Investigating the Use of Various Parts of Moringa oleifera for Removal of Mercury and Arsenic from Contaminated Water

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Abstract

Industrialization and civilization has increased the level of toxic metal ions found in the various spheres of the environment including the hydrosphere. Hence, emergent technologies that are economically feasible and highly efficient must be studied and explored, with the aim of promoting better environmental and human life quality. Plants are known to remove heavy metal ions from wastewater. In this study, the potential of various parts of *Moringa oleifera*; dry pod, seed, fresh pod, root, bark, and leaves to remove As (III) and Hg (II) ions were investigated and the chemical composition involved in this biofiltration were characterized using Fourier Transform Infrared (FT-IR) Spectroscopy. The various parts of *Moringa oleifera* used as biosorbent were pretreated before used as a biofilter. The removal of Hg (II) by the pretreated *Moringa oleifera* various parts biosorbents were higher compared to the removal of As (III) ions by the same parts. This research showed that *Moringa oleifera* various parts have the potential to be used in the removal of Hg (II) metal ions from wastewaters.

Keywords

Biosorbent; adsorption; concentrations; mercury ion; arsenic ion.

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1. Introduction

Heavy metals, which are high density metallic elements are nowadays considered to be the most important pollutants in surface and groundwater (Kinuthia et al., 2020). Although heavy metals may exist in their background concentrations in the environment, their presence particularly in the aquatic environment is attributed to human inputs such as emissions from municipal incinerators, discharges of wastes from industrial activities, as well as relatively minor sources of contamination such as small-scale industries that process metals (Asamoah et al., 2021). High levels of the heavy metals in the various spheres of the environment defy goal 3 and 6 of the sustainable development goals (SDGs). That is, they represent a serious threat to human health, living resources and ecological systems because of their increased discharge, toxic nature, and other adverse effects on receiving waters (DeGhetto et al., 2016). Unlike most organic pollutants which can be degraded, heavy metals are generally non-biodegradable and have high solubility in water. Hence the safe and effective disposal of wastewater containing heavy metals is always a challenge to industrialists and environmentalists, since cost-effective treatment alternative are not available (Berisha et al., 2020). Several chemical and physical technologies including: precipitations (Alluri et al., 2007), solvent extraction (Sekar et al., 2004), ion exchange (Kapoor et al., 1999), reverse osmosis (Araújo et al., 2010), electrochemical treatment (Da Rocha Ferreira et al., 2019), filtration (Kumari et al., 2006), flocculation (Kumari et al., 2005), and solid phase extraction (Alves et al., 2010), etc. have been described for removing toxic heavy metals effluent.

However, these technologies except ion exchange have considerable disadvantages, including incomplete metal removal, requirements for expensive equipment and mon-

itoring system, high reagent or energy requirements or generation of toxic sludge or other waste products that require disposal (Pehlivan and Cetin, 2009). This has prompted researchers to find alternative economical methods for wastewater treatment. Currently, the trend in metal adsorption research is focused on seeking new lowcost adsorbent materials with high adsorption capacity. For instance, studies elsewhere are focused on removal of cadmium ions from aqueous solution employing adsorbents obtained from inexpensive tree leaves such as Moringa oleifera leaves (Eman, et al., 2015). Biological materials have attracted great attention as adsorbents (biosorbents), mainly because of their natural availability in environment, high efficiency, minimization of chemical and/or biological sludge, regeneration of the biosorbent, and low costs (Araújo et al., 2010). Thus, natural sorbents constitute an excellent alternative for chemical remediation. Both methyl mercury and elemental mercury are poisonous even at low concentrations. Exposure to mercury causes severe health problems in humans such as ataxia, sleep disturbances, numbress, as well as tremor (Nakazawa et al., 2016; Gyamfi et al., 2021). Inorganic arsenic is soluble in water and can pose acute toxic effects in humans and other biota. Oral ingestion of high levels of arsenic can lead to gastrointestinal symptoms such as severe vomiting, blood circulation disorder, destruction of the nervous system, and eventually death (Asare et al., 2019).

Moringa oleifera (MO) is the most common species of the Moringaceae family. It is a plant native of northwest India but currently spreads all over the world, very typical in tropical locations. MO seeds biomass have not only been studied for their ability to remove turbidity and other impurities in water but also for their ability to remove heavy metals from aqueous solutions. Studies by Nand Maata et al. (2012) showed that M. oleifera seed was capable of adsorbing heavy metals more than other seed types. Percentage of removal was 90% for copper, 80% for lead, 60% for cadmium, and 50% for zinc and chromium. Many other several studies reported in literature have shown that heavy metals such as Ag (I) (Moringa Lam seed), Cu, Ni, Cr and Zn ions (Matouq et al., 2015), Pb (Mataka et al., 2006), and Pb (II) (Reddy et al., 2010), could be adsorbed by Moringa oleifera various parts, but no work has been published on comparing the various parts to know their adsorption efficiencies and also apart from the seed the part that adsorbs best. In this present work, the aim is to evaluate the adsorption capacities of six different parts of Moringa oleifera: dry pod, seed, fresh pod, root, bark and leaf in respect to two different heavy metals: mercury and arsenic, at two different concentrations; 5 ppm and 50 ppm. Characterization by Fourier Transform Infrared (FT-IR) of the various parts were also conducted.

2. Materials and Methods

2.1 Biomass Material

Moringa oleifera various parts were obtained from trees cultivated in University of Energy and Natural Resources, Ghana. The parts (leaves, Dry pod, Seed, Fresh pod, Root, Bark) were washed first with tap water, distilled water, pulverized and oven-dried at 60°C for 24h, pre-treated with a buffer and stored at 4°C prior to analysis.

2.2 Reagents and Solutions

All reagents used were of analytical grade standard. Mercury (II) sulphate (HgSO₄) and Arsenous oxide (As₂O₃) were purchased from BDH Laboratory, England, and used without further purification. Solutions of Mercury (Hg²⁺) and Arsenic (As³⁺) metal ions were prepared through appropriate dilutions from 1000 ppm stocks. The solutions were prepared with deionized water and to enhance complete dissolution, solution was supplemented with 5-10 mL nitric acid (HNO₃) in order to stabilize the metal ions in solution. All glass wares were soaked in 5% HNO₃ for 24 hours and rinsed with distilled water before and after use to remove trace metal ions.

2.3 Instruments and apparatus

A Nexion 2000 ICP-MS was used for the metal ions determination, DRAGONLAB SK-0330-PRO Mechanical Shaker for agitation and a pH/Conductivity meter 901 (BANTE INSTRUMENTS) was used for pH measurement.

2.4 Moringa various parts IR characterization

The functional groups present in the various parts of M. oleifera were characterized using a Fourier transform Infrared (FT-IR) spectrometer (Shimadzu, IR Prestige-21, Tokyo, Japan). The powdered samples were mixed with KBr in the ratio of 1:100 (w/w) to produce tablets. The spectral range varied from 3500 to 500cm-1, with twenty-eight scans and a resolution of 4cm-1. The purpose of using this procedure was to assess the main functional groups present in the material for adsorption.

2.5 Experimental procedures

Batch adsorption experiments were performed at room temperature $(25\pm1^{\circ}C)$ and conducted under shaking conditions (300rpm) in 50-mL volumetric flasks. The cation solutions (25mL) of known concentrations (5mg L⁻¹ and 50mg L⁻¹), followed by a known amount of biosorbent (0.5g). The flasks were then mixed thoroughly. After allowing sufficient time for adsorption equilibrium to be reached, the biosorbents were separated by filtration (filter paper). The metal ion concentration in the filtrate was then determined by ICP-MS. FTIR analysis were performed on biosorbents before and after metal ion sorption. The adsorption efficiency of the biosorbent with respect to the percentage removal of adsorbed metal ions was calculated using the equation below according to (Emahi, Sakyi et al. 2019)

$$\% of Removal = \left[(Co - Ce)/Co) \right] \times 100 \tag{1}$$

Where C_o is the initial metal ion concentration, and C_e is the concentration of metal ion remaining after biofiltration.

3. Results and Discussion

3.1 Characterization of M. oleifera various parts

The FT-IR technique was used to determine the main functional groups present in each of the various parts of *Moringa oleifera* responsible for the biofiltration of Hg^{2+} and As^{2+} in artificially contaminated water prepared in the laboratory. Fig 1-6 shows the FT-IR spectra of the various parts. According to (Pagnanelli, Mainelli et al. 2003) agricultural remains are generally made up of similar functional groups including phenol, aldehydes, carboxyl groups, ether, alcohols and ketones which bind to metal ion as pollutants.

The FTIR spectrum of the inactivated and activated leaf sample, showed no significant change in their wavenumbers but a change in the intensities of their peaks, with the activated leaf sample having intensified and more pronounced peaks. The activated leaf sample had a broad band around $3500 - 3000 \text{ cm}^{-1}$. This band is due to O–H stretching vibration of alcohol/carboxylic acid functional group. After adsorption of Hg (II) and As (III), these bands shifted to $3300-3350 \text{ cm}^{-1}$ and $3400-3200 \text{ cm}^{-1}$ respectively with a decrease in band broadness indicating the involvement of the O–H functional group in the adsorption of the two metals.

The peak at 1603.24 cm⁻¹ was due to the presence of C=C of aromatic ring/alkene or C=O from carbonyl vibrational stretching. After binding to the Hg (II) and As (III), the peaks at 1731.64 cm⁻¹ are due to C=O stretching vibration, 1624.50 cm⁻¹ and 1543.98 cm⁻¹ for C=C stretching vibrations of aromatic ring or alkene groups were observed indicating the involvement of C=O and C=C functional groups in the adsorption process.

The FT-IR spectrum of the Fresh pod sample inactivated and activated (Fig. 3) showed virtually no differences. Around $3500 - 3100 \text{ cm}^{-1}$, was a broad band due to O-H stretching vibration from the spectrum of the activated fresh pod sample (Fig. 4). There was however an increase in peak intensity, broadness and shift in wavenumber of this stretching vibration to 3400– 3200cm^{-1} and $3300 - 3100\text{cm}^{-1}$ respectively after adsorption of Hg (II) and As (III). This indicates that the O-H functional group was used in the adsorption. At 1595.61cm^{-1} of the activated fresh pod spectrum, was a peak indicating the presence of C=C of aromatic rings. After adsorption of As (III) were peaks at 1726.69cm^{-1}



Figure 1. Visitors to BFMS in 2018



Figure 2. Comparison of the FT-IR spectra of Leaf activated and inactivated Blank

and 1638.29 cm⁻¹ for C=O and C=C stretching vibrations respectively which didn't appear in the adsorption of Hg (II).

A C=C stretching peaks at 1507.49cm⁻¹ was observed for both the adsorption of Hg (II) and As (III). Functional groups O-H, C=C, -COOH and C=O was involved in the adsorption of As (III) but O-H and C=C functional groups were involve in the adsorption of Hg (II).



Figure 3. Comparison of the FT-IR spectra of Fresh pod activated and inactivated

A broad band around 3400cm⁻¹- 3100cm⁻¹ due to O-H stretching vibration from the FT-IR spectrum of the inactivated sample of the root didn't differ as compared to the activated sample. After adsorption of the Hg (II)



Figure 4. Comparison of the FT-IR spectra of Fresh pod activated Raw, As and Hg

and As (III), this band shifted to around $3200 - 3100 \text{cm}^{-1}$ and $3250 - 3050 \text{cm}^{-1}$ respectively. This indicates that O-H functional group was involved in the adsorption.

A peak at 1597.30 cm⁻¹ for C=C stretching vibration of aromatic ring appeared on the spectrum of the activated root sample. After adsorption of Hg (II), was a peak at 1604.12 cm⁻¹ for C=C of aromatic ring but for adsorbed As (III) was two peaks assigned to C=O and C=C stretching at 1735.66 and 1637.74 cm⁻¹ respectively.

[At 1020.75cm⁻¹, 1019.55cm⁻¹ and 1020.48cm⁻¹ were peaks from the spectrum of the activated root sample, Hg (II) and As (III) adsorbed root sample respectively for C-O stretching. The intensities of these bands as compared to the activated sample increased for both the Hg (II) and As (III) after adsorption indicating its involvement in the biofiltration process. The spectrum of both the raw and activated bark sample showed peaks around 3331 – 3100cm⁻¹ assigned to O-H stretching vibration, two set of peaks at 2916.02 and 2850.01cm⁻¹ for C-H stretching, a peak at 1599.44 cm⁻¹ for C=C of aromatic ring and a peak at 1026.94cm⁻¹ for C-O bond stretching.

After adsorption of Hg (II) and As (III), were broad with high intensity band around $3500 - 3000 \text{cm}^{-1}$ indicating the use of O-H functional group in the adsorption process.

[Two peaks which wasn't present in the activated sample were observed for both the Hg (II) and As (III) adsorbed bark samples, at 1734.91 and 1637.75cm⁻¹ and 1731.56 and 1633.59cm⁻¹ respectively. These peaks indicate the presence of C=O and C=C functional groups in the biofiltration process. At 1029.07cm⁻¹ from the Hg (II) adsorbed spectrum, was a decrease in its peak intensity as compared to the activated bark sample. For the As (III) adsorbed sample, was a prominent peak at 1016.19cm⁻¹ as a result of C-O stretching been involved in the biofiltration process.

The FT-IR spectrum of the inactivated and activated dry pod showed a difference between the region of 1650-1500 cm⁻¹ where C=C stretching of aromatic rings are observed. On the inactivated dry pod spectrum there was a peak at 1613.13cm⁻¹ which was completely absent

after activation of the dry pod sample.

Around 3500-3300 cm⁻¹ was a broad band for O-H stretching observed on the activated dry pod spectrum. Adsorption of Hg (II) and As (III) increased the band broadness and intensity 3600 - 3000 cm⁻¹ indicating the use of O-H functional group in the adsorption process.

A decrease in intensity of the claw-like double peak on the activated dry pod spectrum at 2914.91 and 2847. 99cm^{-1} was observed after adsorption of Hg (II) and As (III) at 2916.29 and 2849.99 cm⁻¹ and 2914.96 and 2848.89 cm-1 respectively indicating the presence of H-C=O of aldehyde functional group. The peak at 1030.22 cm⁻¹ on the activated dry pod spectrum, which is due to C-O stretching was found to decrease to 1028.43 cm⁻¹ and 1029.75 cm⁻¹ with an increase in peaks intensities after adsorption of Hg (II) and As (III) respectively. This confirms the use of C-O functional group in adsorbing the metal ions.



Figure 5. Comparison of the FT-IR spectra of Dry pod Activated, As and Hg



Figure 6. Comparison of the FT-IR spectra of Dry pod Activated and inactivated

3.2 Metal ions removal from contaminated wastewater using the various parts of *M. oleifera*

The objective of this study was to investigate the potency of the various parts of M. *oleifera* as a biosorbent for removal of Hg (II) and As (III) in contaminated wastewater. In order to obtain the natural optimum conditions for the removal of the metal ions studied, the pH of the raw samples was measured and pretreated with a buffer at the same pH after activation. This was done for complete interaction of the functional groups with the metal ions, which is pH dependent.

From figure 7&8, all the various parts adsorbed Hg (II) more than As (III) at both 5 ppm and 50 ppm. This can be explained by the differences in the ionic radius of the two chemical species, which for single metal ion solution, larger ionic sizes are mostly adsorbed (Matos and Arruda 2003). At 5 ppm, the percentage removal of Hg (II) and As (III) by Fresh pod was observed to be 96.04% and 0.00% respectively. From the FTIR spectrum of this metal ions, it can be observed that after adsorption of Hg (II) there was a C-H stretching peak at 2918.47 and 2851.39cm which aided in the adsorption of Hg (II) since a single peak at 2916.17cm⁻¹ was observed after adsorption for As (III), hence its poor adsorption capacity. For the seed, all of the Hg (II) was adsorbed.

Relatively, the root was a bad biosorbent for Hg (II) as compared to the other parts due to the difference in peak around $2920 - 2830 \text{cm}^{-1}$ which is brought about by stretching from C-H groups. Uniquely a single peak at 2918.07 cm⁻¹ was observed for the Root as compared to the two peaks of the other parts at 2918.47 and 2851.39cm^{-1} for Fresh pod, Dry pod at 2916.29 and 2849.99cm^{-1} , Seed at 2922.37cm^{-1} and 2853.05cm^{-1} , Bark at 2915.94 and 2851.14cm^{-1} and leaf at 2917.15 and 2849.11cm^{-1} respectively.

And finally, the leaf showed interesting adsorption capacity for As (III) as compared to the other parts with a removal of 24.14%, 16.59% for root, 7.33% for seed, 0.00% for fresh pod, 4.38% for dry pod and 3.69% for the bark. From the FT-IR spectrum of all the various parts after As (III) adsorption, the difference in the adsorption capacity can be noted from the C-H stretching around 2950-2800 cm⁻¹. For which there was a more intensified and a well-defined claw - like double peak which aided in the adsorption process. At 50 ppm from the bar graph, it can be observed that the leaf adorbs 100% of the Hg (II), indicating the larger surface available for adsorption at higher concentration as compared to the other parts which showed lower adsorption capacity which was almost to half as compared to their adsorption at 5ppm. This is generally due to the large atomic radius of the mercury ion and its presence in large amount enhancing interaction with the functional groups. It is also observed that, percentage of removal of As (III) was higher at 50 ppm than at 5 ppm confirming the smaller atomic radius of As (III) which decreases its interaction with the functional groups hence the lower adsorption capacity.



Figure 7. Comparing the adsorption efficiencies of each part at 5 ppm of As (III) and Hg (II)



Figure 8. Comparing the adsorption efficiencies of each part at 50 ppm of As (III) and Hg (II)

4. Conclusion

The study has successfully investigated the adsorption capacities of the various parts of *Moringa oleifera* at 5 ppm and 50 ppm for Hg (II) and As (III) ions. Generally, the six parts of the *Moringa oleifera* are not a good adsorbent for arsenic ion at both 5 ppm and 50 ppm, since all of the parts percent removal were less than 50%. In contrast, the various parts of the biosorbent showed an appreciable percent removal for mercury ion at both concentrations. The decreasing adsorbing efficiencies of the parts at 5 ppm are in the order; Seed > Fresh pod >Bark > Dry pod > Leaf > Root. At 50 ppm the efficiencies were also as follows Leaf > Fresh pod > Seed > Bark > Dry pod > Root. It is therefore concluded that *Moringa oleifera* is a good adsorbent for Hg (II) at both lower and higher concentrations.

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