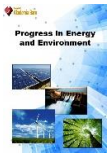
Progress in Energy and Environment

Journal homepage: https://www.akademiabaru.com/submit/index.php/progee/
Link to this article: https://doi.org/10.37934/progee.17.1.3241



Volume 17 (2021) 32-41

Original Article

Potential of fatty acid-modified spent tea leaves as adsorbent for oil adsorption

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Abstract

Treatment of oil pollution remains a challenge due to the growing urbanisation. Thus, there is an increasing number of global studies on exploiting simple and effective methods to remove oil from water. In the present work, spent tea leaves (STL) have been modified using oleic acid (OA) and free fatty acids from waste cooking oil (FFA-WCO). The aim was to enhance the hydrophobicity of the STL so that they can act as an oil adsorbent. The functional groups of the fatty acids within the modified STL were identified using the Fourier Transform Infrared (FTIR) Spectroscopy analysis, while the surface morphology of STL was characterised using a Scanning Electron Microscope (SEM). The performance of the synthesised adsorbents for oil adsorption was tested in batch adsorption experiments. The FTIR results revealed that free fatty acids have been successfully impregnated onto the surface of STL. SEM analyses showed that the surface of the fatty acid-modified STL has smoother surfaces compared to the rougher surface of unmodified STL. From the batch adsorption test, the highest adsorption capacity was observed using 1:10 ratio of STL to WCO, with 120 min of contact time, 1 g of adsorbent dosage, and under the temperature of 45 °C. The adsorption capacity of STL@FFA-WCO at the optimum condition was 1.800 ± 0.15 g/g. For the effect of modification agents, STL that were modified using oleic acid (STL@OA) showed greater adsorption capacity of 2.267 ± 0.21 g/g. These findings proved that the fatty acid-modified STL have the potential of becoming green adsorbents for oil removal.

Article Info

Received 5 June 2021
Received in revised form 30 November 2021
Accepted 15 December 2021
Available online 3 January 2022

Keywords

Oil removal
Biomass adsorbent
Adsorption capacity

1 Introduction

Oil is one of the major pollutants that has led to significant environmental problems. Recent industrial developments have caused an increase in the amount of oil utilised worldwide, especially by food industries. As a food product, oils are widely used in various forms, such as butter, margarine, shortening, and cooking oil [1]. However, the accumulation of waste created from cooking oil is increasing because frying has become the more popular cooking method. The direct discharge of oily

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wastewater into the sea will then create various issues. For example, the disintegration of oxygen in water will be inhibited by the oily layer that lay over the water surface. Consequently, it will devastate the aquatic ecological constellation, and ruin the habitat and breeding areas of wildlife [2]. Generally, oily wastewater from households and industrial premises are treated using wastewater treatment before it is discharged into a water body. Even so, it is reported that 44% of global household wastewater is not safely treated [3]. Therefore, an alternative method is necessary for the elimination of oil from water.

Several treatment methods for oil removal have been implemented, including adsorption, froth flotation, membrane filtration, biological treatment, coagulation, and flocculation. Nevertheless, except for adsorption, all of these treatments have drawbacks, such as high operating costs and less ecologically friendly [4]. Biological treatment is indeed environmentally friendly, but biodegradation methods are not always simple because biological processes depend on suitable environmental growth and appropriate levels of nutrients for microorganisms to grow [5]. Amongst these treatments, adsorption is the most efficient method for eliminating oil due to its advantageous traits, such as non-destructive operation, easy to handle, and sustainable [6,7]. The adsorption method was also reported to have a high oil elimination efficiency, yet low in operation and treatment costs [8].

The adsorbent is the essential element for the adsorption of oil. Various studies have evaluated the application of STL as adsorbents for eliminating effluents, such as azo dyes [9], copper ions [10], chromium ions [11], and endocrine [12]. Tea is a very popular beverage, as it is widely consumed around the world. Tea leaves will become solid waste once they are brewed. Hence, the utilisation of this abundantly available solid waste is the best way to deal with oil waste [13,14].

STL are hydrophilic-based material. The hydrophilicity of the STL is due to the higher content of polyphenols in their chemical structure. The hydrophilic functional group, hydroxyl (-OH) belong to the polyphenol compounds [15]. Since oil is a hydrophobic-based material, the hydrophilic properties of STL must be modified to hydrophobicity to enable them to act as an adsorbent for oil removal.

Based on previous research, fatty acids are frequently used as modification agents for biomass adsorbents, for example, sawdust [16], coconut coir [17], and banana trunk fibre [18]. Fatty acids consist of long hydrocarbon chains connected with a carboxyl group (COOH) [19]. This carboxyl group will react with the hydroxyl group in STL to form long hydrocarbon chain capped with carbonyl group (C=O). The substitution of an alkyl group for a hydroxyl group reduces the density of hydrogen bonding because the long hydrocarbon chain of an alkyl group offers a bulkier branch. Subsequently, the ability of hydrogen bonds to form will be decreased [16,17]. Hence, the fatty acid-modified STL will be more hydrophobic; suitable for oil adsorption.

Waste cooking oil is an abundant resource, which has a higher free fatty acids content because of the repeated heating process [20]. Free fatty acids consist of high oleic acid content. In this study, the STL were modified using waste cooking oil (WCO) and oleic acid (OA). This study aimed to determine the potential of using fatty acid-modified spent tea leaves as an adsorbent for oil adsorption.

2 Materials and Chemicals

2.1 Materials

Spent tea leaves (STL) and waste cooking oil (WCO) were collected from a restaurant in Kampung Paya, Butterworth, Penang, Malaysia.

2.2 Chemicals

Concentrated sulphuric acid (98%, H_2SO_4), n-hexane (95%), and oleic acid (97%) were purchased from Fisher Scientific, Malaysia.

3 Procedures

3.1 Raw material collection and preparation

STL collected from the restaurant were repeatedly washed using boiled water until the filtered water was clear. Then, the washed STL were dried in the oven at 60 °C for 48 h. Finally, the dried STL were sieved to obtain particles with sizes ranging from 0.50–1.00 mm [21].

3.2 STL Modification

The pre-treated STL were modified using waste cooking oil (WCO) and oleic acid (OA) as modification agents. In this study, STL were modified by adding 1 g of STL to 10 g of WCO and 10 g of OA, with 2 mL of concentrated H_2SO_4 as the catalyst [18].

These mixtures (WCO + STL and OA + STL) were continuously stirred in a solution of n-hexane for 30 min at 60 °C. Different ratios (1:10, 1:20, and 1:30) of STL and modification agents were procured after the modified STL have been washed with n-hexane to remove any excess residue. The modified STL were then dried in the oven at 70 °C for 36 h [18].

3.3 Characterisation of STL

The unmodified and modified STL were characterised using the Fourier Transform Infrared (FTIR) Spectroscopy (Perkin Elmer Spectrum-65, USA) and the Scanning Electron Microscope (SEM) (JOEL, JSM-7600F, USA). The functional groups of raw STL, STL@FFA-WCO, and STL@OA were analysed using FTIR in the range of 650 and 4000 cm^{-1}. The surface morphology of adsorbent samples was analysed using SEM at 500× magnification.

3.4 Batch adsorption test

Adsorption tests were performed by varying the parameters that affect the adsorption performance. The process parameters were studied based on the effects of modification on STL, namely, the different ratios of STL to modification agent, contact time, adsorbent dosage, temperature, and modification agents. Before running the tests, the required amounts of adsorbents were sealed in teabags with 4.5 × 3.5 cm dimension. The mass of the adsorbents before and after undergoing the adsorption test was measured and recorded for data analysis. The test was conducted in triplicate under the same conditions and the mean values were calculated for data analysis.

3.4.1 Effect of modification on STL

First, 1 g of unmodified STL and 1 g of modified STL (STL@FFA-WCO), at a 1:10 ratio to modification agent, were sealed in teabags. These teabags were put into beakers, which contained 20 g of palm cooking oil. This test was conducted at 25 °C and the samples were left in the oil for 90 min without agitation.

3.4.2 Effect of ratio of STL to modification agent

In this test, 1 g of STL@FFA-WCO was sealed in teabags at different ratios of 1:10, 1:20, and 1:30 to modification agent. These teabags were put into beakers, which contained 20 g of palm cooking oil. This test was conducted at 25 °C and the samples were left immersed in oil for 90 min without agitation.

3.4.3 Effect of contact time

In this test, 1 g of STL@FFA-WCO at a 1:10 ratio was sealed in a teabag. The teabag was put into a beaker containing 20 g of palm cooking oil. This test was conducted at 25 °C, and the sample was then left for 30 min without agitation. This test was repeated for different contact times (60, 90, 120, 150, 180, 210, and 240 min).

3.4.4 Effect of adsorbent dosage

The adsorption studies were conducted at 25 °C using different amounts of STL@FFA-WCO (0.5, 1.0, 1.5, 2.0, and 2.5 g). The required amounts of STL@FFA-WCO were sealed in teabags and then, each teabag was put into a beaker with 20 g of palm cooking oil for 120 min without agitation [11].

3.4.5 Effect of temperature

In this test, 1 g of STL@FFA-WCO was sealed in a teabag and then, put into a beaker with 20 g of palm cooking oil. This test was conducted at different temperatures of 25, 35, 45, 55, and 65 °C by placing the beakers in a water bath. These samples were left in the beakers for 120 min without agitation [22].

3.4.6 Effect of modification agents

In this test, 1 g of STL@FFA-WCO and 1 g of STL@OA were sealed in teabags and then, put into beakers with 20 g of palm cooking oil. This test was conducted at 45 °C by placing the beakers in a water bath. These samples were left in the beakers for 120 min without agitation.

3.5 Data Analysis

3.5.1 Adsorption capacity (g/g)

Adsorption capacity is the amount of adsorbate taken up by the adsorbent per unit mass of the adsorbent. The adsorption capacity, q, was determined using Eq. (3.1) [23]:

$$q = \frac{M_F(g) - M_I(g)}{M_I(g)} \tag{3.1}$$

Where M_I is the mass of adsorbent before adsorption; and M_F is the mass of adsorbent after adsorption.

4 Results and Discussion

4.1 Characterisation of STL

4.1.1 Functional groups

Fig. 1 (A-C) shows the FTIR spectra of unmodified STL, free fatty acid-modified STL (STL@FFA-WCO), and oleic acid-modified STL (STL@OA). Fig. 1 (A) shows the broad band at 3344.5 cm^{-1} that can be attributed to the hydroxyl groups on the unmodified STL. Based on this figure, two peaks can be observed between 2920 cm^{-1} and 2850.5 cm^{-1} that correspond to the asymmetric and symmetric C-H stretching of CH_2 groups. The peak observed at 1731.5 cm^{-1} can be attributed to C=O stretching. The strong band at 1624.5 cm^{-1} corresponded to the C=C stretching vibration.

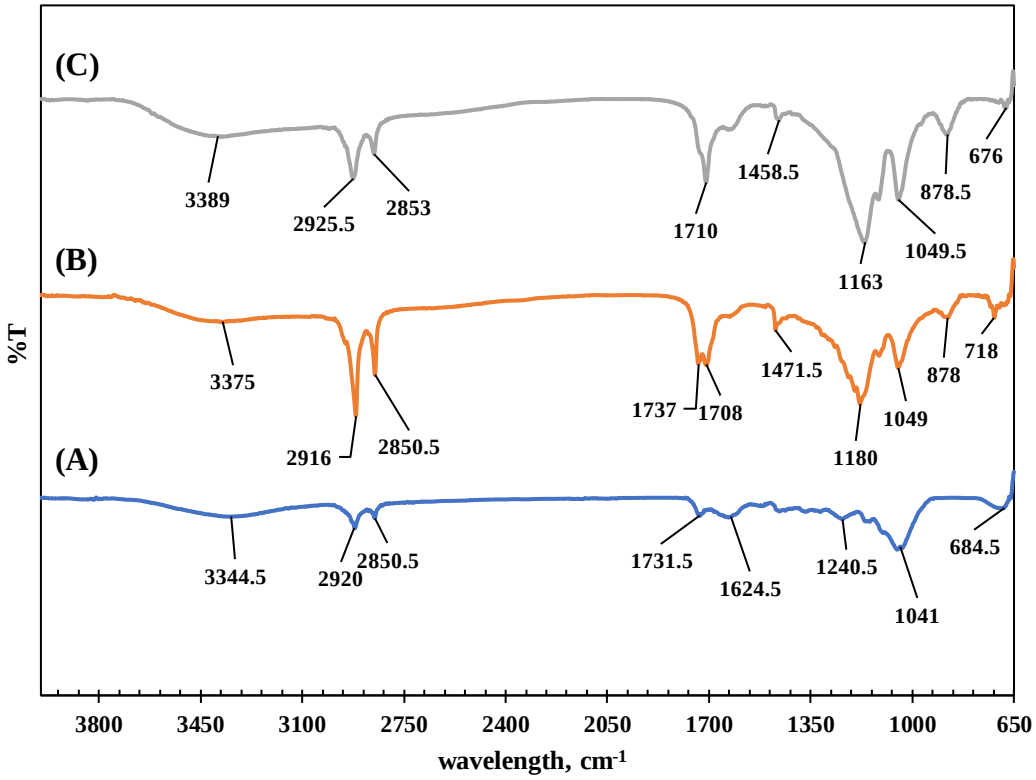

Fig. 1 FTIR spectra: (A) unmodified STL; (B) waste cooking oil-modified STL (STL@WCO); and (C) oleic acid-modified STL (STL@OA)

Fig. 1 (B) shows that the absorption band at 3375 cm^{-1} is weaker in the modified STL, which indicates that the hydroxyl groups have been reduced by the modification process. The increased peak intensity of CH_3 and CH_2 groups after STL modification indicated the successful impregnation of free fatty acids from waste cooking oil into the STL. The presence of two peaks observed at 1737 cm^{-1} and 1708 cm^{-1} corresponded to the stretching of carbonyl groups (C=O) of fatty acids from WCO in the modified STL.

Fig. 1 (C) shows that the peak intensity of the band at 3389 cm^{-1} is weaker than the band at 3375 cm^{-1}. This shows that most of the hydrophilic functional groups (−OH) in the STL@OA have been replaced by the oxalate group ($C_{17}H_{33}COO^-$) of the oleic acid. The presence of the adsorption band at 1710 cm^{-1} corresponded to the stretching vibration of carbonyl groups (C=O). The peak intensities of the bands at 2925.5 and 2853 cm^{-1} (alkane stretching) in the STL@OA spectrum are stronger compared to the peaks of unmodified STL, which indicate hydrophobic characteristics in the STL@OA.

Table 1 lists a summary of FTIR spectra of unmodified STL, STL@WCO, and STL@OA.

Table 1 FTIR summary

Material	Functional Groups	Absorption (cm^{-1})
STL	-OH stretching	3344.5
	Asymmetric and symmetric CH stretching	2920, 2850.5
	C=O stretching	1731.5
	C=C stretching	1624.5
	C=C bending	1041
STL@WCO	-OH stretching	3375
	Asymmetric and symmetric CH stretching	2916, 2850.5
	C=O stretching	1737, 1708
	C-O stretching	1180
	C=C bending	1049
	C-H bending	878
STL@OA	-OH stretching	3389
	CH stretching	2925.5, 2853
	C=O stretching	1710
	C-O stretching	1163
	C=C bending	1049.5
	C-H bending	878.5

4.1.2 Surface morphological analysis

SEM micrographs of unmodified STL and free fatty acid-modified STL (STL@FFA-WCO) are shown in Fig. 2 (A and B). Fig. 2 (A) shows that the unmodified STL have a rough and porous surface. These pores are useful for oil adsorption. The surface of the STL@FFA-WCO, as seen in Fig. 2 (B), is smooth and covered, indicating that free fatty acids from WCO have been successfully introduced onto the surface of STL. According to Rozi et al. [25], WCO mainly consists of long alkyl chains (C16 and C18) that have a hydrophobic interaction with oil. Therefore, modified fatty acids on STL will increase the rate of adsorption for oil pollutant.

4.2 Performance of the unmodified and modified STL for oil adsorption.

Table 2 shows the comparison between the adsorption capacity of the unmodified STL and STL@FFA-WCO, with a 1:10 ratio of STL to modification agent (WCO). The adsorption capacity of STL@FFA-WCO was 3.62 times higher than the unmodified STL. As discussed in the previous section, the presence of alkane bands after the WCO modification process has enhanced the hydrophobic

interactions between oil and the adsorbent's binding sites. This behaviour has increased the adsorption capacity of STL@FFA-WCO.

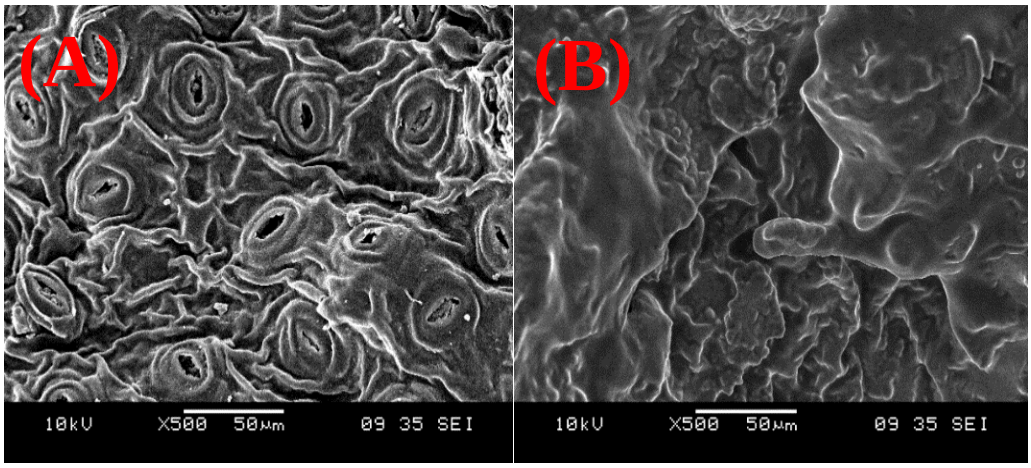

Fig. 2 SEM images: (A) unmodified STL; and (B) STL@FFA-WCO

Table 2 Comparison of adsorption capacity between unmodified STL and modified STL

Condition	Mass of adsorbent after adsorption, m_F (g)	Adsorption Capacity, q (g/g)
Unmodified STL	1.267 ± 0.058	0.267 ± 0.058
STL@FFA-WCO	1.967 ± 0.058	0.967 ± 0.058

Data represent the mean ± standard deviation (n = 3)

4.3 Effect of process parameters

4.3.1 Effect of ratio of STL to modification agent

Table 3 shows the adsorption capacity of STL@FFA-WCO at different ratios of 1:10, 1:20, and 1:30 STL to WCO. The adsorption capacities of STL@FFA-WCO at 1:10, 1:20, and 1:30 ratio of STL to WCO were 0.967 ± 0.058, 0.833 ± 0.058, and 0.66 ± 0.058 g/g, respectively.

Table 3 Adsorption capacity of STL@FFA-WCO at different ratios

Ratio	Mass of adsorbent after adsorption, m_F (g)	Adsorption Capacity, q (g/g)
1:10	1.967 ± 0.058	0.967 ± 0.058
1:20	1.833 ± 0.058	0.833 ± 0.058
1:30	1.667 ± 0.058	0.667 ± 0.058

Data represent the mean ± standard deviation (n = 3)

Waste cooking oil (WCO) consists of free fatty acids, as well as other components, such as boron, sodium, and phosphorus [26]. Boron molecules in the WCO will bind with the hydroxyl groups in STL to form borate compound [27]. Hence, when WCO ratio was increased, of the formation of carbonyl groups on the adsorbents was decreased. The carbonyl group would be capped with a long hydrocarbon chain, which would provide hydrophobic characteristics to STL for oil entrapment. Therefore, the STL sample that was modified using the ratio of 1:10 has a higher adsorption capacity compared to samples modified using the ratio of 1:20 and 1:30 of STL to modification agent.

4.3.2 Effect of contact time

Based on Table 4, the adsorption capacities of STL@FFA-WCO for different contact time of 30, 60, 90, and 120 min are 0.467 ± 0.06, 0.700 ± 0.10, 1.033 ± 0.06, and 1.400 ± 0.10 g/g, respectively. For the first 120 min, the adsorption capacity was increased from 0.467 ± 0.06 to 1.400 ± 0.10 g/g with

increasing contact time. This increment was due to the internal and external diffusion of palm oil particles onto the surface of the adsorbents via hydrophobic interaction. However, when the contact time was further increased to 150 min, the adsorption capacity was decreased from 1.400 ± 0.10 to 1.333 ± 0.15 g/g due to the desorption of oil from the active sites [28]. This decrement occurred because the active sites for oil to bind on to were decreasing on the surface of the adsorbent. This finding is in agreement with the findings reported by [29]. The adsorption capacity remained constant at 1.333 ± 0.15 g/g starting from 150 until 240 min.

Table 4 Adsorption capacity of STL@FFA-WCO at different contact times

Contact time (min)	Mass of adsorbent after adsorption, m_F (g)	Adsorption Capacity, q (g/g)
30	1.467 ± 0.06	0.467 ± 0.06
60	1.700 ± 0.10	0.700 ± 0.10
90	2.033 ± 0.06	1.033 ± 0.06
120	2.400 ± 0.10	1.400 ± 0.10
150	2.333 ± 0.15	1.333 ± 0.15
180	2.333 ± 0.15	1.333 ± 0.15
210	2.333 ± 0.15	1.333 ± 0.15
240	2.333 ± 0.15	1.333 ± 0.15

Data represent the mean ± standard deviation (n = 3)

4.3.3 Effect of adsorbent dosage

Table 5 shows the adsorption capacity of the STL@FFA-WCO at different adsorbent dosages. This table shows that when the adsorbent dosage was increased from 0.5 to 2.5 g, the adsorption capacity was also increased from 0.800 ± 0.20 to 1.587 ± 0.06 g/g. An increase in adsorbent dosage would result in more active sites on the surface of the adsorbent to become available for oil molecules to be adsorbed. Thus, at a higher dosage, the adsorbed oil content was higher because more empty active sites were available compared to at lower adsorbent dosages, which would have fewer active sites to adsorb oil.

Table 5 Adsorption capacity of STL@FFA-WCO at different adsorbent dosages

Adsorbent Dosage (g)	Mass of adsorbent after adsorption, m_F (g)	Adsorption Capacity, q (g/g)
0.5	0.900 ± 0.10	0.800 ± 0.20
1.0	2.400 ± 0.10	1.400 ± 0.10
1.5	3.667 ± 0.15	1.444 ± 0.10
2.0	4.967 ± 0.15	1.483 ± 0.08
2.5	6.467 ± 0.15	1.587 ± 0.06

Data represent the mean ± standard deviation (n = 3)

4.3.4 Effect of temperature

Table 6 shows that the adsorption capacities were increased when the temperature was increased from 25 to 45 °C. Then, the adsorption capacities began to decrease when the temperature was increased from 45 to 65 °C. The most efficient temperature, with the highest adsorption capacity (1.800 ± 0.15 g/g) was at 45 °C. This behaviour could be due to the increase in the rate of adsorption as the temperature was increased. However, beyond 45 °C, the adsorption capacity of STL@FFA-WCO began to decrease because of the increasing speed of its Brownian motion [30]. Therefore, more energy was required to adhere the oil molecules onto the adsorbent's surface. In addition, at a higher temperature, the randomness of the oil adsorption-desorption began to increase, which then may cause less probability of oil attachment on the adsorbent surface [31].

Table 6 Adsorption capacity of the adsorbent at different temperatures

Temperature (°C)	Mass of adsorbent after adsorption, m_F (g)	Adsorption Capacity, q (g/g)
25	2.400 ± 0.10	1.400 ± 0.10
35	2.467 ± 0.06	1.467 ± 0.06
45	2.800 ± 0.15	1.800 ± 0.15
55	2.467 ± 0.10	1.467 ± 0.10
65	2.200 ± 0.15	1.200 ± 0.15

Data represent the mean ± standard deviation (n = 3)

4.4 Adsorption performance of STL@FFA-WCO and STL@OA at optimum condition

As shown in Table 7, the adsorption capacity of STL@FFA-WCO and STL@OA was 1.733 ± 0.15 and 2.267 ± 0.21 g/g, respectively. The STL that was modified by OA (STL@OA) has 30% higher adsorption capacity compared to STL@FFA-WCO. This is because of the different alkyl chain of free-fatty acids (C12, C14, C16, and C18) present in waste cooking oil, which can also bind to the hydroxyl groups of STL during the modification process. However, the standard oleic acid used in this study was 100% C18, which provided the hydrophobic characteristics in the STL@OA. Thus, the percentage of oil uptake of STL@FFA-WCO was less than STL@OA.

Table 7 Effect of modification agents on the adsorption capacity

Modification agents	Mass of adsorbent after adsorption, m_F (g)	Adsorption Capacity, q (g/g)
FFA-WCO	2.733 ± 0.15	1.733 ± 0.15
OA	3.267 ± 0.21	2.267 ± 0.21

Data represent the mean ± standard deviation (n = 3)

4 Conclusion

Raw STL are originally hydrophilic-based material. To enhance its usage as an oil adsorbent, STL was modified with fatty acids. Modified STL have unique features, which led to the hydrophobic interactions, and thus, affected the rate of adsorption for oil removal for the pores of the adsorbent. The adsorption capacity of STL@FFA-WCO and STL@OA were significantly increased compared to the unmodified STL. The performance of these adsorbents was tested using various process parameters, such as the ratio of STL to the modification agents, contact time, adsorbent dosage, and temperature. The best modification ratio was 1:10 (STL:WCO). The maximum adsorption capacity was observed at 1 g of STL@FFA-WCO, with a temperature of 45 °C and contact time of 120 min. In a comparison between the types of modification agents in the optimum condition, STL@OA has the highest adsorption performance compared to STL@FFA-WCO. The adsorption capacity for STL@OA was 2.267 ± 0.21, which was 30% higher compared to STL@FFA-WCO. Thus, this present work has proven that STL modified by fatty acids has the potential to act as a green and innovative adsorbent for oil adsorption, even though STL are originally hydrophilic.

Declaration of Conflict of Interest

The authors declared that there is no conflict of interest with any other party on the publication of the current work.

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Acknowledgement

The authors would like to acknowledge the research funding for research activities through the National Collaborative Research Grant (4B514 and 9023-00014).

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