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Comparison between the Effect of multi-walled carbon nanotubes and single walled carbon nanotubes in removing pb and cd in contaminated bio medical ash

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Abstract: Heavy metal contamination is a highly significant environmental issue. Primarily, this current study is focusing the application of developing Nano technologies which have capacity to remove or reduce toxins from the environment. The extraction and preconcentration abilities of both types of carbon nanotubes, multi-walled (MWCNTs) and single-walled (SWCNTs), were accomplished by using water-acid, acetate, and deghassed water acid EDTA solutions from neuteuche semi-total solid extract from anthropogenic and naturally in origine samples. The results of the study indicated that the adsorption rate was even faster when the solution temperatures were higher in both types of adsorbents. These increases were greater when multi-walled carbon was used. This may be due to the fact that in endothermic nature of adsorption the increase in temperature leads to a faster rate of adsorption. In order to understand the kinetics of Cadmium (II) and Lead(II) adsorption, analysis was executed utilizing the fractional power function model and Lagergren pseudo-first-order and pseudo-second-order methods. A pseudo-second-order model well described the adsorption kinetics of heavy metal ions onto both bioadsorbents. For both biosorbents, the adsorption capacity increased as the solution was heated. The percentage of Pb adsorbed on ultra-sonically assisted MWCNTs was 7.93%, for MWCNTs= 8.33%, 73.50% for swCNTs and 92.46% for amine-MWCNTs (Table 2). Similarly, the percentage of Cd adsorbed on ultra-sonically assisted MWCNTs was 67.92%, 29.30% for MWCNTs, 6.30% for swCNTs and 2.27% for amine-MWCNTs. As far as SWCNTs, they provided some interesting results in terms of adsorption capacity. For example, the adsorption capacity with respect to Pb was about 6.53% with SWCNTs 1, about 7.12% with SWCNTs 2, about 61.33% with SWCNTs 3, and about 79.31% with SWCNTs 4. Similarly, for Cd it was about 61.32% with SWCNTs 1, about 27.13% with SWCNTs 2, about 5.88% with SWCNTs 3, and about 1.53% with SWCNTs 4. The elution tests for single-walled carbon nanotubes and multiwalled carbon nanotubes have been carried out and the results have been shown as binding isotherms and pH effects in Figures 3 and 4. For the study of the adsorption capability of single-walled carbon nanotubes for metal ions, same order of binding to the metal ions has been observed for both the types of the carbon nanotubes-studied, lead (II) < cadmium (II). For multi-walled carbon nanotubes, however, the binding of metal ions was found to depend on the carbon nanotubes type and the binding order, as for carbon nanotubes, were found to be, lead (II) > cadmium (II). Nanotechnologies have facilitated the ability of researchers to address challenges more effectively. This research focuses on the development of Nanotechnologies for the purpose of remediating heavy metals, including cadmium and lead. These metals are considered highly toxic and require urgent attention.

Keywords: *Bio medical ash, Cadmium remediation, Exposure, Lead remediation, Multi-walled carbon nanotubes, Nanotechnologies, Single walled carbon nanotubes.*

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1. Introduction

Urbanization and industrialization have had a significant impact on the environment. Consequently, the pollution and deterioration of the entire ecosystem have emerged as a significant issue and a peril to all parts of life, including humans [1]. The research in this subject has also accelerated. Heavy metals can originate from various sources, including by-products from the medical sector, particularly Bio Medical Ash produced by incinerators. Consequently, researchers have shown a strong interest in extracting heavy metals from effluents, soil, and ash. Cadmium is utilized in various medical sectors. Fig. 1 illustrates the various applications of cadmium. Several removal approaches, including as active sludge, bioremediation, and reverse osmosis treatments, have been developed and used by numerous researchers. However, these strategies also come with numerous drawbacks, such as a decrease in biodiversity and inhibitory impacts on plant growth [1], [2]. Utilizing microorganism's capabilities appears more advantageous as it alters or transforms heavy metal poisons into a less detrimental form. One of the main limitations of bioremediation is the use of Nanoparticles, such as Carbon Nano tubes, as a method. Nevertheless, this technology surpasses other options in terms of its efficiency, environmental sustainability, and economic viability. The utilization of carbon nanotubes (CNTs) in medical applications is currently under investigation, and their safety and effectiveness must be extensively assessed. However, incorporating CNTs into medical ash shows promising potential as a reinforcement material, capable of enhancing the mechanical properties, strength, and durability of the ash [2].
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 [3][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 [4][5]. The background value of (bp) and (Cd) in ash was respectively 1.13 mg/kg and 0.21 mg/kg

2. Materials and Methods

2.1. Reagents, MWCNTs and SWCNT

All reagents, including cadmium chloride [$CdCl_2 \cdot 2.5H_2O$] and antimony potassium tartrate [$C_4H_4KO_7Sb \cdot 0.5H_2O$], were of analytical quality and were purchased from bioreasearch Chemical Reagent Company., Ltd. The pH of the solution was calibrated using a pH meter (SOONDA S-1). The water utilized in the experiment was ultrapure, with a resistivity of 20 megaohm centimeters. Prior to holding extractants, all containers were immersed in a solution of dilute HNO_3 (4% v/v). The MWCNTs and SWCNTs employed in this investigation were unaltered, as indicated by the available literature on ash ecotoxicology and metal extraction [6]. The nanoparticles were acquired from Sigma Aldrich Technology Co., Ltd. They were manufactured using the mechano-chemical process and have the following characteristics: outer diameter of 10–30 nm, inner diameter of 5–15 nm, length of 10–40 μm, purity greater than 98%, and specific surface area greater than 360 m^2/g.

2.2. Ash Sampling

The test Ash was obtained from a bottom incinerator chamber site at Al Murjan Specialist Hospital in Baghdad. It consisted of 47.5% organic materials, 38.5% inorganic materials, and 14% hazardous materials. The ash had the following physical and chemical characteristics: pH of 9.75, total organic carbon of 31.92 g/kg, total calcium of 15.54 g/kg, total iron of 48.3 g/kg, total zinc of 11.92 mg/kg, total lead of 1.13 mg/kg, total antimony of 0.82 mg/kg, and total cadmium of 0.21 mg/kg. The Ash samples were dried by exposure to air, sorted, manually crushed using a mortar, and further pulverized into a fine powder using a continuous feeding pulverizer (Alwathiq for soil test, soil laboratory Co., Ltd, Iraq). The pretreatment method for heavy metal elements used mixed-acid digestion using a toxicity

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characteristic leaching process (TCLP). This procedure utilized two types of extraction solutions: solution A, consisting of acetic acid with a pH of 4.90 ± 0.05, and solution B, consisting of acetic acid with a pH of 2.88 ± 0.05 [7]. The ratio of liquid to solid was 20:1, and the process of stirring lasted for 16 hours in a rotary tumbler rotating at a speed of 30 revolutions per minute. Following the extraction process, the samples were further passed through a GF/C glass fiber filter paper with a pore size of 0.45 μm. The samples were acidified using 1 M HNO3. Then, ICP-OES was used to determine the metal content in the samples.

2.3. Incubation Experiment

The levels of content (pb and Cd nominal content in spiked Ash, mg/kg) and the fraction of MWCNTs injected into spiking Ash were distributed logarithmically in a 2:1 ratio. Ash powder was contaminated with CdCl2·2H2O and C4H4KOpb·2H2O to get concentrations of 500 mg/kg and 250 mg/kg for lead (pb500 + Cd250), 250 mg/kg and 125 mg/kg for lead (pb250 + Cd125), and 125 mg/kg and 62.5 mg/kg for lead (pb125 + Cd62.5). The ash that has electric with the help of manual are mix with the CNTs in the mixer for 24 hours in condition to get an even mixture. The moisture of the ash has 25% of its total dry weight. For that purpose, the ultrapure water is added in the ash. All ash samples were incubated for a period of 35 days in an automated incubator under controlled laboratory lighting and a relative humidity of 75%. After the aging procedure, the Ash samples were air-dried and subsequently pulverized into powder. The powder was then subjected to digestion in order to get the determined concentration of lead (Pb) and cadmium (Cd). The treatment concentrations of MWCNTs and SWCNTs in this investigation were based on the information provided in previously published works [6][7]. In a 500-mL glass beaker, 250 g of spiked Ash was supplemented with MWCNTs and SWCNTs to achieve the desired concentrations of control check (CK, no CNTs added), 0.1%, 0.5%, and 0.10%. The mixture was thoroughly blended until the treated Ash had a uniform texture. The Ash started to be softened of the treated Ash with increased concentration of the treatment. Three replicates of each treatment were applied to each CNTs type. Then, the treated Ash was moistened with ultrapure water at the moisture content covered approximately 25% of its dry weight, as paper towels soaked off the water in the cup. Then, the plastic bag was placed to a glass beaker, and put × g of the treated Ash separately into the plastic bag. Afterward, the plastic bag was tied, and covered the plastic box of known weight. After that, they were kept the beakers in a warm place or into an automated incubator at temperature for 28 days. When 28 days later, the treated Ash collected from each beaker, then began to step-by-step. First, the treated Ash was dried 102 ℃ for 8 ˚C to low moisture content for 8 hours. Following drying f exercise, the treated Ash was ground into powder for subsequent extraction operations [8].

2.4. Characterization the Morphologies of MWCNTs and SWCNTs

To analyze the gel and the Ash MWCNTs and SWCNTs the investigators used Field Emission Scanning Electron Microscope, FE-SEM (S-6840, Sony, Korea).

2.5. Extraction Procedure

2.5.1. Sequential of Three-Steps of Extraction Procedure

This method is used to describe the binding of heavy metals contained in differ- ent ash compositions [7]. The speciation lab was carried forward by the three-step sequential extraction technique [9,10] in accordance with Chinese Standard GB/T 25 282–2010. The speciation was classified as follows: F1, mild acid soluble fraction; F2, reducing fraction; F3, oxidizing fraction, and R, residual percent. In more details, 1.00 g fresh ash was taken and weighed in 100 mL polyethylene tube. The extract was filtered by a 0.45 μm membrane filter and was collected in a glass tube. Then, the sample was stored in a refrigerator at a temperature of −4◦C. The contents of Pb and Cd in all extraction processes were analyzed using the Laser-Induced Breakdown Spectroscopy (LIBS). To

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facilitate data comparisons, the residual content was calculated using the sum of F1, F2, and F3, subtracted from the total content.

2.5.2. Toxicity Characteristic Leaching Procedure

Through the usage of Toxicity Characteristic Leaching Procedure (TCLP) to determine the leaching propensity of heavy metal in the solid under acid rain. We combine sulfuric acid and nitric acid in proportion of 75%, mass fraction (w/w) of sulfuric acid is 97% and mass fraction (w/w) of nitric acid is 70% which the dilution water is 25 %. Mixing 16 mL of extract ant with 4 g of ash. (solution pH 1.83). Adding 1.7 g of sodium hydroxide into the solution (solution pH 4.2) and adding 1.43 mL nitric acid into solution (pH 1.83). Before each unit weight of 15 g of waste propped up by 0.6 g of ash was placed in a conical flask with 100 mL and put the cap on. To take 25 mL of extract ant solution to be added to the flask by cased comes with a 3-neck flask round bottom flask with reflux condenser, the solution within the flask were mixed and oscillated for 16 h at 150 rpm at 25 ℃ after filtration Vacuum filtration for 2 h to remove the unsolved and extraction solution from the residue, the residue was incubated at constant temperature cabinet for 6 h. The remaining precipitation of the heavy metals will get the retention in the column. Calculations of each metal were similar to the previous part.

2.5.3. Physiologically Based Extraction Test (PBET)

Established for bioaccessibility studies (used to find out the dissolution in digestive fluid) of heavy metals by digestive fluid [10].

2.6. pH and Organic-Matter Analysis

The data obtained from the measurements of pH and organic matter content are presented in Table 1. The pH of MWBA ranged from 8.8 to 10.6. The organic matter concentration in the ash is very high, ranging from 22.57% to 47.14%. This indicates that the ash contains a substantial amount of unburned organic matter, which can be attributed to the low operating temperature of the incinerator during combustion.

Table 1.
Measured pH and OM content (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%

3. Results

3.1. Measured Total Cd and pb Content

During the calculation, we set the denominator as the measured content of pb and Cd. This decision was made since the distribution of lead and cadmium in the Ash was uniform, which made it easier to compare horizontally. In general, the measured Pb content was approximately 92% of the expected amount, while the measured Cd concentration was approximately 96% of the expected content. The variations in the percentage of recovery may be attributed to system defects resulting from a combination of processes. In a separate investigation, the use of bismuth citrate ($C_6H_5BiO_7$) resulted in a recovery rate ranging from 75% to 85% for all tested elemental samples [10]. Thus, it was possible to determine the proportion of heavy metals in the extractant by referring to Table 2.

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3.2. Morphology

The shape of multi-walled carbon nanotubes (MWCNTs), single-walled carbon nanotubes (SwCNTs), and the ash treated with both sulfur/molybdenum (S/M) WCNTs was revealed by scanning electron microscopy (SEM) and Fourier-transform infrared spectroscopy (FTIR) images. Based on the observations depicted in Figure 1(a) and Figure 3(a), it was established that MWCNTs possess thinner, more curved, and longer nanostructures compared to SWCNTs. These features align with the description provided by the supplier's company. In addition, consider the case of Ash pb500+Cd250. The arrangement of MWCNTs on the surface of Ash particles may be shown in Figure 2(b)(c)(d).The positioning of SWCNTs on the identical surface is illustrated in Figure 4(b)(c)(d).The increase in the amount of M/S WCNTs on the particle's surface was clearly evident with the additions. Specifically, the addition of 0.10% indicated a significant accumulation of MWCNTs on the surface, whereas the addition of 0.1% resulted in a more uniform distribution. SWCNTs exhibited a similar behavior. Therefore, the study's findings suggest that increasing the amount of carbon nanotubes in the Ash would cause them to stack, resulting in a reduction in the effective contact area between the particle surface and the S/MWCNTs that are in touch.

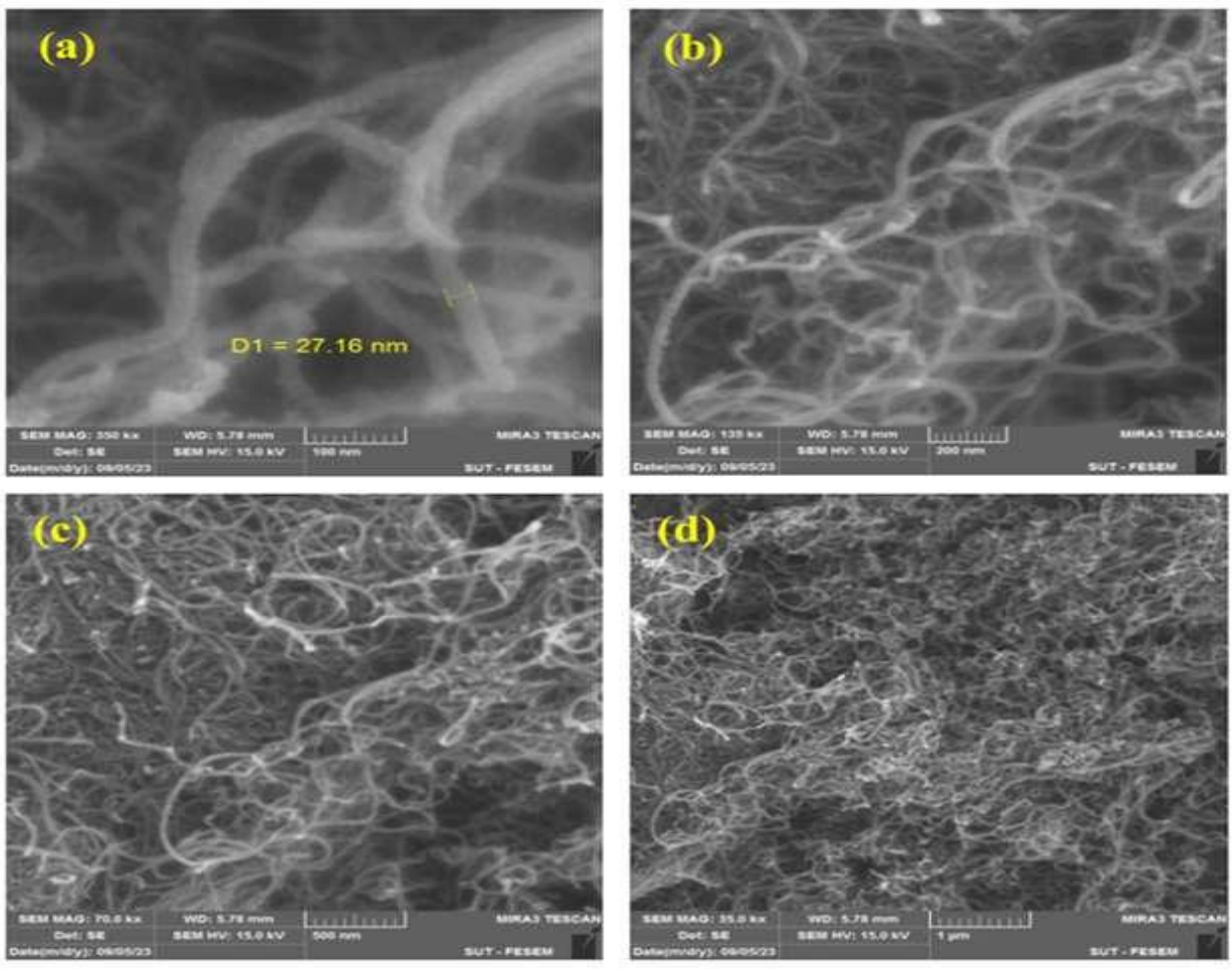

Figure 1.
Morphology of multi wall carbon nanotubes (MWCNTs) tested by scanning electron microscopy (a) under 100 nm magnification, (b) under 200 nm magnification, (c) under 500 nm magnification and (d) under 1µm magnification.

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3.3. Lead and Cadmium Speciation

The distribution of lead (Pb) exhibited notable dissimilarities compared to that of cadmium (Cd). Based on the data presented in Figure 1, the measured total content of F1–Pb, F2–Pb, F3–Pb, and R–pb accounted for 6.65%–9.21%, 7.43%–9.23%, 0.464%–0.80%, and 81.11%–93.81%, respectively. The measured total content of F1–Cd, F2–Cd, F3–Cd, and R–Cd accounted for different percentages for MWCNTs and the overall content. Specifically, for MWCNTs, the content ranged from 63.51% to 72.31% for F1–Cd, from 28.09% to 31.03% for F2–Cd, from 3.78% to 8.81% for F3–Cd, and from 0.52% to 4.00% for R–Cd. For the total content, the percentages ranged from 5.78% to 7.48% for F1–Cd, from 6.22% to 8.53% for F2–Cd, from 0.401% to 0.73% for F3–Cd, and from 78.01% to 89.63% for R–Cd. Meanwhile, the proportions of F1–Cd, F2–Cd, F3–Cd, R–Cd in SWCNTs were 61.56%–70.44%, 26.89%–30.14%, 3.22%–7.93%, and 0.44%–3.61%, respectively. The residual fraction primarily consists of heavy metals that are bonded to aluminosilicate, making them challenging to be ingested or transferred by organisms. The improvement of the residual fraction was a more reliable measure for evaluating the efficiency of immobilization in MWCNTs compared to SWCNTs [11].

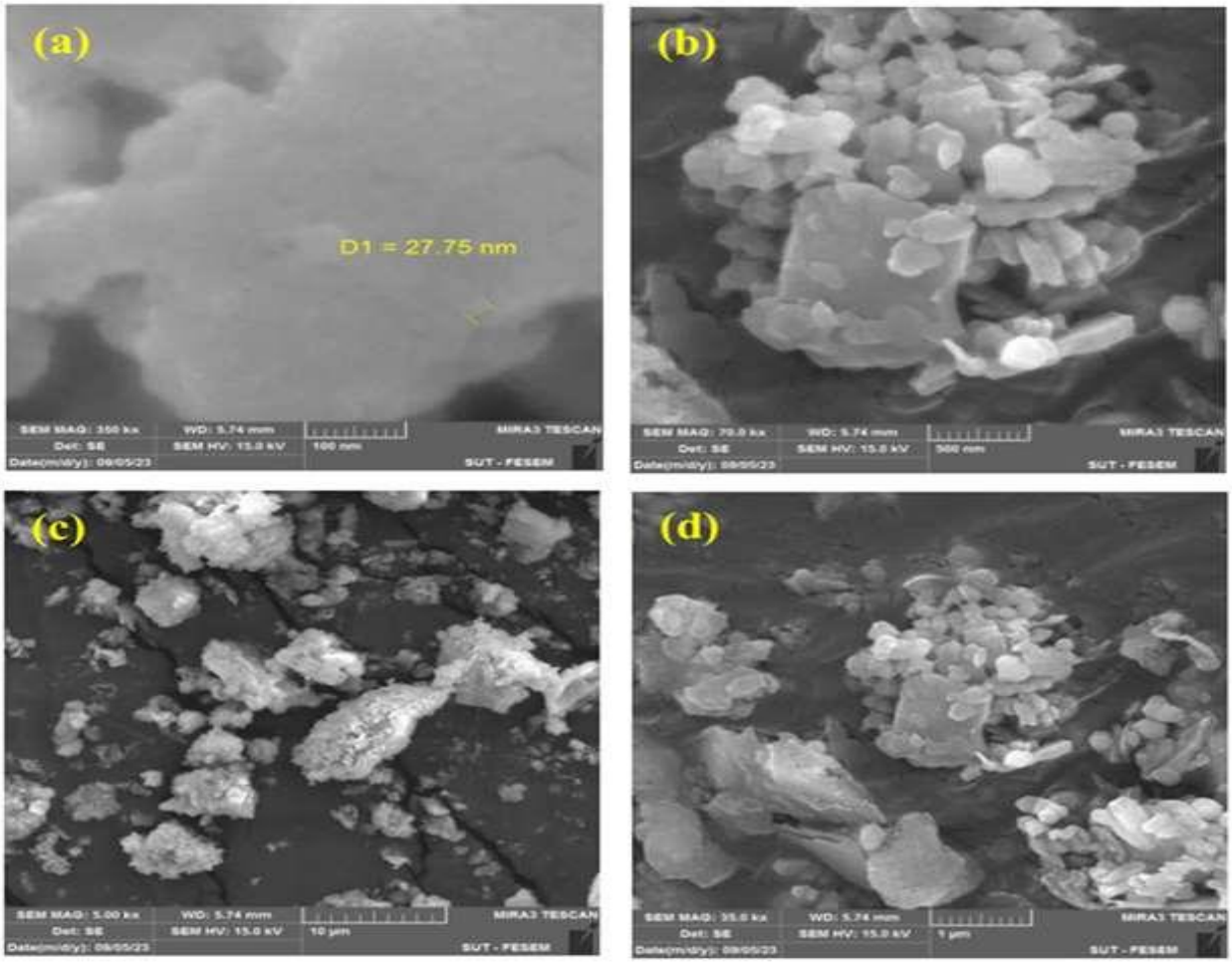

Figure 2.
Morphology of MWCNTs used in this study and the Ash treated with MWCNTs. (a) MWCNTs, (b) Ash pb 500 + Cd 250 treated with 0.10% MWCNTs, (c) Ash pb 500 + Cd 250 treated with 0.5% MWCNTs and (d) Ash pb 500+Cd 250 treated with 0.1% MWCNTs.

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Table 2.
Measured total Pb and Cd content by MWCNTs (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

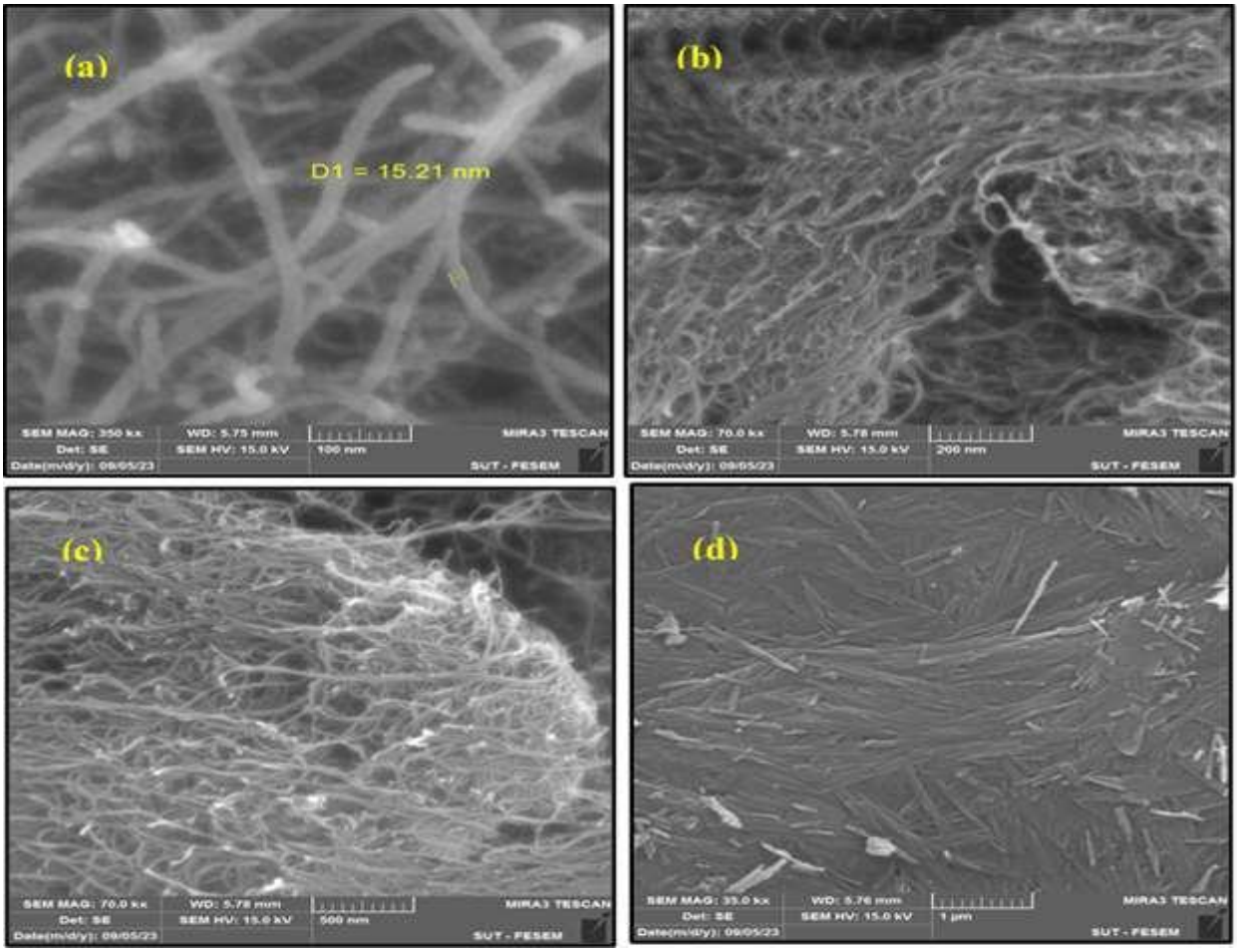

Figure 3. Morphology of single wall carbon nanotubes (SWCNTs) tested by scanning electron microscopy (a) under 100 nm magnification, (b) under 200 nm magnification, (c) under 500 nm magnification and (d) under 1µm magnification.

Table 3.
Measured total Pb and Cd content by SWCNTs (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	460.21	250	214.26
250 + 125	250	239.05	125	106.15
125 + 62.5	125	120.42	62.5	58.68

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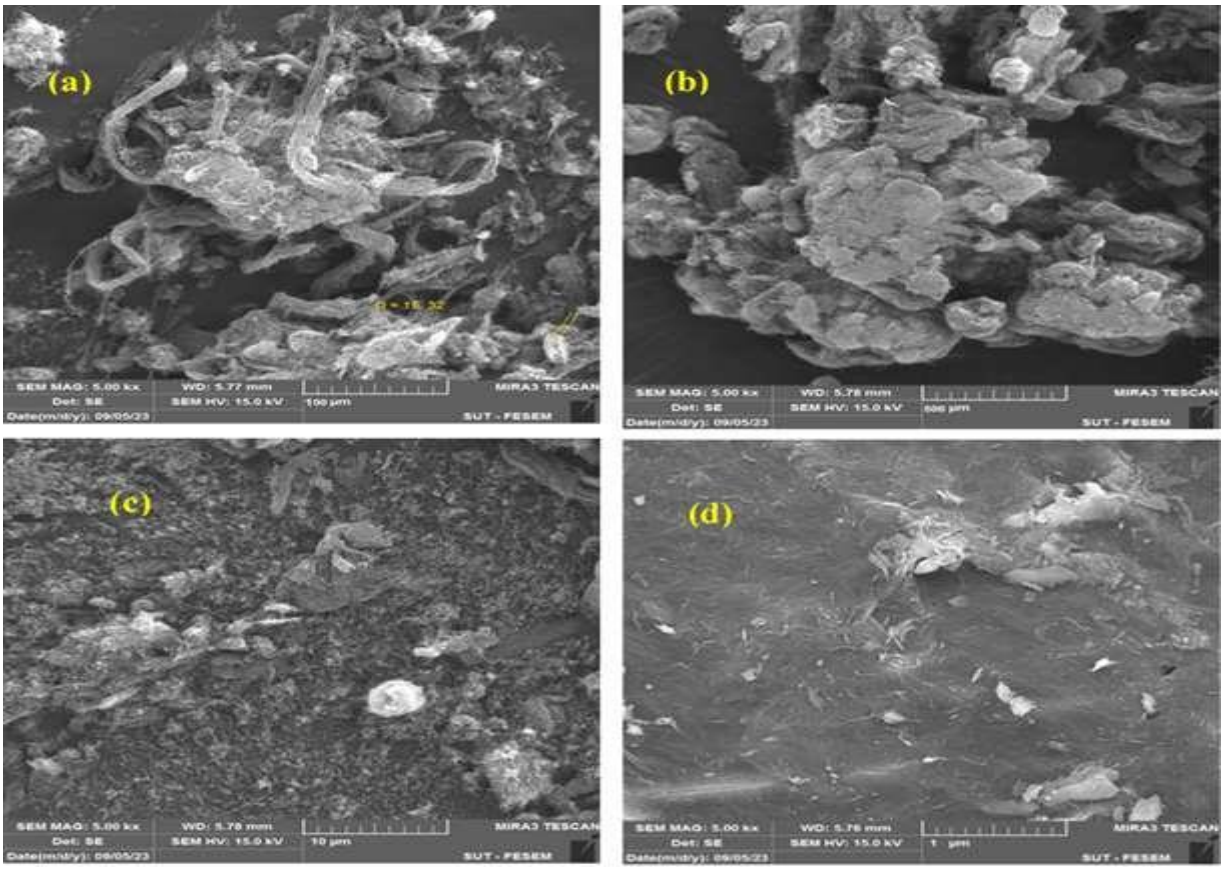

Figure 4.
Morphology of MWCNTs used in this study and the Ash treated with SWCNTs. (a) SWCNTs, (b) Ash pb 500 + Cd 250 treated with 0.10% SWCNTs, (c) Ash pb 500 + Cd 250 treated with 0.5% SWCNTs and (d) Ash pb 500+Cd 250 treated with 0.1% SWCNTs.

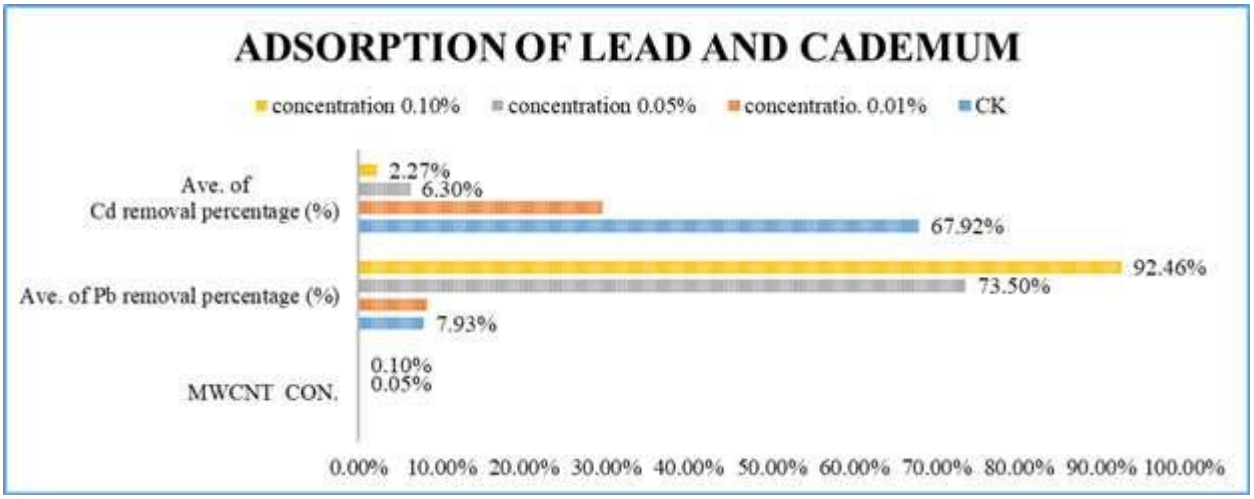

Figure 5.
The speciation of Pb and Cd in aSH treated with multi-walled carbon nanotubes (MWCNTs) and the control ash (CK, without MWCNTs treatment) were analyzed. The abscissas were represented by the sum of the heavy metal nominal content and the treatment level of MWCNTs. The data were presented as the average of three repeated measurements (M±AD, n=3).

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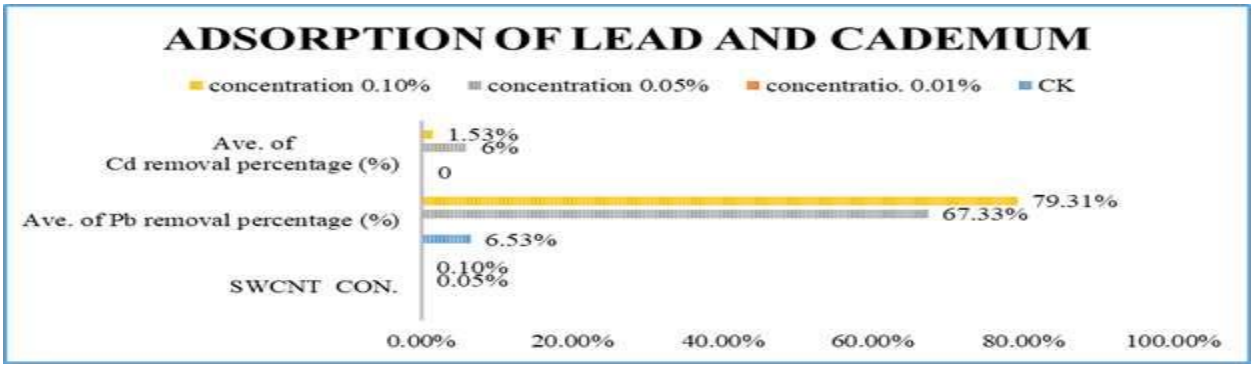

Figure 6.
The speciation of Pb and Cd in aSH treated with SWCNTs and in the control Ash (CK, without MWCNTs treatment) was analyzed. The abscissas were represented by the combined values of the heavy metal nominal content and the treatment level of SWCNTs. The data were presented as the average of three repeated measurements (M±AD, n = 3).

The fractions of pb that were controlled were F1–pb and F4–pb. The inclusion of 0.1% multi-walled carbon nanotubes (MWCNTs) has resulted in an 18.00% rise in F1–pb content under pb500 ($p < 0.05$). However, this effect was not observed under pb250 and pb125, nor with the addition of 0.5% and 0.10% MWCNTs. Furthermore, when considering all the additions, it was shown that multi-walled carbon nanotubes (MWCNTs) did not have a consistent impact on the four lead (pb) samples. Thus, it can be inferred that the increase in F1–pb occurred only in two specific situations. When SWCNTs were used, the pattern of F1 remained unchanged but with a reduced percentage (about 15.66%), following the same trajectory as MWCNTs. The elevated lead (Pb) concentration and limited incorporation of single/multi-walled carbon nanotubes (S/MWCNTs). When Cd125 was added at concentrations of 0.5% and 0.10%, the R–Cd increased by a factor of 2.6 and 2.5, respectively, for MWCNTS. For SWCNTs, the R–Cd increased by a factor of 1.8 and 2.1, respectively. However, the difference in content was only 2.04 mg/kg and 2.45 mg/kg for MWCNTs, and 1.88 mg/kg and 2.13 mg/kg for SWCNTs. When Cd250 was present, adding 0.1% resulted in a 6.52% rise in F1-Cd for MWCNTs and a 4.57% increase for SWCNTs. This suggests that adding lesser amounts of S/MWCNTs increased F1-Cd in spiking Ash with high levels of Cd. From the information provided, it can be inferred that only in Ash with a high concentration of pb and Cd, a smaller amount of S/MWCTNs was sufficient to enhance their mild acid-soluble fractions. As an intriguing consequence, this infers that decreasing the loading of S/MWCNTs may just slightly magnify the eco-hazard of Ash-pb and Ash- Cd.

4. Conclusion

The purpose of this experiment was to add S/MWCNTs to bio medical contaminated Ash and evaluate their efficiency in extracting pb and Cd. We tested three different levels of pb and Cd content (pb500 + Cd250, pb250 + Cd125, pb125 + Cd62.5) and four different levels of S/MWCNTs treatment (0%, 0.1%, 0.5%, and 0.10%). A three-step extraction procedure was used and three single extraction procedures too [12]. The SEM images and FTR images revealed a reduction in the contact area between S/MWCNTs and particle surface as the amount of S/MWCNTs increased. The rate of the two impact is MWCNTs>SWCNTs, this is a new observation in the analysis of "A" treated with S/MWCNTs [13]. MWCNTs at Pb content levels for 125 mg·kg-1 decreased pb bioavailability by 26.67% and SWCNTs decreased by 28.65% respectively, because the more gastric fluid, b (OH) adsorption sites may be found on MWCNTs while adsorption of SWCNTs. The addition of 0.5% to 0.10% for MWCNTs decreased pb bioavailability by 26.67% and 22.34% and SWCNTs decreased by 28.65% and 26.28%, respectively, at the Pb content level of 125 mg·kg-1. In Ash with a low concentration of pb (500 + Cd250), a negligible increase in the contents of mild acid-soluble pb and Cd by addition of 0.1% of SWCNTs and MWCNTs 16.30% and 11.40% of Cd. The increase of additions to

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SWCNTs and MWCNTs increase was 14.16% and 9.81%. This could potentially lead to an elevated ecological risk and impact [14]. It was concluded that the increased surface area of the ash particles facilitated better contact with the surrounding environment. However, no notable trend of extractability was seen in terms of TCLP-extractable pb and Cd. Upon data comparison, it was observed that MWCNTs exhibited a higher degree of selectivity in extracting ash heavy metals compared to SWCNTs. In conclusion, our work found that the presence of S/MWCNTs had an impact on the extractability of pb and Cd in the solution condition [15], [16].

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