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Treatment of Hydrocarbon Contaminated Drinking Water in Niger Delta, Nigeria Using Low-Cost Adsorbents (Coconut Shell)

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Abstract

The basic aim of this research was to explore how to effectively treat hydrocarbon contaminated drinking water using activated carbon made from coconut shell, in the context of Niger Delta region of Nigeria. While adsorption was seen as a viable option for easy removal of hydrocarbon contaminated drinking, simulation of contaminated drinking water was carried out using diesel as contaminant. The results indicated that there was more of a physical process in the tests/analysis rather than the anticipated chemical process. When fitted into existing Isotherm models, values charts and values obtained showed that the adsorption was not a favorable one for treatment of hydrocarbon contaminated drinking water. The study identified huge hydrocarbon contamination in drinking in Niger Delta and concluded that further studies be carried out in order to effectively curb the problem, in a more economical and viable manner.

Keywords: Hydrocarbon contaminated drinking water, Pollution, Activated Carbon, Water Treatment, Niger Delta, Nigeria

1. INTRODUCTION

Intake of contaminated drinking water poses severe risks to both human and other living organisms. With over 884 million people, or one out of eight people all over the world unable to have access to clean and treated drinking water, contaminated sources such as streams, local ponds, irrigation canals and unprotected dug wells have been seen as the only viable source of water (Abulencia et al., 2010). Given the potentiality of disease outbreak associated with the intake of contaminated drinking water, World Health Organisation (WHO) have insisted that water must meet certain requirement and standard before it can be certified as usable. The categories of water required to meet the stipulated standards include water used for drinking, domestic, food production and recreational purposes. However, these standards as required by WHO are never met in the Niger Delta region of Nigeria where there are widespread oil contamination and pollution due to activities of oil companies. There is increasing recognition that 9.1% of disease and 6.3% of all death globally occurred as a result of poor water sanitation and hygiene, and calls have been made for the incorporation of high level treatment of water at the point of use (POU) given that it increases the microbiological quality of drinking water and in turn reduce the risk of diseases (Boisson et al., 2009). This research responds to the calls for improved understanding of how to use high level treatment of water in solving contaminated water especially in developing nations (Niger Delta region of Nigeria). In the light of the challenges currently facing the Niger Delta region under consideration, this research aimed at proffering a suitable solution for the treatment of hydrocarbon contaminated drinking water using low cost adsorbent; and in doing so, assess if coconut activated with Sodium Chloride (NaCl) is viable in the treatment of hydrocarbon contaminated water. The results from the research offers valuable insights into practical management of hydrocarbon contaminated water in the Niger Delta region of Nigeria.

Notably, despite the effort that has been put in place by WHO and United Nations International Children's Emergency Fund (UNICEF) to achieve sustainable water quality across different countries, much is yet to be done in developing countries like Nigeria. For example, the Niger Delta region of Nigeria known for oil exploration and production is constantly under threat from oil exploration, hydrocarbon contamination, oil pollution and degradation (Mafimisebi & Nkwunonwo, 2014; Mafimisebi & Thorne, 2015). This have led to deterioration of environmental conditions in Niger Delta region of Nigeria, particularly in Ogoniland (one of the Niger Delta areas heavily impacted by oil exploration) where hydrocarbon contamination in water was found 1000 times higher than Nigerian drinking water standard of 3 µg/l in seven wells (UNEP, 2011). In another context, although oil exploration and production in the Niger Delta region of Nigeria accounts for more than 80 percent of government revenues, employment generations and development (Mafimisebi & Nkwunonwo, 2015), activities of oil companies within the region make the environment and people vulnerable through constant gas flaring, oil spillages, environmental waste and climate change (Nkwunonwo & Mafimisebi, 2013; Mafimisebi & Thorne, 2016).

Furthermore, the Niger Delta region is regarded as a strategic region in Nigeria, evident from the

proceeds which accrued from the exportation of gas and oil because of the huge amount of hydrocarbon reserves found within the region (Anejionu et al., 2015). For example, it has been noted that the Niger Delta region has over 600 oil fields, 5000 oil wells, 275 flow stations, 10 crude export terminals, 3 refineries and a pipeline network over 2700 miles running through it (Olawoyin et al., 2012). In a related study, Zabbey & Uyi (2014) maintained that the huge exploration in the Niger Delta region of Nigeria have given way to almost daily occurrence of oil spillage; and there are increasing evidences from empirical studies that the Niger Delta region of Nigeria is one of the most severely impacted region in the world (Steiner, 2010; Zabbey, 2009; Olsson, 2012). However, the way and manner the proceeds of this exploration as well the residual effects (water and land contamination) are handled have brought about conflict and agitation within and around the host communities (Obi, 2014). See Figure 1 for the position of Niger Delta Region in Nigeria.

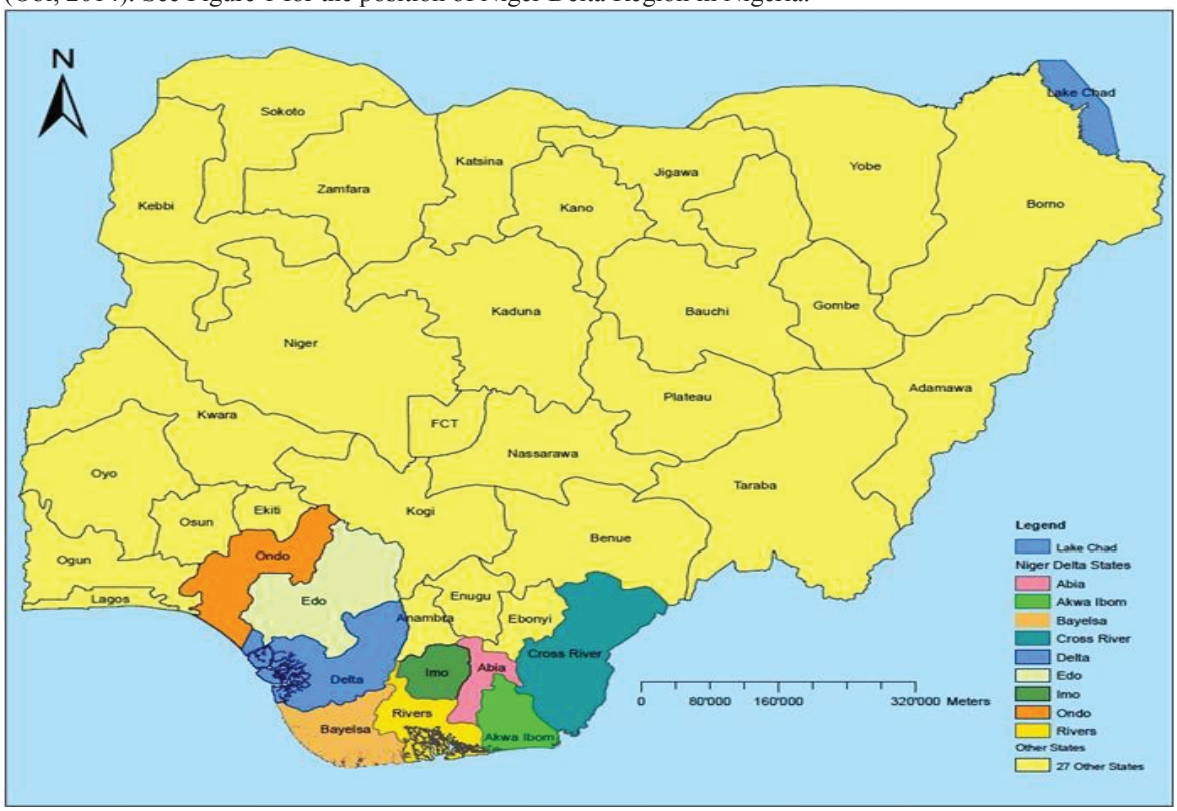

Figure 2 Map of the states that make up Niger Delta

Source: Ite et al. (2013)

In order to concentrate on all the challenges and constraints contained in the background information, this study as part of the objectives determined the conditions and materials required to adequately activate coconut shell and establish the adsorption isotherm from the experiments carried out. Furthermore, this study shed more light on statistical modelling of the obtained data using Langmuir and Freundlich Isotherm models and at the end, make recommendations based on the final outcome of the study. It is on the basis of the quality drinking water challenges ravaging Niger Delta that an in-depth review of literature that has been published with respect to water treatment techniques; adsorption of contaminants using activated carbon and coconut activated carbon in particular will be undertaken. Additionally, information gathered as a result of this review, was evaluated with an aim to see how it can be applied to improve the quality of drinking water in the study area.

2. CHALLENGES FACING NIGER DELTA REGION OF NIGERIA

There are several challenges facing the Niger Delta region of Nigeria ranging from frequent oil spillages, gas flaring, climate change, acid rain, rise sea level, environmental degradation and pollution, vandalism and militancy, and intra-communal crises among others (cf. Nkwunonwo & Mafimisebi, 2013; Mafimisebi & Thorne, 2015; 2016; Mafimisebi & Nkwunonwo, 2015). However, our focus in this research is restricted to hydrocarbon contaminated water within the Niger Delta region of Nigeria. Hydrocarbon contamination and pollution is seen to be an overriding predicament to the inhabitants of Niger Delta region of Nigeria due to exploration of crude in the area (Omofonmwan & Odia, 2009). The effects from hydrocarbon contamination and pollution have adversely undermined the quality of drinking water and consequently left local residents with the challenges of sourcing for clean drinking water. Ntukekpo (1996) cited by Aroh et al. (2010), maintained that 50 percent of oil spills in Nigeria is caused by corrosion of pipeline, with 28 percent and 21 percent attributed to sabotage and oil

production operation respectively. This is contrary to the claim that majority of the oil spills are as a result of vandalism caused by local inhabitants of areas where these pipelines are situated (Nwilo & Badejo, 2005).

There are often conflicting data regarding the extent of oil contamination and pollution in the Niger Delta region of Nigeria. Nevertheless, it has been found that remedial measures carried out by oil companies and multinationals in polluted sites do not meet the required threshold, and in most cases, these companies decide to neglect the stipulated remedial standards. One of such activity in Niger delta is the negligence of the contaminated Ogoni land which paved way for the recommendation of improvement of contaminated sites by United Nations Environment Program (UNEP, 2011). As a result of sheer negligence and unethical form of operation of oil companies, ethnic cleavages and youth bodies have embarked on invasion of multinationals as well as kidnapping and abduction of foreign nationals working in these oil companies (Chukwuemeka & Aghara, 2010). This practice of kidnapping, vandalism and militancy is not a sustainable solution to compel multinational oil companies responsible for environmental damages and pollution. In a recent research, it has been found that militancy approach as a form of protest escalate the problems of the Niger Delta region of Nigeria, and that militarization approach do not sufficiently addressed the problem (Mafimisebi & Thorne, 2015).

It is critical to note that several attempts have been made to address the problems of Niger Delta region of Nigeria but hydrocarbon contamination and spillages have increased in spite of the Nigeria oil pollution Act of 1990, National Environmental Standards and Regulation Enforcement Agency (NESREA) Act 2007, Environmental Impact Assessment Decree No. 86 of 1992, The Department of Petroleum Resources (DPR) Environmental Guidelines and Standards of 1991, establishment of National Oil Spill Detection and Response Agency (NOSDRA) and the formation of Niger Delta Development Commission (NDDC) to curb oil spill and monitor the clean-up of oil spillage (Mafimisebi & Thorne, 2015; 2016; Nkwunonwo & Mafimisebi, 2013; Nwilo & Badejo, 2006). The positive and sustainable impact of the various government interventions and regulations in the Niger Delta have not been felt even with the declaration of amnesty for ex-militants in 2009 aimed at attenuating the rate of oil terrorism, vandalism and oil theft (Al Chukwuma & Sunday, 2013; Mafimisebi & Thorne, 2015; 2016), pollution as result of oil exploration activities and vandalism by multinationals and local residents have been on the rise (Kadafa, 2012). The resultant effects of these activities have led to decline in economic activity for women who depend on fishing and other sea foods for trade, in addition to the risk of deteriorating water quality to communities that depend on well and river for drinking and bathing water (Nwidu et al., 2008). The failure of relevant Nigerian government agencies and regulatory bodies (e.g. NESREA, DPR, NDDC, etc.) put in place to curb activities that leads to water pollution, families in rural communities have been saddled with the responsibility of providing the quality of water they desire. In the next section, we will specifically discuss the issue of pollution and reveal the different forms of pollution as it affect the Niger Delta region of Nigeria.

3. POLLUTION

Pollution is a global problem encountered by both the developed and the underdeveloped countries but different countries react to the problem in different ways. The Organisation for Economic Co-operation and Development (OECD) in a bid to give a clear distinction defined pollution as a direct or indirect introduction of energy or substances by human activity into the environment with the tendencies of causing harm to human health, living resources and the ecosystem (Larsson, 1999). Jain et al. (2009), in a related definition defined pollution as those industrial, agriculture, household, vehicular and other human activities which render the air, water and the environment uncomfortable to human and other living organisms. Ravi (2011) in another development defined pollution as an undesirable change in our surroundings, partially or wholly with the tendencies of affecting water as well as land. Despite the similarities of the definitions above, Ravi (2011) failed to acknowledge the fact that these changes in the environment are directly or indirectly connected to human activities whether past or present. In this research, we acknowledge this basic distinction by arguing that human activities contribute largely to pollution problem especially in developing countries like Nigeria.

There are different forms of pollution. These forms include air pollution, water pollution, soil pollution, marine pollution, noise pollution, nuclear pollution (radioactive pollution), thermal pollution and chemical pollution. Given the context of this study, water pollution as it relates to drinking water will be considered, since the main aim of this work is to treat hydrocarbon contaminated water.

3.1 Water Pollution

Water environment in developing countries like Nigeria is increasingly faced with pollution problems impelled by population growth and economic development amidst unsound water policies that abound (Yang et al., 2015). Particularly in the Niger Delta region, crude oil (Hydrocarbon) exploration by the oil companies operating in the region is regarded as the major source of pollution in the water environment (Obinaju et al., 2015; Sojinu et al., 2010). Although hydrocarbon products are considered as the major pollutants, effluents from textile industries as well runoff during rainfall events are other forms through which the water environment is polluted. These

pollutants in addition to its effect on aquatic bodies and ecosystem, also affect individuals in rural areas who directly depend on river as source of water for cooking, drinking and other domestic purposes.

3.2 Major Pollutants

Pollutants in a broader sense allude to those materials/substances that alter the natural quality of the environment. These changes could be through physical, chemical or biological means. These changes occur as a result of the activities indulged by man. These activities include domestic, agricultural and industrial activities. These pollutants get to human through air, diet, water and contacts with polluted materials (Enault et al., 2015). Most of these contaminated water (predominantly consisting of hydrocarbon contaminants) through infiltration gets into ground water aquifer which is in turn taken to serve as drinking water. The quality of groundwater is strongly influenced by the type of surface water above it (Krishna et al., 2009). The review of these pollutants was considered necessary given that infiltration during rainfall events transfer them to ground water which in turn serves as drinking water source. These pollutants are of different forms which are described in detail below.

3.2.1 Heavy Metals

Majority of the water generated from the industries are predominantly contaminated with heavy metals. Though heavy metals like cobalt, zinc and copper are requisite for human existence as well as that of other living organism (Muhammad et al., 2011). Other heavy metals like lead and arsenic have been said to be problematic to the existence of living organisms (Yusuf et al., 2003; Karve & Rajgor, 2007; Citak & Tuzen, 2010). This heavy metal concentration when found in either drinking or waste water, exceed the allowable limit and, therefore, needs to be adequately taken care of to eliminate the risk it poses.

3.2.2 Phenols

Phenol and phenolic compounds are associated with high level of severe toxicity and bio-recalcitrant (Ye et al., 2015). Intake/exposure to phenolic compound have been associated with extreme health conditions such as; shock, delirium, phenolic breath, pulmonary distress, scanty dark urine and in extreme cases could result to damage of kidney, liver and eyes (Srivastava et al., 2015). Phenol as a harmful substance have been considered as a precursor or indicator of other toxic compounds containing phenol rings, especially highly toxic pentachlorophenol (Teixeira et al., 2015). As a result of problematic nature of phenol, it is necessary to limit/eliminate the concentration level of this compound in drinking water and effluents before it is discharged into the environment.

3.2.3 Dyes

With applications in various areas such as textile, paper, printing, colour photography, pharmaceutical, leather, cosmetic and plastic industries, dye is regarded as one of the significant contaminants found in water in addition to its toxicity and ability to accumulate in the environment (Zhang et al., 2015). Dyes are regarded as a source of non-aesthetic pollution and eutrophication in water. A release of contaminated water containing dye can generate hazardous by products through hydrolysis, oxidation and other chemical reaction that occur in contaminated water (Kordouli et al., 2015). Taking into cognisance the mutagenic and carcinogenic nature of dye to living organisms, and the fact that about 15% of the world's total dye production is lost during the dyeing process and released to textile effluents (Cai et al., 2015), it is necessary that an adequate treatment be taken to eliminate the associated risk. This will either affect drinking water quality through flow into river and streams during rainfall event or through infiltration into ground water table.

3.2.4 Hydrocarbon Contaminants

With the presence of several companies in Niger Delta, whose primary activities are centered on exploration of hydrocarbon products, the quality of drinking water as well the lakes and streams have deteriorated and this have become a major problem. This is because these activities which are associated with spillages and oil theft adversely contaminate the river for those who depend on it as their available source of drinking water and other domestic purposes (Nriagu, 2011). Release of hydrocarbon products such as gasoline, diesel and fuel oil from above the ground level have been considered as the major pollutant of ground water given that they infiltrate into the ground during rainfall events (Chen et al., 2010). In most cases, this underground contaminated water ends up as drinking water through underground well. As a result of the toxic nature of polyclic aromatic hydrocarbon (Wu et al., 2011), a class of hydrocarbon, it is necessary to eliminate its presence in drinking given the fact that it is dangerous to health even at low dosage.

4. TREATMENT OF CONTAMINATED DRINKING WATER

Unfavourable environmental conditions such as threat to human life, wellbeing and economic activity have been attributed to inadequate management of contaminated water (UN Water, 2015). Taking into account the role

water plays in the existence of human and other living organisms, it is imperative to properly treat water that has been termed as contaminated. Northcott et al. (2010) in search of a material suitable for biodegradation of hydrocarbon contaminated ground water which also serves as drinking water source, discovered that zeolite can be regenerated and reused for sorption of hydrocarbon for up to three times in addition to the ability to be used as a bioremediation. Obtaining these materials and implementation might not be realistic in rural areas in Niger Delta region of Nigeria given the technicality and knowledge required.

However, Philips (2009) argued that permeable reactive barriers (PRB) can be used as a better alternative in the treatment and cleaning of ground water that are contaminated. Irrespective of the potential of water treatment, a tendency of contamination still exists given the fact that PRBs are functions of ground hydrology. Biofilter when tested to check how viable it would be in hydrocarbon contaminated water treatment considering its cost effectiveness, it was discovered it can treat contaminated water through the transportation of microorganisms, its initial adhesion colonisation and firm attachment with a potential for large scale production (Srivastava & Majumder, 2007).

In a recent study, Huang et al. (2015) experimented on a combination of polyvinylidene fluoride (PVDF) membrane with polyvinylpyrrolidone (PVP) membrane to treat contaminated water, it was discovered that the combination of the membranes effectively treated oily contaminated water in addition to an improved performance of the membranes compared to each of the membrane tested separately. Despite the successful outcomes of these studies, authorities in areas like Niger Delta region of Nigeria have not been able to replicate these studies. This has led to situations where individuals are saddled with the responsibility of sourcing for forms of water treatment affordable to them. Although there are several forms of low cost treatment of contaminated drinking water, coconut shell in the form of activated carbon was considered for this study considering its abundance in the region under consideration. Irrespective of the choice of low cost material used in contaminated water treatment, several techniques/mechanisms are in existence. Nevertheless, it is pertinent to choose the adequate technique/mechanism in order to achieve a desired result.

As a way to develop several techniques for treatment of specific hydrocarbon contaminants as well as organic pollutants; Thuy et al. (2012) and Tran et al. (2015) discovered that, advanced oxidation, aerobic degradation, filtration, ozonation, coagulation, flocculation, distillation, extraction, precipitation, and adsorption could all be adopted as a method for treatment. Considering the treatment methods, Chen et al. (2011) and Xi & Chen (2014), placed adsorption method as the most superior because of its ability to remove pollutants at low concentration with respect to the environment, in addition to economy and high efficiency. Despite the attractive nature of ion exchange and reverse osmosis due to its ability to recover pollutant value along with removal of pollution from effluent, it have been discovered that it is not economically viable (Rashed, 2013). Although, various chemical treatments have been suggested, they have certain inherent disadvantages and limitations for practical applications. Methods such as reverse osmosis, precipitation and solvent extraction are associated with high capital costs, sludge generation and residual toxicity. Adsorption method with its simplicity of design, among other chemical and physical methods that have been proposed for contaminated water treatment have been proven to be competent in terms of its capability and efficiency to adsorb a wild range of adsorbates (Tan et al., 2008). From the economic and environmental point of view, adsorption provides outstanding advantages like profitability, availability, low cost, efficiency and ease of operation (Owlad et al., 2009).

4.1 ADSORPTION

Adsorption as a form of water treatment involves a phase transfer process which is widely applied in practice to expunge substances from fluid phases (Worch, 2012). In this process, the contaminants or pollutants are separated from its liquid or gaseous surrounding and concentrated at the solid surface adsorbent (Gupta et al., 2009). The phenomenon of adsorption was discovered by Scheele in 1773 when he discovered the ability of charcoal to uptake gas (Brenner, 2013). Wood charcoal also possessed decolourising property; with decades of application in the alcohol industry. Notwithstanding, it was Kayser who first mentioned the term adsorption in 1881 adding that it may occur at liquid/solid and gas/solid interface (Dune & Manos, 2009). Adsorption as a form of water treatment is efficient in the removal of contaminants without producing detrimental by-products.

There are two types of adsorption in existence; the physical adsorption, which are known as van der Waals forces due to its weak attractive forces and chemisorption which is as a result of chemical bonding properties possessed by the adsorbent. On the forces between organic adsorbate and adsorbent, Wei et al., (2015), summarised the forces involved as van der Waal forces, hydrophobic bond, hydrogen bond, ligand-exchange, dipole-dipole interaction and chemical maintaining that enthalpy of activation can be applied to glean the forces between organic adsorbate and adsorbent. Similar forces have been found to be involved in the adsorption process when carried out on inorganic surfaces. The forces involved in the adsorption of amino acid onto inorganic surface like kaolite, hydroxyapatite and polymer resin have been characterised as; electrostatic interactions, specific covalent bond formation, hydrogen bond formation, ligand-exchange and ion-exchange reactions, hydrophobic effects, and the superimposed interactions of other adsorption mechanisms (He et al.,

2015).

However, the ability of activated carbon to adsorb various compounds in an aqueous system have been characterised as a time-dependent process. For the adsorption of aqueous organic compounds, the surface chemistry and the pH of the solution are the most important factors that regulate the adsorption process (Wibowo et al., 2007). It is also pertinent to know that the dosage of the adsorbent involved in the adsorption process is of significant impact in the determination of the adsorbent-adsorbate equilibrium of the system (Deveci & Kar, 2013).

5. COCONUT SHELL ACTIVATED CARBON FOR WATER TREATMENT

Over the years, several studies have come up with different materials suitable to be used as low cost adsorbents, most of which have been seen as a scarce commodity in rural areas and most of the under developed countries (e.g. Huang et al., 2015; Guzel et al., 2015; Santos & Boaventura, 2015). However, coconut has been seen as one of the materials that have remediated this challenge because of its availability in rural areas. The availability of this material in Niger Delta region of Nigeria mean it can be assessed to know if it is viable for treatment of hydrocarbon contaminated water. Sekar et al. (2004), explored the adsorptive efficiency of coconut shell activated carbon when used to filter a lead contaminated water and found that the time required for equilibrium during removal of ion metal is very short and that the removal efficiency of coconut activated shell increase with increase in carbon content, aligning to the fact that pH also affects the adsorption efficiency. This was supported by Amuda et al. (2007), who determined that pH has an effect on adsorption owning to the fact that it affects the solubility of metal ions, concentration of counter ions on the functional groups of the adsorbent and the degree of the adsorbate during reaction.

Taking into cognisance the effect of flow rate, initial concentration of pollutant, percentage of saturation and the particle size of the adsorbent, Kulkarni et al. (2013) discovered that coconut activated carbon was able to adsorb phenol from contaminated water. The adsorption isotherm aligned to the Freundlich adsorption isotherm, with the significant effect attributed to the particle size and the initial pollutant concentration. Similarly, Zhu & Kolar (2014) employed activated carbon derived from coconut shell for the removal of unwanted odour discovered in drinking water. The adsorbent exhibited a high level of adsorption by effectively adsorbing p-cresol with the pH, adsorbent dosage and the initial concentration playing a major role in the adsorption process

As a result of the infeasibility of reliable power supply to support the required temperature for activation in developing country like Nigeria (Oparaku, 2002), Cobb et al. (2012) alternated $CaCl_2$, $ZnCl_2$ and NaCl in a two-step activation process with coconut shell and discovered that partial pyrolysis accompanied with chemical activation yielded a high level of activated carbon suitable for treatment. Mohammed et al. (2015) conformed to this when a two-step pyrolysis (carbonisation and addition of KOH) of coconut shell was adopted to adsorb volatile organic compounds (VOCs), it reaffirmed that chemical saturation of a carbonised (600^0C to 700^0C for two hours) coconut shell gave a high yield. Despite the outcome of several projects, little or no research have been done using coconut shell as an adsorbent for treatment of hydrocarbon contaminated water in developing countries as well as rural areas like the Niger Delta region of Nigeria. With the employment of technology and the use of chemicals in the activation process of the activated carbon found in the review of literature, this research focus on developing activated carbon (partial pyrolysis and activation) column capable of treating hydrocarbon contaminated drinking water. Unlike other activation processes, this research made use of Sodium Chloride (NaCl) because of its availability in the region covered in this research. The primary focus of this research is to assess the viability of coconut activated carbon (activated with sodium chloride) in the treatment of hydrocarbon contaminated drinking water, with the main purpose of proposing a more simplified method for treatment of hydrocarbon contaminated drinking water using coconut shell.

6. MATERIALS AND METHODS

Coconut was chosen for this study because of its abundance in the study area. The coconut used for the experiment was purchased from a local shop in Portsmouth. The coconut was processed in the green house, University of Portsmouth with a hammer and a cleaver to separate the shell from the fruits. The resulting shell was broken into smaller sizes with the hammer. The broken shell was washed with deionised water and dried for 24 hours. The particle size of the adsorbent has been proven to enhance the adsorption capacity of activated carbon (Siangsai et al., 2015). In line with the objective of this study, the coconut shell was further broken down into smaller particles with a mortar. See Figure 2 for how the shell was broken. Although other methods of breaking down the particle existed, the mortar was chosen given its simplicity and availability in developing countries.

Figure 3 Breaking coconut shell into smaller particles with mortar

6.1 Adsorbent

According to Kong et al., 2012, a two stage process; carbonisation (650^0C) and activation (dehydrating agent) was adopted. The carbonisation was carried out for 1 hour and 2 hours respectively to ascertain which temperature was adequate. The carbolite CMF 12 – 7 model furnace with a maximum operating temperature of 1200^0C was used. It took 45 minutes for the furnace to rise to the required temperature.

6.2 Adsorption Test

Given that one of the objective of the research was geared towards water treatment using less cumbersome techniques while achieving the desired result, three columns were set up to carry out the adsorption test. The contact time for the three columns during the adsorption test was less than 2 minutes. With a diameter of 5cm, the columns had a length of 500 cm, 460 cm and 530 cm (A, B and C respectively). Column A and B activated carbon that was carbonised for 2 hours and 1 hour respectively, while column C which served as the control was coarse sand. See Fig. 3 for the column set up.

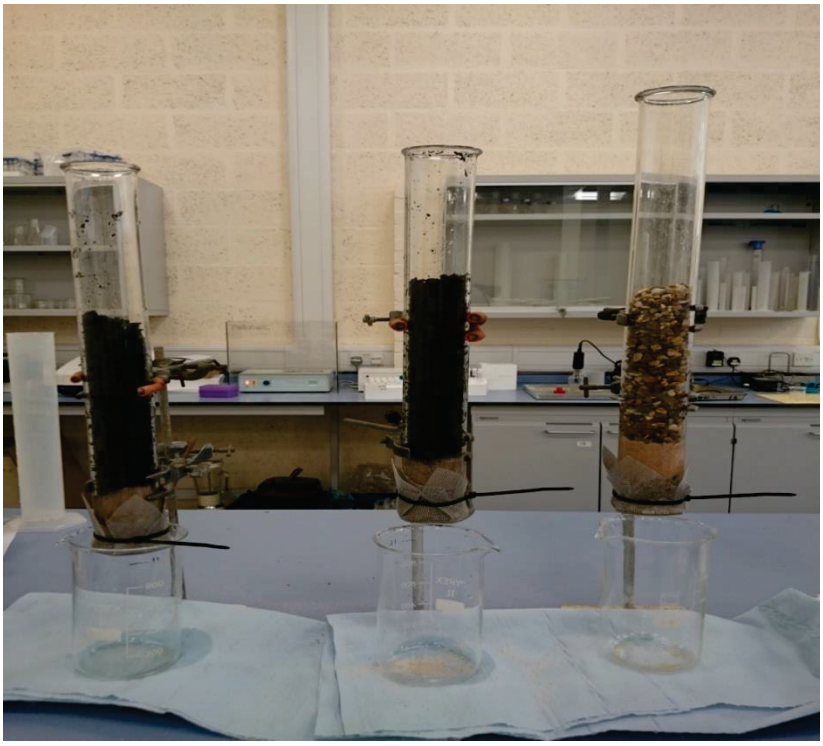

Figure 4 Set of Columns and beaker to collect water sample

1000ml of distilled water which was contaminated (simulated contamination) with diesel was manually shaken and poured through each of the columns. 2ml of the contaminated water was taken at every pass and put into a Mercury (Hg) free camlab vial which was heated before for 2 hours. The heated sample was allowed to cool off for 30 minutes before testing for the chemical oxygen demand (COD) using the Hach cholorimeter. See Fig 4.

Figure 5 Covered sample with COD value after test

The amount of hydrocarbon contaminated drinking water that was adsorbed by the adsorbent was calculated using the formula

$$q(e) = \frac{(Co - Ce)V}{W} \quad (1)$$

Where qe is the equilibrium adsorption (mg/g), Co is the initial concentration of hydrocarbon solution (mg/l), Ce is the equilibrium concentration (mg/l), W is the weight of the adsorbent (g) and V is the volume of the solution. The study of adsorption isotherm was carried out on two different isotherm models: Freundlich and Langmuir Isotherm models. The application of the isotherm models to the adsorption study that was done was compared by judging the correlation coefficients, R^2 values of the isotherm models.

6.3. LANGMUIR ISOTHERM MODEL

Langmuir isotherm surmises a monolayer of adsorption onto a surface containing a limited number of sites for adsorption as a uniform strategy of adsorption with no transmigration of the adsorbate in the plane of the surface. The Langmuir isotherm equation in its linear form is given as:

$$\frac{Ce}{qe} = \frac{1}{Qob} + \frac{1}{Qo} Ce \quad (2)$$

Where Ce is the equilibrium concentration of the adsorbate (mg/l), qe is the amount of adsorbate adsorbed per unit mass of the adsorbent (mg/g), b and Qo are Langmuir constants related to rate of adsorption and adsorption capacity, respectively.

The R^2 value of obtained to see how the data fitted into Langmuir isotherm model differed between the different activation time adopted in this research work. The Langmuir constants Qo and b were calculated using Eq. (2). One of the important characteristics of the Langmuir isotherm used in ascertaining the nature of the adsorbent is expressed in the form of a dimensionless equilibrium parameter (R_L). It is defined in the form

$$R_L = \frac{1}{1+bCo} \quad (3)$$

Where b is Langmuir constant and Co is the highest hydrocarbon concentration (mg/l)

6.4. FREUNDLICH ISOTHERM MODEL

Unlike the Langmuir isotherm model where the equation varies as a function of the coverage at the surface, the Freundlich isotherm model on the other hand assumes that the surface energy is heterogeneous. The Freundlich model is well known for its logarithmic form, and is given by

$$\log q_e = \log K_F + (1/n)\log Ce \quad (4)$$

where Ce is the equilibrium concentration of the adsorbate (mg/l) q_e is the amount of adsorbate adsorbed per unit mass of the adsorbent (mg/g), K_F and n are Freundlich constants, with n indicating how favourable an adsorption process is and KF (mg/g (l/mg)1/n) denotes the adsorption capacity of the adsorbent. K_F in order was used to define the adsorption or distribution coefficient.

7.0 Results and Discussion

7.1 Adsorption Studies

From the activation test carried out, the values obtained showed that the adsorbent carbonised for two hours was activated while the one carbonised for one hour showed little or no sign of activation. Although it was physically observed that the methyl orange (5mg/l) colour which was used for the activation test was not all removed, a transmission value of 44.4% from 24.4 at 5mg/l indicated that adsorption occurred.

Table 1Methyl orange concentration and transmission after 24 hours activation test

Concentration (mg/l)	Transmission (%)
1.000	76.8
2.000	57.6
2.964	44.4
3.000	43.9
4.000	32.8
5.000	24.7

As shown in Table 1, it was seen that 2.964mg/l of methyl orange was adsorbed by the adsorbent after 24 hours contact time. Furthermore, a linear relation was established between transmission and concentration as a proof of adsorption occurrence in the activation test process. The linear relationship is presented in Figure 1.

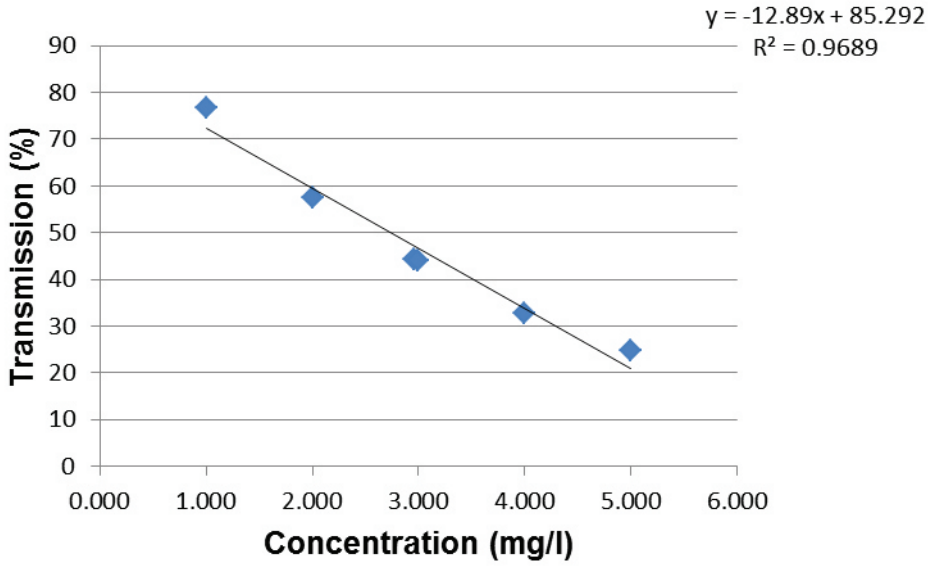

Figure 6 Methyl Orange Calibration Chart

7.2 Adsorption Isotherm

The adsorption isotherm of hydrocarbon contaminated drinking water onto activated carbon (coconut activated with NaCl), are shown in Table 2, Table 3 and Table 4.

Table 2 Weight of carbon and concentration of water sample from column A

Sample	Mass of Carbon(g)	Concentration (mg/l)
A_0		137
A_1	208.77	71
A_2	208.77	53
A_3	208.77	80
A_4	208.77	49
A_5	208.77	76
A_7	208.77	98
A_9	208.77	59
A_{11}	208.77	46
A_{13}	208.77	40

Table 3 Weight of carbon and concentration of water sample from column B

Sample	Mass of Carbon(g)	Concentration (mg/L)
B_0		137
B_1	208.77	88
B_2	208.77	64
B_3	208.77	61
B_4	208.77	63
B_5	208.77	70
B_7	208.77	57
B_9	208.77	46
B_{11}	208.77	45
B_{13}	208.77	39

Table 4 Concentration of water sample from column C

Sample	Concentration (mg/L)
C_0	137
C_1	87
C_2	89
C_3	97
C_4	89
C_5	109
C_7	61
C_9	75
C_{11}	50
C_{13}	56

From the data obtained from the adsorption test, it is possible to obtain valuable information from the process that was adopted in this study. It is possible to conclude that the adsorption is unfavourable given that the control column (made of gravel and coarse sand) also showed signs of adsorption as seen in Table 4 and Figure 2 (Column1 representing 1 hour carbonisation, Column 2 representing 2 hours carbonisation and Colum C representing the control) .

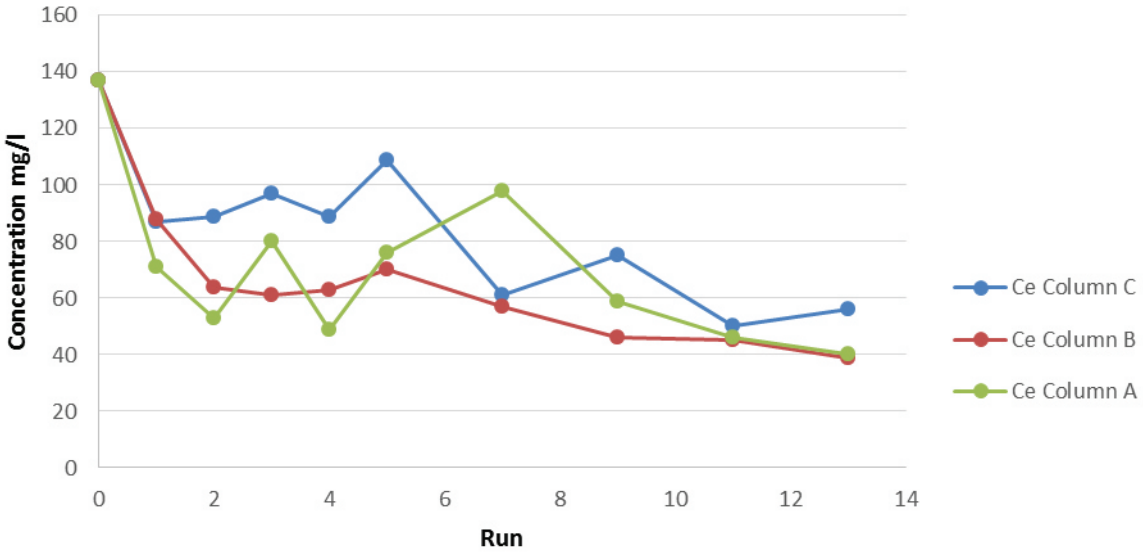

Figure 7 Distribution of concentration from Columns A, B and C

Table 2 and Table 3 represent the data used to carry out the adsorption studies using the Langmuir Isotherm model and the Freundlich Isotherm model.
Table 5 represents the parameters generated using the Langmuir Isotherm model to fit the experimental adsorption data of hydrocarbon contaminated drinking water onto coconut shell activated carbon.

Table 5 Adsorption Calculation Using the Langmuir Isotherm model

Run 1	Ce (Con)	C_e 1	C_e 2	q_e 1	q_e 2	C_e 1/q_e1	C_e 2/q_e2
0	137	137	137				
1	87	88	71	0.235577	0.317308	373.551	223.7576
2	89	64	53	0.350962	0.403846	182.3562	131.2381
3	97	61	80	0.365385	0.274038	166.9474	291.9298
4	89	63	49	0.355769	0.423077	177.0811	115.8182
5	109	70	76	0.322115	0.293269	217.3134	259.1475
7	61	57	98	0.384615	0.1875	148.2	522.6667
9	75	46	59	0.4375	0.375	105.1429	157.3333
11	50	45	46	0.442308	0.4375	101.7391	105.1429

Although a linear relationship was established from the adsorbents used, it can be seen in Figure 7 and Figure 8 that the regression (R^2) values (0.9434 and 0.9232) obtained from both adsorbents was not close to 1. This is contrary to Abkenar et al., 2015 and Zhu et al., 2015 who maintained that R^2 values very close to 1 denotes a more favourable adsorption.

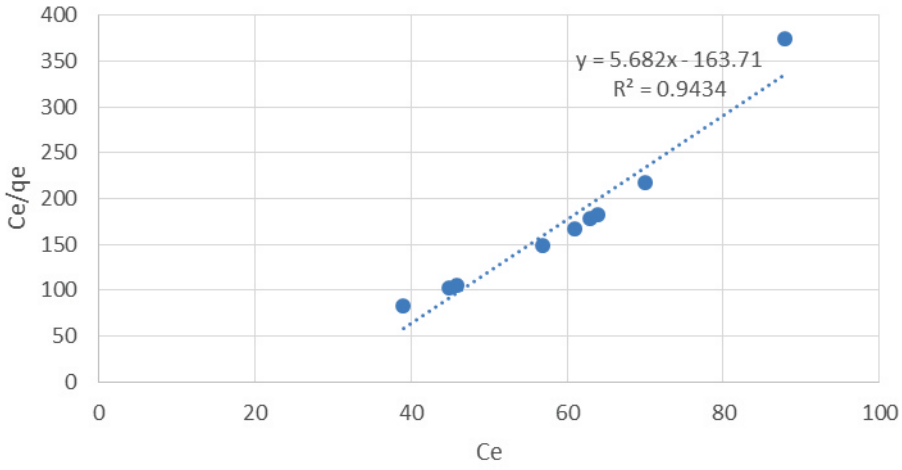

Figure 8 Langmuir Isotherm for 1 hour activated carbon

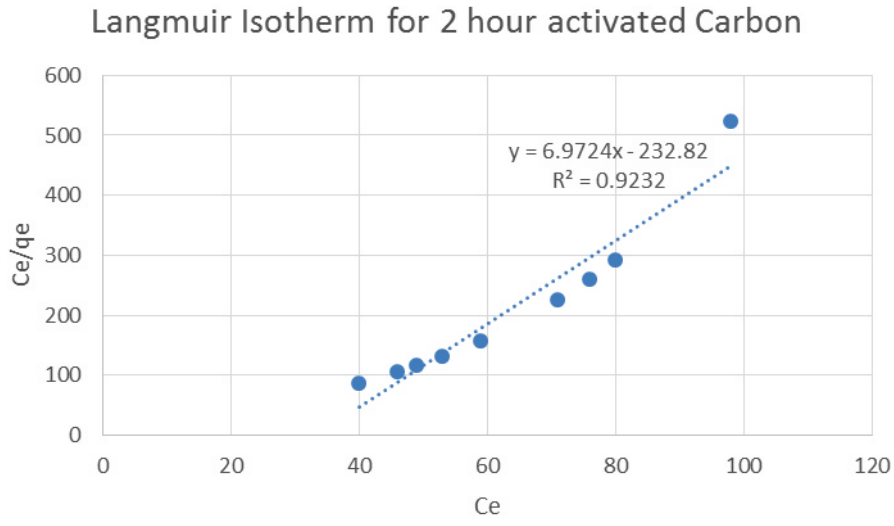

Figure 9 Langmuir model distribution for 2 hours activated carbon sample

Langmuir constants as derived from both adsorbents as shown in Table 6 also show that the adsorption was not a favourable one given that it did not measure up to the condition required using Langmuir constants ;($R_L = 0$) irreversible, (R_L=1) linear, ($R_L > 1$) unfavourable, and ($0 < R_L < 1$) favourable (Mittal et al, 2007)

Table 6 Langmuir Constants

Parameter	2 Hours Activation	1 hour Activation
Qo	0.143	0.176
b	-0.03	-0.034
RL	-0.32	-0.273

Table 7 represents the parameters generated using the Freundlich Isotherm model to fit the experimental adsorption data of hydrocarbon contaminated drinking water onto coconut shell activated carbon.

Table 7 Adsorption Isotherm Calculation Using Freundlich Model

Run	Ce (Con)	C_e 1	C_e 2	q_e 1	q_e 2	Log C_e1	Log C_e2	Log (q_e1)	Log (q_e2)
0	137	137	137						
1	87	88	71	0.235577	0.031731	1.944483	1.851258	-0.627867255	-0.498519399
2	89	64	53	0.350962	0.040385	1.80618	1.724276	-0.454740475	-0.393784049
3	97	61	80	0.365385	0.027404	1.78533	1.90309	-0.437249743	-0.562188479
4	89	63	49	0.355769	0.042308	1.799341	1.690196	-0.448831615	-0.373580663
5	109	70	76	0.322115	0.029327	1.845098	1.880814	-0.491988532	-0.5327335
7	61	57	98	0.384615	0.01875	1.755875	1.991226	-0.414973348	-0.726998728
9	75	46	59	0.4375	0.0375	1.662758	1.770852	-0.359021943	-0.425968732
11	50	45	46	0.442308	0.04375	1.653213	1.662758	-0.354275508	-0.359021943
13	56	39	40	0.471154	0.046635	1.591065	1.60206	-0.326837259	-0.331291601

Similar to the Langmuir Model, data presented in table 7 suggest further upholds the fact that the adsorption of hydrocarbon contaminated drinking onto coconut shell activated with NaCl as carried out in this work as unfavourable. This can be seen in the R^2 value in Figure 5 and Figure 6 which is not very close to 1 as well as the –ve value of n for Freundlich constant in Table 6. The –ve value of n in this case also means that the adsorption shown between the adsorbent and the adsorbate is unfavorable. See Figure 9 and Figure 10 to see the regression values obtained using Freundlich isotherm model.

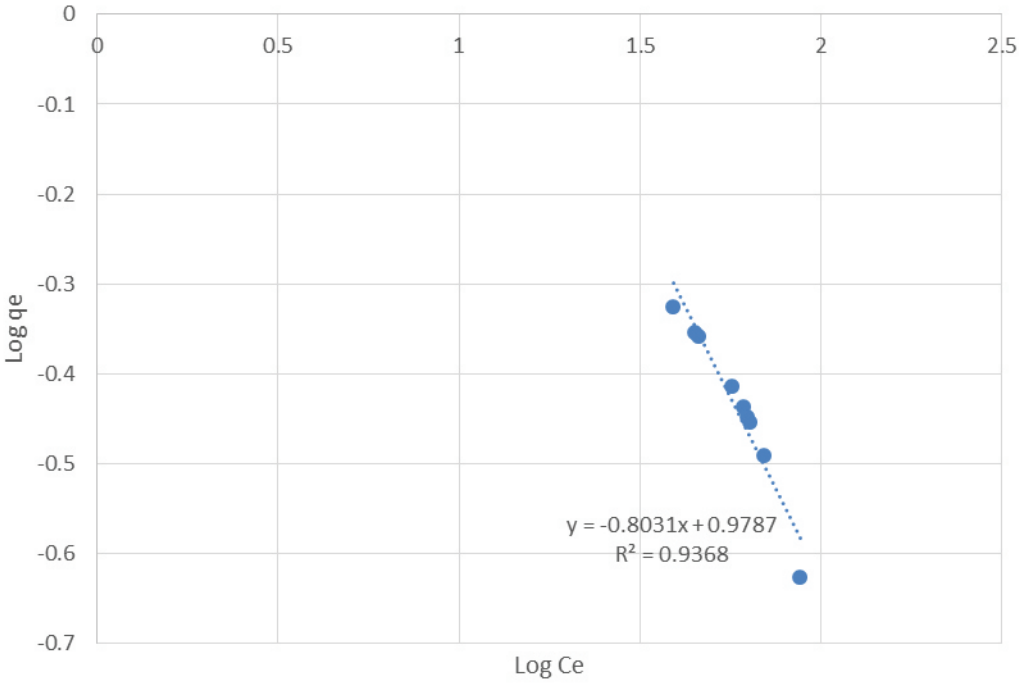

Figure 10 Freundlich model distribution for 1 hour activated carbon sample

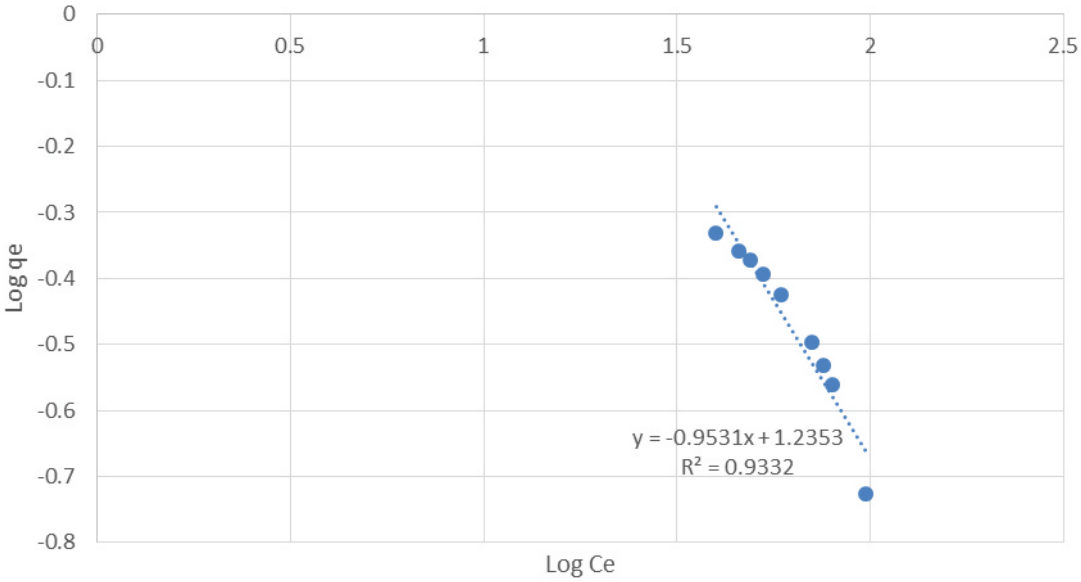

Figure 11 Freundlich model distribution for 2 hours activated carbon sample

Table 8 Freundlich Constants

Parameter	2 Hours Activation	1 hour Activation
n	-1.245	-1.05
K_F	9.51	17.17

From the outcome of the results after analysis of adsorption using Langmuir and Freundlich Isotherm models, it was gathered that all the parameters (R^2 and constants} required to ascertain how favorable the adsorption was did not meet the stipulated standard.

Further analysis suggested that there was more of a physical process as against the anticipated chemical process given that the control column also showed signs of adsorption whereas there was no carbon material present in the control column.

8.0 Conclusion

In this study, coconut shell activated with NaCl after being carbonised at two different temperature (1 hour and 2 Hours) were tested for adsorption of hydrocarbon from drinking water samples. Although one of the carbon material showed sign of activation during the activation test, it was discovered that there more of a physical process than chemical process. This was evident in the fact that the control column made up of sand and coarse sand also showed sign of adsorption during the adsorption test. As a result of this development, none of the isotherm model that was used in the analysis gave a result that was in tandem with existing literatures. What this entails for the body of study moving forward in this area of research, is that a true representation of hydrocarbon contaminated drinking water should be incorporated in future studies while paying attention to other parameters such as contact time and carbonisation time in order to improve the adsorption capacity.

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