RESEARCH ARTICLE

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# Tea Wastes Efficiency on Removal of Cd(II) From Aqueous Solutions

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**ABSTRACT**

**Background & Aims of the Study:** Heavy metals, such as cadmium (Cd(II)), enter into the environment and cause health hazard due to their toxicity and bioaccumulation in the human body. Therefore, they must be removed from water. In recent years, much attention has been focused on the use of material residues as low-cost adsorbents for the removal of heavy metal ions from aqueous solutions. The aim of this paper is the assessment of tea wastes efficiency on removal of Cd(II) from aqueous solutions.

**Materials and Methods:** The present study was conducted in experimental scale. In this paper, tea wastes were prepared and used as an adsorbent for the removal of Cd(II) ions from water. In batch tests, the effect of parameters like pH (1.0-8.0), initial metal concentration (100-800 mg $L^{-1}$), contact time (15-120 min), adsorbent dose (1.0-5.0 g) and temperature (25-55 °C) on the adsorption process was studied.

**Results:** The results demonstrated that the maximum percentage of Cd(II) adsorption was found at pH 6.0 and the equilibrium was achieved after 60 min with 3.0 g tea wastes. The experimental isotherm data were analyzed, using the Langmuir and Freundlich models and it was found that the removal process followed the Langmuir isotherm. In addition, the adsorption kinetics followed the pseudo-second-order kinetic model. The maximum adsorption capacity calculated by Langmuir fitting was 71.4 mg $g^{-1}$.

**Conclusion:** The results suggest that tea wastes could be employed as an effective material for the removal of Cd(II) ions from aqueous solutions and the maximum adsorption capacity was found to be 71.4 mg $g^{-1}$.

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## Background

The presence of heavy metals in waste water and surface water is becoming a severe environmental problem and because of their non-biodegradability, they can accumulate in the food chain, posing a significant danger to human health (1). Cadmium is one of the most toxic metals ions causing environmental pollution specifically in water. It is extensively used by many industries such as electroplating, painting, machinery and steel making industries. Effluents discharged from these industries contain undesired amounts of cadmium (2). Cadmium is accumulated in the human body, causing erythrocyte destruction, nausea, salivation, diarrhea and muscular cramps, renal degradation, chronic pulmonary problems and skeletal deformity. The World Health Organization (WHO) has set a maximum guideline concentration of 0.005 $mgL^{-1}$ for Cd in drinking water (3).

Several treatment techniques for the removal of heavy metals from water/waste water have been investigated; such as chemical precipitation, electrochemical treatment, ion exchange, membrane separation, adsorption, etc (4). Among these technologies, adsorption is recognized as an effective and economic method for heavy metal removal (5-7). Activated carbon has been widely employed as the most popular adsorbent for the effluents treatment. Besides its extensive use, activated carbon of high quality remains costly (8). Due to the high costs of the adsorbents, there is an increasing interest in the obtainment of low-costs adsorbents, which are able to remove metallic ions from water (9). Agriculture wastes or by-products have been investigated extensively for the removal of heavy metals due to their abundance in nature (10). The utilization of agricultural waste materials is increasingly becoming a vital concern because these wastes represent unused resources and in many cases present serious disposal problems. Numerous waste biomass sources are available in different parts of the world, on which some experimental adsorption properties have been reported e.g. rice husk, peanut shells, corn cobs, saw dust, coir dust, dry tree leaves and barks, tea and coffee waste, rice and wheat bran and sea weeds (11,12). Some studies have demonstrated the ability of tea wastes to remove synthetic dyes and some types of heavy metal ions from water. Mondal (2009) discussed about tea waste for the removal of Pb(II) ions from aqueous solution. Sabrina and Hasmah (2008) reviewed heavy metal removal from water, using tea waste (13-16).

**Aims of the study:**

The aim of this study is assessment of tea wastes efficiency on removal of Cd(II) from aqueous solutions.

## Materials & Methods

### Materials

$Cd(NO_3)_2.4H_2O$, hydrochloric acid (HCl) and sodium hydroxide (NaOH) with the highest available purity were purchased from Merck (Darmstadt, Germany). Double-distilled water was used in all experiments. Stock solution (1000 mg $L^{-1}$) of Cd(II) was prepared by dissolving $Cd(NO_3)_2.4H_2O$ in double-distilled water. The solutions of different initial concentrations were prepared by diluting the stock solution in appropriate proportions.

All Cd(II) concentrations were measured, using an inductively coupled plasma mass spectrometry (Verian710-Es Australia). All pH measurements were made with a 780 pH-meter (Metrohm Switzerland) with a combined glace-calomel electrode.

### Preparation of the tea wastes

Tea wastes were prepared from black teas which were available in Iranian market. This tea waste has been washed at the first step and then rinsed with double-distilled water. After drying at 85°C, it was ground and screened (using screen with mesh size 10). In order to preserve, it was kept in plastic stopper bottle (containers), and to minimize in contact with humidity, all these bottles were preserved in desiccators before the time of use. Some experiments were performed to determine the physical and chemical properties of adsorbent. Chemical and physical properties of the adsorbents as follows: humid (%): 10.5, density (g $cm^{-3}$): 0.353, dissolved material (%): 81, solution particles total (mg $^{1-1}$): 108, organic matters (%): 85, ash content: 2.85, pH ZPC: 5 (12,17).

### Batch adsorption experiments

3 grams of tea wastes were added to 100 mL of Cd(II) solution (100-800 mg $L^{-1}$), and the pH of the solution was adjusted at 6 with 0.1 mol $L^{-1}$ HCl or/and 0.1 mol $L^{-1}$ NaOH solutions and then the mixture was agitated, using a magnetic stirrer with a the stirring speed 160 $r.min^{-1}$ for 60 min. Subsequently, the tea wastes with adsorbed Cd(II) were separated from the mixture, using Whatman filter paper with a diameter of 42µm. The concentration of the Cd(II) ions which remained in the solution were determined by an inductively coupled plasma mass spectrometry (Verian710-Es Australia) and the concentration of the Cd(II) ions remained in the adsorbent phase ($q_e$, mg $g^{-1}$) was calculated, using Eq. (1);

$$q_e = \frac{(C_0 - C_e)V}{W} \qquad (1)$$

Where $C_0$ and $C_e$ are the initial and equilibrium metal concentrations in solution, respectively (mg $L^{-1}$), V (L) is the volume of solution and W (g) is the weight of adsorbent (18).

Finally, metal removal efficiency was calculated, using Eq. (2);

$$R\,(\%) = \frac{C_0 - C_e}{C_0} \times 100 \qquad (2)$$

Where $C_0$ and $C_e$ (mg $L^{-1}$) were the initial and final metal concentrations (19).

## Results

The effect of an initial pH on the removal percentage of Cd(II) in the pH range of 1 to 8 at temperature 25±1 °C and the initial Cd(II) concentration fixed at 100 mg $L^{-1}$ were investigated. Result is presented in Fig. 2. Also, the zero point charge (pHZPC) for the tea wastes was determined around 5 (Fig. 1). As Fig. 2 shows, the adsorption percentage was increased by increasing the pH and reached a maximum at pH 6.

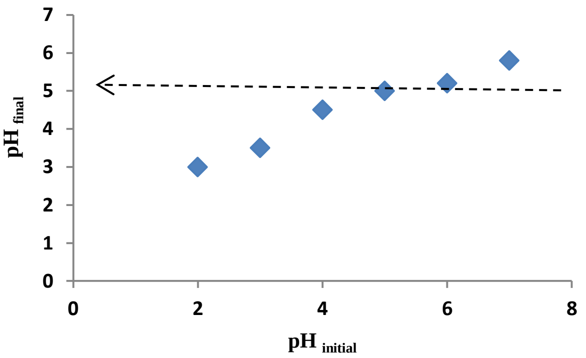

**Figure 1) The determination of the point of zero charge of the tea waste**

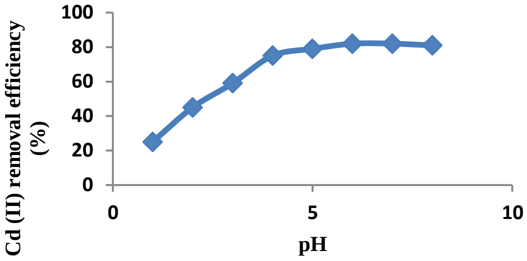

**Figure 2) Effect of pH solution on the removal of Cd(II) from aqueous solution by tea wastes (Initial Cd(II) concentration 100 mg $l^{-1}$, contact time 60 min, adsorbent dose 3 g, temperature 25±1 °C)**

The removal of Cd(II) onto tea wastes as a function of their concentrations was studied at constant temperature (25±0.1°C) by varying the Cd(II) concentration from 100 to 800 mg $L^{-1}$ while keeping all other parameters constant. The adsorption results of Cd(II) by tea wastes are shown in Fig. 3. Equilibrium uptake has been decreased with increase in the initial metal concentration in the range of concentrations 100-800 mg $L^{-1}$.

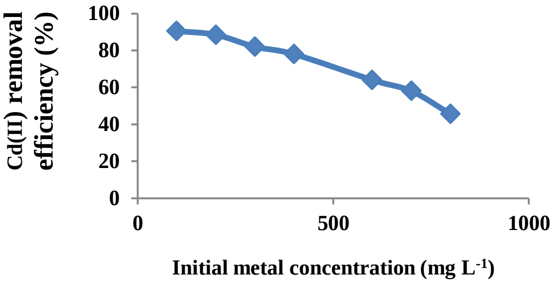

**Figure 3) Effect of an initial metal concentration on the removal of Cd(II) from aqueous solution by tea wastes (pH=6, contact time 60 min, adsorbent dose 3 g, temperature 25±1 °C)**

The effect of contact time on the adsorption of Cd(II) was studied to determine the time taken by tea wastes to remove 100 mg $L^{-1}$ Cd(II) solution at pH 6 (Fig. 4). The removal percentage of Cd(II) by tea wastes was increased by increasing the contact time and then became constant. Therefore, agitation time of 60 min was selected for further works.

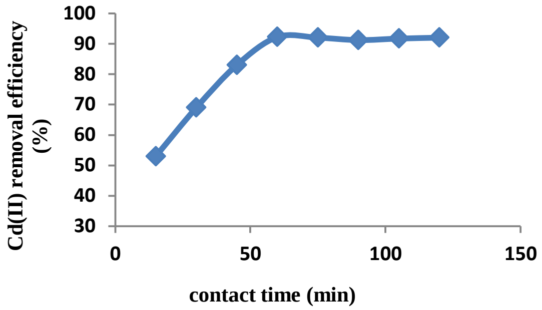

**Figure 4) Effect of contact time on the removal of Cd(II) from aqueous solution by tea wastes (pH=6, initial Cd(II) concentration 100 mg l$^{-1}$, adsorbent dose 3 g, temperature 25±1 °C).**

The dependence of the adsorption of Cd(II) ions to the amount of tea wastes was studied at temperature 25±0.1°C and at pH 6 by varying the adsorbent amount from 1 to 5 g in contact with 100 mg L$^{-1}$ of Cd(II) solution. The results are shown in Fig. 5. Apparently, the percentage removal of Cd(II) ions increased by increasing the amount of adsorbent.

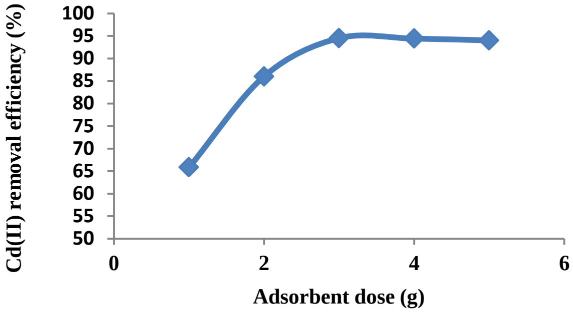

**Figure 5) Effect of adsorbent dose on the removal of Cd(II) from aqueous solution by tea wastes (pH=6, initial Cd(II) concentration 100 mg l$^{-1}$, contact time 60 min, temperature 25±1 °C).**

In order to observe the effect of temperature on the adsorption capacity, some experiments were carried out for the Cd(II) concentration of 100 mg L$^{-1}$ and at different temperatures in the range of 25–55 °C. The results are shown in Fig. 6.

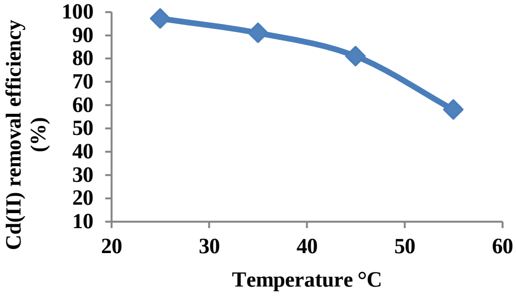

**Figure 6) Effect of temperature on the removal of Cd(II) from aqueous solution by tea wastes (pH=6, initial Cd(II) concentration 100 mg l$^{-1}$, contact time 60 min, adsorbent dose 3 g).**

The relationship between the removal ability of the material and the concentration of the contaminant solution could be illustrated by adsorption isotherms. In the present study, the data obtained from the equilibrium adsorption experiments, at temperature of 25±0.1°C were analyzed, using the Langmuir and Freundlich models. Table 1 showed the values of Langmuir and Freundlich constants, and the correlation coefficients R$^2$ obtained from the linear regression (Fig. 7a,b).

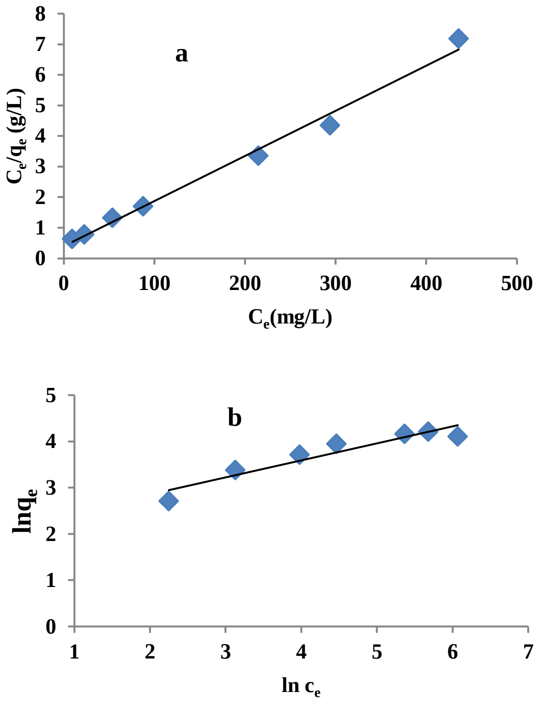

**Figure 7) (a) Langmuir and (b) Freundlich isotherms for Cd(II) adsorption onto tea wastes.**

**Table 1) Isotherm parameters of adsorption of Cd(II) onto tea wastes**

| | **Langmuir** | | | **Freundlich** | | |
|---|---|---|---|---|---|---|
| | b (L mg$^{-1}$) | $q_m$ (mg g $^{-1}$) | $R^2$ | $K_f$ (mg$^{1-(1/n)}$ L$^{1/n}$ g$^{-1}$) | n | $R^2$ |
| **Cd (II)** | 0.035 | 71.4 | 0.989 | 8.38 | 2.73 | 0.895 |

**Table 2) Pseudo-first order and pseudo-second order kinetic model parameters for the adsorption of Cd(II) onto tea wastes.**

| | | **Pseudo-first-order kinetic** | | | | **Pseudo-second-order kinetic** | | |
|---|---|---|---|---|---|---|---|---|
| | $C_0$ (mg L$^{-1}$) | $q_e$ exp(mg g$^{-1}$) | $q_{e1}$(mg g$^{-1}$) | $k_1$ (min$^{-1}$) | $R^2$ | $q_{e2}$ (mg g$^{-1}$) | $k_2$ (g mg$^{-1}$ min$^{-1}$) | $R^2$ |
| **Cd (II)** | 100 | 15.4 | 10.64 | 0.042 | 0.871 | 17.54 | 0.004 | 0.994 |

In order to elucidate the Cd(II) adsorption process on tea wastes, particularly the potentially rate-controlling step, the adsorption data were analyzed, using the pseudo first-order and pseudo-second-order models. The kinetic parameters and the correlation coefficients ($R^2$) were determined by linear regression (Fig. 8a,b), and the results are given in Table 2.

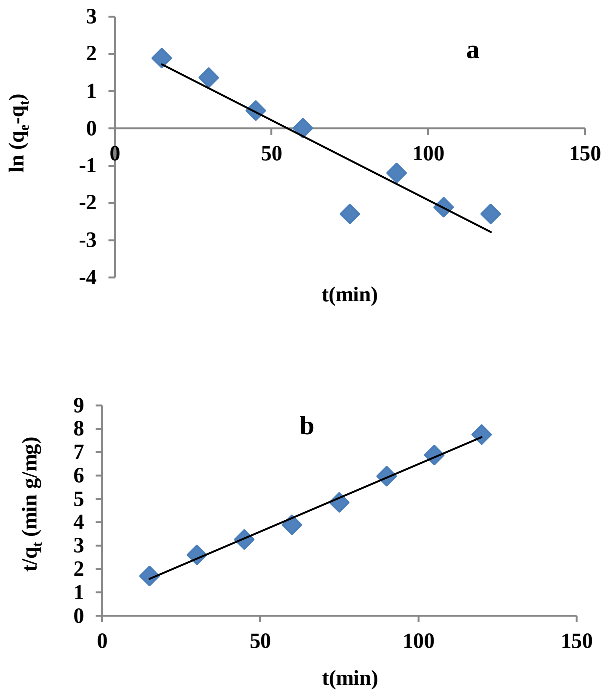

**Figure 8) (a) Pseudo-first-order and (b) pseudo-second-order kinetics plots of Cd(II) adsorption onto the tea wastes**

## Discussion

PH of the solution in the adsorption process of dye molecules can affect both aqueous chemistry and surface binding-sites of the adsorbent. As was seen in the Fig. 2, adsorption capacity increased by increasing the pH and reached maximum at pH 6 and then remained nearly constant at higher pHs. At lower pH (<pHzpc), the tea wastes surface will be positively charged, creating electrostatic repulsion between the positively charged surface of the tea wastes and the metal ions. At higher pH (>pHzpc), the number of positively charged sites are reduced and raised the number of negatively charged sites, which increases the electrostatic attractions between metal ions and tea wastes surface. Moreover, It can be observed that in strong acidic solutions, the removal percentage decreases, because of competition between $H^+$ and Cd(II) ions. Similar results have been found by Şerife et al (20).

As in Fig. 3 was seen, by increasing the initial concentration of Cd(II) ions in aqueous solution, removal percentage was decreased, so, in the concentration of 100 mg L$^{-1}$ and constant contact time, observed highest adsorption percentage and with increasing in concentration amount from 100 to 800 mgL$^{-1}$, decreased adsorption percentage from 90% to 45% for Cd(II) was seen. At a constant tea wastes dose, the decrease in the adsorption percentage was seen which is probably due to the saturation of the active sites on the tea wastes surface at higher Cd(II) concentrations. On the other hand, by increasing the initial Cd(II) concentration, the actual amount of Cd(II) adsorbed per unit mass of the tea wastes was increased. The higher initial concentration of Cd(II) provides an important driving force to overcome the mass transfer resistance for Cd(II) transfer between the solution and the surface of the tea wastes. Similar results were observed by Chang (2011) who investigated the effect of initial concentration on removal of Rhodamine 6G from aqueous solution by hexadecyl functionalized magnetic silica nanoparticles and indicated that adsorption decreases with increasing the initial concentration of metals (21).

The contact time between adsorbate and adsorbent is the most important design parameter that affects the performance of adsorption processes. The equilibrium was found to be nearly 60 min when the maximum Cd(II) adsorption capacity was reached. The results are shown in Fig. 4. The rapid adsorption at the initial contact time was due to the availability of more active surface of the adsorbents, which leads to fast adsorption of the Cd(II) from the solution. The later slow rate of Cd(II) adsorption probably occurred due to the less availability of active site onto the surface of adsorbent as well as the slow pore diffusion of the solute into the adsorbent. Similar results were observed by Gupta who investigated the effect of contact time on removal of lead from aqueous solution by alumina-coated carbon nanotubes and indicated that adsorption increases with increasing contact time (22).

Fig. 5 shows that the removal efficiency increased with the increase in tea wastes dose at constant initial metal concentration. A rapid increase in adsorption with the increasing in tea wastes dose was found due to the availability of more adsorption sites. For Cd(II) concentration of 100 mg $l^{-1}$, 94% of the Cd(II) removal efficiency was obtained, using tea wastes dose of 3 g.

Fig. 6 shows that the adsorption amount of Cd(II) ions onto tea wastes decreased with an increase in temperature indicating that the process is exothermic in nature (Fig. 6). The decrease in the adsorption capacity of tea wastes with temperature may be results of the weakening of bonds between metal ions molecules and tea wastes of adsorbents at high temperatures. Similar results were observed by Ribeiro who investigated the effect of temperature on removal of dyes from aqueous solution by an activated carbon xerogels and indicated that adsorption decreases with increasing the temperature (23).

Adsorption of Cd(II) ions onto tea wastes was analyzed with the Langmuir and Freundlich models. The Langmuir isotherm Eq. (3) and Freundlich isotherm Eq. (4) can be expressed as follows (24, 25);

$$\frac{C_e}{q_e} = \frac{C_e}{q_m} + \frac{1}{q_m b_l} \qquad (3)$$

$$\ln q_e = \frac{1}{n} \ln C_e + \ln k_f \qquad (4)$$

here $C_e$ (mg $L^{-1}$) is the equilibrium concentration of Cd(II) ions in solution, $q_e$ (mg $g^{-1}$) is the equilibrium adsorption capacity of tea wastes, $q_m$ (mg $g^{-1}$) is the maximum adsorption capacity of tea wastes for monolayer coverage, b (L $mg^{-1}$) is a constant related to the adsorption free energy, $K_f$ ($mg^{1-(1/n)\ L1/n\ g-1}$) is a constant related to adsorption capacity, and n is an empirical parameter related to adsorption. The higher correlation coefficient for Langmuir model indicates that the Langmuir model fitted the adsorption data better than the Freundlich model, which indicated the homogeneous distribution of active sites on the surface of tea wastes. The results showed that the maximum adsorption capacity value for the adsorption of Cd(II) ions by the tea wastes was 71.4 mg $g^{-1}$. The Cd(II) adsorption capacity was compared with other adsorbents in the literatures. Adsorption capacity of Cd(II) was achieved by Bagasse fly ash 6.19 (26), Multi-carboxyl-functionalized silica gel 43.8 (27), Graphenes magnetic material 27.83 (28) and Chitosan 58.8 (29), respectively. However, more efficient removal and higher adsorption capacity were observed by the adsorbent (tea wastes) prepared in this work.

In order to explain the adsorption mechanism and adsorption characteristics, two adsorption kinetics models, pseudo-first-order model and pseudo-second-order model were used to analyze the adsorption kinetic data. The linear forms of pseudo-first-order Eq. (5) and pseudo-second-order Eq. (6) models are expressed as follows (30,31);

$$\ln( q_e - q_t) = \ln(q_e) - \frac{k_1 t}{2.303} \qquad (4)$$

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \qquad (5)$$

Where $q_e$ and $q_t$ are the amount of Cd(II) and Pb(II) ions adsorbed (mg/g) at equilibrium and time t (min); $k_1$ is the rate constant of pseudo-first-order ($min^{-1}$); $k_2$ is the rate constant of pseudo-second-order ($g\ mg^{-1}\ min^{-1}$) for adsorption. The results indicated that an experimental data better fits to pseudo-second-order kinetic model in terms of higher correlation coefficient values (Table 2).

## Conclusion

Results of adsorption experiments revealed that removal efficiency of Cd(II) is pH dependent and the maximum removal was attained at pH 6. Moreover, the adsorption capacity of Cd(II) on tea wastes adsorbent decreased with increase the initial metal concentrations and temperature. Also, by increasing the contact time and adsorbent dose, the amount of removal was increased. Adsorption of Cd(II) ions onto tea wastes follows Langmuir model and kinetic of adsorption process follow pseudo-second-order model. This study shows that tea wastes can be used in an industrial wastewater treatment for removal of heavy metal.

## Footnotes

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### Conflict of Interest:

The authors declared no conflict of interest.

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