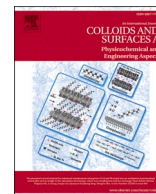




Contents lists available at ScienceDirect

Colloids and Surfaces A: Physicochemical and Engineering Aspects

journal homepage: www.elsevier.com/locate/colsurfa

A hydrophobic PMS-biochar-coated sponge sorbent for pre-purification of heavily oil-polluted waters

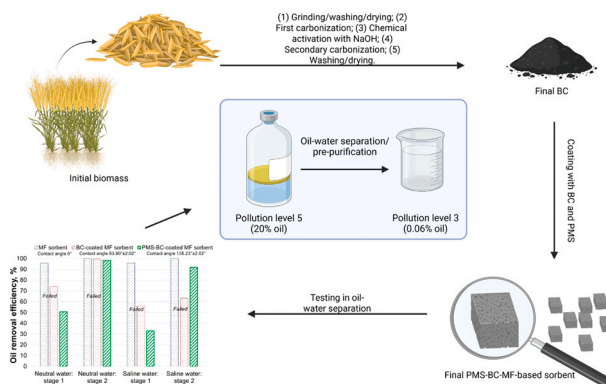
Aizhan Adylkanova^a, Makhabbat Kunarbekova^b, Aitolkyn Uali^{a,*}^a L.N. Gumilyov Eurasian National University, 13 Kazhymukan St., Astana 010000, Kazakhstan^b Satbayev University, 22a Satbayev St., Almaty 050026, Kazakhstan

HIGHLIGHTS

- Waste wheat has been transformed into biochar with a unique composition, molecular structure, and morphology texture.
- A hydrophobic PMS-biochar-coated sponge sorbent with a contact angle of $135.23^\circ \pm 2.02^\circ$ has been proposed to pre-purify heavily oil-polluted waters.

GRAPHICAL ABSTRACT

A hydrophobic PMS-biochar-coated sponge sorbent for pre-purification of heavily oil-polluted waters



ARTICLE INFO

Keywords:

Waste wheat
Biochar
Melamine sponge
Hydrophobic sorbent
Oil-water separation

ABSTRACT

This study primarily focuses on environmental concerns and protection by providing a beneficial use of waste biomass from crops and a low-cost polymer sponge to produce a low-cost and eco-friendly sorbent effective for oil-water separation. Research has indicated that waste wheat-derived biochar (BC), with a yield of 24.14 %, is predominantly composed of a graphite-like phase, featuring an advanced surface morphology and porous structure. This biochar can be combined with the hydrophobic agent polymethylsiloxane (PMS) to coat the melamine sponge, creating an effective sorbent for oil-water separation. The contact angle measurements indicate that once the sorbent surface is coated with BC and PMS, it becomes more hydrophobic, achieving a contact angle of approximately 135.23° . As the experimental study demonstrates, oil-contaminated water can be purified through a two-stage adsorption process with a chemisorption mechanism behind it, achieving total oil removal efficiencies of 99.13 % and 97.47 % from neutral and saline waters, respectively. The GC-MS analysis shows that while the first stage removes most non-polar and hydrophobic compounds, the second stage captures a higher proportion of moderately polar compounds. The impressive effectiveness of the relatively chemically, mechanically, and thermally stable PMS-BC-coated-MF-based sorbent in pre-purifying water from substantial oil contamination highlights its potential for addressing heavily polluted water sources. This finding reinforces the viability of sustainable approaches to oil spill remediation.

* Corresponding author.

E-mail address: uail_as@enu.kz (A. Uali).<https://doi.org/10.1016/j.colsurfa.2025.137154>

Received 11 January 2025; Received in revised form 28 April 2025; Accepted 6 May 2025

Available online 7 May 2025

0927-7757/© 2025 Elsevier B.V. All rights are reserved, including those for text and data mining, AI training, and similar technologies.

1. Introduction

The volume of oil extracted and consumed globally is staggering. Our dependence on oil intensifies as our economy evolves, making it imperative to address oil pollution and avert environmental catastrophe. Hundreds of oil spills occur daily, endangering our oceans and marine life. Marine oil spills involve catastrophic releases of oil, encompassing various types of crude oil, heavy and persistent fuel oil, lighter yet highly toxic fuel oil, and numerous processed products [1]. The sediment's chemical composition, the spilt oil's volumetric properties, and atmospheric reactions determine their fate, behaviour, and impact on the marine environment. Oil spills present significant concerns regarding substantial economic losses and long-term harm to aquatic ecosystems, local economies, and coastal communities [2]. Therefore, the environmental issues stemming from oil spills have emerged as one of the global challenges that the international community is addressing [3]. Numerous methods for combating oil spills include in-situ burning [4–6], mechanical recovery [7,8], solid sorbents [9], dispersion [10,11] and biodegradation [12]. However, these methods have limitations: they require time, entail high costs, and may cause adverse environmental effects. Adsorption is an effective method for removing organic pollutants due to its low cost and simplicity compared to traditional oil spill cleanup technologies [13,14].

Many sorbents have been developed for oil spill cleanup, including activated carbons [15,16], glass and cellulose fibres [17], hollow carbon fibres, organoclays, and nonwoven materials [18,19]. However, these sorbents still require improved hydrophobicity, adsorption capacity, and reusability in practical applications. Thus, developing a recyclable sorbent with high adsorption capacity, good hydrophobicity/lipophilicity, excellent buoyancy, and pressure resistance to combat oil spills is essential. Numerous 2D-3D-structured adsorbent materials have recently been developed for oil spill remediation, including porous rubber materials, nanotubes, aerogels, and others [20, 21]. However, research indicates these materials have drawbacks, such as low adsorption capacity, weak mechanical strength, and insufficient reusability. Melamine foam and polyurethane foam with 3D porous structures show promise as suitable sorbent materials due to their ability to absorb large amounts of liquid and their strength [22,23]. However, the surface of melamine foam is highly hydrophilic, hindering the separation of oil and water. Several methods exist to modify this surface to make it superhydrophobic, but developing environmentally friendly and affordable materials for this purpose remains a significant challenge.

Recent studies [24,25] have shown that biochar possesses excellent adsorption capability for organic and inorganic pollutants due to its large surface area, pore volume, and variety of functional groups. Compared to graphene, biochar (BC) is cost-effective, and its toxicity decreases with increasing pyrolysis temperature, thus holding the potential to replace graphene in adsorption processes. Various modification methods have been applied to enhance biochar's pollutant adsorption capacity, but complex processes and hazardous waste generation limit their widespread application. Therefore, the modification methods of biochar and the application of such materials are important directions for future research. This study aims to obtain biochar from local waste wheat and to design a hydrophobic carbonaceous melamine sponge-based sorbent for oil-water separation.

2. Materials and methods

2.1. Materials

All chemicals used in this study (NaOH, HCl, EtOH, NaCl, KCl, Na₂SO₄, MgSO₄, and CaCl₂) met analytical grade standards. Deionised (DI) water and 90 % ethanol (EtOH) were employed as solvents. Industrial diesel oil has a density of 9.1 kg/m³, and industrial melamine sponge has a density of 10 kg/m³. Polymethylsiloxane lubricant (PMS-100) was employed as a hydrophobic agent. In salt effect measurements,

to model natural seawater with its characteristic ionic balance, a solution with a pH of 8.33 was utilised, containing Cl[−] ions (19.87 g/L), Na⁺ ions (11.05 g/L), SO₄^{2−} ions (2.78 g/L), Mg²⁺ ions (1.32 g/L), Ca²⁺ ions (0.42 g/L) and K⁺ ions (0.40 g/L). NaOH and HCl solutions adjusted the pH to levels corresponding to seawater.

Benzene was employed as a solvent to prepare the engine oil sample by extracting it from an aqueous solution for gas chromatography-mass spectrometry (GC-MS) tests.

2.2. Synthesis of biochar

The biomass used in this study underwent multiple processing stages. The wheat husk, which served as the base material for this biochar, was subjected to primary carbonisation at T = 500 °C, v = 10 °C/min, for t = 1 h, in a nitrogen (N₂) atmosphere. Following the primary carbonisation, chemical activation with sodium hydroxide (NaOH) was conducted according to the procedure outlined in [26]. This was followed by secondary carbonisation at T = 600 °C, v = 10 °C/min, for t = 1 h, also in a nitrogen atmosphere. Firstly, this temperature regime was chosen to provide functional groups on the biochar's surface, enhancing its sorption capacity. Secondly, prior studies [27–30] indicated that higher temperatures reduce yield and lower oil sorption index. The total yield of biochar was 24.14 %.

2.3. Preparation of a sorbent

The superhydrophobic carbon-melamine sorbent was prepared following the procedure [31]. The biochar sample was mixed with diluted water at a 5 mg/mL ratio and subjected to ultrasonic treatment for 30 min to create a biochar suspension. Melamine sponges measuring 1 cm³ were washed with ethanol in an ultrasonic cleaner for 30 min and then rinsed with pure water for 20 min. After drying in an oven at 80 °C for 10 h, the melamine sponges were immersed in the AC suspension for 2 h of ultrasonication. The biochar-loaded melamine sponges were subsequently removed and dried in an oven at 80 °C for 10 h to produce biochar-wrapped melamine sponges. PMS was mixed with ethanol at an 8 mg/mL ratio to create a PMS/EtOH solution. The biochar-loaded melamine sponges were placed in the PMS/EtOH solution and subjected to triple release; they were then removed and subjected to a reaction at 160 °C for 12 h. The following three washes were done with a 90 % EtOH solution, and the material was dried in an oven at 60 °C for 10 h.

2.4. Material characterisation

A comprehensive analysis of the biochar sample was conducted using CHNS, BET, FTIR, Raman, XRD and SEM-EDX methods to determine its elemental composition and structural characteristics.

The CHNS analysis was conducted using a Unicube organic elemental analyser. The samples' ash content was determined by calculating them at 750 °C for five hours in a muffle furnace [32]. The oxygen content (%) was then calculated on a dry basis using the material balance equation: O_{subs} = 100 - (C + H + N + S + A), where C, H, N, S, and A represent the weight percentages of carbon, hydrogen, nitrogen, sulfur, and ash, respectively.

The BET surface area and pore parameters were measured using the N₂ adsorption-desorption isotherm, obtained at 77 K employing the BSD-660S A3 machine. Before the analysis, samples were degassed at 300 °C for 70 min.

The IR Tracer-100 (Shimadzu) machine collected spectra from 4000 cm^{−1} to 400 cm^{−1} with a resolution of 4 cm^{−1} and 75 scans. Before each measurement, the background was collected.

The Raman spectra were recorded from 1000 to 1800 cm^{−1} using a micro-Raman spectrometer (Horiba LabRam Evolution) with a 633 nm laser.

X-ray diffraction (XRD) analysis was conducted using a Bruker D8

Table 1
The elemental composition of biochar (CHNS analysis) and the ash content.

C, %	H, %	N, %	S, %	A, %	O _{subs} , %	Σ, %	H/C	O/C
72.77	1.70	3.51	0.04	15.00	6.98	100.00	0.023	0.096

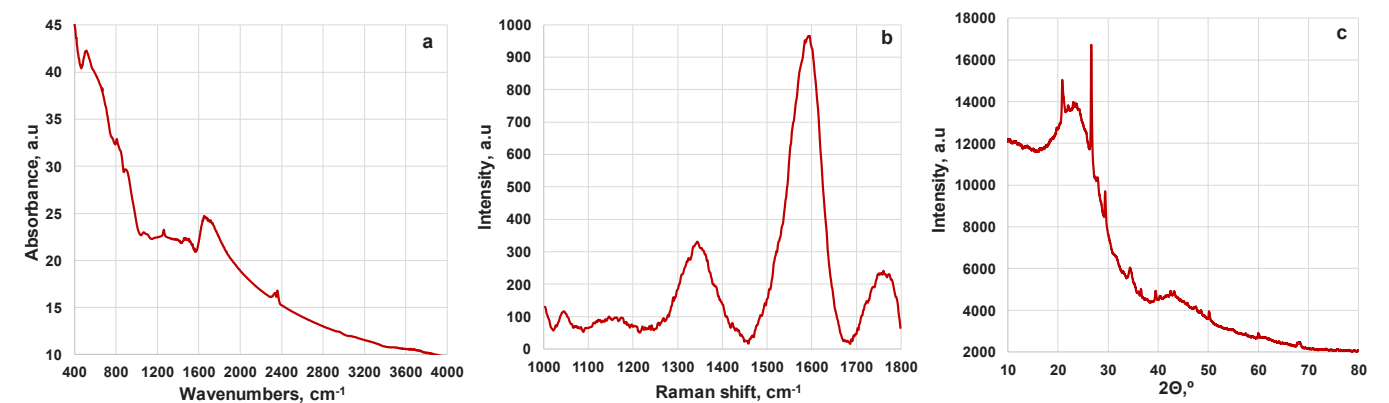


Fig. 1. Biochar's FTIR- (a) and Raman (b) spectra, X-ray diffractogram (c).

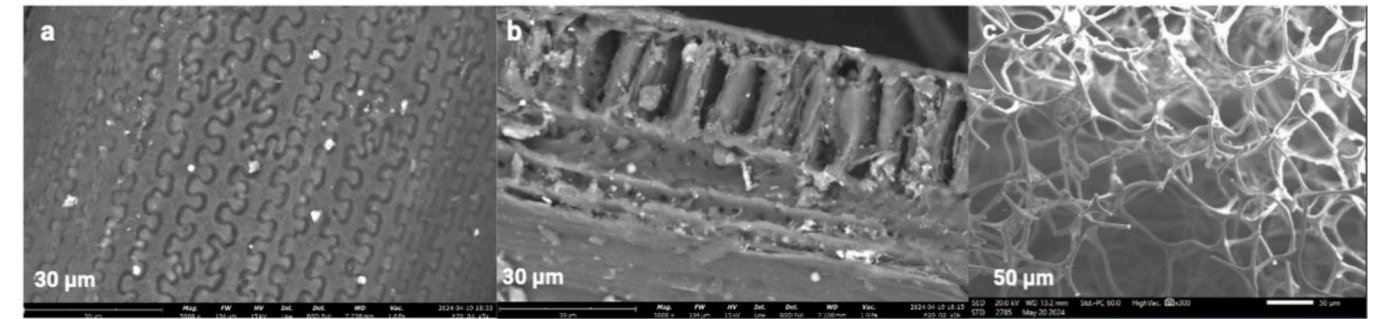


Fig. 2. SEM images of biochar's surface at 30 μm (a-b) and PMS-BC-coated MF sorbent (c).

Advance Eco diffractometer in a θ -2 θ scanning configuration at ambient temperature. CuK α radiation ($\lambda = 1.54060 \text{ \AA}$) served as the X-ray source. The sample was rotated at 15 rpm to reduce the texture effect. The diffraction patterns were recorded over a 2θ angle range from 10 to 80°,

with a step size of 0.05° and a collection time of 1.5 sec/step. Using the backscattered electron detector, the samples were imaged using SEM on a Scanning electron microscope JEOL JSM-IT200 (LA) at a high voltage of 20 kV.

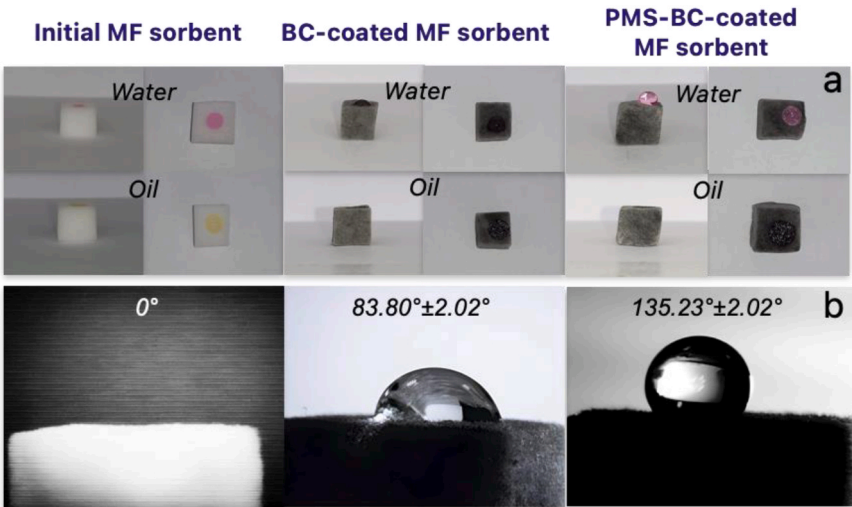


Fig. 3. Wettability of sorbents (a) and contact angles of water drop on the sorbent surface (b).

The thermal properties of the samples were studied using a Labsys Evolution TG-DTA/DSC (Setaram) machine in dynamic mode in the temperature range of 30–900°C while heating in an Al₂O₃ crucible at a rate of 10°C/min in a N₂ atmosphere with a flow rate of 30 mL/min.

The gas chromatography-mass spectrometry (GC-MS) analysis was conducted using the Clarus 600 gas chromatograph coupled with a mass spectrometer (Perkin-Elmer, USA) and an Elite-PONA capillary column (100 m x 0.25 mm x 0.50 µm) to analyse the qualitative content of hydrocarbons in the initial oil and oil extracts. The temperature gradient of the GC involved an initial isotherm of 40°C, followed by heating up to 280°C at a rate of 10 °C/min for 50 min (total analysis time: 50 min).

2.5. Contact angle measurements

The measurement setup employed a camera that recorded images of the drop to determine the contact angles of all the materials of interest. The contact angle measurements were then conducted using the ImageJ analysis software (DropSnake plugin).

2.6. Adsorption tests

Commercial engine oil 5W30 was chosen as a petroleum product to evaluate the material's uptake capacity. The adsorption tests were conducted in two stages, with the initial oil concentrations set at 20 % and 8.6 %, respectively. To study the adsorption kinetics, the adsorption values were calculated at various time points up to 24 h to construct the kinetic curves (at room temperature). The adsorption value was determined as follows (Eq. 1):

$$q_e = \frac{(C_0 - C_t)V}{m} \quad (1)$$

where q_t is the adsorption value [g/g]; C_0 and C_t are oil concentrations before and after adsorption in an emulsion under the study, respectively [g/L]; m is the adsorbent sample mass, taken for the single test [g]; V is the volume of the solution, where the adsorption takes place [L].

Different kinetic models were applied to evaluate adsorption kinetics based on these data.

2.7. Sorbent recyclability study

The stability of the sorbent was assessed by immersing the sample in an oil-water mixture for 5 min. Subsequently, the sample was removed and subjected to a pressure of 200 g for 5 min. It was reused multiple times as a sorbent and passed this test 10 times.

3. Results and Discussion

3.1. The characterisation of waste wheat-derived biochar

Based on the CHNS analysis and the ash content in biochar in this study, the oxygen content and the H/C and O/C ratios are evaluated (Table 1).

One of the characteristics of biochar is its O/C and H/C ratios [33]. The H/C ratio indicates the degree of aromaticity of the molecule: the higher the H/C ratio, the greater the aromaticity. The O/C ratio reflects the predominance of oxygen-containing functional groups in the molecule, such as carboxylic acids, alcohols, and ketones. Similarly, a higher O/C ratio corresponds to a greater prevalence of these functional groups, while a lower O/C ratio suggests a lesser prevalence [33,34]. In this instance, biochar exhibited low values for both the H/C and O/C ratios, confirming a limited content of aromatic and oxygen-containing groups [35–37].

According to the BET analysis, the specific surface area (SSA), total pore volume, and average pore diameter are 244.47 m²/g, 0.1367 mm³/g, and 2.24 nm, respectively. Regarding pore size distribution, the biochar exhibits 4.49 % macropores, 35.89 % mesopores, and 59.62 % micropores. Based on this data, it can be concluded that biochar is predominantly mesoporous [38]. This relatively low SSA is most likely due to the softer mode of chemical activation; KOH was substituted with a less aggressive NaOH at a low concentration and chosen to produce a more cost-effective and environmentally sustainable biochar.

The overall FTIR spectrum of biochar (Fig. 1a) corresponds to those reported in the literature [39–42].

Despite adsorption bands noted in [39,41,43] for biochar activated chemically slightly with sodium hydroxide, the FTIR spectrum did not exhibit those signals. In the current case, the adsorption peak at 2330 cm^{−1} corresponds to the aliphatic C-H functional group, as reported by [40,43]. Additionally, peaks at 1027 cm^{−1} and 871 cm^{−1} are assigned to aromatic C-H functional groups [39,44,45]. A relatively featureless (flat) FTIR pattern has been previously reported for different biochars that have undergone high thermal treatment with a temperature of 3 600°C [46,47]. These studies show that high-temperature carbonisation forms more aromatic and graphite-rich phases.

The Raman spectrum in Fig. 1b exhibits two distinct peaks labelled "D" and "G." These peaks are characteristic of carbon-based materials, and the position and intensity of the "G" peak indicate the degree of graphitisation. A higher "G" peak signifies a more crystalline and graphitic structure, typically at 1580 cm^{−1} [48,49]. The D band, positioned around 1330 cm^{−1}, reflects the presence of defects in the graphite structure of biochar [49]. In [50], based on the D band, it was suggested

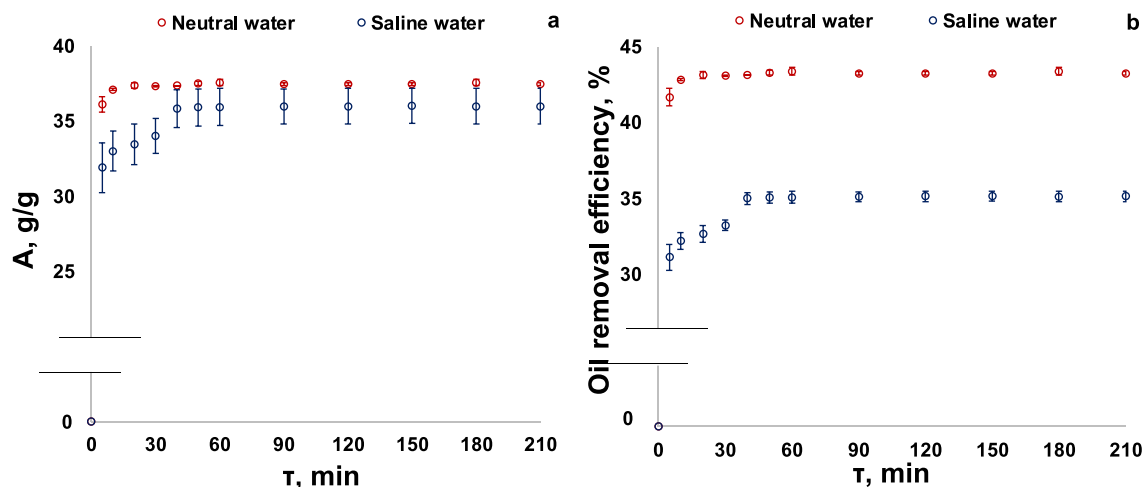


Fig. 4. Kinetic curves of oil sorption onto a PMS-BC-coated MF sorbent (a) and oil removal (b) during the first stage.

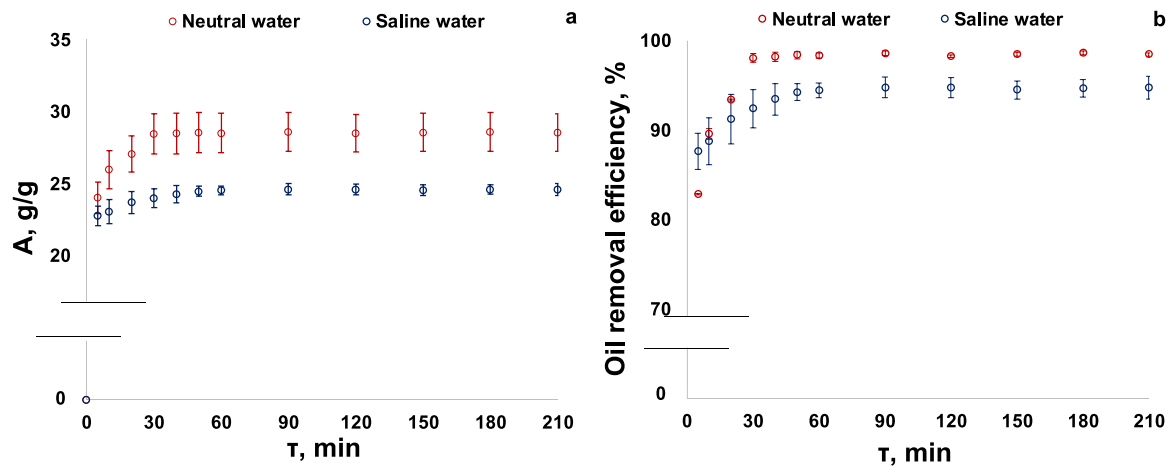


Fig. 5. Kinetic curves of oil sorption onto a PMS-BC-coated MF sorbent (a) and oil removal (b) during the second stage.

Table 2
Kinetic parameters of oil sorption from neutral water.

Kinetic model	The first stage			The second stage		
	k_1	q_e	R^2	k_1	q_e	R^2
PFO	0.0008	4.769	0.0547	0.0008	1.033	0.0025
PSO, type 1	k_2	q_e	R^2	k_2	q_e	R^2
	0.039	37.879	1	0.055	28.653	1
PSO, type 2	k_2	q_e	R^2	k_2	q_e	R^2
	0.136	37.594	0.926	0.036	28.011	0.974
PSO, type 3	k_2	q_e	R^2	k_2	q_e	R^2
	0.271	36.879	0.161	0.038	24.831	0.318
PSO, type 4	k_2	q_e	R^2	k_2	q_e	R^2
	0.137	37.63	0.921	0.034	28.897	0.966
Intraparticle diffusion	K_{Diff}	C	R^2	K_{Diff}	C	R^2
	0.0242	37.115	0.295	0.252	25.785	0.496
Elovich (linear)	α	β	R^2	α	β	R^2
	4.632	$2.3 \cdot 10^{73}$	0.774	0.945	$5.048 \cdot 10^9$	0.736

that aromatic systems with more than six connected rings exist, similar to those found in graphene sheets. A low I_D/I_G ratio of 0.34 indicates a graphitic material with minimal defects, which is advantageous for various advanced applications [50].

The XRD study showed that the biochar diffractogram corresponds with previously reported results [51–53]. The XRD spectrum exhibits two broad peaks at approximately 23° and 43° (in 2θ) (Fig. 1c), which correspond to the (002) plane of disordered graphite and the (100) plane, respectively. According to [26,52], the intensity of the (002)-assigned peak indicates a high degree of graphitisation in the biochar.

Figs. 2a–2b show SEM images of biochar. These images

demonstrate that the biochar surface has advanced, different-sized pores, which contribute to its increased sorption qualities.

Notably, an unusual, uniform distribution of patterned structures is observed on the surface of the biochar (Fig. 2a). The presence of macropores of varying sizes is also apparent (Fig. 2b). Furthermore, Fig. 2c presents an SEM image of the PMS-BC-coated MF-based sorbent, illustrating that the sponge possesses an intricate cavity morphology, which enhances its uptake capacity.

To summarise, all structural features of biochar—specifically its high carbon content, low H/C and O/C ratios, porosity, surface area, and aromatic domains—are essential for both single- and multi-functional applications. Due to its structure, feedstock availability, and low cost, biochar is highly regarded for its diverse uses across various fields. Its effectiveness in long-term carbon sequestration is attributed to its stability and structural integrity, which arise from a high degree of aromaticity and graphitisation, combined with low H/C and O/C ratios and a microporous structure [54]. For effective adsorption applications, a high specific surface area (SSA) with a well-developed micro-, meso-, and macroporous structure, along with functional groups, is critical, depending on the characteristics of the targeted substances [55,56]. Furthermore, biochar’s stability in both basic and acidic media and its other properties enhance its applicability in catalysis [57,58].

However, this study employs waste wheat-derived biochar as a binder that adheres PMS to the melamine surface. This transformation alters the material’s surface from hydrophilic to hydrophobic, thereby enhancing its affinity for non-polar components during oil-water separation.

Table 3
Kinetic parameters of oil sorption from saline water.

Kinetic model	The first stage			The second stage		
	k_1	q_e	R^2	k_1	q_e	R^2
PFO	0.0029	1.389	0.0379	0.001	1.250	0.512
PSO, type 1	k_2	q_e	R^2	k_2	q_e	R^2
	0.03153	35.336	1	0.0701	24.691	1
PSO, type 2	k_2	q_e	R^2	k_2	q_e	R^2
	0.03905	35.088	0.8313	0.0876	24.57	0.8866
PSO, type 3	k_2	q_e	R^2	k_2	q_e	R^2
	0.029	31.686	0.470	0.132	23.075	0.492
PSO, type 4	k_2	q_e	R^2	k_2	q_e	R^2
	0.0384	35.141	0.8168	0.0869	24.588	0.8817
Intraparticle diffusion	K_{Diff}	C	R^2	K_{Diff}	C	R^2
	0.292	31.816	0.6536	0.132	23.081	0.682
Elovich (linear)	α	β	R^2	α	β	R^2
	0.880	$2.6468 \cdot 10^{10}$	0.8347	1.9338	$1.998 \cdot 10^{18}$	0.8793

3.2. Wettability and contact angle measurements

Firstly, the pure melamine foam (MF) sponge and the biochar-loaded one exhibit similar abilities to absorb water and oil (Fig. 3).

However, the PMS-BC-coated MF-based sorbent did not absorb moisture like the previous two; when in contact with oil, the sponge treated with a hydrophobic agent absorbed oil more quickly than the original sponge. Contact angle measurements (Fig. 3) also confirm that from the first to the third sorbent, the hydrophobicity of the sorbent surface increases when the surface of the melamine sponge (0°) is treated with biochar ($83.80^\circ \pm 2.02^\circ$) and subsequently with a hydrophobic agent ($135.23^\circ \pm 2.02^\circ$).

3.3. Adsorption kinetics

Fig. 4 shows kinetic adsorption curves with an initial oil concentration of 20.00 %. The equilibrium was reached at an adsorption value of $A = 37.5$ g/g, and thereafter, the plateau exhibited little to no change. Although the experiment was extended for 24 h, the adsorption results remained relatively unchanged, indicating that the sorbent retains oil and does not desorb. The oil removal efficiency attained 43.24 % (Fig. 4b).

After the first treatment, the second dose of freshly prepared sponge sorbents was loaded into the intermediate oil-water mixture (with an oil concentration of 11.31 %). Equilibrium with A, equal to 28.44 g/g, was achieved in the first 30 min (Fig. 5a), and the oil removal efficiency was 98.46 % (Fig. 5b). In total, it has demonstrated a 99.13 % oil removal efficiency.

Comparing oil adsorption (g/g) and oil removal efficiency (%) from neutral and saline waters demonstrates that the latter case's efficiency is lower than the former (Figs. 4a-b, 5a-b). The adsorption tests showed that the oil removal efficiency decreased to 35.15 % in the first stage and 94.74 % in the second stage, for a total of 97.47 %. This phenomenon can be explained by the fact that in the presence of salts, the surface tension at the adsorbent-adsorbate interface increases as surface potential shifts; together, these factors lead to wettability alteration and reduced hydrophobicity [59,60]. Furthermore, salt ions can block the active sites on the sorbent's surface, decreasing the oil uptake capacity.

To understand the sorption kinetics more deeply, different kinetic models such as the pseudo-first-order (PFO) (Eq. 3) and pseudo-second-order (PSO) (eq-s 4–7 for the PSO types 1–4, respectively) models, as well as the intraparticle diffusion model (Eq. 8), the Elovich model (Eq. 9) were employed:

$$\ln(q_e - q_t) = \ln(q_e) - k_1 t \quad (3)$$

$$\frac{\tau}{q_\tau} = \frac{1}{k_2 q_e} + \left(\frac{1}{q_e}\right) \tau \quad (4)$$

$$\frac{\tau}{q_\tau} = \left(\frac{1}{k_2 q_e^2}\right) \left(\frac{1}{\tau}\right) + \left(\frac{1}{q_e}\right) \quad (5)$$

$$\frac{\tau}{q_\tau} = k_2 q_e^2 - k_2 q_e q_\tau \quad (6)$$

$$q_\tau = q_e - \left(\frac{1}{k_2 q_e}\right) \frac{q_\tau}{\tau} \quad (7)$$

$$q_\tau = K_{diff} \tau^{\frac{1}{2}} + C \quad (8)$$

$$q_\tau = \frac{1}{\alpha} \ln(\alpha\beta) + \frac{1}{\alpha} \ln \tau \quad (9),$$

where q_e and q_τ are adsorption values of oil in the sorbent at a particular time and in an equilibrium state [g/g]; k_1 [1/min] and k_2 [g/(g·min)] are the rate constants for PFO and PSO models, respectively; the first-order process speed and the rate constant, t is the time [min]; C is the interception coefficient [g/g], K_{diff} is the constant of the diffusion rate inside the particles [g/(g·min^{1/2})], α is the initial adsorption rate [g/(g·min)], and β is the desorption constant [g/g]. The calculation results are given in Tables 2–3.

Based on the experimentally observable results in Tables 2–3, the most suitable kinetic model is the PSO-1 due to its regression coefficient, which is nearly 1.00. In contrast, the PFO and PSO-3 models cannot be applied to the kinetic curves because of their relatively low R values, indicating that sorption is not limited by the diffusion properties of the sorbent itself and can proceed through chemical reactions and intermolecular bonding [61–63]. Likewise, the other two models, namely the intraparticle diffusion model and the Elovich model, have not demonstrated a strong correlation with experimentally measured adsorption kinetics. This can be explained by the fact that diffusion within the particles does not significantly influence the sorption rate; consequently, the sorption rate is not considerably affected by the occupancy of active centres in the sorbent [64]. Notably, the neutral-to-saline water replacement does not alter the kinetic model for oil sorption. Thus, it can be suggested that the sorption process is a complex pseudo-second-order reaction with chemisorption as the rate-limiting step. To compare, the PSO-behaved chemisorption on the oil sorption from water was also demonstrated by acetylated kapok fibres and leaf-based biowaste with the rate constants of 0.1246 g/(g·min) [65], and 2.338 g/(g·min) [66], respectively.

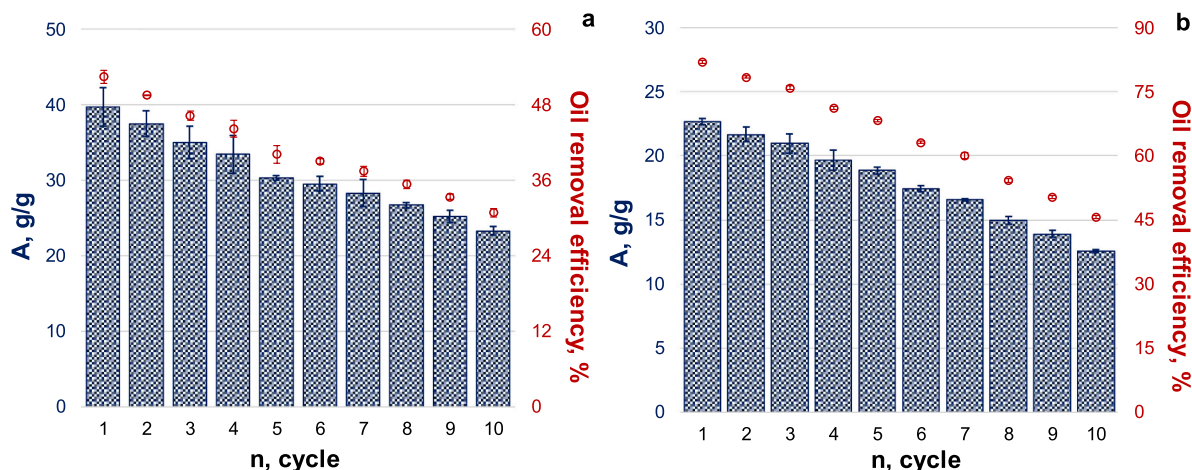


Fig. 6. Recyclability of the proposed sorbent for oil-neutral water separation in the first (a) and second (b) stages.

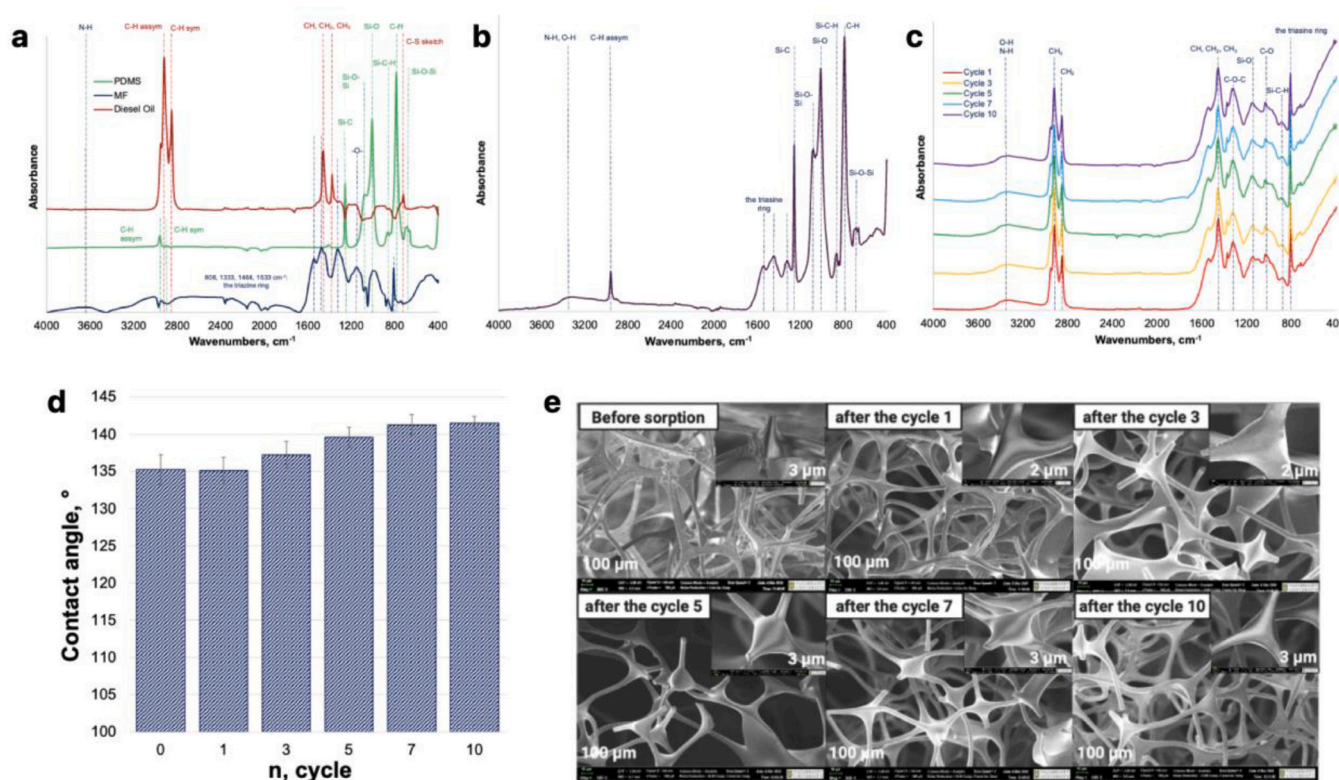


Fig. 7. FTIR spectra initial MF, PMS and engine oil (a), initial PMS-BC-MF-sponge based sorbent (b) and FTIR spectra after using the sorbent n-times for oil separation (c), the contact angles of the sorbent surface after n-times cycling (d), SEM images of the sorbent surface before the sorption and after n-times cycling (e).

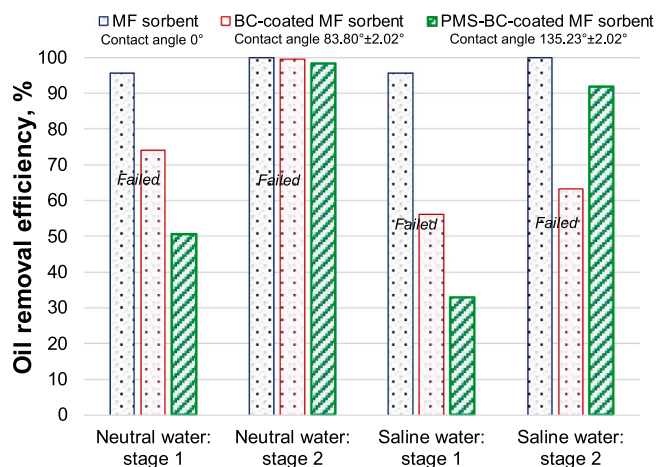


Fig. 8. Comparison of sorption abilities of different MF-based sorbents.

3.4. GC-MS study of initial engine oil and oil extracts

Table 1S (the supplemental file) demonstrates the GC-MS results for the qualitative content of initial engine oil and the contents of organic extracts after the first and second stages of oil adsorption from neutral water. The GC-MS results demonstrate that the initial engine oil contains a mix of hydrocarbons (alkanes, alkenes, esters, ketones, nitriles, etc.). Analysis of the oil extracts, separated after the first stage of compound detection, shows that most nonpolar or weakly polar hydrocarbons are absorbed [67]. Based on the oil extract content, which includes 12 compounds, the sorbent captures additional low-polarity but possibly smaller or more volatile compounds after the second stage.

A deeper analysis reveals that highly hydrophobic and non-polar hydrocarbons, such as alkanes and branched alkanes (isobutane, 2,2,4-

trimethyl-pentane, 2,2,3-trimethyl-hexane), are prime targets for PMS interaction due to their high hydrophobicity and molecular weight. The residual amount remaining in the second stage might be explained by diffusion limits or structural branching reducing initial adsorption.

Regarding the other alkenes and unsaturated hydrocarbons (3,5,5-trimethyl-1-hexene, 3-buten-2-one-propene), less polar and hydrophobic compounds are removed efficiently in the first stage. At the same time, those that remain are more volatile or oxygenated, persisting until the second stage. In the case of esters with moderate polarity (hexyl esters of acetic acid, esters of oxalic acid and propanoic acid), long-chain or bulky esters are removed in stage 1; however, some shorter-chain or branched representatives persist into stage 2, likely due to steric hindrance or partial hydrophilicity. Many polar carbonyl compounds, like ketones (3-pentanone) and diones (2,3-butanedione, 2,3-butanedione), are removed in the second stage, indicating weaker interaction with the sorbent and, most likely, contributing to slower sorption kinetics. Nitriles and nitroalkanes (butane nitrile, propane nitrile, nitropropane), which have moderate polarity and are toxic, persist into the second stage because their polarity weakens interaction with the hydrophobic sorbent. Another group of hydrophilic hydrocarbons, including alcohols (1-butanol) and polyols (3-buten-2-ol), are mostly not retained well, as they are polar and water-soluble, yet may still be removed in stage 2 via adsorption to biochar. Finally, aromatic and heterocyclic compounds, such as thiophene and 2H-Pyran-2-one, generally hydrophobic, interact through π - π stacking or dispersion with the sorbent and are removed early. Based on this separation pattern, it can be suggested that the PMS layer adsorbs non-polar hydrocarbons most effectively. Additionally, larger molecules are trapped more efficiently in terms of molecular size and structure; branching and aromaticity enhance van-der-Waals or π - π interactions. Biochar also appears to add porosity and adsorption for semi-polar compounds while also serving as a linker between PMS and MF.

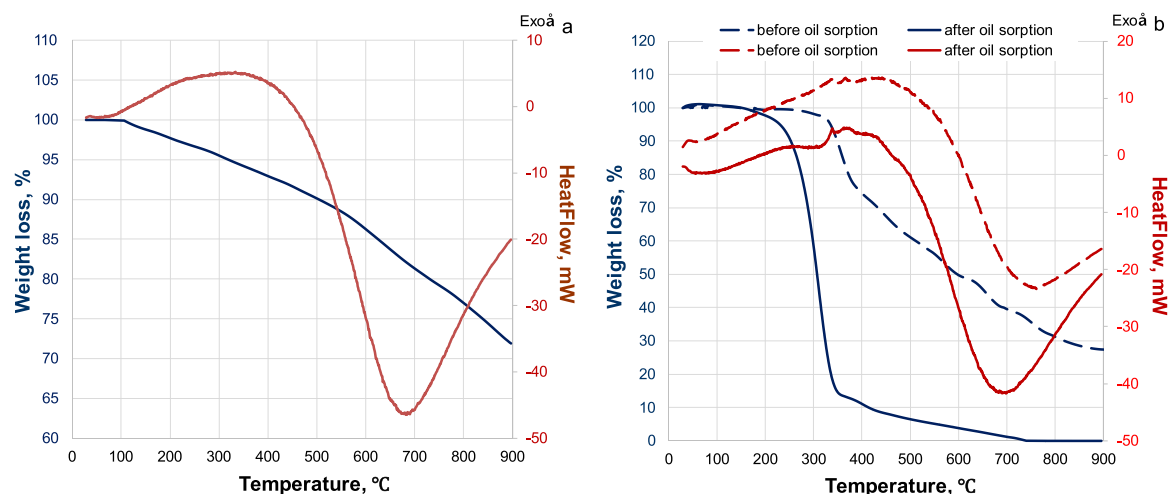


Fig. 9. TGA and DSC curves for initial biochar (a) and PMS-BC-coated MF sorbent before and after oil sorption (b).

Table 4

Characteristics of sorbents for oil-water separation.

Main platform Binder Hydrophobic coating agent	SSA of biochar/AC, m ² /g	Contact angle of the sorbent, °	Initial oil concent- ration, g/L	Initial sorption capacity, g/g Oil removal efficiency, % Recyclability, number of cycles	Ref.
Melamine Celery-derived AC Silicone	240	159.34	n.r.	54–143 n.r. 10	[80]
Melamine Wood chips-based AC Octadecylamine	n.r.	155	1:1 (oil/water in vol. ratio)	137 n.r. 11	[31]
Melamine Coconut shell-based AC Hexadecyltrimethoxy-silane	n.r.	159.53	1:2 (oil/water in vol. ratio)	69.04–158.27 > 98 50	[81]
P(St-EGD-DTD _x) foams No binder No hydrophobic agent	n.r.	153	1:99 (oil/water in vol)	49.6–112.8 > 98 10	[82]
P(St-EGD-DTD _x) foams No binder No hydrophobic agent	n.r.	153	1:99 (oil/water in vol)	49.6–112.8 > 98 10	[82]
Melamine Carbonised soot Dichloroethane	175.7	~145–150	3:35 (oil/water in mL)	~24 times its weight n.r. > 19 times	[83]
Layered double Ni-Co hydroxides (LDH) Wood chips-based Stearate	92.80	156.00	50	5.34–11.60 n.r. n.r.	[84]
Wood aerogel Wood Methyl-trimethoxy-silane	n.r.	151.80	n.r.	55.60–119.50 98.50 20	[85]
Melamine Waste wheat PMS	244.47	135.23	182	37.48/36.01 99.13/97.47 10 Neutral/ saline water	The present study

3.5. Recyclability of a hydrophobic PMS-BC-coated MF sorbent

The lifespan of a sorbent is ultimately defined by the degree to which its oil absorption capacity, selectivity towards target contaminants, and mechanical integrity deteriorate beyond established acceptable thresholds. An in-depth analysis of these parameters is necessary to accurately predict performance degradation under varying operational conditions, thus facilitating the development of more durable and efficient sorbent materials for practical applications. In this work, the analysis included a 5-minute oil absorption phase, followed by a 5-minute application of pressure to assist in oil removal. Observations indicated a decline in adsorption capacity with increasing reuse cycles (Fig. 6, a–b), suggesting a gradual reduction in the sorbent's effectiveness. In the initial trial, oil removal efficiencies were recorded at 53.51 % during the first stage of

oil purification and 82.10 % during the second stage of oil separation from water. However, the oil removal efficiency fell to 30.85 % and 45.59 % after the tenth use.

Fig. 7a presents the FTIR spectra of MF, PMS, and engine oil separately. All the FTIR spectrum patterns align with the reference ones [68–70]. A comparison with the initial PMS-BC-MF sorbent spectra (Fig. 7b) demonstrates that the sorbent is coated with PMS, as indicated by the presence of its functional groups. Furthermore, a broad signal centred at approximately 3400 cm⁻¹ can be attributed to the N-H and O-H groups. It is evident that after sorption, signals appear in the spectra confirming the presence of engine oil within the sorbent's structure, specifically related to the symmetric stretching of C-H in CH₂, the asymmetric deformation of the CH, CH₂, and CH₃ bonds, as well as the deformation of the C-O bond. Given that after the adsorption of engine

oil, no signals associated with PMS are detectable in the spectrum, and pronounced signals from hydrocarbons emerge (Fig. 7c), it can be inferred that the sorbent surface is saturated with engine oil. As the cycles increase, bands with nearly identical intensities are recorded while maintaining the same sorbent after sorption, pointing to the chemisorption mechanism behind the oil uptake.

This suggests that a residual quantity of engine oil remains within the sponge's bulk structure, diminishing the efficiency of the sponge sorbent for subsequent tests (cycles 3, 5, 7, 10) compared to the initial cycles. This observation, the presence of residual hydrocarbons in the sponge's bulk structure and surface, also correlates with the changes in the contact angles (Fig. 7d), illustrating a slight increase in the contact angle from cycle 1 to cycle 10. This aligns with the SEM results (Fig. 7e), demonstrating that the sorbent surface, coated with BC and PMS before the performance, is rough. However, when we move from the 1st cycle to the 10th one, the sorbent surface is still mechanically integrated (no cracks); only the roughness diminishes, and inter-fibre spaces in the melamine sponge structure increase. The same result has been reported in [71], where hydrophobic BC-based material recommended itself as a sorbent with mechanical abrasion resistance and chemical stability. Based on this, the sorbent of interest in the current study is a relatively chemically and mechanically stable composite material. At the same time, a decrease in oil uptake is most probably rooted in the adhesion failure at moving from the 1st to the 10th cycle using the same sorbent, which might be explained by the saturation of the sorbent with hydrocarbons (Fig. 7c) due to the chemisorption mechanism behind the oil sorption.

Comparative tests on oil sorption using a melamine sponge (MF sorbent), an intermediate sorbent (BC-coated MF sorbent), and a final sorbent (PMS-BC-coated MF sorbent) demonstrate that neither the melamine sponge nor the intermediate sorbent effectively targets petroleum and its derivatives (Fig. 8); they demonstrate both water and oil uptake, which makes them ineffective for oil separation from both neutral and saline waters.

In comparison, PMS-BC-coated MF sorbent demonstrates relatively high oil removal efficiency; however, substituting neutral water with saline water reduces the oil removal efficiency (30 min) from 50.67 % to 32.92 % and from 98.33 % to 91.82 % for the first and second stages, respectively.

3.6. Thermal analysis

Thermal analysis, including thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC), was performed to study the thermal stability and decomposition stages of initial biochar and MF sorbent before and after oil adsorption. The thermogravimetric curves of all samples show characteristic weight-loss stages corresponding to various thermal processes. For biochar (Fig. 9a), weight loss occurred smoothly and evenly over the entire temperature range, which indicates its high thermal stability compared to other components.

For the sorbent taken before the oil uptake (Fig. 9b), the initial mass loss (~30–300°C) was insignificant and was mainly associated with the evaporation of adsorbed moisture and residual volatile compounds. In this temperature range, all significant components remain stable. The main weight loss from 300°C to 400°C is due to the destruction of the methylene bridges of melamine, which releases gases such as formaldehyde and ammonia. In this same range, PMS decomposition begins, releasing CH₄, H₂, and other compounds [72–74]. Then, in the 400–1000°C range, further decomposition of sponge residues, biochar oxidation, and decomposition of PMS with the formation of SiO₂ and volatile compounds are observed [72]. For comparison, the heat resistance of dimethoxy(methyl)(3,3,3-trifluoropropyl)silane and trimethoxy(1 H,1 H,2 H,2 H-heptadecafluorodecyl)silane coatings are up to 400°C [75] and 650°C [76], respectively.

In the case of the sorbent, after oil adsorption (Fig. 9b), a slight initial mass loss (~30–220°C) is explained by the evaporation of light

hydrocarbons and the removal of moisture [77]. The primary decrease in weight in the range of 220–350°C is due to the thermal decomposition of melamine. Also, as mentioned before, PMS contributes to the thermal decomposition, releasing gases [72–74]. Weight loss occurs in the 350–800°C range due to the gradual decomposition of more stable components, like BC and PMS [78,79]. Besides this, the sorbent sample, before and after the oil uptake in the temperature ranges of 30–600°C and 30–450°C, respectively, predominantly exhibits exothermic thermal behaviour. However, a transition from exothermic to endothermic processes is observed at higher temperatures. As expected, the sorbent after oil sorption requires more energy for thermal decomposition, indicating altered thermo-oxidative properties. On the strength of these data, it might be suggested that the sorbent and its PMS-BC-coating keep their thermal stability in the operational temperature range for oil-water separation, usually an average of < 40°C.

3.7. Comparison with reference sponge-based sorbents

Table 4 summarises the average uptake capacities, removal efficiencies, and recyclability of hydrophobic sponge-based sorbents for oil-water separation: the reference sorbents compared to the one of our interest.

Table 4 indicates that nearly all sorbents can be characterised by (super)hydrophobic surfaces due to the high contact angles (>150°), resulting in a high sorption capacity and over 90 % oil removal efficiency. The recyclability of the sorbents, which varies from 5 to 50 cycles, is a critical indicator of their economic and environmental viability. In comparison, it has been shown that the PMS-BS-coated hydrophobic sorbent of interest, despite that it has relatively lower water contact angle compared to other alternatives, has effectively separated oil from heavily polluted oil-water systems, achieving a removal efficiency of up to 99.13 % and 97.47 % in neutral and model marine conditions, respectively.

Generally, oil pollution in water is categorised into different types based on the level of harm [86]. The first category is almost pure water, which has minimal effect. The second category, representing low pollution levels (oil concentration: from trace to 0.01 %), results in minimal chronic effects on marine life, such as slight impacts on growth and hatching rates. The third level, with oil concentrations ranging from 0.01 % to 0.1 %, corresponds to moderate pollution that noticeably affects aquatic organisms, posing potential long-term environmental risks. The fourth category entails high pollution levels (0.1–1 % oil), significantly harming marine life and causing acute toxicity and ecosystem disruption. The fifth level of severe pollution (over 1 % oil) indicates extreme environmental damage, leading to a high mortality rate among aquatic organisms and enduring ecological consequences. Based on the findings outlined above, the sorbent of interest in this study effectively purifies heavily oil-contaminated water (level 5) down to a moderate pollution level (level 3). According to [87], this water, which is moderately contaminated with oil, can be used in specific industrial processes that do not require highly purified water [88]. It may also be suitable for applications where additional filtration is permitted before use, such as irrigation for mining and quarrying, washing and processing ore, and dampening roads to control dust.

Regarding the commercial prototypes used in real-world emergency spill responses to oil spills, several options are available on the market. The first option is the 3 M™ Oil Sorbent [89], made from polypropylene inorganic sorbent with an oil uptake capacity of 8–12 g/g. Two other options include the modified-polyolefin-based OPEC™ Oil absorbent [90] and the sorbent known as «Zorbix®» [91] from modified polyolefin. These have been characterised by the same oil sorption capacity, roughly 12–18 g/g. To compare, the sorbent of interest seemed more effective, with approximately 36 g/g and 24.61 g/g oil uptake in the first and second stages, respectively. Based on this, it can be assumed that the studied sorbent is a promising material that is comparable to commercialised for sustainable oil spill remediation.

3.8. Other possible environmental applications for the sorbent

The literature analysis demonstrates that such hydrophobic carbonaceous material and/or sponge-based sorbents are highly recommended for concentrating heavy metals, organic compounds [92], toxic solvents [93], or pharmaceuticals [94] and for simultaneously capturing multiple contaminants [95,96] in complex wastewater. The PMS-biochar-coated sponge demonstrates versatility beyond just oil cleanup; it can also be utilised for various environmental applications, such as the adsorption of organic pollutants. However, effective water purification modelling requires advanced deep learning techniques, including prediction, control, sensing, and data analysis [97].

3.9. Recommendations on the spent sorbent disposal

Recycling oil-absorbing materials, such as sponges, is a complex process requiring environmentally friendly solutions. The most common method for managing traditional sponge-based oil absorbents is combustion. While this process eliminates petroleum products, it also produces emissions such as nitrogen oxides (NO_x), ammonia (NH₃), formaldehyde (HCHO), silicon dioxide (SiO₂), carbon dioxide (CO₂), polycyclic aromatic hydrocarbons, etc. To lessen their environmental impact, used sorbents, including oil-soaked sponges, are typically incinerated in specialised facilities [9,98]. First, sponges are collected and transported to processing facilities, where they undergo high-temperature burning under controlled conditions. This process helps completely decompose hydrocarbons and prevents the formation of harmful byproducts.

4. Conclusion and perspectives

This study has developed an inexpensive and easy-to-make, relatively chemically, mechanically, and thermally stable PMS-BC-coated MF sorbent for oil spill remediation, combining a melamine foam (MF) matrix, biochar (BC), and polydimethylsiloxane (PMS). The contact angle of the sorbent is found to be $135.23^\circ \pm 2.02^\circ$, which provides an efficient oil separation performance from heavily polluted neutral and saline (model) waters. Experimental results indicate this renewable sorbent's remarkable chemisorption-driven capacity for absorbing oil products. The GC-MS analysis shows that while the first stage removes most non-polar and hydrophobic compounds, the second stage captures a more significant number of moderately polar compounds. The proposed two-stage method significantly transformed heavily polluted water into water with moderate oil contamination, achieving total oil removal efficiencies of 99.13 % from neutral medium and 97.47 % from saline medium. This makes it suitable for pre-oil-water separation and contributes to the growing field of sustainable solutions for environmental oil cleaning-up challenges. However, much remains to be learned regarding the practical aspects of the sorbent to understand the mechanism and optimise its operation. Further investigation is needed, including melamine-biochar and biochar-PMS interfaces regarding tensile adhesion tests, the pore size distribution of biochar and oil adsorption kinetics and thermodynamics of the sorption. In addition, the investigation into the contact angle of the sorbent of interest under various experimental conditions, including temperature, humidity, and the oil type, as well as surface energy measurements and zeta potential analyses, is necessary. Moreover, in the future, artificial intelligence and machine learning may enhance the design of sorbents for oil-water separation.

CRediT authorship contribution statement

Adykanova Aizhan: Writing – original draft, Validation, Investigation, Formal analysis. **Kunarbekova Makhabbat:** Writing – review & editing, Investigation. **Uali Aitolkyn:** Writing – review & editing, Validation, Methodology, Formal analysis, Conceptualization.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

The authors thank Dr. Gulnur Tatykhanova (Laboratory of Engineering, Satbayev University) for conducting the GC-MS analysis.

Notes

A graphical abstract was created with BioRender.com (<https://BioRender.com/o21p689>).

Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at [doi:10.1016/j.colsurfa.2025.137154](https://doi.org/10.1016/j.colsurfa.2025.137154).

Data availability

Data will be made available on request.

References

- [1] B. Zhang, E.J. Matchinski, B. Chen, X. Ye, L. Jing, K. Lee, Marine oil spills-oil pollution, sources and effects, : World Seas: Environ. Eval. Vol. III: Ecol. Issues Environ. Impacts (2018) 391–406, <https://doi.org/10.1016/B978-0-12-805052-1.00024-3>.
- [2] C.H. Oppenheimer, Chapter 1 oil ecology, Elsevier Oceanogr. Ser. 27 (1980) 21–35, [https://doi.org/10.1016/S0422-9894\(08\)71378-3](https://doi.org/10.1016/S0422-9894(08)71378-3).
- [3] A. Carpenter, Oil pollution in the North Sea: the impact of governance measures on oil pollution over several decades, Hydrobiologia 845 (2019) 109–127, <https://doi.org/10.1007/s10750-018-3559-2>.
- [4] A.O. Ekperusi, A.P. Onyena, M.Y. Akpudo, C.C. Peter, C.O. Akpoduado, O. H. Ekperusi, In-situ burning as an oil spill control measure and its effect on the environment, : Soc. Pet. Eng. - SPE Niger. Annu. Int. Conf. Exhib. 2019, NAIC 2019 (2019) 1–11, <https://doi.org/10.2118/198777-MS>.
- [5] J.V. Mullin, M.A. Champ, Introduction/overview to in situ burning of oil spills, Spill Sci. Technol. Bull. 8 (2003) 323–330, [https://doi.org/10.1016/S1353-2561\(03\)00076-8](https://doi.org/10.1016/S1353-2561(03)00076-8).
- [6] J. Fritt-Rasmussen, S. Wegeberg, P. Lassen, L.B. Wilms, L. Renvald, M.B. Larsen, O. Geertz-Hansen, J. Wiktor, K. Gustavson, Coastline in-situ burning of oil spills, analysis of a Greenland field experiment, J. Hazard Mater. 441 (2023) 129976, <https://doi.org/10.1016/j.jhazmat.2022.129976>.
- [7] D.S. Etkin, T.J. Nedwed, Effectiveness of mechanical recovery for large offshore oil spills, Mar. Pollut. Bull. 163 (2020) 1–15, <https://doi.org/10.1016/j.marpolbul.2020.111848>.
- [8] V. Broje, A.A. Keller, Improved mechanical oil spill recovery using an optimized geometry for the skimmer surface, Environ. Sci. Technol. 40 (2006) 7914–7918, <https://doi.org/10.1021/es061842m>.
- [9] A.T. Hoang, X.P. Nguyen, X.Q. Duong, T.T. Huynh, Sorbent-based devices for the removal of spilled oil from water: a review, Environ. Sci. Pollut. Res. 28 (2021) 28876–28910, <https://doi.org/10.1007/s11356-021-13775-z>.
- [10] S. Kleindienst, J.H. Paul, S.B. Joye, Using dispersants after oil spills: Impacts on the composition and activity of microbial communities, Nat. Rev. Microbiol. 13 (2015) 388–396, <https://doi.org/10.1038/nrmicro3452>.
- [11] H. Imanian, M. Kolahdoozan, A.R. Zarrati, Vertical dispersion in oil spill fate and transport models, J. Hydrosol. Environ. 1 (2017) 21–33, <https://doi.org/10.22111/JHE.2017.3355>.
- [12] L. Vergeynst, S. Wegeberg, J. Aamand, P. Lassen, U. Gosewinkel, J. Fritt-Rasmussen, K. Gustavson, A. Mosbech, Biodegradation of marine oil spills in the arctic with a greenland perspective, Sci. Total Environ. 626 (2018) 1243–1258, <https://doi.org/10.1016/j.scitotenv.2018.01.173>.
- [13] N. Haridharan, D. Sundar, L. Kurrupasamy, S. Anandan, C.H. Liu, J.J. Wu, Oil spills adsorption and cleanup by polymeric materials: a review, Polym. Adv. Technol. 33 (2022) 1353–1384, <https://doi.org/10.1002/pat.5636>.
- [14] E.C. Emenike, J. Adeleke, K.O. Iwuozor, S. Ogunniyi, C.A. Adeyanju, V.T. Amusa, H.K. Okoro, A.G. Adeniyi, Adsorption of crude oil from aqueous solution: a review, J. Water Process Eng. 50 (2022) 1–15, <https://doi.org/10.1016/j.jwpe.2022.103330>.
- [15] U. Anwana Abel, G. Rhoda Habor, O. Innocent Oseribho, Adsorption studies of oil spill clean-up using coconut coir activated carbon (CCAC), Am. J. Chem. Eng. 8 (2020) 36–47, <https://doi.org/10.11648/j.ajche.20200802.11>.
- [16] T.K. Das, S. Ganguly, S. Ghosh, S. Remanan, S.K. Ghosh, N.C. Das, In-situ synthesis of magnetic nanoparticle immobilized heterogeneous catalyst through mussel

- mimetic approach for the efficient removal of water pollutants, *Colloid Interface Sci. Commun.* 33 (2019) 100218, <https://doi.org/10.1016/J.COLCOM.2019.100218>.
- [17] C.J. Singh, S. Mukhopadhyay, R.S. Rengasamy, A sustainable approach to oil spill cleanup by kapok and waste cotton needle punched nonwoven blends, *Ind. Crops Prod.* 191 (2023) 1–17, <https://doi.org/10.1016/j.indcrop.2022.115939>.
- [18] S. Mantripragada, S. Gbewonyo, D. Deng, L. Zhang, Oil absorption capability of electrospun carbon nanofibrous membranes having porous and hollow nanostructures, *Mater. Lett.* 262 (2020) 1–11, <https://doi.org/10.1016/j.matlet.2019.127069>.
- [19] L. Zang, Z. Bu, L. Sun, Y. Zhang, Hollow carbon fiber sponges from crude catkins: an ultralow cost absorbent for oils and organic solvents, *RSC Adv.* 6 (2016) 48715–48719, <https://doi.org/10.1039/c6ra08183e>.
- [20] M.A. Riaz, G. McKay, J. Saleem, 3D graphene-based nanostructured materials as sorbents for cleaning oil spills and for the removal of dyes and miscellaneous pollutants present in water, *Environ. Sci. Pollut. Res.* 24 (2017) 27731–27745, <https://doi.org/10.1007/s11356-017-0606-x>.
- [21] T.K. Das, S. Basak, S. Ganguly, 2D nanomaterial for microplastic removal: a critical review, *Chem. Eng. J.* 492 (2024) 152451, <https://doi.org/10.1016/J.CEJ.2024.152451>.
- [22] Y. Guan, F. Cheng, Z. Pan, Superwetting polymeric three dimensional (3D) porous materials for Oil/Water separation: A review, *Polym. (Basel)* 11 (2019) 1–34, <https://doi.org/10.3390/polym11050806>.
- [23] J. Pinto, A. Athanassiou, D. Fragouli, Surface modification of polymeric foams for oil spills remediation, *J. Environ. Manag.* 206 (2018) 872–889, <https://doi.org/10.1016/j.jenvman.2017.11.060>.
- [24] J.U. Ani, K.G. Akpomie, U.C. Okoro, L.E. Aneke, O.D. Onukwuli, O.T. Ujam, Potentials of activated carbon produced from biomass materials for sequestration of dyes, heavy metals, and crude oil components from aqueous environment, *Appl. Water Sci.* 10 (2020) 2701–2714, <https://doi.org/10.1007/s13201-020-1149-8>.
- [25] O.N. Ngofa, E.V. Liakos, A.N. Papadopoulos, G.Z. Kyzas, Activated carbon from bamboo and banana wood fibers as adsorbent materials for the removal of oil samples, *Biointerface Res Appl. Chem.* 12 (2022) 2701–2714, <https://doi.org/10.33263/BRIAC122.27012714>.
- [26] D. Hsu, C. Lu, T. Pang, Y. Wang, G. Wang, Adsorption of ammonium nitrogen from aqueous solution on chemically activated biochar prepared from sorghum distillers grain, *Appl. Sci.* 9 (2019) 5249, <https://doi.org/10.3390/app9235249>.
- [27] S. Zeng, E. Kan, E. Kan, Chemical activation of forage grass-derived biochar for treatment of aqueous antibiotic sulfamethoxazole, *ACS Omega* 5 (2020) 13793–13801, <https://doi.org/10.1021/acsomega.0c00983>.
- [28] Y. Wei, H. Qi, X. Gong, S. Zhao, Specially wetttable membranes for oil–water separation, *Adv. Mater. Interfaces* 5 (2018) 1–27, <https://doi.org/10.1002/admi.201800576>.
- [29] H.N. Nguyen, J.J. Pignatello, Laboratory tests of biochars as absorbents for use in recovery or containment of marine crude oil spills, *Environ. Eng. Sci.* 30 (2013) 374–380, <https://doi.org/10.1089/ees.2012.0411>.
- [30] K. Crombie, O. Mašek, S.P. Sohi, P. Brownsort, A. Cross, The effect of pyrolysis conditions on biochar stability as determined by three methods, *GCB Bioenergy* 5 (2012) 122–131, <https://doi.org/10.1111/gcbb.12030>.
- [31] H. Duan, H. Lyu, B. Shen, J. Tian, X. Pu, F. Wang, X. Wang, Superhydrophobic-superoleophilic biochar-based foam for high-efficiency and repeatable oil-water separation, *Sci. Total Environ.* 780 (2021) 1–11, <https://doi.org/10.1016/j.scitotenv.2021.146517>.
- [32] R.R. Domingues, P.F. Trugilho, C.A. Silva, L.C.N.A. De Melo, L.C.A. Melo, Z. M. Magriotis, M.A. Sánchez-Monedero, Properties of biochar derived from wood and high-nutrient biomasses with the aim of agronomic and environmental benefits, *PLoS One* 12 (2017) 1–9, <https://doi.org/10.1371/journal.pone.0176884>.
- [33] W. Bird, M. Keitel, C. Meredith, Analysis of biochars for C,H,N,O and S by elemental analyser, *Biochar: A Guide Anal. Methods* (2017) 39–50, (<https://nottin.gham-repository.worktribe.com/output/970299>) (accessed April 9, 2025).
- [34] S. Nanda, P. Mohanty, K.K. Pant, S. Naik, J.A. Kozinski, A.K. Dalai, Characterization of North American lignocellulosic biomass and biochars in terms of their candidacy for alternate renewable fuels, *Bioenergy Res* 6 (2013) 663–677, <https://doi.org/10.1007/s12155-012-9281-4>.
- [35] K. Jindo, H. Mizumoto, Y. Sawada, M.A. Sanchez-Monedero, T. Sonoki, Physical and chemical characterization of biochars derived from different agricultural residues, *Biogeosciences* 11 (2014) 6613–6621, <https://doi.org/10.5194/bg-11-6613-2014>.
- [36] M. Markič, Z. Kalan, J. Pezdič, J. Faganeli, H/C versus O/C atomic ratio characterization of selected coals in Slovenia, *Geologija* 50 (2007) 403–426, <https://doi.org/10.5474/geologija.2007.028>.
- [37] X. Sun, R. Shan, X. Li, J. Pan, X. Liu, R. Deng, J. Song, Characterization of 60 types of Chinese biomass waste and resultant biochars in terms of their candidacy for soil application, *GCB Bioenergy* 9 (2017) 1423–1438, <https://doi.org/10.1111/gcbb.12435>.
- [38] F.M. Jais, C.Y. Chee, Z. Ismail, S. Ibrahim, Experimental design via NaOH activation process and statistical analysis for activated sugarcane bagasse hydrochar for removal of dye and antibiotic, *J. Environ. Chem. Eng.* 9 (2020) 1–41, <https://doi.org/10.1016/j.jece.2020.104829>.
- [39] D.G. Atinafu, B. Yeol Yun, Y. Uk Kim, S. Wi, S. Kim, Introduction of eicosane into biochar derived from softwood and wheat straw: Influence of porous structure and surface chemistry, *Chem. Eng. J.* 415 (2021) 1–16, <https://doi.org/10.1016/j.cej.2021.128887>.
- [40] Y. Liu, Z. He, M. Uchimiya, Comparison of biochar formation from various agricultural by-products using FTIR spectroscopy, *Mod. Appl. Sci.* 9 (2015) 246–253, <https://doi.org/10.5539/mas.v9n4p246>.
- [41] H. Xu, B. Wang, R. Zhao, X. Wang, C. Pan, Y. Jiang, X. Zhang, B. Ge, Adsorption behavior and performance of ammonium onto sorghum straw biochar from water, *Sci. Rep.* 12 (2022) 1–11, <https://doi.org/10.1038/s41598-022-08591-5>.
- [42] B. Song, M. Chen, L. Zhao, H. Qiu, X. Cao, Physicochemical property and colloidal stability of micron- and nano-particle biochar derived from a variety of feedstock sources, *Sci. Total Environ.* 661 (2019) 685–695, <https://doi.org/10.1016/j.scitotenv.2019.01.193>.
- [43] X. Sun, H. Fu, M. Bao, W. Liu, C. Luo, Y. Li, Y. Li, J. Lu, Development of a new hydrophobic magnetic biochar for removing oil spills on the water surface, *Biochar* 4 (2022) 1–17, <https://doi.org/10.1007/s42773-022-00184-9>.
- [44] A. Kumar, T. Bhattacharya, Removal of arsenic by wheat straw biochar from soil, *Bull. Environ. Contam. Toxicol.* 108 (2022) 415–422, <https://doi.org/10.1007/s00128-020-03095-2>.
- [45] T.J. Purakayastha, S. Kumari, H. Pathak, Characterisation, stability, and microbial effects of four biochars produced from crop residues, *Geoderma* 239–240 (2015) 293–303, <https://doi.org/10.1016/j.geoderma.2014.11.009>.
- [46] R.R. Nair, M.M. Mondal, D. Weichgrebe, Biochar from co-pyrolysis of urban organic wastes—investigation of carbon sink potential using ATR-FTIR and TGA, *Biomass—Convers. Biorefin* 12 (2022) 4729–4743, <https://doi.org/10.1007/s13399-020-01000-9>.
- [47] S. Li, G. Chen, Thermogravimetric, thermochemical, and infrared spectral characterization of feedstocks and biochar derived at different pyrolysis temperatures, *Waste Manag.* 78 (2018) 198–207, <https://doi.org/10.1016/j.wasman.2018.05.048>.
- [48] A.M. Khalil, H. Msaadi, W. Sassi, I. Ghanmi, R. Pires, L. Michely, Y. Snoussi, A. Chevillot-Biraud, S. Lau-Truong, M.M. Chehimi, Facile diazonium modification of pomegranate peel biochar: a stupendous derived relationship between thermal and Raman analyses, 1519–529, *Carbon Lett.* 32 (2022), <https://doi.org/10.1007/s42823-022-00374-2>.
- [49] D. Torsello, G. Ghigo, M. Giorcelli, M. Bartoli, M. Rovere, A. Tagliaferro, Tuning the microwave electromagnetic properties of biochar-based composites by annealing, *Carbon Trends* 4 (2021) 1–9, <https://doi.org/10.1016/j.cartre.2021.100062>.
- [50] G.M. Bhalarao, A.K. Sinha, V. Sathe, Defect-dependent annealing behavior of multi-walled carbon nanotubes, *Phys. E Low. Dimens. Syst. Nanostruct.* 41 (2008) 54–59, <https://doi.org/10.1016/j.physe.2008.06.006>.
- [51] L. Cai, Y. Zhang, Y. Zhou, X. Zhang, L. Ji, W. Song, H. Zhang, J. Liu, Effective adsorption of diesel oil by crab-shell-derived biochar nanomaterials, *Materials* 12 (2019) 1–15, <https://doi.org/10.3390/ma12020236>.
- [52] N.M. Keppetipola, M. Dissanayake, P. Dissanayake, B. Karunarathne, M. A. Dourges, D. Talaga, L. Servant, C. Olivier, T. Toupance, S. Uchida, K. Tennakone, G.R.A. Kumara, L. Cojocar, Graphite-type activated carbon from coconut shell: a natural source for eco-friendly non-volatile storage devices, *RSC Adv.* 11 (2021) 2854–2865, <https://doi.org/10.1039/d0ra09182k>.
- [53] R. Siburian, H. Sihotang, S. Lumban Raja, M. Supeno, C. Simanjuntak, New route to synthesize of graphene nano sheets, *Orient. J. Chem.* 34 (2018) 182–187, <https://doi.org/10.13005/ojc/340120>.
- [54] L. Luo, J. Wang, J. Lv, Z. Liu, T. Sun, Y. Yang, Y.-G. Zhu, Carbon sequestration strategies in soil using biochar: advances, challenges, and opportunities, *Environ. Sci. Technol.* 57 (2023) 11357–11372, <https://doi.org/10.1021/acs.est.3c02620>.
- [55] J. Bayar, N. Ali, Y. Dong, U. Ahmad, M.M. Anjum, G.R. Khan, M. Zaib, A. Jalal, R. Ali, L. Ali, Biochar-based adsorption for heavy metal removal in water: a sustainable and cost-effective approach, *Environ. Geochem Health* 46 (2024) 428, <https://doi.org/10.1007/s10653-024-02214-w>.
- [56] J. Qu, Q. Meng, W. Peng, J. Shi, Z. Dong, Z. Li, Q. Hu, G. Zhang, L. Wang, S. Ma, Y. Zhang, Application of functionalized biochar for adsorption of organic pollutants from environmental media: synthesis strategies, removal mechanisms and outlook, *J. Clean. Prod.* 423 (2023) 138690, <https://doi.org/10.1016/J.JCLEPRO.2023.138690>.
- [57] H. Lyu, Q. Zhang, B. Shen, Application of biochar and its composites in catalysis, *Chemosphere* 240 (2020) 124842, <https://doi.org/10.1016/J.CHEMOSPHERE.2019.124842>.
- [58] M. Bartoli, M. Giorcelli, A. Tagliaferro, A comprehensive overview on biochar-based materials for catalytic applications, 1336 13, *Catalysts* 13 (2023) (2023) 1336, <https://doi.org/10.3390/CATAL13101336>.
- [59] L. Zeng, Y. Chen, M.M. Hossain, A. Saedi, Q. Xie, Wettability alteration induced water uptake in shale oil reservoirs: a geochemical interpretation for oil-brine-OM interaction during hydraulic fracturing, *Int. J. Coal Geol.* 213 (2019) 103277, <https://doi.org/10.1016/J.COAL.2019.103277>.
- [60] C.A. Franco, N.N. Nassar, F.B. Cortés, Removal of oil from oil-in-saltwater emulsions by adsorption onto nano-alumina functionalized with petroleum vacuum residue, *J. Colloid Interface Sci.* 433 (2014) 58–67, <https://doi.org/10.1016/J.JCIS.2014.07.011>.
- [61] M.A. Hashem, S. Payel, S. Mim, M.A. Hasan, M.S. Nur-A-Tomal, M.A. Rahman, M. I. Sarker, Chromium adsorption on surface activated biochar made from tannery liming sludge: A waste-to-wealth approach, *Water Sci. Eng.* 15 (2022) 328–336, <https://doi.org/10.1016/j.wse.2022.09.001>.
- [62] Y.S. Ho, G. McKay, Pseudo-second order model for sorption processes, *Process Biochem.* 34 (1999) 451–465, [https://doi.org/10.1016/S0032-9592\(98\)00112-5](https://doi.org/10.1016/S0032-9592(98)00112-5).
- [63] G. Wójcik, Z. Hubicki, Application of Pyrolox sorbent for vanadium(V) ions removal, *Physicochem. Probl. Miner. Process.* 58 (2022) 1–11, <https://doi.org/10.37190/PPMP/147974>.

- [64] O.A. Aworanti, S.E. Agarry, O.A. Aworanti, S.E. Agarry, Kinetics, isothermal and thermodynamic modelling studies of hexavalent chromium ions adsorption from simulated wastewater onto parkia biglobosa-sawdust derived acid-steam activated carbon, *Appl. J. Environ. Eng. Sci.* 3 (2017) 58–76, <https://doi.org/10.48422/IMIST.PRSM/aje-es-v3i1.7948>.
- [65] J. Wang, Y. Zheng, A. Wang, Investigation of acetylated kapok fibers on the sorption of oil in water, *J. Environ. Sci.* 25 (2013) 246–253, [https://doi.org/10.1016/S1001-0742\(12\)60031-X](https://doi.org/10.1016/S1001-0742(12)60031-X).
- [66] K.G. Akpomie, J. Conradie, Batch sorption of vegetable oil from simulated oil-polluted water onto *Populus nigra* leaf waste, *Int. J. Environ. Sci. Technol.* 20 (2023) 5377–5390, <https://doi.org/10.1007/s13762-022-04328-z>.
- [67] W. Basuki, K. Syahputra, A.T. Suryani, I. Pradipta, Biodegradation of used engine oil by acinetobacter junii TBC 1.2, *Indones. J. Biotechnol.* 16 (2015) 131–138, <https://doi.org/10.22146/ijbiotech.16374>.
- [68] Y. Wang, Z. Chen, Y. Lu, L. Yang, T. Xu, H. Wu, J. Zhang, L. He, A review of application, modification, and prospect of melamine foam, *Nanotechnol. Rev.* 12 (2023) 20230137, <https://doi.org/10.1515/ntrrev-2023-0137>.
- [69] M.A. Coronado, G. Montero, C. García, B. Valdez, R. Ayala, A. Pérez, Quality assessment of biodiesel blends proposed by the new Mexican policy framework, *Energ. (Basel)* 10 (2017) 631, <https://doi.org/10.3390/en10050631>.
- [70] M.G. Nespeca, R.R. Hatanaka, D.L. Flumignan, J.E. De Oliveira, Rapid and Simultaneous Prediction of Eight Diesel Quality Parameters through ATR-FTIR Analysis, *J. Anal. Methods Chem.* 2018 (2018) 1795624, <https://doi.org/10.1155/2018/1795624>.
- [71] M.E. Mohamed, O. Adel, E. Khamis, Fabrication of biochar-based superhydrophobic coating on steel substrate and its UV resistance, anti-scaling, and corrosion resistance performance, *Sci. Rep.* 13 (2023) 9453, <https://doi.org/10.1038/s41598-023-36589-0>.
- [72] Z. Cheng, K. Zheng, S. Zhou, Superhydrophobic silk fibroin-silica melamine sponge for efficient oil–water separation, *J. Porous Mater.* 29 (2022) 279–289, <https://doi.org/10.1007/s10934-021-01166-1>.
- [73] S. Ullah, M.A. Bustam, M. Nadeem, M.Y. Naz, W.L. Tan, A.M. Shariff, Synthesis and thermal degradation studies of melamine formaldehyde resins, *Sci. World J.* 2014 (2014) 1–6, <https://doi.org/10.1155/2014/940502>.
- [74] H.M. Ng, N.M. Saidi, F.S. Omar, K. Ramesh, S. Ramesh, S. Bashir, Thermogravimetric Analysis of Polymers, *Encycl. Polym. Sci. Technol.* (2018) 1–29, <https://doi.org/10.1002/0471440264.pst667>.
- [75] Y. Liu, Q. Lyu, Z. Wang, Y. Sun, C. Li, S. Sun, L.C. Lin, S. Hu, A flame-retardant post-synthetically functionalized COF sponge as absorbent for spilled oil recovery, *J. Mater. Sci.* 56 (2021) 13031–13042, <https://doi.org/10.1007/s10853-021-06006-w>.
- [76] B. Lin, J. Chen, Z.T. Li, F.A. He, D.H. Li, Superhydrophobic modification of polyurethane sponge for the oil-water separation, *Surf. Coat. Technol.* 359 (2019) 216–226, <https://doi.org/10.1016/j.surfcoat.2018.12.054>.
- [77] M. Kozak, A comparison of thermogravimetric characteristics of fresh and used engine oils, *Combust. Engines* 178 (2019) 289–292, <https://doi.org/10.19206/ce-2019-350>.
- [78] S. Elkhailifa, P. Parthasarathy, H.R. Mackey, T. Al-Ansari, O. Elhassan, S. Mansour, G. McKay, Biochar development from thermal TGA studies of individual food waste vegetables and their blended systems, *Biomass-- Convers. Biorefin* (2022) 1–8, <https://doi.org/10.1007/s13399-022-02441-0>.
- [79] T.H. Thomas, T.C. Kendrick, Thermal analysis of polydimethylsiloxanes. I. Thermal degradation in controlled atmospheres, *J. Polym. Sci. Part A-2: Polym. Phys.* 7 (1969) 537–549, <https://doi.org/10.1002/pol.1969.160070308>.
- [80] H. Shayesteh, M.S. Khosrowshahi, H. Mashhadimoslem, F. Maleki, Y. Rabbani, H.B. M. Emrooz, Durable superhydrophobic/superoleophilic melamine foam based on biomass-derived porous carbon and multi-walled carbon nanotube for oil/water separation, *Sci. Rep.* 13 (2023) 1–16, <https://doi.org/10.1038/s41598-023-31770-x>.
- [81] L. He, X. Qi, W. Wei, X. Zhang, J. Wang, Z. Gao, Biomass-activated carbon-based superhydrophobic sponge with photothermal properties for adsorptive separation of waste oil, *J. Hazard Mater.* 477 (2024) 135222, <https://doi.org/10.1016/J.JHAZMAT.2024.135222>.
- [82] Q. Zhang, K. Li, J. Li, Y. Li, Fabrication of hierarchically porous superhydrophobic polystyrene foam for self-cleaning, oil absorbent, highly efficient oil–water separation, *Chem. Eng. J.* 483 (2024) 149338, <https://doi.org/10.1016/J.CEJ.2024.149338>.
- [83] M. Nazhipkyzy, D. Assylkhanova, N. Araylim, A. Seitkazanova, G. Özsin, E.A. Varol, Effective separation of petroleum oil-water mixtures via flexible and re-usable hydrophobic soot-coated melamine sponge, *J. Water Process Eng.* 49 (2022) 103032, <https://doi.org/10.1016/J.JWPE.2022.103032>.
- [84] X. Pan, S. Kuang, X. Wang, Effective strategy for alleviation of oil contamination in marine via the superhydrophobic biochar: Performances, mechanisms and environmental recalcitrance, *J. Clean. Prod.* 462 (2024) 1–8, <https://doi.org/10.1016/J.JCLEPRO.2024.142695>.
- [85] Y. Zhu, H. Li, W. Huang, X. Lai, X. Zeng, Facile fabrication of superhydrophobic wood aerogel by vapor deposition method for oil-water separation, *Surf. Interfaces* 37 (2023) 102746, <https://doi.org/10.1016/j.surfint.2023.102746>.
- [86] I. Okafor-Yarwood, The effects of oil pollution on the marine environment in the Gulf Of Guinea—The bonga oil field example, *Transnatl. Leg. Theory* 9 (2018) 254–271, <https://doi.org/10.1080/20414005.2018.1562287>.
- [87] M. Fingas, C. Brown, Review of oil spill remote sensing, *Mar. Pollut. Bull.* 83 (2014) 9–23, <https://doi.org/10.1016/j.marpolbul.2014.03.059>.
- [88] K.K. Kesari, R. Soni, Q.M.S. Jamal, P. Tripathi, J.A. Lal, N.K. Jha, M.H. Siddiqui, P. Kumar, V. Tripathi, J. Ruokolainen, Wastewater treatment and reuse: a review of its applications and health implications, *Water Air Soil Pollut.* 232 (2021) 208, <https://doi.org/10.1007/s11270-021-05154-8>.
- [89] 3MTM Oil Sorbent Pillows | 3M United States. (https://www.3m.com/3M/en_US/p/d/b00020432/).
- [90] OPEC: OPEC Share of World Crude Oil Reserves (https://www.opec.org/opec_web/en/data_graphs/330.htm).
- [91] Zorbix | Diversified Minerals Inc. (<https://dmicement.com/products/absorbents/zorbix/>).
- [92] A.S. Khan, T.H. Ibrahim, M.I. Khamis, M.A. Jaoude, A. Nasrullah, Hydrophobic deep eutectic solvent-impregnated sponge for efficient phenol adsorption from aqueous media, *J. Mol. Liq.* 422 (2025) 126934, <https://doi.org/10.1016/J.MOLLIQ.2025.126934>.
- [93] Sh.K. Amerkhanova, R. Shlyapov, A.S. Uali, The active carbons modified by industrial wastes in process of sorption concentration of toxic organic compounds and heavy metals ions, *Colloids Surf. A Physicochem Eng. Asp.* 532 (2017) 36–40, <https://doi.org/10.1016/J.COLSURFA.2017.07.015>.
- [94] A.M. Turarbek, F.Zh Abilkanova, A.S. Uali, Development of novel wheat waste-derived biochar and its potential in pharmaceutical wastewater treatment, *EURASIAN J. Chem.* (2025) 76–88, <https://doi.org/10.31489/2959-0663/1-25-2>.
- [95] X. Lu, P. Wu, C. Chi, W. Shi, F. Liu, Y. Yang, H. Liu, J. Fu, A recyclable elastic foam composite with spatially separated adsorption sites for simultaneous capture of multiple contaminants in complex wastewater, *Sep Purif. Technol.* 354 (2025) 129551, <https://doi.org/10.1016/J.SEPUR.2024.129551>.
- [96] N. Erdeanna, B. Surat'man, X.L. Quek, N. Wang, E. Ye, J. Xu, Z. Li, B. Li, Sustainable nanofibrous membranes for air filtration, water purification and oil removal, *Nanoscale* 17 (2025) 6427, <https://doi.org/10.1039/d4nr04673k>.
- [97] M. Alvi, D. Batstone, C.K. Mbamba, P. Keymer, T. French, A. Ward, J. Dwyer, R. Cardell-Oliver, Deep learning in wastewater treatment: a critical review, *Water Res* 245 (2023) 120518, <https://doi.org/10.1016/J.WATRES.2023.120518>.
- [98] A. Patalano, F. Villalobos, P. Pena, E. Jauregui, C. Ozkan, M. Ozkan, Scaling sorbent materials for real oil-sorbing applications and environmental disasters, *MRS Energy Sustain.* 6 (2019) 1–13, <https://doi.org/10.1557/mre.2019.3>.