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Full Length Research Paper

Removal of heavy metals (Pb^{2+}, Cu^{2+} and Cr^{3+}) from aqueous solutions using five plants materials

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The hazardous ill effects of heavy metals on the environment and public health is a matter of serious concern. Biosorption is emerging as a sustainable effective technology. Heavy metals in water resources are one of the most important environmental problems of countries. The intensification of industrial activity and environmental stress greatly contributes to the significant rise of heavy metal pollution in water resources making threats on terrestrial and aquatic life. The toxicity of metal pollution is slow and interminable, as these metal ions are non bio-degradable. The aim of this study was to investigate the removal of heavy metals (Pb^{2+}, $Cu^{2+}$and Cr^{3+}) using five plants materials. These materials were used as sorbent for the removal of Pb, Cu and Cr ions from aqueous media. The five plants were collected different region in Ardabil province and after being dried, were cut into small pieces of size 5 mm. The materials were washed with distilled water, filtered and finally dried overnight at 60°C. They were homogenized to mesh 100 in a mortar and bolter and subsequently used for adsorption experiments. Adsorption studies were performed by batch experiments as a function of process parameters (such as sorption time and pH). Freundlich model fitted best with the experimental equilibrium data among the all tasted adsorption isotherm models. Experimental results showed that the best pH for adsorption was 4 and time was 60 min. This study also showed that absorbent prepared from *Arnebia linearifolia* and *Artemisia fragrans* species were suitable adsorbent to remove the Pb and Cr heavy metal ions.

Key words: Plant material, heavy metals, adsorption, removal of metals, adsorption isotherm.

INTRODUCTION

Heavy metals in water resources are one of the most important environmental problems of countries. Due to modernization, the industrial use of metals especially, heavy metals, has risen alarmingly, thus becoming of prior concern because of their toxicity to flora and fauna. Recovery of heavy metals from industrial waste streams is becoming increasingly important as society realizes the necessity for recycling and conservation of essential metals (Jothinagi and Anbazhagan, 2009).

The intensification of industrial activity and environmental stress greatly contributes to the significant rise of heavy metal pollution in water resources making threats on terrestrial and aquatic life. The toxicity of metal pollution is slow and interminable, as these metal ions are non bio-degradable.

Until now, various methods of removing of these metals have been considered, including the use of low price materials (Devaprasath et al., 2007; Al- Subu et al., 2001; Mehrasbi et al., 2008; Uzun and Guzel., 2000). The use of natural materials for heavy metal removal has become a concern in all countries. Natural materials that are available in large quantities or certain waste from agricultural processes may have potential as low cost adsorbents, as they represent unused resources, widely available and are environmentally friendly (Afkhami et al., 2008). To see the decrease of polluted water, most researches were concentrated with treatment of heavy metals from industrial wastewater. This uses normal material to removal metals from different sides because it is valid largely in agriculture processes in addition to their low price as adsorbent materials (Deans and Dixon, 1992).

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Table 1. Effect of contact time of 5 plants (H_1, H_2, H_3, H_4 and H_5) on the removal percent of Pb^{2+}, Cu^{2+} and Cr^{3+} ions.

Time (min)	Pb					Cu					Cr				
	H_1*	H_2	H_3	H_4	H_5	H_1	H_2	H_3	H_4	H_5	H_1	H_2	H_3	H_4	H_5
30	52.1	48.9	51.3	49.3	49.1	49.7	47.2	39.9	24.1	51.1	33.2	34.9	32.9	12.5	14.3
45	84.5	73.7	82.2	80.7	81.1	69.1	61.3	53.7	34.5	63.5	49	47.6	48.2	16.4	17.9
60	96	86.1	95.7	93.7	93.9	76.7	70.2	58	39	72.2	54.3	54.3	56.7	18.2	19.7
90	96.1	86.1	95.7	93.6	93.9	76.8	70.3	58	39.1	72.2	54.4	54.3	56.7	18.3	19.7

*H_1= *Arnebia linearifolia*, H_2 = *Phlomis aucheri,* H_3 = *Artemisia fragrans,* H_4 = *Zosimia absinthifolia* and H_5= *Hypericum scabrum.*

Chromium, lead and copper are in the waste waters of industries such as, electroplating, plastic and paint manufacturing, mining, metallurgical processes, petrochemical processes, batteries, paper and pulp and e-wastes. Higher concentration of copper causes neurotoxicity called Wilson's disease. Cadmium is carcinogenic in nature. Lead has been the most dreaded toxic heavy metal causing anaemia, encephalopathy and hepatitis. Conventional procedures include chemical precipitation, lime coagulation, ion exchange, reverse osmosis and solvent extraction which are cost prohibitive and are associated with slurry formation hindering filtration. Adsorption onto activated carbon is very effective method of metal removals but its use is limited because of cost. Biosorption is emerging as a sustainable effective technology using low cost biosorbent such as bamboo dust (Kannan and Veemraj, 2009), saw dust (Adouby et al., 2007), algae (Lodiero et al., 2006), silk cotton hull (Shanmugavalli et al., 2009).

This study aimed to investigate the adsorption potential of 5 plants (*Arnebia linearifolia, Phlomis aucheri, Artemisia fragrans, Zosimia absinthifolia* and *Hypericum scabrum*) for the removal of Pb^{2+}, Cu^{2+} and Cr^{3+} ions from wastewater within various experimental conditions. At the same time, this can be considered a way of recycling such environmentally harmful materials.

MATERIALS AND METHODS

The five plants H_1 to H_5 were collected from different region in Ardabil province and after being dried were cut into small pieces of size 5 mm. The materials were washed by tap water, then filtered and finally dried overnight at 60°C. They were homogenized to mesh 100 in a mortar and bolter and subsequently used for adsorption experiments. Adsorption studies were performed by batch experiments as a function of process parameters (such as sorption time and PH). Freundlich model fitted best with the experimental equilibrium data among the all tasted adsorption isotherm models.

Experimental reagents and apparatus

Reagents used were lead standard solution 1000 ppm (Merck), copper standard solution 1000 ppm (Merck) and chromium standard solution 1000 ppm (Merck), sodium hydroxide (Merck), sulphoric acid (Merck). A Varian (240AA) flame atomic absorption spectrophotometer (air/acetylene flame) was used for metal ions determinations. A Metrohm (model 780) digital pH meter equipped with a combined glass electrode was used for the pH adjustments. IKA stirer and centrifuge were also used.

Preparation of synthetic wastewater

The mixed metal ions solution from cations (Pb^{2+}, Cu^{2+} and Cr^{3+}) was prepared from Merck – analytical grade stock standard of concentration 1000 ppm. The solution of wastewater was introduced for each metal according to the method of continuous dilution from the sample solution earlier mentioned. The pH of the wastewater was adjusted by using HNO_3 1% (V/V) and/or NaOH. The final concentration of metal ions in wastewater was analyzed by atomic absorption spectrophotometer type Varian – 240AA.

Bach sorption experiment

Capped tubs containing 50.0 ml of solution test of metal ions with known ion concentration was mixed with a known amount of dried untreated plant and the capped tubs were thoroughly mixed, allowing sufficient time for adsorption equilibrium. The mixtures were filtered through filter paper Whatman no. 1 and the metal ions were determined in the filtrate solution by atomic absorption spectrophotometer type Varian – 240AA. Each experiment was carried out at room temperature and was repeated two times, and the results are given as averages.

Estimation of removal percentage of metal ions by plant materials

Amount of removed material by plants through series of batch investigations were determined by the following equation: Removal %= $[(C_o - C_f)/ C_o] \times 100$, where C_o and C_f are the initial and equilibrium concentration (ppm) of metal ions in solution, respectively.

RESULTS AND DISCUSSION

The effect of different operating conditions (contact time and pH) on the removal by adsorption onto plant materials from *A. linearifolia*, *P. aucheri, A. fragrans, Z. absinthifolia* and *H. scabrum* were investigated. The metals removal studies are shown in Tables 1 and 2,

Table 2. Effect of pH on the removal percent (%) of Pb^{2+}, Cu^{2+} and Cr^{3+} ions by 5 plants (H_1, H_2, H_3, H_4 and H_5).

pH	Pb					Cu					Cr				
	H_1*	H_2	H_3	H_4	H_5	H_1	H_2	H_3	H_4	H_5	H_1	H_2	H_3	H_4	H_5
2	95.1	83.9	93.5	92.9	90.1	74.8	69.3	57.5	36.9	71.5	53.3	51.2	55.8	18.1	19.5
4	96	86.1	95.7	93.7	93.9	76.7	70.2	58	39	72.2	54.3	54.3	56.7	18.2	19.7
6	92.8	84.1	92.1	91.4	92.6	75	68.9	56.3	38.7	70.5	51.1	53.7	54.1	17.8	19.4

*H_1= *Arnebia linearifolia*, H_2 = *Phlomis aucheri,* H_3 = *Artemisia fragrans,* H_4 = *Zosimia absinthifolia* and H_5= *Hypericum scabrum*

which indicated that their removal was strongly affected by the different operating conditions.

Effect of contact time

Adsorption of Pb, Cu and Cr ions were measured at given contact times for initial Pb^{2+}, Cu^{2+} and Cr^{3+} concentrations of 10 mg/L. The plot revealed that the rate of percent lead, copper and chromium removal are higher at the beginning. This was probably due to larger surface area of the plants being available at beginning for the adsorption of Pb^{2+}, Cu^{2+} and Cr^{3+} ions. As the surface adsorption sites become exhausted, the uptake rate was controlled by the rate at which the adsorbate is transported from the exterior to the interior sites of the adsorbent particles. Most of the maximum percent lead, copper and chromium removal was attained after about 90 min of shaking time.

Effect of pH on the removal of the heavy metal ions

The adsorption of Pb^{2+}, Cu^{2+} and Cr^{3+} ions were found to be strongly dependent on the pH of the solution. It was demonstrated that the optimum pH for the adsorption Pb^{2+}, Cu^{2+} and Cr^{3+} ions were about 4 which were rather acidic. At low pH (below 3), there was excessive protonation of the active sites at H_1-H_5 powder surface and this often refuses the formation of links between Pb^{2+}, Cu^{2+} and Cr^{3+} ions and the active site. At moderate pH values (3 to 6), linked H^+ is released from the active sites and adsorbed amount of metal ions is generally found to increase (Table 2). Moreover, at higher pH values (above 6), the precipitation was dominant or both ion exchange and aqueous metal hydroxide formation may become significant mechanisms in the metal removal process. This condition is often not desirable as the metal precipitation could result in copper and chromium having confusion for the adsorption capacity. In practice, metal precipitation is generally not a stabilized form of heavy metal as the precipitation can sometimes be very small in size, and upon the neutralization of the effluent from the wastewater treatment plant, the solubility of the metals increases resulting in a recontamination of the waste outlet stream.

Conclusion

Based on the present study, it could be concluded that some low cost materials like plants can be used efficiently in the removal of heavy metal ions (Pb^{2+}, Cu^{2+} and Cr^{3+}) from aqueous solutions. The removal of heavy metal ions was pH dependent as the adsorption capacity increases with increasing the pH value of the solution, and at a particular pH the order of increasing the removal percentage was $Pb^{2+} > Cu^{2+} > Cr^{3+}$. Experimental results showed that the best pH for adsorption was 4 and time was 60 min. The metal ions showed different behaviors towards adsorption on plants by increasing the initial concentration of the metal ions. This investigation also showed absorbent prepared from H_1 and H_3 species to be suitable adsorbent for removing the Pb heavy metal ions.

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