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REVIEW ARTICLE

BIOSORPTION OF Ni(II), Zn(II), Cd(II) And Pb(II) IONS FROM WATER AND WASTE WATER USING DIFFERENT BIOSORBENTS: A REVIEW

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ABSTRACT

On account of our distorted belief system and vague philosophy there is excessive industrialization, disproportionate traffic congestion in our cities and alarming increase in the use of ammunition. This trend is visible worldwide. Our indulgence in such reckless acts is not only spoiling the environment in an irreversible manner but is also responsible for many such suffering of the mankind which was not there earlier. All these activities are responsible for the emission or surge of heavy metals such as Cadmium, lead, Zinc, Mercury, Chromium, Nickel, Arsenic, Copper, Cobalt, Thallium, Iron, Barium, Selenium and Vanadium etc. As these heavy metals are non-biodegradable, means not capable of being decomposed to harmless substance by bacteria or other living organisms therefore through air or water these metals attack the human being and the outcome are visible in the form of different kind of cancers, Genetic disorder, Rheumatic disorder or malfunctioning of bones etc. The current study is an attempt to explore the possibility of tackling these heavy metals in the waste water through the process of adsorption using economically visible alternatives.

INTRODUCTION

With increasing human *modernity* or *civilization*, industrial activities have gradually reshuffled several toxic pollutants from earth crust to the environment, we cannot deny it. Beside this, no one is actually interested in environmental issues, as their disperse life schedule keep them occupied and people always keep environmental issues at a bay from them, we cannot rule out the possibility. Among all environmental issues, water pollution demands a great emphasis or concern. As water is a constitutive treasure for all living organisms. We cannot speculate our life without water. It is playing a vital role to fulfilling the criteria of life or we can say that water is working just like a '*life preserver'*. Our body consists of about 60-70% of water. A person cannot be pertinent to live for a week without water. It is only the water which makes our planet peerless in this Universe. Several pollutant known to pollute water, out of them one is the presence of heavy metals. Because, the existence of heavy metals in water is a kind of venom which works slowly in destroying the living organisms. Our environment includes some pollutants that can be destroy, but heavy metals are known as the "*non-biodegradable*" source means we cannot destroy them. So their removal from water and waste water demands a separate study as it became a concern of global issue. A venom "*heavy metals/semi metals (metalloids*)" are relatively dense metals that are known for its toxicity especially concerning to environment (Banfalvi, 2011). The term "*heavy metal*" is generally applied to a class of metals/metalloids having density more than 5 g/cm^3 and atomic number greater than 20 (Raut *et al*., 2012).The presence of heavy metals in the eco-system leads to a numerous environmental issues or eco-toxicities like diminution due to its persistent accumulation. Ni, Pd, Cd, Cu, Zn, Fe, Co, Au etc. these metals are highly toxic when their concentration crosses there acceptable limit in this eco-system, intake of venom metals may cause diverse health disputes like kidney damage, skin dermatitis, anaemia, minimata disease and itia-itai disease (Borba *et al*., 2006). Increasing accumulation of toxic heavy metals in eco-system or in food chain lead to tremendous force for their purification and eradication. Several scientists have made various attempts for their removal from waste water, but the techniques adopted by them have some limitations like cost effectiveness, production of high sludge, handling, disposal problem, technical requirements and the main thing regarding this is that it is not Eco friendly anymore.To overcome the limitations of conventional methods it becomes urgent to develop a process which should be superior on these methods. It have been proved by Various scientists that "*bio-sorption technique*" is more desirable method for the remediation of heavy metals such as Pt(II), Ni(II), Pb(II), Cd(II), Cr(II), Hg(II), Cr(III), Zn(II), and Cr(VI) etc.

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"*Bio-sorption*" can be defined as the characteristic property of certain type of non-living, inactive bio molecules which have adequate capacity as metal sequester even from very dilute one (Ahluwalia & Goyal, 2007). Over recent years, research or the outcome obtained by using biosorption process evoke it an ideal alternative for eradication of heavy metals. It is a physiochemical process. The accruement of chemical substance between two immiscible faces (solid-liquid, liquid-gas and liquid-solid) is called adsorption. It is a surface phenomenon. It has been found that larger the surface area more will be the adsorption. The process of removal of adsorbed substance from the surface is called desorption. Due to high efficiency, production of minimum biological sludge, lesser energy requirement, ecofriendly, cost effectiveness, easiest and safest, possibility of metal recovery and easy handling adsorption process has been found to be more applicable as compared to other methods for the treatment of heavy metals from discharge water (Gholizadeh & Ziarati, 2016).

The presence of functional group (hydroxyl and carboxylic groups), mainly cellulose, lignin and other components such as lipids, hemicelluloses, proteins, starches, simple sugar, water and hydrocarbons (Ahluwalia & Goyal, 2005; Calace *et al*., 2003; Joshi *et al*., 2003; Low *et al*., 1995; Marin & Ayele, 2002; Vaughan *et al*., 2001;Verma & Shukla, 2000; Villaescusa *et al*., 2004) on the surface of agriculture waste or adsorbents plays an vital role in binding of adsorbent metals to remove heavy metals from waste water (Sud *et al*., 2008). Agricultural wastes adsorbents has raised great interest in the field of environmental study and are characterized by affordability, availability, eco friendliness and high removal capacity (Kehinde *et al*., 2009). Many researchers have conducted experiment emphasizing on decreasing the pollutants in the polluted water. The main focus of their study is that how to treat the industrial waste water with the help of low cost adsorbent. Different kind of bio- products like Sugarcane bagasse (Mohan & Singh, 2002; Khan *et al*., 2001; Ayub *et al*., 1998; Ayub *et al*., 2001; Ayub *et al*., 2002), Rice husk (Srinivasan *et al*., 1988; Ajmal *et al*., 2003; Suemitsu *et al*., 1986; Khan *et al*., 2003), Sawdust (Ajmal *et al*., 1996; Ayub *et al*., 2001; Kadirvelu *et al*., 2003; Khan *et al*., 2003; Selvi *et al*., 2001; Tan *et al*., 1993), Oil palm shell (Khan *et al*., 2003), Coconut husk (Tan *et al*., 1993), Neem bark (Ayub *et al*., 2001),Wool, Pine needles, Olive cake, Almond shells, Charcoal, Cactus leaves (Dakiky *et al*., 2002), Hazelnut shells (Cimino *et al*., 2000; Demirbas, 2003; Dakiky *et al*., 2002), Banana and Orange peels (Annadurai *et al*., 2003), different Agro waste materials (Qaiser *et al*., 2007), Activated carbon (Marzal *et al*., 1996), Granulated blast-furnace slag (Dimitrova & Mehandgiew, 1998), Okra waste (Hashem, 2007), Marine algae (Holan & Volesky, 1994), Seaweed biosorbent (Lee &Volesky, 1997), Olive pomace (Pagnanelli *et al*.,2003), Olive residue (Gharaibeh *et al*., 1998), Sunflower stalks (Hashem *et al*., 2006), Peanut hulls (Hashem *et al*., 2005) etc. have been investigated for the heavy metals treatment.

Tan and Xiao (2009) carried an experiment for Cadmium removal from aqueous solution using ground wheat stems. Results indicated that 0.1032 mmole of Cadmium uptake per gram of ground wheat stems and this uptake is powerfully affected by pH; at pH 5.0 maximum uptakes have been observed. Single and binary removal of Copper(II), Nickel(II) and Methylene blue (MB) from aqueous solution have been investigated on raw and pretreated Spirogyra sp. using various parameters like initial pollutant concentration, pH, contact time, ionic strength (NaCl) and biosorbent dosage. Results showed that raw and pretreated spirogyra sp. has adequate removal efficiency is on the order of MB> Cu > Ni and Ni > MB > Cu (Guler & Sarioglu, 2013). Hazelnut shell was used to investigate the removal capacity of Cd(II), Zn(II), Cr(III) and Cr(VI). Batch experiment showed that metal adsorption was pH dependent and maximum removal was attained at a specific range of pH. While highest Cr (VI) removal was observed at pH range of 2.5-3.5 (Cimino *et al*., 2000). In 2003, Demirbas carried an experiment to remove Co(II) using activated carbon obtained from hazelnut shell with varying parameters such as the agitation speed (50-200 rpm), initial metal ion concentration (13.30-45.55 mg/l), pH (2-8), particle size (0.80-1.60 mm) and temperature (293-323 K). It has been observed from this study that adsorption of Co (II) is pH dependent and kinetics follows pseudo-second-order equation. A particle size of 1.00-1.20 mm, at pH 6 adsorption capacity (Q_0) calculated from the Langmuir isotherm was 13.88 mg Co(II)/g at 303K. Dakiky *et al*., (2002) carried an experiment for the treatment of Cr(VI) from industrial waste water by using Cactus leaves, Wool, Sawdust, Almond shells, Olive cake, Pine needles and Charcoal at different metal ion ratios and significant results were obtained. An experiment was examined at 30 ^{0}C for the treatment of Cu(II), Ni(II), Zn(II), Co(II), and Pb(II) having concentration range of 5- 25 mg/L. It has been revealed that adsorption increased with increasing pH as Pb(II)> Ni(II)> Zn(II)> Cu(II)> Co(II). Adsorption was best described by the Freundlich equation (Annadurai *et al*., 2003). Many aquatic plants(Salviniales, Azolla filiculoides, Blechnum indicum, Blechnum cartilagineum, Blechnum chambersii) have capacity for accumulation of heavy metals (called hyper accumulators) among them *Azolla caroliniana* was used to treat Cr(VI), Hg(II), and Cr(III) (Bennicelli *et al*., 2004). The aquatic fern *Azolla* have adequate removal capacity for various heavy metals. *Azolla filiculoides* has been used for the treatment of Au(III) from aqueous solution and 86-100% removal was obtained. It has been observed that with increasing metal ion concentration adsorption also increases (Antunes *et al*., 2001). Azolla filiculoides has been also utilized for the removal of lead. Result showed that the removal capacity ranging from 30% of the initial lead concentration at pH 1.5 to approximately 95% at pH of 3.5 and 4.5 (Sanyahumbi *et al*., 1998). Algae Chlorella has been used to treat Au(II), Ag(I), Hg(II) and Cr(VI) ions and results showed that algae biomass have good metal uptake capacity (Darnall *et al*., 1986; Zhao & Duncan, 1997). In 1999 an experiment was carried out to investigate Cu (II) removal using immobilised and non-immobilised Azolla filiculoides. Result showed that efficiency of Cu (II) removal was in the order as epichlorohydrin-immobilised Azolla> milled-sieved Azolla> untreated Azolla (Fogarty *et al*., 1999). On the basis of these investigations Azolla has been proven a good metal sequester. Removal of Lead, Zinc, and Copper from *Ceratophyllum demersum* (C. demersum) has been investigated. Data obtained from this study indicated that *C. demersum* was significantly used to remove Copper, Lead and Zinc from solution. Data obtained from batch studies revealed that within 20 minute equilibrium was attained, adsorption can be well explained with Langmuir Model and the process was best delineated by pseudo second-order kinetics (Keskinkan *et al*., 2004).

Treatment of heavy metals using magnetic Nano material has also been reported (Gautm *et al*., 2014). Nano particles of $CuFe_2O_4$ and $NiFe_2O_4$ have been investigated to treat heavy metals Zinc, Nickel, and Copper from synthetic wastewater (Sezgin *et al*., 2013). The uptake of Zn(II), Ni(II) and Cu(II)by using $CuFe_2O_4$ as Nano particles was calculated as 99.80%, 98.85% and 83.50%, respectively and by using $NiFe_2O_4$ as Nano particles the uptake of Zn(II), Ni(II) and Cu(II) found to be 99.91% , 36.56 % and 92.55%, respectively. It was found that wheat bran a side-product of wheat milling industries has better adsorption capacity of Copper ions. Magnetic polyrhodanine Nano particles has been used to treat Hg(II) ions. The kinetic study followed a pseudo-second-order equation and the adsorption equilibrium study was best delineated by Freundlich isotherm model (Song *et al*., 2011). Humic acid (HA) coated with Fe_3O_4 nanoparticles (Fe_3O_4/HA) have been used to treat toxic metals such as Cd(II), Pb(II), Hg(II) and Cu(II) from waste water. Equilibrium attained Within 15 minutes and adsorption is best described by Langmuir adsorption isotherm model with highest adsorption capacities from 46.3 to 97.7 mg/g. Results showed that at optimized pH, 99% removal was obtained for Hg(II) and Pb(II), and 95% removal was attained for Cu(II) and Cd(II) in natural and tap water (Liu *et al*., 2008). Some low cost adsorbent after *chemical modification* such as Jackfruit, Pecan shells, Rice husk, Hazelnut shell, Maize cob are also used as an adsorbent for heavy metal removal. H_2SO_4, HCl, HNO_3, NaOH, KOH, acetone, polymerization, chloroform, H_2O_2, Fenton reaction, ethanol, tetra ethylene glycol, diethyl ether, and glycol have been used as a modifying agent to enhance the adsorption capacity of wood based adsorbents. For modified Pine bark maximum adsorption capacities were obtained by NaOH (98 and 99%), HCl (85 and 88%), Fenton (94 and 96%), polymerization (94 and 97%), tetra ethylene glycol (97 and % 98%), KOH (96 and 98%)and diethyl ether (95 and 97%), acetone (65.5 and 69%), chloroform (82 and 85%), HNO_3 (75 and 77%), H_2SO_4 (76 and 76%), ethanol (61 and 63%) and glycol (90 and 94%) for Cd(II) and Pb(II) uptake respectively (Argun & Dursun, 2006). As a result of modification adsorption capacities of adsorbents increases with increasing active surface area of adsorbents. Oak Sawdust modified by HCl has been used to treat heavy metal ions Cu, Ni, and Cr from aqueous solutions by Argun *et al*., (2007).This study revealed that adsorption kinetics follows pseudo-second-order reaction, Langmuir as well as D-R adsorption isotherms. Results indicated that the adsorption process was endothermic and spontaneous in nature under natural conditions. The maximum removal competency found to be 93% for Cu (II) at pH 4, 84% for Cr(VI) at pH3 and 82% for Ni(II) at pH 8.Chemically modified biomass of *Rhizopus nigricans* has been investigated for the Biosorption of Cr(VI) by Bai and Abraham (2002), they found that treatment with formaldehyde (10%, w/v) and mild alkalis (0.01 N NaOH and ammonia solution) decrease the biosorption efficiency. However, (0.1 N HCl and H_2SO_4), acetone (50%, v/v) and alcohols (50% v/v, CH_3OH and C_2H_5OH) enhanced the biosorbent uptake capacity for Cr(VI). Adsorption of divalent heavy metal ions specially Cu(II), Zn(II), Co(II), Ni(II) and Pb(II) on acid and alkali treated Orange and Banana peels was accomplished by Annadurai *et al*., (2003). Tomato factory waste treated by HCl for the treatment of copper (II) ion from aqueous solutions was investigated to identify metal ion removal efficiency of Tomato biosorbent. On the basis of pH factor, the maximum % removal was achieved at pH 8 as 92.08% (Yargıc *et al*., 2015). Chemically treated and untreated Olive stone was used to treat Lead ion and later reused as a fuel in Pyrolysis process (Ronda *et al*., 2015).

Study based on use of these biosorbents revealed that low cost adsorbent can be fruitfully used for the treatment of heavy metals. The aim of this study is to contribute low cost adsorbents in the search and to ascertain possibilities of utilization of various agricultural waste by-products and to compare removal tendencies of various heavy metals such as Ni(II), Zn(II), Pb(II) and Cd(II) using different by-products mainly *Tea* and *Coffee* wastes.

Nickel (Ni)

Nickel is the chemically active hard and ductile transition metal with atomic number 28. Ni is corrosion resistant that prevents the surface from further corrosion by forming a protective metal oxide layer. ^{58}Ni, ^{60}Ni, ^{61}Ni, ^{62}Ni, and ^{64}Ni, are the naturally occurring isotopes of Ni with ^{58}Ni being the most abundant with natural occurrence 68.077%. Nickel can form various oxidation states from -1 to +4; however (II) is the most common oxidation state of Ni (Denkhaus & Salnikow, 2002). Ni can be extracted by both *Pyro* and *Hydro* Metallurgy, however "*Mond process*" is predominantly used for Ni extraction from its ores because Mond process being the most efficient and ensure purity of Ni as high as 99.99 %. Ni is used in many industrial and consumer applications, including coinage, stainless steel, rechargeable batteries, electric guitar string, and microphones capsules and specially for making alloys with Copper, Aluminum, Chromium, Lead, Cobalt, Gold and Silver. *Nickel* plays a biological role in some plants like Archaebacteria, Fungi and Eubacteria. Urease, an enzyme of Ni is virulence that is severity factor in some organisms that helps to catalyze the hydrolysis of urea to yield Ammonia and Carbamate. Ni uptake can affect human health by Nickel-dependent bacteria. Food and water are the natural source of Nickel exposure. Mining, smelting, Nickel plated utensils, batteries manufacturing, electroplating, volcanic eruption, spark plugs, fossil fuel consumption, jewelry, tobacco, shampoo, coins, Nickel-Cadmium batteries and detergents are the common sources of Nickel absorption by human being (Peng *et al*., 2004; Kandah & Meunier, 2007; Malkoc,2006). Cigarettes also contain Nickel as a Nickel carbonyl (Gautum *et al*., 2014). Excess intake of Ni may be very toxic, even carcinogenic and results into lung and nasal cancer (Peng *et al*., 2004; Kandah & Meunier, 2007; Malkoc, 2006; Kyzas & Kostoglou, 2015). Based on the World Health Organization (WHO) guidelines, the maximum allowable concentration of Ni in industrial wastewater is 2 mg/l equivalent to 2ppm or 2000ppb while that in drinking water the permissible limit should be less than 0.1 mg/l equivalent to 0.1ppm or 100ppb.

Health effects

- Cardiovascular, contact dermatitis, allergic sensitization, lung fibrosis and kidney diseases and respiratory tract cancer are the cause of excess intake of Nickel (Oller *et al*., 1997; Mcgregor *et al*., 2000; Seilkop & Oller, 2003).

- Nickel toxicity may cause lung and nasal cancer.
- Short-term exposure can be responsible for miscellaneous clinical symptoms (nausea, headache, vomiting, diarrhea, giddiness, visual disturbance, cough, and abdominal discomfort).

Cadmium (Cd)

Cadmium is a very noxious metal. It can be found in soils, coal and mineral fertilizers. It is used extensively in electroplating, including batteries, metal coatings, pigments and plastics. The main source of Cadmium (Cd) in surface and ground waters are welding, nuclear fission plants, paints and plastics, electroplating, Cd and Ni batteries, fertilizers etc. (Moreno-castilla *et al*., 2004; Horiguchi *et al*., 1994; Kadirvelu & Namasivayam, 2003). "*Itai- itai*" disease (A painful, degenerative bone disease) first occurred in *1912 in Japan* due to chronic Cadmium toxicity and the source of Cadmium was discovered to be from mitsui mining and smelting company (Horiguchi *et al*., 1994). An excess exposure of Cd also causes many acute problems such as high blood pressure, kidney damage, osteoporosis and destruction of testicular tissue (Moreno-Castilla *et al*., 2004; Horiguchi *et al*., 1994; Kadirvelu & Namasivayam, 2003; Kobya *et al*., 2005; Nawrot *et al*., 2010; Ghorbani & Eisazadeh, 2013; Jarup *et al*., 2000; Karthik & Meenakshi, 2015). Zinc is an essential element for the effective functioning of *enzymes* however there will be change in the stereo-structure of the enzyme if it get replaced by Cd resulting in impairing of catalytic activity (Jarup *et al*., 2000).

Health effects

Cadmium and Cadmium compounds have life threatening effects on human and is known as human carcinogens.

- Ingestion of high level of Cadmium causes damage to the lungs irritates the stomach, resulting to vomiting and diarrhea.
- Long-term exposures can build-up possible kidney disease, lung damage, and fragile bones.
- Itai-itai disease caused by intake of excessive amount of Cadmium.

Lead (Pb)

Lead is malleable and soft unreactive post transition metal having low melting point. It is amphoteric in nature. It occurs mostly in (II) oxidation state rather it can form +4 oxidation states with lighter member of the carbon group. Galena (PbS) is the chief ore of Lead, mostly found with Zinc ores. High density, high abundance, low cost, ductility, low melting point and inertness towards oxidation make Lead a vital and useful in plumbing, construction, bullet and shot, weight, construction, white paints and radiation shielding. ^{204}Pb, ^{206}Pb, ^{207}Pb and ^{208}Pb are the most stable naturally occurring isotopes of Lead with ^{208}Pb is the heaviest stable isotope and all undergoes α decay to isotope of Hg with the emission of energy. Inorganic lead (Pb) contaminates water system. This is there as a consequences of human activities, such as mining, manufacturing and burning of fuel causing leaded gasoline (Bhowmick *et al*., 2014; Jouad *et al*., 2005), Lead and Lead compounds to get spread out in all parts of our environment. Lead can enter the body by ingestion, inhaling or skin absorption. Ingestion of food and water contaminated with Pb is the cause of lead poisoning in body. Acute Pb poisoning is dangerously harmful to kidneys, liver and also adversely affects the reproductive system and the toxic prodromes are anemia, irritability, insomnia headache, dizziness, hallucination, weakness of muscles and renal damages (Bhowmick *et al*., 2014; Jouad *et al*., 2005; Ding & Bao, 2014; Awual & Hasan, 2014; Naseem & Tahir, 2001; Nevin, 2009). Hence, Pb has been categorized as a priority pollutant by the United States Environmental Protection Agency (US EPA). The maximum contaminant level (MCL) of Pb ions has been set as 0.015 mg/l equivalent to 0.015ppm or 15ppb in drinking water whereas WHO limits it to 0.05 mg/l equivalent to 0.05ppm or 50ppb (Griffiths *et al*., 2012).

Health effects

US-EPA has determined that Lead is a human mutagen, an agent causing change in the structure of gene. Lead poisoning can disturb the function of body and affects the organs as it can damage the nervous system, causing an increase in blood pressure, weakness in fingers and wrists.

- High lead dosage can severely scathe the brain and kidneys.
- Lead poisoning is also known to cause mental abnormality, particularly in children (Nevin, 2009; Ding & Bao, 2014).
- Lead poisoning is also responsible for miscarriage in pregnant women.
- A high Lead level can damage the organs responsible for sperm production in man.

Zinc (Zn)

Some metal ions are highly essential for proper functioning of human organs such as Zinc (Zn), Copper (Cu), Manganese (Mn), Magnesium (Mg) and cobalt (Co), (Zhang *et al*., 2014; Kozlowski *et al*., 2009), However the excess intake of these ions causes serious health issues to living organisms as they are highly toxic, carcinogenic and get bioaccumulated in food chain (Zhang *et al*., 2014; Kozlowski *et al*., 2009; Sebastian & Srinivas, 2015; Omraei *et al*., 2011). On surface and in ground water Zinc is one of the most common pollutants (Omraei *et al*., 2011). Liquid and solid contaminated with Zn are referred as hazardous wastes because of its non-biodegradability (A substance which can't be changed to a harmless end product or state by dint of bacteria and may therefore ruin the environment) and acute toxicity. According to WHO the satisfactory concentration of Zn ions in drinking water should be 5.0 mg/l, equivalent to 5ppm or 5000ppb.

Health effects

Zinc toxicity can be caused due to excess intake of Zinc; however, its small doses are essential for a healthy body. It can be seen occurring at ingestion of greater than 225 mg of Zinc (Fosmire, 1990; Rout & Das, 2009).

- Presence of excess Zinc can lower the Copper and Iron absorption resulting in genetic disorder on account of Wilson Disease (WD) or Menkes Disease (MD) and anemia respectively.

- The solution containing Zinc ion is highly toxic to invertebrates (any animal missing of backbone), bacteria, plants, and even vertebrate fish (aquatic life having a backbone) (Smith & Lakson, 1946; Muyssen *et al*., 2006)
- Excess or prolong Zn exposure can lead to severe health problems, such as skin irritations, stomach cramps, nausea, vomiting, and anemia (Zhang *et al*., 2014; Kozlowski *et al*., 2009; Sebastian & Srinivas, 2015; Omraei *et al*., 2011).

Removal of Nickel

By using different by-products&
By using chemically modified by-products

Profusion use of Nickel and its compounds in industrial and commercial applications raising its demand. The industrialization advancement has led to emission of pollutants into ecosystems. Nickel can be released into environment due to manmade and natural activities like fossil fuel consumption, disposal of Nickel containing alloys, volcanic eruption and weathering of soil and rocks etc.

Many low cost biosorbent have been used such as Agriculture waste of Black gram husk (Saeed *et al*., 2005); Petiolar felt sheath palm (Iqbal *et al*., 2002); Maple saw (Shukla *et al*., 2005); Rice bran, Soybean & Cottonseed hulls (Marshall & Johns, 1996); Baker's yeast (Padmavathy *et al*., 2003); Mustard oil cake (Ajmal *et al*., 2005); Orange peel (Ajmal *et al*., 2000); Activated carbon from coirpith (Kadirvelu *et al*., 2001); Rambai stem (Khuzaimah *et al*., 2011); Coconut leaves (Gowda *et al*., 2012); Peanut hulls (Periasamy & Namasivayam, 1995); Moringa oleifera seeds (Marques *et al*., 2012); Casia fistula biomass (Hanif *et al*., 2007); Tamarind fruit shell (Pandharipande & Kalnake, 2013); Powder babbul bark (Patil *et al*., 2006); Delonix regia bark (Gulmohar) (Patil & Shrivastava, 2010); Leaf, Bark and Seed of Moringa Stenopetala (Aregawi & Mengistie, 2013); Orange peel (Gonen & Serin, 2012); Teak tree bark powder (Patil *et al*., 2012); Rice straw (Brahmaiah *et al*., 2015); Banana and orange peels (Annadurai *et al*., 2003); Almond husk activated carbon (Hasar, 2003); Mango peel (Iqbal *et al*., 2009) and Ficus Religiosa (peepal) leaves (Zaheer *et al*., 2010) etc. for the remediation of various heavy metal ions such as Cr(VI), Ni(II), Pb(II), Cd(II), Cu(II), Zn(II) and Cr (III) etc. (see table 1).

Table 1 List of agricultural waste biomass for the removal/remediation of Ni(II) and other heavy metal ions

Adsorbents	Metal ion	% removal or uptake (mg/g)	References
Black Gram Husk	Cd(II), Cu(II), Ni(II) and Zn(II)	>93%	Saeed *et al*., 2005
PFP (Petiolar Felt Sheath Palm)	Pb(II), Cd(II), Cu(II), Zn(II), Ni(II) and Cr(III)	Pb(II) > Cd(II) > Cu(II) > Zn(II) > Ni(II) > Cr(III)	Iqbal *et al*., 2002
Casia Fistula Biomass	Ni(II)	100%	Hanif *et al*., 2007
Maple Saw	Ni(II)	75%	Shukla *et al*., 2005
Baker's Yeast	NI(II)	11.4 mg/g	Padmavathy *et al*., 2003
Mustard Oil Cake	Ni(II), Zn(II), Pb(II), Mn(II), Cd(II), Cu(II) and Cr(II)	Upto 94% for Cu(II)	Ajmal *et al*., 2005
Orange Peel	Zn, Ni, Cu, Pb and Cr	Good results	Ajmal *et al*., 2000
Activated Carbon From Coirpith	Ni(II)	62.5 mg/g	Kadirvelu *et al*., 2001
Rambai Stem (Baccaureamotleyana)	Ni(II)	51%	Khuzaimah *et al*., 2011
Coconut Leaves	Ni(II)	93.18%	Gowda *et al*., 2012
Peanut Hulls	Ni(II)	53.65 mg/g	Periasamy & Namasivayam, 1995
Moringa OleiferaSeeds	Ni(II)	>90%	Marques *et al*., 2012
Tamarind Fruit Shell	Ni(II) and Cr(VI)	50-85.36% For Ni(II)& 22-90.35% for Cr(VI)	Pandharipande & Kalnake, 2013
Powder Babbul Bark (PBB)	Ni(II)	High adsorption	Patil *et al*., 2006
Delonix Regia Bark (Gulmohar)	Ni(II)	High adsorption	Patil & Shrivastava, 2010
Leaf, Bark And Seed Of Moringa Stenopetala	Ni(II)	93.90%, 96.25%, 97.50% for leaf, bark & seed respectively	Aregawi & Mengistie, 2013
Orange Peel	Ni(II)	High adsorption	Gonen & Serin, 2012
Teak Tree Bark Powder (Tectona Grandis)	Ni(II)	4.975 to 10.101 mg/g	Patil *et al*., 2012
Rice Straw	Ni(II)	Good results	Brahmaiah *et al*., 2015
Banana and Orange Peels	Ni(II)	Banana peel-6.88 mg/g Orange peel-6.01 mg/g	Annadurai *et al*., 2003
Almond Husk Activated Carbon	Ni(II)	97.8%	Hasar, 2003
Mango Peel	Cu(II), Ni(II) and Zn(II)	Cu(II)> Ni(II)> Zn(II)	Iqbal *et al*., 2009
Pine Dust	Ni(II)	High adsorption	Moodley *et al*., 2011
Rice Husk	Fe(II), Pb(II) and Ni(II)	High adsorption	Hegazi, 2013
Rice Husk	Ni(II)	High adsorption	Manjeet *et al*., 2009
Maize Cob	Ni(II)	High adsorption	Muthusamy *et al*., 2012
Chlorella Sorokiniana	Ni(II)	98%	Akhtar *et al*., 2004
Ficus Religiosa (peepal) Leaves	Ni(II)	High adsorption	Zaheer *et al*., 2010
Orange Peel	Ni(II)	158 mg/g	Kurniawan *et al*., 2006
Chlorella Vulgaris	Ni(II)	58.4 mg/g	Aksu & Donmez, 2006
Sphagnum Moss Peat	Ni(II)	Good results	Ho *et al*., 1995
Rice Bran, Cottonseed Hulls &Soybean	Cu(II), Ni(II) and Zn(II)	High adsorption	Marshall & Johns, 1996

An experiment was conducted on Cassia fistula biomass to treat Nickel and 99-100% removal efficiency has been observed (Hanif *et al*., 2007). Wastetea leaves were also reported for the treatment of Nickel from aqueous solutions (Ahluwalia & Goyal, 2005). Saw dust of black locust, maple and oak, Agricultural waste materials as modified cotton seeds, coir fibers, soybeans, corncobs, pecan, peanut, walnut, hazelnut and groundnut shells have been also used for the Nickel uptake (Sciban *et al*., 2006; Shukla *et al*., 2005; Shukla & Pai, 2005; Marshall & Johns, 1996; Vaughan *et al*., 2001; Johns *et al*., 1998; Demirbas, 2003; Kurniawan *et al*., 2006). *Zygnema sp. biomass was used to treat Ni(II)* ions from electroplating industrial wastewater using different parameters such as pH, time and sorbent dosage. Optimum value of pH found to be 3. It has been observed that biosorption efficiency affected by the sorbent dosage and 76.4 % removal was obtained at a dosage of 7.5 g/L (Sivaprakash *et al*., 2015). In 2000, an experiment was carried out to compare the adsorption efficiency of Chlorella vulgari and Chlorella Minimata to treat Ni(II) ions. Results showed that Chlorella Miniata (more than 99%) has grater adsorption rate as compare to Chlorella vulgari (around 33-41%) for the treatment of Ni(II) ions (Wong *et al*., 2000). In 2011, powder of mosambi fruit peelings (PMFP) has been used as an low costadsorbent for the remediation of Ni(II) ion from aqueous solutions (Krishna & Swmay, 2011). Adsorption process was study at different parameters like pH, agitation time, particle size, initial metal ion concentration, and adsorbent dosages. They found that PMEP has great metal recovery capacity and Adsorption process follows Langmuir and Freundlich isotherm models. Maize cob has been used to study Ni(II) removal by using atomic absorption spectroscopy (AAS) for metal assessment (Muthusamy *et al*., 2012). Adsorption process follows Langmuir and Freundlich isotherm models and they also found maize cob as low cost waste material, has significant metal recovery potential. 100% Ni(II) removal was observed by using low cost fly ash. It has been reported that increase in temperature decreases the adsorption of Ni(II). At pH of 9.4 to 10 highest metal removal was obtained (Popuri *et al*., 2015).

In order to enhance *chelating efficiency*, agricultural and plant wastes are pretreated with some *modifying agents* which can be minerals, organic solutions, tartaric acid, thioglycollic, citric acid or base solutions (calcium hydroxide, sodium hydroxide, sodium carbonate, oxidizing agent, organic compounds (ethylenediamine, epichlorohydrin, methanol, formaldehyde) and dye (reactive orange 13) etc. These have been widely used to treat heavy metals ions (Hanafiah *et al*., 2006). Shukla and Pai (2005) conducted an experiment using modified lignocelluloses fiber/jute for the treatment of heavy metal ions like Cu(II), Zn(II) and Ni(II) from aqueous solution. The dye containing jute fiber showed adsorption capacity of 8.4, 5.95 and 5.26 mg/g for Cu(II), Zn(II) and Ni(II) respectively. The equilibrium data followed Langmuir model. Peat and Cellulose adsorbents are successfully used to remove Nickel from aqueous solution at different pH values (Babel & Kurniawan, 2003). Equilibrium data fits well in Langmuir isotherm model. Coir pith was used by various researchers to treat Ni ion, it is an agriculture solid waste. Lignin (36%) and cellulose (44%) are the chief components of coir pith that plays a key role for the heavy metal removal (Kadirvelu*et al*., 2001; Redded *et al*., 2002; Shukla & Pai, 2005). Modified coir pith (treated with NaOH) has shown good metal recovery by increasing metal binding sites (Shukla & Pai, 2005; Li *et al*., 2007). Various chemically treated agriculture wastes have been used for heavy metals biosorption or remediation (see table 2).

Table 2 List of Chemically modified agricultural wastes for the treatment/remediation of Ni (II) heavy metal ions from aqueous solution

Adsorbents	Modifying agent	% removal or uptake (mg/g)	References
Walnut Shell	NaOH	8.57 mg/g	Karimi-Jashini & Saadat, 2014
Peat Coil, Cow Dung And Digested Paddy Husk	Ethanol	86.23%	Zhang & Ismail, 2012
Coconut Husk (CH) And Teak Tree Bark (TTB)	HCI	CH- 24.1% and TTB-29.9%	Kehinde *et al*., 2009
Babul Bark	NaOH	80%	Patil *et al*., 2006
Jute Fiber	H_2O_2	5.57 mg/g	Shukla & Pai, 2005
Activated Carbon From Almond Husk	H_2SO_4	97.8%	Hasar, 2003
Coconut Coir Pith	H_2SO_4	High adsorption	Ratan *et al*., 2016
EggShell Waste	Vinegar	78.70%	Stevens & Batlokwa, 2017
Coir Fiber	Hydrogen peroxide	Good results	Shukla *et al*., 2005
Saw Dust and Dye Loaded Groundnut Shells	Cu(II), Ni(II), Zn(II)	> 90%	Shukla & Pai, 2005
Rice Straw	Acid and alkali	Good results	Brahmaiah *et al*., 2015
Rice Bran, Soybean & Cottonseed Hulls	HCl and NaOH	High adsorption as compare to untreated waste	Marshall & Johns, 1996
Peanut	Formaldehyde	Good results	Randall *et al*., 1978
Sawdust	Sodium hydroxide	10.47mg/g	Rehman *et al*., 2006
Walnut Sawdust	Formaldehyde in sulfuric acid	6.63mg/g	Bulut & Tez, 2003
Sawdust	Reactive Orange 13	9.87mg/g	Shukla & Pai, 2005
Groundnut Shells	Reactive Orange 13	7.49mg/g	Shukla & Pai, 2005
Coirpith	Sulfuric acid and ammonium persulphate	62.5mg/g	Kadirvelu *et al*., 2001
Red Onion Skin	Formaldehyde	7.55 mg/g	Kumar & Dara, 1981

Removal of Cadmium

Byusing different by-products&
By using chemically modified by-products

Cadmium and its compounds are highly noxious as compared to other heavy metal ions. These are water soluble and tend to accumulate in food chain. Cadmium can enter into our ecosystem because of excess use in electroplating, welding, Cd and Ni industries, paints and nuclear power plants etc. In the human body accumulation of Cadmium is predominantly there in kidneys resulting in their malfunctioning (Volesky & Holan, 1995). Wheat bran and rice bran was investigated for the treatment cadmium and significantly encouraging results were reported (Farajzadeh & Monji, 2004; Montanher *et al*., 2005; Singh *et al*., 2005). A natural low cost biosorbent, Pomelo peel, was used to treat Cd(II) ions from aqueous solution using various parameters such as pH, contact times and initial metal ion concentrations. Cadmium uptake directly depends upon pH ranging from 1 to 5 and at pH 5, maximum Cadmium removal has been observed. Adsorption equilibrium process was well described by the Langmuir adsorption isotherm and adsorption capacity found to be 21.83mg/g (Saikaew *et al*., 2009). low-cost adsorbents: grapes bunches, sunflower leaves, almond rinds, eucalyptus barks, bananas rinds and maize leaves have been used to check the ability to remove Cadmium from aqueous solutions. A sorption isotherm was used to evaluate Cadmium uptake on these biosorbents. Results indicated that the equilibrium process is well described by the Langmuir model than the Freundlich equation and kinetics follow pseudo second order rate model. Maximum removal was observed by sunflower leaves 153.85 mg/g followed by eucalyptus barks (99.30 mg/g), almond rinds (104.60 mg/g), grapes bunches (75.64 mg/g), maize leaves (57.84 mg/g), bananas rinds (69.35 mg/g) (Benaissa, 2006). In 1993, brown marine algae *Sargassum natans, Ascophyllum nodosum (*A. *nodosum*) and *Fucus vesiculosus* have been examined to remove Cd(II) ions from aqueous solutions. Results demonstrated that A. *nodosum* accumulated have the highest Cd(II) removal capacity at pH 3.5 (Holan *et al*., 1993). Saudi activated clay (bentonite) have been demonstrated for Cd(II) removal under various operating variables such as initial metal concentration, contact time, solution pH and clay dosage. Results indicated that bentonite has the significant Cd(II) uptake capacity and within 20 min equilibrium was achieved and this uptake significantly depend on pH (Al-Shahrani, 2012). Bark of the plants such as Abies sachalinensis, Pecia glehnii, dried plant biomass, Wheat bran, Husk of black gram etc. have been tried for the removal of Cadmium (see Table 3). Natural and chemically modified rice husk, rice polish, ground nut husk, red onion skin and black gram husk have been used for the removal of Cadmium (Tarley & Arruda, 2004; Saeed *et al*., 2005; Singh *et al*., 2005; Kumar & Bandyopadhyay, 2006; Ajmal *et al*., 2006; Seki *et al*., 1997; Karnitz *et al*., 2007; Okieimen *et al*., 1991; Kumar & Dara, 1981) (see table 3).

Table 3 List of agricultural wastes for the treatment/remediation of Cd(II) and other heavy metal ions from aqueous solution

Adsorbents	Metal ion	% removal or uptake (mg/g)	References
Coniferous Barks	Cd(II)	High adsorption	Seki *et al*., 1997
Rice Polish	Cd(II)	9.72 mg /g	Singh *et al*., 2005
Green Coconut Shell Powder	Cd(II)	High adsorption	Pino *et al*., 2006
Sugar-Cane Bagasse Pith	Cd(II)	24.70 mg/g	Krishnan & Anirudhan, 2003
Sugarcane Bagasse, Rice Straw, Soybean Hulls, Peanut Shells, Walnut Shells And Pecan	Cd(II), Cu(II), Ni(II), Pb(II) and Zn(II)	Pb(II)> Cu(II)> Cd(II)> Zn(II)> Ni(II)	Johns *et al*., 1998
Black Gram Husk	Cd(II)	High adsorption	Saeed & Iqbal, 2003
Parthenium Weed	Cd(II)	99.7%	Ajmal *et al*., 2006
Bagasse Fly Ash	Cd(II) and Ni(II)	High adsorption	Srivastava *et al*., 2006
Bagasse	Cd(II) and Zn(II)	Cd(II)>Zn(II)	Mohan & Singh, 2002
Bagasse Fly Ash (Waste Of SugarIndustry)	Ni(II) and Cd(II)	90.0%	Gupta *et al*., 2003
Rice Bran	Cd(II), Cu(II), Pb(II) and Zn(II)	>80.0%	Montanher *et al*., 2005
Wheat Bran	Cd(II), Hg(II), Pb(II), Ni(II), Cr(III) and Cu(II)	Cr(III)>Hg(II)>Pb(II)>Cd(II)> Cu(II)>Ni(II)	Farajzadeh & Monji, 2004
Mango Peel	Cd(II) and Pb(II)	68.92 and 99.05mg/g	Iqbal *et al*., 2009
Saudi Activated Bentonite	Cd(II)	14.9 mg/g	Al-Shahrani, 2012
Sun Flower Head Carbon (SHC)	Cd(II)	1.22 mg/g	Jain *et al*., 2015
Sun Flower Stem Carbon (SSC)	Cd(II)	1.48 mg/g	Jain *et al*., 2015
Golden Apple Snail	Cd(II)	81.301 mg/g	Zhao *et al*., 2016
Sago Waste	Pb(II) and Cu(II)	46.6 and 12.4 mg/g	Quek *et al*., 1998
Pine Bark	Cd(II), Pb(II), Ni(II) and Cu(II)	Pb(II) > Cd(II) > Cu(II) > Ni(II)	Al-Asheh & Duvnjak, 1997
EggShell	Cd(II)	100%	Park *et al*., 2007
Fluted Pumpkin Waste	Pb(II) and Cd(II)	49.53 and 65.50 mg/g	Horsfall & Ayebaemi, 2005
Fly Ash	Cd and Cu	96.034%	Hegazi, 2013
Chlorella Vulgaris	Cd(II)	86.6 mg/g	Aksu & Donmez, 2006
Pomelo Peel	Cd(II)	21.83mg/g	Saikaew *et al*., 2009
Grapes Bunches, Sunflower Leaves, Almond Rinds, Eucalyptus Barks, Bananas Rinds And Maize Leaves	Cd(II)	Sunflower> almond rinds> eucalyptus barks> grapes bunches> bananas rinds> maize leaves	Benaissa, 2006
Rice Husk	Cd(II)	69%	Kulkarni & Kaware, 2015
Saccharomyces Cerevisiae	Cd(II)	Good results	Volesky *et al*., 1993
Citrus Peels (Orange Peels, Lemon Peels And Lemon-Based Protonated Pectin Peels)	Cd(II)	90% at high sorbent dosage	Schiewer & Patil, 2008
Agave Bagasse	Cd(II)	13.27mg/g	Velazquez-Jimenez *et al*., 2013
Ananas Comosus Peel	Cd(II)	18.21mg/g	Foo *et al*., 2012
Activated Carbon Of Apricot Stone	Cd(II)	33.57 mg/g	Kobya *et al*., 2005
Psidium Guajava Peel	Cd(II)	39.68 mg/g	Foo *et al*., 2012
Parkia Speciosa Pods	Cd(II)	25.64 mg/g	Foo *et al*.,2012
Walnut Sawdust	Cd(II)	4.39 mg/g	Bulut & Zeki, 2007

Table 4 List of Chemically modified agricultural wastes for the treatment/remediationof Cd(II)ions from aqueous solution

Adsorbents	Modifying Agent	% removal or uptake (mg/g)	References
Sugarcane Bagasse	Succinic anhydride	Good results	Karnitz *et al*., 2007
Rice Husk	Sodium hydroxide	20.24 mg/g	Kumar & Bandyopadhyay, 2006
Base Treated Juniper Fiber	NaOH	29.54 mg/g	Min *et al*., 2004
Ground Nut Husk	EDTA	85%	Okieimen *et al*., 1991
Red Onion Skin	Formaldehyde	6.39 mg/g	Kumar & Dara, 1981
Peanut	Formaldehyde	Good results	Randall *et al*., 1978
Ground Nut Husk	EDTA	Good results	Okieimen *et al*., 1985
Rice Husk	Sodium bicarbonate	16.18 mg/g	Kumar & Bandyopadhyay, 2006
Rice Husk	Epichlorohydrin	11.12 mg/g	Kumar &Bandyopadhyay, 2006
Sawdust (Cedrus Deodar wood)	Sodium hydroxide	73.62%	Memon *et al*., 2007
Sawdust (Pinus Sylvestris)	Formaldehyde in Sulfuric acid	9.29 mg/g	Taty-Costodes *et al*., 2003
Walnut Sawdust	Formaldehyde in sulfuric acid	4.51 mg/g	Bulut & Tez, 2003
Cassava Tuber Bark	Thioglycollic acid	26.3 mg/g	Horsfall *et al*., 2006
Wheat Bran	Sulfuric acid	101 mg/g	Ozer & Pirincci, 2006
Spent Grain	Hydrochloric acid	17.3 mg/g	Low *et al*., 2000
Azolla Filiculoides (Aquatic Fern)	Hydrogen peroxide-Magnesium chloride	86 mg/g	Khosravi & Rakhshaee, 2005
Corncorb	Nitric acid	19.3 mg/g	Leyva-Ramos *et al*., 2005
Corncorb	Citric acid	55.2 mg/g	Leyva-Ramos *et al*., 2005
Agave Bagasse	NaOH	18.32 mg/g	Velazquez-Jimenez *et al*., 2013
Agave Bagasse	HCl	12.5 mg/g	Velazquez-Jimenez *et al*., 2013
Agave Bagasse	HNO_3	13.5 mg/g	Velazquez-Jimenez *et al*., 2013
Honeycomb	HNO_3	61.16 mg/g	Reddy *et al*., 2012

Removal of Lead

By using different by-products &

By using chemically modified by-products

Contamination of water by lead is of great concern as it gets tightly attached to particles of sediments, oil and waste sludge. Lead can enter into our body through food chain resulting into a variety of harmful biological effects depending upon the duration of exposure and concentration. High Pb removal efficiency has been observed by various scientist (see table 5) using different agriculture wastes in their natural form viz. soybean hulls, peanut shells, rice straw, walnut shells (Johns *et al*., 1998); tree bark (Bankar & Dara,1985); black gram husk, waste tea leaves, flowers of Humulus lupulus, water hyacinth and Petioler felt sheath palm (Gardea- Torresdey *et al*., 2002; Kamble & Patil, 2001; Iqbal *et al*., 2002, 2005; Saeed *et al*., 2005; Ahluwalia & Goyal, 2005). Activated carbons produced from agricultural wastes, such as bagasse, walnut and hazelnut shells and shells of apricot stones with great removal efficiency and large surface area have been used to treat heavy metals ions (Wilson *et al*., 2006; Ozdemir *et al*., 2011; Kadirvelu *et al*., 2001; Dolas *et al*., 2011; Saka, 2012; Gajghate *et al*., 1991; Vaughan *et al*., 2001). In 1998, 65% Lead removal competency has been reported using bagasse fly ash (Gupta *et al*., 1998). Maple Saw dust, Pinus sylvestries and rubber wood saw dust have shown 85-90% proficiency for Lead removal (Taty-Costodes *et al*., 2003; Raji *et al*., 1997). For biosorption of Lead optimizes value of pH is ranging from 5-6 according to literature review. In 2009 an experiment was investigated for Pb(II) removal on Orange peels by Schiewer and Balaria (2009). They compared the removal efficiency of Orange peel and protonated Orange peel and more than 90% removal was observed using these biosorbents (Schiewer & Balaria, 2009). In 2004 Coconut-shell was investigated to treat Lead from aqueous solutions. It has been found that adsorption was dependent on pH and at pH 4.5 maximum removal was obtained. Adsorption equilibrium data fitted well to the Freundlich, Tempkin isotherm and Langmuir isotherm models. At pH 4.5, 26.50 mg/g removal efficiency has been observed with the Langmuir model (Sekar *et al*., 2004). Modified form of Apple residue with modifying agent phosphorous (V) oxychloride (Lee *et al*., 1999); rose petals with modifying agent NaOH (Karnitz *et al*., 2007); sugarcane modified with succinic anhydride (Tsui *et al*., 2006) and calcium treated sargassum (Nasir *et al*., 2007) have been utilized as excellent adsorbent for Lead removal (see table 5).

Table 5 List of agricultural wastes for the treatment/remediation of Pb(II) and other heavy metal ions from aqueous solution

Adsorbents	Metal ion	% removal or uptake (mg/g)	References
Peanut Shells	Pb(II), Zn(II), Cu(II), Ni(II) and Cd(II)	>75%	Wilson *et al*., 2006
PFP (Petiolar Felt Sheath Palm)	Pb(II), Cr(III), Cd(II), Cu(II), Zn(II) and Ni(II)	Good results	Iqbal *et al*., 2002
Oriza Sativa Husk	Pb(II)	>98%	Zulkali *et al*., 2006
Humulus Lupulus	Pb(II)	74.2 mg/g	Gardea-Torresdey *et al*., 2002
Coir Pith	Hg(II), Pb(II), Cd(II), Ni(II), and Cu(II)	Hg-100%, Pb-100%, Cd- !00%, Ni-92% and Cu-73%	Kadirvelu *et al*., 2001
Apple Residue Waste	Pb(II), Cu(II) and Cd(II)	Pb(II) -95.3%, Cu(II)-91.2% and Cd(II)- 91%	Lee *et al*., 1999
Waste Of Black Gram Husk	Pb(II), Zn(II), Cd(II), Ni(II) and Cu(II)	Pb> Cd> Zn> Cu> Ni	Saeed *et al*., 2005
Soymida Febrifuga Bark	Pb(II)	100%	Bankar & Dara, 1985
Coconut Char	Pb(II)	100%	Gajghate *et al*., 1991
Rice Bran	Pb(II),Cd(II), Cu(II) and Zn(II),	>75%	Montanher *et al*., 2005
Saw Dust	Pb(II) and Cu(II)	3.19 and1.79 mg/g	Yu *et al*., 2001
Sugar-Beet Pectin Gels	Cu, Pb and Cd	High metal recovery	Mata *et al*., 2009
Rice Husk	Fe, Pb and Ni	94.885% to 96.954%	Hegazi, 2013
Sunflower Seed Peel	Pb(II)	99%	Ozdemir *et al*., 2004
Orange Peel	Pb(II)	>90%	Schiewer & Balaria, 2009
Coconut-Shell	Pb(II)	26.50 mg/g	Sekar *et al*., 2004
Marine Green Ulva Fasciata Sp.Carbon	calcium chloride-treated (CCUC)	22.93mg/g	Jeyakumar & Chandrasekaran, 2014
Marine Green Ulva Fasciata Sp. Carbon	sodium sulphate-treated *U.fasciata* carbon (SSUC)	24.15mg/g	Jeyakumar & Chandrasekaran, 2014
Marine Green Ulva Fasciata Sp. Carbon	sodium carbonate-treated *U. fasciata* carbon (SCUC))	23.47 mg/g	Jeyakumar &Chandrasekaran, 2014
Casuarina Glauca Tree Leaves	Pb(II) and Cr(III)	High adsorption	Abdel-Ghani *et al*., 2008
Nipah Palm Shoot Biomass	Pb(II)	15.59 mg/g	Wankasi *et al*., 2006
Agave Bagasse	Pb(II)	7.84 mg/g	Velazquez-Jimenez *et al*., 2013
Activated Carbon Of Apricot Stone	Pb(II)	22.85 mg/g	Velazquez-Jimenez *et al*., 2013
Activated Carbon Of Coconut Buttons	Pb(II)	94.35 mg/g	Anirudhan & Sreekumari, 2011
Passion Fruit Skin	Pb(II)	85.68 mg/g	Gerola *et al*., 2013
Sugarcane Bagasse	Pb(II)	11.63 mg/g	Dos *et al*., 2010
Walnut Sawdust	Pb(II)	6.54 mg/g	Bulut & Zeki, 2007

Table 6 List of Chemically modified agricultural wastes for the treatment/remediation of Pb(II) heavy metal ions from aqueous solution

Adsorbents	Modifying Agent	% removal or uptake (mg/g)	References
Apple Residue	Acidic methanol	Good results	Lee *et al*., 1999
Rose Biomass	NaOH	87.74 mg/g	Nasir *et al*., 2007
Ground Nut Husk	EDTA	>98%	Okieimen *et al*., 1991
Red Onion Skin	Formaldehyde	10.9 mg/g	Kumar & Dara, 1981
Peanut	Formaldehyde	Good results	Randall *et al*., 1978
Ground Nut Husk	EDTA	Good results	Okieimen *et al*., 1985
Rice Husk	Tartaric acid	120.48 mg/g	Wong *et al*., 2003
Sawdust (Pinus Sylvestris)	Formaldehyde in Sulfuric acid	9.78 mg/g	Taty-Costodes *et al*., 2003
Walnut Sawdust	Formaldehyde in sulfuric acid	4.48 mg/g	Bulut & Tez, 2003
Peanut Husk	Sulfuric acid	29.14 mg/g	Li *et al*., 2006
Banana Stem	Formaldehyde	>99%	Noeline *et al*., 2005
Spent Grain	Hydrochloric acid	35.5 mg/g	Low *et al*., 2000
Imperata Cylindrica Leaf Powder	Sodium hydroxide	13.50 mg/g	Hanafiah *et al*., 2006
Azolla Filiculoides (Aquatic Fern)	Hydrogen peroxide-Magnesium chloride	228 mg/g	Khosravi & Rakhshaee,2005
Alfalfa Biomass	Sodium hydroxide	89.2 mg/g	Tiemann *et al*., 2002
Bagasse Fly Ash	Hydrogen peroxide	96-98%	Gupta & Ali, 2004
Nipah Palm Shoot Biomass	Mercaptoacetic acid	21.85 mg/g	Wankasi *et al*., 2006
Agave Bagasse	NaOH	20.54 mg/g	Velazquez-Jimenez *et al*., 2013
Agave Bagasse	HCl	12.40 mg/g	Velazquez-Jimenez *et al*.,2013
Agave Bagasse	HNO_3	14.43 mg/g	Velazquez-Jimenez *et al*., 2013
Honeycomb	HNO_3	74.17 mg/g	Reddy *et al*., 2012
Passion Fruit Skin	NaOH	112.99 mg/g	Gerola *et al*., 2013
Passion Fruit Skin	NaOH and citric acid	204.08 mg/g	Gerola *et al*., 2013
Red Mud	H_2O_2	88.20 mg/g	Gupta *et al*., 2001
Sugarcane Bagasse	NaOH	30.68 mg/g	Dos *et al*., 2010
Sugarcane Bagasse	citric acid	52.63 mg/g	Dos *et al*., 2010
Sugarcane Bagasse	NaOH and citric acid	33.09 mg/g	Dos *et al*., 2010

Removal of ZINC

By using different by-products &

By using chemically modified by-products

Zinc is one of the most precious element or *nutrient* for biological function of body. It is believed that it contain anti-oxidant properties which protects our skin from aging effects. It is used as a catalyst during rubber manufacturing. As a pigment, Zinc is used in cosmetics, plastics, wallpaper, photocopier paper, printing inks etc. Zinc is the 23rd most abundant element on the Earth's crust but due to industrialization its concentrations are rising unnaturally in ecosystem particularly in drinking water resulting in severe health problems such as *Teratogenesis* (Process by which congenital malformation are produced in an embryos or fetus),

Table 7 List of agricultural wastes for the treatment/remediation of Zn(II) and other heavy metal ions from aqueous solution

Adsorbents	Metal ion	% removal or uptake (mg/g)	References
Coconut (Cocos Nucifera) Coir Dust	Zn(II)	High adsorption	Israel & Eduok, 2012
Azadirachta Indica Bark	Zn(II)	33.49 mg/g	King *et al*., 2008
Lemon Peel	Zn(II)	87.5%	Rajoriya & kaur, 2014
Banana Peel	Zn(II)	90.5%	Rajoriya & kaur, 2014
Bagasse	Cd(II) and Zn(II)	High adsorption	Mohan & kumar, 2002
Cork Powder	Zn(II), Cu(II) and Ni(II)	High adsorption	Chubar *et al*., 2003
Tectona Grandis Leaves	Zn(II)	High adsorption	Kumar *et al*., 2006
Rice Husk Ash (RHA),	Zn(II), Cd(II)	High adsorption	Srivastava *et al*., 2008
Mature Leaves Of The Neem Tree	Zn(II)	147.08 mg /g	Arshad *et al*., 2008
Stem Bark Of The Neem Tree	Zn(II)	137.67 mg/g	Arshad *et al*., 2008
Orange Wastes	Zn(II), Cd(II) and Pb(II)	Pb(II) >Zn(II) > Cd(II)	Perez Marin *et al*., 2008
Olive Oil Mill Solid Residues	Zn(II)	>95%	Hawari *et al*., 2009
Physic Seed Hull (PSH)	Cd(II) and Zn(II)	47% and 36%	Mohammad *et al*., 2010
Activated Sludge	Zn(II)	Good results	Mishra & Chaudhury, 1996
Waste-Reclaimed Adsorbent	Cu(II) andZn(II)	1.68 mg/g and 0.06 mg/g	Jo *et al*., 2010
Cork Powder	Zn(II)	98% in synthetic wastewater and electroplating industrial wastewater was 91%.	Kanawade & Gaikwad, 2011
Alga *Chlorella Vulgaris*	Zn(II) and Cd(II)	Good results	Ting *et al*., 1989
Olive Oil Mill Residues	Zn(II)	Good results	Hawari *et al*., 2009
Sugar Beet Pulp	Pb(II), Cu(II), Zn(II), Cd(II), and Ni(II)	Pb(II)> Cu(II) > Zn(II) > Cd(II)> Ni(II)	Reddad *et al*., 2002
Bagasse-Based Activated Carbon	Zn(II) and Cd(II)	Good results	Mohan & Singh, 2002
Date Pits-Based Activated Carbon	Pb(II), Zn(II), Co(II) and Fe(III)	100%	Awwad *et al*., 2013
Microalgae	Zn(II) and Cu(II)	Good results	Chan *et al*., 2013
Industrial Waste Sludge (Steel Plant Waste Sludge)	Zn(II)	7.26 mg/g	Mishra *et al*., 2013
Aquatic Moss Fontinalis Antipyretica	Zn(II) and Cd(II)	28.0 and14.7 mg/g	Martins *et al*., 2004
Bentonite	Zn(II)	Good results	Bellir *et al*., 2013
Botrytis Cinerea	Zn(II)	Good results	Tunali & Akar, 2006
Black Gram Husk	Zn(II), Cd(II), Cu(II), Ni(II)and Pb(II)	Pb(II) > Cd(II) > Zn(II) > Cu(II)> Ni(II)	Saeed *et al*., 2005
Carrot Residues	Cr(III) ,Cu(II) and Zn(II)	Cr(III)>Cu(II)>Zn(II)	Nasernejad *et al*., 2005
Cassava Waste	Cd, Cu and Zn	Good results	Abia *et al*., 2003
Cassava Waste	Zn(II) and Cd(II)	55.82, 86.68 mg/g	Horsfall & Abia, 2003
Dried Marine Green Macroalga Caulerpa Lentillifera	Zn(II), Cu(II) , Pb(II) and Cd(II)	Pb(II)> Cu(II)> Cd(II) > Zn(II)	Pavasant *et al*., 2006
Dried Marine Green Macroalga(*Chaetomorpha linum*))	Cu(II)and Zn(II)	Good results	Ajjabi & Chouba, 2009
Rice Bran	Zn(II), Cd(II), Pb(II) and Cu(II)	Good results	Montanher *et al*., 2005
Oflignite-Based Fly Ash	Zn(II) and Cd(II)	Good results	Bayat, 2002
Walnut Shell	Cu(II), Zn(II) and Cd(II)	14.53, 7.47 and 7.29 mg/g	Najam *et al*., 2016
Immobilized Stem-Bark (IMSB)	Zn(II)	91.60%	Osemeahon *et al*., 2015
Solid Residue Of Olive Mill Products	Zn(II)	5.40 mg/g	Gharaibeh *et al*., 1998
Sun Flower Stalks	Zn(II)	30.73 mg/g	Sun & Shi, 1998
Red Mud	Zn(II)	12.59 mg/g	Lopez *et al*., 1998
Apricot Stones Carbon	Zn(II)	13.21mg/g	Budinova *et al*., 1994
Coconut Shells Carbon	Zn(II)	12.76 mg/g	Budinova *et al*., 1994
Lignite Coal Carbon	Zn(II)	11.11 mg/g	Budinova *et al*., 1994
Almond Shell (Carbon)	Zn(II)	6.65 mg/g	Ferro-Garcia *et al*., 1988
Olive Stone (Carbon)	Zn(II)	5.10 mg/g	Ferro-Garcia *et al*., 1988
Peach Stone Carbon	Zn(II)	5.0 mg/g	Ferro-Garcia *et al*., 1988
Bagasse Carbon	Zn(II)	31.11 mg/g	Ferro-Garcia *et al*., 1988
Clarified Sludge	Zn(II)	15.53 mg/g	Bhattacharya *et al*., 2006
Rice Husk Ash	Zn(II)	14.30 mg/g	Bhattacharya *et al*., 2006
Activated Alumina	Zn(II)	13.69 mg/g	Bhattacharya *et al*., 2006
Neem Bark	Zn(II)	13.29 mg/g	Bhattacharya *et al*., 2006
Sugar Beet Pulp	Zn(II)	35.6 mg/g	Pehlivan *et al*., 2006
Fly ash	Zn(II)	7.84 mg/g	Pehlivan *et al*., 2006
Agave Bagasse	Zn(II)	35.6 mg/g	Velazquez-Jimenez *et al*., 2013
Carrot Residues	Zn(II)	29.61 mg/g	Nasernejad *et al*., 2005

carcinogenesis and *Mutagenesis* (Process causing changes in the gene structure). To overcome these affects caused by intake of excess amount of Zinc it becomes urgent to remove this metal from waste water using different by-products. Various attempts have been carried out for the eradication of Zn ion from industrial waste water by using different by-product (see table 7).An experiment was conducted to study the sorption capacity of immobilized plant stem-bark (IMSB) to discharge Zn(II), Cd(II), Pb(II), Mn(II), Cr(II) andFe(II) ions using different parameters such as contact time, ionic strength, temperature, pH and initial metal ion concentration. The result showed that the sorption capacity of Zn(II), Cd(II), Pb(II), Mn(II), Cr(II) and Fe(II) by IMSB were 91.60%, 85.08%, 97.85%, 65.20%, 78.46%, and 78.52% respectively (Osemeahon *et al*., 2015). The biomass of *Azadirachta indica*bark has been used to remove Zn(II) ions from aqueous solutions using different parameters such as contact time, initial metal ion concentration, pH, biosorbent dosage and average biosorbent size. At pH 6 maximum Zinc biosorption occurred and it has been noted that percentage biosorption increases with increase in the biosorbent dosage. Data obtained from the experiment were tested with the adsorption models like Freundlich, Langmuir and Redlich-Peterson isotherms. Langmuir isotherm best fitted on the experimental data with maximum biosorption capacity of 33.49 mg/g of Zinc ions on *A. indica* bark biomass (King *et al*.,2008). Aloe Vera leaves powder (AV), multi walled Carbon nanotube (MWCNTs) and activated Aloe Vera powder (AAV) as an adsorbents have been used for the treatment of Zn(II) from aqueous solutions using various parameters such as adsorbent dosage, pH and contact time.

Table 8 List of Chemically modified agricultural wastes for the treatment/remediation of Zn(II) heavy metal ions from aqueous solution

Adsorbents	Metal ion	% removal or uptake (mg/g)	References
Rose (Rosa Centifolia) Petals	NaOH	73.8 mg/g	Nasir *et al*., 2007
Oil Palm Frond (OPF)	NaOH	90%	Salamatinia *et al*., 2010
Natural And Activated Bentonite	H_2SO_4	4.54mg/g	Pradas *et al*., 1994
Cassava Waste	Thioglycollic acid	Good results	Abia *et al*., 2003
Cassava Waste	Thioglycollic acid	559.74 mg/g	Horsfall & Abia, 2003
Peanut	Formaldehyde	Good results	Randall *et al*., 1978
Hardwood Sawdust	Formaldehyde in acidic medium	Good results	Sciban *et al*., 2006
Hardwood Sawdust	Sodium hydroxide	Good results	Sciban *et al*., 2006
Sawdust	Sawdust Reactive Orange 13	17.09 mg/g	Shukla & Pai, 2005
Cassava Tuber Bark	Thioglycollic acid	83.3 mg/g	Horsfall *et al*., 2006
Azolla Filiculoides (Aquatic Fern)	Hydrogen peroxide-Magnesium chloride	48 mg/g	Khosravi & Rakhshaee, 2005
Carrot Residues	HCl	29.61mg/g	Nasernejad *et al*., 2005
Groundnut Shells	Reactive-Orange 13	9.57 mg/g	Shukla & Pai, 2005
Red Onion Skin	Formaldehyde in an acidic medium	9.42 mg/g	Kumar & Dara, 1981
Banana Peel	Acid- treated	2.75 mg/g	Annadurai *et al*., 2003
Banana Peel	Alkali- treated	2.25 mg/g	Annadurai *et al*., 2003
Orange Peel	Acid-treated	2.75 mg/g	Annadurai *et al*., 2003
Orange Peel	Alkali-treated	2.65 mg/g	Annadurai *et al*., 2003
Agave Bagasse	NaOH	50.12 mg/g	Velazquez-Jimenez *et al*., 2013
Agave Bagasse	HCl	42.31 mg/g	Velazquez-Jimenez *et al*., 2013
Agave Bagasse	HNO_3	54.29 mg/g	Velazquez-Jimenez *et al*., 2013

This study revealed that the removal efficiency of Zn(II) onto each of AV, MWCNTs and AAV increases with the increase of the adsorbent dosage, pH and contact time. the maximum adsorption efficiency were examined to be 58.54% for AV at (2.2 g, pH 5 and 6 hr),96.27% for MWCNTs at (0.06 g, pH 5 and 6 hr) and 80.83% for AAV at (1.6 g, pH 5 and 6 hr) (Moosa *et al*., 2016).

Removal/Remediation of Ni(II), Pb(II), Cd(II), Zn(II) Using Indian Tea And Coffee Wastes

Over the last few years, tea and instant coffee have taken a valuable place in most popular kinds of beverage drunk by millions of people all over the world as a result of which the tea and instant coffee industries are touching the zenith of success day by day. In India, yearly production of tea is approximately 857000 tons (Wasewar, 2010) and the production of coffee is 6 million tons per year. During this generation and consumption plenty of tea wastes (From the Restaurant and Cafeteria or tea - processing industries) and coffee wastes are usually recycled into the environment without any remediation or treatment, causing environmental⁄hygiene problems and contaminate our ecosystem by emitting organic matter. To resolve these problems, strategies are being scrutinized to appraise their better usage as energy source. On account of its low cost and abundant availability, tea and coffee wastes have been utilized as convenient way of treatment of industrial wastewater. The composition of fresh tea contains various components, such as, tannin, caffeine, amino acids, polyphone (include catechins), lignin, flavonoids, other phenolic compounds are mainly aromatic carboxylate, carboxylate, fluorine, phenolic hydroxyl and oxyl groups, vitamins and polysaccharides. The cell wall of tea waste consists of carbohydrate, cellulose, lignin having hydroxyl groups in their structures (Aikpokpodion *et al*., 2010). Dry tea leaves contain 30-35% Polyphenols and behave as a metal scavenger from solution and wastewater. Coffee waste and coffee pulp contains Tannins, caffeine, Polyphenols, proteins, Carbohydrates, Minerals, Caffeine, Potassium, and some organic solid residue (Padmapriya *et al*., 2013).

An experiment was carried out in which Vigna radiata has been grown in soil contaminated with chromium using tea waste as an suitable adsorbent or as a fertilizer mixed with soil is used to protect plant from the phytotoxicity (It is toxic effect of a compound on plant growth) of Cr(III) (Azmat & Akhter, 2010). Another experiment was investigated to check the Competency and characterization of wasted black tea (WBT) for the treatmentl of Zn(II) and Ni(II) ion from aqueous solution. Maximum capacity found to be upto 90.91 mg Ni/g adsorbent and 166.67 mg Zn/g (Malakahmad *et al*., 2016). Shaikh *et al*. (2011) at the end of experiment they found that tea waste has good capacity as an adsorbent for the Arsenic removal and is highly dependent on pH. At pH 7 maximum arsenic removals was noticed as 92.5%. Aikpokpodion *et al*. (2010) study revealed that with increasing metal ion concentration the uptake of Nickel ions by the tea wastes was increased. Tea waste and coconut husk could be served as an acceptable low cost adsorbents for the treatment of Cr (VI) ions from wastewater (Dave *et al*., 2012). Thakur and Parmar (2013); Wasewar *et al*. (2008), have investigated tea waste as a good metal sponger for Cu, Ni and Fe metal ions. A list containing recovery of heavy metals Zn(II), Pb(II), Cd(II) and

Ni(II) using tea and coffee wastes can show the effectiveness of tea and coffee wastes for the treatment of heavy metals according to previous research (see table 9). To enhance the adsorption efficiency, adsorbent can be pre- treated with chemical such as bases, acids, detergents and dyes etc (Ajmal *et al.*,1998).

Table 9 List of agricultural wastes for the treatment/ remediation of heavy metals Ni(II), Cd(II), Pb(II) and Zn(II)using Tea and Coffee wastes

Waste	Metals	% removal or uptake (mg/g)	References
Tea Leaves (Waste)	Zn(II), Ni(II), Pb(II) and Fe(II)	Pb(II)> Fe(II)> Zn(II)> Ni(II)	Ahluwalia & Goyal, 2005
Tea Waste	Ni(II) and Zn(II)	71% and 79%	Kamali, 2010
Tea Waste	Ni(II)	15.26mg/g	Malkoc & Nuhoglu, 2005
Waste Tea (Camella Cinensis)	Ni(II)	98%	Aikpokpodian *et al.*, 2010
Tea Waste	Pb(II) and Cd(II)	High adsorption	Sreedevi & Parameswari, 2016
Tea Waste	Cd(II), Pb(II) and Ni(II)	Pb(II)> Ni(II)> Cd(II)	Mahvi *et al.*, 2005
Tea Waste	Cu(II) and Pb(II)	48 and 65 mg/g	Amarasingh & Williams, 2007
Tea Waste	Cd(II) and Cu(II)	Good reults	Cay *et al.*, 2004
Wasted Black Tea	Ni(II) and Zn(II)	90.91 and 166.67 mg/g	Malakahmad*et al.*, 2016
Tea Leaves (Hydrazine Monohydrate-Exhausted)	Pb(II) and Zn(II)	120.8 and 79.76 mg/g	Shrestha *et al.*, 2013
Waste Tea (Formaldehyde-Treated)	Zn(II)	120.50mg/g	Shah *et al.*, 2015
Waste Tea Leaves	Pb(II), Cd(II) and Zn(II)	Pb(II)> Cd(II) > Zn(II)	Tee & Khan, 1988
Modified Green Tea Waste	As(III) and Ni(II)	0.4212 and 0.3116 mg/g	Yang *et al.*, 2016
Tea Waste	Cd(II), Pb(II) and Cu(II)	16.87, 33.49 and 21.02 mg/g	Wan *et al.*, 2014
Activated Tea Waste	Pb(II)	99.7%	Mondal, 2010
Tea Waste Adsorbent	Cu(II) and Cd(II)	Cu(II)> Cd(II)	Dwivedi *et al.*, 1970
Spent Tea Leaves	Pb(II)	85-100 mg/g.	Lavecchia *et al.*, 2010
Waste Tea Leaves	Pb(II)	166.6 mg/g	Cheraghi *et al.*, 2015
Tea Waste	Pb(II)	70% to 100%	Yeo *et al.*, 2013
Spent Leaves Of Green And Black Tea	Pb(II)	83-130 mg/g Black tea > green tea	Zuorro & Lavecchia, 2010
Tea Wastes	Ni, Cr, Cr, Cu and Pb	100%	Nandal *et al.*, 2014
Coffee Grounds	Cu(II), Zn(II), Cd(II) and Pb(II)	High adsorption	Utomo & Hunter, 2006
Coffee Grounds	Cu(II) and Pb(II)	Pb(II)> Cu(II)	Seniunaite *et al.*, 2014
Spent Coffee Ground	Cd(II), Cu(II) and Pb(II)	Pb(II)>Cu(II)>Cd(II)	Davila-guzman *et al.*, 2016
Tea factory waste	Zn(II)	Good results	Wasewar *et al.*, 2009

CONCLUSION

Presently these days water pollution is a curse to mankind and it has become essential to develop alternate approach which is not so expensive but at the same time equally effective to treat the waste water to eliminate heavy metals. This study is an attempt in that direction and the outcome is quite encouraging In terms of efficiency of removal/eradication of heavy metals from waste water stream by making use of very inexpensive medium of adsorption. Industrial scale replication of such lab studies is worth considering as at macro level it not only save the national medical outlay against the treatment of those who are suffering of poisoning of heavy metals but it also saves the most precious human resource of the country which otherwise can be utilized effectively.

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