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Letter

Simultaneous Removal of Oil and Bacteria in a Natural Fiber Filter

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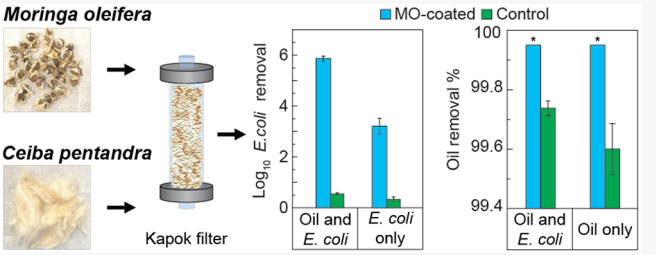
ABSTRACT: Microbes and oil occur together in produced waters (PW) and must be removed. Their removal prior to discharge, reinjection, or reuse is necessitated by downstream challenges and regulations. In particular, microbial removal is required prior to additional treatment, distribution, or reuse to avoid biofouling, biocorrosion, or well clogging/souring. We demonstrate simultaneous removal of oil (>99.95%) and *Escherichia coli* (6 $\log_{10}$) in a *Ceiba pentandra* (kapok) fiber filter with adsorbed *Moringa oleifera* (MO) proteins for sustainable PW treatment under moderate salinity conditions (ionic strength of 90.1 mM). This filter is compared to the industry standard, walnut shell media filters, which remove smaller amounts of oil (99.3%) and a negligible amount of *E. coli* (0.29 $\log_{10}$). MO antimicrobial cationic proteins were adsorbed onto the naturally occurring oleophilic kapok fibers, providing *E. coli* removal while oil sorption occurs on the hydrophobic fibers. Additionally, synergistic removal of *E. coli* by bare kapok fibers in the presence of oil was observed and the effect of MO proteins on enhancing this removal was demonstrated. This work provides a unique framework for the evaluation of sustainable PW treatment using plant-based fiber filters.



INTRODUCTION

Approximately 250 million barrels of produced water (PW) are generated by the global oil and gas industry each year as a byproduct.[1] PW must undergo treatment to remove oil and grease to comply with discharge regulations, allow reinjection into wells, or enable reuse in oil and gas operations.[2] Additionally, it is essential to remove microorganisms from PW to prevent microbe-induced issues in treatment trains, such as biofouling,[3] and production and/or transport issues, such as reservoir souring during reinjection reuse[4,5] and microbially induced corrosion (MIC).[6,7]

The characteristics of PW vary considerably depending on the production method and geographic area, but PW typically contains hydrocarbons, dissolved solids, and bacteria.[8,9] Traditional treatment technologies, for example, gravity or cyclone separators, do not provide sufficient contaminant removal to meet strict regulations, thus requiring advanced treatment to remove organics, microorganisms, and colloids.[10] Advanced treatment frequently uses membrane technologies and multistep trains to reach treatment goals.[11]

PW treatment trains, in general, focus primarily on separate oil removal and disinfection steps. Adsorption processes are often utilized for oil removal, such as walnut shell filters or granular activated carbon (GAC).[1,9] Membrane bioreactors are employed following media filtration and can utilize activated sludge bioprocesses in combination with membrane filtration to achieve high rates of disinfection and particulate removal.[12] Depending on the end use of treated water, various membranes such as ultrafiltration (UF), microfiltration (MF), and reverse osmosis (RO) membranes are used for further contaminant reduction.[9] Reliance on membrane technology is increasing as discharge and reuse standards become more stringent and water scarcity requires recycling of PW.[11,13]

Pretreatment processes are necessary for membrane applications in PW treatment.[10,11] Despite its relative maturity, membrane technology still suffers from challenges, including its high cost and the deterioration of its performance due to fouling, specifically biofouling. Biofouling, prevalent in oil field applications, can cause significant operational issues, decreased permeate production, decreased salt rejection, and increased energy consumption,[8] leading to costs associated with membrane cleaning, decreased lifetime, replacement costs, and increased energy consumption.[14,15] Thus, removing microorganisms prior to membrane treatment is crucial to avoid biofilm formation. Current operational methods[16] and proposed membrane modifications[17−20] to address and prevent biofouling utilize solvents and chemicals, making these options costly and not environmentally friendly.[21] Overall, current challenges with membrane technology for applications in PW treatment provide an excellent example of the need to achieve simultaneous oil and microbial removal with sustainable technologies. Here, we present a sustainable functionalized

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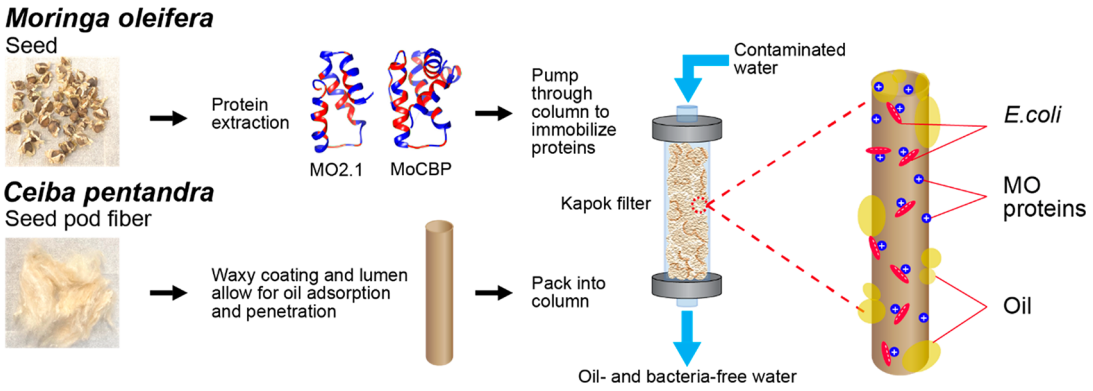

Figure 1. Natural fiber filter made from *Moringa* seed extract and *C. pentandra* (kapok) fibers for removal of oil and bacteria. Cationic antimicrobial proteins are easily extracted from *Moringa* seeds and can onto adsorb to oleophilic kapok fibers, creating an antibacterial−oleophilic filter. Previous work has shown MO2.1 and MoCBP are the primary proteins responsible for antimicrobial activity.[28,30] Kapok fibers provide a hydrophobic surface for oil interaction and adsorption that occur simultaneously with *E. coli* removal.

natural fiber filter for simultaneous removal of dispersed oil and bacteria, a potential upgrade of, or an alternative to, conventional and advanced oil removal technologies.

Ceiba pentandra, commonly termed kapok, is a tropical tree found in Central and South America, parts of west Africa, and Asia. The fibers from kapok tree seedpods have traditionally been used as an inexpensive filling for furniture and upholstery and also as an insulating material.[22] Kapok fibers are highly hydrophobic and have been studied previously as oil sorbents for oil spill and oily wastewater applications.[23−25] These studies primarily focus on kapok's ability to absorb pure oil in static tests, and no work has been reported on simultaneous removal of oil and bacteria in a flow-through filtration setting.

Moringa oleifera (MO) seeds have been studied extensively for water treatment applications because of their coagulation,[26,27] antibacterial,[28,29] and, more recently, antiviral properties.[30] Sand filters coated with MO seed extract have been shown to remove 7 $\log_{10}$ of MS2 virus[30] and 8 $\log_{10}$ of *Escherichia coli*.[31] Microbial removal was shown to occur through interaction with two cationic proteins, MO chitin-binding protein (MOCBP) and MO coagulant protein (MO2.1), and its application was proposed for use in low-resource settings for filtration of drinking water. In these previous reports, cationic MO proteins were adsorbed to negatively charged sand or glass particles, with removals determined only at low salinity (ionic strength of 10 mM).[30,31] In this work, we find that MO proteins can adsorb to hydrophobic kapok fibers and provide 7 $\log_{10}$ *E. coli* removal even under moderate-salinity conditions (82.8 mM NaCl, ionic strength of 90.1 mM, and TDS of 5237 mg/L). By combining the oleophilic properties of kapok fibers with the antimicrobial activity of MO proteins (Figure 1), we produce an anti-biofouling filter that can simultaneously remove oil and bacteria for PW treatment applications, oil/hydrocarbon contamination mitigation, point-of-use (POU) filters for households, membrane pretreatment, and emergency water supply filters.

MATERIALS AND METHODS

***Moringa* Serum and Functionalized Kapok Preparation.** Whole unshelled *M. oleifera* seeds from Echo Global Farm in Florida were ground with a coffee grinder and mixed with modified 10-fold diluted phosphate-buffered saline (PBS) with total dissolved solids (TDS) of 4574 mg/L (75 mM NaCl, 0.27 mM KCl, 1 mM Na_2HPO_4, and 0.18 mM KH_2PO_4), herein termed modified PBS. Modified PBS was used instead of deionized (DI) water (which was used in previous work[30,31]) to demonstrate the ability to use a more realistic water for protein extraction. For example, the typical groundwater salinity ranges from 100 to >50000 mg/L.[32] Crushed seeds (2 g per 100 mL) and modified PBS were mixed for 5 min followed by filtration through a 1.5 μm glass microfiber filter (VWR Inc.). An *in situ* coating process was employed for coating kapok. In this process, the seed extract (100 mL) was coated onto 2 g of kapok fibers packed into a disposable column at a rate of 2 mL/min using a peristaltic pump (Masterflex L/S Variable-Speed Digital Drive, Cole-Parmer).

Walnut Shell Media. Walnut shell (WS) media (20−30 mesh) was purchased to provide a baseline for oil removal due to its use in the PW industry.[33,34] The preparation of WS was adapted from similar work.[35,36] Medium was washed in DI water until the water ran clear. It was dried at 105 °C overnight and stored in an airtight flask.

Model Oily Saline Water. Canola oil was used as the experimental oil in this study due to the use of vegetable oils in previous research related to oil fouling.[37,38] The oil-water emulsion was prepared by mixing 1% (w/w) canola oil in modified PBS using a blender. Constant mixing using a magnetic stir bar was used to keep the oil-water emulsion stable for the duration of the experiments. The particle size distribution of oily water using dynamic light scattering (DLS) is included in Figure S1. Experiments for the removal of oil with and without bacteria were conducted at a flow rate of 10 mL/min (loading rate of 3.39 m/h).

For simultaneous *E. coli* and oil removal experiments, the oil-water emulsion was prepared by mixing 1.167% (w/w) canola oil in modified PBS using a blender. *E. coli* strain TG1 containing red fluorescent protein (pCA24N-rfp-lasR[39]) was used as the model bacterium at an approximate influent concentration of 10^8 colony-forming units (CFU)/mL suspended in PBS buffer (137 mM NaCl, 2.7 mM KCl, 10 mM Na_2HPO_4, and 1.8 mM KH_2PO_4). Culture medium chemicals were removed from the cell suspension by rinsing pellets twice with PBS buffer. Culturing details were described in a previous study.[40] *E. coli* was added to the oil-water emulsion to dilute oil to a final concentration of 1% (w/w) and final salinity of 90.1 mM ionic strength (82.8 mM NaCl, 0.617

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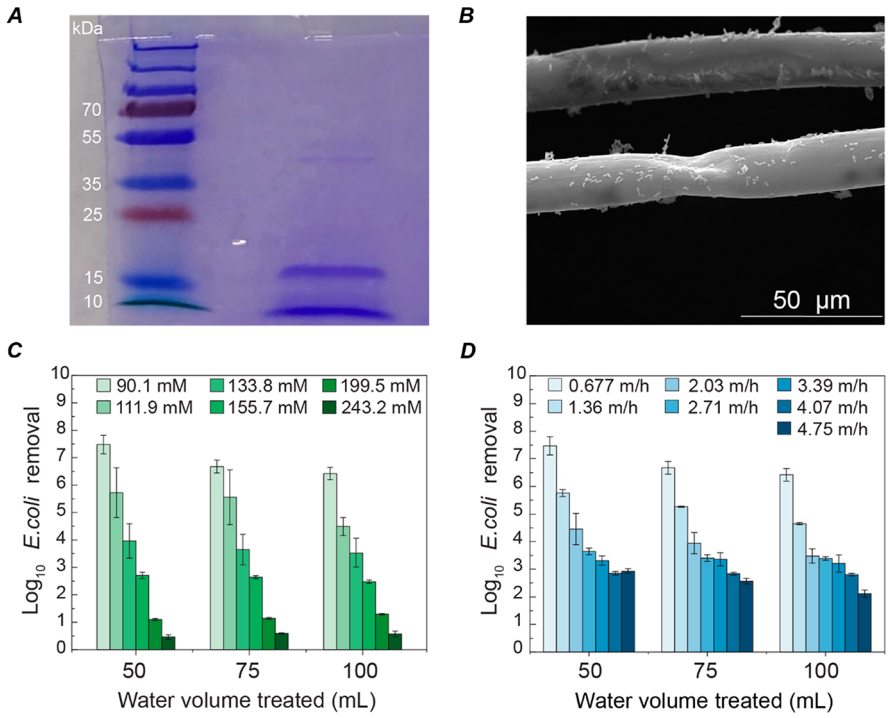

Figure 2. (A) MO proteins adsorbed onto kapok fibers are shown to desorb in a 600 mM NaCl wash, which is confirmed via SDS−PAGE (full size gel shown in Figure S2). (B) SEM image of MO-coated kapok following the *E. coli* filtration experiment that confirms adsorption of MO proteins to kapok (control image in Figure S6B) and the adherence of *E. coli* to MO-coated kapok. (C) Increasing salinity is shown to decrease the rate of *E. coli* removal, but a high level of removal persists (99.9%) at an ionic strength of 155.7 mM. Salinity values shown represent the ionic strength of the *E. coli* solution filtered through the columns. Note that increasing salinity experiments were conducted at a loading rate of 0.677 m/h (2 mL/min). (D) An increasing flow rate is shown to decrease the rate of *E. coli* removal but maintain >99% removal up to a loading rate of 4.75 m/h. Note that increasing flow rate experiments were conducted at an ionic strength of 90.1 mM. Error bars denote the standard error from triplicate experiments.

mM KCl, 2.28 mM Na_2HPO_4, and 0.411 mM KH_2PO_4). A conventional plate counting method was used to quantify cell concentrations.[41]

Column Experiments. The filter columns used were 10 cm disposable columns with an internal diameter of 1.5 cm. Column adapters (Bio-Rad) were used to eliminate the head space in the columns. For kapok filtration media, raw kapok fibers were packed into the column evenly at a packing density of 0.11 g/cm^3 (mass of fibers/column volume). For WS media, an equivalent volume was used (~17.7 cm^3). Peristaltic pumps were used to pump the solution through columns with the feed entering from the top of the column at a constant flow rate. The packed media filter was rinsed with 100 mL of DI water to set the flow rate. For MO-coated kapok experiments, 100 mL of *Moringa* serum was pumped through columns to functionalize kapok. Columns were equilibrated with background ionic strength solutions (100 mL) before switching to appropriate influent solutions. Sterilized vials were used to collect effluent samples (1 mL) at designated bed volumes of 2.78, 4.17, and 5.56 (50, 75, and 100 mL, respectively).

Gel Electrophoresis. To characterize the protein adsorbed on kapok, sodium dodecyl sulfate−polyacrylamide gel electrophoresis (SDS−PAGE) was conducted, and details are provided in the Supporting Information.

Oil Content Analysis. Fatty acids in vegetable oils exist in triglyceride molecules, which are not volatile enough to use gas chromatography−mass spectroscopy (GC-MS) for direct analysis. Fatty acid methyl esters (FAMEs) are a common derivative used to convert triglycerides to a molecule better suited for GC-MS analysis.[42,43] The FAME derivatization protocol used in our work was adapted from ref 44 (details in the Supporting Information). The GC-MS detection limit was determined to be 5 ng/μL for the target compound, methyl oleate. Examples of GC-MS total ion chromatograms are shown in Figures S3 and S4, and the standard calibration curve is shown in Figure S5.

Scanning Electron Microscopy (SEM) Analysis. The morphology of kapok fibers before and after *E. coli* removal was characterized by SEM (details in the Supporting Information).

RESULTS AND DISCUSSION

Protein Adsorption onto Kapok Fibers and *E. coli* Removal. Initial *E. coli* removal experiments maintained a low (ionic strength of 15 mM) salinity and a low (0.677 m/h) loading rate (Figure S6A). Adsorption of protein to kapok fibers was evaluated via SDS−PAGE (Figure 2A), adsorbed protein quantification, and *E. coli* removal (Figure 2B). Liquid chromatography−mass spectrometry (Table S1) confirmed the presence of MO proteins adsorbing to kapok fibers. The protein concentration in the 600 mM NaCl wash from the fluorometric assay was 8.64 ± 0.51 mg, resulting in an average of 4.32 mg of protein/g of fiber. In previous work with MO-coated sand filters, the electrostatic interaction between negatively charged "sand" particles (silica beads) and cationic peptides provided the mechanism for protein adsorption.[30,31] In this work, we show that MO proteins can also adsorb onto the hydrophobic, waxy coating on kapok fibers, based on *E. coli* adsorption shown in Figure 2B. An SEM image of kapok before *E. coli* removal is shown in Figure S6B.

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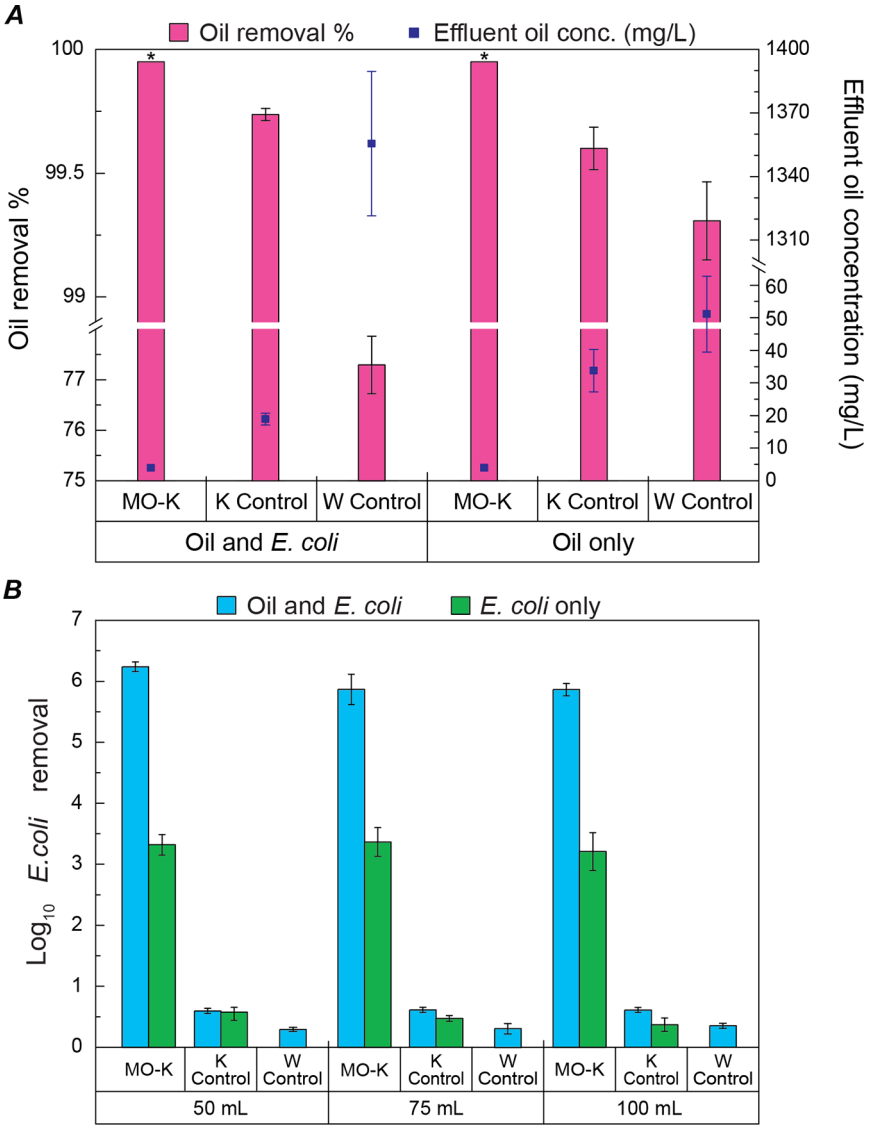

Figure 3. Kapok columns achieve a high level of oil and bacterial removal under all experimental conditions. (A) MO-coated kapok (MO-K) slightly increases the rate of oil removal from control kapok (K Control) during oil filtration. When bacteria are present, K Control filters achieve a higher rate of removal than when filtering oil only, most likely due to coalescence and flocculation. WS media (W Control) achieve rates of oil removal similar to, but slightly lower than, that of K Control when bacteria are not present. The rate of oil removal is significantly decreased when bacteria are present. An asterisk indicates triplicates fell below the GC-MS detection limit of 5 ng/μL, resulting in >99.95% oil removal and a concentration of <4 mg/L in effluent samples. (B) Rates of *E. coli* removal are shown for filtration experiments using MO-K, control (uncoated) kapok (K Control), and WS media (W Control). MO-K shows increased rates of bacterial removal in the presence of oil. This is likely due to flocculation and/or coalescence of bacteria and oil that allows more bacteria to interact with MO proteins. Control kapok (K Control) experiments show little difference in bacterial removal when oil is added to filtration. This shows the significance of MO proteins on kapok to further increase the rate of removal of bacteria in the presence of oil. WS media show very low rates of removal of bacteria. Error bars denote the standard error from triplicate experiments. Note that results shown here were all conducted at a filtration loading rate of 3.39 m/h (10 mL/min) with an ionic strength of 90.1 mM.

The Rate of *E. coli* Removal Decreases as Salinity Increases, but a High Rate of Removal Persists. PW salinities vary on the basis of the extraction method and geographic location but can be anywhere from a few parts per million (or milligrams per liter) to >100000 mg/L.[1,10] In the Permian basin, the Delaware, Devonian, and Leonardian region PW salinity is <8000 mg/L TDS.[45] To understand how *E. coli* removal would be affected under saline conditions similar to those in the low-TDS range of PW, experiments were performed with different TDS concentrations at a constant flow rate of 2 mL/min (0.677 m/h). The results, shown in Figure 2C, show that the rate of removal at a low ionic strength is high at approximately 7 $\log_{10}$ removal, and as the TDS and ionic strength increase, the rate of *E. coli* removal decreases. However, >99% removal persists at an ionic strength of 155.7 mM (9076 mg/L TDS). Previous work has shown the interaction between MO2.1 and *E. coli* is primarily electrostatic.[28] Thus, a large decrease in the rate of removal was expected because of increasing ionic strength, due to electrostatic screening.[46] The high rate of removal seen at high ionic strengths indicates that this highly effective filter can still operate at high TDS values.

Three $\log_{10}$ (99.9%) *E. coli* Removal Is Achieved at Filter Loading Rates Approaching the Rapid Sand

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Filtration Range. In previous work with MO-coated sand filters,[30,31] filter loading rates were quite low and typical of slow sand filtration (0.1−0.4 m/h[47]). Rapid sand filtration (RSF) operates anywhere from 5 to 15 m/h[48] and is a preferred mode of operation because of its low cost and practical implementation. We investigated flow rates approaching the RSF range, and the results indicate that an increasing flow rate decreases the rate of *E. coli* removal but remains around 3 $\log_{10}$ removal at flow rates near the RSF range (Figure 2D). The decrease is likely due to the decrease in contact time between the *E. coli* and adsorbed MO proteins with an increase in loading rate.

Kapok Fiber Filters Achieve 99.6% Removal of Oil from Oil-in-Water Emulsions. The waxy coating on kapok fibers allows for hydrophobic interaction and oil sorption as shown in previous static oil-sorption studies.[23,24,49,50] In the filtration tests we conducted with 1% (w/w) oil-in-water emulsions through kapok fiber filters, 99.6% of canola oil in water was removed. Filtration using kapok fibers has not been extensively reported in the literature, but some studies exist for oil-water separation.[25,51,52] Similar oil removal for kapok filters has been achieved for diesel oil (>99% removal[25,51,52]), hydraulic oil (99.6% removal[25]), and vegetable oil (>99% removal[51]). The filtrate from control (uncoated) kapok (K Control) filters has an average effluent concentration of 33.7 mg/L oil, as shown in Figure 3A, providing sufficient removal to satisfy the U.S. Environmental Protection Agency discharge regulation.

WS Medium Filters Provide 99.3% Oil Removal. WS medium has been well-studied as an oil sorbent[36,53,54] and is commonly employed in produced water treatment to remove oil.[33,55] WS media removed 99.3% of oil from the oil-in-water emulsion (Figure 3A), corresponding to an average oil effluent concentration of 51.1 mg/L, when filtering oil only. This does not meet the U.S. Environmental Protection Agency's produced water discharge regulation of 48 mg/L oil and grease.[56]

MO-Coated Kapok Fiber Filters Show Enhanced Oil Removal. When kapok fibers were coated with MO proteins, the rate of oil removal increased to >99.95%, which correlates to an effluent concentration of <4 mg/L (Figure 3A). Canola oil is composed primarily of triacylglycerols with ∼1% free fatty acids (FFAs).[57] Therefore, the interaction between kapok and oil is predominantly hydrophobic. Under experimental conditions at a pH of ∼7, FFAs of canola oil (62% oleic acid, 20% linoleic acid, and 9% α-linoleic acid)[58] exist in the deprotonated form (pK_a values of 5.02 and 4.77),[59] revealing a negatively charged region. MO proteins, specifically MO2.1, have been shown to possess flocculating activity due to the presence of several positively charged residues on the protein.[60] An electrostatic interaction between deprotonated FFAs and cationic MO proteins may explain the slight increase in the rate of oil removal when MO protein is present on kapok fibers. A jar test experiment was conducted to determine the MO serum flocculating capacity for the canola oil-water emulsion, and the results showed a >99% decrease in turbidity (due to oil) in the suspensions with MO serum (Figure S7). Electrostatic attraction, coupled with hydrophobic interaction between oil and MO proteins, may have contributed to the decrease in turbidity of the oil-water emulsion.

WS Media Provide Low Rates of Removal of Oil and Bacteria When Filtered Simultaneously. The rate of WS oil removal decreased to 77.3% when bacteria and oil were filtered simultaneously. In addition, WS media provided very little bacterial removal (0.29 $\log_{10}$ removal), which is less than control kapok bacterial removal (0.59 $\log_{10}$ removal). The corresponding effluent oil concentration is 1355 mg/L, which does not satisfy U.S. Environmental Protection Agency discharge regulations without further treatment.

Functionalized Kapok Fibers Achieve Simultaneous 6 $\log_{10}$ (99.9999%) *E. coli* Removal and >99.95% Oil Removal under Moderately Saline Conditions. Functionalized kapok columns achieve 5−6 $\log_{10}$ (99.999−99.9999%) removal of *E. coli* and >99.95% removal of oil with a TDS concentration of 5237 mg/L at a loading rate of 3.39 m/h. Interestingly, at 3.39 m/h *E. coli* removal in the presence of oil is higher than removal without oil (Figure 3B). The addition of *E. coli* in oil-water emulsions has been shown to coalesce oil and cause clustering among droplets, suggesting bridging between emulsion droplets.[61] This may allow a hydrophobic interaction between bacterial cell membranes and oil, creating larger negatively charged particles that interact with cationic MO proteins. This could explain the increase in the rate of *E. coli* removal as more bacterial cells interact with oil and MO proteins resulting in their retention on the filter. Control kapok columns do not provide a high rate of *E. coli* removal but can provide a high rate of oil removal with *E. coli* (99.74%) and without *E. coli* (99.6%). This work reveals the importance of MO proteins in increasing the rate of *E. coli* removal of kapok filters during simultaneous filtration. In comparison, GAC can remove up to 1 $\log_{10}$ of *E. coli*[62] and provide 90% oil removal,[63] while no work has reported on simultaneous removal. Microfiltration membranes for oil and bacterial separation from water demonstrated 85% oil rejection and 2 $\log_{10}$ *E. coli* rejection.[64] An overview of current technologies for oil and bacterial removal from produced water is shown in Table S3. Compared to similar technologies, functionalized kapok filters show increased rates of removal for both oil and bacteria under close to rapid sand filtration operating conditions.

MO-Coated Kapok Filters Provide a Sustainable Option for Oily Wastewater Treatment. MO-coated filters can simultaneously remove large amounts of bacteria and oil at a moderate salinity and a loading rate approaching RSF rates. This filter combines traditionally separate water treatment processes (disinfection and removal of organics) into one natural and sustainable technology. While MO-coated kapok can treat higher-salinity waters such as PW, it can also act as an oily wastewater technology for contamination mitigation, POU filters for disinfection, membrane pretreatment, and emergency water supply filters for lower-salinity waters. Overall, MO-coated kapok filters improve upon existing oil/water separation technology by providing concurrent removal of oil and *E. coli* while being comprised of completely plant-based components.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.estlett.1c00733.

Additional information about oil droplet size analysis, oil content method, SDS−PAGE method, proteomics facility results, SEM analysis and other data, including preliminary *E. coli* removal and MO serum coagulation ability, and a table comparing oil and bacterium removal technologies (PDF)

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Notes

The authors declare no competing financial interest.

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