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Waste Paper-Sawdust Composites for the Clean-up of Oils with Varied Viscosities: An Experimental Study

[1]Kolajo T. E., [1]Erabhahiemen L. O. and [2]Kadiri A. O.

[1]*Department of Wood Products Engineering, University of Ibadan, Ibadan, Nigeria.*
[2]*Department of Agricultural and Environmental Engineering, University of Ibadan, Ibadan, Nigeria*

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Corresponding Author:
tolukolajo@yahoo.com

ABSTRACT

Over the years, oil spills have remained a predominant global occurrence. The improper disposal of dusts, flakes and shavings generated from the primary and secondary wood conversion processes, as well as waste paper and paper products has resulted in the release of greenhouse gases into the environment. In view of these environmental dangers, the use of waste papers and sawdust to remediate oil spills presents an ingenious way of utilizing their absorbent properties for environmental benefits. In this study, varying proportions (100:0, 80:20, 60:40, 40:60) of print waste papers and Gmelina arborea sawdust were made into pellets and evaluated for the clean-up of light, medium and heavy oils. The composite pellets were tested for their physical (Thickness Swelling (TS), Water Absorption (WA)) and their impact strength properties. The mean WA was 279.01%, showing excellent absorbent properties, while TS has a mean value of 11.79% and ranged from 8.13% to 16.67% across all samples. The mean low impact velocity strength was 16.64 N/m^2, which is considered adequate for normal handling conditions. The pellet composition of 100:0 had the highest remediation (21.43, 17.44 and 16.09%) while 40:60 had the lowest values (11.90, 12.79 and 9.2%) for light, medium and heavy oil viscosities, respectively. Composite pellets from waste papers and sawdust have potential in the clean-up of oil spills of varying viscosities, which can be applicable to other oil-based effluents from industries like paint and preservatives.

Graphical abstract

INTRODUCTION

Oil is one of the most common pollutants in the ocean, with an estimated three million metric tons entering marine environments each year (Thakur and Koul, 2022). Oil spills in water bodies have long posed serious environmental hazards, causing significant harm to ecosystems and human health due to the release of toxic substances. These spills involve crude oil or a spectrum of refined products such as lubricating oil, gasoline, diesel, jet fuel and kerosene (Hettithanthri *et al*., 2024), each with varying viscosities. Oil pollution can occur in both large and small quantities. In recent years, oil spills have continued to pose a global threat. In 2021, the MV X-Press Pearl spill off the coast of Sri Lanka released hundreds of tonnes of oil and chemicals, causing one of the worst marine disasters in the country's history (Mogollon *et al*., 2023). In 2022, a pipeline rupture in Peru spilled more than 11,000 barrels (~1.7 million litres) of crude oil into the Pacific Ocean, severely affecting marine biodiversity and local livelihoods, causing losses in millions of dollars (Jayathilaka *et al*., 2022). Another major incident occurred in 2023 in Nigeria's Bayelsa State, where an estimated 1 million litres of oil leaked from a wellhead over several weeks, impacting mangrove forests and aquatic life (Oborie *et al*., 2024). These recent spills reflect the ongoing frequency and severity of oil pollution and reinforce the urgent need for cost-effective, biodegradable remediation technologies.

Oil spilled in water undergoes a number of physical and chemical changes which includes spreading, dissolution, biodegradation, evaporation, formation of tar balls, emulsification, sinking, and photo-oxidation (Mogollon *et al*., 2023). These changes make cleanup efforts more complex. It is therefore essential to remove oil spills as quickly as possible. The main goals during an oil spill response are to prevent the oil from reaching the shoreline, reduce its impact on marine life, and accelerate the breakdown of any remaining oil. As oil spills continue, several methods have been employed in its clean-up and these have been grouped into four broad classes according to Dhaka and Chattopadhyay (Dhaka and Chattopadhyay, 2021). Physical/mechanical remediation which involves the removal of oil from water bodies by the use of materials such as sorbent materials for oil mop-up (Tuchman *et al*., 2018; Kolajo and Quadri, 2023), vacuum recovery using vacuum systems (Abidli *et al*., 2020), the use of booms to contain and barriers to control the spread of oil on water surface (Sharma *et al*., 2021), skimmers to remove oil from water surface (Suresh *et al*., 2020; Madhubashani *et al*., 2021). Chemical remediation makes use of dispersants and solidifiers for the breakup/solidification of the oil spills present (Hoang *et al*., 2018). Biological remediation can either be bioremediation which uses micro-organisms that accelerate the natural degradation of individual oil components or phytoremediation which uses plants to absorb and detoxify the polluted water. In-situ burning off of oil on water surface is another method employed for clean-up (Aurell *et al*.,2021). Other methods include emulsification (Balogun *et al*, 2025), the use of nanotechnology and the use of genetically engineered micro-organisms (Mishra *et al*., 2022; Rafeeq *et al*., 2023). The amount of oil spilt, its characteristics, the atmosphere and oceanic conditions (temperature, tidal wave pattern), and the threat the spill poses to human life, the environment, and sites of economic and cultural significance are some of the variables that affect the clean up of oil spills (Wang *et al*., 2021). However, the various methods of oil clean-up are also influenced by the viscosities of the oil to be remediated. For instance, booms, vacuum recovery, barriers and skimmers are more efficient for low viscosity oils but impractical for oils of high viscosities due to their thickness, stickiness and tendency to sink (Dhaka and Chattopadhyay, 2021). Chemical dispersants are more practical for low to medium viscosity oils, as these tend to form smaller droplets. Some oils with high viscosity may require pretreatment in form of heating and addition of surfactants before dispersants can be effective in their clean-up (Kalia *et al*., 2022). This is also true for bio- and phytoremediation methods, as microbes and plants are more likely to be effective against low- compared to high- viscosity oils (Moreira *et al*., 2021).

Sorbents have been identified as suitable for oils of all viscosities (Liu *et al*., 2022). These compounds use either adsorption or absorption to recover oil. They are employed as a principal recovery method for very tiny spills, as a backup to other containment methods like sorbent booms, to clean up the last remnants of oil spills on land or in water, and as a passive cleanup method. They are crucial to the cleanup process. Among all remediation options, sorbents have proven effective for absorbing oils of all viscosities (Hoang *et al*., 2021). These materials work through absorption or adsorption and are used in various capacities, including mopping up residual oil, supporting other containment systems, managing small spills, or serving as a passive remediation solution. Due to their high selectivity of water and oil, rate of recyclability and their absorption ability, in-depth research has been carried out on organic synthetic materials over the years (Ouyang *et al*., 2023). Veering from normal assumptions and trends, outcomes of research carried out on organic synthetic materials produced no significant results in aspects such as cost as they remained expensive to produce and the production process remains complicated, with the emergence of secondary pollution as the materials cannot be naturally degraded even after use (Liu *et al*., 2020). In a bid to staunch the effects associated with some of the current alternatives available in the oil spill clean-up methodologies, there has been a change in the type of materials used.

The search for affordable, abundant, and efficient materials to act as oil spill sorbents in water is currently of increasing interest, with specific emphasis on natural organic sorbents primarily derived from agricultural wastes. Numerous cellulosic materials, including cotton (Cao *et al*., 2017), palm fibres (Abdelwahab *et al*., 2017), rice husks (Kenes *et al*., 2012), Wheat straw (Ly *et al*., 2017), Loofa (Annunciado *et al*., 2005) and Sugarcane bagasse (Behood *et al*., 2016) have shown promise as effective in oil spill treatment. Other biodegradable organic natural materials such as stalks, plant fibres, water hyacinth and sawdust are now serving as replacements (Ouyang *et al*., 2023; Khondoker *et al*, 2024). These materials offer significant advantages with respect to environmental compatibility, biodegradability, and cost-effectiveness, making them attractive options for sustainable oil spill remediation strategies (Subramoniapillai, and Thilagavathi, 2022). Among these natural materials, paper pellets and sawdust represent particularly promising candidates due to their unique structural properties and availability. Papers have a porous structure making them a suitable sorbent material and can be adapted for oils of different viscosities. Sawdust is a by-product of wood-working operations composed of small chippings of wood. It is an organic porous material allowing for the absorption of oil and pollutants that contaminate waters with oil spills. It is a versatile material that is easy to obtain and possesses properties making it ideal for the removal of pollutants from environments affected by oil spills. The use of organic biodegradable waste materials in the production of pellets for oil spill remediation presents a cost-effective and environmentally friendly option. Paper-sawdust composite pellets can be shaped to suit specific remediation needs such as river channels and canals where heavy equipment may be impossible. Additionally, they can be tailored through additives and various treatments to enhance absorption and suit specific remediation purposes. It presents a recycling opportunity for large volumes of waste papers and sawdust which would otherwise end up in landfills. There is the opportunity of oil recovery from the pellets, thus presenting reduced environmental impact when compared to non-biodegradable sorbents.

MATERIALS AND METHODS

Sample collection

Waste print papers were sourced from the University of Ibadan and sorted into different grades. Non-paper elements such as staples, cords, and spiral binding spines were carefully removed to ensure material homogeneity. *Gmelina arborea* sawdust particles were obtained from a local sawmill.

Sample preparation

The sorted papers were then shredded, weighed, and soaked in water for 24 hours to facilitate pulping. The soaked papers were refined at 50% consistency using a mechanical pulp refiner in accordance with TAPPI T248 standards. This step was essential to improve the formation of composite pellets by ensuring that the paper pulp could effectively fill the void spaces within the sawdust matrix.

The sawdust was cleaned by physical inspection to remove visible impurities and then sieved. Only particles sizes between 2.00 - 3.35 mm sieve were selected for pellet production to ensure uniformity in particle size. Three types of oils with varying viscosities were used in the study: light oil (premium motor spirit/gasoline), medium oil (SAE 20 engine oil), and heavy oil (thick palm oil), as categorized in Table 1. These variations in oil viscosity were selected to represent a wide range of oil types encountered in real-world spill scenarios, including spills from industrial processing facilities that use specific oils. To enhance the binding strength of the composite pellets, starch was incorporated as a natural binder during pellet production. A combination of quantitative and qualitative research methods was employed to investigate the production of the remediation composite pellets. These were done to control and measure the ratios of each component of the composite pellets (quantitative method). The ratios of waste paper and *Gmelina* sawdust are intentionally varied in order to explore the influence of the varied compositions on the performance and properties of the composite pellets. Qualitative methods made use of visual and mechanical observations for the evaluation of composite pellets in order to evaluate their physical properties such as compatibility with oil, absorption, buoyancy and porosity. The different varying combination ratios of waste papers and *Gmelina arborea* sawdust in the composite pellets are as given in Table 2.

Production of composite pellets

The mould used for the production of the composite pellets is a modified design of a machine intended for the production of wall panels.

Table 1: Categories of Oil Viscosities

Oil viscosity	Light	Medium	Heavy
Viscosity range (Ns/m^2)*	< 0.01	0.01 – 0.1	> 0.1
Type used**	PMS (Gasoline)	SAE 20 (Engine oil)	Heavy palm oil

* Ilyin *et al.*, 2016 **Keshvadi *et al.*, 2011; Baird *et al.*, 2022; *Alimova et al.*, 2022

Table 2: Ratio of Waste Paper and *Gmelina* arborea Particles in the Pellets

S/N	Waste paper (%)	*Gmelina arborea* sawdust (%)
Sample A	100	0
Sample B	80	20
Sample C	60	40
Sample D	40	60
Sample E	20	80

Starch binder constitutes 40% of the composite mix

The machine is manually operated, containing the mould and counter mould, compacted using a screw mechanism (Kolajo *et al*., 2020). The composite mix was formed in the mould, the counter mould is lowered and force is manually exerted by the counter mould, aided by the screw shaft, with an average force of about 70MPa. Each pellet mix contains an average of 300g, with an additional 40g of starch as the binder. Excess water was drained and the pellets were compacted by the counter mould. The pellets were air-dried for a period of 3-4 days at room temperature.

Composite Testing

Water absorption and Thickness swelling tests

Water Absorption (WA) capacity was determined to evaluate the ability of the composite material to absorb water over a specific period, while the Thickness Swelling (TS) test measured the increase in the thickness of the composite when exposed to moisture. These tests are essential indicators of the dimensional stability and durability of the composite under wet conditions. Both tests were in accordance with ASTM D570 standards, using distilled water. Water absorption was calculated using:

$$WA = \frac{W_2 - W_1}{W_1} \times 100 \quad (1)$$

Where: W_1 = initial weight, W_2 = final weight

Thickness swelling was evaluated as:

$$TS = \frac{T_2 - T_1}{T_1} \times 100 \quad (2)$$

Where: T_1 *= initial thickness,* T_2 *= final thickness*

Low-impact velocity test

The low-impact strength test was determined using a Universal Testing Machine (Instron-3369) in accordance with the ASTM D4812-19 standards. This was necessary to evaluate the impact strength of the pellets and their capacity to absorb energy at low velocity in the process of fracture during transportation and when deployed for remediation. All tests were in three replicates and the impact strength was evaluated as:

$$Impact\ Strength = \frac{Energy\ of\ fracture\ (joule)}{Cross\ sectional\ area\ (m^2)} \quad (3)$$

Fourier Transform Infrared Spectroscopy Analysis

The test was carried out using the Perkinelmer UATR Spectrum Two Spectrometer. The FTIR test was carried out to investigate the molecular and chemical composition of the composite pellets, and detect the presence or otherwise of toxic substances in the pellets. The samples were exposed to different wavelengths of infrared light, using a spectra range between 400 and 4000cm^{-1} to measure the wavelengths absorbed. Each pellet composition and the sawdust sample were ground into fines and the raw absorption data was obtained from one gram each of the specimen pellet fines and the Fourier transform was used to generate the absorbance spectrum.

Remediation Test

The remediation test comprises estimating in parts per million (ppm), the oil absorption capacity of each pellet using the colorimeter extraction method. The reagent (1-1-1 trichloroethane) was used. Oils used in the remediation process were purchased from local outlets in Ibadan, Nigeria. A HACH DR Spectrophotometer of 320-1100 nm wavelength was used for obtaining the ppm values and determining the quantity of oil left in the simulated water body after remediation. A controlled standardised test was held to ascertain the ppm value of oil in water and this procedure was repeated for each of the samples to determine from which ratio the best remediation results emerged, the controlled test recorded values of 84 ppm, 86 ppm and 87 ppm for light oil, medium oil and heavy oil, respectively. Other materials used in this procedure included distilled water, 500 mL separatory funnel, 25 mL collection bottles, a timer and clamps.

A 200 mL of water and 20 mL of oil were mixed together and stirred thoroughly with a stirring rod for proper oil dispersion in the oil-water mixture. Pellets weighing 25 g each then were introduced into the mixture and left untouched for a period of 5 min for remediation of oil. After the remediation period, the remaining water-oil mixture was transferred into the separatory funnel and the reagent was introduced for the separation of oil and water. Vigorous shaking of the mixture ensued, followed by the 10 min of period of rest to allow for the separation of different phases based on immiscibility and density of the concerned liquids. 25 mL of the separated liquid was transferred to a collection bottle which was placed into the spectrophotometer to determine its absorbance of specific light wavelengths. The ppm values obtained indicate the concentration of oil in the water sample.

$$Remediation = \frac{change\ in\ oil\ content}{initial\ oil\ content} \times 100 \quad (4)$$

Results and Discussion

The pellets produced are shown in Figure 1, produced with the compositions defined in Table 2.

Sample A

Sample B

Sample C

Sample D

Sample E

Figure 1: Air-dried composite pellets of varying compositions

Water Absorption and Thickness Swelling Results

The WA and TS capacities of the waste paper and *Gmelina* sawdust fibre composites are shown in Figures 1 and 2, respectively. The pellets demonstrated a high absorption capacity, absorbing more than three times their initial weight which is due to the hygroscopicity of both paper and wood fibres. This observation aligns with findings in previous studies (Kolajo and Quadri, 2023; Aziz *et al*., 2023).

The WA data indicate that the pellets reached a pseudo-equilibrium absorption state. There was a gradual increase in the WA with increase in paper content, with the highest absorption recorded in Sample A (342.15%) and the lowest recorded for Sample E (228.68%). This is attributable to the presence of lignin in sawdust, which confers some hydrophobicity on the pellets as the sawdust content increases. The TS values ranged from 4.37% to 13.16% across the samples. The overall high absorption capacities of the composite pellets confirm their suitability for absorbing oil in water. Pellets with higher proportions of waste paper generally showed increased WA and TS values, indicating improved interfacial bonding between homogenous paper fibres.

Additionally, paper fibres are pretreated wood fibres containing cellulose which has high affinity for water. This, coupled with high surface area makes it more susceptible to WA and TS.

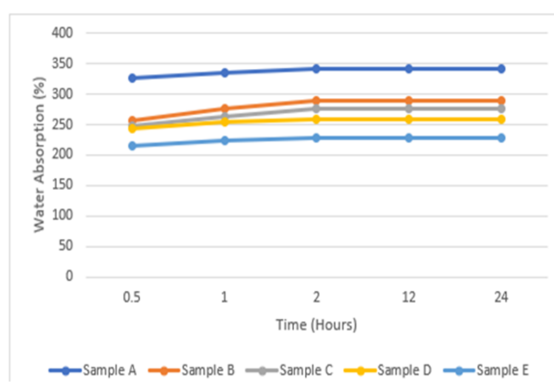


Figure 2: Water absorption across pellet compositions

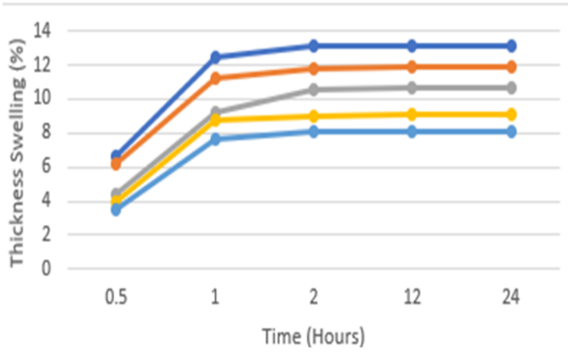


Figure 3: Thickness swelling across pellet compositions

Impact Strength Test Results

The velocity impact was evaluated and the results are presented in Figure 3. The impact strength results ranged from 14.27 N/m² to 18.71 N/m² (Samples A to E respectively) for the varying pellet composition ratios. This is comparable to values obtained by Kolajo and Quadri (2023) in the production of pellets from waste papers and Kenaf fibres. Based on the obtained results, it can be inferred that the highest values of the impact strength were observed in Sample A pellets, and the lowest values were recorded in Sample E pellets. Overall, composite pellets with higher paper content had higher impact strength values than those with higher Gmelina sawdust content. This shows that the structural integrity was increased as a result of inter-fibre bonding, the homogeneity of the paper composite, and the presence of starch in the composite, all of which led to higher performance in the composite pellets. These were absent in the sawdust composites.

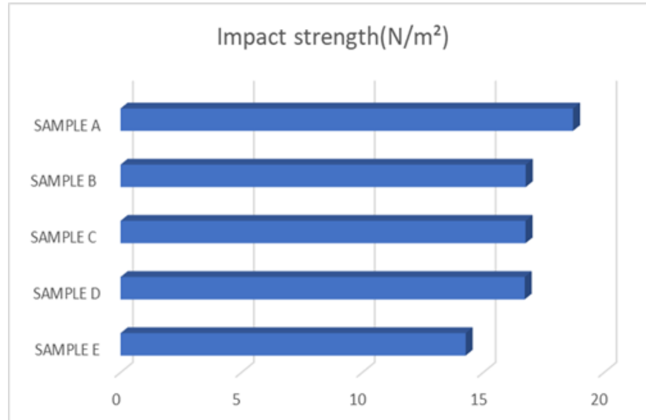
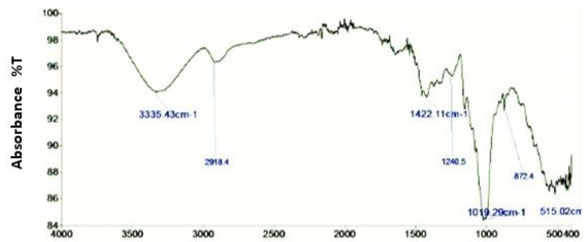


Figure 4: Low-impact Velocity Strength Test Result

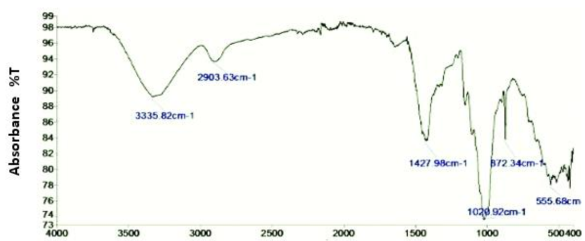
FTIR Results

The infrared spectrum of the pellet composites indicated absorption bands at 3335.82 and 3333.97 cm⁻¹ corresponding to O-H stretching vibrations which alludes to the ability of the glue to bond with other substances (Siddique, 2024). The alkane C-H stretching vibration occurred at 2924.90 and 2903.63cm⁻¹ which alludes to the strength, stiffness and the ability of the material to undergo further modification. The alkene C=C occurred at 1641.88 cm⁻¹. The aromatic C-C stretch in rings correspond to the positions 1427.98 and 1417.05 cm⁻¹. The C-O stretching vibration occurred at 1240.50 and 1014.76 cm⁻¹ which alludes to the biodegradability of the material and the mechanical strength. The aromatic C-H bend occurred at 872.40 and 872.29 cm⁻¹ while the C-Br stretching vibrations occurred at 555.68 and 515.02 cm⁻¹ respectively. These results are similar to those obtained by Kolajo and Quadri (2023) and in consonance with Nandiyanto *et al.*, (2019). The infra-red bands for the different ratios of the pellets are in Figures 4a to 4e. Figure 4f represents the infrared spectrum of sawdust fibres.

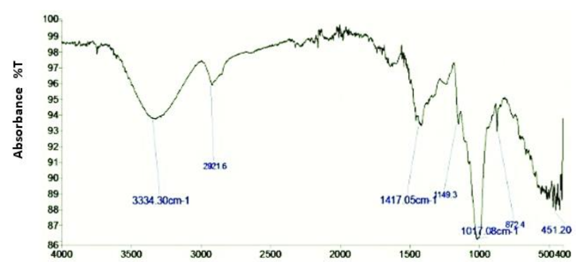
(a) Wavelength cm^{-1}



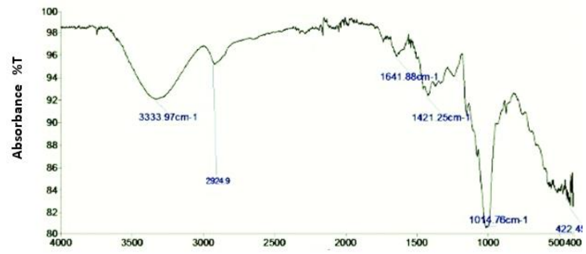
(b) Wavelength cm^{-1}



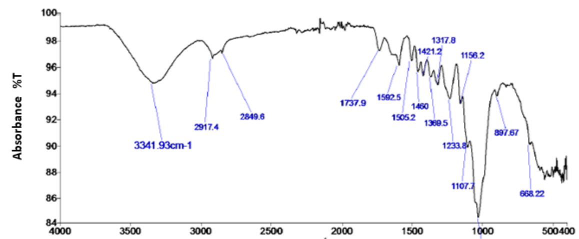
(c) Wavelength cm^{-1}



(d) Wavelength cm^{-1}



(e) Wavelength cm^{-1}



(f) Wavelength cm^{-1}

Figure 5: Infrared bands for waste paper to sawdust in ratio (a) 100:0 (b) 80:20 (c) 60:40 (d) 40:60 and (e) 20:80 (f) 100% Sawdust

Remediation Test Results

The composite pellets demonstrated buoyancy throughout the duration of the experiment, remaining afloat on the water surface. This flotation behaviour not only indicates effective oil remediation capabilities but also highlights the ease with which the pellets can be retrieved from water bodies after the clean-up process. A notable trend was observed with increasing waste paper fibre concentration in the composite matrix: there was a corresponding decrease in the residual oil content in the water. This suggests that the inclusion of waste paper fibres enhances the oil absorption efficiency of the pellets, thereby improving the separation process and resulting in lower oil concentrations post-remediation. Among the samples tested with light oil, Sample A displayed the highest percentage of remediation, with a reduction in oil concentration of 18 ppm and a remediation capacity of 21.43%, while Sample E had the lowest, with 11.9% remediation capacity (Figure 6). This trend is similarly observed in medium and heavy oil viscosities. This may be attributed to the higher surface area of the paper fibres compared with the sawdust fibres, as materials with greater surface area have higher sorption capacities. Furthermore, the presence of lignin in the sawdust fibres is known to repel penetration of the fibres by oil and water. When compared to other lignocellulosic materials such as kenaf fibres, which have been used in the remediation of crude oil spills, *Gmelina arborea* sawdust fibres exhibit relatively lower oil absorption capacities (Kolajo and Quadri, 2023). This finding suggests that the absorptive properties of lignocellulosic materials differ by species, with kenaf fibres outperforming *Gmelina arborea* in oil absorption efficiency. This may be explained by the chemical and morphological differences between the fibre species, especially in the content of the hydrophobic and oleophobic lignin, which prevents penetration of the fibres.

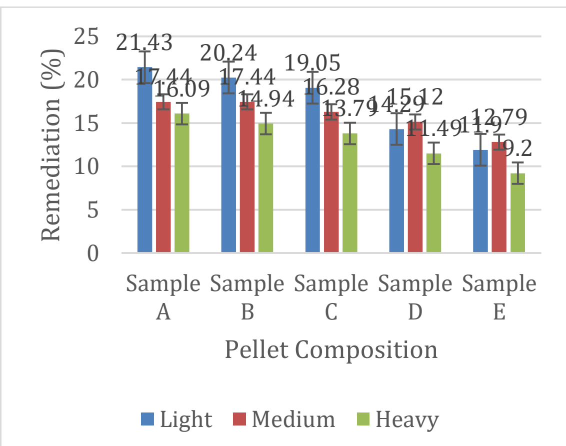


Figure 6: Pellets remediation performance in different oils

Conclusion

This study evaluated the absorptive capacities of composite pellets made from waste print papers and *Gmelina arborea* sawdust for the remediation of oils with light, medium, and heavy viscosities. The different pellet compositions demonstrated oil absorption capabilities across the range of oil viscosities tested. The highest remediation performance was observed in 100:0 pellets composed entirely of waste print papers, showing superior absorption capacity of paper fibres relative to the *Gmelina arborea* sawdust, attributable to the higher surface area of the paper fibres. The impact resistance tests confirmed that the composite pellets can withstand mechanical stresses typically encountered during handling, packaging, and transportation, which also ensures compact handling during remediation. Additionally, the pellets were lightweight and exhibited buoyancy throughout the remediation process, facilitating easy recovery from water bodies. This composite pellet developed has presented a cost-effective, biodegradable, and environmentally sustainable approach for oil spill clean up, and has been found suitable for oils of different viscosities.

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