



Utilisation of rice straw activated carbon in treatment of POME using adsorption method

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Abstract

Rice straw activated carbon (RSAC) was produced and used to decolourise organic dyes and palm oil mill effluent (POME) via adsorption processes. Rice straw was carbonised and activated using potassium hydroxide (KOH) and sodium hydroxide (NaOH) to produce activated carbon. The properties of activated carbon (AC) were studied through surface and morphology analyses. The regeneration of the adsorbents by adsorption/desorption processes shows that the AC can be regenerated up to five cycles while still providing adsorption capacity at maximum of 67.26% at first regeneration and as low as 2.72% at fifth regeneration. The RSAC was also used for the treatment of POME for decolourisation and the reduction of total organic carbon (TOC) as well as chemical oxygen demand (COD). This study suggested that the RSAC can be potentially used as an adsorbent for the removal of different types of positives charges adsorbates and POME from aqueous solution.

Keywords: rice straw, POME, KOH, NaOH, adsorption

Introduction

The processing of palm oil produces two types of by-products, namely solid biomass and liquid sludge known as POME. Per tonne of oil palm fruit processing, about 5.5% of oil kernel, 13.5% of mesocarp fibre and 23.4% of oil palm fruit bunches are produced as solid biomass waste. In addition, approximately 67% of POME is also produced (Eleanor et al. 2018). POME is produced from three (3) manufacturing activities namely sterilisation (0.2 – 0.25 tonnes/t of oil palm fruit), clarification (0.4 – 0.45 tonnes/t of oil palm fruit) and hydrocyclone (0.03 – 0.05 tonnes/t of oil palm fruit) at a ratio of 6:15:1 (Ma 1999).

The characteristics of POME depend on the quality of the oil palm fruit and the manufacturing activities at the palm oil mill. It is a concentrated, viscous and brownish liquid that consists of 95 – 96% water, 0.6 – 0.7% oil and 4 – 5% solids. Besides that, it has value of BOD (>18,000 mg/L) and COD (> 45,000 mg/L), which are drained at a temperature of 80 – 90 °C at acidic pH. It also contains nitrogen, potassium, magnesium, calcium, chromium, copper, iron, organics, phosphorus and cellulose waste content which is a mixture of carbohydrates and oils. The high organic content in POME

is contributed by some sugars such as xylose, glucose, galactose and mannose (Ohimain et al. 2012; Singh et al. 2010). POME is not a harmful contaminant because no chemicals are included during oil palm fruit processing, it contains a good source of nutrients, however untreated POME affects the environment because its organic and nutrient content causes a reduction in oxygen content in aquatic systems (Kamyab et al. 2014). There are several methods of treating POME namely membrane technology (Amosa 2017; Ghani et al. 2018), coagulation/flocculation (Poh et al. 2014; Zahrim et al. 2017), oxidation process (Ng et al. 2016) as well as adsorption using AC (Abdulsalam et al. 2018; Alkhatib et al. 2015; Kaman et al. 2017).

Common practice in POME treatment is through pond systems, which require large soil areas and long exposure times (Kaman et al. 2017). In addition, treated POME still contains high levels of contaminants due to the low efficacy of such treatment methods. Therefore, adsorption is often used to treat treated POME prior to discharge into rivers to meet wastewater discharge standards. There are limited studies on the use of AC adsorbents from biomass in treating POME as shown in *Table 1*.

AC tends to interact with non-polar particles on the adsorbed material in the hydrophobic solution (Adeleke et al. 2016). Hydrophobic particles have a tendency to interact with similar particles as compared to oil droplets. This process results in the breaking of hydrogen bonds between water particles. Similarly, hydrophilic adsorbents can be effective for the reduction of polar solutions in wastewater. Combinations of hydrophilic particles have better thermodynamic interactions than interactions with hydrophobic ones such as oils. The level of hydrophobic or hydrophilic particles is determined by the surface tension of the materials in the aqueous phase and the combination of adsorbent materials can be very effective for the reduction of high strength wastewater containing both polar and non-polar solutions (Adeleke et al. 2016). POME was also analysed by total organic carbon (TOC) and chemical oxygen demand (COD). TOC aims to determine the amount of carbon present in organic matter. TOC is a non specific determinant of water quality. While COD is able to determine the amount of oxygen consumed by the reaction in the solution under study.

Regeneration of the adsorbent is able to reduce the cost of producing the adsorbent after the adsorbent is able to be removed. Nevertheless, the potential for regeneration of adsorbents is one of the distinctive properties of adsorbents. The adsorption ability and efficiency of the adsorbed material are among its advantages. A decrease in adsorption capacity was found to occur when the regeneration cycle increased. AC regeneration requires a specific procedure to remove the molecules of the adsorbed material from the AC surface. There are three main methods of regeneration for AC namely thermal, chemical and biological (Foo and Hameed 2012; Shah et al. 2013; Regti et al. 2017; Yu et al. 2017; Sanyirek and Yonten 2018). Each method has its limitations such as high thermal energy, carbon loss, use of expensive chemicals and longer time. Regeneration of the adsorbent after saturation is done by the adsorption process. Adsorption studies are necessary for the recovery of adsorbed material and to understand the adsorption mechanism. The adsorption process is performed by mixing saturated adsorbents with adsorbents (NaOH, HCl, CH₃COOH, C₃H₆O, C₂H₅OH) for removal of adsorbed materials and separated by filtration (Salleh et al. 2011; Berenguer et al. 2010; He et al. 2013; Lata et al. 2014).

The adsorbed dye can be adsorbed using neutral water, the dye is adsorbed with the adsorbent through a weak bond. Methylene blue (MB) that used in this study was cationic dyes that have a positive charge on the molecule and is also known as a basic dye. This dye is capable spread colour and dissolve in water (Ahmed 2016). If the dye can be absorbed with sulfuric acid or alkaline water, the dye adsorption through ion exchange. The absorption of dye via organic solvents such as acetone or acetic acid can be done through chemical adsorption process. Whereas with mineral acids such as HCl, the dye can be adsorbed through physical adsorption (Bharathi and Ramesh 2013). Hydrophobic functional groups such as methyl in organic solvents are able to be mobilized by etching forces to deactivate cationic MB molecules that are also hydrophobic on the surface of AC (Gerard and Orjan 2005). Therefore, this study is aim to analyse the surface and morphology of RSAC. The regeneration of the rice straw activated carbon (RSAC) by adsorption/desorption processes also be studied. The RSAC was also used for the treatment of POME for decolorisation and the reduction of total organic carbon (TOC) and chemical oxygen demand (COD).

Materials and methods

Preparation of rice straw

Rice straw was taken from a paddy field in Sekinchan, Selangor, Malaysia. It was cleaned with water to remove dirt and was oven dried at 105 °C for an overnight.

Preparation of AC from rice straw

The rice straw was pyrolysed at 400 °C for 4 h to produce rice straw charcoal (RSC). Then, RSC was sieved to collect the particle with 60 mesh in size. The RSC was immersed with the KOH solution (13 M) at a ratio of 1:4 (RSC: KOH or NaOH solution) for 24 h. It was filtered and oven dried overnight at 105 °C. The activation process was done in a tube furnace with a nitrogen gas flow (100 mL/min) for 2 h at 850 °C. Sample was cooled and washed with hydrochloric acid (HCl) solution (1 M) and deionised water until a pH of 6 to 7 in the rinse was obtained. The above procedure was also performed for NaOH solution. The samples

Table 1. POME adsorption using AC adsorbents

Biomass	Pollutants test	Disposal efficiency (%)	Reference
Coconut shell AC (steam method)	TSS, COD and POME colour	TSS (39%), COD (66%), colour (61%)	(Kaman et al. 2017)
Oil palm mesocarp fiber bioadsorption (steam method)	COD and SS POME	SS (80%), COD (70%)	(Ibrahim et al. 2017)
Banana skin AC	TSS, BOD, colour	TSS (96%), BOD (100%), colour (95%)	(Mohammed and Chong 2014)
Commercial AC	COD and TSS	COD (73.08%), TSS (98.33%)	(Parthasarathy et al. 2016)
Oil palm skin AC	SS	71.26%	(Azmi and Yunus 2014)

were labelled as RSC (rice straw charcoal). Meanwhile, RSACK and RSACNa meaning the RSC activated with KOH and NaOH, respectively.

Characterisations of the AC samples

The RSC, RSACK, and RSACNa samples surface area were studied using N₂ adsorption-desorption analysis (Micromeritics ASAP, 2010) then followed by the nitrogen adsorption-desorption standard at 77 K. The samples were degassed prior to analysed for 10 h under vacuum at 110 °C. Surface morphology properties of the samples were obtained using scanning electron microscope (SEM) of the FEI model Quanta 400.

Study of discoloration, COD and TOC POME

Oil palm industry effluent (POME) was obtained from KPSB's oil palm plant in Gua Musang, Kelantan, Malaysia. The properties of the original POME are as in Table 2. The POME was first filtered to remove dirt and sediment and stored in the refrigerator before undergoing discoloration analysis with AC. POME (pH 8.5) performed a decolorization analysis (Equation 1) using the Platinum-Cobalt (Pt-Co) method analysed by a Spectrophotometer (HACH DR 3900) at a wavelength of 450 nm. Analysis was carried out with 100 mL of POME stirred with AC adsorbents at 0.1 g, 0.4 g and 0.6 g and the Pt-Co values of POME were taken at set intervals until the equilibrium point of POME color adsorption was reached. The analysis of chemical oxygen demand (COD) and total organic carbon (TOC) followed the 5220D standard method and the TNT method, respectively, which were analysed with the HACH DRB 200 instrument.

Discoloration (%) = $\frac{(C_o - C_e)}{C_o} \times 100$ (1)
Where C_o and C_e are the initial and final discoloration values of POME, respectively.

AC adsorption and regeneration study

Methylene blue (MB) solution with a concentration of 450 mg/L was prepared for the adsorption and dehydration study process of MB with RSACK and RSACNa samples. AC powder weighing 0.05 g was stirred together with 50 mL of MB solution using magnetic stirrer at 300 rpm overnight. The concentration of the MB solution before and after the adsorption process was obtained using a UV spectrophotometer. The AC powder that has undergone the adsorption process was filtered from the MB solution and rinsed with distilled water to clean the residue of the MB solution on the surface of the AC. The dehydration process by chemical method was carried out by soaking AC in a solution of dehydration agent (3 M) for 3 h. The three types of dehydrating agents are acetone (CH₃H₆O), acetic acid (C₂H₄O₂) (both are organic solvents) and sulfuric acid (H₂SO₄) (inorganic solvents). The concentration values of the absorbed solution were taken with a UV spectrophotometer. The AC was filtered and rinsed with

Table 2. The properties of the original POME

Parameter	Value
pH	9.5
DO (mg/L)	7.32
BOD5 (mg/L)	11.5
COD (mg/L)	280
SS (mg/L)	17
Color (Pt-Co)	888
NH3-N (mg/L)	0.5
Oil and Grease (mg/L)	5
TOC (mg/L)	96

distilled water before being dried overnight in an oven at 60 °C. The above process is repeated for subsequent regeneration up to five (5) cycles. The percentage value of desorption of adsorbed material is obtained based on Equation 2.

Desorption percentage of adsorbate = $A/B \times 100\%$ (2)
where A is the concentration of the desorption solution (mg/L), B is the concentration of the adsorbate solution (mg/L).

Results and discussion

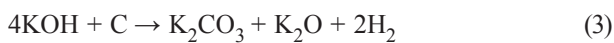
Table 3 shows that the RSACK and RSACNa samples had high surface area values at 1048.3 m²/g and 332.81 m²/g, respectively as compared to RSC which is 1.16 m²/g. These results may be driven by the effects of internal heating and pore opening (Foo and Hameed 2011a). The increase in surface area may be due to the release of volatile components during the activation process. KOH as an activating agent has reacted with the reactive centres of carbonaceous materials such as unstructured carbon, carbon with heteroatoms and carbon on the graphene side producing new pores and dilating existing ones, where it increases with increasing of KOH (Zhang et al. 2008). The total volume of the AC pores has increased with increasing activation temperature. For example, the pore volume of RSC at 0.0028 cm³/g increased to 0.64 cm³/g for RSACK and 0.23 cm³/g for RSACNa. An increase in surface area contributes to an increase in meso pore sized volume (Sricharoenchaikul et al. 2008). The mean pore sizes of RSACK and RSACNa were 2.46 nm and 2.82 nm, respectively. It shows that the pores are between 2 – 50 nm in size, i.e., meso pore sized. According to IUPAC 1972, pore sizes can be classified into three types namely micro pores (below 2 nm), meso pores (2 – 50 nm) and macro pores (over 50 nm) (Lu et al. 2015).

The surface area value of RSACK is 1048.3 m²/g as compared to RSACNa 332.81 m²/g. It was found that KOH acts as a better activating agent than NaOH. It causes pore formation and pore volume and in turn produces porous carbon with a higher surface area (Lillo-Ródenas et al. 2001). These results are equivalent to a study by

Table 3. The results of surface analysis

Sample	Surface area (m ² /g)	Pore volume (cm ³ /g)	Average of pore size (nm)
RSC	1.16	0.0028	9.72
RSACK	1048.30	0.64	2.46
RSACNa	332.81	0.23	2.82

Guo et al. (2003). The increase in porosity was found to be related to the alkaline cations of the activating agent according to the sequence: Li < Na < K < Rb < Cs, in this case KOH has more alkaline cations than NaOH, therefore KOH reacts more with carbon to produce pores (Guo et al. 2003). The possible chemical reactions between alkali hydroxide and carbon occurs during the activation process as in *Equations 3, 4 and 5* (Chunlan et al. 2005).



The activation process produces hydrogen gas, carbonate and potassium (K or K₂O). Potassium metal is removed during the washing process and K₂CO₃ decomposes during the activation process and CO₂ gas is produced. The reaction between the activating agent and the carbon causes the decomposition of organic volatiles and forms the shaft surface of the AC sample. The process of producing gas during activation using an appropriate activating agent is very important in the process of pore formation (Muniandy et al. 2014).

Figure 1 (a) shows the SEM micrographs of RSC, which appear to have almost no pores on their surface. The effect of the presence of NaOH and KOH on the surface of the impregnation sample before the activation process is carried out and were shown in *Figure 1 (b)* and *Figure 1 (c)*, respectively. Chemical activation produces a porous carbon structure which opens pores on the surface of the air conditioner. Many pores are produced on high AC surface area values as discussed in the previous chapter. Similar results were also reported by Zhang et al. (2008); Cheenmatchaya et al. (2014) and Pandey et al. (2015). The difference of AC generated from this study compared to the above three researchers was at the ratio of carbon charcoal: activating agent which was 1: 4 (w/v) and the activation temperature (650 – 850 °C).

Figure 2 shows the effect of RSACK dose as an adsorbent on the percentage of POME dyeing. Increasing the dose of adsorbent has reduced the percentage of POME colour. A total of 75.73% (193 Pt-Co) of POME colour was reduced by using an adsorption dose of 1 g/L. The percentage of POME dye continued to increase by 89.33% (21 Pt-Co) to 99.62% (3 Pt-Co) as the dose of RSACK adsorbent increased from 4 g/L to 6 g/L as shown in *Figure 2 (a) and (b)*. Next, the increased of RSACNa dose showed an increased in the percentage of POME dilution which were 87.62% (90 Pt-Co) at 4 g/L, 96.79%

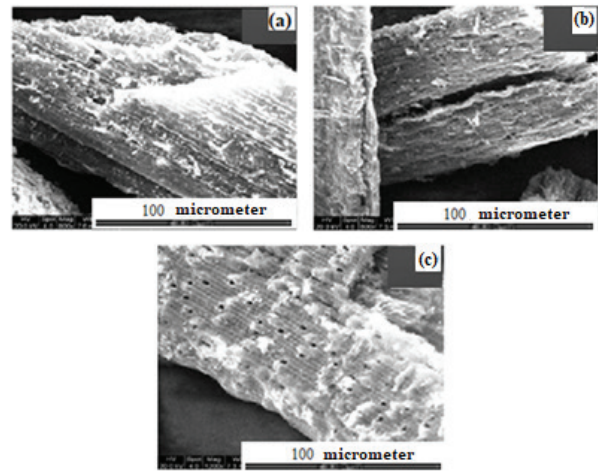


Figure 1. SEM image of (a) RSC, (b) RSACNa and (c) RSACK

(23 Pt-Co) at 6 g/L and 98.90% (8 Pt-Co) at 10 g/L as shown in *Figure 2 (c) and 2 (d)*. In the first stage, the adsorption increases rapidly, indicating that there is a large active area of the adsorbent surface. The results of the increase in the dose of the adsorbent cause an increase in the percentage of dyeing of POME where it is in line with the previous researchers' report (Alkhatib et al. 2015; Gobi and Vadivelu 2013). It is eventually showing a horizontal graph after reaching the equilibrium, possibly due to the mono-organic layer of contaminants covering the surface of the adsorbent (Mohammed 2013). Overall, it can be summarised that a dose of adsorbent at 0.4 g in 100 mL of POME is sufficient to dye POME.

It was found that the dyeing rate increased in line with the time and dose of adsorbent material (Mohammed and Chong 2014) where from the analysis, it was found that a balanced adsorption rate occurred after 420 min (1 g/L) for RSACK sample. The balance increased rapidly for the adsorption dose of 4 g/L at 60 min and 30 min for 6 g/L. RSACNa provides an equilibrium after 300 min for the adsorbent dose of 1 g/L and 4 g/L, while the dose of the adsorbent 6 g/L occurs after 60 min. This is because the RSACNa sample has a low porosity and causes a low adsorption capacity compared to RSACK which has a high value. Researchers such as Abdulsalam et al. 2018; AremuMuyibi et al. (2014); Kaman, Tan and Lim (2017) have reported similar results on the dyeing rate and the effect of AC dosing in the POME dyeing experiment using AC.

In general, AC-KOH samples showed a better percentage of staining than AC-NaOH. It is most likely due to the higher surface area factor formed on the surface of the sample that allows contaminated organic matter to fill it. This is supported by the pictures in *Figure 3 (a)*

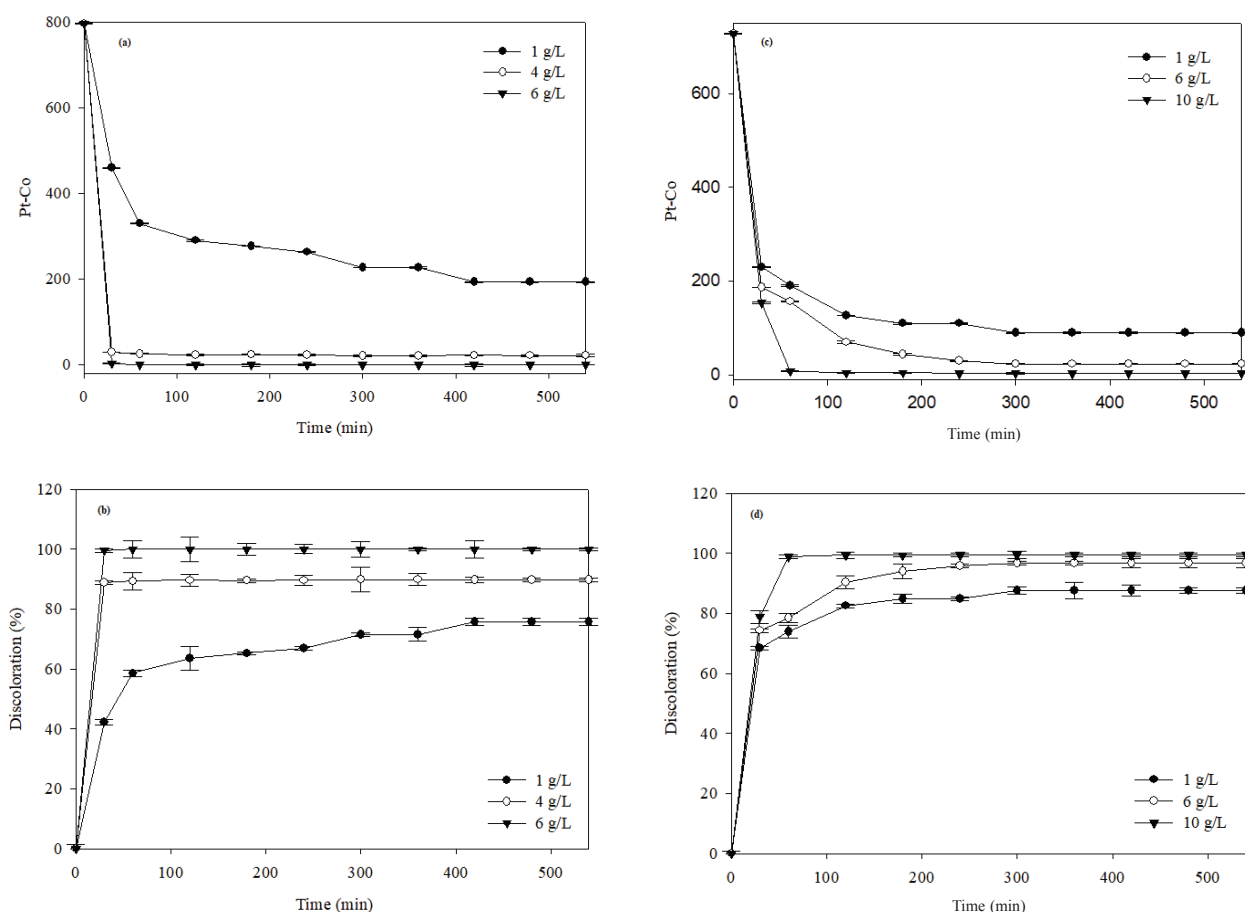


Figure 2. Colour graph (Pt-Co) and percentage of discoloration of RSACK (a) and (b) as well as RSACNa (c) and (d)

where the clarity of the RSACK sample is formed faster than the AC-NaOH sample (Figure 3 (b)). The use of RSACNa samples that have a low BET causes POME color adsorption to be less and slow to become bright. While the RSACK sample has faster adsorption and POME color change due to a higher porosity or BET.

Table 4 is the results of the analysis of chemical oxygen requirements (COD) and the amount of organic carbon (TOC) against POME before and after the adsorption experiment with 0.4 g of the adsorbent sample of RSACK and RSACNa samples. All four AC samples have successfully lowered the COD percentage from as low as 47.88% to as high as 72.82%. This is likely because AC is able to adsorb organic contaminants on active surfaces. The TOC test results also showed a decrease in the TOC value after the POME adsorption process with AC. The percentage loss of TOC value ranged from 73.33% to 95.24%. AC is able to reduce the content of contaminated organic matter by reducing the carbon and oxygen content in POME. According to Adeleke et al. (2017) COD POME has been successfully reduced by 89.60% and $\text{NH}_3\text{-N}$ 75.61% by using AC as adsorbent. Reduction of COD and TOC occurs during the adsorption process where the main area for adsorption takes place on the outer surface of the AC. If the adsorption occurs on the inner surface of the pore, then the adsorption process takes longer (AremuMuyibi et al. 2014). The RSACK sample was able to reduce the TOC value of POME compared to RSACNa

due to its higher BET or surface area. This allows more contaminants to be adsorbed. But the RSACNa sample was found to be quite different because it had a better COD value than RSACK even though it had low BET.

The study of Othman et al. (2013) found that AC at a dose of 10 g/L has caused a drastic reduction in COD and suspended solid (SS). This finding is consistent with a study by Siebdrath et al. (2012) in which AC is proven to reduce dissolved organic carbon (DOC). AC reduces organic matter through very fine pore size during adsorption process. As an adsorbent, AC has a porous surface area to trap organic molecules that contribute to reduced BOD and COD concentrations (Idris et al. 2013). AC can therefore be used as an initial treatment step leading to reduced clogging/coagulant material and better organic removal (Siebdrath et al. 2012).

The adsorption result and the adsorption percentage for RSACK in Figure 4 found that the increased number of regenerations has reduced the adsorption capacity and the adsorption percentage of the sample. The diffusion efficiency of AC by different adsorption agents results in different results. Percentage of MB adsorption on RSACK (Figure 4) with acetone adsorption agent was highest from 67.26 to 11.90% and adsorption capacity from 224.82 to 70.92 mg/g from first to five regenerations, followed by acetic acid with adsorption percentage from 39.23 to 10.45% and adsorption capacity from 174.31 to 75.52 mg/g, while the lowest was sulfuric acid with

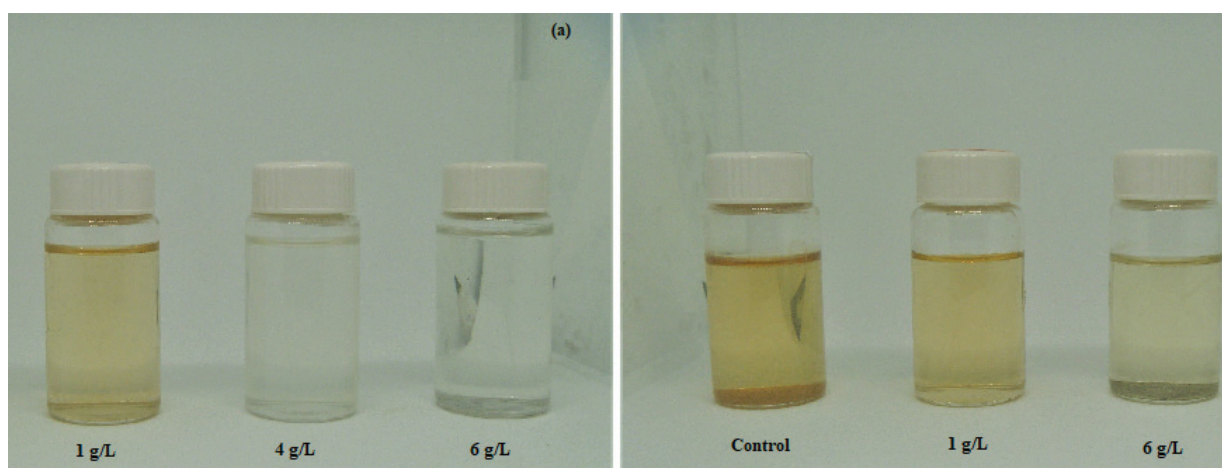


Figure 3. Adsorption of POME (a) RSACK, (b) RSACNa

Table 4. Results of COD dan TOC

Sample	COD (mg/L)	COD (%)	TOC (mg/L)	TOC (%)
POME (control)	401	-	105	-
RSACK	209	47.88	11	89.52
RSACNa	130	67.58	28	73.33

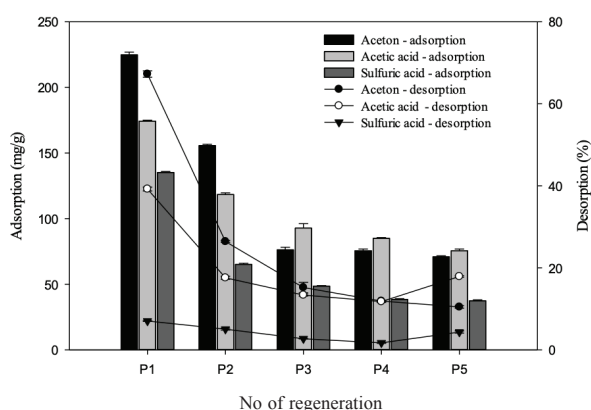


Figure 4. Adsorption results and percentage of RSACK desorption on regeneration analysis

adsorption percentage from 7.02 to 2.72% and adsorption capacity from 135.21 to 37.38 mg/g. The results show that the adsorption agent of organic solvents, namely acetone and acetic acid, is more efficient than the inorganic solvent of sulfuric acid on RSAC-MB. The adsorbed dye can be adsorbed using neutral water, so the dye is adsorbed with the adsorbent through a weak bond. The use of various adsorbents to adsorb dye on the surface of AC is possible through either ion exchange (sulfuric acid), chemical (acetone or acetic acid) or physical adsorption (HCl) (Gerard and Orjan 2005; Bharathi and Ramesh 2013). Since the acetone was the best to adsorb dye so this shows that adsorption chemistry is an AC-MB adsorption mechanism. Organic solvents are more efficient in the process of cationic dye adsorption on RSACK than inorganic solvents. The influence of the type of solvent/dehydrating agent is a causal factor to the dehydration

of AC-MB.

The molecular weight factor of the adsorption agent is likely to influence the results. The molecular weight of the adsorbent agent is smaller than that of the adsorbed material (MB) will provide the physical transfer (dissolution) of the MB molecule by the adsorption agent molecule (Lu et al. 2011). Acetone has a smaller molecular weight of 58 g/mol followed by acetic acid (60 g/mol) and sulfuric acid (98 g/mol) compared to MB of 319.86 g/mol. It is likely to produce a better percentage of adsorption by acetone than acetic acid and acid sulfuric.

Most organic solvents have hydrophobic groups ($-\text{CH}_3$, $-\text{CH}_2\text{CH}_3$) and hydrophilic ($-\text{OH}$, $-\text{CN}$, $=\text{O}$). Organic solvents have a hydrophobic group such as the methyl group ($-\text{CH}_3$) that will be adsorbed on the aromatic hydrophobic group of the carbon structure, while the hydrophilic group interacts with the functional group of the adsorbent material. Because the methyl group is more closely related to the carbon hydrophobic group, there is a driving force for the inactivation of MB molecules than filled AC-MB (Gerard and Orjan 2005). Acetone has a greater methyl group which makes it easier to carry out the aeration process. Xing et al. (2017) stated that the hydrophobic group of the adsorption agent will be more adsorbed than the hydrophilic group with the hydrophobic part of the AC, which in turn forms the impulse to the molecular process of adsorption of adsorbed material.

Studies of the adsorption change for RSACK samples showed an increase in the adsorption percentage after the temperature was increased from 25 °C, 40 °C to 50 °C (Figure 5). The first stage regeneration at 25 °C for adsorption percentage was 21.73% (adsorption 133.23 mg/g), 40 °C by 38.53% (adsorption 189.86 mg/g) and 50 °C increased to 44.29% (adsorption 206.14 mg/g). The adsorption percentage for the next cycle decreases with each temperature. In the fifth regeneration process, it was found that at 25 °C it was 2.23% (adsorption 69.12 mg/g), 40 °C by 8.18% (adsorption 101.28 mg/g) and 50 °C at 9.90% (adsorption 104.41 mg/g). The role of temperature is able to enhance the adsorption process. It is likely to reduce the viscosity of the adsorption agent and make it easier for the agent to enter the pore where the MB molecule is located. The increase in temperature

has increased the percentage of adsorption due to the decrease in viscosity of the adsorption agent and makes it easier to penetrate into the AC pores containing adsorbed material (Hassan and Elhadidy 2017).

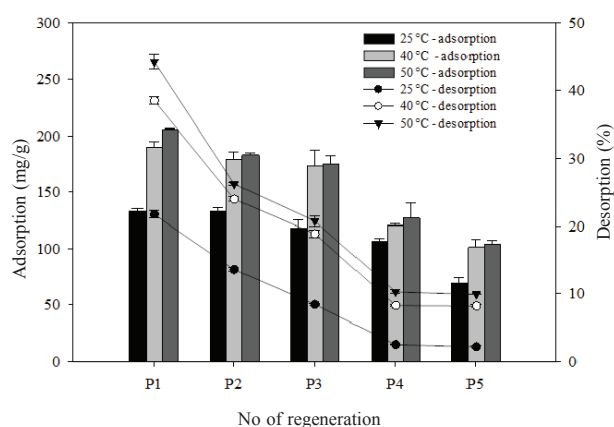


Figure 5. Adsorption results and percentage of RSACK dehydration in acetone with increasing temperature during the regeneration process

Conclusion

Increasing the dose of the AC adsorbent resulted in an increase in the percentage of POME discoloration. Dose of adsorbent (0.4 g) and time of 60 min was able to decolorise POME. AC-KOH showed a better decolorisation percentage than AC-NaOH due to its pore surface area factor. AC samples were able to lower COD 47.88 – 72.82% due to the ability of AC to absorb organic contaminants. The TOC value was lowered to 73.33 – 95.24% of the effect of AC which was able to reduce the presence of C and O in the organic matter content of POME. A five-round regeneration study of adsorption-desorption analysis of MB-AC found that desorption using organic solvent like acetone produced the best adsorption and desorption percentage for RSACK samples compared to using acetic acid and sulfuric acid. Among the three types of dehydration agents, acetone produced a higher percentage of dehydration of MB adsorbed on the AC surface followed by acetic acid and sulfuric acid. Molecular weight factors and hydrophobic groups on the dehydrating agent are likely among the reasons acetone is better. Temperature also plays a role in promoting the dehydration process where the viscosity of the dehydration agent is reduced and makes it easier to enter the pores where there are MB molecules for more dehydration process.

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