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Application of eggplant peels powder for the removal of oil from produced water

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ABSTRACT

The removal efficiency of oil from simulated samples of produced water (SPW) was studied using a low-cost adsorbent eggplant peel powder (EPP). The effects of pH, adsorbent dosage, contact time, and temperature on the removal efficiency were investigated. The optimum conditions for maximum removal of oil from produced water (PW) are found to be: pH 10.00, adsorbent dosage = 1.75 g/L, contact time = 40 min, and temperature = 55˚C. The results showed that the removal efficiency increases with increasing adsorbent dosage, salinity, and pH. The maximum removal efficiency of oil on EEP, at the optimum condition, is greater than 90% by weight. The study showed that EPP is a fast and excellent adsorbent for this oil removal. The crude oil adsorption on EPP is found to follow Langmuir adsorption isotherm, whereas the adsorption kinetics is best described by Pseudo-second-order kinetic model.

Keywords: Produced water; Eggplant peel; Biosorbent; Equilibrium isotherms; Kinetic model; Crude oil

1. Introduction

The Arabian Gulf is known to be one of the largest production areas in the world for petroleum products. In addition to its presence with oil and gas in the underground natural reservoirs, water is a common byproduct of the drilling and refinery processes. Moreover, the use of surfactant materials is very common during oil extraction and separation processes leading to the release of highly contaminated water with organic and inorganic matter that do not meet the EPA specification for consumable water and therefore termed as produced water (PW) [1,2]. Alkaline surfactants and polymers are usually added to enhance oil recovery where they play a key role in lowering the surface tension at the interface between crude oil and water resulting in lower zeta potential

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on the droplets surface. Oil industry is challenged to develop treatment methods to manage the PW. The amount of PW produced during the drilling and separation processes of a field of oil is expected to be equivalent to about 80% of the volume of the entire field from which the oil was drilled [3]. The PW have negative impact on the environment leading to the contamination of the surrounding water and soil. Different techniques are used to manage and treat PW including filtration, flocculation, coagulation, adsorption, and de-emulsification [4].

Physical, chemical and biological procedures have been employed for PW treatment [1,3]. The most common techniques for PW treatment includes gravity separator, corrugated plate separator, induced gas floatation, hydroclone, coalescers, media and membrane filters, adsorption, extraction, and centrifugation [5]. Arthur et al. presented a technical summary of the technologies used for the treatment of the oil and gas PW [6]. The authors classified the technologies based on the type of treatment into de-oiling, disinfection, desalination, membrane treatment, and other miscellaneous treatment. In that report, they described the advantages and disadvantages of each technology and the ranges of field applicability. Most of the available processes associated with PW treatment are costly and not environment-friendly [7]; therefore, they need to be tackled through more innovative economic and environmental approaches [8]. Among all techniques, adsorption seems to be the most efficient in lowering the amount of oil in water to negligible levels [9]. Although activated carbon is one of the promising adsorbents, its use is challenged by its high initial and regenerative costs. Several other naturally occurring adsorbents were utilized or evaluated to separate oil from PW [10–24]. Many of them were found to be promising including eggshells [15], modified barely straw, banana peels [13], and surface-modified ball media fibers [25].

Activated carbon prepared from eggplant peel was found to be an effective adsorbent for the removal of heavy metals and organic compounds from water [26–28]. Eggplants are produced on large scale in India, Turkey, Iran, Spain, Tunisia, Afghanistan, Greece, Japan, United States of America, China, France, Armenia, Cyprus, Egypt, Italy, Syria, Lebanon, and Palestine. India produced about 33,415 tons during the year of 2009–2010 [29]. It is worth mentioning that the presence of surfactants in PW may results in modifying the zeta potential of adsorbents and hence likely to change adsorption capacity toward oil. Since the surfactants have hydrophobic part, which will solubilize the nonpolar components of the oil that are extracted by hexane, it is likely that their removal will be enhanced. This concern should be considered during the application of these adsorbent in real samples.

The objective of this work is to investigate the ability of desiccated eggplant peel powder (EPP) in removing crude oil and other pollutants from PW. Furthermore, the adsorption mechanism and adsorption isotherms are studied.

2. Experimental

2.1. Materials and instrumentation

A crude oil sample was obtained from Abu Dhabi National Oil Company (ADNOC), UAE. *n*-Hexane (95% pure) was purchased from J.T. Baker. Double distilled water (Water Still Aquatron A4000D, UK) was used for all preparation and analysis steps. The eggplant samples were purchased from a Dubai (UAE) local market and their skin were peeled to be used as biosorbent material. A fluidized bed dryer was used to dehydrate eggplant peels, while a high-precision oven equipped with a vacuum pump was employed to dehydrate the powdered sorbent (Model WOV-30, DAIHAN Scientific Co., Ltd, Korea). A digital reciprocating shaker was used for adsorption studies and the temperature was regulated using hotplate cum magnetic stirrer (Model MSH-20D, DIHAN Scientific Company, Korea). The homogeneity of the oil/water suspension was attained using a mechanical stirring source (Model MSH-20D, DIHAN Scientific Company, Korea). Oil analysis was done using spectrophotometric technique (HACH DR-5000). pH measurements were made using a pH meter (model 3320, JENNWAY Ltd, UK). The pH was adjusted to the desired values using 0.1 M HCl or 0.1 M NaOH. Particle size distribution was obtained using mechanical sieves of mesh sizes in the 150–500 microns range (Stainless steel; Aperture 150–500 μm; Pascal Engineering Company, UK). The surface of the sorbent material was evaluated before and after sorption of oil using scanning electron microscopy (SEM: TESCAN VEGA.3-LMU, USA). The high-resolution SEM images were used to explore the changes in the chemical nature and the topographic features of the surface of the sorbent material as a result of its interaction with oil. In addition, elemental analysis of the relative carbon content on the surface was measured using energy dispersive X-ray spectroscopy (EDAX: INCAX-ACT, Oxford Instruments, UK).

2.2. Biosorbent preparation

To remove stains and impurities form the eggplant peels, the samples were washed with double distilled water followed by air drying for 24 h. The air-dried samples were then chopped down into particle sizes in the range of 2–3 mm. The produced adsorbent particles were subjected to fluidized bed drier at a temperature of 60˚C. The particles were then treated with *n*-hexane for 2 h to remove hydrophobic dissolved organic matter and to decolorize the peel. The sorbent particles were then dried, washed with deionized water, and then dried again followed by further grinding and sieving to obtain samples of particle size in the 150–500 μm. The produced powdered samples, labeled as EPP, was then washed and dried at a temperature of 80˚C in a vacuum oven for 24 h. The final product was stored in moisture-free glass containers at ambient temperature.

2.3. Sorption experiments

To determine the optimum contact time for the sorption process of oil on EPP powders, simulated produced water (SPW) samples were used [13]. The SPW solutions were prepared using eight oil-in-water concentrations in the range of 200–1,600 mg/L. A total volume of 150 mL of each solution was transferred into separate 200 ml conical flask where 1.5 g of EPP was added to each flask. The pH of each sample was adjusted to 9.5 using 0.1 M HCl and NaOH solutions. The suspensions were mixed at 120 rpm. Five milliliter was collected from each suspension at defined time intervals, filtered and extracted with 150 ml *n*-hexane. The absorbance of *n*-hexane fraction was measured at 450 nm and the concentration of oil was determined using a calibration curve [13]. Three replicates for each set in the experiment were done to assure reproducibility and conduct statistical analysis.

2.4. Effect of pH, salinity, temperature, and EPP dosage

The impacts of pH, salinity, temperature, and EPP dosage on the efficiency of oil removal were examined. The pH was varied in the range of 0.1–13.7 at fixed EPP dosage of 1.75 g/L, oil concentration of 1,200 mg/L and contact time of 40 min. The effect of salinity on the sorption process was evaluated using NaCl solutions of concentrations in the 100–2000 mg/L range. The pH was adjusted to 9.5 while maintaining the other parameters constant as given above. The temperature effect and the EPP dosage were studied separately in the 25–70˚C and 0.33–2.64 g/L ranges while keeping all other experimental conditions constant as described above.

3. Results and discussion

3.1. Sorbent characterization

3.1.1. Scanning electron microscope

The surface of EPP treated with *n*-hexane was examined using SEM before and after oil sorption. The SEM images displayed in Fig. 1(a) and (b) reveals that oil is adsorbed on the surface initially followed by filling the spongy EPP surface structure. The energy dispersive X-ray spectrometry analysis results showed the noticeable increase in the carbon content on the surface of the EPP sample after oil adsorption; therefore, supporting the physical conclusion revealed by the SEM images.

3.2. Adsorption isotherms

The effect of contact time on oil removal efficiency was studied at initial pH of 6.5 ± 0.2, temperature of 25 ± 2˚C, and adsorbent dosage of 1.75 g/L. The removal efficiency increased with time as shown in Fig. 2. It is clear that the maximum removal capacity is achieved after 40 min. It is important to note that the EPP removal efficiency for oil decreased with the increase in oil concentrations for a given time interval. Based on the results, the oil removal efficiency is expected to decrease from 95.71 to 77.1% when the concentration of oil is increased from 400 to 1,600 mg/L for a fixed time interval of 120 min. Such trend might be associated with the variation in the interfacial tension within SPW and at the surface of the EPP at high concentrations.

3.3. Effect of pH

Variation in the pH of the SPW solution is expected to significantly influence the sorption process of oil on EPP. The change in the pH of the solution dictates its acidic or the basic nature which results in changing the surface characteristics of the biosorbent and its reaction/adsorption sites [30,31]. Therefore, pH is considered as a critical parameter in crude oil removal process, which is expected to be a critical factor when EPP is used. The effect of pH on the sorption efficiency is shown in Fig. 3. Initially, the efficiency seems to decrease with the increase in the pH of the SPW from 2 to 4; however, the trend reverse in the pH 4–12 range where the EPP oil removal efficiency starts to increase linearly with pH. EPP reaches its maximum oil removal efficiency at pH 12 followed by slight decrease at further higher pH values. The results reveal the presence of strong electrostatic forces of attraction between the oil molecules and the EPP-

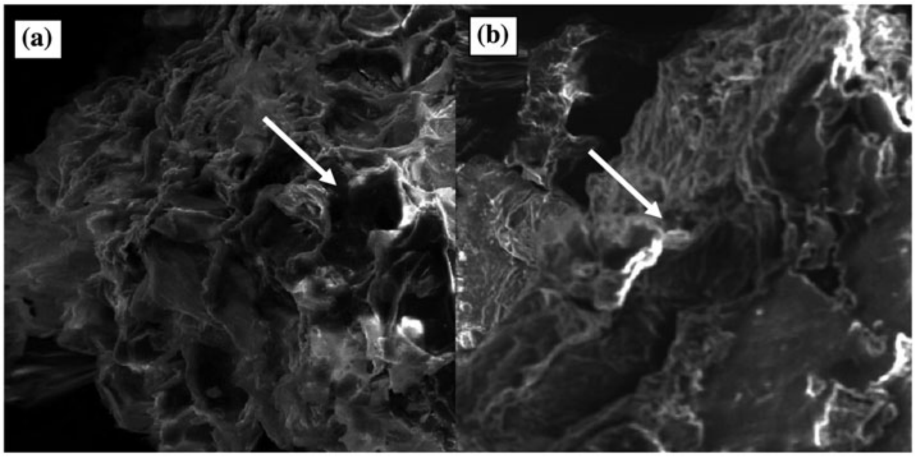

Fig. 1. SEM of EPP (a) before and (b) after sorption of oil.

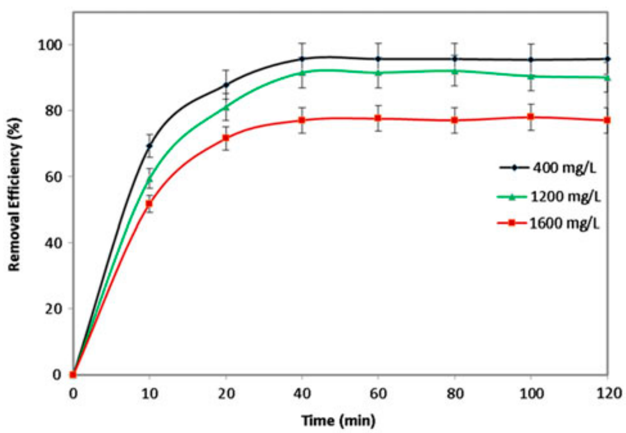

Fig. 2. Effect of contact time and initial oil concentration on the EPP oil removal efficiency.

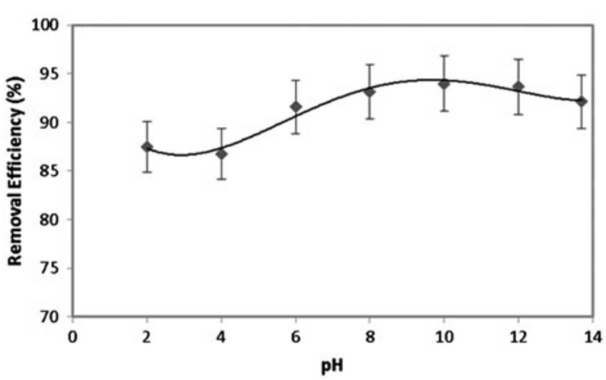

Fig. 3. Effect of pH on adsorption of crude oil over EPP.

protonated surface sites resulting in an increase in oil removal capability. It seems the surface sites exhibit an amphoteric behavior at pH of 4.0; consequently, the oil removal efficiency is lowered. Two factors seem to be accountable for the increase in efficiency at higher pHs. At higher pH values, flocking phenomena seems to occur at the surface of the EPP sorbent where the oil and water emulsion starts to form small oil droplets that will deposit at the surface of the EPP sorbent; therefore, increasing its efficiency for oil removal [18]. The second factor is associated with the behavior of water itself at the surface of the EPP adsorbent. At high pH values, the surface of the EPP is expected to be negatively charged imposing repulsion forces on water molecules. Such behavior would result in the separation of oil at the EPP surface increasing the removal efficiency to its maximum at pH 12 [32]. At higher pH values, the surface of the biosorbent starts to repel the oil molecules and hence result in the observed decrease in the efficiency at pH 13.7. Based on the statistical error analysis and the simulated trend shown in Fig. 3, it is concluded that oil removal sorption study is best measured in the pH range of 10–12.

3.4. Effect of salinity

The effect of the salinity of the PW on oil removal was studied. Several saline 1,200 mg/L SPW solution was prepared with different NaCl concentrations that ranged between 0 and 2000 mg/L. The effect of salinity was studied at the optimized experimental conditions. The results are shown in Fig. 4. The oil removal efficiency for EPP increases with the salinity reaching its maximum removal capacity of 95.5% when the NaCl concentration is 2,000 mg/L. The oil solubility is known to vary inversely with the amount of NaCl in

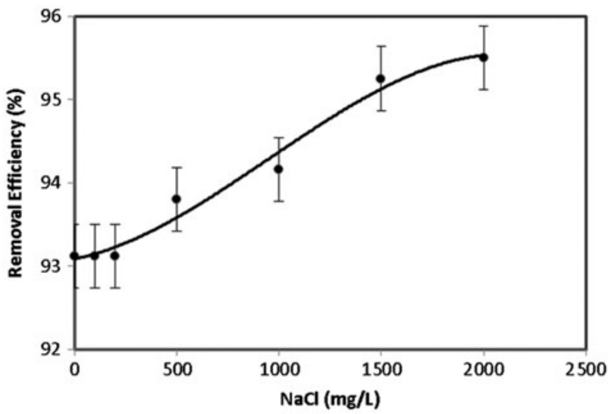

Fig. 4. Effect of salinity on the crude oil removal efficiency.

SPW [33]. As far as the reaction feasibility is concerned, salinity casts negligible impact on experimental procedure, thus it is not a critical experimental factor to be considered.

3.5. Effect of temperature

Temperature is expected to be another essential parameters in the oil removal processes. The effect of temperature was studied in the temperature range of 25–70˚C. As the temperature increases from 25 to 50˚C, the amount of oil removed by the SPP increased by about 4% (Fig. 5). The oil removal efficiency starts to slightly decrease with further increase in temperature. The oil viscosity is known to vary inversely with temperature. As the oil gets thinner due to increase in temperature, the oil molecules start to diffuse into the pores within the EPP surface. The mass transfer coefficient as well as the diffusion rate starts to increase with decrease in viscosity [17,34]. Such decrease in viscosity would increase the free energy of the oil molecules in SPW; therefore, possess an increase in their random motion. The interaction between the oil molecules and the EPP surface sites become more probable leading to the greater oil removal efficiency. The oil removal phenomenon is being facilitated within optimum temperature range while above 60˚C exothermic process is the controlling parameter for adsorption. The optimum temperature observed for oil removal from SPW is room temperature because no prominent benefits are seen at higher temperatures, thus making it the most suitable process in the context of cost and expenditures.

3.6. Effect of biosorbent concentration

The effect of the EPP biosorbent dosage on oil removal efficiency was studied in the 0.05–2.5 g/L range. The results are presented in Fig. 6 showing an increase in the oil removal capability with EPP dosage reaching its maximum efficiency of 95.13% at 1.75 g/L beyond which no major change is observed. The results are expected since the adsorption process should be enhanced due to the increase in the mass transfer factor from solution into the EPP surface.

3.7. Adsorption isotherms models

The relationship between the amounts of oil adsorbed per unit mass of the EPP sorbent can be represented in terms of an adsorption isotherm generated at the optimum experimental conditions defined in the previous sections. In this study, the concentration of oil in SPW was varied while the amount of the EPP sorbent was kept constant. The experimental data were fitted to the Langmuir (Eq. (1)), Freundlich (Eq. (2)), and Temkin (Eq. (3)) adsorption isotherms equations:

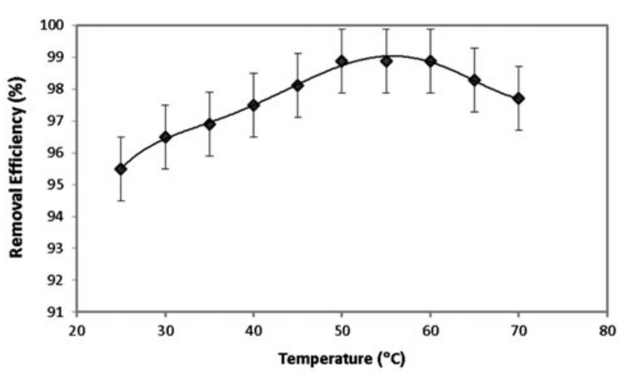

Fig. 5. Effect of temperature on the crude oil removal efficiency.

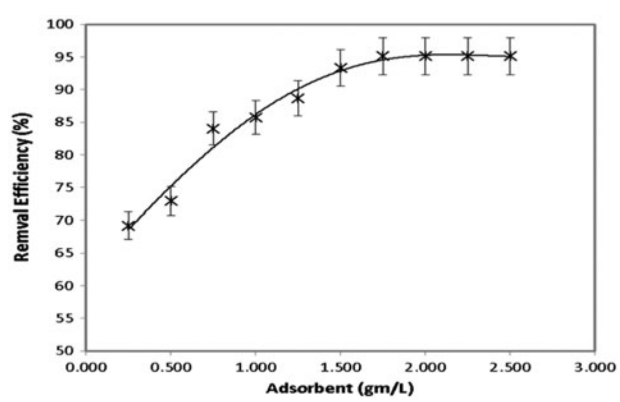

Fig. 6. Effect of adsorbent dosage on the removal efficiency of oil.

$$\frac{C_e}{q_e} = \frac{1}{k_a q_m} + \frac{C_e}{q_m} \tag{1}$$

$$\log q_e = \log k_f + \frac{1}{n} \log C_e \tag{2}$$

$$q_e = B \log k_t + B \log C_e \tag{3}$$

where q_e represents the amount of oil adsorbed at equilibrium per unit weight of adsorbent; q_m denotes the monolayer adsorption capacity; C_e expresses the equilibrium concentration; k_a is the Langmuir adsorption equilibrium constant; n and k_f are the Freundlich constants and B is used for Temkin equilibrium adsorption constant.

The regression coefficients (R^2) were calculated for the three models using a logarithmic plotting. The R^2 values were found to be 0.997, 0.916, and 0.963 for Langmuir, Freundlich, and Temkin isotherms, respectively (Fig. 7). An overview representing the isotherms adsorption feasibilities is given in Table 1. It is clear that data fits well using Langmuir adsorption model.

3.8. Analysis of adsorption kinetics

The kinetics of the adsorption of crude oil on EPP was fitted to both pseudo-first-order and pseudo-second-order reaction models [3]. Eqs. (4) and (5) represent the linear representation of both equations:

$$\ln(q_e - q_t) = -k_I t + \ln q_e \tag{4}$$

$$\frac{t}{q_t} = \frac{t}{q_e} + \frac{1}{k_{II}(q_e)^2} \tag{5}$$

where q_t (mg/g) represents the instantaneous amount of oil adsorbed for a given time interval; q_e represents the amount of oil adsorbed at equilibrium phase; k_I is pseudo-first-order rate constant (1/h), and k_{II} is the pseudo-second-order rate constant (g/mg h).

The regression coefficients for the two kinetic models (0.997 and 0.947 for pseudo-first-order and pseudo-second-order models, respectively) reveal that the adsorption kinetics follows pseudo-second-order model (Fig. 8). The adsorption kinetics constants are listed in Table 2. It is clear that $q_{e,cal}$ (833.34 mg/g) calculated using pseudo-second-order is excellent agreement with the experimental value (715.89 mg/g) as shown in Fig. 9.

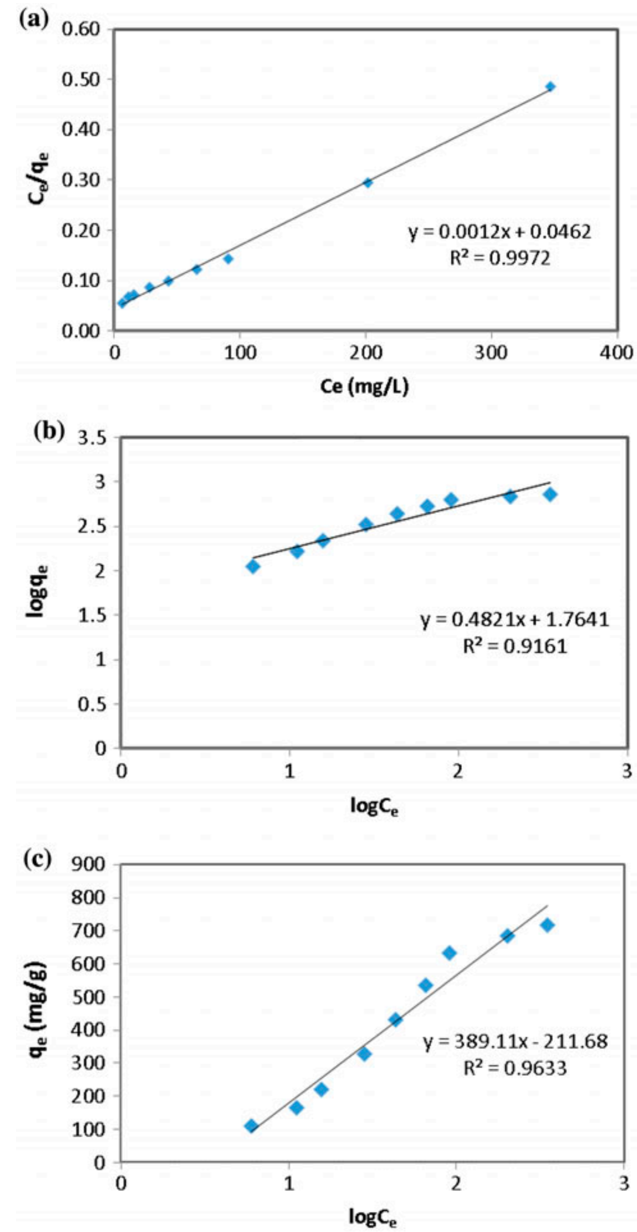

Fig. 7. Adsorption isotherms for the removal of oil from PW using EPP: (a) Langmuir, (b) Freundlich, and (c) Temkin.

3.9. Desorption studies

The crude oil desorption from EPP sorbent was examined experimentally using n-hexane as a solvent. The experimental procedure was carried out using three consecutive aliquots of 100 ml n-hexane. The EPP sample with the adsorbed crude oil on its surface was used. Post to the n-hexane extraction process, the EPP was dried and reused for oil removal from SPW.

Table 1
Equilibrium adsorption parameters of three isotherms for the removal of oil from PW using EPP

Isotherm model	q_m (mg/g)	k_L (L/mg)	k_f	k_t	n	B (mg/g)	R^2
Langmuir	833.3	0.02570	–	–	–	–	0.997
Freundlich	–	–	5.836	–	2.074	–	0.916
Temkin	–	–	–	0.2857	–	389.1	0.963

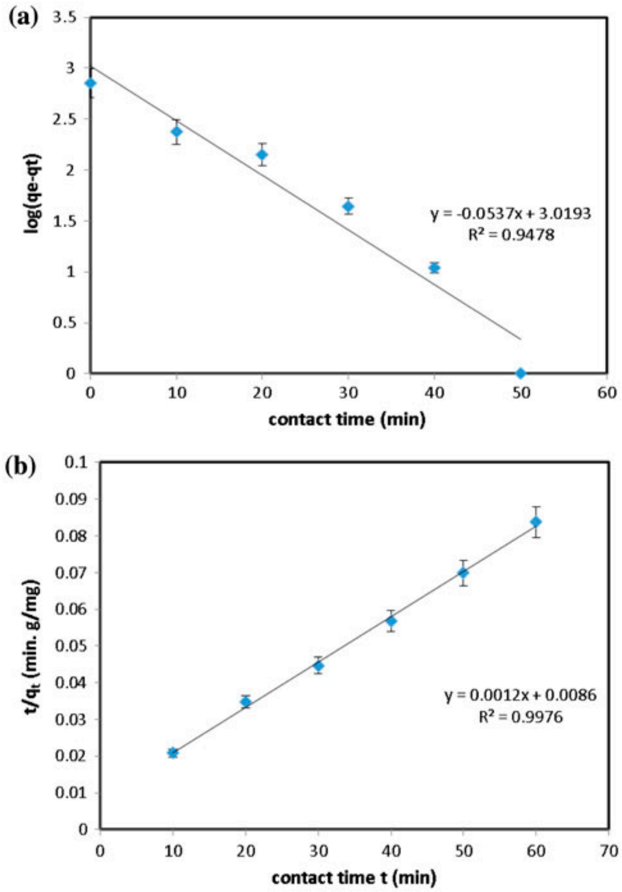

Fig. 8. Kinetic analysis for the adsorption of oil by eggplant peal powder: (a) pseudo-first-order model and (b) pseudo-second-order model.

The reuse of EPP as an adsorbent for the second time lowers its efficiency from 95.2 to 88.8%. However, the reduction is not significant and hence this regeneration method proved to be valuable for practical applications.

3.10. Oil removal of oil from actual produced water

A sample of an actual produced water was obtained for evaluation from Sharjah National Oil Company, Sharjah. Using n-hexane for oil extraction, the amount of oil concentrations extracted from the PW sample was found to be 230 mg/L. The efficiency of EPP for oil removal from the actual produced water was also studied at the optimum experimental conditions. The EPP efficiency for oil removal was measured to be about 95.5%. The calculated efficiency explains vividly the suitability of the process in real applications.

Table 2
Kinetic model parameters for sorption of oil onto the surface of EPP

Kinetic model	Parameters	Values
Pseudo-first-order	k_I (g/mg h)	0.0537
	$q_{e,cal}$ (mg/g)	1,045
	R^2	0.947
Pseudo-second-order	k_{II} (g/mg h)	0.000167
	$q_{e,cal}$ (mg/g)	833.3
	R^2	0.997

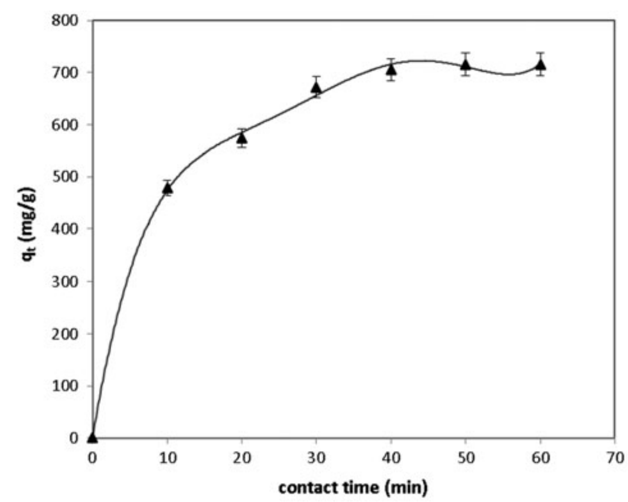

Fig. 9. The relationship between the amounts of crude oil adsorbed per amount of adsorbent and equilibrium concentration.

4. Conclusion

The oil removal process was made easier compared to other available procedures by the introduction of newly developed feasible and suitable naturally occurring adsorbent. EPP is the raw material used to develop this proposed adsorbent. The efficiency of the developed adsorbent for oil formal from PW was found to be about 95% at the optimum parameters. The efficiency of the process was optimized by adjusting the experimental conditions. The optimum conditions for EPP dosage, pH, contact time, and temperature were found to be 1.75 g/L, 10.0, 40 min, and 55˚C, respectively. The salinity was found to have a positive effect on the efficiency. Langmuir adsorption curve played a dominating role during the oil removal process showing maximum adsorption capacity of 834 mg/g. Adsorption proceeded through pseudo-second-order kinetics expressing a rate constant of 1.67×10^{-4} g/mg h. The newly proposed adsorbent is expected to make an excellent contribution to the oil separation techniques since it can be easily regenerated without significant alterations in its efficiency, therefore, making it the most economical and pollution-free method to adopt.

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