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Water Purification through a Novel Electrospun Carbon Nanofiber Membrane

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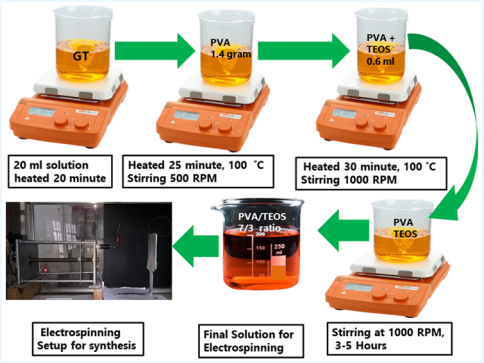
ABSTRACT: Here, we report water purification through novel polyvinyl alcohol (PVA)-based carbon nanofibers synthesized through the electrospinning technique. In our novel approach, we mix PVA and tetraethyl orthosilicate (TEOS) with green tea solutions with different concentrations to synthesize carbon-based nanofibers (CNFs) and further calcine at 280 °C for carbonization. The scanning electron microscopy (SEM) results show the diameter of the nanofibers to be ∼500 nm, which decreases by about 50% after carbonization, making them more suitable candidates for the filtration process. Next, using these carbon nanofibers, we prepare filters for water purification. The synthesized CNF filters show excellent performance and successful removal of contaminants from the water by analyzing the CNF-based filters before and after the filtration of water through SEM and energy-dispersive X-ray (EDX) spectroscopy. Our SEM and EDX results indicate the presence of various nanoparticles consisting of different elements such as Mg, Na, Ti, S, Si, and Fe on the filters, after the filtration of water. Additionally, the SEM results show that PVA and TEOS concentrations play an important role in the formation, uniformity, homogeneity, and particularly in the reduction of the nanofiber diameter.



1. INTRODUCTION

We have been racing against time to devise an economical and sustainable technology for water purification due to the rapid growth in population. Noteworthy is the fact that at the turn of the millennium there had been over 1 billion individuals who had no access to safe drinking water with 4000 children below the age of 5 who died because of unclean water. However, with the joint efforts of the World Health Organization and United Nations, access to clean water to a significantly large percentage of the population has improved; however, still a large number of people in the world have no access to clean and safe drinking water, especially in developing countries.[1]

Nowadays, many consumer products contain heavy metals, metal oxides, and non-metallic nanoparticles (NPs), the occurrence of which increases their probability to be released into natural water and generate an unhealthy environment.[2] Natural water is often contaminated by nanosized inorganic materials, such as arsenic, iron, lead, mercury, copper, nickel, zinc, cobalt, silver, sodium, potassium, magnesium, and so forth, making it unsafe for consumption. It is estimated that around 1300 fiber-based products are being marketed and hence are likely to enter the aquatic systems causing health concerns due to their persistency in water systems.[3]

The nanofibers synthesized by the electrospinning technique have remarkable characteristics such as a large surface to volume ratio, excellent morphology, and chemical activity rendering them a suitable candidate for various applications such as sensors, composites, and for air and water purification.[4] There are several polymeric fibers, including natural (silk, cotton, etc.) and synthetic (polyester, polyamide, etc.) that can be used as adsorbents for the elimination of dyes and metal ions from wastewater.[5] These include fibers based on polyvinyl alcohol (PVA), polyacrylic acid, and pullulan, excellent adsorbents for Cr(VI) removal.[6] FIBAN ion-exchange fibers show the best performance for water softening and removal of heavy metal ions including Cu^{2+}, Co^{2+}, Zn^{2+}, Ni^{2+}, Pb^{2+}, and Cd^{2+}.[7] The composite of cotton fibers and the ZrO^{2+} adsorbent shows high capacity for Cr(VI) ion removal.[8] The carboxyl group-containing hydrazine fibers are excellent candidates for the extraction of zinc, copper, cadmium, lead, chromium, nickel, and cobalt.[9] The polyacrylonitrile (PAN)-based fiber contains a bis-amide group having significant capacity for the removal of Cu^{2+}, Co^{2+}, Zn^{2+}, Mn^{2+}, Mg^{2+}, Cr^{3+}, and Hg^{2+}.[10] A poly(acrylphenyl amidrazone-phenyl hydrazide) chelating fiber showed excellent performance in the removal of V(V), Bi(III), Ga(III), Ti(IV), and In(III) from

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wastewater.[11] Similarly, PAN fibers were used as an adsorbent for the extraction of copper, lead ions, and humic acid from aqueous solutions.[12] A poly(acryl benzoylamidrazone-acryl-benzoyl hydrazine) chelating fiber is reported to be highly efficient for the removal of Pd(IV) and Au(III) ions from the solution.[13] Polyamide fibers displayed a maximum adsorption capacity of the adsorbent for Cu^{2+} and Pb^{2+} ions.[14] PAN−polyamidoamine-based nanofibers are being used for the elimination of Direct Red 80 (DR80) and Direct Red 23 (DR23) dyes.[15] Composite nanofibers modified with dendrimer-coated CeO_2 NPs manifested remarkable efficiency to get rid of chromium and phenol.[16] The N6-PPI-based nanofibers displayed successful and efficient extraction of anionic AR252 organic and inorganic dyes from water.[17]

Carbon nanofibers (CNFs) have attracted considerable attention in the field of water filtration, as they exhibit unique features such as high porosity, large surface to volume ratio, low density, and high flexibility.[18] The fiber-based membranes have small pore sizes, low cost, and high efficiency in terms of the removal of contaminants from water.[19,20] While the production of a free-standing non-polymeric nanofiber mat is still a technological challenge due to their intrinsic brittleness, large-area CNF sheets with high mechanical strength and flexibility can be easily fabricated. In addition, their higher chemical stability compared to polymeric filters makes them suitable for water filtration.[21] Polyvinyl alcohol (PVA) is a semi-crystalline fiber, which in comparison with other fibers has a large carbon content (54.5%) and easily splits the hydroxyl groups in the polymer chain that make PVA a favorable precursor for the fabrication of carbonaceous materials. So far, PVA cross-linked to cellulose nanofibers from a hybrid aerogel has been used as a superabsorbent for oils, organic solvents, and different heavy metals and, hence, are potential candidates to be used for water purification.[22] Additionally, outstanding characteristics such as high mechanical strength, high flexibility, high thermal stability, and unique morphology of the PVA-based nanofibers makes them preferable over conventional nanofibers based on poly(furfuryl alcohol), polyvinylidine chloride, polyimide, PAN, polyvinylidine fluoride, and PVA.[23] However, the existing PAN-based nanofibers have low performance due to laborious synthesis protocols which result in an inefficient membrane morphology with a large pore size, thereby leading to inadequate filtration.[24] To overcome these issues in PAN-based CNFs, we propose the novel PVA-based CNFs, prepared through green nanotechnology and are more flexible, adhesive, and have high tensile strength as compared to CNFs based on PAN.[19,25]

Electrospinning has been a pioneering technology for the production of ultra-thin membranes.[26] It is very simple process, which can also be used for the fabrication of nanofibers on an industrial scale.[27] Its various parameters, that is, different operating voltages, jet diameters, and the distance between the needle and the collector affect the morphology of nanofibers.[28] Polyvinyl chloride was added to polyvinylpyrrolidone (PVP) using an electrospinning process with different percentages so that the surface of the nanomembrane becomes hydrophilic and gives the best performance.[29] In this study, we report the development of efficient CNF membranes for the removal of toxic NPs from aqueous solutions. Free-standing CNF membranes were fabricated by electrospinning the PVA precursor solution followed by thermal treatment of the fibers. Moreover, tetraethyl orthosilicate (TEOS) was added for the flexibility and to increase the specific surface area of the CNFs.[30] To the best of our knowledge, deriving CNFs from PVA/TEOS and the synthesis of the CNF filter through green nanotechnology to provide safe and healthy drinking water at affordable prices have not been reported elsewhere. Our results reveal the removal of Mg, Na, Ti, S, Si, and Fe by filtration of contaminated water through the CNF filter. Interestingly, we also find that these membranes could efficiently reject various NPs of different sizes and nature from the contaminated water due to the small size of the membrane pores. Additionally, we also present the synthesis of continuous and uniform diameter carbon nanofibers and its nanofilter. These filters are supposed to be cost-effective, portable, and can be easily installed on domestic taps and pipes, and so forth.[31]

2. RESULTS AND DISCUSSION

2.1. X-ray Diffraction Analysis. Figure 1a shows the X-ray diffraction (XRD) analysis of the composite and carbonized nanofibers based on PVA/TEOS. As shown in Figure 1a, there is only one main peak, where the values of 2θ are 22.67 and 20.48°, while the intensities recorded are 415 and 595 for the composite and carbonized nanofibers, respectively. The overall structure is amorphous as is evident from the XRD pattern.[32] In carbonized nanofibers, the peaks increased, due to the arrangement of different atoms. The small amount of crystallinity in carbonized nanofibers is due to carbonization.[33]

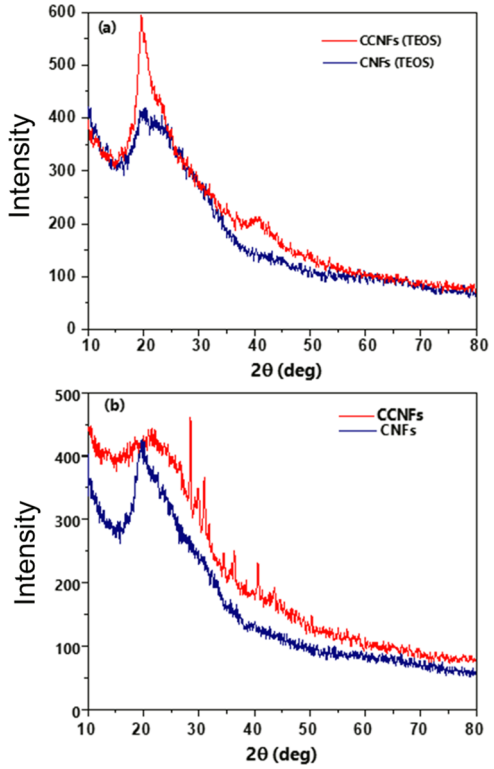

Figure 1. XRD patterns of carbonized and composite CNFs based on (a) PVA/TEOS and (b) pure PVA.

Figure 1b shows the XRD analysis of the carbonized and composite nanofibers based on pure PVA. As shown in Figure 1b, only one peak at 2θ values of 19.61° is observed where the intensity is recorded at 425 for the composite and two peaks at

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2θ values 22.51 and 29.5° are observed where the intensities are recorded at 440 and 460, respectively, for carbonized fibers. Small peaks at 32.3 and 39.5° are observed in the carbonized fibers at different angles which is attributed to carbonization.[34] The overall structure is amorphous.[35]

2.2. Fourier Transform Infrared Spectroscopy Analysis. Figure 2 shows the Fourier transform infrared (FTIR) spectrum of the carbonized and composite nanofibers. The material was used in the powder form for FTIR analysis. The resolution of FTIR was 4 cm^{-1}. The composite nanofibers were heated at 280 °C in a furnace. The spectrum of carbonized nanofibers showed the O−H stretching vibration peak at ∼3360 cm^{-1} of the bonded H_2O molecules (which is a very small quantity in the solid sample) with PVA. The shift in the peak from ∼3280 cm^{-1} (in composite nanofibers) to ∼3360 cm^{-1} and less intensity of the peaks may be caused by water vaporization.[36] The vibration band at ∼2915 cm^{-1} (in composite nanofibers) of C−H stretching in alkanes appears at ∼2909 cm^{-1} in the carbonized nanofiber spectrum, which is due to the decomposition of PVA molecules.[36] The absorption band from ∼2300 to 2050 cm^{-1} is due to the presence of the C≡C stretching vibration, indicating the presence of polyphenols from the green tea extract, and it is slightly shifted (carbonized nanofibers) due to the decomposition of green tea.[37]

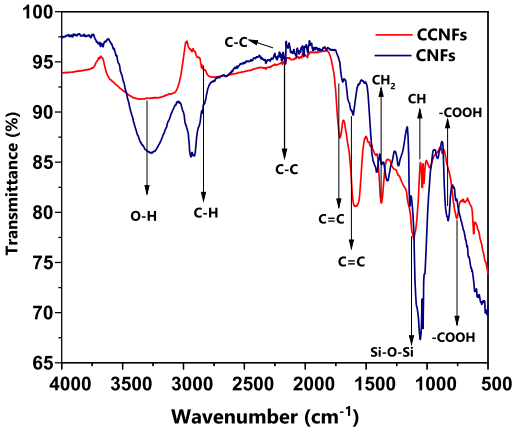

Figure 2. FTIR patterns of the carbonized and composite nanofibers in the 500 to 4000 cm^{-1} range.

The green synthesis of CNFs has been published by many researchers using a green tea extract, which is a cost-effective and domestic source. The green tea extract was preferable because of its environment-friendly nature, solubility in water at room temperature, and does not give any toxic byproducts. The green tea plant extracts are affluent in polyphenols and caffeine, which have excellent antioxidant characteristics. Hoag et al. reported the synthesis of stable nanoscale zero valent iron (nZVI) at room temperature by using a green tea extract without the addition of any surfactant or polymer.[38] The polyphenols in green tea leaves act as a reducing agent and a capping agent, which results in stable green CNFs with unique properties. Similarly, Shahwan et al. prepared nZVI by the electrospinning technique using a 0.10 M iron chloride solution to green tea in 2/3 volume ratios. The prepared nZVI fiber having a 40−60 nm diameter is used as a catalyst for the degradation of methylene blue and methyl orange dyes.[39] Ponder et al., in another study, fortunately, synthesized the nZVI with a diameter of 10−30 nm on a nonporous, hydrophobic polymer resin support. The synthesized nZVI shows a high reactivity toward the removal of metal-ion impurities in aqueous solution.[40] Keeping these points in view, we employed green tea as a starting material in our study. It shows a better adsorption effect on our synthesized CNFs, as discussed in the forthcoming sections.

The peak at ∼1715 cm^{-1} is due to the C−O stretching vibration from the remaining alcoholic group in the carbonized nanofiber sample.[41] The peak at ∼1615 cm^{-1} of the C═C stretching vibration is slightly shifted to ∼1550 cm^{-1} due to the carbonization of composite nanofibers. The peaks at ∼1415 ∼1319, ∼1230, and ∼1140 cm^{-1} observed in the composite nanofiber spectrum have been attributed to C−O and C−C stretching vibrations which disappear after carbonization, and a flat peak at ∼1350 cm^{-1} appears due to the C−C stretching vibration.[37] The peak at ∼1110 cm^{-1} after carbonization is ascribed to the C−C stretching vibration, which is an indication of the crystalline region in the composite nanofibers. The sharp peak at ∼1110 cm^{-1} corresponds to the Si−O−Si stretching vibration, but it is slightly shifted due to carbonization.[42] The absorption peaks at ∼872 cm^{-1} and the less intense absorption band from ∼745 to ∼620 cm^{-1} are attributed to the characteristic vibrations of Si−O−Si and C−Si asymmetric stretching in carbonized nanofibers, respectively.[43]

2.3. Morphological Analysis. *2.3.1. Effects of Heating on the Morphology of Nanofibers.* The scanning electron microscopy (SEM) images of the composite and carbonized nanofibers are shown in Figure 3a−d. The composite nanofibers were heated overnight at 100 °C. Then, it was collected from an Al collector for carbonization. For a morphological and roughness study of the synthesized nanofibers, the samples were prepared in a powder form. Subsequently, the samples were coated with a platinum thin layer via the sputtering technique to make the surface conductive and to avoid any possible charging effect while performing SEM analysis. The coated samples were then loaded into a SEM measurement chamber under high vacuum and were examined at a high voltage of about 3−10 kV.

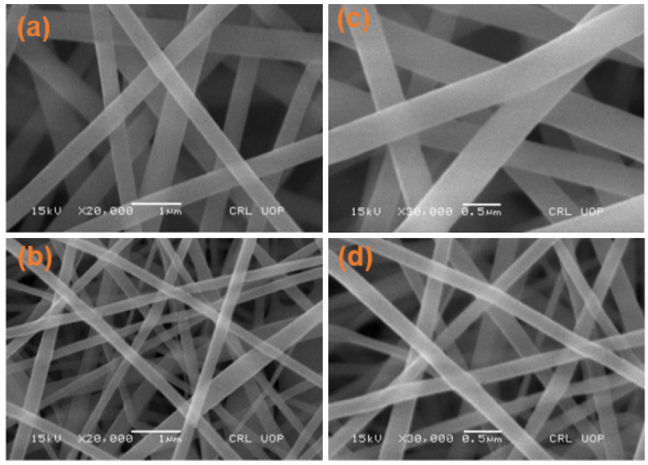

Figure 3. SEM images showing the effect of carbonization on nanofibers at different resolutions for: (a,c) composite nanofibers with diameters of 500 (a) and 630 (c) at 1 and 0.5 μm resolutions, respectively; (b,d) carbonized nanofibers with diameters of 230 (b) and 290 (d) nm at 1 and 0.5 μm resolutions, respectively.

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The SEM images of the nanofibers are taken at different resolutions of 500 nm, as shown in Figure 3a, and 1000 nm, as shown in Figure 3c. The diameters of nanofibers were almost uniform and aligned. No cracks, beads, or branches in the fiber are observed. However, the joint is due to the alignment of one fiber on another owing to a fixed collector. The diameters of the composite nanofibers reduced from 500 to 230 nm (a,b) and from 630 to 290 nm (c,d). The shrinkage in diameters of fibers is due to the decomposition of PVA and TEOS after the carbonization of composite nanofibers.[44]

2.3.2. Effects of PVA Concentrations on the Morphology of Nanofibers. The SEM images of the carbonized nanofibers are shown in Figure 4a–d. Upon varying the amount of PVA in the solution, the diameter of the nanofibers varies significantly. It can be seen from the SEM images that the CNFs are distributed uniformly throughout the materials and there is no agglomeration. The diameter of CNF was found to be 400 nm with the addition of 2 g of PVA, which further decreased to 350, 240, and 200 nm at 1.8, 1.6, and 1.4 g, respectively.

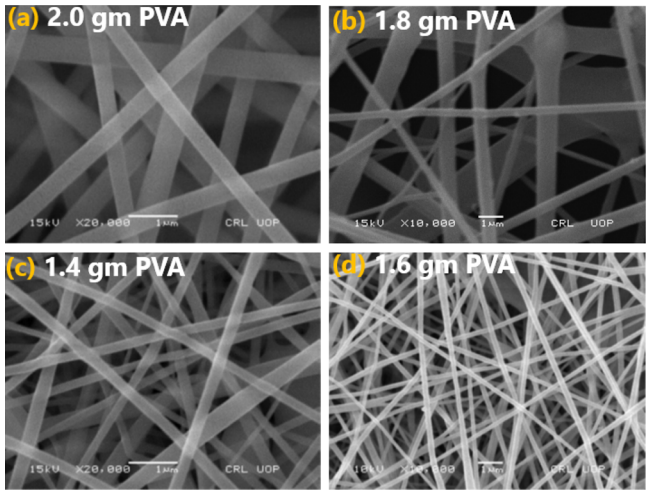

Figure 4. SEM images showing effect of addition of different amounts of PVA on nanofiber diameter: (a) with 2 g, the diameter is 400 nm, (b) with 1.8, the diameter is 350 nm, (c) with 1.6 g, the diameter is 240 nm, and (d) with 1.4 g, the diameter is 200 nm.

2.4. Carbon Nanofiber Filter Performance Tests. *2.4.1. SEM and EDX Analyses of the Filter before Filtration of Water.* Figure 5 shows the SEM and energy-dispersive X-ray (EDX) analyses results of the CNF-based filter. The EDX results show the presence of elements; C, O, Al, and Si. Due to the carbonization of fibers, the quantity of carbon has increased. The presence of oxygen is due to the surface oxidation nanofibers in air during handling. The Al is due to the use of a collector in electrospinning.

2.4.2. SEM and EDX Analyses of the Filter after Filtration of Water. Figure 6 shows the SEM and EDX images of the CNF filter after filtration of water, in which the contaminated particles are clearly shown. The CNF filter trapped the contaminated particles and purified the water. The overall morphology of fibers is the same as before; however, now the contaminated particles appeared on the fibers, as shown in Figure 6. The presence of oxygen is due to the surface oxidation of nanofibers in air during handling. Furthermore, the results showed the presence of some other elements, that is, Mg, Na, Ti, S, Si, and Fe. These elements are due to the contaminated water which appeared on the filter after the process of filtration. This was also confirmed by the SEM image of the filter (Figure 6b). The quantity of Si has also increased due to the silica present in contaminated water. Thus, our post-filtration analysis of CNFs clearly revealed the removal of contaminations to purify the water.

3. EXPERIMENTAL SECTION

3.1. Materials. The synthesis of carbon nanofibers involves two major steps: first is the preparation of the solution and second is electrospinning. In solution preparation, green tea (GT, locally available), PVA (M_w = 66,000, Junsei), and TEOS (98% purity, Sigma-Aldrich) were used. PVA and GT were used as precursor materials. TEOS was added for the flexibility and to increase the specific surface area of the CNFs. In solution preparation, GT (150 g) and distilled water (20 mL) were mixed. The mixed solution was then heated at 100 °C for 20 min. The GT solution was filtered through the silicon filter and the filtered solution was heated at 100 °C for 30 min again. Then, PVA (1.4 g) was mixed with the GT solution and stirred for 45 min at 500 rpm. When the PVA and GT had been mixed thoroughly, then TEOS (0.6 mL) was mixed with it. The concentration of the prepared solution was 1.589 mol/L . Then, the solution was stirred for 3.0 to 5.0 h to make the desired viscosity of the solution.

The whole process during the solution preparation is shown in the block diagram, as shown in Figure 7a. Electrospinning was used for the synthesis of carbon nanofibers. It is the second most important step for the preparation of CNFs. The basic principle of this technique is electrostatic forces. In this work, we used the electrospinning setup, which operates on 17 kV. An 18 mm gauge needle was used. The distance between the collector and the needle was 10 in. The composite nanofibers dried overnight at 100 °C in an oven (Thomas Scientific Model, 605) and then collected from aluminum foil. These dried nanofibers were kept in a tube furnace (Nabertherm Model, LHT 04/18) at 280 °C for 5 h with a 3 °C/min rise in temperature within the air atmosphere. For carbonization at 700 °C for 2 h in a nitrogen atmosphere with a heating rate of 3 °C/min, the organic contents were evaporated and PVA was pyrolyzed to carbon, and through this CNFs were obtained.[23,45] After synthesis of CNFs they are used as a filter for the purification of dusty water. The wastewater was collected from the local river in which many houses and industries' wastewater was mixed. In the purification procedure, we take a strip of CNFs and put it on a beaker. Wee attached CNFs to the beaker with the help of tape so that the CNF strip does not fall into the beaker with water. In this way, we smoothly filtered the wastewater through the CNF strip. The CNF strip, that is, the filter was analyzed, through SEM and EDX before and after the filtration of wastewater in order to clarify the purification function of CNFs. In Figure 7b, the whole procedure is shown.

3.2. Characterization Techniques. The crystallinity of the composite and carbonized nanofibers was investigated through a powder X-ray diffractometer (Rigaku Miniflex II X-ray diffractometer, Ni-filtered Cu Kα radiation, λ = 1.5406 Å). For morphological and roughness studies of the synthesized nanofibers, the sample was prepared in the powder form. Subsequently, the samples were coated with a platinum thin layer via a sputtering technique to make the surface conductive and to avoid any possible charging effect while performing SEM analysis. The coated samples were then loaded into the SEM measurement chamber under high vacuum and were

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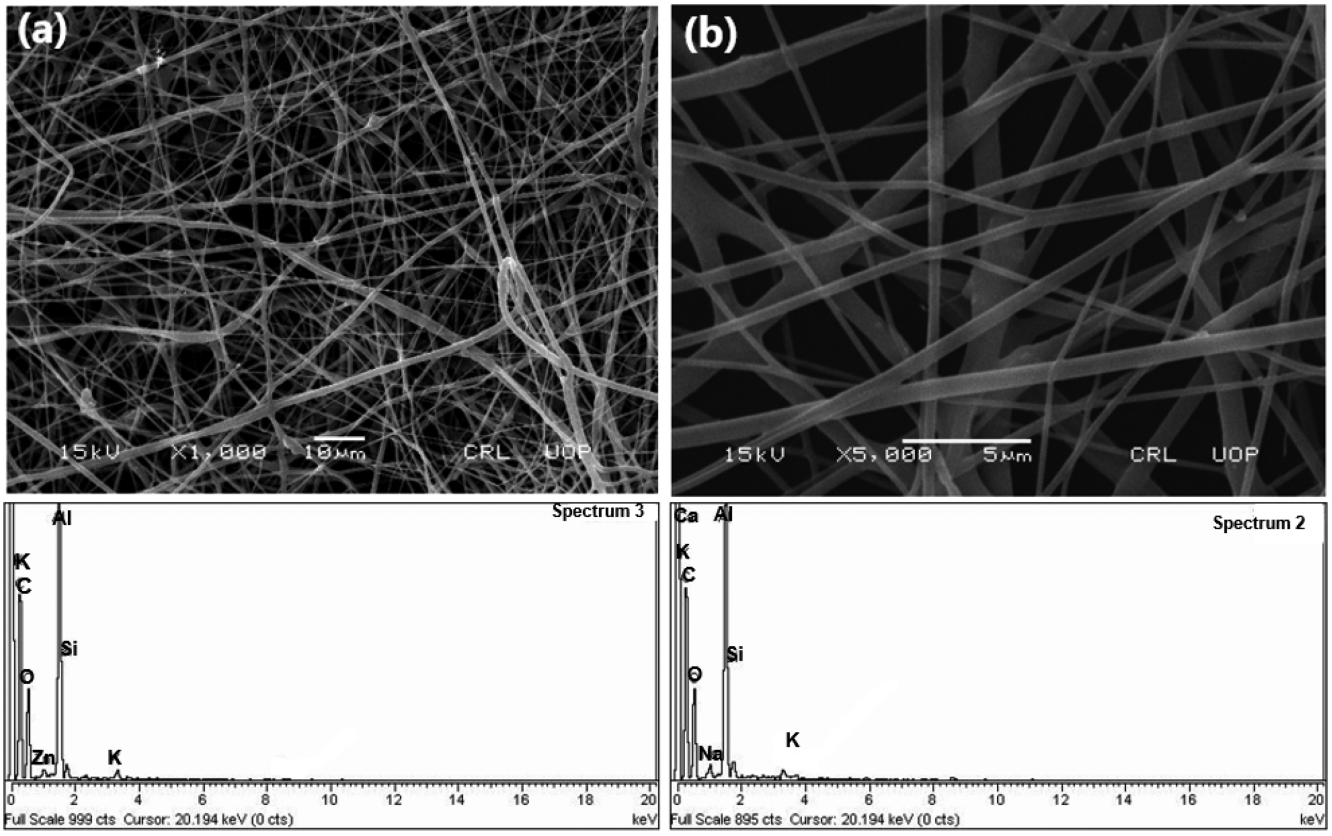

Figure 5. SEM and EDX results of the CNF filter before filtration: (a) at 10 μm resolution and (b) at 5 μm resolution.

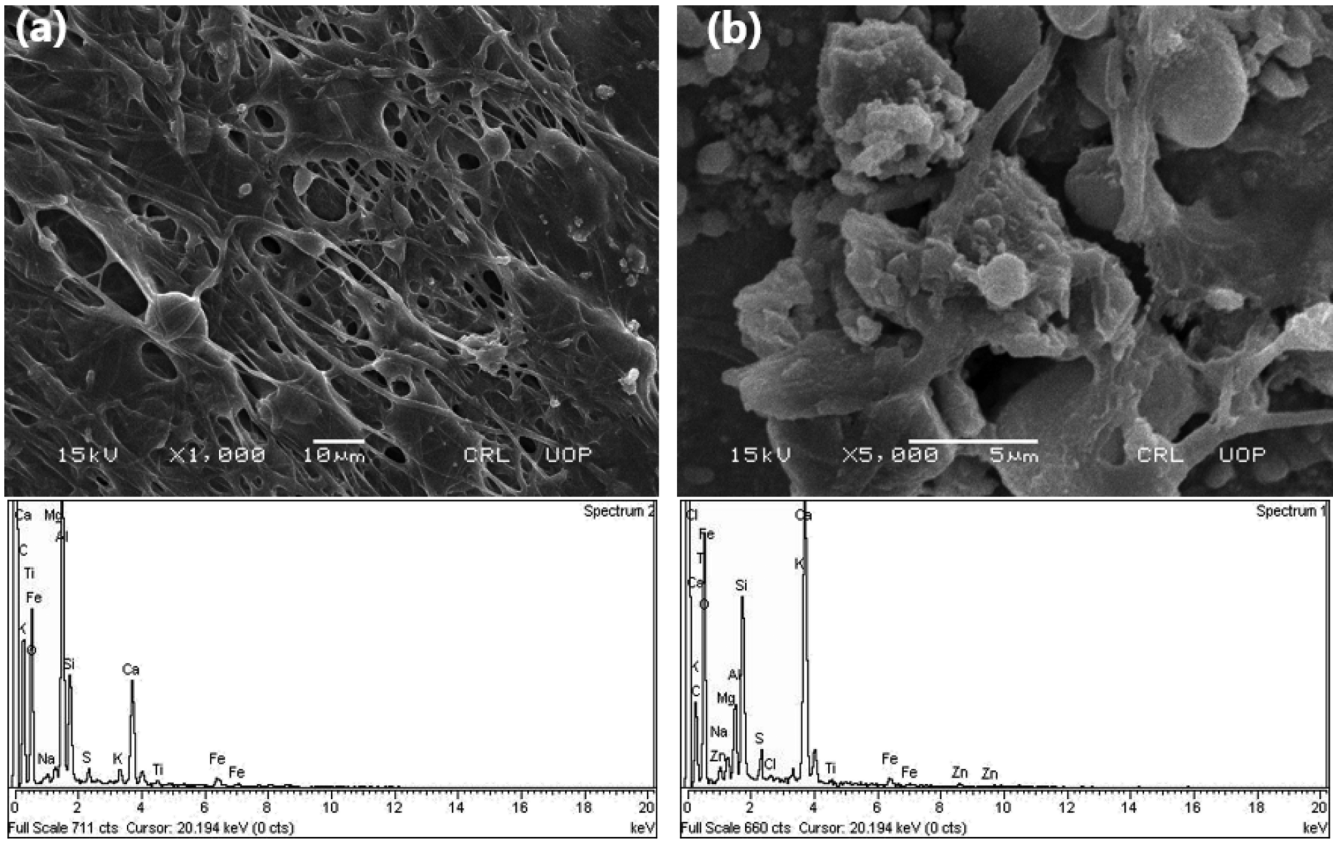

Figure 6. SEM and EDX results of the CNF filter after filtration: (a) at 10 μm resolution and (b) at 5 μm resolution.

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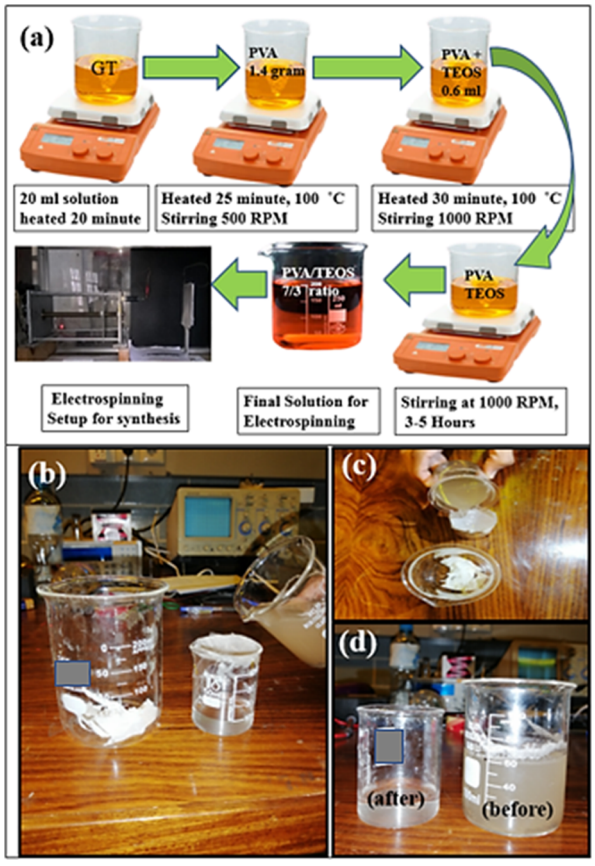

Figure 7. (a) Schematic of the solution preparation and electrospinning setup. (b) Water purification process, (c) top view of the purification process, and (d) water before and after filtration.

examined at a high voltage of about 3−10 kV. The morphology was examined using a scanning electron microscope (JSM-5910 JEOL Japan). IR transmission spectra were collected in the range of 400−4000 cm^{-1} using a PerkinElmer Spectrum Two FTIR spectrometer, equipped with a universal attenuated total reflection accessory. An EDX electron spectrometer (INCA 200, Oxford Instruments, UK) was used for elemental analysis and their compositions.

4. CONCLUSIONS

CNFs were successfully synthesized via the electrospinning technique for water purification and filtration. The synthesized CNFs were characterized via XRD, FTIR, SEM, and EDS analyses. The XRD results showed that the CNF structure is amorphous in nature; however, a small crystallinity was observed after carbonization. The SEM analysis confirmed that the diameter of CNFs was in the range of 500 nm, which further decreased to almost 50% thereby resulting in the reduction of the pore size and making it more suitable for the filtration process. The SEM analysis revealed that different concentrations of PVA and variable parameters of the electrospinning setup affect the morphology of nanofibers. Finally, the CNF filter was successfully synthesized. The SEM and EDX analyses showed excellent filtration results, clearly indicating the removal of contamination and purification of water.

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Notes

The authors declare no competing financial interest.

Data Availability Statement: The authors confirm that the data supporting the findings of this study are available within the article [and/or] its Supporting Information.

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