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Single and Binary sorption of lead(II) and zinc(II) ions onto *Eichhornia Crassipes* (water hyacinth) ash

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Abstract— *Eichhornia crassipes* ash was used for the removal of Pb(II) and Zn(II) ions from mono and binary systems. Batch equilibrium studies were carried out under optimum conditions and the results were analysed by the Langmuir and Freundlich adsorption isotherms for single metal systems and by Langmuir competitive model for binary metal systems. The equilibrium data fitted well to both mono sorption isotherm models with $R^2 \geq 0.99$. The ash had greater affinity for Pb(II) ions ($q_m = 50 \text{ mg g}^{-1}$ and $n = 1.05$) than Zn(II) ions ($q_m = 16.67 \text{ mg g}^{-1}$ and $n = 1.04$) in mono sorption systems. There existed competition between Pb(II) and Zn(II) ions during their simultaneous removal by the ash ($R^2 \geq 0.97$). The ash showed preference for Pb (II) ion sorption ($q_m^* = 50 \text{ mg g}^{-1}$ and $q_m^*/q_m = 1$) than Zn(II) ions ($q_m^* = -2.94 \text{ mg g}^{-1}$ and $q_m^*/q_m = -1.53$) that was attributed to physicochemical properties of the cations. Pb (II) and Zn (II) ions can be significantly sorbed onto *Eichhornia crassipes* ash from single metal systems. However, in multi-metal systems, the removal of Zn (II) ions is inhibited by Pb (II) ions, while Pb(II) ions removal is not affected by the presence of Zn(II) ions.

Index Terms— Mono sorption, binary sorption, water hyacinth ash, lead and zinc.

I. INTRODUCTION

The accumulation of heavy metal contaminants in the environment has become a concern due to growing health risks to the public. These contaminants such as Pb(II) and Zn(II) ions enter the environment through industrial waste, mill tailings and landfill run off [1]. Lead poisoning causes various health hazards, such as damage to the liver, kidney and reduction of haemoglobin formation, mental retardation, infertility and abnormalities in pregnant woman [2]. Reference [3] reported that zinc causes non-fatal fume fever, pneumonitis and is a potential hazard as an environment pollutant. It was against this background that the two metal ions were targeted for removal.

The extensive literature available on metal-ion adsorption shows that many researchers focused on the use of plants in treating synthetic solutions containing a single metal ion [4]-[11]. However in practice, industrial waste waters usually contain more than one metal ion [12]-[14]. Reference [15] indicates that when more than one metal at a time is present the adsorbent selectivity becomes a precise requisite to be found in metal removal since there may be interference and competition for the adsorption sites. Preliminary investigation on the composition of the real effluent from heavy industries along Bristol Road of Gweru City of Zimbabwe indicated the following concentrations as determined by Flame Atomic Absorption Spectrometry (FAAS); $3.6 \pm 0.2 \text{ mg L}^{-1}$ for Pb and $1.93 \pm 0.03 \text{ mg L}^{-1}$ for Zn as the major heavy metal ions. The successful application of adsorption technology would require the knowledge of the interaction between these ions for the sorption sites on *Eichhornia crassipes* (*E. crassipes*) ash.

Research has demonstrated that *E. crassipes* is capable of accumulating trace amounts of Ni, Fe, Co, Pb, Cu, Cd and Zn. Most studies have focused on living plants [16]-[20] and non-living forms of the plant [10], [12]-[14], [21]-[25]. None of these studies have focused on the removal of Pb(II) and Zn(II) ions from binary systems using the ash of *E. crassipes* plant. In our earlier work [14] a report was made that the weed is a perennial problem at Lake Chivero, the main water source for the capital city of Zimbabwe, Harare. Chemical treatment of *E. crassipes* was unsuccessful and the city authorities have since resorted to its physical removal from the water surface to pave way for recreation activities. This has led to the accumulation of decaying weed on and around the banks of the Lake. Better ways of managing this abundant waste material have to be explored. Incineration of the waste is a potential method to manage this waste and the ash produced can be investigated for heavy metal ion removal from waste waters.



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Reference [26] reports a high efficiency removal of cadmium ions by *Ulmus* leaves ash and [27] obtained similar results on cadmium adsorption by wood ash. The ash particles offer the advantages of having large surface area and offering minimum disposal problems since the metal-ion loaded ash can be used as an additive to cement in concrete making. The aim of this study was to generate *E. crassipes* ash and investigate its potential use in the removal of Pb(II) and Zn(II) ions from mono and binary metal ion systems.

II. MATERIALS AND METHODS

Biomass preparation and activation

Samples of mature *E. crassipes* plants were harvested from Lake Chivero near Harare, Zimbabwe. The plants were washed three times with lake water before they were packed in polythene bags [14]. The plants were washed thrice with running tap water and twice with de-ionized water before they were sun dried. Complete drying was achieved by oven-drying the sample at 105 °C for six hours. 300 g of the dried sample were burnt at 800 °C in an electric muffle furnace (Clifton GB 004). The ash produced was acid-treated with 1000 cm³ of 1 M HCl over a shaker set at 150 rpm for 24 hours followed by washing with distilled water before it was oven dried to constant mass at 65 °C. It was crushed and ground with a mortar and pestle and then sieved using a polythene sieve (size fraction of 0.5 mm). The ash was stored in airtight containers and subsequently used in the adsorption experiments.

Determination of optimum conditions

Batch sorption studies were conducted to determine the optimum pH, contact time and ash dosage in the sorption of Pb(II) and Zn(II) in single metal ion systems. These were identified as the most critical parameters that affect the adsorption process. Temperature, shaking rate and solution volume were maintained constant throughout the study at 25 ± 2 °C, 150 rpm and 100 cm³ respectively. Repeatability precision was ascertained by carrying out replicate analysis on each investigation. Systematic instrument errors were reduced by proper instrument calibration and determination of both initial and residual metal ion concentration by Flame Atomic Absorption Spectrometer (FAAS)

Sorption equilibrium studies

(a) Mono sorption studies.

The study was carried out under the identified optimum conditions. In each case, the volume of solution was maintained at 100 cm³, a temperature of 25 ± 2 °C, pH of 6, shaking rate of 150 rpm, biomass doses of 6 g L⁻¹ (Pb) and 8 g L⁻¹ (Zn) and contact time of 120 minutes. The initial metal ion concentration was varied from 0.4 to 2 and 5 to 25 ppm for Zn(II) and Pb(II) ion concentrations respectively. The initial and residual metal ion concentrations were determined by FAAS.

(b) Binary sorption studies

The investigation was done to examine the interactions between Pb(II) and Zn(II) ions during their sorption onto the surface of *E. crassipes* ash. Solutions of Zn-Pb combinations were prepared in the concentration ratios of 0.4:8, 0.8:12, 1.2:16, 1.6:20 and 2.0:24 as in [28]. These were transferred to 250 cm³ Erlenmeyer flasks and contacted with 0.7 g of *E. crassipes* ash at pH 6, contact time of 120 minutes and a shaking rate of 150 rpm. The mixtures were vacuum filtered and the initial and residual metal ion concentrations determined by FAAS.

III. RESULTS AND DISCUSSION

Isotherm studies

Adsorption isotherms have constants that are used to infer surface properties and the potential of the adsorbent to remove the sorbate. The isotherm studies were investigated using equilibrium sorption data generated under the optimum pH (6), contact time (120 minutes) and ash dose (7g L⁻¹). The isotherm models, Freundlich, Langmuir and Langmuir competitive (Table 1), were used to analyse data of metal sorption by *E. crassipes* ash in single and binary systems respectively.

(a) Mono sorption systems

The Langmuir and Freundlich adsorption isotherms were used to analyse the equilibrium data. The generalized and linearized isotherm equations are shown in Table 1.



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Table 1. Adsorption isotherm equations [10]

Isotherm	Generalized form	Linearized form
Langmuir	$q_e = q_m b C_e / (1 + b C_e)$	$1/q_e = 1/q_m + 1/(b q_m C_e)$
Freundlich	$q_e = K_f C_e^{1/n}$	$\ln q_e = \ln K_f + 1/n \ln C_e$
Langmuir Competitive	$q_{ei} = q_{mi} b_i C_{ei} / (1 + \sum_{j=1}^{j=n} b_j C_{ej})^{-1}$	$C_{e1}/(C_{e2} q_{e1}) = C_{e1}/(q_{m1} C_{e2}) + b_2/(b_1 q_{m1})$ $C_{e2}/(C_{e1} q_{e2}) = C_{e2}/(q_{m2} C_{e1}) + b_1/(b_2 q_{m2})$

Figures 1 and 2 show the linearized Langmuir plots for the Pb(II) and Zn(II) ions adsorption onto the *E. crassipes* ash.

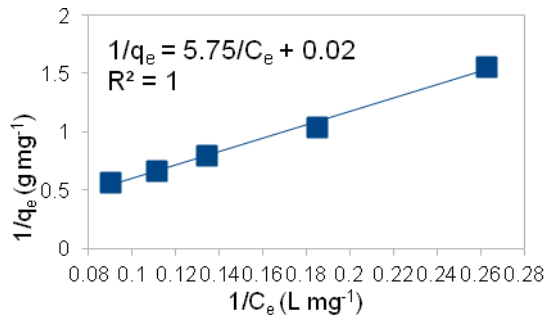


Fig 1. Langmuir plot of Pb sorption onto ash

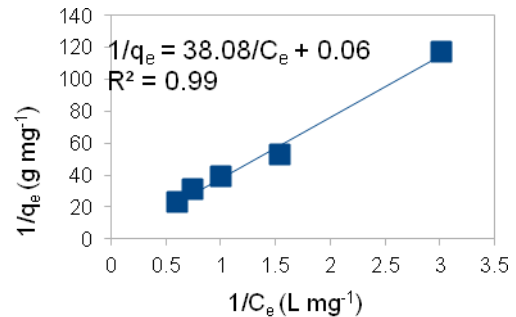


Fig 2. Langmuir plot of Zn sorption onto ash

Table 2. Monosorption isotherm parameters

Langmuir				Freundlich		
Metal ion	q_m (mg g ⁻¹)	b (L mg ⁻¹)	R^2	K_L (mg ¹⁻ⁿ g ⁻¹ L ⁿ)	n	R^2
Pb(II)	50	3.48×10^{-3}	1	0.18	1.05	1
Zn(II)	16.67	1.58×10^{-3}	0.99	0.03	1.04	0.99

Table 2 shows the monosorption parameters. The correlation of determination ($R^2 \geq 0.99$) shows that the data for the adsorption of both metal ions fitted to both adsorption isotherms. However, the Pb(II) ($R^2 = 1$) adsorption data fitted both the Langmuir and Freundlich adsorption models better than that of Zn(II) ions ($R^2 = 0.99$). The applicability of the Langmuir isotherm indicated the monolayer coverage of Pb(II) and Zn(II) ions on the homogeneous surface of the *E. crassipes* ash [29] while the applicability of the Freundlich isotherm suggested the heterogeneous nature of the ash surface hence adsorption was random due to existence of a random distribution of energetically different active sites [30]. The fitting of the adsorption equilibrium data to the two models revealed the complex nature of the surface structure of the *E. crassipes* ash. The maximum sorption capacities (q_m) were 50 mg g⁻¹ and 16.67 mg g⁻¹ and the free energy of sorption (b) of 0.0035 and 0.0016 L mg⁻¹ for Pb(II) and Zn(II) respectively were obtained. The Freundlich isotherm parameters also pointed to a similar trend with K_f values of 0.18 and 0.03 and the adsorption intensity (n) of 1.05 and 1.04 for Pb(II) and Zn(II) ion sorption respectively. Both model parameters revealed that *E. crassipes* ash could be used successfully to remove Pb(II) and Zn(II) ions from waste waters. The results were comparable to those obtained by [31] who obtained the trend; Cu = Pb > Zn using water hyacinth ash. Reference [32] investigated metal-ion sorption using *Lagenaria vulgaris* shell and got the sorption order; Pb²⁺ > Cd²⁺ > Zn²⁺ that was in agreement with the current findings.

(b) Binary sorption system

The Langmuir competitive model (LCM) was used to analyse the equilibrium data obtained during the simultaneous sorption of Pb(II) and Zn(II) ions onto *E. crassipes* ash. The model helped to identify the nature of

interaction between the metal ions to occupy the adsorbent sites [33]. Reference [34] reported that the existence of competition between any given sorbates can be established by LCM. Quantitative evaluation of sorbate interaction was achieved by using the ratio q_m^*/q_m (where q_m^* is the metal sorbed at equilibrium in the presence of another metal ion and q_m is the biosorption capacity in single metal systems). When the ratio $q_m^*/q_m > 1$, synergism; adsorption is promoted by the presence of the other metal ion; if $q_m^*/q_m = 1$, non-interaction; the mixture has no effect on the adsorption of the adsorbate and if $q_m^*/q_m < 1$, antagonism; the effect of the mixture is less than that of each of the individual adsorbate in the mixture [10], [15].

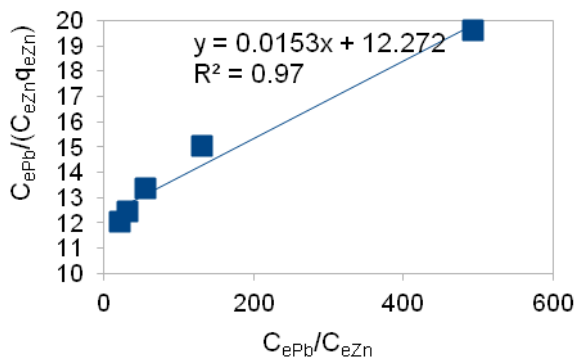


Figure 3. Binary sorption of Pb(II) in Zn(II) by ash

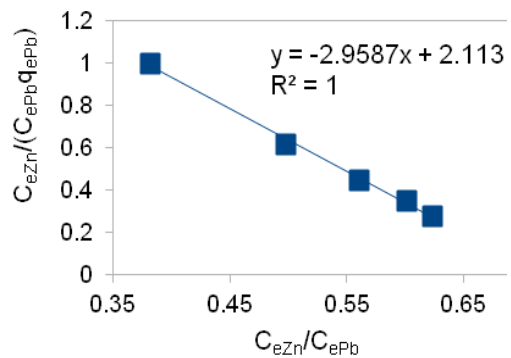


Figure 4. Binary sorption of Zn(II) in Pb(II) by ash

The binary sorption data for both metal ions fitted well to the LCM with R^2 values of 0.97 and 1 for Pb(II) and Zn(II) ions respectively (Figs 3 and 4). This implied that there was competitive interaction in the sorption of these ions in the concentration ranges investigated [14]. The total adsorption capacities for the mixtures (47.06 mg g^{-1}) was found to be higher than the adsorption capacity of Zn(II) alone ($q_m = 16.67 \text{ mg g}^{-1}$) but was lower than the adsorption of Pb(II) ($q_m = 50 \text{ mg g}^{-1}$) in a single component system. This indicated that the binding sites for Zn(II) might not be the same as those for Pb(II) ions and the adsorption of Pb(II) was more favourable than the adsorption of Zn(II) ions onto *E. crassipes* ash [35].

Table 3. Langmuir competitive model parameters

Metal ion	q_m^* (mg g^{-1})	$*b/b$	R^2	q_m^*/q_m
Pb/Zn	50	613.5	0.97	1
Zn/Pb	-2.94	-2.09	1	-1.53

* b/b specifies the ratio of the b parameter of the interfering metal ion to that being adsorbed (b).

Figure 4 and Table 3 show that the removal of Zn(II) ions in the presence of Pb(II) ions fitted well to the LCM ($R^2 = 1$). However, the maximum sorption capacity and affinity ratio were both negative and of low magnitudes. This indicated the inhibition of/or improbable sorption of Zn(II) ions as a result of the presence of Pb(II) ions under the range of concentration investigated. The negative q_m and b parameters implied the desorption of the sorbate, Zn(II), from the sorbent (ash) into the solution.

Negative Langmuir parameters have been interpreted in different ways. Reference [36] obtained negative Langmuir constants during the biosorption of Pb(II) and Zn(II) ions using unmodified and EDTA-modified maize husk. They attributed the result to the non-fitting of the data to the model. Reference [37] obtained the negative Langmuir b parameters relating to multi-metal system that was attributed to a reverse reaction or tendency of Zn(II) ions to desorb when in competition with Pb(II) ions. The meaning of the negative Langmuir sorption parameters needs to be established through further investigations. It was deduced that no significant sorption of Zn(II) ions is possible from Pb(II)-rich waste waters.



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There was no change in the maximum sorption capacity of the ash for Pb(II) ions in single and binary systems ($q_m^*/q_m = 1$ in Table 3). The mixture had no effect on the adsorption of Pb(II) ions confirming the existence of different sorption sites for Pb(II) and Zn(II) ions on the ash. This implied that *E. crassipes* ash could be used to remove Pb(II) ions in the presence of Zn(II) ions from waste waters. The binary sorption of Pb-Zn performed by [38] using activated sludge concluded that the equilibrium concentration of Zn would have to be 70 times greater than that of Pb to obtain the same proportion of uptake for each metal. The trend, $Pb \gg Zn > Cd$, was obtained by [14] using *E. crassipes* root powder. Reference [2] arrived at the same trend to that of [14] using orange wastes as the biosorbent. Other researchers who have investigated multi-metal systems include [37], who used polyurethane-immobilised *Sphagnum* moss and found the metal uptake capacity of the biomass to be in the order: $Pb > Cu > Zn > Ni$. Basing on current results and previous findings on multi-metal sorption systems, the order of metal uptake capacities are independent of the biosorbent used but depends on the physicochemical properties of the ions under investigation. Table 4 shows the ionic properties of Pb(II) and Zn(II) ions.

Table 4. Metal ionic properties

Metal ion	Pb(II)	Zn(II)
Atomic weight	207.2	65.39
Coordination number	(4,6)	4.00
Electronic structure	$[Xe]4f^{14}5d^{10}6s^26p^2$	$[Ar]3d^{10}4s^2$
Electronegativity	2.33	1.65

The preference of *E. crassipes* ash for Pb(II) ions over Zn(II) ions can be attributed to the differences in the ionic properties. A metal ion with unpaired electrons could be attracted by a magnetic field originating from the adsorbent and a high ionic weight implies greater momentum that results in an effective collision between the metal ion and the solid surface. Electrostatic attraction of the metal ions by the cationic sorbent increases with increase in the electronegativity of the metal ions [2]. Pb(II) ion has the larger atomic weight, two coordination numbers (4,6), is paramagnetic and more electronegative compared to Zn(II) ion [13]. Reference [37] presented that lead ion's first hydrolysis constant is much greater than that of zinc ions ($\log K_{Pb} = -7.71 > \log K_{Zn} = -8.96$) implying that Pb(II) are readily hydrolysed compared to Zn(II) ions. These are the ionic properties that gives rise to the favourable sorption of Pb(II) ions over Zn(II) ions by *E. crassipes* ash.

IV. CONCLUSION

Batch adsorption experiments for the removal of Pb(II) and Zn(II) ions both in single and binary systems by *E. crassipes* ash were simulated. The adsorption was investigated at optimum pH values, contact time and ash dose at a constant temperature of 25 ± 2 °C and a constant flask shaking rate of 150 rpm. The results from the investigation of the effect of initial concentration were fitted to Freundlich and Langmuir and Langmuir competitive models for the single and binary systems respectively. The results obtained are summarised below:

- The experimental equilibrium data in single metal ion systems fitted the Langmuir ($R^2 = 1$) and Freundlich ($R^2 = 1$) for Pb(II) sorption. R^2 value was 0.99 for Zn(II) ions in both isotherm models. The sorption of metal ions was both homogeneous and heterogeneous in nature.
- Maximum sorption capacity (q_m) for Pb(II) (50 mg g^{-1}) was much higher than that of Zn(II) (16.67 mg g^{-1}) and the same difference was observed on the binding energy parameters (b) Pb(II) (0.0035 L mg^{-1}) and Zn(II) (0.0015 L mg^{-1}). The Freundlich isotherm parameters K_L and n indicated that Pb(II) ($K_L = 0.18$ and $n = 1.05$) were better than those of Zn(II) ($K_L = 0.03$ and $n = 1.04$) in single metal ion systems.
- There existed competition for the sorption sites during the adsorption of Pb(II) ions in the presence of Zn(II) ions ($R^2 = 0.97$) and Zn(II) ions in the presence of Pb(II) ions ($R^2 = 1$) by *E. crassipes* ash with Pb(II) ions emerging as the more preferred ion ($Pb \text{ } q_m^* = 50 \text{ mg g}^{-1}$ while $Zn \text{ } q_m^* = -2.94 \text{ mg g}^{-1}$).
- *E. crassipes* ash could be successfully used as an adsorbent of Pb(II) and Zn(II) ions from single metal systems. The binary sorption results showed that the ash had a greater preference for the removal of Pb(II) ions than Zn(II) ions that the sorbent would remove Pb(II) significantly prior to the removal of Zn(II) ions.



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- Utilisation of Coal Fly Ash Zeolites in the Removal of Copper (II) Ions from an Aqueous Solution [Accepted for publication: Engineering Science and Technology: An International Journal (ESTIJ)].
- Synthesis of Zeolites and Their Applications in Heavy Metals Removal: A Review [Accepted for publication: IRACST–Engineering Science and Technology: An International Journal (ESTIJ)].
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