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Potential application of carbon nanotubes in wastewater treatment compared with conventional methods

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ABSTRACT

Over the last decade, water consumption has grown rapidly due to the increase in population growth, industry, and agriculture. The scarcity of clean water has become one of the biggest problems due to its large demand. Recently, carbon nanotube has exhibited remarkable potentials in water treatment due to their excellent adsorption capacity. In this study, carbon nanotubes (CNT) sponge was investigated for the treatment of wastewater in comparison with the conventional method (physical, chemical, and biological processes). The purification of wastewater was performed by the vacuum filtration technique. The experimental results have shown that the carbon nanotube sponge was very effective and efficient in eliminating harmful microorganisms with superior removal efficiency (100%). The analysis results revealed that the quality of the wastewater treated by carbon nanotube sponge was within the recommended Kuwait Environment Public Authority standards limits in irrigation for pH, total suspended solids (TSS), total dissolved solids (TDS), nitrate–nitrogen and phosphate, and microorganism for influent samples. Further treatment with CNT sponge revealed an additional reduction in the water quality for TSS, biological oxygen demand, TDS, conductivity, nitrate–nitrogen, nitrite–nitrogen, ammonia–N, sulfate, phosphate, and microorganisms for effluent samples. The merits of incorporating carbon nanotube with or without the conventional water-treatment material are highlighted and the challenges are discussed. The positive results confirmed that carbon nanotube has the potential to be a leading technology in water treatment for microorganism removal.

Keywords: Application; Carbon nanotubes; Filtration; Microorganisms; Separation; Sponge; Wastewater treatment; Water analysis

1. Introduction

All forms of living things depend heavily on the safe consumption of water. The lack of clean and affordable water causes around 2 million deaths a year, either directly through dehydration or indirectly through serious illnesses. Worldwide, in 41 countries, one-fifth in populated areas, such as Africa and other under-developed countries, people drink water from an unregulated source of water [1]. All water sources are gradually becoming polluted [2,3]. The main cause of pollution is widely attributed to human activities which have a degrading impact on the environment [4]. Water pollution is on the rise in urban areas due to population growth. On the other hand, agricultural, domestic, and industrial waste is a major source of pollutants affecting people, habitats, and the environment [5]. In addition, sewage is the main pollutant source of freshwater when it is pumped back into freshwater sources. Also, sewage is generated from residences, institutions, hospitals, commercials, and industrials enterprises, as well as household waste liquids from toilets, baths, showers, kitchens, and sinks [6]. The various types of water pollutants can be categorized

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into physical (i.e., color, temperature, suspended solids, sediments), chemical (organic and inorganic), biological (all pathogenic organisms), and radioactive pollutants [7].

Such contaminants affect living creatures and their environment. The pollutants can cause many different diseases such as respiratory diseases, cancer, diarrhea, cardiovascular diseases, and neurological disorder. In fact, water pollution demonstrated risks to marine life, human health, and crop production. The lack of water resources and, at the same time, the increase in the demand for fresh and clean water for human usage and for environmental usage such as agricultural purposes, lead to the development of alternative ways of treating contaminated water to meet the needs of humans and others without causing any harm [8]. It should be noted that various traditional and advanced water treatment technologies such as coagulation and flocculation, membrane filtration, microfiltration, ultrafiltration, adsorption, reverse osmosis, ozonation, and many more have been adopted for wastewater treatment [9]. Conventional wastewater treatment plants approach requires various physical, chemical, and biological processes in which the outcomes could be constrained due to high operating costs, poor treatment efficiency, companies are focusing on new technologies that are more advanced than existing ones, that is, nanotechnology. Nanotechnology has become one of the most innovative technologies in the world. Nanomaterials exhibited additional features upon fabrication and can be utilized in catalysis, adsorption, sensing and, optoelectronics [10]. This technology provides opportunities to develop next-generation water supply systems [11]. These nanometer particles were also produced which could greatly improve the water filtration and desalination processes. The nanomaterials have a high surface area, making treatment of the contaminants much less time-consuming and more effective than the conventional method treatment. In addition, nanotechnology has the potential to develop the environment through the direct application of nanomaterials to track and eliminate contaminants [12]. After numerous studies, carbon nanotube (CNT) has been found to be incredibly attractive due to their fascinating properties such as specific fibrous structure, astounding tensile strength, excellent thermal stability, low density, electrical conductivity and, in particular, super-elasticity [13]. They are one of the most effective nanomaterials for water treatment. Due to their nano-sized properties, they have shown super efficiency [14]. CNTs have special electrical characteristics, such as structural, chemical and physical, which make them a good water treatment component [5]. CNT showed the high performance to absorb microorganisms due to its fibrous structure and high external surface accessibility. It shows a high potential to adsorb a wide range of contaminants through different physical and chemical surface-active groups. It also has a high hydrophobic property which allows water to flow faster with less friction. CNT displays selectivity to certain pollutants based on the conditions used. CNT adsorption technology has the potential for the removal of bacterial pathogens, natural organic matter, and cyanobacterial toxins from water systems [15–18].

Adsorption is one of CNT's key mechanisms in water purification. This process includes the removal of the sorbate when a sorbate is attracted by the sorbent surface. Most of the organic pollutants are adsorbed by CNTs at the surface and inner sites. The Van Der Waals forces between the tubes also make interstitial channels (hollow channels between the tubes) that also make a positive contribution to the process of adsorption [17]. There are many forms of adsorptive activity taking place at these potentially CNT sites, where the non-covalent bond is created an intermolecular force between the CNT and contaminants [18]. CNT cavity has a major effect on capturing contaminants larger than internal pores [14]. It is noteworthy that there are some factors that may affect the adsorption capability of the CNTs. The first factor is the CNTs surface area, where the smaller CNT nano-size powder has more surface area which produces a higher adsorption rate [19]. The smallest size for the nano-diameter enhances the elimination of the microorganism contaminants such as *E. coli*, the bacteria that were trapped in CNT due to the size of its capturing and exclusion feature [17].

The uniqueness of CNT has initiated the appearance of numerous applications for water treatment. However, lots of challenges still need further research [20]. One of the main challenges is the regeneration of CNTs after use. Another challenge is the separation of CNT particles that could be released into the environment with the water and thereby disturbing the environment [21]. Therefore, a lot of efforts were focused on making fixed and immobilized CNTs to stop them from being released into water [22]. The immobilization of CNTs could be solved by the incorporation of CNTs into membranes and filters [23]. Another application is the manufacture of CNT sponges, which are aggregated nano-sized CNT powders. Such sponges maintain CNT's great functions in water treatments [20].

CNT sponges have recently attracted considerable attention in numerous fields. CNT sponges are extremely lightweight nanomaterials and can be distorted into any shape and compressed recurrently in liquid or air without collapsing. CNT sponges are way lighter than water because of their low density, and because the sponges naturally repel water, saturated sponges can be collected for recycling. CNT sponges display structural flexibility that was rarely observed in other high-porosity materials (e.g., aerogels) or aligned CNT arrays. CNT sponges can be used as sensors, electrodes, compressible, flexible conductors, and can soak up water contaminants [2].

The CNT-coated sponge has kept the mechanical characteristics of the uncoated sponge. It has preserved its flexibility and could be bent arbitrarily to any degree with stretchable and compressible structure. These excellent characteristics, for example, large surface area, high porosity (>98%), thermal stability, and great sorption capacity make CNT-sponges a promising candidate as an adsorbent for the pre-concentration of environmental contaminants [24,25]. CNT sponge electrode appears superior compared to textiles, for microbial colonization and catalytic decoration [26]. This study aims to investigate the feasibility of using CNT sponges as an adsorbent for the enrichment and analysis of organic pollutants in water samples.

In the present work, Kabd WWTP was chosen for investigation. Kabd WWTP is a domestic WP, its 35 km away from Kuwait City with 800 m × 800 m fence area and 180,000 m^3/d loading capacities. The conventional treatment process in Kabd includes Conventional treatment in Kabd WWTP include pretreatment (coarse screening, grit chamber), primary treatment (primary clarifier), secondary treatment (bacterial decomposition), and tertiary treatment (extra filtration and disinfection). All samples treated by conventional and CNT methods were analyzed before and after the treatment.

In this study, the adsorption behavior towards CNTs of different contaminants was studied on two samples. The first sample was the raw sewage (influent), and the second sample was the conventional treated tertiary water (effluent). Both samples were taken from Kabd Wastewater Treatment Plant (WWTP) in Kuwait (its initial and final products) by passing it through the CNT sponge type. To accelerate this process, a vacuum filtration system was used to drive the influent and effluent water into the sponge in a less time-consuming and more efficient manner. Moreover, a comparison was made between the findings of both influent and tertiary effluent before and after CNTs sponge filtration through laboratory tests. The reduction percentage was calculated to provide a clear outlook of the efficiency of contaminant elimination between conventional and nanotechnology (CNTs sponge) methods. The quality of CNT-treated water was checked for its suitability for irrigation using the recommended standards of the Kuwait Environment Public Authority (KEPA).

2. Materials and methods

All chemicals were obtained from Mushrif Trading and Contracting Company in Kuwait. CNT (CAS-No.: 1333-86-4) sponge was bought from Advance Chemical Supplier (ACS) Material Advance Chemicals Supplier. CNT tube sponge was purchased from ACS Material Advanced Chemicals, LLC959 E Walnut Street #100, Pasadena, CA 91106.

2.1. CNT specification

CNT sponges are lightweight, elastic, chemically stable, and highly porous. Shape: black block, outer diameter: 30–50 nm, inner diameter 10–20 nm, porosity ratio: 99%, density: 10 mg/cm^3, L × W × H: 10 cm × 5 cm × 0.4 cm. The sponge structure is uniform, good mechanical strength, and good flexibility with low density. In addition, they float on water, have a stable structure, and have a high surface area that is hydrophobic. Potential applications include environmental clean-up. CNT sponges show great promise for use in environmental issues such as water purification and oil spill clean-up.

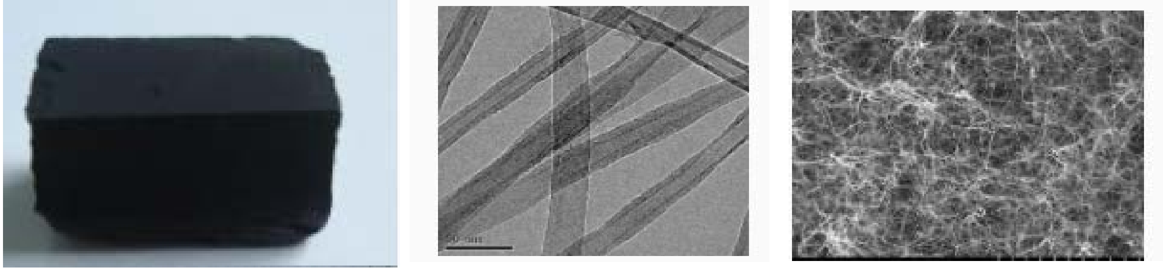
2.2. CNT sponge in water purification

One of the challenges that face the utility of CNT in water treatment is the challenge of retaining the CNT material and reuse of it. The solution to this problem was to use a fibrous, freestanding aggregate of CNT instead of direct suspension. The CNT sponge with high mechanical strength and toughness and the adsorption feature of CNT material has shown great ability in purifying the contaminated water [27]. Fig. 1 shows the inner configuration of the CNT sponge that has been utilized.

2.3. Experimental setup and procedure

In a period of three months (March, April, and May), six triplicate samples of 4 L each were collected from Kabd WWTP for investigation. Samples were collected in a labeled plastic container and transferred to the laboratory following the standard method [28]. Samples were preserved immediately at the time of sample collection. All samples preserved were checked using pH strips, to ensure that they were adequately preserved. The water samples were analyzed before and after the filtration for both the conventional and the CNT sponge.

The six triplicates samples were, three from the raw sewage sample after sand and gravel filter (influent). The other three samples were taken from the effluent sample released after the tertiary treatment by ultraviolet radiation. Quantitative analysis was investigated for both influent and effluent before and after passing it through the CNT sponge and analyzed. All experiments were performed at 25°C. The analysis was repeated for each of the three triplicate samples two times and average values were presented with standard deviation for the accuracy of the results (Table 1). The quality of the water was tested in Kabd WWTP laboratories. The water analysis procedure was following the standard methods for the examination of water and wastewater [29].



(a) (b) (c)

Fig. 1. Morphology of the ACS material CNT sponge, (a) ultralight porous carbon nanotube sponges, (b) transmission electron microscopy image, and (c) scanning electron microscopy image.

Fig. 2 shows the vacuum filtration setup that was utilized in the treatment. 4 L of raw sewage (influent) water was poured into the funnel containing CNT sponge (with the aid of vacuum filtration). The pressure difference in the system will eventually suck the water for the removal and capturing of contaminants. Thus, outflowing filtrate represents influent after CNT. The same procedure was repeated for the tertiary water (effluent) from a conventional method to give the effluent after CNT. The configuration of the system is shown in Fig. 2.

The difference between the influent water before and after passing it through the CNT sponge is shown in Fig. 3. Figs. 3a and b show the filter paper for total suspended solids (TSS) analysis for influent and effluent before and after the CNT treatment respectively.

2.4. Characterization of raw and treated wastewater

pH and conductivity were measured using HACH (model HQ11D) meter and HACH (HQ14D) meter respectively. Glass microfiber filter (model 934-AH, 47 mm diameter) was used for TSS and total dissolved solids (TDS) measurements and the samples were heated using Fisher Scientific Isotemp Oven (model 655G). The vacuum pump for the filtration process was Vacuubrand (model MZ 2 C NT).

Biological oxygen demand (BOD_5) was tested by 300 mL incubation glass bottle having a ground glass stopper and a flared mouth, water was added to the flared mouth of special BOD_5 bottles, then a plastic cup was used over the flared mouth of bottle to reduce evaporation of the water seal during incubation. The samples were placed in an incubator (Lab-Line Instruments) at 20°C ± 1°C for 5 d then the dissolved oxygen (DO) was determined using HACH DO Meter (model HQ30D Flexi) before and after the incubation period.

HACH (model DR3900) spectrophotometer was used for measuring nitrate–nitrogen, nitrite–nitrogen, ammonia–nitrogen, phosphate and sulfate. The reagent used for ammonia was mineral stabilizer, alcohol dispersing agent, and Nessler Reagent. The reagents used for nitrate–nitrogen and nitrite–nitrogen were NitraVer 5 and NitriVer 3 respectively. Reagents used for sulphate, and phosphate were SulfaVer 4 and amino acid reagent (Cat No.193432), molybdate reagent (Cat No. 223632), respectively. All the reagents were permachem reagents (PK/100).

Fig. 2. Vacuum filtration of raw sewage using CNT sponge as filtering media.

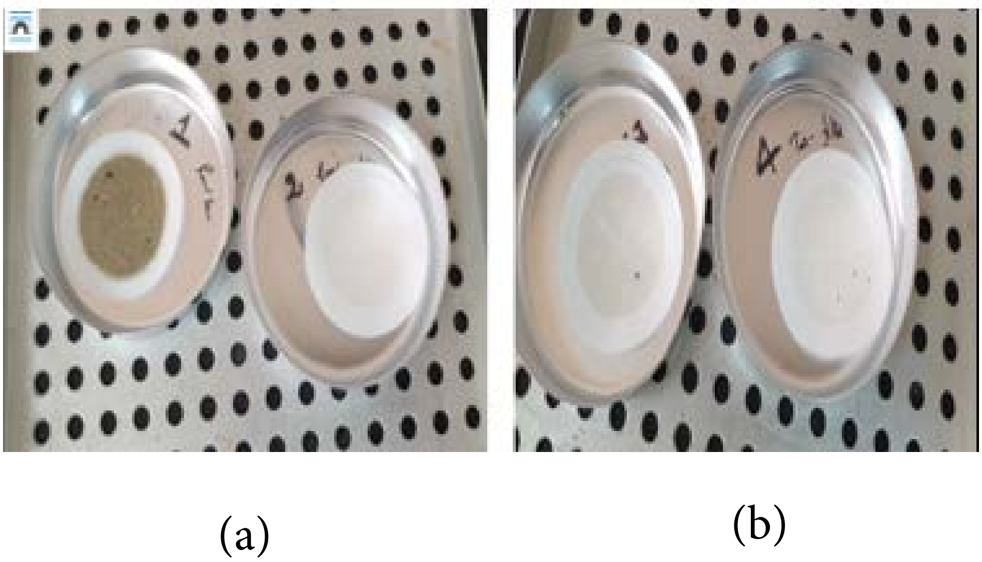


Fig. 3. Dried filter paper of influent and effluent after filtration without and with CNT sponge (a) raw sewage and (b) effluent.

Chemical oxygen demand (COD) was measured by HACH DR3900 Spectrophotometer using LCK 514 (100–2,000 mg/L) and LCK 314 (15–150 mg/L) reagent. All reagents performed in the HACH spectrophotometer were from HACH Lange (Gmbh). Lovibond (model ET125) COD reactor was used for heating the sample. Samples were heated for 120 min.

In the microbiological analysis, an appropriate volume (100 mL) of an influent and effluent water sample from Kabd WWTP was filtered through a 47 mm, 0.45 μm pore size cellulose ester membrane filter that retains the bacteria present in the sample. The filter is placed on a 5 mL plate of Salmonella–Shigella agar using HiMedia (M108-500 G) agar was used for culturing Salmonella and Shigella where plates were incubated at 36.9°C ± 0.1°C. Similarly, HiMedia Endo Agar M029-500 G was used for testing the presence of fecal coliform at 36.9°C ± 0.1°C while HiMedia Endo Agar, M1122-500 G was used to culture fecal coliform at 44.5°C ± 0.1°C. All samples were then placed in the bicategorical (Raypa) incubator. Dilution was performed for the influent sample 60 times for total coliform, 50 times for fecal coliform, and 40 times for Salmonella.

3. Results and discussions

Municipal wastewater (after sand and gravel) with a high concentration of contaminants (influent) and conventional treated tertiary water (effluent) have been treated using a CNT sponge to determine its efficiency for the removal of pollutants from wastewater. Table 1 demonstrates the chemical and biological analysis of influent, CNT treated influent, conventionally treated effluent, CNT treated effluent. All findings were compared with both conventional methods and CNT treatment. Both methods were checked for their suitability for irrigation using KEPA irrigation standard limits [30].

3.1. Comparison of water quality for influent and effluent, using CNT and conventional method

Table 1 shows the average results with the standard deviation of water quality for pH, TSS, COD, BOD_5, TDS, conductivity, DO, nitrate–N, nitrite–N, ammonia–N, sulfate, phosphate and microbiological tests (total coliform, fecal coliform, and Salmonella).

The results in Fig. 4a showed that the pH was 7.49 for the influent sample and was increased to 7.85 after filtration with a CNT sponge. Whereas, the conventional method revealed 7.48 for the effluent. Further treatment of the effluent sample by CNT raised further the pH to 7.56. This finding can reflect on the pH stability of the CNT sponge toward water pollutants. The pH is of major importance in determining the corrosivity of water. In general, the lower the pH, the higher the level of corrosion [31].

In Figs. 4b–d the results indicated that the TSS, COD, BOD_5 for the influent sample were 164, 528 and 344 mg/L respectively. The results were reduced to 15 mg/L 90.85%), 500 mg/L (5.30%), 330 mg/L (4.07%) after CNT sponge treatment.

Superior removal efficiency for influent was indicated for TSS using a CNT sponge. The slight reduction was shown in COD and BOD_5 concentration, this indicated that further treatment is required to oxidize the organic matter present. Recently, the capability of the CNTs-sponge to degrade the

Table 1
Chemical and biological analysis of raw, and CNT-treated and conventionally-treated wastewater

Test	Influent	Influent after CNT treatment	Effluent by conventional method	Effluent after CNT treatment	KEPA standard limits
pH	7.49 ± 0.5	7.85 ± 0.6	7.48 ± 0.5	7.56 ± 0.55	6.5–8.5
TSS, mg/L	164 ± 4.97	15 ± 1.1	12 ± 0.86	11 ± 0.83	15
COD, mg/L	528 ± 12.6	500 ± 11.8	31 ± 2.3	138	100
BOD_5, mg/L	344 ± 8.2	330 ± 7.8	14.31 ± 0.95	14.31 ± 0.95	20
TDS, mg/L	860 ± 19.8	855 ± 19.1	850 ± 18.6	750 ± 16.3	1,500
Conductivity, μS/cm	1,550 ± 34.4	1,540 ± 32.3	1,542 ± 32.5	1,520 ± 29.1	NA
Dissolved oxygen, mg/L	8.61 ± 0.77	8.65 ± 0.8	8.18 ± 0.76	8.48 ± 0.78	>2
Nitrate–N, mg/L	32.2 ± 2.4	4.3 ± 0.48	1.3 ± 0.21	1.1 ± 0.20	15
Nitrite–N, mg/L	0.017 ± 0.1	0.017 ± 0.1	0.017 ± 0.1	0.017 ± 0.1	NA
Ammonia–N, mg/L	45 ± 0.65	27.2 ± 2.2	40 ± 2.9	24.4 ± 1.77	15
Sulfate, mg/L	202 ± 3.9	108 ± 1.85	202 ± 3.9	108 ± 1.85	NA
Phosphate, mg/L	20.1 ± 1.5	17.3 ± 1.4	16.2 ± 1.38	15 ± 1.23	30
Total coliform (MPN/100), CFU/mL	$7.7 \times 10^9 \pm 30.4$	0	$9.8 \times 10^1 \pm 4.6$	0	400
Fecal coliform (MPN/100), CFU/mL	$5.5 \times 10^8 \pm 20.4$	0	$7.7 \times 10^1 \pm 4.1$	0	20
Salmonella (MPN/100), CFU/mL	$6.8 \times 10^7 \pm 15.4$	0	$1.1 \times 10^1 \pm 3.6$	0	NA

NA: not available

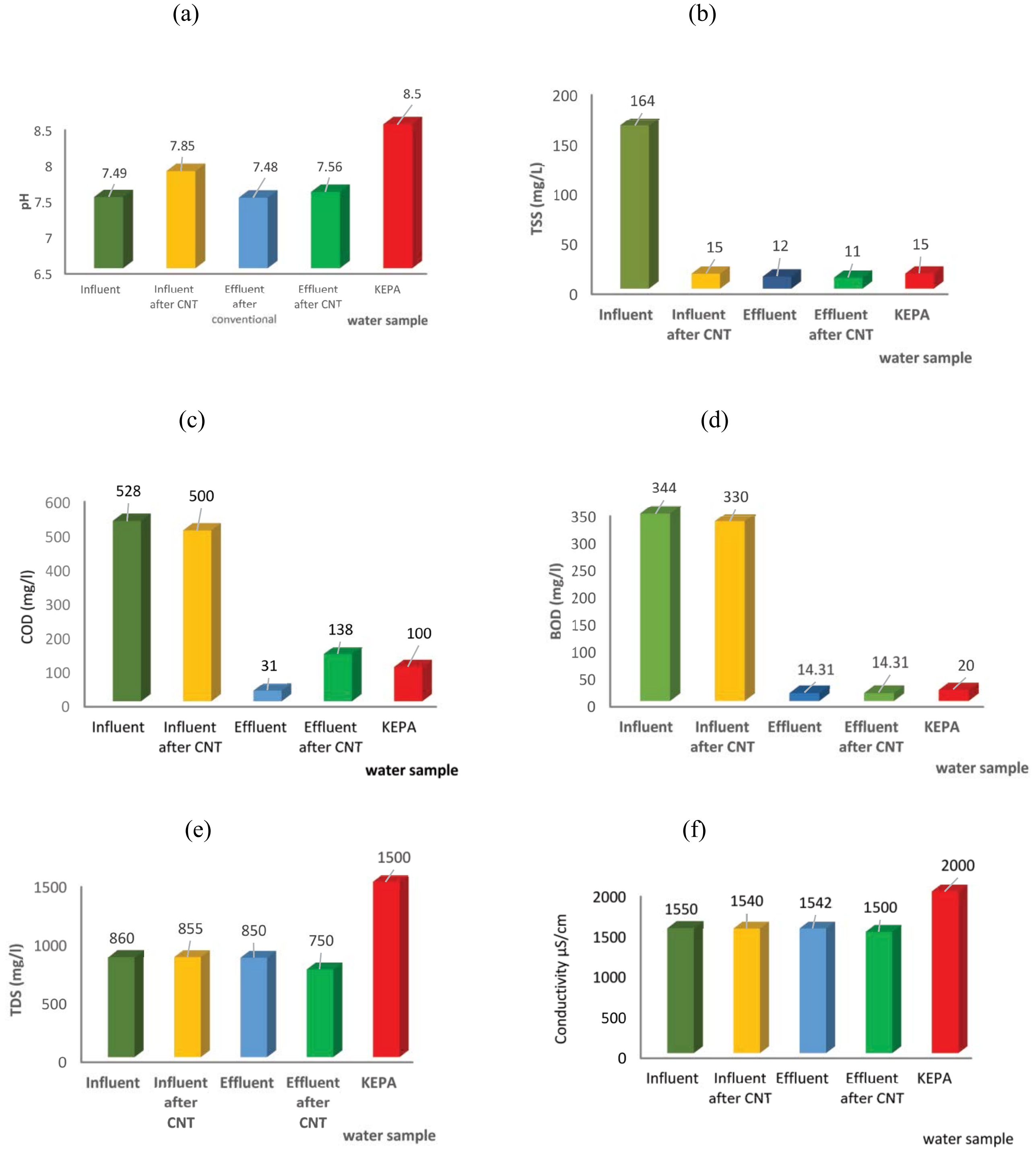


Fig. 4. Analysis for influent, effluent by CNT and conventional treatment: (a) pH, (b) TSS, (c) COD, (d) BOD_5, (e) TDS, and (f) conductivity.

COD concentration (600–800 mg/L) to 28%–52% using a different load of CNTs load (0.1 to 0.3 mg/mL) was demonstrated, further enhancement of the removal efficiency of COD in the electro-bioreactor zones revealed 85%–95% which was due to the role of the electro-coagulation and CNT-sponge adsorption during stage [24].

However, the effluent by conventional method was reduced to 12 mg/L (92.7%), 31 mg/L (94.1%), 14.31 mg/L (95.8%) for TSS, COD, BOD_5, respectively. The results indicated that the conventional method showed better performance than the CNT treatment for the influent sample.

Further treatment of the effluent with CNT treatment revealed that the TSS reduced to 11 mg/L, COD value was increased to 138 mg/L, and no reduction for BOD_5. The results indicated that CNT was inefficient in reducing COD and BOD_5 values. This could be due to a reduction in sponge efficiency due to the blockage of CNTs' active sites by adsorbed by-products resulted from organic oxidation [24] or could be, that some of the sponge particles pass to the filtrate. Additional study could be performed to minimize COD and BOD_5 levels by the addition of a high-pressure ozone and oxygen stream [32].

TDS (Fig. 4e), conductivity (Fig. 4f) for the influent sample was 860 mg/L and 1,550 µS/cm. The results were reduced slightly with CNT treatment to 855 mg/L (0.58%) and 1540 µS/cm (0.65%) respectively.

Quite similar results were shown by the conventional method that slightly reduced to 850 mg/L (1.16%) and 1,542 µS/cm (0.52%). The TDS of effluent treated by CNT showed a slight further reduction to 750 mg/L (11.76%). However, the conductivity was reduced on small scale to 1520 mg/L (1.43%). The analysis indicated that the CNT sponge is inefficient in removing a high concentration of salts, previous results were obtained using the CNT adsorption column [31]. Both CNT and conventional treatment method TDS result falls below KEPA standard limits for irrigation as shown in Table 1.

DO reading as shown in Fig. 5a for the influent sample was increased from 8.61 to 8.65 mg/L using CNT, while for the effluent conventional method was increased to 8.18 mg/L. Whereas the DO of the treated effluent by CNT did not change as shown in Table 1 limits for irrigation.

The concentration of DO is an important parameter for the characterization of natural and wastewater and the general assessment of the global state of the environment. DO is needed for the good quality of the water. The analysis shows that the CNT treatment was quite similar to the conventional treatment for influent and effluent samples. The results for both methods were within the recommended levels of KEPA standards limits.

Nitrate–nitrogen (Fig. 5b) concentration for influent sample was at 32.2 mg/L. The value of nitrate–nitrogen was reduced to 4.3 mg/L (86.6%) by the CNT treatment, whereas, the reduction for nitrate–nitrogen by the conventional method was higher and reduced to 1.3 mg/L (95.9%). However, the value was slightly reduced to 1.1 mg/L (15.38%) for the nitrate–nitrogen of the treated effluent by CNT. Similar reduction efficiency to 80% for NO_3 was demonstrated recently using CNT sponge [24].

The readings indicate the capability of CNT in treating the water with a high concentration of nitrate–nitrogen. Nitrate is vital for growth and reproduction, but excessive nitrogen levels delay the ripening of some crops and prolong the vegetation stage, making the crop more vulnerable to pests and diseases and lower yields. Wastewater that contains a high level of nitrate–nitrogen considers health concern and it is important to know the nitrate content of irrigation waters before their use [33].

As illustrated in Fig. 5c, the results of nitrite–nitrogen in the influent and effluent sample were 0.017 mg/L and did not change for both CNT and conventional treatment methods. Furthermore, no reduction of nitrite–nitrogen in the effluent sample by CNT treatment was observed.

Ammonia–nitrogen for the influent sample (Fig. 5d) indicated that CNT has decreased the concentration of ammonia–nitrogen in the influent from 45 to 27.2 mg/L (39.5%). Whereas, the conventional method was slightly reduced to 40 mg/L (11.11%). This indicated a higher reduction with the CNT method. The ammonia–nitrogen of treated effluent by CNT showed a further reduction to 24.4 mg/L (39%). CNT reveals great potential in lowering ammonia levels than conventional treatment. Recent studies showed that the sponge-CNT was capable of eliminating nitrogen by 81%. Other studies indicated the increased biological phosphorus removal, as well as excess phosphorus uptake, can be achieved by the accumulation of phosphate organisms on the sponge layer. Furthermore, microorganisms attached to the sponge remove a portion of the phosphorus biologically, as P is an essential nutrient for biomass growth [34].

Sulfate concentration (Fig. 5e) and phosphate (Fig. 5f) for influent sample was 202 and 20.1 mg/L respectively. The concentration was decreased to 108 mg/L (46.5%) and 17.3 mg/L (13.9%) respectively by CNT treatment. Recently, phosphorous was reduced using a CNT sponge submerged membrane bioreactor to >90.6% [24]. Phosphate concentration was within KEPA standard limits for irrigation (30 mg/L).

No reduction percentage was observed for the conventional method for sulfate value. The conventional effluent showed a slightly higher reduction to 16.2 mg/L (19.4%) for phosphate value. Further treatment of the effluent by CNT showed that the sulfate and phosphate reduced further to 108 mg/L (46.5%) and 15 mg/L (7.41%) respectively.

The results revealed that CNT was more efficient in minimizing the sulfate and phosphate concentration than the conventional method. The results for both methods for influent and effluent were within the recommended levels of KEPA standard limits for irrigation. The high porosity, and high flexibility, make CNT-sponges a favorable candidate as an adsorbent for the pre-concentration of environmental contaminants.

3.2. Effect of CNT treatment on microorganisms in comparison with conventional method

Pathogenic microorganisms enter the water bodies either through sewage as a major source or through the wastewater from industries like slaughterhouses. Viruses and bacteria can cause waterborne diseases, such as cholera, typhoid, dysentery, polio, and infectious hepatitis in humans [35].

Table 1 shows the results for the effect of the CNT sponge on total coliform, fecal coliform, and Salmonella.

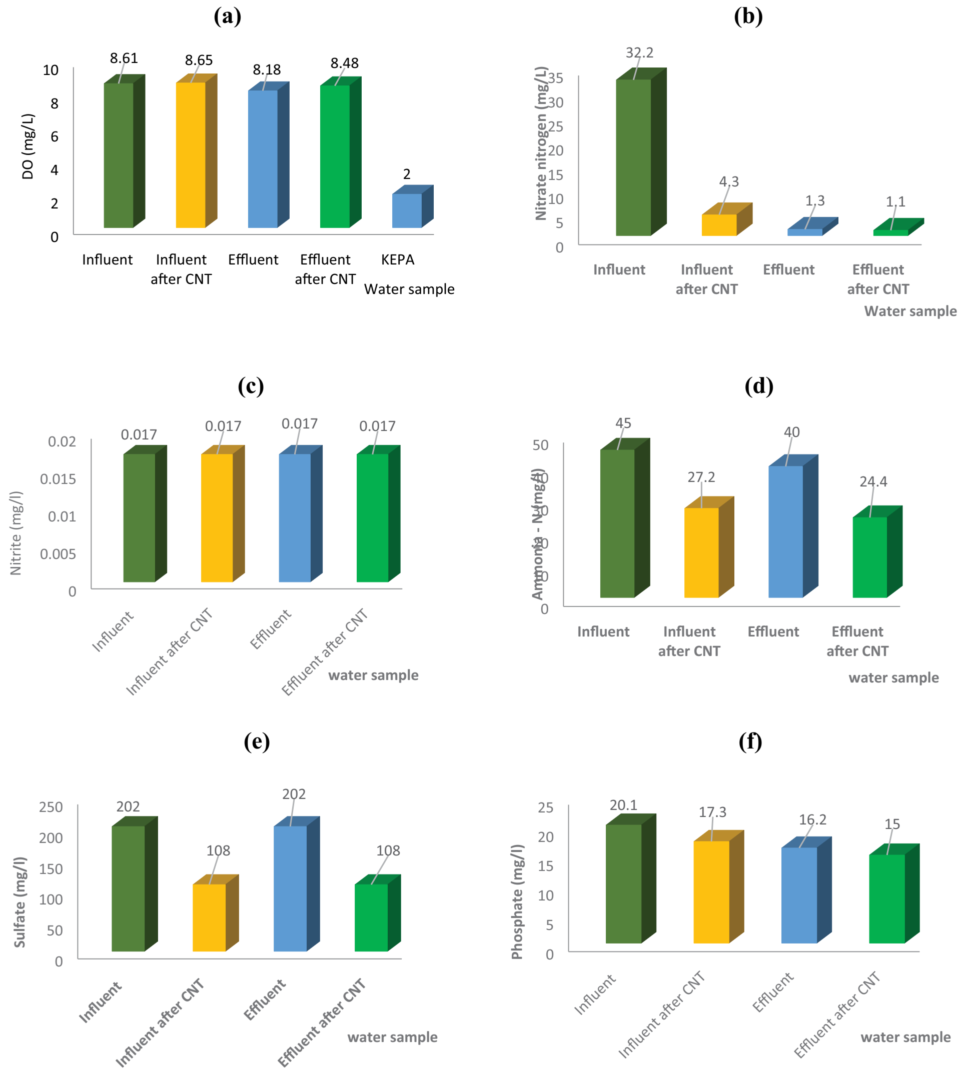


Fig. 5. Analysis for influent, effluent by CNT and conventional treatment: (a) DO, (b) nitrate–nitrogen, (c) nitrite–nitrogen, (d) ammonia–nitrogen, (e) sulfate, and (f) phosphate.

The results for influents for total coliform, fecal coliform, and Salmonella were 7.7×10^9, 5.5×10^8, and 6.8×10^7 colony-forming unit (CFU)/mL, respectively. After the treatment with the CNT sponge, all the microorganism (total coliform, fecal coliform, and Salmonella) for the influent by the CNT method was reduced superiorly to 0 CFU/mL (100%). Quite similar results were shown by the conventional treatment that diminished total coliform, fecal coliform, and Salmonella to 9.8×10^1, 7.7×10^1, and 1.1×10^1 CFU/mL respectively, with a reduction of 99.99%. Fecal coliform is up to 400 most probable number (MPN) /100 mL and 20 MPN/100 mL respectively in water [36]. The tests results indicated that CNT has remarkable effect on the removal of microbiological contaminants in comparison with chemical contaminants and with the conventional method. The test results indicated that CNT has a remarkable effect on the removal of microbiological contaminants in comparison with chemical contaminants and with the conventional method. The superior removal efficiency of the bacteria was due to the high porosity and large surface area of the CNT sponge and due to the strong interaction between CNT and the microorganisms present in the wastewater.

This generated a great interest in CNT sponges in their use as sorbent materials for microorganisms. Similarly, previous studies using CNTs showed superior adsorption capacities in the removal of a diverse range of biological contaminants including bacteria, viruses, and cyanobacterial toxins from water systems [17]. Similarly, complete removal of bacteria by multiwalled carbon nanotube through filtration was published [37,38]. Another comparative study indicated that the sponge prevents the transport of the bacteria with high-efficiency removal (96.6%) was achieved using a carbon sponge with a large pore size [34].

Comparing the conventional method and CNT treatment, it has been clearly shown that CNT treatment could meet the most demanding standards of water quality compared to the conventional treatment methods for bacterial removal.

3.3. Comparison of influent and effluent treatment by CNT with KEPA standards limits

To further address the possibility of using CNT sponge material in wastewater treatment, the results of the CNT have been compared to KEPA standards limits published for irrigation.

Table 1 results showed that the pH values of the treated influent according to the standards were in the range of the recommended KEPA standard limits for irrigation. The TSS of both influent filtrate and effluent were acceptable and with the KEPA limits after CNT sponge treatment. The COD values of the influent samples were not in the desirable range, which means further treatment methods should be used. Water quality analysis using CNT sponge for TSS (influent and effluent) and BOD_5 (effluent) were within KEPA standard limits for irrigation. In contrast, COD (influent and effluent) and BOD_5 (influent) were higher. However, for the conventional method, the results fall within the KEPA standard limits. The COD level can be minimized by adding hydrogen peroxide to the water in small concentrations (300–500 mg/L) and subject it to UV treatment which can oxidize the organic and inorganic matter present [39].

The TDS for both influent and effluent were within the range. The concentration of nitrate in the treated influent and effluent in both methods, was less than the KEPA standard limit. The concentration of nitrate in the treated influent and effluent in both methods, was less than the KEPA standard limit.

The results for total coliform and fecal coliform were within KEPA standard guidelines which indicate that the CNT can be used for the disinfection stage in the conventional method. The high adsorption capacity was due to the strong interaction of the CNT sponge surface to the adsorbed microorganism.

3.4. Challenges

One of the biggest challenges in the use of CNTs sponge for water treatment is the fact that the structural material of CNT is highly dependent on the synthesis methods and storage of the material. CNTs sponge was inefficient for COD and ammonia. Further treatment is required to minimize it. The COD level can be minimized by adding hydrogen peroxide to the water in small concentrations (300–500 mg/L) and subject it to UV treatment which can oxidize the organic and inorganic matter present [39]. One of the major challenges is the escape of nanosized CNT material into the filtrate for both influent and effluent. CNTs are hard to handle and eventually get lost or dispersed. This revealed that further treatment is required. The discharge of ammonia from wastewater treatment plants (WWTPs) has become a challenging issue in wastewater treatment. Ammonia can be reduced by nitrification, this can be achieved through biological oxidation of ammonia to nitrite followed by oxidation of the nitrite to nitrate [40]. Ammonia can also be reduced by adding nitrification inhibitors which are applied to soil as fertilizers [41].

4. Conclusion

The application area of CNTs is continuously growing. In this study, the feasibility of CNTs column adsorption in the removal of pollutants in terms of physicochemical parameters was investigated. The parameters such as pH, TSS, TDS, COD, BOD_5, conductivity, DO, nitrate–N, nitrite–N, ammonia, sulfate, phosphate, total coliform, fecal coliform, and Salmonella of all samples were studied before and after the treatment with CNT sponge and conventional method. The present study has conclusively indicated that CNT sponge with porous structure has performed an excellent efficiency in the removal of suspended solids and nitrate–N with a reduction of 90.85%, 86.6% respectively. A slight reduction was observed for sulfate (46.5%) and phosphate (13.9%) using a CNT sponge. Influent and effluent treated with CNT sponge revealed remarkable reduction efficiency for bacterial removal (100%). The reason is due to its nano-size, high surface area, and adsorption capacity, which reflects the strong interaction between CNT particles and the microorganisms present in wastewater. This superior removal efficiency of microorganisms indicated that CNT sponge can be utilized with or in combination with the conventional method in the disinfecting stage instead of using chemicals. The water quality analysis results (influent and effluent) for pH, TSS, TDS, conductivity, and phosphate were within KEPA standard limits for irrigation using a CNT sponge. However, problems have arisen when trying to handle the fine powders and eventually retrieve them from the water. Further studies could improve the porous structure of the CNTs to avoid leakage to the water body after filtration. CNT sponge could be developed by changing the pore size inner structure, morphology by functionalization of the sponge to enhance adsorptive filtration of waterborne contaminants.

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