REMOVAL OF HEXAVALENT CHROMIUM FROM CONTAMINATED WATERS USING SYZYGIUM CUMINI SEED BIOSORBENT

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Abstract- Hazardous metals such as chromium in industrial wastewater followed by leaching to ground water are threat to the environment because of their toxicity and their impact on our ecosystem. Among the different heavy metals, chromium (VI) is a common and very toxic pollutant being introduced into natural waters from a variety of industrial wastewaters. Chromium (III) is an essential element for organisms that can disrupt the sugar metabolism and cause heart conditions, when the daily dose is too low. Chromium (VI) is mainly toxic to organisms. It can alter genetic materials and cause cancer. Adverse health effects associated with Cr (VI) exposure include occupational asthma, eye irritation and damage, perforated eardrums, respiratory irritation, kidney damage, liver damage, pulmonary congestion and edema, upper abdominal pain, nose irritation and damage, respiratory cancer, skin irritation, and erosion and discoloration of the teeth. Some workers can also develop an allergic skin reaction, called allergic contact dermatitis.

The Chromium (VI) discharged from various industries has potential to contaminate drinking water sources and is one of the most important environmental problems due to its health impacts on human. Adsorption is one of the effective techniques for chromium (VI) removal from wastewater. In the present study, adsorbent has been prepared from Syzygium cumini (L.) seed and studies are carried out for chromium (VI) removal. The parameters investigated in this study are contact time, adsorbent dosage, temperature, variable initial chromium (VI) concentration and pH using Diphenyl carbazide as color developing reagent and taking the absorbance at 540 nm spectrophotometrically. The adsorption process of chromium (VI) is tested with Linear, Langmuir and Freundlich isotherm models. Application of the Langmuir isotherm to the systems yielded maximum adsorption capacity of 13.15 mg/g at a solution pH of 7 having Cr (VI) concentration 50 mg/L and biosorbent dose 1g/L. The adsorption of chromium (VI) was found to be maximum 71.58 % at low pH values of 2 having Cr (VI) concentration 50 mg/L and biosorbent dose of 1g/L and can used for removal of chromium (VI) from the polluted water as a very low cost biosorbent.

Key Words: Biosorption, Syzygium cumini seed, Hexavalent chromium

Introduction

Industrialization is the major cause of inclusion of heavy metals in to the environment especially in the water bodies all over the world. Presence of chromium (VI) in surface and ground water is hazardous to the environment because of its high potentiality to contaminate drinking water sources (Noorbakhsh *et al*, 1994), leads to toxicity, pollution effect on our ecosystem, possible human health risk. In recent years, increasing awareness of water pollution and its far reaching effects has prompted concerted efforts towards pollution abatement. Among the different heavy metals, chromium is a common and very toxic pollutant introduced into natural

waters from a variety of industrial wastewaters (Srivastava *et al*, 2013; Tripathi *et al*, 2013). The two major sources of contamination are tanneries (trivalent chromium) and electroplating, metal finishing industries (hexavalent chromium). Chromium occurs most frequently as Cr (VI) or Cr (III) in aqueous solutions (Dakiky *et al* 2002). Both valency of chromium are potentially harmful but hexavalent chromium possesses a greater risk due to its water soluble nature, fast permeability to living cells and carcinogenic properties (Dakiky *et al* 2002). Hexavalent chromium, which is primarily present in the form of chromate CrO_4 ⁻⁻ and dichromate Cr_2O_7 ⁻⁻, has significantly higher levels of toxicity than the other valence states (Sharma and Forster, 1995).

Various conventional methods for chromium (VI) is removal from waste water such as chemical precipitation, electrochemical reduction, sulfide precipitation, ion-exchange, reverse osmosis, electro dialysis, solvent extraction, and evaporation, etc. are reported. (Namasivayam and Yamuna, 1995; Sharma and Bhattacharyya, 2004; Jianlong *et al*, 2000; Gupta and Babu, 2006). These conventional methods for the removal of metals from waste water however, are often cost prohibitive and having inadequate efficiencies at low metal concentrations.

The search for new technologies involving the removal of toxic metals from waste water has directed attention to biosorption, based on metal binding capacities of various biological materials (Singh *et al*, 2013; Ozer *et al*, 1997; Lotfi and Adhoum, 2002; Mauri *et al*, 2001; Padilla and Tavani, 1999; Rengaraj *et al*, 2003; Namasivayam and Yamuna, 1995). The major advantage of biosorption over traditional methods includes low cost, high efficiency of metal removal and eco friendly behavior.

Therefore, biosorption is an effective and versatile method for removing chromium. Most of the low cost biosorbent have the limitation of low sorptive capacity hence, there is need to explore low cost biosorbent having high contaminant sorption capacity. Several recent publications utilized locally available adsorbents (Bai and Abraham, 2003; Nourbakhsh *et al*, 1994; Singh *et al*, 2013) and agricultural byproducts (Bailey *et al*, 1999) for heavy metal removal. However, the literature is still insufficient to cover this problem and more work and investigations are needed to deal with other locally available and cheap biosorbent to eliminate Cr (VI) from industrial contaminated waters.

The objective of this study is to develop inexpensive and effective biosorbent that is easily available in large quantities. Syzygium cumini (L.) is the widely used medicinal plant in the treatment of various diseases particularly in diabetes. It has been viewed as an antidiabetic plant since it became commercially available several decades ago. The plant is rich in compounds containing anthocyanins, glucoside, ellagic acid, isoquercetin, kaemferol and myrecetin (Ayyanar and Pandurangan 2012). The vast number of literatures found in the database revealed that the extracts of different parts of S. cumini showed significant pharmacological actions (Lal and Choudhuri, 1968; Shrotri et al, 1963). Phytochemical constituents isolated from S. cumini (L.) seed has been found to contain Jambosine, gallic acid, ellagic acid, corilagin, 3,6-hexahydroxy diphenoylglucose, 1-galloylglucose, 3-galloylglucose, quercetin, β -sitoterol, 4,6 hexahydroxydiphenoylglucose (R. Rastogi and B. Mehrotra, 1990). From all over the world, the fruits have been used for a wide variety of ailments, including cough, diabetes, dysentery, inflammation and ringworm

Literature survey reveals that in most of the peer reviewed journals, the adsorption study of Cr (VI) with Syzygium cumini seed as a biosorbent has not been investigated and this is the first such study undertaken by the authors. Based on their efficacy, Syzygium cumini seed was selected for further study. The effect of pH, contact time, temperature, initial hexavalent chromium concentration, variable adsorbent doses and adsorption equilibrium were investigated.

Methods & Material

All the chemicals used were of analytical reagent grade. The standard stock Cr(VI) solutions was prepared by weighing 2.8287 g of Potassium dichromate in one liter double distilled water and it was further diluted to desired concentrations ranging from 1 to 200 mg/L of chromium (VI) in aqueous phase standard solutions. The estimation of hexavalent chromium was carried out by using Diphenyl carbazide method as per standard methods (APHA, 1995). Shimadzu UV-VIS Spectrophotometer at 540 nm was used for measurement. The Cr (VI) loadings on sorbents were computed based on mass balance through loss of metal from aqueous solution. 430 <u>www.ijergs.org</u>

The pH of solution was maintained using 0.5 N HCl and 0.5 N NaOH solutions. The temperature of the solutions was maintained by using temp. regulatory oven.

Preparation of Biosorbent (Syzygium cumini seed powder)

The sorbents used was Syzygium cumini seed powder. The materials were obtained from local area. Material was washed, dried and then pulverized in pulverizer and air-dried in the sun for five days. After drying, the materials were kept in air tight plastic bottles. The powdered material was used as such and no pretreatment was given to the materials. The particle size was maintained in the range of $212-300 \mu m$ (geometric mean size: $252.2 \mu m$).

Screening of Biosorbent

The experiments were carried out in 150 mL borosil conical flasks by agitating a pre-weighed amount of the Syzygium cumini seed powdered adsorbent with 10 - 100 mL of the aqueous chromium (VI) solutions for a predetermined period at 10-40°C in an ice bath / oven. The biosorbent doses were maintained 1-5 g/L for different experiments. The adsorbent is separated with whatman filter paper no 41. Adsorption isotherm study was carried out with different initial concentrations of chromium (VI) from 1 to 200 mg/L with the adsorbent dosage of 1-5 g/L. The effect of pH on Cr (VI) biosorption was studied at 30°C with chromium (VI) concentration of 50 mg/L and an adsorbent dosage of 1 g/L. The effect of adsorbent dosage was studied by varying the adsorbent amount from 1 g/L to 5 g/L with chromium (VI) concentration of 50 mg/L. The time duration 10-300 min was maintained for impact of time at Cr (VI) concentration of 50 mg/L and biosorbent dose of 1 g/L.

The concentration of free chromium (VI) ions in the solution was determined spectrophotometrically by developing a purpleviolet color with 1, 5-diphenyl carbazide in acidic solution as complexing agent. The absorbance of the purple-violet colored solution was read at 540 nm after 20 min.

Results and Discussion

In the present study, Syzygium cumini seed has been used for chromium (VI) removal from aqueous solutions. Table-1 shows the adsorbent capacity of various adsorbents. When compared with other non-conventional adsorbents, the results of the study indicate that adsorbent prepared from Syzygium cumini seed has better adsorption capacity in many cases (biomass residual slurry, Tamarind seed, Fe(III)/Cr(III) hydroxide, Waste tea, walnut shell), comparable adsorption capacity with palm pressed-fibers, maize cob, sugar cane bagasse and lower adsorption capacity with Ficus racemosa bark, Mangifera indica bark, activated carbon, saw dust for chromium (VI) ions (Namasivayam and Yamuna, 1995; Huang and Wu, 1977; Sharma and Forster, 1994; Tan *et al*, 1993; Gupta and Babu, 2008; Orhan and Buyukgungur, 1993; Namasivayam and Ranganathan, 1993; Singh *et al*, 2013; Singh *et al*, 2013). Based on the above conditions the results obtained indicating the effect of various experimental conditions such as equilibrium time, pH, amount of adsorbent etc. has been studied.

Table No. 1: Summary of adsorbent capacity of various adsorbents

Adsorbent N	Aaximum Adso	rbent Capacity,	<i>q</i> m (mg/g) Reference		
Walnut shell		1.33	[Orhan and Buyukgungur, 1993]		
Fe (III)/Cr (III) hyd	lroxide 1.43		[Namasivayam and Ranganathan, 1993]		
Waste tea		1.55	[Orhan and Buyukgungur, 1993]		
Biomass residual sl	lurry 5.87		[Namasivayam and Yamuna, 1995]		
Tamarind seeds	11.08		[Gupta and Babu, 2008]		
Syzygium cumini s	leed 13.15		[Present Study]		
Sugar cane bagasse		13.4	[Sharma and Forster, 1994]		
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Maize cob		13.8	[Sharma and Forster, 1994]
Palm pressed-fibers		15.0	[<i>Tan et al, 1993</i>]
Mangifera indica bark		19.64	[Singh et al, 2013]
Ficus racemosa bark		25.9	[Singh et al, 2013]
Syzygium cumini bark	31.51		[Singh et al, 2014]
Sawdust		39.7	[Sharma and Forster, 1994]
Activated Carbon		57.7	[Huang and Wu, 1977]

Effect of Contact Time on Chromium (VI) Adsorption

The effect of contact time up to 300 min. on chromium VI adsorption was studied using Biosorbent dose of 1 g/L and hexavalent Chromium concentration of 50 mg/L.(Fig -1). The extraction process was carried out with standard Cr (VI) 100 mL solution of 50 mg/L in 150 mL conical flask with biosorbent dose of 1g/L and the concentration of hexavalent chromium in the solution was measured by filtration through whatman filter paper followed by development of colour using Diphenyl carbazide at 540 nm in time interval of 10, 20, 30, 60, 120, 180, 240 and 300 minutes. Most of the adsorption takes place in first hour of contact and longer contact time has negligible effect on extraction of chromium (fig-1)

Effect of Increasing Concentration of Cr (VI) on Adsorption

Standard Cr (VI) solutions of 100 mL having initial concentration of 10, 20, 30, 40, 50 mg/L were treated with biosorbent 0.1 g dose in each solution. The concentration of Cr (VI) in the solution was determined using the standard methods (APHA, 1995). The percentage adsorption slowly decreases from 47.5 for 10 mg/L to 23.46 for 30 mg/L and further increases to 27.24 for 50 mg/l Cr (VI) concentration solutions (figure-2).



Effect of Temperature on Cr (VI) biosorption

The 100 mL samples of 50 mg/L hexavalent chromium concentration in 150 mL conical flasks were treated with 0.1 g of biosorbent (Syzygium cumini seed powder) maintained at 10, 20, 30 & 40°C. The solutions were kept for 120 min. with gentle shaking at periodical intervals and the concentration of Cr (VI) was measured in the solution after filtering through Whatman filter paper and developing the colour using Diphenyl carbazide at 540 nm spectrophotometrically. The percentage biosorption of Cr (VI) was found maximum at 40°C and minimum at 10°C showing an increasing trend with temperature. (Figure-3)

Effect of pH on Cr (VI) biosorption

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The experiments using 100 mL of 50 mg/L Cr (VI) solutions for 120 min time and adsorbent dose of 0.1 g were carried out at

pH 2, 4, 7, 10 & 12 and the biosorption of Cr (VI)



is depicted in figure-4. The acidic medium (pH-2) has been found to show maximum biosorption up to 71.58% of initial chromium (VI) which decreases to 26.3% at neutral (pH-7) and further decreases to 39.24% in basic medium at pH-12 (Figure-4)

Effect of Biosorbent Concentration on Cr (VI) Adsorption

The 100 mL samples of 50 mg/L hexavalent chromium concentration in 150 mL conical flasks were treated with 0.1, 0.2, 0.3, 0.4, 0.5 g of biosorbent (Syzygium cumini seed powder) maintained at room temperature 25 °C. The solutions were kept for 120 min. with gentle shaking at periodical intervals and the concentration of Cr (VI) was measured in the solution after filtering through Whatman filter paper and developing the colour using Diphenyl carbazide at 540 nm spectrophotometrically. The percentage biosorption of Cr (VI) was found maximum with biosorbent dose of 0.5g and minimum at 0.1g showing an increasing trend with increasing biosorbent. (Figure-5)



Adsorption Isotherms

The equilibrium of sorption is one of the important physico-chemical aspects for the evaluation of the sorption process as a unit operation. The sorption isotherm studies were conducted by varying initial concentration of chromium (VI) from 10-50 mg/L, constant adsorbent dosage of 1 g/L; constant initial concentration of 50 mg/L with varying biosorbent doses of 1-5 g/L and maintaining the temp.(Fig-6 &7). The adsorption isotherm (qe versus Ce) shows the equilibrium between the concentration of chromium (VI) in the aqueous solution and its concentration on the solid (mass of chromium (VI) per unit mass of Syzygium cumini seed). It is evident that adsorption capacity increases with increasing equilibrium chromium (VI) concentrations. Fig. - 6 shows that 433 <u>www.ijergs.org</u>

the adsorption capacity increases slowly from 0 to 13.62 mg/g for the equilibrium concentration of 0 to 36.38 mg/L. The linear isotherm with variable biosorbent (Fig-7) shows a linear increase in adsorption capacity up to 12.86 mg/g for the equilibrium concentration of 37.14 mg/L.

In order to model the sorption behavior, adsorption isotherms have been studied. The adsorption process of chromium (VI) is tested with Langmuir and Freundlich isotherm models. Langmuir and Freundlich equations are given in equation (1) and (2), respectively.

qe - Amount of Cr(VI) adsorbed by the adsorbent (mg/g) and qm - Maximum adsorption capacity (mg/g)]

 $\ln q_{\underline{e}} = \ln K_{\mathrm{F}} + (1/n) \ln C_{\mathrm{e}}$

(2)

[where $K_{\rm F}$ - Freundlich constant (mg/g) and *n* - Freundlich constant (L/mg)]



The isotherm data has been linearized using the Langmuir equation and shown in Fig. 8 to 11. The regression constants are tabulated in Table-2. The high values of correlation coefficient ($R^2 = 0.874$, 0.972, 0.993, 0.913, 0.911) indicated a good agreement between the parameters. The constant qm, which is a measure of the adsorption capacity to form a monolayer, can be as high as 13.15 mg/g at pH 7. The constant b, which denotes adsorption energy, varies from 0.099 to 0.392 L/mg. The same data also fitted with the

Freundlich equation and shown in Fig. 12, 13 & 14. The regression constants are listed in Table-2. The values of correlation coefficient ($R^2 = 0.827, 0.821, 0.996$) showed that the data confirm well to the Freundlich equation also.

Lang	muir Isotherm		Freundlich Isotherm		
Constants		Correlation	Constants		Correlation
Constants qm (mg/g)	b (L/mg)	Coefficient (R^2)	$K_{ m F}$	1/n	Coefficient (R^2)
47.7	0.249	0.972	1.986	0.472	0.827
28.12	0.345	0.993	2.517	0.421	0.821
71.58	0.099	0.913	2.477	3.374	0.996
28.14	0.392	0.991			

Table No. 2: Isotherm constants for adsorption of chromium (VI) on Syzygium cumini seed.



Conclusions

Based on present study and scientific information derived from literature, the Following conclusions are made -

- Adsorbent prepared from Syzygium cumini seed can be used for removal of chromium (VI) from aqueous solutions due to its remarkable higher biosorption capacity of 13.53 mg/g at 30 °C.
- The adsorption rate of chromium (VI) on the adsorbate prepared from Syzygium cumini seed in the present study from aqueous solutions (50 mg/L solution and adsorbent dose of 1 g/L) is maximum for the first hour (27%) thereafter it increases very slowly up to 28% in the next four hours. Since maximum adsorption of chromium (VI) on Syzygium cumini seed powder takes place within first hour so the equilibrium time is found to be 60 min. in the present experimental conditions.
- The adsorption process of chromium (VI) can be described by Langmuir isotherm and Freundlich isotherm models. However, Langmuir isotherm model shows a good agreement with the equilibrium data.
- Adsorption of chromium (VI) on Syzygium cumini seed yielded maximum adsorption capacity of 71.58 mg/g at solution concentration of 50 mg/L, pH 2, temperature 30 °C and adsorbent dose of 1g/L.
- Removal of chromium (VI) increases from 25% to 79% with increase of adsorbent dosage of 1 to 5 g/L using 50 mg/L Cr (VI) solution.
- The maximum adsorption (71.58%) of chromium (VI) took place in acidic medium at pH 2.
- The increase in temperature increases the biosorption up to 40 °C, showing the chemisorption behavior.
- The maximum adsorption takes place in 60 minutes and further increase in duration of contact time has negligible effect on biosorption.
- The Syzygium cumini seed has better efficiency in Cr (VI) removal as compared to biomass residual slurry, Tamarind seed, Fe(III)/Cr(III) hydroxide, Waste tea, walnut shell powders; hence it can be used as low cost biosorbent for treatment of hexavalent chromium from contaminated waters.

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