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Non-woven sorbent based on recycled jute fibers for efficient oil spill clean-up: From production to biodegradation

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ABSTRACT

Loose assemblies of cellulose fibers could be employed in the clean-up of oil spills, but the difficulty to separate them from the spill after use makes them impractical. In an effort to tackle this issue, a non-woven sorbent based on recycled jute fibers from the carpet industry was developed. To enhance the porosity and hydrophobicity/oleophilicity of the sorbent, carbonization in an inert atmosphere was carried out. A comparison in oil sorption performance between non-carbonized and carbonized sorbents was made by evaluating the oil capacity in a water medium, buoyancy, oil retention and reusability. Carbonization of sorbent resulted in more than doubled oil sorption capacity independent of oil viscosity. Oil viscosity did not affect the oil sorption capacity of non-carbonized sorbent. Carbonized sorbent showed superior buoyancy in water even after 24 hours, oil retention of approximately 60–80% after 3 hours and only 12–20% decline of oil sorption capacity after five repeated sorption/desorption trials. However, the ability of oiled non-carbonized sorbent to efficiently biodegrade in model compost (up to 45% weight loss after 10 weeks) makes it a sustainable candidate for oil spill clean-up.

1. Introduction

Oil spills often result in disasters with long-lasting environmental, social and economic impacts. One of the latest accidents happened in January 2022, when 1500 cubic meters of Bezios crude oil was spilled while the tanker was at a single buoy mooring (SBM) near the La Pampilla terminal in Peru (ITOPF, 2022). As reported, tens of kilometers of oiled sandy beaches and rocky shores required an urgent mobilization of many international entities to assist in making the strategy for oil clean-up. Generally, the clean-up of oil spills demands a quick and extensive response combining variety of remediation approaches. Various sorbents are used especially in the last phase of shoreline clean-up or for retrieving smaller oil pools (ITOPF, 2014). Oil sorbents should be inexpensive, simple to use on oiled foreshores or floating slicks, capable to absorb large amounts of oil, possess good retention, and degrade quickly with adhered oil (Hoskin et al., 2001).

Synthetic sorbents made of polyurethane, polyethylene, polypropylene, cross-linked polymers or rubber materials are very efficient in capturing oil due to hydrophobic/oleophilic nature and may be applied repeatedly, but their

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environmental impact is a major cause for concern. A poor degradation of synthetic polymers limits their disposal options. Additionally, if they are not entirely removed from the ocean, they may cause microplastic pollution (Hoskin et al., 2001). Recently, low-cost and easy-to-fabricate surface engineered sponges have emerged as efficient alternatives for oil–water separation (surface oil removal by absorption, immiscible oil–water mixture separation, oil microdroplet adsorption from wastewater) due to large oil capacity, excellent reusability and selectivity (Cherukupally et al., 2017, 2020, 2021, 2022). On the other side, agro-industrial residues such as banana and orange peel, sugarcane bagasse, rice husk, peat moss, saw dust, barley straw, or natural fibers such as cotton, jute, kapok, wool, coconut etc. are biodegradable, abundant and inexpensive (Choi and Kwon, 1993; Lim and Huang, 2007; Radetić et al., 2007; Husseien et al., 2008a; Dong et al., 2015; Thilagavathi et al., 2018; Thilagavathi and Karan, 2019; Asadpour et al., 2021; Vijayasekar and Saravanan, 2022; Cardoso et al., 2021; Hakeim et al., 2022; Wang et al., 2022; Singh et al., 2023) and have been examined as sorbents. These biosorbents can be disposed in many ways, including composting (Husseien et al., 2008b).

Cellulose and lignocellulose fibers have been extensively investigated as a valuable, abundant and inexpensive resource (Renuka et al., 2016; Thilagavathi and Karan, 2019; Vijayasekar and Saravanan, 2022). However, their relatively small oil sorption capacity and hydrophilicity are the most commonly reported drawbacks. Therefore, number of strategies have been applied to overcome these disadvantages. Silylation was utilized to convert the surface hydroxyl groups on the hydrophilic cotton fibers to alkylsilyl ethers forming the hydrophobic surfaces (Jarrah et al., 2018). Extremely large oil sorption capacities can be obtained by roughening kapok fibers with MoS_2 nanoparticles (Zhou et al., 2020). Functionalization of kapok fibers by acetylation, grafting, bio-adhesion, hydrothermal modification and pyrolysis for improvement of their oil sorption properties was also proposed (Zheng et al., 2021). Acetylation of jute fibers in the presence of N-bromosuccinimide as a catalyst resulted in good sorption of machine oil (Teli and Valia, 2013). Thermal treatment of cellulose materials such as carbonization is one of the most favorable methods to produce carbon-based sorbents and enhance their oil to water selectivity (Angelova et al., 2011; Bi et al., 2013; El Gheriany et al., 2020; Husseien et al., 2009, 2008b; Inagaki et al., 2002; Lam et al., 2018; Phan et al., 2006; Song et al., 2020).

In this study, an effort has been made to: (i) produce a cheap non-woven sorbent (NWS) made of recycled jute fibers, (ii) examine its oil sorption behavior; (iii) explore how the carbonization in inert atmosphere affects its oil sorption performance, and (iv) evaluate the biodegradation of oiled NWS in compost. Disposed carpet fringes made of jute fibers as a post-industrial waste were exploited for the production of NWS by needle punching. Taking into account that the most commonly spilled oils into water are crude oil (35%), diesel oil (20%) and marine oil (10%) (Zamparas et al., 2020), the oil sorption behavior (oil sorption capacity in water, oil retention, buoyancy, reusability) of fabricated NWS was evaluated by testing the crude oil, diesel oil and two motor oils of different viscosities.

2. Experimental

2.1. Materials

Oil sorbent was produced by a local company "Meteks" (Serbia). Carpet fringes made of jute with small polyamide (PA) patches were collected as a post-industrial waste from the carpet industry. Fibrous webs formed after garneting the waste were needle-punched to fabricate the NWS with the area density and the thickness of 800 g/m^2 and 6.3 $\pm$ 0.4 mm, respectively. The bursting strength of NWS is 380 $\pm$ 57 N. By performing a quantitative chemical analysis in accordance with ISO 1833-11: 2017-Textiles-Quantitative chemical analysis-Part 11: Mixtures of certain cellulose fibers with certain other fibers (method using sulfuric acid), the NWS was found to contain 90% of jute and 10% of PA fibers.

The carbonization of NWS was performed in a quartz furnace tube, in an inert atmosphere. Constant argon flow was maintained through the entire carbonization process. To produce carbonized NWS (CNWS) and retain the integrity of the sorbent, the pieces of NWS (6 cm $\times$ 6 cm), were put in the quartz furnace tube, and the temperature was raised to 650 °C at a heating rate of 10 °C/min. The samples were kept at 650 °C for 15 min, when the furnace was cooled down to room temperature.

Four types of oil were tested in this study: crude oil (NIS, Serbia, density at 15 °C: 0,8703 g/mL, kinematic viscosity at 40 °C: 10.87 mm/s^2), diesel oil (NIS, Serbia, density at 15 °C: 0,8307 g/mL, kinematic viscosity at 40 °C: 2.525 mm/s^2), commercial synthetic motor oil 1 (Nisotec autoline motor oil SINT SAE 5W-30, NIS, Serbia, density at 15 °C: 0,8526 g/mL, kinematic viscosity at 40 °C: 66.69 mm/s^2) and commercial mineral motor oil 2 (Nisotec autoline motor oil SF SAE 15W-40, NIS, Serbia, density at 15 °C: 0,8828 g/mL, kinematic viscosity at 40 °C: 113.2 mm/s^2). Keeping in mind that lighter-weight hydrocarbons evaporate in the early phases of an oil spill before any possible remediation activity, oils were stored in a fume hood for 48 h before the use.

2.2. Characterization of NWS and CNWS

The morphology of fiber surface of NWS and CNWS was analyzed by field emission scanning electron microscope (FESEM, Mira3 Tescan, Czech Republic). The samples were sputter coated with a thin layer of Au before the analysis. Obtained FESEM images are utilized for determination of jute fiber diameter using the open-access imaging software tool "ImageJ". EDX analysis was assessed by FESEM (FEI Scios 2 Dual Beam, ThermoFisher Scientific, Czech Republic).

Fourier transform infrared (FTIR) spectra of NWS and CNWS were recorded in the attenuated total reflectance (ATR) mode using a Nicolet iS5 FTIR Spectrometer (ThermoFisher Scientific, USA) at 4 cm^{-1} resolution, over a range of 500–4000 cm^{-1}.

The dynamic contact angle between water and the samples were measured using the Kruss Drop Shape Analyzer DSA100M (Hamburg, Germany, GmbH). The sterile syringe needles (NE 44, Kruss GmbH) was used for each new measurement. Kruss DSA100M uses an optical microscope and a digital camera (20 fps) which quickly takes images of the object and then uses a digital image processing algorithm to calculate the droplet's contact angle by tangents or Laplace–Young approximations. The temperature (22 $\pm$ 0.3 °C) and humidity (30 $\pm$ 5%) of the test chamber were controlled. For each sample, at least five successive measurements were taken.

2.3. Oil sorption testing of NWS and CNWS

The water uptake (q_w) and buoyancy of the sorbents were studied under static and dynamic conditions (Cooper and Keller, 1992; ASTM F726-99, 1999). In static test, 4 samples (5 cm ×5 cm) were left to stay 15 min (q_w-15 min) and 24 h (q_w-24 h) in a beaker filled with an approximately 7.5 cm deep layer of distilled water. In order to mimic a real water flow (dynamic test), identically sized samples were placed in a 3 L beaker that was half filled with distilled water. They were agitated in a shaker for 15 minutes at a frequency of 110 cycles min^{-1}. The water uptake q_w (g/g) after 30 s long vertical drainage was calculated in accordance with Eq. (1):

$$q_w = \frac{m_{wt} - m_0}{m_0} \qquad (1)$$

where m_o is the initial weight of the sample (g) and m_{wt} is the weight of the wet sample after 30 s of drainage (g). The sorbents that sink before the experiment is over are considered unsuitable for the intended use.

The oil sorption capacity in water was evaluated by pouring 40.00 g of oil into a beaker filled with 400 mL of distilled water. Circular sample with a diameter of 4 cm was placed in the beaker, which was further shaken for 15 min in a laboratory shaker at a frequency of 110 cycles min^{-1}. A wet sample was drained for 30 s, and weighed. The amount of sorbed oil, i.e. oil sorption capacity of sorbents (q, g/g) was calculated according to Eq. (2):

$$q = \frac{m_f - (m_0 + m_w)}{m_0} \qquad (2)$$

where m_f is the weight of the wet sample after 30 s of drainage (g), m_o is the initial weight of the sample (g) and m_w is the water content in the sample after drainage (g). The method used to determine the water content in tested samples was based on the one described in literature (Renuka et al., 2016). The volume of sorbed water was measured by squeezing the wet samples between the rolls and collecting the content in graduated cylinder. By adding the n-hexane (Fisher chemical, Germany), water is supposed to sink to the bottom of the cylinder leaving the dissolved oil in upper layer.

To evaluate the oil retention, the samples cut into squares (5 cm × 5 cm) were dipped into 150 mL of oil. After 15 min of sorption, the samples were removed and hung vertically to drain. The weight of the samples was measured after 15, 30, 60, 120, 300, and 1800 s of draining. The amount of retained oil was calculated as a difference between the weight of the wet sorbent after drainage and the initial weight of the sorbent (Kovačević et al., 2023).

The reusability was evaluated in accordance with a method described for oil sorption capacity in oil without water (Radetić et al., 2007), where reusability is defined as the quantity of sorbed oil in a particular cycle per weight of dry sample. The sorption process was performed five times. After each cycle, NWS was squeezed between rolls while the CNWS samples were immersed in 25 mL of n-hexane for 5 min, and left to dry in a laboratory oven at 80 °C. The amount of sorbed oil after each cycle was calculated using the Eq. (3):

$$q = \frac{m_f - m_r}{m_0} \qquad (3)$$

where m_f is the weight of the wet sample after 30 s of drainage (g), m_o is the initial weight of the sample (g) and m_r is the weight of the sample after excessive oil being removed (g).

2.4. Biodegradation testing in model compost

The biodegradability of NWS and oiled NWS (ONWS) was evaluated by soil burial degradation test in model compost according to previously adopted procedure with some adaptations (Ponjavic et al., 2017; Tomšič et al., 2022). The samples saturated with crude oil were selected for the test because the crude oil is the most commonly spilled into water. Samples (300 mg to 500 mg) were buried approximately at half the depth of the Petri dish (diameter of 10 cm, depth 3 cm) filled with 200 g of compost soil to ensure aerobic degradation and incubated at 37 °C over 10 weeks. One experimental setup included addition of water (20 mL) at intervals of seven days in order to keep soil wet, alternatively compost was augmented by culture (20 mL) of bacterial consortium isolated from the petrochemically polluted river sediments containing *Rhodococcus* sp. TN101, *Pseudomonas* sp. TN301 and *Acinetobacter* sp. TN302 (Narancic et al., 2012). After 10 weeks the samples were withdrawn from the soil, cleaned with water, washed with 70% ethanol, then soaked in water for

one hour and finally dried to constant weight. Composted samples were photographed, and weight loss was calculated according to Eq. (4):

$$weight\ loss\ (\%) = \frac{m_c - m_0}{m_0} \times 100 \qquad (4)$$

where m_0 represents initial weight of the sample before degradation (g) and m_c corresponds to the weight of dried residual of samples after composting (g). All the experiments were done in duplicates and the presented data referred to the average values.

To determine the number of microbial cells per g of soil of model compost, aliquots (1 g) before and after the burial of NWS and ONWS were taken and standard culture-dependent enumeration procedure was applied (Lawlor et al., 2000; Lee et al., 2021). Soil micro-organisms were cultured at 30 °C on Luria-Bertani agar (LA, Oxoid, UK). Bacterial counts were made 48 h after plating and expressed as colony forming units per gram of soil (CFU/g). In addition, CFU/g of NWS and ONWS was also determined.

3. Results and discussion

3.1. Morphological and chemical properties of NWS and CNWS

The morphological changes of jute fibers induced by carbonization of NWS were assessed by FESEM analysis. FESEM micrographs of NWS and CNWS under different magnification are shown in Fig. 1. Coarse jute fibers with typical multicellular structure are seen in Fig. 1a, b and c. Fig. 1d, e and f reveal that carbonization of NWS leads to a reduction of jute fiber diameter. The average jute fiber diameter in the NWS was 47.3 ± 10.0 μm, with a minimum of 30.3 μm and a maximum of 62.1 μm. After carbonization, the average fiber diameter decreased to 28.9 ± 6.0 μm, with a minimum of 17.2 μm and a maximum of 45.4 μm. The diameter reduction of almost 40% is attributed to material degradation. This reduction resulted in larger voids between fibers.
Originally smoother fiber surface altered to rather rough and the appearance resembled a crumpled paper. However, the formation of pores on the jute fibers is not visible which is in line with literature data (Phan et al., 2006). The pyrolysis of cellulose or lignin during the carbonization lead to elimination of mostly non-carbon elements such as hydrogen, oxygen, and nitrogen in the form of gases and tars, leaving behind a rigid carbon structure (Rodriguez-Reinoso and Molina-Sabio, 1992). The results of EDX analysis shown in Fig. 2g confirm the increase of the carbon content by 75% after carbonization. The same decrease in carbon content induced by carbonization of jute fibers was reported by Chen et al. (2018). At the same time, a sharp drop of oxygen content (by 50%) occurred. In addition, jute fibers were defibrillated and a structure with larger amounts of fine fibers was formed (Fig. 1d). Significantly less severe fiber splitting is sporadically noticed in NWS, which likely occurred due to intense mechanical action during the recycling and production of NWS (Radetić et al., 2007).

FTIR spectroscopy spectra of NWS and CNWS are presented in Fig. 1h. Obviously, carbonization caused a radical change in the chemical structure of jute fibers. The FTIR spectrum of NWS shows a chemical structure characteristic for lignocellulose. A broad band in the region between 3500–3200 cm^{-1} is assigned to O–H stretching vibrations in cellulose and hemicellulose (Alvarez-Murillo et al., 2016). Another broad band with the peaks located at 2922 and 2852 cm^{-1} are associated with an existence of asymmetric C–H stretching in aliphatic hydrocarbons (alkane stretch) and symmetric stretching, present in lignocellulosic constituents (Alvarez-Murillo et al., 2016; Tripathi et al., 2022). The peak centered at 1728 cm^{-1} in the spectrum of NWS ascribed to C=O groups of acetyl ester in hemicellulose and aldehyde in lignin, disappeared in CNWS (Ahmed et al., 2014). The peaks corresponding to C=C vibrations of skeletal aromatic rings in lignin appear at 1595 and 1505 cm^{-1} in the spectrum of NWS. The peak at 1422 cm^{-1} is attributed to skeletal aromatic vibration combined with the C–H in-plane deformation in –OCH_3 (Dong et al., 2018). The bands at 1366 and 1315 are related to C–H bending (deformation stretch) vibrations and C–H wagging vibrations in cellulose, respectively (Chung et al., 2004; Oh et al., 2005). Additionally, the bands at 1160 and 1105 cm^{-1} (asymmetric bridge C–O–C), 1054 cm^{-1} (asymmetric in plane ring stretching), 1030 cm^{-1} (C–O stretching) and band at 899 cm^{-1}(asymmetric out-of-phase ring stretching at C_1–O–C_4 β glucosidic bond) confirm the presence of cellulose in NWS (Chung et al., 2004; Oh et al., 2005; Ciolacu et al., 2011). Reduction of the intensity of almost all mentioned bands after carbonization suggests that the pyranose structure of cellulose is opened (Alvarez-Murillo et al., 2016; Volpe et al., 2020; Bengtsson et al., 2022).

An absence of band between 3500–3200 cm^{-1} in CNWS indicates the occurrence of dehydration during the carbonization, which is in agreement with elemental analysis. In addition, a complete disappearance of the band located at 2922 and 2852 cm^{-2} is strong indication of the aromatic structure formation during pyrolysis (Miliotti et al., 2020; Bengtsson et al., 2022). The presence of the carbon-dioxide (doublet at 2632 and 2338 cm^{-1}) as a product of carbonization is also detected in CNWS (Oh et al., 2005). Evident bands with maxima at 1570 and 1374 cm^{-1} correspond to G band (graphite-like) and D band (disordered graphite), respectively (Socrates, 2001). The peak at 1570 cm^{-1} is attributed to C=C vibrations of aromatic skeletal rings mixed with aromatic C–C stretching vibrations (Miliotti et al., 2020), while a broad band centered at 1374 cm^{-1} corresponds to vibrations observed for mono- and di-substituted naphthalenes (Socrates, 2001). The peak at 873 cm^{-1} in CNWS is related to aromatic out-of-plane bending vibrations due to ring substitution (Tripathi et al., 2022). It also confirms the presence of lignin in the form of lignin-containing aromatic rings and indicates that lignin possesses higher thermal stability than cellulose-hemicellulose (Tripathi et al., 2022). Several bands between ∼850 and ∼550 cm^{-1} (810, 750, 700, 669, 650, 620 and 580 cm^{-1}) are ascribed to C–H vibrations of mono, di, tri and tetra substituted aromatic rings and naphthalene structures produced during the carbonization process (Socrates, 2001).
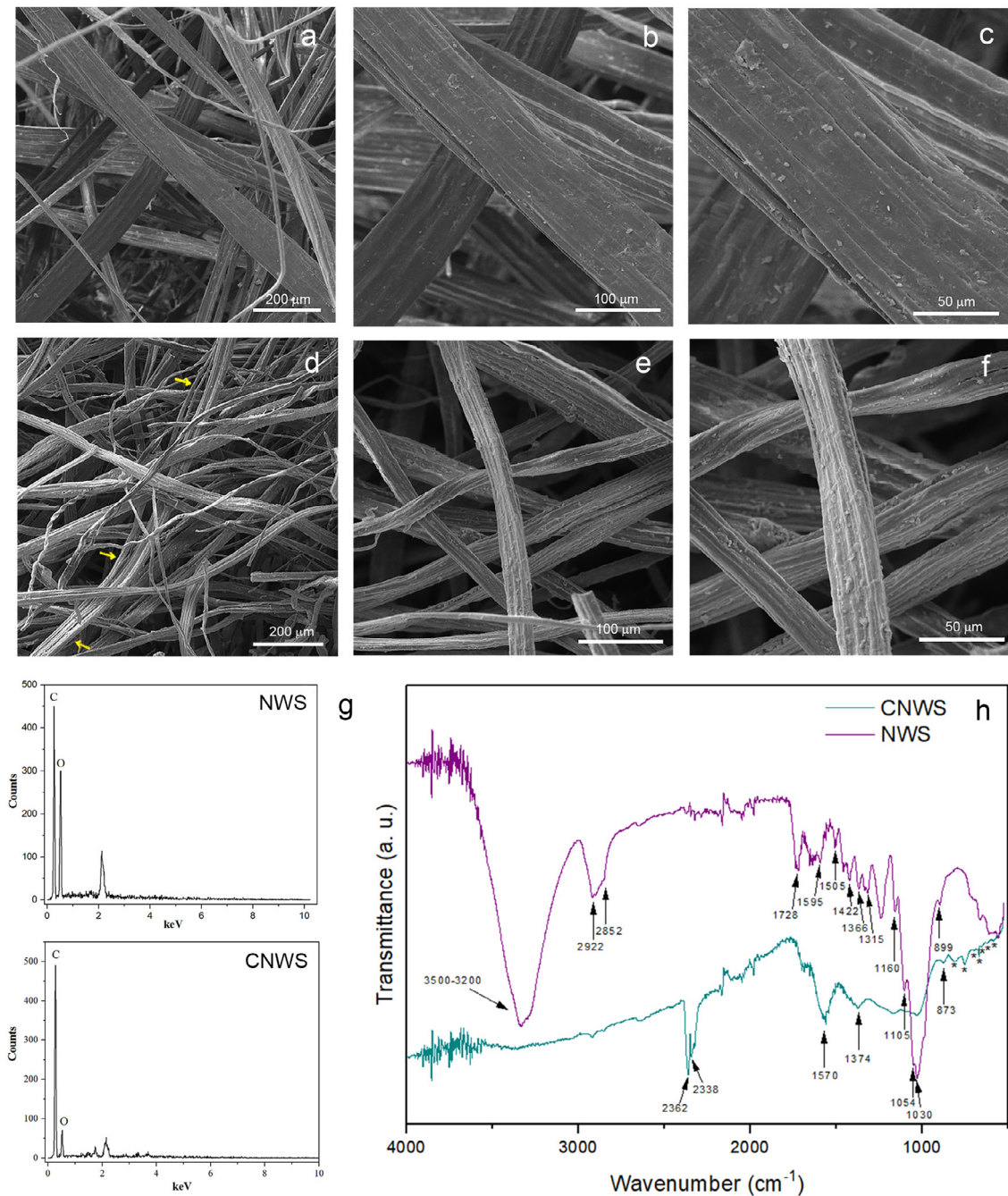
Fig. 1. FESEM micrographs of NWS (a, b, c) and CNWS (d, e, f) under different magnification; EDX (g) and FTIR (h) spectra of NWS and CNWS.

3.2. Water and oil sorption performance of NWS and CNWS

An efficient sorbent should be able to float on the surface of the water for extended periods of time (ITOPF, 2014). This behavior is mostly governed by the sorbent attraction to water and oils. Having a strong affinity for water is the major disadvantage of organic natural sorbents composed of cellulose. As they quickly become waterlogged and sink, they are often not suitable for use. The water uptake of NWS (0.3 g/g under static and 0.4 under dynamic conditions) and CNWS (0.4 g/g under both static and dynamic conditions) is barely different after 15 min of contact independent of applied conditions. The surface of the jute fibers in NWS is partially hydrophobic due to the presence of waxes and pectin, and thus, practically no water was sorbed under static as well as under dynamic conditions within 15 min. They showed a good
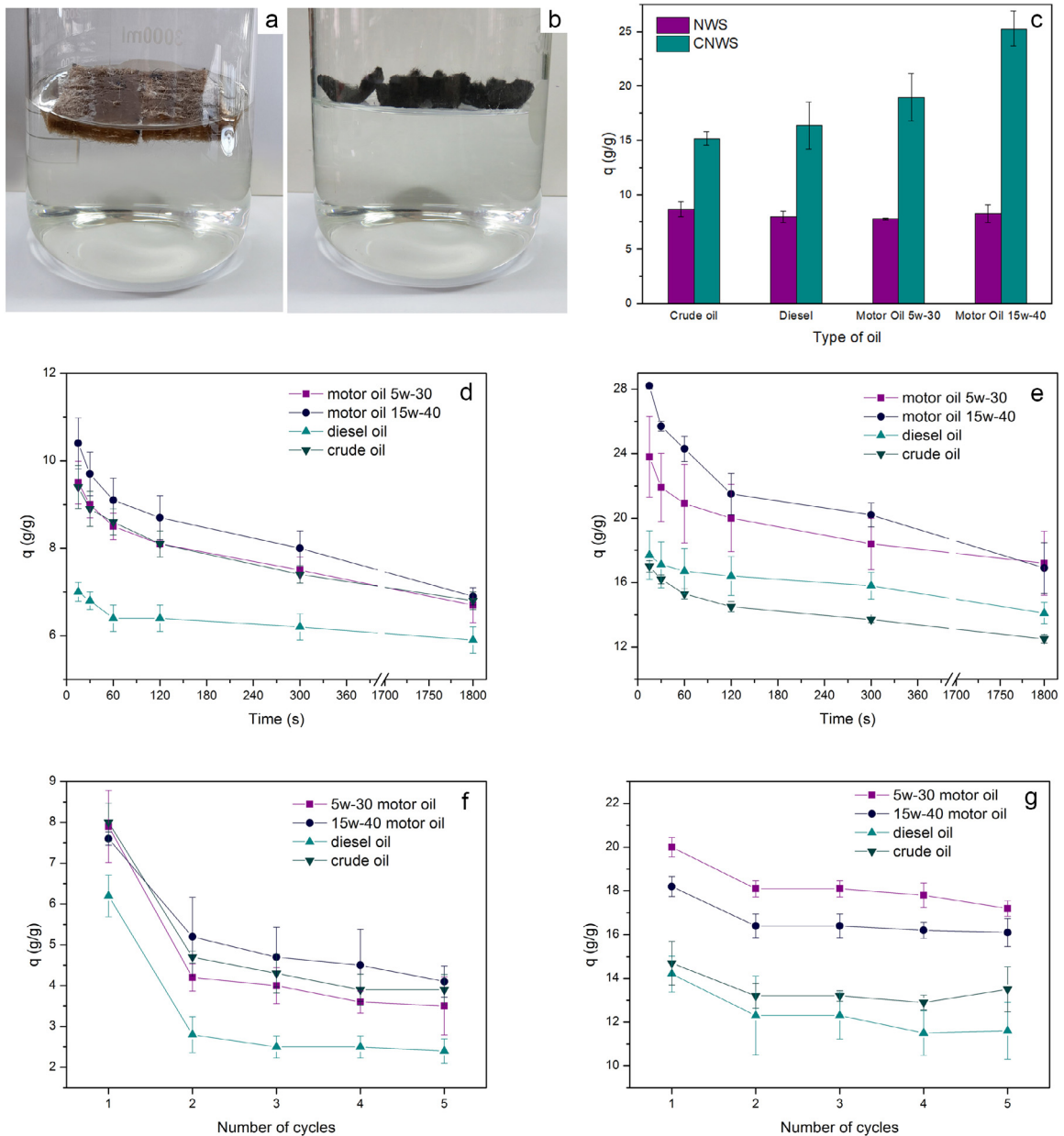
Fig. 2. Buoyancy of NWS (a) and CNWS (b) under static conditions after 24 h long contact with water; oil sorption capacities of NWS and CNWS in water (c); oil retention of NWS (d) and CNWS (e), and reusability of oiled NWS (f) and CNWS (g).

buoyancy during this time, staying well above the water surface. Although NWS did not sink even after 24 h long contact with water under static conditions (Fig. 2a), considerably larger amount of water was sorbed (9.0 g/g). In contrast, the CWNS did not absorb any additional water and it also remained on the water surface (Fig. 2b). A small water uptake (0.4 g/g) even after long contact with water is attributed to increased hydrophobicity of the sorbent and decrease in hydrophilic groups induced by carbonization (Huang, 2009). Increased hydrophobicity was confirmed by measuring the water contact angles. NWS and CWNS exhibited water contact angles of approximately 103° and 132° (Fig. S1), respectively.

High oil uptake and large oil retention capacity are key features of efficient sorbents. Oil sorption capacity of NWS and CNWS was evaluated in water. Fig. 2c reveals that oil sorption capacity of the NWS is rather low ranging between 7.8–8.7 g/g depending on tested oil. Both NWS and CNWS did not absorb measurable amount of water during the sorption process. This is explained by the aforementioned partially hydrophobic surface of NWS originating from jute fiber waxes.

Another reason for such behavior is oil adsorbed on the NWS before the sample even reached the water–oil interface, which prevents water sorption (Thilagavathi et al., 2018).

Oil sorption capacity of NWS is insignificantly affected by the viscosity of tested oils. Such behavior disobeys generally accepted trend that oil sorption capacity of fibrous materials increases with an increase of oil viscosity (Tan et al., 2021). Namely, high viscosity oils are more easily adhered, forming a thick layer on the fiber surface (Radetić et al., 2007; Zhou et al., 2017; El Gheriany et al., 2020). Unlike them, low-viscosity oils tend to diffuse quickly into inter-fiber voids passing through the fibrous network into lumen where the oil becomes trapped due to capillary forces (Tan et al., 2021). Taking into account a high area density of NWS (800 g/m^2) and its compact structure with closely packed fibers, noticed behavior is not surprising because of the lack of space for capillary bridging between the neighboring fibers, which was prominent in the case of jute NWSs of smaller area densities (400–530 g/m^2) (Kovačević et al., 2023).

The influence of oil viscosity becomes more prominent during the oil sorption on CNWS. Fig. 2b indicates that high-viscosity motor oils 5w-30 and 15w-40 were considerably better sorbed than diesel and crude oil. Increased fiber surface roughness (Fig. 1) and hydrophobicity (Fig. 2a, Fig. S1) induced by carbonization facilitate the adherence of higher-viscosity oils resulting in larger oil sorption capacities. In addition to fiber surface changes, the decrease in fiber diameter, defibrillation and increased voids between fibers in CNWS led to a drastic increase in oil sorption capacities (15.2 to 25.3 g/g) independent of oil type.

Beside the large oil sorption capacity, efficient oil sorbent is supposed to provide sufficiently good oil retention i.e. to suppress excessive oil dripping from saturated sorbent. The oil retention on NWS and CNWS is presented in Fig. 2d and e, respectively. NWS and CNWS followed the same pattern: after the most intensive oil dripping within the first minute of draining, the oil release slowed down without reaching the steady-state. Similar behavior of fibrous sorbents is reported in literature (Radetić et al., 2007; Kovačević et al., 2023). NWS retained 84, 72, 70 and 66% of initial diesel oil, crude oil, motor oils 5w-30 and 15w-40 uptake after 30 min of draining, respectively. Similar trend appeared with CNWS: 80, 73, 72 and 64% of diesel, crude oil, motor oils 5w-30 and 15w-40 was retained, respectively. Apparently, motor oil 15w-40 with the highest viscosity is better sorbed on CNWS, but the retention of sorbed oil is the worst. This is in line with the fact that highly porous materials have a greater potential for sorption but worse oil retention (Wei et al., 2003). On the contrary, the largest amounts of diesel oil are retained on both NWS and CWNS. In general, the oil release take place because of the immediate dripping from NWS or CNWS external surface and slow draining of extra-lumen oils since the capillary pressure is insufficient to hold retained oil (Lim and Huang, 2007).

Following the sustainable path, after the simple removal of used material provided by non-woven structure, the further focus was on recovery of the adsorbed oil and the reusability of the sorbents. Recovery methods of different efficacy are proposed, such as distillation and extraction for removal of valuable contaminants or those with low boiling points, squeezing for costly or nonflammable contaminants with high boiling points, and combustion for flammable and valueless contaminants (Bi et al., 2013). Fig. 2f and g demonstrate the possibility to reuse the NWS and CNWS after five sorption/desorption cycles, respectively. The majority of the oil sorbed by the NWS could be removed and collected with simple squeezing between rolls. The same pattern is evident independent of tested oil: a sharp decrease in sorption capacity (approximately 50%) after the first cycle, followed by an almost constant value in all subsequent cycles. The worst reusability occurred in the case of diesel oil with the lowest viscosity where reduction in oil uptake decreased about 60% after the first cycle. Observed trend can be explained primarily by a change in non-woven structure caused by the pressure applied during the squeezing. The load compressed the voluminous structure, reducing the voids between the fibers that were accessible to oil sorption (Jarrah et al., 2018). The decrease of capacity is partially due to the residual oil, entrapped in the lumen of jute fiber or in the voids of densely packed fibers.

CNWS showed a similar trend during five cycles of sorption/desorption, with considerably smaller decrease in oil uptake after the first cycle (approximately 12–20% depending of tested oil). This could be partially due to differences in the desorption method. Namely, squeezing between rolls under the pressure was not a suitable option for carbonized samples because of their fragility. Therefore, the oils from CNWS were dissolved in n-hexane. This approach of oil recovery is also reported for high-viscous oils, which are hard to collect by filtration under suction (Inagaki et al., 2002). In this case, the residual, insufficiently dissolved oils are the major reason for the decrease in oil uptake after the first cycle, since the CNWS structure has not been changed.

3.3. Biodegradation of NWS and ONWS

The biodegradability of NWS and oiled NWS (ONWS) was evaluated in model compost over 10 weeks. The photographs of the samples after 10 weeks in compost (Fig. 3a) clearly reveal that the structure of sorbents started to disintegrate. The damage of jute fibers and their disintegration were confirmed by FESEM analysis. FESEM images shown in Fig. 4 indicate much faster and more severe biodegradation of NWS compared to ONWS. Not only that plenty of fibers became shortened or broken (Fig. 4a and c), but the attack of microorganisms resulted in disappearance of cells which is particularly prominent on NWS_C (compost) sample (Fig. 4b). Unlike jute fibers, PA fibers remained almost intact. Oiled samples showed slower biodegradation rate, what was further corroborated by lower weight loss of oiled samples to ones without oil (Fig. 3b). The appearance of large holes as a result of microorganism action is evident in Fig. 4f and h. It seems that augmentation with microorganisms isolated from the contaminated sites with versatile metabolic potential is beneficial for biodegradation of oiled samples. Differences in weight loss of samples were not observed between augmented and
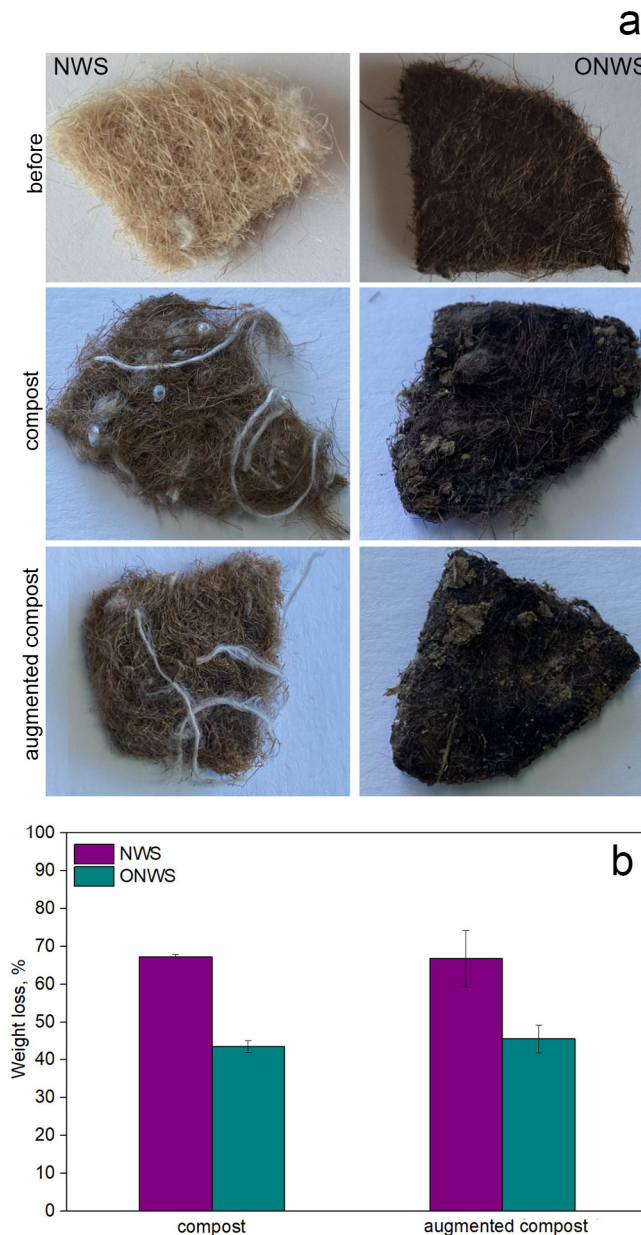
Fig. 3. Photographs of NWS and ONWS before and after biodegradation in compost and augmented compost (a) and weight loss of NWS and ONWS after 10 weeks in model compost at 37 °C (b).

non-augmented compost (Fig. 3b). Worth mentioning is that these samples contained considerable and comparable load of bacteria prior to burial of 1.5–2 $\times$ 10^7 CFU/g. Incubation of NWS and ONWS in model compost has not significantly changed microbial cell counts apart in augmented soil, where bacterial culture aliquots were added (Table S1, Fig. S2, Supplementary data).

4. Practical applications and future research prospects

Keeping in mind the fact that NWS based on post-industrial textile waste was manufactured in industrial conditions, its mechanical and sorption properties, it could be applied for oil spill clean-up at scale without further examination. In contrast, CNWS requires optimization. Further research should be directed towards the influence of area density of NWS on its sorption performance and recovery after carbonization. Namely, the possibility to recover the CNWS should
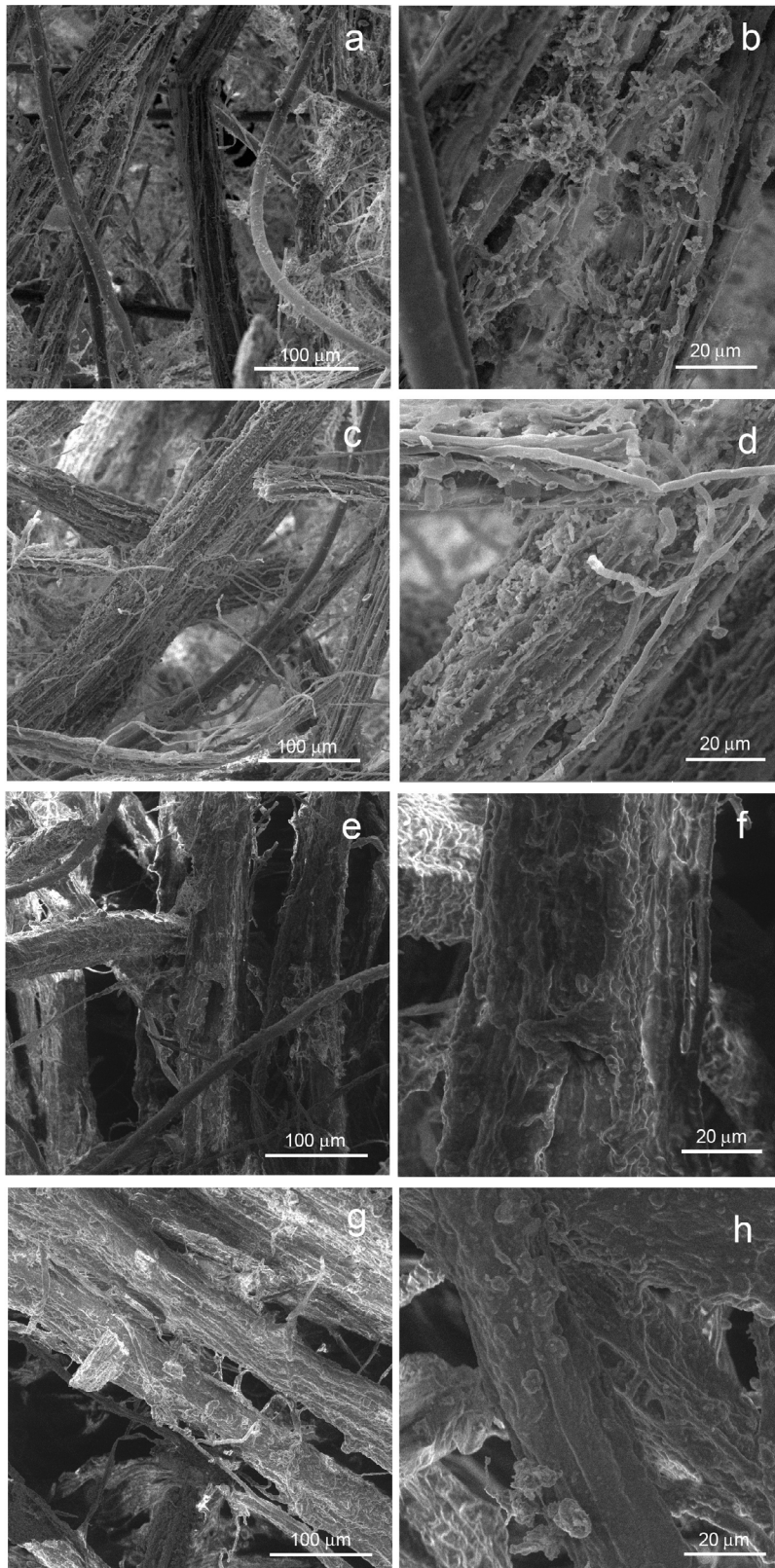
Fig. 4. FESEM images of (a, b) NWS_C, (c, d) NWS_AC (augmented compost), (e, f) ONWS_C and (g, h) ONWS_AC (augmented compost).

be evaluated with green solvents. The research should also be extended with other investigated oils and the possibility to adjust the compost composition to pollutant should be investigated in more detail.

5. Conclusion

Recycled jute fibers from carpet fringes as a post-industrial waste can be efficiently exploited for the manufacturing of non-woven sorbent suitable for oil spill clean-up on water surfaces. Although the produced sorbent passed the buoyancy test under static and dynamic conditions for short times and provided good oil retention, the oil sorption capacity was low. Oil sorption capacity did not significantly depend on oil viscosity. Carbonization of the sorbent caused the defibrillation of jute fibers and chemical changes leading to an increase in fiber surface hydrophobicity and voids between neighboring finer fibers. Consequently, the oil sorption capacity significantly increased particularly for the high-viscosity motor oils. Particular advantage of carbonized sorbent is efficient reusability as it retained 80–88% of its initial oil sorption capacity after 5 sorption/desorption cycles. Non-carbonized oiled sorbent showed a tendency to efficiently biodegrade in compost, which makes it a sustainable candidate for cleaning of oil spills.

CRediT authorship contribution statement

Aleksandar Kovačević: Investigation, Writing – original draft. **Marija Radoičić:** Investigation, Writing – review & editing. **Darka Marković:** Investigation, Writing – review & editing. **Marijana Ponjavić:** Investigation, Writing – original draft. **Jasmina Nikodinovic-Runic:** Investigation, Writing – original draft. **Maja Radetić:** Conceptualization, Supervision, Writing – review & editing.

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.

Data availability

Data will be made available on request

Ethical approval

This article does not contain any studies with human participants or animals performed by any of the authors.

Appendix A. Supplementary data

Supplementary material related to this article can be found online at https://doi.org/10.1016/j.eti.2023.103170.

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