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Single and binary adsorption of Cd (II) and Zn (II) ions from aqueous solutions onto bottom ash

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Abstract−Bottom ash, a waste obtained from coal-burning power plant, was used as a low cost adsorbent for the removal of Cd (II) and Zn (II) ions from single and binary systems in batch experiments. The results of adsorption capacity showed that bottom ash could be considered as a potential adsorbent. The uptake of Zn (II) ion was greater than that of Cd (II) ion. For single adsorption, based on the correlation coefficient (R^2) values, both Langmuir and Freundlich isotherms suitably described the adsorption equilibrium data in the initial metal ion concentration range of 10-50 mg/L. The multicomponent isotherms, including the extended Langmuir and IAST-Freundlich isotherms, were used to predict the binary adsorption of Cd (II) and Zn (II) ions. Furthermore, the appropriate multicomponent isotherm was investigated by minimizing the average relative error (ARE) function. It should be confirmed that the extended Langmuir isotherm fitted the binary adsorption equilibrium data satisfactorily.

Keywords: Bottom Ash, Binary Adsorption, Multicomponent Isotherm, Cadmium (II), Zinc (II)

INTRODUCTION

Heavy metal contamination of wastewater is an important environmental problem due to their toxicity, persistence and non-biodegradability. Heavy metals such as lead, copper, nickel, chromium, cadmium, mercury and zinc are common pollutants widely found in industrial wastewaters from many industrial activities including electroplating, mining, battery manufacturing, chemical manufacturing, metallurgical processes, pigments, leather industries, fertilizer and pesticides industry etc. The presence of heavy metals in the environment can be harmful to human health because of accumulation via food chain [1-3]. Among heavy metals, Cd (II) and Zn (II) ions are high toxicity and relatively widespread in the environment. Cd (II) is a non-essential and non-biodegradable element which slowly accumulates in the human body. The toxic effects of Cd (II) may cause acute and chronic disorders such as Itai-Itai disease, renal degradation, emphysema, hypertension, and testicular atrophy [4,5]. Zn (II) is an essential element which participates in several metabolic processes and regulates many biochemical processes. But excessive ingestion of Zn (II) may lead to serious health problems like stomach cramps, skin irritations, vomiting, nausea and anaemia [6,7]. Therefore, it is necessary to remove Cd (II) and Zn (II) from wastewater prior to its discharge into the environment.

The conventional methods used for the removal of heavy metals from wastewater include chemical precipitation, coagulation, solvent extraction, reverse osmosis, ion exchange, filtration and adsorption. Among these methods, adsorption is the most attractive method because of its high efficiency, low cost and simplicity [8-11]. Activated carbon is regarded as a highly effective adsorbent for wastewater treatment but the high cost of activated carbon restricts its application especially in developing countries. Hence, low cost adsorbents, developed from industrial and agricultural waste materials such as clay, rice husk, wood, peat, sewage sludge, sawdust, fly ash, etc., have been used for the removal of heavy metals [12-15].

Bottom ash is a waste material obtained from electricity generation through combustion of coal. Its physical properties are grayish, porous, glassy, coarse and granular materials. Major chemical compositions of bottom ash are SiO_2, Al_2O_3, Fe_2O_3 and CaO. Bottom ash is used in several beneficial applications such as construction, structural fill, road base material and concrete aggregate [16-18]. Recently, some studies have been interested in the usage of bottom ash as an adsorbent for the removal of various heavy metals. Bottom ash has been successful used as a potential adsorbent due to its physical characteristics for example high porosity, small particle size and large surface area [19,20].

Most of the previous works on the adsorption of heavy metal ions by various adsorbents considered only single metal ion. Actually, industrial wastewater can contain several metal ions. For this reason, it is important to study the simultaneous adsorption of two or more metal ions and also to investigate the interactive effect of one metal ion on the sorption of other metal ions [21,22].

Preliminarily, the effect of important factors such as pH and contact time has been previously reported by Hatairat et al. 2014 [23], who studied the adsorption of Cd (II) and Zn (II) ions from aqueous solutions onto bottom ash. The experimental results showed that the optimum pH was 6 and the equilibrium contact time was

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achieved within 60 min for the removal of Cd (II) and Zn (II) ions. In the present work, the single and binary adsorption of Cd (II) and Zn (II) ions from aqueous solutions onto bottom ash was investigated. The single adsorption equilibrium data was analyzed using the Langmuir and Freundlich isotherms. The complex mathematical isotherms, namely, extended Langmuir and IAST-Freundlich isotherms were proposed for representation of experimental isotherm in the binary system. The parameters for these isotherms were obtained from the single adsorption isotherms based on adsorption equilibrium data. Furthermore, binary adsorption studies could be conducted to investigate the competitive effect between two metals ions in the adsorption process.

MATERIALS AND METHODS

1. Adsorbent

Bottom ash used as an adsorbent was collected from Mae Moh power plant in Thailand. It was dried in an oven at 110 °C for 24 h. After drying, the dried bottom ash was ground and sieved through 80 mesh sieve. The sample was kept in a desiccator and ready for use as adsorbent. The physical properties of bottom ash were carried out by standard methods [23]. Some of its average physical properties such as pH, density, porosity, particle size distribution and specific surface area are presented in Table 1. The particle size distribution analyzer (Mastersize 2000, Malvern instruments, UK) was used to determine particle size distribution. The specific surface area was measured using a Quantachrome autosorb automated with nitrogen gas (version 2.46). The chemical compositions of bottom ash as characterized by X-ray fluorescence (XRF) Spectrometer (Phillips MagiX PRO PW 2400) are given in Table 2.

Scanning electron microscopy (SEM) (JSM 6335F, Oxford instruments, UK) was used to examine the surface morphology of bottom ash. SEM photograph in Fig. 1 shows that bottom ash was irregular particles and consisting of hollow spheres. Furthermore, at higher size ranges, porous particles were detected. The results of physical and chemical properties demonstrated that bottom ash could be used as a potential adsorbent for the removal of Cd (II) and Zn (II) ions.

Table 1. Physical properties of bottom ash

Property	Value
pH	8.84
Density (g/cm^3)	2.43
Porosity (%)	38.05
Particle size distribution (μm)	24.55
Specific surface area (m^2/g)	5.14

Table 2. Chemical compositions of bottom ash

Compound	Weight percent (%)
SiO_2	38.33
CaO	18.01
Al_2O_3	16.98
Fe_2O_3	15.78
SO_3	5.62
MgO	2.46
K_2O	2.15
TiO_2	0.55
Mn_2O_3	0.12

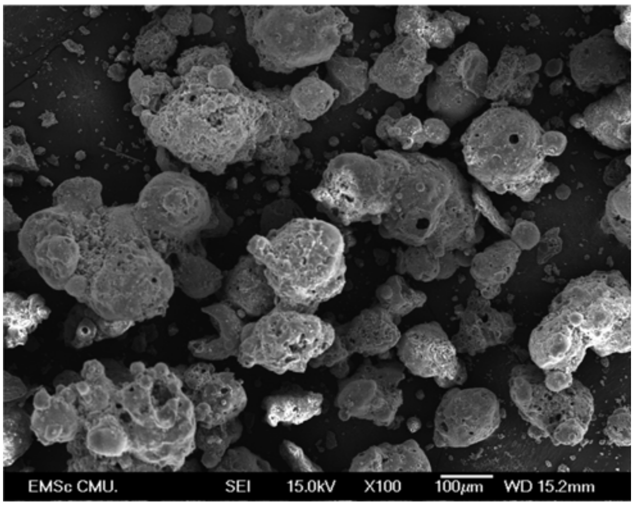

Fig. 1. SEM photograph of bottom ash.

2. Adsorbate

All reagents used were of analytical grade. Stock solutions containing 1,000 mg/L of Cd (II) and Zn (II) ions were prepared by dissolving of accurate amount of $Cd(NO_3)_2 \cdot 4H_2O$ and $Zn(NO_3)_2 \cdot 6H_2O$, respectively, in deionized water. In this study, different concentrations of Cd (II) and Zn (II) ions ranging from 10-50 mg/L were obtained by dilution of the stock solutions. The pH of the solutions was adjusted to the desired value by addition of 1 M HNO_3 solution. The solutions pH was measured by using pH meter (Model pHTestr20, Eutech instruments, Singapore).

3. Batch Adsorption Experiments

Batch experiments were conducted to investigate the adsorption of Cd (II) and Zn (II) ions onto bottom ash in single and binary systems. The amount of adsorbent (0.1 g) and 100 mL of Cd (II) and Zn (II) solutions were placed in 250 mL Erlenmeyer flask under shaking at 150 rpm and at controlled temperature of 30 °C using a water bath shaker (Model WSB-45, Daihan scientific, Korea).

To select the optimum conditions for the adsorption process, the effects of pH, contact time and temperature were investigated in the previous study [23]. The adsorption of Cd (II) and Zn (II) ions as a function of pH increased with increasing pH from 4 to 6. The removal of metal ions was poor at pH lower than 4 due to the competition between H^+ and metal ions on the adsorption sites. The effect of contact time for the adsorption of Cd (II) and Zn (II) ions onto bottom ash were investigated from 10 min to 300 min. It was observed that the adsorption of metal ions increased with increasing contact time and reached equilibrium in about 60 min. The effect of temperature on the adsorption of two metal ions was also studied at temperature between 10 °C and 40 °C. The adsorption of these metal ions increased with an increase in temperature, thereby indicating an endothermic adsorption process. However, the values of adsorbed amount at 30 °C and 40 °C are not much significantly different. Therefore, in this study, all experiments were at pH value of 6, contact time of 60 min and temperature at 30 °C.

In adsorption isotherm studies, the concentration was varied while other parameters were retained constant. For the single system, the initial concentration of Cd (II) and Zn (II) ions was varied from 10 to 50 mg/L. In the binary system, for each initial concentration of Cd (II) ion (10, 20, 30, 40 and 50 mg/L), the Zn (II) ion concentration was varied in the range of 10 to 50 mg/L. After reaching equilibrium, the supernatant was separated by filtration through the filter paper. The residual concentrations of Cd (II) and Zn (II) ions in single and binary systems were analyzed by atomic absorption spectrophotometer (AAS) (Model 3110, Perkin Elmer, USA) at 213.9 nm and 228.8 nm for Zn and Cd, respectively. The calibration curves of each ion in binary systems were investigated at various concentrations and the remaining concentrations of each ion were determined.

The amount of metal ion adsorbed per unit mass of the adsorbent was calculated according to the following equation:

$$q_e = \frac{(C_0 - C_e)V}{m} \tag{1}$$

where q_e is the amount of metal ion adsorbed at equilibrium (mg/g), C_0 is the initial metal ion concentration (mg/L), C_e is the equilibrium metal ion concentration (mg/L), V is the volume of solution (L) and m is the mass of the adsorbent (g).

RESULTS AND DISCUSSION

1. Single Adsorption Isotherms

Adsorption isotherm describes the relationship between the amount of metal ion adsorbed per unit mass of bottom ash and the equilibrium metal ion concentration. Fig. 2 shows the isotherms for the single adsorption of Cd (II) and Zn (II) ions onto bottom ash. The amount of metal ions adsorbed increased gradually with increasing metal ions concentration. Also, the adsorption capacity of bottom ash for Zn (II) ion was greater than that of Cd (II) ion. Although Cd (II) has larger radius (97 pm) than Zn (II) (74 pm), the hydrated form of these two ions is similar (430 pm for Cd (II) and 426 pm for Zn (II)) [24]. In general, the smaller the ionic radius, the easier it is for a metal ion to penetrate through the boundary layer and adsorb on the surface. The lower adsorption of Cd (II) might be because its ions do not compete effectively for variable charge surfaces. As a result, its adsorption is more restricted to permanent charge sites. The difference in the adsorption affinity in this study compared with other studies could be attributed to the unique features of each of the adsorbents as the production methods were vastly different [25].

Several isotherm equations have been applied to describe the equilibrium characteristics of adsorption. In this study, Langmuir and Freundlich isotherms were used to evaluate the adsorption equilibrium data in single system.

The Langmuir isotherm is expressed by the following equation [26,27]:

$$\frac{C_e}{q_e} = \frac{1}{bq_m} + \frac{C_e}{q_m} \tag{2}$$

where C_e is the equilibrium metal ion concentration (mg/L), q_e is the amount of metal ion adsorbed at equilibrium (mg/g), q_m is the maximum adsorption capacity (mg/g) and b is the Langmuir constant related to the energy of adsorption (L/mg). The plot of C_e/q_e versus C_e was used to calculate the values of q_m and b from slope and intercept, respectively (Fig. 3).

The Freundlich equation is given as [28,29]:

$$\log q_e = \log K_F + \frac{1}{n}\log C_e \tag{3}$$

where K_F and n are the Freundlich constants related to adsorption capacity and adsorption intensity, respectively. The Freundlich constants, n and K_F, can be obtained from slope and intercept of the plot log q_e versus log C_e, respectively (Fig. 4).

The Langmuir and Freundlich constants along with the correlation coefficients (R^2) are reported in Table 3. For the single adsorption of Cd (II) and Zn (II) ions, the values of R^2 approaching unity showed that both Langmuir and Freundlich isotherms were able

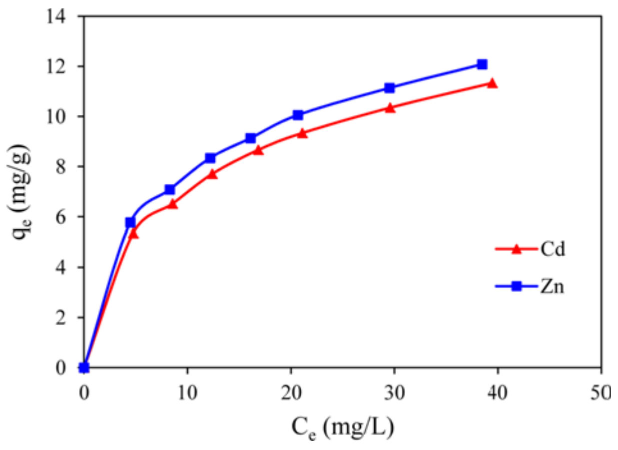

Fig. 2. Equilibrium isotherms for the single adsorption of Cd (II) and Zn (II) ions onto bottom ash (pH: 6, contact time: 60 min, temperature: 30 °C).

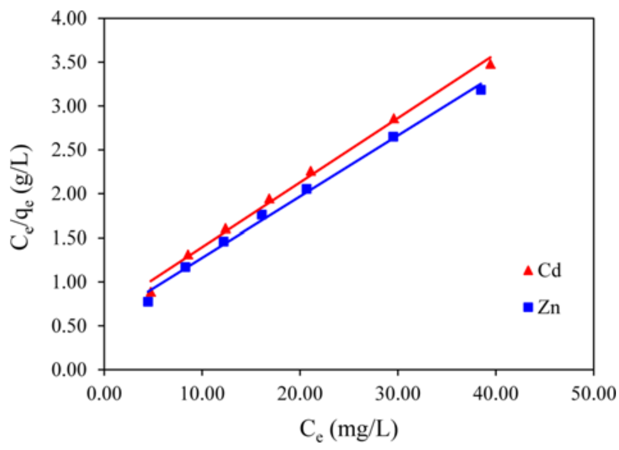

Fig. 3. Langmuir isotherms for the single adsorption of Cd (II) and Zn (II) ions onto bottom ash (pH: 6, contact time: 60 min, temperature: 30 °C).

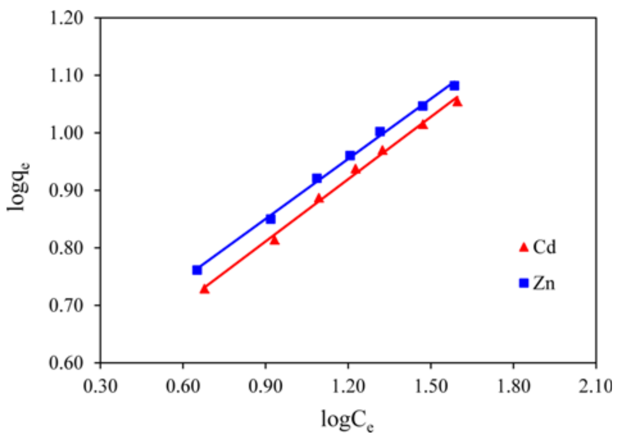

Fig. 4. Freundlich isotherms for the single adsorption of Cd (II) and Zn (II) ions onto bottom ash (pH: 6, contact time: 60 min, temperature: 30 °C).

Table 3. Langmuir and Freundlich constants for the single adsorption of Cd (II) and Zn (II) ions onto bottom ash

	Langmuir isotherm			Freundlich isotherm		
	q_m (mg/g)	b (L/mg)	R^2	K_F	n	R^2
Cd (II)	13.7	0.111	0.994	3.06	2.78	0.996
Zn (II)	14.5	0.120	0.994	3.44	2.87	0.997

to correlate adsorption equilibrium data satisfactorily, since the adsorption isotherms increased slowly with increasing concentration. This describes quantitatively the formation of a monolayer adsorbate on the heterogeneous surface of the adsorbent. Various mechanisms have been forwarded to explain specific sorption of metals by soil colloids. These include sorption of the metal in a hydrolyzed form as pH increases, and metal complexation with surface OH groups. In this study the maximum pH obtained (pH 6) was well below the metal hydrolysis constants of Zn (II) and Cd (II) ions. Only a very small proportion of the added metals would have been in the hydrolyzed form. The possible explanation may be that the metals may have undergone specific interaction with surface OH-(i.e., OH--M^{2+}) groups as suggested by Kinniburgh et al. [30]. The maximum adsorption capacity (q_m) obtained from Langmuir isotherm was 13.7 mg/g for Cd (II) ion and 14.5 mg/g for Zn (II) ion. The q_m values confirmed that the adsorption capacity of bottom ash for Zn (II) ion was higher than that of Cd (II) ion. Normally, the Freundlich constant, n values, which greater than unity can be indicating that the adsorption process is appropriate. From the results, the values of n for the single adsorption of Cd (II) and Zn (II) ions onto bottom ash were found to be greater than unity, characterizing as a favorable adsorption. 1/n is a heterogeneity parameter: the smaller 1/n, the greater the expected heterogeneity.

The maximum adsorption capacities values of several low cost adsorbents for adsorption of Cd (II) and Zn (II) ions have been reported in the literature. A comparison between these values and the values obtained in this study is listed in Table 4. As can be seen from the q_m values, bottom ash shows a reasonable capacity and can be a good adsorbent for Cd (II) and Zn (II) ions adsorption.

Table 4. The maximum adsorption capacities of various low cost adsorbent for adsorption of Cd (II) and Zn (II) ions

Metal ion	Adsorbent	q_m (mg/g)	Reference
Cd (II)	Maize stalks	18.05	[1]
	Rice husk ash	3.04	[21]
	Bagasse fly ash	6.19	[22]
	Clay mineral beidellite	42.01	[28]
	Sugarcane bagasse	6.79	[31]
	Peat	5.16	[32]
	Lignin (black liquor)	25.40	[33]
	Bottom ash	13.70	This study
Zn (II)	Maize stalks	30.30	[1]
	Fly ash	5.82	[14]
	Blast furnace slag	3.25	[14]
	Rice husk ash	5.88	[21]
	Wheat straw	11.40	[34]
	Peat	11.70	[35]
	Lignin (black liquor)	11.24	[33]
	Bottom ash	14.50	This study

2. Binary Adsorption Isotherms

The simultaneous adsorption of Cd (II) and Zn (II) ions from binary aqueous solutions by bottom ash was examined at pH 6 and constant temperature of 30 °C. The adsorption isotherms of Cd (II) ion at varying concentrations of Zn (II) ion and the adsorption isotherms of Zn (II) ion at varying concentrations of Cd (II) ion are presented in Fig. 5 and Fig. 6, respectively. As can be seen in Fig. 5, the equilibrium uptake of Cd (II) ion decreased continuously with increasing the concentration of Zn (II) ion. For example, at 50 mg/L initial concentration of Cd (II) ion, the amount of Cd (II) ion adsorbed was diminished from 11.34 to 6.06 mg/g when the Zn (II) ion concentration was raised from 0 to 50 mg/L.

Also, the adsorption isotherms of Zn (II) ion (Fig. 6) show that the equilibrium uptake of Zn (II) ion decreased with an increase in the Cd (II) ion concentration. For example, at initial concentration of Zn (II) ion of 50 mg/L, the amount of Zn (II) ion adsorbed reduced from 12.08 to 7.72 mg/g with increasing the Cd (II) ion

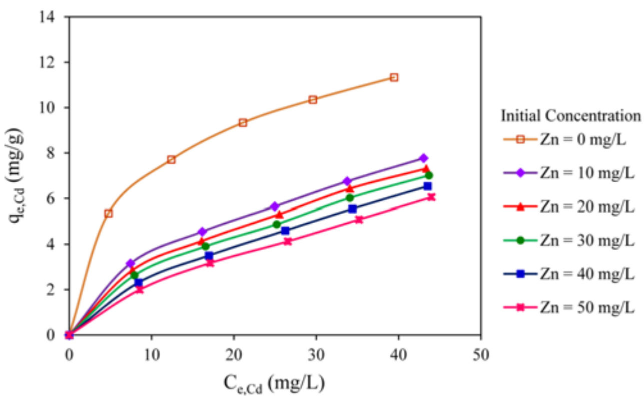

Fig. 5. Adsorption isotherms of Cd (II) ion at various initial concentrations of Zn (II) ion (pH: 6, contact time: 60 min, temperature: 30 °C).

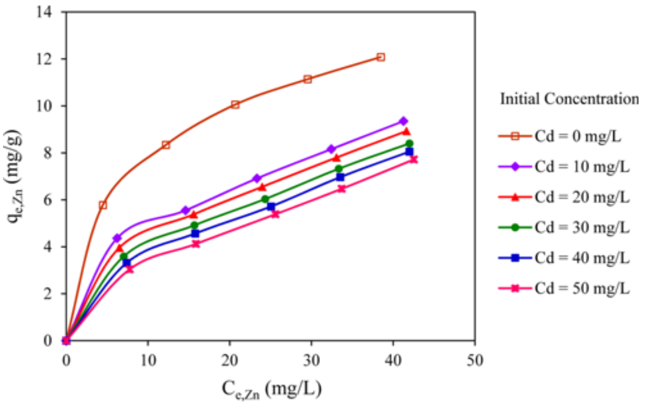

Fig. 6. Adsorption isotherms of Zn (II) ion at various initial concentrations of Cd (II) ion (pH: 6, contact time: 60 min, temperature: 30 °C).

concentration from 0 to 50 mg/L. The results clearly revealed that the adsorption capacity of each ion was affected by the presence of the other due to the competitive adsorption between them. Note that the adsorption isotherms of both ions in the binary system were nearly straight lines. The adsorbed amount increased slowly with increasing concentration due to the small value of the Langmuir constant b and, therefore, it was directly proportional to the equilibrium concentration.

When several metal ions appear in aqueous solutions, there is interference and competition among the different metal ions for adsorption sites. For this reason, the adsorption isotherms for single system are inapplicable. A more complex mathematical isotherm is used to describe a multicomponent system. In this work, the adsorption equilibrium data of Cd (II) and Zn (II) ions from binary system were fitted to the multicomponent isotherms, i.e., the extended Langmuir and IAST-Freundlich isotherms. Other multicomponent isotherms were rejected due to the increase of modifying parameters involved in its use. The mathematic expression of the extended Langmuir and IAST-Freundlich isotherms is presented below.

Extended Langmuir isotherm for multicomponent system is expressed as follows [36-39]:

$$q_{e,i}=\frac{q_{m,i}b_iC_{e,i}}{1+\sum_{j=1}^{N}b_jC_{e,j}} \tag{4}$$

where $q_{e,i}$ is the equilibrium amount of component i adsorbed in the multicomponent system, $C_{e,i}$ is the equilibrium concentration of component i, $C_{e,j}$ (j=1, 2, …, N; N is the number of the components) is the equilibrium concentration of each component in the system, b_i and b_j are the Langmuir constants for component i and j, respectively and $q_{m,i}$ is the maximum adsorption capacity of component i. The parameters $q_{m,i}$, b_i and b_j are obtained from the single adsorption isotherm.

For binary system, the extended Langmuir isotherm can be written as:

$$q_{e,1}=\frac{q_{m,1}b_1C_{e,1}}{1+b_1C_{e,1}+b_2C_{e,2}} \tag{5}$$

$$q_{e,2}=\frac{q_{m,2}b_2C_{e,2}}{1+b_1C_{e,1}+b_2C_{e,2}} \tag{6}$$

where the subscripts 1 and 2 represent Cd (II) and Zn (II) ions, respectively in the binary system used in this study.

The ideal adsorbed solution theory (IAST)-Freundlich isotherm is a simplification of the multicomponent Freundlich isotherm. For component i in N-component system, the IAST-Freundlich isotherm can be described by the following expression [9,13,34,40]:

$$C_{e,1}=\frac{q_{e,i}}{\sum_{j=1}^{N}q_{e,j}}\left(\frac{\sum_{j=1}^{N}n_jq_{e,j}}{n_iK_{F,i}}\right)^{n_i} \tag{7}$$

In the case of binary system, Eq. (7) can be written separately for each component as follows:

$$C_{e,1}=\frac{q_{e,1}}{q_{e,1}+q_{e,2}}\left(\frac{n_1q_{e,1}+n_2q_{e,2}}{n_1K_{F,1}}\right)^{n_1} \tag{8}$$

$$C_{e,2}=\frac{q_{e,2}}{q_{e,1}+q_{e,2}}\left(\frac{n_1q_{e,1}+n_2q_{e,2}}{n_2K_{F,2}}\right)^{n_2} \tag{9}$$

where $C_{e,1}$ and $C_{e,2}$ are the equilibrium concentrations of Cd (II) and Zn (II) ions, respectively, $q_{e,1}$ and $q_{e,2}$ are the equilibrium amount of metal ions adsorbed in the multicomponent system, while $K_{F,1}$, $K_{F,2}$, n_1 and n_2 are the Freundlich constants for the Cd (II) and Zn (II) ions in the single system, respectively.

The adequacy and accuracy of the extended Langmuir and IAST-Freundlich isotherms for adsorption equilibrium data were estimated using the average relative error (ARE) [34,41,42]. ARE can be represented by the following equation:

$$ARE(\%)=100\cdot\frac{1}{N}\sum_{i=1}^{N}\left|\frac{X_{exp}-X_{pred}}{X_{exp}}\right|_i \tag{10}$$

where X_{exp} and X_{pred} are the experimental and model predicted values, respectively, and N is the number of the experimental data points.

The ARE values for the extended Langmuir and IAST-Freundlich isotherms are given in Table 5. The suitable isotherm for binary adsorption would be selected by the minimum ARE value of both isotherms used. As shown in Table 5, ARE of the extended Langmuir isotherm was 16.28% for Cd (II) ion and 12.23% for Zn (II) ion, whereas ARE of IAST-Freundlich isotherm was 31.74% for Cd (II) ion and 31.59% for Zn (II) ion. Based upon the values of ARE, the extended Langmuir isotherm fitted adsorption equilibrium data better than the IAST-Freundlich isotherm for the binary adsorption of Cd (II) and Zn (II) ions onto bottom ash.

The comparisons of the model predicted and experimental val-

Table 5. ARE between the experimental and model predicted values for the extended Langmuir and IAST-Freundlich isotherms

Model	ARE (%)	
	Cd (II)	Zn (II)
The extended Langmuir isotherm	16.28	12.23
The IAST-Freundlich isotherm	31.74	31.59

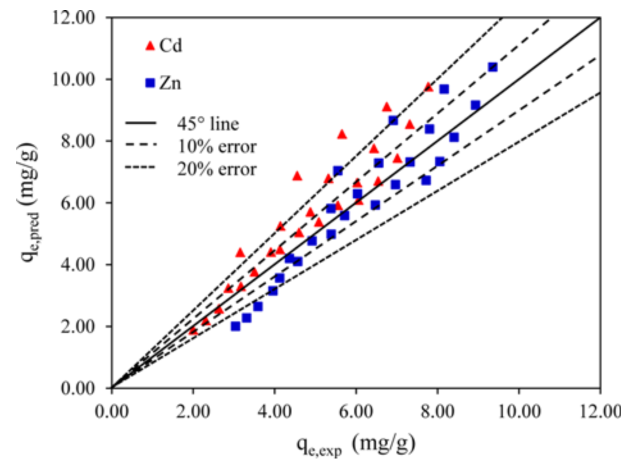

Fig. 7. Comparison of q_e values predicted from extended Langmuir isotherm and experimental values of Cd (II) and Zn (II) ions in binary system.

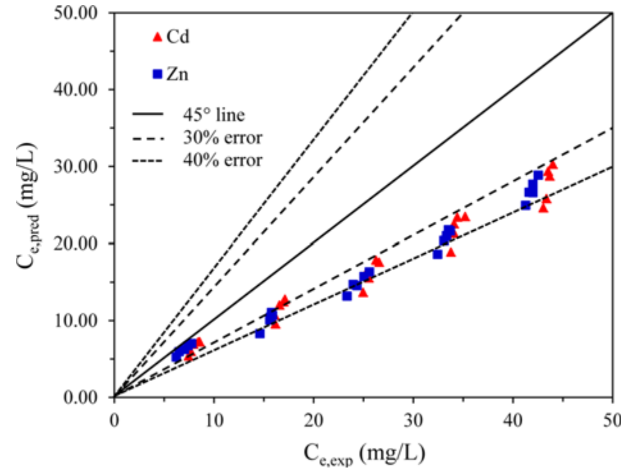

Fig. 8. Comparison of C_e values predicted from IAST-Freundlich isotherm and experimental values of Cd (II) and Zn (II) ions in binary system.

ues of Cd (II) and Zn (II) ions in binary system are shown in Figs. 7 and 8. Basically, if most of the data points are distributed around the 45° line, this indicates that the multicomponent isotherm represents well the adsorption equilibrium data for binary system.

For the extended Langmuir isotherm, the results plotted in Fig. 7 clearly showed that most of the data points distributed around the 45° line. The uptake of Cd (II) ion was over-predicted, while the uptake of Zn (II) ion was both over-predicted and under-predicted by the extended Langmuir isotherm. In the case of the IAST-Freundlich isotherm, all the data points were far away from the 45° line, as shown in Fig. 8. The uptake of both Cd (II) and Zn (II) ions was under-predicted. Therefore, these results confirmed that the extended Langmuir isotherm was adequately used to predict the adsorption of Cd (II) and Zn (II) ions onto bottom ash in binary system.

CONCLUSIONS

Bottom ash was considered as an effective adsorbent for the removal of Cd (II) and Zn (II) ions from aqueous solutions. The capacity of bottom ash for adsorbing Zn (II) ion was greater than that of Cd (II) ion. In single system, both Langmuir and Freundlich isotherms showed good fit with the adsorption equilibrium data based on their R^2 values. For binary system, the adsorption of Cd (II) and Zn (II) ions onto bottom ash was predicted by the extended Langmuir and IAST-Freundlich isotherms. A comparison of ARE values for both isotherms indicated that the extended Langmuir isotherm fitted the adsorption equilibrium data of Cd (II) and Zn (II) ions satisfactorily and adequately. Binary adsorption studies showed that the presence of Cd (II) and Zn (II) ions in aqueous solutions created competition between them in the adsorption process. These results confirmed that bottom ash can be used for the individual and simultaneous adsorption of Cd (II) and Zn (II) ions from aqueous solutions. Finally, bottom ash is a low cost adsorbent so it will be useful for the economical treatment of wastewater contaminated with heavy metals.

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