The use of metal oxide-polymer nanocomposite was also reported. Here microgels of TiO_2 prepared through in situ hydrolysis were loaded with Ag nanoparticles (polymer- TiO_2/Ag) (Fig. 8b-e) and were used to reduce

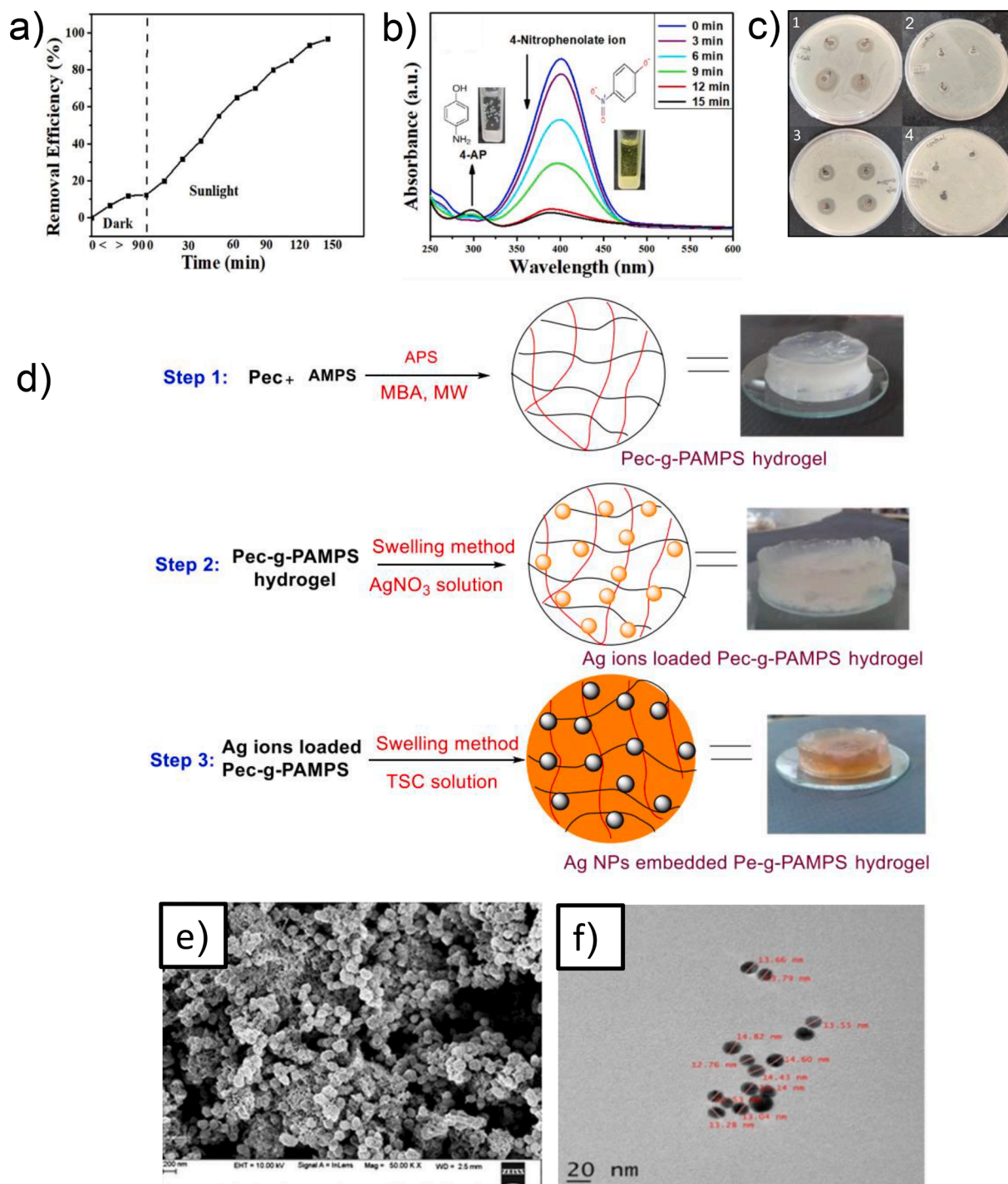


Fig. 7. a) Dye removal efficiency, b) The adsorption, c) Antibacterial properties (1 and 3: sample, 2 and 4: control) of Ag-NPs (Rani et al., 2020). d) Pec-g-PAMPS-SN synthesis stages, e) SEM and f) TEM image of Pec-g-PAMPS-SN2. Reprinted with permission from (Babaladimath and Badalamoole, 2019). License Number: 5385780960507.

4-nitrophenol. During this process of catalytic reduction, it was found that pseudo-first-order kinetics was followed. This catalytic reduction is favoured by the organic-inorganic network chains of the polymer microgels (Yan et al., 2019).

The effects of capping agents on silver nanocomposites and their media-induced release and catalytic activity were also studied in a specific study. Here, Ag nanoparticles were green synthesized from *L. indica* extract. These were then stabilized using capping agents like alginate and β -cyclodextrin using the ionotropic gelation method and were found to alter Ag-NPs morphology. Degradation ability was

assessed against methyl orange and determined using a UV-vis spectrophotometer. It was analyzed and concluded that pseudo-first-order kinetics had been followed (Nguyen et al., 2019). Another significant drawback was the development of an Ag nanocomposite sponge for dye removal. Herein, Ag nanoparticles were fastened onto a poly(vinyl alcohol) sponge with dopamine as an intermediate. Catalytic activity against 4-nitrophenol was evaluated and found to follow pseudo-first-order kinetics. It was also noted that the presence of dopamine helps to anchor the Ag-NPs onto the sponge (Guo et al., 2019).

Devi et al. (2016) put forth a simple process for methylene blue

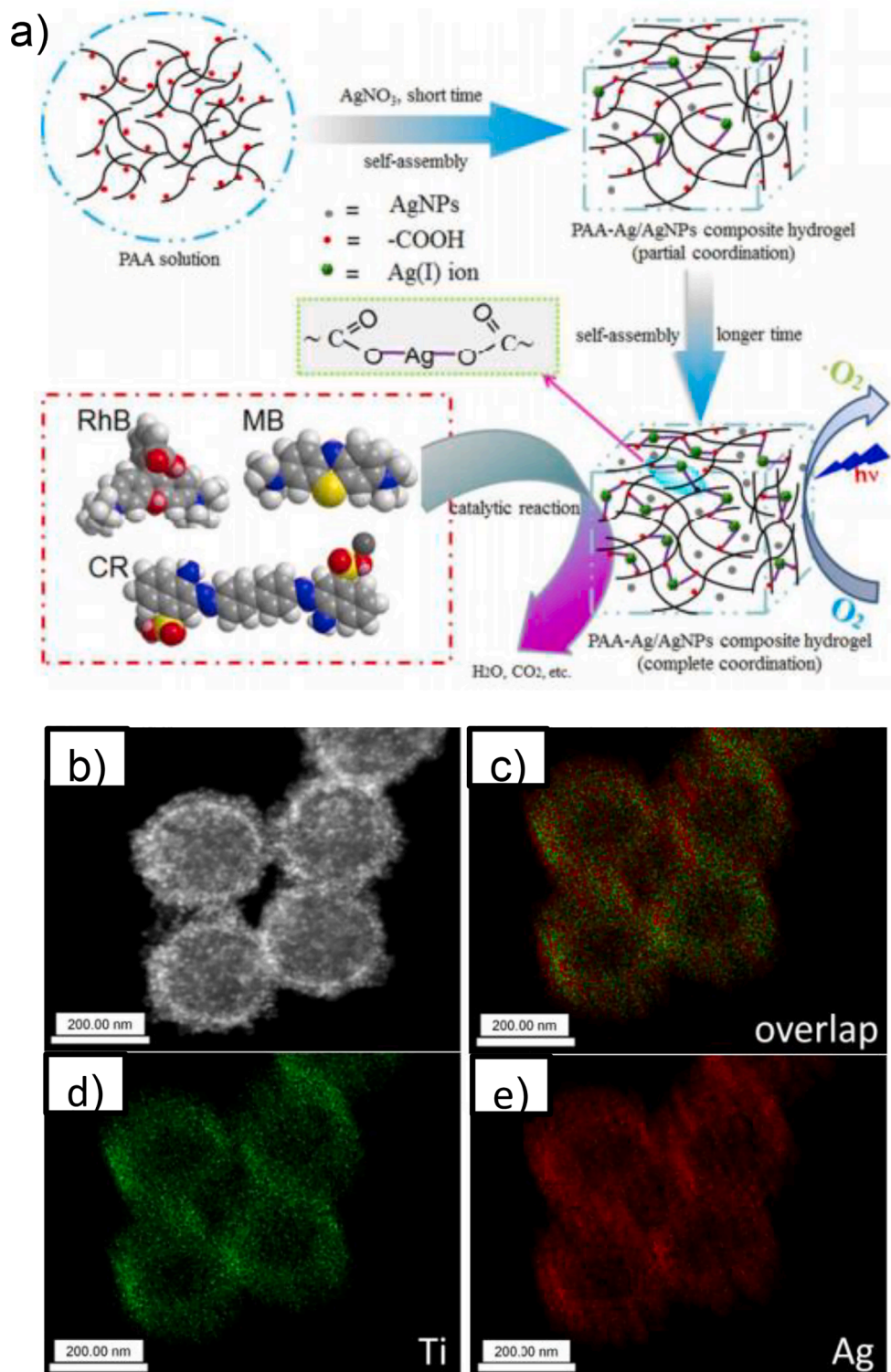


Fig. 8. a) The formation process of PAA-Ag/Ag-NPs hydrogels. Reprinted with permission from Hou et al. (2016). b) STEM image, c) overlap mapping image, d) Ti element image, e) Ag element images of polymer-TiO₂/Ag composite microgels (Yan et al., 2019).

removal from aquatic systems. *Mukia maderaspatana* extract was used as a reducing agent for nanocomposite hydrogel synthesis, and its adsorption capacities were excellent. The adsorption capacity was 213.7 mg and followed pseudo-first-order kinetics. Likewise, silver nanocomposites of poly (2-isopropenyl-2-oxazoline-co-N-vinylpyrrolidone) were synthesized with potassium persulphate as an initiator. The catalytic activity against 4-nitrophenol was determined and

found to be satisfactory (Palem et al., 2018). AgNP-calcium alginate beads were prepared using *Walsura trifoliata* bark extract as both reducing and capping agents, exhibited very high catalytic activity in reducing 4-nitrophenol and did not lose their catalytic activity even after 10 cycles validating its reusability. This was found to follow the pseudo-first-order kinetics (Saran et al., 2018).

Ag nanoparticle decorated holocellulose (Ag-NPs/HCNF) was

synthesized through the microwave-assisted green process and was freeze-dried to produce aerogels (Fig. 9 a-b), which are great catalysts against Congo red and methylene blue. The adsorption activity followed pseudo-first-order kinetics and was reusable for up to five cycles. The presence of holocellulose helped in the stabilization of Ag nanoparticles (Bandi et al., 2020). Moreover, Ag nanoparticles embedded into poly-(*N*-isopropyl methacrylamide-*co*-methacrylic acid (p(NMA) microgels were found to be used for the catalytic degradation of azo dyes like Congo red, methyl orange, and Alizarin yellow. The catalytic degradation of methyl orange was higher, while those of Congo red and Alizarin yellow were satisfactory. They were found to follow pseudo-first-order kinetics (Iqbal et al., 2020). The formation mechanism of AgNPS-p(NMA) hybrid microgels is demonstrated in Fig. 9c. Albukhari et al. synthesized silver nanoparticles using *Durantaerecta* leaves, and nanocomposites were formed with the help of cellulose acetate filter paper and titanium dioxide through a facile biological route. The composites showed multi-functional activities, especially catalytic activities, against methylene blue and rhodamine B dyes and followed pseudo-first-order kinetics (Albukhari et al., 2019). Ag nanoparticles supported onto polyurethane foam were used to remove methyl orange from tap water. Adsorption

was more efficient and followed pseudo-second-order kinetics, and adsorption capacity was found to be 1.256 mg g^{-1} (Ali and El-Sheikh, 2017). The cationic dye absorption (crystal violet) using κ -carrageenan and nanosilver chloride composite hydrogel was found to be more than 98 %. The kinetic model followed here is the pseudo-second-order, and it was also noted that the addition of nano-silver chloride into hydrogel made it a superabsorbent (Dargahi et al., 2018).

El-Sharkawy (2019) immobilized Ag-NPs onto orange peel functionalized with sulphuric acid (AgNPs@OP/A), orange peel functionalized with sodium hydroxide (AgNPs@OP/B), and onto functionalized MWCNTs, their adsorption kinetics were analyzed. The adsorption of brilliant green dye using AgNPs@OP/A and AgNPs@OP/B followed pseudo-first-order kinetics while the nanoparticles attached to multi-walled carbon nanotubes (AgNPs@MWCNTs) followed pseudo-second-order kinetics. In another study polyester textile membranes containing silver nanoparticles, polyvinylidene fluoride, polyethylene glycol, and zeolitic-like framework-67 with ethylenediamine as a cross-linker was prepared. The membrane was used to remove Rose Bengal dye and was found to selectively remove the same even in the presence of interfering dyes like methylene blue, and amido black. Adsorption

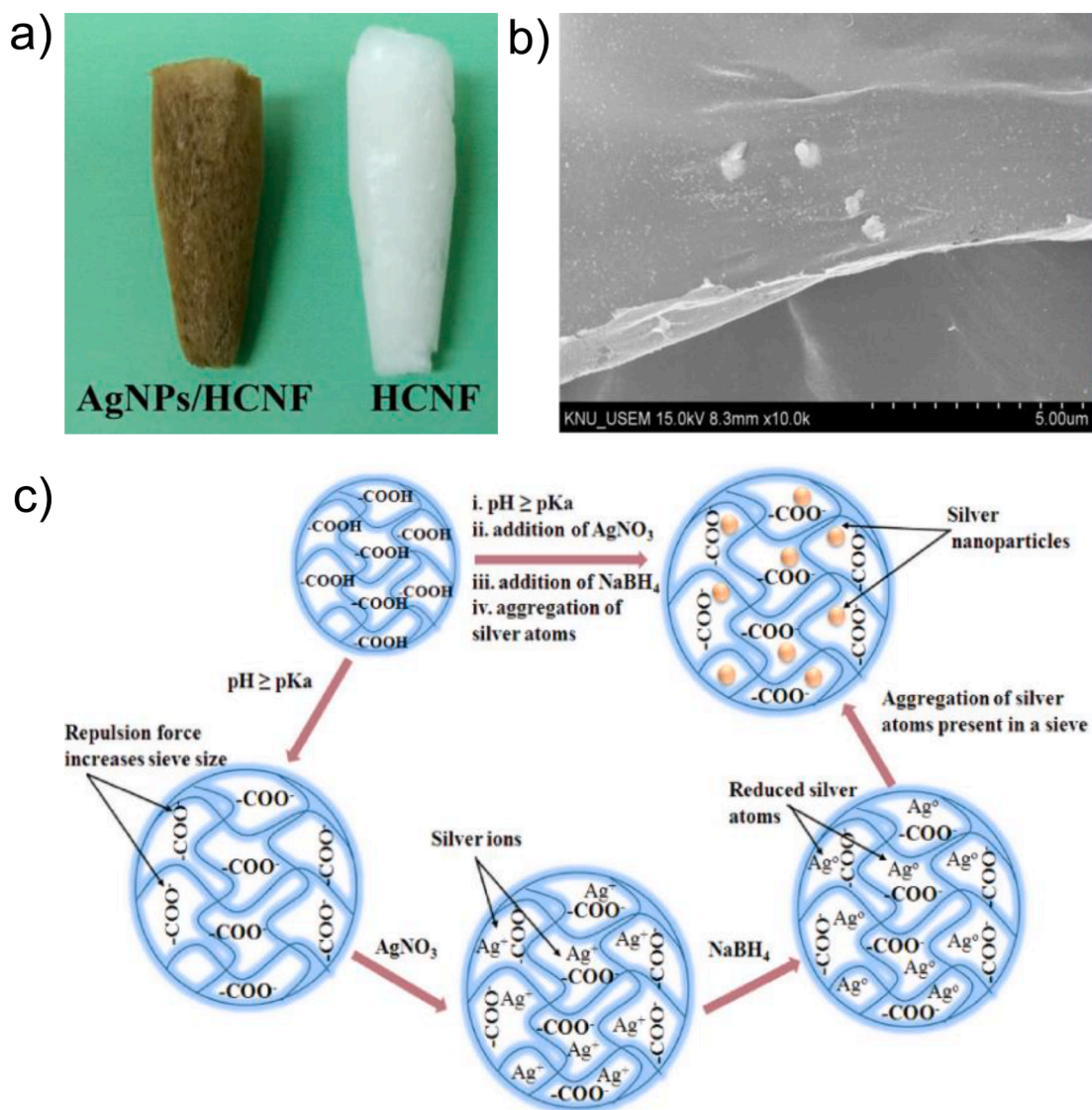


Fig. 9. a) Ag-NPs/HCNF and HCNF (Aerogels have taken the form of the centrifuge tube and remain intact), b) SEM images of Ag-NPs/HCNF. The pore walls are smooth in the case of HCNF Aerogel, whereas in the case of Ag-NPs/HCNF aerogel, it is rough, showing silver deposits. Adapted with permission from Bandi et al. (2020). License Number: 5,385,791,417,028 c) The mechanism illustrates the formation of AgNPs-p(NMA) hybrid microgels. Adapted with permission from Iqbal et al. (2020). License Number: 5385800151357.

studies calculated the removal of 96.41 % of the Rose Bengal dye at pH 6 (Mofradi et al., 2020). Nguyen et al. (2018) biosynthesized hybrid nanocomposites based on silver nanoparticles capped with 2-hydroxypropyl- β -cyclodextrin/alginate (Fig. 10). The novel nanocomposite showed catalytic activity against 4-nitrophenol, methyl orange, and rhodamine B. The adsorption reactions were quick and followed pseudo-first-order kinetics. Nanoparticles of silver chloride embedded into adsorbents constructed by co-polymerizing acrylic acid and hydroxyethyl methacrylate with sodium alginate through the co-precipitation method were used for the adsorption of brilliant cresyl blue. This gel showed a high adsorption capacity of greater than 95 % and was followed by pseudo-second-order kinetics (Bhangi and Ray, 2020).

Mane and co-workers synthesized Ag nanoparticles loaded poly-pyrrole nanotubes by biogenic reduction method using *Scadoxumultiflorus* leaf extract and in situ chemical oxidative polymerization. The composites reported higher photo-catalytic activity against methyl orange and the degradation of dye followed pseudo-first-order kinetics (Mane et al., 2020). Mohan et al., (2020) synthesized pH-responsive poly(methacrylic acid) functionalized SBA-15 through free radical polymerization and were embedded with Ag nanoparticles. The nano hybrid catalyst was further used for the reduction of anionic and cationic dyes and followed pseudo-first-order kinetics. Silica-supported Turkevich silver nanocomposites prepared through electrostatic interactions and were analyzed for the photocatalytic degradation of the methyl orange dye. This study shows that the metal nanoparticle in the presence of incident light causes irradiation, resulting in energy transfer with in the metal nanoparticles. The electrons transferred generate $\cdot\text{OH}$ radicals which degrade methyl orange (Das et al., 2020). Certain studies also employed the use of magnetic silver nanoparticles modified with β -cyclodextrin- maleic anhydride polymer nanocomposites for the reduction of organic dyes like methylene blue, which followed pseudo-first-order kinetics with a rate constant of 0.1587 min^{-1} (Nariya et al., 2020). Silver nanoparticles embedded in graphene oxide sheets were used in the catalytic degradation of indigo carmine and methylene blue in a study carried out by Jose et al. (2018). The researchers suggested that dye degradation was due to photocatalytic activity. Synthesis of silver-doped polyaniline-polyvinyl chloride composite films using the non-solvent-induced phase inversion method showed excellent photocatalytic activity against methylene blue. The degradation of the dye followed first-order kinetics (Ismayil, et al., 2019). Microgels of poly(*N*-isopropyl methacrylamide-co-acrylic acid) synthesized by the

precipitation-polymerization method were embedded with uniformly dispersed Ag nanoparticles using in situ reduction method was used as a catalyst against 2-Nitroaniline, and the reduction process followed pseudo-first-order kinetics (Begum et al., 2018).

The use of bi-metallic composites is also reported. In a certain study, Tripathy et al. synthesized Ag-Au bimetallic nanocomposites using graft copolymer hydroxyethyl starch-g-poly (acrylamide-co-acrylic acid). The reduction of Congo red, Sudan-1, and methyl orange was reported in the presence of composites (Tripathy et al., 2017). Biocomposite with chitosan-Guar gum blend and Ag nanoparticles prepared from palm shell extract was studied against dyes reactive blue-21, reactive red -141, rhodamine 6G, and their combinations. The composites reported high catalytic activity for the degradation of the dyes (Vanaamudan et al., 2018). Green synthesized AgNPs embedded into the alginate-guar gum blends were used to degrade methylene blue dye showing a degradation capacity of 92.33 %. The kinetics followed a pseudo-second-order mechanism (Hasan et al., 2020). The cryogels embedded with silver or gold nanoparticles were also used for the degradation of dyes. In one such experiment, Haleem et al. (2020) used cryogels of poly (*N*-isopropylacrylamide-co-acrylamido-2-methylpropane sulfonic acid) produced through redox-initiated cryo-polymerization and immobilized Ag and Au nanoparticles. These were used to reduce p-nitrophenol and were found to follow pseudo-first-order kinetics; moreover, it was noted that Au-containing cryogels had higher degradation capability than Ag hydrogels. Another study used polymer microspheres through seeded swelling polymerization and embedded with Ag nanoparticles in the presence of polyvinylpyrrolidone as both stabilizing and reducing agents for the degradation of methylene blue dye. The rate constant was found to be $2.6 \times 10^{-3} \text{ s}^{-1}$ and followed pseudo-first-order kinetics (Tian et al., 2017).

Silver nanoparticles decorated onto the surface of ZnO/SiO₂ nanocomposites act as catalysts in the chemical reduction process. The photocatalytic degradation of the methylene blue dye was found to be 81 % with this nanocomposite. Pseudo-first-order kinetics was observed. The nanocomposite attracts light effectively so that transfer of electrons from the valence band to the conduction band occurs. This results in the transmittance of electrons to the surface to absorb O₂ resulting in superoxide radical anions along with the formation of hydroxyl radicals (due to the reaction of holes with H₂O). These hydroxyl radicals lead to the degradation of methylene blue (Govindhan and Pragathiswaran, 2019). Nanocomposite hydrogel beads with chitosan/AgCl/ZnO

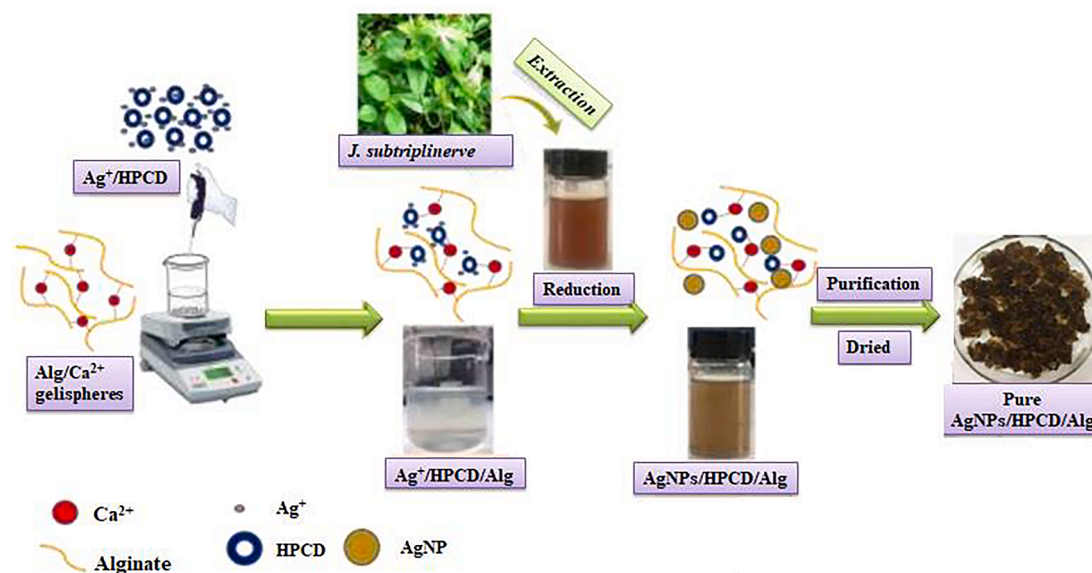


Fig. 10. Ag-NPs/HPCD/Alg synthesis process. Ag⁺ ions can be capped on the nanocomposite by chemical bonds of -OH and -COO⁻ groups in alginate and HPCD molecules. Reprinted with permission from Nguyen et al. (2018). License Number: 5385800435815.

successfully degraded methylene blue and followed pseudo-first-order kinetics (Taghizadeh et al., 2020).

Multi-functional guar-gum/Ag nanocomposite hydrogels were synthesized using rhubarb stem extract as a bio-reductant. The catalytic activity of the composites against p-nitrophenol was found to be around 85 % with a rate constant of $121.8 \times 10^{-3} \text{ min}^{-1}$, and followed pseudo-first-order kinetics (Palem et al., 2020). Another study used a nanosilver embedded into lignin/polyacrylamide hydrogel to remove p-nitrophenol and was found to follow a pseudo-first-order kinetic model (Gao et al., 2019). The use of different shapes of hydrogels was also noted. Narayanan and co-workers used polyacrylic acid/amido-diol fibrillar hydrogels embedded with Ag nanoparticles by situ polymerizations of acrylic acid and amido-diol as cross-linker and reducing agents. Catalytic activity against methylene blue, rhodamine 6G, and crystal violet showed first-order kinetics, and a complete reduction of the dyes was noticed (Narayanan et al., 2014). The highly porous carbon matrix, which is N-doped and derived from chitosan embedded with Ag nanoparticles, is used as a catalyst against 4-nitrophenol. The catalytic reduction was found to follow pseudo-first-order kinetics. There was a much lesser loss of the composite even after seven cycles (Alhokbany et al., 2019). Ag-NPs incorporated into the Ag-2-Amino-5-mercapto-1,3,4-thiadiazole polymer matrix exhibited remarkable catalytic activity against 4-nitrophenol and were found to follow a pseudo-first-order reaction (Cheng et al., 2017). A sonochemical approach for synthesizing nanocomposites was noticed in a particular study (Chang, et al., 2018). Here a cobalt (II) coordination polymer and Ag-loaded coordination polymer nanocomposite were used to study its catalytic activity against methylene blue, and the photodegradation studies followed a pseudo-first-order kinetic model.

Emam et al. (2017) deposited nanoparticles (i.e., Ag, Au, and Ag-Au alloy) onto the cellulosic solid support, through redox reactions, to fabricate metallic/bi-metallic nanocomposites. The reduction reaction of p-nitro-aniline was carried out using NaBH_4 in the presence of nanocomposites. The reduction process followed pseudo-first-order kinetics with a k-value of $33.5 - 37.7 \times 10^{-3} \text{ m}^{-1}$ (in the presence of bi-metallic composites). Another study utilized chitosan-Ag nanocomposite compounds for the photocatalytic degradation of methyl orange, the degradation followed pseudo-first-order kinetics (Nithya et al., 2015). Saeed et al. prepared polyacrylonitrile nanofibers through electrospinning with modified amidoxime groups, and silver nanoparticles were embedded into them. Photodegradation results of methyl orange showed around 80 % degradation (Saeed et al., 2015). In a study, polyaniline/silver nanocomposite was produced by oxidative polymerization. The adsorption of brilliant green dye on polyaniline/silver nanocomposite followed the pseudo-second-order kinetic model (Salem et al., 2016). Photocatalytic degradation of methyl orange with the help of Ag/AgCl nanocomposites was also observed (Zou et al., 2019).

6. Conclusions and perspectives

There is no clarity on the fate of textile dyes in the sediments of rivers and seas. Some suggest that these aromatic compounds can be naturally degraded by bacteria over time, while the period and amount of degradation are not yet exact. Also, the success rate of this under anaerobic conditions is still a question. Hence it is always better to treat the wastewater before discharging it into the water bodies. This is where novel adsorbents come for effective dye removal. This article analyzed the use of biopolymers with embedded silver particles as adsorbents to remove different kinds of dyes. At the practical level, it is found that these composites effectively remove different kinds of dyes, even multiple pollutants. The fact that nanocomposites are reusable makes them more feasible. The use of engineered nanocomposite membranes is in research for applications in different fields. A particular fact that needs research is the implementation of different metal nanoparticles into a single polymer to achieve enhanced wastewater treatment, where one influences the other metal/metal oxide nanoparticle. Nanomaterial

implied water treatment processes are still at the research level and are seldom used on a large scale and hence not employed as widely as conventional methods. This scenario needs to change. Also, the synthesis of Ag nanoparticles through chemical methods involves different hazardous reagents, but this is eliminated now with more research focusing on biogenic nanomaterials. However, the purity in the case of biogenically synthesized Ag nanoparticles is of concern as nanoparticles used for dye treatment need to be pure as they are released into the environment. In contrast to all these, some studies prove silver-mediated toxicity even at low concentrations and their biological systems effects. There are also reports of silver nanoparticles affecting the microbial biome of wastewater, as silver is a proven antimicrobial agent, a matter of concern that needs to be looked into as chances of superbugs that adversely affect are envisioned. Hence the presence of silver in treated water is an aspect that needs to be considered.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

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