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Study of Equilibrium and Kinetics of Pb(II) in Solution Using Persimmon Tannin Gel as an Adsorbent

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Abstract

Heavy metals concentrations exceeding the recommended threshold are hazardous for the environment, so there is a need for handling it safely. The purpose of this research was to determine the adsorption capacity and kinetics of adsorption. This research uses an adsorption method in the process of removing Pb(II) heavy metal ions using persimmon tannin gel. Besides the influence of contact time, pH, and the concentration of Pb(II) metal ions on adsorption, a kinetics study was also carried out. The adsorption rate is obtained through the adsorption rate constant (K) and the reaction order generated from the kinetics model. Based on the results of the research, showed the optimum adsorption process, which is 20 minutes and at a pH of 5. The maximum adsorption capacity of 17.62 mg/g with the value of the standard energy changes of Gibbs adsorption ($-\Delta G^{\circ}$ = -14.274 kJ/moll) indicates that the adsorption takes place through physical interaction. The kinetic adsorption gives the adsorption rates a constant of, k = 0.008231 g/mg.min.

1. Introduction

The rapid growth of the world's population and the rapid development of industry causes the more and more toxic waste material to be discharged into the environment. These waste materials can become toxic waste and pollutants to the environment if it is released in large quantities that become difficult to control precisely. In Indonesia, the source of pollutants comes from household waste, companies, mining, industry, and others. Among these toxic waste materials are heavy metals [1]. The nature of heavy metals is that they are difficult to degrade and are cumulative; large quantities of toxic waste that accumulated is hazardous. Heavy metals include cadmium (Cd), cobalt (Co), chromium (Cr), copper (Cu), iron (Fe), lead (Pb), Zinc (Zn) and others.

Lead metal is widely used in the battery, cable, and paint industry (as a coloring agent), electroplating pesticides, and it is widely used as an antipyretic agent in gasoline. Lead is also used as a pipe joint formulation, which results in household water have a high possibility of containment from lead itself [2]. The process of removing heavy metals from liquid waste has been done in several ways, such as chemical precipitation [3], extraction using certain solvents, ion exchange, reverse osmosis [4], and adsorption [5]. The adsorption process with the right choice of adsorbent type compared to other methods is a more straightforward and effective process in removing heavy metals from liquid waste [6] and does not give side effects in the form of toxic gases [7].

The natural material is a potential adsorbent because it is cheap and environmentally friendly [8]. Tannin is a natural material that has the potential to be developed into an adsorbent. Tannins are polyphenol compounds found in many plants, where they function as a means of self-defense from predators. Tannins can form complexes and settle with macromolecules such as proteins, fats, polysaccharides, and heavy metals [9, 10].

Persimmon (*diospyros kaki* L.), which is rich in condensed tannins (proanthocyanidins) which are soluble in water and have a low molecular weight, generally have a sour feeling (astringency). Water-soluble tannins with low molecular weight cause the taste of sepimmon fruit. The main structure of tannins from persimmon fruit consists of 2 types of flavan-3-ol, namely catechin (CA) and gallocatechin (GC), and its gallate form, CA-3-O-gallate (CG) and GC-3-O- gallate (GCG), with an interfluve carbon-carbon bond at position C-4 from one unit to position of C-8 (or C-6) at another group. The flavan-3-ol unit stereochemistry of persimmon tannin is dominated by 2,3-cis, which is a type of epi-catechin, as shown in Figure 1 [11].

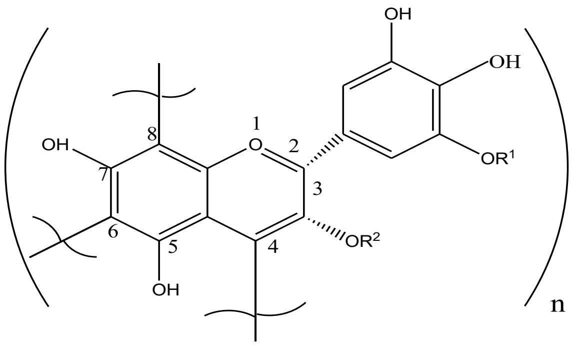

Figure 1. Chemical structure of persimmon tannin, R1 = H or OH, R^2 = H or galloyl

Several methods have been reported in the preparation of tannin gels, mostly using formaldehyde or aldehydes in acid or base media and autoxidation [12]. Furthermore, the gel has been made from persimmon tannins using formaldehyde cross-linking agents and shows a high affinity for methylene blue dyes [5]. In this research, persimmon tannin gel will be used to adsorb lead metal ions in aqueous systems. The equilibrium and kinetics adsorption of lead metal ions were studied. The effect of several parameters that affected the adsorption is further analyzed.

2. Methodology

The tools and materials used in this research are as follows.

2.1. Equipment and Material

The tools in this research are atomic absorption spectrophotometer (Perkin Elmer Analyst 400), jar test (Flocculator SW6 Stuart), pH meter. The materials used in this study were persimmon tannin gel, lead anhydrous nitrate ($Pb(NO_3)_2$, E.Merck), Whatman filter paper No. 40, hydrochloric acid (37% HCl, E. Merck), sodium hydroxide (sodium hydroxide (NaOH, E. Merck).

2.2. Preparation of Pb(II) Solution 1000 mg/ L

A 1000 mg/L Pb(II) ion solution was prepared by dissolving 0.799 grams of anhydrous Pb(NO3) 2 in 500 mL of distilled water.

2.3. Making a Standard Curve

To make a standard curve for Pb(II) metal ions, a series of standard solutions of Pb(II) metal ions with a concentration of 0; 2; 4; 6; 8; and 10 mg/L are required. The series of standard solutions absorption capacity is measured using atomic absorption spectrophotometers; then, by channeling absorbance to the concentration of standard solutions, a standard curve can be obtained. From the standard curve above we get the linear regression equation:

$$y = a + bx \quad (1)$$

where a is the intercept, b is the slope value, x is the concentration, and y is the absorbance.

2.4. Effect of Contact Time [13]

An amount of 250 mL Pb(II) metal ion solution with a concentration of 200 mg/L mixed with 0.1-gram persimmon tannin gel. Then the mixture is stirred in a test jar at a speed of 120 rpm. Contact time varies with successive intervals of 10, 20, 30, 40, 50, and 60 minutes. The filtrate was further separated by filtering using Whatman filter paper and then analyzed by atomic absorption spectrophotometer to determine the concentration of Pb(II) metal ions which were adsorbed using equation (2).

2.5. Effect of pH Solution [14]

Forty milliliters of Pb(II) solution at a concentration of 200 ppm was put into five 500 mL beaker with the pH being varied (variation of pH 3; 5; 7; 9 and 11) with the addition of 0.1 M HCl and 0.1 M NaOH and added drop by drop. The pH value is measured using a pH meter. Furthermore, the Pb(II) metal solution with a different pH is contacted with 0.1 gram of persimmon tannin gel. Then stir using the test jar at a speed of 120 rpm at the optimum contact time. After that, the mixture is filtered using Whatman filter paper. The resulting filtrate was analyzed using an atomic absorption spectrophotometer to determine the metal ions that are absorbed by the adsorbent.

2.6. Effect of Pb(II) Metal Ion Concentration

A total of 40 mL of Pb(II) metal ion solution with a variety of concentration at 150; 200: 300 and 400 mg/L mixed with 0.1-gram persimmon tannin gel, then stirred using the test jar at a speed of 120 rpm with the optimum contact time and optimum pH. The filtrate was separated by filtering using Whatman filter paper and then analyzed by SSA to determine the concentration of Pb(II) metal ions, which were not adsorbed. The results obtained are plotted to the Freundlich Equation and the Langmuir Equation to determine the capacity and energy of adsorption of tannin persimmon gel.

2.7. Determination of Pb(II) Metal Ion Adsorption Capacity

The concentration of Pb(II) metal ions adsorbed by persimmon tannin gel can be calculated using the following equations :

$$[Pb(II)]\ adsorbed = [Pb(II)]\ initial - [Pb(II)]\ end \quad (2)$$

$$\%\ [Pb(II)]\ adsorbed = \frac{[Pb(II)]\ adsorbed}{[Pb(II)]\ initial}\ x\ 100\ \% \quad (3)$$

$$Adsorption\ capacity = \frac{weight\ of\ adsorbed\ Pb(II)\ (mg)}{adsorbent\ weight\ (g)} \quad (4)$$

The adsorption equation in this research is determined through the Freundlich or Langmuir isotherm equation. For the Freundlich isotherm, a log graph C_e versus log q_e (equation 6) was made. The slope value is the KF log, and the intercept is 1/n. For the Langmuir isotherm, a graph of C_e versus C_e/q_e (equation 5) was made. The value of slope (b) is 1/qm, while intercept (a) is 1/Ka.qm.

Langmuir equation

$$\frac{C_e}{q_e} = \frac{1}{q_m K_L} + \frac{C_e}{q_m} \quad (5)$$

With q_e is the amount of substance absorbed per unit weight of the adsorbent (mg/g), C_e is the concentration of adsorbate at equilibrium (mg/L), qm is the maximum adsorption capacity representing the Langmuir constant (mg/g), and KL is the Langmuir constant (Langmuir (constant) L/mg).

Freundlich equation

$$\log\ q_e = \log K_F + \frac{1}{n}\log C_e (6)$$

With q_e is the amount of substance absorbed per unit weight of adsorbent (mg/g), while C_e is the concentration of adsorbate at equilibrium (mg/L), n is the maximum adsorption capacity (mg/g), and K_F is the Freundlich constant (L/mg).

The data obtained was done by analyzing its reaction order with pseudo-first-order formulas and pseudo-second-order formulas so that the *k* value can be determined. By connecting log (q_e-qt) to t and t/qt to t, a straight-line equation will be obtained. Pseudo-first-order kinetics and pseudo-second-order kinetics are compared to find similarities between them:

$$log\ (q_e - q_t) = \log q_e + \frac{K_1}{2{,}303} t \quad (7)$$

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{1}{q_e}\ t \quad (8)$$

2.8. Thermodynamic study (ΔG) [15]

The occurrence of the adsorption process involves intermolecular forces such as; electrostatic force, London forces, and the interaction of ions contained in the adsorbent or adsorbate so that the adsorption process will involve changes in energy. This energy change can be calculated using the equation:

$$E = \Delta G = -\ RT \ln K \quad (9)$$

With the value of ΔG being the change in Gibbs free energy (J/moll), R is the general gas constant (8.314 J/moll K), T is the temperature (Kelvin), and *k* is the adsorption equilibrium constant(L/moll).

3. Result and Discussion

3.1. Effect of Contact Time

Contact time is very instrumental in achieving adsorption equilibrium. The optimum time of adsorption of Pb(II) metal ions by tannin gel persimmon is determined by counting the number of Pb(II) metal ions adsorbed as a function of time. The results showed that the amount of Pb(II) metal ions adsorbed increased with increasing contact time between the adsorbent and the adsorbate until equilibrium was reached, as shown in Figure 2.

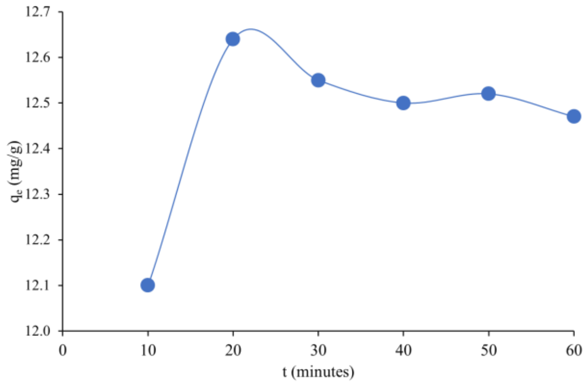

Figure 2. Effect of Pb(II) Metal Contact Time on Pb(II) Metal Adsorption Capacity (weight of adsorbent = 0.1 g, adsorbate concentration of 200 mg/L)

Adsorption of Pb(II) metal ions occurs very quickly in the 20th minute because there are still quite a lot of active sites available so that interactions take place immediately. This interaction occurs between the acidic OH group and the nucleophilic phenolic group on the surface of the persimmon tannin gel with the Pb(II) metal ion [16, 17, 18]. After the 20th minute, the amount of Pb(II) metal ions adsorbed slowly decreases. That is due to the saturated adsorbent surface; hence, some of the Pb(II) metal ions are desorbed.

3.2. The effect of pH

Adsorption of metal ions in solution is strongly influenced by pH. The pH value of the solution is significant in the adsorption process because changes in pH can cause changes in the charge on the surface of the adsorbent or metal ion species in the solution. The optimum pH value of the adsorption of Pb(II) metal ions by tannin gel persimmon can be determined by counting the number of Pb(II) ions adsorbed as a function of pH. The pH value greatly influences the condition of hydroxyl groups in tannin gel persimmon and the speciation of Pb(II) metal ions, thus determining the effectiveness of adsorption. The relationship between pH and the amount of Pb(II) metal ions adsorbed by persimmon tannin gel can be seen in Figure 3.

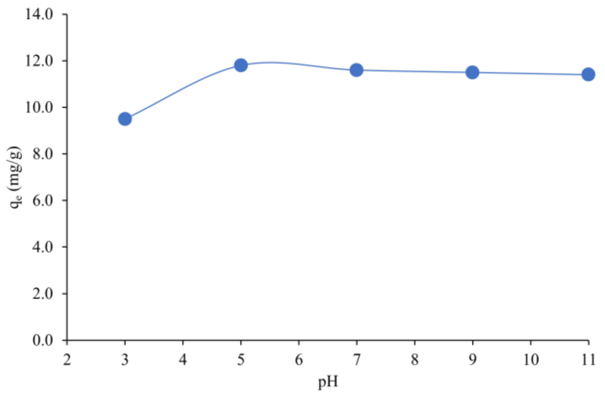

Figure 3. Relationship between pH and the amount of Pb(II) metal ions adsorbed (q_e) by persimmon tannin gel (adsorbent weight 0.1 g, adsorbate concentration 200 mg/L with 20 minutes)

The number of Pb(II) metal ions absorbed at pH 3 is minimal at only 9.656 mg/g and tends to increase until it reaches pH 5. This tendency is thought to be due to competition between H^+ and Pb(II) metal ions to react with active groups of persimmon tannin gel.

The higher the acidity, the more H^+ in the solution. As a result, the amount of Pb(II) metal ions adsorbed is minimal. At pH five, the number of Pb(II) metal ions adsorbed reached 11.826 mg/g with an efficiency (%) of adsorption of 14.77%. The efficiency (%) of adsorption is obtained from the substitution of initial concentration and concentration adsorbed into equation (3). Based on the results of this study indicate that the most significant adsorption capacity occurs at pH 5, then in the next treatment that is the adsorption of Pb(II) metal ions to the effect of the concentration of Pb(II) metal ion solutions used pH 5.

3.3. Effect of Concentration

Other factors affecting adsorption is the adsorbate concentration. Increasing the concentration of Pb(II) metal ions, which is contacted with a fixed amount of adsorbent, will result in decreased absorption of Pb(II) metal ions. That happens because the active adsorbent site is already saturated.

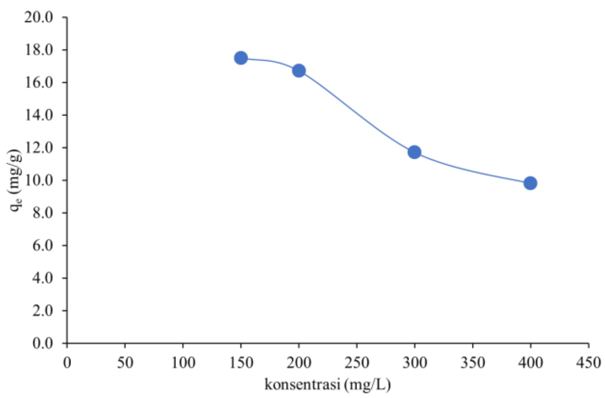

Figure 4. Relationship between concentration and the amount of Pb(II) metal ion adsorbed (q_e) by persimmon tannin gel (adsorbent weight 0.1 g, contact time 20 minutes and pH 5)

The effect of concentration on the amount of Pb(II) metal ions adsorbed can be seen in Figure 4. The number of Pb(II) metal ions adsorbed continues to decrease with the increase of concentration.

The results showed that the highest Pb(II) metal ion was adsorbed at 17.62 mg/g at a concentration of 150 ppm. That is due to the large number of OH groups and phenolic groups found in the tannin persimmon gel, so the adsorption capacity of the Pb(II) ions is also higher. The OH group and the presence of a phenolic group can exchange or form interactions with Pb(II) metal ions.

3.4. Maximum Adsorption Capacity

The adsorption capacity of the Pb(II) metal ion can be determined based on the Freundlich and Langmuir isotherm equations. The isotherm model that is suitable for adsorption is determined by comparing the linearity values of the two equations. The data in Figures 5 and 6 are then processed to obtain the Langmuir and Freundlich equation graphs. Based on Figure 5, the equation of the line $y = -0.225x + 66.4428$ is obtained while based on Figure 6, the equation of the line $y = 3.7181x - 7.445$. From the line equation in Figure 5 the slope value = -0.225 and intercept = 66.442 while the slope value of Figure 6 obtained the slope value = 3.7181 and intercept = 7.445. Furthermore, the slope and intercept values are substituted into equations (5) and (6).

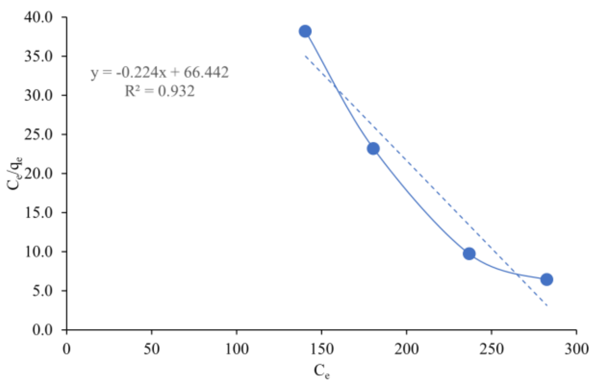

Figure 5. Adsorption of Langmuir metal ion Pb(II) by persimmon tannin gel

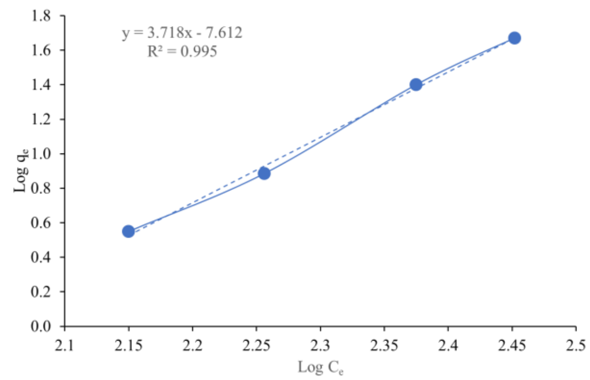

Figure 6. Adsorption of Freundlich Pb(II) metal ion by Persimmon tannin gel

Based on Figures 5 and 6 show that the adsorption of Pb(II) metal ions in the tannin gel persimmon follow the Freundlich isotherm. The value of the adsorption constant capacity (KF) and the adsorption intensity (n) were 1.146 (mg/g) (L/mg) 1/n and n = 0.314 L/mg. The value of n above one indicates that the adsorption process takes place physically [19].

The KL and qm constants can be used to predict the tensile strength between the absorbing material and the adsorbent using a separation factor or dimensionless balance parameter, RL. The RL value for Pb(II) at a different concentration is 0.985. The value of the RL separation factor is <1, indicating that the isotherm shape obtained is in an excellent category.

The adsorption capacity of tannin gel persimmon against Pb(II) metal ions was 17.62 mg/g. This value is smaller than the activated carbon adsorption of 151.52 mg/g [20] but higher than the adsorption by natural clay which is 4.0263 mg/g [21], and natural zeolite adsorbent which is 23.21 mg/g [22] so that the tannin gel persimmon has enormous potential to be developed and used as an adsorbent.

3.5. Thermodynamic Study (ΔG°)

Thermodynamic parameters in the adsorption system can be calculated using the Van, not Hoff equation ΔG° = -RT ln K. The *k* value is obtained from the value of the adsorption intensity (n) in the Freundlich isotherm equation. The value ΔG is used to determine the nature of the adsorption process. Spontaneous adsorption process, if at a constant temperature, gives a negative ΔG value.

The value of ΔG obtained was -14,274 kJ/mol. According to Ye *et al.* [23] Gibbs free energy for chemical adsorption is in the range of -80 kJ/moll to -400 kJ/moll and physical adsorption of -20 kJ/moll to 0 kJ/mol. The results of this study indicate that the adsorption takes place physically. That further strengthens the assumption of physical adsorption based on the n value described previously.

3.6. Adsorption Kinetics

The adsorption rate can be determined from the adsorption rate constant (K) and the reaction order generated from an adsorption kinetics model. Data of adsorbed metal concentration (q_e) can be processed to determine the reaction kinetics equation. The testing phase of the adsorption rate of Pb(II) metal ions can be carried out by estimating the reaction order, either pseudo-first-order or pseudo-second-order.

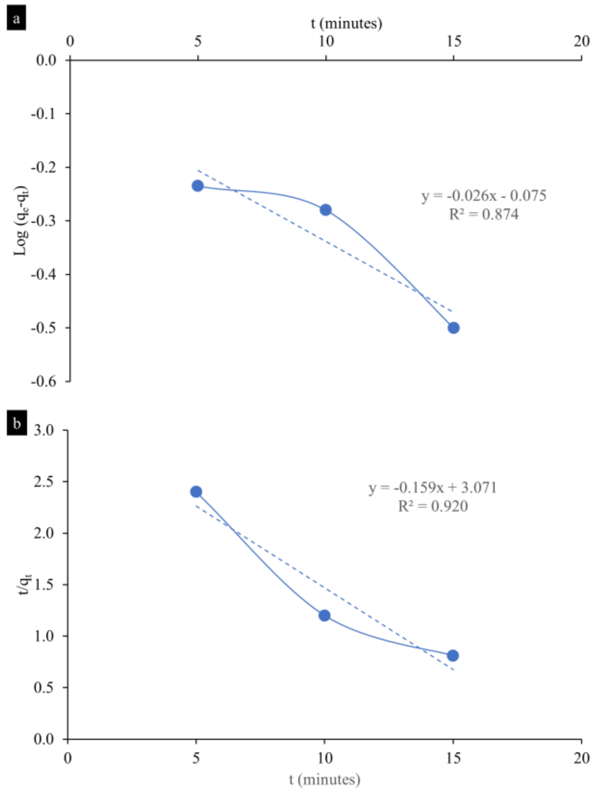

Figure 7. Kinetic adsorption model of Pb(II) metal ion by tannin persimmon gel: (a) pseudo first order and (b) pseudo-second-order

Figure 7 shows the kinetic adsorption model of Pb(II) metal ion by tannin gel persimmon based on pseudo-first-order and pseudo-second-order. Pseudo order reaction means that the Pb(II) metal ion adsorption reaction behaves like a first or second-order reaction. This situation is because the adsorbed substance exists in a fixed amount at a certain level, and the reaction rate is determined by one reactant. Based on Figure 7, the value of R^2 and the adsorption rate constant for pseudo-first-order equations are 0.8741 and k_1 = 0.05987 g/mg, respectively. For pseudo-second-order kinetics models, the value of R^2 = 0.9204 with k_2 = 0.008231 g/mg.min is obtained. The suitable adsorption model is determined by comparing the value of the curve linearity, where the greatest linearity is chosen as the corresponding model. Based on the R^2 values of the two models, which are close to 1, it is a pseudo-second-order equation. Thus the adsorption of Pb(II) metal ions by tannin gel persimmon follows a pseudo-second-order kinetics model. That means that the addition of the concentration of adsorbate or the amount of adsorbent will double the rate of adsorption.

4. Conclusion

The optimum time for the adsorption process is at 20 minutes, while the optimum pH value for the adsorption of Pb(II) metal ions is pH 5. Variation in concentration causes a decrease in adsorption as the concentration of Pb(II) metal ions in the solution increases. The adsorption capacity of the persimmon tannin gel is 17.62 mg/g. The kinetics that fulfills the adsorption process for Pb(II) metal are pseudo-second-order kinetics.

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