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Improvement of organic matter removal in water produced of oilfields using low cost moringa peels as a new green environmental adsorbent

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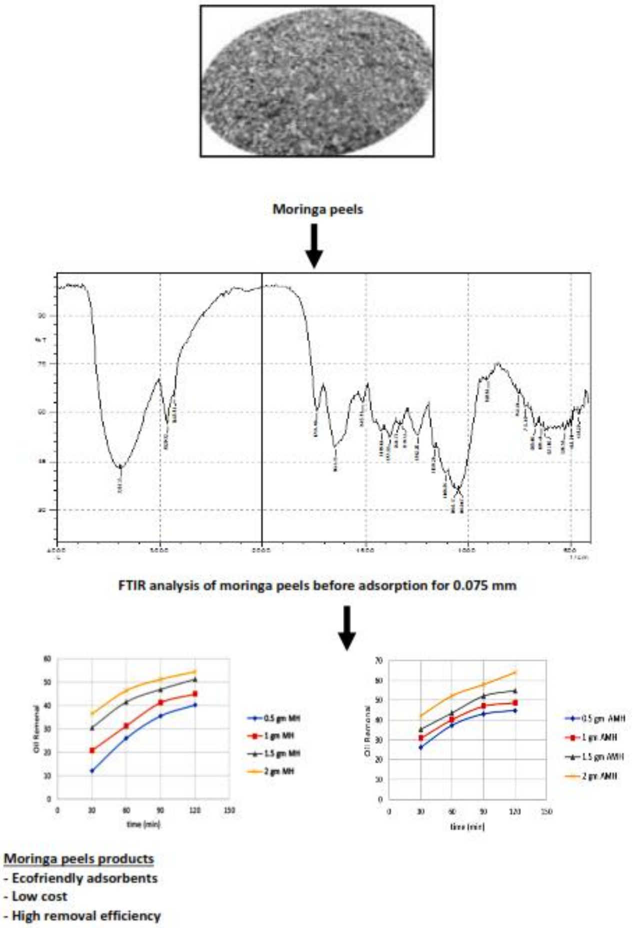
Graphical abstract



Abstract

The present study observed the evaluation of organic content removal from produced water by adsorption onto moringa and activated moringa peels. the removal of the organic content from the real effluent water produced by the oilfields. The proposed treatment process was the adsorption technique using the moringa peels as adsorbing agent. The effect of some parameters such as: adsorbent dose, pH and contact time on adsorption process has been investigated using the batch adsorption method. FTIR spectral examination shows a presence the bands of carboxyl, hydroxyl and amine on the surface of MH. FT-IR device type shimadzu (4000–400 cm^{-1}) was approved out to classify the functional groups and structural in the moringa peels that strength be complicated in the bio-sorption process. The Langmuir isotherm provided a better fit for the adsorption equilibrium data than the Freundlich isotherm. It was observed that organic content adsorption was dependent on pH. The maximum removals of organic content were 65.4 % and 94.1% for moringa and activated moringa peels respectively at pH = 3, 120 min, 1.5 gm dose. The outcome of this study exposes that the moringa and activated moringa peels are ecofriendly adsorbents can suggest for the treatment of produced water. In addition, moringa peels are available in large amount and its treatment technique is very economical.

Keywords: Oilfields, produced water, organic material, moringa peels, adsorption.

1. Introduction

Like most production activities, during crude oil extraction process, water from the reservoir gas and oil production processes generate huge amounts of liquid waste identified by means of produced water (PW) (Boudrahem *et al.*, 2015). PW is the main waste stream produced in oil and gas trades. Produced water comprises of a diverse mixture of organic and inorganic chemicals, cations (e.g. magnesium, calcium and iron), anions (e.g. carbonate and bromide sulfate) and other components such as heavy metals (e.g. barium, uranium, cadmium, chromium and lead) (Li *et al.*, 2005),(Ali A. Hassan *et al.*, 2018). Discharging produced water to environment lead to contaminate soil, surface and underground water (Fakhru'l-Razi *et al.*, 2009).

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Furthermore, release of the produced water with high chemical oxygen demand (COD) value has harmful ecological penalties. However, many countries were implemented a severe guidelines for discharging a least level permissible of their produced water (Aziz and Daud, 2012). The release of huge amounts of contaminated waste water to the environment motivates the researchers to develop different techniques and implementing appropriate treatment skills, capable of eliminating dangerous pollutants present in numerous of oil and gas manufacturing streams (Hernández-Francisco *et al.*, 2017)-(Diya'uddeen *et al.*, 2011). Treatment of these wastes might result in enhanced water quality, water reclaim, oil recovery, defense of downstream amenities and ecological permit obedience (Okiel *et al.*, 2011). PW is treated by numerous physical means earlier release and rules put severe restrictions on levels of pollutants which can be discharged to the sea (Bakke *et al.*, 2013). Presently, properly preserved water can be recycled and used for water inundating. These helpful recycles directly reduces the withdrawal of potable water, an extremely appreciated commodity in numerous areas of the biosphere (Fathy *et al.*, 2017). The elimination of organic residual from oilfield produced water has been attained by numerous processes such as a phase separator (Almarouf *et al.*, 2015), sedimentation (Mohammed and Shakir, 2017), coagulation/flocculation (Amuda and Amoo, 2007), (Khalid M. Mousa and Ali. A. Al-Hasan, 2017), electrocoagulation (Fouad, 2014), flotation(da Silva *et al.*, 2015), filtration and membrane processes (Zoubeik *et al.*, 2017)-(Zsirai *et al.*, 2018), biological operations (Benitez *et al.*, 2001) and chemical processes (Saien and Nejati, 2007)-(Lee and Park, 2013). Most of these approaches suffers from some drawbacks such as the high capital or operative cost and there are difficulties in disposal of remaining organic sludge (Al-Othman *et al.*, 2012). In contrast, the adsorption method has developed one of the most favored methods for the elimination of organic owing to its high competence and low cost. Various agricultural products had straight been used as adsorbent for organic adsorption from waste water which comprised Palm fibers (Abdelwahab *et al.*, 2017), banana peels (El-Din *et al.*, 2017), bentonite (Okiel *et al.*, 2011) soybean hull (Steevensz *et al.*, 2014), olive (Al-Anber and Al-Anber, 2008), barley straw (Ibrahim *et al.*, 2010), cocoa shells (Fiset *et al.*, 2002), tea leaves (Ahsan *et al.*, 2018) and orange peels (AbdurRahman *et al.*, 2013). Activated carbon is showed to be an active adsorbent in waste treatment process. Though, there is a possibility to discover an available and inexpensive sorbents from agro-industrial origin (Ratnamala *et al.*, 2016). Moringa peels are non-toxic usual organic polymer. The tree is usually recognized in the emerging world as medicinal plant, herbal, and a foundation of vegetable oil (Vieira *et al.*, 2010). The aim of this work is to evaluate organic content removal from produced water by adsorption onto moringa and activated moringa peels. The effect of some parameters such as: adsorbent dose, pH and contact time on adsorption process has been considered.

2. Materials and methods

2.1. Preparation of moringa peels

Moringa peels (MH) used in the current study was collected from Iraqi farms. The MH was crumpled and dried in the sunlight for 48 hour and used as a fresh adsorbent. The dried peels were preserved with HCL concentration for 4 hour and then wash methodically with distilled water till it reached neutral pH, then the adsorbent peels were washed with distilled water. AMH used as adsorbent was prepared according to a procedure that described in literature (Bello *et al.*, 2017). The dried MH were carbonized in the department of chemical Laboratory of Al-Muthanna University, Iraqi. 100 gm of the dried moringa peels was heated in a muffle furnace (Mikrotex, Turkey) set at 250 °C for 2 h. Throughout, the procedure, the moringa peels was thermally disintegrated to absorbent materials and hydrocarbon compounds. The activated samples were rinsed with distilled water until the neutral pH and then keep them at room temperature. The washed activated moringa peels dried later in an oven at 110 °C. The carbonized moringa peels were sieved with 0.075 mm mesh sieve (Besmak sieves from 2.36 mm to 0.075 mm). The specific surface area of MH and AMH is estimated Brunauer Emmat and Teller (BET) technique, whereas, the MH and AMH is strongly minded through a micrometric apparent (ASAP, 2010) via nitrogen adsorption at 77 K.

2.2. Experimental procedure

The produced water (PW) was collected from Al-Ahdab oilfield, located in Iraqi. The waste was exposed to filtration to eliminate most of its total solids then reserved at 5 °C, to safeguard that its characteristics of waste water will not be tapering or weathered. The description of produced water is assumed in Table 1.

Table 1. Characteristics of produced water (PW)

Parameter	value
Oil	210 (ppm)
Turbidity	85.64 NTU
pH	6.2
Solution oxygen content	0.053 (ppm)
Specific gravity	0.998
conductivity	70688.64 µs/cm
TDS	64346.24 (ppm)
Viscosity	1.301 m Pa/S
İron	0.36 (ppm)
Sulphate	65.2 (ppm)
TSS	20 ppm
Manganese	2.5 ppm
Chrome	0.15 ppm

The organic adsorption tests were performed by a batch technique. The PW was agitated by responding shaker at room temperature (25 ± 2 °C) with 130 rpm. Samples were occupied at limited intervals and then filtered with Whatman No. 2041 filter paper (150 mm). The filtrates were examined for remaining organic content in the PW. The influence of the solution pH on the organic sorption performance was examined in the similar way but that the initial pH of the solutions was adjusted to values varying from 2.0 to 10.0 with the adding of either 0.1 M NaOH or 0.1 M H_2SO_4, the pH values were also recorded after the moringa husks-organic suspensions had reached equilibrium. Different amounts of MH and AMH in the range (0.5–2 mg) were added to PW solution. The quantity of PW at equilibrium (q_e) was estimated using an assumed mass balance equation as below (Ali Saleh Jafer *et al.*, 2019):

$$q_e = V(C_o - C_e)/M \qquad (1)$$

where q_e (mg/g) is the total of organic in PW per mass unit of watermelon absorbent at certain time t, V is the solution volume (cm^3), M is the mass of adsorbent (mg) and C_0 and C_e (ppm) are the original and at time t concentration of PW correspondingly. The oil elimination by MH and AHM was estimated at each equilibration by the following equation as:

$$\text{Adsorption}(\%) = (C_0 - C_t)/C_0 \times 100 \qquad (2)$$

2.3. Chemical analysis and tools

The organic content of oilfield produced water was monitored by a UV-spectra meter (UV-1800 Shimadzu, Japan) spectrophotometer associated to a PC at maximum absorption wavelength (312 nm). The turbidity was estimated using turbid meter (Lovibond, SN 10/1471, and Germany). The pH measurements were performed by pH meter (Model 2906, Jenway LTd, UK). All chemicals used in this study are an analytical grade, H_2SO_4 (98% purity), NaCL and NaOH (98% purity) were purchased from India. Oil was Tested using a UV–spectra meter, where NaCl (0.25 mg) was added to 50 mL PW in the unraveling funnel with the intention of disruption the emulsion of organic. 5 ml of CCL_4 was supplementary and shadowed by vigorous shaking for 1 min. After 25 min, once the solution separated into two separate layers, the inferior (organic) layer was occupied for the absorbance dimension, and after the calibration curve, organic content in PW was found. The characterization of CCL_4 is given in Table 2.

Table 2. Properties of CCl_4 solvent

Property	Value
Physical state	Liquid
Molecular weight	153.82
Color	Colorless
Odor	Aromatic, sweet
Melting point	-23 °C
Boiling point	76.5 °C
Formula	CCl_4
Purity	99%

3. Results and discussion

3.1. FTIR analysis

The natural moringa peels functional groups characterized using FTIR measurements with spectra in the variety of 4000–400 cm^{-1} as shown in Figure 1. FTIR spectral examination shows a presence the bands of carboxyl, hydroxyl and amine on the surface of MH. FT-IR device type shimadzu (4000–400 cm^{-1}) was approved out to classify the functional groups and structural in the moringa peels that strength be complicated in the bio-sorption process. The comprehensive band at 3381.33 cm^{-1} in moringa peels is credited owing to hydroxyl (–OH) stretching or amine (–NH_2) widening of polymeric compounds. This band seemed in the inferior region 2920.32–2852.81 cm^{-1} in the FTIR spectra of MH and it is credited to the attendance of the C-H bond. The C O widening of moringa peels by away at 1735.99 cm^{-1} (Kebede *et al.*, 2019). The C O widening removed to higher incidence as a result of participation of carboxyl (–C O) group during adsorption process of the organic content with MH. 1510.31 cm^{-1} indicates to the aromatic rings, while 1419.66 and 1377.22 cm^{-1} related with the C-O of phenols and –CH_3 correspondingly. Then the bands less than 800 cm^{-1} are finger print zone of phosphate and sulphur functional groups (Ali Saleh Jafer *et al.*, 2019).

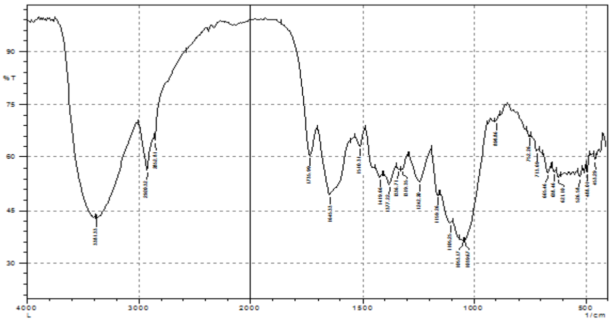

Figure 1. FTIR analysis of moringa peels before adsorption for 0.075 mm.

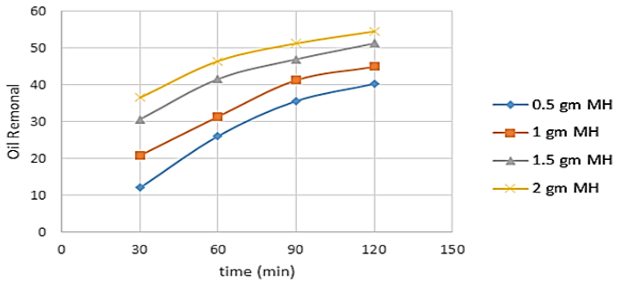

Figure 2. Effect of dosage on organic removal in 0.075 mm moringa peels size, pH = 6.2, room temperature.

3.2. Effect of moringa peels doses

Wide range of dosages has been used 0.5–2 mg to investigate the removal of organic content from PW solution by MH and AHM. In these series of trials, the adsorbent dosages effect on the produced water by the moringa and activated moringa peels are exposed in Figures 2 and 3. It is obvious, the increase of adsorbent dosages has a significant impact on the process performance. Whereas, the raised of MH and AMH dosages from 0.5 to 2 mg led to improve the removal percent from 40.2%, 44.5 % to 54.5%, and 64 % respectively. This may be attributed to the fact that availability of exchangeable sites for the pollutants increases with an increase in the adsorbent dosage. However, the increased of MH and AMH doses from 1.5 to 2 mg didn't have a significant effect on the removal efficiency of organic pollutant for both adsorbents (MH and AMH) used. The acceptance of PW that (organic content) augmented with upsurge in the adsorbent (HM and AHM) doses can be credited to the increase of surface area, active sites, pores, the number of unsaturated places and the adsorption places (Gupta *et al.*, 2016).

A similar tendency was stated by Rehab M (2016) in his studies about removal of heavy metal from aqueous solution by peanut hull (Ali *et al.*, 2016). Furthermore, the removal efficiency of organic pollutant from PW when use the AHM adsorbent was higher than that of HM adsorbent. This marked is attributed to the AHM adsorbent has a specific surface area (913 m^2/g) which higher than that of HM (713 m^2/g).

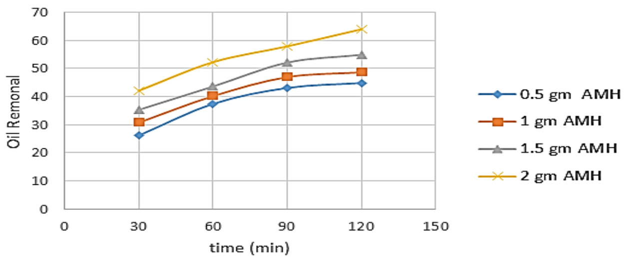

Figure 3. Effect of dosage on organic removal in 0.075 mm activated moringa peels size, pH = 6.2, room temperature.

3.3. Reaction time effect

Contact time considered one of the most significant factors during adsorption process. It is well known, the removal rate of the pollutant and the adsorbent efficiency are increased with the increase of the contact time. During treatment process, the MH and AMH surface is continuously loaded by the pollutant molecules which lead to saturate the active sites of the adsorbent. After saturation, it is difficult for the adsorbent to uptake any more organic pollutant from the PW. The contact time effect on the removal of organic pollutant by MH and AMH is observed in Figures 4 and 5. The results show the quantity of adsorbed organic pollutant is augmented by the beginning of the contact times. However, the adsorption rate is increased with contact time until 120 min, later the increase in contact time has a limited effect on organic pollutant removal. This may be attributed to the huge free active sites are obtainable at an early stage of treatment then these available sites are reduced as result of organic pollutant adsorption (Mohammed and Baytak, 2016). Subsequently attainment the plateaus, the equilibrium are attained about 35 min at 2 gm adsorbed organic. All the tests are approved out through 2 h of contact time to find equilibrium at the solid/liquid interface. It is obvious that there is no change in organic removal when the time is lengthy (Hosny *et al.*, 2016) and (Haider T. Naeem and Ali A. Hassan, 2018).

3.4. pH effect

Solution pH is an important variable to consider during the adsorption treatment process**.** The adsorption of organic content from PW aqueous solutions on the MH and AMH were investigated in the pH range of 2–10. Figures 6 and 7 demonstrate the effect of pH on the removal of organic content from PW aqueous solutions. The result reveals that adsorption process is favored the acidic medium solution. The removal efficiency of organic pollutant increased as a result of pH decreasing. While organic exclusion was 83.2%, 94% at pH 2 and 20.7%, 43% for HM and AHM at pH 10. This result is in agreement with previous reported works, where similar trend was stated for adsorption of organic content on moringa peels waste (Dan and Chattree, 2018) and (George *et al.*, 2016). The surface of adsorbents was negatively charged as the pH solution increased from 2.0 to 6.0. The functional groups of the HM and AHM were more deprotonated and therefore it is available for the organic content. Reduction in bio adsorption harvest at higher pH (pH > 6) is not only connected the formation of solvable hydroxylated complexes of the organic compounds (Munagapati *et al.*, 2010).

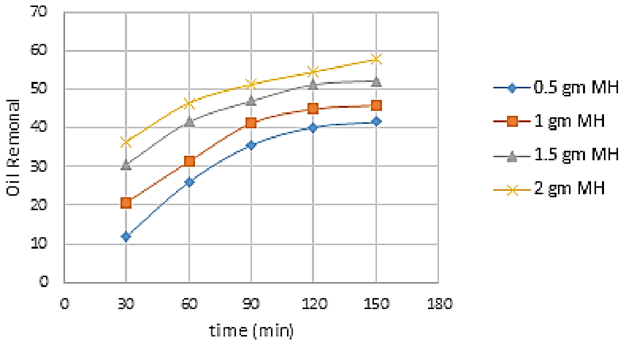

Figure 4. Effect of contact time on organic removal in 0.075 mm moringa peels size, pH = 6.2, room temperature.

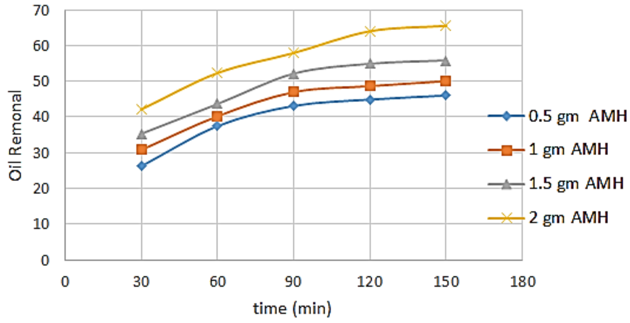

Figure 5. Effect of contact time on organic removal in 0.075 mm activated moringa peels size, pH = 6.2, room temperature.

3.5. Adsorption isotherms

Reactions between solutes and the surfaces of solids play a vital character in the waste treatment chemistry. Adsorption equilibria provide fundamental physicochemical information to evaluate the applicability of the adsorption process. The adsorption isotherms exemplify the relations between equilibrium concentrations of adsorbate in the solid phase and in the liquid phase at constant temperature. The delivery of contaminant such as organic between the adsorbent and the PW solution below equilibrium conditions is significant in sympathetic the capacity of the adsorbent for the organic elimination (Ardejani *et al.*, 2007) and (Ali Saleh Jafer *et al.*, 2019). The relationship between the quantities of the oil field pollutant adsorbed and the concentration at equilibrium was investigated using the linear form of the Langmuir and Freundlich isotherms, the most commonly used adsorption isotherm models (Yu *et al.*, 2016) and (El Messaoudi *et al.*, 2016).

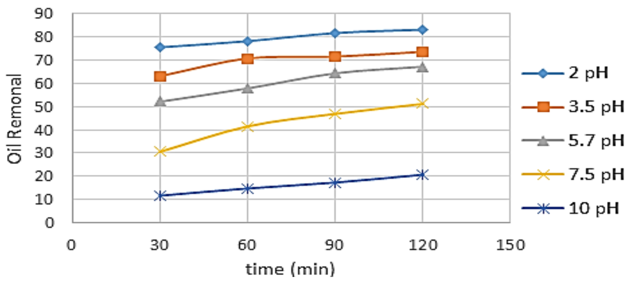

Figure 6. Effect of pH on organic removal in 0.075 mm moringa peels size, 1.5 gm dosage, room temperature, and 120 min contact time.

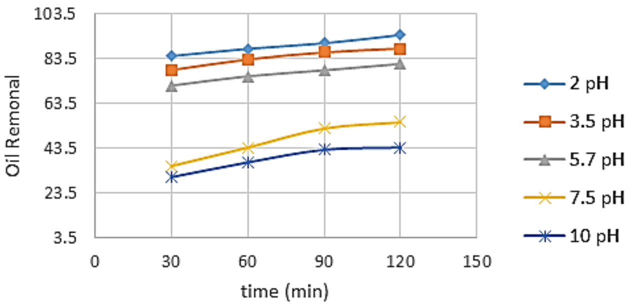

Figure 7. Effect of pH on organic removal in 0.075 mm activated moringa peels size, 1.5 g dosage, room temperature, and 120 min contact time.

Langmuir isotherm:

$$q_e = (q_m K_L C_e)/(1+K_L C_e) \tag{3}$$

Freundlich isotherm:

$$q_e = K_F (C_e)^{1/n} \tag{4}$$

where, q_e (mg/g) is the quantity of organic content per unit weight of adsorbent at equilibrium concentration, Ce (ppm). The q_m (mg/g) and k_L (L/mg) are the Langmuir coefficients connected to the maximum monolayer capacity and energy of adsorption, correspondingly. The K_F and 1/n are Freundlich coefficients connected to adsorption capacity and intensity of adsorption correspondingly. Langmuir isotherm shoulders a monolayer adsorption surface deprived of slightly lateral contact between adsorbed molecules (Anirudhan and Sreekumari, 2011). The adsorption data for HM and AHM were fitted into Langmuir and Freundlich isotherm equations. The maximum adsorption on moringa peels and activated moringa peels were found to be 22.88 and 26.88 mg/g. It seems that the Langmuir model provided a better fits than Freundlich model over the experimental range with correlation coefficient ($R^2 > 0.99$). This result is similar to Aseel M. *et al.*, (Aljeboree *et al.*, 2017). Figures 8 and 9 show the Langmuir model for HM and AHM respectively. The Freundlich model correlation coefficient is lower than that of Langmuir model with R^2 (0.9545). The limits of K_F and 1/n showed penetrating alteration at higher temperatures. The standards of 1/n (0.1<1/n < 1) provide a promising adsorption of organic compounds at experimental conditions as shown in Figures 10 and 11.

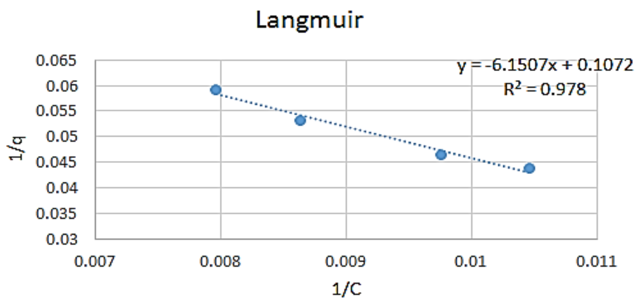

Figure 8. Langmuir isotherm plot for moringa peels adsorption of organic content on produced water sample.

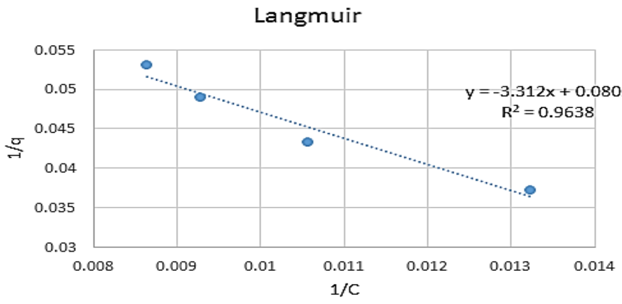

Figure 9. Langmuir isotherm plot for activated moringa peels adsorption of organic content on produced water sample.

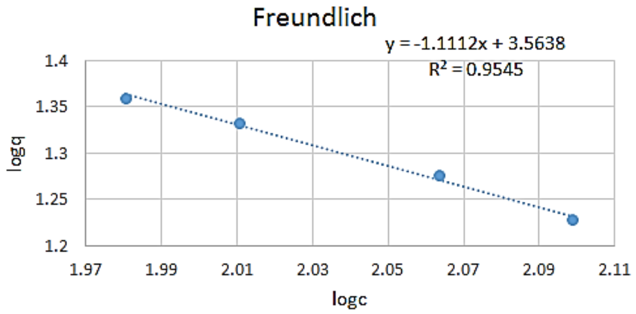

Figure 10. Freundlich isotherm plot for moringa peels adsorption of organic content on produced water sample.

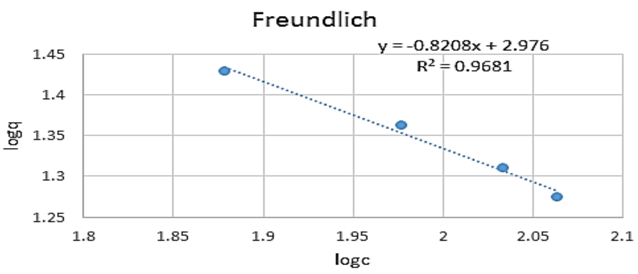

Figure 11. Freundlich isotherm plot for activated moringa peels adsorption of organic content on produced water sample.

4. Conclusions

Activated moringa peels were found to have a good adsorbent for the elimination of organic content from oilfield produced water. Results show the sorption was depended on solution pH, adsorbent dose and contact time of the adsorbent. The quantity of adsorption capacity (mg/g) was improved with increase the adsorbent dosage. Adsorption of organic content follows the Langmuir isotherm. This work showed that the moringa and activated moringa peels are ecofriendly adsorbents can suggest for the treatment of produced water. In addition, moringa peels are available in large amount and its treatment technique is very economical.

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