**Phytoremediation in Removing Selected Heavy Metals from Aqueous Solutions**

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**Abstract**

Bioaccumulation of *Ulothrix zonate* was compared for cadmium, copper, nickel, and zinc removal at various concentrations and contact time. The results showed that lowest amount of Cd, Cu, Ni and Zn was adsorbed when the initial heavy metal concentration was 10 ppm. whereas Cd showed highest concentration at 20 ppm and Cu exhibited greatest removal at 30 ppm. Ni and Zn highest removals were at 40 ppm The value of Freundlich model constant (1/n) for different metals ranged from 0.334 to 0.721 and the values of Langmuir separation factor values (RL) varied between 0.111 and 0.722 which indicated favorable biosorption by the biomass of the algae. The order of metals uptake was found to be Ni>Zn>Cu>Cd.The finding of the study showed that *U*. *zonate* has much potential as a biosorbent for the sorption of Cd, Cu, Ni and Zn and indicated that the metal uptake was concentration-independent for Cd and Zn whereas for Cu and Ni an increase in initial metal concentration resulted in higher metal uptake. This study is leading for future studies on the use of algae as a cheap bioadsorbent for heavy metals removal in Iraq which suffer from huge waste water in its land.

**Key words: Heavy Metals, Wastewater, Ulothrix zonata, Biosorbent**

**Introduction**

The contamination by heavy metals from industrial waste water to the water resources effect life significally, because heavy metal ions can accumulate in the environment and into food chains (Chevron and Costa, 2012). Therefore, the process of heavy metal removal is very important and industries should be aware of it.

Many methods tried by the researchers around the world to remove heavy metals from aqueous solutions include adsorption, biological methods, electro coagulation, electro dialysis and various membrane separation techniques among others. Biological methods are very important area of research with huge potential for research and applicability for removal of heavy metals. Various biological methods include trickling filter, biosorption, activated sludge process and various anaerobic processes.

The old technologies of heavy metal removal have several disadvantages such as high cost, chemical uses and large volume residual sludge. The phytoremediation is a new alternative method for heavy metal removal process because of low cost, low biological sludge, high efficiency and environmental friendly [Farooq *et al*., (2010), Srivastava and Majunder (2008), Abdel-Ary, *et al* ., (2013)].

The bioaccumulation process is known as an active mode of metal accumulation by living cells which depends on the metabolic activity of the cell (Volesky, 1990; Wase and Forster, 1997). Biosorption is a term that describes the removal of heavymetals by the passive binding to non-living biomass from an aqueoussolution (Davis *et al*., 2003). Algae are important agents and are already been used by many wastewater facilities. The contamination of Shatt Al-Arab, southern Iraq by heavy metals was reported by many researchers as ( Al-Saad *et al*.,1996,Al-Saad *et al*.,1997, DouAbul *et al*.,1587) and it was attributed to agricultural, industrial, anthropogenic activities around the river beside the ground water effects.

The indiscriminate discharge of chemical toxins especially Cd, Cu, Ni and Zn etc into the environment ensure their transfer into plants, animals and man, and it was also reported that high concentrations of heavy metals in irrigation waters could effects plants and crops growth, interfere with uptake of other essential nutrients or form dust deposit on fruits and render the edible portion of plants toxic to humans and grazing animals (Dan'Azumi, 2010). Algae have been proven efficient biological vectors for heavy metal uptake. Biosorption potential of two strains *Spirogyra sp.* and *Spirulina sp*. have been studied under different initial ofmetal concentrations (Mane and Bhosle 2012)..*Spirulina sp*. treated with different metal ions have been employed to understand the sorption for management of industrial waste water(Doshi *et al* 2007). Deng e*l al*.,( 2007) reported that the green algae *Cladophora fascilcularis* is highly efficient for the biosorption of copper from aqueous solution. Results showing high sorption of pb+2 from solution by biomass of commonly available filamentous green algae *Spirogyra* species ( Gupta and Rastogi,2008). Khalaf (2008) observed biosorption of textile dye from textile water by no viable biomass of *Aspergillus niger* and *Spirogyra* species. Brahmbalt *el al*., (2012) use the filamentous algae *Pithophora* species for the removal of cadmium, chromium and lead from industrial waste water. Gao and Yan (2012) observed the response of *Chara globularis* and *Hydrodictyon reliculatum* to lead pollution. Sheng *et al*., (2004) used the locally harvested brown marine algae *Sargassum soecies* and *Padina* species for the removal of cation of Cd+2and Cr+3 and anion ( Cr+2) from diluted aqueous solutions.

The high Biodegradation and biosorption capacity of some potential cyanobacterial species: *Oscillatoria* sp., *Synechococcus* sp., *Nodularia* sp., *Nostoc* sp. and *Cyanothece* sp. Dominated the effluents and mixed cultures showed varying sensitivity. Contaminants were removed by all the species either as individuals or in mixtures (Dubey *et al,* 2011). Lee and Chang(2011) observed the biosorption capacity from aqueous solutions of the green algaespecies, *Spirogyra* and *Cladophora*, for lead (Pb (II)) and copper (Cu (II)). In comparing the analysis of the Langmuir and Freundlich isotherm models, the adsorption of Pb (II) and Cu(II) by these two types of biosorbents showed a better fit with the Langmuir isotherm ). In Saudi Arabia the accumulation of heavy metals by the green algae *Chaetomorpha aerea, Enteromorpha Clathrata* and *Ulva Lactuca* were carried out to measure the level of iron, nickel, cobalt, zinc, cadmium and lead in three site of the Saudi coast of the Arabian Gulf (Al-Homaidan, 2007) .A[-Mayaly, (2011) studied the use of filamentous algae *Mougoutia sp* to remove lead from contaminated water under laboratory conditions. It was found that the algae was able to remove this metal with high efficiency.

It was found that the filamentous green algae are more efficient in removing heavy metals than micro algae like *chlorella vulgarise*. This may relate to the increasing of active sites according to the multiply of cell numbers in each filament (Nassam *et al*, 1996). Further more, the chemical nature and polarity of the adsorbent surface can influence the attractive forces between the adsorbent and adsorbate (Babel and Kurniawan, 2003). Prado *et al* (2010) observed the rate of biosorption of cadmium and copper ions by nonliving biomass of the brown macroalga *Sargassum sinicola* under saline conditions. They concluded that presence of salt did not significantly affect the rate of biosorption and there is antagonistic effect on biosorption when both these metals are present in the solution .Michael *et al*.,(2015) studied the lower and upper tolerance levels of *Cladophora* to abiotic conditions (PH ,salinity and nutrients concentrations*)*and found that *cladophora* could be a possible candidateas anew West Water Treatment tool in these experimental conditions.

**Materials and Method**

*U. zonate* was collected from waste water of the polluted Shatt Al-Arab channels ,southern of Iraq. Algae was washed under running tap water and double distilled water to remove other algae and any epiphytes and adhering foreign particles like sand and debris.The washed biomass was first air dried for 24hrs and then in an oven at 80Co to constant weight. The dried biomass was then ground in an analytical mill and then sieved through 2mm mesh size sieve and stored in polyethylene bottles. Experiments were performed at room temperature in 250ml Erlenmeyer glass flaks containing aqueous solution of Cd, Cu, Ni and Zn of known concentrations, i.e. 10, 20, 30, 40 ppm which found sub lethal for *U. zonata* in these contact times. Analytical grade cadmium nitrate[Cd(No3)2], copper chloride (Cucl2), nickel chloride ( Nicl2),and zinc chloride ( Zncl2) were used to prepare the controls. 250mg portion of biomass was added to each flask and the mixtures were agitated in a rotary shaker at 180r.The contact times were 60, 120, 180, 240 minutes.

Aliquots of the powdered samples were analysed for heavy metals using the methods described by Kurelshy (1991). heavy metals contents were quantified using Inductively Coupled Plasma-optical emission spectroscopy (ICP-OES; Perkin Elmer Optima-3300 RL). The amount of metal sorbed at equilibrium, q (mg g-1), which represents the heavy metal uptake was calculated from the difference in metal concentration in the aqueous phase before and after adsorption according to Basha *et al*., (2006). Adsorption from aqueous solutions at equilibrium is usually correlated by Freundlich and Langmuir isotherm (Freundlich, 1907).

**q = K Ceq1/n**

In this model, K (l g-1) and 1/n are the constants to be determined from the data while q is the amount adsorbed and C is the equilibrium concentration in the solution, For a good adsorbent, 0.2<1/n<0.8 and a smaller value of 1/n indicates better adsorption and formation ofrather strong bond between the adsorbate and adsorbent. Langmuirequation (Langmuir, 1916,1917) is expressed as

**q = qmaxbCeq/1+bCeq**

Where qmax is the amount of adsorption corresponding tocomplete monolayer coverage, i.e., the maximum adsorption capacity and b (l mg-1) is the Langmuir constant.

**Results**

Bioaccumulation capacities of the green algae *U. zonata* were studied for cadmium, copper, nickel and zinc removal at various concentrations( 10ppm, 20ppm, 30ppm, and 40ppm) at different intervals of times (Fig 1, 2 ,3 and 4). Cadmium showed lowest metal uptake at 10 ppm (q=1.5 ) in 60 min and highest at 20 ppm (q=8.23) with contact time of 180 min. The minimum metal uptake for Cu was recorded at 10 ppm ( q=2.5) in the contact time of 60 min which increased into 7.21 (q) at 30 ppm in 180 min. For Ni and Zn the metal uptake increased with increasing initial concentration of the metal, with lowest uptake at 10 ppm (q=6.11) and (q= 2.45) at 60 min contact time and highest at 40 ppm (q=16.17) and (q=9.22) at 120 min. The order of metal uptake for the dried biomass was found to be Zn>Ni>Cd>Cu.

**Fig 1: Cadmium uptake by *U. zonata*.**

**Fig 2: Copper uptake by U. zonata.**

The metal uptake capacity of the biomass was evaluated using Freundlich and Langmuir isotherms. The Langmuir isotherm (Langmuir, 1916,1917) represents the equilibrium distribution of metal ions between the solid and liquid phases . qmax and b determined from the slope and 2intercept of the plot. Highest qmax value was observed for Ni (30.998 mg g-1). ). The linear form of Freundlich adsorption isotherm was used to evaluate the sorption data The values of 1/n ranged between 0.334 and 0.701 which indicates good adsorption..The separation factor values (RL) indicated (Table 1) . For Zn, Ni and Cu Freundlich model fitted satisfactorily as depicted by high values of correlation coefficients R2**.**

The results of the study indicated that the metal uptake was concentration-independent for Cd and Zn whereas for Cu and Ni an increase in initial metal concentration resulted in higher metal uptake ( Fig1,2,3.4).

Fig 3: Nickel uptake by U. zonata.

**Fig 4: Zinc uptake by U. zonata.**

**Table 1: Isotherm constant for (Cd),( Cu), ( Ni) and ( Zn) bioabsorption**

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| **Isotherm constants** | **Cd** | **Cu** | **Ni** | **Zn** |
| **qmax (mg g-1)** | **4,799** | **12.811** | **30.998** | **4.808** |
| **b (l mg-1)** | **0.122** | **0,061** | **0.098** | **0.059** |
| **RL** | **0.246±0.110** | **0.518±0.299** | **0.477±0.288** | **0.722±0.922** |
| **R2** | **0.397** | **0.454** | **0.877** | **0.922** |
| **Freundlich** |  |  |  |  |
| **1/n** | **0.334** | **0.565** | **0.682** | **0.701** |
| **R2** | **0.082** | **0.639** | **0.966** | **0.991** |

**Discussion**

Evident from the results, for all the metals studied ,the least metal uptake was recorded at 10 ppm while initial concentration for highest uptake varied in different metals. Among all the metals studied, Cd recorded decrease in the metal uptake at higher concentrations after attaining the maximum values. This appears to be the same results obtained by Ahalya *et a*l.,( 2005) for the same metal and explained due to the increase in the number of ions competing for the available binding sites in the biomass and also due to the lack of binding sites for the complexation of these ions at higher concentration levels. At lower concentrations, all metal ions present in the solution would interact with the binding sites and thus facilitate maximum adsorption. At higher concentrations, more ions are left unabsorbed in solution due to saturation of binding sites(Ahalya *et al.,* 2005). Also, contact time for minimum uptake was found to be 60 min for all the metals. However, contact time for maximum uptake also differed for different metals.The separation factor values (RL) indicated (Table 1) that metal sorption onto the biomass was favorable (Hall *et al*., 1966). According to Kadirvelu and Namasivayam (2000), n values between 1 and 10 represent beneficial adsorption. The results values of 1/n ranged between 0.334 and 0.701 which indicates good adsorption (Table 1). The. magnitude of K and n shows easy separation of heavy metal ion from wastewater and high adsorption capacity (Ahalya *et al*.,2005). The value of n, which is related to the distribution of bonded ions on the sorbent surface, was found to be greater than unity indicating that adsorption is favorable. Tien (2002) found that the magnitude of K and n showed easy uptake of surface area and dry weight of algal cells. It was found to be the main factor influencing metal sorption and indicates favorable Huge load of wastes from industries.

Biosorption using biomass derived from fresh water algae, marine seaweeds and fungi has recently attracted growing interest of researchers. Many potential binding sites occur in algal cell walls and alginate matrices (Saitoh *et al*., 2001; Tam *et al*.,1998). Algal cell surface has several kinds of functional groups with varying affinity for an ionic species. Low and high affinity functional groups are involved in sorption of metal ions at high and low concentrations of metal ions, respectively (Mehta and Gaur, 2001).Cell wall of green algae contains heteropolysaccharides, which offer carboxyl and sulfate groups for sequestration of heavy metalions*. U. zonata* was found to record high uptake values for Ni ,Zn and Cd. Algal cells have showed considerable potential in removal of heavy metal from aqueous solutions. Alpana *et al* (2007) observed 97% removal of Pb2+ by *Pithophora odeogonia* and 89% removal by *Spirogy neglecta*in at 30 min from a solution containing 5mgl-1 initial concentration of Pb2+ by a biomass concentration of 1 g l-1. Further, higher removal of 133.3 mg Cu (II) g-1 of dry weight of Biomass of *Spirogyra* species was observed in 120 min contact period with an algal dose of 20 mg l-1 by Gupta *et al*. (2006). The results of this study indicated that the metal uptake was concentration-independent for Cd and Zn whereas for Cu and Ni an increase in initial metal concentration resulted in higher metal uptake, which is in agreement with the results obtained by Ahuja *et al*. (1999) for cobalt where, increase in cobalt concentration resulted in the increased up take of the metal**.**

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**إزالة بعض العناصر الثقيلة من المحاليل بواسطة المعالجة بالطحالب**

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الخلاصة

لقد تم مقارنة الأستهلاك الحياتي للطحلب *U.zonete* للعناصر الكادميوم و النحاس و النيكل و الخارصين في تراكيز مختلفة و فترات مختلفة . أوضحت النتائج بأن أقل كمية تم امتصاصها من العناصر Zn ,Ni, Cu and Cd كانت عند تركيز ppm 10 ، بينما كان أعلى امتصاص للكادميوم عند تركيز ppm 20 ، و كان أعلى امتصاص للنحاس عند تركيز ppm 30 ، أما النيكل و الخارصين فكان أعلى امتصاص لهما عند تركيز ppm 40 ، و كانت قيمة ثابت موديل Freundlich ( 1/n ) للعناصر المختلفة تتراوح بين 0.334 الى 0.721 ، وكذلك قيمة عامل الفصل ل Langmuir يتراوح بين 0.111 الى 0.722 موضحاً امتصاصاً لصالح كتلة الطحالب المستخدمة . ان ترتيب أخذ العنصر كان كالآتي ،cd>Cu>Zn>Ni . ان النتائج أوضحت بأن الطحلب *U. zonata*  له القابلية الكبيرة للأمتصاص الحياتي للعناصر Zn ,Ni, Cu and Cdوكان عنصري الكادميوم Cd و الخارصين Zn غير متأثرين بالتركيز الأولي لهما في المحلول ، بينما عنصري النيكل Ni و النحاس Cu فقد كانا متأثرين بزيادة تركيزهما في المحلول حيث ان أي زيادة في التركيز تبعها زيادة في أخذ العنصر . تعتبر الدراسة فاتحة لدراسة مستقبلية بأستخدام الطحالب كمصدر امتصاص حياتي رخيص لأزالة العناصر الثقيلة في نهرشط العرب في البصرة –العراق الذي يشكو من كثرة المياه الملوثة.