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Eradication of Oil Wastes Using Chempedak Stone Powder as Natural Adsorbent: Study on Adsorbent Dosage and Type of Water Sources Effect

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Abstract
Oil pollution has become a water pollution problem in the world. An eco-friendly adsorbent need to be produced to resolve this problem. This study focuses on removing oil waste using Chempedak Stone (CS) powder as a natural adsorbent. The purpose of the study is to observe the effects of adsorbent dosage and the effects of several type of water sources on adsorbing different types of oils (vegetable oil, lubricant oil and diesel oil). Different adsorbent dosage (treated and untreated) of 0.2g, 0.4g, 0.6g, 0.8g and 1.0g was examined in different types of water which are tap water, sea water and lake water. The treated adsorbent was introduced to 0.5M NaOH and 0.5M nitric acid (HNO_3). The CS was collected from a local market and went through several processes to become a powder such as rinsing, drying, crushing and sieving. The results show that the increasing adsorbent dosage decreased the percentage of oil removal. It was proven that the effectiveness of treated and untreated adsorbent depended on the type of oil and the type of water. The highest percentage of oil removal was found using untreated CS powder on vegetable while treated CS powder was effective for adsorbing lubricant oil at the dosage of 0.2g. The results thus showed that untreated CS powder has the most effective adsorbent to adsorb various types of oils on various types of water compared to treated CS powder.
Keywords: Chempedak Stone, Oil Removal, Adsorbent, Oil Pollution, Oil Treatment

Introduction

Oil is defined as a viscous liquid derived from petroleum, especially for use as a fuel or lubricant. It has wide applications such as for cooking, cosmetic industries, lubrication industries, fuel, painting and many more. The oil industry can be traced back to 1859 in the USA and as early as 600 BC in the Zoroastrian religion of Persia and India. The first oil drilled well was done by the Chinese in the year 347 AD (Prasad and Anuprakash, 2016). Since then, the demand for oil has kept increasing and has escalated more than it can be produced. It has been reported that by 2030, the world population oil demand will reach 105.5 MBPD. The global oil and gas industry value in 2009 was recorded at $2,119.3 billion while in 2014, the global oil and gas market forecasted a value of $3,192.8 billion. In 2014, the global oil and gas market was forecasted to have a volume of 41 billion BOE (Barrels of Oil Equivalent), an increase of 10.2 per cent (Prasad and Anuprakash, 2016). Hence, this shows the importance of oil to humans, and the reason why the production of oil is in very high demand.

Although oil has many uses, it can have an adverse effect on the environment such as pollution. Pollution can be defined as contaminated water, air and the environment with harmful or poisonous substances (Adeyi et al., 2019). The largest money earning industry in the world today is the oil and gas industry but, in fact, it is the dirtiest and the largest environment destructive industry in the world (Prasad and Anuprakash, 2016). Oil-water separation has recently become a worldwide challenge due to expanding industrial oil wastewater and frequent oil spill accidents. As such, the development of a low-cost, bio-based, and environmentally friendly material has been attempted to treat environmentally sensitive water pollution caused by oils (Li et al., 2016, Abdullah et al., 2016). Not only because of expanding industrial oil wastewater and oil spill, but human activities such as discharging oil improperly into the water stream has also contributed to pollution.

Large amounts of oil on any water streams give many adverse effects to the environment, human economy, tourism industry and to other living things. Oil spilled or found discharged in wastewaters, seas, rivers, lakes or any water streams can be lighter oils or heavy oils. For example, the mortality rates of seabirds are decreasing because of oil spills on seawaters. Not only that, but it may also have long-term impact on their diet (Schulz et al., 2017). In late November 2010, it was reported that almost 510km of shoreline was covered with crude oil spill in Louisiana (Liu et al., 2016) and this caused great damage to marine and wildlife habitats. The fishing and tourism industries were affected by the spills (Liu et al., 2016). Therefore, action needs to be taken to tackle oil pollution so that the natural environment can be preserved.

It has become priority amongst environmental scientists and technologists to solve the problem of oil spills and oil contaminated waters (Li et al., 2016, Abdullah et al., 2016). Among various adsorbents developed, activated carbon is one of the most widely used. Due to a high demand for activated carbons in other advanced applications such as catalyst supports, air filters and gas storage, it has resulted in increased costs for it in the market (Hasan et al., 2018). Adsorption process using commercial activated carbon is very effective but requires high cost (Dahri et al., 2015). So, this leads to research on how to introduce alternative activated carbon using cost-effective materials (Dahri et al., 2015). Among available methods, sorption process is commonly used to remove oil because it is cheap, simple and effective (Abdullah et al., 2021).

Previous research provided evidence that biological wastes could act as activated carbon to remove oil such as banana peel (Abdullah et al., 2016) and *parkia speciosa* pod [9]. Other than that, biological waste such as Chempedak and Durian (Artocarpus sp.) peels can also be used to eradicate Methyl Violet color (Dahri et al., 2015). This technique is expected to replace the expensive and hazardous chemical treatment to remove oil as since the 1970s, the environmental awareness and concern for oil pollution has increased (Othmah et al., 2020).

In the present study, Chempedak (Artocarpus integer) stone is used to act as an activated carbon or as the adsorbent to remove oil. Chempedak is a species of tree in the Moraceae family, and it is of the same genus as breadfruit and jackfruit (Monica et al., 2018). More than 50 % of Artocarpus fruits' skin and core are discarded as wastes when they can be an effective low-cost bio sorbent (Dahri et al., 2015). With the increase in the production of processed fruit products by the world population, abundant amounts of Artocarpus integer stones are disposed from food processing sectors or by the residences causing a serious problem of disposal. The idea of utilizing it as an adsorbent at a low-cost is the alternative way that could increase its economic value. Moreover, it can help to reduce the cost of disposal and can reduce environmental pollution (Hasan et al., 2018). As far as the knowledge of the researchers, Chempedak (Artocarpus integer) stone has never been used as an adsorbent before. This has led to discovering the potential and possibilities of using it as an adsorbent. Thus, the main aim of this paper is to study and observe the potential of Chempedak (Artocarpus integer) stones in adsorbing oil (used vegetable oil, lubricant oil and diesel oil). The experiments are conducted by varying parameters such as the adsorbent dosage (0.2g, 0.4g, 0.6g, 0.8g, 1.0g), type of water solution (tap water, the seawater and lake water) and untreated and treated adsorbents.

Methodology

Raw Material Preparation

Chempedak Stones (CS) were collected as solid waste at Pasar Peladang, Skudai, Johor. The wastes were then washed several times using tap water to remove physical and adhering impurities such as dirt and the fruit's flesh. After washing with tap water, the CS were then dried under the sunlight for 2 days. After that, the CS were oven dried at 60°C for about 4 hours to make sure they were moisture free.

Chemicals Preparation

About 1000 mL of 0.5M of sodium hydroxide (NaOH) and 0.5M of nitric acid (HNO_3) were prepared for the chemical treatment for CS.

Adsorbent Preparation

The untreated Chempedak Stones (CS) were prepared by drying the CS and then crushing them using mortar (Chaidir et al., 2015) and grounding them into fine powder using a knife mill (Model: Retsch GM200). Then, the ground powder is sieved by using a laboratory sieve (Model: Retsch) to the particle size of 63µm. On the other hand, the treated adsorbent was prepared by soaking the dried CS in a 1000mL beaker containing 0.5M NaOH for about 24 hours. After that, the soaked CS were rinsed with distilled water before they were sun dried for 3 days. The dried CS were then soaked again in a 1000ml beaker containing 0.5M HNO_3 for about 30 minutes. Next, the CS were then rinsed with distilled water and extra pure water before sun dried for 2 days and oven dried for 4 hours at 60°C to make sure that they were moisture free. Then, the dried treated CS were crushed and grounded into fine powder and sieved for particle size of 63µm.

Effect of Adsorbent Dosage

The effect of adsorbent dosage in oil removal was prepared for different concentration of CS 0.2g, 0.4g, 0.6g, 0.8g and 1.0g in powder form. After measuring the weight using analytical mass balance, the adsorbent was transferred into the small tea bags. At the same time, about 200mL of oil was prepared at 5 different beakers. Then the adsorbent in the tea bag was dipped into the oil and set aside for 30 minutes. After that, the adsorbent weight in the tea bag was measured and observed. The steps were repeated for treated CS.

Type of Water Solution

Three different types of oil were introduced in different types of water solution which are seawater, lake water and tap water to study the effectiveness of untreated and treated CS powder in oil removal. About 20 mL of oil was added in 60 mL of water solution and at the same time the adsorbent was dipped into the solution. After 30 minutes, the adsorbent tea bag was removed. The process was repeated for different adsorbent dosage (0.2g, 0.4g, 0.6g, 0.8g and 1.0g). Small tea bags were used for the entire experiment to make sure the powder stayed in place. The initial and final mass of the tea bag were measured to be used to calculate the percentage of oil removal using the equation as below (Abdullah et al., 2019):

$$\boldsymbol{Percentage\ of\ oil\ removal}\ (\boldsymbol{R}\%) = \frac{\boldsymbol{Final\ mass}\ (\boldsymbol{g}) - \boldsymbol{Initial\ mass}\ (\boldsymbol{g})}{\boldsymbol{Final\ mass}\ (\boldsymbol{g})} \times \mathbf{100}\ \% \qquad (1)$$

Fourier-Transform Infrared Spectroscopy (FTIR) Analysis

For the characterization process, the treated and untreated CS were characterized using Fourier-Transform Infrared Spectroscopy (FTIR). The peaks produced in the spectra indicates the functional group of CS (Hasan et al., 2018). The spectra were taken between 700 cm^{-1} and 4000 cm^{-1} using Bruker Vertex 70v FTIR spectrometer.

Scanning Electron Microscope (SEM) Analysis

The morphology of surface area of the CS powder was observed by using JSM-IT200 In Touch Scope™ SEM with high vacuum (2000X) magnification before and after the chemical treatment of CS (Hasan et al., 2018) and after the oil absorption.

Results and Discussion

Fourier-Transform Infrared Spectroscopy (FTIR)

The results show that there were slight differences between the wavenumber at the peak for untreated (Figure 1) and treated CS (Figure 2). The wavenumbers of untreated CS at a certain pick have a higher number compared to the treated CS powder. The wide range of wavenumber is observed at 1000 and 1300 cm^{-1}in the fingerprint region because of the difficulty in picking out the infra-red spectrum. At this region, the carbon-oxygen (C-O) single bond is predicted (Mohd Jopery et al., 2020). The O-H bond was present in the samples at 2500 – 3300 cm^{-1} which is a useful bond for adsorption depending on its environment (Mohd Jopery et al., 2020). For untreated to treated, the peak was shifted from 3277.67 cm^{-1} and to 3280.15 cm^{-1} (Chaidir et al., 2015). At between 2850 – 2950 cm^{-1} it shows sp^3 C-H bonding. From the graph, for untreated CS powder, the peak was at 2924.95 cm^{-1} and after treatment, the peak shifted to 2925.42 cm^{-1}.

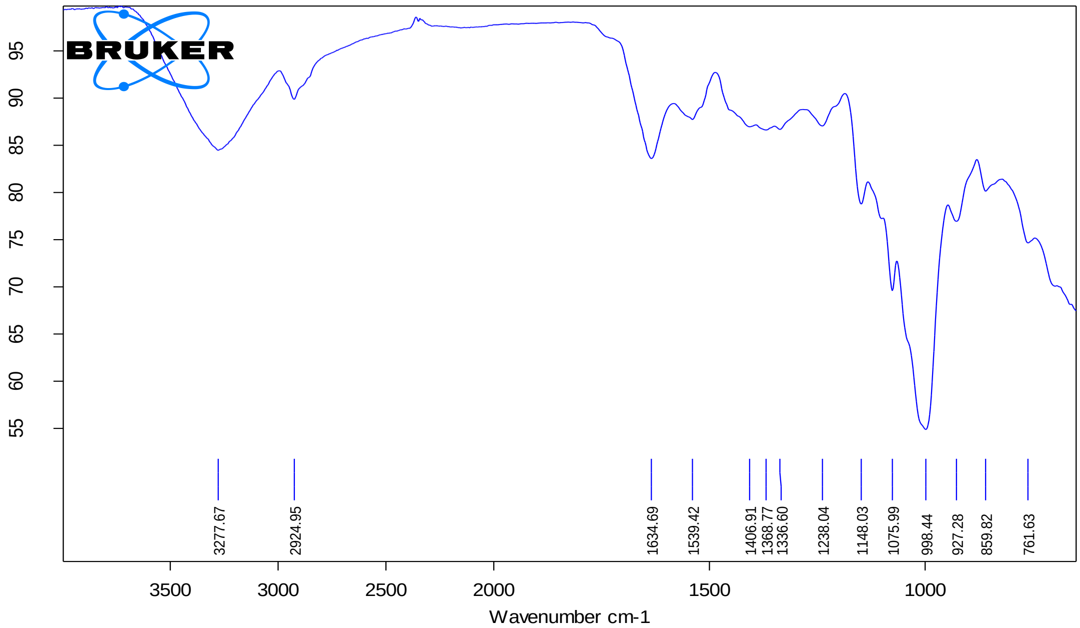

Figure 1. FTIR Analysis of untreated CS powder.

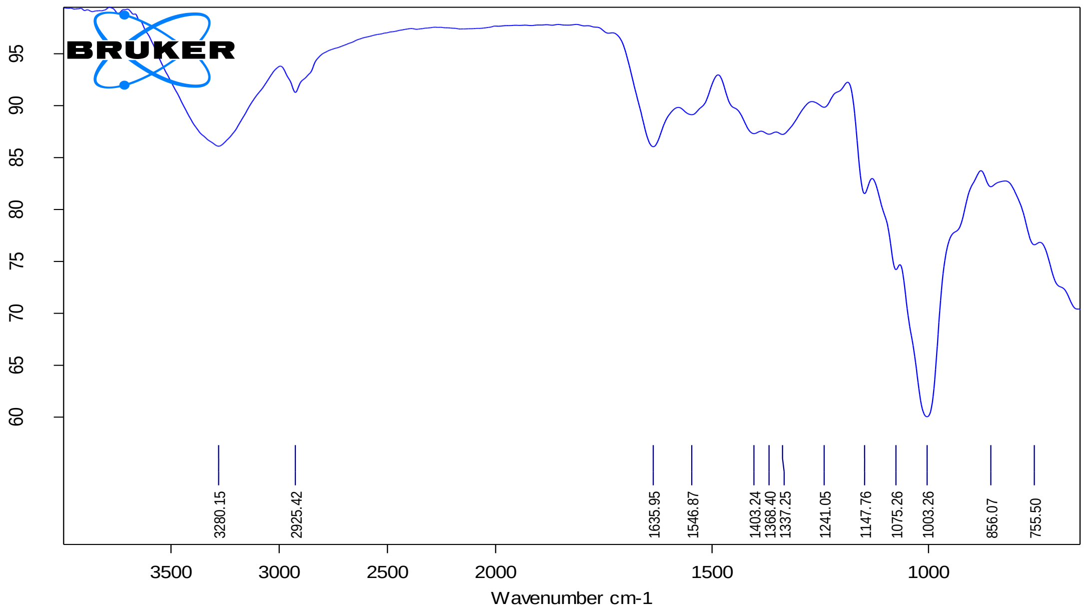

Figure 2. FTIR Analysis of treated CS powder.

Scanning Electron Microscope (SEM)

Scanning Electron Microscope (SEM) analysis was performed to analyze and observe the morphology of surface area of the CS powder before and after treatment (Chaidir et al., 2015). Figure 3 (a) and (b) above show that treated CS powder have larger porous surface area which can adsorb more oil compared to the untreated CS (Mohamad et al., 2019). The surface of the treated CS powder looks rougher with different degree of wrinkles and grooves.

This indicates that the alkali and acid used for sample treatment and pre-treatment caused serious damage to the structure of CS (Wang et al., 2012). At Figure 3 (c) and (d), the beads of the adsorbent after oil adsorption seem to have swollen. Both of untreated and treated CS powder can trap and adsorb the oil because the pores at both samples were occupied after the adsorption of oil. These show that the oil was adsorbed by the CS powder.

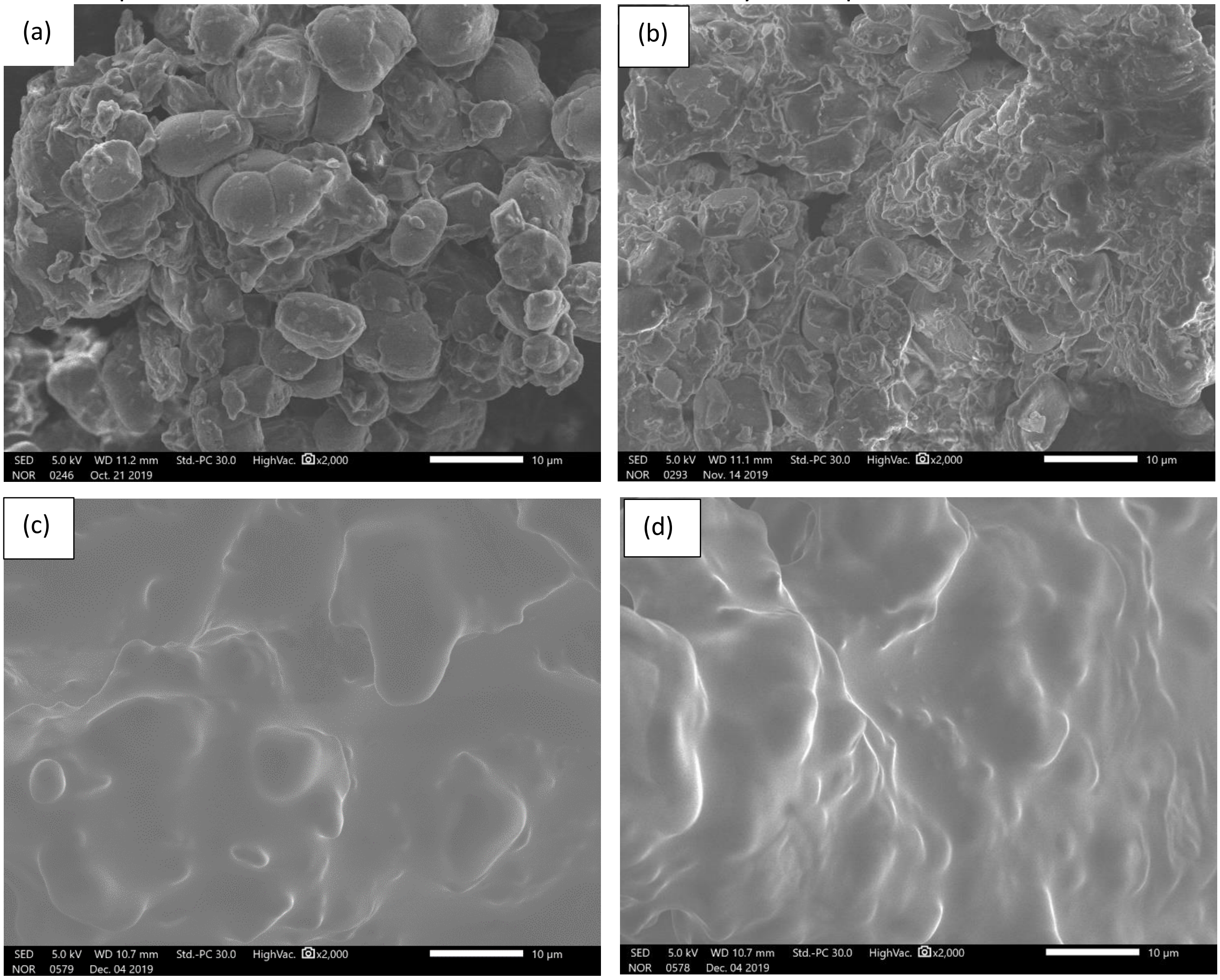

Figure *3.* SEM micrograph of the (a) untreated CS; (b) treated CS; (c) after oil adsorption of untreated CS; (d) after oil adsorption of treated CS .

Adsorbent Dosage Experiment

Figure 4 show that the percentage of used vegetables oil removal for both untreated and treated CS at different adsorbent dosages (0.2g, 0.4g, 0.6g, 0.8g and 1.0g). From the result obtained, there is an increase in percentage of removal before it decreased with an increasing adsorbent dosage which is contrary with previous study. Previous studies indicated that the adsorbent dosage is directly proportional to the oil removal efficiency in terms of percentage (Abdullah et al., 2019). This is due to the full adsorption of the adsorbent. As the adsorbent dosage increase, particles attract each other and causes coagulations. This can reduce total surface area of the adsorbent (Abdullah et al., 2016). The low removal at high adsorbent dosage can contribute to concentration of oil which is lower than the available binding sites of adsorbent. Since the adsorbent has reached its maximum limit for the oil uptake, there will be some spaces left for the available binding site in adsorbent (Wang et al., 2012).

The present study indicated that using the same adsorbent dosage provided different results of absorption. For the untreated CS, the highest percentage of oil removal is obtained at 0.2 g and 0.4 g for vegetable oil which is 89.76 per cent and 84.85 per cent respectively while the lowest oil removal at the same dosage is diesel (Figure 6) which is 81.08 per cent and 74.70 per cent respectively. These happened because of the factor of morphology structure of natural sorbent in oil adsorption. Surface roughness with suitable microstructure influence the oil sorption capacity of natural sorbent because the available structure can maximise the oil entrapment. Besides, for natural adsorbent, the surface of adsorbent contains lipid-binding surface of hydrophobic proteins subunits which have the ability to adsorb the oil (Mohamad et al., 2019).

The overall results showed that treated CS powder was different from untreated CS powder. Clearly, the highest percentage of oil removal for treated CS at 0.2g and 0.4g is obtained for lubricant oil (Figure 5) which are 89.01 per cent and 81.08 per cent respectively compared to used vegetable oil and diesel oil. In terms of removal, diesel oil has the lowest percentage removal compared to the others. Mostly, treated CS powder has lower percentage of oil removal compared to the untreated CS powder. Previous studies showed that treated adsorbent had better removal than the untreated because it had improved their pore surface area and micro porosity (Ndifor-Angwafor et al., 2017). However, the decrease in the adsorption capacity is contributed by unsuitable pH of treatment. At low pH, it can decrease the adsorption capacity due to acidic properties which deteriorated the removal efficiency (Abdullah et al., 2017).

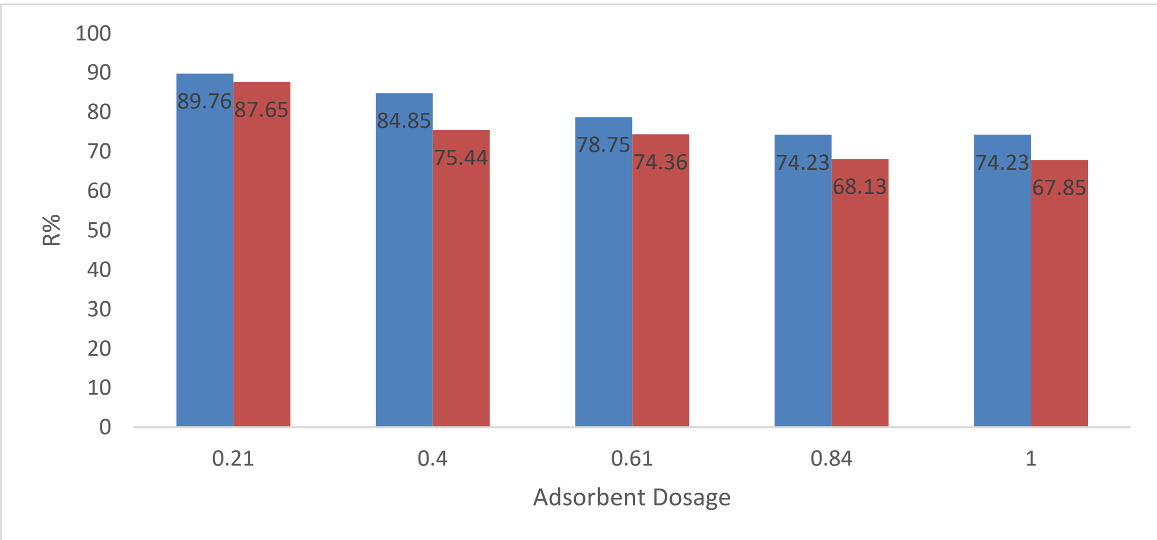

Figure 4. Effect of Adsorbent Dosage (used vegetable oil) (Note: Blue represent treated CS and red represent untreated CS).

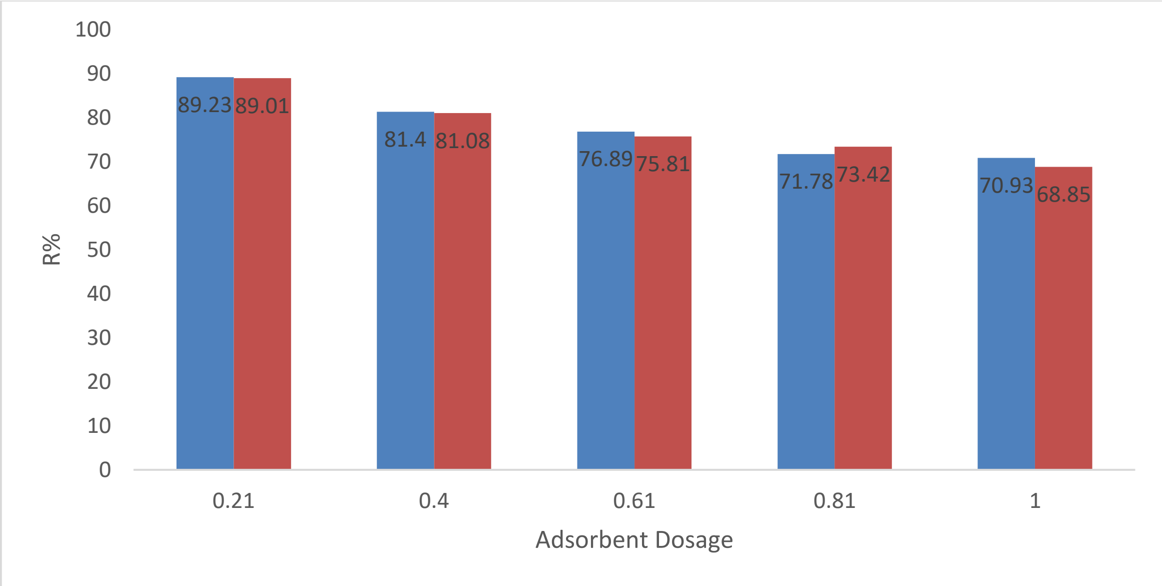

Figure 5. Effect of Adsorbent Dosage (lubricant oil) (Note: Blue represent treated CS and red represent untreated CS).

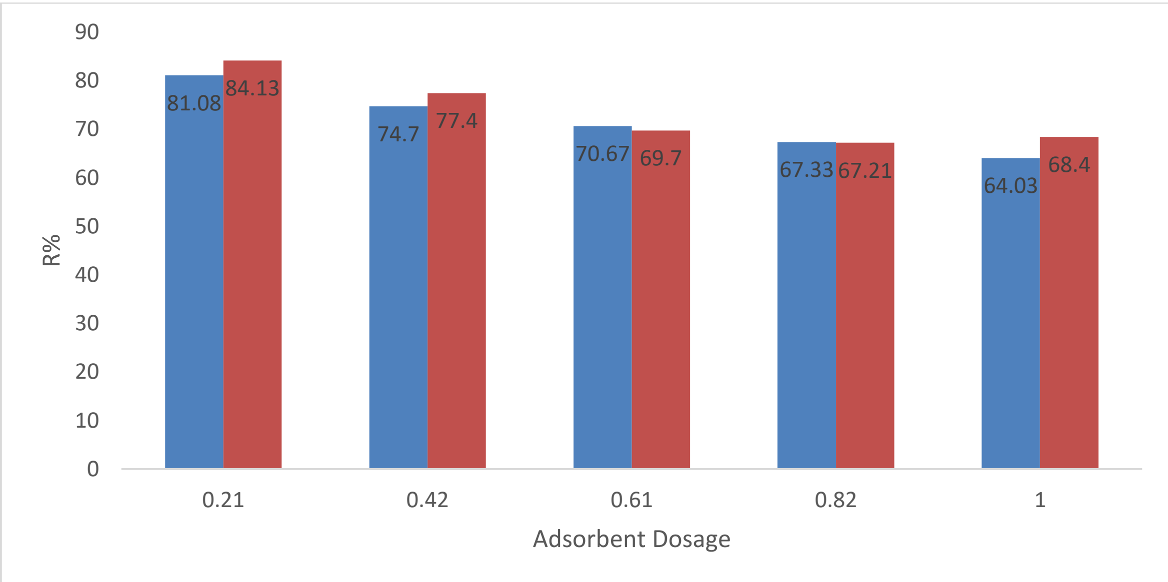

Figure 6. Effect of Adsorbent Dosage (diesel) (Note: Blue represent treated CS and red represent untreated CS).

Types of Water Experiment

Figure 7 shows that the highest percentage of oil removal was observed at 0.4g untreated CS on vegetable oil in the tap water which is 82.14 per cent. Unlike treated CS, it is more effective to adsorb used vegetable oil on sea water with 81.31 per cent of oil removal. As for lubricant oil (Figure 8), the oil removal is effective for untreated CS on tap water with 84.31 per cent as compared to sea water at the same absorbent dosage, 0.4g. Using the same lubricant oil, the removal from sea water is more effective than lake water using treated CS powder with 83.47 per cent of oil removal. Under different circumstances, for diesel oil (Figure 9), untreated CS has the more effective adsorbent than treated CS to adsorb the oil on lake water compared to tap water and sea water with 76.05 per cent of oil removal. In addition, the percentage removal for treated CS is similar for both tap water and lake water which is 74.03 per cent and ineffective to adsorb the oil from sea water. It can be concluded that the effectiveness of the untreated and treated adsorbent is based on the type of oil and water introduced.

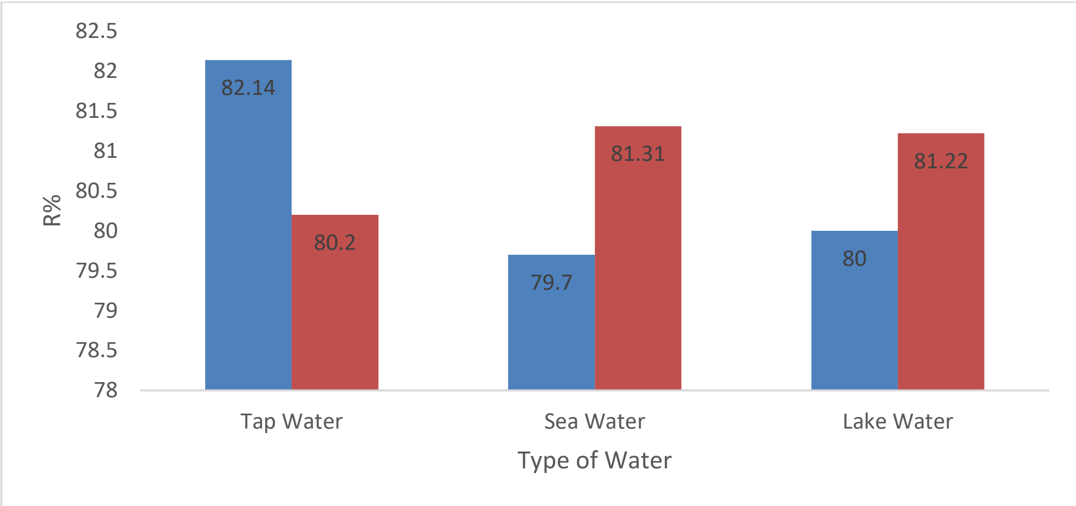

Figure *7.* Effect type of water source for used vegetable oil.

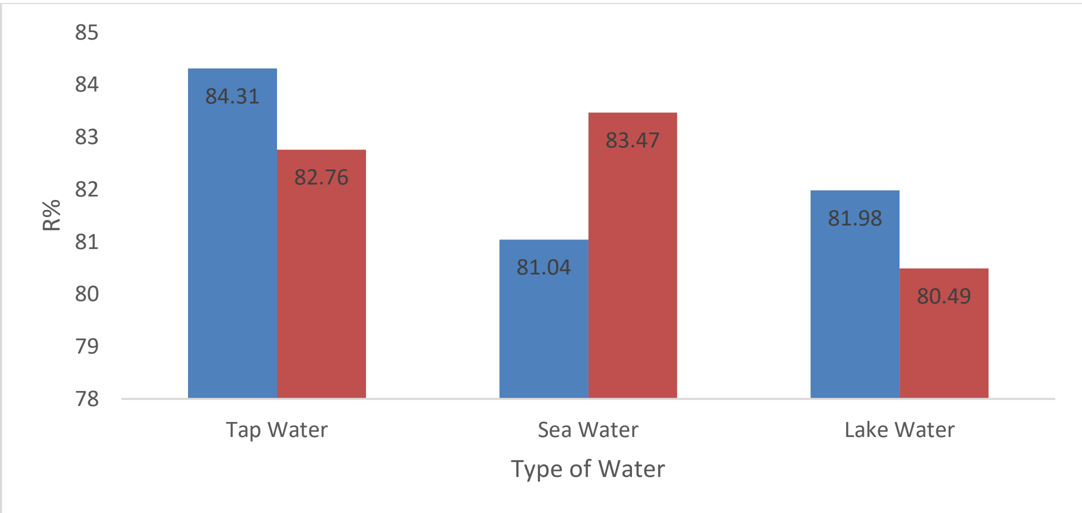

Figure *8*. Effect type of water source for lubricant oil.

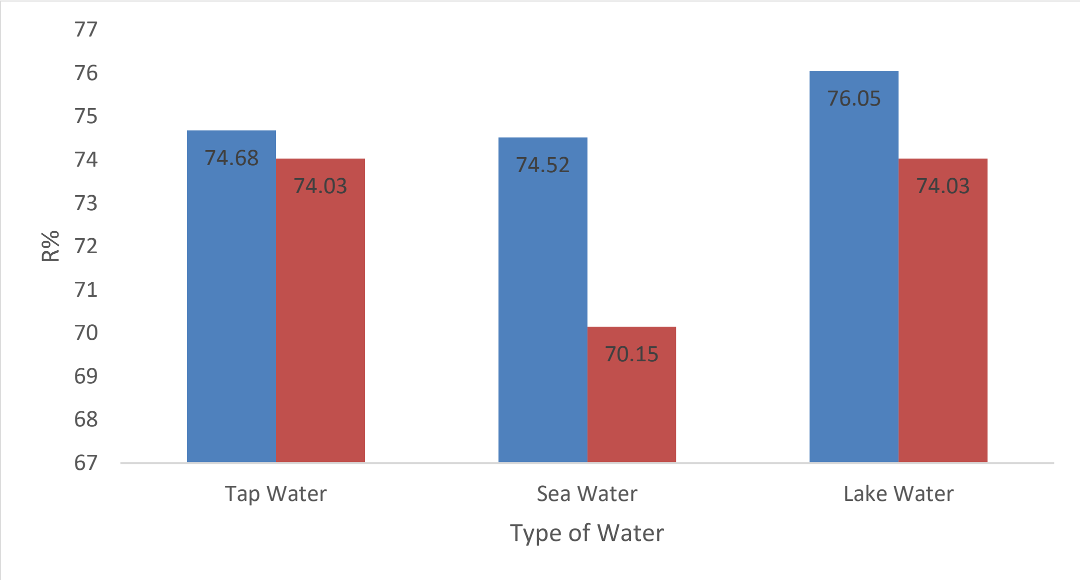

Figure *9*. Effect type of water source for diesel.

Conclusions and Recommendations

In conclusion, the objective of the present study to produce a low cost and eco-green adsorbent of Chempedak Stone (CS) in cleaning and removing oil spill is successfully achieved. This proved the capability of natural adsorbent; CS powder has a good potential as alternative economic adsorbent in clean up oil spill. Moreover, because of its widespread availability characteristics from local markets, it can confirm constant supply of adsorbent instead of being dumped as waste. In addition, it can help to reduce environmental problems such as land pollution and water pollution since CS powder is biodegradable, disposable and can be available at low cost. In fact, current conventional methods in clean up oil spill using activated carbon seem to be of higher cost and is not environmentally friendly. The present study proves that the pretreatment using NaOH and HCl improves the absorption capacity. Additionally, the types of oil and water can influence the percentage of removal and there is no absolute result to determine which condition of adsorbent is the most effective to remove oil. Other than that, the percentage of oil removal (R%) is not directly proportional to the adsorbent dosage. In effect, as the adsorbent dosage increase, percentage of oil removal (R%) decrease.

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References

Prasad, D. R. G., & Anuprakash, M. M. V. V. S. (2016) .Pollution Due to Oil Spills in Marine Environment And Control Measures. *IOSR J. Environ. Sci. Toxicol. Food Technol.*, *10*(9),01–08.

Adeyi, A. A., Jamil, S. N. A. M., Abdullah, L. C., Choong, T. S. Y., Lau, K. L. & Abdullah, M. (2019). Adsorptive removal of methylene blue from aquatic environments using thiourea-modified poly(acrylonitrile-co-acrylic acid). *Materials*, 12, 1734.

Li, J., Li, D., Yang, Y., Li, J., Zha, F., & Lei, Z. (2016). A prewetting induced underwater superoleophobic or underoil (super) hydrophobic waste potato residue-coated mesh for selective efficient oil/water separation. *Green Chem.*, *18(*2), 541–549.

Abdullah, M., Sukor, M. Z., Roslan, A., & Tumar, M. (2016). Preliminary Study of Adsorption Copper (II) Ions from Aqueous Solution Using Aromatic Green Dwarf Waste as an Biosorbent. *J. Appl. Environ. Biol. Sci*., *6*(7S):58-62.

Schulz, M., Fleet, D. M., Camphuysen, K. C. J., Schulze-Dieckhoff, M., & Laursen, K. (2017). Oil pollution and seabirds. In: Wadden Sea Quality Status Report. Eds.: Kloepper S. et al., Common Wadden Sea Secretariat, Wilhelmshaven, Germany. Last updated 01.03.2018. Downloaded DD.MM.YYYY. qsr.waddensea-worldheritage.org/reports/oil-pollution-and-seabirds.

Hasan, R., Razifuddin, A. M., Wahyuny, N., Jusoh, C., Jusoh, R., & Setiabudi H. D. (2018). Artocarpus integer peel as a highly effective low-cost adsorbent for methylene blue removal: Kinetics, isotherm, thermodynamic and pelletized studies. *Malaysian J. Fundam. Appl. Sci*., *14*(1) 25–31.

Dahri, M. K., Chieng, H. I., Lim, L. B. L., Priyantha, N., & Mei, C. C. (2015). Cempedak durian (artocarpus sp.) peel as a biosorbent for the removal of toxic methyl violet 2B from aqueous solution. *Korean Chem. Eng. Res*., *53* (5) 576–583.

Abdullah, M., Azmi, A. N. F., MuzakharAzahar, M. Z. M., Amri, M. F. N., Kadiran, K. A., Che Lat, D., Abu Kasim, H., & Khalid, A. H. (2021). Application of CasuarinaEquisetifolia needle for the removal of heavy andlight oil waste. *AIP Conference Proceedings 2339, 020156* (2021); https://doi.org/10.1063/5.0044212.

Othmah, N. Q., Abdullah, M., Muhammad, A., Ismail, A. M. M., & Mustam, M. M. (2020). Preliminary Study of Chemically Modified Parkia speciosa as an adsorbent for oil treatment. *Malaysian Journal of Industrial Technology*, *4* (1) 1-5.

Chaidir, Z., Zein, R., Sagita, D. T., & Munaf, E. (2015). Bioremoval of methyl orange dye using durian fruit (Durio zibethinus) Murr seeds as biosorbent. *J. Chem. Pharm. Res*., *7*(*1*) 589–599.

Abdullah, M., Shah, N. A. F. N., Saadun, M. A., Kadiran, K. A., Zaiton, S. N., Azman, H. A., Othman, Z. S., & Osman, M. S. (2019). Comparative Study of Acid-Treated and Alkali-Treated Carbonised Kapok–Fibres for Oil/Water Absorption System. *J. Phys. Conf. Ser*., 1349, 012104.

Wang, J., Zheng, Y., & Wang, A. (2012). Effect of kapok fiber treated with various solvents on oil absorbency. *Ind. Crops Prod., 40* (1) 178–184.

Jopery, M. N. S. A., Abdullah, M., KumYoke S., & Mustaffa, A. R. (2020) The preliminary study of oilremoval using lemon peel waste. M*alaysian Journal of Chemical Engineering and Technology (MJCET)*, *3* (1) 56-61.

Abdullah, M., Roger, S. L., & Michael, D. D. (2017). Preliminary Study of Response Surface Methodology for Optimization of Oil Removal by Adsorbent-based Pineapple Peel Waste. M*alaysian Journal of Industrial Technology*, *2*, (1) 1-5.