

Asam Gelugur-based Carbon Aerogels for Highly Recyclable Oils Adsorption

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ABSTRACT

Rapid development of the oil industries and improper oil management caused a huge amount of oil pollutant released into the water environment. Oil pollutant is toxic and may harm aquatic biodiversity. The present work highlights the facile preparation of carbon aerogels based from Asam Gelugur (CA@Ga) for oil remediation from aqueous. Analysis on the structure of CA@Ga using Fourier Transform Infrared Spectroscopy (FTIR), Energy Dispersive X-ray (EDX) and Scanning Electron Microscopy (SEM), confirmed the successful preparation of CA@Ga through hydrothermal carbonization. The main parameters affecting the adsorption of oil such as the sorption time, pH and mass of sorbent were screened and oil adsorption studies revealed that the optimum conditions were at contact time of 3 minutes, pH of solution at 7 and the adsorbent mass of 2 g with the highest adsorption capacity of 0.82 ± 0.01 g/g. CA@Ga displayed a good stability towards thermal treatment and exhibited good adsorption ability towards several types of oil. Recyclability study depicts that CA@Ga could be regenerated by simple physical treatments and retained a high sorption after 10 cycles with adsorption capacity of 0.80 ± 0.01 g/g. Therefore, the prepared CA@Ga has potential in application of oil recovery and environmental protection.

Keywords: Asam Gelugur; Carbon aerogel; Adsorption; Oil remediation

1. INTRODUCTION

Oil spillage is defined as the unintentional or purposeful discharge of large quantities of crude oil hydrocarbons into the environment. The formation of the oil slick occurs when oil disperses out and creates a thin layer floating on the water's surface. Oil slick becomes thinner when continues spreading and this is alluded as sheen. This thin layer always looks like a rainbow (Ndimele *et al.*, 2018). Crude oil exploitation, seepage of oil from the pipeline, transfer of oil into vessels or tanks, vandalization are the most argumentative aspects of oil exploration. Oil spillage in the sea will go through a series of biological, chemical, and physical processes that result in degradation of oil. Characteristics of oil, condition of environment and ambient temperature will affect the speed of oil spillage. Heavy oils deteriorate more slowly than lighter oils as they contain a higher proportion of hydrocarbons and have a higher molecular weight. The degradation process will be faster at high temperatures as compared to low temperature. Evaporation, dispersion, dissolution, and sedimentation are processes involving in the breakdown of an oil spill (Huz *et al.*, 2018).

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A recent incident that has been reported on July 2018, where six tonnes of diesel and hundreds litres of engine oil drifted from Penang seas towards Perak when a ship sank. The stench of fuel was overpowering, and it left a thin rainbow sheen. It threatened the fish and shrimp farms and the coastal environment. Another incident that has been reported on 8 March 2019 where around 20 to 40 tonnes of oil waste and toxic chemicals were disposed into Kim Kim River, Pasir Gudang. The illegal action led to a serious impact not only to humans but also to marine life. The toxic chemicals included benzene, acrolein, acrylonitrile, hydrogen chloride, toluene, methane, d-limonene, xylene, and ethylbenzene.

Oil pollution has resulted in serious consequences on the marine ecosystem (Zhu *et al.*, 2017). It affects the qualities of water for fishing, swimming, consumption, and domestic use. Humans will tend to consume seafood with bioaccumulated oil toxins and breathing oil vapor that leads to health impacts. The presence of oil on the surface of the ocean blocks adequate amounts of sunlight from penetrating the surface hence decreasing the level of dissolved oxygen which is detrimental to marine life (Doshi *et al.*, 2018). The chemical properties of the oil will be altered during the reaction between oxygen and ultraviolet (UV) radiation from the sun. Hence, denser and viscous oil slicks that consist of large hydrocarbon molecules will be formed and affecting the marine life. Fishers will also suffer from economic consequences if the fish are contaminated. Although the national legislation has been strengthened to protect the aquatic life, however, oil pollution in the world still achieves a high index due to the high demand for industry supply, development in economic and population to fulfil these requirements. Hence, it is very important to recover the ecosystem from oil spillage to minimize danger to human health, aquatic life, and natural resources.

Many conventional oil spill cleaning treatments including physical, chemical, thermal and biological have been used for the recovery of oil and organic solvents. The efficiency of these technologies, however, are restricted by high waves, currents, and adverse weather. Some of the traditional techniques performed reveals either low purification efficiency, high operating costs or leading to second contamination. Among various techniques, the adsorption process is the most effective and economical technique for oil remediation. Adsorbent facilitates the adsorption process and considered as the most promising approach due to its high uptake rate, greater potential for recyclability and large sorption capacity. Agricultural biomass waste is one of the raw materials that could be used to develop adsorbents for oil adsorption. Since biomass possesses great abundance in nature, eco-friendliness, and low cost, it is better to utilize biomass for fabrication of carbon aerogel for oil remediation instead of discarding in landfills where they will release carbon dioxide and resulted in environmental pollution. Carbon aerogel derived from biomass resulted in the wide application of wastewater treatment due to its natural porous structure, strong sorption ability, easy to obtain, large surface area and environmentally friendly (Grosjean *et al.*, 2018).

Zhu *et al.* (2017) reported the preparation of the carbon aerogel derived from waste pomelo peels (PPCA). They found that, PPCA exhibits excellent adsorption capacity ranged from 5 g/g for castor oil to 36 g/g for sunflower oil. In addition, the sorbent could be easily regenerated by simple physical treatments and kept a high sorption rate after five sorption-regeneration cycles. Other study by Wang and coworkers shows durian shell-based carbon aerogel (DSCA) exhibited good absorptive effect to organic solvents and oils studied in our work and the adsorption capacities ranged from 3 to 19 times as heavy as the mass of the materials for different organic liquids, which depended on some properties of the adsorbent and adsorbate, such as the specific surface area, pore volume and pore size of the aerogels, the density, surface tension and hydrophobicity of the liquids (Wang *et al.*, 2017). Their work proved that DSCA is a cheap available novel absorbent with great adsorption capacity, superior hydrophobicity and good recyclability, showing a promising application in the field of wastewater treatment. Furthermore, the preparation of carbon aerogels (CAs) using eggplant as raw material for oil adsorption shows that it could exhibits adsorption capacities for oils and organic solvents could reach 35–45 times its own weight. In addition, the adsorbed oil and organic solvents could be recovered by distillation, and

the regenerated carbon aerogels samples exhibited the stable performance and outstanding reusability (Yin *et al.*, 2016). Li *et al.* (2014) have prepared three-dimensional carbon aerogels via a hydrothermal and post-pyrolysis process using winter melon as the only raw materials. The winter melon carbon aerogel (WCA) shows a low density of 0.048 g/cm³, excellent hydrophobicity with a water contact angle of 135°, and selective absorption for organic solvents and oils. The absorption capacity of WCA for organic solvents and oils can be 16–50 times its own weight. These studies proved that biomass derived carbon aerogel might be an alternative and superior adsorbent for removal of organic solvents/oils in polluted waters.

Inspired by excellent adsorption of biomass derived carbon aerogel towards oil adsorption, the newly carbon aerogel based Asam Gelugur (CA@Ga) was fabricated via the simple hydrothermal carbonization for oil remediation in this research. Since the main utilization of Asam Gelugur (Ga) is as one of the ingredients for slimming product and has been rarely reported for environmental application, it is a good idea to explore this biomass as the raw material to develop an adsorbent for decontamination of oil. In addition, the selection of Asam Gelugur for fabrication of carbon aerogel, suggesting the source of preparation of this adsorbent is economical and easily available. Multiple analysis techniques, such as FTIR, SEM, and EDX were used to investigate the characteristics of carbon aerogel based Asam Gelugur. The sorption capacity of the as-prepared CA@Ga samples for the removal of common organic solvents were studied and compared. Moreover, reusability experiment was carried out by using oil as the adsorbate to assess the recycling performance of the synthesized CA@Ga.

2. MATERIALS AND METHODS

2.1 Materials

The organic solvents including *n*-hexane, toluene, benzene, pentane, heptane, hydrochloric acid, and sodium hydroxide were purchased from Sigma Aldrich company (Missouri, USA). The Asam Gelugur was purchased from Giant Superstore, Kangar, Perlis, Malaysia. The commercial oil including grease lubricating oil, engine oil, olive oil, corn oil, and palm cooking oil were bought from Giant Superstore Kangar, Perlis, Malaysia.

2.2 Instrumentations

The functional group of CA@Ga was analyzed by using Fourier Transform Infrared Spectroscopy (FTIR) (Perkin Elmer Spectrum-65, USA) in the range between 400 and 4000 cm⁻¹ with a resolution of ± 4 cm⁻¹. The elemental and morphological of CA@Ga were investigated using JEOL JSM-7600F Scanning Electron Microscopy (SEM) equipped with Energy Dispersive X-ray (EDX) Spectroscopy.

2.3 Preparation of (CA@Ga)

The preparation of CA@Ga (Figure 1) was adapted from the standard procedure of previously reported work by Zhu *et al.* (2017). Before synthesizing CA@Ga, Ga was washed with water for several times to eliminate dirt particles. First, Ga was cut into small pieces with a dimension of about 4 cm x 2.2 cm x 1 cm. Subsequently, small pieces of Ga were put into stainless-steel autoclave and undergo hydrothermal treated at 121°C for 20 minutes. Then, Ga hydrogels were taken out and washed repeatedly with hot water about 70°C for several times to remove soluble impurities. Next, the brown monoliths were dried in the oven at 70°C for 48 hours. Finally, Ga hydrogels were pyrolyzed in a furnace at 800°C for 1 hour to obtain CA@Ga.

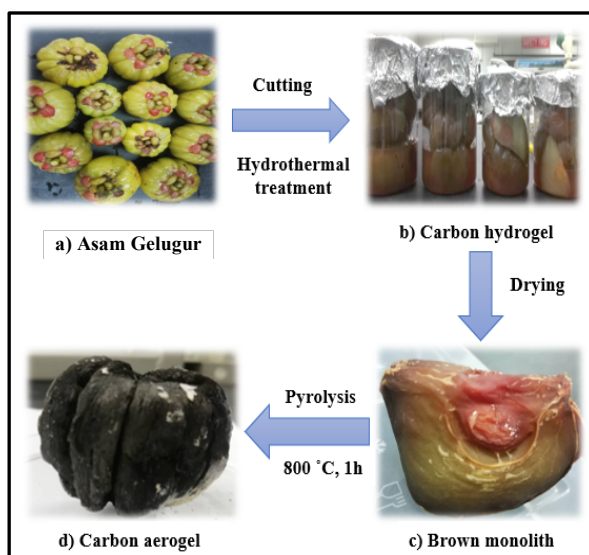


Figure 1. Schematic diagram (a-d) of preparation of CA@Ga

2.4 Analysis stability of CA@Ga

The stability of CA@Ga was examined by undergoing heat and corrosive treatments. First, 2 g of CA@Ga was kept in the oven at 70°C and 90°C for 3 days to undergo heat treatment. Another 2 g of CA@Ga was kept under ambient temperature. For corrosive medium treatment, 2 g of CA@Ga was kept under different pH of solutions with pH 2, 4, 6, 7, 8, 10, 12 and 14 for 24 hours. After that, the adsorption capacity of these treated CA@Ga was examined by conducting oil adsorption.

2.5 Oil adsorption using CA@Ga as adsorbent

The schematic diagram of oil adsorption experiment is shown in Figure 2. Briefly, 2 g of CA@Ga was placed in 20 mL of water (pH 7) at 25°C with 3 mL of lubricating oil for 3 minutes. Gravimetric analysis was used to determine the adsorption capacity, k of CA@Ga towards oil's adsorption. The weight of the adsorbent before and after oil adsorption was measured by using a weighing balance. The adsorption capacity of CA@Ga was evaluated using Equation 1 (Wang *et al.*, 2017),

$$k = \frac{(m_2 - m_1)}{m_1} \quad (1)$$

where m_1 = weight of the adsorbent before oil adsorption; m_2 = weight of the adsorbent after oil adsorption; $m_2 - m_1$ = the weight of adsorbed oil. After oil adsorption, the used CA@Ga was rinsed with *n*-hexane to remove adsorbed oil and dried in the heating chamber oven at 90°C for 24 hours. The regenerated CA@Ga adsorbent was reused for oil adsorption from water surface up to 10 times. The parameter tests for lubricating oil removal were included contact time, mass of adsorbent and pH of adsorbent. All the experiments were repeated three times.

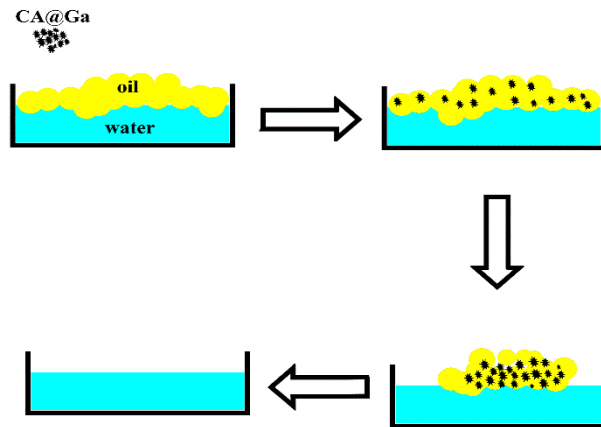


Figure 2. Schematic diagram of oil adsorption experiment

3. RESULTS AND DISCUSSION

3.1 Characterization of adsorbents

3.1.1 Vibrational Analysis

The functional groups of the adsorbent were characterized using FTIR. The spectra of Asam Gelugur, carbon hydrogel and carbon aerogels based Asam Gelugur (CA@Ga) are shown in Figure 3 (A-C). Spectrum in Figure 3 (A) shows the presence of broad and intense band at 3376.6 cm^{-1} resulting from hydroxyl groups on the Asam Gelugur (Jing *et al.*, 2019). The bands at 1645 cm^{-1} , 1106.5 cm^{-1} , and 1062.5 cm^{-1} corresponded to C=O stretching vibration (Wang *et al.*, 2017). These results depict that raw Asam Gelugur rich in hydrophilic functional groups likes hydroxyl and carbonyls. Spectrum (Figure 3 (B)) shows that the carbon hydrogel shows certain functional groups at peaks of 2922.3 cm^{-1} for C-H stretching, 1631.1 cm^{-1} (C=C stretching vibration), 1747.9 cm^{-1} , 1210.7 cm^{-1} , 1164 cm^{-1} and 1060.6 cm^{-1} (stretching of C-O bonds), 786.98 cm^{-1} and 620.16 cm^{-1} (C-H bending) (Long *et al.*, 2021).

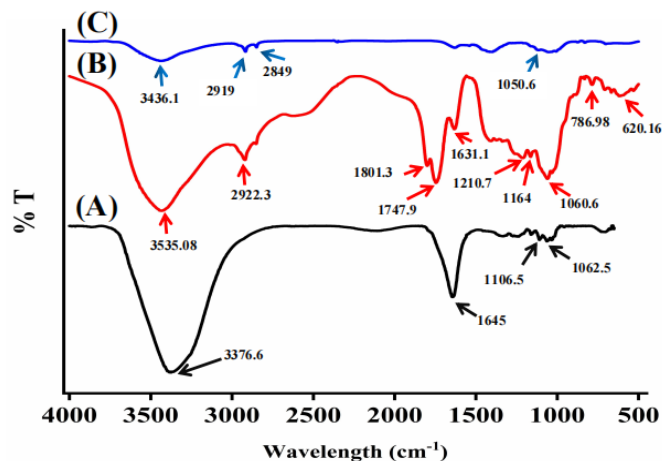


Figure 3. FTIR spectrum of (A) Asam Gelugur (B) Carbon Hydrogel (C) Carbon Aerogel from Asam Gelugur (CA@Ga)

According to research by Wang and co-researchers, the intensity of adsorption peaks around 3435.08 cm^{-1} (stretching of hydroxyl group) was weaker in carbon hydrogel showing that the hydrophilic functional groups were decomposed in hydrothermal treatment (Wang *et al.*, 2017). The characteristic bands corresponded to stretching vibrations of hydroxyl groups (3436.1 cm^{-1})

and carbonyl groups (1050.6 cm^{-1}) in the spectrum of CA@Ga (Figure 3 C) were decreased and the former adsorption band at 1747.9 cm^{-1} due to carbonyl groups stretching disappeared in the spectrum CA@Ga because these functional groups were further decomposed during the pyrolysis process (Zakaria *et al.*, 2021). This phenomenon enhanced the hydrophobicity of CA@Ga which might be favourable for oil adsorption.

3.1.2 SEM-EDX analysis

The morphological surface of the CA@Ga at different magnifications was analyzed using SEM analysis and the result is shown in Figure 4 (A-B). From these images, it can be seen that CA@Ga has interconnected porous microstructure after pyrolysis at 800°C which a large amount of irregular flaky skeleton as circled in Figure 4(A). The similar result was obtained from published work by Wang *et al.*, (2017).

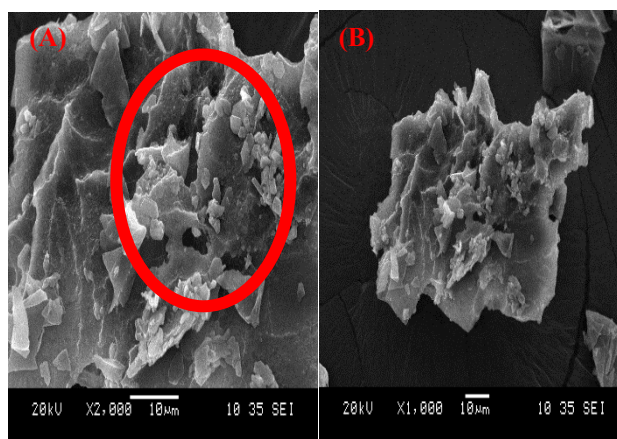


Figure 4. SEM images of CA@Ga at different magnifications of (A) x2000 (B) x1000

EDX analysis was employed to confirm the presence of elements of interests in CA@Ga and result shows that the high content of carbon in CA@Ga, proved that the surface of CA@Ga has been successfully pyrolyzed (Figure 5). This finding suggests that the surface of CA@Ga is hydrophobic and could be a potential as oil adsorbing agent.

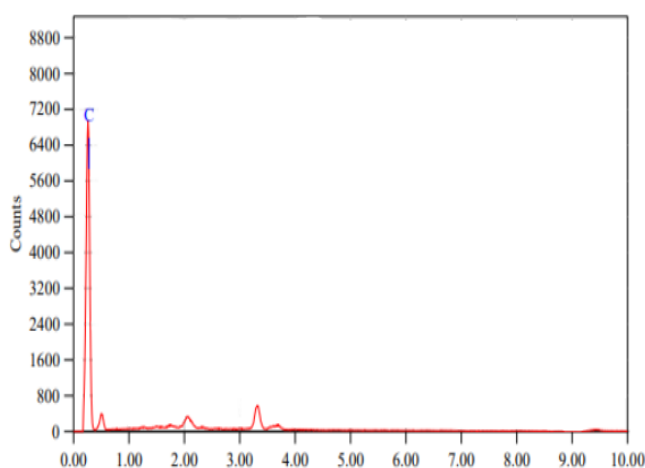


Figure 5. EDX spectrum of CA@Ga

3.1.3 Thermal stability

Thermal stability is the significant property for a hydrophobic material. It is defined as the ability of the hydrophobic material to resist heat energy by maintaining the chemical structure of its hydrophobicity. As seen in the result, the adsorption capacity of CA@Ga remained stable after undergoing heat treatment at 70°C and 90°C for 72 hours and comparable with adsorption capacity Ca@Ga without any heat treatment (ambient temperature) (Figure 6). This phenomenon can be explained by the fact that the surface of the CA@Ga was resistant towards heat in which it retained its hydrophobic properties even though kept under a heated environment (Wang *et al.*, 2015). This finding is in agreement with study by Mohamed *et al.*, (2015). Hence, it was concluded that CA@Ga exhibits high thermal stability.

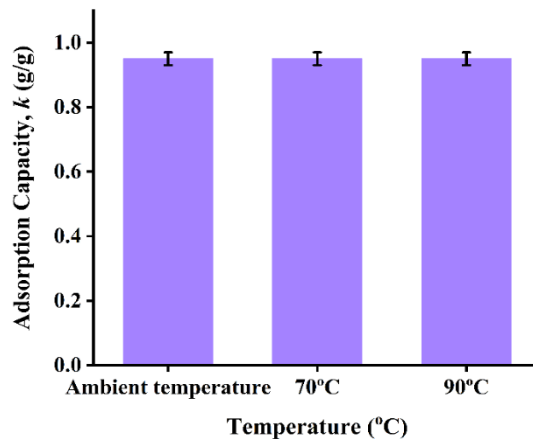


Figure 6. Effect of thermal stability against adsorption capacity of (CA@Ga)

3.1.4 Corrosive medium stability

The stability of the adsorbent in corrosive media is crucial for real water application. Thus, the stability of CA@Ga was evaluated in acidic and basic media. Experimental result shows that CA@Ga has the highest adsorption capacity in the neutral medium as compared to acidic and basic media (Figure 7). The result reveals that the surface of CA@Ga was affected by immersed in acidic and basic solution for 24 hours. At acidic conditions, a huge amount of proton was available and caused the protonation of the functional group that leads the surface of CA@Ga to become more cationic, thus the structure of adsorbent turn hydrophilic (Rozi *et al.*, 2016). At basic conditions, the decrease in adsorption capacity was directly related to the repulsion between negative charge of CA@Ga surface (Bernal *et al.*, 2017). The effect of deprotonation making the adsorbent became anionic and enhanced hydrophilic properties of CA@Ga. However, in this study, the application of CA@Ga for the real-life environment is still acceptable as literature research from Kulthanan *et al.* (2013) stated that the pH of water at normal environment between 6.9 and 8.

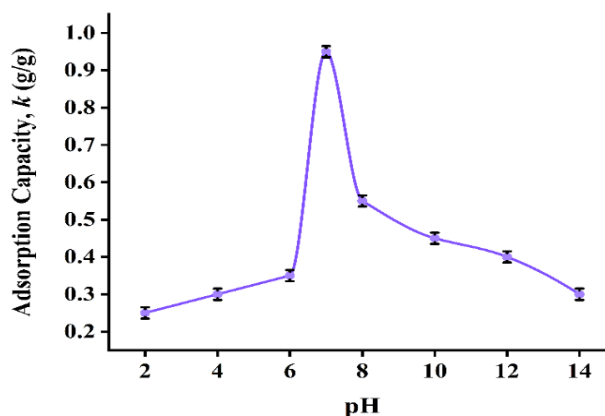


Figure 7. The effect of corrosive media on the adsorption capacity of CA@Ga

3.2 Optimization of oil adsorption using CA@Ga

3.2.1 Effect of Contact Time

The effect of contact time was studied in the range of 1 to 5 minutes towards oil adsorption. The adsorption capacity lower at the initial stage indicates that contact time provided for oil adsorption was not adequate. It has been observed that amount of oil adsorption increased with increasing contact time due to a longer time allocated (Anisuzzaman *et al.*, 2019) for lubricating oil to be adsorbed by CA@Ga through hydrophobic interaction (Figure 8). The oil adsorption capacity increased rapidly as the adsorption sites still available. The adsorption phase achieved equilibrium within 3 minutes, owing to the unavailability of a reaction site for oil adsorption. The oil adsorption capacity decreases with the increase of contact time due to the adsorption site was fully occupied (Rozi *et al.*, 2017) (Figure 9 (A)). Aggregation of oil restricts the adsorption that contribute to decreasing in adsorption capacity. Hence, oil could not be further removed by the adsorbent. Therefore, 3 minutes with an adsorption capacity of 0.89 ± 0.01 g/g was considered as the best contact time for further studies.

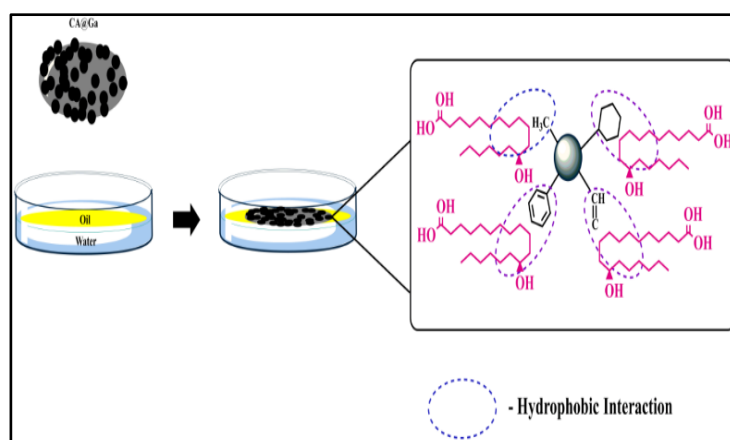


Figure 8. Schematic mechanism adsorption of CA@Ga towards lubricating oil (assume that the main composition in lubricating oil is ricinoleic acid ($C_{19}H_{34}O_3$) (Fundamentals, 2013).

3.2.2 Effect of pH

Figure 9 (B) illustrates the effect of pH on the oil adsorption. The results indicated that the lower adsorption capacity was obtained in slightly acidic and alkaline medium. In acidic medium, there were large amounts of protons available at acidic medium which saturated the adsorbent sites and increase the cationic properties of the surface of CA@Ga, making the adsorbent less hydrophobic. Furthermore, oil coalescence might happen thus increase the size of oil droplets subsequently under a strong acidity environment (Cai *et al.*, 2019). Under an alkaline condition, the deprotonation of an excess hydroxyl group on the surface CA@Ga occurs, therefore increase the anionic properties of the adsorbent surface which enhanced hydrophilicity properties of CA@Ga (Rozi *et al.*, 2016, Rozi *et al.*, 2017). Thus, the adsorption capacity decreased under unfavourable pH conditions mainly acidic and basic medium due to electrostatic force between the oppositely charged of adsorbate and adsorbent. Hence, it can be seen that pH 7 was more favourable with a maximum adsorption capacity of 0.89 ± 0.01 g/g which was chosen for the rest of the experiment.

3.2.3 Effect of mass of adsorbent

The effect of the mass of adsorbent, CA@Ga ranging from 1 to 14 g in oil adsorption (Figure 9 (C)) towards adsorption of oil was examined. It was revealed that oil adsorption capacity increased with an increase in the mass of CA@Ga. This result is due to adsorption sites of CA@Ga increased as the adsorbent dosage increase, thus making the penetration of lubricating oil molecules into adsorption sites is easier (Panda *et al.*, 2017). An equilibrium was achieved with further increase the mass of

CA@Ga revealing that the adsorption site was fully filled with the lubricating oil. Beyond the 2 g of CA@Ga, the adsorption capacity was slightly decreased due to the aggregation of adsorbent, thus the number of accessible sites decreases as less surface area is exposed (Shakoor and Nasar, 2016). Similar trend was found by work from Lazim *et al.* (2015). As a result, 2 g of CA@Ga was selected as the optimized mass that exhibited the highest adsorption capacity of 0.82 ± 0.01 g/g.

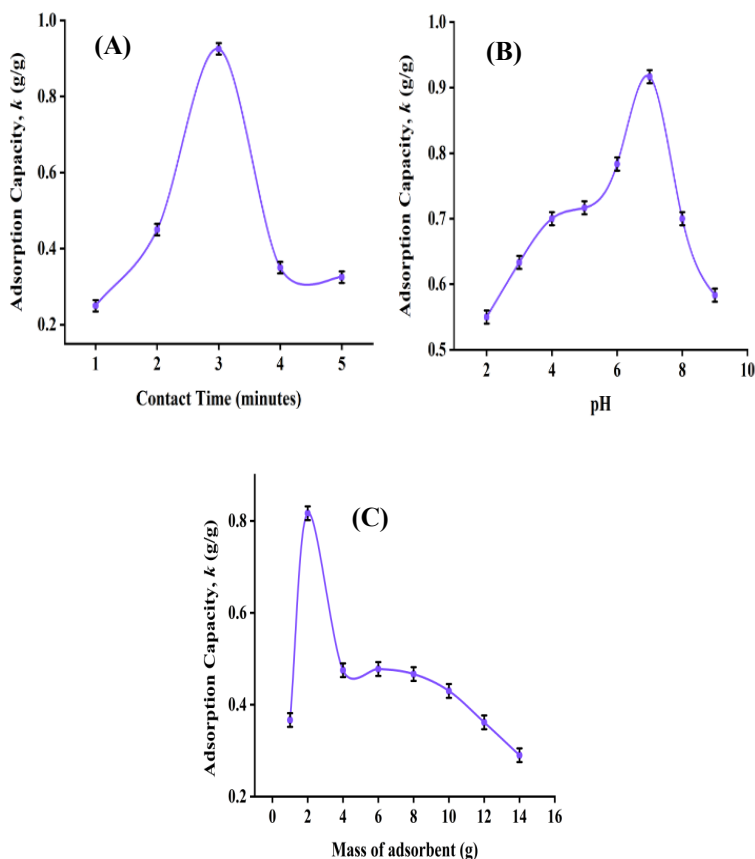


Figure 9. The effect of (A) contact time, (B) pH and (C) adsorbent dosage towards oil adsorption using CA@Ga

3.3 Adsorption capacity of CA@Ga towards several types of commercial oils and organic solvents

The adsorption capacity of an adsorbent towards oil and organic solvents depends on the surface tension, density, viscosity, molecular dimension and hydrophobicity of oils and organic solvents (Shi *et al.*, 2019). By adopting optimum conditions such as 3 minutes of contact time, pH of 7 and mass of adsorbent of 2 g, the adsorption capacities of CA@Ga toward series of commercial oils (grease lubricating oil, engine oil (SAE 30), olive oil, palm. cooking oil and corn oil) and water-immiscible solvents (benzene, toluene, heptane, *n*-hexane and pentane) were determined. The adsorption capacities of CA@Ga for the remediation of several types of oil and organic solvent is illustrated in Figure 10. Grease lubricating oil (1.75 ± 0.01 g/g) and benzene (0.90 ± 0.01 g/g) depict highest adsorption capacity against a series of commercial oil and organic solvents, respectively. The obtained results revealed that pollutants with high viscosity displayed high sorption capacity. The result obtained in this work is similar to the results reported in other published work by Rozi *et al.* (2018). The viscosities of different types of oil and organic solvent used in this study were summarized in Table 1. The occurrence of oil and organic solvent adherence to the surface of CA@Ga is better for more viscous oils and organic solvents.

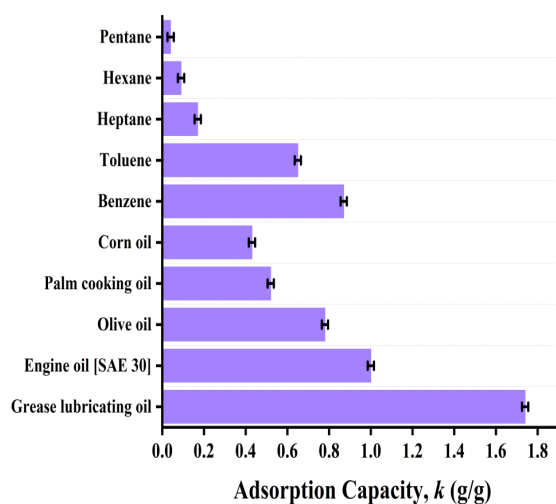


Figure 10. Adsorption capacity for different types of oil and organic solvents

Table 1. Viscosity of oils and organic solvents

Oils/ Organic Solvents	Viscosity (cP)
Grease lubricating oil	580.00
Engine Oil [SAE 30]	420.00
Olive Oil	85.00
Palm Cooking Oil	77.19
Corn Oil	52.30
Benzene	0.603
Toluene	0.560
Heptane	0.408
<i>n</i> -hexane	0.280
Pentane	0.214

3.4 Recyclability of CA@Ga

Recyclability of CA@Ga is important in clean-up applications due to environmental protection and pollution control. The saturated CA@Ga after oil adsorption was rinsed with *n*-hexane to release the adsorbed oil and then the adsorbent was dried in a heating chamber oven at 90°C for 24 hours. The regenerated CA@Ga was collected for recycling. The structure of CA@Ga was retained the same after regeneration. As shown in Figure 11, CA@Ga has possessed a high adsorption capacity of 0.80 ± 0.01 g/g and retained identical after 10 cycles of oil adsorption. There was no significant decrease in adsorption capacity, suggesting that CA@Ga exhibited a highly stable recycling performance. This finding is in agreement with study by Cai *et al.* (2019).

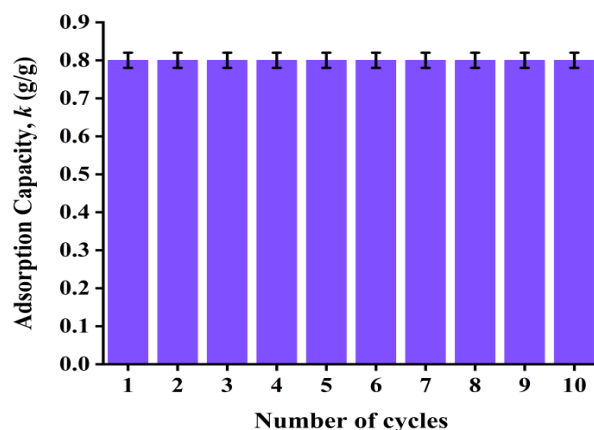


Figure 11. Recyclability of CA@Ga for oil adsorption

3.5 Oil loaded CA@Ga analysis

3.5.1 FTIR analysis

FTIR spectrum of CA@Ga before and after oil adsorption is shown in Figure 12 (A-B). It was found that the bands at 2925 cm^{-1} and 2855 cm^{-1} are more intense for the oil-loaded CA@Ga, proved that there is adsorption of oil onto the hydrophobic alkyl chain layer of CA@Ga. This result is similar with the work published by Rozi *et al.* (2018).

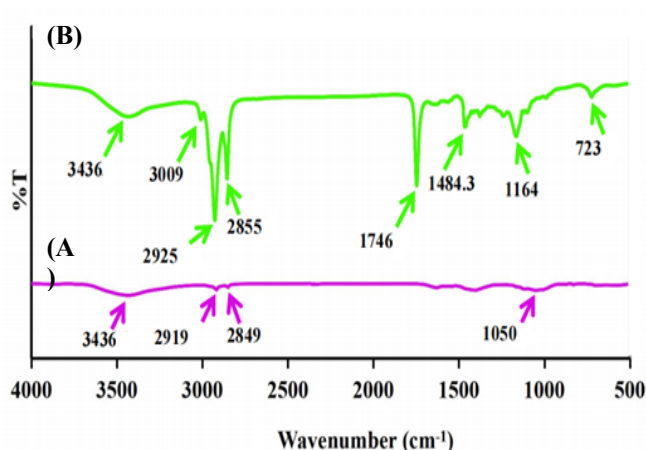


Figure 12. FTIR spectra of (A) CA@Ga, and (B) CA@Ga after oil adsorption

3.5.2 SEM analysis

To further confirm the oil adsorption by the prepared CA@Ga, SEM images were taken before and after adsorption as shown in Figure 13 (A-D). The SEM images of oil loaded CA@Ga depict the significant changes as compare to original CA@Ga as the surface of the prepared CA@Ga was spread and covered with a muddy-line (as circled in Fig. 13 (D)) due to adsorbed oil molecules. These results proved that oil was adsorbed by CA@Ga into its pores and developed a layer of oily substance on the surface (Sokker *et al.*, 2011) which is in agreement with FTIR result for oil loaded CA@Ga.

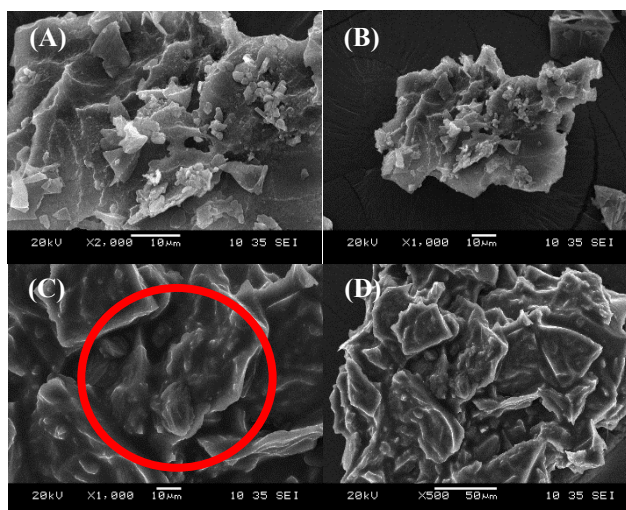


Figure 13. SEM images of CA@Ga at different magnifications of (A) x2000 (B) x1000 and oil loaded CA@Ga at different magnifications of (C) x1000 (D) x500

4. CONCLUSION

CA@Ga was successfully fabricated through the hydrothermal carbonization process. The structure of CA@Ga was characterized by FTIR, EDX and SEM analyses and showed successful preparation of CA@Ga. CA@Ga possessed some unique features such as hydrophobic and high thermal stability which showed potentials as adsorbent in the remediation of oils and organic solvents. Several parameters were applied during oil adsorption such as contact time, pH and mass of adsorbent. The highest adsorption capacity of 0.82 ± 0.01 g/g was obtained at optimum conditions of 2 g of adsorbent, solution at pH 7 and contact time of 3 minutes. CA@Ga expressed excellent recyclability through simple physical treatment due to it maintained effective adsorption capacities for oils after ten adsorption-desorption cycles. It could be an alternative oil adsorbent since the source of its preparation is available, cheap, and environmentally friendly, offering easy removal and operation.

ACKNOWLEDGMENT

We gratefully acknowledge the financial support for this project by FRGS RACER/1/2019/STG01/UNIMAP//1 from the Ministry of Higher Education Malaysia.

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