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Experiment of treating polluted wastewater resulting from petroleum refineries using pyramid solar still distillation system to eliminate hydrocarbon toxicity

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ABSTRACT

The current research contributes to addressing the exploitation of the abundance of solar energy in providing potable water treatment and reviewed a green and inexpensive mechanism for treating wastewater from the petroleum oil refinery industries that is contaminated with organic polluting hydrocarbon compounds that are harmful to the environment. The experiment was conducted in the city of Samawah in Iraq during the month of January for the treatment of petroleum wastewater by using a local pyramidal solar distillation system aided with a filtration basin made of recycled materials at varying weights of charcoal (100–2,000 g), time (1–18 h), pH (6–7), and temperature (28°C–88°C). The results showed a high performance of the passive solar system in treating wastewater from pollutants corresponding to 99.5% biological oxygen demand removal efficiency at optimum conditions of pH (6), 2,000 g of charcoal, 18 h contact time, and internal and external temperatures of 28°C and 48°C, respectively. The statistical test confirmed that the pH and weight of charcoal have a significant effect on the overall performance of the pyramidal solar distillation system at a 95% confidence interval (p-value < 0.05). The key characteristics of the finished waterfalls are within the minimum permissible limits for safe and clear water discharge.

Keywords: Oil wastewater; Solar energy; Pyramidal solar still distillation system; Solar wastewater treatment; Water purification; Desalination

1. Introduction

Since the beginning of the 2000s, many countries and societies began to sense the environmental hazard that human exert on natural resources, including water pollution caused by hydrocarbon exploration and petroleum refining activities [1,2]. The rate of human consumption of petroleum and natural gas for energy and transportation is an indication of the depletion of these natural resources at a rate that exceeds nature's ability to produce. The increasing exploration, production, and refining of crude oil and petroleum processing to satisfy human needs contributes severely to ground and surface water pollution [1,3]. To redress the mentioned problem, it is important to consider sustainable solutions to the treatment of petroleum refining wastewater with low damage to the environment [2,4] along with rationalizing consumption to reduce the negative environmental impact of industrial wastewater on nature. Around 80% of industrial wastewater is thrown into the environment without any treatment [5] at a time

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when 15% of the world population suffers from drinking unsafe water [5,6]. Thus, the impact of polluted water containing high concentrations of organic pollutants extends to all living organisms, apart from humans, and affects the diversity and biological balance on earth [6,7].

The world population is witnessing a steady increase in urbanization, accompanied by an increase in industrialization as a result of a consumer lifestyle [7]. Large quantities of solid and liquid oily wastes, dissolved hydrocarbon produced from petroleum industrial wastewater during extraction, transportation, refining, dumping, and storage operations [8–11]. The continuous and increasing work of the crude oil exploration and petroleum refining industrial sectors generates a large amount of waste, including wastewater, whose rates reached alarming levels, at a time when that wastewater or effluent is disposed of without proper treatment [3,4]. Wastewater from petroleum industries also contains a high level of chemical oxygen demand (COD), and biological oxygen demand (BOD), high concentration of total solids, and oil impurities [1,2], in addition to toxic hydrocarbons and other wastes including heavy metals, high total dissolved solids (TDS), catalyst, sludge, and volatile organic compounds [2,8,12,13]. Wastewater from petroleum industries includes different types of organic and inorganic pollutants, such as BOD, heavy metals, hydrocarbons, phenols, and hydrocarbon sulfides [11,12]. Through the activities of the hydrotreating, desalting, cooling, and distillation operations, large amounts of stubborn organic pollutants and toxic hydrocarbons are released that have the potential to cause serious damage to the environment and human health [13–15]. Asphalt, aromatic and aliphatic petroleum hydrocarbons, and compounds containing oxygen, nitrogen, and sulfur are the four basic types of oily wastewater. They also contain organometallic complexes with lead, nickel, vanadium, and cadmium [15,16–19]. The wastewater is classified, depending on the proportion of solids and water, into sludge, crude oil, or simple wastewater. The pH value of oily sludge varies according to the source of the crude oil, the treatment methods, and the type of reagents used. It usually ranges from 6.5 to 7.5 [15,18], and it can lead to acute and chronic problems for the natural ecosystem, both now and in the future [19–22]. Although refineries and petrochemical industries play an important role in economic growth, the problem of getting rid of polluted industrial effluent is a major challenge for these industries [15,16].

Several methodologies for remediation of petroleum refinery wastewater such as Fenton-oxidation [4], advanced oxidation process [6], coagulation–flocculation [1,2], adsorption techniques [23], gravity techniques, and the use of solar still system [24,25] have been reported in the literature. Most water purification technologies rely mainly on electric power and batteries using complex technical parts such as reverse osmosis (RO), vapor compression (TVC), multi-stage flash desalination (MSF), multi-stage distillation (MED), mechanical vapor compression (MVC), among others [24,25]. The conventional and biological methods of treatment of petroleum wastewater are limited in wide areas of application due to the complex phase, small size of oil droplets, and relatively high viscosity [26,27]. Most conventional methods were associated with secondary pollution [26], the cost of chemicals required for absorbents, and their interaction with substrates in the water [1]. There are also associated challenges of low separation efficiency [26], susceptibility of coagulation–flocculation processes to changes in water chemistry [2], and challenges of fouling and inability of membrane systems to handle emulsified water [27,28].

Recent advancement in the application of solar still, and solar still distillation systems (SDSs) as green and energy-efficient approach for industrial wastewater operations are beginning to gain relevance in water remediation processes [24,25,29,30]. Decisively, it was found that SDSs effectively can remove pollutants [30]. The COD elimination rates for different types of wastewaters, including sludge, textile, kitchen, petroleum, palm oil, municipal wastewater, water, and wastewater plant, were found to be 98.13%, 96.84%, 97.85%, 87.99%, 96.71%, 85.67%, and 86.99%, respectively [24,25,30]. The SDSs treatment achieved a salt reduction rate of 99.99% for seawater [24–25]. However, very limited research work reported on pyramid solar still distillation systems (PSDS) for the remediation of petroleum industry wastewater and effluent [32,33]. Investigation into the application of the pyramid solar still distillation system aided-filtration process for removal of BOD and other stubborn organic pollutants from petroleum refinery effluent is very limited in the literature.

This research examines the results of treating polluted water from oil refineries using the PSDS during the month of December in Iraq in the city of Samawah (31.3188° N, 45.2806° E), where the solar radiation is at a low level to reveal the effectiveness of the solar distillation process during these climatic conditions. In this research, the performance of the PSDS would be examined and optimized by posting methods of the inner heat transmission, the difference of the temperature between the water and the inlet surface of the system cover, and to increase the efficiency of solar radiation absorption-aided filtration system. The findings of a BOD test were used to predict the degradation and removal of organic compounds outcomes for a particular sample of industrial wastewater. There is an empirical connection between BOD and COD, and can further be employed to ascertain the biodegradability index of the industrial effluent for clean discharge.

2. Materials and methods

2.1. Collection and preservation of water sample

The petroleum refinery wastewater sample was collected from the remnants of the Samawah oil refinery during the month of December in the city of Samawah, located in Iraq. The wastewater sample was placed in a 20 L keg and stored in a refrigerator maintained at 0°C–4°C to prevent microbial and chemical degradation. The examination of the industrial wastewater, sampling, and preservation was carried out following the methodology reported in the literature [31,32], and by procedure recommended by the Al-Furat Al-Awsat Technical University, Department of Mechanical Engineering, and the Technical Institute of Samawah Iraq. The experimental design in this research consisted of 17 experimental runs carried out using a PSDS aided with a filter basin containing layers of fine sand and charcoal for the remediation of petroleum refinery wastewater treatment. The PSDS used in this research was manufactured locally by the researchers. The experimental design was aimed at oil-water separation and removal of organic and toxic hydrocarbons pollutants from the industrial wastewater sample.

2.2.1. Pyramid still distillation system

Fig. 1 shows a simplified illustration of a PSDS. Solar distillation stills are common water treatment solutions. It is one of the most promising utilizing of renewable energy sources (solar energy) [33]. Although the traditional single slope systems are amongst the least expensive and the simplest, the solar still system can refine polluted water with concentrated salt up to 104 ppm [34]. Mainly, its performance relies on climatic circumstances and many other factors which include the angle of inclination of the cover of the container, the depth of the water in the container, direction, vapor concentration, sizes, and materials of the still [24,25]. The process of purifying water by solar distillation is green, similar to what occurs naturally in what is called the water cycle in nature [35], where the sunlight functions to evaporate the water to the surface where clouds formed, and then it condenses in the cold layers of the atmosphere to fall in the form of clean water, then it mixes with surface water and groundwater [36]. The solar distillation device is characterized as a fixed device without moving parts, does not need membranes or filters, does not contain complications, and does not need electricity. It is a wooden, plastic, or metal box with a transparent cover, usually glass. This system works on the principle of global warming to the dirty water inside the box to generate thick steam through a rapid condensation process to form pure water that is safely thrown out to the environment, leaving behind salts, impurities, and pollutants deposited at the bottom of the device. The solar still distillation system enabled the long-wavelength portion of the solar spectrum to be used for the production of high-quality potable clean water. The system is designed to enhance 98% water recovery, with a solar-to-water evaporation efficiency of about 72%, and subsequent removal of dissolved hydrocarbon salts, toxic heavy metals, and removal of organic via integrated photocatalytic and photo-thermal pathway [36–38]. Even at low temperatures, the solar still system can reportedly inhibit the transmission of pathogens through pollutants and steam [39].

2.2.2. Oil-water separation via filtration drum in PSDS

The industrial wastewater treatment system employed in this research work consists of two (2) parts. The first part consisted of the wastewater compartment for filtering impurities associated with dirty water, while the second part consisted of three distinct layers as shown in Fig. 2. The first layer is composed of gravel functions as enhancement to aid the distribution of water flow, and support the finer filter particles to reduce suspended solids present in the oily water [50,51], the second layer is sand whose function is to trap and remove smaller sized oil droplet during oil-water separation, prevent colloidal particles in the water from entering the outlet stream, and contribute to the reduction of beneficial bacteria, which helps in a biological breakdown of organic matter during the treatment process [51]. The third layer contains crushed charcoal with a large surface area that functions as an aid in the adsorption of dissolved salts, toxic hydrocarbons, heavy metals, and volatile organic compounds in industrial wastewater [51].

2.3. Experimental design of the wastewater treatment

The industrial wastewater sample was characterized following the standard procedure to determine the initial properties of the wastewater parameters before treatment. In this research work, the experiment was carried out in two stages. The pictorial representation of the components of the PSDS and the structure of the filter basin used for the treatment system is presented in Fig. 3. In the first stage, the

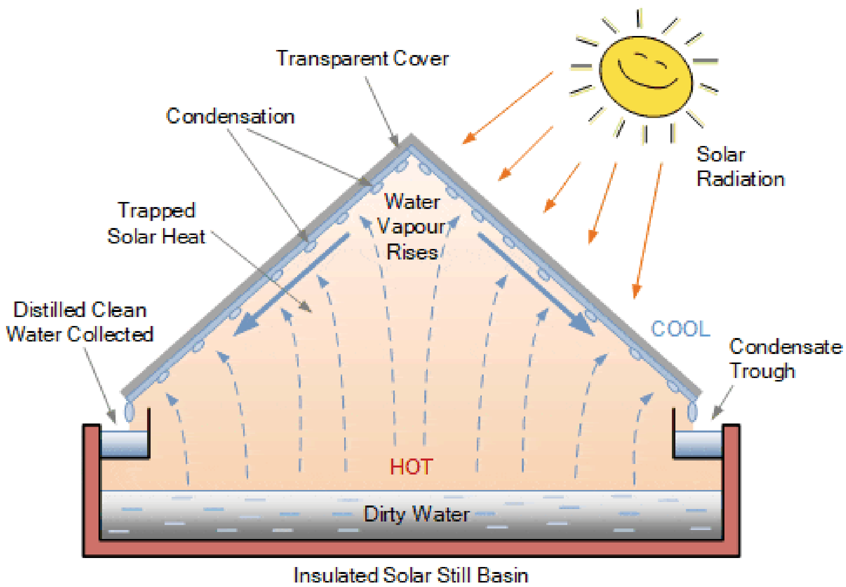

Fig. 1. Basic model of a pyramid solar still distillation system. Source: Alternative energy tutorials. Source online (Alternative Energy Tutorials).

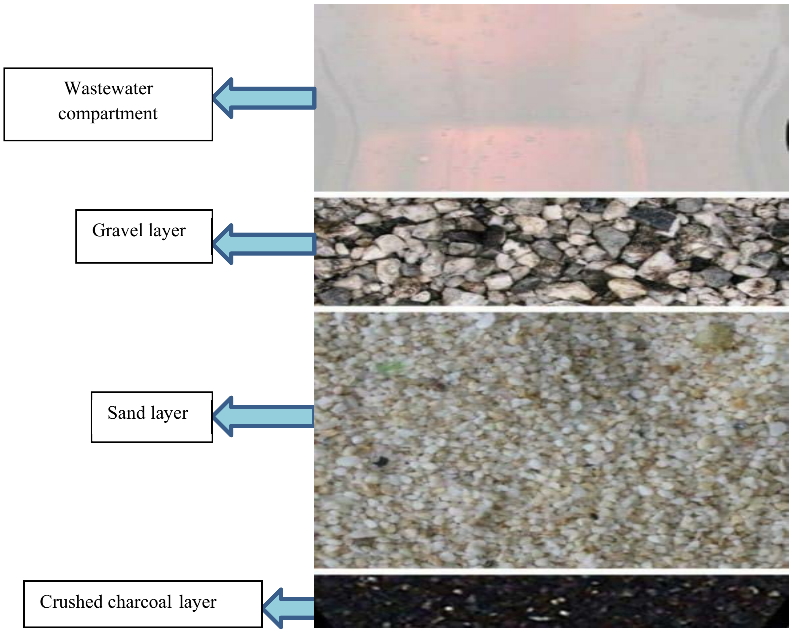

Fig. 2. Schematic representation of the different layers of the filter basin.

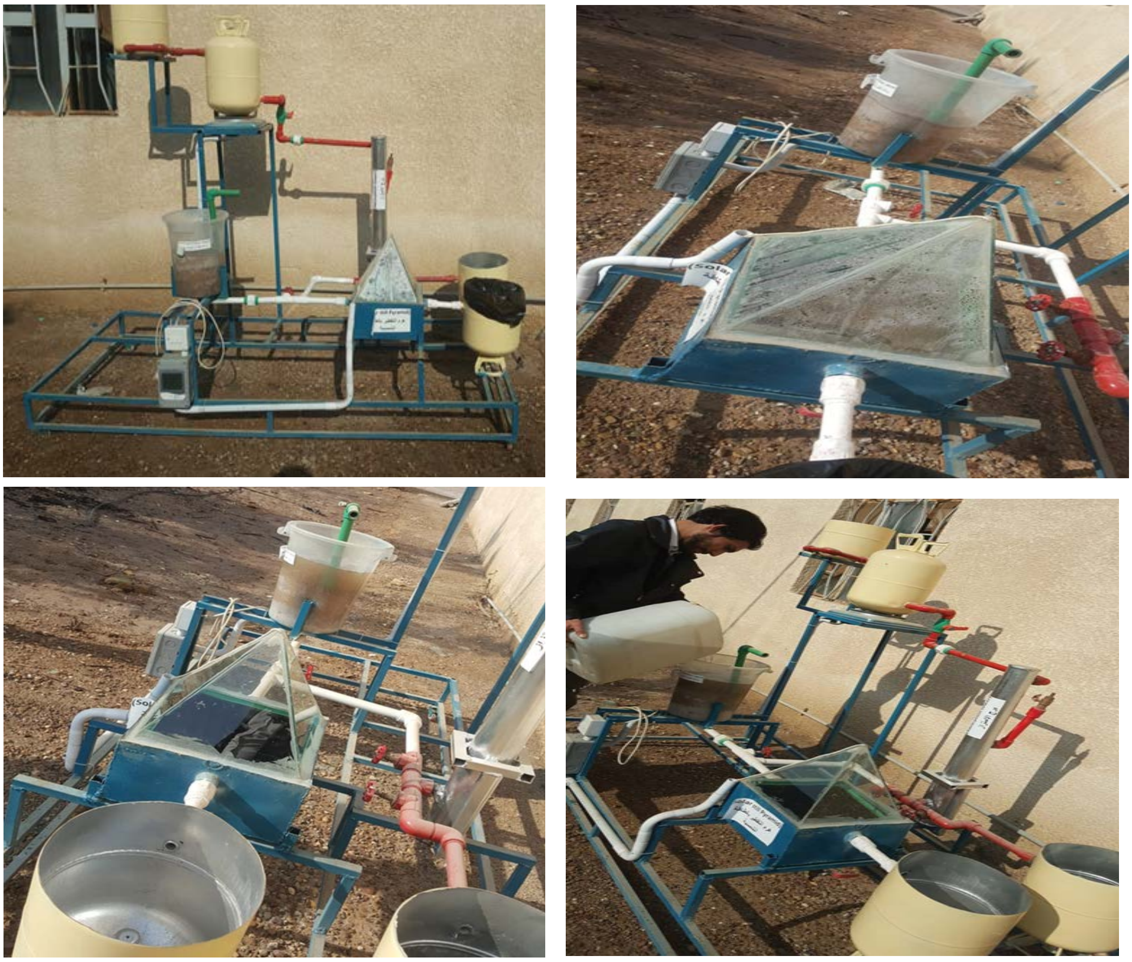

Fig. 3. Pictorial images of the PSDS aided filtration basin produced locally for the experiment.

wastewater sample was introduced into the filtration basin at a constant flow rate of 0.015 m^3/s through a hose. The flow rate was monitored using a flowmeter. The removal of oil BOD was monitored at the time interval of contact time (1–18 h). The process was repeated for 17 experimental runs. In each experimental run process, the layer of charcoal varied from 100 to 2,000 g was weighed and added to the filtration basin to improve the treatment process. The percentage of BOD removed from the wastewater sample at specific time intervals was recorded. The supernatant (water sample collected from the outlet of the filter basin) was transferred into the PSDS system through an outlet valve.

In the second stage of the experiment, the initial temperature of the wastewater sample entering the PSDS was measured using a thermometer. The water was then introduced into the distillation chamber of the PSDS. The temperature inside the PSDS was monitored and recorded via a sensor positioned within the system, while the external temperature was read from a thermometer. The PSDS consisted of a translucent glass lid to trap UV radiation and solar energy from sunlight straight into the solar still. The PSDS by design enables UV light and solar energy to penetrate through, thereby heating the water sample below in the pool. The solar energy trapped within the still causes the water to evaporate, while the contaminants in the water are retained in the pan. Moisture-filled air rises to the slanting glass sheet and condenses after cooling from the ambient air outside. The accumulation of clear water through still was monitored for the removal of stubborn organics at varying contact times, and internal and external temperature of the PSDS system. The BOD concentration (mg/L) of the accumulation was read at regular time intervals and was measured in milligrams of oxygen per liter ($mg \cdot O_2/L$) using a BOD meter [31,32]. The residual concentration of oil-water separation from the filtration process was determined by Eq. (1), while the percentage of organic removal was evaluated from Eq. (2).

$$\text{Residual Oil after separation} = C_0 - C_t \tag{1}$$

$$\text{BOD}_2 \text{ Removal Eff} = \frac{\left[C_0 - C_t\right]}{C_0} \times 100 \tag{2}$$

where C_0 is the initial concentration in (mg/L) of the organic (BOD) contaminant present in the hydrocarbon wastewater sample before treatment, and C_t is the final concentration (mg/L) of the organic contaminant present in the water sample at time t (h) after the treatment process.

2.4. Statistical evaluation of the wastewater treatment process

The optimization of the industrial wastewater treatment process was evaluated following the application of one-variable-at-a-time (OVAT) statistical analysis by employing the trial version of the NCSS software version 20. The analysis of variance (ANOVA) was employed to investigate the significance of experimental variables at a 95% confidence level. In this case, the significant effect of contact time, pH of solution, and weight of charcoal on the efficiency of the filtration basin-aided PSDS will be investigated at probability statistics ($p < 0.05$).

3. Results and discussion

3.1. Characteristics properties of the petroleum refinery wastewater

The results obtained before the characteristics of the industrial wastewater are presented in Table 1. The result shows that the industrial wastewater under investigation contains 59.7 NTU of turbidity content at pH 7.3. This value is greater than the 10 NTU minimal threshold required for industrial effluent discharge [1,2]. The BOD content of the wastewater is 3.10×10^6 mg/L. The BOD content is higher than the residual 30 mg/L stipulated for clear discharge [2], and considerably higher than the 125 mg/L stipulated baseline for the minimal permitted threshold [1]. The higher concentration of BOD indicates a higher level of organic pollutants in the water medium. The concentration of dissolved oxygen (DO) content in the wastewater amounts to 60 mg/L. The low level of DO is consistent with the high concentration of organic matter in the wastewater, confirming a degradation in the water quality with a tendency of the water to stimulate the growth of bacteria and algae [48], limit photosynthetic activities, and disrupt the balance in the aquatic ecosystem [45,49]. The TDS contents in the water amount to 2.35×10^6 mg/L with an electrical conductivity value of 1,479 μS/cm. The TDS is higher than the 30 mg/L stipulated threshold required for sustainable effluent discharge [2]. The electrical conductivity value of the wastewater is high and confirms the presence of dissolved hydrocarbon salts in the industrial wastewater. It can thus be concluded that the physicochemical parameters of the petroleum refinery wastewater are highly polluted and confirmed the need for the environmentally sustainable discharge.

3.2. Effect of process parameters on the treatment process

3.2.1. Effect of weight of charcoal on BOD reduction

The outline of Fig. 4a and b shows the effect of the mass of charcoal on BOD removal in the filtration-aided PSDS system. It can be observed from the outline of 4a in Fig. 4 that BOD concentration was reduced significantly from 3.10×10^5 mg/L initially to less than 2×10^3 mg/L of residual BOD content in the petroleum refinery wastewater medium. The outcome proves that the adsorptive uptake of organic matter is very effective with charcoal [48]. Also,

Table 1
Characterization of wastewater before and after treatment

Water parameter	Before treatment value	After treatment value
Turbidity	59.7 NTU	Nil
pH	7.3	6.5
Dissolved oxygen (DO) content	6×10^{-2} mg/L	Nil
Density	1.04×10 g/cm^3	1.01 g/cm^3
Conductivity	1.48×10^3 μS/cm	–
Total dissolved solids	2.35×10^6 mg/L	1.3×10^4 mg/L
Biological oxygen demand	3.10×10^5 mg/L	1.0×10^3 mg/L
Total oil content	100 mg/L	Nil

the curvature of the graph in 4b in Fig. 4 confirmed that the BOD removal increased significantly as the mass of charcoal added to the filtration basin increased. The increase in the mass of charcoal provides a larger surface area for adsorption, resulting in a proportionate decrease in residual BOD [49]. The outcome corresponds to a maximum 99.5% BOD removal efficiency recorded with an optimum 2,000 g of charcoal and transcends to a minimum residual BOD concentration of 1×10^3 mg/L in the water medium as shown in the outline of 4b in Fig. 4. The result confirmed the effectiveness of the filtration unit as efficient in removing a substantial portion of organic compounds present in the water medium [50]. Considering the effectiveness of charcoal in the adsorption of organic compounds, a further increase in the mass of charcoal might diminish BOD removal efficiency due to the saturation of active sites on the adsorbent medium. It becomes imperative to investigate the impact of other parameters, such as pH, and exposure time on the overall performance of the filtration-aided PSDS. The statistical evaluation of the treatment process based on p-value statistics confirmed the effect of the weight of charcoal has a significant impact on the BOD reduction at a p-value of 0.0001 at 95% confidence interval ($p < 0.05$).

3.2.2. Effect of charcoal mass on pH of the medium

The gradient of Fig. 5a and b illustrates the effect of charcoal on the pH of the solution, and the removal of organic compounds from petroleum refinery wastewater. The result proved that the pH of the water medium decreased significantly from an initial pH of 8.5 to pH 6 as the mass of charcoal added to the filtration basin increased from 200 to 2,000 g. This outcome indicates a change in water chemistry [2] while decomposing organic compounds in the medium [17]. The acidic ions released by charcoal confirmed the presence of acidic salts characterized by the high conductivity recorded in the industrial wastewater. The ionic exchange between the hydrocarbon salts and the constituents of charcoal probably caused a change in the pH of the solution from alkaline to acidic solution under the effect of increasing charcoal mass [43]. This outcome suggests that an acid-neutralizing effect probably occurred on the surface of charcoal [2,44], with a tendency to lower the pH of petroleum wastewater [6]. The water medium was affected by the low pH of charcoal [43,44], as the mass of charcoal increased leading to the increase in H^+ ions in solution [1,6]. Thus, the optimum pH of 6.0 corresponds to the overall performance of the treatment process and transcends to 99.5% organic removal efficiency. The optimum pH of 6 in the finish effluent is acidic [1], thus in biological systems, the pH of the environment has a profound effect on microbial growth [45]. The optimum pH reported in the PSDS system is consistent with the result of petroleum refinery effluent treatment reported in reference [1]. The finding from the p-value statistical result at 95% C.I. ($p < 0.05$) confirmed the weight of charcoal has a

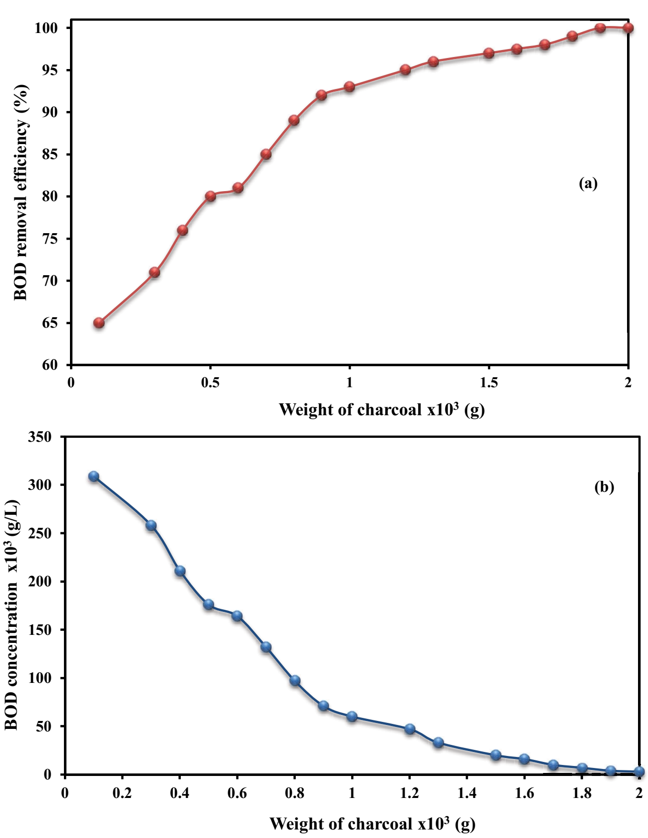

Fig. 4. Effect of weight of charcoal on (a) the BOD removal efficiency and (b) residual BOD concentration following the PSDS aided filtration treatment of Industrial wastewater.

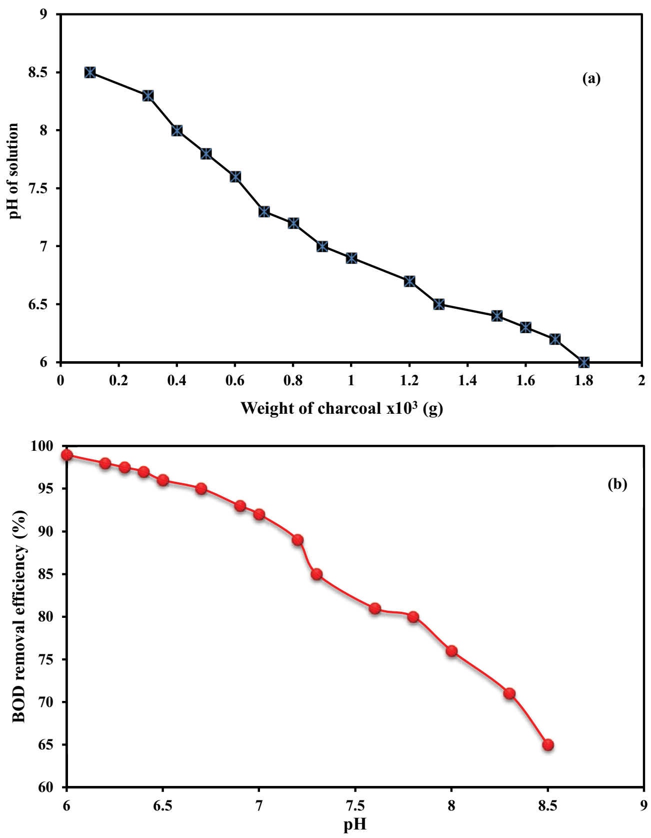

Fig. 5. Effect of pH of solution on (a) weight of charcoal and (b) BOD removal efficiency following the filtration treatment of industrial wastewater.

significant influence on the pH of the medium at a *p*-value of 0.0006.

3.2.3. Effect of exposure time on performance of PSDS system

Fig. 6a and b illustrate the effect of contact time on the overall performance of charcoal in PSDS. The results show an intermittent increase in the weight of charcoal with contact time [46]. The trajectory of the line in 6a of Fig. 6 shows an increase in the contact time of charcoal with organic constituents present in the industrial wastewater. The outline of the 6a in Fig. 6 confirmed the reduction of BOD present in petroleum refinery wastewater from 3.10 × 10^5 mg/L to residual 1 × 10^3 mg/L as contact time increased significantly from 6 to 18 h. This outcome established that 3.09 × 10^5 mg/L in concentrations of BOD was removed from the industrial wastewater. The curvature of 6b in Fig. 6 confirmed that the best performance of the PSDS system was recorded at a contact time of 18 h. The outcome proved that the decomposition and subsequent removal of organic matter increased significantly with contact time in the PSDS system [33–35]. This result corresponds to a 99.5% increase in the removal of organic matter that consumes oxygen in the petroleum refinery wastewater. Comparatively, the findings proved that the PSDS system for the industrial effluent remediation process outperformed the bio-remediation methodology reported in [47] for BOD removal in petroleum refinery wastewater using (*Bacillus cereus*) AKG 1 requiring a longer period of 20 d. Additionally, the PSDS remediation approach to petroleum refinery wastewater outperformed the 87.68% BOD removal from 110 to 13 mg/L after 12 weeks via the phytoremediation process using *Echinochloa pyramidalis* in phytoremediation drum system [51].

3.2.4. Effect of radiation temperature on performance of PSDS system

The effect of solar radiation on the petroleum wastewater treatment process. Fig. 7 illustrates the internal and external temperature variations with time investigated. Outline of 7a in Fig. 7 confirmed that the temperature within and outside the PSDS system increases intermittently and then decreases as exposure time to UV from sunlight increases from 2 to 18 h. The curvature of 7b in Fig. 7 confirmed the external temperature increased intermittently with the BOD removal. The BOD removal decreased as the external temperature increased from 28°C to 45°C. The best performance corresponding to minimal residual BOD concentration of less than 2 × 10^3 mg/L was obtained at an external temperature of PSDS less than 38°C, the output corresponds to residual BOD less than 1.2 × 10^3 mg/L and translates to 99% BOD removal efficiency. The same trend was observed with internal temperature variation with residual BOD concentration in the PSDS system. The best performance was recorded at an internal temperature of less than 50°C and translates to a residual BOD concentration of 1 × 10^3 mg/L in petroleum

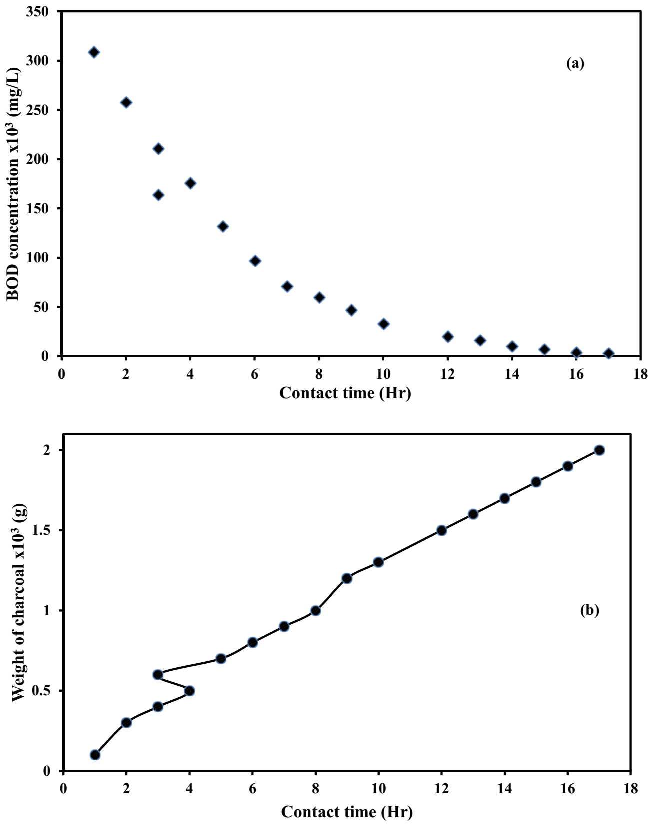

Fig. 6. Effect of contact time on (a) weight of charcoal and (b) BOD removal efficiency following the PSDS aided filtration treatment of petroleum wastewater.

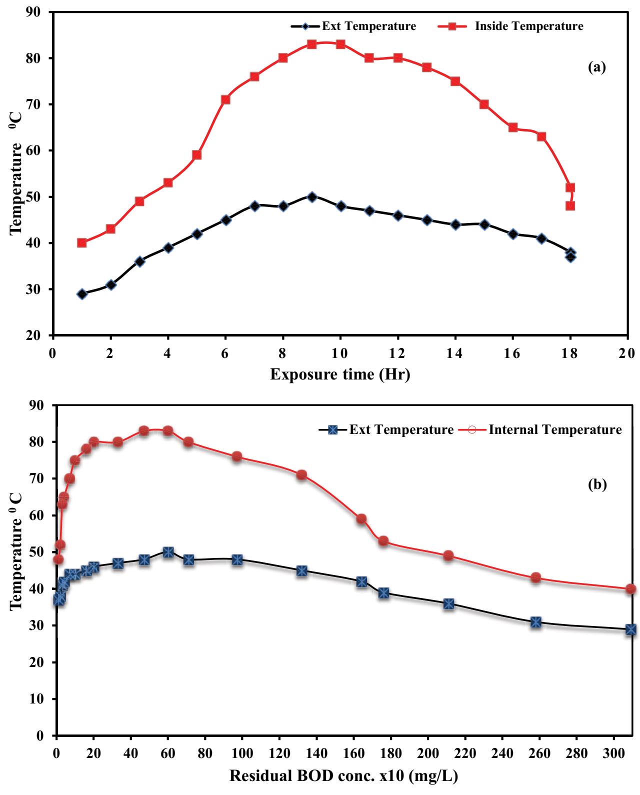

Fig. 7. Effect of temperature variation (a) with exposure time and (b) BOD removal following the PSDS aided filtration treatment of wastewater.

(a). (b)

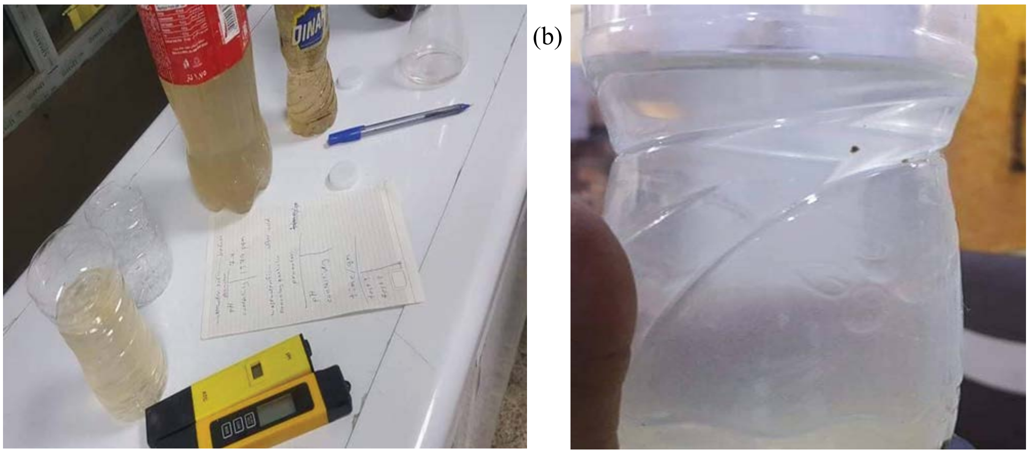

Fig. 8. Pictorial representation of the petroleum refinery wastewater (a) before treatment and (b) after the PSDS-aided filtration treatment of wastewater.

refinery effluent. The outcome proved that the optimum performance of the PSDS system is recorded at 37°C external temperature and 48°C internal temperature, respectively. The findings were consistent with the report in published reference [52,53]. The BOD removal efficiency of 99.5% recorded in the present study was significantly higher than the 87.65% reported for the solar distillation system in reference [53], and 99.2% reported in the work of reference [52]. Thus, it can be concluded that a narrow margin of temperature difference is most effective for the overall performance of a solar still system.

3.3. Characteristics of the water after treatment process

The pictorial representation of the finished water sample is shown in Fig. 8. Final characteristics of the water recorded after the PSDS aided-filtration treatment of petroleum wastewater shown in Table 1 confirmed that the PSDS system reduced the BOD contents under investigation to a minimal level. The total oil content (TOC) content was reduced significantly to meet the minimum threshold for natural disposal into the environment [1,2]. The PSDS reduced significantly the turbidity content in the finished petroleum refinery effluent to less than 0.5 NTU. Furthermore, the treatment process eliminated all oil content from the water and reduced the TDS concentration in the finished water to below 100 mg/L. The final water treated by the PSDS process met the criteria for clean discharge, including pH between 6 and 8.5, turbidity of less than 5 NTU, and TOC levels below 1.0 mg/L, as reported in [1].

4. Conclusion

During this study, the possibility of treating polluted water from the oil refinery was examined by using a solar distillation apparatus to remove dissolved solids and the content of organic matter by examining the BOD level. The results proved the ability of PSDS to get rid of organic matter at a rate >98% and reached BOD value reached a residual concentration of 1×10^3 mg/L. Although there are many limitations, including the intensity of solar radiation and the thickness of the glass cover of the distillation device, the solar still has proven to be highly efficient in terms of the efficiency of water purification and the isolation of pollutants and toxic substances. We recommend the application of the PSDS system for the investigation of oil content removal in future studies.

Disclosure statement

Conflict of interest

The authors solemnly declare that there are no conflicts of interest and okay the manuscript for publication.

Compliance with ethical standards

This research article does not contain any studies involving human or animal subjects.

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Data availability

The authors declare that the data used for this research were obtained from laboratory experimentation. The data stored in a repository will be made available prior to request to the corresponding author.

Contribution of the authors

(I.H.A): Data Curation, Conceptualization, Writing Original Draft, Project Supervision. (T.F.A): Resources, Writing original draft, Visualization. (R.A.M): Writing Original Draft, Project Supervision, writing review (A.K.H): Resources, Writing Original Draft, Project Supervision (P.E.O): Data Visualization, Software, Writing reviews & Editing.

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