REMOVAL OF SOME HEAVY METALS FROM INDUSTRIAL WASTEWATER BY USING DRY BIOMASS OF *HYDRILLA VERTICILLITA* (L. F.) ROYLE

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Abstract

The aim of this research work is to reduce the concentration of Cd^{2+}, Cr^{3+} and Pb^{2+} ions in three industrial wastewater samples from dry cell battery factory, nickel plating factory and leather factory in industrial zones I and II, Mandalay Region, Myanmar. The aquatic plant namely *Hydrilla verticillata* (L. F.) Royle (Mhaw Gyan) was used as dry biomass for the removal of heavy metal ions from three industrial wastewater samples within the optimum experimental conditions. The sorption parameters (optimum contact time and loading weight) were determined using Langmuir isotherm. According to the results, the experimental data for sorption of Cd^{2+} and Cr^{3+} ions were fitted to the Langmuir model except Pb^{2+}. The optimum contact time and optimum loading weight were used for the removal of selected metal ions from three industrial wastewater samples. The result revealed that the removal order of metal ions from three industrial wastewater samples using dry biomass was found to be $Cd^{2+} > Cr^{3+} > Pb^{2+}$.

Keywords: dry biomass, industrial wastewaters, Langmuir isotherm, sorption, heavy metal ions

Introduction

The increasing of urbanization and industrialization has dramatically led into the production of the intensity of wastewater around the world (Asia *et al*., 2006). Heavy metal pollution comes in streams from industrial sectors. The industrial sectors produces harmful heavy metal waste, as a result contaminating water resources and ground resources (Tariq *et al*., 2018). With increasing environmental awareness and legal constraints being imposed on the discharge of effluents, a need for cost-effective alternative technologies are essential. In this endeavor plant biomass can emerged as an option for developing economic and eco-friendly wastewater treatment through a process called biosorption (Dixit *et al*., 2015). Biosorption is a physiochemical process that occurs naturally in certain biomass which allows it to passively concentrate and bind contaminants onto its cellular structure (Ramasubramaniam *et al*., 2012). It is a biological method of environmental controlled and can be an alternative to conventional contaminate wastewater treatment facilities (Dixit *et al*., 2015). Biosorbent materials are derived from raw microbial, seaweed or even some plant biomass through different kinds of simple procedures. They may be chemically pretreated for better performance and suitability for process applications. Biosorbents are capable of directly sorbing metal ionic species from aqueous solutions (Ramasubramaniam *et al*., 2012). Studies on biological methods are very important area of research with huge potential for research and applicability for removal of heavy metals (Dhokpande and Kaware., 2013). Adsorption is the adhesion of atoms, ions, biomolecular or biomolecules of gas, liquid, or dissolved solid to a surface (Itodo *et al*., 2013). Several mathematical models can be used to describe experimental data of adsorption isotherms. The Freundlich, Langmuir and Temkin models are employed to analysis of adsorption occurred in the experiment (Rahimi and Vadi., 2014). In this research work, the optimum contact time and

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optimum loading weight were determined and the removal of selected metal ions (Cd^{2+}, Cr^{3+}, Pb^{2+}) with dry biomass *Hydrilla verticillata* (L.F.) Royle was investigated.

Materials and Methods

Samples Collection

Two selected wastewater samples from dry cell battery factory and nickel plating factory were collected from Industrial Zone I in Pyigyitagun Township, Mandalay Region (Figure 1). Another wastewater sample from leather factory was collected from Industrial Zone II in Amarapura Township, Mandalay Region (Figure 2).

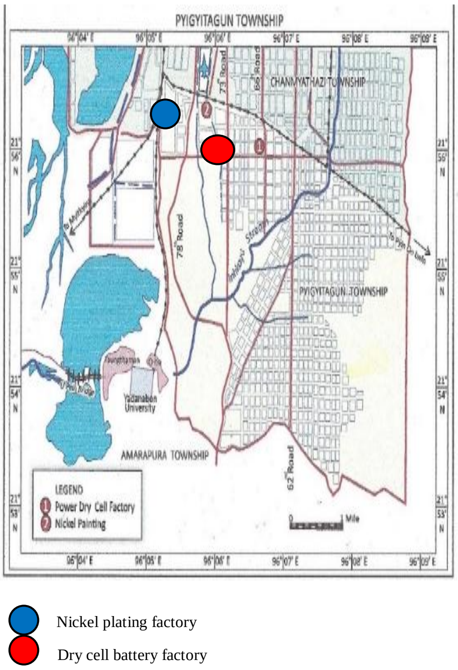


Figure 1 Location map of the study area (1)

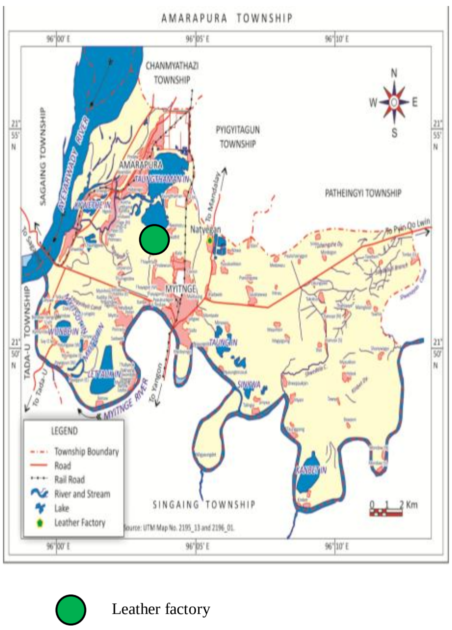


Figure 2 Location map of the study area (2)

Hydrilla verticillata (L. F.) Royle (Mhaw Gyan) was collected from Mandalay moat in Mandalay, Myanmar. It is shown in Figure 3.

Figure 3 *Hydrilla verticillata* (L. F.) Royle (Mhaw Gyan)

Wastewater Treatment by Using Dry Biomass

Adsorption method was applied for the wastewater treatment by using dry biomass (Baig*et al*., 2015).

Preparation of Dry Biomass

The collected aquatic plant was washed with pure water and dried in an oven at 60ºC for 36 h. The dried sample was ground and sieved through a 100 mesh sieve.

Effects of Adsorption Time of Dry Biomass

2.5 g of dry biomass was weighed and washed twice with 0.1 M HCl and once again with distilled water. The biomass was suspended in 500 mL of 0.01 M sodium acetate to obtain a tissue concentration of 5 mg/mL. After adjusting the pH to 5, 50 mL of the suspension was transferred to conical flask. The suspension was centrifuged and the supernatant was discarded. And then, 50 mL of wastewater samples was added to each precipitate in 100 mL conical flask with various time intervals (5, 10, 15, 30, 60, 90 and 120 min). All the conical flasks were shaken on a shaker for 90 min. After the mixtures were shaken, centrifuged and filtered, the supernatants were analyzed by atomic absorption spectrophotometer.

Effect of Loading Weight of Dry Biomass

Different weights (0.20 g, 0.25 g, 0.30 g, 0.35 g and 0.40 g) of dry biomass were weighed and washed twice with 0.1 M HCl and once again with distilled water. The biomass was suspended in 50 mL of 0.01 M sodium acetate to obtain a tissue concentration of 5 mg/mL.

After adjusting the pH to 5, 50 mL of the suspension was transferred to 100 mL conical flask and settled for 15 min. The suspensions were centrifuged and the supernatants were discarded. And then, 50 mL of wastewater sample was added to each precipitate in 100 mL conical flask. All the conical flasks were shaken on a shaker for 90 min. After the mixtures were shaken, centrifuged and filtered, the supernatants were analyzed by atomic absorption spectrophotometer.

Adsorption Study

The phenomena of adsorption is generally applied for the heavy metal (Cd^{2+}, Cr^{3+} and Pb^{2+}) ions removal from industrial wastewater. The sorption studies of the wastewater can be carried out by the sorbent-sorbate interaction with the dry biomass. In the sorption studies, the different loading weights and the different contact times were used.

Adsorption Study on Effect of Loading Weight of Dry Biomass

In the sorption studies the loading weight (mass of sorbent (dry biomass)) was varied and concentration of wastewater (5 mg/mL) was fixed. The different mass of sorbents (0.20 g, 0.25 g, 0.30 g, 0.35 g and 0.40 g) were used.

Wastewater Treatment by Using Dry Biomass

Each 0.25 g and 0.35 g of dry biomass were weighed and washed twice with 0.1 M HCl and washed again with distilled water. The biomass was suspended in 50 mL of 0.01 M sodium acetate for 15 min and 90 min to obtain a concentration of 5 mg/mL. The pH was adjusted to 5. After 15 min and 90 min later, the suspensions were transferred to conical flasks. The suspensions were centrifuged and the supernatants were discarded. And then, 50 mL of wastewater sample was added to each precipitate in 100 mL conical flask. All the conical flasks were shaken on a shaker for 90 min, centrifuged and filtered. Finally, the supernatants were analyzed by atomic absorption spectrophotometer.

Results and Discussion

Wastewater Treatment by Using Dry Biomass

The optimum experimental conditions (the contact time and loading weight) were used for the removal of selected heavy metal ions from dry cell battery factory, nickel plating factory and leather factory wastewater samples using dry biomass.

Effects of Contact Time on the Removal of Cd^{2+}, Cr^{3+}, Pb^{2+}Ion from Three Industrial Wastewater by Dry Biomass

In this study, the effects of contact time using dry biomass with initial concentration 5 mg/mL at 0.25 g adsorbent weight on the removal of Cd^{2+}, Cr^{3+} and Pb^{2+} from dry cell battery factory, nickel plating factory and leather factory wastewater samples were investigated. The resulted data is shown in Table 1 and Figure 4, the optimum time for the removal percentage of Cd^{2+} ion from wastewater samples of dry cell battery factory (52.99 %), nickel plating factory (56.68 %) and leather factory (58.33 %) were observed at 90 min using the dry biomass. Furthermore, the optimum time for the removal percentage of Cr^{3+} ion from dry cell battery factory (33.45 %), nickel plating factory (35.96 %) and leather factory (33.60 %) were also found at 90 min using the dry biomass (Table 2 and Figure 5). As shown in Table 3 and Figure 6, dry biomass could remove Pb^{2+} ion from dry cell battery factory (3.30 %), nickel plating factory (4.35 %) and leather factory (3.33 %), after 15 min.

Table 1 Effect of Contact Time on the Removal of Cd^{2+} Ion from Three Industrial Wastewater Samples by Dry Biomass

Contact time (min)	Removal percent of Cd^{2+} ion (%)								
	Dry cell battery factory			Nickel plating factory			Leather factory		
	Before treatment	After treatment	Removal (%)	Before treatment	After treatment	Removal (%)	Before treatment	After treatment	Removal (%)
5		0.270	40.13		0.271	41.59		0.029	39.58
10		0.245	43.68		0.268	42.24		0.027	43.75
15		0.235	47.89		0.261	43.75		0.025	47.91
30	0.451	0.216	52.11	0.464	0.245	47.19	0.048	0.024	50.00
60		0.214	52.54		0.227	51.08		0.022	54.16
90		0.212	52.99		0.201	56.68		0.020	58.33
120		0.217	51.88		0.215	53.66		0.203	52.08

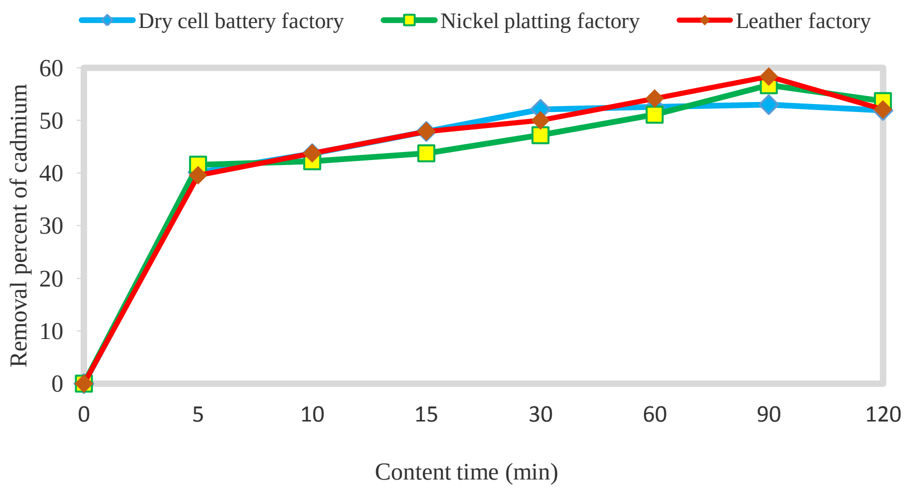


Figure 4 Effect of contact time on the removal of Cd^{2+}ion from three industrial wastewater samples by modified dry biomass

Table 2 Effect of Contact Time on the Removal of Cr^{3+} Ion from Three Industrial Wastewater Samples by Dry Biomass

Contact time (min)	Removal percent of Cr^{3+} ion (%)								
	Dry cell battery factory			Nickel plating factory			Leather factory		
	Before treatment	After treatment	Removal (%)	Before treatment	After treatment	Removal (%)	Before treatment	After treatment	Removal (%)
5		0.200	28.10		0.375	23.22		0.283	24.53
10		0.194	30.21		0.321	30.96		0.261	30.40
15		0.193	30.58		0.317	31.83		0.256	31.73
30	0.278	0.190	31.65	0.465	0.315	32.26	0.375	0.255	32.00
60		0.189	32.01		0.304	34.62		0.252	32.80
90		0.185	33.45		0.299	35.96		0.249	33.60
120		0.187	32.73		0.303	34.83		0.252	32.80

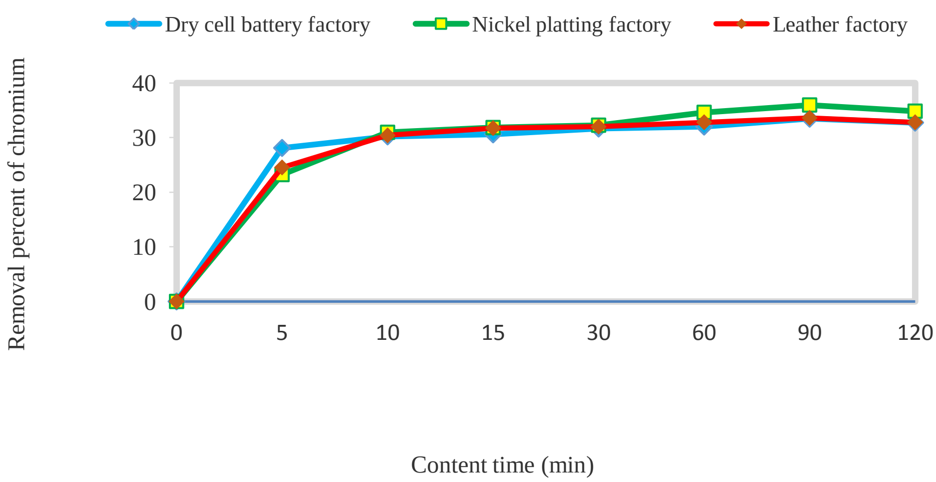


Figure 5 Effect of contact time on the removal of Cr^{3+} ion from three industrial wastewater samples by dry biomass

Table 3 Effect of Contact Time on the Removal of Pb^{2+} Ion from Three Industrial Wastewater Samples by Dry Biomass

Contact time (min)	Removal percent of Pb^{2+} ion (%)								
	Dry cell battery factory			Nickel plating factory			Leather factory		
	Before treatment	After treatment	Removal (%)	Before treatment	After treatment	Removal (%)	Before treatment	After treatment	Removal (%)
5		1.930	1.91		0.910	3.39		0.762	2.23
10		1.935	3.10		0.901	3.93		0.751	2.34
15		1.925	3.30		0.905	4.35		0.654	3.33
30	1.991	1.931	3.01	0.942	0.908	3.61	0.981	0.685	3.02
60		1.929	3.12		0.923	2.02		0.670	3.17
90		1.926	3.16		0.924	1.91		0.675	3.12
120		1.927	3.21		0.925	1.80		0.673	3.14

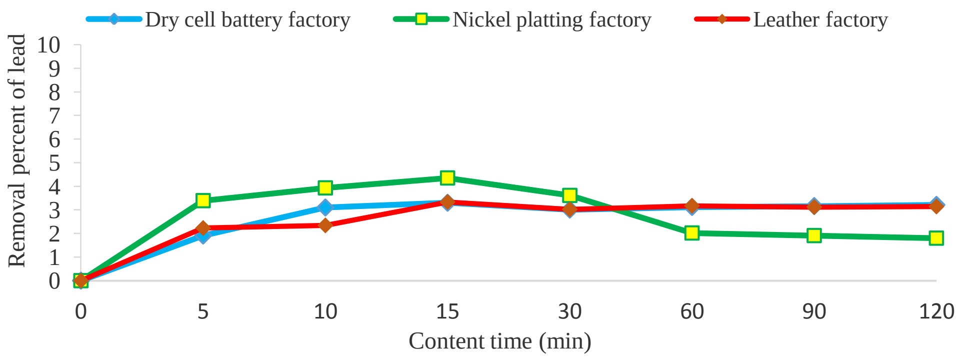


Figure 6 Effect of contact time on the removal of Pb^{2+} ion from three industrial wastewater samples by dry biomass

Effects of Loading Weight of the Removal of Cd^{2+}, Cr^{3+}, Pb^{2+} Ions from Three Industrial Wastewater by Dry Biomass

In this study, the effect of loading weight of dry biomass in initial concentration 5 mg/mL taking the range of the weight 0.20 g to 0.40 g for dry cell battery factory, nickel plating factory and leather factory were investigated at contact time 90 min.

In resulted data indicated in Table 4 and Figure 7, dry biomass at 0.35 mg showed the highest removal percent of Cd^{2+} ion (71.37 %, 70.90 % and 72.92 %) respectively. Furthermore, it was also found that the weight of biomass at 0.35 mg showed the highest removal percent Cr^{3+} ion (67.27 %, 67.52% and 66.93 %) from dry cell battery, nickel plating and leather factories wastewater samples (Table 5 and Figure 8). As shown in Table 6 and Figure 9, dry biomass at 0.25 mg could remove the highest removal percent of Pb^{2+} ion (3.10 %, 4.56 % and 4.99 %) from dry cell battery, nickel plating and leather factories wastewater samples.

Table. 4 Effect of Loading Weight of Dry Biomass on the Removal of Cd^{2+} Ion from Three Industrial Wastewater Samples

Loading weight (g)	Removal percent of Cd^{2+} ion (%)								
	Dry cell battery factory			Nickel plating factory			Leather factory		
	Before treatment	After treatment	Removal (%)	Before treatment	After treatment	Removal (%)	Before treatment	After treatment	Removal (%)
0.20	0.451	0.245	45.68	0.464	0.265	42.88	0.048	0.028	41.67
0.25		0.220	51.22		0.225	51.50		0.026	45.83
0.30		0.190	57.78		0.198	57.33		0.023	52.08
0.35		0.129	71.37		0.135	70.90		0.013	72.92
0.40		0.135	70.06		0.150	67.67		0.015	68.75

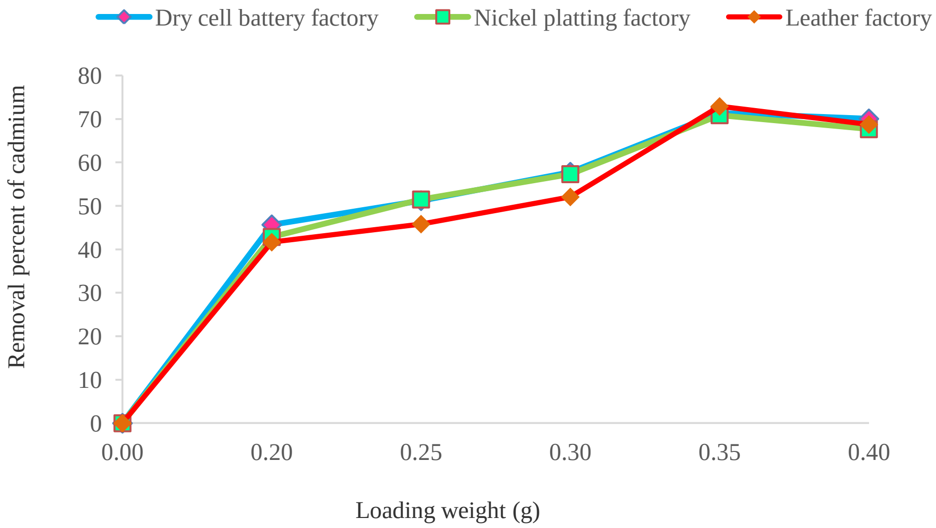


Figure 7 Effect of loading weight of dry biomass on the removal of Cd^{2+} ion from three industrial wastewater samples

Table 5 Effect of Loading Weight of Dry Biomass on the Removal of Cr^{3+} Ion from Three Industrial Wastewater Samples

Loading weight (g)	Removal percent of Cr^{3+} ion (%)								
	Dry cell battery factory			Nickel plating factory			Leather factory		
	Before treatment	After treatment	Removal (%)	Before treatment	After treatment	Removal (%)	Before treatment	After treatment	Removal (%)
0.20	0.278	0.215	22.67	0.465	0.355	23.66	0.375	0.300	20.00
0.25		0.197	29.13		0.341	26.67		0.277	26.13
0.30		0.181	34.89		0.298	35.91		0.250	33.33
0.35		0.091	67.27		0.151	67.52		0.124	66.93
0.40		0.101	63.66		0.162	65.16		0.129	65.60

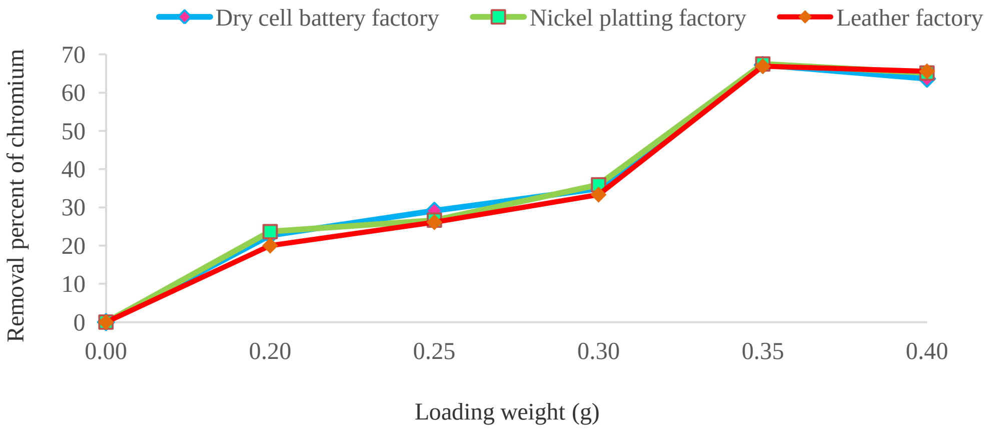


Figure 8 Effect of loading weight of dry biomass on the removal of Cr^{3+} ion from three industrial wastewater samples

Table 6 Effect of Loading Weight of Dry Biomass on the Removal of Pb^{2+} Ion from Three Industrial Wastewater Samples

Loading weight (g)	Removal percent of Pb^{2+} ion (%)								
	Dry cell battery factory			Nickel plating factory			Leather factory		
	Before treatment	After treatment	Removal (%)	Before treatment	After treatment	Removal (%)	Before treatment	After treatment	Removal (%)
0.20	1.991	1.984	2.15	0.942	0.911	3.30	0.981	0.957	2.45
0.25		1.930	3.10		0.899	4.56		0.932	4.99
0.30		1.935	2.81		0.907	3.71		0.949	3.26
0.35		1.946	2.20		0.921	2.23		0.960	2.14
0.40		1.941	2.51		0.925	1.81		0.961	2.04

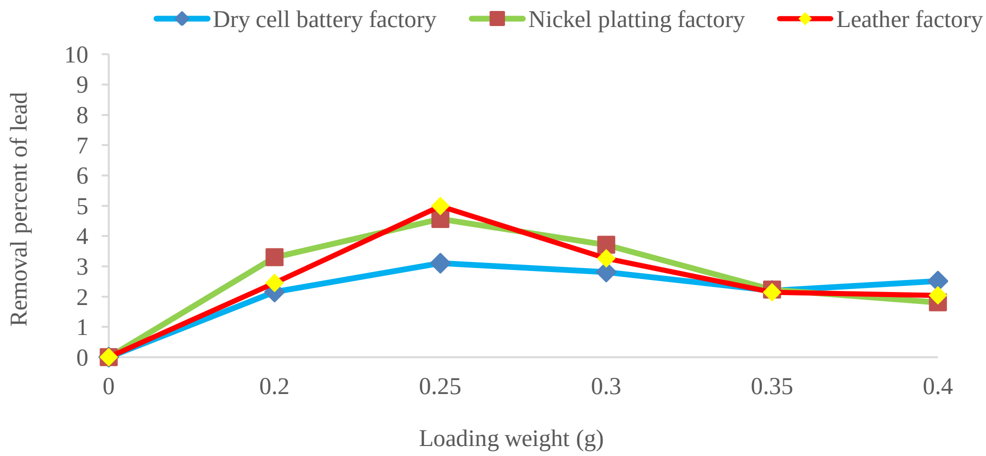


Figure 9 Effect of loading weight of dry biomass on the removal of Pb^{2+} ion from three industrial wastewater samples

Study on Adsorption Isotherm

The amount of Cd^{2+}, Cr^{3+} and Pb^{2+} adsorbed onto the dry biomass from *Hydrilla verticillata* (L. F.) Royle, q_e (mg g^{-1}) was calculated by

$$q_e = (C_0 - C_e)\ \frac{v}{w}$$

where C_o and C_e are the initial and equilibrium concentrations of wastewater (mg g^{-1}), 'v' the volume of the solution (L), and 'w' the weight of the plant biomass (g) (Nethaji*et al.*, 2013).

There are two well-known types of adsorption isotherm; the Langmuir isotherm and, Freundlich isotherm.

The Langmuir adsorption isotherm is mathematically expressed as;

$$\frac{x}{m} = \frac{X_m bC}{1+bC}$$

$$q_e = \frac{X_m bC_e}{1+bC_e}$$

$$\frac{1}{q_e} = \frac{1}{X_m} + \frac{1}{X_m bC_e}$$

$$\frac{C_e}{q_e} = \frac{C_e}{X_m} + \frac{1}{X_m b}$$

where X_m = Langmuir monolayer capacity parameter

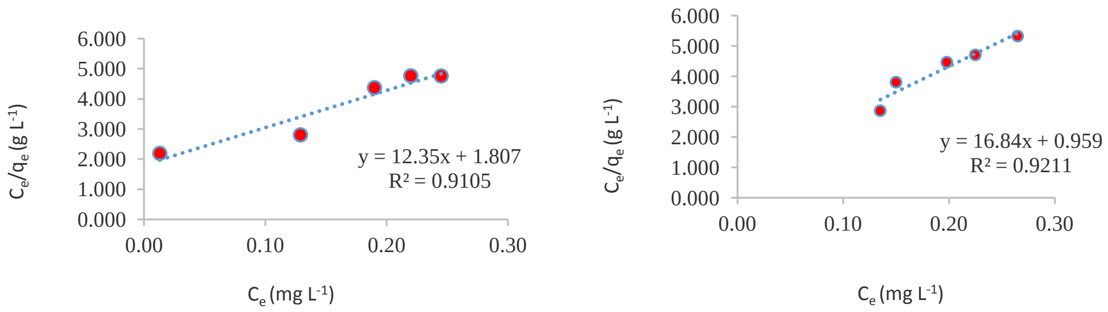
b = constant for a given adsorbate and adsorbent at a particular temperature.

From the isotherm model plot, Langmuir and model constants are calculated to know its isotherm model fit to the adsorption of biomass on adsorbent Cd^{2+}, Cr^{3+} and Pb^{2+} ions.

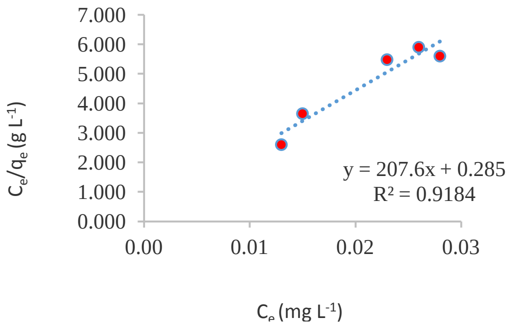
Adsorption Isotherm from Three Industrial Wastewater Samples onto the Dry Biomass

The adsorption of heavy metal ions such as Cd^{2+}, Cr^{3+} and Pb^{2+} ions from three industrial wastewater onto dry biomass were determined by using Langmuir's adsorption isotherm. The optimum contact time and optimum loading weight were determined. The adsorption nature of sorbent-sorbate interaction with dry biomass was studied by using the optimum time (90 min for Cd^{2+} and Cr^{3+}) and (15 min for Pb^{2+}).

As shown in Figure 10, the linear coefficient values (R^2) from wastewater samples of dry cell battery, nickel plating and leather factories were 0.9105, 0.9211 and 0.9184 for Cd^{2+} ion. Therefore, for the adsorption of Cd^{2+} ion from three industrial wastewater samples the data fitted with the Langmuir isotherm.



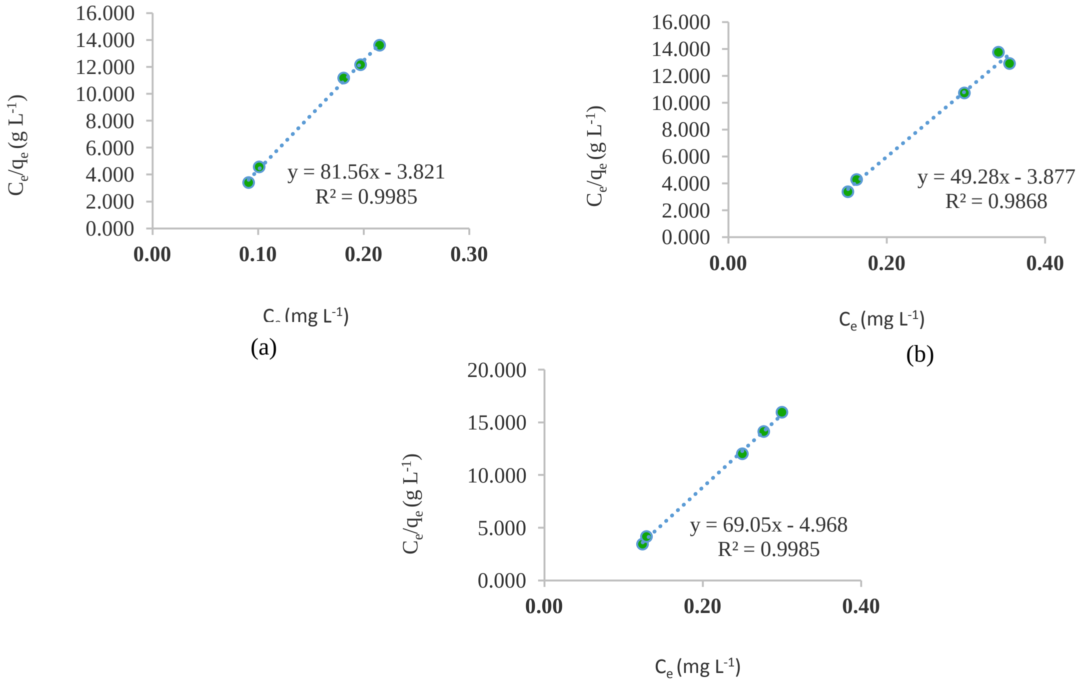
(a) (b)



(c)

Figure 10 Langmuir isotherm for Cd^{2+} ion from three industrial wastewater samples onto the dry biomass (a) dry cell battery factory (b) nickel plating factory and (c) leather factory

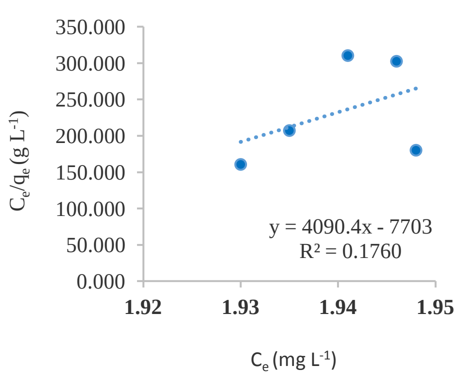
According to Figure 11, the linear coefficient values (R^2) from wastewater samples of dry cell battery, nickel plating and leather factories were 0.9985, 0.9868 and 0.9985 for Cr^{3+} ion. Therefore, for the adsorption of Cr^{3+} ion from three industrial wastewater samples the data fitted with the Langmuir isotherm.



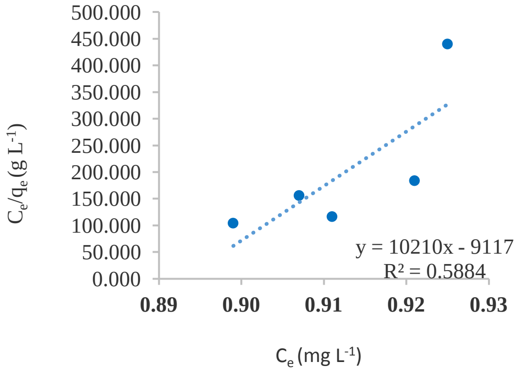
(c)

Figure 11 Langmuir isotherm for Cr^{3+} ion from three industrial wastewater samples onto the dry biomass (a) dry cell battery factory (b) nickel plating factory and (c) leather factory

The linear coefficient values (R^2) from wastewater samples of dry cell battery, nickel plating and leather factories were 0.1760, 0.5884 and 0.6354 for Pb^{2+} ion in Figure 12. Therefore, the adsorption of Pb^{2+} ion from three industrial wastewater samples were found to be deviation from the Langmuir isotherm.



(a)



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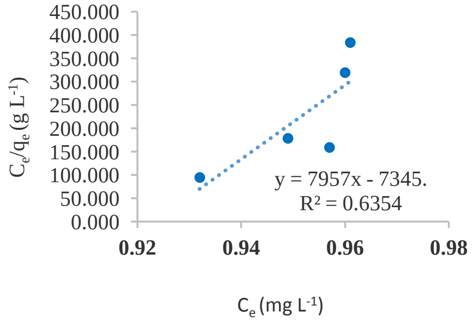


(c)

Figure 12 Langmuir isotherm for Pb^{2+} ion from three industrial wastewater samples onto the dry biomass (a) dry cell battery factory (b) nickel plating factory and (c) leather factory

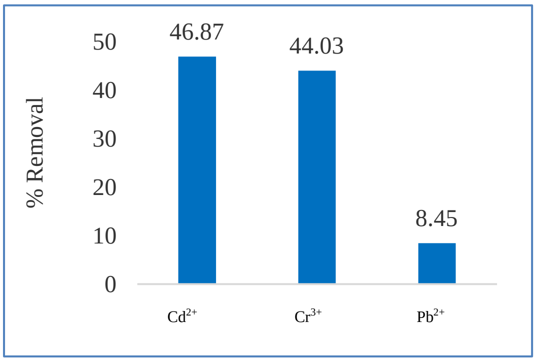
Removal of Metal Ions from Three Industrial Wastewater Samples by Using Dry Biomass

In this study, the optimum time was 90 min for the removal of Cd^{2+}, Cr^{3+} ions and 15 min for the removal of Pb^{2+} ion from dry cell battery, nickel plating and leather factories wastewater samples. The optimum loading weight of dry biomass at 0.35 g was used for the removal of Cd^{2+}, Cr^{3+} ions whereas the loading weight at 0.25 g was used for the removal of Pb^{2+} ion.

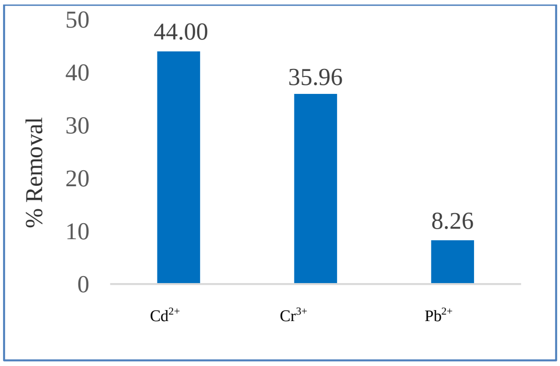
As shown in Table 7 and Figure 13, dry biomass could remove metal ions, 46.87 % of Cd^{2+}, 44.03 % of Cr^{3+} and 8.45 % of Pb^{2+} ions from dry cell battery factory wastewater sample. Moreover, dry biomass could remove metal ions, 44.00 % of Cd^{2+}, 35.96 % of Cr^{3+} and 8.26 % of Pb^{2+} ions from nickel plating factory wastewater sample. This study revealed that the dry biomass could reduce 46.99 % of Cd^{2+}, 43.39 % of Cr^{3+} and 10.83 % of Pb^{2+} ions from leather factory wastewater sample.

Table 7 Comparative Data for Removal of Metal Ions from Three Industrial Wastewater Samples Using Dry Biomass

Metal ions	Dry cell battery factory			Nickel plating factory			Leather factory		
	Before treatment	After treatment	Removal (%)	Before treatment	After treatment	Removal (%)	Before treatment	After treatment	Removal (%)
Cd^{2+}	0.064	0.034	46.87	0.050	0.028	44.00	0.583	0.309	46.99
Cr^{3+}	0.604	0.338	44.03	0.570	0.365	35.96	0.053	0.030	43.39
Pb^{2+}	0.142	0.130	8.45	0.109	0.100	8.26	0.120	0.107	10.83



(a)



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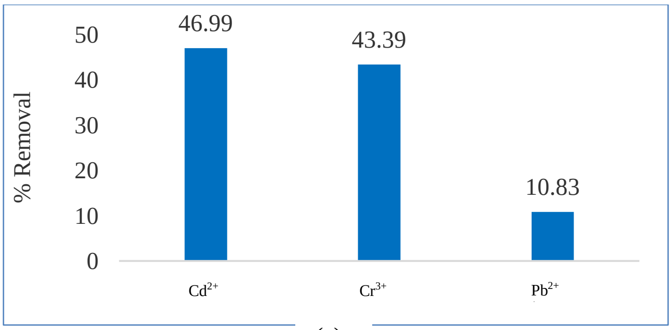

(c)

Figure 13 Removal of metal ions from three industrial wastewater samples using dry biomass (a) dry cell battery factory (b) nickel plating factory and (c) leather factory

Conclusion

In this research work, the reduction of the concentration of metal ions (Cd^{2+}, Cr^{3+} and Pb^{2+}) from three industrial wastewater samples using dry biomass was studied. Initially, aquatic plant namely *Hydrilla verticillata* (L.F.) Royle was introduced as dry biomass to study the sorption of heavy metal ions from wastewater sample under different times and different loading weights. The experimental conditions (contact times 5, 10, 15, 30, 60 and 90) min and loading weights (0.20 g, 0.25 g, 0.30 g, 0.35 g and 0.40 g) were used for the process of removal metal ions from dry cell battery, nickel plating and leather factories wastewater samples. It was found that the optimum time for removal of Cd^{2+}, Cr^{3+} and Pb^{2+} ions were 90 min, 90 min and 15 min, respectively using dry biomass. The loading weight of dry biomass at 0.35 g showed that the highest removal percent of Cd^{2+} and Cr^{3+} ions from three industrial wastewater samples. And then, the loading weight of biomass at 0.25 g showed that the highest removal percent of Pb^{2+} ion from three industrial wastewater samples. In accordance with Langmuir isotherm, the linear coefficient values, R^2 were found to be 0.9105, 0.9211 and 0.9184 for Cd^{2+} ion, 0.9985, 0.9868 and 0.9985 for Cr^{3+} ion and 0.1760, 0.5884 and 0.6354 for Pb^{2+} ion from dry cell battery factory, nickel plating factory and leather factory wastewater samples respectively. According to the results given by Langmuir isotherm, the sorption of Cd^{2+} and Cr^{3+} ions were fitted to the Langmuir model but the experimental data for the Pb^{2+} was not fitted to the Langmuir model. Then, the dry biomass was studied for the removal of selected metal ions from three industrial wastewater samples at their optimum experimental conditions. The results revealed that the dry biomass could remove 46.87 % (Cd^{2+}), 44.03 % (Cr^{3+}) and 8.45 % (Pb^{2+}) ions of wastewater sample from dry cell battery factory. Furthermore, the percentage of metal ions removed by the dry biomass decreased up to 44 % (Cd^{2+}), 35.96 % (Cr^{3+}) and 8.26 % (Pb^{2+}) ions of wastewater sample from nickel plating factory. In addition, the dry biomass could remove 46.99 % (Cd^{2+}), 43.39 % (Cr^{3+}) and 10.83 % (Pb^{2+}) ions of wastewater sample from leather factory. The removal order of metal ions in wastewater samples from dry cell battery factory, nickel plating factory and leather factory using the dry biomass were found to be $Cd^{2+} > Cr^{3+} > Pb^{2+}$.

According to the experimental data, the dry biomass could be applied in the wastewater treatment for removal of Cd^{2+} and Cr^{3+}.

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