

**BIODEGRADABLE NATURAL FIBRE-BASED COALESCENCE FILTER
AND SORBENT FOR OIL SEPARATION FROM OILY WASTEWATER**

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by

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Submitted

in fulfilment of the requirements of the degree of Doctor of Philosophy

to the



INDIAN INSTITUTE OF TECHNOLOGY DELHI

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.....dedicated to my beloved parents.

For their endless love, support, and encouragement.

&

*In loving memory of
elder brother (Late Shri Udai Bhan Singh)*

CERTIFICATE

This is to certify that the thesis entitled “**Biodegradable Natural Fibre-Based Coalescence Filter and Sorbent for Oil Separation from Oily Wastewater**”, being submitted by **Mr. Chandra Jeet Singh** to the Indian Institute of Technology Delhi for the award of the degree of **Doctor of Philosophy**, is a record of bonafide research work carried out by the candidate in the Department of Textile and Fibre Engineering, IIT Delhi. He has worked under my guidance and supervision and fulfilled the requirements for the submission of the thesis, which has attained the standard required for a Ph.D. degree at this institute.

The research work carried out to complete the thesis has not been submitted for the award of a degree or diploma in any institute in part or full.

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Chandra Jeet Singh

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ABSTRACT

Oil-in-water emulsion is released into the wastewater by many industries, frequently in quantities that are hazardous to the environment. Oily wastewater is treated using porous media like coalescence beds and membranes. Membrane filtration is limited to the removal of very fine droplets. Larger oil droplets lesser than 100 μm are removed from secondary emulsions using coalescence bed filters, which are made of either fibrous or granular packing. Fibrous media have greater surface areas and porosities than coarse granular media, which results in greater oil removal effectiveness. It is critical to comprehend the method of oil separation in order to develop a fibre coalescence bed filter that treats industrial discharge effectively.

Furthermore, porosity, bed height, pore size, and fibre diameter in relation to influent oil concentration and droplet size are interaction characteristics that influence the effectiveness of coalescence. Coalescence filtering performance is assessed using the oil concentrations and D50 (median value of droplet sizes at 50% in the cumulative distribution) droplet sizes in the influent and effluent. In this investigation, a non-ionic surfactant was used to create a stable emulsion of engine oil in water at a concentration of 10%. As filter beds, kapok fibres were utilised to separate the oil from the oil-water emulsion. SEM analysis was employed to study the surface morphology of fibres, and Fourier transform infrared (FTIR) analysis to investigate the chemical bonding of fibres. For the coalescence filter, three different bed heights (10 mm, 20 mm, and 30 mm) and four different porosities (0.90, 0.92, 0.95, and 0.98) were used to create kapok filter beds. The filter column was pumped with the oil-water emulsion (influent). The effectiveness of the oil separation process was assessed in terms of the droplet size (D50) change and oil concentration from influent to effluent. Apart from a porosity of 0.92, the separation efficiency increases with the

increases in porosity and bed height. At lower porosities, raising the bed height has no positive impact on the process efficiency. The highest filtration performance in terms of oil separation efficiency and D50 droplet ratio was achieved with 0.98 porosity and a bed height of 30 mm. When the bed height was increased from 10 to 30 mm at 0.98 porosity, the D50 droplet ratio decreased from 0.25 to 0.14, indicating a considerable reduction in the size of the droplets in the effluent and, consequently, an enhances the oil separation efficiency from 91.3 to 99.63 %.

Further, coalescence filters were prepared using milkweed fibres for oil separation from oil-water emulsion. The chemical groups, microstructure, morphology, and surface hydrophobicity of the fibres were analysed using FTIR, SEM, and contact angle techniques, respectively. A non-ionic surfactant was used to create a stable emulsion of engine oil in water. Deep bed coalescence filters were constructed at three filter bed heights (10 mm, 20 mm, and 30 mm) with a combination of three porosities (0.92, 0.95, and 0.98). Maximum filtration performance in terms of oil separation efficiency (99.73%) and D50 droplet ratio was achieved at a porosity value of 0.98 and bed height of 30 mm. Fouling was not observed while continuously operating the filter bed having 30 mm depth and 0.98 porosity for a long duration.

We investigated coalescence filtering on a bed produced from kapok and milkweed fibres and how they separated oil from emulsions at different porosities and bed heights. The ability of filter beds to separate oil improves with the increases in porosity and bed height. The filter bed with lower porosity saturates more oil in the bed. Increasing the bed height at lower porosity (0.92) could not significantly improve the oil separation efficiency, as the high-pressure drop eject the larger droplets into the effluent, coupled with fewer available fibre surfaces for the coalescence to take place effectively. The potential for using kapok and milkweed filters to coalesce fine oil droplets in stable oily emulsions is significant. The

separation efficiency for kapok and milkweed filter beds of up to 89.6% and 93%, respectively was achieved at a porosity of 0.98 and bed height of 30 mm. The lower amount of oil in the influent shows a higher separation efficiency because the filter bed retained most of the oil in its structure. The smaller size of the droplets (2 μm) makes it difficult for them to stick together inside the structure or intra-fibre pores. In this case, the separation efficiency decreases with the increase in porosity and bed height. When the droplets are 2 μm in size rather than 5 μm in size, the oil separation efficiency of kapok and milkweed filters is reduced.

Recently, there has been a lot of interest in using natural materials that can absorb a lot of oil as sorbents to clean up oil spills in seawater. Therefore, we developed a nonwoven sorbent; by utilizing a carding and needle-punching process, kapok fibres were combined in various ratios with cotton fibres received as blow room waste from the textile industry. We investigated the potential of kapok/waste cotton nonwovens for oil sorption, oil retention, and water sorption. Since the physical and chemical characteristics of the fibres affect their ability to absorb oil, FTIR, SEM, and surface contact angle measurements were performed. The behaviour of a nonwoven made of kapok and waste cotton with regard to oil absorption and oil retention was investigated. Blended nonwovens (kapok/cotton, 90/10) having an areal density of 150 GSM or g/m^2 exhibited maximum oil sorption capacities of 45.36 g/g, 43.97 g/g, and 39.92 g/g for engine oil, vegetable oil, and diesel oil, respectively. According to the findings of the reusability test, the oil absorbency of 150K/C 90/10 against engine oil, vegetable oil, and diesel oil was 42.55 g/g, 40.61 g/g, and 36.45 g/g, respectively, after ten cycles. A comparison of the kapok/waste cotton blends to absorb oil with that of other synthetic (Polypropylene) sorbents was also presented. Our research suggests that nonwovens made of waste cotton and kapok fibres could play a significant role in the cleanup of oil spills.

Finally, all significant findings discussed in earlier chapters are summarised, and the future scope of the research work is proposed.

सार

कई उद्योगों द्वारा पानी में तेल का पायस अपशिष्ट जल में छोड़ा जाता है, जो अक्सर ऐसी मात्रा में होता है जो पर्यावरण के लिए खतरनाक होता है। तैलीय अपशिष्ट जल का उपचार सहसंयोजक बिस्तरों और झिल्लियों जैसे छिद्रपूर्ण माध्यम का उपयोग करके किया जाता है। झिल्ली निस्पंदन बहुत महीन बूंदों को हटाने तक सीमित है। 100 माइक्रोमीटर से कम बड़ी तेल की बूंदों को कोलेसेंस बेड फिल्टर का उपयोग करके द्वितीयक पायस से हटा दिया जाता है, जो रेशेदार या दानेदार पैकिंग से बने होते हैं। रेशेदार मीडिया में मोटे दानेदार मीडिया की तुलना में अधिक सतह क्षेत्र और छिद्र होते हैं, जिसके परिणामस्वरूप तेल हटाने की प्रभावशीलता अधिक होती है। फाइबर कोलेसेंस बेड फिल्टर विकसित करने के लिए तेल पृथक्करण की विधि को समझना महत्वपूर्ण है जो औद्योगिक निर्वहन का प्रभावी ढंग से इलाज करता है। इसके अलावा, प्रवेशी तेल सांद्रता और बूंद के आकार के संबंध में सरंधता, बिस्तर की ऊंचाई, छिद्र आकार और फाइबर व्यास परस्पर क्रिया विशेषताएं हैं जो कोलेसेंस की प्रभावशीलता को प्रभावित करती हैं। कोलेसेंस फ़िल्टरिंग प्रदर्शन का मूल्यांकन तेल सांद्रता और D50 (संचयी वितरण में 50% पर बूंद के आकार का औसत मूल्य) और प्रवाह में बूंद के आकार का उपयोग करके किया जाता है। इस अनुसंधान में, एक गैर-आयनिक सर्फैक्टेंट का उपयोग करके पानी में इंजन तेल के स्थायी विमलन के लिए 10% सांद्रता में पायस बनाया गया। फिल्टर बेड के रूप में, कपोक रेशा का उपयोग तेल-पानी के पायस से तेल को अलग करने के लिए किया जाता था। एसईएम विश्लेषण को तंतुओं की सतह आकृति विज्ञान का अध्ययन करने के लिए नियोजित किया गया था, और फूरियर ट्रांसफॉर्म इंफ्रारेड (एफटीआईआर) विश्लेषण को तंतुओं के रासायनिक बंधन की जांच के लिए नियोजित किया गया था। कोलेसेंस फ़िल्टर के लिए, कपोक फ़िल्टर बेड बनाने के लिए तीन अलग-अलग बेड ऊंचाई (10 मिमी, 20 मिमी, और 30 मिमी) और चार अलग-अलग सरंधता (0.90, 0.92, 0.95, और 0.98) का उपयोग किया गया। फ़िल्टर कॉलम को तेल-पानी पायस (प्रवेशी) के साथ पंप किया गया था। तेल पृथक्करण प्रक्रिया की प्रभावशीलता

का मूल्यांकन बूंद के आकार (डी50) में परिवर्तन और प्रवेशी से प्रवाह में तेल की सांद्रता के संदर्भ में किया गया था। 0.92 की सरंध्रता के अलावा, सरंध्रता और बिस्तर की ऊंचाई में वृद्धि के साथ पृथक्करण दक्षता बढ़ जाती है। कम सरंध्रता पर, बिस्तर की ऊंचाई बढ़ाने से प्रक्रिया दक्षता पर कोई सकारात्मक प्रभाव नहीं पड़ता है। तेल पृथक्करण दक्षता और डी50 बूंद अनुपात के संदर्भ में उच्चतम निस्पंदन प्रदर्शन 0.98 सरंध्रता और 30 मिमी की बिस्तर ऊंचाई के साथ हासिल किया गया था। जब बिस्तर की ऊंचाई 0.98 सरंध्रता पर 10 से 30 मिमी तक बढ़ाई गई, तो डी50 बूंद अनुपात 0.25 से घटकर 0.14 हो गया, जो कि प्रवाह में बूंदों के आकार में काफी कमी का संकेत देता है और, परिणामस्वरूप, तेल पृथक्करण दक्षता 91.3 से 99.63% तक बढ़ जाती है।

इसके अलावा, तेल-पानी पायस से तेल को अलग करने के लिए मिल्कवीड रेशो का उपयोग करके कोलेसेंस फ़िल्टर तैयार किए गए। रेशो के रासायनिक समूह, सूक्ष्म संरचना, आकृति विज्ञान, और सतहीय जलरहितता का विश्लेषण क्रमशः एफटीआईआर, एसईएम और संपर्क कोण तकनीकों का उपयोग करके किया गया। एक गैर-आयनिक सर्फैक्टेंट का उपयोग करके पानी में इंजन तेल के स्थायी विमलन के लिए स्थिर पायस बनाया गया। तीन फ़िल्टर बेड ऊंचाई (10 मिमी, 20 मिमी, और 30 मिमी) के साथ तीन सरंध्रता (0.92, 0.95, और 0.98) के संयोजन से गहरे बेड कोलेसेंस फ़िल्टर बनाए गए। तेल पृथक्करण दक्षता (99.73%) और डी50 बूंद अनुपात के संदर्भ में अधिकतम निस्पंदन प्रदर्शन 0.98 के सरंध्रता मूल्य और 30 मिमी की बिस्तर ऊंचाई पर हासिल किया गया था। 30 मिमी गहराई और 0.98 सरंध्रता वाले फ़िल्टर बेड को लंबे समय तक लगातार संचालित करते समय कोई गड़बड़ी नहीं देखी गई। हमने कपोक और मिल्कवीड रेशो से बने बिस्तर पर कोलेसेंस फ़िल्टरिंग का अध्ययन किया और देखा कि कैसे उन्होंने अलग-अलग सरंध्रता और बिस्तर की ऊंचाई पर पायस से तेल को अलग किया। सरंध्रता और बिस्तर की ऊंचाई में वृद्धि के साथ तेल को अलग करने के लिए फ़िल्टर बेड की क्षमता में सुधार होता है। कम सरंध्रता वाला फ़िल्टर बिस्तर बिस्तर में अधिक तेल संतृप्त करता है। कम सरंध्रता (0.92) पर

बिस्तर की ऊंचाई बढ़ाने से तेल पृथक्करण दक्षता में उल्लेखनीय सुधार नहीं हो सका, क्योंकि उच्च दबाव वाली बूंद बड़ी बूंदों को, प्रभावी ढंग से कोलेसेंस के लिए कम उपलब्ध रेशो की सतहों के साथ मिलकर, प्रवाह में बाहर निकाल देती है। स्थिर तैलीय पायस में महीन तेल की बूंदों को एकत्रित करने के लिए कपोक और मिल्कवीड फिल्टर का उपयोग करने की क्षमता महत्वपूर्ण है। कपोक और मिल्कवीड फिल्टर बेड के लिए पृथक्करण दक्षता 0.98 की सरंध्रता और 30 मिमी की बेड ऊंचाई पर क्रमशः 89.6% और 93% तक हासिल की गई थी। प्रवेशी में तेल की कम मात्रा उच्च पृथक्करण दक्षता को दर्शाती है क्योंकि फ़िल्टर बिस्तर ने अपनी संरचना में अधिकांश तेल को बरकरार रखा है। बूंदों का छोटा आकार (2 माइक्रोमीटर) उन्हें संरचना या अंतः- रेशा छिद्रों के अंदर एक साथ मिलने में कठिनाई पैदा करता है। इस स्थिति में, सरंध्रता और बिस्तर की ऊंचाई में वृद्धि के साथ पृथक्करण दक्षता कम हो जाती है। जब बूंदें 5 माइक्रोमीटर के बजाय 2 माइक्रोमीटर आकार की होती हैं , तो कपोक और मिल्कवीड फ़िल्टर्स की तेल पृथक्करण दक्षता कम हो जाती है।

हाल ही में, समुद्री जल में फैले तेल को साफ करने के लिए सॉर्बेंट के रूप में प्राकृतिक सामग्रियों का उपयोग करने में बहुत रुचि देखी गई है जो बहुत सारे तेल को अवशोषित कर सकते हैं। इसलिए, हमने एक गैर बुना हुआ सॉर्बेंट विकसित किया; कार्डिंग और सुई-छिद्रण प्रक्रिया का उपयोग करके, कपोक रेशा को कपड़ा उद्योग से ब्लो रूम अपशिष्ट के रूप में प्राप्त कपास फाइबर के साथ विभिन्न अनुपातों में जोड़ा गया था। हमने कपोक/ अपशिष्ट कपास के मिश्रण से बने गैर बुने हुए कपड़े की तेल सोखने, तेल प्रतिधारण और पानी सोखने की क्षमता का अध्ययन किया। चूंकि फाइबर की भौतिक और रासायनिक विशेषताएं तेल को अवशोषित करने की उनकी क्षमता को प्रभावित करती हैं, इसलिए एफटीआईआर, एसईएम और सतह संपर्क कोण मापन का उपयोग किया गया। तेल अवशोषण और तेल प्रतिधारण के संबंध में कपोक और अपशिष्ट कपास से बने गैर बुने हुए कपड़े के व्यवहार की जांच की गई। 150 जीएसएम या ग्राम/मीटर² के क्षेत्रीय घनत्व वाले मिश्रित गैर बुने हुए कपड़े (कपोक/कपास, 90/10) ने अधिकतम तेल सोखने की क्षमता 45.36 ग्राम/ग्राम, 43.97 ग्राम/ग्राम और 39.92 ग्राम/ग्राम

क्रमशः इंजन तेल, वनस्पति तेल, और डीजल तेल के लिए प्रदर्शित की। पुनर्प्रयोग परीक्षण के निष्कर्षों के अनुसार, इंजन तेल, वनस्पति तेल और डीजल तेल के लिए 150K/C 90/10 की तेल अवशोषण क्षमता दस चक्रों के बाद क्रमशः 42.55 ग्राम/ग्राम, 40.61 ग्राम/ग्राम, और 36.45 ग्राम/ग्राम थी। कपोक/अपशिष्ट कपास मिश्रण की अन्य कृत्रिम (पॉलीप्रोपाइलीन) सॉर्बेंट्स के साथ तेल को अवशोषित करने के लिए तुलना भी प्रस्तुत की गई थी। हमारे शोध से पता चलता है कि अपशिष्ट कपास और कपोक रेशा से बने गैर बुने हुए कपड़े तेल रिसाव की सफाई में महत्वपूर्ण भूमिका निभा सकते हैं।

अंत में, पहले अध्यायों में चर्चा किए गए सभी महत्वपूर्ण निष्कर्षों का सारांश संक्षेप में प्रस्तुत किया गया है, और भविष्य के अनुसंधान कार्य के लिए प्रस्तावना प्रस्तुत की गई है।

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List of Abbreviations

SEM	Scanning Electron Microscope
FTIR	Fourier Transform-Infrared Spectroscopy
GSM	Gram Per Square Meter
PP	Polypropylene
<i>S</i>	Spreading Coefficient
K	Kapok Fibres
C	Waste Cotton Fibres
K/C	Kapok/Cotton Fibres Blended Nonwoven
D50	Median value of droplet sizes at 50% in the cumulative distribution
D50out	Median value of droplet sizes at 50% in the cumulative distribution in outlet
D50in	Median value of droplet sizes at 50% in the cumulative distribution in inlet
DWH	Deepwater Horizon
BP	British Petroleum
4D	Four-Dimensional
CCD camera	Charge-coupled Device
η	Oil Separation Efficiency
D_h	Hydraulic Inter-fibre Pore Diameter
d_i	Inner (lumen) Diameter
d_o	Outer (lumen) Diameter
D10	Median value of droplet sizes at 10% in the cumulative distribution
D90	Median value of droplet sizes at 90% in the cumulative distribution
SP	Smallest Pore () Diameter
MFP	Mean Flow Pore
NBS	N-Bromo Succinimide