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Marine Algal Species as Biosorbents for Sequestering Toxic Heavy Metals: A Review

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ABSTRACT: Seaweed biomass; especially marine macro algae have been studied for their potential to remove the heavy metals from aqueous solution which attributed their use in environmental cleanup. The prevalent remediation procedures include oxidation/reduction, chemical precipitation, ion exchange, reverse osmosis etc. However, researchers and environmental engineers are hoping this phenomenon will provide an economical alternative to get rid of toxic heavy metals from industrialized wastewater so as to aid in environmental remediation. The point sources of toxic heavy metals are industrial effluents coming from various industries like leather, paper, dairy, tannery, electrical, sugarcane, automobiles, mining etc. Such toxic heavy metals can be removed by using biosorption techniques with the help of seaweeds; mostly macro algal species; as these are abundantly found growing on the areas like rocky coast. This attribute of being used as biosorbents make them cost effective and ecofriendly alternative for conventionally used costly techniques. Earlier research reports have contributed in reviewing the utilization of major marine macro algal species for detoxification of polluted water bodies. Besides the conventional techniques; the present review article highlights the use of marine macro algal species as biosorbents for removal of toxic heavy metals along with the major parameters influencing the process of biosorption.

Keywords: Marine macro algae, biosorption, toxic heavy metals.

INTRODUCTION

Heavy metals constitute a group of contaminants belonging to inorganic compounds including elements lighter than carbon. These occur with large variations in concentration and enter an environment through various ways that can be natural and or anthropogenic; such as industrialization, weathering of rocks, burning of petroleum, non-ferrous metal working etc. (Gupta *et al*. 2016). An environment is characterized by prevalence of toxic heavy metals that are accumulating quite consistently through various small and large scale industrial, medical and agro-domestic technological units. Consequently, their ill effects on ecology and human health have made researchers to find sustainable approaches for the effective treatment of toxic heavy metals such as Arsenic (As), Cadmium (Cd), Chromium (Cr), Lead (Pb), Mercury (Hg) etc. (Tchounwou *et al.*, 2012). Unlike organic contaminants, heavy metals are non-biodegradable and tend to accumulate in living organisms and many heavy metal ions are known to be toxic or carcinogenic. In the developing countries; direct or indirect discharge of heavy metals has found to be increasing rapidly (Jyothi *et al.,* 2015). Among the toxic metals; three having major impact on ecosystems are mercury, cadmium and lead. Cadmium, lead and chromium are dangerous to humans and also for environment (Hogan, 2010). Unlike organic pollutants, heavy metals do not decay and thus pose a different kind of challenge for remediation.

CONVENTIONAL TECHNIQUES IN HEAVY METAL REMOVAL

The most commonly discharged heavy metal ions as industrial effluents are Calcium, Magnesium, and Sodium ions (Ca^{+2}, Mg^{+2}, and Na^{+}). The prevalent remediation procedures utilized are oxidation/reduction, chemical precipitation, ion exchange as well as reverse osmosis (Ghoneim *et al*., 2014). Most of these conventional methods are not convenient for small scale industries due to their high cost (Lee and Volesky 1997). Earlier; various conventional techniques were used to remove toxic metals, such as ion exchange and precipitation, lack specificity and are ineffective at low metal ion concentrations. Some of the technologies employed for industrial effluents often create secondary problems with metal-bearing sludge. Most of these techniques are expensive, usually dependent on the concentration of the waste water and also not environment friendly (Table 1).

Table 1: Some conventional techniques used to remove heavy metals.

Technique	Principle involved	Suitability	Drawbacks/Limitations	Reference
Chemical precipitation	Heavy metals are removed by using coagulants such as alum, lime, iron salt and other organic polymers.	used by 75% plating companies.	Production of large amount of sludge, containing toxic compounds.	(Charerntanyarak, 1999; Ahalya *et al.*, 2003)
Ion exchange	Metal ions from dilute solutions are exchanged with ions held by electrostatic forces on the exchange resin.	Metal finishing industries.	High cost and partial removal of certain ions.	(Da¸browski *et al.*, 2004)
Electrowining	Metallic ions are removed from concentrated rinse water, ion exchange, regenerate and spent process.	Suitable for metals like copper, gold and silver etc. and solutions containing a moderate to high concentration of metal ions (73,000 mg/l).	Not applicable to solutions containing hydrochloric acid, formation of chlorine gas, the recovery metal is not pure enough for reuse as anode material.	(Yuan and Weng 2006)
Reverse osmosis	Heavy metals are separated by a semi-permeable membrane at a pressure greater than osmotic pressure caused by the dissolved solids in wastewater.	Metal finishing industry for the purpose of recovery of metal salts and reuse of the water.	Expensive process, formation of metal hydroxides, which clog the membrane is the main limitation.	(Ahalya *et al.*, 2003)
Ultra filtration	The Porous membranes are used for removal of heavy metals.		Generation of sludge.	(Da-Qi Cao *et al.*, 2020)

BIOSORPTION METHODS FOR HEAVY METAL REMOVAL

Biosorption can be defined as physico-chemical binding of metal species in solution to the cellular component of biomass. The mechanism of biosorption is complex involving few independent metal uptake processes that can be broadly categorized into active and passive bioaccumulation which is carried out by living and dead biomass respectively (Brown *et al.*, 2000). All these algal based metal uptake processes need to be optimized in order to adopt the most economical, sustainable and efficient biosorption mechanism (Mack *et al.,* 2007; Salam, 2019). Biosorption is mainly used to treat waste water, when more than one type of metal ions is present, the removal of one metal ion may be influenced by the presence of other metal ions. According to Das *et al.* (2008); it is a process which represents a biotechnological innovation and a cost-effective excellent tool for removing heavy metal ions from aqueous solution. Among the different biological methods, bioaccumulation is the phenomenon of living cells while biosorption is a cost effective process carried out by using inactive biosorbents for the removal of heavy metal ions (Volesky, 2003).

A. Heavy metal binding in algal biosorption

Biosorption mechanisms are classified on the basis of cellular metabolism and the site where biosorption process occurs. Several mechanisms have been postulated to explain the process of metal binding during algal biosorption. (Table 2).

Table 2: Mechanisms involved in heavy metal binding process.

Mechanism	Principle involved	Algal material used	Reference
Precipitation	The metal uptake may take place both in the solution and on the cell surface. It is associated with active defence mechanisms.	Red macroalgae.	(Ibrahim 2011; Ahmad *et al.*, 2018)
Physical adsorption	Through electrostatic interaction between the metal ions in solutions and cell wall molecules.	*Laminaria digitata* and *L. japonica*	(Pohl and Schimmack 2006)
Transport across the cell membrane	Metabolism dependent intracellular uptake, whereby metal ions are transported across the cell membrane.	*Neochloris oleoabundans*	(Rashidi and Trindade 2018)

MAJOR PARAMETERS INFLUENCING HEAVY METAL BIOSRPTION

The parameters that influence efficiency of algae based metal uptake process are mainly hydrogen ion concentration, contact time, initial concentration of metal, effect of biomass and temperature.

A. Hydrogen ion concentration

P^H is an important factor for the adsorption of metal by the biological material (Kapoor *et al.,* 1999). There is a relationship between the amount of metal adsorbed and the magnitude of negative charge on the surface of the biosorbent which is related to the surface functional groups (Selania *et al.,* 2004). It depends on the ionic form of metal in solution and electric charge on the biological material. The P^H value of the medium affects the equilibrium of the system (Romera *et al.,* 2007). Removal of Cadmium by dead biomass of *Fucus vesiculosus* and *Fucus serratus* found to be increasing with increase in pH (Herrero, 2006).

B. Contact time

It has been reported that the metal sorption completes in about less than one hour. The fast adsorption rate at the initial stage may be explained by an increased availability in the number of active binding sites on the adsorbent surface (Murugan and Subremanian 2006).

C. Initial concentration of metal

Metal uptake is strongly influenced by its initial concentration. The removal of metal depends upon the availability of specific surface functional groups and their ability to bind metal ions of high concentration. However, by increasing the amount of the available metal the fraction of metal bound is decreased (Blanco *et al.,* 1999).

D. Effect of Biomass

With an increased amount of biomass; the number of available adsorption sites or functional groups also increases. It is observed that removal efficiency of the adsorbents generally increases with increasing the quantity. It is due to the fact that the availability of exchangeable sites for the ions (Dubey and Gopal 2007).

E. Temperature

An increase in temperature influence the surface of adsorbent which tends the detachment of metal ions. At lower temperatures; metal removal efficiency gradually increases due to increase in the physical adsorption process. (Adeniyi and Ighalo 2019).

ALGAE AS A BIOSORBENTS

Considerable amount of research work has been initiated for utilization of seaweed-based biosorption for heavy metals removal. Seaweeds are extremely efficient biosorbents with the ability to bind various metals from aqueous effluents (Davis *et al.,* 2003; Tsui *et al*., 2006). Raize *et al.* (2004) highlighted the mechanism of biosorption of different heavy metals like cadmium, nickel and lead by *Sargassum vulgaris.* Loredana *et al.* (2007) used marine micro and macro algal species as biosorbents for heavy metal. Murphy *et al.,* (2008) studied on Chromium by red, green and brown seaweed biomass. Most of the algal members require minimum growth inputs and cultivated commercially on a large scale having promising biosorptive potential (Jyothi *et al*., 2015). Latinwo *et al*. (2015) studied the potential of green marine algal biomass for the removal of heavy metals like Silver (Ag), Chromium (Cr), Iron (Fe) etc. from the textile waste water. Among the marine macroalgae; Phaeophyceae members can serve as better biosorbents for heavy metal removal; than Cholorophyceae and Rhodophyceae members. Higher uptake capacities have been found for brown algae than for red and green algae. The recent studies have shown that Brown algae is highly effective for the purpose of recovery of heavy metals from industrial effluents or waste water bodies. This may be due to an interaction that occurs on the algal cell walls that are rich in nutrients, polysaccharides etc. (Ankit *et al*., 2022; Sreevani and Anierudhe 2022). It is found that the four different marine algal species namely *Ulva lactuca*, *Janiarubens*, *Pterocladia capillacea* and *Colpomenia sinosa* can be used for removing toxic heavy metal ions like lead and Nickel from synthetic wastewater. Of these, the red macro alga, *J. rubens* is found as potential inexpensive algal species for sequestering heavy metals form waste waters (Ibrahim *et. al.,* 2018). According to them the surface treatment improves the reduction capacity of the biosorbents. Apart from the studies on biosorption mechanism many researchers have examined adsorption processes extensively with reference to contact tests (Lee *et al.,* 2000; Bishnoi *et al.,* 2007). Adsorption onto the cell surface occurs through cell wall polysaccharides, cytoplasmic ligands phytochelatins, other intracellular molecules etc. (Azhar Uddin and Lall 2019). Considerably significant attention is drawnby algae biosorbent materials especially marine algal strains due to their abundance, easy harvesting and ability to accumulate heavy metal in high amount (Pan *et al*., 2018; Hamad *et al*., 2022).

Also, recent advancements in terms of developing genetically modified heavy metal tolerant algal strains specifically designed for removal of specific heavy metal removal (El-Sayed *et al.,* 2019). Also, comparative studies on few types of marine algal species are being done so as to assess their bioremediation potential. Likewise; in Egypt; two types of marine macroalgae namely *Colpomenia sinuosa* and *Ulva fasciata* were studied for their biosorption performance in cobalt contaminated aqueous solutions. It revealed that the presence of alginate in the cell walls of marine algal species as well as presence of metal ions in water bodies; can influence biosorption capacity. Removal of such metal ions can result in better the biosorption potential as observed in case of *C. sinuosa* for the removal of Co(II) ions and remediation (Dalia *et al.,* 2021). Various marine macro algal strains like *Laminaria japonica, Ulva lactuca* are being investigated for their bioremediation potential (Castiglia *et al.,* 2021; Sarker *et al*., 2021). Recently; numerous studies have been conducted by many researchers across the world; that highlighted significance of various algal strains as a sustainable biotechnological approach. Such algae based bioremediation i.e. 'Phycoremediation' can be an effective tool towards reduction of anthropogenic carbon footprints thereby realizing the green economy (Kaur and Reddesen 2022). Toxic heavy metals like Lead (Pb) is found to be leaching out of industrial effluents is considered as potential threat for the estuaries and aquatic ecosystems (Rehana *et al.,* 2022).

The World Health Organization (WHO) has set the permissible level of heavy metals like Mercury, Lead, Copper, Cobalt, Cadmium, Nickel, Arsenic, Zinc etc. and these can be removed successfully by various micro and macro algal strains with varying removal efficiency at a particular optimum operating conditions (Znad *et al*., 2022) (Table 3).

Table 3: Different algal biosorbents used in heavy metal removal.

Heavy metals	Allowed values by WHO (mg/l)	Biosorbent material (Algal strain) used	Removal Efficiency (%)/Adsorption Capacity (mg/g)	References
Mercury	0.05	*Ulva intestinalis,*	95%	(Fabre *et al.,* 2020)
		Ulva lactuca,	90%	
		Fucus spiralis,	85%	
		Fucus vesiculosus,	80%	
		Gracilaria sp.	90%	
		Osmundea pinnatifida	80%	
Lead	0.01	*Gelidium amansii, Sargassum natans, Ascophyllum nodosoum, Gracilaria corticata, Polysiphonia violacea*	100%	(Holon *et al*., 1993); (Jalali *et al.,* 2002); (El-Naggar *et al.,* 2018)
Copper	1.0	*Sargassum* spp., *Caulerpa lentillifera, Ulva reticulata*	80%	(Antunes 2003); (Vijayaraghavan *et al.,* 2004); (Madacha *et al*., 2006)
Cadmium	0.003	*Sargassum filipendula, Sargassum muticum*	0.43 mmol/g	(Nishikawa *et al.,* 2018); (Lodeiro *et al.,* 2005)
Chromium	0.05	*Cystoseira barbata, Padina boergesenii*	Cr(III):70.70% Cr (VI):35%	(Yalçın and Özyürek 2018; Thirunavukkarasu and Palanivelu 2007)
		Cystoseira crinite	Cr(III):73.34%	
Cobalt	0.08	*Hypnea Valentiae*	Raw adsorbent 10.98 mg/g Modified Adsorbent 16.66 mg/g	(Vafajoo *et al*., 2018)
Arsenic	0.01	*Sargassum muticum*	100%	(Vieira *et al*., 2017)
Nickel	0.015	*Sargassum Filipendula*	45%	(Moino *et al.,* 2017)
Zinc	3.0	*Ulva lactuca, Caulerpa scalpelliformis, Chaetomorpha antennia*	83.3 mg/g	(Jayakumar *et al.,* 2021); (Lahari *et al*. 2010)

FUTURE PROSPECTS IN ALGAL BIOSORPTION

The algal biosorption studies have focused the need of bioprospecting novel algal strains for treating raw industrial waste waters on commercial basis as well as evaluating novel techniques for low-cost seaweed based adsorbent regeneration. Also, few studies have shown that the metal binding capacity can be improved through modification in molecular and chemical extraction, use of nanoparticles etc. (Cheng *et al.*, 2019; Znad *et al*., 2022). Due to the better genetic abilities future studies need to be focused on biosorption of heavy metals from the soil through the use of genetically modified algal strains. Thus the omics based techniques should be emphasized so as to produce algal strains that are more tolerant to prevalent environmental conditions (Hemmat-Jou *et al*., 2018; Pande *et al*., 2022). Marine macro algal species; due to their rapid growth and regeneration; are being utilized for detoxification of polluted waters which is reported recently by many workers and is considered as easy and efficient way to achieve environmental protection. Likewise; researchers have conducted studies to examine the potential of marine algal strains like *Gelidium amansii* for biosorption of Ni^{2+} ions from aqueous solution and assessed its ability against independent variables like temperature, contact time, agitation etc. (Noura *et al.*, 2022). Recently; algal strains like *Caulerpa racemosa* which have been known for their invasion in Mediterranean ecological niche investigated for its potential in recovery of heavy metal pollutants like Zinc (Zn) (Landi *et al*., 2022). Likewise, use of algal strains like *Prorocentrum triestinum* for Cadmium removal from the waste water; as well as for preventing secondary disasters like sea water hypoxia; was attempted successfully (Xu *et al*., 2022).

The ability of different algal species to remove metals varied with algal group and morphology. Also, the morphological characteristics and surface composition of sea weeds after heavy metal bio sorption changes have also been explored and morphologically significant difference is found between raw seaweed and seaweed that been treated by metal sorption (Mingu *et al*., 2022). Recently; attempts are being made to ascertain detailed information about the hazardous health effects of toxic heavy metal pollution including isotherms, kinetics. Moreover; apart from the recovery and reuse of biosorbents; their use for sustainable developmental goals are being undertaken.

CONCLUSIONS

The biosorption techniques using marine algal species, is an effective tool for removal of toxic heavy metals from the different aqueous solutions and water bodies. Today world is witnessing industrial and biotechnological advancements in bioremediation process; However, numerous challenges still need to addressed; such as release of novel toxic elements, reliable methods to detect, monitoring and eco-friendly approaches for removal of such pollutants etc. In this context, the multidisciplinary research involving all stakeholders and policymakers; is needed in future. At global level; these considerations will be helpful to highlight degradation and or removal of life-threatening heavy metals and other pollutants from environment at a global level.

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