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Research Article

ADSORPTION OF Co (II) IONS FROM AQUEOUS SOLUTION ONTO TEA AND COFFEE POWDER: EQUILIBRIUM AND KINETIC STUDIES

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ABSTRACT

Adsorption of Co(II) ions from aqueous solutions onto Tea and Coffee powders has been examined in a batch adsorption process. The adsorption of Co(II) ions was found to be dependent on contact time, pH of solution, initial metal ion concentration, and adsorbent dose. The experimental equilibrium adsorption data were studied by Langmuir, Freundlich, Temkin and Dubinin–Radushkevic isotherms models. The Langmuir model display demonstrated a superior fit than the other three models by higher correlation coefficient, R^2. The maximum adsorption capacities calculated from the Langmuir isotherm model were 244 mg/g, for both coffee and tea at optimum circumstances. The kinetic studies denoted that the adsorption process of Co(II) ions followed well pseudo-second-order model. According to the adsorption capacity, coffee and tea powder considered as an effective, low cost, and environmentally friendly adsorbent for the removal of Co(II) ions from aqueous solutions.

Keywords: Adsorption; Cobalt; Isotherms model; Kinetic study.

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1. INTRODUCTION

The release of heavy metals in industrial waste water is of great consideration because their accumulation have a toxic impact on living species [1]. Industrial wastewater containing metal ions such as lead, cadmium, nickel, cobalt, copper, and zinc are widespread because these metals are widely used in industries such as batteries manufacture, mine, metal finishing, electroplating, brewery, pharmaceutical, and others. Heavy metals are poisonous to aquatic organisms even at extremely low concentration. Most of these metals were present in our environment only in trace levels until recent centuries, when the tendency toward manufacturing and production caused our many technological advances. But technology, such as medicine, has its lateral influences. Currently, heavy metals have contaminated our waters, atmosphere, soil, and food chain [2].

A common technique for separating metal ions from industrial waste water involves solvent extraction, chemical precipitation, coagulation, membrane separation, ion– exchange electrolysis, and adsorption. A large portion of these techniques endure with high capital and recovery expenses of the materials [3-15]. Therefore, there is presently a demand for novel, innovative and cost effective techniques for the removal of toxic metals from wastewaters. The use of biosorbents is an effective and multitalented process and can be simply adopted with minimum cost to remove heavy metals from great amount of industrial wastewaters. Recent studies have shown that heavy metals can be removed using plant materials such leaves of indigenous biomaterials, Tridax procumbens, peat moss, cork and yohimbe bark wastes, an palm pressed fibers and coconut husk, water fern Azolla filiculoidis, duck weed Wolffia globosa, and Rhizopus nigricans [5-18].

In our previous work, we have studied the efficiency of olive leaves and orange peels powders as an adsorbent for removal of cobalt from aqueous solutions [19,20]. In this paper, the adsorption of cobalt onto coffee and tea powders has been investigated. The adsorption capacity was evaluated under various conditions of pH, contact time, initial copper concentration, and adsorbent dosage. The adsorption isotherms were also investigated to explain the probable mechanism of adsorption and to provide several information such as maximum sorption capacity, energy of sorption, homogeneity/heterogeneity, and affinity between sorbent and adsorbent.

2. RESULTS AND DISCUSSION

By UV VIS Spectrophotometer, the concentrations of Co (II) as thiocyanate complex in the solutions were determined [21]. The range of calibration curve of cobalt ions prepared from stock solution varies between 20-80 ppm. From the calibration curve, the concentrations of cobalt (II) in the solutions before and after equilibrium adsorption were determined.

2.1 Effect of pH

The biosorption and removal of metal ions from wastewater and aqueous solutions are strongly controlled by pH changes. The pH of the solution is considered to be as the most important parameters governing metal up taking and adsorption on adsorbent substrate. The adsorption of cobalt ions was monitored over a range of pH from 2.00 to 6.50 of separate solutions Figure 1. Low percent removal were observed at low pH values because of H3O+ ions will compete with Co(II) for binding and also surrounded (H_3O^+) will prevent the cobalt ions from approaching the binding sites. However, at higher pH, (above pH 6), the Co (II) started precipitating, so the removal was not achieved only by adsorption. The maximum percent removal % R of cobalt ions by both absorbents were observed at pH 5.5 - 6.0. Therefore, the remaining all biosorption experiments were carried out at this pH values.

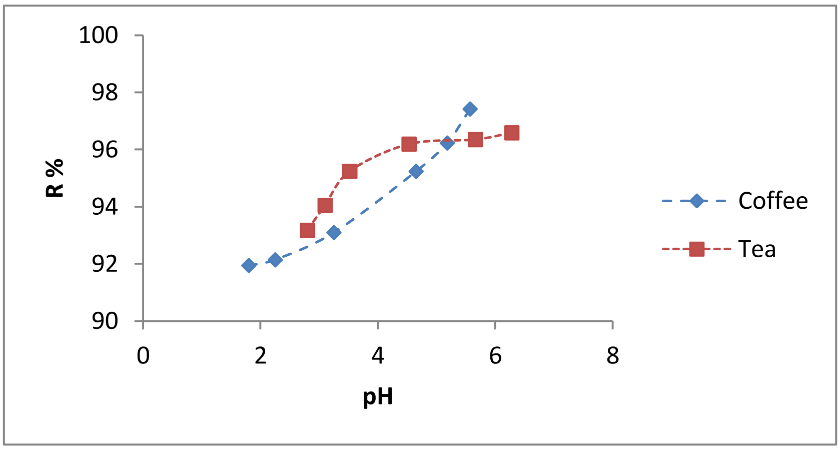

Fig.1. Effect of pH on percent removal of Co (II) (% R) onto both adsorbents

2.2 Effect of Initial Metal Concentration

The effect of different initial cobalt ion concentrations on equilibrium of adsorption experiments were investigated from 40 to 240 ppm at pH 5.50 – 6.00. The relation between cobalt uptake Q_m ($mg.g^{-1}$) and initial metal ion concentration is shown in Figure 2. The cobalt ion adsorption is possible at lower concentrations, but as the concentration is increased, the driving force also increased, which favored the adsorption at higher concentrations. The equilibrium uptake of the adsorbent was observed increasing gradually with an increasing the initial concentration of Co ions and then reach a maxima at a concentration of 240 ppm. The increasing of adsorption capacity with the increasing in metal ions concentration is probably due to higher interaction between the metal ions and sequestering sites of biosorbent.

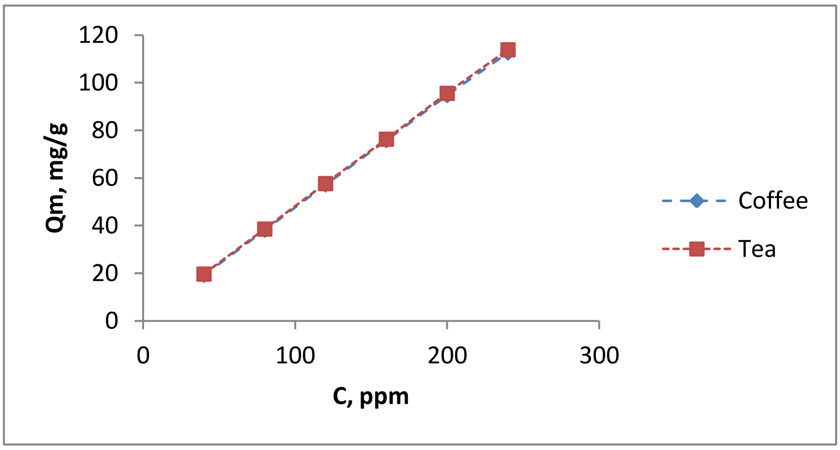

Fig.2. Effect of initial concentration on cobalt uptake Q_m onto both absorbents

2.3 Effect of contact time

The rate of biosorption is an important for designing batch biosorption experiments. Therefore, the effect of contact time of cobalt adsorption was investigated as shown in Figure 3. The adsorption of Co(II) ions increased considerably until the contact time reached 40 min at 25°C**.** Further increase in contact time did not enhance the adsorption, so, the optimum contact time was selected as 40 min for further adsorption experiments.

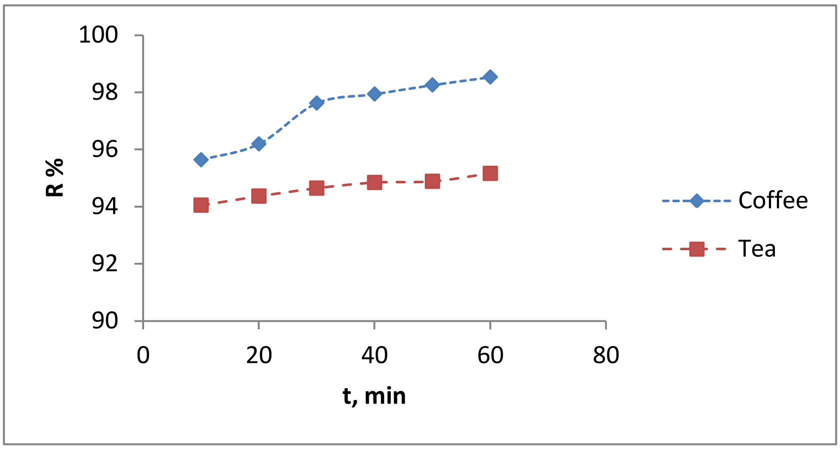

Fig.3. Effect of contact time on percent removal of Co (II) (% R) onto both absorbents

2.4 Effect of adsorbent dose on adsorption

The adsorbent dosage of coffee and tea powders is an important parameter because this determines the suitable capacity of an adsorbent for a given metal concentration. The adsorption efficiency for Co(II) ions as a function of adsorbent dosage was investigated Figure 4. The percentage of the metal adsorption increases with the adsorbent loading up to 1 g/L. However, the metal uptake decreases with adsorbent dose. This result can be explained by the fact that the adsorption sites in remain unsaturated during the adsorption reaction whereas the number of sites available for adsorption site increases by increasing the dose of adsorbent. The further experiments were taken as 1 g/L because the maximum percentage of the metal biosorption was attained at 1.0 g/L of coffee and tea powders dosage as in Figure 4.

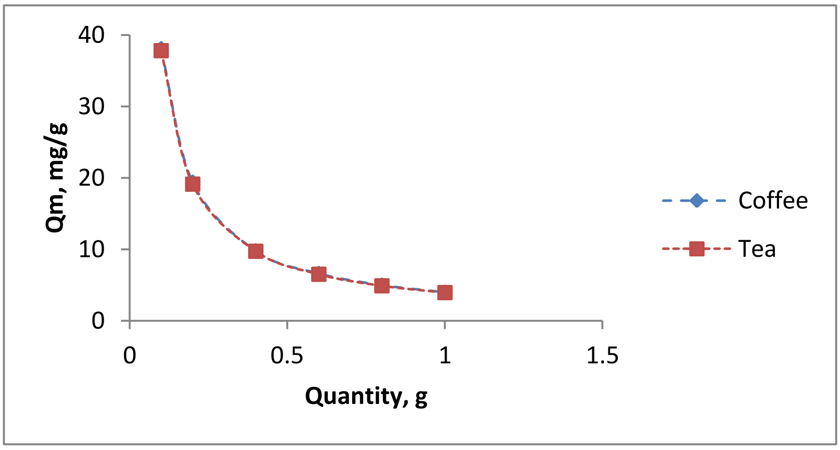

Fig.4. Effect of contact time on cobalt uptake Q_m onto both absorbents

2.5 Adsorption Isotherms

An adsorption isotherms characterized the portion of sorbate molecules that are partitioned between liquid and solid phases at equilibrium**.** Adsorption of Co(II) ions onto tea and coffee powders was modeled using four adsorption isotherms.

The Langmuir isotherm supposes a monolayer adsorption on a uniform surface with a finite quantity of adsorption sites. Once a site is become full, no further adsorption can happen at that site. As such the surface will finally attain a saturation point where the highest adsorption of the surface will be achieved. The linear form of the Langmuir isotherm model is represented as [22]:

$$\frac{1}{Q_e}=\frac{1}{Q_m}+\frac{1}{b\,Q_mC_e} \qquad (3)$$

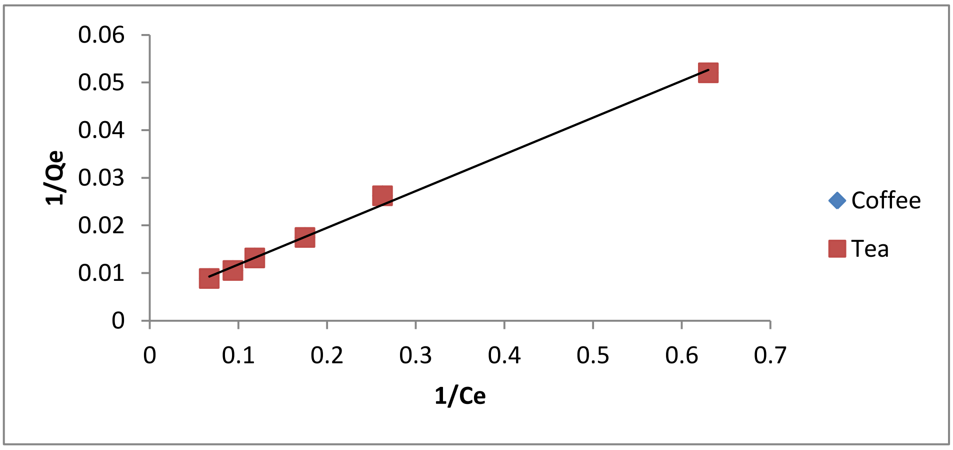

Fig.5. Langmuir adsorption isotherm for the removal of Co(II) by adsorption on coffee and tea powders

where b is the Langmuir constant related to the energy of adsorption (L.mg^{-1}) and Q_m is the maximum biosorption capacity (the monolayer adsorption saturation capacity mg/g), Q_e (mg/g) the amount of metal ion bound per gram of the adsorbent at equilibrium; and C_e, the residual (equilibrium) metal ion concentration left in the solution after binding. Values of Langmuir parameters Q_m and b were calculated from the slope and intercept of the linear plot of $1/Q_e$ versus $1/C_e$ as shown in Figure 5**.** Values of Q_m, b and regression coefficient R^2 are listed in Table 1. These values indicated that Langmuir model describes the adsorption phenomena favorable. The level of conformity for both adsorbents is identical according to the correlation coefficients (R^2). The essential characteristics of the Langmuir isotherm parameters can be used to predict the affinity between the sorbate and sorbent using separation factor or dimensionless equilibrium parameter, R_L expressed as in the following equation:

$$R_L = \frac{1}{1 + b\,C_0} \qquad (4)$$

The value of R_L indicated the type of Langmuir isotherm to be irreversible (R_L=0), favorable (0 <R_L< 1), linear (R_L=1) or unfavorable (R_L>1). In concentration range of 40 to 240 mg/L of Co ions, the values R_L of adsorbed were found to be 0.20 for both adsorbents. They are in the range of 0.0-1.0 which indicates the favorable adsorption.

The Freundlich isotherm model is the well-known earliest relationship describing the adsorption process. This model applies to adsorption on heterogeneous surfaces with the interaction between adsorbed molecules and the application of Freundlich equation. Also suggests that sorption energy exponentially decreases on completion of the sorption centers of an adsorbent. This isotherm is an empirical equation and can be employed to describe heterogeneous systems and is expressed as follows in linear form[23]:

$$\text{Log } Q_e = \text{Log } K_f + \frac{1}{n}\,\text{Log} C_e \qquad (5)$$

where K_f is the Freundlich constant related to the bonding energy. 1/n is the heterogeneity factor and n (g/L) is a measure of the deviation from linearity of adsorption. Freundlich equilibrium constants were determined from the plot of log Q_e versus log C_e, Figure 6 on the basis of the linear of Freundlich equation. The n value indicates the degree of non-linearity between solution concentration and adsorption as follows: if $n = 1$, then adsorption is linear; if $n < 1$, then adsorption is a chemical process; if $n > 1$, then adsorption is a physical process. The n value in Freundlich equation was found to be 1.23 and 1.66 for coffee and tea respectively (Table 1). Since n lie between 1 and 10, this indicates the physical adsorption of Co ions onto tea and coffee is suitable.

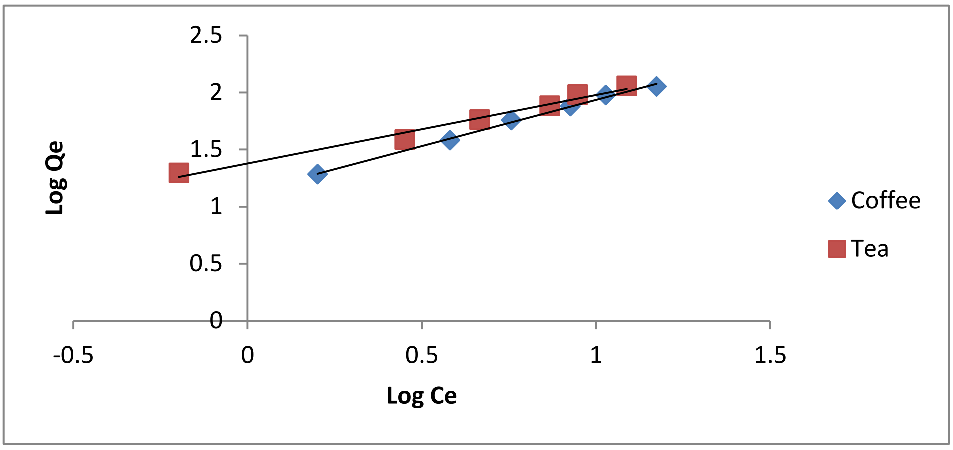

Fig.6. Freundlich adsorption isotherm for the removal of Co(II) by adsorption on coffee and tea powders

Temkin isotherm equation [24] assumes that the heat of adsorption of all the molecules in the layer decreases linearly with coverage due to adsorbent-adsorbate interactions and that the adsorption is characterized by a uniform distribution of the binding energies up to some maximum binding energy. The Temkin isotherm has been used in the linear form as follows:

$$Q_e = B \text{ Log } A + B \text{ Log } C_e \qquad (6)$$

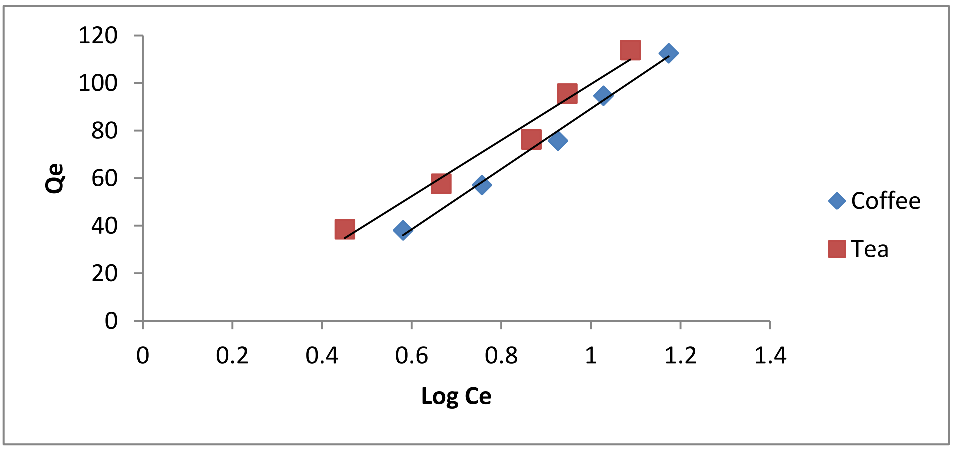

Fig.7. Temkin adsorption isotherm for the removal of Co(II) by adsorption on coffee and tea powders

The plot of Q_e versus log C_e enables the determination of the isotherm constants B and A obtained from the slope and the intercept, Figure 7. Where A (L/mg) is the equilibrium binding constant corresponding to the maximum binding energy and constant B is related to the heat of adsorption, Table 1.

Dubinin-Radushkevich (D-R) model, which does not assume a homogenous surface or a constant adsorption potential as the Langmuir model, was also used to test the experimental data [25].

$$\mathrm{Log}\ Q_e = \mathrm{Log}\ Q_d - \beta\, \varepsilon^2 \qquad (7)$$

Where ε can be correlated to C_e (mg/L) by the following equation:

$$\varepsilon = R\,T\ \mathrm{Log}\left(1 + \frac{1}{C_e}\right) \qquad (8)$$

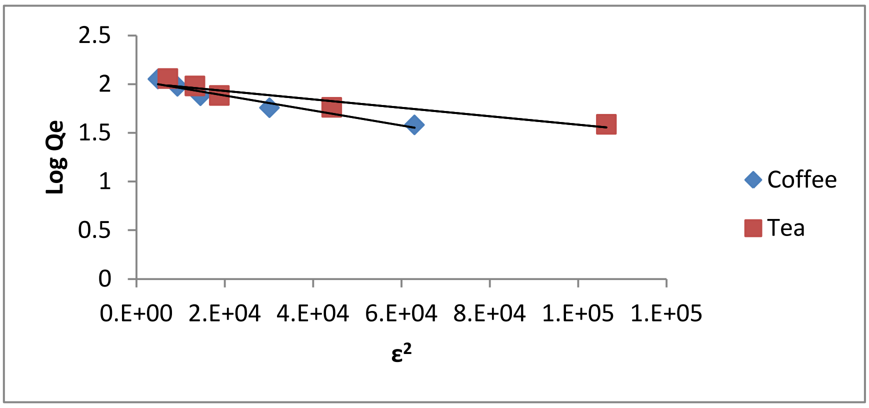

Fig.8. Dubinin-Radushkevich (D-R) adsorption isotherm for the removal of Co(II) by adsorption onto coffee and tea powders

Where R is the universal gas constant (8.314 J/mol K) and T is the absolute temperature (K) . The D-R isotherm parameters β and Q_d were obtained from the slope and intercept of the plot of log Q_e versus ε^2, Figure 8. The calculated vales are listed in Table 1. The values of correlation coefficient were lower than that of other three isotherm values. In all cases, the D-R model represents the least fit to experimental data than the other isotherms models.

The mean free energy of adsorption, E defined as the free energy change when 1 mole of ion is transferred to the surface of the solid from infinity in solution and calculated from the β value using the following equation:

$$E = \frac{1}{\sqrt{2\beta}} \qquad (9)$$

If the magnitude of E is between 8 to 16 KJ/mol then the sorption process is supposed to proceed via chemisorption reaction, while for values of E is less than 8 kJ/mol, the sorption process is of physical nature. The value of E calculated from equation (9) for the adsorption of Co ions by tea and coffee powders were 0.25 and 0.35 KJ/mol, respectively. This indicates that the biosorption process is of physical nature.

2.6 Adsorption Kinetics

To examine the adsorption kinetics of metal uptake onto coffee and tea powders, the two kinetic models (pseudo first-order and pseudo second-order) were fit to experimental data. The pseudo-first order equation of Lagergren [26] is generally expressed as follows:

$$\frac{dQ_t}{dt} = k_1 \, (Q_e - Q_t) \qquad (10)$$

Where Q_e and Q_t are the sorption capacities at equilibrium and at time t, respectively and k_1 is the rate constant of pseudo-first order sorption. The integrated form of equation (10) at boundary conditions, from $Q_t = 0$ to Q_t , and $t = 0$ to t; becomes:

$$Log \, (Q_e - Q_t) = Log \, Q_e - k_1 t \qquad (11)$$

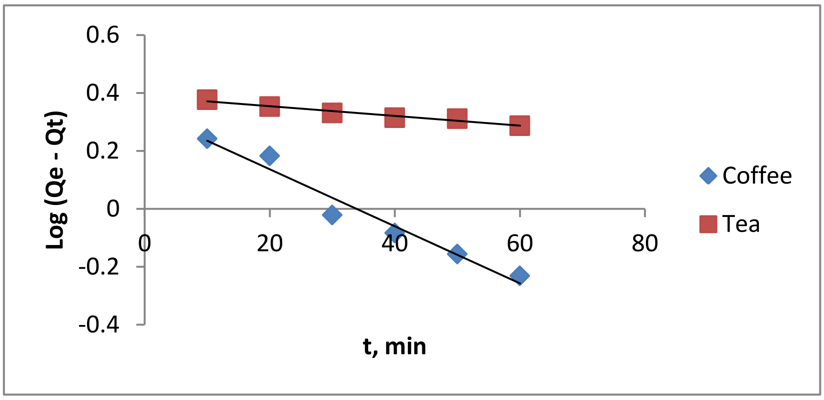

Fig.9. Pseudo first order kinetic plots for the removal of Co(II) by adsorption on coffee and tea powders

In order to fit the experimental data by integrated form of pseudo-first order equation, the equilibrium sorption capacity, Q_e must be known. Furthermore, one has to find some means of extrapolating the experimental data to $t = 0$, on treating Q_e as an adjustable parameter to be determined. For this reason, and for analyze the pseudo-first order model kinetics it is therefore necessary to use trial and error to obtain the equilibrium sorption capacity. The equation applicable to experimental results generally differs from a traditional first order equation in two ways: the terms k_1 and (Q_e – Q_t) does not represent the number of available sites; and the log Q_e is an adjustable parameter which is often not found equal to the intercept of a plot of log (Q_e– Q_t) against *t*, where as in a true first order sorption reaction log Q_e should be equal to the intercept and rate constant can be obtained from the slope Figure 9. In most cases in the literature, the amount sorbed is still significantly smaller than the equilibrium amount and the Lagergren equation does not fit well for the whole range of contact time process. The calculated values and their corresponding linear regression correlation coefficient values are listed in Table 2. R^2 was found to be 0.9607 and 0.9675 which shows that this model cannot be applied to predict the adsorption kinetic model.

The pseudo second-order rate expression, which has been applied for analyzing sorption kinetics rate, is expressed as [27]:

$$\frac{dQ_t}{dt} = k_2\,(Q_e - Q_t) \qquad (12)$$

For the boundary conditions from $Q_t = 0$ to Q_t and t = 0 to t; the integrated form of equation becomes:

$$\frac{t}{Q_t} = \frac{1}{k_2 Q_e^2} + \frac{1}{Q_e} t \qquad (13)$$

Where t is the contact time (min), Q_e and Q_t are the amount of the solute adsorbed at equilibrium and at any time t (mg/g) respectively and k_2 is the rate constant of pseudo-second order sorption, (g/mg.min).

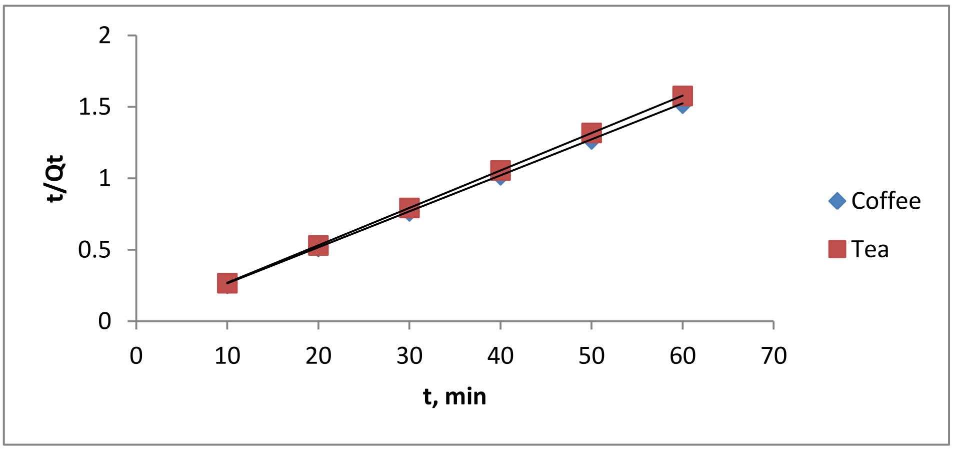

Fig.10. Pseudo-second order kinetic plots for the removal of Co(II) by adsorption on coffee and tea powders

If pseudo-second order kinetics is applicable, the plot of t/Q_t versus t of the equation should give a linear relationship, from which Q_e and k_2 can be determined from the slope and intercept of the plot, Figure 10. The pseudo-second order rate constant k_2, the calculated Q_e value and the corresponding linear regression correlation coefficient value R^2 are given in Table 2. At all initial metal concentrations, the linear regression correlation coefficient R^2 values were high (equal 1.00). The higher values confirm that the adsorption data are better represented by pseudo-second order kinetics and the calculated Q_e values agreed with the estimated experimental Q_e values 38.17 – 37.81 (Table 2). This suggests that the adsorption of Co ions follows pseudo-second order kinetics

Table1. Langmuir, Freundlich, D-R and Temkin isotherm constants for adsorption of Co(II) onto coffee and tea powders

Langmuir	**Q_m**	**b**	**R_L**	**R^2**
Coffee	243.9	0.05	0.20	0.9964
Tea	243.9	0.05	0.20	0.9964
Freundlich	**K_f**	**n**		**R^2**
Coffee	13.31	1.23		0.9962
Tea	23.85	1.66		0.9815
Temkin	**B**	**A**		**R^2**
Coffee	126.88	0.30		0.9920
Tea	117.79	0.16		0.9724
Dubinin-Radushkevich	**Q_d**	**β**		**R^2**
Coffee	108.49	8.0×10^{-6}		0.9421
Tea	104.18	4.0×10^{-6}		0.9040

Table 2. Kinetic parameters of Co(II) adsorption onto coffee and tea powders

First Order	**Q_e**	**k_1(1/min)**	**R^2**
Coffee	2.15	$9.8x10^{-3}$	0.9607
Tea	2.44	$1.7x10^{-3}$	0.9675
Second Order	**Q_e**	**k_2 (g/mg min)**	**R^2**
Coffee	Graphically (39.68) Estimated (38.10)	0.050	1.000
Tea	Graphically (38.17) Estimated (37.81)	0.137	1.000

3. EXPERIMENTAL

3.1 Reagents

All chemicals used were of analytical reagent (AR) grade. 1000 ppm Co (II) stock solution was prepared by dissolving an appropriate amount of $CoCl_2$ salt in deionized water. The stock solution was diluted to the required concentrations using deionized water. The solution pH was adjusted using 0.10 M HCl or 0.10 M NaOH.

3.2 Determination of Co (II)

The concentration of Co(II) ions in the solutions before and after equilibrium was determined by Molecular Absorption Spectrophotometer 6305 from JENWAY. The pH of the solution was measured with pH Meter 3505 from JENWAY. The determination of cobalt (II) as thiocyanate complex was carried out according to the published work [21]. The range of calibration curve concentrations of Co(II) ions prepared from stock solution varies between 20-80 ppm.

3.3 Preparation of Adsorbents

The coffee and tea powders were collected from a coffee shop, washed with double distilled water and then dried in an oven at 70^o C for 24 hrs. The dried materials were sieved through 500 µm size fraction using an American Society for Testing and Materials (ASTM) standard sieve.

3.4 Adsorption Experiments

The adsorption experiments were carried out in a series of 150 mL Erlenmeyer flasks containing 50 ml of metal ions solution, 0.100 g adsorbent powder and if necessary, an appropriate volume of HCl or NaOH solutions was used to adjust the pH of the solution. The solutions were shaken (175 rpm) at 25°C. Then solutions were filtered by Whatman filter paper. The removal percentage (% R) was calculated according to the following equation:

$$\%R = \frac{C_o - C_e}{C_o} \times 100 \qquad (1)$$

Where: C_o and C_e are initial and final concentrations in ppm, respectively. The amount of adsorbed metal Q_e mg/g (mg metal per gram adsorbent) was calculated based on the difference between the initial (C_o, ppm) and final concentration (C_e, ppm) in every flask, as follows:

$$Q_e = \frac{C_o - C_e}{M} \times V \qquad (2)$$

where Q_e is the metal uptake capacity (mg/g), V the volume of the metal solution in the flask (L) and M is the dry mass of biosorbent (g).

3.5 Parameters Affecting Adsorption Experiments

The effects of experimental parameters such as pH, biosorbent dose, contact time, and metal ion concentration were investigated for cobalt adsorption onto coffee and tea powders. The pH dependent study was carried out in 50 ml aqueous solution of 80 ppm Co (II) and 0.10 g powder. The investigated pH values were 2.00 to 6.50. The effect of contact time on Co (II) adsorption was examined for 50 ml solutions with 80 ppm concentration and containing 0.10 g powder. The effect of biosorbent doses on metal uptake was investigated with adsorbent masses of 0.10, 0.20, 0.40, 0.60, 0.80 and 1.0 g per 50 ml of 80 ppm Co (II) solution. Finally, various concentrations of Co (II) were investigated to study the effect of metal ion concentration which were: 40, 80, 120, 160, 200, 240 ppm per 50 ml solution.

4. CONCLUSION

In our present work, we show that the coffee and tea powders could be used as a adsorbent for the removal of Co (II) ions from aqueous solutions. The removal efficiency and metal capacity were found to be dependent on pH, contact time, initial metal ion concentration, and adsorbent dose. adsorption isothermal data could be well simulated by Langmuir, Freundlich, Temkin and then Dubinin-Radushkevich (D-R) models. The maximum metal capacities determined by using the Langmuir isotherm were 243.9mg/g for coffee and tea powders. The values of metal uptake capacities at equilibrium Q_e calculated from equation (2) and pseudo-second-order plot were found comparable .The adsorption kinetics could be well predicted by pseudo-second-order kinetic. The results of our investigation indicate that the coffee and tea powder has a potential for use in removing Co (II) from aqueous solutions.

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