Review

The Use of Silver Nanoparticles in Environmental Remediation

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Abstract: The invention of nanoscience not only brings a revolutionary change in the field of science but also changed the direction of research. Today the whole world is under the trigger of nano and nanoparticles have multidimensional applications in every aspect of life including environmental point of view. Till today a plethora of nanoparticles have been synthesized and have also been applied for multiple purposes and hence grabbed the attention of researchers all over the world. Among the bunch of NPs discovered to date, we have a particular interest in silver nanoparticles (AgNPs) because of their cost-effectiveness and huge abundance in the earth's crust. With respect to every passing day, due to various kinds of anthropological activities, the quality of the environment such as air, water and soil is depleting which ultimately hampers the human civilization. To encompass the growing environmental issues, many techniques have been adopted. Among the many strategies, tackling the current issues regarding environment through nanoscience is highly worthy as because of its cost-effectiveness, less time consuming and easy handling process. This article reviews the potential of nanoparticles, particularly silver nanoparticles, for a wide range of environmental applications, such as soil, air, and water remediation.

Keywords: applications; biogenic; environment; remediation; silver; soil

1. Introduction

Now-a-days environmental pollution is standing as a major problem is a standing as the biggest thing of concern for the growing civilization. In the name of development, the current generation is deteriorating the quality of the environment day by day by doing various geological works [1-5]. The most important environmental issue is the water and air pollution which arises primarily due to industrialization. It may make us economically enrich but at the same time it brings high-rate air, water and soil pollution by the huge amount of release of carcinogenic gases, heavy metal ions, organic wastes, nuclear wastes. Due to the discharge of the various types industrial and agricultural wastes into the environment, it ultimately results in different kinds of air, water and soil borne diseases [6-9]. The most recent air born disease is corona which causes the death of thousands of people and millions of peoples suffered by this disease all over the world. Also, due to the release of the heavy metal ions, carcinogenic gases into the environment, it results various kinds of life killing diseases like cardiac disease, cancer, failure of multiple organs like excretory system, nervous system, liver etc [10,11]. Simultaneously, aquatic animals are also getting sufferer due to the discharge of industrial effluents into the water bodies. They increase the turbidity of the aqueous medium, thus limiting the sunlight penetration and oxygen dissolving capability. The problem persists for a long period of time as the effluents have a long half-life period. Though, several strategies have been invented emphasizing the huge demand of environmental issues, all they have their own merits and somehow suffer with some sort of limitations i.e., high cost, long time duration, necessity of large manpower etc. [12,13]. Thus, the development of new, cost effective and eco-friendly approach to handle the various environmental issues is profoundly important and it is the call of the day.

Nanotechnology is the most emerging field of science which provides a bunch of applications for every aspect of life. Particularly, nanomaterials have gained a lot of attention due to their multidimensional use in every field starting from environment to life saving drug molecule synthesis [14, 15]. Easy method of preparation, low cost, avoidance of harmful reagents and solvents, high yield, small size with large surface area are the most charming features found to be associated with the nanomaterials which makes them more demandable [16-20]. Among the variety of nanomaterials invented, silver nanoparticles are quite unique and desirable as because of their cost-effective synthesis, environmentally sustainable and most significantly their high toxicity only towards the multidrug resistant micro-organisms over healthy cells and soil at low dose. Therefore, it has gained multidimensional application [21-23]. Among the different applications, the pages of the literature are enriched with enormous number of environmental applications of silver nanoparticles [24-27]. Considering the safe and biocompatible nature of the AgNPs, it becomes worthy to apply it for water, soil and air remediation purpose in the form electro sensor, photocatalyst, fluorogenic probe etc.

This review summarizes the well documented applications of AgNPs towards the environment. It briefly describes the physical, chemical, and biological pathways followed for the synthesis of the nanoparticles and its application as electro sensor, photocatalyst, fluorogenic probe etc. towards the detection of heavy metal ion, dyes, micro-organisms etc. The developed nano-reagents not only help to wipe out the desired pollutants from environment but also these exhibit good reusability and recyclability.

2. Application of silver nanoparticles for the environment

AgNPs are synthesized thermally using graphite carbon sheets and used as an electrochemical sensor for the detection of organic materials in water samples. The structure, composition and morphology of the developed nanomaterial were studied through Scanning electron microscope (SEM), X-ray diffraction technique (XRD) and IR spectroscopy. The sensor extremely exhibited sensitivity towards the detection of nitrofurazone with very low detection limit values i.e., LOD value of 1.2×10^{-8} M and LOQ value of 1.3×10^{-7} M. The sensor displayed a reduction peak at -0.57V as well as a calibration curve was constructed using the concentrations 10^{-4} M to 2×10^{-7} M which indicates the reduction of nitrofurazone. It was experimentally verified that application of AgNPs on the carbon nanosheets increases its electrocatalytic ability and enhances its potential towards the reduction of nitrofurazone. The developed sensor worked well and can be applied for the potential detection of nitrofurazone in real water samples like tap water, human urine, and commercial milk etc. It showed high reproducibility and reusability [28].

PBT (phenyl benzotriazole) functionalized AgNPs (PBT-|) were synthesized calorimetrically at room temperature through reduction by $NaBH_4$ for the selective detection of Cr (III) ions in the water samples. Developed NPs were successfully characterized through UV-Vis, Fourier Transform infrared Spectroscopy (FTIR), Zetasizer, Atomic Force Microscopy (AFM), SEM, etc. Reaction parameters such as temperature, pH, ionic strength etc. were found to have negligible influence upon the stability of the NPs. The prepared supramolecular stabilized NPs were not only found to be highly sensitive but also it showed high selectivity towards the detection of Cr^{3+} ions in the tap water samples. The detection was achieved by selective binding of the NPs with the Cr^{3+} ions which was confirmed through Jobs plot which showed a binding ratio of 1:2. The sensing probe showed a linear response having $R_2 = 0.9992$ with LOD vale of 0.2 μM [29].

Diversified AgNPs were synthesized biogenically from capsid structural proteins of bacteriophage (a naturally occurred bacterial virus) and applied both for the detection of heavy metal ions from the water samples as well as studied their antibacterial and antibiofilm properties. The AgNPs were prepared by the bacteriophage mediated reduction of AgNO3. NPs were found to have a particle size of 10 to 30 nm as confirmed by TEM. The developed NPs worked tremendously well ad were found to successively inhibit the bacterial biofilm of *S. sciuri* and detect specifically Cd^{2+} ions in the real water samples in a concentration of 100 μM [30].

Targeting indoor air purification, photocatalytic films were prepared which are composed of glass supported AgNPs fabricated over graphitic carbon nitride (g-C_3N_4) which acts as photocatalyst. The prepared photocatalyst performed outstandingly and found to catch the volatile organic compounds such as ethylene, toluene, p-xylene, benzene, acetaldehyde, formaldehyde etc as well as other bacterial derivatives like *E.coli*, *S.aureus*, *C.albicans* etc efficiently. It took advantage of the visible light to degrade these types of air pollutants. Due to the strong ability of Ag to absorb the visible light through SPR effect and helps in the charge separation through the formation of Schottky barrier with the semiconductor, the Ag decorated g-C_3N_4 sheets worked well as photocatalyst. The major generated species during the degradation process include superoxide radical anions and holes rather than hydroxyl radicals. The developed photocatalytic probe exhibited excellent recyclability even after ten successive cycles of application [31].

Targeting dual oxidation state determination of heavy metal ions in the water samples, an electrochemical sensor consisting of nano silver and gold modified electrodes was developed. The sensor was designed in such a way that i.e., the surface of the screen-printed carbon electrodes acting as working electrode was coated with the layers of Ag and AuNPs so that it can be able to detect the dual oxidation of Cr i.e., Cr^{3+} and Cr^{6+}. Further to detect the Cr^{3+} state, the bimetallic NPs were oxidized to form stable Ag-Au metallic oxides. Merging of the AgNPs with Au provided stability to the formed oxide coatings was proved experimentally. The sensor was characterized successfully and applied for the detection of heavy metal ions in the water samples. It showed excellent performance towards the detection of both the oxidation states of the Cr species having LOD value 0.1 ppb for 0.5 to 5 ppm concentration towards Cr^{6+} ions and 0.05 to 1ppm concentration towards Cr^{3+} ions in different types of water samples including tap water, artificial sweat samples, artificial saliva etc. [32] (Fig. 1).

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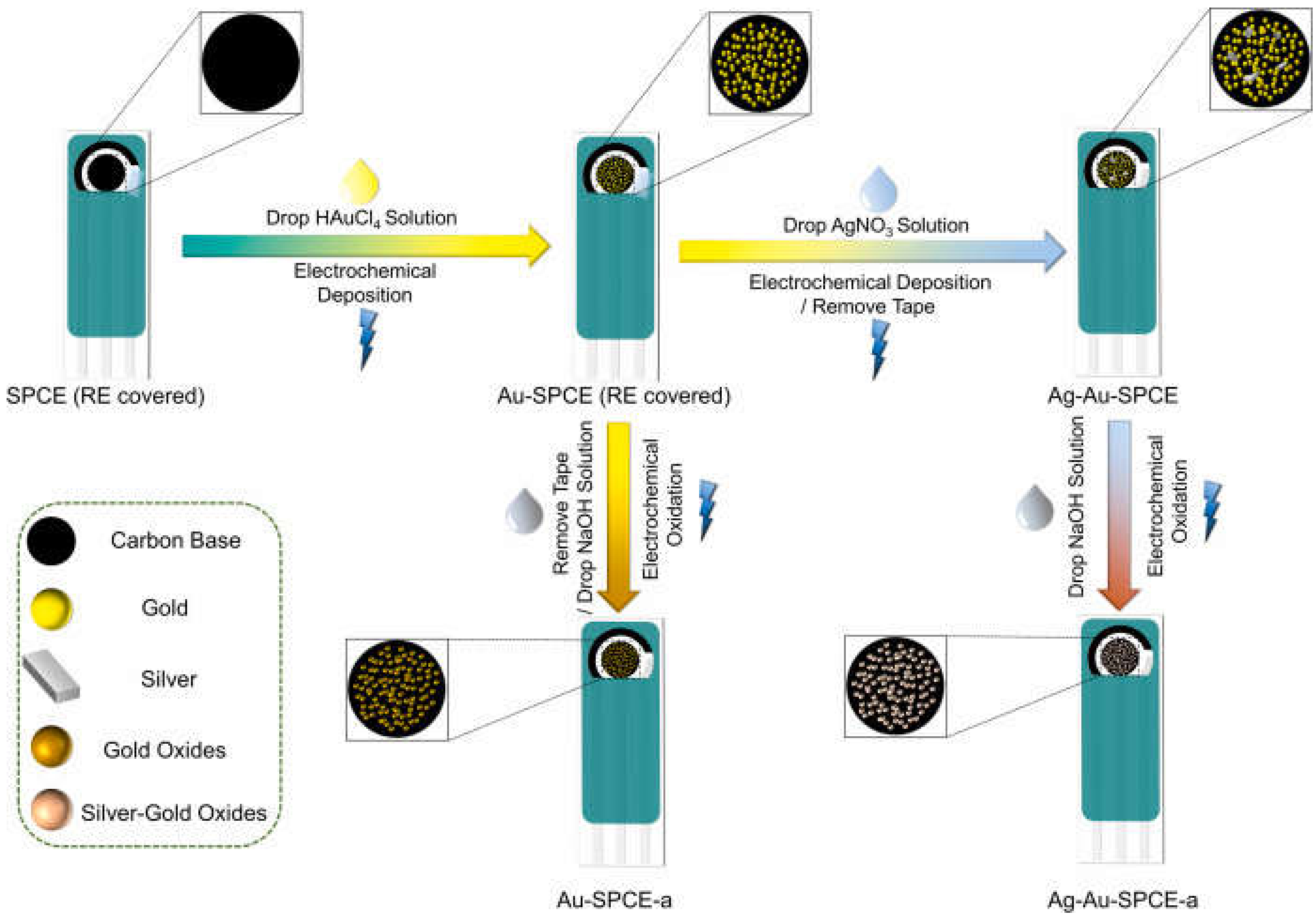

Figure 1. Electrodes preparation "Reprint and Copyright permission obtained from Elsevier publication" [32].

To detect the growing water and soil pollution due to rapid use of glyphosate, a most used herbicide for cultivation purpose, rGO rapped AgNPs over TiO_2 nanotubes were prepared as SERS sensor for the first time. The prepared sensor worked promising well towards the detection of methylene blue and glyphosate in water samples having LOD value of 10^{-14} M and 3µg/L respectively. Moreover, the sensor exhibited good reproducibility and repeatability with 2.0 % and 4.0% relative standard deviation [33].

Photochemical degradation of the organic pollutants like reactive yellow 186 and reactive blue 19 from water medium by using Ag nanoparticles prepared biogenically using renewable source i.e., from the leaf extracts of *Trigonella foenum-graecum* was reported by the group of Kim and Rawat [34]. 5mL of the leaf extract with pH 10 was set as the optimum conditions for the generation of the nanoparticles. Prepared nanomaterials were successfully characterized through various spectrometric analyses which indicated the average particle size of the nanoparticles was 5 to 20 nm with 428 nm as absorption edge. The NPs showed 88% degradation potential towards RB 19 and 86% towards RY186 in180 mins which was confirmed by the decrease in the chemical oxygen demand value as the degradation of the organic materials resulted the formation of CO_2 and H_2O which reduced the oxygen percentage of the water and as a result decreased the COD value. The rate of photocatalytic degradation of the dyes followed pseudo-first order kinetics having rate constant K values 0.01230 and 0.01034 min^{-1} and R2 values 0.98446 and 0.96881 respectively. Mechanistically, in the presence of UV light, excitation of the electrons occurs. The excited electrons interacted with O_2 and OH^- ions to form O_2 and OH radicals. Interaction of both radicals with the dyes leads to their degradation. The NPs synthesized through environmentally sustainable approach exhibited good reusability as well as

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photochemical activity and hence, it can act as an effective candidate for wastewater treatment (Fig 2-4) [34].

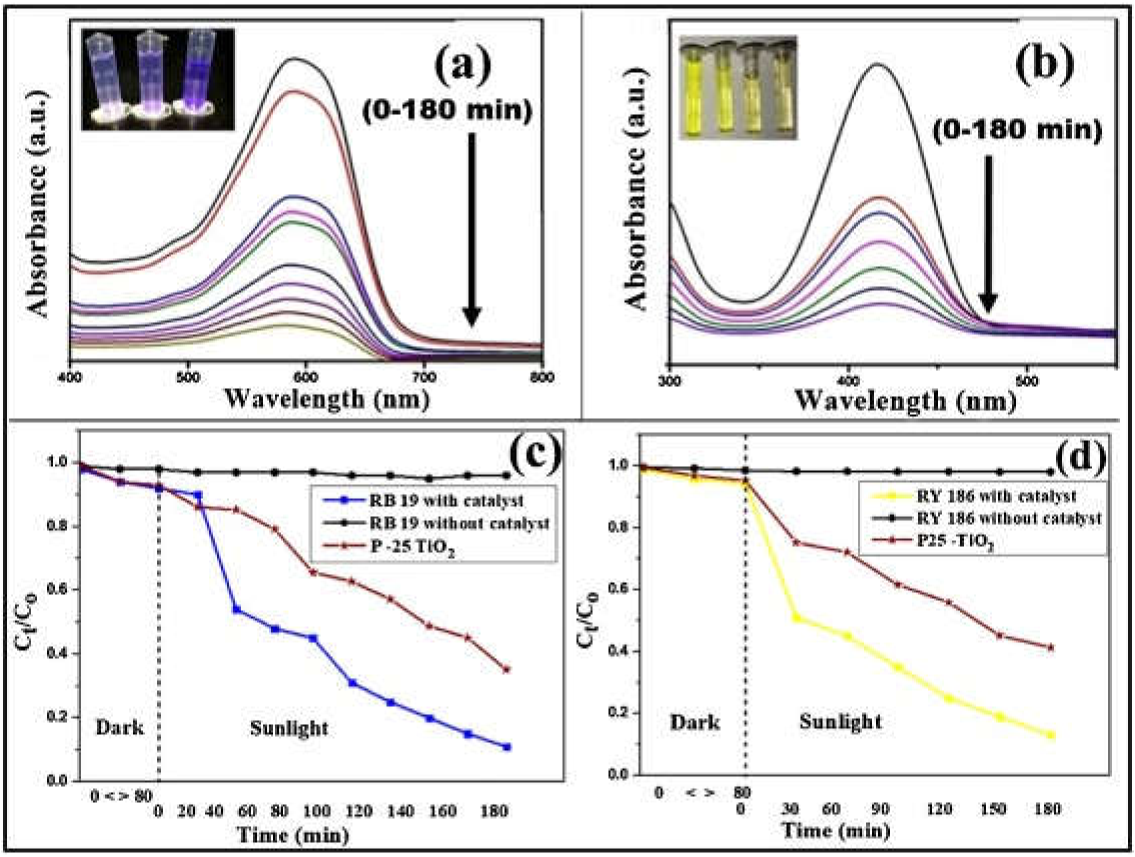


Figure 2. UV-visible spectra of the degradation patterns for the two model dyes (RB19 and RY186). Time dependent decrease in the absorbance of (a) RB19 and (b) RY186 dye in presence of AgNPs. (c) and (d) show normalized concentration of RB19 and RY186 dyes, respectively in the presence and absence of AgNPs/commercial P-25 TiO2 under dark and sunlight conditions "Reprint and Copyright permission obtained from Elsevier publication" [34].

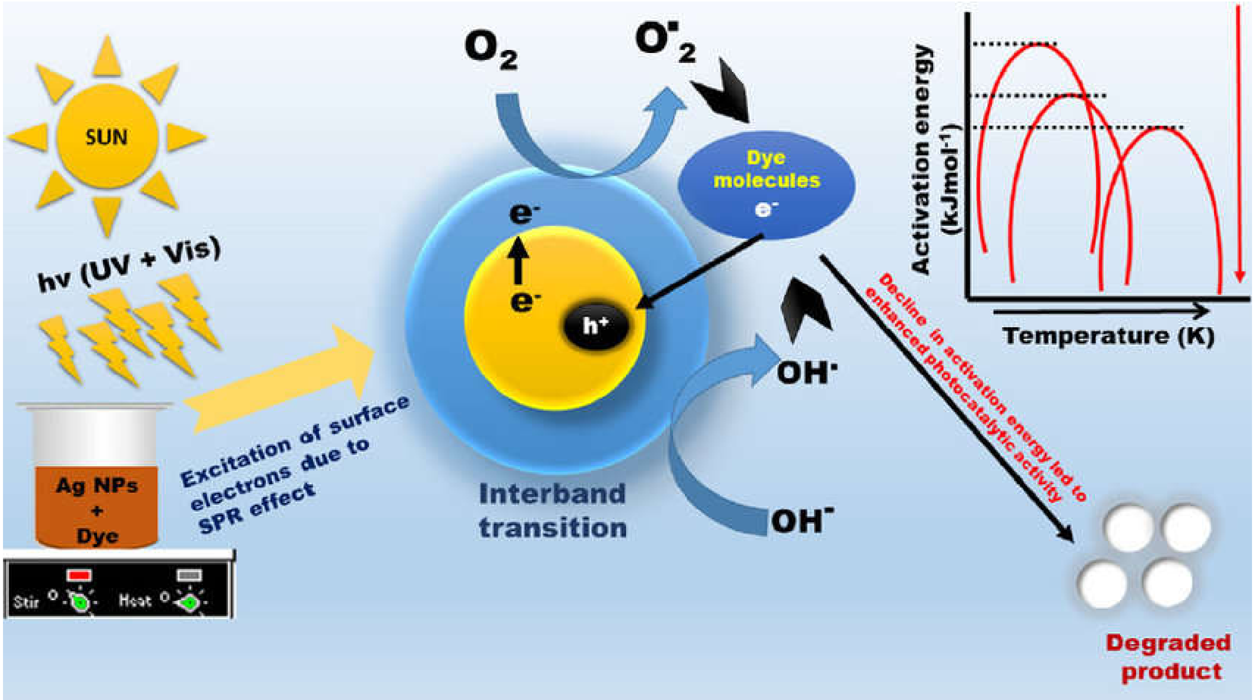


Figure 3. Schematic for the proposed mechanism of photocatalytic dye degradation by biogenic AgNPs and describing the role of activation energy "Reprint and Copyright permission obtained from Elsevier publication" [34].

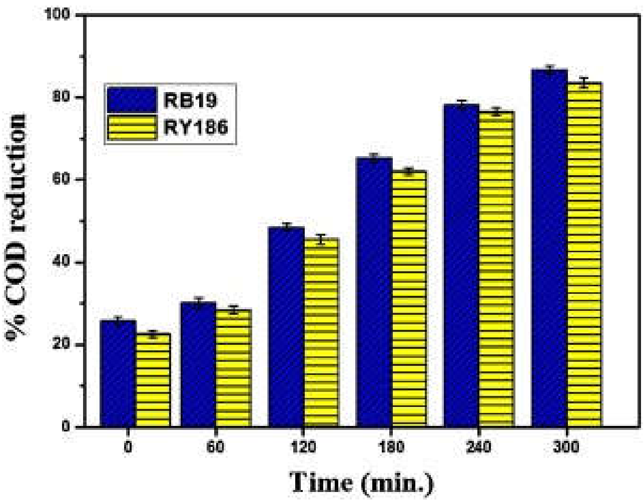


Figure 4. COD reduction studies of RB 19 and RY186 dyes after photocatalytic process "Reprint and Copyright permission obtained from Elsevier publication" [34].

Biogenic recyclable core hell nanoparticles consisting of FeO/AgNPs and FeO/Au NPs were synthesized using pomegranate fruit peel extraction. The formation of the dual types of the nanoparticles was confirmed by different spectral methods. In the UV-visible spectrum, absorbance peaks at 465 nm and 530 nm confirmed the formation of FeO/AgNPs and FeO/AuNPs. Through EMR analysis it was found that in case of FeO/AgNPs, 14 nm shell of AgNPs were found to surround the 13 nm Fe core whereas the average size of FeO/Au NPs were found to be less than 100 nm. The in vitro antimicrobial and antifungal abilities of the NPs were determined through zone of inhibition and mycelium inhibition method. The results indicated that the NPs possess a good range of antimicrobial properties against all types of micro-organisms. Moreover, the NPs displayed wonderful dye degradation ability. Aniline blue dye can be degraded by the NPs significantly within 150 min at 70 °C at pH 10. The biocompatibility natures of the NPs were tested on Vero cell lines which signified that the core shell NPs were highly biocompatible in nature up to 500 μg/ml concentration. The NPs were found to follow pseudo-second order adsorption kinetics. Therefore, NPs associated with these many numbers of versatile properties make them a valuable material for the implementation of wastewater treatment (Fig. 5-6) [35].

Figure 5. Morphological analysis of FeO/AgNPs, FeO/AuNPs, and FeO NPs by TEM "Reprint and Copyright permission obtained from Elsevier publication" [35].

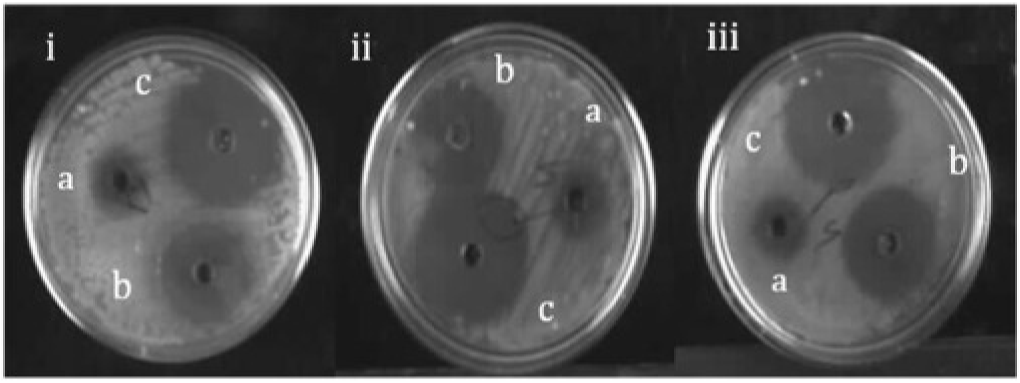


Figure 6. Antimicrobial activity of a) FeO NPs, b) FeO/Au NPs and c) FeO/AgNPs against (i) *P. aeruginosa* (ii) *S. enterica* (iii) *M. luteus* "Reprint and Copyright permission obtained from Elsevier publication" [35].

In the light of wastewater treatment from bacterial infection, nanocomposites composed up of multi-walled carbon nanotubes fabricated with FeO and AgNPs were prepared. TEM, SEM, XRD, XRF, EDAX confirmed the structure, crystallinity, composition, morphology, and surface properties of the NPs. The NPs displayed significant antibacterial potential towards *E. coli* with minimum bactericidal concentration of 200 μg/ml within 8 hours as growth inhibition time [36].

A greener, atom economic and eco-friendly approach towards the synthesis of AgNPs was developed by the group of Jamal. The NPs were synthesized from the leaf extracts of *Cinnamomum tamala* (*Tejpata*) [37]. The characterization of the NPs was done through UV-visible, FTIR, SEM, TEM, XRD etc. The average particle diameter of the NPs was found to be 25 to 30 nm. The prepared recyclable NPs were applied to test for their bacterial inhibition ability against *P. aerginosa* isolated from the hospital's drainage water. The antimicrobial ability of the NPs was studied through agar diffusion method. The results of the antimicrobial study indicated that the AgNPs possess tremendous range of antimicrobial ability against the studied bacterial strain with 17.67 ± 0.5777 as the maximum zone of inhibition over aqueous leaf extract which had only 5.67 ± 0.5777 bacterial zone of inhibition ability. The authors predicted a tentative mechanistic pathway followed by the AgNPs to inhibit the bacterial growth i.e., as the bacteria cell wall is made up of many carboxyl, amino and phosphate groups, there occur a positive charge electrostatic attraction between the AgNPs and the cell wall (Fig. 7-8) [37].

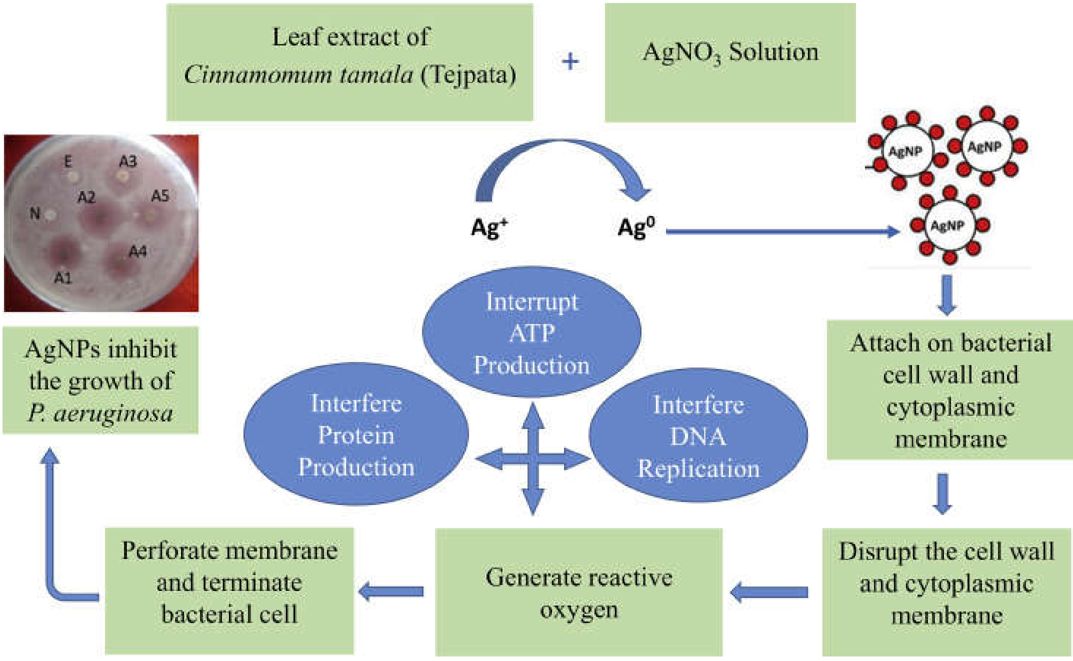


Figure 7. Mechanism of green synthesis of AgNPs and its antimicrobial action "Reprint and Copyright permission obtained from Elsevier publication" [37].

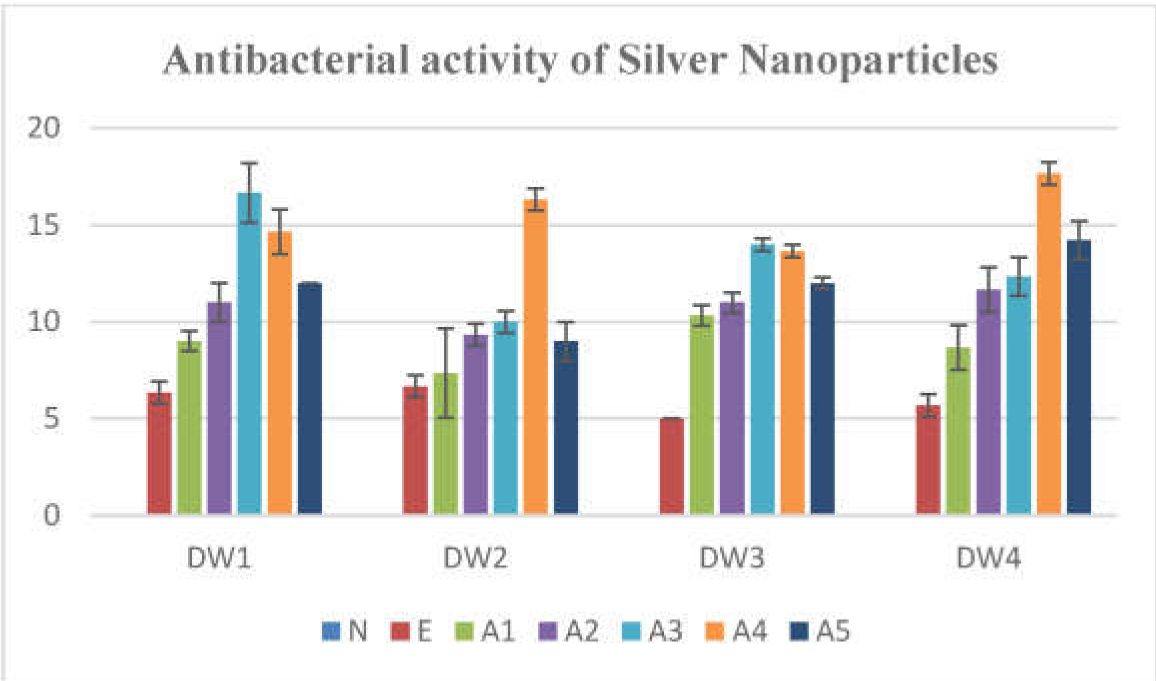
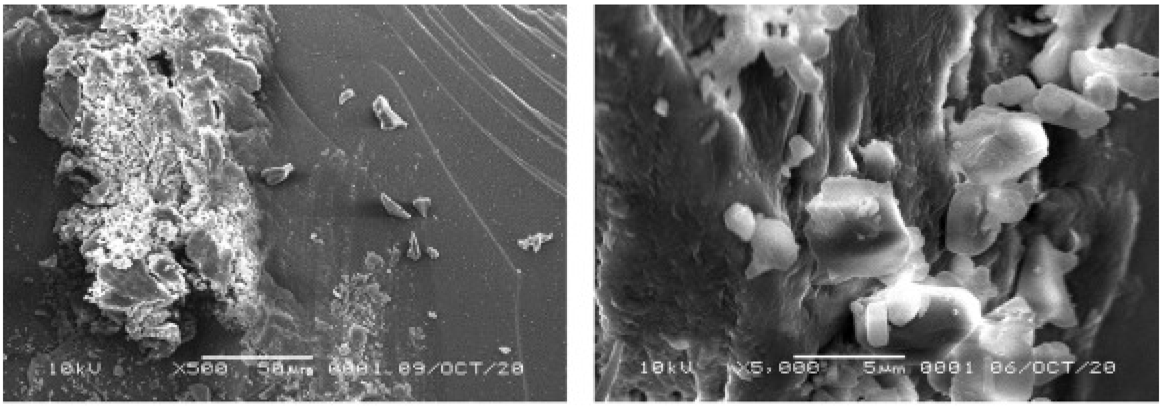


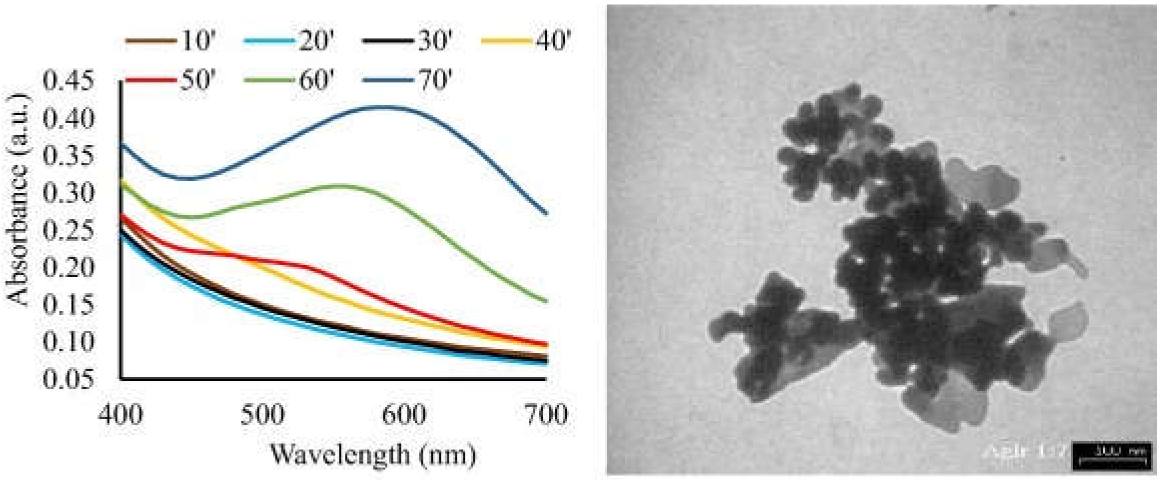
Figure 8. Antibacterial efficacy of AgNPs against multidrug resistant bacteria strain "Reprint and Copyright permission obtained from Elsevier publication" [37].

Chitosan stabilized AgNPs immobilized upon solid silica gel support was prepared following atom economic and environmentally sustainable approach. The chi-AgNPs were generated using MeOH under visible light irradiation and thereafter immobilized using white silica gel beads, which were earlier coated with chitosan i.e., chi-SiG. The formation of the stable, solid, dispersed form of the nanoparticles was confirmed through SEM, TEM, V-visible, FTIR etc. Interaction of different functional groups on the chitosan surface with Ag+ ion makes the NPs highly stable. The prepared nanomaterial was used as air filter to fight against the multidrug resistant bacteria *S. aureus*, *E. coli* and *B. subtilis* resent in the air. The results of the antibacterial study revealed that the NPs work as a strong inhibitory agent the bacterial cell growth in agar medium. Including also, they also exhibited greater antibacterial activity in the air against *B. subtilis* bacterial strain as air filter. Mechanistically, the Ag nanoparticles interacted with the proteins present on the bacteria cell wall and the phospholipids which results in the rupture of the cell wall inhibiting the bacteria cell growth (Fig. 9-11) [38].



(a) (b)

Figure 9. SEM images of AgNPs-[chi-SiG] "Reprint and Copyright permission obtained from Elsevier publication" [38].



(a) (b)

Figure 10. UV-Vis and TEM image of AgNPs "Reprint and Copyright permission obtained from Elsevier publication" [38].

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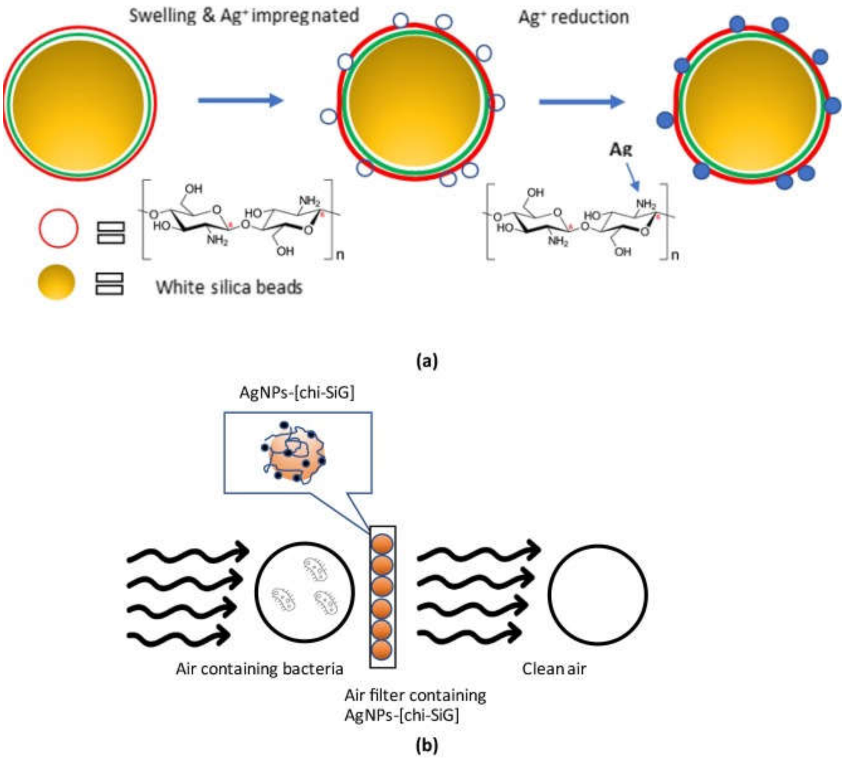

Figure 11. Preparation and antibacterial action of AgNPs "Reprint and Copyright permission obtained from Elsevier publication" [38].

Silver nanoparticle doped on aminated polyether sulfone-based nanocomposite membrane was prepared and was applied for wastewater treatment. Initially the NH_2-PES i.e., APES was formed by the functionalization of $–NH_2$ on polyether sulfone. Subsequent immobilization of the AgNPs on the NH_2-PES surface resulted in the formation of the AgNPs- APES. The formation of the nanocomposite membrane was confirmed by XRD, SEM, EDAX, TEM, FTIR etc. Experimental results suggested that the NPs were formed having diameter 5 nm to 40 nm which were immobilized on the surface of the APES. The nanoparticles were subjected to study for their bacterial inhibition ability against wastewater. It was found that, as compared to unfunctionalized AgNPs-PES, the amino functionalized AgNPs-APES based nanocomposite membrane displayed higher antibacterial potential which might be due to their longer lifetime (nearly about 25 days) due to improved and controlled release of the Ag+ ion. The nanoparticles work against the pathogens by inhibiting their biofouling ability i.e., accumulation of AgNPs on the cell wall of the micro-organisms leads to rupture of their cell wall and ultimately inhibits bacterial multiplication. (Fig. 12 &13) [39].

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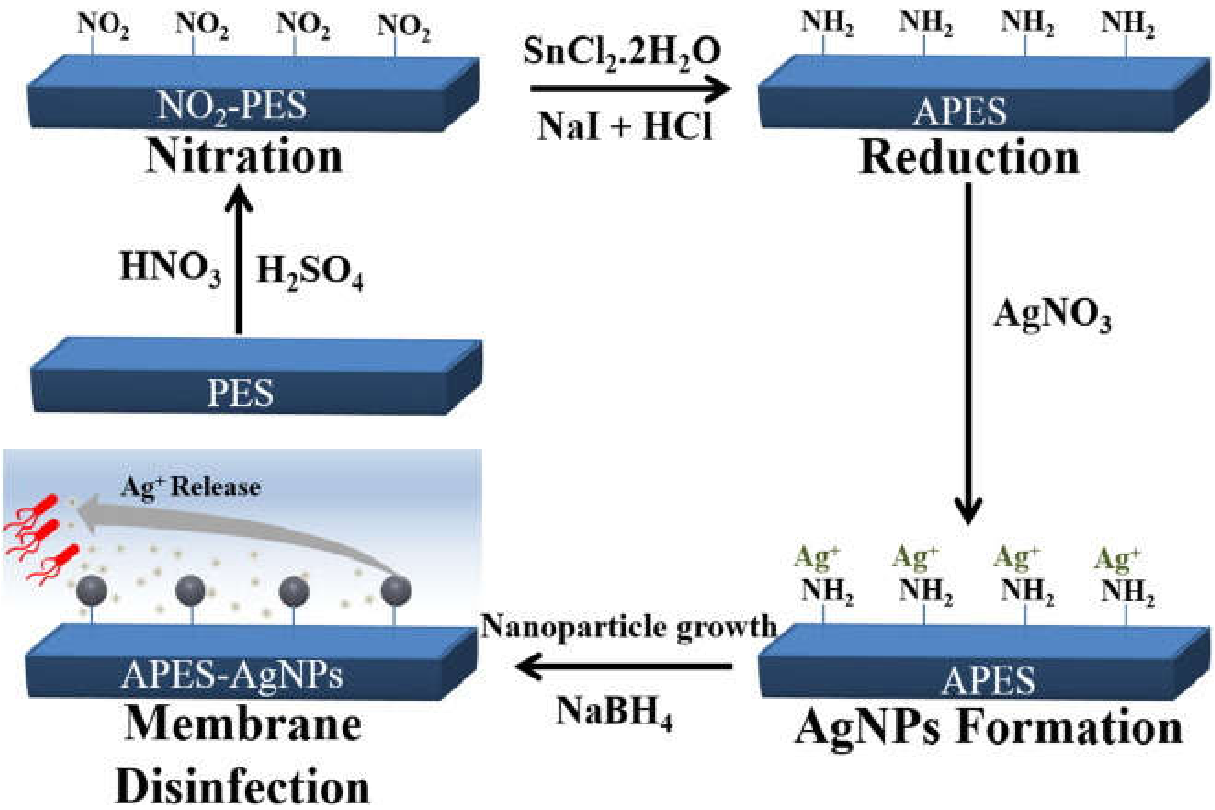

Figure 12. Preparation of aminated polyethersulfone decorated by AgNPs "Reprint and Copyright permission obtained from Elsevier publication" [39].

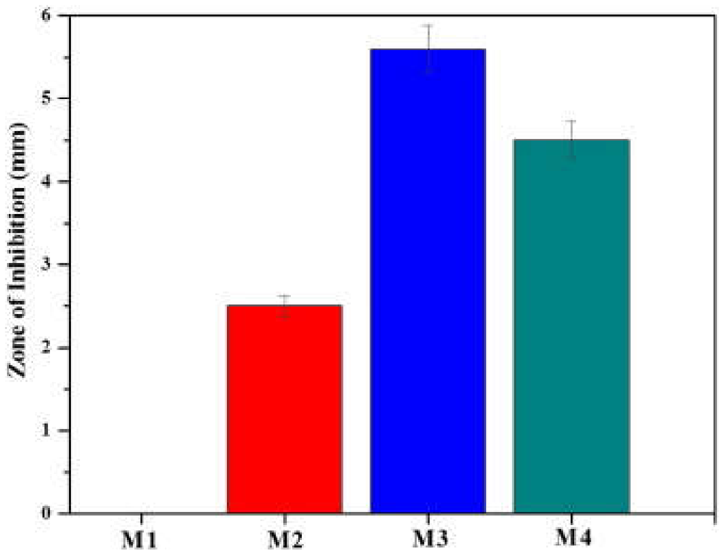

Figure 13. Zone of inhibition of AgNPs against different bacteria strains "Reprint and Copyright permission obtained from Elsevier publication" [39].

Ag nanoparticle decorated PDA/M-ATP/PCN nanocomposites as photocatalyst was developed by reduction of AgNO3 to detect the presence of organic dyes and heavy metal ions in the water samples. According to the experimental results, due to the high surface area of the nanocomposites formed after doping of Ag atoms upon M-ATP and PDA, reduced the bandgap, promoted the photogenerated species separation, improved the synergistic effects of charge separation and ultimately enhanced the photocatalytic efficacy. In addition to this, the as prepared photocatalyst displayed superior catalytic performance and high stability over the conventional g-C3N4 based photocatalysts. The nanocatalysts found to be quite efficient towards the detection of heavy metal ions like Cr (VI) (98.4%)

as well as organic dyes i.e. methylene blue (90.3%) from the waste water samples. the degradation of the MB and Cr (VI) was done by the electrons and OH radicals generated by the Ag based PDA/M-ATP/PCN nanocomposite (Fig. 14 & 15) [40].

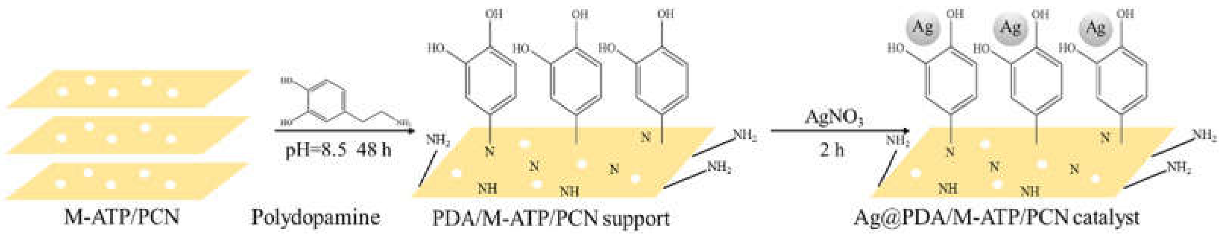

Figure 14. Mechanism of Ag@PDA/M-ATP/PCN synthesis "Reprint and Copyright permission obtained from Elsevier publication" [40].

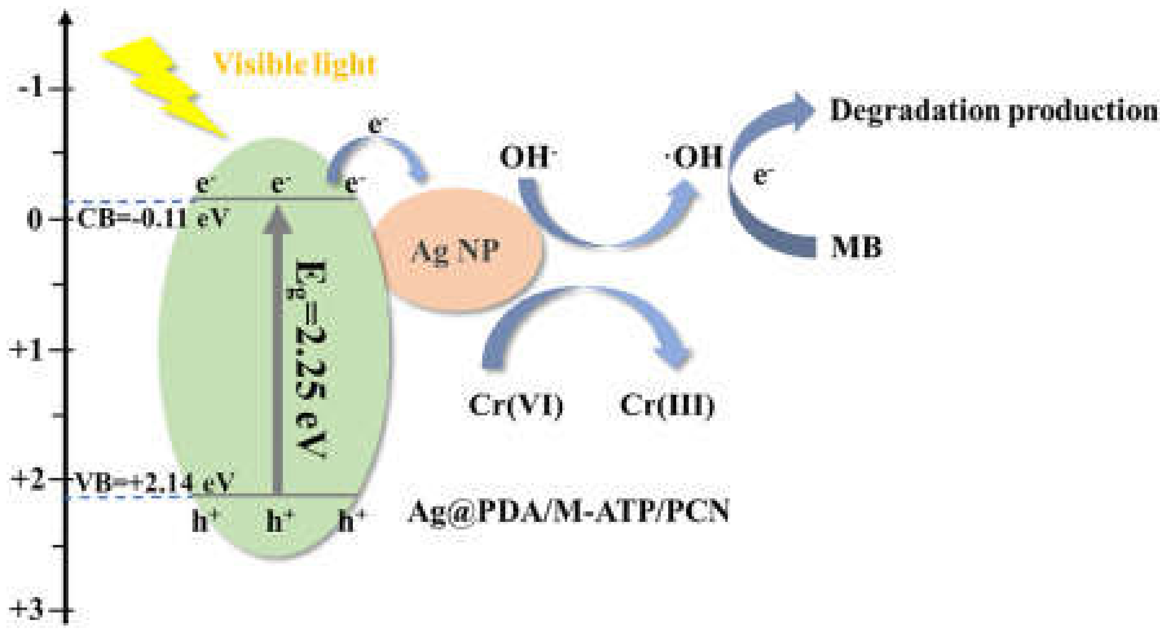

Figure 15. mechanism of photocatalytic action of AgNPs towards Cr (VI) and MB "Reprint and Copyright permission obtained from Elsevier publication" [40].

Synthesis of the AgNPs was achieved by using the aqueous leaf extracts of *Ginkgo biloba* by avoiding the use of toxic chemicals. The optimum reaction condition for the synthesis of AgNPs was found to be 0.5 mL of the leaf extract, 0.5 mM $AgNO_3$ solution 5.0 mL, at 80 °C for 30 mins under alkaline medium i.e., pH 9. Spherical shape of the NPs with average particle size of 14.14 ± 4.44 nm was confirmed through SEM, TEM, EDX, SEAD and DLS study. The synthesized nanoparticles displayed diversified environmental applications such as hindered the growth of both G+ and G- bacterial strains, ten times faster azo-dye degradation following pseudo-first order kinetics. Moreover, it also acted as a fluorescent probe towards the detection of Cr (VI) [41].

3. Conclusion and Future Perspective

In conclusion, this review signifies the importance of the silver nanoparticles towards environmental remediation. Environmental pollution is increasingly globally day by day due to various types of manmade activities which results multiple kinds of air, water, and soil borne diseases. Nanomaterials made up of AgNPs have found a great deal of environmental applications because of their safe and effective synthesis, tiny particle size with high surface area as well as biocompatibility. At low concentration they are found to be more harmful towards the pollutants including dyes, micro-organisms, pathogens, and heavy metal ions, having no adverse effect towards the healthy cells including soil micro-organisms. In this review the authors briefly elaborate the versatile synthesis and applications of AgNPs in the form of photocatalyst, electrochemical sensor, fluorogenic sensor etc. towards the detection of hazardous pollutants in the air, water, and soil medium. All the synthesized nanomaterials possess low LOD value with high sensitivity and selectivity towards the detection of pollutants. Due to their biogenic nature, it can also be further recycled and reused.

This review will help the future researchers towards the more economic and efficient innovation of Ag NP carrying nanoprobes which can be efficient to detect multiple groups of pollutants at the same time with low LOD value along with good reproducibility.

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