Remediation of Oil Spills Using Pellets from Waste Papers and Kenaf (Hibiscus cannabinus L) fibres

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ORIGINAL RESEARCH

Abstract- The clean-up of oil spills from water bodies is a global problem. Enormous quantities of waste are generated from paper and paper products yearly, which constitutes to serious environmental pollution when disposed inappropriately. Consequently, it is essential to repurpose waste paper in a sustainable manner to address the remediation of oil spills. In this study, absorbent composite pellets of varying waste paper to kenaf fibers compositions were produced for oil remediation and evaluated for their physical (water absorption (WA) and thickness swelling), mechanical (modulus of elasticity (MoE), modulus of rupture (MoR) and impact strength) and remediation capabilities. The WA ranged from 404% to 546% with the highest obtained at 100% paper and the lowest obtained at 20% paper. Mechanical strengths increased with paper composition from 20% (2.19 N/mm^2, 0.08 N/mm^2, and 0.26 N/mm^2) to 100% (6.23 N/mm^2, 0.17 N/mm^2, and 0.86 N/mm^2) for MoE, MoR and Impact strength respectively. The pellets having 20% paper and 80% Kenaf pellet were recorded as optimum for the remediation of oil spills.

Keywords- Kenaf paper pellets, absorbents, remediation, oil spill cleanup, waste paper recycling

1 INTRODUCTION

Bioremediation involves using biological agents to clean the environment. This process involves using plants, trees, and microorganisms to degrade, reduce, or detoxify waste products and pollutants. An increase in water pollution due to oil spills has increased the toxic substances in the environment. Currently, the primary methods for managing oil spills include in situ combustion, chemical dispersants and flocculants, microbiological degradation, physical interception, and a variety of adsorbents (Hubbe et al., 2013; Xue et al., 2016). Adsorbents are sometimes the most effective and optimized treatment among these methods because other approaches may have detrimental effects on the environment, such as resource waste (in situ combustions), long treatment times (micro-biological degradation), and secondary pollution (chemical dispersants and flocculants) (Liu and Wang, 2019).

Mechanical, chemical, physicochemical, biological, and other techniques can be used to control spills (Wahi *et al*., 2013). According to Deymeh *et al*. (2012) and Seyedi *et al*. (2013), dispersants, sorbents, solidifiers, booms, and skimmers are frequently used. Skimmers are used to collect the oil, and booms are used to prevent it from spreading. Another option for collecting oil spills is to use sorbents (Hoang *et al*., 2018; Kukkar *et al*., 2020). A huge variety of natural and synthetic materials have recently been put to the test as potential oil sorbents (Okoh *et al*., 2019; Ossai *et al*., 2020). Currently, organic natural materials, organic synthetic products, and inorganic mineral materials are generally categorized as the most widely used oil-absorbing materials.

Inorganic oil absorption materials are inexpensive, but they have poor oil/water selectivity and reusability, which restricts their use. Because of their excellent oil/water selectivity, reusability, and absorption capacity, research on organic synthetic products has recently advanced quickly (Li *et al*. 2012; Pham and Dickerson 2014; Oribayo *et al*. 2017). However, the creation of organic synthetic products requires a complex and expensive process. The main problem with these products is that they cannot be broken down naturally, which frequently results in secondary pollution problems (Liu and Wang, 2019). As a result, scientists have gradually shifted their attention to biodegradable organic natural materials, such as biomass fibres, cotton grass mats, plant fibres, cotton grass fibre, and stalk.

The remediation pellets are designed to be buoyant, porous, bio-degradable, and non-toxic to ensure the effective clean-up of oil spills without adverse effects to the environment. Kenaf is a biomass, an annually renewable source of industrial fibre excellent for its natural absorbency. It has been explored in the remediation of spills (Oloruntoyin and Olayinka 2016; Siwayanan *et al*., 2020; Tan *et al*., 2021). In this research, kenaf particles are made into pellets using waste paper slurry as binder, thereby providing an alternative method of recycling waste papers. The use of paper and paperboard results in the annual production of tons of waste paper, with over 100 million tons of paper

waste generated worldwide (Guo, 2018). Conventional methods of disposing of paper waste are bad for the environment because they are frequently buried in landfills or burned in incinerator plants, which increases CO_2 emissions and may cause cancer (Kolajo and Odule, 2021). By preserving forest resources and maximizing the use of waste paper, recycling can support the development of a circular economy (Van *et al*., 2018). This waste paper can be recycled to address and resolve the issue of oil spills in water bodies in an environmentally friendly approach.

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Section D- MATERIAL/ CHEMICAL, ENGINEERING & RELATED SCIENCES
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Kenaf is a multipurpose annual herbaceous plant native to Africa and a member of the *Malvaceae* family (Ogunniyan, 2016). It is a plant of economic interest because it is drought tolerant and has a high cultivation density of up to 220,000.00 plants per ha (Asim *et al.* 2018). It is made up of a variety of beneficial parts, including stalks, leaves, fibers, seeds, fiber strands and some allelopathic chemicals (Akinrotimi and Okocha, 2018). The commercial uses of kenaf have expanded from its traditional use as rope and twine to include absorbents, paper goods, building materials, and animal feed. Its roots also contain a variety of microorganisms, and the symbiotic relationship between plant and microbes' aids in the remediation process (Arbaoui *et al*., 2013; Chen *et al*., 2018). Many metals with a high potential for toxicity, including Lead and Mercury (Hg), as well as kenaf's potential to stabilize cadmium in soils with low fertility, acidity, and sand content have already been studied. In wastewater treatment, the kenaf's extraction effect on chromium has also been reported (Abioye *et al*. 2012; Chen *et al*. 2017; Fitria and Dhokhikah 2019). It is on this basis that it is considered for remediation of crude oil spills on water bodies.

2 Materials and Methods

2.1 Material Collection and Preparation

Waste papers collected within the University of Ibadan were sorted to separate the waste print paper from other grades of paper. They were cleaned to remove contaminants like staple pins. For each specific ratio being investigated, the waste print paper was weighed and then manually shredded into smaller pieces. The shredded waste print paper was then soaked in water for 24 hours and then turned into slurry using a hydropulper. The slurry was further refined using a laboratory refiner to improve the bonding ability of the fibres by creating increased contact area and better fibre suspension in water. The kenaf stems were obtained from Ibadan metropolis were first manually reduced into chips and then into smaller particles using a hammer mill. The particles obtained were sieved and the acceptable size were of particles that could pass through a 2mm screen.

The research methodology employs a combination of quantitative and qualitative methods to investigate the production of these composite pellets. By varying the ratios between the waste print paper and Kenaf particles, the influence of these varying compositions on the properties and performance of the composite pellets are examined. The pellets are evaluated for their physical and mechanical properties, as well as remediation potentials. The waste paper to Kenaf particles composite pellets were produced in 5 distinct ratios (Table 1).

Table 1. Percentage composition of waste paper and Kenaf particles in varying ratios

	Waste papers (%)	Kenaf fibres (%)
Sample A	100	0
Sample B	80	20
Sample C	60	40
Sample D	40	60
Sample E	20	80

2.2 Formation Process of the Composite Pellets

For the purpose of creating the composite pellets, a mould and counter-mould were created. These were modified to create a manually operated device that was created for the manufacture of wall panels (Kolajo *et al*., 2020). The screw shaft lowering mechanism, support frame, mould, and counter mould make up the machine. The moulds and counter mould design for the pellet production are in Figs 1 and 2.

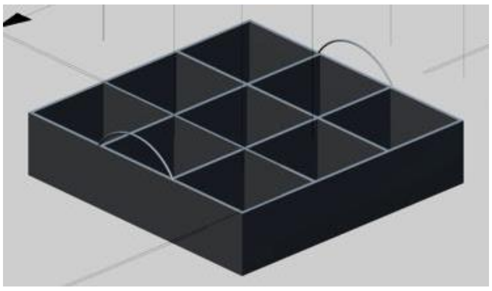


Fig. 1: Pellet Mould

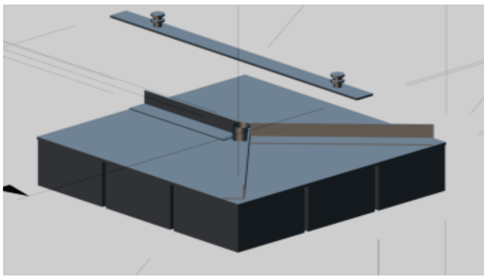


Fig. 2: Counter Mould

The refined waste paper slurry and the pre-weighed Kenaf particles are combined, ensuring a homogeneous mixture without any noticeable clusters. The mixture, well-blended is measured into each mould compartment, the counter mould is lowered unto the mould and compressed using a screw shaft. Water from the slurry is drained as the counter mould compresses the mixture to form the pellets. The pellets were air dried at room temperature for 3 days and thereafter detached from the moulds (Plate 1).

Plate 1: Composite Pellets Produced

2.3 Testing and Performance Evaluation

The composites produced were evaluated for their water absorption (WA) and thickness swelling (TS) properties according to the ASTM D570. The impact strength and flexural stress-strain test were conducted using the Instron Universal Testing Machine at the Forestry Research Institute of Nigeria (FRIN). Mechanical test was to evaluate the resistance of the pellets towards shocks and/or abrasion due to handling and transportation processes. The evaluation was as follows:

$$Water\ Absorption = \frac{W_2 - W_1}{W_1} \times 100\% \quad (1)$$

$$Thickness\ Swelling = \frac{T_2 - T_1}{T_1} \times 100\% \quad (2)$$

$$Stress = \frac{P}{A} \quad (3)$$

$$Strain\ (E) = \frac{\Delta L}{L} \quad (4)$$

$$MOE = \frac{S}{E} \quad (5)$$

$$MOR = \frac{3PI}{2PH^2} \quad (6)$$

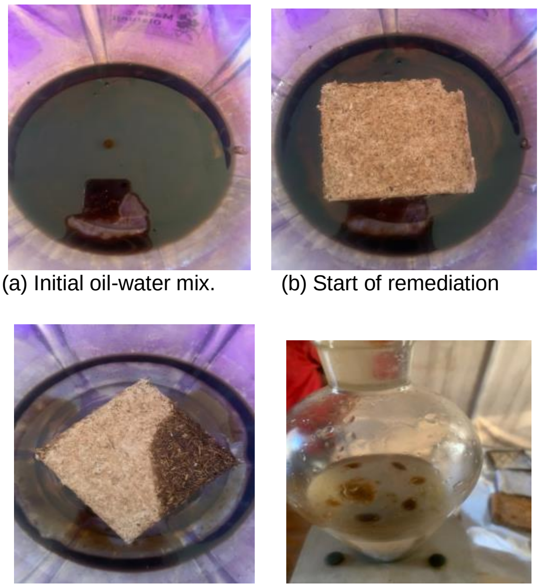
Where:
W_1 is the initial weight of composite
W_2 is the final weight after immersion T_1 is the initial thickness of the composite
T_2 is the final thickness after immersion
P = Applied Load (N)
A = Cross-sectional Area (m^2)
B = width (m)
L = Length (m)
ΔL = Change in length (m)
h = height (m)
y = deflection (m)

2.4 Remediation Test

The remediation test involves determining the oil extraction capacity of each pellet in parts per million (ppm) using the calorimeter extraction method. Crude oil would be obtained from Conoil regional office in Port Harcourt, Rivers State while 1,1,1-trichloroethane was obtained from a store in Ibadan. A control standard test was first conducted to determine the ppm value of oil in water and the procedure was repeated for the 5 pellet samples. A HACH DR/2000 Spectrophotometer was used for this experiment and the control test recorded 85 ppm of oil and water.

200 ml of water and 50 ml of crude oil was measured and stirred vigorously for 1 minute using a stirring rod to achieve proper dispersion. 25 g composite pellets sample was introduced into the oil-water mixture and left undisturbed for absorption and remediation for 5 minutes. The pellet was removed and the remaining solution transferred to a separating funnel. 35 ml of 1,1,1-trichloroethane reagent is added to the separating funnel, thoroughly mixed for about a minute and then left to settle for 10 minutes, allowing for separation based on density and immiscibility. Subsequently, 25 ml of the separated sample was released from the separating funnel into a collection bottle. The collection bottle, containing the extracted sample, was placed into a spectrophotometer to measure the absorbance of specific wavelengths of light. The parts per million (ppm) value, indicating the concentration of oil in the water sample, was recorded from the analysis of the sample.

$$Remediation = \frac{Change\ in\ oil\ content}{Initial\ oil\ content} \times 100\% \quad (7)$$



(a) Initial oil-water mix. (b) Start of remediation
(c) End of remediation (d) Remediated water

Plate 2: Pellet Remediation Process

3 Results and Discussions

3.1 Water Absorption and Thickness Swelling Results

The results of the water absorption (WA) and thickness swelling (TS) tests of the waste paper and Kenaf fibre composites are shown in Figs. 3 and 4. The pellets displayed great absorption capacity, absorbing more than four times their original weight. The WA ranged from 470 to 546% with the highest value recorded for Sample A and the lowest for Sample E. range in Fig. 3 shows the water absorption rate of the pellets reaching a pseudo-equilibrium state. This trend was noticed in TS which ranged from 3.7 to 42.8% (Samples E and A, respectively). The pellet composites with a higher percentage of paper generally recorded higher WA and TS, due to the high affinity of cellulose fibres for water (Ali and Sylvian, 2013; Etale *et al.*, 2023) and the absence of lignin which confers some hydrophobicity to lignocelluloses (Lisy *et al.*, 2021).

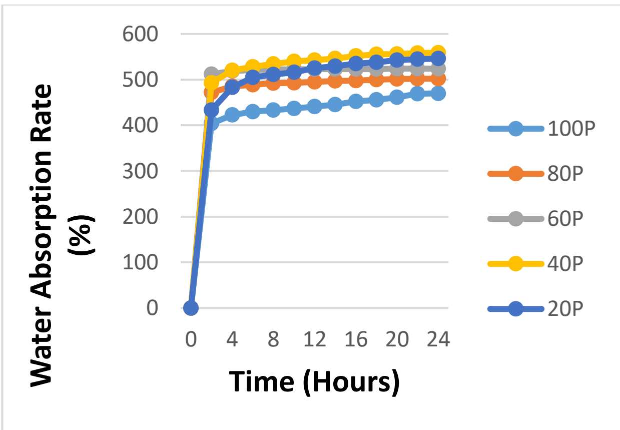


Fig. 3: Water absorption of composites

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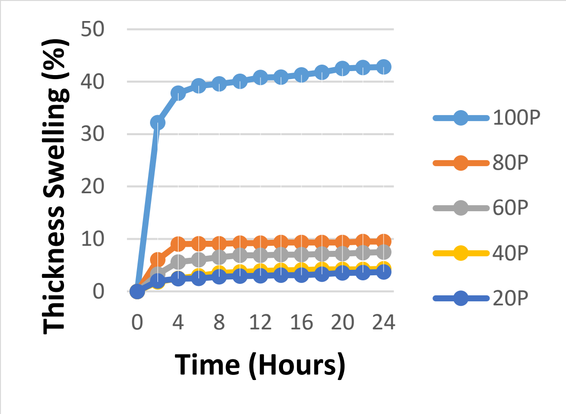

Fig. 4: Thickness swelling of pellets

3.2 Mechanical Tests Results

The Modulus of Elasticities (MoEs) ranged from 6.23 N/mm² to 2.192 N/mm², while the Modulus of Ruptures (MoRs) ranged from 0.17 to 0.08 N/mm². The obtained MoRs indicate that the composite pellets may not be suitable for load-carrying structural applications, but are within permissible strength for pellets (Kubik and Kažimírová, 2015). However, they show potential for use as insulating components like wall panels or ceiling boards. The impact strength results ranged from 0.86 N/mm² to 0.26 N/mm² for the varying paper-Kenaf ratios. Generally, it was observed that composite pellets with higher paper content exhibited higher MoRs, MoEs, and impact strength compared to those with higher Kenaf concentrations. This finding suggests that the structural integrity and inter-fibre bonding were enhanced through the refining process of the paper, leading to improved performance in the composite pellets.

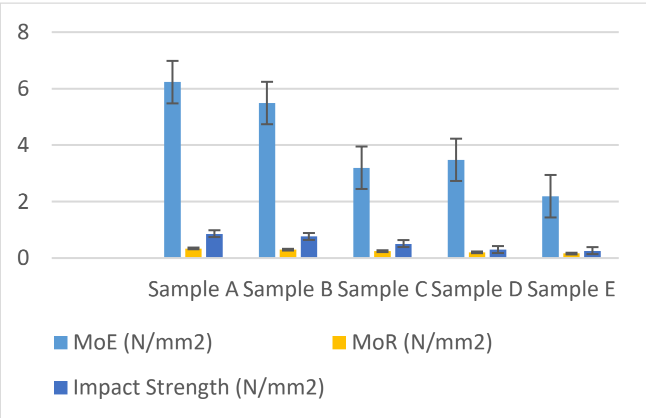

Fig. 5: Mechanical properties of Pellets

3.3 Remediation Test Results

It was observed that the oil extraction capacity varied depending on the composition of the pellets. As the concentration of Kenaf fibres increased in the composite matrix, there was a noticeable decrease in the oil remnants in the water. This suggests that the inclusion of Kenaf fibres enhances the oil absorption and separation process, leading to a lower oil concentration in the water after remediation. Among the tested ratios, Sample E demonstrated the highest oil extraction. These pellets recorded a change in oil content of 30ppm and a remediation capacity of 35.3%. On the other hand, Sample A exhibited the least oil concentration after the remediation process, with a recorded remediation capacity of 12.9%. However, these results show that waste papers also have the capacity for oil remediation, and has not been detrimental but complimentary to the remediation capacity of the kenaf fibres. Table 2 shows the recorded oil extraction capacities for each sample, as well as the percentage of oil extracted when compared to the initial control test of 85ppm oil in water. Fig. 10 depicts the increased remediation potential of the composite pellets with higher Kenaf fibre content.

Table 2. Remediation effects of the pellets

Samples	Unit of oil left (ppm)	Change in oil content (ppm)	Oil spill remediation (%)
Sample A	74	11	12.9
Sample B	67	18	21.2
Sample C	63	22	25.8
Sample D	61	24	28.2
Sample E	55	30	35.3

The pellets displayed buoyancy throughout the experiment by remaining afloat on the water surface, indicating their effective remediation properties (Plate 3). This floating behaviour is also advantageous as it allows for easy collection and removal of the pellets once the remediation process is complete. An interesting observation was also made regarding the thickness of the oil films in the separating funnel. As the concentration of Kenaf in the pellet mixture increased, there was a noticeable decrease in the thickness of the oil films. This observation suggests that the presence of Kenaf fibres in the composite matrix aids in reducing the thickness of the oil films, enhancing the separation efficiency and facilitating better oil extraction. These results and observations indicate the promising potential of the composite pellets for remediation applications, specifically in extracting oil from water sources.

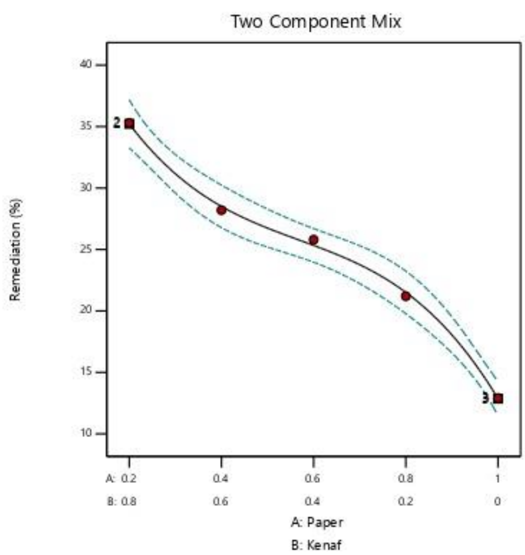

Fig. 6: Analysis of the pellets' remediation performance using Design Expert®

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4 Conclusion

The composite pellets produced from waste paper and Kenaf particles proved successful in the remediation of oil spills in water, with all test results indicating the pellets containing 20% paper and 80% Kenaf fibres is most recommended for efficient oil spill cleanup, attributable to the higher absorption capacities of kenaf fibres compared to fibres in the waste print papers. Therefore, remediation approach using composite pellets shows promise for the remediation of oil spills and can be further explored for other remediation applications, such as remediation of light and heavy oils in industrial waste water clean-up. The composites can be explored for non-structural in-door applications such as wall panels and room dividers.

Authors' Contributions; Kolajo, T.E.: Conceptualisation, Experimentation, Supervision, Writing – Review and Editing. Quadri, Q.L.: Experimentation, Writing – Original Draft

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