



Effect of grafted polyurethane on oil spill remediation in the aquatic environment

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ABSTRACT

The aquatic environment may be seriously harmed when cargo gases, diesel, and their compounds leak or are accidentally spilled onto the water's surface. Oil exploration also leads to water contamination. The remaining oil coats the water's surface, resulting in the formation of a thin emulsion of water and oil. Three novel compounds with long chains of linear alkyl groups were grafted to flexible polyurethane (R-FPU) to clean up oil spills. The sorption testing was thoroughly examined and contrasted with the aid of crude oil, diesel fuel, and water-oil systems. It was found that as compared to ungrafted FPU, the chemical sorption of crude oil and diesel fuel was boosted by modified FPU cubes, while water sorption was reduced by roughly 57%. The sorption competence of the modified FPU was highly correlated with the length of the alkyl chain, with the longer alkyl chain significantly increasing sorption capacity. The results demonstrate that oil may be successfully cleaned using modified FPU cubes.

1. Introduction

Today, petroleum products and their derivatives have become major sources of environmental pollution [1]. The crude oil may be discharged into the environment through its processing, from extraction to storage, polluting the ecosystem [2]. The oil spill had to be removed early before it could degrade the ecosystem a possibility before the oil slicks could become an emulsion. In the case of emulsification, oil stains merge with water, and the mixture can turn into heavy sludge. Emulsified crude oil is difficult to clean by normal methods; moreover, it contains many chemicals that are very toxic and harmful to the environment as they can affect living organisms [3]. A diversity of methods

have been utilized to clean up oil from water surfaces: mechanical techniques (oil skimmers and oil containment booms), in situ combustion, and use of dispersants, biodegradation, and the use of absorbent materials [4]. Absorbent materials have gained importance because they can be easily applied with superior efficiency and effectiveness [5]. Absorbent materials can be classified into the three main categories below: inorganic absorbents (zeolites, silica aerogels, graphite, active carbon, perlite, vermiculites, and absorbent clay). Natural organic absorbents include straw, corn corb, wood fiber, cotton fiber, cellulosic kapok fiber, and kenaf. Synthetic organic absorbents include polypropylene, polyurethane foam, polyesters, polycarbonates, styrene-alkyl acrylate polymers, hydrophobic aerogels, cellulosic fibers, polyelectrolyte gels, cryogels based on rubber, poly (orthocarbonate) [6]. Synthetic organic sorbents like polymeric

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materials (polyurethane, polypropylene, polyethylene, and cross-linked polymers) are the most commonly utilized commercial sorbents in oil spill cleanup due to their oleophilic and hydrophobic properties [3, 5]. However, in general, the oil sorption capacity of these synthetic organic sorbents is not very high. It is known that open-cell, low-density, and high-porosity polyurethane foams (PU) are thought of as good oil sorbents. PU has many ether, carbamate, and amide groups, which absorb oil and water. The research focused on the surface modification of PU foams to make them more oleophilic and hydrophobic for oil spill cleanup. In the in situ chemical reaction method, the commercial sponge 3D structure can serve as a model, and the functional groups on the sponge surface point out that it can act as a reactant to interact with modification materials to form a hydrophobic sponge surface [6, 7]. The polymerization method is also a common way to produce a hydrophobic sponge. In this interaction, the polymer is cross-linked to form polymer chains, which can serve as crosslink agents to connect hydrophobic materials with sponge surfaces to manufacture hydrophobic sponge surfaces. Wang et al. created hydrophobic and oleophilic carbon nanotubes (CNTs) supported by a PU sponge using dopamine oxidative self-polymerization, as it is the more commonly used cross-linked agent [8,9]. Hydrophobic polymers can be used as amendment materials to convert hydrophilic to hydrophobic sponge surfaces. Wang et al. improved the hydrophobicity of polyurethane (PU) sponge by grafting the copolymerization method onto a layer of polymer molecular brush [10,11]. The grafting copolymerization method is an attractive way to impart functional groups to polymers. Graft polymer is also known as graft copolymer since it includes at least two types of monomer units, such as the grafted side chains structurally distinct from the main chain. The monomer to be grafted may be of one type or more than one type; thus, the graft chains in the grafted copolymer may be homopolymers or

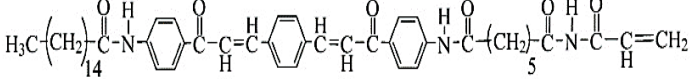
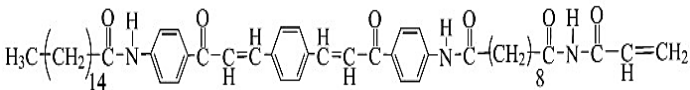
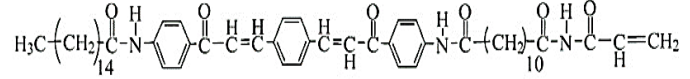
copolymers [12,13]. Polymer chemists use the graft copolymerization method of hydrophobic vinyl monomers to significantly modify the chemical and physical properties of synthetic or natural starting materials. Graft copolymer materials are prepared by generating free radicals on the polymer backbone and then allowing these radicals to serve as macro-initiators for the vinyl monomers [14]. While previous research on oil-sorbent substances focused primarily on the sorption capability of oil, some studies have shown variations in sorption capacities in water, oil phase, and oil-water systems [15]. Hydrophobic and oleophilic properties play a fundamental role in the applications of sponge adsorbents to the separation of water from oil. Recently, hydrophobic materials have been widely used to modify the surface of hydrophilic sponges, and the modified sponges have significantly improved adsorption performance. To improve the oleophilic/hydrophobic properties of commercial flexible PU sponge, grafted copolymerization with three new oleophilic compounds ending in a vinyl group [16] was used, with divinyl benzene (DVB) as the cross-linker, benzoyl peroxide (BPO) as the initiator, and toluene as the solvent. The crude oil and commercial oil (diesel fuel) samples were chosen for the tests, and the sorption capacities of the ungrafted FPU cubes and modified FPU cubes in pure water, pure oil (the crude oil and diesel fuel), and water–oil systems were scrutinized and compared in detail.

2. Experiment

2.1. Materials

A commercial flexible PU sponge was purchased from Arix S.p.A. Co., Italy, and cut to 1cm×1cm×1cm before use. Divinylbenzene (DVB), benzoyl peroxide (BPO), toluene, and ethanol were purchased from Sigma-Aldrich Company. Three hydrophobic/oleophilic compounds were pre-prepared and fully characterized, as reported in our previous work [16] are exhibited in Table 1.

Table 1. Names and structures of the prepared compounds

Symbol	Names of compounds	structures of compounds
D1	N ¹ -acryloyl-N ⁷ -(4-(3-(4-(3-oxo-3-(4-palmitamidophenyl) prop-1-en-1-yl) phenyl)acryloyl)phenyl) heptane diamide	
D2	N ¹ -acryloyl-N ¹⁰ -(4-(3-(4-(3-oxo-3-(4-palmitamido phenyl) prop-1-en-1-yl) phenyl)acryloyl)phenyl) decane diamide	
D3	N ¹ -acryloyl-N ¹² -(4-(3-(4-(3-oxo-3-(4-palmitamido phenyl) prop-1-en-1-yl) phenyl)acryloyl)phenyl) dodecane diamide	

2.2. Grafting copolymerization

The interaction was performed in a 100 ml three-necked round bottom flask equipped with a thermometer, nitrogen gas atmosphere inlet, and reflux condenser. 0.01 g of initiator Benzoyl peroxide (BPO) dissolved in 50 ml toluene solvent was added into the flask. 0.2 g of ungrafted FPU cubes were immersed into the stirred reactive solution for 20 min at 80 °C. Subsequently, a mixture containing 0.3 g of each new compound, pre-prepared (D1, D2, and D3), and 0.003 g of cross-linker divinyl benzene (DVB) was added to the flask at once. After 6 hours of interaction under stirring (500 rpm) and nitrogen gas atmosphere at 80 °C, the cubes were washed with ethanol and deionized water several times, then dried at 60 °C under vacuum for 48 hours [17, 18]. The sorbents were designated FPU-g-Dn when (n=1, 2, and 3). The grafting reaction is shown in Scheme 1.

2.3. Sorption capacity test

The sorbent oil sorption capacity measurement method was based on AstM F726-99 [19]: standard Test Method for Sorbent Performance of

Adsorbents. The procedures applied for the water sorption tests, crude oil sorption tests, diesel fuel sorption tests, and oil-water system sorption tests are shown in Figures 1 to 3.

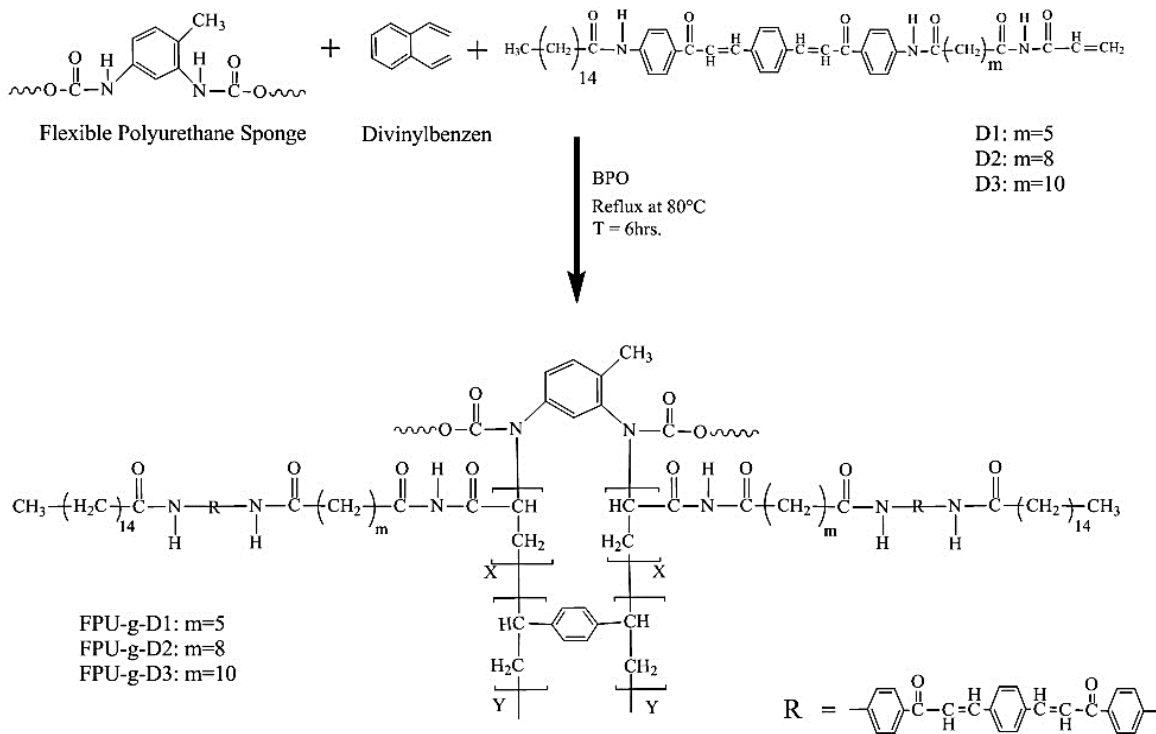
2.3.1. Water sorption tests

For water sorption tests, the sorbent (FPU cubes) was initially weighed and then placed into a 100 ml Erlenmeyer flask containing 50 ml of deionized water. The closed Erlenmeyer flask was then put in a shaker (150 rpm) and shaken for 15 min. The contents of the flask were allowed to settle for 2 min. The sorbent was removed and allowed to drain for 30 s, then instantly relocated to a pre-weighed Erlenmeyer flask and weighed (Fig.1). The water sorption was calculated using Equation 1.

$$\text{Water sorption} \left(\frac{g}{g} \right) = \frac{(S_{wt} - S_0)}{S_0}$$

(Eq.1)

Where S_0 is the initial dry weight of the sorbent and S_{wt} is the wet weight of the sorbent (after water sorption).



Scheme 1. Reaction route for grafting polymerization of (D1-D3) compounds onto the FPU surface

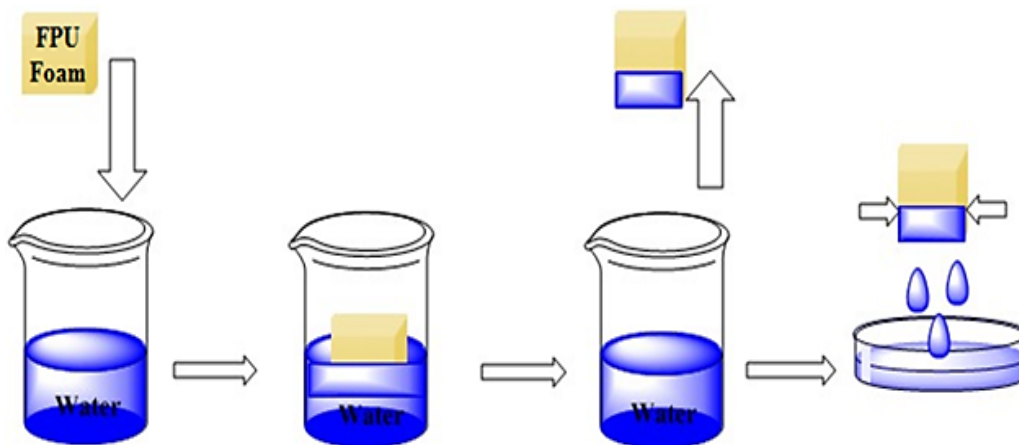


Fig. 1. Scheme of the procedure applied for the water sorption tests

2.3.2. Oil sorption tests

For oil sorption tests, crude or diesel fuel (50ml) was decanted into a 100 mL beaker. The sorbent (FPU cubes) were weighed and the value recorded, then immersed into the crude oil or diesel fuel. Generally, after 20 min±20 s of inundation, the sorbent (FPU) cubes were removed and allowed to drain for 30 s±3 s. The saturated sorbent was then immediately transferred to a pre-weighed weighing bottle and weighed (Fig.2). The oil sorption of sorbent (FPU) cubes on a weight basis was calculated as Equation 2.

$$\text{Oil sorption (g/g)} = \frac{(S_t - S_o)}{S_o}$$

(Eq.2)

Where S_o is the initial dry weight of a sorbent and S_t is the weight of the sorbent with oil absorbed. Solvent sorption capacity measurements were carried out similarly.

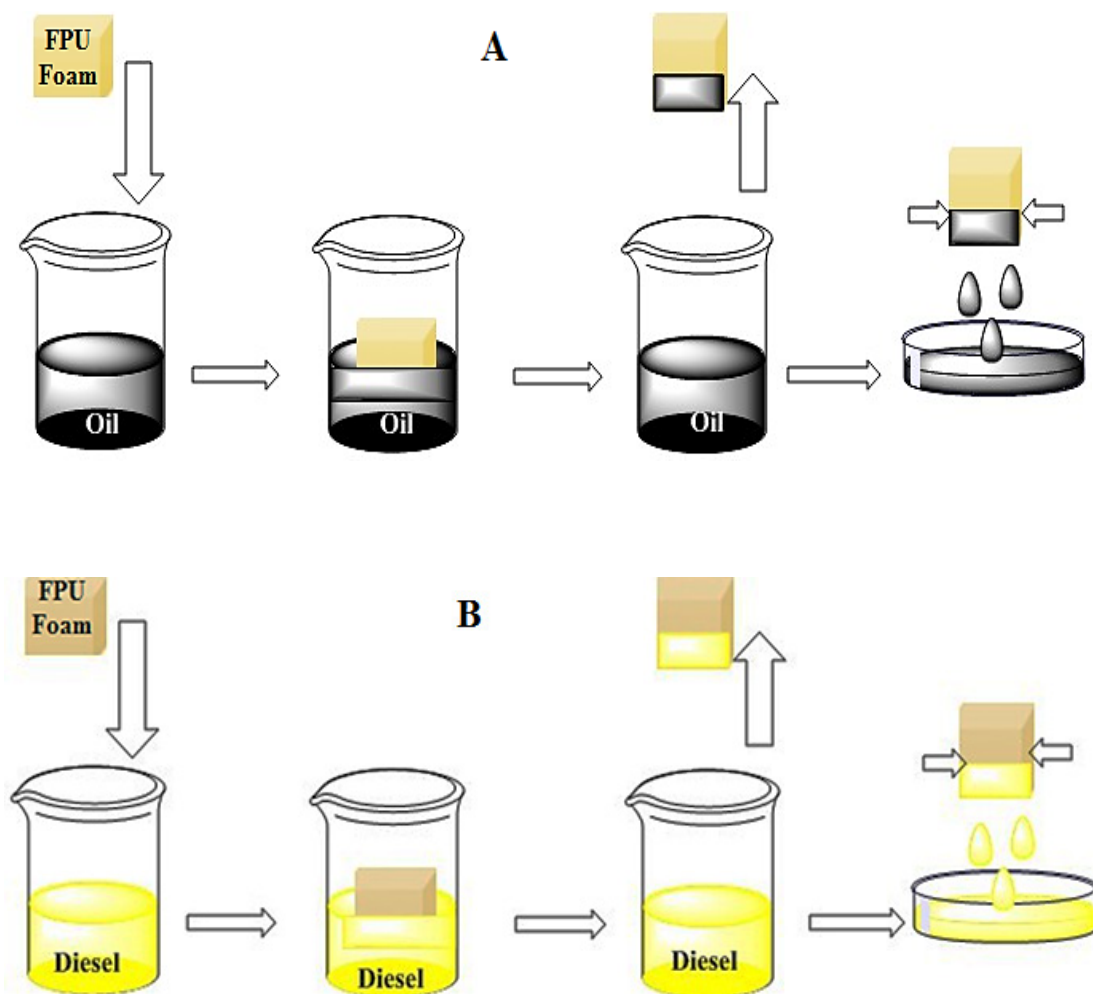


Fig. 2. Schematic of the procedure applied for the oil sorption tests using crude oil (A) and diesel fuel (B)

2.3.3. Water–oil system sorption tests

In water–oil system sorption tests, 4.0 g of (crude oil or diesel fuel) was decanted into a 100 mL Erlenmeyer flask which was filled with 50 ml of deionized water, and the thickness of the oil stratum was 2–3 mm. The sorbent (FPU) cubes were initially weighed and put into the Erlenmeyer flask. The closed Erlenmeyer flask was placed in a shaker (150 rpm) and shaken for 30 min. The contents of the closed Erlenmeyer flask were allowed to settle for 2 min. Then sorbent (FPU) cubes were removed to an Erlenmeyer flask and then extracted by petroleum ether (boiling range 30–60 °C) several times; subsequently, the sorbent was removed and squeezed out the absorbed petroleum ether. Anhydrous sodium sulfate was added to the petroleum

ether solution until there was no agglomeration, and then the Erlenmeyer flask was sealed for 30 min. to dehydrate. Next, after filtration, the filtrate was collected by a dry beaker with a constant weight. The beaker was placed in a 65°C water bath to vaporize petroleum ether. Then the beaker was put in a 65°C drying oven until it reached constant weight (Fig.3). Finally, the oil sorption in water–oil system was calculated using Equation 3.

$$\text{Oil - water sorption (g/g)} = \frac{(M_t - M_o)}{S_o}$$

(Eq.3)

Where M_o is the constant weight beaker, M_t is the weight of the constant weight beaker with absorbed oil, and S_o is the initial dry weight of sorbent.

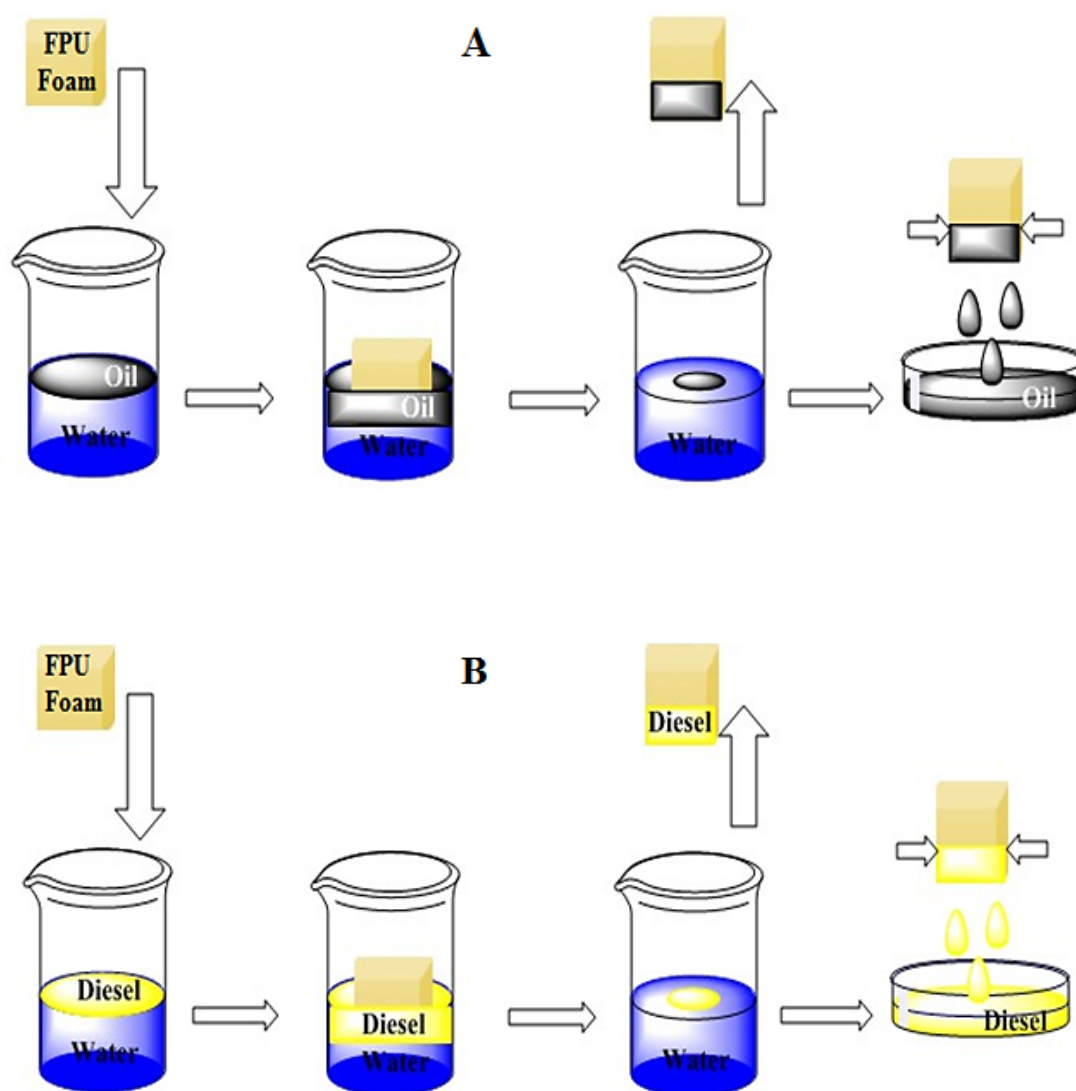


Fig. 3. Schematic of the procedure applied for the water-oil sorption tests using water-crude oil (A) and water-diesel fuel (B)

3. Results and Discussion

The testing used crude oil and diesel fuel as oil samples, and the sorption properties of the ungrafted and grafted PU cubes in water, pure oil phase, and water-oil systems were thoroughly investigated and compared.

3.1. Sorption Capacity Test

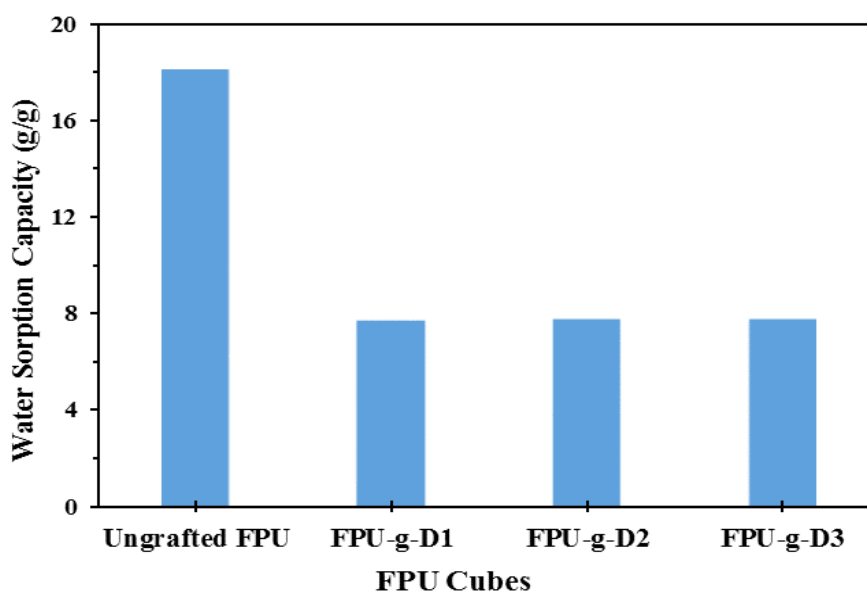
3.1.1. Water sorption capacity

In water-oil system sorption tests. The water sorption was calculated using Equation 1. The results obtained for the water sorption capacity of

ungrafted FPU and grafted FPU cubes are shown in Table 2 and Figure 4. The water sorption capacity is higher for ungrafted FPU cubes than the water sorption capacity for grafted FPU cubes, which decreased by more than 57% after the grafting process. This may be attributed to the long alkyl chains of the grafted organic compounds [D1-D3] that exhibit oleophilic and hydrophobic properties [20], and it may also be due to the pore geometry and the heterogeneous distribution of pores in flexible polyurethane as reported in our previous work [21].

Table 2. The water sorption capacity of the ungrafted FPU cubes and grafted FPU

Cubes	S_o (g)	S_{wr} (g)	Water Sorption Capacity (g g ⁻¹)
Ungrafted FPU	0.1033	1.9674	18.0454
FPU-g-D1	0.0998	0.86937	7.7111
FPU-g-D2	0.105	0.91645	7.7280
FPU-g-D3	0.1013	0.8868	7.7541

**Fig. 4.** The water sorption capacity of the ungrafted and grafted FPU cubes

3.1.2. Oil sorption capacity

The oil sorption capacities of crude oil and diesel fuel were calculated using Equation 2, and the

results obtained are shown in Table 3 and Figure 5 for crude oil and Table 4 and Figure 6 for diesel fuel.

Table 3. The crude oil sorption capacity of the ungrafted and grafted FPU

FPU Cubes	S_o (g)	$S_{c.o}$ (g)	Crude Oil Sorption Capacity (g/g)
Ungrafted FPU	0.0916	2.7536	29.0611
FPU-g-D1	0.0966	3.5263	35.5041
FPU-g-D2	0.0853	3.6231	41.4747
FPU-g-D3	0.113	5.1227	44.3336

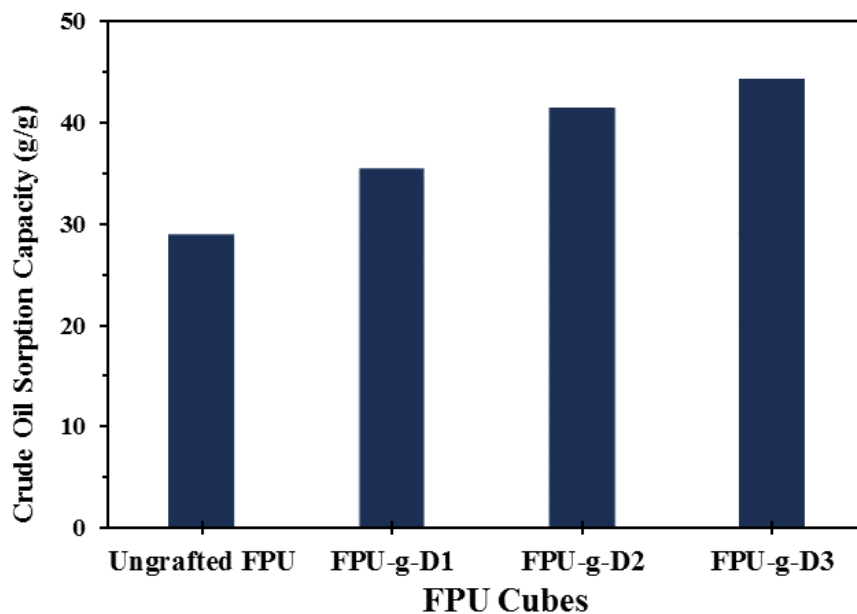


Fig. 5. The crude oil sorption capacity of the ungrafted and grafted FPU cube

Table 4. The diesel fuel sorption capacity of the ungrafted and grafted FPU

FPU Cubes	S_o (g)	S_{die} (g)	Diesel fuel Sorption Capacity (g/g)
Ungrafted FPU	0.1082	3.0587	27.2689
FPU-g-D1	0.0985	3.57545	35.2989
FPU-g-D2	0.1058	4.2285	38.9669
FPU-g-D3	0.1028	4.3053	40.8803

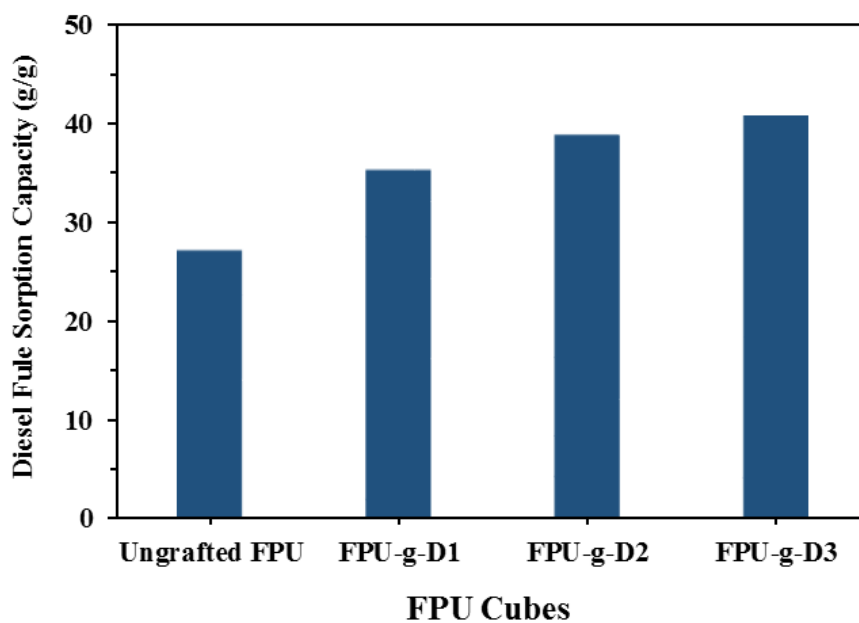


Fig. 6. The sorption capacity of diesel fuel of the ungrafted and grafted FPU cubes

As expected, grafted FPU cubes performed better in absorbing crude oil and diesel fuel, as shown in Figures 5 and 6, respectively, than ungrafted FPU cubes. Furthermore, incorporating long-chain alkyl groups into the PU backbones improves the hydrophobic properties of the FPU cubes [21]. The sorption capacity of grafted PU cubes is shown in the following order, from highest to lowest sorption capacity:

$$\text{FPU-g-D3} > \text{FPU-g-D2} > \text{FPU-g-D1}$$

In contrast, the absorption capacity of the grafted PU cubes for crude oil and diesel fuel increased more than that of the ungrafted FPU cubes due to long aliphatic non-polar groups ($-\text{CH}_3$ and $-\text{CH}_2-$). These are hydrophobic groups in nature, which can be explained by the increased oleophilic and

hydrophobic characteristics of grafted FPU cubes [22]. Table 5 and Figure 7 show the obvious differences between the sorption capacities of water.

3.1.3. Water-oil system sorption tests

Finally, the oil sorption in water–crude oil systems and water–diesel fuel systems are calculated using Equations 3; the results are shown in Table 6 and Figure 8. It has been noted that the oil sorption values increased and that water–crude oil had a better capacity for sorption in the water system of diesel fuel. Since there are stronger adhesion forces between the absorbent and the oil surface as the oil viscosity increases, these results can be explained [24, 25].

Table 5. The water, crude oil, and diesel sorption capacities of the ungrafted and grafted FPU cubes

FPU Cubes	Water Sorption Capacity (g/g)	Crude Oil Sorption Capacity (g/g)	Diesel fuel Sorption Capacity (g/g)
Ungrafted FPU	18.0454	29.0611	27.2689
FPU-g-D1	7.7111	35.5041	35.2989
FPU-g-D2	7.7280	41.4747	38.9669
FPU-g-D3	7.7541	44.3336	40.8803

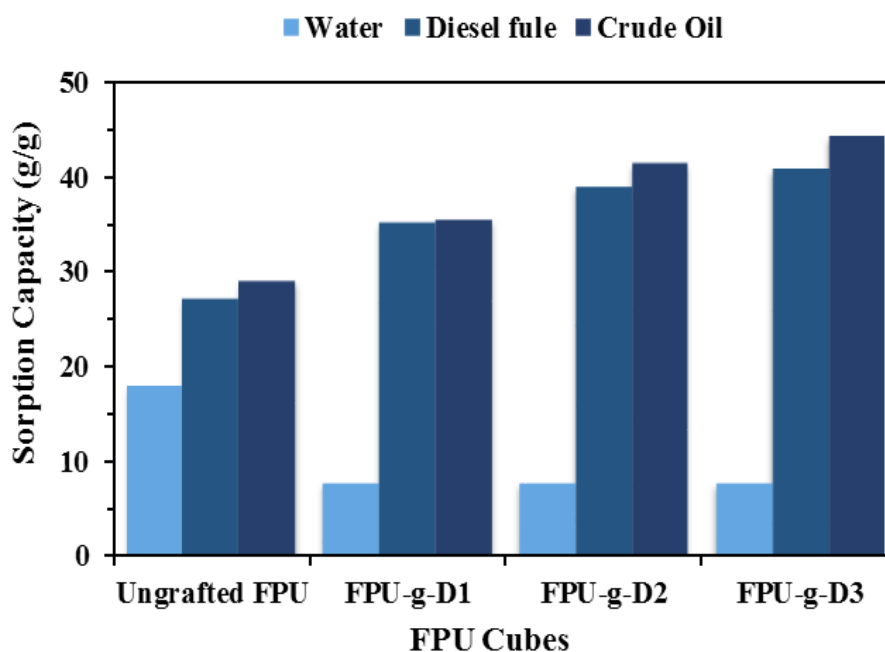
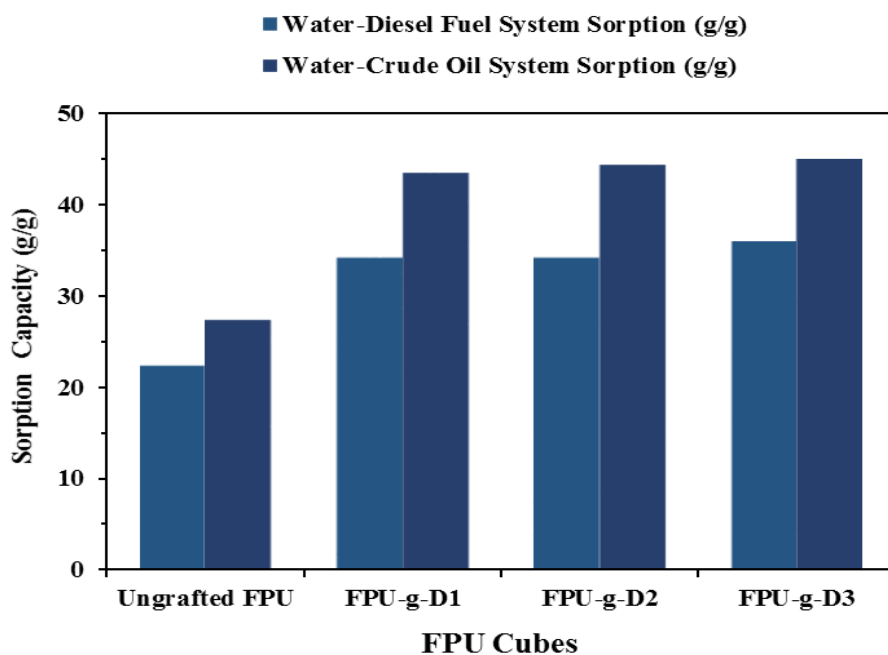


Fig. 7. Water, crude oil, and diesel fuel sorption capacities of the ungrafted and grafted FPU cubes

Table 6. The crude oil sorption capacity of the ungrafted and grafted FPU cubes in water–crude oil system

FPU Cubes	Crude Oil Sorption Capacity (g g ⁻¹)	Diesel fuel Sorption Capacity (g g ⁻¹)
Ungrafted FPU	27.2873	22.2841
FPU-g-D1	43.2986	34.0547
FPU-g-D2	44.2361	34.0837
FPU-g-D3	44.7937	35.8290

**Fig. 8.** The crude oil sorption in water–crude oil system tests and the diesel fuel sorption in water–diesel fuel system sorption tests of the ungrafted and grafted FPU cubes

The modified FPU sponge material shows high selectivity, efficiency, and recyclability for oil/water separation. The modified sponge could be synthesized from a commercial PU sponge with standard chemical modifications to lead to the required super-hydrophobic properties [25,26]. The obvious differences between the sorption of crude oil and diesel fuel capacities can be explained by the modified FPU cubes' higher oleophilic and hydrophobic features [26]. This may be attributed to the increase in the length of the alkyl chain from five carbon atoms in D1 to eight carbon atoms in D2 to ten carbon atoms in compound D3. This may be seen clearly in Table 5 and Figure 4 for the water, crude oil, and diesel fuel sorption

capacities of blank FPU and modified FPU cubes. The same conclusion was made by Santucci and Fiore [27] by considering the application of FPU as contingency equipment for absorbing oil spills. Due to its interesting properties (high porosity and hydrophobicity), FPU is used as the primary raw material in manufacturing industrial absorbents, which can absorb crude oil and related by-products. Hydrophobic properties decrease in cubes (FPU-g-D1 to FPU-g-D3) due to an increase in polarity caused by the resonance that occurs between the amide group, aromatic rings, and the two (-CO-CH=CH-) groups in bis-chalcone. In addition to the increased polar group number (-NH-C=O) in the monomer backbone for (D1-D3) [28]. The other

reason is the decrease in the length of the alkyl chain from the number of carbon atoms (10) in monomers D3 to the number of carbon atoms (8) in monomers D2 to the number of carbon atoms (5) in monomers D1 [29]. Furthermore, molecules with aliphatic (linear) chains have better hydrophobicity than branched or aromatic molecules, owing to the steric effect of the neighboring branches and aromatic rings that reduce the hydrophobic interactions with the water molecule. On the other hand, the hydrophobic surfaces interact less with water because of non-polar functional groups at the surface. In general, hydrophobicity is affected by the length and shape of the functional groups; increasing the length of an alkyl chain makes the surface more hydrophobic [30]. The FPU sponge's absorption capacity for crude oil and diesel fuel depends on the density and viscosity of the crude oil and diesel fuel. The absorption rate of crude oil was higher, as shown in Tables and Figures. The crude oil and diesel fuel were stored in the pores formed by the interconnected skeleton of the sponge, exhibiting a high oil absorption capacity. By combining the special wettability and high porosity, the sponges exhibited high oil-absorption capacity and high selectivity when they were employed as absorptive materials for cleaning crude oil and diesel fuel on the water surface [31].

4. Conclusions

A modified flexible polyurethane was developed from the commercially grafted FPU with aliphatic chains to have better hydrophobicity and less interaction with water because of non-polar functional groups at the surface. In general, hydrophobicity is affected by the length and shape of the functional groups; by increasing the length of an alkyl chain, the surface would be more hydrophobic. The grafted FPU sorption capacity for crude oil and diesel fuel depends on the density and viscosity of the crude oil and diesel fuel. The crude oil and diesel fuel were stored in the pores formed by the interconnected skeleton of the sponge, which exhibited a high oil absorption capacity. By combining the special wettability and high porosity,

the sponges exhibited high oil-absorption capacity and high selectivity when they were employed as absorptive materials for environmental remediation by cleaning crude oil and diesel fuel on the water surface.

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