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The Removal of Ibuprofen Drugs Residues from Municipal Wastewater by *Moringa Oleifera* Seeds

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ABSTRACT
Municipal wastewater may contain residues of different drugs causing severe chemical contamination of water bodies. However, the microbial degradation of Wastewater Treatment Plants (WWTP) may not eliminate such drug residues completely. The current work was designed to remove the Ibuprofen drug residues by using the *Moringa Oleifera* seeds. Various testing methods such as Brunauer, Emmett and Teller (BET), Transmission Electron Microscopy (TEM), scanning electron microscopy (SEM), Fourier transform infrared spectroscopy (FT-IR) were applied to assess the efficiency of such plant seeds in bioremoval of ibuprofen residues from municipal wastewater The batch reactor was used to find the optimum operating conditions using various parameters with different pH values, duration time, Ibuprofen concentration and various quantities of plant seeds. In the batch reactor, the operation conditions were: pH 7, duration time 150 min, Ibuprofen dose of 1000 mg/l, activated adsorbents and *Moringa Oleifera* seeds in the amount of 1000 mg/l. Moreover, the packed bed reactor was used to examine different parameters such as initial Ibuprofen concentration, flow rate and bed depth for 6 hours. It was found that the best conditions were 2 cm depth, and 25 l/hr flow rate. Meanwhile, the kinetic constants were studied by adsorption equilibrium with the isothermal Langmuir and Freundlich models. The best results were shown with the Freundlich isotherm, and the first pseudo order was more suitable for the removal of Ibuprofen by adsorbed activation of *Moringa Oleifera* seeds.

Keywords: adsorption, activated *M. Oleifera*, Batch reactor, Ibuprofen, packed bed reactor.

INTRODUCTION

It has been reported that the residues of various pharmaceuticals and xenobiotic compounds were found in drinking and wastewaters are another public health concern (Ippolito et al., 2011; Touraud et al., 2011; Szymonik et al., 2017). Little is known about the potential chronic health effects associated with long-term ingestion of mixtures of these compounds through drinking water (Kümmerer, 2001; Patneedi and Prasadu, 2015). However, the occurrence of such pharmaceuticals drugs residues in water is due to pharmaceutical industry waste, personal hygiene products, hospital waste and therapeutic drugs (Hee-Jong and Seong-Ho, 2011). It has been well documented that such drug residues had a significant impact on both public health and environment (Yao et al., 2010; Proesser and Sibly, 2015; Adegoke et al., 2018).

The residues of pharmaceutically active compounds (PhACs) and related contaminants were investigated thoroughly worldwide (Lin et al., 2018; He et al., 2018; Omar et al., 2019; Duan et al., 2020). A detailed work (Heberer et al., 2002) has examined a new long-term monitoring program of sewage, surface, ground- and drinking water and found that in the course of the monitoring program, PhACs and some other polar compounds were detected at the concentrations up to the micro g/L-level in all compartments of the Berlin water cycle.

However, various studies have focused on the removal of ibuprofen and other pharmaceutical residues from fresh and wastewaters by using various techniques such as chemical coagulation

(Vieno et al., 2006), aerobic and anaerobic process (Drewes, 2007), membrane bioreactor (Smook et al., 2008), microbial materials (Langenhoff et al., 2013), electrochemically generated ferrate (Ljilian et al., 2016), Ozonation process (Oghazyan et al., 2017); activated carbon (Nourmoradi et al., 2018), advanced oxidation process (Soudabeh et al., 2018), and plant materials (Leon et al., 2019).

On the other hand, *M. oleifera* seeds were used intensively in water purification (Amagloh and Benang, 2009), river water disinfection (Ali et al., 2019), water treatment (Ali et al., 2009; Tan et al., 2013; Okuda and Ali,2018; Narender et al., 2019), wastewater treatment (Ramesh and Mekala, 2018) and as biosorbent for the removal of fluoride (Agnihotri et al., 2013), various heavy metals (Beltran and Sanchez-Martin, 2008; Limmatvapirat et al., 2015; Abbas, 2018) and the toxic pollutants (Shirani et al., 2018) from all water types.

The *M. Oleifera* (Moringaceae) plant belongs to the *Moringa* genus (Morton, 1991). This plant is now cultivated in all tropical and subtropical regions (Ali et al., 2009). This is due to its resistance to different climates, as well as poor and moderately dry soils (Ali et al., 2009). It reaches 15 m in height, with a diameter of 20–40 cm (Odee, 1998). Many parts of the plant show pharmacological properties, recognized by popular use and corroborated by the scientific community while other parts of the plant (seeds and leaves) are regarded as low cost and environmentally sound biosorbents (Ali et al., 2015).

Ibuprofen drug, a carboxylic acid. Anti-inflammatory drugs or NSAIDs are frequently used to relieve certain pains. The Ibuprofen medicine has a powdery white appearance and is produced in the form of capsules, tablets, or powder (Carlo, 2013; Pehlic et al., 2013).

Therefore, the current work was designed to use the *Moringa Oleifera* seeds as biosorbent for the removal of Ibuprofen drug residues from municipal wastewater.

MATERIALS AND METHODS

Preparation of activated carbon

M. Oleifera seed pods were collected and washed thoroughly with distilled water and dried to constant weight under sun light. Thereafter, the seed pods were ground and sieved to a size of 106 μm, left to dry in an air oven at 105 °C for 6 h. Finally, they were stored in vacuum desiccators.

According to the procedure in ASTM, D 4442 for moisture content, the activating carbon of *M. Oleifera* seed (ACMO) was prepared following the previous work (Abdallah, 2017). After pretreatment, zinc chloride ($ZnCl_2$) was added to the chemical activation method, as follows:

The sulfuric acid (H_2SO_4) was used to activate the raw material with ratio 1:10; 60 g of raw material was weighted and impregnated in 600 ml 10% H_2SO_4 (v/v) and 10% $ZnCl_2$ (w/v) for 24 hours with continuous mixing then left to dry at 100–105°C in oven and stored until it was used as shown in Figure 1. (Dahham, 2018).

The experimental work consisted of two parts where the first was performed using a batch reactor to study the operation conditions and the second used a packed bed reactor to study the breakthrough.

Batch Experiments

The different adsorption experiments were carried out in a batch reactor to obtain the best

M. Oleifera crude Activated carbon

Fig. 1. *M. oleifera* crude before and after activated carbon

operation conditions, such as Ibuprofen concentration (100, 250, 500, 1000 mg), adsorbent material (100, 250, 500, 1000 mg) and pH (2, 5, 7, 9). The tests were performed in a beaker with the capacity of 1 L with rotation speed of 200 rpm for detention time 3 hour at room temperature of 22°C.

Packed bed reactor was designed and constructed to be used for continuous operation. The reactor consists of a glass tube with 60 cm length and internal diameter of 2.1 cm. The Ibuprofen solution was pumped using submersible water pump from feed solution to the bed during the valve that controls the flow rate after passing during flow meter. Different sizes of glass beads were used to provide inert zone as well as to catch any impurities and to assist inflow distribution of the reactants through the bed as stated by (Satterfield, et al., 1996), As shown in Figure 2, the flow pattern used in this experimental was down flow and the effluent was collected in a plastic container.

The operation conditions obtained from batch reactor experiment were used in a continuous system (backed bed reactor). The parameters involved different bed heights (1, 1.5, and 2 cm), different flow rates (25, 30, and 35) l/hr, and different Ibuprofen initial concentrations (100, 400, and 625 mg/l). The experiment was performed at room temperature. The sample was withdrawn from effluent each 30 minutes, filtered and stored to analyze later. The high performance liquid chromatography (HPLC) instrument was used to measure Ibuprofen concentration.

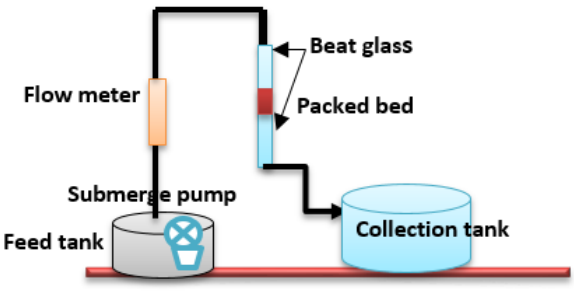

Fig. 2. Schematic diagram of the packed bed system

RESULTS AND DISCUSSION

BET Instrument Results

Many factors are important on adsorbent process one of them surface area, Table 1 shows the results of *M. oleifera* seed activated carbon surface area.

The results of BET instrument show that there was a clear increase in the surface area of activated *M. oleifera* compared to normal seeds. According to the International Union of Pure and Applied Chemistry (Nicoleta et al., 2013), the mesoporous (20 to 500 nm) was the characteristic of the activated *M. oleifera* pore size. The chemical composition of the *M. oleifera* seed, and the activation during Transmission Electron Microscopy (TEM) test are given in Tables 2 and 3, while the results of Energy Dispersive Spectroscopy (EDS) are presented in Figures 3 and 4. It was observed that sulfur, and phosphorus and nitrogen appeared in certain amounts, but these quantities were decreased after the carbonization process where the carbon reduction during carbonization process was due to the oxidation of the organic content.

In the images of the *M. oleifera* before and after activation during the scanning electron microscopy (SEM) test, which are shown in Figures 5 a-b as well as 6 a-b, the pores and surface roughness in different magnification also show the boundary of crystals regions, different materials that are mixed with nanomaterials in different sizes.

From Fourier transform infrared spectroscopy (FT-IR), the IR spectrum of natural and activated *M. oleifera* with function groups are shown in Table 4 and in Figures 7 and 8.

The results of FT-IR spectrum of natural and activated *M. oleifera* are shown in Figures 7 and 8. A wide function group range was about 3912.50–3282.84 cm^{-1} but low peaks were attributed to the surface hydroxyl group and chemisorbed water. The narrow peak varied from 2331.92 cm^{-1} to 2312.65 cm^{-1} and hydroxyl group from 1905.09 cm^{-1} to 1789.94 cm^{-1} that between two groups which

Table 1. BET Instrument results

Adsorbent	Surface area (m^2/gm)			Pore volume cm^3/gm(Å)		Pore size	
	before	after	after packed bed reactor	before	after		
M. oleifera seed activated carbon	612	720	250	–	0.88739	–	30.029 nm

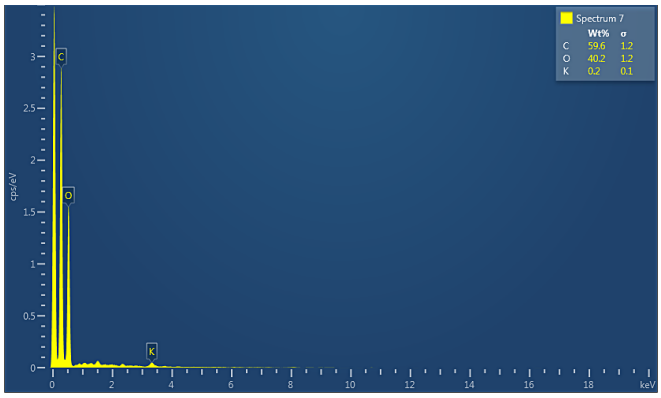

Fig. 3. EDS for natural *M. oleifera*

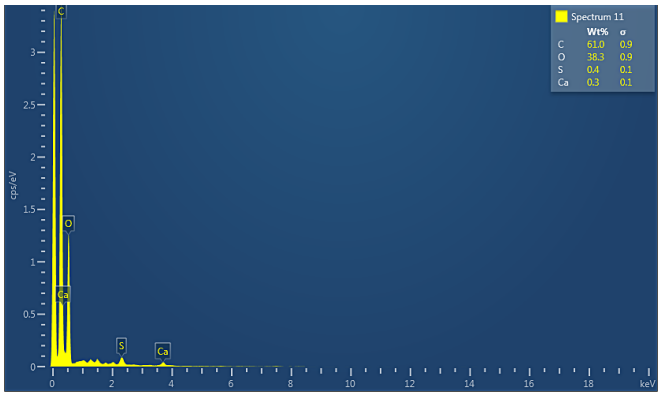

Fig. 4. EDS for active of *M. oleifera*

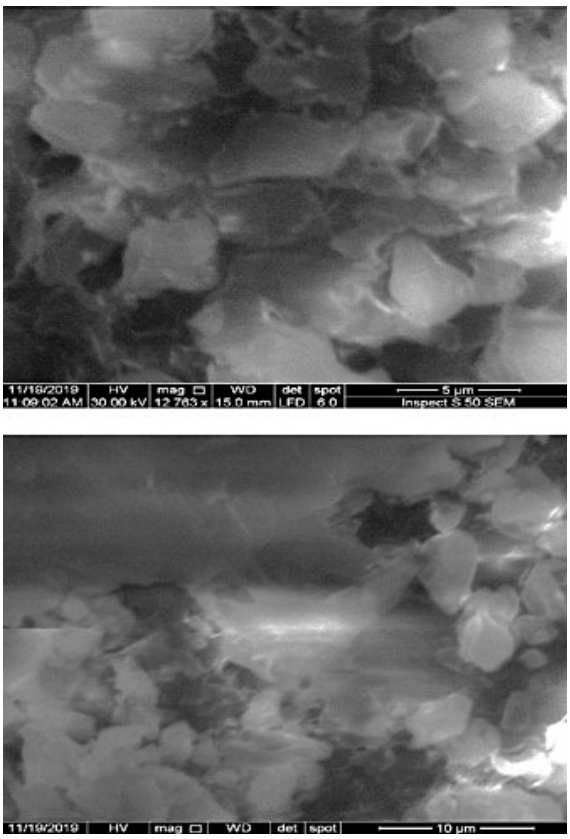

Fig. 5. SEM image for natural *M*. Oleifera with magnification of (a) (5 µm scale), (b) (10 µm scale)

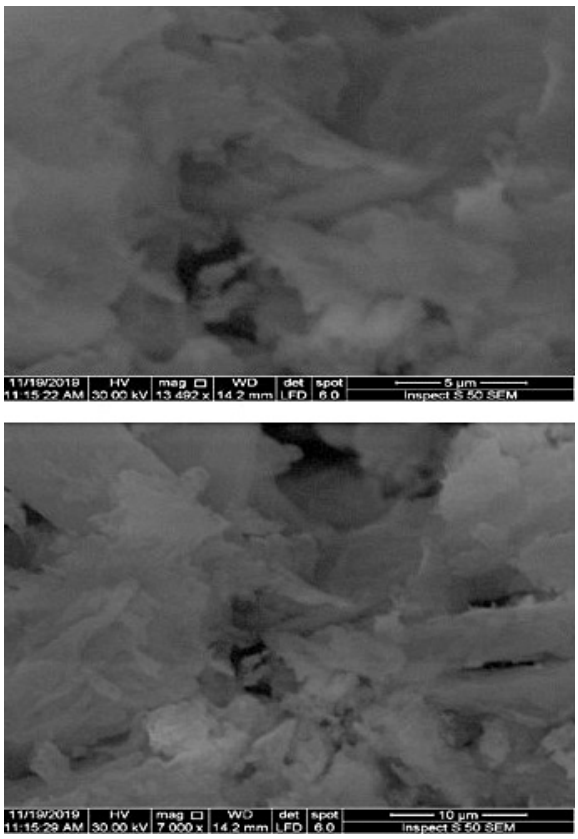

Fig. 6. SEM image for active of M. Oleifera with magnification of (a) (5 µm scale), (b) (10 µm scale)

Table 2. TEM for natural *M. oleifera*

Spectrum 7	Wt%	σ
C	59.6	1.23
O	40.2	1.22
K	0.2	0.11
N	10.22	2.5
S	1.95	0.12
P	1.3	0.09

Table 3. TEM for active of *M. oleifera*

Spectrum 11	Wt%	σ
C	61.0	0.9
O	38.3	0.9
K	0.43	0.06
N	3.2	1.81
S	0.9	0.55
P	0.8	0.06

Table 4. The groups of spectrum and this function

IR spectrum	Group function	Reference
3903.92 cm^{-1} to 3745.76 cm^{-1}	hydroxyl group and chemisorbed water	
3375.43 cm^{-1} to 2866.22 cm^{-1}	the stretching vibration of C-H alkane group	
2308.79 cm^{-1} to 1876. 74 cm^{-1}	C=O ketonic and aldehyde group	(Kalavathy et al., 2010).
1649.14 cm^{-1} to 1066.64 cm^{-1}	C=C alkene aromatic ring and COO- Carboxylate groups	
671.13 cm^{-1} to 499.56 cm^{-1}	functional groups, the band can be assigned to strong C-O bond	the basic structure of *Moringa oleifera* (Pehlic et al., 2013)

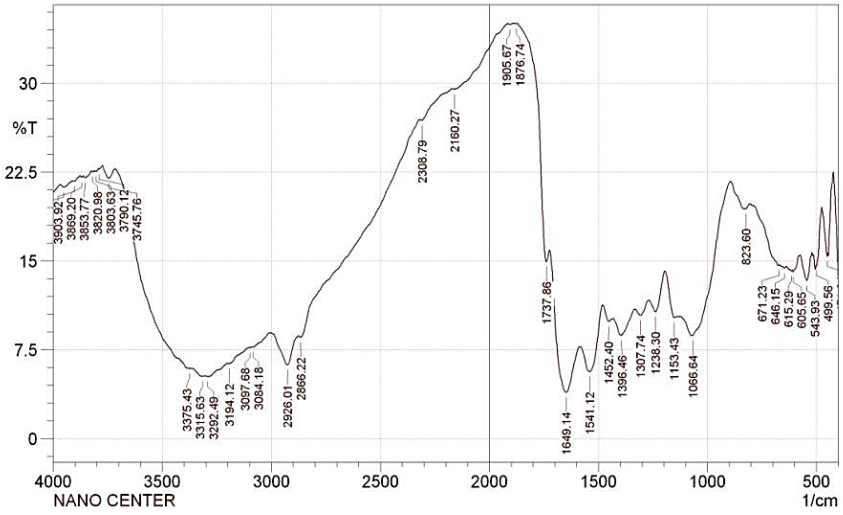

Fig. 7. FT-IR spectrum of the MO seed

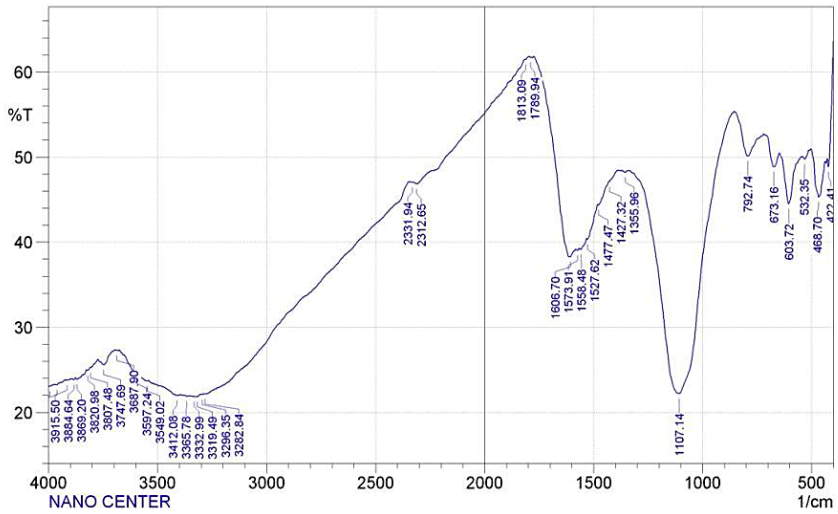

Fig. 8. FT-IR spectrum of the ACMO seed

were C-H and C=O. 1606.70 cm^{-1} to 1355.96 cm^{-1} and the range between C=O to C-O peak appeared in 1107.14 cm^{-1} (Kalavathy et al., 2010). The last function group of peaks was between 792.74 cm^{-1} to 422.14 cm^{-1}. The basic structure of *M. Oleifera* was shown in the peaks between 792.74 cm^{-1} to 422.14 cm^{-1} (Munajad et al., 2018).

Results of batch reactor

In order to estimate the Ibuprofen concentration in the experimental work of batch reactor, the Ibuprofen concentration was measured in the wastewater collected from the AL Rustomyia treatment plant -Baghdad city and it was found very high (650 mg/l). The removal efficiency rate in this station was 60% due to the use of drugs in large quantities through medical and non-medical prescriptions. Moreover, an animal skin tanning factory (Syadia), near the treatment plant, discharged industrial wastewater carrying significant quantities of Ibuprofen with diclofenac used in tanning animal skins.

Change in pH

Figures 9 and 10 represent the effect of pH on the adsorption of Ibuprofen by activated *Moringa Oleifera*. An improved removal of Ibuprofen along with increasing pH up to 7 was observed, where at low pH, the adsorbent surface (activated *M. oleifera* seed powder) was surrounded by hydronium (H_3O^+) ions, and this has decreased the interaction of Ibuprofen with the sites of activated *M. oleifera* seed powder by repulsive forces resulting in low adsorption.

Different Ibuprofen concentration

In this study, the temperature and pH were kept constant at 22°C , and pH 7 while the activation Moringa oleifera equaled to (100 mg/l), and

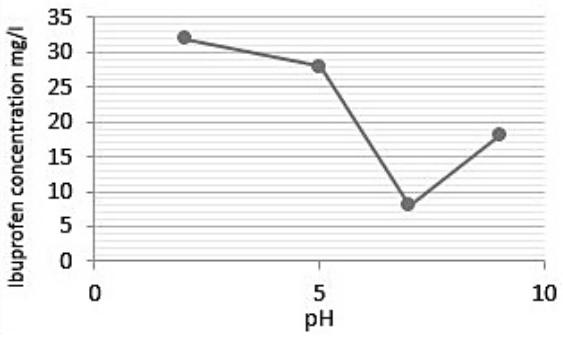

Fig. 9. The relation between pH and removal of Ibuprofen with different pH and constant temperature of 22°C, activated *M. Oleifera* seed concentration (100 mg/l), and the Ibuprofen concentration in (100 mg/l)

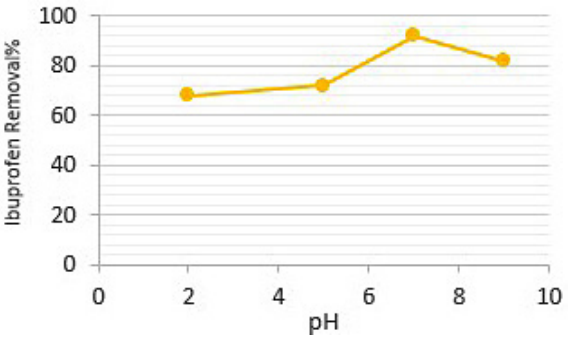

Fig. 10. The relation between pH and Removal efficiency of Ibuprofen with different pH, and constant temperature of 22°C, activated *M. Oleifera* seed concentration (100 mg/l), and the Ibuprofen concentration in (100 mg/l)

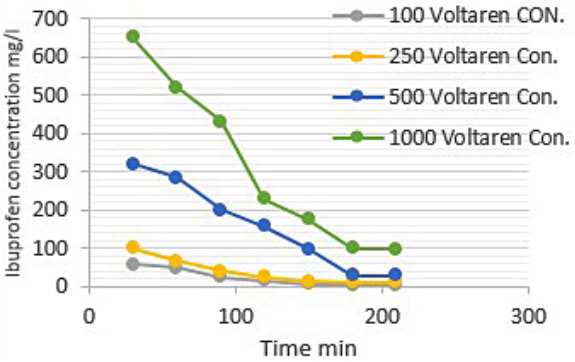

Fig. 11. the relation between time and Ibuprofen concentration, with constant temperature of 22°C, pH 7 and the activated *M. oleifera* seed in (100 mg/l)

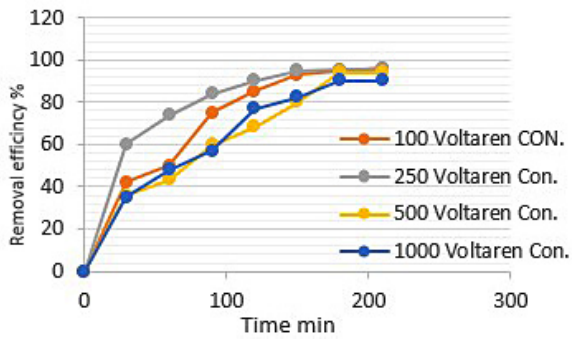

Fig. 12. The °between time and removal efficiency of Ibuprofen, with constant temperature of 22°C, pH 7 and the activated *M. oleifera* seed in (100 mg/l)

only the concentration of Ibuprofen was changed systematically (100–250–500–1000mg/l). The Ibuprofen efficiency was calculated by Eq. (1):

$$\text{Efficiency} = (C0 - Ce)/\ C0 \quad (1)$$

where: $C0$ = Initial Ibuprofen concentration (mg/l).
Ce = Effluent Ibuprofen concentration (mg/l).

Figure 11 shows the relation between time (min) with Ibuprofen concentration (mg/l) where the Ibuprofen concentration was decreased with time and the high removal concentration was 250 mg/l. This is due to the available space of activated Moringa Oleifera. However, Figure 12 shows that the Ibuprofen concentration was increased with decreasing the time for all adsorbent tests.

Different mass of *Moringa oleifera*

The relationship between time and the concentration of activated *M. oleifera* is shown in Figure 13. The Ibuprofen concentration was decreased with the time causing a clear increase in the removal efficiency, along with the concentration of the adsorbent substance activated *M. oleifera*, as shown in Figure 14. This was due to the adsorption sites, which were initially opened and the Ibuprofen was interacted easily with the site where the concentration difference between the bulk solution and the solid liquid interface was initially higher and this may lead to higher rate of adsorption observed after 150 minutes. At this time, the reaction has reached the steady state. The best removal efficiency was achieved at the *M. oleifera* concentration of 1000 mg / liter. Because there was sufficient space for the reaction and the last period, the reaction was completed because the Ibuprofen concentration has become a very small amount. Thus, ensuring uniformed spaces with high interlayer where the Ibuprofen entered to become eliminated (Kurniawan et al., 2006- Sirocki, et al., 2013).

The kinetic models

An important step in studying the adsorption processes is the study of kinetics (Wang, 2008). The kinetic equation is reported in the n-th order of absorption as in equation (2):

$$(dc / dt) = - k\ C^{n} \quad (2)$$

where: k – adsorption rate coefficient,
C – Ibuprofen concentration,
t – time, and
$n\text{-}th$ – reaction order.

From the previously equation, the pseudo-first and pseudo–second-order kinetic equation were shown as in Equations (3) (4), respectively (Emami et al., 2010):

$$(\text{First} - \text{order})\ \log (q_e - q_t) = \log q_e - K_1 t \quad (3)$$

where: qt, q_e is the adsorbed at time t and equilibrium (mg/g),
t is the time of adsorption process (min), and,
k_1, and k_2 is the rate constant for pseudo-first, and pseudo-second-order reaction, respectively.
K_1 is a constant (min-1), which is determined by plot ln $(q_e - q_t)$ versus t.

$$(\text{Second} - \text{order})\ \frac{t}{qt} = \frac{1}{K2\ qe\text{ʌ}2} + \frac{t}{qe} \quad (4)$$

The initial sorption rate is defined by the following equation:

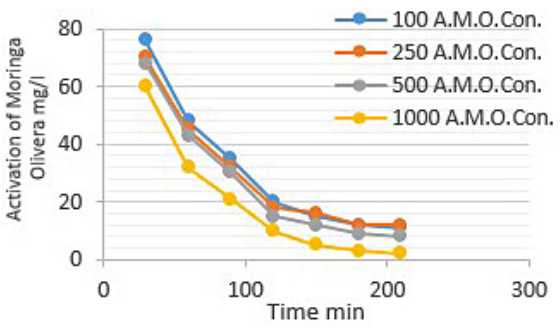

Fig. 13. The relation between time and Ibuprofen concentration, with constant temperature 22°C, pH 7 and the Ibuprofen concentration in (100 mg/l)

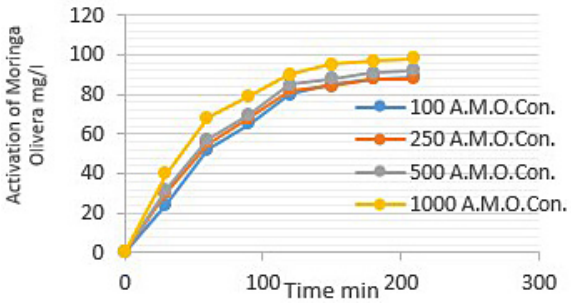

Fig. 14. The relation between time and removal efficiency of Ibuprofen, with constant temperature 22°C, pH 7 and the Ibuprofen concentration in (100 mg/l)

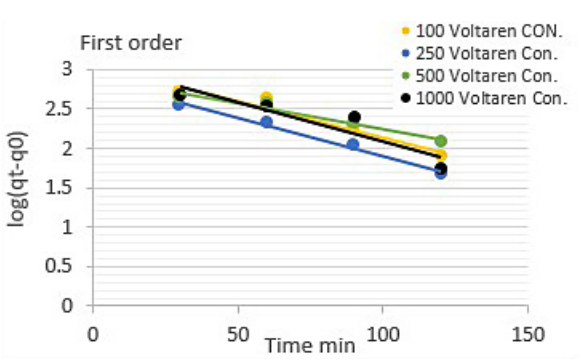

Fig. 15. Linear plots of kinetics data of the first order model at various dosage of Ibuprofen

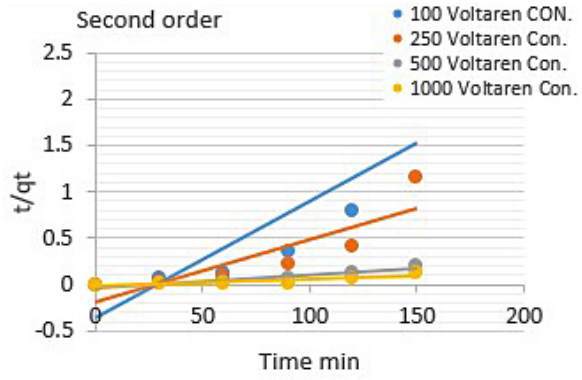

Fig. 16 Linear plots of kinetics data of the second order model at various dosage of Ibuprofen

Table 5. Kinetic parameters

	First order			Second order			
	R^2	k_1 (min^{-1})	calculated eqilb. uptake qe (mg g^{-1})	k_2 (g mg^{-1} min^{-1})	h (mg g^{-1} min^{-1})	R^2	Calculated eqilb. uptake qe (mg g^{-1})
100	0.9402	0.00286	794.3	0.003	19.2	0.8741	80
250	0.9887	0.0051	891.3	0.0053	107.16	0.9339	142.8
500	0.9596	0.0013	501.2	0.00147	235.2	0.7718	400
1000	0.8369	0.00432	398.1	0.00123	136.6	0.8896	333.33

$$h = k_2 * q2^2 \quad (5)$$

This kinetic study on different Ibuprofen concentration due to large value of R^2

The result k and R^2 for first and second order were shown in Figures 15, 16 and in Table 5. From the results, it was found that the pseudo-first order was the best result because of the high R^2.

The results showed that the pseudo-first-order equation has fitted the experimental data well with a correlation coefficient (R^2) which was more close to one than the second-order. Table 5 shows the result of first-order and second-order models and also to those observed in Figures 15 and 16. The deviation from the straight line of sorption as in the pseudo-first- and second-order model, and the adsorption in this study was a slow especially in the initial period of the reaction. On the basis of the correlation coefficients, the first-order model was a reaction pathway for sorption of Ibuprofen by activated Moringa oleifera (Kowanga et al., 2016).

Packed bed reactor

The breakthrough was studied in packed bed reactor and the experimental tests were performed with different parameters such as bed height of adsorbent (1, 1.5, 2cm), initial Ibuprofen concentration (100- 400- 625 mg/l), the flow rate (25-30–35 l/hr), constant pH up to 7 and temperature of 22°C.

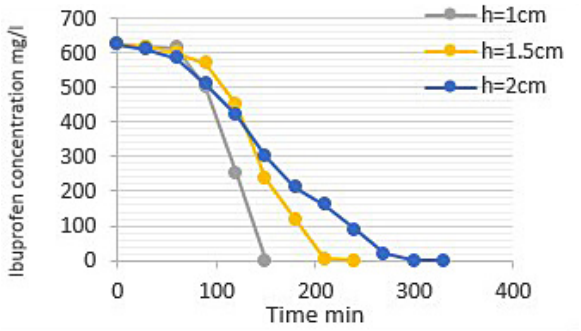

Fig. 17. Ibuprofen concentration adsorption on activated *M. oleifera* seed with different depth and flow rate 25 l/hr. and pH 7

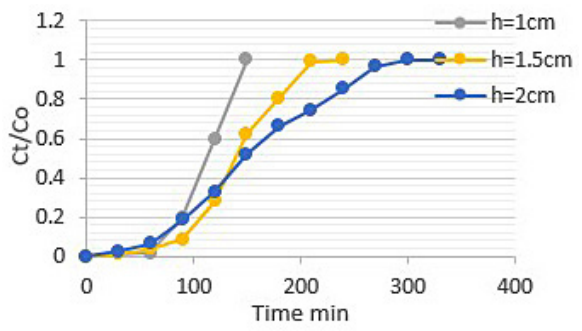

Fig. 18. Breakthrough curve of Ibuprofen adsorption on activated *M. oleifera* seed with different depth and flow rate 25 l/hr. and pH 7

Different height bed

The results showed that the breakthrough was increased along with bed height, as shown in Figures 17 and 18 for Ibuprofen removal efficiency with time. Moreover, it was found that the breakthrough observed in bed height 1 cm was not clear because the bed was small and not enough to complete adsorption, but in beds of 1.5, 2 cm, the breakthrough was very clear due to increase in the surface area for adsorption and increase in detention time that referred to high capacity to adsorb the Ibuprofen solution. These results are supported by those of recent study (Al Ani et al., 2019). The breakthrough has begun approximately from the same time, but it was finished at different times.

Different flow rate

In order to study the effect of flow rate, different flow rates (25, 30, and 35 l/hr) were applied with constant of bed height at 2 cm, pH at 7, Ibuprofen concentration at 625 mg/l, and temperature at 22°C. The effect of changing flow rate is shown in Figure 19. The results showed that with increasing the flow rate, the breakthrough has become steeper depending on resident time of Ibuprofen passing on adsorbed bed. Thus, the residence time was decreased with increasing the flow rate. Additionally, Figure 20 shows that the Ibuprofen concentration was decreased with increasing operation time of the bed for removal and decreasing flow rate.

Different initial Ibuprofen concentrations

The effect of the initial Ibuprofen concentrations (100, 400, 625mg) on breakthrough with other parameter kept constant is shown in Figure 21, where the bed height was 2 cm, flow rate 25 l/hr, pH 7, and temperature – 22°C. It seems that with increasing the initial Ibuprofen concentration, the breakthrough increased too. Figure 22 shows that the Ibuprofen concentration decreased with time.

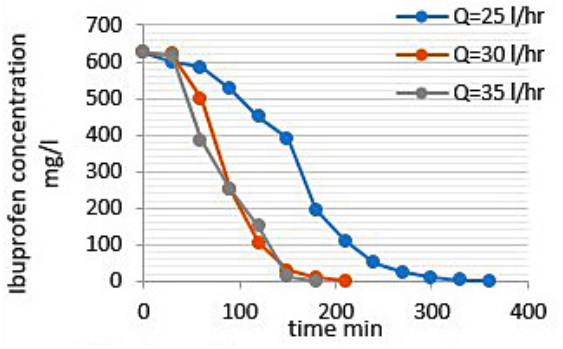

Fig. 19. Ibuprofen concentration adsorption on activated *M. oleifera* seeds with depth 2 cm, and pH 7, Ibuprofen concentration 625 mg, and different flow rate

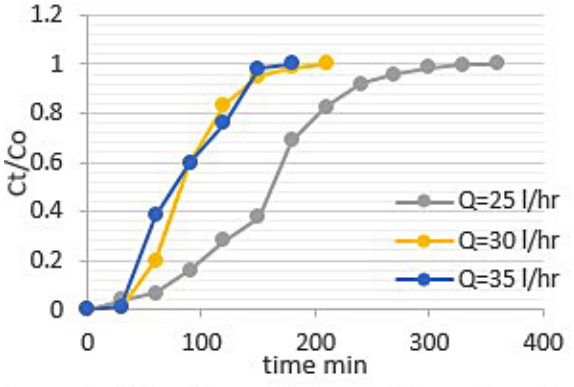

Fig. 20. Breakthrough curve of Ibuprofen adsorption onto activated M. oleifera seeds with depth 2 cm, and pH 7, Ibuprofen concentration 625 mg, and different flow rate

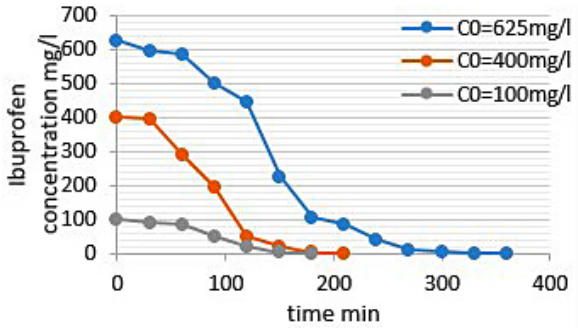

Fig. 21. Ibuprofen concentration adsorption on activated *M. oleifera* seeds with depth 2 cm, pH 7 and different Ibuprofen concentration

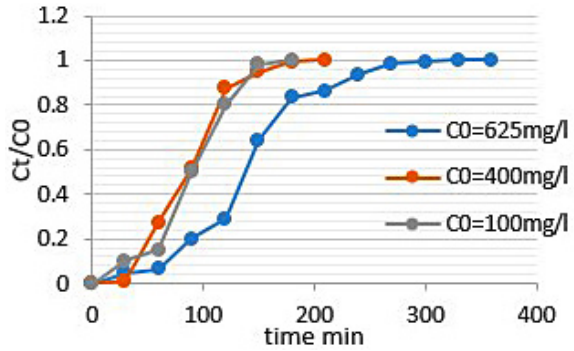

Fig. 22. Breakthrough curve of Ibuprofen adsorption on activated *M. oleifera* seeds with depth 2 cm, pH 7 and different Ibuprofen concentration

Kinetic isotherm model

The isotherm analysis was studied through applying a fitting technology for several isotherm models to achieve a suitable model that it is applied for designing objectives. In the Langmuir and Freundlich adsorption models, the constant of Freundlich equation was determined by slope and the linearized was done by using the equation 5.

$$\ln q_e = \ln K + \frac{1}{n} \times \ln C_e \quad (6)$$

where: *k, n* constant, 1/n range between (0–1)

However, the constant of Langmuir equation was determined by slope, interest linearized by using the equation 7,

$$\frac{1}{q_e} = \frac{1}{q_{max}} + \frac{1}{q_{max}.b} * \frac{1}{C_e} \quad (7)$$

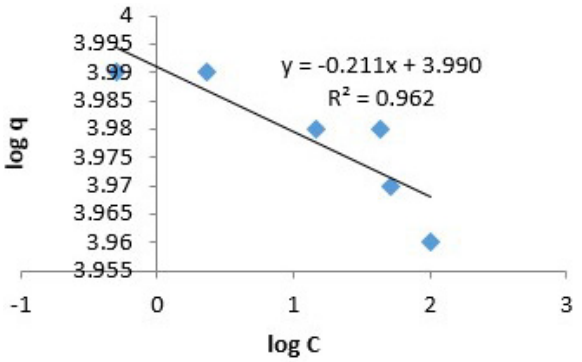

Fig. 23. Freundlich mode (C_e=mg/l, and q_e= mg/g) for Activated Moringa Oleifera seed adsorbent

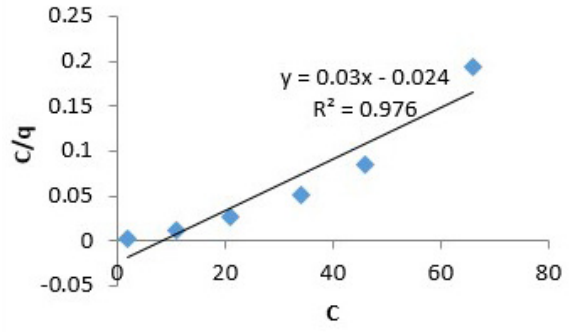

Fig. 24. Langmuir mode (C_e=mg/l, and q_e= mg/g) for Activated Moringa Oleifera seed adsorbent

This equation was used to determine the value of q_{max} (mg/g) and b (L/mg) through plot $1/q_e$ versus $1/C_e$. The activated *Moringa oleifera* seeds adsorbent through Freundlich adsorption models showed that during R2, the Ibuprofen concentration was more effective on the model, so that the kinetic was adapted to the change in Ibuprofen concentrations.

The activated *M. oleifera* seeds adsorbent through the Langmuir adsorption models showed that the change in the concentrations of activated *M. oleifera* seed at 100 mg was more effective on Ibuprofen removal, so that the kinetic was adapted on change in weight of activated *M.oleifera* seed as shown in Figures 23 and 24. The results of the Freundlich and Langmuir model constant are given in Table 6. It was seen that:

- q_e which represents the solute obtained in unit mass of local adsorbent was found to be related directly with value of concentration C_e that represented the equilibrium concentration
- The type of equilibrium isotherm that considers a favorable type and there due to the high weight of adsorbent for Langmuir and Freundlich which can be applied. Both models give approximately the same results, but the Langmuir model gives slightly higher R2 values.

CONCLUSION

The activated *M. oleifera* seeds were found to be the best adsorbent and a good alternative, being a cheap adsorbent that can be used for the Ibuprofen removal from wastewater. Using a batch reactor, it was found that the operation conditions were as a follows: pH 7, Ibuprofen concentration 1000 mg/l, adsorbents (activated *M. oleifera* seed – 1000 mg/l. From the results, the first order was found the best adsorption kinetic result in batch reactor. The break points of the breakthrough curve for adsorbents (activation of M. oleifera seed) increased along with the bed height and initial flow rate. In the case of a continuous reactor, the Freundlich and Langmuir isotherm models were found to be favorable for activation of *M. oleifera* seed as adsorbents.

Table 6. Freundlich and Langmuir model constant

Adsorbent	Models	Model constant		
		R^2	K	n
M. Oleifera seeds activated Carbone	Frundlish model	0.962	0.59	4.7 1/n = 0.212
	Langmuir model	0.976	Q = 0.33	b = 0.013

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