A Review on Low-Cost Adsorbents for Cadmium Pollutant Removal from Industrial Effluents

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Cadmium is biopersistant. Significant concentration of cadmium is released in the environment by various industrial activities worldwide. Therefore, removal of cadmium from waste water is now receiving greater attention from researcher various techniques for cadmium metal removal from industrial effluents has been developed. These techniques are: chemical precipitation, chemical reduction, ion exchange, evaporation, membrane processes, and adsorption. Each method has its own advantages and disadvantages among these techniques; adsorption of cadmium metal by using various materials was proved to be a very effective method. Moreover, research is needed to find the commercial and practical utility of low-cost adsorbents.

From the current review, adsorption seems to be the best alternative, help to develop new metal remediation technology and to study the scientific advances of recent years in this area.

Introduction

Cadmium occurs naturally in the environment by the erosion process, abrasion of rocks, volcanic eruptions and from forest fires. Cadmium has been widely dispersed into the environment by its mining, smelting as well as by other man-made activities.

Cadmium is a heavy metal with a high toxicity at very low exposure levels. It has acute and chronic effects on health and environment. Cadmium is not degradable in nature and thus stays in circulation once released to the environment.

When cadmium enters in human bodies, above certain level it may cause liver damage, renal damage, hyper tension, anemia, disturbances of calcium metabolism and formation of stones in the kidney. High exposure can lead to lung cancer and prostate cancer.

In India the maximum permissible limit for cadmium concentration in drinking water is 0.003 mg/l (Government of India, 2000). Cadmium classified as B1 carcinogen and category I carcinogen by US Environment protection agency in 1999, and International Agency for Research on Cancer respectively. Department of Environment, UK listed Cd (II) in the red list of priority pollutants because of its adverse health effect also Cadmium is black listed in EEC dangerous substance directive (EEC black list substance, 1976) (Pandey *et. al.*, 2008)

There are various techniques for removal of cadmium such as chemical precipitation, chemical reduction, ion exchange, evaporation, membrane processes, and adsorption. These processes have its own some disadvantages like produce large quantities of sludge which, when disposed unscientifically, can cause even more acute problems (Sahu *et. al.*, 2018). Therefore, it is require developing a cost effective, sludge free and prominent technique for the removal of cadmium from waste water. In light of all these requirements, adsorption is one of the best processes due to its ease in operation and high binding capacity with the metals (Sekar *et. al.* 2004). Hence this paper focuses on to provide a historical review and comparison between the conventional and non-conventional techniques of adsorption, there pros and cones in the actual practical environment.

Different techniques for cadmium remediation

There are various methods used to treat mining and industrial effluents containing Cd (II). These methods can be broadly divided into the following categories:

- Chemical methods
- Membrane separation technique
- Ion exchange technique
- Solvent extraction technique
- Adsorption technique

Chemical methods

The chemical methods for cadmium removal include precipitation and cementation techniques (**Fig. 1.**).

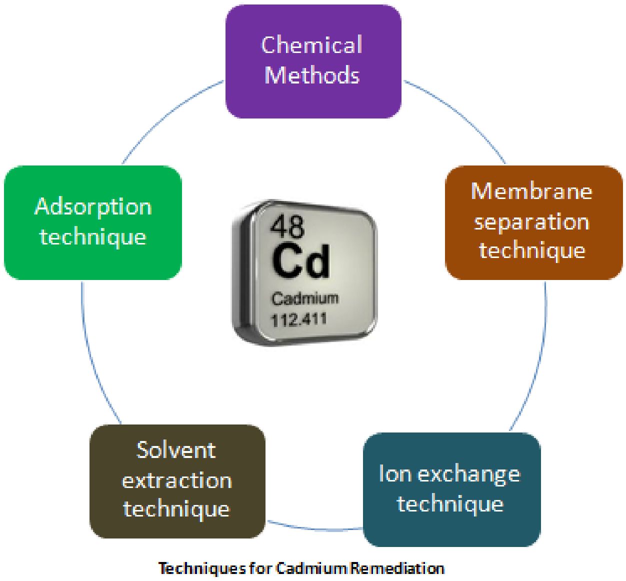

Fig. 1. Technique for Cadmium Remediation.

Precipitation

Chemical precipitation is the most widely used method for heavy metal removal from inorganic effluents because of its low cost and simplicity. Chemicals react with heavy metal ions to form insoluble precipitates of metal hydroxide, metal carbonates, or metal sulfides. Carbonates and phosphates (insoluble solid particles) that can be simply separated by sedimentation or filtration. (Fu. and Wang, 2011) (**Table 1**).

Table 1. Cadmium removal data using Precipitation Technique.

Techniques	% Recovery	Cd Concentration		Reference
		Initial	Final	
Aerophine 3481A	99.7%	18mg/L	0.05mg/L	Rickelton, 1998
Lime followed by Na2S	98%	150 mg/l	3.0 mg/l	Charerntanyarak (1999)
lime/Mg(OH)2	~100%	1mM	nil	Lin *et al., 2005*
Na2S	99.9%	7500mg/L	10 mg/L	Islamoglu *et. al., 2006*
Electro coagulation	>99%	50-250mg/L.		Bazrafshan *et. al.*, 2006
Pyrite and synthetic iron sulfide	99.8%	20 g/l		Özverdi and Erdem (2006)
Dithiophosphate	99.9%	200 mg/l		Ying, & Fang (2006)

Cementation

The cementation process was successful for recovery of Cd^{2+} ion into industrial wastewater sample, as well as verity of heavy metals. Cementation is a type of precipitation, a heterogeneous process in which ions are reduced to zero valences at a solid metallic interface. The process is often used to refine leach solutions [**1**] (**Table 2**).

Table 2. Cadmium removal data using Cementation Technique.

Techniques	% Recovery	Cd Concentration		Reference
		Initial	Final	
Magnesium	½ Order	<25mM		Gould et al., 1986
Magnesium	1st Order	>25mM		Gould et al., 1986
Zinc powder	1st Order	<500mg/L		Ku *et al., 2002*
Zinc powder+ SDS	95.6%	6.5 µg/L	0.28µg/L	Taha and Ghani, 2004

Membrane separation technique

Membrane separation is promising technique for the removal of heavy metals; also, this technique is space-saving and easy to operate. Nano filtration, ultra-filtration reverse osmosis are some of the common membrane separation techniques. (Fu. and Wang, 2011) (Table 3)

Ultra filtration

Ultra filtration (UF) is useful for the removal of dissolved and colloidal materials; pore size of UF membrane is larger than dissolved metal ion so the ion passes through UF membranes. Ultra filtration is use for the range of 0.1-0.001 micron suitable for water and low molecular weight solute. (Kanamarlapudi *et. al.*, 2018). To increasing the removal efficiency, the polymer enhanced ultrafiltration (PEUF) and micellar enhanced ultrafiltration (MEUF) was proposed. (Fu. and Wang, 2011)

Nanofiltration

Pore size of nanofilters is in between 0.5-2 nm, so this can be use for the size of molecule with the range of 300 to 500 Da. (Kanamarlapudi *et. al.*, 2018).

Many researchers have tried the electro-dialysis technique and electro coagulation as effective method for cadmium removal. The advantage of this method is no sludge formation and no need of additional chemicals. However, it is inefficient at low metal concentration. (Rao *et. al.*, 2010).

Table 3. Cadmium removal data using Membrane separation Technique.

Techniques	% Recovery	Cd Concentration		Reference
		Initial	Final	
TOPS 99	99%	0.89mM		Swain *et. al., 2006*
D2EHPA+TRPO	98.6%	0.18mM	2.5µM	He *et. al.*, 2007
Cyannex 923	85.8%	0.89mM	0.13mM	Alguacil and Navarro, 2001
MEUF Polysulfone	92-98%	50 mg/L		Huang *et. al.*, 2010
MEUF Amicon regenerated Cellulose	99%	0.5 mM		Landaburu-Aguirre *et. al.*, 2010
PEUF Polysulfone	99%	112.4 mg/L		Ennigrou *et. al.*, 2009
Commercial NF membrane from aqueous solutions	82.69%,	5 mg/l		Murthy and Chaudhari, 2009

Ion exchange Technique

Ion-exchange techniques have been widely used to remove heavy metals from industrial effluent water due to its many advantages, such as high treatment capacity, high removal efficiency and fast kinetics (Kang *et. al.*, 2004). Ion exchange process is a chemical reaction between an electrolyte in solution and an insoluble electrolyte with which the solution is contacted. The ion exchange between the heavy metal ion and hydrogen ion is shown in Eq. (2)

$$M2^{+} + 2RH = R2M + 2H^{+} \quad (2)$$

where M is bivalent metal and R is the aliphatic portion of the cation exchanger.

Due to the use of chemicals for the generation of resin, ion exchange requires high initial cost equipments as well as high operational cost. Hence it is not a popular method for removal of cadmium metal ions from industrial wastewaters. (Rao *et. al.*, 2010). (**Table 4**)

Table 4. Cadmium removal data using Ion exchange Technique.

Techniques	% Recovery	Cd Concentration		Reference
		Initial	Final	
S-950	83.9%	1mM	0.16mM	Koivula *et. al.*, 2000
Amberlite IRC-718	~100%	0.25mg/L	nil	Malla *et. al.*, 2002
Na-Amberlite IR 120	93.4%	20mg/L	1.32mg/L	Kocaoba and Akcin, 2005
Amberlite IRC-718	99.5%	20mg/L	0.1mg/L	Fernandez, 2005
Resin A	91%	1060mg/L	96.46mg/L	Wang and Fthenakis, 2005
Dowex 50W	97%	5mM	0.15mM	Pehlivan and Altun, 2006
Amberlite IR 120	97.4%	20mg/L	0.52mg/L	Kocaoba, 2007
Purolite C-160	97.7%	31.1mg/L	0.719mg/L	Bożęcka, *et. al.*, 2016

Solvent extraction technique

Solvents extraction technique is a method to separate metal ions or compounds from aqueous solutions having higher concentrations to obtain pure solution based on their relative solubility in two different immiscible liquids, usually water and organic solvent.

The disadvantage of this method is that the large amount of solvent of the extracted phase should be refreshed in a costly stripping step. This method may not remove heavy metals whose concentration is very less as the recovery cost will be very high. (Rao *et. al.*, 2010). (**Table 5**)

Adsorption technique

Conventional techniques have their own disadvantages like incomplete metal removal, generation of sludge, high reagent and energy requirements, and aggregation of metal precipitates and fouling of the membrane, production of secondary sludges and costly for their disposal. (Maran and Protton, 1971).

Table 5. Cadmium removal data using Solvent extraction technique.

Techniques	% Recovery	Cd Concentration		Reference
		Initial	Final	
1M D2EHPA	99.7%	30g/L	0.09g/L	Nogueira and Delmas., 1999
0.2M Cyanex 923	98±1%	1mM		Gupta *et. al., 2001*
D2EHPA+TPEN	99.9%	0.89mM		Takeshita *et. al., 2003*
20%TBP	99.9%	0.5mM		Mellah and Benachour, 2007
Cyanex471	>97%	0.541g/L	<0.016	Reddy *et. al.*, 2008
D2EHDTPA	99.1%	1g/L	<2 µg/L	Touati *et. al.*, 2009
0.15M D2EHPA	~100%	4.45mM	Nil	Kumar *et. al.*, 2009
Cyanex301	99.95%	90g/L	0.045g/L	Qian *et. al.*, 2018

Adsorption is powerful technique for heavy metal wastewater treatment. The adsorption process are associated with various advantages like: low cost of adsorbent, easy availability, utilization of industrial, biological and domestic waste as adsorbents, low operational cost, ease of operation compared to other processes, environmentally friendly, cost effective and can be regenerated by suitable desorption process.

The last few decades have witnessed remarkable interest in development of new adsorbents and to modify the performance of existing ones. The use of different materials as biosorbents is explained in detail.

Activated carbon adsorbents

Activated carbon adsorbents are widely used in the removal of heavy metal contaminants. Its usefulness derives mainly from its pore size, pore distribution and number of surface oxygen. A large number of researchers are studying the use of AC for removing heavy metals (Jusoh *et. al.*, 2007; Kang *et. al.*, 2008).

Some suitable oxidizing agents and thermal treatments were used to manipulate the surface oxygen to get the surface functional groups such as carboxyl, phenolic and lactonic group attached to carbon (Toles et al., 1999; Park and Jang 2002). These groups can improve the adsorption capacity and selectivity on certain adsorbate in the gaseous or liquid phase. (Barton *et. al.*, 1997; Pradhan and Sandle, 1999).

Activated carbon such as high carbonaceous materials was used to remove cadmium ions from wastewater. (Rao *et. al.*, 2006; Cheung *et. al.*, 2001; Ricordel *et. al.*, 2001; Ferro-García *et. al.*, 1988). Adsorption capacity of activated carbon was improved by treating with sulfur (Gomez-serrano *et. al.*, 1998), sulfur dioxide (Macías-

García *et. al.*, 2003), surfactant (Nadeem *et. al.*, 2009) and electrochemical oxidation (Rangel-Mendez *et. al.*, 2000). Activation was also done by using $ZnCl_2$ (Kula *et. al.*, 2008). Activated carbon was prepared from coconut coir pith (Kadiravelu and Namasivayam 2003). Activated carbon was derived from bagasse (Mohan and Singh 2002). The study on cadmium removal was carried out using fly ash (Visa and Duta 2008). The various experimental parameters studied for optimizing uptake capacity included contact time, adsorbate concentration, adsorbent dosage, pH and temperature.

Synthetic, naturally occurring and waste oxidic materials as adsorbents

Iron oxides/hydroxides/oxyhydroxides form an important category of low-cost adsorbents for removal of heavy metals and organic compounds form wastewater (Fendorf *et. al.*, 1997; Heijman *et. al.*, 1999; Venema *et. al.*, 1998). (Sen and Sarzali 2008) used synthetic Al_2O_3 (Merck) to find out its adsorption capacity for the removal of cadmium from solutions. Mixed oxides of iron and aluminum were used by (Srivastava *et. al.* 1988) to study the adsorption behaviour of cadmium. Manganese oxides/hydroxides are known to be good scavengers for toxic metal ions. Cornelis and Weijden (1976) carried out experiments on the uptake of zinc and cadmium by manganese oxides.

A number of low-grade ores containing oxide minerals, sludges and residues have been used to treat Cd (II) contaminated water. Mining waste like chromite overburden has been reported (Mohapatra and Anand, 2006; Mohapatra and Anand, 2007 (a) to be a good adsorbent for Cd (II).

Biosorbents

The research on removal of cadmium by using various biological methods has been attempted by many researchers. Biosorption of heavy metals from aqueous solutions is a relatively new process that has been confirmed a very promising process in the removal of heavy metal contaminants. It is desirable to focus on the generation, search of higher adsorbent biomass and to develop a practical and environmental friendly method for the removal of heavy metal, with the capacity to bind metal ions with greater affinities. Biosorption processes are particularly suitable to treat dilute heavy metal wastewater. Wide ranges of materials available in environment can be used as biosorbents for the removal of metals from contaminated water. Biosorbents can be derived from different sources as follows (Apiratikul and Pavasant, 2008 (b): (1) non-living biomass such as stems, peels, husks, shells, leaves bark, lignin, shrimp, krill, squid, crab shell, etc.; (2) algal biomass; (3) microbial biomass, e.g., bacteria, fungi and yeast. Biosorption of Cadmium metals is a complex process that affected by several factors including pH of the solution, initial metal ions concentration, contact time, temperature and adsorbent dose (**Fig. 2**).

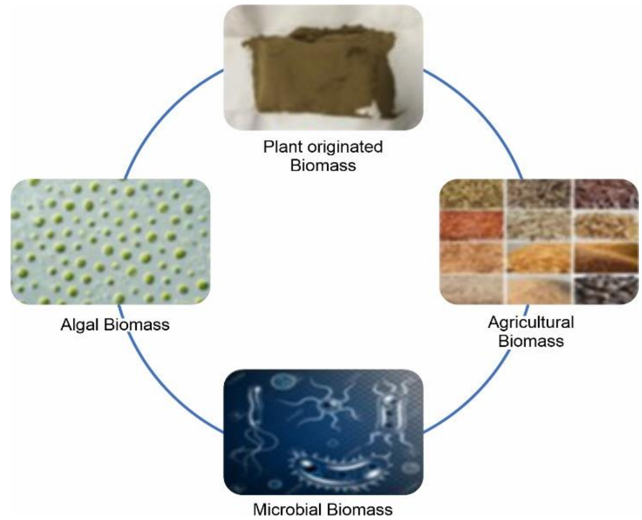

Fig. 2. Different types Biosorbents.

Non-living biomass /Agricultural based biosorbents such as stems, peels, husks, shells, leaves bark, lignin, shrimp, krill, squid, crab shell, etc.

The waste material obtained from agriculture is generally high in lignin and cellulose. The functional groups present in this type of material are mainly carbonyl, ether, alcoholic, amino and phenolic groups with good binding ability. (Hossain *et. al.*, 2012) These groups have the ability to bind heavy metal by replacement of hydrogen ions for metal ions in solution or by donation of an electron pair from these groups to form complexes with the metal ions in solution.

The abilities of 15 coniferous barks for removing toxic heavy metal ions were investigated (Seki *et. al.*, 1997). Of the barks tested, high adsorption ability for heavy metal ions was found in *Picea abies* (Norway Spruce). The continuous column experiments using *P. jezoensis* bark for Cd(II) adsorption indicated that packing had retained 10.1-14.2 mg/g adsorbent until the column broke through (**Fig. 3**).

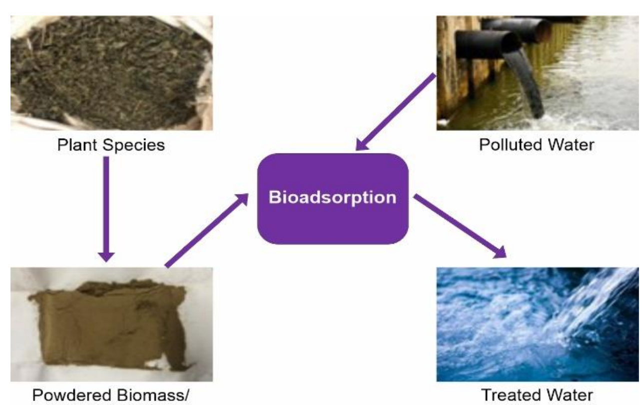

Fig. 3. Process of Biosorption.

As low-cost adsorbent eucalyptus bark is well fitted with nonlinear method to obtained langmuir equation in batch mode study. (Ghodbane *et. al.*, 2007). The Kinetics of cadmium adsorption on tree fern has been well represented psudo-langmuir and psudo Redlich-Peterson data. (Ho and Wang 2004). According to Tan and Xiao (2009) ground wheat steam is effective for removal of cadmium. FTIR and XPS studies indicate that the COO^- present in adsorbent mainly active group for adsorption of cadmium. It is also observed that the as the functional group increases the binding capacity of adsorbent also increases. The adsorption capacity of Grapefruit peel was reported 42.09 mg/g from aqueous solution which on equilibrium study better fit with Frendulich isotherm. (Torab *et. al.*, 2013). Sun and Shi 1998 were reported that the sunflower stalks with finer size adsorbent particle adsorbed maximum 42.18 mg/g cadmium, adsorption rate decreases as the temperature rise. The adsorption of lead and cadmium by grape stalk waste was reduced due to presence of NaCl and $NaClO_4$ and other metals in the solution. (Martinez *et. al.*, 2006). Calotropis Procera is a wild perennial plant shows high uptake capacity of Cadmium in between Ph 5.0 and 8.0. Maximum absorption was found to be 40 and 50.5 mg/g. (Pandey *et. al.*, 2008).

Hazelnut shell, an agricultural waste shows a good efficiency in removing from aqueous solution toxic ions such as cadmium, zinc, chromium (III) and (V) from wastewaters. (Cimino *et. al.* 2000).

The biosorption capacity of Acacia leucocephala bark powder, a low-cost bio adsorbent is 167.7 mg/g from the aqueous solution. Hydroxyl, amine and carboxyl groups present on the surface of biosorbent are involve in this absorption process. (Munagapati *et. al.*, 2010) (**Table 6**).

Table 6. summarizes the type of the biosorbent and maximum biosorption capacity of the different agriculture wastes as biosorbents for removal of cadmium from aqueous solutions.

Adsorbent	Biosorption Capacity/efficiency (mg/g or %)	Reference
Grapefruit peel	42.09 mg/g	Torab *et. al.*, 2003
peas peel	118.91 mg/g	Benaissa, H. (2006)
Sawdust of cedrus deoda	97%	Memom *et. al.* (2007)
sawdust of Pinus sylvestris	96%	Costodes *et. al.* (2003).
Poplar wood sawdust	31.9%	Sciban *et. al., (*2006)
Papaya wood	94.9%	(Saeed *et. al., (*2005)
Green coconut shell (powder)	99%	Pino *et. al.*, (2006)
medlar peel	98.14 mg/g	Benaissa, H. (2006)
Date pit	39.5 mg/g	Al-Ghouti *et. al.*, (2010)
Cassava Peelings	119.6 mg/g	Horsfall *et. al.*, (2004)
Hazelnut shell	5.42 mg/g	Cimino *et. al.*, (2000)
Grape stalk	27.88	Martinez *et. al.*, (2006)
Rice husk	103.09	(Ajmal *et. al.*, (2003)
Acacia leucocephala bark	167.7mg/g	Munagapati *et. al.*, (2010)
fig leaves	103.09 mg/g	Benaissa, H. (2006)
Calotropis procera	>90%	Pandey *et. al.*, (2008)

Algal biomass

Algae's having high absorption capacity with bountiful availability attracted the attention of researches. The several advantages of using algae as biosorbent are: (Fu, F., & Wang, Q. 2011, Pahlavanzadeh *et. al.*, 2010)

1. They are renewable natural biomass
2. High surface area to volume ratio
3. Less sludge deposition
4. Grow faster
5. Potential for metal regeneration and recovery
6. Economic and eco-friendly

Microalgae are very effective for the removal of heavy metal due to high binging affinity and large surface area (Roy *et. al.*, 1993). The composition of algae cell wall is differing for different groups of algae. The major constituents of brown algae are alginic acid polymer of mannuronic & guluronic acid with its salt, cellulose and sulfated polysaccharide with carboxylic group which are responsible in metal absorption. (Romera *et. al.*, 2007) Fucus Vesiculosus a verity of brown algae has greater metal uptake capacity than red green algae. (Mata et al., 2008) Arkipo *et. al.*, 2004 has been reported that the green algae Chlorella emersonii is used for removal of cadmium from waste water (**Table 7**).

Table 7. summarizes the type of the biosorbent and maximum biosorption capacity of the different Algal biomass as biosorbents for removal of cadmium from aqueous solutions.

Adsorbent	Biosorption Capacity/efficiency (mg/g or %)	Reference
Ulva lactuca sp.	35.72 mg/g	Lupea *et. al.*, (2012)
Enterobacter sp.	46.2 mg/g	Lu *et. al.*, (2006)
Algae, marine, dead Biomass	80 mg/g	Herrero *et. al.*, (2006)
Algae, Nile water	37.43 mg/g	Sherif *et. al.*, (2008)
Spirogyra	90%	Singh *et. al.*, (2012)
Ascophyllum nodosum	38	Lodeiro *et. al.*, (2005)

Microbial biomass

Removal of heavy metal with the help of microbial biomass is a highly effective technique. These are the low cost bioadsorbents and still going through the experimental phase. Separation of biosorbent is quite difficult after adsorption process. Biosorption of heavy metals from bacterial biomass include a number of bacteria such as Pseudomonas aeruginosa (Gabr *et. al.*, 2008), Escherichia coli (Souiri *et. al.*, 2009), Bacillus cereus (Pan *et. al.*, 2007). Yeast and fungi are easy growing, high yield biomass. Aspergillus niger (Amini *et. al.*, 2009), Rhizopus arrhizus (Aksu & Balibek 2007), Trametes versicolor mycelia (Arica *et. al.*, 2001) are the common spcies that are used as fungai biomass. (**Table 8**)

Table 8. Summarizes the type of the biosorbent and maximum biosorption capacity of the different microbial biomass as biosorbents for removal of cadmium from aqueous solutions.

Adsorbent	Biosorption Capacity/efficiency (mg/g or %)	Reference
Saccharomyces Cerevisiae (yest)	15.63 mg/g	Göksungur *et. al.*, (2005)
Mucor rouxii	76%	Yan & Viraraghavan (2000)
Alcaligenes eutrophus(bacteria)	122	Mahvi & Diel, (2004)
Aspergillus niger (Fungi)	15.50 mg/g	Liu *et. al.*, (2006)
Desulfovibrio alaskensis 6SR	99.9%	Perez *et. al.* (2015)

For the removal of cadmium from aqueous solution, number of effective low-cost biomass have been reported, but still more attention required in the evolution of commercial use of microbial biomass.

Desorption of biosorbents

To keep the biosorption process economic and Regeneration of precious metals after biosorption, desorption of biomass play an important role. (Volesky, 2001)

Desorption process must satisfied the following conditions

- No change in physical form of biomass.
- No change in metal uptake capacity of bio mass
- Regeneration of metal in concentrated form

Mechanism of desorption involve ion exchange and complexation process, that is very similar to bioadsorption. Although removal of heavy metal from biosorbent by suitable eluting agent involves different desorption mechanism. Mineral acid such as HCl, H_2SO_4 and HNO_3 are able to displace the metals from sorbent binding site. (Aldor *et. al.* 1995) Another strong chelating agent i.e., EDTA is considered as efficient to regenerate biomass which is loaded by cadmium. (Chu *et. al.* 1997)

In desorption process solid to liquid ratio (S/L) is an important parameter in which mass of used or loaded biomass is taken in grams and volume of eluting agent is taken in liters. Higher will be the S/L ratio, higher the metal concentration in eluting agent. (Aldor *et. al.* 1995) the S/L ratio is also related to Concentration ratio (CR). Higher CR ration indicate the effective sorption process that makes the recovery of metal more advantageous. (Volesky, 2001). Therefore, desorption is economical and ecofriendlly process (*Tewari et. al.,* 2005).

Conclusion

Cadmium is hazardous and toxic ions for living organisms, reported to cause several health-related issues. High exposure may lead to cancer also. Removal of cadmium from waste water can be a challenge for researchers. The review addresses some conventional methods for cadmium removal with their pros and cones. Main category of adsorbents is generally: Activated carbon adsorbents, Synthetic and naturally occurring and waste oxidic materials, Non-living biomass or Agricultural based Biosorbents, algae and microbial biomass. Above all the plat originated biomass are specially recommended for removal process, however there are several plat species are reported to adopt and resist heavy metals in highly contaminated zone. Biosorbent obtained by the plat originated species are beneficial over most of the conventional methods because most of them are local species, economic, reusable, safe disposal and effective in removal process, but there is gap between the available literature of use and reuse, regeneration or disposal of biosorbents. An encouraging environment that supports the commercial use of biosorbents needs to be created and in near future it is expected that the situation is changed and biosorption technology is frequently used for removal of heavy metals than the physiochemical technology of removal.

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