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Adsorption and Treatment of Petroleum Products Contaminated Water using Activated Carbon Produced from *Hura crepitans* Linn Seeds

Yelwa Abubakar Sani[1], **Ogbiko Cyril**[1] **and Muhammad Aliyu**[2,*]

[1] Department of Pure and Applied Chemistry, Usmanu Danfodiyo University, Sokoto, Nigeria

[2] Department of Microbiology, Ambrose Alli University, Ekpoma Edo State, Nigeria
e-mail: muhammadaliyufla@gmail.com

[*] Corresponding author

Abstract

Industrial waste water arising from crude oil exploration and processing activities poses a serious environmental problem in Nigeria. Consequently, a study of the use of activated carbon in the clean up was designed by this study with the aim of reducing the water contamination to a more acceptable level. Locally produced activated carbon produced from *Hura crepitans* seeds of different mass was employed as adsorbent with its adsorption potentials compared with standard commercial activated carbon. A 2h contact time was allowed after which physicochemical parameters such as temperature, odour, appearance, density, pH, chemical oxygen demand, dissolved oxygen, biological oxygen demand, conductivity, turbidity, total dissolved solids, chloride, sulphate, total hardness, calcium and magnesium concentrations were determined. Results of the analyses showed that the produced activated carbon is an excellent means for the removal of the contaminants as seen by the decrease in the contaminant concentration from an initial concentration to that within or close to both WHO and refinery specifications. The results of this study revealed that the powdered form of the activated carbon from *H. crepitans* seed would be very effective in the remediation of petroleum-hydrocarbon contaminated ground water and its use is therefore recommended.

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Introduction

Industrial wastewater is often contaminated with various compounds such as: phenol, chromium, suspended solids, dissolved organic compounds among others hence it is imperative that it should be treated to an environmental acceptable limit. The current problems in wastewater treatment stem primarily from the increasing pollution of waters by organic compounds that are difficult to decompose biologically, because these substances resist the self-purification capabilities of the rivers as well as decomposition in conventional wastewater treatment plant [1]. Oilfields like most production activities generate large volumes of liquid waste called produced water [2, 3]. It is estimated that globally over seventy billion barrels of produced water is generated by the petrochemical industries annually [2, 4]. The constituents of discharged water from petroleum activities are broadly classified into organic and inorganic compounds which includes dissolved and dispersed oils, grease, heavy, metals, radionuclides, treatment chemicals, formation solids, dissolved gases, scale products, waxes, microorganisms and dissolved oxygen [2, 5, 6]. The discharge of emerging wastes and pollutants in water bodies is on the rise with increase in industrialization. Wastes from petroleum refining are not only carcinogenic but are bio-accumulative which are not easily biodegraded and hence if discharged into the environment, they could cause environmental and human health effects in addition to the pollution of surface water, ground water and soils [2, 7]. If extensive discharge is done into water bodies, it could cause "chronic pollution" capable of limiting the breeding and reproduction of seabirds [8]. There is therefore the need to treat adequately waste water from petroleum production processes before their discharge to protect water resources and avoid acute and chronic toxicities [8, 9]. The treatment choice is however dependent on factors such as regulatory acceptance, site location, technical feasibility, cost of equipment and availability of infrastructure [2, 8, 10] with the management practices suggested to be environmentally friendly in itself.

Hura crepitans Linn. is a tropical plant belonging to the family Euphorbiacea. In Nigeria, it is known as "*odan mecca*" by the Kabba people of Kogi State and "*aroyin*" by the Ijesha people of Osun State. *H. crepitans* is often planted in towns and villages as a shade tree. It has short, densely crowned spines on the trunk and branches; the long stalked leaves with prominent, closely parallel pinnate nerves, the purple flower spikes, and the large fluted flattened fruits are highly distinctive. This tree is up to 90-130 feet high with a wide spreading crown, branching low down. Leaves are 5-20 cm long by 5-

15 cm broad, ovate, shortly and abruptly acuminate, and dark green in color. This tree flowers usually at the beginning of and again at the end of rainy season. One nut is a flattened and fluted disc with five to 20 lobes about 2.5 cm deep and 7.5 cm wide on a stout stalk. The capsule splits explosively, releasing one flattened circular seed about 18 mm across from each chamber [11, 12].

The traditional treatment of the wastewater effluents from the refinery and petrochemical plants are usually based on the mechanical, physicochemical and biological methods. The conventional processes that are applied for the treatment of petrochemical wastewater can only partially remove the contaminants hence the need for diverse and more advanced treatment techniques [9, 13]. Among the advanced treatment techniques adsorption on activated carbons for waste water treatment has been found superior compared to other chemical and physical methods in use in terms of its capability for efficiently adsorbing a broad spectrum of pollutants, and its simplicity in design [14]. This study was undertaken with the aim of ascertaining the effectiveness of activated carbon from *Hura crepitans* seeds in adsorbing pollutants from industrial waste water from petroleum production processes viz-a-viz ascertaining the level of certain physicochemical parameters in the effluent waste water.

Materials and Methods

Sample collection

Fresh and matured sample of *Hura crepitans* fruits were collected randomly behind College of Agriculture, Agenebode Etsako East Local Government Area of Edo State Nigeria. The samples were transported to the laboratory in clean polythene bag with its identity confirmed by a taxonomist

Sample preparation

The plant fruits were broken manually to release its seeds. The seeds were washed with running tap water to remove the contaminants before being shade dried for seven days. Good and matured seeds of variable sizes were manually selected and grounded into powder using an electric milling machine. The powered seeds sample was stored in an air tight glass container until use.

Preparation of activated carbon

The procedure as outlined by Ali et al. [15] was adopted with slight modification.

500 g of the pulverized *H. crepitans* seed powder was placed in a crucible and covered by 600 g of sand. The crucible was heated in a furnace oven at 800°C for 2h with the resulting products of weight 202 g were separated from the sand, placed in a crucible and further heated in a steam sterilizer at 120°C for 2h. The resulting product was then crushed and sieved in mesh size No 40 (425 µm) and mesh size 8 (2.36 mm).

Chemical activation of the activated carbons

The procedure for the chemical activation of the produced activated carbon was according to the procedure outlined by Ademiluyi et al. [16]. The produced activated carbon was carefully weighed and put in beaker containing 120 cm^3 of 0.1M HCI. The content of the beaker was thoroughly mixed until it forms a paste. The paste was then transferred to a crucible and placed in a Muffle furnace heated to 800°C for 2 h. The sample was then cooled at room temperature, washed with distilled water until the pH of the draining water is in the range of 6-7. The sample was then dried in an oven at 105°C for 3 h. The final product was kept in an air tight glass container until ready for use.

Procurement of discharged petroleum product contaminated industrial water

Industrial waste water used for the adsorption studies were collected thrice (coded A, B and C) at different times (7 days intervals) from a petroleum refining industry in Port Harcourt, Rivers State in Nigeria. The sample was collected after mechanical and biological treatment has been carried out on sample in a pre-rinsed 10 litre plastic containers, sealed and transported to the laboratory for analysis.

Adsorption of organic contaminants using the produced activated carbon (PAC)

100 ml of the waste water samples in batches were mixed with 1.00, 2.00, 3.00, 4.00 and 5.00 g respectively of the produced activated carbon in 250 ml Erlenmeyer flasks. The mixtures were shaken thoroughly and intermittently for 2 h before being filtered using a Whatman filter paper No 1 size to remove the carbon. The characteristics of the industrial waste water was analysed before (negative control) and after treatment with the produced activated carbon (PAC). Physicochemical parameters of the representative samples such as temperature, odour, appearance, density, pH, chemical oxygen demand (COD), dissolved oxygen (DO), biological oxygen demand (BOD), conductivity, turbidity, total dissolved solids (TDS), chloride, sulphate, total hardness, calcium and magnesium concentrations at the varying dosages of the activated carbon used were performed within a period of 7 and 10 days using methods described by the American

Public Health Association (APHA) [17, 18]. The same procedure was performed for sample A, B and C and the procured commercial activated carbon (CAC) (positive control) with the mean result taken to serve as the representative result.

Results and Discussion

The physicochemical characteristics of the discharged industrial water treated with different dose of the adsorbent with reference to the commercial sample is presented in Table 1.

Table 1. Physicochemical characteristics of the discharged industrial water

Parameters	PAC Mean Value (Before Analysis)	PAC Mean Value (After Analysis)	5.00 g of CAC Value	Standard
Temperature (°C)	34	28	28	38^RS^
Odour	Offensive	Totally removed	Totally removed	NS
Appearance	Not clear	Clear	Clear	NS
Density (kg/m^3)	843	(800), (800), (820), (820), (840)	NS	NS
pH	5.2	(5.4), (5.8), (6.5), (6.9), (7.3)	7.7	6.5–8.5^WHO^
COD (mg/l)	373	(371), (343), (311), (287), (261)	148	150 ^RS^
DO (mg/l)	4.1	(6.1), (6.9), (7.4), (8.3), (8.8)	9.1	13 – 14^RS^
BOD (mg/l)	52.6	(45.6), (41.8), (37.4), (33.3), (28.8)	15.5	10 – 20^RS^
Conductivity (µS/cm)	767	(694.5), (672.4), (666.2), (658.9), (641.7)	41.4	0 - 40^WHO^
Turbidity (NTU)	2.5	(2.3), (1.9), (1.7), (1.5), (1.2)	1.1	1^WHO^

TDS (mg/l)	878	(857), (821), (798), (746), (664)	354	0 - 500[WHO]
Chloride (mg/l)	765.1	(672.8), (522.5), (482.5), (442.5), (346.9)	248	250[WHO]
Sulphate (mg/l)	235.2	(222.8), (197.6), (187.3), (172.4), (165.7)	153.6	150[WHO]
Total hardness (mg of $CaCO_3$ per litre)	17.56	(24.4), (39.6), (41.6), (47.9), (57.7)	195.2	200 - 250[WHO]
Calcium (mg/l)	397.60	(391.1), (387.6), (381.3), (379.7), (377.7)	197,5	75 - 200[WHO]
Magnesium (mg/l)	12.80	(19.4), (21,7), (23.8), (25.7), (28.9)	31.3	30 - 150[WHO]

Key: PAC = Produced activated charcoal, CAC = Commercial activated carbon, WHO = World Health Organization [19], RS = Refinery specification. NS = No specification. The first, second, third, fourth and fifth brackets represent the mean results for 1.00, 200, 3.00, 4.00 and 5.00 g of the produced adsorbent respectively.

All the examined parameters except pH were found to be outside the prescribed ranges set by the World Health Organization (WHO) [19]. This is line with similar observations made by Horsfall and Spiff [20] and Odu et al. [21]. The results however are evidence of the ability of the adsorbent to remove the contaminant from the industrial waste samples by attaching to the contaminant molecules at an available adsorption site when higher concentrations of PAC were employed. The result showed that there was a decrease in the concentration of organic pollutants with increasing concentration of the activated carbon dose used. The adsorptive property of the PAC may have been influenced by factors such as particle-size distribution, concentration of the adsorbent, surface area and contact time. The CAC showed a better adsorbing potential at the highest concentration of PAC employed for the study since it possess a better surface area to volume ratio and adsorbing potentials similar to the report of Olafadehan and Aribike [1] and Snoeyink and Summers [22] who treated industrial waste water with commercial activated carbon.

The temperature of the wastewater after adsorption process was constant due to the fact that, the analysis was carried out at room temperature. The result showed that the PAC corrected the pH to specification as against the effluents before analysis. There was a remarkable decrease in the amount of total dissolved solid after treatment. It was reduced from 878 before treatment with the PAC to 857 and 664 when 1.00 g (the lowest) and 5.00 g (the highest) concentration of the produced adsorbent was used. With respect to COD in the discharged waste water before and after treatment, it can be observed that the COD was reduced from 373 mg/l to 371 mg/l and 261 mg/l when 1.00 g and 5.00g of the PAC was used respectively. The result obtained shows that the dissolved oxygen was increased from 6.1 mg/l before treatment to 8.8 mg/l when the highest concentration of the adsorbent was used for 2 h contact duration. The BOD was reduced from 52.6 mg/l before treatment to 28.8 mg/l when the highest concentration of the adsorbent was used for 2 h contact duration. This thou higher compares favorably with the standard adsorbent used. The result presented in Table 1 showed that the turbidity was relatively removed. As regards other parameters like conductivity, chloride, sulphate among others, the results from this show showed the potential of the PAC to correct their concentration to specifications. The activated carbon produced from *H. crepitans* seeds completely removed the offensive odour in the industrial waste water, with clear appearance.

Conclusions

H. crepitans seeds used as precursor successfully produced activated carbon locally. The adsorption tests clearly revealed that the locally prepared activated carbon can be used as effective and low cost adsorbent. Though the contaminants removal efficiency increases with a corresponding increase in the concentration of the locally prepared activated carbon, its potentials were less but comparable to that of the commercial procured activated carbon used as standard. Therefore, the effectiveness of the activated carbon produced from *H. crepitans* seeds in the removal of organic contaminants has further been validated.

Conflict of Interest Statement

The authors declare that there is no conflict of interest that could be perceived as prejudicating the impartiality of research reported.

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