



Research article

Polypropylene/lignin blend monoliths used as sorbent in oil spill cleanup

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ABSTRACT

With increasing industrial development, frequent oil spillages in water; therefore, it is imperative and challenging to develop absorbents materials that are eco-efficiency, cost-effective, and pollution prevention. In this study, sorbents obtained from Lignin incorporated with Polypropylene in different levels loading 0, 10, 20 % wt using thermally induced phase separation Technique (TIPS). The Polypropylene/Lignin blend monoliths were fabricated and compared in terms of morphological, thermal, and wetting characterizations. The successfully blending of different lignin concentrations with preserved the chemical structure of the polymer was confirmed by FTIR analysis. Thermogravimetric tests displayed that the existence of Lignin has changed the onset temperature (Tonset) of the blending sorbents, decreasing as the loading of Lignin is increased. The contact angle measurement showed a decrease in the hydrophobicity of sorbents with increasing lignin loading. Polypropylene/Lignin blend monoliths showed better absorption toward oils (soybean – engine) as compared to Polypropylene itself. PP10L showed an improvement in the oil sorption capacity around 2 times compared to the Polypropylene. These excellent features make Polypropylene/Lignin blend monoliths more competitive promising candidates than commercial absorbent.

1. Introduction

Polyolefin polymers are derived from fossil fuels, which have dangerously adverse on the ecological systems and humankind's health. Sources of energy resources are limited, and their stockpile is decreasing very fast, which makes this source very expensive. Recently, scientists have given great attention to biomass-based resources [1, 2, 3, 4, 5]. From these biomass resources, Lignin considers as the second abundant natural attractive material in Plantae (second to cellulose) [6]. Further, Lignin is available as industrial waste material from the pulp industry and paper making sector in large quantities; a small portion (about 2%) is effectively utilized, and most of the Lignin is burned or discharged into rivers, thereby causing serious environmental pollution and grievous waste of renewable resource [7]. The unique properties of Lignin, such as environmentally-friendly, sustainable, biodegradable, inexpensive material, thermal stability, and better mechanical features, these attractive properties have motivated recent interest of using Lignin in the industry that can contribute to the reduction of the environmental problem [8, 9]. Besides, Lignin is an irregular three-dimensional complex and amorphous polyphenolic molecule, which contains a great number of polar functional units making it promising renewable materials for being unitized in polymer blends and composites [10, 11, 12]. Related functional groups, chemical components, and inter-unit

linkages existing in Lignin are alkyl-aryl ether, aliphatic and aromatic hydroxyl units, and low polydispersity, which present the possibility for significant value-added applications in renewable sources development [13].

These prominent properties have driven to the utilize of Lignin in various applications, including oil-water separation [14]. Lignin has been recognized as a compatibilizer in polymer matrix composites. The impactful use of Lignin-based thermoplastic polymers has been prepared with Polypropylene [15, 16], Polyethylene [17], Polylactide acid [18], Polyurethane [19], Polyvinyl alcohol [20], Polyamide [21], and Polystyrene [22] as the polymer matrix. Among them, Polypropylene is widely used in the world in many applications and processed in several ways [23]. One of these practical applications, using Polypropylene as sorbent materials in oil spillage clean up, attracted increasing attention [24, 25, 26, 27]. Sorbents consider one of the most efficient physical methods used in oil-water cleanup according to unique properties [16, 28, 29, 30]. The ideal features of sorbent material used to remove the spilled oil from the contaminated area should contain hydrophobicity/oleophilicity surface, high sorption capacity, buoyancy, and reusability [31, 32]. So, Polypropylene with above advantages are a potential candidate for the sorption of oil. Despite the wealth of information about Polypropylene advantages. Polypropylene sorbents are

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Table 1. Properties of the test oils.

Oil	Soybean oil	Engine oil (15 W-40)
Viscosity (mPa.s)	65.30 (1.14)	234.50 (0.89)
Density (g.cm ⁻³)	0.92 (0.016)	0.87 (0.013)
Surface tension (mN.m ⁻¹)	33.41 (0.50)	30.78 (0.53)

considered to be mainly responsible for this phenomenon known as white pollution, which cause in the gathering of plastics on earth [33].

To overcome this problem and to reduce the dependence of oil feedstocks, there is an increased demand to use renewable sources for bio-economic perspective consideration [34]. Consequently, partial replacement of Polypropylene by Lignin is economically and ecologically gained attention for versatile applications [35]. Lignin combination in the Polypropylene matrix at various loading levels was presented [36, 37, 38, 39]. The development of novel synthetic strategies to simplify miscible polymer/lignin blends with massive efforts has been made to recognize the potential advantages of Lignin for the growth and develop new thermoplastic polymer mixture [40]. From current techniques, the thermally-induced phase separation (TIPS) is a template-free process by simply dissolving a polymer in a suitable high-temperature solvent, accompanied by the polymer solution cooling. The phase separation takes place in the cooling cycle, creating an interconnected porous structure. TIPS define the approach as easy operation, energy-saving, and adaptable process [41, 42, 43, 44].

To the extent of our knowledge, the incorporation of Lignin in Polypropylene blends monoliths through using the thermally induced phase separation technique (TIPS) method has not been reported yet. In this work, different loading levels of Lignin were incorporated in Polypropylene via using the TIPS method to fabricate low cost and environmentally friendly sorbents. The objective of this experimental analysis examined the impact of Lignin to improve the properties of Polypropylene/Lignin blends monolith in terms of thermal and wetting properties. Further, the spreading of oils was characterized by plotting dynamic contact angles of oils as a function of contact time. The purpose of this experimental work was to assess the impact of Lignin on the thermal and wetting properties of Polypropylene/Lignin blends monolith. Moreover, the spreading of oils was plotted the dynamic contact angles of oils as a function of time.

2. Materials and methods

2.1. Materials

Polypropylene pellets (PP) with melt flow index (MFI = 1.65 g/10 min), Acetone, Decalin, 1-butanol, and Lignin (L) were purchased from (Aladdin, China). Two different oils, namely engine oil, and soybean oil were used. This represents medium-viscosity and high-viscosity oils. The densities and surface tensions of oils were examined using dynamic contact angle tester (DCAT1), and ASN2 Digital Rotary Viscometer tested the viscosities. Every test was repeated three times to obtain an average value. The test temperature was maintained at 22 ± 2 °C. The properties of the studied oils are reported in Table 1.

2.2. Fabrication of polypropylene/lignin blends monoliths

The monolith PP has been fabricated via using the thermally induced phase TIPS method accomplished as follows. PP pellet 2.8 g dissolved in the mixed solvent of decalin 16 mL and 24 mL 1-butanol at temperature 115 °C. After that, the solution is cooled at temperature 20 °C to form the phase separation of the polymer as a monolith. The monolith was immersed with acetone and subsequently dried under vacuum. For the synthesis of the PP/Lignin blend monolith during making PP solution with solvents, dried Lignin was added with different total mass percentages 10 wt%, and 20 wt% of Lignin, which was labeled PP10L and PP20L, respectively. Figure 1 demonstrates the general procedure for the manufacture of monolithic.

2.3. Characterization

The morphology of PP/lignin blends surface was performed using scanning electron microscopy (SEM-TM3000, Hitachi, Japan). For SEM observation, Samples were cut then fixed on double tape after that plated with a thin film of gold before measurement. The acceleration voltages value used in SEM analysis was 15KV. The tested samples were characterized by Fourier-transform infrared spectroscopy (FTIR) (Nicolet 6700, Thermo Fisher, USA). The FTIR spectra were recorded in range 500–4000 cm⁻¹ wavenumber using attenuated total reflection (ATR) method with a resolution of 4 cm⁻¹. The thermal stability was investigated using Thermogravimetric Analyzer (TGA 4000, PerkinElmer, USA). Samples

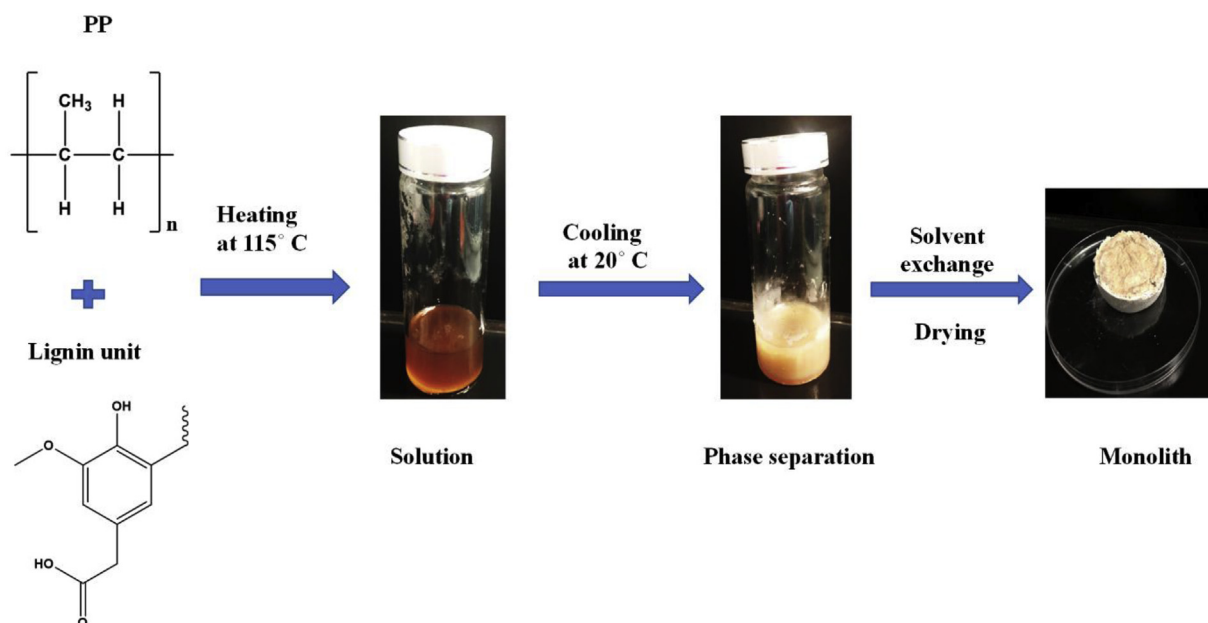


Figure 1. A general protocol for the preparation of PP/Lignin blend monoliths by a thermally induced phase separation technique.

masses (~10mg) were heated in the range from 30 °C to 700 °C at a heating rate 10 °C/min under nitrogen flow (20 mL/min). The characterization of wettability properties (hydrophobic - oleophilic) of tested samples was measured using contact angle values of oils and water. The contact angle was taken from optical contact angle device (OCA15EC, Data physics, Germany). Tested samples were attached to a glass slide by double side tape and fixed in the form of a plane. The static sessile drop method was used to measure water contact angles. Oil droplets would spread out immediately by using a static sessile drop method. Hence, the captive bubble method was adopted by making an air bubble on the immersed tested monolith surfaces in oil by using the “U” shaped needle. Each liquid was repeated five times and took the average. The test temperature was maintained at 22 ± 2 °C [45].

2.4. Analysis of dynamic oil spreading

PP/Lignin blends have oleophilic nature; once oil touches the surface of samples; the oil will spread on the surface. In this study, oil droplets (5ul) instantly spreading on each tested sample, which attached to a glass slide and mounted in the form of a plane. The spreading process of oils on the surface of each sample was recorded by a CCD camera installed in OCA15EC. The spreading rate of two oils was characterized by plotting the dynamic oil contact angles as a function of contact time.

2.5. Determination the oil absorption

Absorption ability in the oil system was studied by weighing the samples before (W_i) and after (W_t) immersing them in oil (Soybean, Engine) as percent weight gain (Q%) per the following equation. The test was repeated several times until the absorption process reached equilibrium [46, 47]:

$$Q (\%) = \frac{W_t - W_i}{W_i} \times 100 \quad (1)$$

The recyclability of samples for oil sorption was determined via using soybean oil, the sample with oil was squeezed, and then squeezed sample was again used with the same procedure as described above. The sorption – squeezing process was repeated many times under identical conditions to evaluate the reusability of the sorbent. The test temperature was maintained at 22 ± 2 °C.

3. Results and discussion

3.1. Morphological observation

In order to further investigate the interaction between Lignin and Polypropylene in blending scanning electron microscopy was done. SEM micrographs of samples were shown in Figure 2. Samples showed that

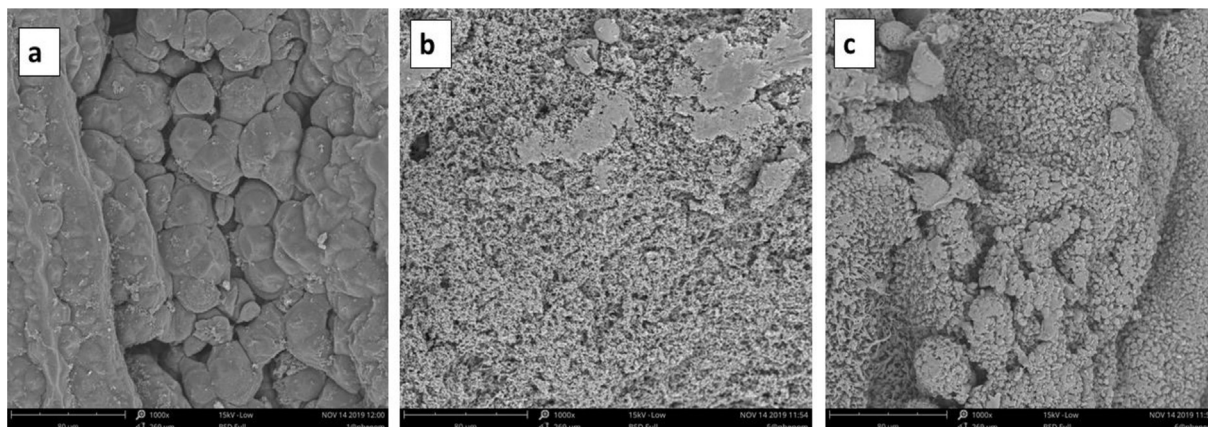


Figure 2. SEM image a. PP b. PP10L c. PP20L.

monolith had three-dimensional interconnected porous structures with small macropores, which might be explained to the phase separation of the polymer solution during the cooling process [48]. This structure allows the good access of oils inside of the monolith. It is clearly visible from images that pure PP had a smooth surface. While with addition Lignin aggregates with a rough surface were observed, which indicates the small lignin particles embedded in PP matrix successfully [49].

3.2. Fourier-transform infrared spectroscopy (FTIR)

FTIR spectrum of tested samples between wavenumbers 4000 cm^{-1} and 500 cm^{-1} was shown in Figure 3 and Table 2. For Lignin, a strong absorbance at range 3500 cm^{-1} - 3400 cm^{-1} assigned to –OH stretching vibrations were caused by the presence of alcoholic and phenolic hydroxyl units involved in hydrogen bonds [50]. The hydroxyl groups considered as functional organic moiety on the Lignin, that was the reactive sites and supported to change the polarity of the Lignin surface that can prohibit their dispersion in hydrophobic polymers composites. Bands with great intensity at 2936 cm^{-1} and 2846 cm^{-1} can be assigned to C–H of methyl and methylene units [40, 51]. Other essential structures in Lignin were medium bands which centered in carbonyl region, bands at 1710 cm^{-1} that is associated with conjugated carbonyl stretching. Aromatic skeletal vibrations were seen at peak 1510 cm^{-1} beside; it was found more intense at higher loading of Lignin. Other bands at 1597 cm^{-1} and 1427 cm^{-1} were representative aromatic rings units due to aromatic skeleton vibration [52]. By comparing the spectrum of PP/Lignin blends monoliths' bands with PP and Lignin, it was observed that peaks come from Lignin became visible in all blend monoliths. PP/Lignin blend monoliths showed peaks at 2956 cm^{-1} , 2916 cm^{-1} , 2866 cm^{-1} and 2836 cm^{-1} which represented (C–H stretching). Two bands were observed at 1373 cm^{-1} (syringyl group) and 1360 cm^{-1} (C–H₃ bonding and C = O stretch) [53]. PP/Lignin blends monoliths have these peaks, like PP. It can be recognized from a typical FTIR spectrum of added Lignin in PP blends, as shown in Figure, the composition of the bonds did not change but increased the intensity of the peaks. Moreover, Lignin was detected at 1250 cm^{-1} , 1210 cm^{-1} , 1115 cm^{-1} , and 1037 cm^{-1} , corresponding to the vibration of syringyl rings and guaiacyl rings, which was observed in the spectrum of PP/Lignin blend monoliths [54]. This indicated the successful process of addition Lignin was achieved in different proportions combined with preserving the chemical structure of PP. Table 2 shows the main characteristic bands observed for tested samples [55, 56].

3.3. Thermal properties

Figure 4 presented the effect of Lignin on the thermal stability of blends under the nitrogen atmosphere. It was observed from figures; the percentage mass loss was given as a function of temperature. Table 3

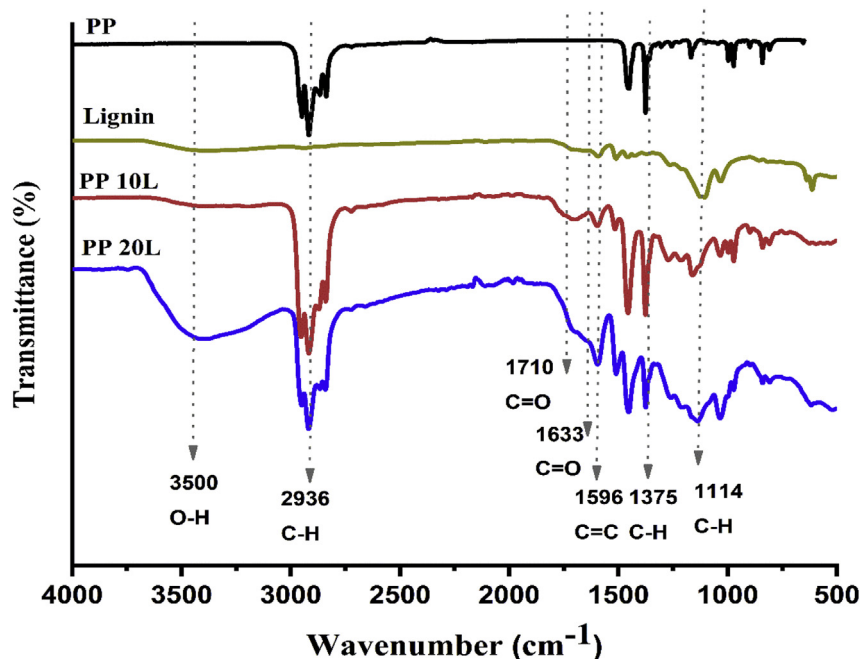


Figure 3. FTIR spectra of Polypropylene, lignin, and Polypropylene/lignin blends monoliths.

Table 2. Functional groups and vibration of the FTIR spectra for Polypropylene, Lignin, and Polypropylene/lignin blends in the range of 4000 cm^{-1} down to 500 cm^{-1}

wavenumbers cm^{-1}	Functional groups	Vibration
3500–3400	Alcoholic and phenolic units	Stretching
2936–2846	Methyl and methylene units	Stretching
1710–1737	Ester units	Stretching
1597–1510 - 1440	Aromatic skeletal	Stretching
1330	Syringl rings	Stretching
1114–1210 - 1328	Syringl units and guaicyl units	Stretching
1036	Guaicyl units	Bending
860–750	Aromatic units	Stretching

summarizes the TGA experimental data, including temperature (Tonset), maximum degradation temperature (Tmax) of weight loss, and carbonized residual yield, respectively. It was clearly observed all tested samples degraded in one single sharp step from $30\text{ }^{\circ}\text{C}$ to $700\text{ }^{\circ}\text{C}$. In the TGA curve of Lignin, the initial weight loss around 4.61% occurred at $30\text{ }^{\circ}\text{C}$ and continued until temperature $120\text{ }^{\circ}\text{C}$. The reason for this weight loss could be mainly explained to the gradual evaporation of moisture [57]. It was observed that Lignin started to degrade at temperature $245.46\text{ }^{\circ}\text{C}$, and this degradation continued until temperature $473.22\text{ }^{\circ}\text{C}$ with a weight loss of 54.57% and produced remarkable char residue [58]. Lignin has high thermal stability, which attributed to the presence of a complex phenyl propanoid unit [59]. In the TGA curve for pure PP showed one step degradation with a weight loss of 96.37% , associated with degraded between temperatures around $430.70\text{ }^{\circ}\text{C}$ and $489.29\text{ }^{\circ}\text{C}$. It

Table 3. Results of TGA and DTG of Polypropylene/lignin blends monolith.

Samples	T onset, $^{\circ}\text{C}$	Tmax, $^{\circ}\text{C}$	Weight Loss, %	Residue at $600\text{ }^{\circ}\text{C}$, %
Lignin	245.46	473.23	34.04	54.56
PP	430.70	489.29	96.37	1.46
PP 10L	428.73	492.71	89.17	6.85
PP 20L	429.81	497.62	79.64	14.79

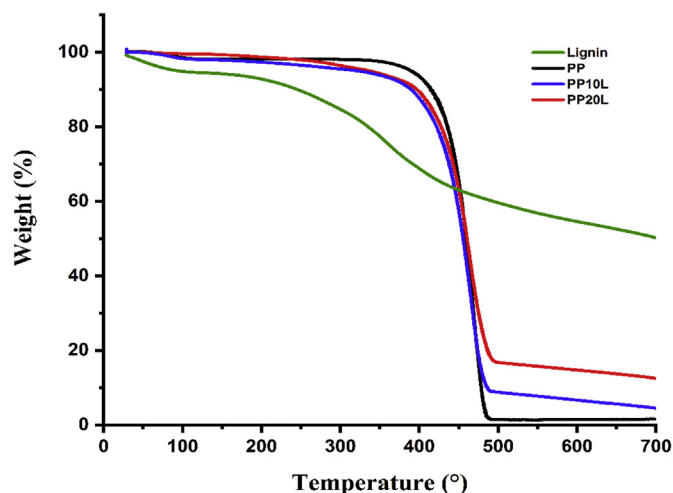


Figure 4. TG curves of Polypropylene/lignin blends monoliths.

was worth noting that the addition of Lignin content in range 10–20 wt% resulted in a slight fall in degradation onset temperature around $428.73\text{ }^{\circ}\text{C}$ and $429.81\text{ }^{\circ}\text{C}$ for PP10L and PP20L. The presence of Lignin in composites acted as a role of lowering degradation onset temperature due to the oxygen-containing groups that accelerate the thermal degradation in the N_2 atmosphere. At the same time, It was noticed that the maximum degradation temperature of the composites increased with the lignin combination around $492.71\text{ }^{\circ}\text{C}$ and $497.62\text{ }^{\circ}\text{C}$ for PP10L and PP20L, with

Table 4. Oil sorption of sorbents.

Sorbent	Oil sorption	Reference
PP sponge	100 % pump oil	[66]
	110 % soybean oil	
Lignin	Up to 70 % carotino oil	[67]
PP	480 %–388 %	Current study
PP10L	983 %–788 % soybean oil – Engine oil	
PP20L	788 %–645 %	

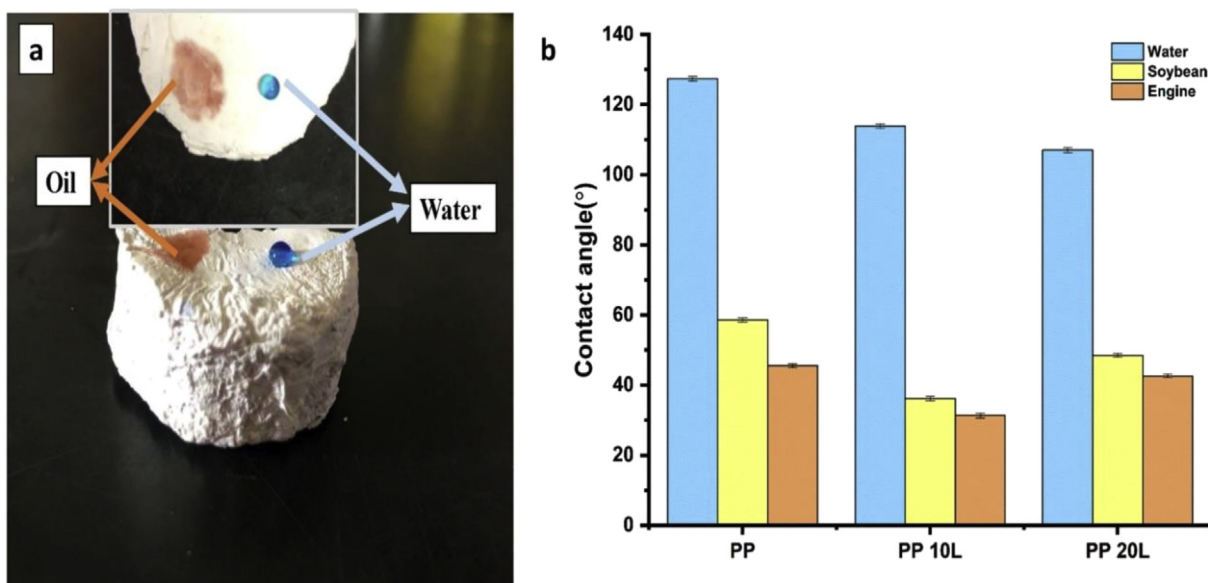


Figure 5. a. Surface wettability of PP Lignin blend sorbent. Engine oil (colored in red) and water (colored in blue with methylene blue) were spotted on the surface of sorbent b. The contact angle of PP, PP10L, and PP20L with liquids (water, soybean oil, engine oil).

total mass loss equal to 89.17% and 79.64%, respectively. This was related to containing a phenyl aromatic group and a hydroxyl group in structures. In the presence of these groups, there will be strong interaction of intermolecular hydrogen to limit the thermal mobility of the polymer molecule. On the other hand, the amounts of carbonized residue

at 600 °C were found to be 6.85% and 14.79% for PP10L, PP20L, respectively, which was higher than PP (1.46%). This property was related to the existence of a high ratio of aromatic subunits in the Lignin structure [58].

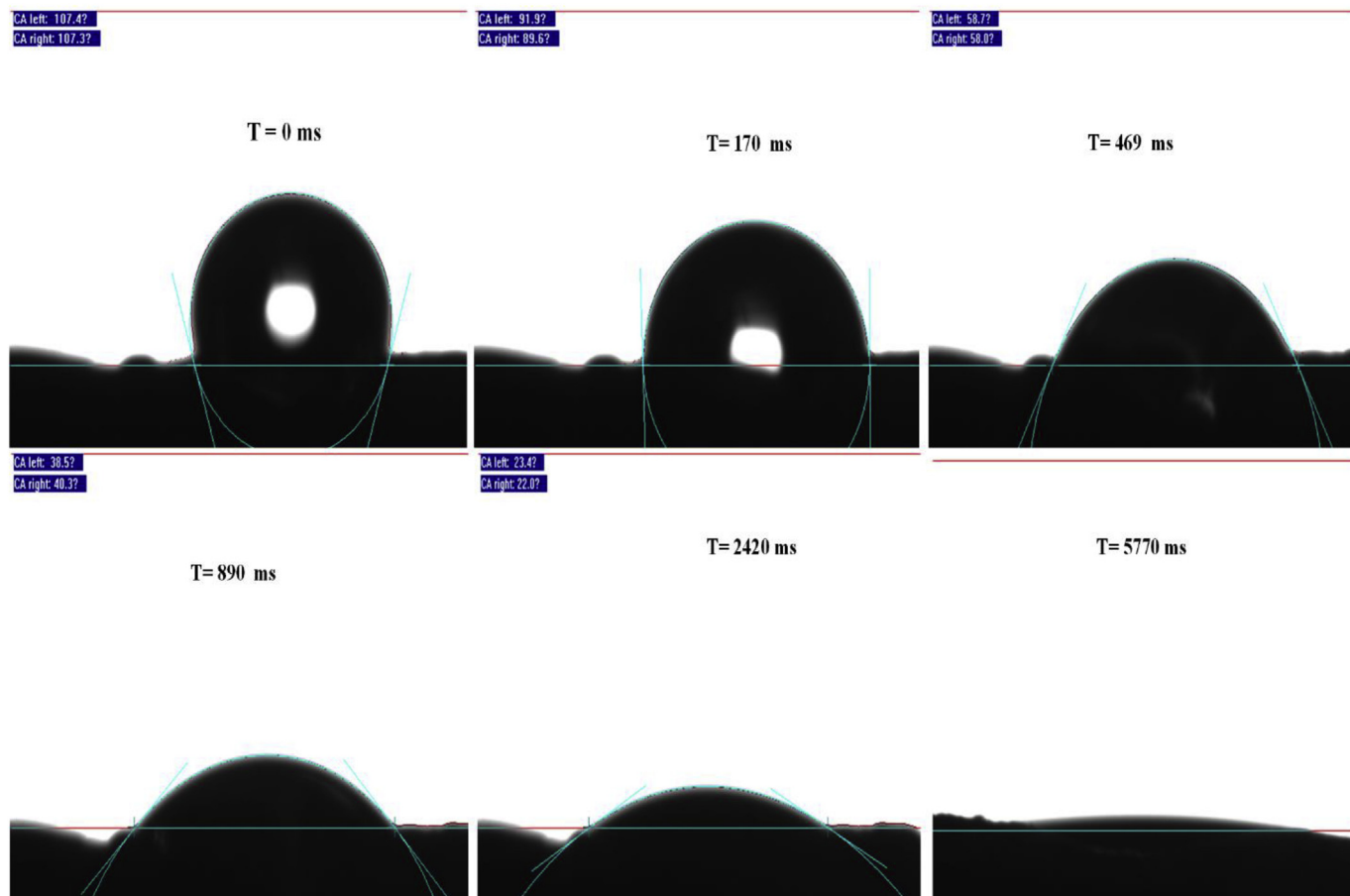


Figure 6. The dynamic spreading process of engine oil on PP10L.

3.4. Hydrophobicity and oleophilicity

Hydrophobicity and oleophilicity of tested samples were carried out through measuring contact angles of the sorbents and liquid (water-oil). It was used the sessile drop method for water and captive bubble method for oil, as seen in Figure 5. The contact angle measurements were registered to analyze the behavior of different lignin loading on PP. It was observed the water contact angle value of PP was 127.40° while the water contact angle values of PP10L and PP20L was 113.89° and 107° . The addition of 10,20 wt% Lignin in the polymer composites demonstrated to reduced the water contact angle. This result may be explained to the presence of polar hydroxyl and carboxylic groups on the lignin surface where the water droplets relatively quickly adsorbed and disappeared [60, 61]. However, the obtained results of angles greater than 90° indicate a hydrophobic feature [62].

In contrast, the static contact angles between oils and sorbent surfaces, as depicted in Figure 5(a), showed oil droplets immediately spreads on the surface with oil contact angle 0° . With using captive bubble methods, all tested samples had low contact angles with oils between 30° and 60° , as shown in Figure 5(b). The consequences for obtained results revealed highly oleophilic in nature for these samples. Generally speaking, engine oil had lower contact angles on tested samples compared to soybean oil. This is attributed to the lower surface tension of engine oil compared to soybean oil, as listed in Table 1.

3.5. The dynamics of spreading of oils

The experimental demand for investigating the spread of oil droplets on the surface of samples as a function between the dynamic contact angles and time was investigated, it was observed that the oil droplets disappeared quickly when applied to tested samples. Figure 6 showed the instantaneously spreading of engine oil on PP10L. The microporous structure, as well as high wetting properties, characterized the spreading behaviors of oil droplets all tested monolith samples. It was noticed that the engine oil and soybean oil droplets absorbed rapidly when it touched the surface on the monolith of PP10L within a few seconds, as shown in Figure 7. The disappearing process of oil droplets deposited on the surface of samples was attributed to penetration and spreading. This phenomenon occurs in parallel, and one of these two was faster than others. The disappearance of oil was accompanied by a wetting surface of samples, which was driven by capillary action. However, it was expected the disappearing times for tested oils were varied because of different viscosity of oils as listed in Table 1. The oil viscosity showed the main role in the rapidity of the spread of the oil drops. As the oil viscosity increased, the contact angles curve shifted to a higher time. All the curves did not plateau at the same value, and the contact angles with engine oil were the highest value. Soybean oil showed rapid wetting behavior, while the process of the spreading of engine oil was slower. The viscosity values for these oils were significantly different soybean oil (65.30 Pa s) had the lowest viscosity compared with engine oil's viscosity (234.50 Pa s) [63].

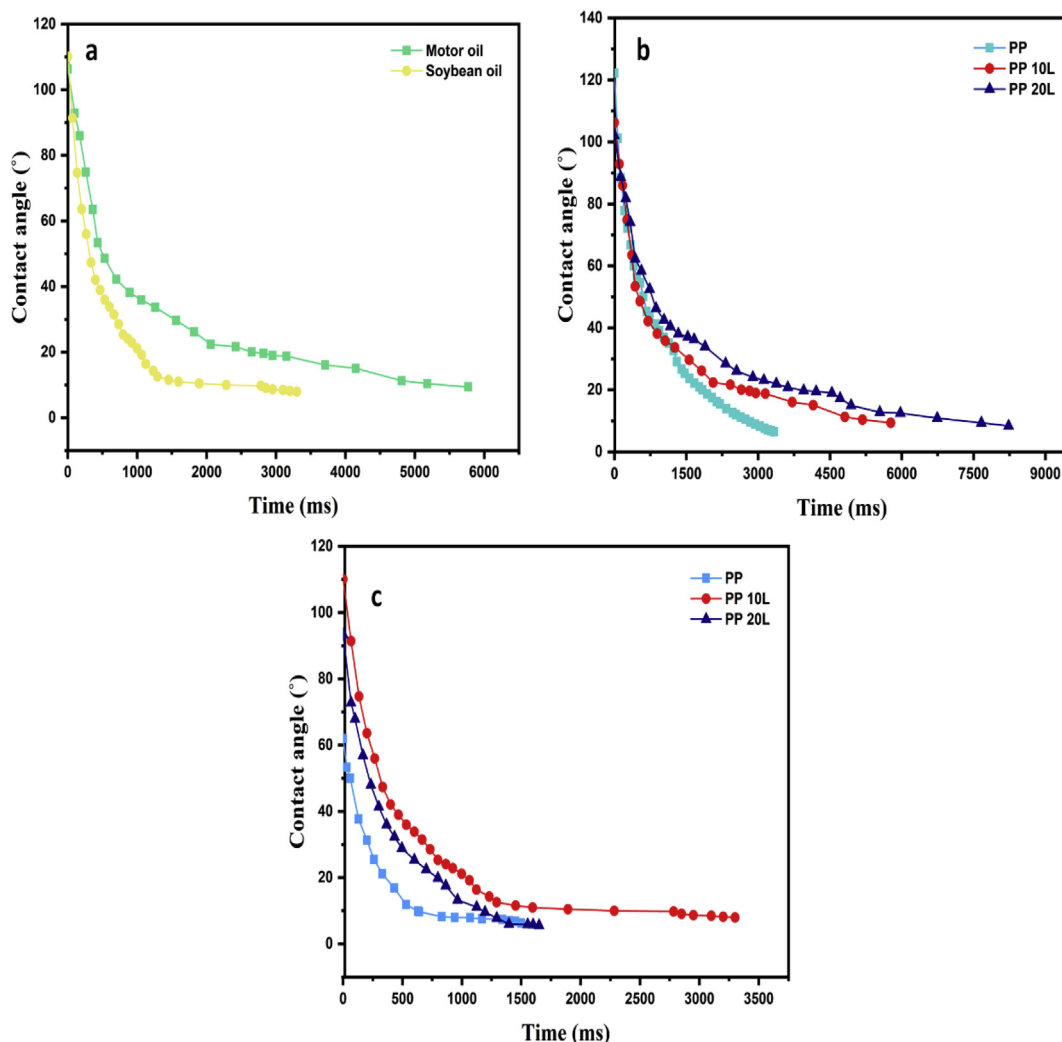


Figure 7. The dynamic oil contact angle as a function of contact time a. Oils spreading on PP10L b. Engine oil spreading, c. Soybean oil spreading.

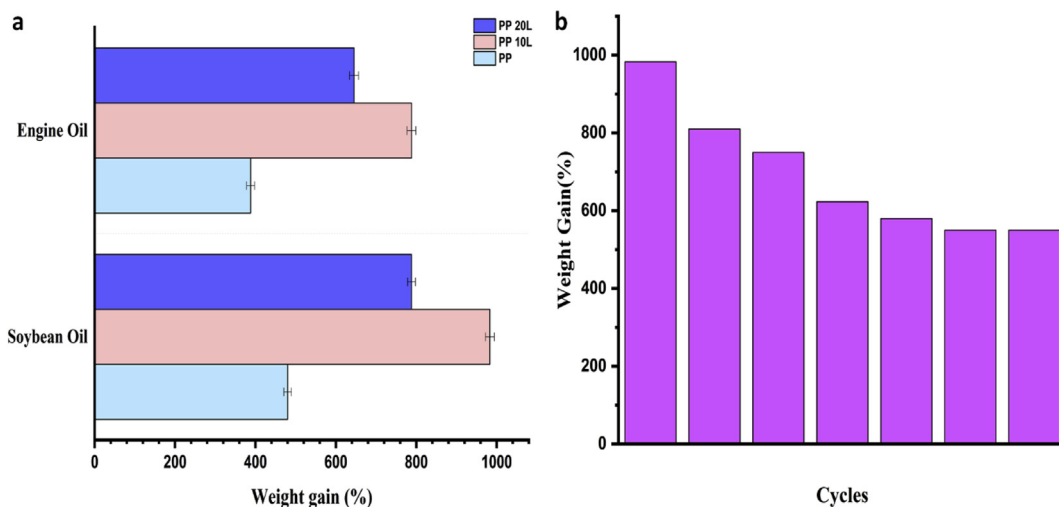


Figure 8. a. Absorption capacities of Polypropylene and Polypropylene/lignin blend monoliths towards oils b. Recyclability of PP 10L monolith toward Soybean oil.

Consequently, Soybean oil exhibited accelerated wetting speed owing to its lower viscosity as compared to engine oil. Figure 7(a) presented the spreading rates for oils that can be transposed on to a master curve when the oil drop dynamic contact angle was plotted versus the contact time on the PP10L surface. It observed that the spreading of two different oils mainly displays similar behavior. After the impact on the surface, the drop started from an initial position with a contact angle of 110.10° and 106.45° for soybean and engine oil, respectively. Then, the oil drops began dropping quickly, and the contact angle reduced within some milliseconds. It was interesting to observe that oil rapidly spread out from 0 ms to 1000 ms after that smooth shift from 1000 ms to 2000 ms with dynamic contact angles of both oils decreased to reach 20° , where the oils continued to spread at vary short rates until the drop reached the equilibrium position.

Comparative evaluation of soybean oil and engine oil spreading rates between PP, PP10L, and PP20L were illustrated in Figure 7(b-c). There were no significant differences observed in oil spreading. It was apparent that the spreading oils on the PP surface were generally faster than on PP10L and PP20L surfaces. This may be explained to have lower static contact angles values with oil, which can result from lower surface energy barrier between the samples surfaces and oils, therefore promote their spreading and sorption into the sample's pores [64].

3.6. Oil absorption

As illustrated in Figure 8(a) Soybean oil absorption capacity of PP20L, PP10L, and PP was 788%, 983%, and 480%, respectively. While engine oil absorption indicated a similar behavior, which was 645%, 788%, and 388%, respectively, it was observed that in the same sorbents had a higher sorption capacity toward soybean oil than engine oil. This could be attributed to the fact that soybean oil is heavier than engine oil within the same unit volume. On the other hand, soybean oil was more ready to drip out from the sorbents than engine oil. The draining took place when the capillary pressure was insufficient to hold the weight of the oils [65].

Furthermore, sorbents PP20L and PP10L possessed higher sorption capacity compared with PP itself. Addition lignin content showed a positive impact on oil sorption and reduced environmental problems through particle replacing of polymer with the sustainable resource according to 3-D interconnected microporous structure, hydrophobicity, and oleophilicity. During the oil absorption test, sorbents after saturated absorption continued to be floating on the surface of the water phase and could be easily removed. The adsorbed oil collected by simply squeezing the monoliths manually. After repeating the process several times, as seen in Figure 8(b). Further, the monolith could be used easily by

washing with volatile liquid and dried at room temperature. The oil sorption of tested sorbents reported in this work was further compared with previous works of other sorbents reported by other researchers, as shown in Table 4.

4. Conclusion

Lignin polypropylene blends were successfully fabricated by using a thermally-induced phase separation (TIPS) technique in the range of 10–20 % by weight. Based on the results, Thermogravimetric measurement presented that the presence of Lignin decreases onset temperature as the concentration of Lignin is increased. At the same time, maximum degradation temperature, and carbonized residue increased, which indicating improved thermal stability, while contact angle measurement showed a decrease in the hydrophobicity. SEM images of monoliths point to three-dimensional interconnected porous structures with an agglomeration of Lignin parties. The obtained PP/Lignin blend monoliths showed a better absorption ability toward oils (Soybean-Engine), which follow a similar trend in both types of oils. Therefore, PP/Lignin blends monoliths used as sorbents, which was based on inexpensive materials and simple methods to fabricate, is expected to be a great promising eco-friendly sorbent material for oil clean up applications in water bodies.

Declarations

Author contribution statement

Abeer Alassod: Conceived and designed the experiments; Performed the experiments; Analyzed and interpreted the data; Contributed reagents, materials, analysis tools or data; Wrote the paper.

Magdi Gibril, Syed Rashedul Islam, Wanzhen Huang: Contributed reagents, materials, analysis tools or data.

Guangbiao Xu: Performed the experiments; Contributed reagents, materials, analysis tools or data.

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Competing interest statement

The authors declare no conflict of interest.

Additional information

No additional information is available for this paper.

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