Combating Water Contamination using Plastic Waste

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ABSTRACT

The disposal of the problematic plastic waste represents a major challenge for all over the world especially in developing countries for waste and environmental management. High valued carbon nanotube (CNT) was synthesized from the noxious plastic grocery bags waste with a simple technique to minimize the disposal problems. Conventional methods for synthesizing carbon nano structured materials involve the complex reaction system design, using toxic chemicals and solvents, high growth temperature, high pressure, poor yield and the production of hazardous wastes. This paper describes an economically viable and environmentally friendly process that converts the LDPE plastic bag waste into the multiwalled carbon nanotube (MWCNT) at 700 °C using a novel self-prepared catalyst in a closed system without applying any extra pressure in the reaction zone and finally the fine sand coated with the prepared CNT’s to create a low cost, rapid, super-efficient, highly recyclable and sustainable solution for the coastal areas salinity problem and industrial synthetic dye’s contaminations. Ultraviolet-Visible Spectroscopy (UV-Vis), Raman spectroscopy, X-Ray Diffraction Analysis (XRD) and Fourier-Transform Infrared Spectroscopy (FT-IR) used in this study to characterized prepared MWCNT structured materials and its filtration capacities.
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KEY WORDS

ABBRERVIATIONS AND ACRONYMS

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<tr>
<th>CNT: Carbon Nano Tube</th>
<th>CSC: CNT Sand Composite</th>
</tr>
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<tr>
<td>MWCNT: Multi Walled Carbon Nano Tube</td>
<td>SWCNT: Single Walled Carbon Nano Tube</td>
</tr>
<tr>
<td>CVD: Chemical Vapor Deposition</td>
<td>PE: Polyethylene</td>
</tr>
<tr>
<td>MB: Methylene Blue</td>
<td>ETP: Effluent Treatment Plant</td>
</tr>
<tr>
<td>GO: Graphene Oxide</td>
<td>RO: Reverse Osmosis</td>
</tr>
<tr>
<td>HDPE: High Density Polyethylene</td>
<td>LDPE: Low Density Polyethylene</td>
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ACKNOWLEDGEMENT
I would like to sincerely thank Bangladesh Council of Scientific and Industrial Research (BCSIR) and Dr. Md. Abdul Gafur from Pilot Plant and Process Development Centre for his guidance and for allowing me to conduct my research work in his lab. I would also like to thank Professor Dr. Tanvir Ahmed of the Department of Civil Engineering at Bangladesh University of Engineering and Technology (BUET) for mentoring me. I would like to thank Professor Dr. Helal Uddin of the Department of Chemistry at Islamic University for helping me to conduct characterization experiment stages. I am also thankful to the Stockholm Junior Water Prize for providing me a platform to represent my research. My parents and my two sisters have always been a constant source of inspiration during my research.

BIOGRAPHY
Didarul Islam is a rising senior at Kushtia Zilla School. He received the Presidential Award at National Children Prize Competition-2017 in Science Project. In the same year, he was named “The Most Talented Student of the Year-Science” in the Creative Talent Hunt Competition-2017 organized by the Ministry of Education, Bangladesh. He was awarded as the “Project of the Congress” in the National Children Science Congress-2017. He represents his research work at the Yokohama National University, Japan in the Sakura Science Exchange Program-2018 organized by the Japan Science and Technology Agency (JST) and Ministry of Education, Bangladesh. He is currently working with two open access journals with his new concept. In his free time, he enjoys giving talks to encourage people all around his community about environmental pollution and water management to increase awareness about the global water crisis. Didarul hopes to continue working in the field of environmental engineering in order to create a novel solution to environmental problems facing around the world.
INTRODUCTION

Every year more than 300 million tons of plastic waste is generated all around the world and only 10% of that waste is recycled [3]. Besides, our country imports more than 150,000 metric tons of plastic bag raw materials each year and only 5% is recycled and other 95% is thrown away our lakes, rivers and seawater surface [4]. This highly toxic plastic waste leads an extreme water, air and soil pollution. In the global perspective about 8 million tons of plastic enter the ocean every year and at least 5.25 trillion pieces of plastic are currently in the ocean [5]. Also, some plastic bags are taken more than 1000 years to decompose in nature. During this process, plastic release toxins and microplastics that impact aquatic species, which impact us. Microplastic pollution directly influences the health of the local human community, wildlife, and aquatic environment. The recent study reported that 92% of the dead seabirds found in the test area had ingested plastic up to 5% of their total body weight and also 80% of microplastics are formed land-based sources like polyethylene bags [5]. Also, the non-biodegradable plastic polymers can cause carcinogenic effect or promote endocrine disruption on human health and also damage our ecosystem and wildlife.

![Cumulative plastic waste generation and disposal](image)

**Source:** Production, use, and fate of all plastics ever made; Roland Geyer, Jenna R. Jambeck, Kara Lavender Law.

More than 2·8 million hectares of area in Bangladesh that is highly affected by the salinity problem [6]. In the coastal areas of our country salinity creates a range of health effects, including increased hypertension rates. Large numbers of pregnant women and children in the coastal areas are being diagnosed with pre-eclampsia, eclampsia, and hypertension due to the increased salinity of drinking water [6]. On the other hand, the rapid growing textile and leather industries synthetic dye contaminated wastewater considerably affecting the aquatic environment, rivers, and groundwater surface. In addition to aquatic environmental damage, these dyes can cause several carcinogenic effects on human health. In our community, the bio sand filters are widely used to remove contamination from the water. But, the bio sand filters are less effective to remove industrial synthetic dye or salinity from water and also produce hazardous sludge due to its non-recyclable ability. So, the aim of this research is to revealed the most environmental-friendly solution to transformed the negative valued plastic waste into the high valued CNT coated sand composite in a cheaper way and also making “the pollution into the solution” by ensuring sustainable plastic waste management and saving our planet from the industrial synthetic dye contamination and salinity of the water. Reverse osmosis, membrane technology, electrochemical technique, effluent treatment plant (ETP), ion-exchange and adsorption are used for treating industrial dye and saline contaminated water which is highly expensive for the people who lived under the poverty line. Industrial grey water is polluting the most of the rivers of our country every day. Besides, most of the people in our country are collecting the river water for drinking and other using purpose.
Without any pretreatment, they used that contaminated water on a daily basis. The slow sand filter is widely used as a pre-treatment medium for the desalination plant. But the slow sand filters are less effective for treating high saline water and cannot completely remove turbidity of the contaminated water. CNT sand composite is highly efficient for reducing salinity of the water which can revolutionized the water desalination plant pretreatment process by reducing cost and saving energy. The novelty of this present work is preparing a high valued carbon nanostructured material from the trash plastic waste as a low-cost feedstock, in spite of chemical feedstock like benzene, ethylene or toluene to minimizing the disposal problems in a cost-effective method.

Carbon is a versatile adsorbent that is heavily used in the removal of various pollutants from aqueous solutions. Carbon nanotubes are allotropes of carbon with a barrel-shaped nanostructure. The carbon nanotube is a rolled-up graphene sheet including single-walled CNT and multi-walled CNT. Carbon nanotubes are used as an effective adsorbent of both heavy metals and natural synthetic compounds due to their strong adsorption affinity. Carbon nanotubes, inferable from their tunable physical, chemical, electrical and structural properties, move inventive advancements to address the water shortage and water contamination issues. CNT is undoubtedly emerging as the most promising nanomaterial because of its unique combination of superb properties and chemical stability compared with other nanostructured material in the water purification. Chemical vapor deposition (CVD), pyrolysis, laser ablation, arc discharge, electrochemical exfoliation, mechanical exfoliation, hydrothermal treatment, templating techniques, heating polymer spheres in an inert atmosphere, reduction of glucose and pressure carbonization are a few convenient methods are employed to synthesized carbon nanostructured materials [7]. Complex reaction system design, high temperature, poor yield and hazardous waste are the main burdens for the large production of carbon nanostructured materials. Solid thermoplastic polymers, such as; polyethylene (PE), polypropylene (PP), polystyrene (PS), polyvinyl alcohol (PVA), polyvinyl chloride (PVC), polytetrafluoroethylene (PTFE) and polyethylene terephthalate (PET) have been studied as a low-cost feedstock for carbon-based nanomaterials synthesis [8]. Due to the higher carbon content and less impurities in the LDPE plastic bags, it can be recycled through greener routes. Also, selecting an appropriate catalyst is an important parameter for the controllable synthesis of nanomaterials. Transition metals and their metallocene have been examined for the production of carbon nanostructured materials including ferrocene, ferrous chloride, cobaltocene or nickel oxides. Fe-Al catalyst is used in this study as a catalyst where Fe is the core catalyst and Al are both co-catalyst and catalyst support. Fe-Al catalyst catalyzes the decomposition of the polyethylene bags during the heat treatment. Alumina particle is the catalyst carrier in this decomposition process. Due to the low cost, easy availability and higher yield; Fe-Al catalyst is an appropriate catalyst for the large-scale and controllable synthesis of CNT-based nanomaterials. Besides, cheaper and locally available river sand is the best medium for the production of CNT coated “super sand composite.” Plastic grocery bags can act as the carbon source where CNT binds on the river sand without any binding agent.

**PRIMARY QUESTION**

Is it possible to create a low-cost, eco-friendly, reusable and super-efficient CNT sand composite made out of waste plastic bags that can remove contaminated synthetic textile and leather industries dye from wastewater and also can take part for reducing the salinity of the drinking water?

**HYPOTHESIS**

A low-cost, recyclable and non-toxic CNT sand composite filtration system made out of real-world waste plastic grocery bags. CNT sand filtration capacity was successfully characterized in both laboratory and field scale experiment. And it can work as a highly effective composite in the industrial wastewater treatment and ensuring the public health security by reducing salinity in the drinking water.
MATERIALS AND METHODS

MATERIALS

All the chemical reagents including Methylene Blue, NaCl, HCl, Acetone, NaOH, Fe (NO3)3-9H2O, Al2O3, River Sand, Ferrocene, Whatman Filter Paper etc. was purchased from the “Silver Scientific and Chemical Company” and “Sigma Aldrich”. The plastic bags sample was collected from the local grocery shop. The entire chemical reagent was purchased under the guidance of the mentor. All solutions were prepared with the ultra-pure deionized water under ambient conditions.

METHODS

Synthesis of Catalyst: The novel Fe-Al catalyst was prepared by adding the Fe (NO3)3-9H2O powder in the deionized water solution and mixing it with the commercial Al2O3 powder in a measuring beaker. Then the solution was magnetically stirred for 30 min to get a homogeneous mixture. After that, the solution was dried at 120 °C in an oven for 10 hours and finally, the dark brown powder was separated by a sieve to get a fine particle sized powder.

Synthesis of CNT: CNT were synthesized by the decomposition of LDPE waste in a closed crucible. In a closed system, 3.545 gm of LDPE waste was added with the 0.512 gm of Fe-Al catalyst and sealed the crucible. The crucible was placed inside an electric furnace and the temperature of the furnace was set at 700 °C for 3 hours. After that, the system was left for few hours to cool down automatically. Finally, the dark powder was crushed with a mortar to get a fine particle sized powder.

CNT Sand Composite Preparation: 10 gm of river sand washed with 50 ml of 1 M HCl (35% purity) solution to remove other impurities from the sand and washed with deionized water few times to remove excess acid residuals. After that, the sand was dried at the room temperature for 2 hours. Then 5ml of prepared CNT solution was added with the sand particle and mixed homogeneously to make the CNT sand composite. Finally, the composite was dried in an oven at 110 °C for 2 hours.

Fig-1: a) LDPE plastic bag added in 3.545 gm portions, b) Fe-Al catalyst added in 0.512 gm portions, c) Heat treatment and d) CSC preparation.
RESULTS AND DISCUSSIONS

Different temperature, catalyst and reaction time are important parameter for the production of carbon nanotube from the plastic waste. The yield of the prepared material was calculated by the equation below–

\[
\text{Carbon Yield (\%)} = \left( \frac{\text{weight of product}}{\text{weight of raw materials}} \right) \times 100 \quad \text{.................} \quad (1) \quad [10]
\]

The burn-off (BO) of carbon nanostructured materials was calculated as the following equation–

\[
\text{Burn-Off (\%)} = \left( \frac{w_1 - w_2}{w_1} \right) \times 100 \quad \text{.....................................................} \quad (2) \quad [10]
\]

Where \(w_1\) is the weight of raw materials and \(w_2\) is the weight of final product.

The prepared Fe-Al catalyst is producing better yield of CNT than commercial Ferrocene catalyst. The experimental results are showing below–

<table>
<thead>
<tr>
<th>Temperature (°C)</th>
<th>Catalyst</th>
<th>Reaction Duration (hour)</th>
<th>Yield (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>600</td>
<td>Fe-Al</td>
<td>1</td>
<td>66</td>
</tr>
<tr>
<td>600</td>
<td>Ferrocene</td>
<td>3</td>
<td>35</td>
</tr>
<tr>
<td>650</td>
<td>Ferrocene</td>
<td>1</td>
<td>18</td>
</tr>
<tr>
<td>650</td>
<td>Fe-Al</td>
<td>3</td>
<td>63</td>
</tr>
<tr>
<td>700</td>
<td>Ferrocene</td>
<td>3</td>
<td>65</td>
</tr>
<tr>
<td>700</td>
<td>Fe-Al</td>
<td>3</td>
<td>93</td>
</tr>
<tr>
<td>750</td>
<td>No Catalyst</td>
<td>6</td>
<td>21</td>
</tr>
<tr>
<td>750</td>
<td>Fe-Al</td>
<td>2</td>
<td>85</td>
</tr>
<tr>
<td>800</td>
<td>Ferrocene</td>
<td>3</td>
<td>40</td>
</tr>
<tr>
<td>800</td>
<td>No Catalyst</td>
<td>1</td>
<td>55</td>
</tr>
</tbody>
</table>

Characterization of Prepared Carbon Nano Structed Materials

The prepared CNT powder sample was characterized using Raman spectroscopy, X-Ray Diffraction Analysis (XRD), UV Visible Spectroscopy (UV-Vis), and Fourier-Transform Infrared Spectroscopy (FT-IR) to understand the structure of the prepared materials.
Raman Spectroscopy

Raman spectroscopy is widely used to analyze the graphene structure and it gives exact information about quality, how many layers, defects, and extent of disorder. The Raman spectrum of CNT is characterized by two main features including the G mode and the D mode. Where the G mode usually observed at 1596 cm−1 and the D mode observed at 1360 cm−1 [12]. In the fig-3 it reflects both D band and G band is present in the prepared CNT structure. D band (1336.8 cm−1) is typical for a hard carbon which is representing a highly disordered (D) graphite arrangement and imperfect structure of amorphous carbons. And G band (1583.9 cm−1) characteristic of a more ordered graphitic (G) structure.

![Raman Spectra of Prepared CNT.](image)

UV-Visible Spectroscopy

The UV-Vis Spectroscopy experiment was conducted to place the CNT powder cuvette inside the sample compartment of the UV-Vis spectrophotometer. After that, set the spectrophotometer in the wavelength scan mode with a wavelength range between 1100 nm to 190 nm. Then the spectrophotometer started scanning. Repeat the number of scans for the better result. The absorption peak around at 262.7 nm in the UV–vis spectrum corresponds to the basic confirmation of the CNT-based nanomaterials.
Fig-4: UV-Vis spectra of prepared Carbon Nano-Materials.

X-Ray Diffraction Analysis (XRD)

Fig-5: XRD Analysis of Prepared CNT Powder.
The X-ray diffraction (XRD) is the most widely used technique for characterizing carbon-based nanomaterials crystallinity. It calculates the average spacing’s between layers or rows of atoms and measure the orientation of a single crystal or grain. The XRD pattern obtained for as-synthesized LDPE plastic waste at 700 °C for 3 hours is shown in the Fig-5. The CNT structure is reflected by the broad X-ray diffraction peaks centered at approximately 26.35 and 44.93 2θ which correspond to the (002) and (101) reflections which are typical for highly amorphous graphite-like carbons. The broad (002) peak corresponding to the highly organized layer structured pure graphite. The presence of the (101) peak suggests a stacking order of graphene sheets [13].

**Fourier-Transform Infrared Spectroscopy (FT-IR)**

![FT-IR Analysis of Prepared CNT Powder](image)

Fig-6: FT-IR analysis of prepared CNT powder.

FT-IR is widely applied to get an infrared spectrum of absorption, emission, and photoconductivity of a solid, liquid or gas. Also, it can be utilized to quantitative analysis of an unknown material. FTIR measurement was employed in this study to characterize the prepared carbon nanostructured material. In the Fig-6, the prepared CNT has three characteristic peaks. FT-IR absorption peak around 1600 cm\(^{-1}\) corresponding to the in-plane C=C vibration, which is one of the basic characteristics of sp2 graphitic materials [16,17]. Besides, IR peak at 3445.39 cm\(^{-1}\) and 1102.92 cm\(^{-1}\) is attributed to the O–H and alkoxy C–O, bond respectively [12,18,19,20].
FILTRATION EXPERIMENTS

Preparation of Methylene Blue Stock Solution

50 ml of commercial MB dye solution was taken in 10 ml proportions using a pipette and added to a 500 ml measuring cylinder. The cylinder was then filled with deionized water up to 500 ml. Then the dye solution was magnetically stirring for 30 minutes to get a homogenous MB dye solution.

Field Scale Study of the Saline Contaminated Sample

The water sample collected from three different districts including Dhaka, Khulna, and Kushtia. The salinity of the individual areas sample was calculated using Automatic Temperature Compensation (ATC) Refractometer. Total Dissolved Solids of the contaminated water is calculated by using high-quality HM Digital TDS Meter.

Stock solution of Methylene was passing through 1cm, 5cm, 7cm and 10cm fixed bed column to calculate the amount of MB dye removal efficiency achieved and compared with bio sand filtration. The effects of regeneration cycle, column depth and pH of the dye removal process were investigated. Desired temperature was controlled by using a temperature-controlled water bath shaker. All pH measurements were carried using a digital pH meter and the pH of the contaminated solution were adjusted using HCl or NaOH solution. The percentage of pollutant removal was calculated using the following equation:

\[
\text{Removal Efficiency (\%) } = \frac{(C_0 - C_t)}{C_0} \times 100 \quad \text{................................................................. (3)}
\]

\(C_0\) = initial concentration of MB solution, \\
\(C_t\) = concentration of MB solution after time t.

Fig-2: a) Sand and CNT Sand Composite, b) Filtration setup, c) Before and after passing the MB stock solution in the filtration column and d) Water sample collected from the three different districts.
Filtration of Stock Solution

The prepared stock solution was passing through the 1, 3, 5, 7 and 10 cm depth fixed bed CSC column and in the case of 10 cm, UV-Vis Spectra were used to calculate the amount of the dye removed after passing the column by using the removal efficiency equation-3.

![Absorbance vs Wavelength](image)

Fig-7: UV-Vis absorbance of contaminated solution and after passing the CSC column. By calculating the removal efficiency equation, it can be concluded that, 10cm depth fixed bed of CSC column has a higher efficiency of 91.6 % for MB contaminated solution.

The CSC efficiency percentage in different bed depth (cm) and pH level were calculated by the removal efficiency equation and the UV-Vis experiment. In the Fig- 8, it reflects the CSC efficiency level in the different pH condition. In the acidic condition the efficiency of the CSC is lower but in the basic condition, CSC shows higher pollutant removal efficiency including synthetic dye solution. Besides, the pollutant removal efficiency was increased if the size of the bed depth increased. 1, 3, 5, 7 and 10 cm fixed bed was set up by CSC filter to calculate its removal efficiency in the different filter column size. The result shows that; 10 cm CSC filter medium is perfect for removing the wide range of pollutant from contaminated water.
Fig-8: CSC fixed bed removal efficiency at different range of pH where the pH of the solution was adjusted by adding HCl or NaOH solution.

Fig-9: Scatter diagram of CSC fixed bed filtration efficiency against columns depth.

**CSC Regeneration**

Regeneration is one of the important parameters for water filtration and sustainable waste management. Besides higher synthetic dye removal efficiency, the CNT Sand Composite regeneration efficiency is also satisfactory. The CSC column bed after exhaustion with synthetic dye pollutant was regenerated using acetone and was reused for five cycles. After five cycles the synthetic dye removal efficiency was decreasing rapidly. The regeneration efficiency was calculated by the previous equation.
Fig-10: Regeneration cycle vs synthetic MB stock solution filtration capacity measurement.

Field Scale Study of the Saline Contaminated Sample

The different water sample was collected from three individual regions and calculating the salinity of the water, TDS level and measured the pH of the water sample to understand the real-world water crisis. Then the CSC was applied as an alternative to bio sand filtration and the filtration capacity is describing below-

<table>
<thead>
<tr>
<th>Before Passing the GSC Filter</th>
<th>Total Dissolved Solids (TDS)</th>
<th>Salinity (%)</th>
<th>pH</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dhaka</td>
<td>312 ppm</td>
<td>0.5</td>
<td>5.5</td>
</tr>
<tr>
<td>Khulna</td>
<td>804 ppm</td>
<td>1.3</td>
<td>6.6</td>
</tr>
<tr>
<td>Kushtia</td>
<td>308 ppm</td>
<td>0.3</td>
<td>6.1</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>After Passing the GSC Filter</th>
<th>Total Dissolved Solids (TDS)</th>
<th>Salinity (%)</th>
<th>pH</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dhaka</td>
<td>206 ppm</td>
<td>0.001</td>
<td>6.1</td>
</tr>
<tr>
<td>Khulna</td>
<td>256 ppm</td>
<td>0.005</td>
<td>6.4</td>
</tr>
<tr>
<td>Kushtia</td>
<td>234 ppm</td>
<td>0.002</td>
<td>6.3</td>
</tr>
</tbody>
</table>
FUTURE WORK

I would like to experiment further with prepared CSC on ultra-filtration and prepared graphene powder in the field of membrane technology. I would also like to experiment about various heavy metal contamination removal from water by prepared CSC. Further research should be conducted on experimenting with SEM, TEM, TGA, EDX and understanding the prepared nano-materials complete structure. In future, I would also like to commercialize my prepared CSC and make it available for the common people with the cheapest budget.

CONCLUSIONS

A person uses a plastic grocery bag on average only for 12 minutes where we only reuse 1 plastic bag in every 200 we produce [26]. In this situation, feasible and sustainable plastic waste recycling is the best option to save our planet from the noxious plastic contaminants. So, it is possible for us to make these trash plastic bag waste by-products into valuable CSC which can save people’s lives and ensure the global water security. In this experiment simple, inexpensive and waste by-product was used to create an alternative option for industrial synthetic dye contamination cleanup technology and costly desalination pretreatment plant. The developed method is safe, simple and inexpensive than conventional chemical vapor deposition (CVD), pyrolysis, hydrothermal treatment and templating techniques to produce carbon-based nanomaterials and also don’t produce any harmful by product or toxic gases compound in the environment during its production. UV-Vis spectroscopy revealed its higher synthetic dye removal capacity which is 91.6% for the contaminated MB solution in a 10 cm fixed bed filtration column. Raman spectroscopy, XRD analysis, Fourier-Transform Infrared Spectroscopy (FT-IR) and UV-Vis spectroscopy used in this study to analyze the graphene structure and its filtration capacities. This project opens numerous possibilities for sustainable, economically viable and effective wastewater purification and also can save our planet from petroleum-based plastic pollution. The advantages of CSC filtration –

1) Simple and Feasible,

2) Cost and Energy Effective,

3) Eco-friendly and Efficient,

4) No Operational Skill Required,

5) Provides a Sustainable Solution to Plastic Waste Management.
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