Coffee Biomass Encapsulated in Calcium Alginate as Material for Lead (II) Adsorption from Aqueous Solutions

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ABSTRACT

This work aims to study the removal of lead (II) from aqueous solutions with Ca-Alginate (CA) and Coffee-Calcium-Alginate (CCA). The coffee biomass were successfully prepared as the material to be encapsulated in calcium alginate. The characterization of the synthesized CA and CCA was performed using fourier transform infrared and scanning electron microscopy. The method used was batch study. Various factors which affected adsorption efficiency of lead (II) ions by CA and CCA, such as pH, agitation time, and adsorbent dose were investigated for determination of optimum experimental conditions. The result showed that CA and CCA had significant effects on adsorption of lead (II) ions at pH = 4, agitation time of more than 120 min, and the adsorbent dose was 0.05 gram. Moreover, the Langmuir isotherm model showed that maximum adsorption capacity (qm) was 163.66 mg/g and 176.99 mg/g respectively for CA and CCA. The Langmuir isotherm was better described the adsorption equilibrium. Both of the adsorbent fitted to pseudo second order equations. These results demonstrated that CA and CCA show great potential to remove Pb(II) ions from aqueous solutions.

Keywords: adsorption, alginate, coffee, encapsulation, lead

INTRODUCTION

Water, an important and abundant resource, plays an important role in our life. According to the ministry of health Indonesian Republic number 492/ Menkes/ Per/ IV/ 2010, the maximum standard of Lead (II) contained in drinking water is 0.01 mg/L [1]. Therefore, the presence of lead poses a big problem to the environment and humans because the characteristic of lead are non-biodegradable and can affect human health, such as hepatitis, anemia, hypertension, and cancer [2]. The growth of industry and chemical uses by the industry has resulted in waste. Those become the major environmental problem since chemicals used by industry. A survey conducted by International Lead and Zinc Study Group (1992) reported that in 1990 lead has been used as the battery materials (64.4%), pigments (10.9%), extruded products (7.8%), metal coatings (3.5%), wire coatings (7.8%), bullet materials (3.8%), fuel additives (3.8%) and et cetera (3.1%) [3]. Several methods have been applied to treat contaminated water such as chemical precipitation, membrane filtration, ion exchange, liquid extraction, and electro-dialysis. These methods are not effective because they require high operational costs and are less suitable for processing water on a small scale [4]. The adsorption method has been known as an effective process for reducing ions metals from liquid waste because it is easy to separate, and has high effectiveness. The effectiveness of the adsorption method gives a thought for researchers to utilize biomass as adsorbent of heavy metal ions.
because of its recyclability, non-dangerous, easy to obtain, and does not require high operational costs. Biomass for ion adsorption has been widely used such as peanut shells for anionic dyes adsorption [5] and marine brown algae for lead adsorption [6]. Biomass that can be used to adsorb heavy metals is coffee powder derived from residual coffee beans. Indonesia is an agrarian country where coffee is one of the main commodities. According to a report from the Directorate General of Plantations (2015), Indonesia coffee production was estimated at 675,881 tons in 2013 [7]. AEKI (Association Indonesian Coffee Exporters and Industry) reported that in 2016 estimated coffee consumption in Indonesia reached 1.15 kg/capita/year [8]. Meanwhile, there are some benefits which coffee biomass offered due to the high presence of cellulose. The coffee biomass powder used in waste adsorption makes it difficult to separate between adsorbent and adsorbate after the adsorption. Encapsulation technique offer an advantage that facilitates separation between the adsorbate and the adsorbent. Biomass encapsulation with an alginate polysaccharide with chains (1 → 4)-linked β-D-mannuronic acid and α-L-guluronic acid can increase adsorption capacity because alginates have active groups that play a role in heavy metal adsorption. The synergistic effect of encapsulated biomass alginate was found to be effective with an adsorption capacity of 171.1 mg/g and 259.8 mg/g for calcium-alginate and calcium encapsulated activated sludge alginate for Cd(II) adsorption [9]. Coffee and alginate are also expected to have a synergistic effect for lead (II) from aqueous solutions with Ca-Alginate (CA) and Coffee-Calcium-Alginate (CCA).

EXPERIMENT

Chemicals and instrumentation
The chemicals used in this study were coffee biomass, a robusta coffee-local coffee from Indonesia, sodium alginate (Sigma Aldrich), hydrochloric acid 37% (Merck), sodium hydroxide (Merck), lead (II) nitrate (Merck), calcium chloride dihydrate (Merck), and distilled water.

Instrumentation applied for analysis were fourier transform infra red (FTIR Shimadzu IR prestige-21), atomic adsorption spectrophotometry (AAS Double Beam GBC®-avanta), pH meter (ORION Model 210A), and scanning electron microscope (AAS JEOL-JSM-6510LV).

Coffee Biomass Preparation
Coffee biomass obtained from household waste was washed with distilled water until the extract turns colorless then dried with oven for 5 hours at 105°C. Dried coffee biomass was sieved with 140 mesh sieve size. Coffee biomass then characterized by FTIR.

Calcium Alginate (CA) and Coffee-Calcium-Alginate (CCA) Preparation
The sodium alginate 2% solution was dropped with the syringe into the calcium chloride solution 0.1 M. The CA beads formed were stirred by a magnetic stirrer. The CA beads then soaked in calcium chloride bath within 2 hours. To remove the excess calcium chloride, the CA beads were washed with a particular amount of distilled water repeatedly until the pH of distilled water reaches 7. The CA beads were dried 48 hours.

To prepare the CCA beads, the coffee biomass 0.25 grams were mix with distilled water and sodium alginate until the mixture resulted in 2% sodium alginate. A similar synthesis procedure was adopted from CA synthesis as it follows the process of dropping the mixture of coffee-calcium alginate, washing the excess calcium chloride, and drying the beads. Both CA and CCA beads were characterized by FTIR and SEM.
Adsorption Studies

To optimize the lead (II) ions removal by CA and CCA, all of the experiments were carried out by batch method. Various factors that affected the adsorption efficiency of lead (II) ions by CA and CCA, such as pH, contact time, and adsorbent dose were investigated for the determination of optimum experimental conditions. The determination of optimum pH was done by placing 0.05 gram CA and CCA in Erlenmeyer flask 100 mL containing 100 ppm lead (II) solutions. The pH of the solutions then adjusted by adding hydrochloric acid 0.1 M and sodium hydroxide 0.1 M to vary the pH from 3 up to 7. The Erlenmeyer flasks were agitated at 200 rpm for 120 minutes. After the samples were filtered, the remaining lead (II) ions were measured using atomic adsorption spectrophotometry (AAS). The percentage of adsorption (% Adsorption) is expressed as:

\[
\text{% Adsorption} = \frac{C_i - C_e}{C_i} \times 100
\]  

where \( C_i \) is the initial concentration of lead (II) solution (mg/mL), \( C_e \) is the final concentration of lead (II) solution (mg/L). The adsorption capacity (qm) was determined by this following equation:

\[
q_e (\text{mg/g}) = \frac{C_i - C_e}{m} \times V
\]

where \( q_e \) is adsorption capacity at the equilibrium (mg/g), \( C_i \) is the initial concentration of lead (II) (mg/L), \( C_e \) is the final concentration of lead (II) (mg/L), \( V \) is the volume of lead (II) and \( m \) is the adsorbent mass (g) used.

The effect of agitation time between CA and CCA with the lead (II) ions was carried out by contacting 0.05 gram CA and CCA respectively at 200 rpm, initial lead (II) concentration 100 ppm, pH = 4 for 60, 120, 180, 240, 300, 360 and 420 minutes. To determine the optimum adsorbent dosage, CA and CCA were placed in Erlenmeyer flasks 100 mL containing 100 ppm lead (II) at the pH = 4, rotation 200 rpm for 2 hours with the adsorbent dosage 0.025; 0.050; 0.100; 0.150; 0.200; 0.250 for both CA and CCA. The effect of the initial lead (II) concentration was carried out by varying the lead (II) concentrations (10, 15, 20, 25, 30, 100, 200, 400 ppm) and contacting them to 0.05 gram CA and CCA respectively for 2 hours, pH = 4 at 200 rpm. The experimental set up for the removal of lead (II) ions from aqueous solutions is shown by Figure 1.

Figure 1. The experimental set up for the removal of lead (II) ions from aqueous solutions
RESULT AND DISCUSSION

Coffee Biomass Characterization

The coffee biomass is washed by the distilled water to remove the dirt and the color before it dried at 105°C for 5 hours. The coffee biomass then sieved by the 140 mesh sieve size to result in a homogenous coffee particle and to enlarge the surface area of lead (II) adsorption. According to Infra-Red (IR) spectrum in Figure 2, there is a vibration of O-H stretching at 3427.51 cm\(^{-1}\). The peak shows that O-H resulted from the carboxylate functional group contained in proteinogenic amino acid such as glycine, glutamate, and aspartate acid which are the main coffee’s organic component \([10]\). The peaks showed at 2944.09 cm\(^{-1}\) and 2664.66 cm\(^{-1}\) indicate the asymmetrical and symmetrical stretching of CH\(_2\) vibrations. The vibration at 1633.00 cm\(^{-1}\) contributes to C=C or C=O stretching and O-H bending. The carboxyl vibration from the xanthenes derivate like caffeine and the secondary amino group (N-H) at the adsorbent surface was indicated at 1744.58 cm\(^{-1}\) and 1460.11 cm\(^{-1}\) \([9]\).

![Figure 2. Coffee Biomass Infra-Red Spectrum](image)

Calcium Alginate (CA) and Coffee-Calcium-Alginate Characterization (CCA)

The Fourier Transform Infra-Red (FTIR) analysis was carried out to 4 samples which were calcium alginate before adsorption (CA), calcium alginate after adsorption (CA-Pb), coffee-calcium-alginate before adsorption (CCA), and coffee-calcium-alginate after adsorption (CCA-Pb). The infra red spectrums are shown in Figure 3.

![Figure 3. Infra-Red Spectrum of CA (left) and CCA (right) before and after adsorption.](image)
The infra red spectrum indicated that several functional groups resulted in wave number shifting after adsorption. Based on the CA and CA-Pb spectrum, the hydroxyl (O-H) functional groups has shifted from 3388 cm\(^{-1}\) to 3408 cm\(^{-1}\), the carbonyl (C=O) which appears at 1610 cm\(^{-1}\) has shifted into 1604 cm\(^{-1}\), the carboxyl (COOH) which appears at 1421 has shifted into 1423 cm\(^{-1}\). As can be seen from CA-Pb and CCA-Pb spectrum, the O-H functional group has shifted its wave number from 3390 cm\(^{-1}\) to 3412 cm\(^{-1}\), the C=O functional group appeared at 1629 cm\(^{-1}\) has shifted into 1610 cm\(^{-1}\) and the COOH appeared at 1427 cm\(^{-1}\) has shifted to 1429 cm\(^{-1}\).

According to 3000 times magnifying morphology of CA and CCA by SEM, the visual image from both adsorbent after adsorption demonstrated that the surface area of the adsorbent has larger pores. The compact ridge-like structure was observed at the surface of CA and CCA before adsorption showed in Figures 4 and 5 which related to the high degree of dehydration during the drying process [11].

![Figure 4. SEM images of CA (left) and CA-Pb (right)](image)

![Figure 5. SEM CCA (left) and CCA-Pb (right)](image)

The targeted chemical analysis of adsorbent surfaces was informed by SEM-EDS. As can be seen from Table 1, the main component of CA and CCA are carbon, oxygen, and calcium. The abundance number of carbon and oxygen indicates the presence of alginate and coffee biomass. The high percentage of calcium represents that cross linker-calcium chloride is the primary source of calcium. The percentage of lead (II) does not reflect how much lead (II) bonded to the CA and CCA surface due to the unhomogenized surface area inferred by SEM-EDS. Therefore, the data doesn't represent the whole surface area of the adsorbent.
Table 1. Percentage of elements in CA and CCA before and after adsorption analyzed by SEM EDS

| No | Elements  | Percentage (%) | CA    | CCA   | CA    | CCA   |
|----|----------|----------------|-------|-------|-------|-------|
|    |          | Before | After | Before | After | Before | After |
| 1  | Carbon (C) | 38.22  | 44.53 | 41.95  | 44.76 |
| 2  | Oxygen (O) | 17.63  | 38.75 | 16.56  | 38.66 |
| 3  | Calcium (Ca) | 44.16  | 1.92  | 41.49  | 2.25  |
| 4  | Lead (Pb)  | 0      | 14.8  | 0      | 14.33 |

Adsorption Studies

The optimum pH condition becomes a crucial factor in the adsorption process because pH affects the charge and the active group on the surface of the adsorbent [12]. Figure 6 shows the result. The adsorption capacity increased significantly from pH = 3 to pH = 4 but slightly decreased at pH = 5. At the low pH, the adsorption capacity is relatively small. The huge number of H⁺ interacted with the functional groups such as carboxyl on the surface area of the adsorbent and the protonated functional group resulted in the small adsorption capacity at the low pH. The pH = 4 become the optimum pH to conduct the lead (II) adsorption with the adsorption capacity (qe) 4.29 mg/g and 4.53 mg/g respectively for CA and CCA. The dissociated β-D-manuronic acid (pKa = 3.38) and α-D-guluronic acid (pKa = 3.65) at pH 4 provide abundance negative charge that effectively interacted with lead (II) ions. [13] The adsorption capacity was slightly decreased at pH 5 and 6 due to the interaction between lead (II) and OH⁻.

Figure 6. Effect of pH on lead (II) adsorption from aqueous solution
(agitation time: 120 min, volume 50 mL, dosage 0.05 g, 200 rpm, Lead (II) 100 ppm)

The result of agitation time represented by fig 7 shows that lead (II) adsorption was rapid at 10 minutes up to 120 minutes and then slightly constant after it reached 120 minutes. It indicates that the adsorption of lead (II) takes place in two stages; the rapid adsorption and the slow adsorption rate until it reaches the equilibrium. The slow adsorption stage was caused by a reduction of the adsorbents’ active site and intraparticle diffusion which dominate the adsorption process.
The adsorbent dosage shows how much the adsorbent needed for a particular adsorption condition. The mass of the adsorbent directly affects the amount of adsorbents’ active site. The percentage of adsorption (% adsorption) increases along with the increase of CA and CCA mass. After the mass of adsorbent reached 0.05 gram, % adsorption tends to be constant due to some active adsorption sites on the adsorbent remained unsaturated because of insufficient lead (II). Therefore, the adsorbent dosage 0.05 gram observed as the optimum dosage with the % adsorption 94.65% and 95.76% for CA and CCA respectively.

The result of the initial concentration of lead (II) shows that the adsorption capacity of CA and CCA were almost similar. It informs that the interaction between lead (II) and alginate is dominant compared to coffee biomass. The alginate functional groups are more dominant in the adsorption. Based on the experiment, the maximum adsorption capacities are 155.00 mg/g and 170.00 mg/g respectively for CA and CCA. Along with the increasing of lead (II) concentration, there is a bigger mass transfer that occurs in lead (II) to the adsorbents’ active site [14]. When the lead (II) concentration reached 400 ppm, the increase of adsorption capacity wasn’t significant. This is due to the active sites of adsorbent that have been saturated by lead (II). The repulsion force of bonded lead (II) to the unbounded lead (II) can also decrease the interaction of Pb(III) in the solution to the adsorbents’ active site.

**Adsorption Isotherm**

The adsorption isotherm describes the distribution of ions in the solid phase, which is the adsorbent surface and in the liquid phase when the adsorption process reaches equilibrium [15]. Two isotherm models that are often used in the adsorption process are Freundlich and Langmuir.
isotherms. The Freundlich isotherm is applicable to both monolayer (chemisorption) and multilayer (physisorption) and assumes that the adsorbate adsorbs onto the heterogenous adsorbent surface [16]. The linear equation of Freundlich equation is expressed as:

\[ \log q_e = \log K_F + \frac{1}{n} \log C_e \]  

(3)

where \( K_F \) and \( n \) are Freundlich isotherm constants related to adsorption capacity and adsorption intensity, and \( C_e \) is the equilibrium concentration (mg L\(^{-1}\)) [16]. The Langmuir isotherm describe monolayer adsorption on a uniform surface with a finite number of adsorption sites (boparai). The linear equation of Langmuir equation is expressed as:

\[ \frac{C_e}{q_e} = \frac{1}{K_L q_m} + \frac{C_e}{q_m} \]  

(4)

Where \( K_L \) is the Langmuir constant related to the energy of adsorption and \( q_m \) is the maximum adsorption capacity (mg g\(^{-1}\)).

Figures 9 and 10 show that the adsorption data for Pb (II) ions by CA and CCA fitted to the Langmuir isotherm better than the Freundlich adsorption isotherm. This can be seen from the correlation value (R\(^2\)) of the adsorption isotherm; 0.992 for CA adsorbent and 0.994 for CCA adsorbent. The Langmuir isotherm on the adsorption of Pb (II) ions by CA and CCA was also strengthened by the results of the substitution of the parameters in the linear equation against the non-linear equation of Langmuir and Freundlich adsorption isotherms. Figure 11 shows that the Ce plot against qe experimental results tend to follow the non-linear equation of the Langmuir adsorption isotherm. The Langmuir isotherm assumes that the monolayer is formed at the adsorbents’ surface. After that, there will be no other layers formed, following by saturated adsorbents’ surface [16]. The qm results that the maximum capacity of CA and CCA are 163.66 mg/g and 176.99 mg/g for lead (II) adsorption. Other studies found that the adsorption capacity to lead (II) was 65.50 mg/g for tea biomass [17], and 37.04 mg/g to coffee husk biomass [18].

**Figure 9.** The Freundlich linier plots of lead (II) adsorption by CA (left) and CCA (right)
Figure 10. The Langmuir linear plots of lead (II) adsorption by CA (left) and CCA (right)

Figure 11. The non linear plots of lead (II) adsorption by CA (left) and CCA (right)

As can be seen in the Table 2, The Freundlich isotherm constant (K_f) is determined from the intercept and slope of the log qe curve against log Ce. The Freundlich isotherm with a value of n> 1 shows a high affinity between the adsorbate and the adsorbent which is classified as a chemisorption [16].

Table 2. The Langmuir and Freundlich isotherm parametric

| Isoterm  | Adsorben | Parameter | R²   |
|----------|----------|-----------|------|
|          |          | K_i       | n    |
| Freundlich | CA     | 8,95      | 1,894 | 0,914 |
|           | CA-K    | 11,56     | 1,992 | 0,873 |
| Langmuir  | CA      | 163,66    | 176,99 | 0,995 |
|           | CA-K    | 170,00    | 0,035  | 0,992 |

Adsorption Kinetics

The adsorption kinetics was found to be important because it informs the adsorption mechanism. Furthermore, the adsorption kinetics illustrated the adsorption rate and agitation time of the adsorbate between the solid phase and liquid phase. The pseudo first-order equation describes adsorption in solid–liquid systems based on the sorption capacity of solids [16]. It indicates that one lead ion is adsorbed onto one sorption site on the CA and CCA surface:
\[ A + Pb_{aq}^{2+} \xrightarrow{k_1} APb_{solid\ phase} \]  \hspace{1cm} (5)

where \( A \) represents an unoccupied sorption site on the CA and CCA and \( k_1 \) is the pseudo first order rate constant (h\(^{-1}\)). The linear form of pseudo first order model can be expressed as:

\[
\log(qe - qt) = \log qe - \frac{k_1}{2.303} t
\]  \hspace{1cm} (6)

where \( qe \) and \( qt \) (mg g\(^{-1}\)) are the adsorption capacities at equilibrium and at time \( t \) (h), respectively. The pseudo second order rate describe the chemisorption kinetics from liquid solution \(^{[16]}\) and it is expressed as:

\[
\frac{t}{qt} = \frac{1}{k_2q_e^2} + \frac{1}{q_e} t
\]  \hspace{1cm} (7)

where \( k_2 \) is the rate constant for pseudo second-order adsorption (g mg\(^{-1}\) h\(^{-1}\)) and \( k_2q_e^2 \) or \( h \) (mg g\(^{-1}\) h\(^{-1}\)) is the initial adsorption rate. This model assumes that one lead ion is adsorbed onto two sorption sites on CA and CCA surfaces:

\[ A + Pb_{sol}^{2+} \xrightarrow{k_2} A_2Pb_{solid\ phase} \]  \hspace{1cm} (8)

The experimental results in Figure 12 and 13 give the value of the correlation \( (R^2) \) for the pseudo second order model is better than the value of pseudo-first-model with the correlation value \( (R^2) \) 0.8221 and 0.9481 for CA and CCA assumed by pseudo-first-order model; 0.9972 and 0.998 for CA and CCA assumed by pseudo second order model. This suggested that, pseudo-second-order model could describe the adsorption of lead (II) on CA and CCA better than the pseudo-first-order model. The adsorption could be chemisorption mainly due to exchange or sharing of electrons between the solid adsorbent; and the adsorbate; which agreed well with previous work (Surchi, 2011).

**Figure 12.** The pseudo first order plots for lead (II) adsorption by CA (left) and CCA (right)
Figure 13. The pseudo second order plots for Lead (II) adsorption by CA (left) and CCA (right)

CONCLUSION
The lead (II) adsorption mechanism by CA and CCA can be described by the isotherm model, adsorption kinetic, and Infra-Red spectrum. Isotherm study and adsorption kinetic state that the adsorption of lead (II) by CA and CCA follows Langmuir isotherm and pseudo-second-order equation. It shows that the adsorption mechanism was dominated by chemisorption where one adsorbate molecule interacts with two of the adsorbents’ active sites. Several functional groups of CA have shifted its wave number mainly to the O-H, C=O, and C-O functional groups. The carbonyl has shifted into a lower wavenumber. It indicates that C=O bonded weakened after having an interaction with lead (II). Electrostatic interactions between C = O and ions lead (II) cause the C = O bond to weaken thereby reducing the amount of energy needed for the C = O group to vibrate. Based on the study, it can be concluded that the adsorption optimizes at pH 4, agitation time 120 minutes, the adsorbent mass of 0.05 grams, and an optimum concentration of 720 ppm. Data isotherms and kinetics of adsorption indicate that the maximum adsorption capacity (qm) respectively was 163.66 mg/g for CA and 176.00 mg/g for CCA.

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