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Biosorption of Cadmium, Lead, and Uranium by Powder of Poplar Leaves and Branches

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Abstract The removal of metal ions from aqueous solutions by biosorption plays an important role in water pollution control. In this study, dried leaves and branches of poplar trees were studied for removing some toxic elements (cadmium, lead, and uranium) from aqueous solutions. The equilibrium experiments were systematically carried out in a batch process, covering various process parameters that include agitation time, adsorbent size and dosage, initial cadmium, lead and uranium concentration, and pH of the aqueous solution. Adsorption behavior was found to follow Freundlich and Langmuir isotherms. The results have shown that both dried leaves and branches can be effectively used for removing uranium, while only branches were found to remove lead and cadmium completely from the aqueous solution. The maximum biosorption capacity of leaves for uranium was found to be 2.3 mg g^{-1} and 1.7 mg g^{-1} and 2.1 mg g^{-1} for lead and cadmium on branches, respectively. In addition, the studied biomass materials were used in removing lead and cadmium from contaminated water and the method was found to be effective.

Keywords Poplar · Uranium · Cadmium · Lead · Biosorption · Wastewater · Pollution

Introduction

The rapid development of various industries has produced large quantities of wastes containing heavy metals that are directly or indirectly discharged into the water environment [1, 2]. Heavy metal pollution has become one of the most environmental and health problems and many attempts to reduce their emissions to environment or remove them from the environment have been studied. Conventional methods for removing metal ions from aqueous solutions include chemical precipitation, ion exchange, electrochemical treatment, membrane technologies, and adsorption on activated carbon [3–5]. These methods are sometimes ineffective, especially when metal ion concentration in aqueous

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solution is as low as 1 to 100 mg l^{-1}. In addition, large quantities of sludge which require treatment and disposal are generated. Ion exchange, membrane technologies, and activated carbon adsorption processes are extremely expensive for treating large quantities of water and wastewater containing heavy metal in low concentration. In recent years, biosorption using natural materials has emerged as a cost-effective and efficient alternative for the removal of heavy metals from low concentration heavy metal wastewaters [1, 6]. This novel approach is competitive, effective, cheap, available in large quantities, and environment friendly. In addition, biosorbents can eliminate heavy metal ions effectively from dissolved metal ions from complex solutions containing very low concentration (10^{-6} and 10^{-9} mg g^{-1}) [1, 6]. Many biosorbents materials such as bacteria, fungi, yeast, and algae have been successfully used for removing heavy metals from aqueous solutions [6–20]. Other materials such as rice husk, maize cobs, coconut and seed hull [21], saw dust [22], maize leaf [23], and olive pomace [24] were studied for heavy metal removal from water bodies. Tae-Young et al. [15] found that the adsorption capacity for lead, copper, and cadmium on brewery yeast reached a value of 96.4 mg g^{-1}, 48.9 mg g^{-1}, and 14.3 mg g^{-1}, respectively. *Larrea tridentate*, a common desert plant, was used to remove toxic heavy metal ions such as copper, cadmium, lead, zinc, nickel, and chromium from contaminated waters [25], while others have used dried lichens for uranium removal from aqueous solutions [26]. In addition, the biosorption capacity of coconut shell powder was studied for different toxic metal ions (Cr, As, and Cd) [27]. While *Saraca indica* leaf powder removed 95.37% of Pb at the pH 6.5 [28]. In addition, Mahvi et al. [29] reported that cadmium uptake was found to be rapid and reached 85% and 92% for both *Ulmus* leaves and their ash, respectively [29]. On the other hand, agricultural by-products such as peat, wood, pine bark, banana pith, soybean and cotton seed hulls, peanut, shells, hazelnut shell, rice husk, sawdust, wool, orange peel, and compost and leaves have been widely used for metal removal from water [11, 24, 30–34].

Poplar (*Populus,* sp.) trees are widely distributed in Syria growing to a height of 20 m with over 0.6 m trunk diameter. It is normally fairly short-lived, but some trees may live for up to 400 years. Poplar wood contains high percentage of lignin (18.1%) and cellulose (49.9%) [35] that are known as biopolymers and admittedly associated with the removal of heavy metals [36]. Therefore, the aim of the present work was to investigate the sorption of U, Cd, and Pb from aqueous solutions by the powder of dried biomass of leaves and branches of the poplar tree.

Material and Methods

Biosorbent Material

The biomass materials were prepared as follows: leaves and branches were collected from poplar trees grown near Damascus city (5 kg from each part). The leaves and branches were washed several times with distilled water and then divided into two subsamples; the first sample was dried at room temperature and the second sample was dried in a vacuum drying oven at 80 °C. The dried biomass was grounded with a mortar and pestle and homogenized. The sample was then passed through standard testing sieves with different particle sizes (200 and 800 µm), stored in a sealed bottle with a silica gel to prevent re-adsorption of moisture.

U, Pb, and Cd concentrations in the collected branches and leaves were determined using the analytical techniques mentioned below and found to be 0.11±0.04, 504±45, and 627±29 mg kg^{-1}, while U and Pb concentrations in leaf samples were 0.042±0.004 and 475±22 mg kg^{-1}, respectively.

Biosorption Experiments

Heavy metal solutions were prepared by dissolving the metal chloride salts (UCl_6, $PbCl_2$, and $CdCl_2$) in distilled water. The mixture containing 1 g of biosorbent and 100 ml of metal chloride solution was agitated on a shaking incubator at 150 rpm at 25 °C. Heavy metal concentrations used in this study varied between 1–10 mg l^{-1}; the toxicity level of Pb and U in water falls within 1–10 mg l^{-1} [19] and for Cd 0.001–0.1 mg l^{-1} [1]. After equilibrium, the biosorbent materials were separated from the solution by filtration using 0.45-μm filters and the residual metal concentration in the solution was analyzed. The amount of U, Pb, and Cd adsorbed by the biomass materials at equilibrium were calculated from the following mass balance equation:

$$q = (C_i - C)\frac{V}{W} \tag{1}$$

where q is the equilibrium amount adsorbed on the biomass (mg g^{-1}), C_i is the initial concentration of bulk fluid (mg l^{-1}), C is the equilibrium concentration of the solution (mg l^{-1}), V is the volume of solution (l) and W is the weight of biomass (g).

The equilibrium of a solute between the liquid and solid phases may be described by various models of sorption. In order to investigate the sorption capacity and isotherm, two equilibrium models have been used, namely, Langmuir and Freundlich.

Langmuir Isotherm

The Langmuir model is probably the best known and most widely applied sorption isotherm [15, 30]. This model supposes a monolayer sorption with a homogeneous distribution of sorption sites and sorption energies, without interactions between the sorbed molecules. It has produced good agreement with a wide variety of experimental data and may be represented as follows [27]:

$$q = \frac{q_{max}\, bC_f}{1 + bC_f} \tag{2}$$

where, C_f is the equilibrium concentration (mg l^{-1}), q is the amount of metal ions sorbed (mg g^{-1}), q_{max} is a maximum amount of metal ion per unit weight of biomass (mg g^{-1}) and b is a sorption equilibrium constant.

The Eq. 1 can be rearranged to the following linear form:

$$\frac{1}{q} = \frac{1}{q_{max}}\left(\frac{1}{b}C_f + 1\right) \tag{3}$$

Freundlich Isotherm

This empirical model can be applied to non-ideal sorption on heterogeneous surfaces as well as multilayer sorption and is expressed by Eq. 4 [27]. The Freundlich isotherm is also more widely used but provides no information on the monolayer adsorption capacity, in contrast to the Langmuir model.

$$q = k_f C^{\frac{1}{n}} \tag{4}$$

where, C is the equilibrium concentration (mg l^{-1}), q is the amount of metal ions sorbed (mg g^{-1}) and k_f and n are Freundlich's adsorption constants.

The Eq. 4 is frequently used in the linear form by taking the logarithm of both sides of Eq. 4 as follows:

$$\log q = \log k_{\mathrm{f}} + \frac{1}{n} \log C \qquad (5)$$

Temperature has an influence on the biosorption of metal ions, but to a limited extent under a certain range of temperature.

Determination of Parameters Affecting the Absorption Process

Effect of pH on Cd, Pb and U Removal

The pH of solution was adjusted at 4, 7, and 10 with 1 N HNO_3 and 1 N NH_4OH, in 250-ml Erlenmeyer flasks. Each flask initially contained 1 g dry wt. biomass, 100 ml metal solution, and 2 mg l^{-1} metal and was shaken for 4 h at 25 °C. The mixtures were filtered through 0.45-μm membrane filters and then the residual metal concentration in the solution was analyzed.

Effects of Temperature on Cd, Pb, and U Removal

Flasks containing 100 ml metal solution (2 mg l^{-1}) and 1 g dry wt. biomass were shaken for 4 h. Four different temperatures were tested, ranging from 5 °C to 35 °C. The mixtures were filtered through 0.45-μm membrane filters and then the residual metal concentration in the solution was analyzed.

Effects of Biomass Concentration on Cd, Pb, and U Removal

The effect of 1, 2, 5, and 10 g l^{-1} of biomass concentration on metal removal from metal solution (2 mg l^{-1}) were evaluated in batch tests. The experiments were shaken for 4 h at 25 °C. The mixtures were filtered through 0.45-μm membrane filters and then the residual metal concentration in the solution was analyzed.

Analytical Methods

The filtrates resulting from leaching experiments were evaporated to near dryness and digested using a combination of mineral acids (nitric, hydrofluoric, and hydrochloric acid) for at least 24 h. Uranium was then separated from the sample using ion exchange column (Dowex 1×4) and measured using Fluorometry instrument (Jerrel-Ash 27000, Advanced Technical Services Gmbh Swiss). The accuracy of this method is 97% and the limit of detection is 1 μg l^{-1} for water samples.

Lead and cadmium were measured using differential pulse anodic stripping voltammetry (DPASV) at the static mercury drop electrode (SMDE) by the method described in Khandekar et al. [37] using 693 VA Processor and 694 VA Stand; (Metrohm, Switzerland). Optimum conditions for the anodic stripping of Pb and Cd were determined using CH_3COONH_4 dilute HNO_3 buffer solution (pH 4) as an electrolyte. The accuracies of these methods are 95% and 92% for Cd and Pb, respectively, and the limits of detection are 1 μg l^{-1} for Cd and Pb for water samples.

Sampling of Wastewater

Two water samples were collected from battery factory disposal site and disposal sites of phosphogypsum piles. U, Cd, and Pb were determined in these two samples.

Results and Discussion

Biosorption Capacity

Table 1 shows values of Langmuir and Freundlich constants for biosorption of U, Cd, and Pb by poplar leaves and branches. Langmuir constants were calculated from Eq. 2 after plotting the relationship between $1/C_f$ and $1/q$, while Freundlich constants were calculated from Eq. 4 after plotting the relationship between log (C) and log (q). Table 1 shows that leaves of poplar can be used for removing U from water (q_{max}=2.3 mg g^{-1}), and poplar branches can be used for removing Cd and Pb (q_{max}=2.1 and 1.7 mg g^{-1}, respectively). These impute to n value where $0<1/n$ (0.83, 0.77, and 0.91 for U, Pb, and Cd, respectively) <1 gives high affinity of biomass to metal. Even through, the capacities of poplar branches and leaves are low in comparison to other biomasses reported in other studies; the values seem to be suitable for removing such ions from natural waters with high concentration. On the other hand, leaves seems to be not effective in removing Cd and Pb from water (q_{max}=−1.3 and −0.1 mg g^{-1}, respectively). This means that Cd and Pb transferred from leaves to water. Table 2 shows a comparison of U, Cd, and Pb uptake capacities of various types of biomasses. It is clear that q_{max} values in this work were much lower than those reported for biomass materials and this may be due to low initial concentrations of the metals employed in the isotherm study.

Effect of Particle Size and Drying Temperature

Particle Size

Particle size is an important parameter that influences metals uptake on biomass. The results presented in Fig. 1 show a reduction in the biosorption of U, Cd, and Pb with particle size of leaf biomass. It is important to stress that larger particles, in general, present higher external mass transfer than smaller particles [1]. In this case, higher metal sorption from

Table 1 Sorption isotherm coefficients of Langmuir and Freundlich models applied to U, Cd, and Pb adsorption by powder of leaves and branches of poplar.

Metal	Poplar part	Langmuir		Freundlich		
		q_{max} (mg g^{-1})	R^2	k_f	1/n	R^2
U	Leaves	2.3	0.91	8	0.83	0.98
	Branches	0.4	0.17	70	1.25	0.62
Pb	Leaves	-0.1	0.87	2×10^{-5}	−2.50	0.47
	Branches	1.7	0.25	23	0.77	0.83
Cd	Leaves	-1.3	0.40	250	1.25	0.96
	Branches	2.1	0.88	29	0.91	0.66

Table 2 A comparison of U, Cd, and Pb maximum biosorption capacity values (mg g^{-1}) of various biomass.

Adsorbent	Maximum uptake capacities			Reference
	Cd	Pb	U	
Brewery yeast	14.3	48.9	–	[15]
Larrea tridentate	36.4	9.61	–	[25]
Coconut shell	295.8	–	–	[27]
Rice husk	0.32	–	–	[32]
M. Spicatum	–	46.69	–	[10]
Dried lichens	–	–	42	[26]
Poplar (leaves)	–	–	2.3	Present study
Poplar (branches)	2.1	1.7	0.4	Present study

these particles is attributed to mass transport inside the sorbent particles, while there is no practical size effect on uptake of Cd by branches (Fig. 1).

Drying Temperature

Table 3 shows the effect of drying temperature on removing metals. Drying leaves at room temperature were found to have more effect on removing the studied metals than oven drying temperature (80 °C) and this is due to the fact that leaves contain high concentration of glycolipids. In the case of metals, the anionic biosurfactant of glycolipids carries a negative charge therefore an ionic strong bond is formed [38]. In addition, these glycolipids are easily destroyed when they are heated so the adsorption will be lower.

Effect of Initial Weight of Biomass

Metal-ion uptake per gram of biosorbent increases as long as the biosorbent is not saturated [39]. In addition, the uptake values depend on the nature and origin of the biosorbent itself [40]. In the present study, sorption of U and Pb increased with increasing the leaf amount (Table 4). This is due to the fact that the glycolipids increase when the mass of biosorption increases, so more biomass is required for concentrated solutions, such as liquid wastes. While the biosorption capacities reached maximum values when the branches were used so no differences were observed.

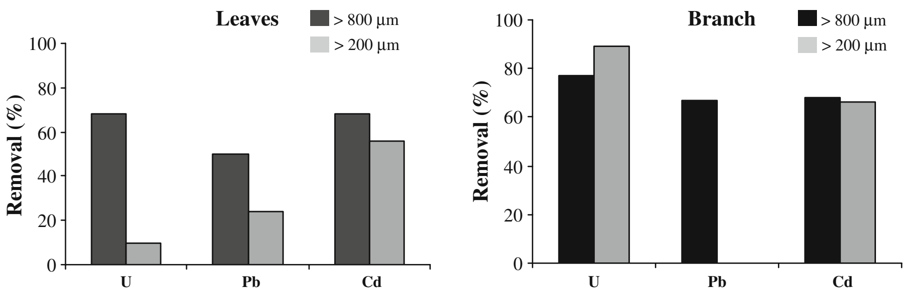

Fig. 1 U, Pb and Cd removal by different particle size of poplar tree parts

Table 3 The effect of plant drying temperature on removing metals.

Poplar part	Drying temp. (°C)	Removal (%)		
		U	Pb	Cd
Leaves	25	55	47	75
	80	33	16	65
Branches	25	87	84	65
	80	90	84	70

The error of measurement is 10% at 1 σ

Effect of pH

Different initial pH values of the solutions (4, 7, and 10) and using a constant amount of plant powder (1 g) have been used to investigate pH effect at room temperature. The highest removal capacities for U and Pb using leaves and branches powder were observed for acidic solution (pH=4) (Table 5), while cadmium removal (by leaves) increased from 68% at pH 4 to 86% at pH 7 and from 68% at pH 4 to 75% at pH 7 where branches were used. At a pH value of 10, cadmium ions precipitate as $Cd(OH)_2$ hence, Cd can not be biosorbed by plants [27].

Effect of Temperature

Biosorption process is usually not operated at high temperature because it will increase the operational cost [2]. The influence of temperature on U, Pb, and Cd removal capacities using different parts of poplar trees is shown in Fig. 2. The maximum U and Cd removal capacity using leaves occurred at a temperature of 25 °C while the maximum U, Pb, and Cd removal capacities using branches occurred at a temperature of 35 °C.

Effect of Contact Time

Generally, the biosorption capacity and the removal efficiency of metal ions by a biomass became higher with prolonging the contact time. However, in practice, it is necessary to

Table 4 Influence of biomass of poplar on metals removal.

Poplar part	Water losing (%)	Biomass wt. (g)	Removal (%)		
			U	Pb	Cd
Leaves	15	1	33	16	65
	14	2	38	39	73
	29	5	82	41	62
	35	10	–	47	75
Branches	13	1	90	84	70
	33	2	83	84	68
	38	5	85	85	79
	60	10	92	84	65

The error of measurement is 10% at 1 σ

Table 5 Influence of pH of poplar on metals removal.

Poplar part	pH	Removal (%)		
		U	Pb	Cd
Leaves	4	76	50	68
	7	37	7	86
	10	57	13	22
Branches	4	90	67	68
	7	47	5	75
	10	41	<4	<3

The error of measurement is 10% at 1 σ

optimize the contact time, considering the efficiency of desorption and regeneration of the biomass [2]. Figure 3 shows that the maximum uptake has occurred in the first 5 h for U using leaves powder, while the equilibrium has occurred at the first hour for removing U by poplar branches. In addition, the maximum Cd uptake has occurred in the first 10 h using leaf sample, while Cd removal by branches powder has increased when contact time increased to reach a maximum value at 25 h (Fig. 3). On the other hand, Pb removal by leaves powder has rapidly increased when contact time increased to reach a maximum value at 15 h then it has slowly decreased when contact time increased (Fig. 3). Different equilibrium agitation times are reported in literature for the removal of cadmium with

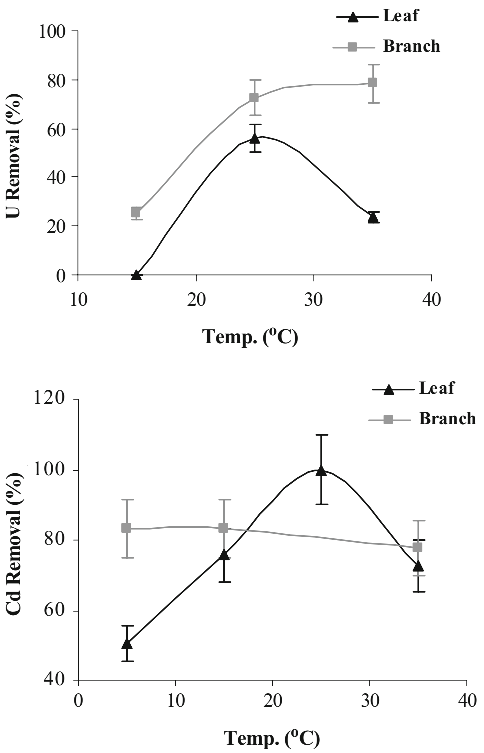

Fig. 2 Effect of temperature on U, Pb and Cd removal

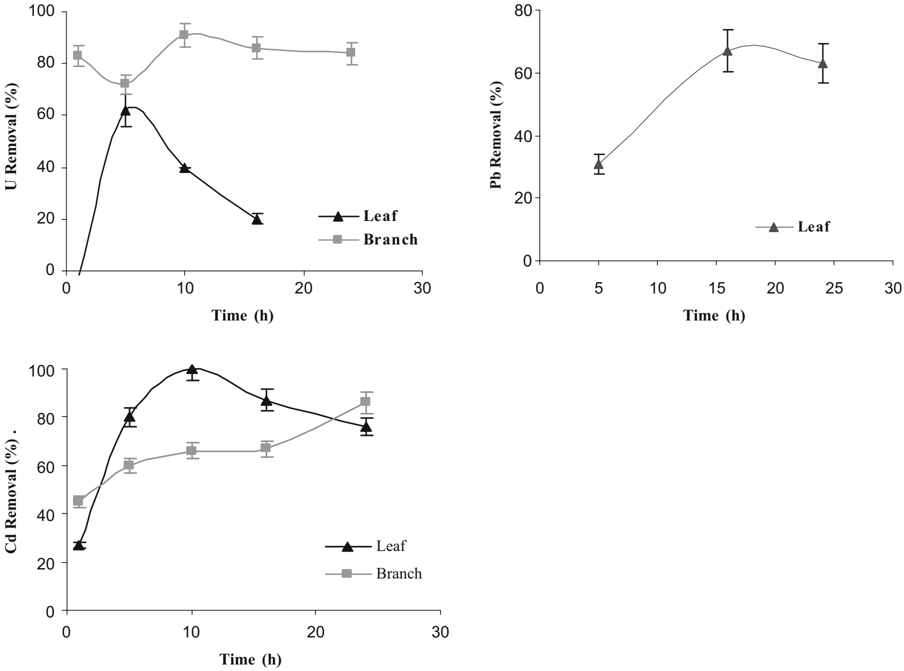

Fig. 3 Effect of contact time on U, Pb and Cd removal

powder of coconut shell [27]; the equilibrium agitation time is found to be 2 h. While Ceribasi and Yetis reported that biosorption of lead by the fungus *Phanerochaete chrysosporium* was rapid in the first 15 min and equilibrium was attained after 3 h [41].

Effect of Competing Ions/Co-Ions

Real industrial effluent usually contains various ionic components, including metal cations and anions. Some studies indicated that cations and anions additional to the ions of interest have a generally detrimental impact on metal accumulation [19]. The metal uptake by non-living biomass is affected significantly by the presence of other cations in solution, depending on the chemical interactions of the other chemical species (co-ions) with the metal of interest and the biomass [42–44]. Many of the functional groups present on the cell wall and membrane are nonspecific and different cations compete for the binding sites, so the biosorption capacity of one metal ion is interfered and reduced by co-ions, including other metal ions and anions presenting in solution; however, the gross uptake capacity of all metals in solutions remains almost unchangeable.

Light metal ions, such as Ca^{2+}, Na^{+}, and K^{+} are present in industrial wastewater. The experimental data has shown that light metal ions and Fe^{3+} have different effect on U, Pb, and Cd biosorption (Fig. 4). As an example, the presence of K^{+} did not have any adverse influence on ions of Pb, while the biosorption capacity of Cd has been decreased in the presence of Fe^{3+}. On the other hand, the presences of anions also affect the biosorption of metal ions. Kapoor and Viraraghavan [10] reported that the biosorption capacity decreased in the presence of sulfate, chloride, phosphate, carbonate, glutamate, citrate, and pyrophosphate. The anions existing in the solution may form a complex with the metal

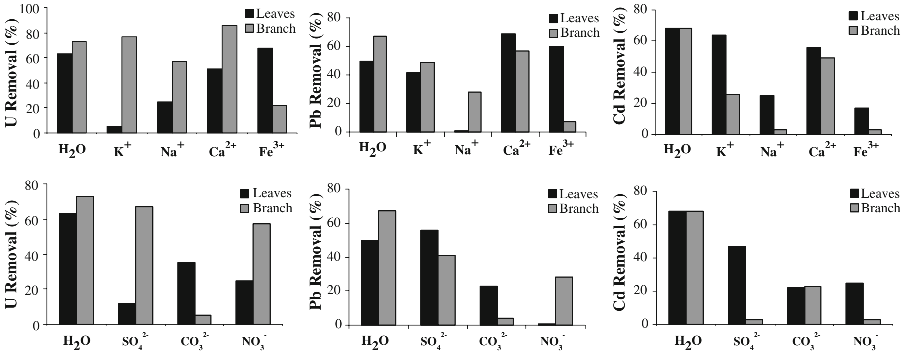

Fig. 4 Effect of competing ions on biosorption

ions, which would reduce the metal biosorption capacity seriously [10]. So the biosorption capacity of Pb was decreased in the presence of CO_3^{2-} and NO_2^{-}, while the presence of SO_4^{2-} and NO_2^{-} were found to have no effect on U (Fig. 4).

Application of Biosorption Technology

In general, there are no big scale applications of biosorption technology in wastewater treatment, it is necessary to continue to explore the various aspects relevant to the application. So, two types of industrial wastewaters were collected and treated using the proposed method. Initial concentration of Pb and Cd were first determined, the biosorption technology was then applied for removing Pb and Cd using branches and leaves of poplar. The results have shown that Pb and Cd removal from the solution can reach 90% (Table 6). More large scale experiments are required.

Conclusions

The results presented in this work showed that dried leaves and branches of poplar tree can be good potential biomass materials for removing U, Pb, and Cd from aqueous solutions. Dried leaves at room temperature are more effective in removing the studied metals than the oven dried materials at 80 °C. The uptake of U, Pb, and Cd by poplar branches and leaves from aqueous solutions depends on time of incubation, temperature, pH, particle size of biosorbent, and the concentration of co-ions. In addition, sorption of U and Pb is with leaves powder amount and both metals are removed with high capacities from acidic

Table 6 Application of biosorption technology on real samples.

Source of the sample	Metal	Initial con. (mg l^{-1})	Removal (%)	
			Leaves	Branches
Pb-battery factory disposal site	Pb	0.17±0.01	100	100
Disposal sites of Phosphogypsum piles	Cd	1.65±0.1	95	92

solution (pH=4) using both leaves and branches powders. Moreover, the best temperature for removing the studied metals ranged between 25 and 35 °C. Furthermore, the dried leaves and branches of poplar tree could be potential biomass materials for treatment of industrial wastewater resulting from phosphate fertilizers and the lead battery factories.

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