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Research Paper

Effect of multi-walled carbon nanotubes in removing Pb and Cd in contaminated bio-medical ash

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ABSTRACT

Heavy metal pollution is one of the most serious environmental problems. As a result, the present research is an overview of the use of nanotechnologies that are being developed to remove or treat such pollutants from the environment. There is no text provided. Multi-walled carbon nanotubes have effectively eliminated cadmium (II) and lead (II) from biomedical ash. The findings demonstrated that the rate of adsorption escalated as the temperature of the solution increased, owing to the fact that the adsorption process is endothermic in nature. An analysis was conducted on the adsorption of cadmium (II) and lead (II) on multi-walled carbon nanotubes. The kinetics of this process were examined using three models: quasi-first-order, quasi-second-order Lagergren, and fractional energy function. The findings indicated that the adsorption of heavy metal ions followed a quasi-second-order mechanism, and the adsorption capacity exhibited a positive correlation with the solution temperature. The binding of metal ions by carbon nanotubes was evaluated through the adsorption capacity and was found to be as follows: 7.93%, 8.33%, 73.50% and 92.46% respectively of Pb adsorption capacities and for Cd the results were 67.92%,29.30%,6.30%and 2.27% after adding three different concentration of MWCNTs (0.0%) as check control (CK) and the other concentrations are (0.01%,0.05% and 0.10%) respectively and it found that the order: Lead(II) <Cadmium(II). The use of nanotechnologies has paved the way for more researchers to cope with problems. The evolution of Nanotechnologies for heavy metal remediation, such as cadmium and Lead is discussed in this research because it is one of the most hazardous heavy metals that needs immediate attention.

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1. INTRODUCTION

Rapid urbanization and industrialization have affected the environment. As a result, pollution and degradation of the entire ecosystem have become a serious problem and a threat to all aspects of life, including humans [1]. Research in this area has also gained great speed. Heavy metals can be produced from several sources, including by-products from the medical sector, especially biomedical ash from incinerators. As a result, researchers have developed great interest in extracting heavy metals from wastewater, soil, and ash. Cadmium is used in many medical industries. Figure 1 shows the uses of cadmium. Several removal techniques have been proposed and practiced by many researchers, including activated sludge, bioremediation, and reverse osmosis treatments. However, these techniques also have several drawbacks, such as reduced biodiversity and inhibitory effects on plant growth [2]. The use of microorganism techniques seems more promising because it changes or converts heavy metal toxins into a less harmful state. Bioremediation has some drawbacks. The most important technology was nanoparticles, such as carbon nanotubes. However, this technology is a better option in terms of process efficiency, environmental friendliness and economical way. The use of CNTs in medical applications is still a subject of research, and their safety and efficacy need to be thoroughly evaluated, but adding CNTs to medical ash, may have very potential applications by using it with medical ash as a reinforcement material to enhance the mechanical properties of the ash and improve its strength and durability [3]. The source of cadmium and lead in the ash resulting from the burning of medical waste is the medical materials themselves that are burned, as cadmium and lead are used in many medical products, such as medicines, medical devices, surgical equipment, as well as intravenous solutions [4]. When these products are burned, the cadmium and lead turn into ash due to degrees of High temperatures, which poses a serious threat to the environment and public health. Because it may reach the environment in several ways, such as leakage into the soil and groundwater, which is the most common risk. Fly ash from burning medical waste can also cause contamination of the soil and groundwater. Inhalation of these toxins by humans and animals can cause serious health problems, such as respiratory diseases [5].The background value of (bp) and (Cd) in ash was respectively 1.13 mg/kg and 0.21 mg/kg (Krenev et al., 2015).

2. LITERATURE REVIEW

Hospitals and other health care facilities are absolutely essential for treating and caring for people with health problems as well as promoting health in the community. As health care services improve and increase in scope even in developing countries, the problem of healthcare waste also develops, as hospitals generate a relatively huge amount of healthcare waste, consisting of general and hazardous waste. People handling healthcare waste are at immediate risk, followed by people residing near waste treatment or dumping areas and the general public. Infectious healthcare waste poses a major threat to the health of humans and animals because it has the potential to spread many infectious diseases between humans and animals. Due to the uncontrolled use of non-biodegradable disposable materials by healthcare systems and their treatment or lack thereof, healthcare waste has become one of the major sources of environmental pollution, including the emission of a large amount of greenhouse gases, ranging from 3 to 10 % of the country's total emissions. Household waste also causes the leakage of chemicals and heavy metals such as lead, cadmium, chromium and radioactive materials, and even the generation of carcinogenic substances such as dioxins into the environment, leading to air, soil and water pollution in general, especially in the areas surrounding the dumping or treatment of household waste, affecting the health and quality of household waste and the life of not only humans but also plants and animals living in those areas. Therefore,

household waste has become one of the main sources of environmental pollution and collectively contributes to the problem of global warming. Household waste should be given the required attention and priority in procedures and policies. The chapter focuses on the different sources, types, environmental and health risks related to household waste, its overall environmental impact and management strategies to reduce the impacts with an environmentally friendly and sustainable approach [6].

The COVID-19 pandemic has caused a significant increase in the production of biomedical waste and plastic waste. The rapid surge in the development of garbage vehicles used for waste disposal has presented substantial obstacles to current waste disposal systems, particularly in developing nations. The significance of appropriate waste management has become increasingly apparent as a result of the health crisis caused by the coronavirus. This review attempts to comprehensively examine all facets of biological waste, encompassing its administration, secure disposal techniques, hazards linked to inadequate waste management, and additional perils to hospitals, laboratories, and the environment. The focus is on the potential contribution of laboratories in hospitals, research facilities, and academic institutions that are directly or indirectly involved in the management of biomedical materials. It is important to highlight that laws concerning the management of biological waste should be regularly revised in order to include new research and development findings into the system. Establishing collaboration among hospitals, laboratories, and research people is crucial for effective waste management in healthcare facilities, as highlighted in the present review. The review focuses on current developments in the treatment of biological waste and the methods used for its safe disposal, with particular emphasis on incineration, sterilization, chemical disinfection, and land disposal. Good laboratory practices and techniques for needle disposal, cutting, packaging, and freezing are also covered. Emphasis is placed on the importance of biomedical waste management policies to promote safe and environmentally responsible practices and on amendments to these policies [7].

3. MATERIALS AND METHODS

3.1 Reagents and MWCNTS

Every single one the reagents, cadmium chloride [CdCl2·2.5H2O] and potassium antimony tartrate [C4H4KO7Sb·0.5H2O], were of analytical quality and were obtained from Bioreasearch Chemical Reagent Company., Ltd. The pH of the solution was calibrated using a pH meter (SOONDA S-1). The experiment utilized ultrapure water with a resistivity of 20 MΩ cm. Prior to preserving the extracts, all vessels were immersed in a solution of dilute HNO3 (4%, v/v). The multiwalled carbon nanotubes utilized in this investigation were not altered, as indicated by the existing literature on ash toxicity and metal extraction [8]. The samples were acquired from Sigma Aldrich Technology Co., Ltd and were produced using the mechanochemical process. Their outer diameter ranges from 10 to 30 nm, their inner diameter ranges from 5 to 15 nm, their length is between 10 and 40 μm, they have a purity level over 98%, and their specific surface area is greater than 360 m2/g.

3.2 Ash Sampling

The test Ash was a solution (at 0–10 cm deep) obtained from a bottom incinerator chamber site of Al Murjan Specialist Hospital in baghdad, wre had those characteristics:) organic materials 47.5%, inorganic materials 38.5%, hazardous materials 14%), and the physical and chemical characteristics: (pH 9.75, Total organic carbon 31.92 g/kg, total Ca 15.54 g/kg, total Fe 48.3 g/kg, total Zn 11.92 mg/kg, total Pb 1.13 mg/kg, total Sb 0.82 mg/kg, total Cd 0.21 mg/kg). The ash samples underwent a process of air drying, screening, hand grinding using a mortar and pestle, and further grinding to a powder using a continuous feed mill (Al-Watheq Soil Testing, Soil Laboratories Co. Ltd., Iraq). The heavy metal element pretreatment involved a mixed acid digestion process employing a toxic leaching approach known as TCLP. This procedure utilized two different extraction solutions: solution A, consisting of HOAc with a pH of 4.90 ± 0.05, and solution B, consisting of HOAc with a pH of 2.88 ± 0.05 [9]. The ratio of liquid to solid was 20:1, and the mixture was stirred for 16 hours in a rotating drum at a speed of 30 revolutions per minute. Following extraction, the samples underwent filtration using GF/C glass fiber filter paper with a pore size

of 0.45 µm. The samples were treated with 1 M HNO3 to acidify them and then analyzed for metal content using ICP-OES.

3.3 Incubation Experiment

The content levels (nominal lead and cadmium content in dentate ash, mg/kg) and (proportion of multiwalled carbon nanotubes added to dentate ash, %) were logarithmically spaced. The content ratio between lead and cadmium was 2:1. (CdCl2·2H2O and C4H4KOpb·2H2O) were added to the powdered ash to achieve lead and cadmium concentrations of 500 mg/kg, 250 mg/kg (pb500 + Cd250), 250 mg/kg and 125 mg/kg (pb250 + Cd125), 125 mg/kg and 62.5 mg/kg (pb125 + Cd62.5). Following a manual shock, the enriched ash was agitated with a carbon nanotube mixer for 24 hours to obtain a uniform mixture. The moisture content of the ash, relative to its dry weight, was around 25% after the addition of ultrapure water. The improved ashes were subjected to a 35-day equilibration process in an automated incubator, where they were exposed to laboratory lighting and maintained at a relative humidity of 75%. Once the aging process concluded, the incubated ashes were dehydrated using air and subsequently pulverized into a fine powder. Subsequently, the powder underwent processing to extract and quantify the levels of lead and cadmium in it. The processing concentrations of multiwalled carbon nanotubes in this study are derived from the results documented in a credible research authored by Radwan et al. in 2016. A 500-mL beaker was utilized to mix 250 g of ash with multiwalled carbon nanotubes. The composition was modified to get several test levels: control (CK) devoid of any nanotubes, 0.1%, 0.5%, and 0.10%. The units were meticulously blended until the cured ash achieved a consistent texture. With an increase in the curing level, the texture of the cured ash progressively grew more polished. There were three replications for each treatment. The moisture level of the treated ash was approximately 25% of its weight when it was dry. This was done by adding ultrapure water. Afterward, the glass beakers were hermetically sealed using a thin polyethylene film and then placed in either a heated building or an automated device designed for promoting growth for a period of 35 days. Following the incubation period, samples of the ash that had undergone treatment in each beaker were gathered. Subsequently, the samples were subjected to a drying process at a temperature of 102 °C for a period of 8 hours. After the drying process, the samples were pulverized into a fine powder to facilitate the extraction procedures [10].

3.4 Characterization the Morphologies of MWCNTS

The conductive gel utilized in this investigation, together with the ash coated with multi-walled carbon nanotubes (MWCNT), were analyzed using field emission scanning electron microscopy (FE-SEM, S-6840, Sony, Korea).

3.5 Extraction Procedure

3.5.1 Sequential of Three-Steps of Extraction Procedure

This method is employed to characterize a metallic element with the ability to form compounds with various concentrations of ash [9]. The species identification test in this investigation was conducted using a three-step sequential extraction process [11], following the Chinese standard GB/T 25.282–2010 [12]. The species can be categorized as follows: F1, the fraction that is soluble in acid and light; F2, the fraction that can be reduced; F3, the fraction that can be oxidized; and R, the residual fraction. In summary, a 1.00 g sample of ash was transferred into a 100 mL polyethylene tube, then filtered using a 0.45 µm membrane filter. The filtered solution was collected in a glass tube and kept at a temperature of 4 °C. The levels of lead and cadmium in all extraction methods were measured by LCP-AES. To compare the results, the residual content was determined by subtracting the sum of the F1, F2, and F3 contents from the measured total content.

3.5.2 Toxicity Characteristic Leaching Procedure

The Toxicity Characteristic Leaching Procedure (TCLP) was employed to ascertain the degree of heavy metal solubility in solid waste under acid rain conditions, as per the specifications given in China in

2011. As per the specifications of HJ/T 299–2007 [13], a mixture of sulfuric acid and nitric acid, with concentrations of 97% and 70% by mass respectively, was combined with water in a mass ratio of 2:1. This resulted in the production of extracts with a pH of 4.2 and a mass of 0.6 grams. The ash was transferred into a 100 mL conical flask with a closed top. Then, 10 mL of the extract was added and the mixture was vigorously shaken for 16 hours at a speed of 150 revolutions per minute at a temperature of 25 °C. The further steps of filtration, incubation, and calculations were carried out in the same manner as previously described.

3.5.3 Physiologically Based Extraction Test (PBET)

The PBET was created to assess the bioaccessibility, which it used to measure the dissolution of heavy metals in digestive fluid [14].

3.6 PH and Organic-Matter Analysis

According to the results obtained from the pH and organic matter quantity measurements shown in Table 1, the pH of MWBA ash was in the range (8.8 to 10.6) and the organic matter content was very high (22.57 to 47.14%), indicating that this type of ash contains a high amount of unburned organic matter, which should be attributed to the low operating temperature of the incinerator during combustion.

Table 1. Measured PH and OMcontent (n = 3, Reported as Mean).

No. of Reported	pH Value	Organic-Matter Content%
Repeat1	8.80	22.15%
Repeat 2	10.60	47.67%
Repeat 3	9.80	31.45%
Average	9.75	23.43%

4. RESULTS AND DISCUSSIONS

4.1 Measured total Cd and Pb content

For the computation, we chose to use the measured Pb and Cd content as the denominator. This decision was based on the assumption that the distribution of Pb and Cd in the ash was uniform, making it easier to compare horizontally. Overall, the measured Pb concentration was approximately 92% of the expected value, whereas the measured Cd content was around 96% of the expected content. The variation in the percentage of recovery may be attributed to system mistakes during the mixing process. According to Saleem et al. [14], the inclusion of bismuth citrate (C6H5BiO7) in a study resulted in a recovery percentage of components ranging from 75% to 85%. Consequently, it was feasible to determine the proportion of heavy metals in the extract (Table 2).

4.2 Morphology

The shape of multi-walled carbon nanotubes and the ash treated with multi-walled carbon nanotubes was shown through the use of scanning electron microscope and FTIR pictures. Figure 1(a) demonstrates that the observation verified the presence of thin, curved, and elongated nanostructures in the multi-walled carbon nanotubes. Furthermore, their characteristics were in line with the description provided by the supply company. Furthermore, let's consider the pb500+Cd250 ash as an example.

Figure 2(b) (c) (d) illustrates the arrangement of multi-walled carbon nanotubes on the surface of the ash particles. The increase in the quantity of multi-walled carbon nanotubes on the particle surface was clearly evident with each addition. Specifically, the addition of 0.10% indicated a dense packing of multi-walled carbon nanotubes on the surface, whereas the addition of 0.1% resulted in an even distribution of the nanotubes. Thus, the study's findings suggest that increasing the amount of carbon nanotubes in the

ash would result in the formation of clusters and a reduction in the efficient interaction between the particle surface and the multi-walled carbon nanotubes that were in touch.

4.3 Lead and Cadmium Speciation

The distribution of lead (Pb) exhibited notable dissimilarities compared to that of cadmium (Cd). According to Figure 1, the proportions of F1–Pb, F2–Pb, F3–Pb, and R–Pb were 6.65%–9.21% and 7.43%, respectively. The percentages of the total measured content are as follows: 9.23%, 0.464% to 0.80%, and 81.11% to 93.81%, respectively. The percentages of F1–Cd, F2–Cd, F3–Cd, and R–Cd were as follows: 63.51%–72.31%, 28.09%–31.03%, 3.78%–8.81%, and 0.52%–4.00%, respectively. The residual component mostly refers to the heavy metal that is chemically bound to aluminosilicate, which poses difficulties for the body in terms of absorption and mobility. Clearly, improving the remaining element proved to be a very dependable method for evaluating the effectiveness of paralysis [15].

Table 2. Measured Total Pb and Cd Content (n = 3, Reported as Mean).

Pb + Cd Content Concentration mg/kg + mg/kg	Nominal Total Pb mg/kg	Measured Total Pb mg/kg	Nominal Cd mg/kg	Measured Cd mg/kg
500 + 250	500	490.81	250	223.31
250 + 125	250	245.25	125	117.76
125 + 62.5	125	122.83	62.5	60.05

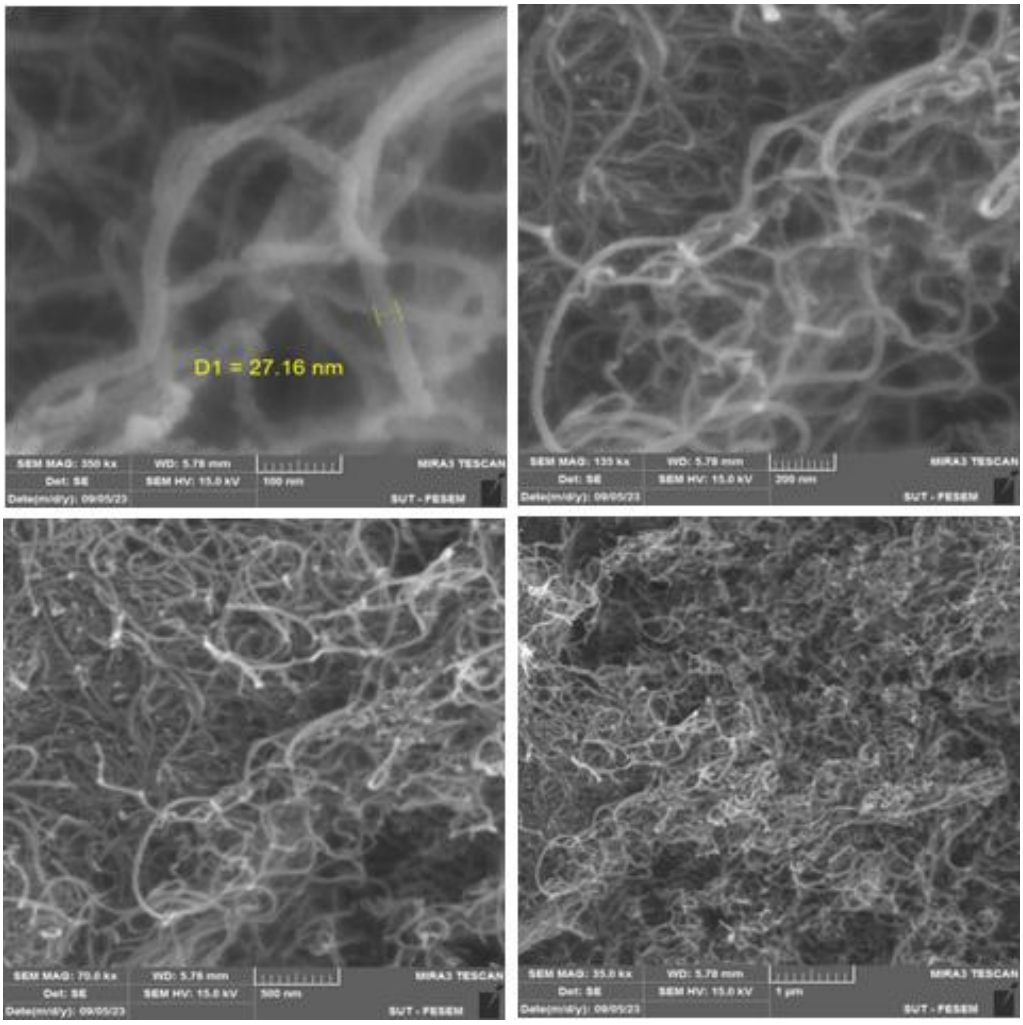


Figure 1. Morphology of Multi Wall Carbon Nanotubes (MWCNTs) Tested by Scanning Electron Microscopy a: under 100nm Magnification b: under200 nm Magnification. C: under 500nm Magnification. D: under 1µm Magnification

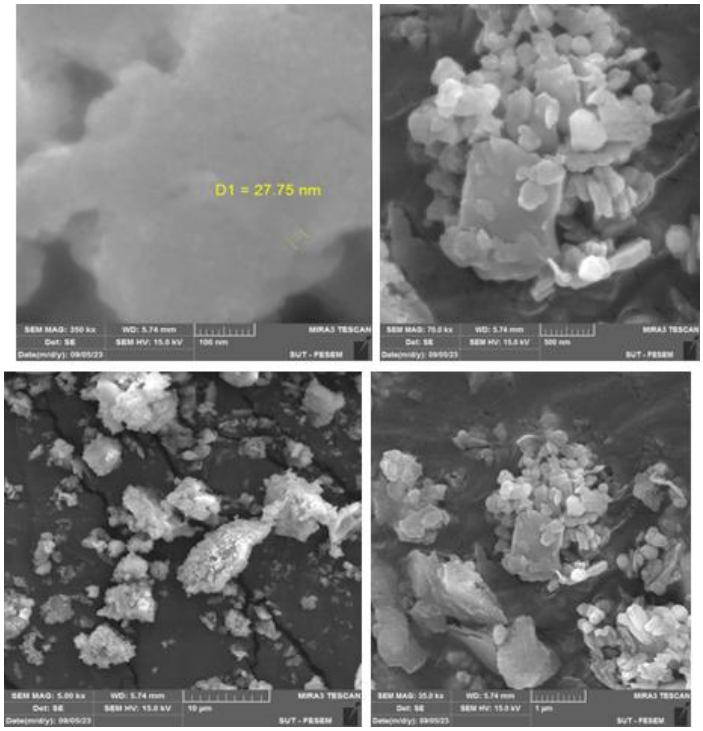


Figure 2. Morphology of MWCNTs Used in This Study and the Ash Treated with MWCNTs. a: MWCNTs. b: Ash pb500 + Cd250 Treated with 0.10% MWCNTs. c: Ash pb500 + Cd250 Treated with 0.5% MWCNTs. d: Ash pb500 + Cd250 Treated with 0.1% MWCNTs

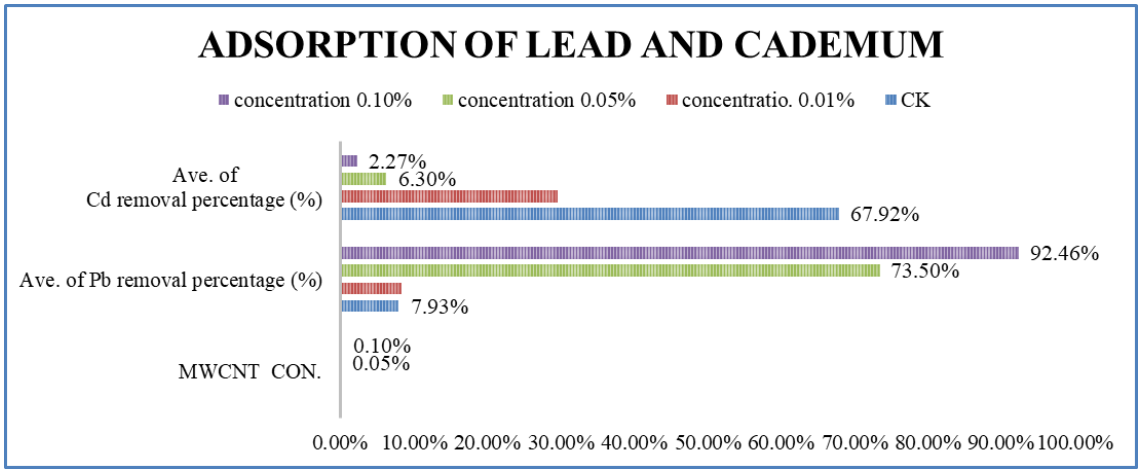


Figure 3. Lead and Cadmium Species in MWC-Treated Ash and Control Ash (CK, no MWC Treatment). the Sigmoidal Deviation Was Expressed as Nominal Heavy Metal Content + MWC Processing Level. Data Are Reported as the Average *of Three Replicates (M±AD, n=3)*

The primary control fractions were F1-Pb and F4-Pb. The inclusion of 0.1% MWC resulted in an 18.00% rise in F1-Pb content at pb500 ($p<0.05$). However, this increase was not observed at pb250 and pb125, nor with the addition of 0.5% and 0.10%. Moreover, with regards to all additives, MWC did not demonstrate any consistent impact on the four lead samples. Therefore, we may deduce that the increase in F1-Pb only occurred under two conditions: when the concentration of Pb was high and when the amount of MWCNT added was limited. When Cd125 was introduced, the addition of 0.5% and 0.10% resulted in a 2.6-fold and 2.5-fold rise in R-Cd, respectively. Nevertheless, the disparity in substance amounted to just 2.04 mg/kg and 2.45 mg/kg. However, the application of Cd250 led to a 6.52% increase in F1-Cd when 0.1% was used, indicating that the addition of smaller MWCNTs improved F1-Cd in high Cd ash. From the information provided, it can be inferred that the light acid-soluble fractions of high Pb and Cd ash were only

increased when a smaller amount of MWCNTs was added. This discovery implies that a reduced quantity of multiwalled carbon nanotubes could potentially elevate the environmental hazards associated with lead and cadmium ash.

5. CONCLUSION

The aim of this study was to assess the efficacy of Multi-Walled Carbon Nanotubes (MWCNTs) in extracting lead and cadmium from ash contaminated with biomedical materials. The study involved three different levels of lead and cadmium content (pb500 + Cd250, pb250 + Cd125, pb125 + Cd62.5) and four different levels of MWCNT treatment (0%, 0.1%, 0.5%, and 0.10%). The extraction process consisted of a three-step extraction procedure and three simple extraction procedures, as described by Zhong et al. [16]. The scanning electron microscopy (SEM) and field emission transmission electron microscopy (FTR) images revealed that increasing the amount of multi-walled carbon nanotubes (MWCNTs) led to a reduction in the effective contact area between the MWCNTs and the surface of the particles. This finding, reported by Chen et al. in 2006, is the initial detection of the morphological changes in ash treated with MWCNTs. Nevertheless, the inclusion of 0.5% and 0.10% resulted in a decrease in the bioavailability of bp by only 26.67% and 22.34% respectively, when the bp content level was 125 mg/kg. This suggests that there may be a greater number of Pb(OH) adsorption sites on MWCNTs due to the presence of stomach fluid. Furthermore, it was observed that only in the ash containing high levels of pb500 + Cd250, the addition of less than 0.1% resulted in a maximum increase of 16.30% and 11.40% in the content of acid-soluble Pb and Cd, respectively. This increase in content could potentially lead to heightened hazards and environmental consequences, as concluded by Britton et al. [17]. It was also determined that the most effective surface areas for interaction with the ash particles were provided. However, there was no discernible pattern of extractability observed for TCLP-extractable benzene and cadmium. The multi-mobile carbon nanotubes exhibited selectivity in extracting heavy metals from ash, as evidenced by the data comparison. In conclusion, this study determined that the presence of multi-mobile carbon nanotubes has an impact on the ability to extract lead and cadmium when they are in a solution form [18].

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Author Contribution Statement

Name of Author	C	M	So	Va	Fo	I	R	D	O	E	Vi	Su	P	Fu
Abbas A. Kanoosh	✓	✓		✓	✓				✓	✓				
Waleed M. Sh. Alabdraba	✓		✓			✓						✓	✓	
Ahmed Al-Obaidi				✓			✓	✓			✓			✓

C : **C**onceptualization
M : **M**ethodology
So : **So**ftware
Va : **Va**lidation
Fo : **Fo**rmal analysis
I : **I**nvestigation
R : **R**esources
D : **D**ata Curation
O : Writing - **O**riginal Draft
E : Writing - Review & **E**diting
Vi : **Vi**sualization
Su : **Su**pervision
P : **P**roject administration
Fu : **Fu**nding acquisition

Conflict of Interest Statement

No conflict of interest.

Informed Consent

Not Applicable.

Ethical Approval

Not Applicable.

Data Availability

The data that support the findings of this study are available from the corresponding author on reasonable request.

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