Impregnation of liquid natural rubber (LNR) foam with activated carbon for enhancing oil removal from water

Azwan Mat Lazim[a,*], M. Hilman[a], S. Saroni[a], N. Muslihuddin[a], A. Kamil[a], A.A. Rahman[a], A. Hafidz Yusoff[b], Mazlan Mohamed[b], N. Haida M. Kaus, Ekwipoo Kalkornsurapranee[c]

[a]*School of Chemical Sciences, Faculty of Science and Technology, The National University of Malaysia, 43600 UKM Bangi, Selangor, Malaysia, emails: azwanlazim@ukm.edu.my (A.M. Lazim), muhdhilman12@yahoo.com (M. Hilman)*
[b]*Faculty of Bioengineering and Technology, Universiti Malaysia Kelantan, 17600 Jeli, Kelantan, Malaysia, emails: hafidz.y@umk.edu.my (A. Hafidz Yusoff), mazlan.m@umk.edu.my (M. Mohamed)*
[c]*School of Chemical Sciences, Universiti Sains Malaysia, 11800, Pulau Pinang, Malaysia, email: ekwipoo@gmail.com (E. Kalkornsurapranee)*
[d]*Department of Materials Science and Technology, Faculty of Science, Prince of Songkla University, Hat-Yai, Thailand*
erifsyarifudin@gmail.com,nikmus94@gmail.com, akhsan199@gmail.com, amir950213@gmail.com, noorhaida@usm.my

ABSTRACT

Macroporous rubber foam made of liquid natural rubber (LNR) was fabricated at ambient temperature by using sodium bicarbonate (NaHCO$_3$) as foaming agent and S$_2$Cl$_2$ as crosslinker. Activated carbon was added through blending process to enhance oil sorption performance. The scanning electron microscopy morphology analysis showed that the LNR-foam has produced pores with diameter sizes between 50 and 150 µm whilst porosity volume was in range of 5.5–6.5 mol/cm^3. The swelling properties of this porous LNR-foam incorporated with activated carbon and their application as various oil types (petrol A, petrol B, diesel, olive, kerosene and engine oil) were evaluated. Result showed that absorption capacity was totally depending on the viscosity of oil. At optimum crosslinker concentration (6.5 w/w %), petrol A has been absorbed up to 5.47 g g^{-1} while olive oil approximately 1.00 g g^{-1}. The LNR-foams are able to be re-used up to 20 absorption cycles which indicates them as good reusability material. Therefore, this natural based foam is highly potential to be used as alternative absorbent that are readily available.

Keywords: Macroporous; Foam; Activated carbon; Rubber; Oil spill

1. Introduction

Oil spill pollution gives negative impact to the environment and living organism as oil itself can be graded as toxic because initially it is a combination of different chemical components. As hazardous material, oil is not only harmful to plants and animals but also endangered to their habitat. Oil spills on land consequently prevent water from being absorbed by soil, lowering the quality and productivity leading to destruction of the crops [1]. In water, oil pollution leads to devastating effects as it quickly spreads over the surface, producing a layer that prohibits oxygen from reaching aquatic plants and animals.

There were established techniques that have been used to manage the oil pollution; as example graphite and chalk, detergent, burning or microbes [2]. Although it has been used for sorbent material, graphite was known as an expensive material which was costly and needed complicated process which normally produced from petroleum coke. Though chalk is used to stop oil dispersant, it works ineffectively as absorbent due to its difficulty to uptake or discharge oil after absorption

* Corresponding author.

process. Moreover, the structure is built as complex block which makes more difficult for the oil to penetrate the voids. Whereas, the untreated detergent consequently caused aquatic organism especially fish to absorb more chemicals than they normally consumed. Making it worst, detergent becomes a catalyst to freshwater algal blooms that may generate toxin which endangered the marine life by reducing the oxygen concentration in water. Despite the fact that the burning technique may be a quick way to solve oil pollution, it creates air pollution from the toxic gases released. Using microbes is possibly an effective green technique to overcome the oil pollution. However, this technique is less favorable due to its low resistance toward temperature, pH and also difficult to produce in a large scale. The techniques previously mentioned perhaps offered solution to solve the oil spilled problem. However, on contrary the methods were tedious, ineffective and expensive [3]. Alternatively, oil sorbent material becomes the most convincing method for oil removal. Modifications are made to the sorbent material in order to achieve good absorbent criteria such as hydrophobic or oleophilic with high sorption capacity, selective, buoyancy and re-usability [4].

Nowadays, most synthetic polymers that are being used as oil sorbents are usually made of engine oil or their derivatives, which is a non-renewable resource. Therefore, current interest to bio-material based polymer becomes a viable alternative to petroleum-based polymer because it is classified as renewable sources. Attention has been given to natural-based sorbents that are made of cotton fibres, wood fibres, paddy husks or straw cellulose which were well known as abundant, renewable, biodegradable and cheap [5]. Although these materials elegantly can be used as candidates of oil sorbent materials, the only drawback of these natural-based materials is their low oil sorption capacity [6].

Preferably to clean up the oil contaminant, absorbent made of synthetic rubber such as polypropylene, polyurethane, butyl rubber and polyvinylchloride are usually used because of their hydrophobicity characteristics. Whilst, this chemically fabricated polymer is slow to degrade and pricey compared with the natural rubber (NR) [7]. There were attempts of using modified natural rubber for oil removal, as an example NR modified with poly(butyl methacrylate) (PBMA) and poly(butyl acrylate) (PBA) [8], NR latex with cationic polyacrylamide [9], grafted NR with polymethyl methacrylate (PMMA) and polybutyl methacrylate (PBMA) via emulsion polymerization [10]. However, the preparation proposed were quite complicated, tedious and demanded more chemicals in the process of producing the absorptive material. Consequently, these weaknesses reduce the necessity of manufacturing industry to use the NR widely although natural polymer is established as an economical material with various applications owing to their vast functional characteristics such as lightweight, good buoyancy, insulation and cushioning [11].

In this study, we proposed a simple modification to produce absorptive NR foams from liquid natural rubber (LNR) that can be used for oil removal. First, the NR was transformed into LNR where its molecular weight lighter with shorter polyisoprene chain compared with solid NR [12]. This privilege hence contributed to uncomplicated mixing

and crosslinking process. The foams were prepared by mixtures comprising of LNR and different amounts of disulfur dichloride (S_2Cl_2). Activated carbon was added to improve the performance of these LNR-based foams. Further investigations of oil uptake capacities of the foams were then measured including other characteristic features of the LNR-based foams.

2. Experimental

2.1. Materials

Natural rubber was obtained from Rubber Institute of Malaysia. Acetic acid, sodium bicarbonate and methanol were bought from Systerm®. The toluene solvent (HmbG® Chemicals) was used to produce LNR. Sulfur dichloride (S_2Cl_2) by EMD Millipore was employed to crosslink between the molecules of LNR polymer. The activated carbon was purchased from Evoaqua Water. The oil testing used in this study were Petrol A, Petrol B, kerosene, diesel, engine oil and olive oil. The characterizations of various oils used in this research are given in Table 1.

2.2. Preparation of liquid natural rubber

Method that is reported by Ibrahim et al. [13] has been used to prepare LNR using photochemical oxidation.

2.3. Preparation of absorbent foam with curing process

About 20 g of LNR was stirred with magnetic stirring bar together with 5 g of sodium bicarbonate at 400 rpm for 3 min. A set of samples have been tested to determine the optimum amount of activated carbon. Result was in agreement with research reported by Inagaki et al. [14]. Based on optimum result obtained, 1 g of activated carbon has been dissolved in 10 mL of acetic acid before being immersed into the mixture of LNR with the same speed. After the solution was fully homogenized, disulfur dichloride (S_2Cl_2) was added with appropriate amount followed by addition of 15 mL of water for curing process in 20 min at ambient temperature. The LNR-activated carbon foam (LNR-foams) was then washed with methanol for several times to remove remaining solvent. The LNR foams with different concentrations of crosslinker are shown in Fig. 1.

Table 1
Properties of oil tests

Type of oils	Origin and properties
Petrol A	Petronas RON95, viscosity = 6.14×10^{-4} PaS (25°C)
Petrol B	Petronas RON97, viscosity = 5.43×10^{-4} PaS (25°C)
Kerosene	Starbrand commercial grade, viscosity = 1.61×10^{-3} PaS (25°C)
Olive	Basso 1904 brand, viscosity = 6.97×10^{-2} PaS (25°C)
Engine	Local service station, viscosity = 1.88×10^{-1} PaS (25°C)
Diesel	Petronas, viscosity = 6.61×10^{-3} PaS (25°C)

Fig. 1. LNR foam with different concentration of crosslinker (w/w %) to LNR.

2.4. Oil sorption test

The oil sorption test was conducted based on ASTM F726-99: Standard Test Method for Sorbent Performance of Adsorbents. A cubic specimen of absorbent (1 cm × 1 cm × 0.5 cm) was cut and immersed in oil at ambient temperature. The absorbent was taken out after first 60 ± 2 s, then excess oil on the surface of absorbent was allowed to drain for 30 ± 3 s. The wet absorbent was then immediately transferred to a pre-weighed petri dish and weighed. The oil absorption has been carried out in range of 5–30 min. The oil sorption capacity (S_{oil}) (w/w) of absorbent was calculated as follows:

$$S_{oil} = \frac{m_{oil} - m_{dry}}{m_{dry}} \tag{1}$$

where m_{dry} is the initial weight of an absorbent while m_{oil} is the final weight of absorbent after immersed in oil at time t.

2.5. Crosslinking density

The crosslink density was measured based on Flory–Rehner equation using ASTM D471. The adsorbent was cut into different sizes, before weighed using analytical balance. The samples were immersed in a glass vessel containing toluene for 6 h. After that, the samples were taken out from solvent, wiped with filter paper to remove excess solvent, then weighed. This test is to determine the swollen weight. The Flory-Rehner equation was calculated as:

$$-\left\{ \ln\left(1 - V_r\right) + V_r + XV_r^2 \right\} = pV_o M_c^{-1} V_r^{1/3} \tag{2}$$

where X = Flory–Huggins interaction parameter, ρ = rubber density, V_o = molar volume of the solvent, V_r = volume fraction of swollen rubber gel, M_c = average molecular weight between crosslinks.

The volume fraction of swollen rubber gel, V_r, was calculated according to Eq. (3) as follows:

$$V_r = \frac{\dfrac{X_r}{\rho_r}}{\left(\dfrac{X_r}{\rho_r}\right) + \left(\dfrac{X_s}{\rho_s}\right)} \tag{3}$$

where ρ_s = density of toluene, ρ_r = density of the rubber, X_s = mass fraction of toluene, X_r = weight of the rubber, X_s and X_r can be obtained from the equation below:

$$X_s = \left(\frac{\text{Weight of swollen sample} - \text{Original sample}}{\text{Weight of swollen sample}} \right) \tag{4}$$

$$X_r = 1 - X_s \tag{5}$$

Then, the value of M_C is used to calculate the physical crosslink density according to the following equation:

$$[X]_{Phys} = \frac{1}{2M_C} \tag{6}$$

2.6. Characterization

2.6.1. Fourier-transform infrared

The functional groups present in the structure of LNR and LNR foams were observed by using PerkinElmer (USA) with attenuated total reflection attachment. This instrument can determine the crosslinking reaction that occurs between the inter-molecule of polymer.

2.6.2. Contact angle test

The water contact angle test was performed to investigate the surface wettability of absorbent with DSA1/Kruss at ambient temperature. The syringe was positioned in such a way that the probe liquid (3 μL) could contact the surface of absorbent.

2.6.3. Swelling test

The swelling test was conducted to determine the sorption capacities of the foam toward suitable organic solvent of polymer such as toluene. The different ratio of filler in LNR foam was observed for the optimum swelling. Results were compared with crosslinking densities of the foam. The swelling test was then calculated as:

$$\text{Swelling ratio} \left(\%\right) = \frac{W_S - W_I}{W_I} \times 100\% \tag{7}$$

where W_S and W_I are the weights of swelled and initial sample, respectively.

2.6.4. Morphology test

Scanning electron microscopy (SEM, LEO 1450 VP) was used to examine and determine the average size of pores composed in LNR foams and all analyses were conducted

at low vacuum at 20 kV. The surface of the absorbent was observed to measure the average size of pores that have been formed via crosslinking reaction. The pore volume was determined through absorbed methanol of dry foams. Methanol was used because it provided a non-solvent system for the polymer which allowed the solvent to penetrate the polymer network pores easily as suggested [15]. The pore volume of absorbent was calculated as the following equation:

$$V_p = \frac{M_m - M_{\text{dry}}}{d_m \cdot M_{\text{dry}}} \tag{8}$$

where M_m(g) is the total weight of absorbent, M_{dry} is the weight of dry LNR foam and d_m is density of methanol (0.792 g mL^{-1}).

3. Result and discussion

3.1. FTIR analysis

Fig. 2 shows the IR-spectra of LNR and prepared LNR foams. In general, FTIR is used to identify the presence of functional groups of polyisoprene chains especially after the vulcanization process. Broad absorption peak was observed for both samples at 3,394 and 3,401 cm^{-1} pointed out the domination of hydroxyl group (OH). This may be also contributed by methanol utilization during the absorbent preparation. The vibration bend at 2,961 cm^{-1}; 2,923 cm^{-1} and stretching at 1,447 cm^{-1}; 1,452 cm^{-1} were referred as methyl group (=C–CH$_3$) which attributed to the main structure of LNR (polyisoprene). Observation at 1,734 and 1,712 cm^{-1} for LNR foam sample was referred to carbonyl group of LNR. The sulfur-carbon stretching exhibited at 755 cm^{-1} which showed the replacement of hydrogen atom (C–H) with sulfur atom (C–S–C) in the polymeric networks during the vulcanization process. The decrement in the intensity of LNR absorbent peaks for IR spectra has represented that there was active interchange of functional groups of the polymers.

3.2. Characterization of foam

3.2.1. Contact angle

The contact angle obtained was 95.9° ± 0.48° which assigned the LNR-based foam as a hydrophobic type material. As a quick reference, when the value of contact angle is more than 90° suggests the rubber foam absorbent has good water repellent characteristics. The hydrophobic behavior thus lowering the LNR foam's affinity toward water compared with oils. Surface composition and the surface topography of a composite affected its wettability or water repellancy in particular for porous material with open-cell structure [11,23]. This clearly can be seen in Fig. 3 where the water droplet steadily maintains approximately about 5 s without quickly dispersed to surface of the LNR foam. Similar results were reported by Chin et al. [11] where their findings suggested that the result obtained has proven that LNR-based material was oleophilic and undeniably suitable for oils clean-up.

For the swelling tests, the dried LNR foam was immersed in organic liquid, toluene. In this study, LNR foam ability to absorb the oil and swell is determined by its mass changes. The different ratio of activated carbon added and crosslinker density to produces foam samples are shown in Fig. 4.

As shown in Fig. 4, LNR foam sample of 6.5 w/w% has demonstrated the most optimum composite in swelling test, which absorbed toluene up to 5.87%. As the crosslinker concentration increased to 7.5 w/w%, the absorbency of LNR foam dropped to 4.5%, which was the lowest intake of toluene. These results suggested LNR foam of 6.5 w/w% sample gave an ideal of crosslinker to fabricate the composite, where the LNR foam forms consummate pores which allows solvent to penetrate easily via the polymeric structure. However, the crosslinking reaction between sulfur molecules and the polymer chain increased by upscaling the amount of crosslinker. As the crosslink density of polymeric material increased, the LNR foam consequently became stiffer, hard to swell and less solvent able to infiltrate the material [16]. Moreover, addition of activated carbon into LNR foam also resulted in lowering down the foam swelling capability. It was owing
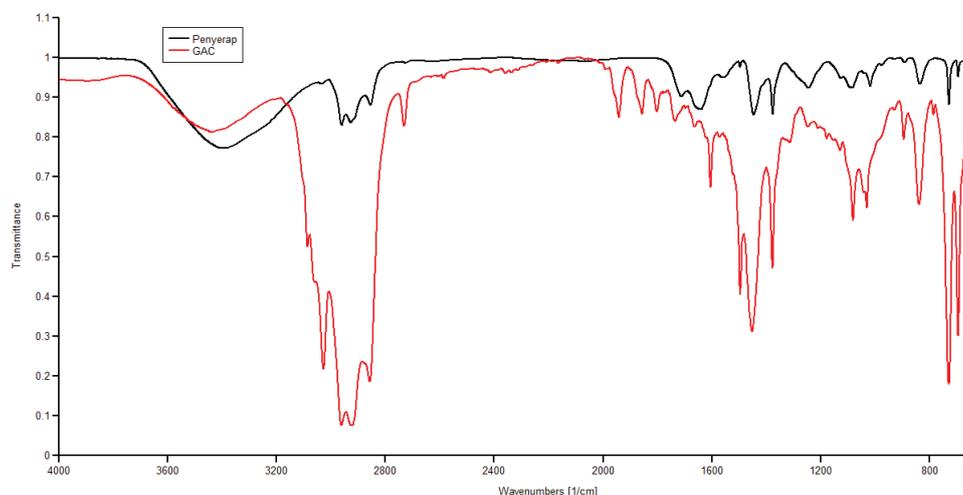

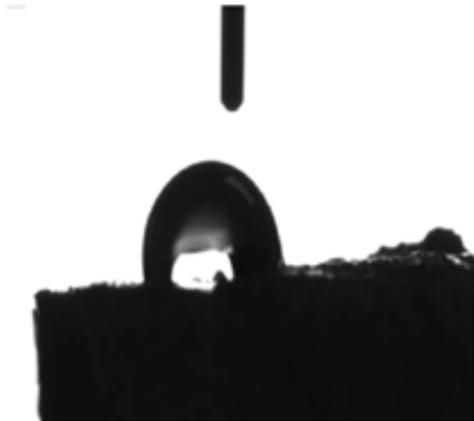Fig. 2. FTIR spectrum of LNR (black) and LNR foam (red).

Fig. 3. Contact angle test of LNR-based selective foams absorbent (6.5 w/w %) which taken in 5 s.

Fig. 5. Pore volume of LNR foams at different concentrations used.
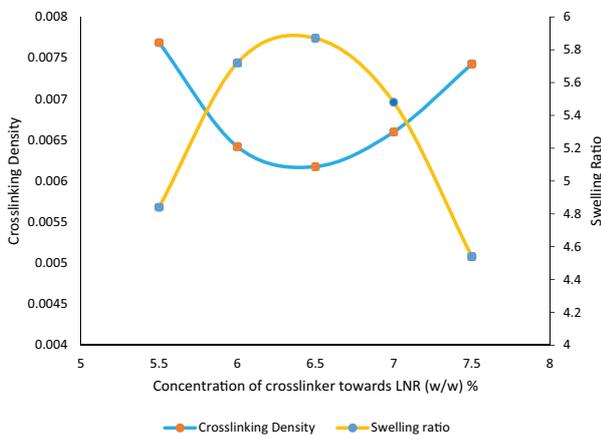

Fig. 4. Swelling ratio and crosslinking density of LNR foam in a function of crosslinker concentration.

to homogenously dispersed activated carbon and filled into polyisoprene polymer networks resulting in increasing of foam density plus reducing the voids availability to interact with solvent [17].

The pore volume of foam was measured in the swollen state which in this study, samples were immersed in methanol. Methanol has been chosen compared with other solvents because of its non-polar property, which able to permeate the pores without any interaction. According to Ibrahim et al. [13] in order to confirm methanol is completely filled into the absorbent, pore volume method is used. In this method, methanol diffused into pores of absorbent caused the absorbent swelled until it achieved a maximum absorption capability which is determined by the constant weight of the absorbent after several hours. As shown in Fig. 5, LNR foam of 6.5 w/w% has the highest pore volume with optimum amount of crosslinker and activated carbon content. Of all samples, rubber foam of 5.5 w/w% exhibited the lowest pore volume properties. According to Chin et al. [11], crosslinker plays important roles in developing networks between the monomers. Below an optimum amount of crosslinker, the
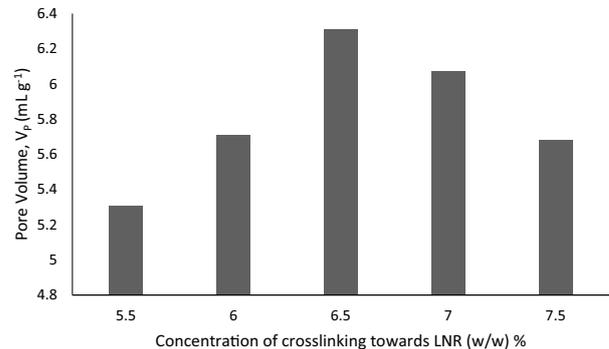
crosslinking process does not occur properly resulting in weak polymeric networks and not able to hold the solvent in its voids.

The efficiency of LNR foams in oil absorption using activated carbon was further investigated using different oil types which was conducted at ambient temperature (Fig. 6). There are two parameters usually used to determine sorbent ability; oil absorption kinetics and absorption capacity. Pertinent pore structure of absorbents helps suitable solvents penetrate easily and reduces time to achieve an absorption equilibrium [18–20]. The results demonstrated that most of oil absorption adequately occurred from 0 to 80 min and gradually achieved the equilibrium from 80 to 180 min, respectively. In general, these LNR foams show excellent absorbencies, depending on the oil density and viscosity. High viscosity will reduce the efficiency of oil sorption. As example, Petrol A and B were the highest of oil sorption whereas olive oil was the lowest. Light viscosity and small molecules allowed the oil to move faster, quick dispersion into the absorbent pore ducts meanwhile simultaneously solvated the foam networks. For high viscosity oil types, however, the molecules diffussed at a very slow rate and retained in pores consequently probihited networks solvation and swelling thus consumed more time to reach an equilibrium [21]. The absorbency of high viscosity oil absorption was below 1.5 g/g⁻¹. Similar trend was observed, where the LNR foam of 6.5 (w/w %) performed the best absorption toward each type of oil samples, while foam of 5.5 (w/w %) demonstrated the lowest efficiency. The performance of each absorbent was related to their porosity which is shown in scanning electronic microscope image (Fig. 7).

SEM is a tool to determine particles sizes, shapes and porosity including surface morphology. SEM images of different ratio LNR foam are presented in Fig. 7 while pore volumes of each sample have been determined using Eq. (7). At high magnification (×100 μm), the LNR foam exhibited as disordered macroporous. Considerable number of pores with irregular sizes allowed the foam to hold oil molecules that are entrapped in these voids. Despite applying higher magnification, the SEM images cannot determine morphology of activated carbon due to its small particles and well dispersed in the foams.

It can be observed in Fig. 7a that LNR foam of 5 w/w % exhibits with minimal pore cells appearance (10–30 μm)
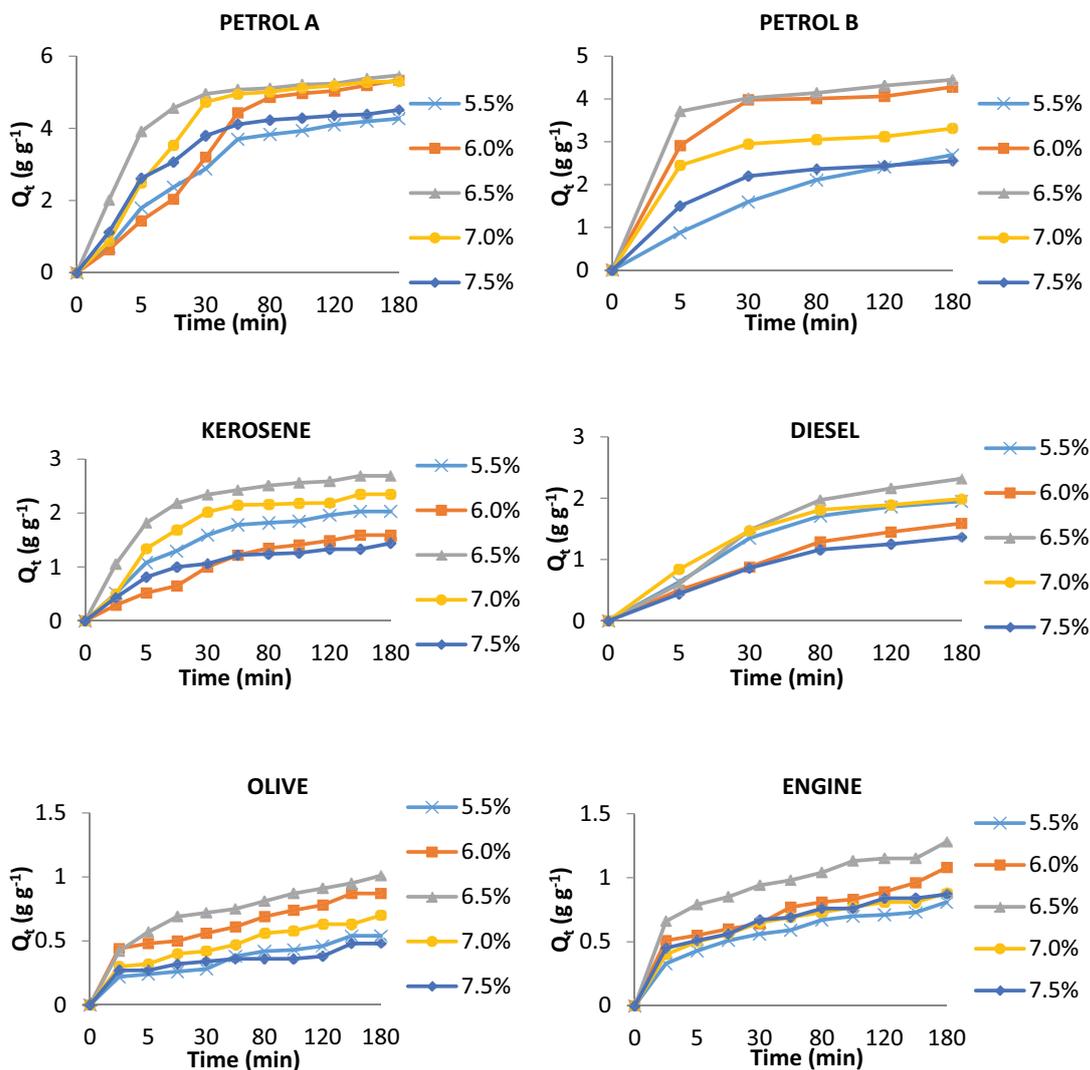
Fig. 6. Capability of LNR foam absorption of different oil types.

because of its low crosslinking formation resulting in the foam aggregations. On the other hand, it was also proved by the lowest pore volume absorbed compared with the other samples. By adding the crosslinker gradually, there was increment of the pore cells and better surface properties of foams obtained. At an optimum amount of mixture components, crosslinking formation between the rubber isoprenes and crosslinker was also maximized. Result demonstrated that LNR foam of 6.5 w/w % has good interconnected, large pore structures (20–120 μm) that helped to amplifying the dispersion and aided quick absorption especially for organic solvents. Due to the increment of crosslinker consequently made the pore cells LNR foam of 7 w/w % and 7.5 w/w % (Figs. 7d and e) collapsed constantly as it was over-cured during the vulcanization process [22–25]. Therefore, formations of pore cells on the surface discontinued, reduced the absorption rate and roughened the physical appearance of the foams.

Good reusability is another important property considered for an absorbent. In real application, saturated absorbents usually went through mechanical press to accumulate the absorbed oil before the same absorbents are ready to be reused. In this study, the reusability test of rubber foam was carried out via repeated absorption and desorption processes. Fig. 8 showed the oil sorption capacities of rubber foams after being subjected to 20 cycles (5 min interval per cycle) of sorption-squeezing process. The oil sorption capacities were found to increase steadily as the sorption-squeezing process was repeated and then decreased gradually upon reaching a certain sorption limit. Reusability of absorbents was performed differently based on oil viscosity.

Results showed that low viscosity oil category such as fuel A, fuel B and kerosene had the average absorption capacity of 5.73, 4.67, 2.77 g g⁻¹ and recovery ability of 92.33%, 73.33% and 51.20%. On contrary, the trend was different to oil that was classified as high viscosity (diesel, engine and olive oils)
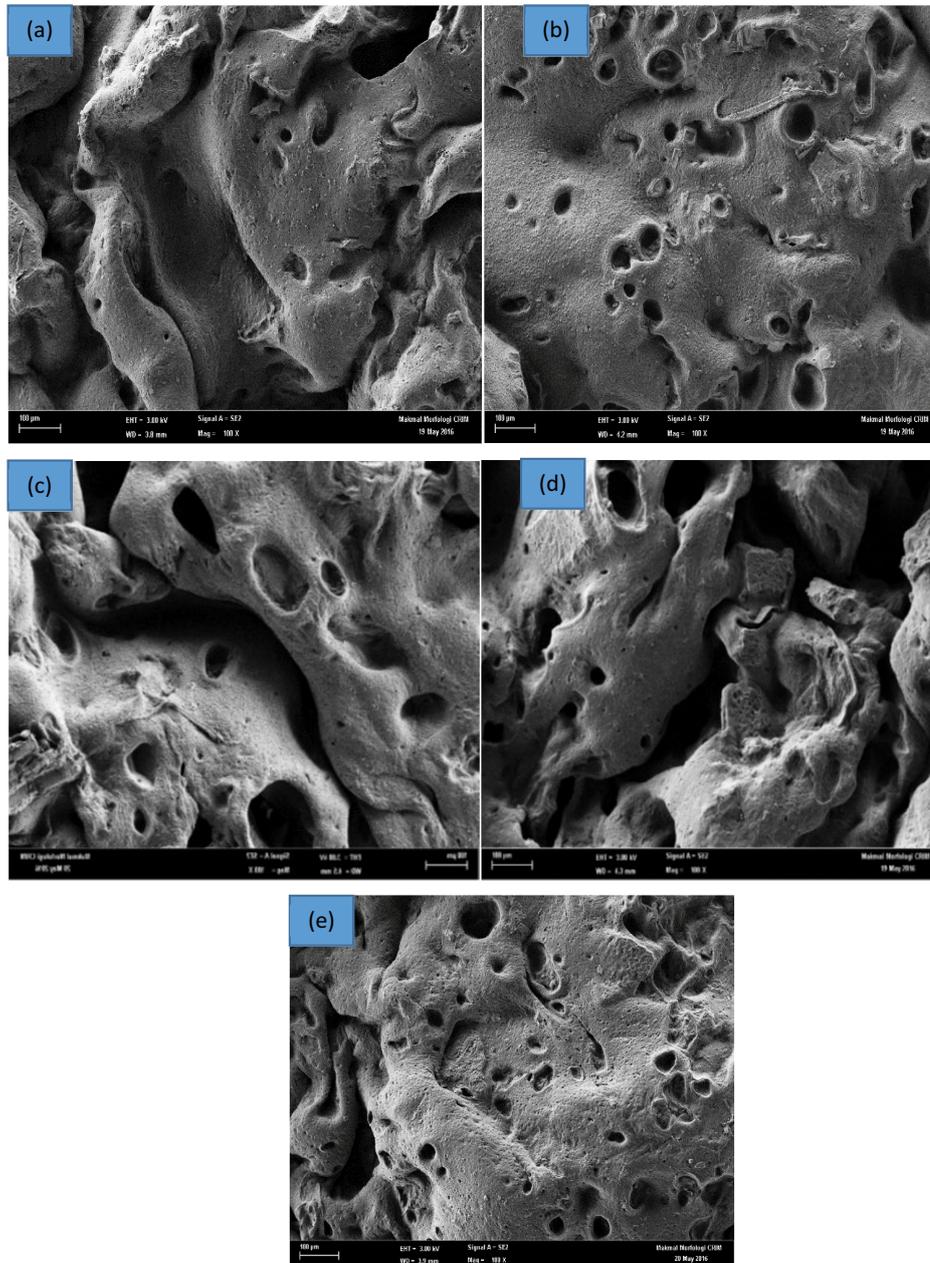
Fig. 7. Micrograph of LNR foams at different crosslinker used (w/w%); (a) 5.5, (b) 6.0, (c) 6.5, (d) 7.0 and (e) 7.5.

3.04, 1.34 and 0.85 g g⁻¹ of average absorption capacity and their recovery was 44.01%, 29.75% and 29.67%, respectively. There was no water absorbed into the foam although it was immersed into the water after the reusability test, confirming that the hydrophobicity behavior still remained.

4. Conclusions

In summary, the modified LNR-foam was successfully prepared by combination curing and foaming method with activated carbon. The activated carbon dispersed through the LNR foam improved the hydrophobicity/oleophilicity of composite. In sorption tests for various types of oil, modified LNR foam incorporated with activated carbon showed excellent absorption performance for Petrol A and B compared with other oil. LNR-foam with the maximum sorption capacity was demonstrated by 6.5 w/w % activated carbon, which could absorb 5.3 g/g Petrol A, 4.45 g/g Petrol B and 2.69 g/g kerosene and below than 2 g/g for other oil. After 20 cycles of sorption-squeezing processes, the sorption capacity was still in excellent condition. This study proposed that a material made of natural rubber has high continuous sorption capacity for different types of oil which can possibly be used in the clean-up of large-scale oil spills.
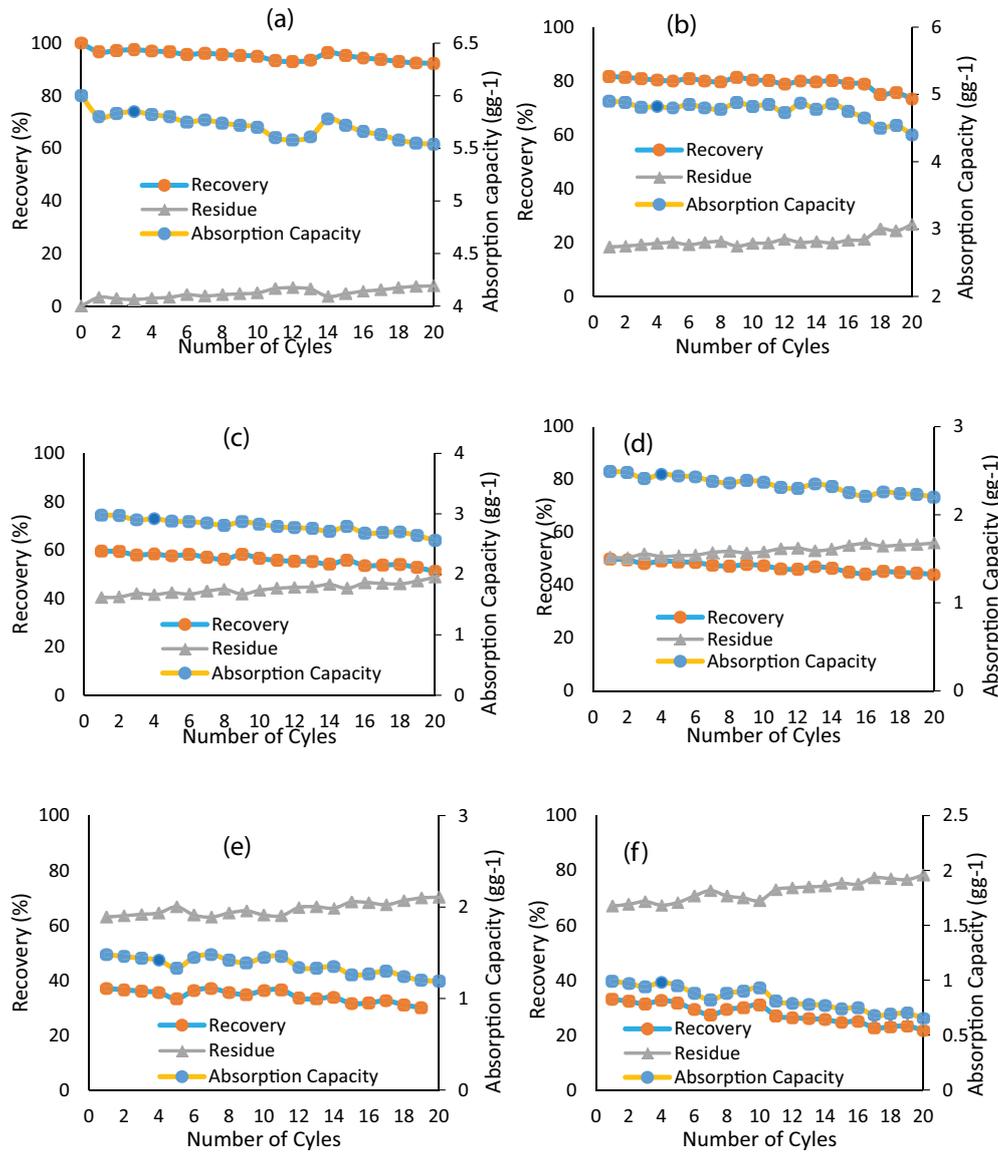
Fig. 8. Reusability of foam after has been applied in different oil samples (a) petrol A, (b) petrol B, (c) kerosene (d), diesel and (e) engine oil.

Symbols

d_m	—	Density of methanol
S_{oil}	—	Oil sorption capacity
m_{oil}	—	Final weight of absorbent after immersed in oil at time t
m_{dry}	—	Initial weight of an absorbent
V_r	—	Volume fraction of swollen rubber gel
V_o	—	Molar volume of the solvent
M_c	—	Average molecular weight between crosslinks
M_C	—	Used to calculate the physical crosslink density
M_m	—	Total weight of absorbent
M_{dry}	—	Weight of dry absorbent
ρ	—	Rubber density
ρ_s	—	Density of toluene

ρ_r	—	Density of rubber
W_s	—	Weights of swelled
W_I	—	Weights of initial sample
X	—	Flory–Huggins interaction parameter
X_s	—	Mass fraction of toluene
X_r	—	Weight of the rubber

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