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Removal of Used Motor Oil from Water Body Using Modified Commercial Activated Carbon

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In this study, modification of commercial activated carbon (AC) was carried out for adsorption of used motor oil (UMO) from wastewater. The modification process involved impregnation of phosphoric acid at ratio 1: 2 followed by 500^{o}C for 2 h for carbonization. Characterization of modified activated carbon (MAC) was done by using scanning electron microscopy with energy dispersive X-ray spectrometry and Fourier transform infrared spectroscopy to study morphology and the structural change and to identify the surface functional group of MAC. Based on the results obtained from this study, the percentage yield of the MAC was 63% with a moisture content of 37%, and pH was 2.37. The adsorption of UMO by MAC was investigated. The effect of contact time, temperature, and pH to the adsorption process was studied, where the results showed that MAC had higher percentage of adsorption compared to AC. The study of the reusability of MAC was also done for 30 min and three cycles. The result showed that MAC could be regenerated and reused. The study of adsorption equilibrium was done by using Langmuir and Freundlich isotherms. The adsorption of UMO proved to fit better to Freundlich isotherm compared to Langmuir isotherm with a R^2 value of 0.9174. The adsorption kinetic study was done using pseudo-first-order and pseudo-second-order models. The adsorption data showed a better fit to pseudo-first-order kinetic model with a R^2 value of 0.9239. Adsorption capacity was found to be 3.951 oil/g (MAC).

Key words: Adsorption; modified activated carbon; used motor oil; kinetics; impregnation of phosphoric acid; SEM-EDX; FTIR

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In the present day, the increasing of wastewater discharged which contains oil to the environment is due to the urbanization and industrial development [1]. Wastewater treatment is an important aspect of investigations to meet the objective of sustainable development and clean technology [2]. One of the major problems encountered by many industries such as crude oil production, oil refinery, petrochemical industry, lubricant, and many other industries is oily wastewater. Fingas [3] reported that the method for oil spill recovery on water is skimmer and sorbent. There are also other methods for oily water treatment including a dispersant and in-situ burning. These methods do not include oil recovery. Nikkhah *et al.* [4] have stated that different techniques have been developed to eliminate oil contaminants from water. Previous studies suggested that the most profitable method to remove oil contaminants is by using adsorption method as it can effectively remove and also recover the oil from water [2, 5-8]. Sorbents are very commonly used during oil spills for their cost-effectiveness and affordability [5]. According to Sarbatly *et al.* [9], the sorbents can be categorized based on its raw material such as synthetic sorbent, natural organic sorbents, or natural inorganic sorbents.

Activated carbon (AC) is commonly made from diverse kind of materials which include agronomic and forestry remainders. According to Prahas *et al.*[10], most of the raw materials used to produce AC are rich in carbon. Typically AC can be produced via two routes, which are physical activation and chemical activation. ACs are also well known for their high adsorptive capacities in which internal porosity and other properties such as surface area, pore volume, and pore size distribution associated. Previous pieces of evidences suggest that AC is an efficient adsorbent for different types of pollutants and is well known for its efficiency in removing organic compounds [11-19]. Modification of AC is further made to increase surface adsorption and removal capacity and adding selectivity to carbon [20,21]. The most commonly used method of modification is through chemical activation. The most widely used dehydrating agents for chemical activation are zinc chloride ($ZnCl_2$) and phosphoric acid (H_3PO_4) [22-24]. However, the usage of $ZnCl_2$ has been declined because it will cause environmental contamination with zinc compounds [24]. Thus, a suitable dehydrating agent is needed to modify the AC further. Although by modifying AC might help in increasing the performance of AC, the adsorption capacity depends on the properties of the modified activated carbon (MAC) [20]. If the properties such as pore size increases, the adsorption capacity also increases as more oil is adsorbed.

In this study, the sorption method or better known as adsorption method has been selected for extraction of oil and grease from used motor oil (UMO). The adsorbent used for this study is commercial AC which was further modified through chemical activation with H_3PO_4.

EXPERIMENTAL

Materials

The UMO sample was obtained from a local Automobile Service Center and was used as adsorbate in this study. The density of UMO was found to be 0.9 g/cm^3. 85% of H_3PO_4 (Merck) was used for impregnation of AC. Sodium hydroxide (NaOH) (Fischer) with more than 97% purity was used for washing of the AC. 99% of purity of n-hexane (C_6H_{14}) (System) was used as a solvent for extraction of residual oil for oil content analysis and to analyse the oil recovery from MAC.

Preparation of AC

The AC (NORIT) was purchased from Sigma-Aldrich, which is coconut-shell based in powder form with ~100 mesh particle size. It is made from charcoal of the hard-shell of the coconut which has some inherent properties such as high micro-porosity, high density, low attrition loss, intrinsic hardness, low ash content, etc.

Impregnation Using H_3PO_4

10 g of AC was impregnated with 40% w/v of H_3PO_4 solution. The conical flasks containing the mixture are enclosed with aluminium foil. A few holes were made in order to let the vapours to evaporate at 30^{o}C. Then, the mixture was set for three days to dissolve the solution by using shaking water bath at 30^{o}C. The chewy liquid formed was further dried at 120^{o}C for 24 h in the oven. After that, the impregnated AC was weighed and put into a muffle furnace [11-16].

Procedure of Modification of AC

The impregnated AC was placed in a muffle furnace (Carbolite RHF 1500) at 500^{o}C for 2 h for carbonization. After the AC heating, the AC cooled to room temperature. Then, the MAC was refluxed 3 times for 3 h to remove any inorganic impurities such as metal ions, on the surface. Lastly, the MAC was dried process at 110^{o}C for 24 h [11-16].

Determination of Percentage Yield

The percentage yield (PY) of the MAC can be obtained by obtaining the final mass of the impregnated AC at the end of the activation process divided by the initial mass of AC. This can be achieved using the following equation:

$$\text{PY}\ (\%) = \frac{\text{Final weight of AC (g)}}{\text{Initial weight of AC (g)}} \times 100 \qquad (1)$$

Determination of Moisture Content

The moisture content (MC) in MAC was obtained by using method recommended by SIRIM [25]. Approximately 1.0 g of MAC was placed in the drying oven for 2 h at 110^{o}C. After that, the MAC was cooled down to room temperature and weighted. The following equation was used:

$$\text{MC}\ (\%) = \frac{\text{Initial weight (g)} - \text{Final weight (g)}}{\text{Initial weight (g)}} \times 100 \qquad (2)$$

Determination of pH

The pH of MAC was determined using pH meter. 200 ml Erlenmeyer flask was used to mix 1.0 g of MAC with 100 ml of distilled water. Then, the mixture was heated under gentle boiling. Then the mixture was cooled to room temperature. The mixture was diluted with 100 ml of distilled water. Finally, after a quick stirring, the pH was determined immediately.

Scanning Electron Microscopy with Energy Dispersive X-ray Spectrometry (SEM-EDX) Analysis

In general, SEM was an instrument which applied a narrow electron beam to scan over the surface of any specimen which coated with a thin layer of metal. The morphology, pore structure, and structural changes of AC were observed via SEM analysis which was coupled with energy dispersive X-ray spectrometer (EDX). Therefore, to determine the characteristics of the MAC, SEM-EDX analysis was used.

Fourier Transform Infrared Spectroscopy (FTIR) Analysis

For this study, the AC was modified via chemical activation using H_3PO_4. Therefore, the alterations in the functional group were compared. As for analysis, both AC, unmodified and modified was analysed to identify the surface functional group. Only a small amount of dried AC, both modified and unmodified, was tested. According to Shaarani and Hameed [26], the spectra were recorded within the range 400 to 4000 cm^{-1}.

Effect of Contact Time

To study the effect of contact time to adsorption of UMO onto AC, the time was varied, ranging from 30 to 180 minutes with constant pH value (5.42 for AC and 2.37 for MAC), and temperature at 30^{o}C.

Effect of Temperature

As to study the effect of temperature for adsorption process, the UMO was heated until a certain temperature ranging from 30°C to 100°C. After that, the AC (pH value – 5.42 for AC, and 2.37 for MAC) was mixed with the heated sample for the adsorption process to take place with contact time ranging from 30 to 90 minutes.

Effect of pH

To study the effect of pH in oil and grease adsorption onto AC, the pH value of the MAC was adjusted to be in the range of acidic to alkaline by going through washing process with sodium hydroxide (NaOH).

Reusability of MAC

Based on the method reported by Ngarmkam *et al.*[27], the AC and MAC can be recovered by using the Soxhletextraction with n-C_6H_{14} as the oil-extraction solvent. The samples were dried for 2 h at 130°C. Then, the samples were transferred to the extraction unit containing n-hexane. After the extraction takes place, the AC and MAC could be used to determine the reusability where 1 g of AC was mixed with 10 ml of UMO for 30 min. Then, the same steps as mentioned earlier were repeated for three times to determine the reusability of AC and MAC.

Adsorption Equilibrium Study

Adsorption equilibrium could be achieved if the amount of adsorbed solute was equal to the amount of desorbed solute. It described the interaction between both adsorbate and adsorbent. The graph of solute phase concentration q_e against liquid phase concentration C_e of solute was plotted to determine the equilibrium adsorption isotherms. According to Gottipati [28], the most commonly used models are Langmuir isotherm and Freundlich isotherm. Therefore, Langmuir isotherm and Freundlich isotherm were studied. The adsorption kinetic study was applied to determine the efficiency of the adsorption process. Kinetic models were applied to investigate the experimental data and to determine the adsorption mechanism. The most widely used models are pseudo-first-order and pseudo-second-order kinetic models. Thus, in this study, pseudo-first-order and pseudo-second-order models were applied. The values are reported with an average deviation of ±0.2.

RESULTS AND DISCUSSION

Percentage Yield

Table 1 shows the data obtained based on the impregnation to the AC. The PY of MAC which was impregnated with a ratio of 1:2 with 40% H_3PO_4 was 63%. According to Girgis *et al.* [29], the usual percentage yield as a result of modifying with H_3PO_4 is 35-50%. The purpose of H_3PO_4 is to constrain the discharge of volatile matter and to produce a higher yield of AC. Higher concentration of H_3PO_4 will result in decreasing of percentage yield as reported by Wan Nik *et al.*[28]. They have indicated that H_3PO_4 usually acts as a dehydrating agent which may inhibit the formation of tar and other liquids which might fill the pores of samples during the activation process. In general, the percentage yield of MAC is quite high, and it signifies the ability of H_3PO_4 to reserve carbon and to prevent the loss of volatile matter [31].

Moisture Content

Based on

Table 1, the moisture content of MAC obtained is 37%. In our previous studies, we reported that there is no significant effect of impregnation ratio and activation temperature to the moisture content of AC [11-19]. They also have reported that moisture content of AC is one of the aspects which influences the adsorption capacity as high moisture content will decrease the adsorption capacity. However, Madhavakrishnan *et al.*[32] has proposed that there is no correlation between moisture content and the adsorption power of AC.

pHValue

The pH value of AC is mainly based on the inorganic substances in the source material or which has been added during production. Based on the result, the acidic pH value of MAC is 2.37. According to Puziy *et al.* (2002) [33], this might be due to the formation of phosphorus-containing compounds such as polyphosphates during the impregnation process. It has been reported that MAC with pH value below 8 is more likely to be used for adsorption since it has higher adsorption

Table 1. Impregnation ratio, temperature, percentage yield, moisture content and pH of MAC

Impregnation ratio, X_p	1:2
Activation temperature, °C	500
Percentage yield (%)	63.00
Moisture content (%)	37.00
pH	2.37

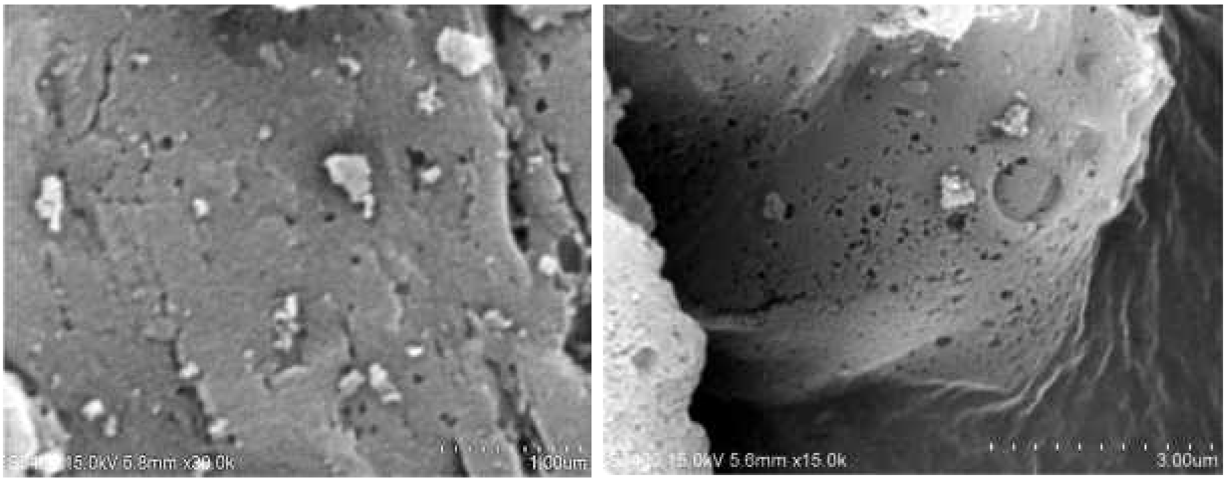
capacity [16-19, 34]. Gottipati [28] also has reported in his study that AC modified with phosphoric acid with pH value ranging from 2-6 has the maximum adsorption capacity.

SEM with EDX Analysis

In this study, SEM-EDX was applied to study the morphology and the pore size of AC and MAC. Based on the SEM images, the dark spots which were considered as the pores could be seen on the surface of both AC and MAC. Figure 1(a) shows the SEM image of unmodified AC. It can be observed that the surface of AC is smooth and has small pores on it. Some salt particles can be found on the surface structure of the AC. This may be due to some metal compounds which are not fully removed from AC [16-19, 34].

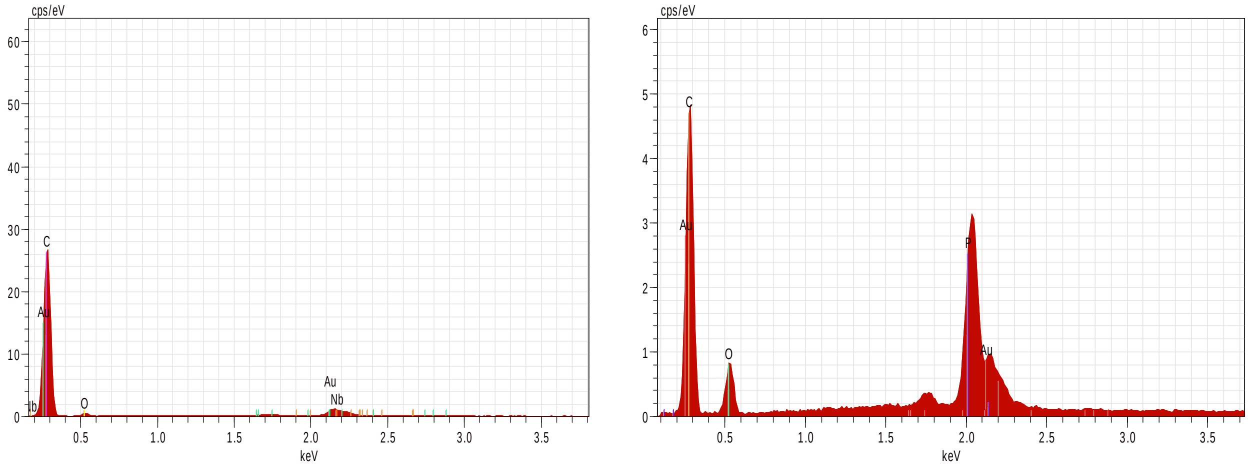
Figure 1(b) shows the surface structure of MAC which has been modified with 40% w/v H_3PO_4 with activation temperature of 500°C. In terms of structure and size of pores, observations of MAC through SEM are commonly known to be easier and traceable. These pores are considered as medium to the microporous network [28]. Based on the observation, it can be seen that the number of pores on MAC is higher compared to AC. This is because the pores on MAC are more visible than on AC. Based on the SEM image, salt particles (white particles) can be found on the surface of MAC as well. This is probably because of some metal compounds or excess H_3PO_4 which are not fully removed from the MAC [20-22].

Figures 2(a) and 2(b) show the EDX data obtained for both AC and MAC. The gold (Au) element detected on the surface of both AC and MAC is due to the sample coated with Au before characterization. Based on the results obtained, the phosphorus element can be found on MAC since it has been chemically modified with H_3PO_4. The other elements, such as carbon and oxygen, can be found on the surface of both AC and MAC. Thus, the modification of activated carbon with H_3PO_4 could be considered as successful based on the results obtained.



(a) (b)

Figure 1. SEM image for (a) AC; and (b) MAC.



(a) (b)

Figure 2. EDX data of (a) AC; and (b) MAC.

FTIR Analysis

In general, FTIR analysis is used to characterize the structure and functional groups that can be found in both AC and MAC. Figure 3 and Figure 4 show the FTIR spectra for both unmodified AC and MAC at the activation temperature of 500°C, respectively. Based on the result obtained, the spectra shared approximately the similar peak area. Some small peaks are detected at the spectra at 3800-3600 cm^{-1} bandwidths which indicate O-H stretching vibration of hydroxyl functional groups in low concentrations. This is due to the acidic treatment which reduces the hydroxide groups and produces more acidic groups on the surface of MAC [35]. The peak found at 2320 cm^{-1}, and 2330 cm^{-1} correspond to the C≡C stretching vibrations in alkyne groups [26].

The small peak at approximately 1700 cm^{-1} bandwidth represents the C=O stretching of carbonyls vibration such as ketones, aldehydes, lactones and carboxylic groups [36]. The peak formed at 1540 cm^{-1} is attributed to C=C stretching. This is because of the tars formed during depolymerisation of cellulose, followed by dehydration and condensation which contribute to the development of more aromatic and reactive products with some cross-linking [23]. Also, peaks formed at 800-700 cm^{-1} may be due to the presence of C-H out of plane bending in benzene derivatives [6]. Even though the functional groups found are similar, MAC could be classified as well developed in porosity which could increase the percentage of adsorption of motor oil compared to unmodified AC.

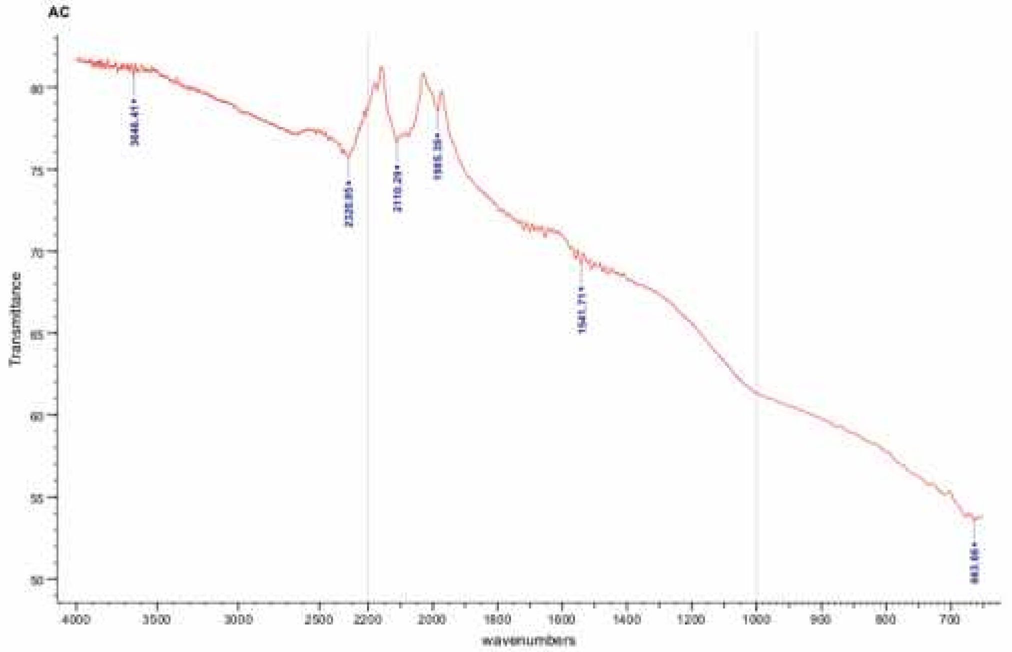


Figure 3. FTIR spectra of AC.

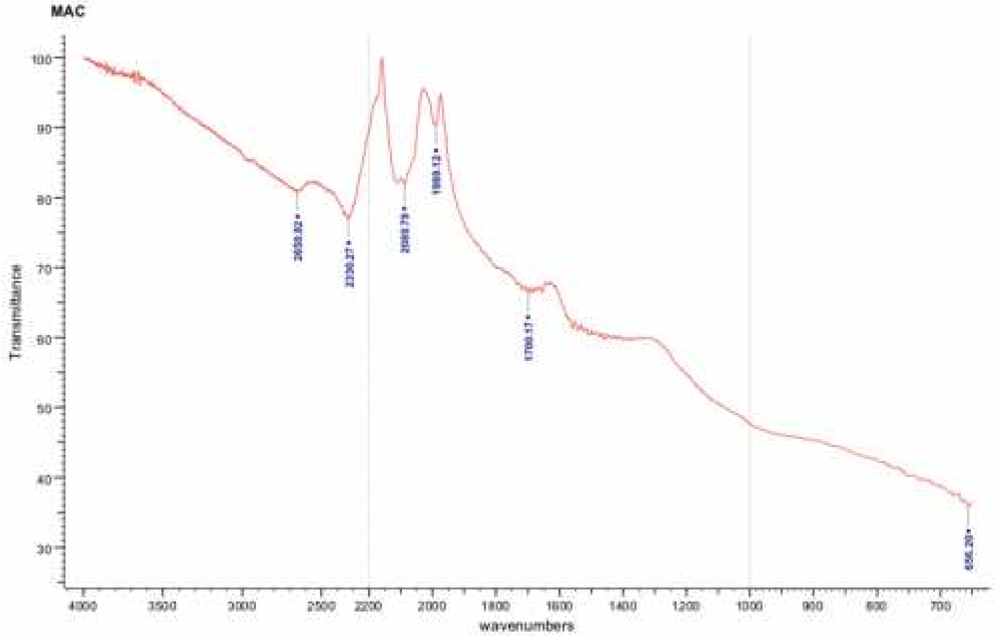


Figure 4. FTIR spectra of MAC.

Table 2. Effect of contact time on oil adsorption onto AC and MAC.

Time (min)	Weight of oil adsorbed (g)		Percentage of adsorption (%)	
	MAC	AC	MAC	AC
30	0.90	0.30	10.98	3.66
60	1.60	0.80	19.51	9.76
90	2.40	1.60	29.27	19.51
120	3.30	2.10	40.24	25.61
150	3.70	2.50	45.12	30.49
180	4.30	2.90	52.44	35.37

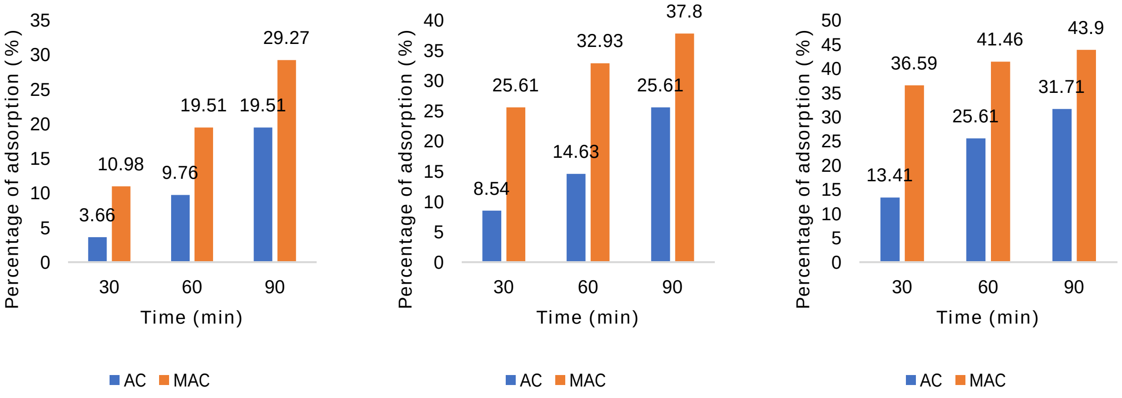
Effect of Contact Time

The study on the effect of contact time to the amount of motor oil adsorbed was studied. The comparison between the ability of AC and MAC to adsorb the UMO was also made. Table 2 shows the weight of oil adsorbed by AC and MAC. Based on the result, generally, the longer the time allocated for oil to be adsorbed on to the AC and MAC, the higher the amount of oil is adsorbed. The result showed that MAC had the higher percentage of adsorption compared to AC, where at 180 min, the percentage of adsorption by MAC was 52.44% while adsorption by AC was 35.37% only. This was due to larger pores of MAC compared to AC. This resulted in a higher adsorption capacity of MAC [16-19].

Effect of Temperature

Figure 5 shows the comparison between AC and MAC on the effect of temperature on the percentage of adsorption for 30°C, 50°C, and 100°C. The experiments were done in the temperature range of 30°C to 100°C at a constant adsorbent dose of 1.0 g, 10 ml of UMO, fixed pH at 5.42 for AC and 2.37 for MAC, and the contact time range of 30 to 90 min. Based on the results obtained for adsorption onto AC, 100°C show the highest percentage of UMO adsorbed, followed by 50°C and 30°C. As for MAC, the results obtained show the same trend as adsorption onto AC in which 100°C shows the highest percentage of adsorption compared to 30°C and 50°C. Gottipati [28] also showed the same trend in which the percentage of adsorption increased when temperature increased. As the contact time rose from 30 min to 90 min, it would also contribute to the increasing of percentage of adsorption onto the AC since the time taken for oil to be adsorbed was increasing. The maximum adsorption capacity was found to be 3.951 g oil/g (MAC).

Based on the result, it was clear that MAC had a higher percentage of adsorption compared to AC. This was due to the number of pores and pore sizes found on MAC were higher and bigger compared to AC. Thus, the adsorption capacity was increased. In conclusion, it could be said that the increase of the percentage of adsorption of AC and MAC showed that it was an endothermic process. This is because, when the temperature increased, the percentage of adsorption also increased which indicates that the diffusion rate of solute within the adsorbent pores is higher [28, 34].



(a) (b) (c)

Figure 5. Effect of temperature on percentage of adsorption for (a) 30°C; (b) 50°C; and (c) 100°C.

Effect of pH

Figure 6 shows the effect of different pH of MAC on the percentage of oil adsorbed. The initial pH for MAC is 2.37. The MAC is then washed with NaOH to obtain pH neutral and alkaline. The pH obtained for neutral was 7.66, while for alkaline, the pH of MAC was 11.08. A series of experiments were done with a fixed amount of adsorbent which is 1.0g of MAC with 10 ml of used motor oil, and contact time range of 30 min to 90 min.

Based on the result obtained, MAC with neutral pH had the lowest percentage of adsorption, followed by alkaline MAC. The acidic MAC, which had been modified with H_3PO_4 showed the highest percentage adsorption even with different range of contact time. The data obtained for the effect of pH on the adsorption process share a similar trend with a research done by Gottipati (2012). Basically, acidic MAC had more visible pores and bigger in size. Therefore, the adsorption performance of acidic MAC was higher compared to neutral and alkaline MAC.

Reusability of AC

Table 3 shows the data obtained from the reusability study for both AC and MAC. This study was done to determine the ability of MAC to be regenerated and reused in comparison with unmodified AC. The experiments were done in which the used motor oil adsorbed by both AC and MAC were extracted using Soxhlet extraction method with n-C_6H_{14} as a solvent. The experiments were done for three cycles. Based on the result obtained, it showed that both AC and MAC could be reused. However, MAC could still adsorb higher amount of UMO compared to AC. Throughout the cycles, the percentage of UMO adsorbed was gradually decreased since the pores were occupied by oil.

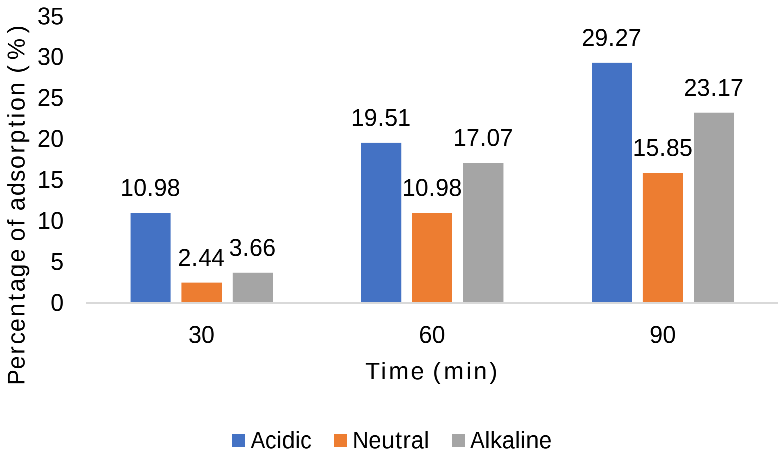


Figure 6. Effect of pH of MAC on the oil adsorption.

Table 3. Comparison of reusability of AC and MAC.

Reusability (cycle)	Weight of oil adsorbed (g)		Percentage of adsorption (%)	
	AC	MAC	AC	MAC
1	0.700	1.300	8.536	15.854
2	0.690	1.250	8.414	15.244
3	0.692	1.270	8.439	15.488

Adsorption Equilibrium

Generally, Langmuir isotherm is applicable to homogeneous sorption in which the sorption of each sorbate molecule onto the surface has equal sorption activation energy. The adsorption data obtained from the experiments were analysed according to the linear form of the following equation:

$$\frac{C_e}{q_e} = \frac{1}{K_L} + \frac{a_L}{K_L} C_e$$

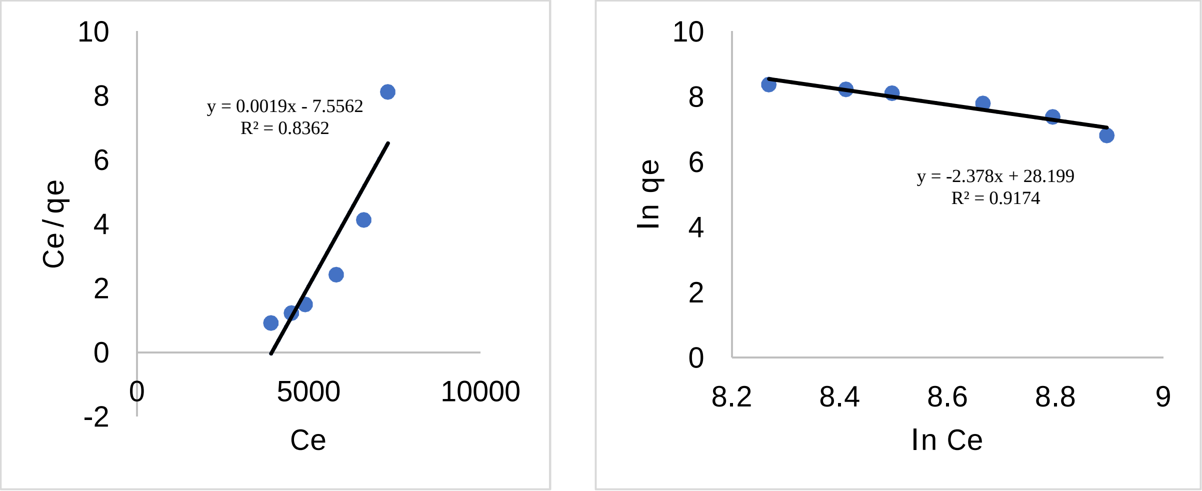
The most significant multilayer adsorption isotherm for heterogeneous surfaces is the Freundlich isotherm in which the heterogeneity factor 1/n characterized it. According to Gottipati [28], Freundlich isotherm is an empirical equation based on an exponential distribution of adsorption sites and energies. The graph was plotted based on the linear equation form:

$$\ln q_e = \ln K_F + \frac{1}{n} \ln C_e$$

Figure 7 shows the adsorption equilibrium of Langmuir isotherm and Freundlich isotherm, respectively. The adsorption of UMO proved to fit better to Freundlich isotherm compared to Langmuir isotherm with R^2 value of 0.9174.

Table 3 shows the Langmuir and Freundlich isotherm constants obtained from this study.

The heterogeneity factor, n value is 0.42 from Freundlich isotherm which indicates that the equation is poor. If the value of n is less than 1, it indicates that the adsorbent has poor adsorption characteristics. Also, K_f value in Freundlich isotherm indicates the intensity of the adsorption, in which the value obtained is 1.765. The regression coefficient, R^2 value of Freundlich isotherm is higher than Langmuir isotherm, where the values are 0.9174 and 0.8392, respectively. Therefore, this adsorption of used motor oil onto MAC is more likely to follow Freundlich isotherm.



(a) (b)

Figure 7. Adsorption equilibrium of UMO on MAC for different contact time (a) Langmuir isotherm, and (b) Freundlich isotherm.

Table 3. Langmuir and Freundlich isotherms constant.

Isotherm parameters	MAC
Langmuir	
[illegible]/g)	0.1323
∝[illegible] (L/mol)	2.514×10^{-4}
[illegible]	0.8392
Freundlich	
[illegible]mol/g)(mol/l)^{12}]	1.765
1/[illegible]	2.378
[illegible]	0.9174

Adsorption Kinetic

Adsorption kinetic study was done to study the adsorption efficiency. The models that were studied are pseudo-first-order and pseudo-second-order. The integral form of pseudo-first equation is as follows:

$$\log(q_e - q_t) = \log q_e - \frac{k_1}{2.303}t$$

Pseudo-second-order kinetic model could be expressed as:

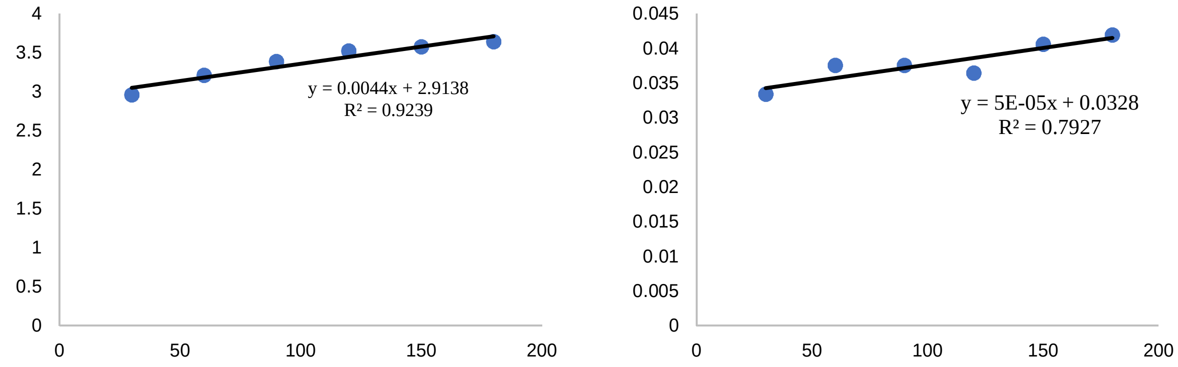
$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{1}{q_e}t$$

Figure 8 shows the adsorption kinetic study of UMO on MAC for different contact time.

Table 4 shows the kinetic parameters data calculated from the Figure 8 for pseudo-first- order and pseudo-second-order models. Both kinetic models showed a good fit of kinetic to the data. However, the regression value for pseudo-first order was higher compared to pseudo-second-order which was 0.9239 and 0.7927, respectively. This indicates that the R^2 value is close to unity and the adsorption is more favourable to follow pseudo-first-order kinetic model [11-19].

CONCLUSION

From the characterization of MAC, the number of pores found on the surface of MAC was higher than AC. It was also found that the pore size of the MAC was higher than AC. The pores on MAC were more visible than that of AC. The functional groups found on MAC were similar with AC. However, MAC had well-developed porosity which could help the adsorption power of used motor oil onto MAC. As for the effect of contact time was concerned, we found that as the contact time increased, the amount of oil adsorbed onto the AC also increased for both AC and MAC and as the temperature increased, the percentage of adsorption of UMO also increased for both AC and MAC. However, MAC showed a higher percentage of adsorption capacity compared to AC due to its characteristics that was mentioned earlier. The effect of pH on the adsorption of oil onto MAC was also studied. It showed that acidic MAC had the higher percentage of adsorption, followed by alkaline and neutral MAC. The reusability of MAC was also studied. Based on the



(a) (b)

Figure 8. Adsorption kinetic of UMO on MAC for different contact time (a) pseudo-first order; and(b) pseudo-second order.

Table 4. Pseudo-first-order and pseudo-second-order parameters.

Constant	MAC
Pseudo-first-order	
[illegible]1/min)	0.0101
[illegible]mg/g)	819.9738
[illegible]	0.9239
Pseudo-second-order	
[illegible]g/mg.min)	7.6219×10^{-8}
[illegible]mg/g)	20,000
[illegible]	0.7927

results, both MAC and AC could be reused. However, MAC had the ability to be reused more than the AC. The study of adsorption equilibrium and adsorption kinetics of adsorption of UMO onto MAC was done. The adsorption data of MAC was more likely to follow Freundlich isotherm and pseudo-first-order kinetic model.

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