

# Adsorption of various heavy metals through dehydrated powder of two aquatic plants (*Lemna minor* and *Eichhornia crassipes*) from aqueous medium

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## Abstract

Different heavy metals can be eliminated by non-living or living biomass. Various aquatic plants species can be applied for the removal of metals. In this research, cadmium, copper, zinc and lead adsorption properties of *Lemna minor* L. and *Eichhornia crassipes* (Mart.) Solms were examined. This work was carried in the laboratory to ascertain the efficiency of aquatic plants *Eichhornia crassipes* and *Lemna minor* on the removal of various metals. Two plants *Lemna minor* and *Eichhornia crassipes* were obtained in a dehydrated powder type. The adsorption procedure of various metals was investigated under different pH ranges, metal ion concentration and reaction times as well as noticed the chemical composition of these plants. The results indicated that both submerged aquatic plant powders can be effectively used for heavy metal elimination under various factors. The FTIR spectra analysis showed a various functional groups in both aquatic plants, they accelerated the adsorption process.

## Keywords

*Aquatic, heavy metals, pH, reaction time, adsorption*

## Introduction

Metals contamination of water is a biggest ecological issue immediately demanding to be resolved. Identified causes of metals pollution in the environment such as agricultural, geogenic, pharmaceutical, industrial and domestic effluents can induce water heavy metal pollution (Algul and Beyhan, 2020; Hubeny *et al.*, 2021; Rai *et al.*, 2019). Metals pollution can be eliminated via aquatic plants but cannot be degraded; consequently these metals accumulate by food web and induce adverse health effects for long term (Rajeshkumar *et al.*, 2018; Zakaria *et al.*, 2021). Adsorption, ion exchange, reverse osmosis, membrane filtration and chemical deposition methods are used to manage and control

water heavy metal contamination; adsorption is a most common and broadly used technique (Qasem *et al.*, 2021). Activated carbon (AC) is a most frequently used adsorbent; conversely cost is immense, and thus inexpensive, environmentally friendly, more attainable and most effective adsorbent are being investigated (Block *et al.*, 2021; Saleem *et al.*, 2019). Pine bark, loofah sponge, mushroom residues, modified chitosan, cassava, seaweed, sawdust, pine leaves and needles have been applied as prospective adsorbents in the research works (He *et al.*, 2020). Aquatic plants have higher surface areas and better efficiency on the adsorption of heavy metals (Elbasiouny *et al.*, 2021). It is also probable to apply adsorbent plants substance in a form of dried powder but studies on their metals elimination

characteristics and effects of the environmental factors on the elimination ratio are exquisite. In this work, 2 submerged plants were produced as dehydrated powders and their removal properties of Zinc (Zn), lead (Pb), cadmium (Cd) and copper (Cu) and impacts of the environmental factors, such as (metal initial concentration, contact time and pH values) that influence adsorption efficiency were equated. The effect of the various functional groups in elimination process was explained, and they would be worthwhile references for selecting and applying metals adsorbent.

## **Materials and Methods**

### **Aquatic plants**

Flourishing submerged plants *Lemna minor* and *Eichhornia crassipes* were taken from green-house and taken back to research laboratory. Both aquatic plants were cleaned by de-ionized water and kept in desiccator at 70 °C for dry, and then made powder of dried material by mortar, separated by a 2mm mesh and kept in the polyethylene bags for use.

### **Chemicals**

5% HNO<sub>3</sub> was used to diluted ZnSO<sub>4</sub>·7H<sub>2</sub>O, Cd(NO<sub>3</sub>)<sub>2</sub>, CuSO<sub>4</sub>·5H<sub>2</sub>O and PbSO<sub>4</sub> to make 1000 mg/l Zn<sup>2+</sup>, Cd<sup>2+</sup>, Cu<sup>2+</sup> and Pb<sup>2+</sup> correspondingly. The stock was unswervingly diluted to required ion level in the experiment. The NaOH and HNO<sub>3</sub> solutions were used for the adjustment of pH ranges.

### **Elemental and chemical composition of both aquatic plants**

Total carbon and total nitrogen contents were determined by CHN analyzer. The amounts of non-structural polysaccharides and soluble sugars were measured colorimetrically with anthrone using a spectro-photometer (Moretti et al., 2020). Non-structural carbohydrates were computed as sum of non-structural polysaccharides and soluble sugars. Organic acids content was determined on the base of nitrate content and ash pH measurement (Wang et al., 2022). All the parameters were determined in three replicates and mean values are exhibited.

### **FTIR Spectra analysis of plant powders**

The FTIR spectra of samples was recorded in FTIR instrument (Model/Make: IFS 25, Bruker, Germany), with PC based software controlled instrument operation

and data processing. A small amount of powdered leaf samples were made into pellets using KBr for FTIR analysis and a thin film was prepared by applying pressure. The data of infrared transmittance was collected over a wave number ranged from 4000 cm<sup>-1</sup> to 500 cm<sup>-1</sup>. All the samples were analyzed in triplicates with plain KBr pellets as blank. The spectral data were compared with a reference to identify the functional groups existing in the sample.

### **Determination of the optimum aquatic plant powder (dose of powder)**

Both plants powder in range of 1-50 g/l were mixed to various bottles comprising 100 mg/L from every metal solution to examine the optimal plants powder for metals sorption. The highest sorption around 98% of metals ions were achieved, when plants powder of solutions approaching 20 g/l attuned at pH 6 within two hours. Consequently, the optimal amount of powders was taken as 20 g/l for next experimentations.

### **Determination of the optimum reaction time**

0.50 g of both plants powder was mixed to the 40 ml metals solutions with initial level of 100 mg/l adapted to the pH 6 employing 1 mol/L nitric acid in a bottle. The samples were taken at 10, 20, 30, 40, 50, 60, 80, 100, 120 minutes, filtered and analyzed through flame atomic absorption spectrophotometer (FAAS). Flame atomic absorption spectroscopy (FAAS) model AA-7000 Shimadzu was used as instrumental detection system using hollow cathode lamps. Absorbance measurements of blank (solvent) were recorded using deuterium lamp. Air/acetylene burner head was used as a carrier gas for all samples (Jasim et al., 2020).

### **Effect of different pH ranges**

The pH for the metals biosorption were examined through mixing 0.50 g with level of 20 g/l plants powder to 40 ml metals solutions with initial level of 100 mg/l attuned to different pH ranges (1-8) employing 1 mol nitric acid solution in the bottle. Samples were shaken at 140 rpm for 2 hours. After 2h samples were filtered by 0.45 μm membrane and analysed by AAS.

### **Effect of different concentrations of metals**

3.5 g plants powder was mixed to 40 ml metals solutions attuned to the pH 6 employing 1 mol/l nitric acid in bottle. The initial levels of metals were 10, 20, 50, 100, 200, as well as 500 mg/l. These samples were shaken

at 180 rpm for 2 hours. After 2 h, the samples filtered and analysed via AAS. The findings from 3 replicate samples.

### Analysis of data

The adsorption mechanism  $q$  (mg/g) was measured conforming to the adopting formula

$$q = \frac{m_{ion}}{m_s} = (C_i - C_f)V/m \quad [1]$$

where, ratio of the metals adsorption  $g$  (%) =  $(C_i - C_f)/C_i$ ,  $m_{ion}$  is mass of metals ions (mg),  $m_s$  is mass of plants sample (g),  $C_i$  is initial level (mg/L),  $C_f$  is reacted mass level (mg/L), as well as  $V$  is reaction solutions volume (L).

Freundlich and Langmuir adsorption models maybe applied to report the metals absorption mechanism. Langmuir model can be stated in the adopting formula (Febrianto *et al.* 2009).

$$C_e / q_e = (1/b q_{max}) + C_e / q_{max} \quad [2]$$

where,  $C_e$  is equilibrium level (mg/l),  $q_e$  and  $q_{max}$  are the equilibrium and highest adsorption capability (mg/g), correspondingly and  $b$  is the equilibrium continual (Gupta and Rastogi, 2008).

Freundlich adsorption model can be stated in the adopting formula,

$$1n q_e = 1n K_F + (1n C_e) / n \quad [3]$$

where,  $K_F$  is Freundlich Co-efficient (mg/g), representing adsorption capability and  $n$  is Freundlich indicant showing amounts of the adsorption.

Lagergren's first and second order equations are applied to explain the kinetics of metals adsorption (Gupta and Rastogi 2008). The linear form of Lagergren first order rate formula is as follows,

$$\log (q_e - q_t) = \text{Log } q_e - K_1 t, \text{ ad} \quad [4]$$

where,  $K_1$ , ad is the first order rate continual ( $\text{min}^{-1}$ ), and  $q_e$  and  $q_t$  are the equilibrium time and adsorption capability during time period  $t$  (mg/g), correspondingly. The linear form of Lagergren's second order formula,

$$t / q_t = 1 / (K_2 \text{ ad } q_e^2) + t / q_e \quad [5]$$

where,  $K_2$  ad is a pseudo 2nd order equilibrium continual (g/mg/min).

## Results and Discussion

### Elemental and chemical composition of both aquatic plants

Carbon contents noticed in *Lemna minor* and *Eichhornia crassipes* 354 and 447 mg/g, respectively. Maximum values were observed in *Eichhornia crassipes*. Maximal amount of C contents is known to be needed for production of cell wall substances such as cellulose and lignin. Similarly, Nitrogen components found higher amount in *Eichhornia crassipes*. Its higher amount of nitrogen contents in *Eichhornia crassipes* may be due to naturally occurrence of nitrate in water. Storage of nitrogen in the environment could be showed via C/N ratio in aquatic plants. The total nitrogen content in both plants is generally related to their photo-synthetic actions because almost 70% of leave organic N is nitrogen of the chloroplast proteins involved in carbon dioxide assimilation (Bassi *et al.*, 2018). In our study showed lower contents of organic acids in both plants. It might be mainly related to particular attributes of N metabolism in the aquatic plants. Hence, lower contents of organic acids in both plants are obviously related to prevalent use of the ammonium N via both plants. Non-structural carbohydrates contents were observed 191 and 246 for *Lemna minor* and *Eichhornia crassipes*, respectively. Also we noticed low level of soluble carbohydrates and higher levels of non-structural polysaccharides.

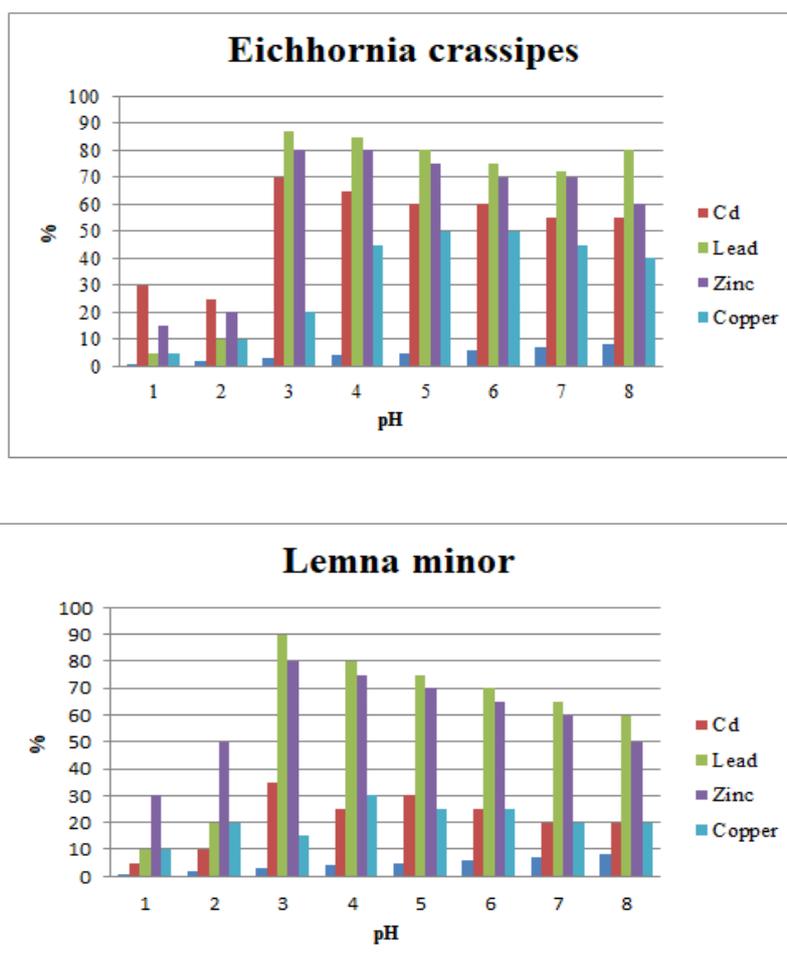
**Table 1.** Chemical composition of both aquatic plants

Aquatic plants	Content (mg/kg)						
	Nitrogen	Carbon	Mineral compound	Soluble carbohydrate	Non-structural carbohydrate	Non-structural polysaccharide	Organic acid
Lemna minor	22	354	59	11	191	180	91
Eichhornia crassipes	38	447	63	17	246	132	53

### Adsorption mechanism

Influences of various factors on metals adsorption in dried aquatic-plants powder under various pH, initial concentration and contact time exhibited a great consistency for each metal. Highest adsorption took place at a pH of 3 for lead and at a pH of 4 for copper (Cu), Zinc (Zn), cadmium (Cd) (Fig. 1), with except of lead (Pb) elimination through *Lemna minor*,

which enhanced slowly with contact time and metals adsorption of both plants powders achieved highest within initial 10 minutes for each heavy metals (Fig. 2). Under various initial levels, lead adsorption of both plant powders enhanced linearly with lead level and did not seem to achieve the saturation even at highest level investigated. Adsorption of the copper (Cu), zinc (Zn) as well as cadmium (Cd) was saturated, while the solution concentration achieved certain degrees (Fig. 3).



**Figure 1.** Influences of different pH values on the metal removal % of both aquatic plant powders

Solution's acidity is a key aspect affecting for metals adsorption and availability. It generally impacts chemical states of the metal ions and adsorption sites for the metals on the surfaces of adsorbents. While pH is lower, linker on the surfaces of adsorbents substance will be tenanted through metals ions and hydrated hydrogen ions getting adjacent because of repulsions. When the pH is enhanced, the level of the hydrogen ions reduces, disclosing more  $-ve$  charged connectors. This expedites the adsorption of the metals ions to the

active sites. Whenever, pH is raise, a lot of metals ions in the solution will organize as  $OH^-$  constituents and adsorption mechanism cannot continue (Egbosiuba *et al.*, 2021; Lusa *et al.*, 2021). In this research, for both plants, the adsorption of lead (Pb) was immensely less when the pH was lower than 3. The adsorption of zinc (Zn), copper (Cu) and cadmium (Cd) was immensely less when the pH range was lower than 4. For both plant powders, adsorption of the zinc, copper, cadmium and lead was considerably reduced when pH value was

above 5, probably this result due to dissolved states of the metals ions.

Initial levels of the metals impacts on adsorption process (Chen *et al.*, 2021). Usually, the adsorption mechanism occurs at the specific adsorption sites, when level is less and the specific adsorption situates for the adsorption process will be saturated with level enhance until it

progressively attains the point, where interchange of the tenanted sites is only mechanism happening (Liu *et al.*, 2019). Under every value of pH, contact time and temperature, enhance in the concentration of Pb adsorbed was proportionate to weight of the plant powder firstly extant and the adsorption of Zn, Cu, and Cd was proportionate to initial levels in the solution.

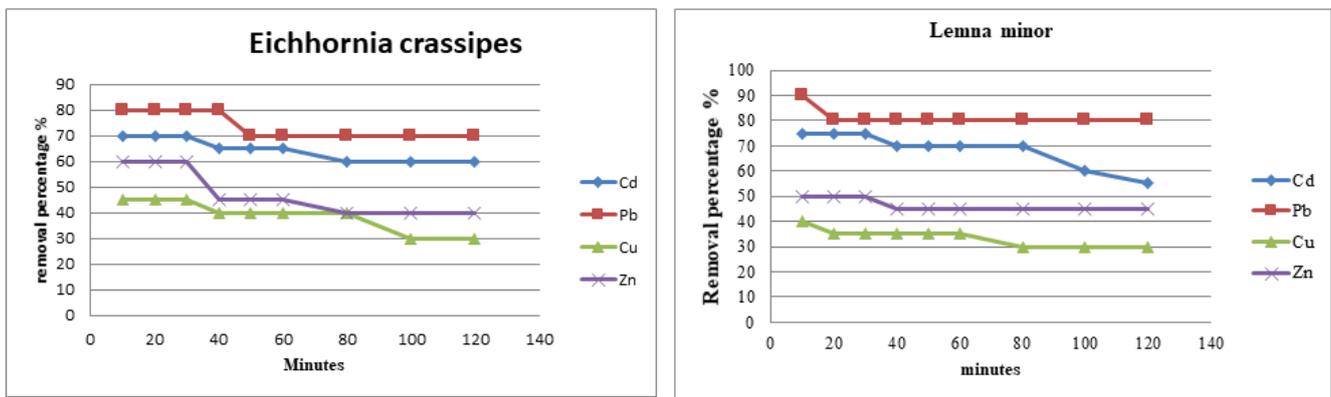


Figure 2. Influences of contact time on the metal removal % of both aquatic plant powders

When level was enhanced progressively, adsorption mechanism would fall-off until it achieved saturation. This might be due to enhance in the adsorbance with enhance in the various functional groups in the solution, where-by progress of the accessibility of particular sorption sites increased the adsorption degree. Enhance in adsorbance enhanced the amount of adsorbent

particles and decreased the SA available for more adsorbance to take place. Moreover, under the higher amount of the adsorbents, particle interface might induce de-sorption of the metals ions which loosely bind on the surfaces of adsorbents and contribute in decrease of adsorption capability (Kalak *et al.*, 2021).

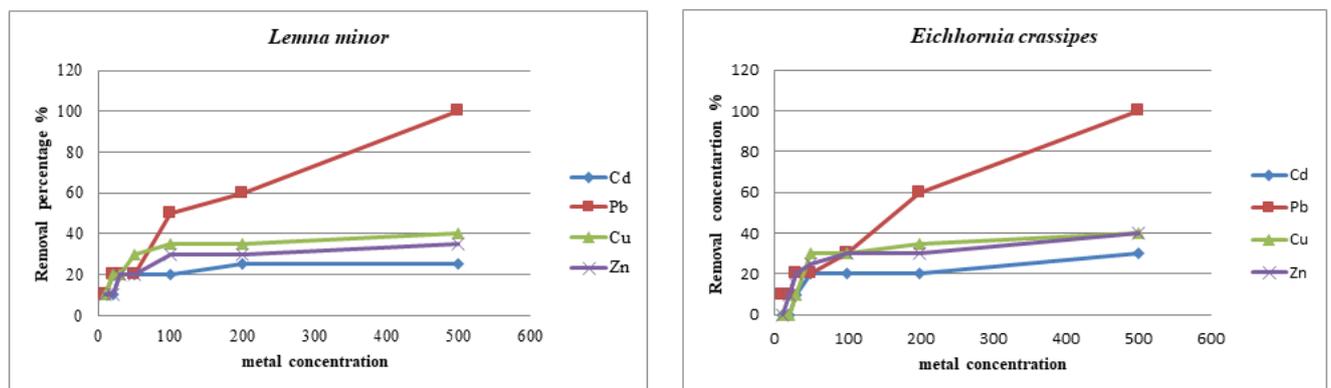


Figure 3. Influences of metal concentration on the metal removal % of both aquatic plant powders

**Isothermal adsorption and kinetics adsorption**

Fitting Langmuir model to elucidate adsorption ratio of Cu, Zn, Cd, and Pb through both plants obtained

wide R<sup>2</sup> difference (showing in Table, 2). Except for zinc Freundlich adsorption R<sup>2</sup> of *Lemna minor* and lead of *Eichhornia crassipes* were 0.7629 and 0.8495,

correspondingly. And all other  $R^2$  were higher than 0.9. First order formula in contrast has lower  $R^2$  (<0.5) and revealed a scanty correlation with experimental results (results not listed). Second order kinetics formula showed a well fit to data compared to first order equation (Table, 3). Langmuir is a monolayer diffusion adsorption model which is applied to present chemically-based adsorption among metal ions and adsorbents. The Freundlich and Langmuir formulas can be applied to describe of metals ions adsorption isotherm of the adsorbents. Assuming the quantity of adsorbent is slight and sorption sites on surfaces are similar, maximum adsorption capability should be determined via the levels of saturation on surfaces of adsorbents. Adsorption's energy remains perpetual. Freundlich is a non-linear model which can be applied for the present of isotherm adsorption (Shikuku *et al.*, 2021). Assuming adsorption is occurring on heterogeneous surfaces with enhance

in adsorption capability, energy of adsorption will logarithmically decrease to the adsorption equilibrium (Leon *et al.*, 2021). For both plants, isotherm adsorption equations of the Zn, Pb, Cd, and Cu are much better fit to Freundlich model which shows the adsorption mechanism is not mere monolayer adsorption. The studies of kinetics adsorption are typically applied to explain the adsorption ratio of adsorbents to adsorbates. Time for adsorbates to remain on the solid-liquid interaction is regulated via the adsorption rate. For both plants, isotherm adsorption equations of the Zn, Pb, Cd, and Cu were a much better to fit with Lagergren 2<sup>nd</sup> order formula, showing adsorption procedure charted Lagergren 2<sup>nd</sup> order process and adsorption ratio was controlled via chemical adsorption and dispersion of particles process played a key role in entire adsorption mechanism.

	Langmuir			Freundlich			<b>Table 2</b> <i>Freundlich and Langmuir isotherm constant</i>
	$q_{max}$ (mg/g)	b	$R^2$	$K_F$	$1/n$	$R^2$	
<b>Lemna minor</b>							
Zn	0.4322	0.0108	0.3982	0.1500	0.7386	0.7629	
Pb	1.5636	1.6357	0.8742	3.8265	0.5335	0.9921	
Cd	0.3401	0.0130	0.5473	1.1209	0.6968	0.9103	
Cu	0.6389	0.0348	0.8483	0.0997	0.8538	0.9673	
<b>Eichhornia crassipes</b>							
Zn	0.2283	0.0098	0.3201	0.5447	0.6764	0.9908	
Pb	0.7872	1.5133	0.9886	2.3672	0.3997	0.8495	
Cd	0.1962	0.0151	0.5963	3.4669	0.8725	0.9642	
Cu	0.3292	0.0176	0.5381	1.0614	0.6762	0.9247	

$q_{max}$  = maximum adsorption capacity -  $R^2$  = correlation coefficient -  $K_F$  = Freundlich coefficient - b = equilibrium constant -  $1/n$  = Freundlich index

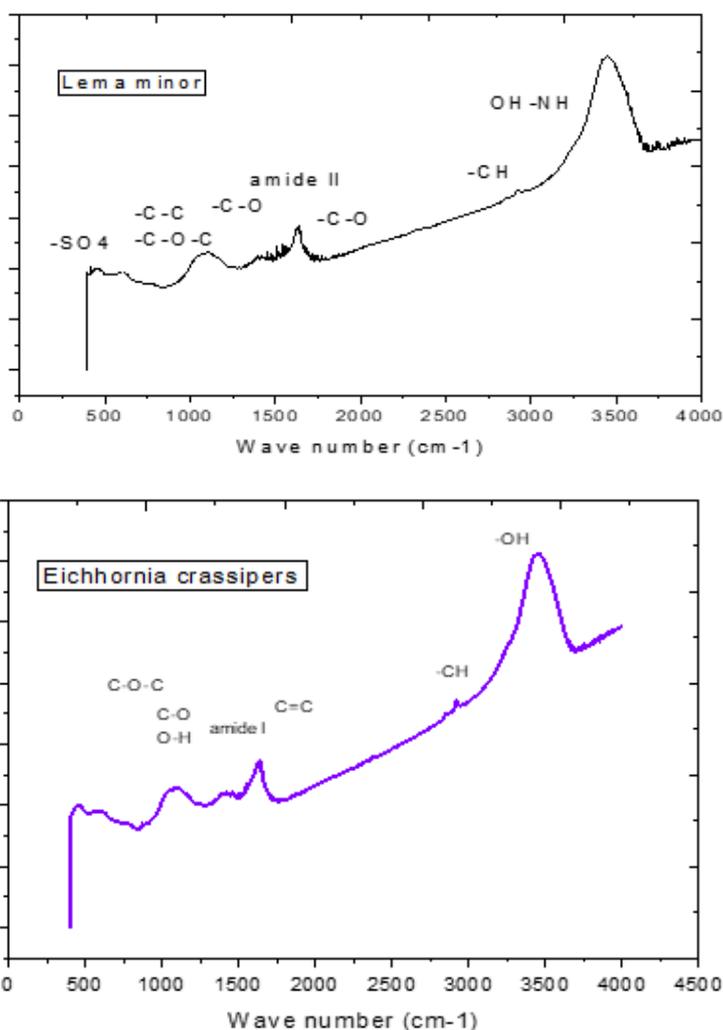
	$q_e$ (mg/g)	$K_2$	$R^2$	<b>Table 3</b> <i>Pseudo second-order kinetic constants</i>
	<b>Lemna minor</b>			
Zn	1.2256	6.3845	0.9998	
Pb	2.0662	0.7879	1.0000	
Cd	1.8132	1.7802	1.0000	
Cu	0.7976	1.3711	0.9997	
<b>Eichhornia crassipes</b>				
Zn	1.7249	4.1781	0.9997	
Pb	2.2834	0.3756	0.9990	
Cd	1.4477	2.8034	1.0000	
Cu	1.3813	0.5998	0.9996	

$q_e$  = adsorption equilibrium -  $R^2$  = correlation coefficient -  $k_2$  = pseudo-second order equilibrium constant

### FTIR Spectra (Characterization of powder)

Spectra of FTIR indicated various functional groups typically included amino, hydroxyl, carboxyl, carbonyl and double and triple bonds. Plants cells are built by lipid composition, protein, and polysaccharide, containing amino, hydroxyl, carbonyl and carboxyl which can bind with the metals ions (Giarikos *et al.*, 2021; Redha, 2020). FTIR analysis showed in Figure 4. Both plants samples had absorption peak at 3300 to 3400  $\text{cm}^{-1}$ , which characterized  $-\text{NH}$  and  $-\text{OH}$  stretching vibrations of the proteins, cell-wall substance and carbohydrate. Absorption peaks around 2900  $\text{cm}^{-1}$  could be  $-\text{CH}$  irregular stretching in  $\text{CH}_2$  and  $\text{CH}_3$ . Joint peaks at 1732  $\text{cm}^{-1}$  were carbonyl peak, which was a ketone and ester compounds observed in the cell-wall pectin and membrane lipid. Other absorption

peak around 1540  $\text{cm}^{-1}$  and 1650  $\text{cm}^{-1}$ , which signified amide I ( $\text{C}=\text{O}$  stretching) and amide II ( $\text{NH}$  and  $\text{CN}$  stretching), correspondingly. Absorption peak at 1400  $\text{cm}^{-1}$  to 1440  $\text{cm}^{-1}$  exhibited  $\text{C}-\text{O}$  stretching in carboxylate ions, which are noticed in the pectin. Absorption peak around 1440  $\text{cm}^{-1}$  to 1300  $\text{cm}^{-1}$  signified symmetrical bending of  $\text{CH}_2$  and  $\text{CH}_3$  in cellulose and protein (Zhuang *et al.*, 2020). Absorption peak around 1150  $\text{cm}^{-1}$  to 1100  $\text{cm}^{-1}$  might be  $\text{CO}$  stretching of ether, alcohol hydroxyl centred, which derived from major constituent of cell wall, for example, starch and cellulose. The acromion around 1060  $\text{cm}^{-1}$  was produced via  $-\text{OH}$  bending and  $\text{C}-\text{O}-\text{C}$  stretching, and  $\text{P}-\text{O}-\text{C}$  stretching vibration, also vibration of  $\text{SiO}_2$  might accelerate to production of acromion (Daghash *et al.*, 2020). There was a peak near 610  $\text{cm}^{-1}$ , signified  $\text{SO}_4^{2-}$ . This feeble peak showed there little quantity of  $\text{H}_2\text{SO}_4$  ester in biological substance.



**Figure 4**  
FTIR spectra

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## Conclusion

In both aquatic plants, the main constituents took part in metal sorption include protein, cell-wall pectin and carbohydrates functional groups namely amine, hydroxyl, carboxyl, and double-triple bonded hydrocarbon chains. Both aquatic plants indicate an efficient and economical material for heavy metals removal mechanism. Metals contamination does not impact the basic chemical constituents of both plants.

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