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Investigation of Absorption Capacity of Surface Modified PU Sponges for Oil Spills Using Different Silane Coating Agents

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Abstract

Today's marine oil spills are causing environmental concerns on a global scale. In order to effectively remove such pollutants, various absorbent materials with two-dimensional (2D) and three-dimensional (3D) structures and super-wetting properties have been developed. However, there are significant difficulties in desorption of the absorbed oil from these materials. The strong adsorption of oil components on the surface of the material limits both the efficiency of sorbent materials and the potential for reuse. In this study, polyurethane (PU) sponge adsorbents coated with different silane agents were fabricated to treat oil spills. Four different types of silanes Polydimethylsiloxane (PDMS), Octadecyltrichlorosilane (ODTCS), Methyltrichlorosilane (MTCS) and Dodecyltrimethoxysilane (DTMS) were used as silane agents. For the prepared test specimens; silane binder ratios of 0.1%, 0.5% and 1% were used in three different ways, respectively. The coating temperatures were 25°C, 40°C and 55°C and the coating times were 15 minutes, 30 minutes and 45 minutes. The contact angles and absorption capacities of the obtained PU sponge adsorbents with water were measured. In addition, their surface morphologies were examined by SEM analysis. The data obtained showed that the best absorption capacity for 81 g was achieved in the coating with PU-MTCS silane agent when 1% silane agent, 25 °C reaction temperature and 30 minutes time were applied.

Keywords: Superoleophilic sponge, silane agent, adsorbent, sorbent

INTRODUCTION

Oil spills have a major impact on the environment and ecosystems and have become a global problem with increasing amounts of oily wastewater from industry and frequent oil spill accidents resulting in energy loss. To treat oil spills, there is an increasing need to develop new materials that can effectively absorb and transport oil spills from water [1,2]. Many oil spills into rivers and seas every year. These oil spill accidents have occurred in more than 21 major incidents between 2019 and 2022 [3]. Such as the Exxon Valdez major oil spill [4], the "Sanchi" oil tanker spill in 2018 [5], etc., have led to huge economic losses and serious environmental pollution. To solve these problems, various techniques have been proposed, such as solidification, stripping, sorption, dispersion, and controlled incineration [6-7]. Among these efforts, oil-absorbing materials including inorganic mineral products, organic natural products and synthetic polymers have been widely used due to their special potential [8-9]. It is well known that the wettability of materials with water and oil can be reasonably controlled by their chemical composition and surface geometrical structure [10]. Especially sorbent materials with special wettability properties are recognized as the most promising candidates for efficient treatment of oily wastewater [11]. Their marked opposite affinity for oil and water is useful to remove one phase from an oil/water mixture and simultaneously repel the other phase, resulting in selective oil/water separation. It is envisioned that an ideal super-wet surface for oil-water separation should be both superhydrophilic and superoleophobic so that water can pass through the materials and oil can still be blocked [12-13]. To achieve this goal, significant research has recently been conducted to obtain superhydrophobic and superoleophilic textured surfaces [14]. Pan et al. described an innovative and general method for the fabrication of ultra-low density magnetic foams with superhydrophobic and superoleophilic properties for oil-water separation [15]. Melamine sponge is a commercially available three-dimensional porous material that has been used in some studies as a substrate for the development of oil-absorbing surfaces. Through surface modification, hydrophobic characteristics can be imparted, enhancing its absorption capacity for organic compounds [16]. In general, superhydrophobic surfaces can be achieved by combining suitable surface roughness with hydrophobic materials [17].

Commercially available sponges include melamine sponge (MS), PU sponge and cellulose sponge (CS). Clean and unmodified sponges have both hydrophilic and oleophilic properties [18]. Among various absorbent materials used for oil-water separation, PU, poly(melamine-formaldehyde) (PMF), PDMS, and carbon/graphene-based sponges have been extensively studied due to their promising properties. Since PU sponges are amphiphilic, they cannot be used directly for oil-water separation. Therefore, in order to effectively separate marine oil spills and industrial or domestic waste oils, PU sponges need to be hydrophobically modified with certain methods and materials. By adding fire retardants during the sponge manufacturing process or by applying fire retardant coatings to the surface of the sponge, these sponges have been given excellent fire retardant properties while maintaining highly efficient oil-water separation capability. At the same time, in order to meet the application requirements, some specific functions are imparted to the sponge according to the specific properties of the modified material. This allows better adaptation to harsh environmental conditions such as high viscosity oils, high acidity or alkalinity [19-20].

The presence of dispersed siloxane groups on the adsorbent surface facilitated oil-water separation. Although these silane modification materials show improved adsorption capacities, the use of silane coupling agents to modify organic materials is generally easier than chemical reactions or grafting methods [21]. After hydrophobic modification, inorganic particles can rapidly adsorb the fine oil phase on the water surface; however, the low adsorption capacity and difficulty of aggregation of such particulate materials limit their application in oil-water separation. Therefore, three-dimensional porous hydrophobic materials offer advantages in terms of in situ sorption and portability due to their large storage volume. Therefore, the effect of silane coatings on the adsorption capacity of superoleophilic PU sponges needs to be investigated [22].

In the existing literature, it is noteworthy that there is a scarcity of studies that systematically investigate the production parameters using both high absorption capacity, different silane agents, different coating temperatures, times and ratios. In this study, PU sponge adsorbents surface modified with different silane agents were developed to provide solutions to the global environmental threats posed by marine oil spills. In addition, the absorption capacities of the adsorbents were increased by using different coating temperatures, times and ratios for PU sponges. Thanks to the SEM analyzes obtained, it was determined that there were improvements in the microstructure. 2D/3D porous structures and surface morphologies of the developed sponge were analyzed. The results of the analysis showed that the developed sponge effectively absorbs petroleum components by showing superhydrophobic properties. This study has contributed to the development of low-cost and effective oil absorbent materials while providing solutions to current problems such as desorption difficulties and reuse limitations.

MATERIAL AND METHOD

Material

In the experimental study, PU sponges had a density of 35 kg/m^3, a medium pore size of 30 PPI and dimensions of 20x20x40 mm. The silane agents used in the coating of PU sponges were obtained from Merck İlaç Ecza ve Kimya Tic. A. Ş., hexane was used as solvent and their physical properties are given in Table 1.

Table 1 Physical properties of the chemicals used in the experimental studies.

Chemicals	Chemical Formula	Melting Point (°C)	Boiling Point (°C)	Flare Point (°C)	Density (g/cm^3)
PDMS	$(C_6H_{18}O_4Si_3)n$	-50	35 (1.013hPa)	321	0.76-0.97
ODTCS	$C_{18}H_{39}Cl_3Si$	22	223 (13hPa)	207	0.984
MTCS	CH_3SiCl_3	-77.8	66	8	1.273
DTMS	$C_{12}H_{25}O_3Si$	-15	315	164	0.875
Hexane	C_6H_{14}	-95	68.7	-22	0.654

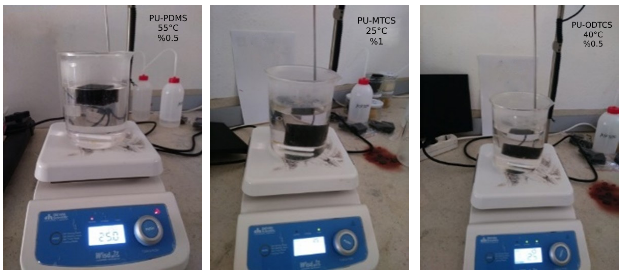
Preparation of Superoleophilic Sponges

In this study, various silane agents—including MTCS, PDMS, ODTCS, and DTMS—were utilized to modify the surface properties of PU sponges. As summarized in Table 2, the coating conditions were systematically optimized by varying the silane concentration (0.1%, 0.5%, and 1%), coating temperature (25°C, 40°C, and 55°C), and reaction time (15, 30, and 45 minutes). These parameters were selected considering their significant influence on silanization efficiency and surface wettability. The optimization of these conditions played a critical role in controlling the surface energy and enhancing the oil-water separation performance of the fabricated materials.

The PU sponges were first washed with acetone and dried in an oven at 100 °C for 45 minutes. Then, the coating solution was prepared by adding hexane as a solvent into the beaker with a simple immersion method. In the experimental studies, the amounts of silane agents in the coating solution were adjusted as 400:4:0.4, 400:20:2 and 400:40:4 for PU-PDMS two-component A and B component, 400:2, 400:4 and 400:8 for PU-ODTCS, PU-MTCS, PU-DTMS agents. Each sample was mixed at different coating temperatures (25°C, 40°C and 55°C) and for different times (15 minutes, 30 minutes and 45 minutes) to prepare the adsorbents as shown in Fig. 1. The prepared silane agents were cured in an oven at 120°C for 60 minutes for binding to the PU sponge surface.

Table 2 Coating parameters of PU sponges produced by applying different coating temperatures silane agents, and coating times

	Coating Temp. [°C]		Rate of Silane Agent [%]		Coating Time [min.]
MTCS	25	MTCS (25°C)	0.1	MTCS (0.5%)	15
	40		0.5		30
	55		1		45
ODTCS	25	ODTCS (40°C)	0.1		
	40		0.5		
	55		1		
PDMS	25				
	40				
	55				
DTMS	25				
	40				
	55				



(a) (b) (c)

Figure 1 Images of the coated (a) PU-PDMS (b) PU-MTCS (c) PU-ODTCS experimental setup.

Absorption Capacity Measurement

In this study, the absorption capacities of PU based sponges against organic solvents and oils were investigated. In order to determine the liquid holding capacity of the sponges, the results were calculated with the weight-based measurement method as indicated in Equation 1 [23], which is the most widely used of the three different measurement methods (weight, volume and actual volume based) used to evaluate the absorption capacity.

$$\text{Weight absorption capacity} = \frac{m_a - m_d}{m_d} \quad (1)$$

Where *ma* is the weight of the sponge after liquid absorption and *md* is the weight of the coated liquid-absorbed sponge. In line with the data obtained, the liquid retention capacities of PU sponges were calculated and comparative analyzes were carried out depending on the material type and silane agent.

Measurement Uncertainty Analysis

One of the important factors affecting the reliability of experiments is the distinction between fixed errors and random errors. Experimental errors are usually divided into two main groups, which are caused by the experimental setup, the measuring instruments and the person performing the experiment. These errors, which affect the uncertainty of the experimental results, are calculated by Equations (1)-(4) [24]. Constant errors occur uniformly for all measured values and can be eliminated by appropriate calibration and corrections. The uncertainties encountered in the experiment include errors due to the reading of tabular values of velocity and physical properties. These errors include in-system temperature (T_{fg}), ambient temperature (T_c), temperature measurement time (T_s), thermo element pairs (a_1), digital thermometer (b_1), fasteners (c_1), SEM device measurement error (e_1), ambient temperature measurement error (l_1), time reading error (a_2), periodic temperature acquisition error (c_2), temperature measurement error (W_{cv}), mixing element error (v_1) and digital sensor error (x_1). Uncertainty analysis measurement results were obtained with error values ranging from 0.7 to 1.22.

$$W_{T_{fg}} = \sqrt{\left[{a_1}^2 + {b_1}^2 + {c_1}^2 + {e_1}^2\right]}$$

$$W_{T_c} = \sqrt{\left[{a_1}^2 + {b_1}^2 + {c_1}^2 + {l_1}^2\right]} \tag{2}$$

$$W_{T_s} = \sqrt{\left[{a_1}^2 + {c_1}^2\right]} \tag{3}$$

$$W_{C_v} = \sqrt{\left[{v_1}^2 + {x_1}^2\right]} \tag{4}$$

Table 3 Derived error analysis parameters. (5)

Parameters Causing an Error	Unit	Total Error
Total error in temperature measurement		
$W_{T_{fg}}$	°C	0.781
W_{T_c}	°C	0.925
W_{C_v}	m/s	0.1
Total error in time measurement		
W_{T_s}	minute	0.02
Other errors	%	0.1

As seen in Table 3, a detailed uncertainty analysis was conducted to ensure the reliability of the experimental results. The total error in temperature measurements was found to be 0.781 °C and 0.925 °C for different components, while velocity and time measurement uncertainties were 0.1 m/s and 0.02 min, respectively. These values fall within acceptable limits and indicate that the variations in absorption capacity are primarily due to material and process parameters rather than experimental error. The low error margins confirm the robustness and reproducibility of the applied methodology [25].

RESULT AND DISCUSSION

In this study, a comprehensive optimization process was conducted to enhance the surface modification a nd absorption capacity of PU sponges. The effects of critical parameters, including temperature, reaction time, silane type, and silane concentration, were systematically investigated, and the individual impacts of these parameters on material performance were evaluated. Contact angle and absorption capacity were used as performance metrics, and the conditions were supported by scanning electron microscopy (SEM) analyses. The iterative optimization process improved modification efficiency and ensured exp erimental reproducibility. The obtained results provide an effective roadmap for enhancing the functionality of PU sponges in environmental applications.

SEM Analysis of Surface Morphology

The morphological changes of PU sponge surfaces after coating at different temperatures (25 °C, 40 °C and 55 °C) using 0.5% silane agent for 30 minutes were evaluated by scanning electron microscopy (SEM) images. Fig. 2 shows SEM images of PU-PDMS, PU-ODTCS, PU-MTCS and PU-DTMS surface morphologies, respectively.

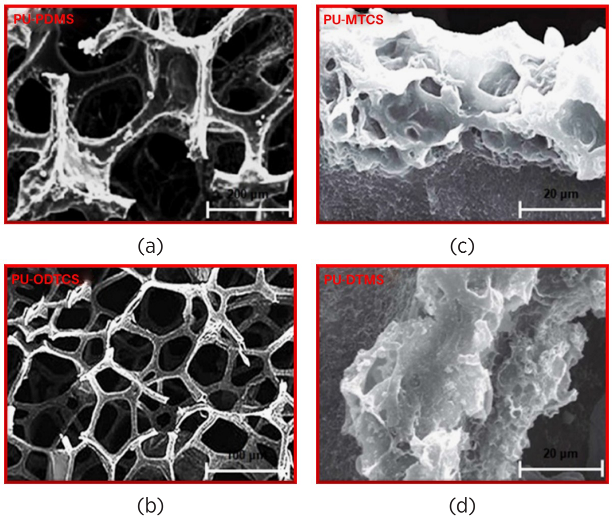
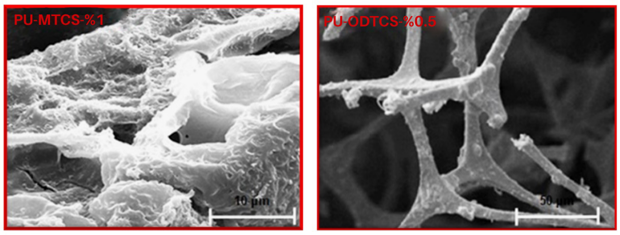


Figure 2 SEM images obtained at different coating temperatures (a) PU-PDMS-55°C, (b) PU- ODTCS-40°C (c) PU-MTCS-25°C and (d) PU-DTMS-25°C.

Fig. 2(a), the PU-PDMS-55°C adsorbent shows that the skeletal structure of the sponge was generally preserved, but irregular film-like formations were observed on the surface due to the PU-PDMS coating. Although the coating spread throughout the porous structure, it caused local accumulations and agglomerations in some regions. This suggests that the viscous nature of PU-PDMS may limit its ability to provide a homogeneous distribution on the surface. Despite the increased micro-roughness of the PU-PDMS coated surface, the coating layer exhibited a thin and heterogeneous morphology, resulting in an average contact angle of approximately 118°. This improved coating uniformity and associated surface energy characteristics led to a reduced oil-water separation capacity compared to sponges modified with other silane agents that formed thicker and more homogeneous coatings, thereby enhancing hydrophobicity and sorption performance. Fig. 2(b), the SEM image of PU-ODTCS-40°C shows that the sponge surface was modified with a more regular, homogeneous and continuous coating. It is observed that the PU-ODTCS coating is evenly distributed throughout the sponge skeleton, preserving the pore structure and forming a uniform film on the surface. This morphological structure indicates that PU-ODTCS develops stronger physicochemical interactions with the sponge surface due to its long alkyl chains and the coating is effectively adhered to the surface. The uniformity of the coating shows that it has a high superhydrophobic character obtained from contact angle measurements. This high contact angle of 154° indicates that PU-ODTCS with a long alkyl chain significantly reduces the surface energy and creates a surface that completely repels water. Furthermore, the absorption capacity was measured to be 61 g. Since low-density organic solvents (e.g., hexane, $\rho \approx 0.66$ g/cm^3; toluene, $\rho \approx 0.87$ g/cm^3) occupy a larger volume per unit mass, this high value indicates that the sponge effectively absorbs such solvents on a volumetric basis. This observation is consistent with previous studies reported in the literature [26]. Fig. 2(c), the surface of the sponge coated with PU-MTCS-25°C is characterized by open and regular pores. The coating is homogeneously distributed on the surface, enhancing the superoleophilic properties of the sponge. The even distribution of the coating throughout the porous structure indicates that the PU-MTCS provides a controlled interaction with the surface and is homogeneously adhered to the structure. In addition, the 152° value obtained in contact angle measurements indicates that the surface has become highly hydrophobic and water drops can slide without adhering to the surface. This indicates that the PU-MTCS bonded effectively to the surface even though the coating was carried out at low temperature (25 °C). The absorption capacity of about 63 g indicates that PU-MTCS sponge can work with high efficiency against organic pollutants (especially petroleum derivatives) and the low temperature production of this material makes it an economical superoleophilic adsorbent alternative. Fig. 2(d), the surface of the PU-DTMS-25°C coated sponge exhibits a more irregular and coarse morphology, with pores less pronounced and closed in some areas. Dense coating deposits were also observed. This causes irregularities in the thickness and surface morphology of the coating, limiting the desired level of superoleophilic properties.

The chemical structure of the coating agent (e.g. chain length and reactive groups) and its dispersion stability in solution are the determinants of the surface morphology and functional surface properties obtained on PU sponges. The coatings obtained using PU-ODTCS and PU-MTCS showed higher contact angles and improved superoleophilic properties thanks to their regular and homogeneous surface morphologies. On the other hand, structural irregularities and homogeneity problems were observed in coatings made with PU-DTMS and PU-PDMS, which resulted in a decrease in functional surface performance. As a result, when the absorption capacities of the adsorbents developed for oil spills using different silane agents at different coating temperatures were evaluated, the most suitable silane agent PU-MTCS 63 g and PU-ODTCS 61 g were measured. Due to the insufficient superoleophilic performance and low absorption capacity of the samples modified with other silane agents, the effect of silane concentration and coating time parameters for these agents were not systematically investigated.

According to the absorption test results, PU-MTCS exhibited its highest absorption capacity of 63 g at a coating temperature of 25 °C, while PU-ODTCS reached its peak capacity of 61 g at 40 °C. These findings suggest that the silanization temperature varies depending on the silane structure and its interaction with the PU matrix. Fig. 3, the SEM images of the samples were examined using 0.5% and 1% silane agents and the coating time was 30 min.

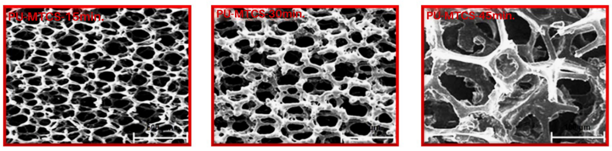


(a) (b)

Figure 3 SEM images of materials produced with (a) PU-MTCS-1%, (b) PU-ODTCS-0.5% with different silane agents at different ratios.

Fig. 3(a), the SEM image of the PU-MTCS-1% sample reveals that an irregular, layered and dense silane layer was formed on the surface of the sponge structure. This dense coating caused partial occlusion of the porous structure and increased surface roughness. The resulting morphology led to a reduction of the surface energy and a high-water contact angle of about 162° was obtained in this sample. This result shows that the MTCS agent effectively binds to the sponge surface at the relevant rate and imparts superoleophilic properties to the surface, and the highest result was achieved with an absorption capacity of 81 g. The results obtained support the information in the literature and are in accordance with the Cassie-Baxter type wetting mechanism [27]. Fig. 3(b) shows that the PU-ODTCS-0.5% sample has a more open and regular porous structure. It is observed that the silane coating is homogeneously distributed on the surface of the sponge structure and the overall structure of the sponge skeleton is preserved. This less layered and uniform structure resulted in a contact angle of approximately 149°. Although the long alkyl chain of PU-ODTCS greatly reduced the surface energy, it did not produce as high a roughness effect as PU-MTCS due to the more uniform and thinner coating, resulting in a slightly lower contact angle. Although highly hydrophobic and oleophilic surfaces were obtained when both silane agents were used, the dense and rough structure observed on the surfaces of the sponges modified with PU-MTCS increase in the contact angle seems to be more pronounced. In contrast, the PU-ODTCS coating provided a more regular structure, preserving the open porosity and significantly maintaining the mechanical and permeability properties of the sponge skeleton. When the contact angle and absorption capacities of both samples were evaluated, it was found that the samples coated with PU-MTCS showed superior performance in general. Specifically, PU-ODTCS exhibited a contact angle of 154° and an absorption capacity of 61 g under the experimental conditions. One of the most common approaches to obtain superhydrophobic surfaces is to reduce the surface energy of the material [27]. In this context, two different silane agents were preferred to create hydrophobic surfaces with low surface energy. The PU-MTCS coating applied to the PU foam surface is more effective than PU-ODTCS and reduces the wettability by reducing the surface energy more successfully. According to the application results, it was determined that the most efficient improvement in surface modification was achieved with PU-MTCS and PU-MTCS was the most industrially suitable silane agent for PU foam, especially in terms of absorption of oil spills.

The surface morphology obtained as a result of the coating process performed with PU-MTCS-1% agent at 25 °C shows significant changes depending on the coating time (15 minutes, 30 minutes and 45 minutes). The SEM images presented in Fig. 4 clearly shows the differences in surface roughness, pore opening and coating homogeneity over time. The effect of these morphological changes on the contact angle with water is critical for understanding the wettability properties of the surface.



(a) (b) (c)

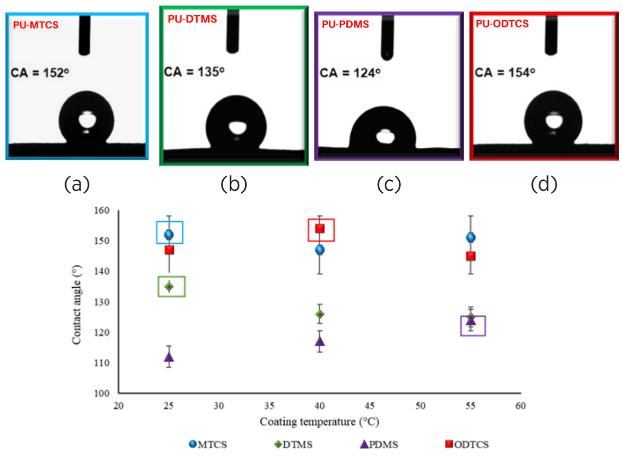
Figure 4 SEM images of PU sponge adsorbent produced using MTCS silane agent at different coating times of (a) 15 minutes (b) 30 minutes (c) 45 minutes.

In Fig. (4a), the surface of the sponge coated with PU-MTCS-15' with a coating time of 15 minutes is relatively smooth and the pores remain largely open. The limited silane distribution observed in the SEM image shows that there is not enough roughness on the surface and therefore the contact angle remains at about 153°. This structure indicates that the MTCS is not sufficiently adsorbed on the sponge surface and the contact with the water drop is more. Fig. 4(b), the coating time of 30 minutes gave the most stable results in terms of modification in the sponge coated with PU-MTCS-30'. In the SEM image, it is seen that the porous structure is preserved but the surface is significantly roughened. The absorption capacity measurement for the oil spill was recorded as 81 g. These results show that the coating time of 30 minutes corresponds to the most efficient time interval for the PU-MTCS sample. Fig. 4(c), the surface coated with PU-MTCS-45' at 45 minutes coating time shows a visibly thick, irregular morphology with some agglomeration. Pore openings decreased and dense silane accumulation occurred in the structure. This structure disrupted the porous structure of the surface, causing the contact angle to decrease to approximately 151°. The excessively dense coating caused the air gaps between the water drop and the surface to disappear, thus increasing the contact surface.

In conclusion, the surface morphology and contact angle of PU sponges coated with PU-MTCS are directly dependent on the coating time. The time of 30 minutes offered the highest superhydrophobic performance by both maintaining the homogeneity of the surface morphology and roughness. These findings suggest that c areful c ontrol of t he time parameter in silane-based surface modifications is critical to achieve the desired surface properties.

Effect of Coating Temperature on the Contact Angle of Superoleophilic PU Sponges

Fig. 5 presents the water contact angles measured on PU sponges treated with various silane agents (MTCS, DTMS, PDMS, and ODTCS) at coating temperatures of 25 °C, 40 °C, and 55 °C, emphasizing the influence of these treatments on water repellency. In this study, the water contact angles of the PU sponges were determined by the static contact angle measurement technique. Fig. 5(a), it is observed that the contact angle of PU-MTCS coating decreases slightly with temperature and reaches the highest contact angle of 152° at 25 °C. In Fig. 5(b), in contrast, PU-DTMS-coated sponges showed a decrease in contact angle measurements with increasing coating temperature, resulting in contact angles in the range of 125°-135° at all temperatures without improvement in absorption capacities. Fig. 5(c), the PU-PDMS coating showed an increase in the contact angle with increasing temperature, but the maximum value was 124°, which was lower than the samples coated with other silane agents. According to the results obtained, in Fig. 5(d), the sponges coated with PU-ODTCS showed the highest contact angles at all coating temperatures, giving the surface superior hydrophobic properties. In particular, it exhibited maximum performance at 40 °C with a contact angle of approximately 154°. The results concluded that the coatings obtained with PU-ODTCS and PU-MTCS created a more effective hydrophobic structure on the surface of the sponge, while PU-PDMS provided moderate hydrophobicity and PU-DTMS provided low hydrophobicity.



(e)

Figure 5 Effect of (a) PU-MTCS, (b) PU-DTMS, (c) PU-PDMS, (d) PU-ODTCS and (e) different silane agents coating temperature on contact angle of superoleophilic PU sponges.

Effect of Silane Agent Ratio on Contact Angle

After measuring the absorption capacities of PU sponges coated with different silane agents depending on the coating temperature, the coating solutions were prepared by adding silane to hexane solvent at 0.1%, 0.5% and 1% to the samples produced using PU-MTCS and PU-ODTCS silane agents which gave the best results and contact angle measurements were made. The results obtained are as shown in Fig. 6.

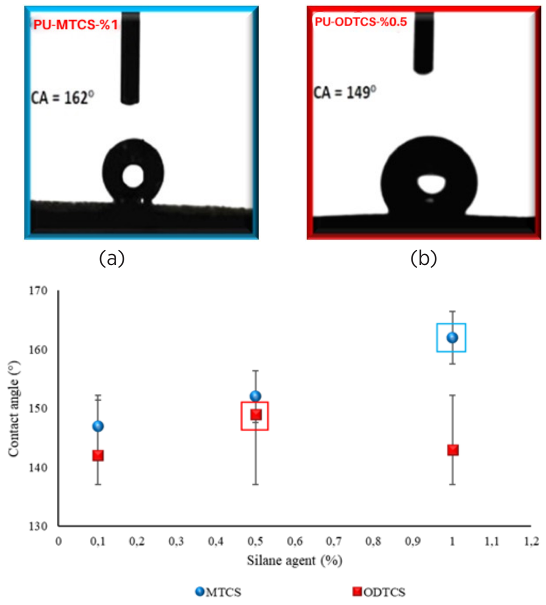


Figure 6 Effect of silane agents and concentration on contact angle (a) PU-MTCS, (b) PU-ODTCS

The effects of PU-MTCS and PU-ODTCS silane agents used at different ratios (0.1%, 0.5% and 1%) on the contact angle on PU surfaces were compared. According to the findings, PU- MTCS-coated surfaces exhibited higher contact angles compared to PU-ODTCS at all ratios, indicating that the surface became more hydrophobic. In particular, the contact angle of about 162° obtained in the coating with PU-MTCS-1% reveals that the surface with this agent has reached a superhydrophobic character Fig. 6(a). On the contrary, the highest contact angle of about 149° was observed at 0.5% for surfaces coated with PU-ODTCS, but this value decreased at 1%, Fig. 6(b). This suggests that PU-ODTCS is effective at a certain optimum concentration, whereas at higher ratios, the hydrophobic efficiency decreases due to the formation of uneven coating on the surface. On the other hand, as shown in Fig. 6(c), PU-MTCS appears to form a more regular surface coating with increasing concentration, which may contribute to a reduction in surface energy. However, since quantitative surface energy measurements were not conducted, it remains unclear whether the observed effects are primarily due to changes in surface morphology, chemical composition, or both. These results reveal that PU-MTCS is a more effective surface modification agent than PU-ODTCS in enhancing the hydrophobic properties of the surfaces. These findings suggest that not only the type of silane agent used in superoleophilic surface coating should be carefully optimized, but also the rate of application.

Effect of Coating Time on Contact Angle

The influence of coating duration on the wettability behavior of the modified PU surfaces was systematically investigated. As seen in Fig. 7, the contact angle values of PU surfaces modified with PU-MTCS varied depending on the coating times of 15, 30, and 45 minutes. All measurements were repeated three times to ensure consistency of the results. The obtained data demonstrate that coating time plays a significant role in modulating the surface's hydrophobic behavior.

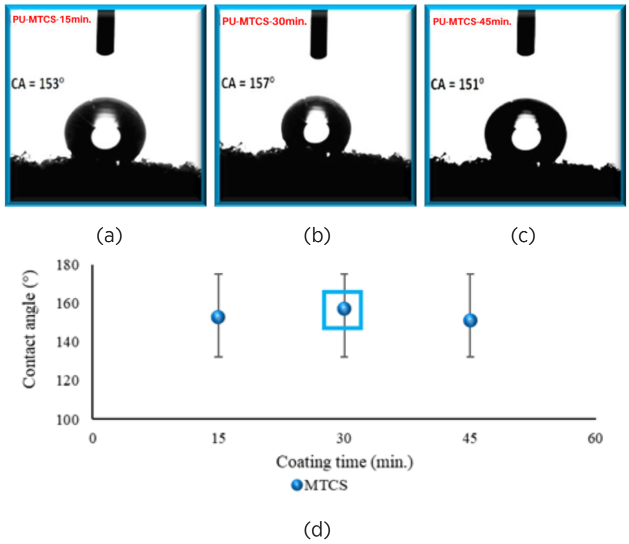


Figure 7 Effect of coating time (a) 15 minutes (b) 30 minutes (c) 45 minutes and (d) coating time-contact angle on contact angle.

In Fig. 7(a), the contact angle of ~153° obtained at a coating time of 15 minutes indicates that the PU-MTCS has successfully bonded to the surface, although the surface coating has not yet reached full saturation. In Fig. 7(b), the contact angle increased to ~157° when the time was increased to 30 minutes. This increase, along with the low standard deviation, was attributed to the PU-MTCS molecules forming a more regular, homogeneous, and dense layer on the surface. This suggests that the surface energy was minimized, leading to minimized water contact and the achievement of superhydrophobic properties. However, when the coating time was extended to 45 minutes, a decrease in contact angle to ~151° was observed, as shown in Fig. 7(c). This reduction, accompanied by slightly higher variability, can be explained by negative effects such as the formation of irregular structures on the surface, agglomeration, or chemical degradation due to excessive PU-MTCS deposition. Additionally, prolonged treatment may lead to rearrangement or deviation of silane molecules from the cured structure, further decreasing hydrophobic activity. Based on these findings, and considering the standard deviations, a coating time of 30 minutes was identified as the optimum treatment duration for achieving maximum hydrophobic performance on PU-MTCS coated surfaces.

Absorption Capacities

The oil absorption capacities of PU sponges coated with different silane agents were evaluated at various coating temperatures. Sponges coated with PU-MTCS, PU-DTMS, PU-PDMS and PU-ODTCS agents at 25 °C, 40 °C and 55 °C respectively, the oil absorption capacities of the sponge samples obtained were examined and the effects of both the coating temperature and the silane agent used on the performance were investigated Fig. 8. Sponges coated with PU- DTMS exhibited moderate absorption performance compared to other sponges and reached the highest value of ~52 g at 40 °C. This parameter suggests that PU-DTMS forms a relatively lower density or shorter chain hydrophobic layer on the surface. The lowest performance was observed for PU-PDMS coated sponges, where the absorption capacity remained low at ~45- 48 g at all temperatures. The low molecular weight structure of PU-PDMS and its limited interaction with the surface resulted in its inability to form a sufficiently thick or effective hydrophobic layer on the surface of the sponge. Sponges modified with PU-MTCS and PU- ODTCS reached the highest absorption capacities at all temperatures, demonstrating that these two agents formed strong hydrophobic properties on the sponge surface. Especially at 40 °C, the absorption capacities of the sponges coated with both PU-MTCS and PU-ODTCS were at the same levels and measured at~ 61 g. Although this indicates that 40 °C is the most suitable temperature range for these agents to bind effectively to the surface and form a uniform coating layer, the absorption capacity of the sponge coated with PU-MTCS at 25 °C was determined as 63 g, which is the absorption capacity with the best improvement. In general, although increasing the coating temperature caused small changes in absorption capacity, the best parameter was obtained at 25 °C in terms of improvement. These findings suggest that both the choice of silane agent and coating temperature are critical for the functionalization of sponge surfaces for degreasing applications.

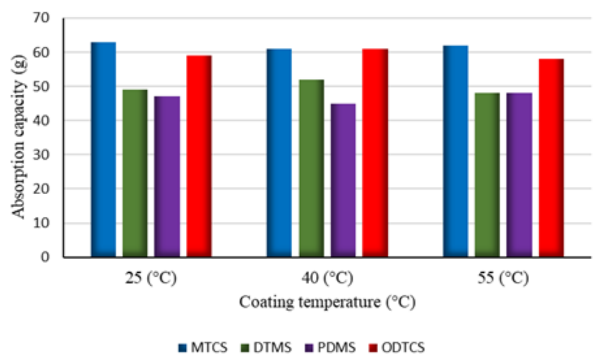


Figure 8 Absorption capacities of PU sponges produced at different coating temperatures.

PU sponges were coated with PU-MTCS and PU-ODTCS at different concentrations and the effects of the obtained surface modifications on the oil absorption capacity were evaluated. The absorption capacities obtained depending on the concentrations of 0.1%, 0.5% and 1% of silane agents applied to the sponge surface were analyzed Fig. 9. In sponges coated with PU-MTCS, a significant increase in absorption capacity was observed as the agent ratio increased. Especially when 1% of PU-MTCS was used, the absorption capacity reached approximately 81 g and the highest performance was achieved. This increase was attributed to the PU-MTCS forming a more effective hydrophobic layer on the surface, which allowed the sponge pores to remain open and the oil molecules to contact the surface more effectively. A different behavior was observed in sponges modified with PU-ODTCS. Coatings applied at 0.1% and 0.5% silane concentrations exhibited similar absorption capacities of approximately 61 g. However, increasing the silane content to 1% resulted in a decreased absorption capacity of about 55 g. This indicates that the long alkyl chains of PU-ODTCS form a dense and thick layer on the surface of the sponge at high concentrations, leading to clogging of the pores and thus limiting the penetration of oil into the sponge. When we look at the difference in agent ratio, high concentrations for PU-MTCS agent have an increasing effect on the absorption capacity, while the optimum ratio for PU-ODTCS can be determined as 0.5%. These findings reveal that not only the chemical structure of the silane agent used, but also the application concentration plays an important role on the surface properties and functional performance of the sponge.

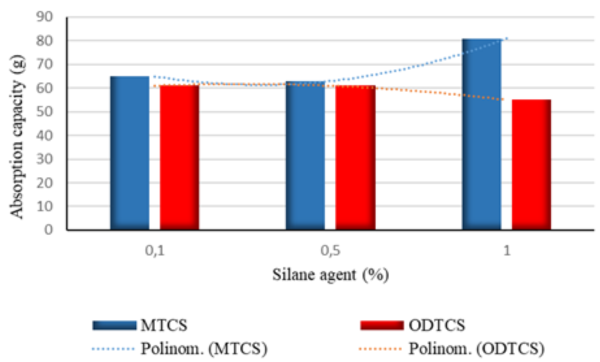


Figure 9 Effect of different coating ratios on absorption capacity.

Considering the effect of different durations of PU-MTCS coating on the oil absorption capacity, the sponges were coated by soaking in PU-MTCS solution for 15 minutes, 30 minutes and 45 minutes, respectively, and the absorption capacities obtained at the end of each coating time were compared Fig. 10. When the coating time was 15 minutes, the sponge~ absorbed 74 g of oil, showing a basic performance compared to the other times. This indicates that PU-MTCS has a limited interaction with the surface in a short time and cannot form a fully continuous hydrophobic layer.

When the coating time was increased to 30 minutes, the absorption capacity reached the maximum level and increased to ~81 g. This increase indicates that PU-MTCS molecules reacted with the sponge surface for sufficient contact time to form a more uniform and effective hydrophobic coating. However, when the time was extended to 45 minutes, the absorption capacity~ was determined as 79 g and a decrease was observed, indicating that partial blockages occurred due to excessive agent accumulation in the porous structure of the sponge. The prolonged coating formed a layer of irregular thickness on the surface, limiting the absorption of oil from the pores. When these data were analyzed, the coating time of 30 minutes was evaluated as the optimum coating time for PU-MTCS agent and was determined as the most suitable condition for both the efficiency of the surface coating and the functional performance of the sponge.

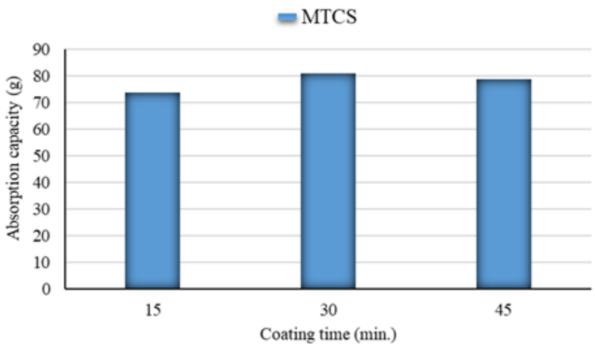


Figure 10 Effect of coating time on absorption capacity.

The parameter values of PU-MTCS, the coated sponge with the best improvement obtained as a result of coating different silane agents on PU sponge in the study, are as shown in Table 4 below.

Table 4 Parameters and results of absorption capacity of PU sponge coated with MTCS.

Parametre	Value
Silan Agent	MTCS
Concentration (%)	1
Temperature (°C)	25
Time (minutes)	30
Absorption Capacity (g)	81

The PU sponge sample coated with PU-MTCS silane agent reached the highest absorption capacity of 81 g under the conditions of 1% concentration, 25 °C temperature and 30 minutes coating time. This result shows that the high silane ratio and low temperature coating creates a more effective and regular hydrophobic structure on the sponge surface, thus providing maximum efficiency in the retention of petroleum-derived liquids.

CONCLUSION

In this study, the performance of superoleophilic PU sponges subjected to surface modification using different silane agents (PDMS, ODTCS, MTCS, DTMS) for the removal of oil spills was investigated in detail. The effects of parameters such as coating temperature, time and silane concentration on surface morphology, contact angle and absorption capacity were systematically investigated. In the experimental study, it is predicted that PU sponges coated with PU-PDMS and PU-DTMS silane agents will not be an ideal adsorbent for oil spills since the lowest values were obtained according to weight absorption capacity measurements. Nevertheless, two different silane agents used for PU-ODTCS sponge compared to PU-MTCS sponge were preferred to create hydrophobic surfaces with low surface energy. The PU-MTCS coating applied to the PU sponge surface is more effective than PU-ODTCS and reduces the wettability by reducing the surface energy more successfully. It is seen that the sponges prepared with PU-MTCS agent at 1% silane concentration, 25 °C temperature, and 30 minutes of coating time exhibited superior superhydrophobic/superoleophilic properties and achieved the highest oil absorption capacity of 81 g. This value significantly exceeds the oil absorption capacities reported for PU sponges modified with direct silane agents such as OTS and C18, which typically range around 25 g and between 60–70 g [30]. The absorption capacity of 81 g/g achieved in this study is notably high, as it is comparable to the range of 14.99 to 86.53 g/g reported for PU sponges modified with OTS in the literature [31]. The observed performance gain clearly demonstrates the effectiveness of the optimized MTCS coating conditions in promoting superoleophilic surface characteristics, thereby maximizing oil uptake. Consequently, this outstanding absorption performance positions MTCS-modified sponges as a highly promising and innovative material for efficient oil spill remediation. This study contributes to the development of reusable and high performance adsorbent materials for the effective and economical removal of oil spills, which pose a great environmental threat. In the literature, studies in this field generally focus on a single silane agent or examine the effect of coating parameters to a limited extent. In this context, this study makes an original contribution to the literature by systematically comparing a large number of silane agents and process parameters.

In the sectoral sense, such sponge-based adsorbents offer cost-effective and feasible solutions, especially for institutions and organizations operating in the maritime and waste management fields. Thanks to their easy manufacturability, low-cost raw materials and high recovery potential, such materials are suitable for future commercial applications.

In future studies, similar surface modification strategies can be applied to different polymer types to develop more comprehensive adsorbent materials. Furthermore, the reuse performance, selectivity and long-term durability of such sponges against different types of petroleum derivatives should also be investigated. Nanomaterial doped hybrid coatings or the use of environmentally friendly, biodegradable silane agents are also potential areas for future research in the literature.

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