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Removal of Hexavalent Chromium from Tannery Effluent Using Algal Extracts - A New Approach

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ABSTRACT

Hexavalent chromium is one of the major pollutants released from the tanneries that have a direct effect on man and animals. In this study, seaweeds such as *Centeroceras clavulatum*, *Enteromorpha flexuosa*, *Grateloupia lithophila*, *Enteromorpha intestinalis,* and *Ulva lactuca,* were collected from Covelong, Chennai. *Sargassum* sp., *Amphiroa* sp., *Ulva* sp., and *Hypnea* sp. were collected from Kanyakumari and *Chaetomorpha antennina* was collected from Puducherry. Different solvents such as methanol, ethanol, water, benzene and chloroform were used for extraction. Then these extracts were used to treat tannery effluent. The reduction in hexavalent chromium was analysed and correlated with phytochemicals of algal extracts. Results revealed that Cr(VI) concentration was highly reduced by benzene extract of *Hypnea* sp. (95%).

INTRODUCTION

Exposure to hexavalent chromium can induce allergies, irritations, eczema, ulceration, nasal and skin irritations, perforation of eardrum, respiratory track disorders and lung carcinoma (Poopal & Laxman 2009, Gibb et al. 2000, Gibb et al. 2000a). Also, Cr(VI) evidences the capability of accumulating in the placenta, damaging fetal development (Saxena et al. 1990). Cr(VI) pollution in the environment changes the structure of soil microbial communities (Zhou et al. 2002), reducing microbial growth and related enzymatic activities, with a consequent persistence of organic matter in soils and accumulation of Cr(VI) (Shi et al. 2002). The toxic action of Cr(VI) is due to its capability to easily permeate into cellular membranes, and cell membrane damages caused by oxidative stress induced by Cr(VI) have been extensively reported, both in eukaryotic and prokaryotic cells (Codd et al. 2001). Furthermore, Cr(VI) enters into the cells using the sulphate transport system of the membrane in cells of organisms that are able to use sulphate (Ohta et al. 1971, Ohtake et al. 1987, Cervantes & Silver 1992). Once Cr(VI) enters into cells, simultaneous reactions takes place with the intracellular reductants as ascorbate and glutathione (Costa 2003, Xu et al. 2004). On the other hand, Cr(IV) is able to bind to cellular materials, altering their normal physiological functions (Pesti et al. 2000). It is a very dangerous chemical form on biological systems as it can induce mutagenic, carcinogenic and teratogenic effects. Moreover, Cr(VI) is able to induce oxidative stress in cells, damaging its DNA. Considering the dangerous effects Cr(VI) can cause to human health, Cr(VI) has been consisted of among the priority pollutants and listed as a class A human carcinogen by the US Environmental Protection Agency (USEPA) (Costa & Klein 2006).

In this study, waste water from leather industries which is drained into Peria Eri located at Nagalkeni, Pallvaram without treatment was collected and treated with algal extracts.

MATERIALS AND METHODS

Collection and preservation of seaweed: Marine algal species such as *Centeroceras clavulatum*, *Enteromorpha flexuosa*, *Grateloupia lithophila*, *Enteromorpha intestinalis, Ulva lactuca,* were collected from Covelong, Chennai. Samples of *Sargassum* sp.*, Amphiroa* sp.*, Ulva* sp., and *Hypnea* sp. were collected from Kanyakumari whereas *Chaetomorpha antennina* from Puducherry. They were identified in Madras Christian College, Tambaram, Chennai. The collected samples were thoroughly washed with water to remove sand and epiphytes present in them. After complete washing, they were dried under sunlight. Then the dried algae were powdered well using a mixer and then stored.

Preparation of algal extracts: Five gram of powdered sample was soaked in 50 mL of non polar solvents such as benzene and chloroform for two days to extract the phytochemical constituents completely. They were also soaked in 50 mL of polar solvents such as methanol, ethanol and water for 24 hrs and were extracted. Table 1 shows the various extracts of seaweed and their abbreviations used in this study for convenience.

Collection of effluent: Leather industry effluent was collected from the canal which was connected to the Peria Eri located at Nagalkeni village, Pallavaram, Chennai, Tamil Nadu and was preserved in a refrigerator to avoid further microbial contamination.

Preparation of algal extracts: Collected samples were soaked in different solvents such as methanol, ethanol, water, chloroform and benzene. Then the extracts were collected.

Phytochemical analysis: Phytochemical analysis of extracts was done by standard methods.

Treatment of effluent: 10 mL of algal extract was added to 100 mL of effluent and was left for 5 days. After five days of treatment, concentration of hexavalent chromium was analysed.

Estimation of Cr(VI) concentration: Concentration of Cr(VI) was analysed by using APHA (1998).

RESULTS AND DISCUSSION

Leather industry effluent is one of the important sources of harmful chemicals that are drained into land and water bodies without treatment, leading to severe impact on environment. In recent studies, marine algae were used to treat industrial effluent effectively (Esmaeili et al. 2010, Thillai Natarajan et al. 2011, Jayaraj et al. 2011). They are mostly used either as an adsorbent or as a biofilter. In the present study, solvent extracts of marine algae collected from Tamil Nadu and Puducherry, were used to treat the effluent.

Phytochemical analysis: Phytochemical results revealed that the terpenoids were absent only in EIEC, UEK, SEK, GLEC, CCCC and CCWC. Phenol was found to be absent in the aqueous extracts such as CAWC, ULWC, AWK, UWK, CCWC, HWK, GLWC, SWK and also in methanolic extracts of *Chaetomorpha* sp., from Covelong and *Amphoroa* sp., from Kanyakumari. Since solubility of phenol is very less at room temperature. Aqueous extracts showed absence of carboxylic acid except in *E. intestinalis* collected from Covelong and in case of chloroform extracts it was found only in ULCC and CACP. Seven extracts of ethanol showed presence of carboxylic acid except EFEC, CCEC and AEK. All the aqueous extracts showed presence of amino acids and it was found to be absent in all benzene extracts except in EFBC and also absent in chloroform extract except in EFCC. Because of high solubility of amino acids in water its solubility decreases towards non polarity (Thomas 1970). Other extracts showed presence of amino acid except in ULEC, CACP, AEK, CAEC, HMK, SMK, CAMP, ULMC, EIMC, GLMC, and UMK. Phytochemical report showed that most of the extracts contained carbonyl compounds except in UEK, CAEP, HEK, AEK, CAEC, HMK, CAMP, CCMC, GLMC, UMK, CACP, CCCC, CACC, ACK, HWK, GLWC, EIWC. All the methanolic extracts showed presence of carbonyl compounds (Data not shown).

Reduction of Cr(VI) concentration: Reduction of Cr(VI) by algal extracts were characterized as Category A (75% - 100%) (Fig. 1), Category B (50%-75%) (Fig. 2) and Category C (below 30%) (Fig.3).

Hexavalent chromium content of untreated effluent was found to be 1250 mg/L which was very high against the permissible limit of The National Environment Regulations, (1999) (0.05 mg/L). Among all extracts, ethanol was found to be the good solvent system, since all the ethanolic extracts showed good reduction, except *Enteromorpha flexuosa* collected from Covelong (Category B) (Fig. 2). Among all samples, extracts of *Ulva lactuca* collected from Covelong yielded Category A type of reduction (Fig. 2). Highest reduction of

Table 1: Seaweed extracts.

S.No	Name of Seaweed	Collection Place	Water	Chloroform	Benzene	Methanol	Ethanol
1	*Enteromorpha intestinalis*	Covelong	EIWC	EICC	EIBC	EIMC	EIEC
2	*Ulva* sp.	Kanyakumari	UWK	UCK	UBK	UMK	UEK
3	*Hypnea* sp.	Kanyakumari	HWK	HCK	HBK	HMK	HEK
4	*Grateloupia lithophila*	Covelong	GLWC	GLCC	GLBC	GLMC	GLEC
5	*Sargassum* sp.	Kanyakumari	SWK	SCK	SBK	SMK	SEK
6	*Chaetomorpha antennina*	Puduchery	CAWP	CACP	CABP	CAMP	CAEP
7	*Centerocerus clavulatum*	Covelong	CCWC	CCCC	CCBC	CCMC	CCEC
8	*Ulva lactuca*	Covelong	ULWC	ULCC	ULBC	ULMC	ULEC
9	*Enteromorpha flexuosa*	Covelong	EFWC	EFCC	EFBC	EFMC	EFEC
10	*Chaetomorpha attenina*	Covelong	CAWC	CACC	CABC	CAMC	CAEC
11	*Amphiroa* sp.	Kanyakumari	AWK	ACK	ABK	AMK	AEK

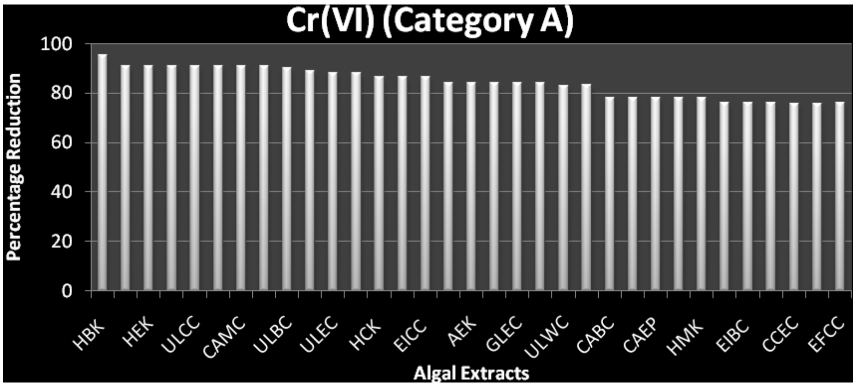

Fig. 1: Percentage reduction in Cr(VI) concentration (Category A).

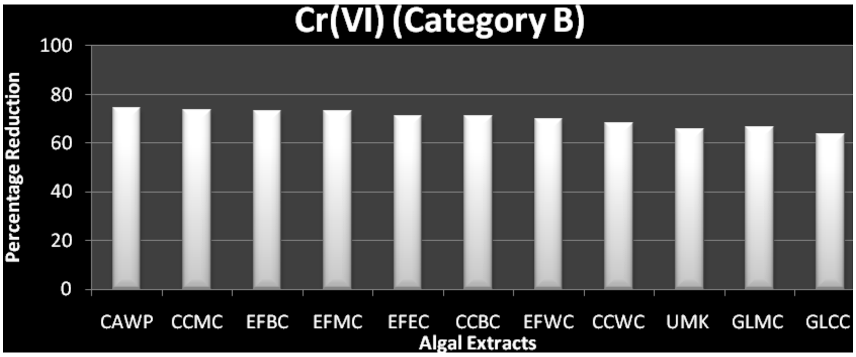

Fig. 2: Percentage reduction in Cr(VI) concentration (Category B).

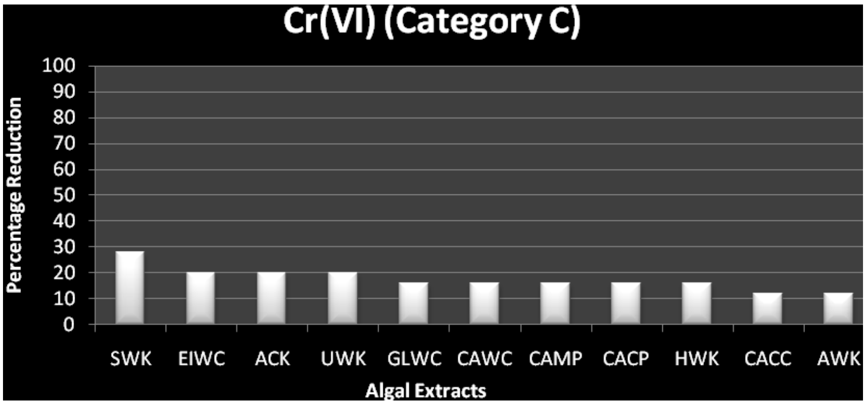

Fig. 3: Percentage reduction in Cr(VI) concentration (Category C).

95 % was found in benzene extract of *Hypnea* sp., collected from Kanyakumari and least reduction of 12 % was found in CACC and AWK (Fig. 3). Fungal isolates of *Fusarium solani* was found to reduce 44% (Sen et al. 2013) of Cr(VI) which was very much lower than this result, in which nearly thirty three extracts were found to be the good reducing agents and eleven were found to be medium reducing agent and another eleven extracts showed poor reduction.

Hexavalent chromium reduction was correlated with phytochemical analysis. Results showed that the presence of carbonyl compounds might be the reason for the hexavalent chromium reduction. Cr(VI) is reduced to Cr(III) during the oxidation of alcohol or aldehyde. Hence the harmful hexavalent chromium in this study might have been reduced to Cr(III).

Aldehyde + Cr(VI) $\rightarrow$ Ketone or Carboxylic acid + Cr(III)

Cr(III) is known to be less toxic than Cr(VI) or nontoxic and an essential nutrient for human and animals, but long term exposure to a high concentration of Cr(III) may cause poisoning symptoms such as allergic skin reactions. Cr(III) is readily precipitated in neutral or alkaline solutions as $Cr(OH)_3$ or adsorbed by various substrates (Nasrallah et al. 2011).

CONCLUSION

Motivating the natural process provides an opportunity for reducing the environmental impact of various pollutants. This method is an effective biological treatment for polluted waters. Many micro and macro algae are being used in various bioremediation techniques particularly in polluted waters. The close association which the algae have with the aquatic habitat makes them an attractive tool for such studies. In this study, new approach with effluent treatment was carried out, instead of algal powder as adsorbent, algal extracts were used for wastewater treatment and the results showed good reduction in all the compounds. In future, optimization of all the parameters may be carried out to nullify the harmful contents. Effect of algal extracts on other pollutants may also be done in our future studies

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