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**SORPTION OF COBALT FROM AQUEOUS SOLUTION
BY WATER HYACINTH ROOTS**

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SORPTION OF COBALT FROM AQUEOUS SOLUTION BY WATER HYACINTH ROOTS

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Abstract

Purpose: In the present study, removal of cobalt (II) from aqueous solutions by using dried roots of water hyacinth has been investigated.

Methodology: The removal of cobalt was examined as a function of initial concentration of cobalt, pH, weight of roots and contact time. Ten different concentrations from $10\mu\text{g.ml}^{-1}$ to $100\mu\text{g.ml}^{-1}$ were used. The obtained results are applied for Langmuir, Freundlich and Timken isotherms.

Findings: The results indicated that the removal of cobalt depend on concentration of cobalt, weight of roots, pH and very rapid. Comparing the regressions and error values of the used isotherms indicated that the removal of cobalt by water hyacinth roots follows a Langmuir isotherm better than Freundlich and Timken isotherms. Langmuir dimensionless constant indicated that the removal of cobalt by water hyacinth roots is favorable.

Unique contribution to theory, practice and policy: This investigation indicated a new material successfully removes cobalt (II) from aqueous solutions. And can be used for removing ^{60}Co from aqueous solutions that might be released into the environment from radioactive waste.

Key words: Cobalt, removal, Water hyacinth roots, Langmuir isotherm

1.0 INTRODUCTION

The transition metal, cobalt has molecular mass of 58.93 u, and atomic weight 27 and belonging to group 8B of the periodic table. The most important cobalt ores are Linneite (Co_3S_4), erythrite ($\text{Co}_3(\text{AsO}_4)_2 \cdot 8\text{H}_2\text{O}$), Safflorite (CoAs_2), and cobaltite (CoAsS). Cobalt has great hardness and being resistant to high temperatures, so it is used in the manufacture of many alloys (iron, nickel, and aluminum), electroplating, surface coatings, in the ceramics industry and chemical industry as a catalyst (Ivanova, *et al*, 2006 & Lee, 1999). These applications produce various liquid and solid wastes and could be a source of contamination for water and soils by cobalt. Pulmonary fibrosis, respiratory sensitization, bronchitis, asthma, and emphysema are diseases resulted from entrance of cobalt into human body (Lombaert, *et al*, 2008). So, removal of cobalt from liquid wastes containing cobalt is very important.

There are several treatment techniques such as precipitation, oxidation/ reduction, ion-exchange, membrane filtration, electrochemical reduction, and adsorption (Keskinan, *et al*, 2003). Adsorption process is highly economical and removes contaminants even at trace levels. The use of this technique in wastewater treatment due its simplicity, low cost of operation and wide end use has been reported before (Naeem *et al*, 2007).

Experimental studies showed that water hyacinth has been successfully used for removing many elements from aqueous solution such as Cd (O'Keeffe, *et al*, 1984, Hasan, *et al*, 2007, Nir, *et al*, 1990, Sadhna, *et al*, 1994, and Lu, *et al*, 2004), Hg (Chattopadhyay, *et al*, 2012, Lenka, *et al*, 1990, Panda, *et al*, 1988, and Tabbada, *et al*, 1990), Zn (Akicin, *et al*, 1993, Hasan, *et al*, 2007 and Lu, *et al*, 2004), Cu (Lee & Hardy, 1987 and Swain, *et al*, 2014), Pb (Akicin, *et al*, 1994, Pereira, *et al*, 2014 and Win, *et al*, 2003), Ni (Hussain, *et al*, 2010 and Turnquist, *et al*, 1990), As (Alvarado, *et al*, 2008, Low and Lee 1990 and Snyder, 2006), Fe (Win, *et al*, 2002), REEs (Chua, 1998) and mixture from ^{137}Cs and ^{60}Co (Saleh, 2012).

Water hyacinth (*Eichhornia crassipes*) used as a scavenger of heavy metal pollutants in the lake Chivero and its associated rivers in Zimbabwe. (Zaranyika & Ndapwadza, 1995). Water hyacinth is a good tool for water monitoring (Ismail, *et al*, 1996), a powerful bio-accumulator for uranium in the River Nile (Aly, *et al*, 2004) and a good tool for monitoring the environmental impact of industrial facilities on the River Nile (Aly, *et al*, 2009). Water hyacinth (*Eichhornia crassipes*) plants acted as an efficient adsorbent for phosphorus and nitrogen from swine waste water (Chen, *et al*, 2010) and being able to grow and clean up extremely polluted environment such as leachates (Akinbile, 2012). Also, water hyacinth was able to remove cyanide from aqueous solution and could be useful in treating cyanide effluents from small scale gold mines (Ebel, *et al*, 2007).

Dead whole water hyacinth plant powder removed Cr (VI) from aqueous solutions (Gude & Das, 2008) and dead water hyacinth whole plant powder (chemically treated) removed of Zn (II), Cd (II), and Cr (VI) from single, bi, tri – metal systems (Saraswat & Rai, 2010) and Methylene blue (El-Khaiary, 2007 & El-Khaiary, *et al*, 2009) from aqueous solutions. Dried water hyacinth roots have ability to remove Cr (III) (Low & Lee, 1997), Cr (VI) (Mohanty *et al*, 2006 and Sarker, *et al*, 2010), Cu (Low *et al*, 1994 and Zheng, *et al*, 2009), As (Al Rmalli *et al*, 2005 and Govindaswamy, *et al* 2011), La (Aly *et al*, 2013), Eu (Kelley, 1999), Th (Aly *et al*, 2014) and U (Bhainsa & D'Souza, 2001, Shawky, *et al*, 2005 and Sadeek, *et al*, 2014) from aqueous solutions. Also, water hyacinth roots (chemically treated) were used in removal of U (Sadeek *et al*, 2014 and Abd El-Mageed, *et al*, 2017), and water hyacinth shoot removed Cr and Cu from aqueous solutions (Sarkar, *et al*, 2017).

Water hyacinth ash removed Ni (Hussain, *et al*, 2010) and Phenol (Uddin, *et al* 2007) from aqueous solutions. Cd (II) was removed from aqueous solutions using activated carbon prepared from water hyacinth (Singh & Verma, 2017). Cu was removed from aqueous solution by cellulose xanthogenate prepared from raw fiber of water hyacinth (Tan, *et al.*, 2007). Magnetic graphene oxide polypyrrole composite synthesized from water hyacinth roots removed Th from aqueous solutions (Gado, 2018). Ethanol extract of water hyacinth roots effective in the removal of As from treated organs rat and may be effective in the removal of As from patients of chronic arsenic poisoning (Quayum, 2007).

No previous published data about removal of cobalt (II) by dried water hyacinth roots, so this work studies the removal of Co (II) from aqueous solution by water hyacinth roots under various experimental conditions such as pH, concentration of Co (II),

contact time, and weight of roots. The obtained data has been analyzed in terms of Langmuir, Freundlich and Timken isotherms. This study is a model for removing ^{60}Co from aqueous solutions that might be released into the environment from radioactive waste.

2.0 METHODOLOGY AND TECHNIQUES

Sample preparation

Water hyacinth roots were collected from the River Nile at Cairo city, Egypt. The roots were cleaned thoroughly to remove dirt and the unhealthy parts. The roots were rinsed in double distilled water for 24 hr then, were boiled for 1 hr using double distilled water to remove any soluble compounds, and were dried at 80 °C for 24 hr using a drying oven. The roots were ground using a gate mortar (Retch) and were passed through a sieve of 1 mm size.

Preparation of cobalt stock solution

A 1000 $\mu\text{g}\cdot\text{ml}^{-1}$ stock cobalt solution was prepared by dissolving analytical grade $\text{CoCl}_2\cdot 6\text{H}_2\text{O}$ in double distilled water and the solution was made up to 1 liter. The pH was adjusted to the desired value using NaOH solution and HNO_3 solution. All the chemicals used were of analytical reagent (AR) grade.

Instrumentation

The pH of the solutions was measured using a digital pH meter model HI 8519 (Hanna). The shaking was carried out in a thermostated shaker bath, GFL.1083 model. ICP-AES was used to measure cobalt in the solution.

Removal experiments

The batch technique was used to study the removal of cobalt on the roots in this study. The used method for this study is described below. The roots were equilibrated with the metal solution of known concentration in a plastic vial at 25 °C in a thermostated shaker water bath for a known period of time. The roots were filtered, and then the residual cobalt in the filtrate was determined using ICP-AES. Controls were simultaneously carried out to ensure that the removal was by water hyacinth roots and not on the wall of the container.

Removal yield (%) was calculated by:

$$\text{Removal yield (\%)} = \frac{C_{O_i} - C_{O_e}}{C_{O_i}} \times 100 \quad (1)$$

Where, C_{O_i} and C_{O_e} are the concentrations of cobalt in the initial and equilibrium solution, respectively.

3.0 RESULTS AND DISCUSSIONS

Cobalt concentration, pH of the solution, shaking time and weight of water hyacinth roots were studied to investigate the removal process of cobalt from aqueous systems by water hyacinth roots.

Effect of pH of solution

In this study the removal of cobalt by water hyacinth roots was investigated in the pH range from 2 to 8 because cobalt precipitated at $\text{pH} > 8$ as a result of formation of a cobalt hydroxide precipitate (Demirbas, 2003). According to our pervious study on the prepared water hyacinth roots, the roots have isoelectric point at 7.53 (Aly, *et al*, 2013). So, the roots surface has a positive charge in the pH range 2-7 and has a negative charge at pH8.

Changing the pH of the solution will change; 1- The degree of protonation or deprotonation of the functional groups of the roots such as carboxylates which will affect the removal process (El-Khaiary, 2007 & El-Khaiary, *et al*, 2009). 2- The types and ratios of cobalt chemical species such as Co^{+2} and $\text{Co}(\text{OH})^+$ which may have different reactivity towards the roots (Kim & Lee 2001, Netzer & Hughes 1984 and Rengaraj & Moon 2002). All of these factors effect on the removal of cobalt by water hyacinth roots.

Fig 1 shows the effect of pH on the removal of cobalt by water hyacinth roots. The effect of pH on the removal of cobalt was investigated in the range of 2-8. 0.1 g of roots were equilibrated in 20 ml of $50 \mu\text{g}\cdot\text{ml}^{-1}$ cobalt solution and shaken for 90 min at 25 °C. This illustrates that the amount of removed cobalt increased until pH 8, the results of the experiments showed that the removed cobalt depends on the pH of the solution. The highest cobalt removal yield (86.9 %) was achieved at pH 8.0.

The effect of pH on removal of cobalt indicates three stages of increment. Elevated increment by changing the pH of solution from 2 to 3 the removal increases from 10.8 % to 70.22 % respectively, i.e. the removal becomes 6.5 times from its value at pH 2. The second stage from pH 3 to pH 5 the removal increases from 70.22 % to 82.96 % respectively, i.e. the removal becomes 1.18 times from its value at pH 3. And in this stage the increase in the removal is constant (6.3%) by increasing the pH of the solution from 3 to 4 and from 4 to 5. The last stage from pH 5 to pH 8 the removal increased very slowly from 82.96% at pH 5 to 86.9% at pH 8.

At pH 2 the removal of cobalt is low (10.8 %) as a result of competition of H^+ ions with the cobalt ions. Increasing the pH (2-8) of the solution enhance removal of cobalt. That may be attributed to increasing pH of the solution enhance the negative charge groups of the water hyacinth roots then the chemical species of cobalt such as Co^{+2} and $\text{Co}(\text{OH})^+$ can be easily removed by the water hyacinth roots (Netzer & Hughes, 1984; Rengaraj & Moon, 2002; Kim & Lee, 2001).

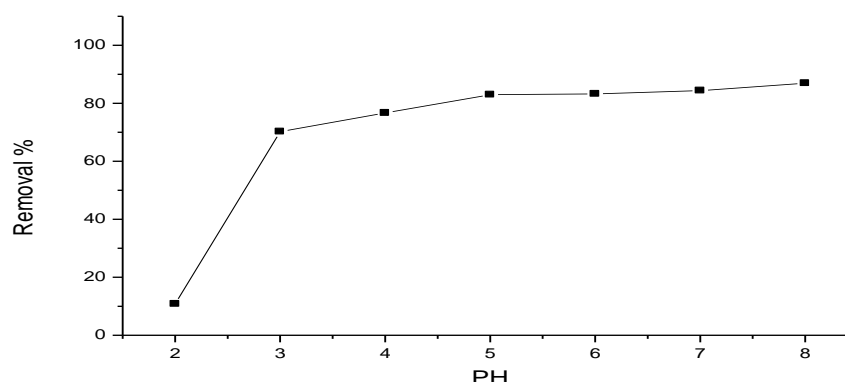


Figure 1: Removal of cobalt on roots as a function of pH

(pH 2-8, V= 20 mL, Co = 50 μ g mL⁻¹, t = 90 min, wt = 0.1 g, and T = 25^oC)

Effect of the weight of water hyacinth roots

Figure 2 shows the effect of weight of water hyacinth roots on the removal of cobalt by water hyacinth roots. The effect of weight of water hyacinth roots on the removal of cobalt was investigated in the range of 0.02 - 0.1 g of roots. Roots were equilibrated in 20 ml of 50 μ g.ml⁻¹ cobalt solution at pH 6 and shaken for 90 min at 25 $^{\circ}$ C. The removal of cobalt increases from 52.6% to 86.9% by increasing the weight of the roots from 0.02 to 0.1 g, due to the greater availability of the removal sites at the roots.

Comparing this study with our previous studies indicated that 0.1 gm of roots removed 86.5 % only from cobalt in spite of the solution concentration is 50 μ g.ml⁻¹, while in our previous studies 0.07 gm removed 95.5 %, 0.03 gm removed 99%, and 0.05gm removed 98% from 100 μ g.ml⁻¹ solutions of La (Aly et al, 2013), Th (Aly, *et al*, 2014), and U (Shawky, *et al*, 2005) respectively. But this study has the highest quantity of removed moles of Co (14.7 μ mole) compared to La (13.7 μ mole), Th (8.23 μ mole), and U (8.5 μ mole).

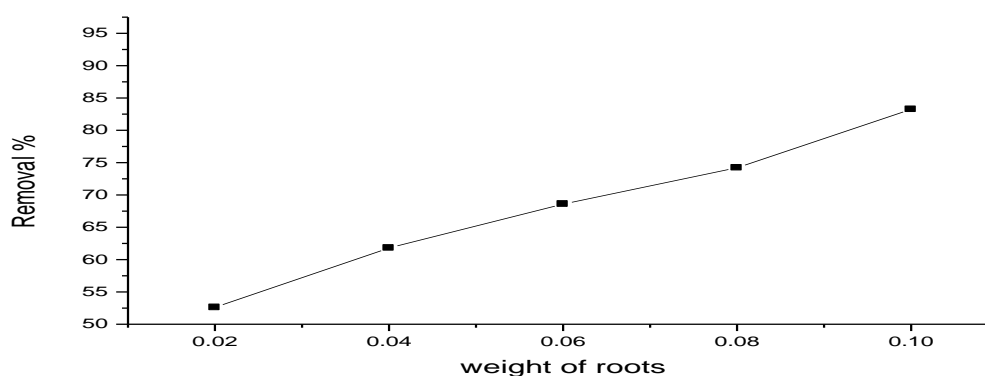


Figure 2: Removal of cobalt as a function of the weight of the roots

(wt. = (0.02-0.1) g, V= 20 mL, Co = 50 μ g mL⁻¹, t = 90 min, pH= 6, and T = 25^oC)

Effect of the initial cobalt concentration

The effect of the cobalt concentration on the removal of cobalt in the range of 10-100 $\mu\text{g}\cdot\text{ml}^{-1}$ was investigated. 0.1 g of dried roots were equilibrated in 20 ml of cobalt solution and shaken for 90 min at 25 °C.

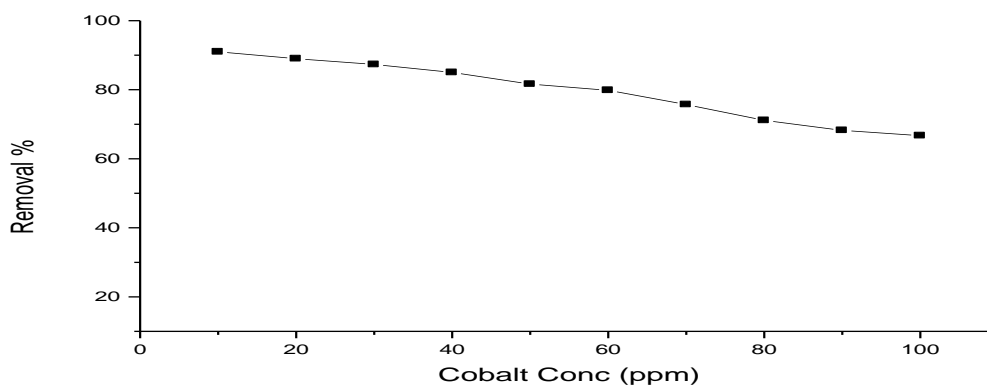


Figure 3: Removal of cobalt on roots as a function of initial cobalt concentration

(wt. = 0.1 g, V= 20 mL, $C_0 = 10\text{-}100 \mu\text{g mL}^{-1}$, $t = 90 \text{ min}$, $\text{pH} = 6$, and $T = 25^\circ\text{C}$)

Figure 3 shows the removal yield at different cobalt concentrations. The obtained results indicate a slight influence of initial cobalt concentration on the removal over the investigated range (10-100 $\mu\text{g}\cdot\text{ml}^{-1}$). The removal reached a maximum of 91% at the lowest cobalt concentration (10 $\mu\text{g}\cdot\text{ml}^{-1}$) i.e., the removal is more efficient from dilute solutions. However, the quantity of the studied element increases by increasing the initial concentration as shown in figure 4. The removed amount of cobalt increased from 182 μg to 1334 μg by increasing the initial cobalt concentration from 10 $\mu\text{g}\cdot\text{ml}^{-1}$ into 100 $\mu\text{g}\cdot\text{ml}^{-1}$ respectively.

For comparison removal of U (Bhainsa & D'Souza, 2001 and Shawky, *et al* 2005), Th (Aly, *et al*, 2014), and La (Aly, *et al*, 2013) is more efficient from diluted solution by water hyacinth dried roots. On the other hand, removal of Cr by water hyacinth (whole plant) increases as the concentration of Cr increases (Mohanty, *et al*, 2006).

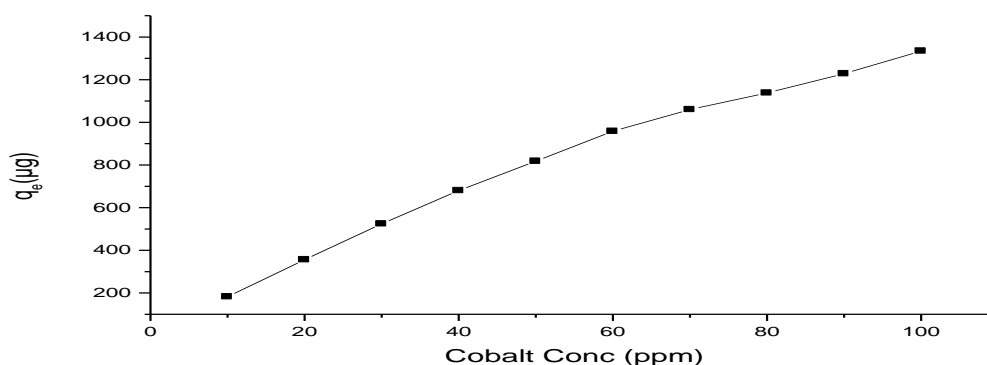


Figure 4: Quantity (μg) of cobalt removed on water hyacinth roots as a function of initial cobalt concentration.

($C_0 = 10\text{-}100 \mu\text{g mL}^{-1}$, $\text{pH} = 6$, $V = 20 \text{ mL}$, $t = 90 \text{ min}$, $\text{wt.} = 0.1 \text{ g}$, and $T = 25^\circ\text{C}$)

Effect of contact time

The effect of contact time on the removal of cobalt in the range of 5-90 min was investigated. 0.1 g of dried roots were equilibrated in 20 ml of cobalt solution and shaken for 5-90 min at 25°C .

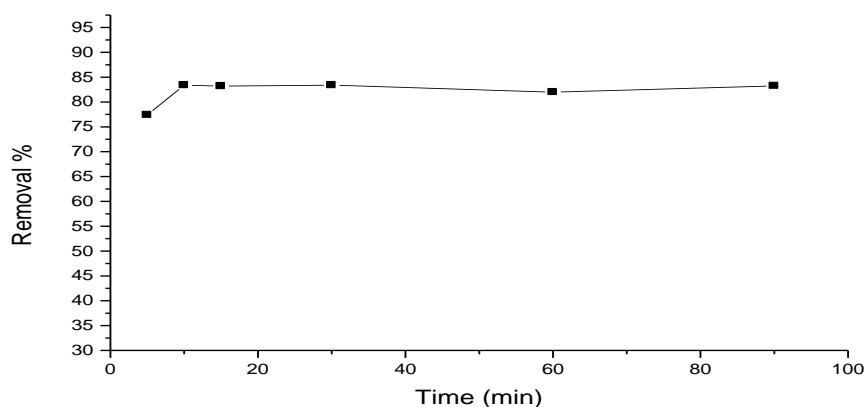


Figure 5: Removal of cobalt as a function of contact time

($C_0 = 50 \mu\text{g mL}^{-1}$, $\text{pH} = 6$, $V = 20 \text{ mL}$, $t = 5\text{-}90 \text{ min}$, $\text{wt.} = 0.1 \text{ g}$, and $T = 25^\circ\text{C}$)

Figure 5 shows the removal yield as a function of contact time. The removal of cobalt from solution is very rapid. It almost reached a maximum after 10 minutes, and almost remained constant up to 90 minutes. Our previous studies indicated the same trend where, 10, 5, and 5 minutes are required to La (Aly, *et al*, 2013), Th (Aly, *et al*, 2014) and U (Shawky, *et al*, 2005) to reach equilibrium with water hyacinth roots respectively.

Sorption isotherms

Adsorption isotherms are important for the description of how molecules of adsorbate interact with adsorbent surface (Lorenc-Grabowska & Gryglewicz, 2007) and also, are critical in optimizing the use of water hyacinth as an adsorbent. Hence the correlation of equilibrium data using both theoretical and empirical equations is essential for the adsorbent interpretation and prediction of the extent of adsorbent.

The most famous isotherm relationship is Langmuir. The Langmuir model was originally developed to describe the removal of gases, the equation may be written in the following form for solid-liquid system:

$$C_{0eq}/y = \left(\frac{1}{a}\right) + \left(\frac{b}{a}\right) \cdot C_{0eq} \quad (2)$$

Where C_{0eq} is the concentration of solute in the solution phase at equilibrium (mg L^{-1}), y is the amount of solute removed per unit weight of sorbent (mg g^{-1}), b and a are

Langmuir constants; b is represented as the ratio of sorption and desorption rate coefficient ($L g^{-1}$), and

$$a = b \cdot Q_{ms} \quad (3)$$

Q_{ms} is the maximum removal capacity ($mg g^{-1}$). At low concentrations the Langmuir equation reduces to a linear relationship, while the maximum removal capacity, Q_{ms} is attained at concentrations corresponding to monolayer coverage. Langmuir data for cobalt is shown in Figure 5.

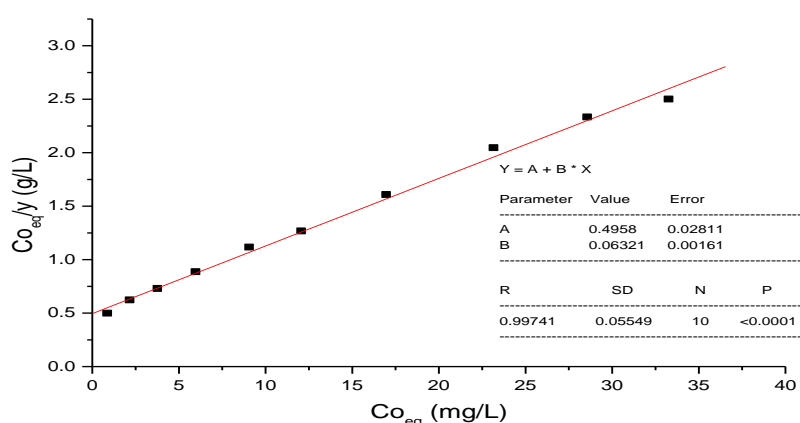


Figure 6: Langmuir removal isotherm $b = 0.13 L g^{-1} L g^{-1}$, $Q_{ms} = 15.8 mg g^{-1}$ and $R^2 = 0.997$

($C_0 = (10-100) \mu g mL^{-1}$, $pH = 6$, $V = 20 mL$, $t = 90 min$, $wt. = 0.1 g$, and $T = 25^\circ C$)

The data were expressed as the amount of cobalt removed per unit weight of sorbent (C_{oeq} / y) versus the cobalt concentration in the solution at equilibrium (C_{oeq}). These data conformed to the Langmuir equation (Equation 2) where the values of b and Q_{ms} were calculated to be $0.13 L g^{-1}$ and $15.8 mg g^{-1}$ respectively. Also, removal of La (Aly, *et al*, 2013), Th (Aly, *et al*, 2014), U (Shawky, *et al*, 2005) and Cu by water hyacinth roots fits Langmuir isotherm (Zheng, *et al*, 2009).

The Langmuir model assumes that the energy of adsorption is the same for all surface sites and doesn't depend on the degree of coverage, indicating monolayer adsorption of the adsorbate, and finite numbers of identical sites. Fitting of Langmuir model is suggesting the occurrence of binding to the biomass with homogenous surface energy with no interaction between the removed metals. Schneider, *et al*, (2001) stated that Langmuir fit may be evidence to ion exchange mechanism.

In order to predict the favorability of the adsorption process, the essential feature of Langmuir model can be expressed in terms of a dimensionless constant factor " R_L " given as following.

$$R_L = \frac{1}{1+bc} \quad (4)$$

Where b is the binding energy constant from Langmuir equation, and C ($mg L^{-1}$) is the initial concentration of the cobalt.

Table 1: the dimensionless constant factor "R_L"

Concentration ($\mu\text{g mL}^{-1}$)	R _L
10	0.43859649
20	0.28089888
30	0.20661157
40	0.16339869
50	0.13513514
60	0.11520737
70	0.10040161
80	0.08896797
90	0.0798722
100	0.07246377

The parameter indicates the type of the isotherm to be irreversible if the R_L is zero, "favorable" if $0 < R_L < 1$, "linear" if R_L=1, or "unfavorable" if R_L>1. The results listed in Table 1 indicate that the removal of cobalt is favorable. Also, our previous studies indicated that the removal of La (Aly, *et al*, 2013), Th (Aly, *et al*, 2014) and U (Shawky, *et al*, 2005) by water hyacinth roots is favorable.

The Freundlich isotherm model assumes heterogeneous surface energies and / or different mechanisms in which energy term in Langmuir equation varies as a function of surface coverage. K and 1/n are Freundlich isotherm constants related to the isotherm constants related to adsorption capacity and intensity of adsorption. It is assumed that the stronger binding sites are occupied first and that the binding strength decreases with the increasing degree of site occupation (Vijayaraghavan, *et al*, 2009). This model describes reversible adsorption and is not restricted to the formation of monolayer.

The logarithmic form of the Freundlich equation is:

$$\text{Log}y = \text{Log}K + \frac{1}{n}\text{Log}C_{o_{eq}} \quad (5)$$

The graphical relation between Log y and Log C_{o_{eq}}, gives a straight line of slope 1/n and the intercept = log K as presented in figure 6.

The results are confirmed to Freundlich equation and the values of K and n were calculated as 2.3 and 1.9 respectively. Fitting to Freundlich model suggests the binding to the roost with different functional groups and / or different mechanisms (El – khaiary, 2007). Also, our previous studies indicated that the removal of La (Aly, et al, 2013), Th (Aly, et al, 2014) and U (Arafat, et al, 2019) by water hyacinth roots fits freundlich isotherm.

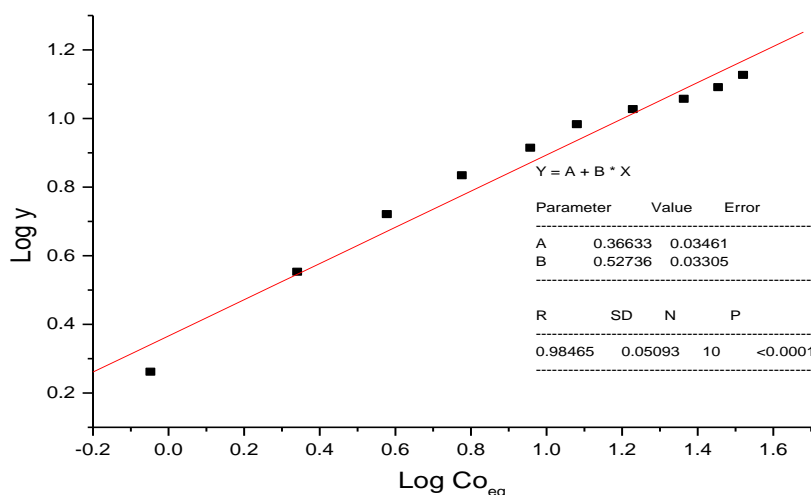


Figure 7: Freundlich removal isotherm $n = 1.9$, $K = 2.3$, and $R^2 = 0.984$

($C_0 = (10-100) \mu\text{g mL}^{-1}$, $\text{pH}=6$, $V= 20 \text{ mL}$, $t=90 \text{ min}$, $\text{wt.} = 0.1 \text{ g}$, and $T = 25^\circ\text{C}$)

Timken isotherm model assumes that the heat of adsorption of all the molecules decreases in linearly rather than in logarithmic scale, as implied in Freundlich isotherm (Bhainsa & D'Souza, 2001). Timken isotherm model is given by the following equations:

$$q_e = RT/b \ln(AC_e) \quad (6)$$

$$q_e = B \ln A + B \ln C_e \quad (7)$$

Where $B = (RT / b)$, R is the universal gas constant ($8.314 \text{ J mol}^{-1} \text{ K}^{-1}$), T is absolute temperature in Kelvin (K), C_e is the residual equilibrium concentration (mg L^{-1}), and q_e is the amount adsorbed per specified amount of adsorbent (mg g^{-1}) at equilibrium. Timken removal data for cobalt was indicated in Figure 8. A and B constants calculated to be 1.53 and 3.24 respectively.

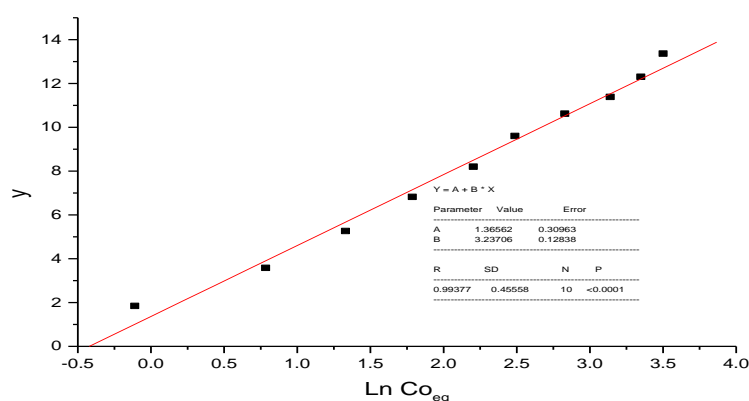


Figure 8: Timken removal isotherm; B =3.24, A= 1.53 (L g-1), and R²= 0.993

(Co = (10-100) µg mL⁻¹, pH=6, V= 20mL, t=90 min, wt. = 0.1 g, and T = 25°C)

Comparing the linear regressions and standard deviations for all the studied isotherms indicated that cobalt is best fitted to Langmuir isotherm. Removal of U (Arafat, *et al*, 2019), La (Aly, *et al*, 2013) and Cr (Sarkar, *et al*, 2010) by water hyacinth roots are best fitted by Langmuir isotherm, while removal of Th (Aly, *et al*, 2014) at pH 1 and pH 5 by water hyacinth roots are best fitted by Langmuir and Freundlich isotherms respectively.

4.0 CONCLUSION

Cobalt was removed successfully from aqueous solutions by water hyacinth roots. The experiments showed that the removal of cobalt from aqueous solution is influenced by some factors such as pH, initial cobalt concentration, contact time and weight of the roots. The amount of cobalt removed increased from 10.8 % to 86.9 % by increasing the pH of the solution from 2 to 8 respectively. The removal of cobalt increases from 52.6% to 86.9% by increasing the weight of the roots from 0.02 to 0.1 g respectively. The removal is more efficient from dilute solutions but the removed amount of cobalt increased from 182 µg to 1334 µg by increasing the initial cobalt concentration from 10 µg.ml⁻¹ into 100 µg.ml⁻¹ respectively. The removal of cobalt by water hyacinth roots is best fitted by Langmuir isotherm. The velocity of removal was fast and need only 10 min for the process to equilibrium. Finally, this study presents a cheap and available biomaterial for the removal of Co and may be used for removal of ⁶⁰Co from radioactive wastes.

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