

REMEDIATION OF COPPER CONTAMINATED SOIL WITH SPENT ENGINE OIL USING *LUFFA CYLINDRICA*

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Abstract. Human activities have caused several pollution activities. It has been established over the years that the remediation method has been successfully utilised to remove these heavy metals. Copper contamination has shown different levels of contamination in the soil over time. Via the batch method, the fibrous sponge of *Luffa cylindrica* (LFC) was used as an adsorbent to determine its potential to remove copper from spent engine oil (SEO). The characterization results show the adsorption peaks at 3294 cm^{-1} , 2922 cm^{-1} and 2855 cm^{-1} by the presence of bonded hydroxyl groups and the C-H bond in the methyl group on the surface of *Luffa cylindrica* respectively. Different masses of *Luffa cylindrica* fiber were used to assess the effect on the adsorption process. The adsorption capacity of 47.2% at 0.056ppm using 3.0g of *Luffa cylindrica* (LFC) for 4 hours, gave the optimum removal of copper. The results obtained demonstrated adsorption of copper on to the surface of *Luffa cylindrica*. The sample plots of different levels of copper in soil contaminated by 30ml of spent oil in three different locations, were created using a randomized technique along a transect at 10-inch intervals: designated as treatment SA-24H, SB-48H, and SC-72H. For each treatment, a composite soil sample was obtained at depth of 10 cm. The highest and lowest values SD (0.21ppm) and SA (0.05ppm), were recorded respectively. *Luffa cylindrica* is a potential adsorbent for the effective adsorption of copper from spent engine oil and can be successfully utilized for remediation of copper contaminated soil.

Keywords: Soil contamination, *Luffa cylindrica*, copper, adsorption, spent engine oil

1. Introduction

Pollution occurs via contamination. Agricultural land products produced from heavy metal ion-contaminated soil led to the accumulation of metal ions, and therefore, are not consumable due to the detrimental issues to human health and food safety (Tripathi et al., 2023). Soil contamination can be introduced into the soil by anthropogenic and natural processes, which affect its chemical properties. It is mandatory to choose the best remedial option for remediation of soil contaminants as these inorganic and organic pollutants pose extreme risk to human health (Murtaza et al., 2014). This consists of chemical, biological, physical, or radioactive elements that, in high enough concentrations, indirectly affect living things by contaminating their food, water, soil, or air. As a result of the natural and manmade sources, such as of volcanic eruptions and the soil weathering of its original components, contaminants are released into the soil (Garrett 2000). Iron, zinc, manganese, chromium, lead, cobalt, copper, vanadium, nickel, mercury, and cadmium are the major trace metals found in the dust crust; however, their occurrence vary widely across the earth's surface (Garrett 2000). Moreover, it has been discovered that metal concentrations rise with decreasing particle size, particularly in severely worn and wind-eroded soils (Meza-Figueroa et al., 2009). High concentrations of lead, zinc and copper are produced during Asian dust storm episodes, demonstrating the significance of these

events in the geochemical cycling of trace metals. Heavy metals and metalloids, such as Cu, Zn, Cd, Pb, Ni, Cr, and As, are the main inorganic contaminants. They generally originate from industrial waste and domestic rainwater, including Cu and Pb leaks. Heavy metals interact with or adhere to building particles during primary treatment, allowing them to enter the sludge; during the second phase, they attach to the cell walls of bacterium and concentrate within the cells of bacteria. The detriment of the heavy metal loading in sewage sediment has led to the creation of guidelines and laws in various parts of the world. The maximum allowed permissible limits of metal concentration in biosolids serve as the foundation for these rules and regulations. Heavy metal toxicities limiting crop growth in biosolids-amended soils are extremely uncommon when agronomic rates are applied (Murtaza et al. 2010). Yet, a significant detrimental effect is that heavy metals could build up in edible plant parts, penetrate the food chain, and depict deleterious effects on animals and humans.

It has been established over the years that the adsorption method has been successfully utilised to remove these heavy metals and organic dyes from aqueous solutions of wastewater. Adsorption is frequently used in water treatment plants to remove heavy metals. Adsorption has been utilized in the removal of heavy metals and dyes from industrial wastewater, recovering dyes from dilute solutions, purifying petroleum oils, inactivating bacteria toxins, hormones, purifying motor spirits, clarifying sugar liquid, softening hard water, drying, decolorizing, and purifying gases (Singh et al., 2005).

Due to sustainable adsorbents, such as *Luffa cylindrica*, which has been utilized to decrease the pollutants in chemical and biological sludge, give a high efficiency process, has potential for metal recovery, flexibility, and straightforward design, metal ions can be removed from the solution even at extremely low concentrations (as low as 1 mg/L), according to Ahalya *et al.* (2003) (Ahalya *et al.*, 2003; Khan *et al.*, 2008). Remediation of contaminated soil is an important factor in environmental study (Li *et al.*, 2022). It has been discovered that adsorption is a more effective technique.

The effective adsorbent is a plant *Luffa cylindrica*, a member of the Curcubitacea family. In Figure 1, its fruit is depicted (a). The sponge gourd also contains an exterior mat core and an interior fiber core, both of which are depicted in Figure 1 (b). Common sponges have sizes ranging from 15 to 25 cm to 1.20 to 1.50 m. (Boynard and Almeida 2000). In the quest for sustainable materials, novel materials have increased the significance of the luffa sponge material. The Luffa sponge material depicts excellent strength, stiffness, and energy absorption capabilities under quasi-static stress (Shen *et al.*, 2012), it possesses characteristics such as cellulose (55–90%), hemicelluloses (8–22%), lignin (10–23%), extractives (3.2%), and ash (0.4%), making suitable for a variety of applications. It is a good alternative for usage as a reinforcing material in a polymer matrix because of this property (Seki *et al.*, 2012). This current research indicates that it might be a suitable substitute material for packaging, water absorption (Bal *et al.*, 2004), wastewater treatment (Demir H. et al., 2006), and other applications (Song et al., 2017). The luffa fibers have also been used in biotechnology to immobilize cells and as a reinforcing fiber for other materials (Ghali et al., 2009). These results suggest that luffa fiber's full potential has not yet been completely realized. The need for sustainable solutions has led to employing novel materials and has increased the significance of the luffa material.

Engine oil as an operational fluid is frequently used in cars and serves as a lubricant for spark-ignition and self-ignition engines. Its function is to dissipate heat, create an engine seal and to clean and protect the metal parts of the engine from corrosion (Landowski and Baran, 2019). Spent engine oil, used engine oil, indicate the value changes in the oil rheological parameters. This spent engine oil will show a change in the viscosity index and presence of metal ions. The pollution of soil with spent engine oil, oil waste derived from engine oils used in cars, in automobile stations has negative effects on agriculture and the economy.

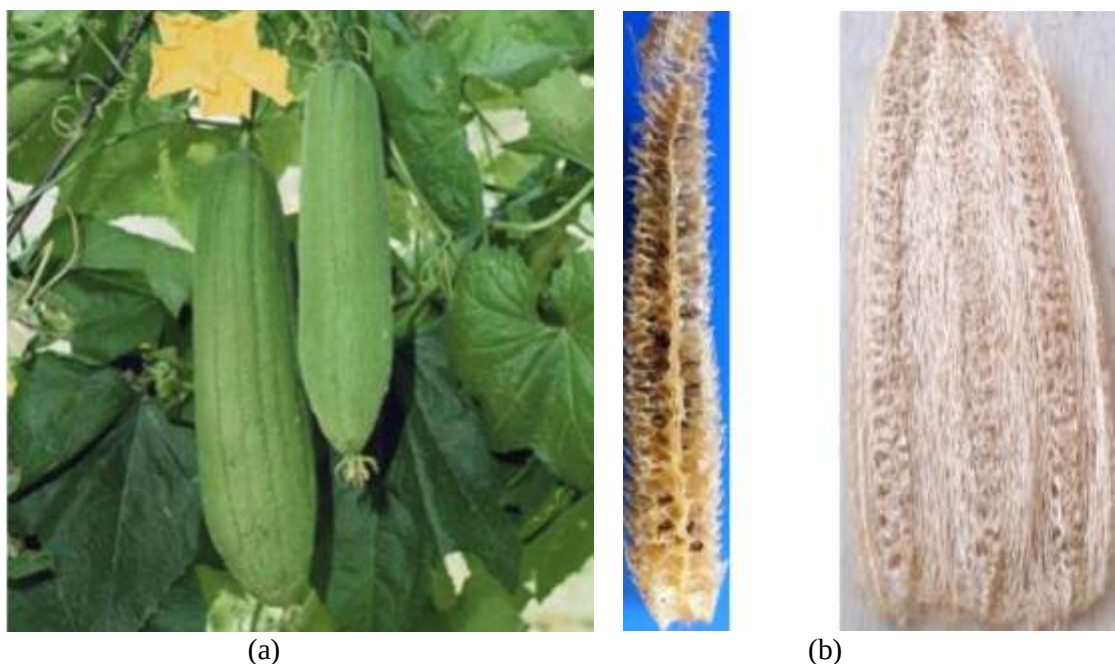


Figure 1 *Luffa cylindrica* fruit plant (a) Inner fiber and external core (b)

This research will determine the contamination of copper in spent engine oil in soil at different depth and time duration; and investigate the removal capacity potential of the adsorbent, *Luffa cylindrica*, and its soil remediation properties. The novelty of the research focuses on the analysis of adsorption of copper ions as a contaminant in soil over a duration of time.

2. Materials and Method

2.1 Preparation of sample

10-30ml of Multigrade Premium engine oil was used having a viscosity index of 127 and flashpoint 234°C. During digestion, 10ml of nitric acid (HNO_3) and hydrogen peroxide (H_2O_2) were used and 500ml of distilled water was used to dilute the samples until neutral. Loofa (*Luffa cylindrica*) was obtained from Nigeria, and the seeds were removed and the loofa was cut into large pieces, washed with distilled water several times until the pH turned neutral and air dried for 24 hours. The cut loofa was grinded and sieved to 425 micrometer size. 30ml of spent engine oil was digested by using the nitric acid which was heated to 120°C on a hot plate. Hydrogen peroxide was then added and heated for 6 hours.

2.2 Characterisation of *Luffa Cylindrica*

Fourier Transform Infra-red (FT-IR) Spectrometer utilised from Kaduna Institute Laboratory was used to detect the functional groups that interact with copper ions. 1.0g of dried *Luffa Cylindrica* were prepared prior to the FT-IR analysis. Measurements were done by 16 scans between the range 4000 cm^{-1} to 1000 cm^{-1} .

2.3 Adsorption studies

By batch method, 0.5g, 1.0g, 2.0g and 3.0g of processed loofa was immersed in 30ml of spent engine oil in an 100ml conical flask, the mixture was shaken for 4 hours at 600rpm to determine the concentration of heavy metal ions using Atomic Absorption Spectrophotometer (A.A.S.). The amount of spent engine oil from the contaminated soil was determined via 10ml of spent engine oil poured on 3 different random isolated spots on soil and labelled SA, SB and SC. After every 24 hours, the soil from SA, SB and SC were collected at a depth of 10cm. Samples of the soil was

then digested, filtered and the concentration of metal ion was determined using AAS and the concentration was calculated. Samples were prepared in 3 replicates.

The percentage of adsorbed copper ions was calculated using:

$$\text{Percentage adsorption (E\%)} = \frac{C_i - C_f}{C_i} \times 100 \quad (1)$$

where C_i is the initial concentration of the copper ions before contact in the spent engine oil, C_f is the final concentration of copper ions after contact. Triple measurements were made to analyze errors.

3. Results and Discussion

Figure 2 shows the FT-IR spectra of *Luffa cylindrica* fibers. The adsorption peaks that occur in the IR spectra of *Luffa cylindrica* demonstrate the intricacy of the plant. Figure 3 shows a variety of distinct bands that are related to functional categories discovered in cellulose and lignin, the two primary chemical components of *Luffa cylindrica*.

The FT-IR spectra of *Luffa cylindrica* before and after copper adsorption clearly exhibit a number of absorption peaks, demonstrating the complexity of *Luffa cylindrica*, as seen in Figures 2 and 3. Figure 3 shows that *Luffa cylindrica* has carbonyl and hydroxyl groups on its surface (Tanobe, 2005). Weak absorption peaks about 3294 cm^{-1} demonstrated the presence of bonded hydroxyl groups on the surface of *Luffa cylindrica* due to SiO_2 . The peaks at 2922 cm^{-1} and 2855 cm^{-1} are a result of the C-H bond in the methyl group stretching vibration, which has been identified as the reason for adsorption of the copper ions. The absorption band of 1729 cm^{-1} was ascribed to carbonyl and carbonyl moieties (C=O). It was confirmed that aromatic rings have stretching vibrational maxima at 1595 cm^{-1} and 1461 cm^{-1} . The lignin structure of loofa is further reinforced by the C-O bond deformation at 1028 cm^{-1} , which is seen in the strong absorption band at wave number 1371 cm^{-1} , the aliphatic bending group (CH), and other factors. Hence, the subsequent adsorption of copper on the surface of *Luffa cylindrica* may be the result of chemical interaction between the molecules of copper and surface functional groups. Via van der Waals interactions, H_2 bonding and electrostatic interactions between the copper molecules and electron-rich regions on the surface of *Luffa cylindrica*, the molecules of *Luffa cylindrica* and copper interact with one another. This shows the attraction of copper ions to the surface of the *Luffa cylindrica*. Hence, the adsorption of copper onto the surface of *Luffa cylindrica* may be caused by the interaction between the carboxyl group of *Luffa cylindrica* and the electronegative groups in copper. According to research, the adsorption of copper onto *Luffa cylindrica* involves a complicated sequence of physical and chemical reactions (Olaseni 2014).

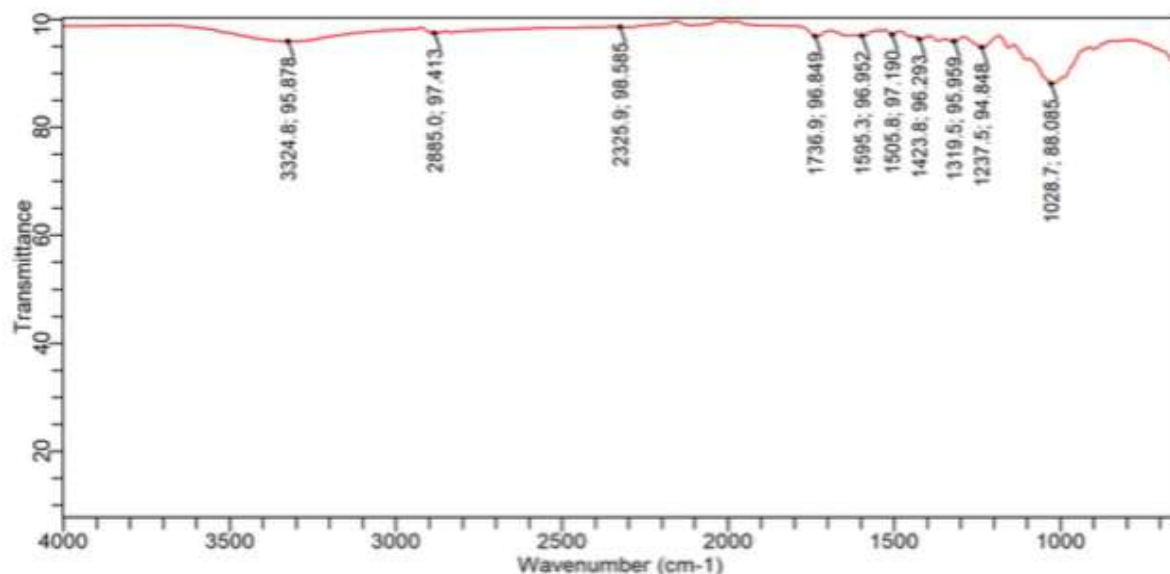


Figure 2: FT-IR of loofa before adsorption of spent engine oil

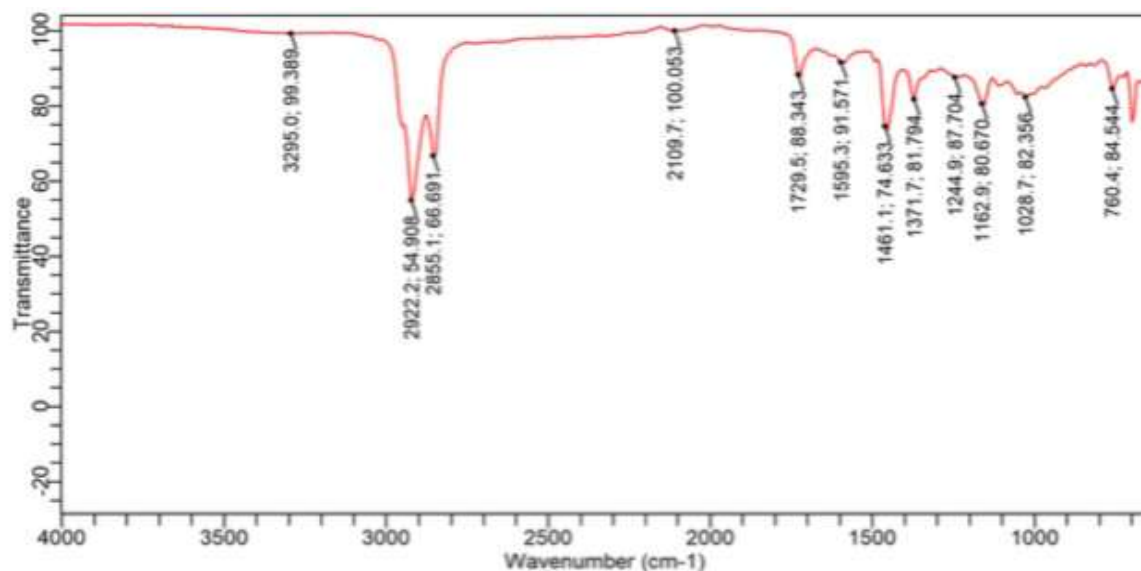
Figure 3.: FT-IR of *Luffa cylindrica* after adsorption of spent engine oil

Figure 4 shows the percentage adsorption of copper ions with a change in the dosage of the adsorbent. The percentage increases as the dosage of the adsorbent increases which indicates that active sites are available for adsorption of the spent engine oil (Nwadiogbu, 2016). Table 3.1 shows the copper ion concentrations and the optimum percentage adsorption capacity at an adsorbent dosage of 3g.

Table 3.1: Dosage of *Luffa cylindrica* in spent engine oil

Dosage (g)	Concentration (ppm)	Percentage %
0.5	0.068	35.849
1	0.063	40.566
2	0.059	44.339
3	0.056	47.169

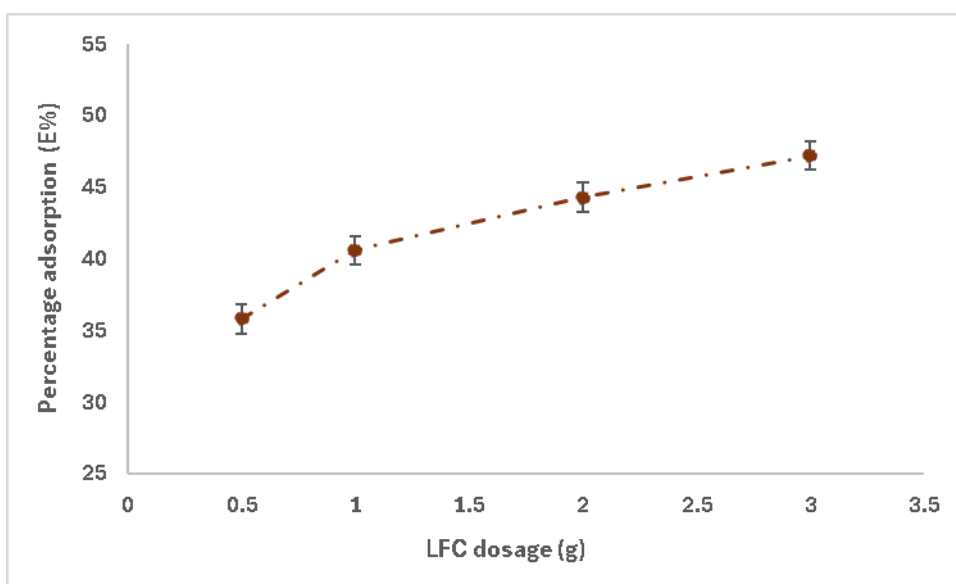


Figure 4 The percentage adsorption of spent engine oil on the different dosages of *Luffa cylindrica* (LFC).

Figure 5 shows that the spent engine oil contaminated soil absorbs more copper ions as the retention time increases. This is because the metal ions are likely to accumulate in the soil and the amount of metal ions increase due to soil penetration over time (Tan et al., 2020). This explains that the soil acts as an adsorbent with a holding and binding capacity for adsorption. Table 3.2 shows each sample taken from the soil after 24hrs, 48 hrs and 72hrs of engine spent oil deposited in the soil, that was digested and absorbed by *Luffa cylindrica*, and the concentration of copper was determined to be 0.047, 0.147, and 0.207ppm respectively. Figure 5 depicts the amount of copper ions in the soil increase because of the retention time of the spent engine oil deposited in the soil.

Table 3.2: Copper concentration in soil samples with variation of resident time

Times (hours)	Soil Sample	Concentration of copper (ppm)
24	Soil A	0.047
48	Soil B	0.147
72	Soil C	0.207

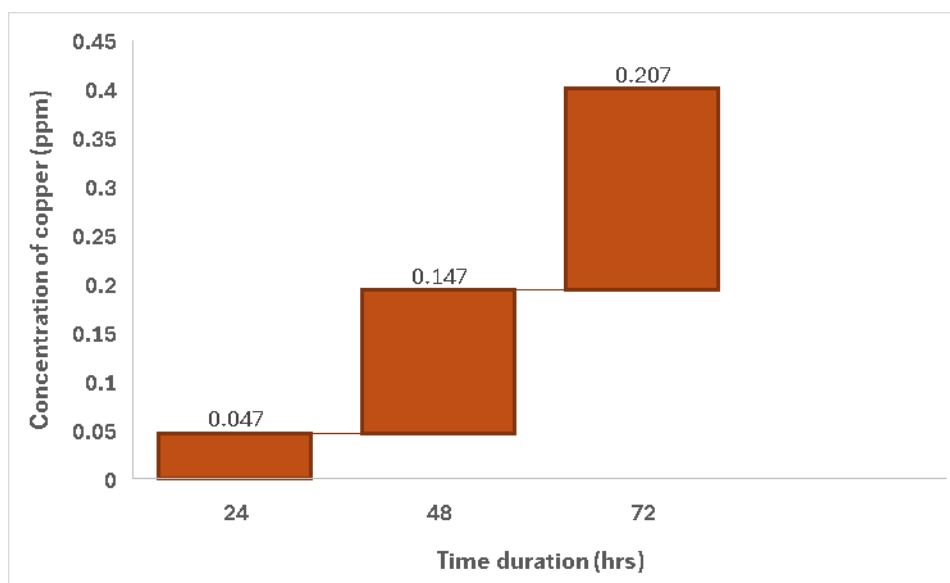


Figure 5: Concentration of copper in soil contaminated with spent engine oil (SEO) at various retention times (initial concentration – 0.08ppm).

At the adsorption zone of 10 cm and as the SEO is flowing through the soil depth, the copper ions are being adsorbed at the adsorption zone in the soil via the mass transfer mechanism and the soil is slowly saturated with copper ions through the adsorption sites of the soil at a transport time of between 24 – 72 hrs. The constant percentage of approximately 32% after every 24 hrs, slowly shows the saturation of the metal ions in the adsorption zone of the soil.

4. Conclusion

The presence of copper ions in the spent engine oil increased in the soil over time and the *Luffa cylindrica* was shown as an efficient adsorbent for removal of copper ions. The percentage adsorption of spent engine oil onto *Luffa cylindrica* (LFC) yields a performance capacity of approximately 47%. This research also demonstrates that as spent engine oil is deposited in the adsorption zone of the soil over time, the concentration of copper in the soil increases. Due to the adsorption capacity of the *Luffa cylindrica* applied on the soil, copper ions present can be effectively adsorbed. Also, the *Luffa cylindrica* material acts as a good adsorbent for removal of copper from aqueous solution and contaminated soil. This effective, environmentally friendly and economical method can be modified and improved by enhancing the properties of the adsorbents and materials that capture these pollutants.

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