Silver nanoparticles loaded on silica nanoparticles and activated carbon are being prepared and developed for use in portable water treatment

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ABSTRACT: A unique method for creating a powerful portable water purifying system was put to the test. The silicate nanoparticles were doped with 3-chloropropyltrimethoxylsilane, coated with silver nanoparticles and activated carbon, and then layers of silica and zeolite were added. This process resulted in the silver nanoparticles loaded on silica nanoparticles and activated carbon composite. For the purpose of measuring water purification in terms of microorganisms, micropollutants, and turbidity, four layers were arranged in a series. Within 6 minutes of the contact time, the system removed about 85% of micropollutants, 97% of microorganisms, and up to 79% of turbidity. The usage of plastic water bottles for activities in remote locations can be reduced by employing this gadget, which does not require an energy source to operate. This research was done to develop a portable, low-cost, and simple-to-use antibacterial water filter.

KEYWORDS: Silver nanoparticles, Activated carbon, Portable water purifier, Water treatment.

■ Introduction

Due to its substantial surface area and exceptional adsorption ability, activated carbon (AC) is the most effective adsorbent for the removal of VOCs [1,2]. While airborne bacteria cannot be successfully collected by AC, certain airborne bacteria can be absorbed by the AC. Because carbon materials have a high biocompatibility, the deposited bacteria can easily proliferate on the AC surface [3]. As a result, indoor airborne bacteria build up in huge quantities on its surface, degrading IAQ as a result of the AC's potential to create bacterial pollution. On the other side, polluted AC may release VOCs due to bacterial metabolism. The secondary indoor air pollution is a result of this. Therefore, antibacterial AC is necessary for optimal IAQ since it may kill germs in addition to removing VOCs. Due to its extensive antibacterial action, silver is well known as a potential antimicrobial agent [4]. Using the single gel approach and the Ag-coated method, numerous silver-containing carbon composites have been created for antibacterial applications. [5–7]. When using the Ag-coated technique, Ag nanoparticles are either directly mixed with AC or deposited as gas-phase Ag nanoparticles. Then, with the solitary gel approach, Ag ions are placed onto the AC and in-situ reduced to Ag nanoparticles. These materials can nonetheless have a good antibacterial effect on deposited germs even when they cannot catch airborne microorganisms. Aggregation of Ag particles and boring reaction process are two major obstacles to the manufacturing of these composites. Because uniform dispersion is a need for sufficient contact and interaction between Ag and microbiological species, the agglomeration of Ag particles may decrease antibacterial effectiveness [4]. Additionally, the large-size Ag particles produced by aggregation have a lower adsorption ability and can obstruct microspores. Currently, the uniform and well-adherent coating of Ag particles on carbon surface to obtain highly efficient

antibacterial action and good adsorption capacity represents a hard issue for silver-containing carbon composites.

Since 97% of the water on earth is salt water, the majority of it is unfit for human consumption. Every continent and about 2.8 billion people worldwide experience water scarcity for at least one month each year. Over 15% of people worldwide do not have access to clean drinking water. If the current trends continue, it is predicted that more than half of the world's population will face water shortages. 162,000 of the 575,000 Indian communities are plagued by unclean water and a lack of water. The goal of this project is to create a portable water purifier that uses nature to reduce the number of microbes in the sample. The inability of low-income individuals to transport pricey electrical water purifiers, together with numerous circumstances or locations where a portable water purifier is required to obtain potable water from untreated sources, served as the impetus for this study. One method cannot be used to solve all problems because different locations have distinct resources and difficulties. Water is frequently pumped from a nearby lake or from the ground, where it may include a variety of viruses, dissolved pollutants, mineral ions, and carcinogens.

It has been established that water purification techniques are insufficient to meet the rising demand for clean water. Additionally, because existing water filtration techniques require a lot of energy, they can only be implemented in certain places. Our goal is to develop an energy-free water filter to address these issues. We intend to address the microbiological concerns in addition to the physical purification aspects. We propose to evaluate and make use of the antibacterial characteristics that readily available plant leaves have.

In this study, functionalized AC and silica were coated with silver to create Ag-coated AC nanocomposites (Ag/ACs), which were then in-situ reduced to produce silver nanoparticles that were evenly disseminated. This method prevents the coagulation and overgrowth of Ag particles in the subsequent reduction process by using Ag's dispersing and stabilizing effect, provided by oxygen-containing groups on the functionalized AC surface. Ag nanoparticles can therefore be evenly distributed on the surfaces of AC and silica. The introduction of uniform Ag nanoparticles on AC not only may provide extremely efficient anti-bacterial activity, but also can retain high adsorption capacity, according to the verification of antibacterial activity and adsorption capacity for toluene as a model VOC.

Our final product will be a water filter (**Fig. 1**) that is capable of removing both biological and physical contaminants from water without the use of energy. This will make it easier for people who live in isolated and impoverished places to benefit from this water filter. Our filter's components are readily available and interchangeable. The filter is divided into compartments so that any element of the filter can be quickly and easily replaced without affecting the integrity of the entire filter. The filter is simple to use and was created with the understanding that the majority of the target demographic is uneducated and lacking in technological skills.
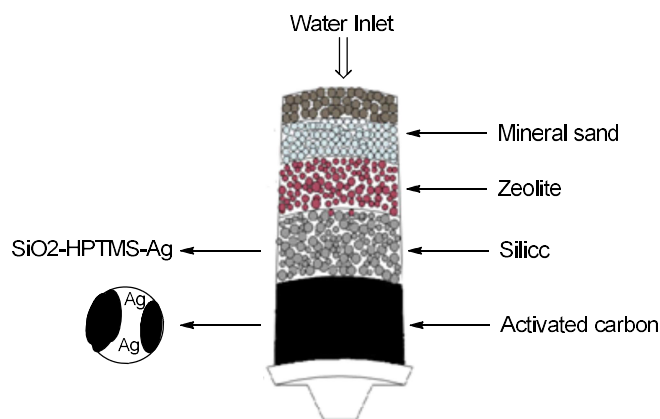
Fig 1. Assembled filter

Design

The four chambers in the water filter's design are easily cleanable and refillable with their corresponding contents. To create a design that is simple to operate and does not require a lot of technical understanding, plastic bottles were cut into sections and used to compartmentalize the filter. The filter includes an open end and a cap on each compartment (Fig. 1) to make it simple for the user to clean the filter without assistance from a professional. Additionally, the filter was created so that it is economical and accessible to regular people.

A layer of muslin cloth in the first compartment, which is designated for primary filtering of the water, aids in removing the large physical pollutants from it. A layer of mesh is added to the bottle's cap for additional filtration. Sand and gravel made up the second chamber, which was then covered in several layers of muslin cloth filter. In order to stop fibers or sand particles from moving with the water, the cap of this container also has a mesh layer on it. A cotton filter is found in the third compartment, and it is followed by a filter made of plant leaf extracts that have antibacterial properties. There is a mesh attached to the cap of this container as well.

The fourth container contains activated charcoal, which aids in eliminating bad odors, ionic dye, as well as gases produced by water-borne bacteria. Multiple layers of non-absorbent cotton are layered over the activated charcoal layer to assist stop the flow of water and charcoal particles.

Sand

It purifies the water by removing coliform bacteria, algae, color, and iron and manganese concentrations. A complex biological layer that naturally forms on the surface of the sand, which only serves as a substrate for the filtration, is how sand filters work. Slow sand filters function by forming a gelatinous layer or biofilm made up of bacteria, fungus, and protozoa on top of a layer of fine sand [8,9]. The layer that provides excellent filtration in drinking water is the surface bio-film.

The underlying sand acts as the support medium for this biological treatment layer, which treats water. Particles of foreign material are caught in the mucilaginous matrix as water exits the middle of the hypogeal layer, and soluble organic material is adsorbed. The bacteria, fungus, and protozoa break down the contaminants in the environment. It is anticipated that water generated by a slow sand filter will be of outstanding quality and significantly lower in bacterial cell count [10].

Testing antibiotic properties of leaves

The antibacterial abilities of the leaves of tulsi (Ocimum tenuiflorum), guava (Psidium guajava), neem (Azadirachta indica), and duranta repens (Duranta repens) were examined. For each of the leaf samples, a leaf extract was made. To create an extract tablet, tiny filter paper tablets were floated in a water extract solution with each leaf. On a Petri plate with Nutrient Agar (2.8 g/100 mL) streaked with serially diluted lake water, this and a full dried leaf were taken to test for the zone of inhibition. These plates were kept in an incubator for 24 hours at 37°C and observed.

Results
pH test

Imam Ali University's water sample was determined to have a pH of 5.38. By lowering the water's pH to 6.92 and bringing it within the permitted range of 6.5-8.4 (**Table 1**), this filter was also helpful in reducing the acidity of the water (**Table 1**).

Presumptive tests

Both lake water and water that had passed through the filter underwent a most probable number (MPN) test. Each water sample was successively diluted to 10 mL, 1 mL, and 0.1 mL, then added to 5 test tubes of each batch of Luria broth. Each test tube also received one of Durham's tubes. Results were obtained after roughly 24 hours of keeping both sets at 37°C.

Confirmation test

On Petri plates streaked with lake water as well as those streaked with water following passage through filter and charcoal layer, colonies had a distinctive green sheen (blue-black in certain cases). Similar to the last test, less colonies appeared in the case of filtered water.

Table 1. Physical analysis of water samples				
Sample taken	pH	Odor	Turbidity	MNP index mL
Lake water	5.31	Foul odor	Particulate matter	240
Filtered water	6.99	No odor	Absence of turbidity	17
MPN: Most probable number				

Completed test

Pink, non-mucoid colonies were discovered, indicating that the majority of the E. coli in the water samples were Gram-negative, lactose-producing strains. The Petri plate holding the lake water sample contained the most colonies of this type. After passing through the filter and charcoal layer, the growing and number of these colonies were seen to be reduced in the Petri plates containing water. It was determined that the sand layer was mostly to blame for a decline in microbial content following the passage of water from the filter

since the number of colonies in a Petri plate having water from the filter was much lower than that of a plate holding water from the charcoal layer.

Testing antibiotic properties of leaves

Due to the low concentration of leaves in the water extract, there are very small zones of inhibition compared to those often discovered by the antibiotic action of conventional antibiotics. Therefore, it was also suggested that a layer of leaf extract in acetone [10] or methanol might be added to the filter, which will not only be simple to change but also exhibit stronger antibacterial activity.

Silver analysis

The concentrations of silver ions in the raw and treated water were determined using inductively Coupled Plasma-Mass Spectrometry (G8421A). The following standard solutions of silver ions were prepared; 0.625 ppm, 1.25 ppm, 2.5 ppm, and 5.0 ppm.

Determination of water hardness

The method for determining the hardness of the water was modified from Nielsen et al. (2017). In a nutshell, 3 g of EDTA disodium salt were weighed into a 500 ml beaker to create a 0.02 M EDTA solution. The salt was dissolved, transferred to a 1000 ml volumetric flask, and diluted with distilled water until the proper consistency was achieved. 100 ml of distilled water was used to dissolve 0.4 g of $CaCO_3$ in a 500 ml volumetric flask to create the calcium chloride standard solution. Once there was no longer any effervescence and the solution was clear, 50% HCl was added. With distilled water, the solution was applied to the mark. By weighing 6.75 g of ammonium chloride in 57 ml of concentrated ammonia in a 100 ml volumetric flask and adding distilled water to the mark, ammonia-ammonium chloride buffer was created. Three 250 ml Erlenmeyer flasks each received 50 ml of the calcium chloride solution in order to standardise EDTA. The flasks were filled with 5 ml of the ammonia-ammonium buffer and 5 drops of the Erichrome Black T indicator. EDTA was titrated into the solution until the colour transitioned from red to blue. Based on the calcium chloride solution's content and the amount of EDTA used to titrate, the average molarity of the EDTA solution was determined. A conical flask was filled with 50 ml of water that had been treated with clay discs without the addition of AgNPs or silver nitrate. Five millilitres of buffer and five drops of Ferrochrome Black T indicator solution were then added. After that, the solution was titrated with 0.01M EDTA until it was sky blue. For the remaining ten water samples from clay discs, the same process was repeated.

Determination of chloride in raw water and treated water

According to earlier literature, the mohr titration was employed to determine the chloride concentration. 0.5g of $AgNO_3$ was, in essence, dried for two hours at 100°C and then allowed to cool. In a conical flask, 200 ml of distilled water were used to dissolve 0.418 g of solid $AgNO_3$ that had been precisely weighed. In a brown bottle, the prepared silver nitrate solution (0.01 molL1) was kept. In order to make potassium chromate indicator solution (0.025 molL-1), 0.1 g of K_2CrO_4 were dissolved in 20 ml of distilled water. First, a silver nitrate solution was poured into the burette. A tidy 50 ml conical flask was used to extract the material in 50 ml. A 1 ml aliquot of potassium chromate indicator was added to provide a white yellow hue. Silver nitrate solution was then used to titrate it. The initial appearance of a reddish silver chromate colour served as the titration's endpoint.

Determination BOD in water samples

It uses the Winkler method as modified by Montgomery et al in 2007. In a 1000 ml volumetric flask, 600 g of manganese (II) chloride (3 M: reagent grade), MnCl2.4H2O, was dissolved in 600 ml of distilled water and diluted to the proper concentration. In a 1000 ml volumetric flask, 600g of NaI was dissolved in 600 ml of distilled water, and 320g of NaOH was added. The solution was then diluted to the proper strength with distilled water. 500 ml of distilled water and 500 ml of sulphuric acid (50% v/v) were slowly combined. A 1000 ml volumetric flask was filled to the proper level with distilled water after adding about 100 ml of sodium thiosulfate working solution (0.018 M, reagent grade). To make the potassium iodate standard (0.00167 M = 0.01 N: analytical grade), 0.3567 g of KIO3 were dissolved in 1000 ml of distilled water, and the solution was then diluted to the proper concentration. A 60 ml reagent bottle was filled completely when the sample was transferred there. Two reagent bottles were utilised for each sample, with the other bottle being tightly closed and incubated at 25 0C in the dark. For the water in the first reagent bottle, the amount of dissolved oxygen was calculated. The reagent bottle was filled with 1 ml of sulphuric acid, followed by 1 ml of potassium permanganate, and everything was thoroughly mixed. To get rid of the orange colour, 1 ml of potassium oxalate was added after 10 minutes. The water sample received 3 ml of potassium iodide; after 30 minutes, a brown colour was seen after the addition. Sodium thiosulphate was then used to titrate the water samples.

■ **Discussion**

The manufacturing of the water filter's economics are getting better as time goes on. According to the Indian government, 22% of the population lives below the poverty level. Any sector or business that decides to produce our product will already have a sizable consumer base that has to be reached. Since there is now a cheap, accessible water filter on the market that doesn't require energy, the impoverished people, who make up the project's main shareholder, gain. The highest documented price for plastic in the last two months is Rs. 25 for 1 kg. To create up to 18 g, 25 polyethylene terephthalate (PET) bottles are required. As a result, 1 kg contains about 1388 plastic bottles. By applying that reasoning, we can make the 347 recommended filters by purchasing 1388 PET bottles for just Rs. 25. One kg of activated charcoal costs Rs. A half kg of charcoal is needed for each filter, which brings the price of the charcoal used in our filter to roughly Rs. 80. River sand has a price of Rs. 43/kg. Sand costs Rs. 22 if around half a kg is utilised. A kg of cotton costs roughly Rs. 220. If roughly half kg is utilised once more, the price comes to Rs. 110.

Advantages of the filter:

-This filter's parts are compartmentalized for ease of replacement.

-The majority of parts are readily available.

-In Indian surroundings, the plants used in the microbiological component are widely encountered.

The final filtrate will be pH neutral, odorless, colorless, and free of microorganisms.

- Extremely frugal.

- Does not require electricity or sophisticated technological expertise to function

Modification of nanosilicc surface

The produced silica nanoparticles were disseminated in 100 ml of EtOH, followed by the addition of 2 ml of toluene, 30 minutes of sonication, and the addition of 2 ml of CPTMS dropwise to the reaction mixer. Under an inert atmosphere, the mixture was swirled for 10 hours. The modified nanoparticles were once more isolated using a filter, and the precipitated particles were then repeatedly washed with MeOH. (Figure 2).
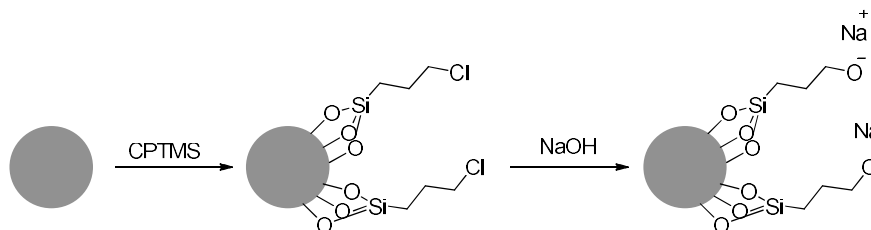
**Figure 2**. Modification of silicc surface with 3-cloropropyltrimethoxysilane and NaOH

Synthesis of SiO2-HPTMS-Ag

The modified nanoparticles were then added to the 0.5 gr of AgNO3 and 10 ml of CHCl3 solution. The cream solid was washed with CHCl3 and dried after the suspension was vigorously agitated and refluxed for 4 hours. (Figure 3).
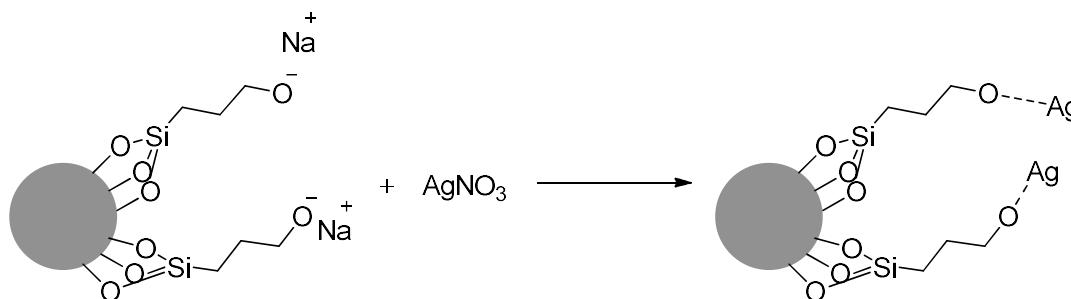

Figure 3. Synthesis of SiO2-HPTMS-Ag

As seen in Fig. 4, Ag-coated AC nanocomposites (Ag/ACs) were created by oxidizing AC and attaching and reducing Ag ions in-situ to the surface of the material. First, concentrated HNO3 was used to oxidize AC, producing functionalized AC (F-AC) that contained oxygen-containing groups like hydroxyl (-OH) and carboxyl (-COOH). Due to its strong affinity for oxygen, Ag ions (Ag) produced from aqueous AgNO3 solution can then be stably anchored on F-AC surface through ion-dipole interactions [11].
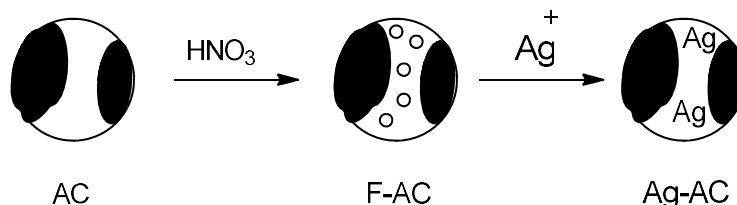

Fig. 4. Preparation of Ag-coated AC nanocomposites.

Then, employing trisodium citrate (Na3 Ct) and dimethylethanolamine (DMAE) as reducing agents in combination, Ag ions on the F-AC surface were in situ reduced to Ag nanoparticles to produce Ag/ACs. Ag ions are stably and evenly dispersed on the surface of F-AC because the oxygen-containing groups on

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its surface can firmly attach Ag through ion-dipole interactions. The generated Ag nanoparticles can be evenly dispersed on the ionized selective sites of the F-AC surface after the Ag ions on its surface were decreased, preventing coagulation. Using SEM and an EDS probe, the surface morphology and elemental makeup of Ag/ACs were investigated. SEM images of AC and Ag/ACs are shown in Figures 5 and 6, respectively.
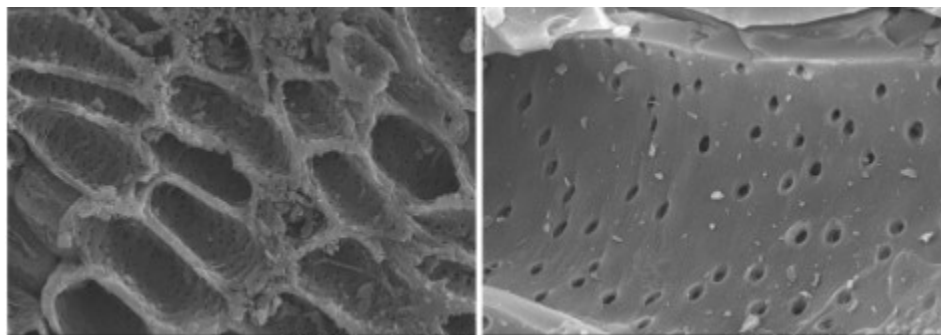
Fig 5. SEM images of AC (right is magnified picture)
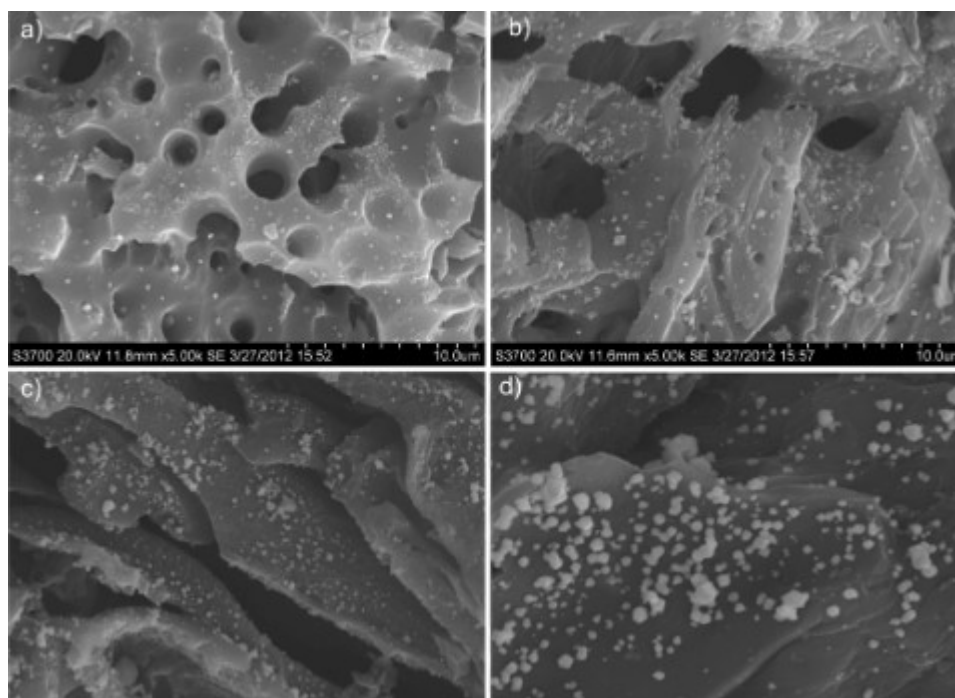
Fig 6. SEM images of AC (right is magnified picture)

Specific surface area (SBET) and pore volume of Ag/ACs were measured in order to assess the impact of Ag particles on the micro-porous structures of AC, and the results of the characterization are shown in Table 2. The specific surface area of ACs is higher than that of Ag/ACs. Nitric acid widens micropores with smaller pore sizes, which results in a decrease in the specific surface area, as seen by SEM images (Figs. 5 and 6). Additionally, a clear trend shows that, below 3% AgNO3, increasing AgNO3 concentration only slightly reduces SBET and pore volume. S BET and pore volume clearly decrease when AgNO3

concentration exceeds 3%. Because the aggregated Ag particles clog interior pores, SBET and pore volume of Ag/AC-4 (3.2%) are 20% lower than those of the immaculate AC. As previously mentioned, the oxygen-containing groups on the surface of the F-AC function to disperse and stabilise the Ag ions through ion dipole interactions. As the concentration of AgNO3 rose, these effects became less noticeable, which allowed extra Ag ions to move around and eventually cause Ag particles to coagulate as Ag ions were reduced. Because the big Ag particles partially obstruct the micropores, the SBET and pore volume reduced as a result.

Atomic adsorption spectroscopy was also used to determine the amount of silver on Ag/ACs. AgNO3 concentration increases the quantity of Ag on Ag/AC surfaces. These experimental findings show that the microporous structure of AC is not considerably altered by the presence of Ag particles with a content of less than 1.65%, maintaining a higher degree of adsorption capability.

Table 2. Microstructural properties and Ag content of AC and Ag/ACs.

Sample	$SBET/m^2g^{-1}$	V/cm^3g^{-1}	Ag/wt%
AC	707	0.363	0
Ag/AC-1	698	0.364	0.6
Ag/AC-2	689	0.357	0.99
Ag/AC-3	672	0.342	1.57
Ag/AC-4	573	0.285	3.26

Experimental section

Each and every one of the chemicals and solvents were bought off the shelf and used right away. Chinese company Tianjin Kemel Chemical Co. Ltd. was used to obtain coconut shell activated carbon (AC). Sinopharm Chemical Reagent Co. Ltd. in China provided the silver nitrate (AgNO3), trisodium 2-hydroxypropane-1, 2, 3-tricarboxylate hydrate (Na3 Ct), and dimethylethanolamine (DMAE). The Chinese company Guangzhou Jian Yang Biotech Co. Ltd. provided the typetone, agar, and yeast extract. The studies were conducted with deionized water.

Preparation of Ag-coated AC nanocomposites (Ag/ACs)

AC was oxidized by concentrated HNO3 (69 wt% HNO3) in accordance with the method described in reference [15] to produce functionalized AC (F-AC) with oxygen-containing groups. The produced F-AC (1 g) was impregnated with a 50 mL solution of AgNO3 and agitated at room temperature for one hour. AC loading Ag (Ag/AC) was obtained following filtration. After that, 50 mL of trisodium citrate (Na3Ct) solution was added together with the resulting Ag/AC. The reaction system was then agitated for 2 hours at 80 C while 1 mL of a dimethylethanolamine (DMEA) solution was added dropwise to the reaction mixture. Following filtering, the solid was separated and cleaned with deionized water. The sample was

then vacuum-dried at 70 C for 12 hours to produce Ag-coated AC nanocomposites, also known as Ag/ACs. **Table 2** contains a list of Ag/AC preparation parameters.

■ Conclusion

We have successfully created a method to create homogeneously distributed Ag-coated activated carbon nanocomposites. Through the attachment of Ag to the functionalized AC with oxygen-containing groups and the subsequent in-situ reduction of Ag, Ag particles were firmly coated on AC surface. By adjusting the proportion of AgNO3, it is possible to create Ag-coated AC nanocomposites (Ag/ACs) with evenly dispersed Ag nanoparticles and various compositions.

Because of the evenly distributed distribution of Ag particles on the surface of the AC, antibacterial tests showed that Ag/ACs may effectively kill bacteria in both an aqueous and an airborne environment. More importantly, the shape of AC and toluene adsorption capacity are not significantly altered by the addition of moderate Ag nanoparticles. In addition to effectively killing airborne bacteria, Ag/AC-2 with optimized Ag content (0.98%) also maintains nearly the same toluene adsorption performance as AC. Ag-coated activated carbon nanocomposites show great potential for removing airborne microorganisms and VOCs in IAQ regulation.

The silicc water filter SWF created in this experiment performed well. By lowering the concentrations of some water parameters, such as nitrates, chlorides, water hardness, pH, and BOD, the silicc/AgNPs composites are able to treat water. When compared to clay alone, which only reduced pH by 1.08%, the manufactured clay/AgNPs composites shown more efficacy in doing so, by a percentage of 12.0%.

67.0% less water hardness was produced compared to 7.70% for filters made only of clay. Silicc/AgNPs nanocomposites reduced BOD by over 100% compared to clay-alone discs, which only reduced BOD by 50%. AgNPs and AgNO3 CWFs shown that they are ineffective at lowering nitrate levels. The silic/AgNO3 filters appear to have leached more silver ions as seen by the AgNO3 percentage rise of 84% compared to AgNPs 73%. Clay filters and silicc/AgNPs filters each reduced chloride concentrations by 5.64%, 54.0%, and 65%, respectively. AgNPs nanocomposites show a larger potential for usage in the purification of water for human consumption when compared to other filter types evaluated in this work.

Acknowledgments

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