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Facile Fabrication of High-Performance Superhydrophobic Reusable Oil-Absorbing Sponges

Rabiga Kudaibergenova [1], Yerzhigit Sugurbekov [2], Gulzat Demeuova [2] and Gulnar Sugurbekova [2,*]

[1] Department of Chemistry and Chemical Technology, Faculty of Technology, Taraz Regional University Named after M.Kh. Dulaty, 60 Tole Bi Street, Taraz 080000, Kazakhstan; rabi_07@bk.ru
[2] Department of Chemistry, Faculty of Natural Sciences, Eurasian National University Named after L.N. Gumilyov, Astana 010000, Kazakhstan; gulnar-sugur@yandex.ru (Y.S.); yerzhigit.sugurbekov@nu.edu.kz (G.D.)
* Correspondence: sugurbekova.g.55@gmail.com

Abstract: Wastewater treatment from oil, oil products and organic mixtures is a very relevant topic that can be successfully utilized to solve problems of severe environmental pollution, such as oil spills, industrial oily wastewater discharges and water treatment in the water treatment process. In this work, we have developed new superhydrophobic magnetic polyurethane (PU) sponges, functionalized with reduced graphene oxide (RGO), $MgFe_2O_4$ nanoparticles, and silicone oil AS 100 (SO), as a selective and reusable sorbent for the purification and separation of wastewater from oil and organic solvents. The surface morphology and wettability of the sponge surface were characterized by scanning electron microscopy (SEM) and a contact angle analysis system, respectively. The results showed that the obtained PU sponge PU/RGO/$MgFe_2O_4$/SO had excellent mechanical and water-repellent properties, good reusability (lasted more than 20 cycles), as well as fast (immersion time 20 s) and excellent absorption capacity (16.61–44.86 g/g), and additional good magnetic properties, which made it easy to separate the sponge from the water with a magnet. The presence of RGO in the composition of the nanomaterial improves the separating and cleaning properties of the materials and also leads to an increase in the absorption capacity of oil and various organic solvents. The synthesized PU sponge has great potential for practical applications due to its facile fabrication and excellent oil–water separation properties.

Keywords: polyurethane sponge; reduced graphene oxide; silicone oil; magnetic; superhydrophobic/superoleophilic; oil/water separation

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1. Introduction

In recent years, there has been an increase in the number of accidental oil spills during oil production, transportation, storage and industrial discharges of oils/organic solvents, which pose a significant threat to marine ecosystems [1,2]. In this regard, the separation of oil–water mixtures is crucial and plays a vital role in the industry dealing with severe environmental pollution, such as oil spills, industrial oily wastewater discharges and oil recovery [3]. In the process of reducing the impact of oil pollution, the main challenge for researchers is to develop low-cost and high-performance oil absorption materials. The preferred oil spill treatment method is absorption [4], which has high efficiency, easy operation and a high absorption capacity [5]. It is vital for an oil absorbent to be hydrophobic as it needs to selectively absorb oil while repelling water [6–8].

In recent years, conventional absorption materials such as activated carbon [9,10], carbon nanotubes [11], fibers [12], zeolites [13,14] and silica [15], as well as synthetic films [16], meshes and fabrics [17,18], have been explored for the removal and collection of spilt oil. However, these materials have several disadvantages, such as low absorbency, low selectivity and poor reusability.

To date, much research has been focused on developing nanomaterials that have significant advantages in oil and organic solvent absorption and high porosity, low density and recyclability, such as membranes [19], carbon nanotubes [20], polymer gels [21], and nanoporous polymers [22]. Unfortunately, the complexity of the preparation process, its high cost and its environmental hazards do not allow the use of these materials in practical applications. Therefore, it is necessary to develop absorbent nanomaterials with better characteristics to remove oils/organic solvents efficiently. Superhydrophobic magnetic nanomaterials and their outstanding absorption capabilities, especially regarding oils and organic solvents, have recently been investigated. Zhu et al. [23] coated polysiloxane on polyurethane sponge, which exhibited a high oil/organic solvent absorption capacity; their use is still limited due to some restrictions such as expensive materials and complex processes.

Currently, much attention is given to producing superhydrophobic and superoleophilic PU sponges, as they are inexpensive, porous, elastic three-dimensional materials with a large internal surface area, ideally used for oil and water separation [24].

Among various superhydrophobic materials, adsorbents coated with reduced RGO have unique hydrophobic properties, due to their chemical stability and hydrophobic interactions [25]. The following study aims to use graphene oxide to produce superhydrophobic materials with superior oil–water separation capabilities. Graphene oxide (GO) is a single-atomic layered 2D material made by the powerful oxidation of graphite, which is cheap and abundant [26]. Graphene-based superhydrophobic composite nanomaterials have become widely used due to their excellent characteristic properties, such as self-cleaning abilities, excellent mechanical/superhydrophobic properties, and effective absorption of oil and organic liquids [27,28].

In the following study, a highly absorbent RGO-, $MgFe_2O_4$ NPs- and SO-based PU sponge was fabricated using the dipping method. Researchers [29] in their work obtained a superhydrophobic magnetic PU sponge of a similar type, but as a sorbent, with polydimethylsiloxane (PDMS) as a gluing agent. SO is a liquid compound with excellent water-repellent properties. When phenyl radicals are introduced into the frame of the organosiloxane chain, a separate large group of SOs (or polymethylphenylsiloxanes) with specific properties is formed. SO differs in the structure of the molecules, as well as the ratio of methyl and phenyl radicals in them. The chains of SO molecules can consist of methylphenylsiloxy units (I) or dimethyl, methylphenylsiloxy units (II) or dimethyl and diphenylsiloxy units. The main difference between SO and PDMS is that the former is softer and more elastic, thermally resistant, low cost and easily attainable. The phenyl radical at the silicon atom, combined with the methyl radical, increases the thermal stability of SO by 50–70 °C. Taking into this consideration, the authors investigated and obtained a cheaper, elastic and thermally stable version of a superhydrophobic magnetic PU sponge, which has excellent potential for practical applications, owing its complete absorbance of absorb oil within a few seconds. In addition, the new super hydrophobic magnetic sponge has excellent oil selectivity and high absorbency. This study proposes a simple and inexpensive method for making a hydrophobic and lipophilic sponge that can be used to separate oil and water.

2. Materials and Methods

2.1. Chemicals and Apparatus

Silicone oil AS 100 (viscosity ~100 mPa·s, neat (25 °C)) and hexane (laboratory reagent, $\geq$95%) were purchased from Sigma-Aldrich (St. Louis, MO, USA). Polyurethane sponge (PU, density 22 kg/m^3) was purchased at a local hardware store. The new superhydrophobic magnetic sponge was characterized using various analysis techniques listed below. Scanning electron microscopy (SEM) images were recorded using a Carl Zeiss Crossbeam 540 with a GEMINI II Scanning Electron Microscope (GmbH, Jena, Germany). Water contact angles (CA) of the initial PU and prepared PU/$MgFe_2O_4$/RGO/SO sponges were measured with a Dataphysics Instrument OCA 15EC (GmbH, Filderstadt, Germany) contact angle analysis system.

2.2. Preparation of GO and RGO

GO was synthesized according to the modified Hummers' method [28], as described in [29]. The resulting GO is chemically reduced with hydrazine monohydrate [29].

2.3. Preparation of Magnetic $MgFe_2O$ Nanoparticles

Magnetic $MgFe_2O_4$ NPs were obtained by the sol–gel method [30], which is described in detail in the work [29].

2.4. Preparation of Superhydrophobic Magnetic PU Sponges

First, the polyurethane sponge was cut into pieces (1.5 × 1.5 × 1 cm) and sonicated with acetone and deionized water, respectively, for 30 min, in order to remove surface stains and oil. The sponges were subsequently placed into an oven at 80 °C for 3 h to dry completely [29].

The dried sponge was immersed in 50 mL of a homogeneous solution containing $MgFe_2O_4$ NPs (130 mg), RGO (0 or 40 mg) and SO (40 mg) in hexane under ultrasound for 7 h. Finally, the sponge was repeatedly washed with distilled water to remove unreacted solutions and then dried at 60 °C for 12 h. Mass loading of $MgFe_2O_4$/SO or $MgFe_2O_4$/RGO/SO ranged from 0 to 40 wt%. The resulting superhydrophobic magnetic sponges were named PU/$MgFe_2O_4$/RGO/SO with RGO and PU/$MgFe_2O_4$/SO without RGO.

The presence of $MgFe_2O_4$, SO and RGO nanoparticles in the prepared superhydrophobic magnetic sponge improved superoleophilic, self-cleaning and oil/water separation properties. With increasing mass loading of $MgFe_2O_4$/RGO/SO and $MgFe_2O_4$/SO, the absorption capacity of the superhydrophobic magnetic sponge increased. Therefore, all absorption studies were conducted with a maximum load of 40%.

2.5. Testing Superhydrophobic Properties, Oil/Water Separation Properties and Oil Absorption Capacity

The hydrophobic properties of the PU/$MgFe_2O_4$/RGO/SO (40 wt% loading) sponge were studied. A piece of sponge measuring 1.5 × 1.5 × 1 cm and of known mass was immersed in distilled water for 10 min, applying an external force using tweezers. The mass of PU/$MgFe_2O_4$/RGO/SO (40 wt% loading) sponge was weighed three times before and after immersion to calculate the mass water absorption. All experiments were performed in triplicate.

To study the oil/water separation property, a modified PU/$MgFe_2O_4$/RGO/SO and PU/$MgFe_2O_4$/SO sponge was immersed in a crude oil/water mixture until completely filled with oil. The sponge was then removed from the water, using a magnetic field to check the weight.

The separation efficiency (R(%)) of the superhydrophobic magnetic sponge for various mixtures of oils and organic solvents was calculated by measuring the weight of the oil before and after absorption according to Equation (1):

$$R(\%) = \left[\frac{n2 - n1}{n0}\right] * 100\% \quad (1)$$

where n1 and n2 are the total mass of oil/water mixed before and after separation, and n0 is the mass of oil before absorption. In total, 20 cycles were carried out, all experiments were performed in triplicate and the mean experimental value was reported.

The absorption capacity was studied by immersing the modified sponge in oil or an organic solvent for 20 s. Subsequently, each piece of sponges PU/$MgFe_2O_4$/RGO/SO and PU/$MgFe_2SO_4$/SO (40 wt% loading) was weighed and the oil absorption amount (Q(g/g)) was calculated by the following Formula (2):

$$Q(g/g) = \frac{m2 - m1}{m1} \quad (2)$$

where m1 and m2 refer to the masses of the sponge before and after oil absorption. All experiments on the absorption capacity of the sponge were carried out three times, and the average absorption value was recorded.

The effectiveness of PU/$MgFe_2O_4$/RGO/SO (40 wt% loading) sponge in separating water from oil or organic solvents was investigated using a 125 mL vacuum filtration flask, a rubber stopper and a diaphragm vacuum pump. A known mixture of water/oil or organic solvent was filtered through a PU/$MgFe_2O_4$/RGO/SO sponge. After filtration, the extracted masses of water in the beaker and oil or organic solvents collected in the filter flask were compared with their masses in the original mixtures to evaluate the separation efficiency of PU/$MgFe_2O_4$/RGO/SO. All experiments were carried out in triplicate.

3. Results and Discussion

3.1. SEM Morphology Analysis of New Superhydrophobic Magnetic Sponges PU/$MgFe_2O_4$/SO and PU/$MgFe_2O_4$/RGO/SO

The morphology of the new superhydrophobic magnetic sponge before and after modification was studied using SEM. Figure 1 shows SEM images of a new PU/$MgFe_2O_4$/RGO/SO sponge prepared with 20 and 40 wt% loadings. As seen in Figure 1a, the sponge has a three-dimensional porous structure, which is advantageous for liquid absorption. At the same time, the prepared superhydrophobic magnetic sponge had the same porous structure after modification [31], which means that the modification did not destroy the porous structure of the sponge (Figure 1b,c). Also, compared with the smooth surface of the original sponge, it is evident that the 3D skeleton of the sponge modified with PU/$MgFe_2O_4$/RGO/SO has become uneven, rough, and has a rougher structure. As the content of $MgFe_2O_4$/RGO/SO increases from 0 to 40 wt%, the 3D sponge skeleton becomes more uneven and rough. The unevenness and roughness of the surface are of great importance for the preparation of an excellent hydrophobic and oleophilic surface. These properties show a good distribution of $MgFe_2O_4$/RGO/SO over the entire surface of the PU sponge, resulting in uniform coverage of the surface of the PU sponge with superhydrophobic (RGO) and magnetic NPs ($MgFe_2O_4$). The results showed that the superhydrophobic $MgFe_2O_4$/RGO/SO magnetic NPs are uniformly distributed over the entire surface of the PU sponge.

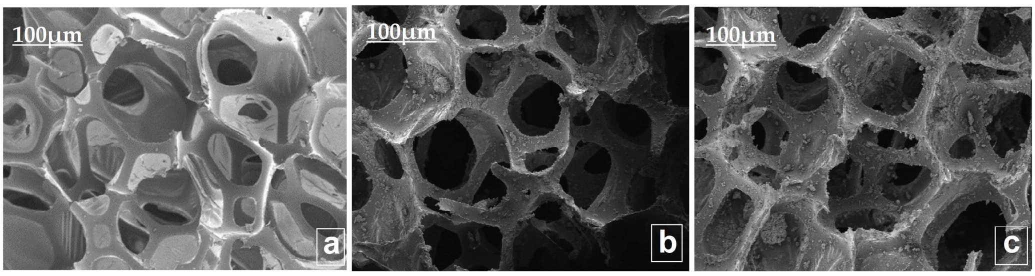

Figure 1. SEM images of (**a**) unmodified PU sponge, (**b**) 20 wt% and (**c**) 40 wt% $MgFe_2O_4$/RGO/SO-modified sponges.

3.2. The Magnetic Properties of the Magnetic $MgFe_2O_4$ NPs, New Superhydrophobic PU/$MgFe_2O_4$/SO and PU/$MgFe_2O_4$/RGO/SO Magnetic Sponges

The magnetic properties of the obtained magnetic $MgFe_2O_4$ nanoparticles and modified PU/$MgFe_2O_4$/SO and PU/$MgFe_2O_4$/RGO/SO (40 wt% loading) sponges were tested using the magnet presented in Figure 2a,b (see Supplementary Videos S1 and S2). As shown in the Videos S1 and S2 in the Supplementary Materials, $MgFe_2O_4$ nanoparticles, as well as modified sponges PU/$MgFe_2O_4$/SO, PU/$MgFe_2O_4$/RGO/SO (40 wt% loading) are ideally attracted to the magnet. Their excellent magnetism indicates that the magnetic $MgFe_2O_4$ nanoparticles were successfully filled into the original PU sponge.

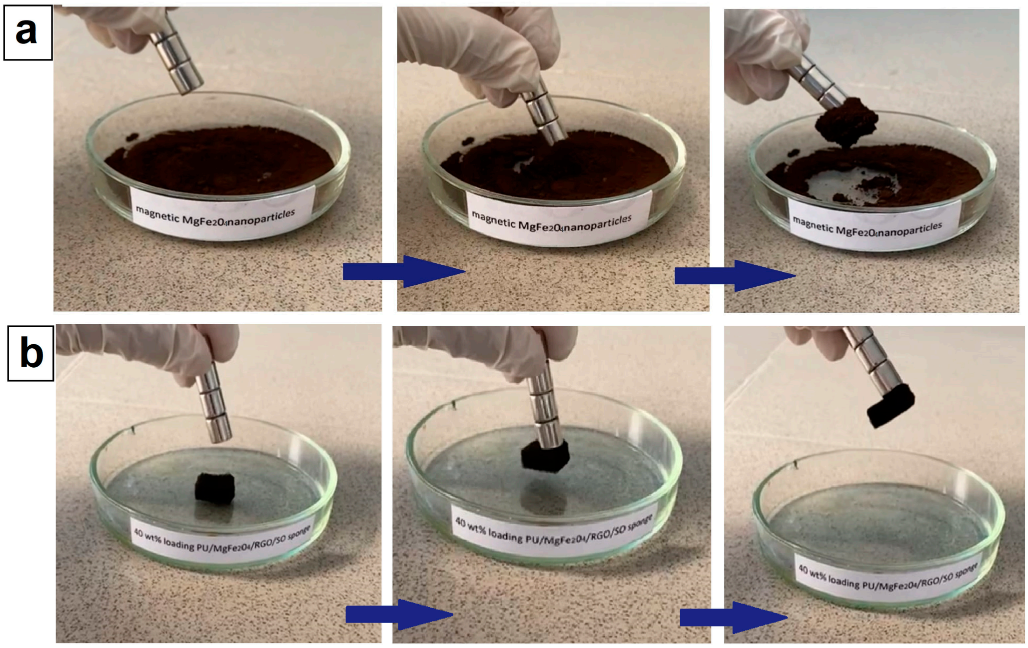

Figure 2. The magnetic properties of the (**a**) magnetic $MgFe_2O_4$ NPs and (**b**) new superhydrophobic PU/$MgFe_2O_4$/SO and PU/$MgFe_2O_4$/RGO/SO magnetic sponges.

3.3. Hydrophobic and Oleophilic Properties of New Superhydrophobic Magnetic Sponges PU/$MgFe_2O_4$/SO and PU/$MgFe_2O_4$/RGO/SO

The separation of oil and water usually affects the interface, and developing new materials with special wettability is an effective strategy. The wettability of the surface of the sponges was assessed using the water contact angle (WCA) measurement, which was conducted by placing drops of oil and water on the surface of an unmodified PU sponge (Figure 3a), 20 wt% (Figure 3b) and 40 wt% (Figure 3c) $MgFe_2O_4$/RGO/SO-modified sponges [32]. The results showed that the unmodified PU sponge had high hydrophilic properties based on the shape of the water drop (Figure 3a). Also, on the surface of the 20 wt% and 40 wt% $MgFe_2O_4$/RGO/SO-modified sponges, the waterdrop remained stable, defining good hydrophobicity. The WCA were about 90° (unmodified PU sponge, Figure 3a) 148.5° (20 wt%, Figure 3b) and 157° (40 wt%, Figure 3c). As shown in Figure 4b, the oil droplet was completely absorbed into the pores of the modified sponge, as the angle of contact with water was 0.0 °C. The rolling angle was 0.3 °C, which in turn indicates its hydrophobic and oleophilic properties.

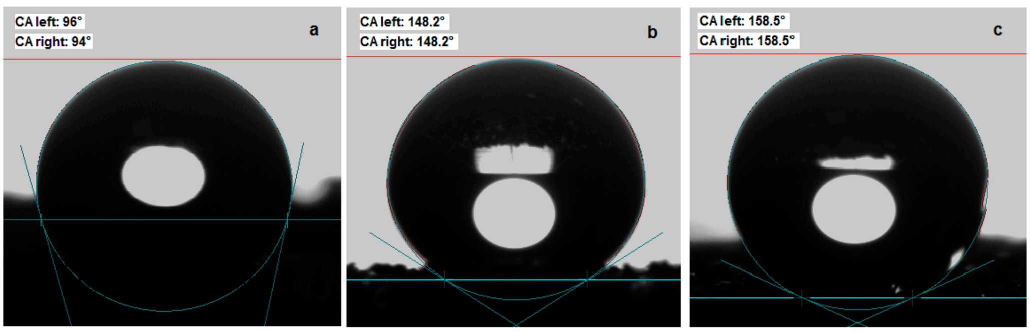

Figure 3. Water contact angle on the (**a**) unmodified PU sponge, (**b**) 20 wt% and (**c**) 40 wt% $MgFe_2O_4$/RGO/SO-modified sponges.

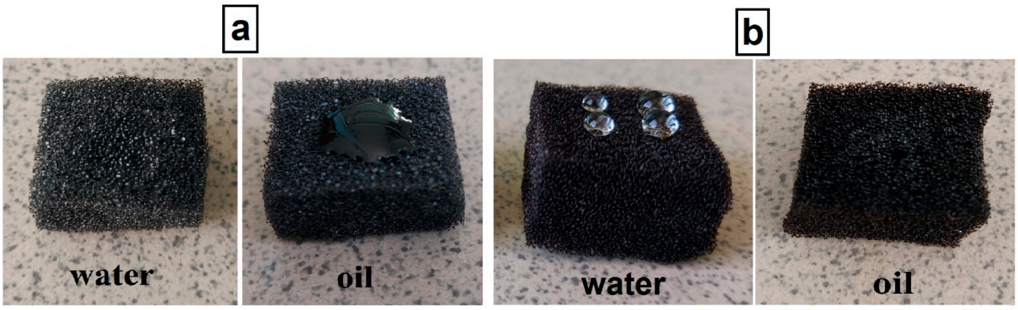

Figure 4. Photographs of drops of water and oil on (**a**) unmodified PU sponge; (**b**) modified sponge PU/$MgFe_2O_4$/RGO/SO ($MgFe_2O_4$/RGO/SO 40 wt% loading).

The hydrophobicity and oleophilic wettability of the $MgFe_2O_4$/RGO/SO-modified sponge were further determined by taking photographs (Figure 4). As seen in Figure 4, a drop of water on the surface of the modified sponge stands in the form of a ball and can easily roll off the surface, showing its superhydrophilic properties [33]. On the contrary, when a drop of crude oil was dropped onto the surface of the sponge, the drop of oil was completely absorbed and penetrated into the interior of the modified sponge, which in turn indicates excellent oleophilic properties. The contact angle of the oil was about 0°. The results also showed that increasing the loading of $MgFe_2O_4$/RGO/SO leads to the enhancement of hydrophobic and oleophilic properties of the sponges.

The hydrophobicity of the modified sponges (PU/$MgFe_2O_4$/SO and PU/$MgFe_2O_4$/RGO/SO) was also investigated by immersing them in water for 10 min at room temperature before applying external force with tweezers. Figure 5 shows a modified sponge immersed in water, where a homogeneous air gap between the hydrophobic surface and water illustrates a typical silver mirror surface [34]. In addition, before immersion, it floated on the water surface due to its lightweight and hydrophobicity (Figure 5). Also, after the action of the external force was stopped, the modified sponge immediately floated to the surface of the water again, as water was not absorbed into the sponge. The water absorption percentage of PU/$MgFe_2O_4$/SO and PU/$MgFe_2O_4$/RGO/SO (40 wt% loading) sponges was investigated by comparing the weight of the sponges before and after immersion. As a result of the experiment, the water absorption of PU/$MgFe_2O_4$/SO (40 wt% loading) was 5.2%, and PU/$MgFe_2O_4$/RGO/SO (40 wt% loading) demonstrated deficient water absorption of about 0.3%. The studies were carried out three times, and the results indicate a positive effect of RGO on the superhydrophobic properties of the modified sponges PU/$MgFe_2O_4$/SO and PU/$MgFe_2O_4$/RGO/SO.

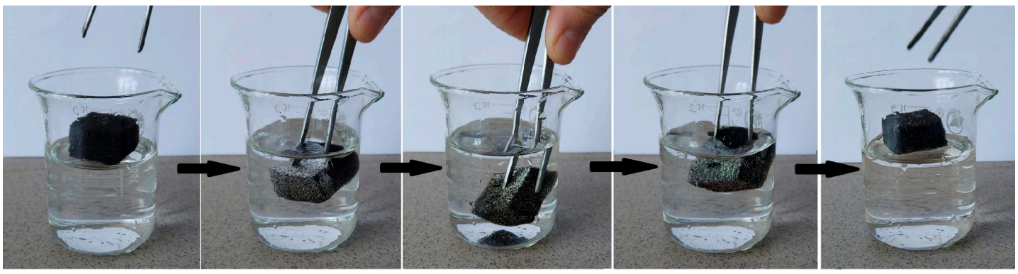

Figure 5. Photographs of a modified PU/$MgFe_2O_4$/RGO/SO sponge ($MgFe_2O_4$/RGO/SO 40 wt% loading) immersed in water using tweezers, showing the silver mirror surface (left to right).

3.4. Oil–Water Separation Tests and Oil Absorption Capacity

The new $MgFe_2O_4$/RGO/SO modified (40 wt% loading) sponge was further tested as an oil-absorbing and recyclable sorbent for wastewater treatment (Figure 6), which are essential factors for its practical application. Also, in order to investigate the positive effect of RGO on the absorption capacity of the newly modified sponge, absorption and recycling experiments were carried out with a sponge without RGO (e.g., PU/$MgFe_2O_4$/SO sponge, 40 wt% loading) [29]. As shown in Video S3 in the Supplementary Materials, the prepared superhydrophobic sponge has excellent sorption properties.

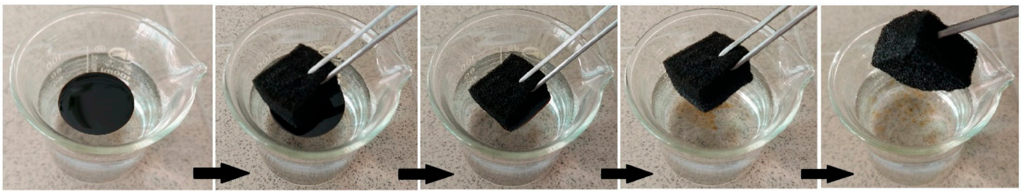

Figure 6. Separation process of oil and water using a superhydrophobic magnetic sponge PU/$MgFe_2O_4$/RGO/SO (40 wt% loading) to collect crude oil from the surface of the water.

We investigated the possibility of reusing $MgFe_2O_4$/RGO/SO- and $MgFe_2O_4$/SO-modified sponges for the selective recovery of crude oil from water by repeated absorption-desorption processes. As shown in the Video S3 in the Supplementary Materials, it is worth noting that the new superhydrophobic sponge had great elasticity and strength, as evidenced by its high absorption capacity after 35 cycles, which gives it a high potential for practical applications [35].

The dependence of the water contact angle on the absorption cycle of PU/$MgFe_2O_4$/RGO/SO (40 wt% loading) was also studied (Figure 7). The results showed that the PU/$MgFe_2O_4$/RGO/SO (40 wt% loading) sponge exhibited a superhydrophobic state after 35 cycles (after 1 cycle 155.5°, 5 cycles 147.5°, 10 cycles 145.6°, 15 cycles 143.8°, 20 cycles 141.9°, 25 cycles 139.0°, 30 cycles 137.6° and 35 cycles 135.7°). The remaining organic solvents after the absorption process were characterized based on appearance (aggregate state, color, smell), density (pycnometer method), boiling point and refractive index (refractometric method). The results showed that the characteristics of organic solvents before and after absorption tests remained unchanged, indicating no leaching of RGO or $MgFe_2O_4$. Additionally, the resulting sponges did not lose their magnetic and hydrophobic properties after 35 cycles of use, also depicting no leaching of RGO or $MgFe_2O_4$.

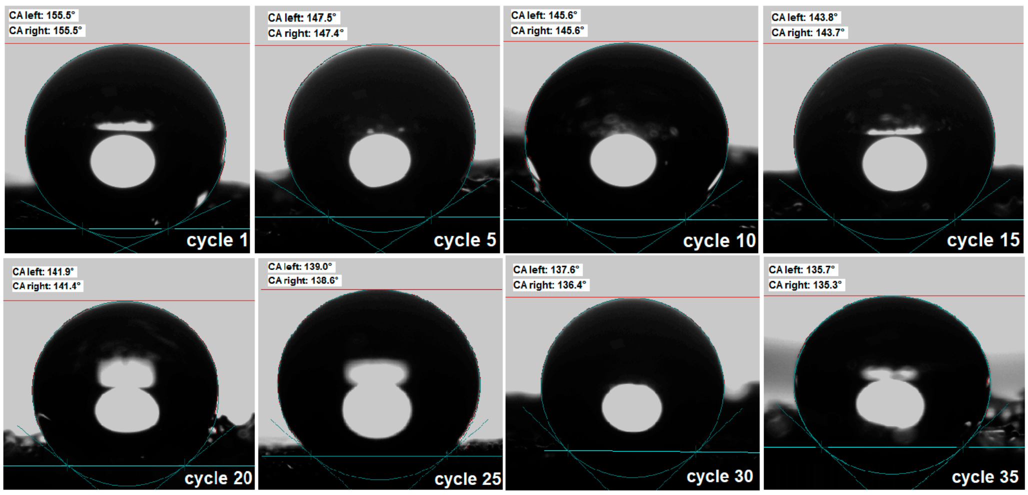

Figure 7. Dependence of water contact angle on the absorption cycle of PU/$MgFe_2O_4$/RGO/SO (40 wt%).

The separation efficiency of the superhydrophobic magnetic sponge was investigated by separating crude oil–water, olive oil–water, toluene–water and ethanol–water mixtures and calculated using Equation (1). Figure 6 shows the successful separation of crude oil–water mixture with high efficiency and an almost imperceptible amount of oil remaining in the water after separation. Also, the magnetic properties of the modified sponge make it easy to place in contaminated areas and remove from the water with a magnet. Calculations of the separation efficiency of the resulting superhydrophobic sponge for various mixtures are shown in Figure 8. It can also be seen that the resulting superhydrophobic magnetic sponge exhibits excellent and efficient crude oil and water separation capability. In particular, the separation efficiency of the prepared superhydrophobic sponges of crude oil–water, olive oil–water, toluene–water mixtures was 97.5%, 89.3% and 96.7%, respectively. The high separating properties of the obtained sponges can be associated with high porosity, since the small pore size creates strong capillary effects [36].

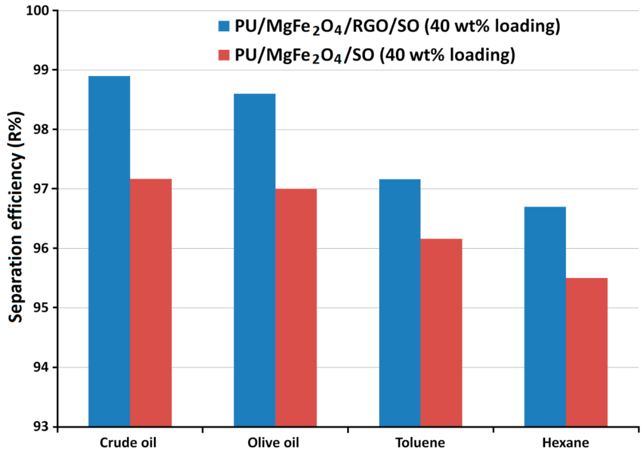

Figure 8. Separation efficiency of prepared superhydrophobic magnetic PU/$MgFe_2O_4$/RGO/SO and PU/$MgFe_2O_4$/SO (40 wt%) sponges for various oil–water mixtures.

These claimed superior properties of the new $MgFe_2O_4$/RGO/SO- and $MgFe_2O_4$/SO-modified PU sponges make them promising candidates for oil–water separation.

Secondly, the absorption capacity of the new $MgFe_2O_4$/RGO/SO- and $MgFe_2O_4$/SO (40 wt% loading)-modified PU sponges for olive oil and organic solvents was investigated using the immersion method for 20 s. The amount of absorbed olive oil and organic solvents was determined by the mass method according to Equation (2). The absorbency of the superhydrophobic sponges containing PU/$MgFe_2O_4$/RGO/SO and PU/$MgFe_2O_4$/SO (40 wt% loading) was tested on various organic solvents with different densities, such as hexane, ethanol, acetone, toluene and chloroform, as well as on olive oil. Figure 9 shows the absorption capacities of the sponges containing PU/$MgFe_2O_4$/RGO/SO and PU/$MgFe_2O_4$/SO (40 wt% loading) during five absorption cycles, with each cycle repeated three times. The results show that the resulting superhydrophobic sponge exhibits excellent absorbency (10–20 times its weight in 20 s of immersion) and after five cycles, the absorbency of the sponges for all solvents and oils was almost unchanged. In addition, the absorption capacity also depends on the density, surface layer and viscosity of the absorbed solvent or oil [37].

The porous structure, super hydrophobicity and oleophilicity, as well as excellent mechanical properties, provide excellent absorbency for organic solvents and oil, as well as high recyclability [38]. Accordingly, it can be concluded that the sponges containing PU/$MgFe_2O_4$/RGO/SO and PU/$MgFe_2O_4$/SO (40 wt% loading) can presumably become a promising and inexpensive material for practical use in the purification of water contaminated with oil and organic solvents.

Finally, the performance of the sponge containing PU/$MgFe_2O_4$/RGO/SO (40 wt% loading), was further investigated for the efficiency of continuous separation of oil–water mixtures using a diaphragm vacuum pump, a rubber tube and a sponge (Figure 10a) [39]. With the help of a diaphragm vacuum, a tube connected with a modified sponge, when placed at the oil/water interface, quickly sucks in oil and forms an oil flow in the tube due to its continuous suction, then the oil flows through the tube into a connected glass flask, and the thickness of the oil layer in the glass gradually decreases (Figure 10c).

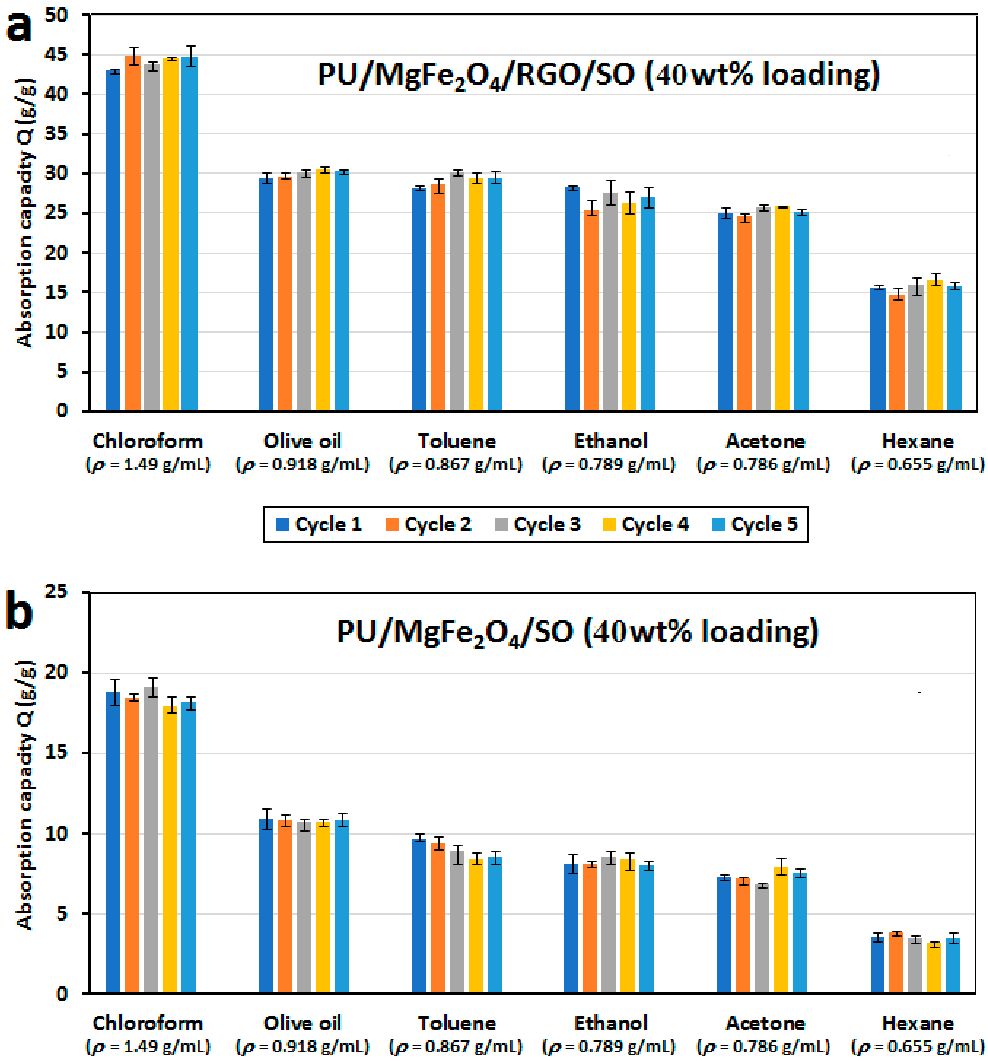

Figure 9. Absorption capacity of superhydrophobic magnetic (**a**) PU/$MgFe_2O_4$/RGO/SO and (**b**) PU/$MgFe_2O_4$/SO (40 wt% loading) sponges for oil and various organic solvents.

Figure 10b shows that no oil remained on the surface of the water and no water was found in the collected oil, and the separation efficiency was about 99.4%. Separation efficiency was evaluated based on the difference between the mass of oil and water before and after separation. All experiments were carried out three times.

Experimental results showed that the sponge containing PU/$MgFe_2O_4$/RGO/SO (40 wt% loading) had excellent separation efficiency and high selectivity, due to its admirable hydrophobicity and lipophilic capacity.

The absorption capacity of absorbents mainly depends on the physical properties of the absorbed substance, such as density and surface tension, as well as on the time it is immersed in these liquids. The new $MgFe_2O_4$/RGO/SO (40 wt% loading)-modified sponge exhibits excellent and fast (immersion time 20 s) absorbency for oil and organic solvents (16.61 to 44.86 g/g), while repelling water (water absorption of about 0.5%), as well as a very high separation efficiency of oil– and organic solvent–water mixtures (up to 98.9%). The absorption capacities of PU/$MgFe_2O_4$/RGO/SO are relatively high compared to other magnetic composites found in the literature (Table 1), indicating competitiveness and high potential interest in the new PU/$MgFe_2O_4$/RGO/SO sponge as a promising candidate for the separation of oil and water.

Figure 10. Continuous separation of oil and water using a superhydrophobic PU/$MgFe_2O_4$/RGO/SO magnetic sponge (40 wt% loading) and a diaphragm vacuum pump, (**a**) before separation; (**b**) after separation; (**c**) during separation.

Table 1. Adsorption capacity of various sorbent materials described in the literature.

Material	Adsorbed Organics	Q (g/g)	References
PU/$MgFe_2O_4$/RGO/SO sponge	Crude oil, olive oil, chloroform, toluene, ethanol, acetone, hexane	16.61–44.86	Present work
PU/$MgFe_2O_4$/SO sponge	Olive oil, chloroform, toluene, ethanol, acetone, hexane	3.5–19	Present work
PU/$MgFe_2O_4$/RGO/PDMS sponge	Crude oil, olive oil, chloroform, toluene, ethanol, acetone, hexane	12–36	[29]
PU/$MgFe_2O_4$/PDMS sponge	Olive oil, chloroform, toluene, ethanol, acetone, hexane	4–22	[29]
Fe_3O_4-PDMS/MWNTs sponge	Dichloromethane, petroleum ether, hexane, chloroform, tetrahydrofuran, toluene, gasoline	8.5–20	[39]
PDMS sponge	Dichloromethane, toluene, transformer oil	4.3–11	[40]
PU Sponge@Fe_3O @SiO_4 @Fluoropolymer sponge	Petrol, toluene, chloroform	17–23	[41]
PU Sponge@Magnesium Stearate@Phenol formaldehyde Resin	Motor oil, food oil, paraffin, gasoline, n-hexane, toluene	19–38	[42]
3D macroscopic superhydrophobic magnetic porous carbon aerogel	Engine oil, chloroethane and corn oil	10.02−10.83	[43]
TiO_2–PVA sponge	Polyethylene glycol, CCl_4, liquid paraffin, *N*, *N*-dimethylformamide, ethanol, edible oil and *n*-hexane	4.3–13.6	[44]
CNT/PDMS-coated PU sponge	Soybean oil, used motor oil, diesel oil, n-hexadecane, gasoline, n-hexane	15–25	[45]

4. Conclusions

Superhydrophobic sponges have significant potential for separating oil and water. The practicality of the current methods used to modify superhydrophobic sponges is compromised due to their environmental footprint, high cost and easy destruction of the hydrophobic layer on the surface of the sponge. This article presented a straightforward method to develop a high-performance super hydrophobic sponge with excellent recyclability. The SEM images showed the high porosity and roughness of the sponge, confirming that the sponge's surface was successfully modified with PU/$MgFe_2O_4$/RGO/SO and PU/$MgFe_2O_4$/SO. The wettability of PU/$MgFe_2O_4$/RGO/SO was studied, and the

WCAs of the sponges increased when loading was increased up to 20 wt% (148.5°) and 40 wt% (157°), respectively, which proves their superhydrophobic properties. And also, experiments on adsorption and cycling showed that sponges with a loading of 40 wt% PU/$MgFe_2O_4$/RGO/SO and PU/$MgFe_2O_4$/SO showed excellent absorption capacity for various organic solvents and oils (16.61–44.86 g/g), with an oil-water separation efficiency of up to 97.5%, and the structure of the obtained sponges remains stable after 20 cycles. In addition, PU/$MgFe_2O_4$/RGO/SO and PU/$MgFe_2O_4$/SO sponges have shown excellent recyclability, and due to their magnetic properties, the sponges can be controlled by a magnet. The new superhydrophobic magnetic material PU/$MgFe_2O_4$/RGO/SO, compared with the material synthesized in [29], shows enhanced performance (the absorption capacity of oil and organic solvents increased by 24%–35%) and cost-effectiveness. The authors suggest that the high absorbance of the PU/$MgFe_2O_4$/RGO/SO sponge is more related to the physical properties of SO; its softness and elasticity give an excellent distribution of components throughout the entire surface of the sponge, making it softer and more flexible than [29], thus enhancing the quality and accelerating the absorption. Therefore, it can be concluded that superhydrophobic and superoleophilic sponges, containing PU/$MgFe_2O_4$/RGO/SO and PU/$MgFe_2O_4$/SO, will be widely used to clean water from oil and organic pollutants in the future.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/coatings13101777/s1, Video S1: The magnetic properties of the magnetic $MgFe_2O_4$ NPs; Video S2: The magnetic properties of the new superhydrophobic PU/$MgFe_2O_4$/SO and PU/$MgFe_2O_4$/RGO/SO magnetic sponges; Video S3: Separation process of oil and water using a superhydrophobic magnetic sponge PU/$MgFe_2O_4$/RGO/SO (40 wt% loading) to collect crude oil from the surface of the water.

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