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Original Article

Equilibrium, kinetics and isotherm studies of Cadmium (II) adsorption from aqueous solution utilizing Cane Papyrus

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ABSTRACT

Cadmium(II) ions is a very toxic element known to cause detrimental effects to human health even at very low concentrations. Cane Papyrus was prepared form the marsh in south of Iraq, as adsorbent to Cadmium(II) ions adsorption from aqueous solution. Completed the analysis by using Fourier transform infra-red and notes presence of amino (–NH), carbonyl (–C=O) and hydroxyl (–OH) functional groups. Batch experiments were performed on simulated aqueous solutions under optimized conditions of adsorbent dosage, contact time, pH and initial Cadmium(II) ions concentration at 25Co. The Freundlich isotherm model more suitably described the adsorption process than the Langmuir model with linearized coefficients of 0.986 and 0.9733, respectively. Pseudo-second order kinetic equation best described the kinetics of the reaction. Furthermore, 1M HCl was a better desorbing agent than 1M NaOH and de-ionized water. The experimental data obtained demonstrated that Cane Papyrus can be used as a suitable adsorbent for Cadmium(II) ions removal from wastewater.

1. Introduction

Persistence, bioaccumulation tendencies and toxicity pollution of heavy metals are the most challenging environmental problems[1]. Many industrial unites are discharge the effluent that containing waste of heavy metals to the rivers, so that causes on the water quality affect by increase the metals concentrations and the water bodies aesthetic[2]. Most activities like melting processes , ore mining ,treatment units of wastewater, different agricultural tasks and castings of metals unit, all these can effects a lot on the increasing heavy metals in environment[3]. At the developing countries , Cadmium(II) ions consider the first causes for the children disabilities and also causes deaths for many people every year[4][5].

Many technological processes have been used to treat contaminated water for disposal of heavy metal ions. The techniques that have been tested are chemical precipitation[6], coagulation[7][8], activated carbon[9], ion exchange[10][11], filtration and membrane[12][13] these techniques have advantages and disadvantages. The disadvantages of these technologies is that it is low efficiency and high cost and it leaves behind large waste difficult to get rid of [14].

The researchers looking for another approach to rid out the heavy metals from aqueous solution. one of these methods, adsorption[15] which is utilizing the materials available in the nature, low cost materials and give the high efficiency to eliminated the heavy metals especially the toxic and hazardous metals ions from the industrials wastewater[15]. There are many materials tested as an adsorbent like naturally plant abundant[16], microbes[17], algae[18], fruit shell[19] and biomass wastes[20]. Most of these materials have high surface area (capacity of sorption)such as bio-sorption, it has the ability to sequester the ions of metal from wastewater [21]. The advantages of adsorption processes are available in the nature, environmentally friendly and very cheap. The disadvantage

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of this technique is the efficiency for some metal ions low, but the researchers continue to developed it to depend on it instead of the traditional processes[22]. Below some adsorbents that used to recovery the Cadmium ions from aqueous solution by modified , it's plant origin such as: olive stone [23], bagasse fly ash-a sugar industry waste [24], rice hush ash[25], grape stalk waste[26], rice husk and modified rice husk [27], agricultural waste rush polish[28], organo-ceramic[24], low cost bio-polymeric [29], Bacteria-Modified Red Mud [30], akaganeite-type nano-crystals[31] and natural and oxidized corncob[32].

The aim of this study is using Cane Papyrus for the recovery the Cd^{+2} ions from the industrial wastewater and aqueous solution samples as a novel, low cost, environmentally friendly and potential adsorbent.

2. Materials and Methods

2.1. Adsorbent Preparation

Cane Papyrus was composed from farmlands in the marshes of Messan, south of Iraq. The grass are cautiously separate the plant stem then, washing by distill water to take out mud, rubbish and dust particles and then drying by sun shine at least l0 days. grinding the dry Cane Papyrus by hammer mill then take the powder to weighed it. Utilizing the analytical sieves for fractionated the powder(Alatabe,2012a). Fine powder in size between l0 to 200 µm using to samples of wastewater. The adsorbent show a feathery and greatly permeable and coarse microstructure contain cracks and a few voids which is appropriate for Cd^{+2} ions adsorption. By Atomic Absorption Spectrometer (AAS) analyzing the aqueous solution samples to determine the Cd^{+2} ions concentration. For agitation time using adjustable speed mechanical shaker, to pH measurements using pH meter. Analyzed the chemical composition of Cane Papyrus powder by X-ray Fluorescence spectrometer. The results of chemical composition is obtainable in Table l.

From (Table 2) the adsorbent surface area is l.96 m^2 /gm, that's equivalent to 2 m^2 /gm for the ash of waste rubber utilize to Cadmium ions recovery from aqueous solutions. The normal Cane Papyrus diameter in the range between $l0 < d < 1000 \times l0^{-8}$ cm also the cane papyrus can classify same as a meso-porous substance. Also, BET surface areas of activated carbon prepared from macademia nuts utilize to phenol recovery as well as maize tassels that used to heavy metal recovery from aqueous solution are l.083 m^2 /g and 2.52 m^2 /g respectively.

Table 1. Chemical Composition of Cane Papyrus.

Compound	Wt %	Compound	Wt %
Na_2O	l.l7	K_2O	ll.7
MgO	7.10	CaO	24.1
Al_2O_3	5.90	SiO_2	23.6
TiO_2	l.33	P_2O_5	2.42
MnO	0.778	Fe_2O_3	l2.8
SO_3	5.68	SrO	l.24
Cl	l.09	BaO	0.52l

Table 2. Characteristics of Cane Papyrus.

Physical Parameters	Obtaining Results
Surface area of Cane papyrus (m^2 /gm)	l.96
Micro pore surface area (m^2 /gm)	l.75
Overall volume (cm^3/gm)	0.01
Micro pores volume (cm^3/gm)	0.0095
Normal diameter($l0^{-8}$ cm)	500

For Cd^{+2} ions adsorption utilize fin powdered of Cane Papyrus in (1 gm) as an adsorbent surface by putting it in speed of 200 rpm of mechanical shaker at 25C°, studying the Cane Papyrus dosage by initial concentration changes of adsorbent dosage among 0.5 to 2.0 gm, for subsequent procedure uses optimum dosage that obtained. Also pH change effects, Cadmium(II) ions primary concentration and changing the time of equilibration from 2 to l2 and from l to 50 mg/l and from 5 to l20 min correspondingly. The Cd^{+2} ions removal percentage from wastewater predictable from the Equation (l):

$$\text{Adsorption}(\%) = [Ci - Cf]/Ci \times 100 \qquad (1)$$

where("C_i " is initial concentration and "C_f " is the final concentrations) for Cd^{+2} ions.

$$qe = (Co - Ce)V/w \qquad (2)$$

when "qe" in mg/g the Cane Papyrus quantity, "C_e " is the Cd^{+2} ions equilibrium concentrations in aqueous phase. Solution volume is "V" in liters(l) and Cane Papyrus weight is "W" in grams (gm).

2.2. Desorption Testing

Adding the Cane Papyrus in weight(1 gm) into a plastic beaker contain Cd^{+2}ions at l0 mg/l initial concentration(C_i), the equilibration appear after 60 min, then the Cane Papyrus recovery, washing the Cane Papyrus surface Three times with distill water to rid out from all the Cd^{+2} ions remaining. Testing the agents like 1M NaOH, 1M HCl and demineralized water as desorbing potential.

Adding one of the agents in (50 ml) volume to the recovered Cane Papyrus that putting in plastic beaker , then waiting the solution about 60 min for equilibrate, then the mixture filtrated the aqueous solutions, after desorption analyzed the supernatant to determine Cd^{+2} ions concentration(final concentration C_f).

3. Results and Discussion

3.1. Spectroscopy (Infra-Red)Analyzes

Figure 1. represent the functional groups for Cane Papyrus by analyzed in the Fourier transform(infra-red), a ultra-peak in high frequency transmittance in 3000cm-1, that impute to one of these groups (-NH) or (-OH). The figure show the (C-H) groups may be at value (2550cm$^{-1)}$because of the aliphatic saturation compounds make vibrations stretching, whilst the primary amines groups(-NH) make winding vibrations at the value (1750cm^{-1}). The aromatic rings groups(C-C) formed vibrations stretching at the value (1350cm^{-1}) in a tiny peak. The carboxylic acids, ethers or esters and alcohols groups(C-O) stretch around the values from (1150cm^{-1}) to (1200cm^{-1}) peaks. The alkyl halide groups operated absorption at the value (500cm^{-1}). Many of the natural plants have an acidic functional groups in their biochemical characteristics. The acidic functional groups presence in the Cane Papyrus excess the Cd^{+2} ions adsorption.

3.2. pH Changes Effect

Generally, the changes of solution pH were effect on the metal ions adsorption, due to the activity of protons and ions. Figure 2 represented the pH changes effect on the Cd^{+2}ions adsorption by Cane papyrus as adsorbent material, test results were showing that the solution pH changing affected slightly on the Cd^{+2} ions adsorption. When (pH=4) the Cd^{+2}ions uptake 98.8% were increases slightly, then pH > 4 decreases slightly. When the solution pH between 5 to 6.5, the adsorption relatively percentage was constant between 98.8% to 99%, but at pH = 7 the adsorption efficiency decreasing to less than 95% because of the Cd^{+2} started precipitate[3].

3.3. Contact Time Changes Effect

The contact time is one of the important parameters that effect on the Cd^{+2} ions adsorption. In the beginning time of adsorption, especially in the first five minutes that seen at equilibration state the adsorption efficiency is very rapid, reach to 85% figure 3. The cadmium(II) ions rapidly transfer to Cane papyrus surface in the empty voids that lead to a fast adsorption of this ions. Then, the equilibrium state started until (60 min)the adsorption of the ions is slow, at this state the Cd^{+2} ions adsorption efficiency is 98.5%. After this time to 90min the adsorption reach to the equilibrium state completely and the change in the adsorption efficiency is neglected because of all the voids in the Cane Papyrus filled with Cd^{+2} ions.

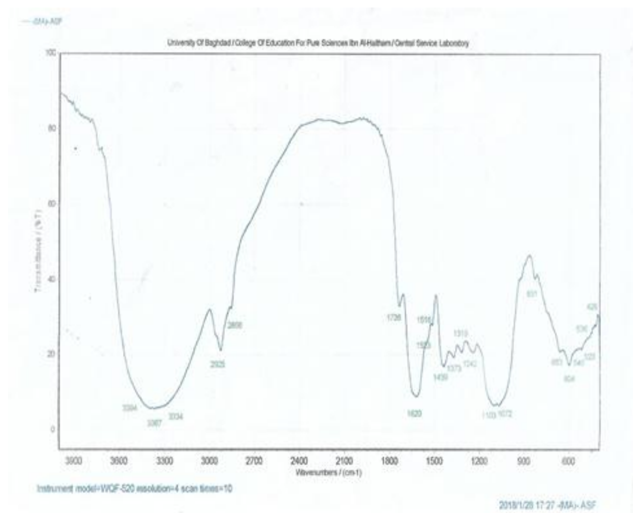

Fig. FTIR for Cane Papyrus.

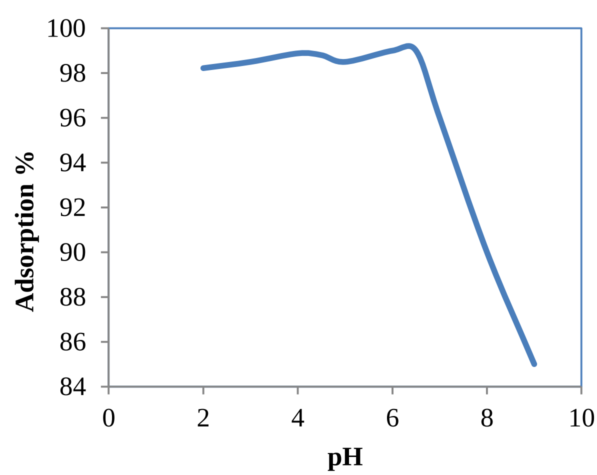

Fig 2. pH changes effect on Cd^{+2} ions adsorption efficiency onto Cane Papyrus.

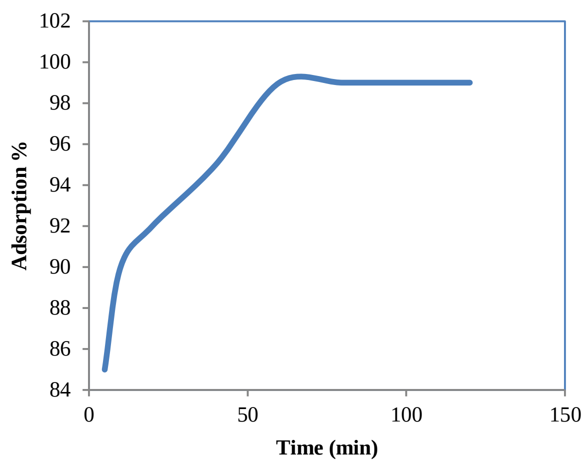

Fig 3. Efficiency adsorption of Cd^{+2} ions with contact time onto Cane Papyrus.

3.4. Effect of Adsorbent Dosage

Effecting dosage of Cane Papyrus on the Cd^{+2} ions adsorption can resolute with change the Cane Papyrus dosage from 10 to 200gm. The percentage removal of Cd^{+2} ions by onto Cane Papyrus adsorbent bigger stridently starting 30% on Cane Papyrus 10gm to 95%on 100gm, however on 200gm the adsorption efficiency decreases near 85% . Figure 4 represented the changes, at the beginning a large surface area of cane papyrus available causes very easy adsorption of ions, then equilibrium state the adsorption efficiency decreases.

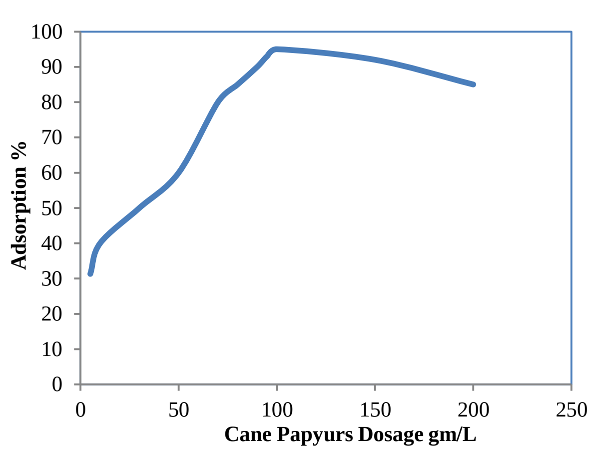

Fig 4. Effect of Cane Papyrus Dosage on the adsorption efficiency of Cd^{+2} ions.

3.5. Adsorption Isotherms

The testing data of Cd^{+2} ions adsorption by Cane Papyrus next to the primary concentration, notice that the adsorption is match with the Freundlich and Langmuir model adsorption isotherms[33]. The Langmuir model [25] "assumes that the ideal gas adsorption onto an ideal surface occurs only at fixed number of sites and every site can only hold one adsorbent molecule (monolayer)". In addition "assumes that all available sites are equivalent and there is no interaction between adsorbed molecules on adjacent sites". The Langmuir model equation is linearized as showing in the following expression (3).

$$\frac{l}{qe} = \frac{l}{qmax} + \{\frac{l}{bqmax}\}\frac{l}{Ce} \qquad (3)$$

when "Ce" in (mg/l) is the Cd^{+2} ions equilibrium concentration, "q_e" in (mg/g) is the Cd^{+2} ions quantity adsorbed at equilibrium, "q_{max}" in(mg/g) is the maximum amount adsorbed and "b" in (l/mg) is the adsorption constant. (Figure 5) is the plot of "l/q_e" next to "l/Ce" get a straight line with a regression coefficient of 0.9733, that's indicate to Langmuir model adsorption conforms. Adsorption efficiency (capacity) and Cd^{+2} ions adsorbed(maximum concentration) determined from the plot intercept and slope, the values in table(3) arranged. By equation(4),these values was calculated for Langmuir model adsorption conforms:

$$Rl = \frac{l}{(1+bCo)} \qquad (4)$$

where, "R_l": separation factor, "Co": initial metal concentration in(mg/l) and "b": Langmuir constant in (l/mg).
when, $R_l > 1$ indicates to an unfavorable monolayer adsorption process,
$R_l = 1$ linear,
$0 < R_l < 1$ favorable and $R_l = 0$ irreversible.
From this study obtained a result that "R_l" values between (zero and one), that's mean adsorption process of Cd^{+2} ions onto Cane Papyrus is a favorable.

Table 3. Freundlich and Langmuir constants for adsorption of Cd^{+2}ions on Cane Papyrus.

Langmuir model	**q_{max} mg/gm**	**b**	**R_l l/mg**	**R^2**
	45.5	0.213	0.0-1.0	0,9733
Freundlich model	K_f	n	R^2	
	0.2	0.091	0.9869	

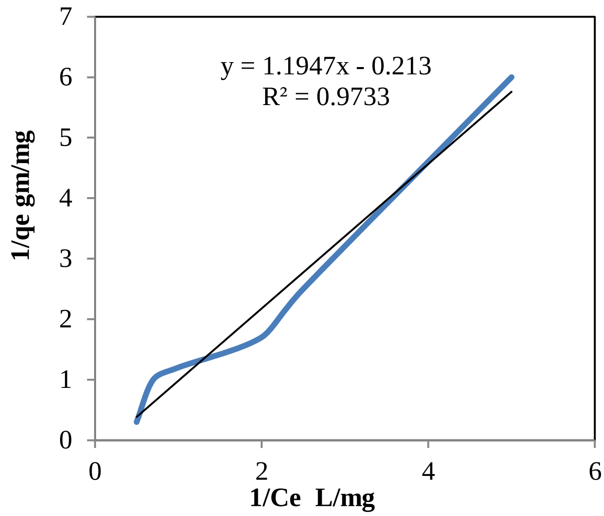

Fig 5. Langmuir for Cd^{+2} ions adsorption onto Cane Papyrus.

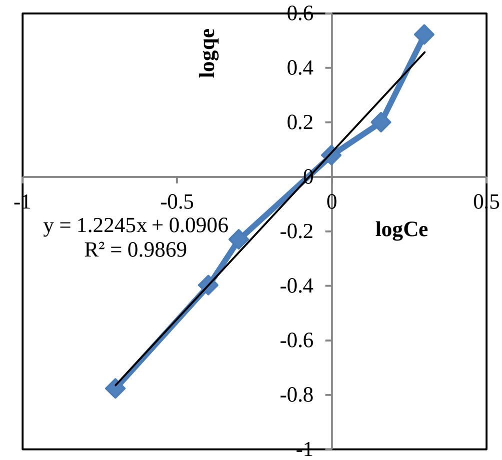

Fig 6. Freundlich plot for Cd^{+2} ions adsorption onto Cane Papyrus.

The Freundlich isotherm model[26] describes a multi-site adsorption for heterogeneous surfaces and can be represented by Equation (5):

$$q_e = K_f \, Ce^{l/n} \quad (5)$$

when "K_f" in (l/mg) is adsorption capacity and "l/n" the adsorption showing the heterogeneity intensity. If taking Equation (5) logarithm, obtain the Equation (6):

$$\log q_e = \log K_l + \frac{l}{n} \log C_e \quad (6)$$

A plot of $\log q_e$ against $\log C_e$ give a linear diagram in 0.9869 as a regression coefficient(Figure 6), showing that Freundlich model adsorption is also conforms. The results from two models (linearized coefficients),indicated that Langmuir model more suitable than Freundlich model for the adsorption process, that mean the chemisorptions process instead of physisorption process is occur. Table 3 illustrated the Langmuir and Freundlich constants, 45.5 mg/g represented the Cane Papyrus maximum adsorption capacity to adsorbed the Cd^{+2} ions from the wastewater.

3.6. The Models of Adsorption Reaction :

Studying both, the diffusion models of adsorption and adsorption reaction models lead to understanding the adsorption reactions mechanism and the reaction kinetics. By using the pseudo first and pseudo second order kinetics Equations (7) and (8) respectively, linearized equations.

$$\log(qe - qt) = logqe - \{\frac{Kt}{2.303}\} \quad (7)$$

$$\frac{t}{qt} = \left\{\frac{1}{K2qe2}\right\} + \frac{1}{qe} \quad (8)$$

when "q_t" and "q_e" are the Cd^{+2} ions amounts adsorbed at time "t" and in equilibrium state,
"k_l" and "k_2" are pseudo first and pseudo second order models rate constants respectively.
(Figure 7) representing pseudo second order, plotting time "t" vs t/q_t values the graph is linear (R_2 = 0.9885). Also, (Figure 8) plotting time "t" next to $\log(q_e – q_t)$ using experimental data, the result from figure, linear coefficient is 0.8681that representing pseudo first order kinetic model [27]. Depending on the results that obtained, the pseudo second order is finest describe to the kinetics adsorption process and the kinetic diagram special treatment chemisorptions mechanistic pathway quite than physisorption[28].

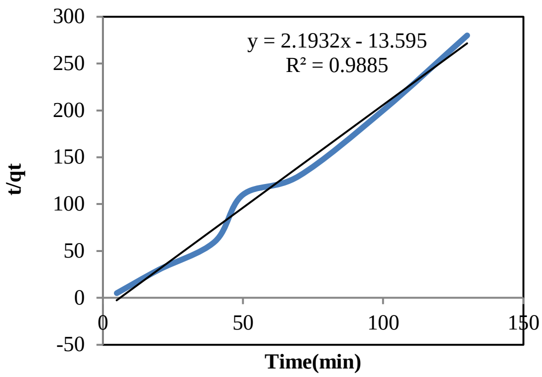

Fig 7. Pseudo second order kinetics for Cd^{+2} ions adsorption onto Cane Papyrus.

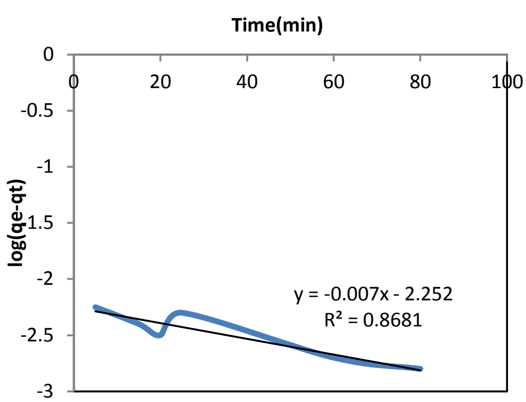

Fig 8. Cd^{+2} ions adsorption onto Cane Papyrus using Pseudo first order kinetics.

Equation 9, explained the Weber-Morris mechanistic model to determine the external film diffusion otherwise intra-particle diffusion :

$$qt=Kd(t)^{1/2}+I \quad (9)$$

when "k_{d}" in (mg/g $min^{-0.5}$) is intra-particle diffusion rate constant and "I" in (mg/g) is a boundary layer thickness constant. The diagram "q_t" against "$t^{1/2}$" is linear, from most studying of mechanism if linearization is obtain not passes through the origin point that's mean the Cd^{+2}ions adsorption controlling by many mechanism. From (Figure 9) the linearization is obtain not pass from origin point, thus the adsorption process mechanism is multi-linearization as well as the adsorption is controlling by intra- particle diffusion and film diffusion[29].

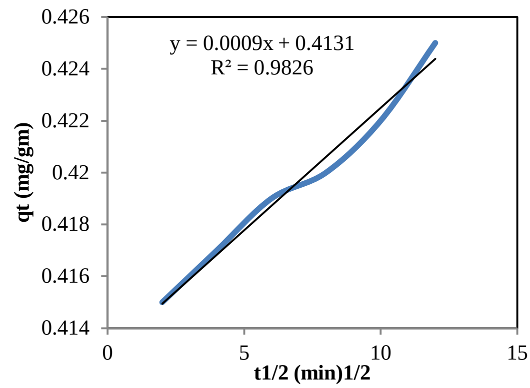

Fig 9. Intra-particle diffusion model plot for Cd^{+2} ions adsorption onto Cane Papyrus.

3.7. Desorption Studies

This study was carried out to assess the most suitable desorbing agent for eluting adsorbed Cd^{+2} ions from the surface of Cane Papyrus. The effects of de-ionized water, 0.2M NaOH and 0.2M HNO_3 solutions were tested for their ability to remove the adsorbed Cd^{+2} ions from the surface of the adsorbent. HNO_3 was a better desorbing agent and was able to recover 50% of Cd^{+2} ions adsorbed to the surface of the adsorbent[30]. NaOH and de-ionized water showed desorption efficiencies of 25% and 2%, respectively. Desorption is beneficial for the separation and enrichment of Cd^{+2} ions as well as the regeneration of the adsorbent.

4. Conclusion

The adsorption ability of powdered Cane Papyrus has been investigated and found effective for the removal of Cd^{+2} ions from wastewater. Acidic functional groups present on the surface Sustainability of the adsorbent is believed to be responsible for the removal of Cd^{+2} ions from aqueous media. The Freundlich isotherm model gave a better description of the adsorption process than the langmuir isotherm model. Pseudo-second order kinetics best described the kinetics of the reaction while 1M HCl was a better desorbing agent than 1M NaOH and de-ionized water.

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Conflict of Interest

The authors declare that they have no conflict of interest.

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