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*Original Article*

# Phytoremediation of synthetic wastewater by adsorption of lead and zinc onto *Alpinia galanga* Willd

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## Abstract

Adsorption of Pb(II) and Zn(II) from aqueous solution onto a phytosorbent mixture of galangal (*Alpinia galanga* Willd) leaves and pseudostem has been investigated in a batch system. Removal efficiency was optimized with respect to pH, contact time, initial Pb(II) and Zn(II) concentration and phytosorbent dosage. Galangal exhibited a higher adsorption efficiency for lead (95.2%) than for zinc (66.9%). Both lead and zinc equilibrium sorption data were better represented by the Freundlich isotherm than the Langmuir isotherm and in each case followed second order kinetics.

**Keywords:** galangal, heavy metal, adsorption isotherm, adsorption kinetics, phytosorbent

## 1. Introduction

Heavy metals are amongst the most dangerous pollutants of groundwater, (Briggs, 2003; Samuding *et al.*, 2009; Momodu and Anyakora, 2010) industrial wastewater (Ziemacki *et al.*, 1989; Sa'idi, 2010; Kapungwe, 2011) and marine environments (Krishna *et al.*, 2003; Boran and Altinok, 2010; Nubi *et al.*, 2011). Because metal ions are not degradable, they have a tendency to accumulate in water streams, endangering human health. Many treatment methods for the removal of heavy metals have been proposed, including ion exchange, adsorption onto activated carbon, chemical precipitation and membrane processes (Sud *et al*, 2008; Demirbas, 2008). However, removal of metals from aqueous solution by plants is particularly interesting as plants are environmentally safe and socially acceptable (Ngah and Hanafiah, 2008; Miretzky and Cirelli, 2010). Various forms of horticultural and agricultural waste have been investigated as potential adsorbents for heavy metals, including bael and cypress leaves, wheat bran, orange peel, and both banana pith and peel. However, the efficiency of metal adsorption by plant matter is affected not only by the quantity and dosage of adsorbent, but also by pH, contact time, temperature and metal concentration (Brown *et al.*, 2000; Demirbas, 2008; Ngah and Hanafiah, 2008; Sud *et al.*, 2008; Farooq *et al.*, 2010). The optimum parameters for metal adsorption vary widely from one phytosorbent to another, so the effect of each of these factors must be determined when a plant by-product is investigated as a metal adsorbent.

Galangal, (*Alpinia galanga* Willd) is a rhizomatous herb in the ginger family (Zingiberaceae) which is distributed throughout Thailand and Southeast Asia. The plant has a distinctive aroma, resulting from the presence of essential oils and phenolic compounds (Jirovetz *et al.*, 2003; Chudiwal *et al.*, 2010; Wungsintaweekul *et al.*, 2010). However, only the rhizomes are widely used as a food additive and in herbal medicine. As herbal remedy, galangal rhizomes act as a carminative, and also treat rheumatism, spleen pain, bronchitis, diabetes mellitus and loss of appetite (Chan *et al*, 2008). However, the leaves and pseudostem are usually discarded as waste products of the food and herbalism industries. This report shows our research into the phytosorption of lead and

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zinc from aqueous solution using the leaves and pseudostems of *A. galanga*, an application which could raise the value of the galangal plant. The optimum conditions of pH, contact time, initial metal concentration and dosage of phytosorbent are also shown.

## 2. Materials and Methods

### 2.1 Preparation of phytosorbent materials

The leaves and pseudostem of galangal were collected from Pattani Province, Thailand. They were used in the form of a phytosorbent powder for the removal of Pb(II) and Zn(II) from aqueous solution. The galangal powder was prepared by washing the leaves and pseudostems with distilled water, and then oven drying them at 70°C for 12 hrs. The dried galangal was then ground and sieved through a 40 mesh filter to give particles of $\leq$ 0.42 mm, and stored in an air tight plastic container for further experiments.

### 2.2 Preparation of synthetic wastewater

Single metal solutions of $Pb(NO_3)_2$ and $Zn(NO_3)_2$ were used to prepare the synthetic wastewater test solutions. They were prepared by diluting with deionized water to the desired concentrations. The acidity of each solution was adjusted to give a range of values between pH 2 and 7 by addition of either 1 M hydrochloric acid or 1 M sodium hydroxide.

### 2.3 Batch phytosorption studies

All experiments were performed in 100 mL Erlenmeyer flasks at room temperature and the flasks were agitated on a mechanical shaker at 125 rpm throughout the study. Initial testing was carried out with 50 mL samples adjusted to pH 2, at an initial metal concentration of 20 mg/mL. Powdered galangal phytosorbent (0.5 g) was then added and the suspension was shaken for 60 min. After shaking, the suspension was centrifuged and filtered under vacuum. The residual metal ion in the filtrate was analyzed using atomic adsorption spectroscopy (Perkin Elmer Analyst 100).

Variation sets were established to experimentally evaluate the optimum pH, contact time, metal ion concentration and dosage. Each experiment was conducted according to a separate batch system using the parameters described in Table 1. All experiments were carried out in triplicate. Only average values were reported, as the maximum error did not exceed 5%.

### 2.4 Metal adsorption

The metal ion uptake capacity of the galangal phytosorbent ($q_t$, mg/g) was calculated from Equation 1.

$$q_t = (C_i - C_t)V/W \qquad (1)$$

where $C_i$ and $C_t$ (mg/mL) are the initial and final metal ion concentrations in the filtrate, respectively. V (mL) is the volume of the solution and W (g) is the mass of galangal phytosorbent used.

The percentage uptake (% Sorption) was determined from Equation 2.

$$\text{Sorption}(\%) = [(C_i - C_t)/C_i]*100 \qquad (2)$$

## 3. Results and Discussion

### 3.1 Effect of pH

The pH optimization study was carried out using 0.5 g phytosorbent/50 mL synthetic wastewater to adsorb Pb(II) and Zn(II) at an initial concentration of 20 mg/L within a pH range of 2-7 for 60 min. Pb(II) and Zn(II) adsorption improved as the pH was raised from 2 to 4. However, above pH 4 the adsorption only increased marginally and reached an optimum value at pH 6 for Pb(II) and pH 7 for Zn(II) (Table 2). Similar variations in adsorption behavior with solution pH have already been reported for phytosorbents (Quek *et al.*, 1998; Dhakal *et al.*, 2005; Homagai *et al.*, 2009 and Chakravarty *et al.*, 2010). Effective adsorption of Pb(II) and Zn(II) occurred in the pH range 4-7. At low pH, it is likely that an association between the adsorbent surface and hydronium ions occurs, giving rise to a repulsive force between the

Table 1. Parameters used in the adsorption assessment with galangal.

| Effect | | pH | Contact time (min) | Dose (g) | Initial concentration (mg/L) |
|---|---|---|---|---|---|
| pH | Pb | 2,3,4,5,6,7 | 60 | 0.5 | 20 |
| | Zn | 2,4,5,6,7 | 60 | 0.5 | 20 |
| Time | Pb | 6 | 30,60,90,120,150,180 | 0.5 | 20 |
| | Zn | 7 | 30,60,90,120,150,180 | 0.5 | 20 |
| Dose | Pb | 6 | 150 | 0.5,1.0,1.5,2.0 | 20 |
| | Zn | 7 | 60 | 0.5,1.0,1.5,2.0 | 20 |
| Concentration | Pb | 6 | 150 | 0.5 | 10,20,30,40,50 |
| | Zn | 7 | 60 | 0.5 | 10,20,30,40,50 |

Table 2. Effect of pH on removal of lead and zinc ions by galangal.

| pH | Removal of Pb(II) | | Removal of Zn(II) | |
|---|---|---|---|---|
| | $q_t$ (mg/g) | sorption (%) | $q_t$ (mg/g) | sorption (%) |
| 2.0 | 1.48±0.01 | 74.11±0.75 | nt | Nt |
| 4.0 | 1.85±0.03 | 92.52±1.87 | 1.20±0.00 | 60.22±0.21 |
| 5.0 | 1.85±0.02 | 92.38±1.10 | 1.23±0.00 | 61.41±0.79 |
| 6.0 | 1.90±0.07 | 95.09±3.56 | 1.28±0.02 | 64.73±1.87 |
| 7.0 | 1.91±0.01 | 95.24±0.21 | 1.34±0.04 | 66.93±2.75 |

nt. = not tested

adsorbent and the metal cation. As the pH increases the concentration of hydronium ions in solution decreases, and the binding between the active sites of the adsorbent and metal ions is enhanced. Additionally, acidic functional groups in phytosorbents begin to dissociate in the pH range 4-7, giving rise to multiple negative charges on the adsorbent surface. The resulting electrostatic attraction / complexation between the surface and metal ions in solution may play a significant role in metal binding (Feng and Aldrich, 2004; Hashem, 2007). Several previous reports have revealed variations in the optimal pH for removal of lead and zinc ions by phytosorbents. For example, the maximum adsorption of lead occurred at pH 5.1 with bael leaves, (Chakravarty *et al*, 2010) at pH 6.7 for cypress leaves, (Salim *et al.,* 1994) and at pH 5 for banana peels (Anwar *et al.*, 2010). For zinc, the best pH for adsorption by carrot residues was 5, (Nasernejad *et al.*, 2005) by bael leaves 6 (Chakravarty *et al.*, 2010), and by wheat bran 6.5 (Dupont *et al.*, 2005). For this study, after the values of pH 6 and 7 were established as optimal for adsorption of Pb(II) and Zn(II), these acidity levels were maintained for subsequent experiments.

### 3.2 Exposure time and adsorption kinetics

The effect of varying the exposure time of Pb(II) and Zn(II) to *Alpinia galanga* Willd was studied using 0.5 g galangal powder/50 mL synthetic wastewater. A metal concentration of 20 mg/L was used at pH 6 for lead, and at pH 7 for zinc adsorption over a period of 30 to 180 min. The experimental results are depicted graphically in Figure 1, and indicate that the adsorption rate for lead increased rapidly over the first 30 minutes and then leveled off, slowly reaching a maximum adsorption after 150 min. However, while the adsorption rate for zinc also increased rapidly over the first 30 min, maximum efficiency ($q_e$) for Zn(II) phytosorption was achieved after just 60 min. From 60 min until 150 min, a gradual decrease in the efficiency of adsorption was observed, until after 150 minutes little change was observed until the final data point, representing a slightly lower adsorption efficiency at 180 min. Most phytosorption studies have indicated that initial adsorption occurs rapidly, but is followed by a slow increase in the rate of adsorption, (Low *et al.*, 1995; Sun and Shi, 1998; Ünlü and Ersoz, 2006; Karthika *et al.*, 2010) as we observed for Pb(II). The adsorption rate not only depends on the structure and chemical properties of the adsorbent, but also on the coordination number and initial concentration of the metal, the solution pH, the adsorbent dose and the temperature. The complexity of the parameters influencing metal adsorption give rise to wide variations in optimal contact time (Nasernejad *et al*, 2005; Li *et al.*, 2008; Deviprasad and Abdullah, 2009; Karthika *et al*, 2010) such as those observed for Pb(II) and Zn(II).

Lagergren's pseudo-first order equation was used to analyze the phytosorption kinetics of galangal. The first order equation is given in Equation 3 and the linearized model is shown in Equation 4.

$$\frac{dq_t}{dt} = K_1(q_e\text{-}q_t) \tag{3}$$

$$\log(q_e\text{-}q_t) = \log q_e - K_1.\ t/2.303 \tag{4}$$

where $q_e$ (mg/g) represents the mass of metal adsorbed onto the galangal powder at equilibrium, while $q_t$ corresponds to the amount of metal adsorbed at time t (min). $K_1$ is the first order rate constant. However, the data failed to fit this model as $R^2$ was low and instead a pseudo-second-order equation was used to model the data.

The pseudo-second-order equation, Equation 5 which was applied to the adsorption kinetics, can also be expressed in the form of a linearized Equation 6.

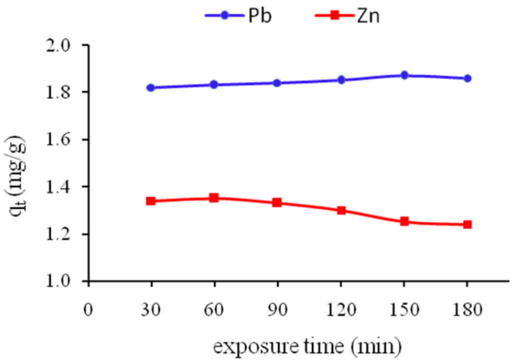

Figure 1. Effect of exposure time on Pb(II) and Zn(II) adsorption by galangal (Galangal dose = 0.5 mg, initial concentration of metals = 20 mg/g at pH 6 for Pb and pH 7 for Zn).

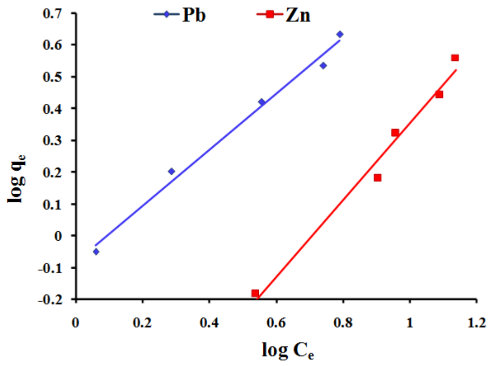

Figure 2. Pseudo-second-order kinetic plots of Pb(II) and Zn(II) adsorption by galangal.

$$\frac{dq_t}{dt} = K_2(q_e - q_t)^2 \qquad (5)$$

$$t/q_t = 1/K_2q_e^2 + t/q_e \qquad (6)$$

In Equation 6, $K_2$ represents the constant for the pseudo-second-order kinetic model. It was found that the adsorption of Pb(II) and Zn(II) could be effectively described as a pseudo-second-order process using Equation 6. The plots of $t/q_t$ versus t exhibited excellent linearity with high regression coefficients ($R^2$ = 0.9999 and 0.9997) as shown in Figure 2. Additionally, the calculated $q_e$ values obtained from the slope agreed well with the experimental uptake values, $q_e$ (exp) as depicted in Table 3. These results proved that the rate of adsorption was proportional to the square of the number of free binding sites $(q_e-q_t)^2$. Correspondingly, each divalent metal ion (Pb or Zn) is likely to be associated with two monovalent active sites on the galangal phytosorbent (see Schiewer and Patil, 2008). Previous reports have indicated that second order models are effective in a variety of cases for describing the binding of divalent metal ions to adsorbents. Examples include the adsorption of Ni(II) by *Cassia fistula* leaves (Hanif *et al*, 2007), of Fe(II) by tamarind bark, and also by potato peel (Prasad and Abdullah, 2009) and the adsorption of Pb(II) by bael leaves (Chakravarty *et al.*, 2010).

### 3.3 Dose-response experiments

Varying the dosage of galangal powder within the range 0.5-2.0 g indicated that metal absorption reached a maximum of 0.5 g galangal for 50 mL synthetic wastewater, a dosage at which sufficient binding sites were available to adsorb the maximum metal ions for both Pb(II) and Zn(II). However, as the mass of galangal adsorbent increased above 0.5 g, a steady decrease in the mass of metal adsorbed ($q_e$) was recorded (Figure 3). These results agree with those of Quek *et al.* (1998); Hanif *et al.* (2007), Kazemipour *et al.* (2008), and Anwar *et al.* (2010), which indicate that at high adsorbent concentration partial aggregation of the adsorbent occurs, decreasing the availability of active binding sites. Accordingly, the optimum dosage of powdered galangal leaves (0.5 g) was not exceeded in subsequent studies.

### 3.4 Effect of initial metal ion concentration on adsorption isotherms

Adsorption of lead and zinc at various initial concentrations, ranging from 10 to 50 ppm was studied under the optimum conditions of phytosorbent dose, pH and contact time. It was found that the efficiency of adsorption improved with increasing initial metal ion concentration, as shown in Figure 4. This relationship has also been reported by other researchers (Li *et al.*, 2008; Javaid *et al.*, 2011; Alfa *et al.*, 2012), who have observed that at high concentrations of the metal ion, affinity towards the active sites of the adsorbent increases.

Langmuir and Freundlich isotherms were used to model the adsorption isotherms of Pb(II) and Zn(II). The linear Langmuir equation is represented in Equation 7:

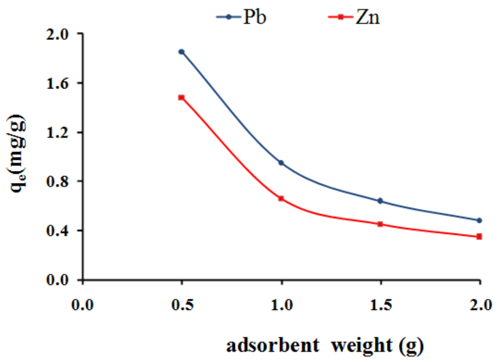

Figure 3. Effect of biosorbent dose on the adsorption of Pb(II) and Zn(II) by *A. galanga*.

Table 3. Pseudo-second-order model parameters, calculated mass adsorbed $q_e$ (cal) and experimental mass adsorbed, $q_e$(exp) for removal of Pb(II) and Zn(II) by galangal.

| Metal | $q_e$ (exp)(mg $g^{-1}$) | Pseudo-second-order parameters | | |
|---|---|---|---|---|
| | | $K_s$ (g/ mg min) | $q_e$ (calc)(mg/g) | $R^2$ |
| Pb | 1.97 | 1.35 | 1.99 | 0.9999 |
| Zn | 1.34 | 0.5 | 1.32 | 0.9971 |

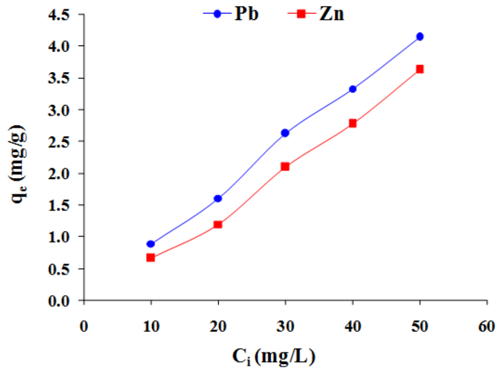

Figure 4. Effect of initial concentration of Pb(II) and Zn(II) on the adsorption capacities of *A. galanga* (dose = 0.5 mg at pH 6, 150 min for Pb and pH 7, 60 min for Zn).

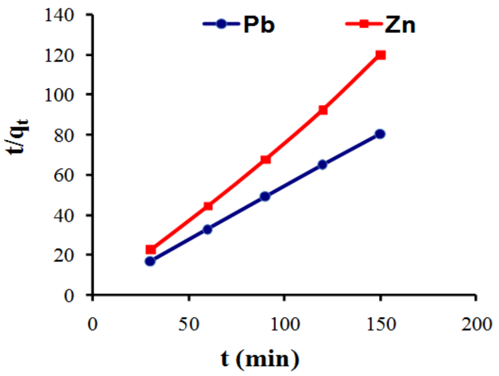

Figure 5. Freundlich isotherm for Pb and Zn (initial metal concentration 10-50 mg/L, sorbent dose 0.5 g, exposure time 150 min, at pH 6 for Pb and exposure time 60 min, at pH 7 for Zn).

$$\frac{C_e}{q_e} = \frac{1}{q_m K_l} + \frac{C_e}{q_m} \tag{7}$$

where $C_e$ is the equilibrium concentration of the remaining metal ion in solution (mg/L), $q_e$ (mg/g) is the amount of metal ion adsorbed by the adsorbent at equilibrium, $q_m$ is the maximum metal uptake capacity, and K is the equilibrium constant. The linearized Freundlich model is represented by Equation 8:

$$\text{Log } q_e = \log K_f + \frac{1}{n} \log C_e \tag{8}$$

where $K_f$ and n are Freundlich constants indicating the adsorption capacity and intensity, respectively. The values of the regression coefficient, $R^2$, $K_f$ and n were obtained from the plot of log $q_e$ against log $C_e$ as in Figure 5 and the data are presented in Table 4. For each metal the adsorption data fit the Freundlich isotherm better (indicated by a higher value for the regression coefficient, $R^2$). The Pb(II) adsorption exhibited a higher $K_f$ value than the Zn(II) adsorption, demonstrating that the phytosorbent has a higher capacity for binding Pb(II) than Zn(II) ions. A 1/n value of <1.0 (Surchi, 2011; Alfa *et al.*, 2012) for Pb(II) adsorption is also a clear indication that adsorption is most effective at an initial concentration range of 10-50 mg/mL .

## 4. Conclusion

This study has proven that a mixture of galangal leaves and pseudostems can be employed as a phytosorbent for the removal of lead and zinc ions from synthetic wastewater. The efficiency of lead adsorption is higher than that of zinc, but for both metals, variations in pH and the duration of the experiment have only a marginal effect on adsorptivity. Hence, galangal remains an effective adsorbent for both Pb(II) and Zn(II) ions over a wide range of pH values and contact times. Maximum adsorption for both metals occurred at a dosage of 0.5 g galangal/50 mL wastewater. For Pb(II) adsorption was most efficient at an initial metal ion concentration of 50 ppm, and a pH of 6 over a period of 150 min. However, with Zn(II) adsorption was most efficient at a pH of 7, with a contact time of only 60 min. The phytosorptions of both lead and zinc by galangal are pseudo-second-order processes which are well-described by the Freundlich isotherm.

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Table 4. Langmuir and Freundlich parameters.

| Metal | Langmuir | Freundlich | | |
|---|---|---|---|---|
| | $R^2$ | $R^2$ | $K_f$(mg/g) | 1/n |
| Pb | 0.908 | 0.988 | 0.877 | 0.71 |
| Zn | 0.388 | 0.925 | 0.131 | 1.22 |

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