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REVIEW

Significance of exploiting non-living biomaterials for the biosorption of wastewater pollutants

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Abstract Industrial effluents from various sectors have become a matter of major environmental concern. The treatment of wastewater in recent year plays a significant role in order to remove the pollutants and to safeguard the water resource. The conventional wastewater treatment is considered costlier and associated with problem of sludge generation. Biosorption methods are considered as the potential solution due to their economical efficiency, good adsorption capacity and eco-friendliness. In this review, an extensive list of biosorbents from algae, bacteria, fungi and agricultural byproducts have been compiled. The suitability of biosorbents towards the eradication of heavy metals, textile dyes and phenolic compounds were highlighted. It is evident from the literature survey of recently published research articles that the biosorbents have demonstrated outstanding removal potential towards the wastewater pollutants. Therefore, biosorbents from the source of dead microbial and agricultural byproduct can be viable alternatives to activated carbon for the wastewater treatment.

Keywords Microbial biomass · Agricultural byproducts · Biosorption · Wastewater

Introduction

The origin of the water contamination includes industrial growth at various sector, civilization, agricultural practices, environmental and global changes (Yang 2011). The industrial wastewaters from battery manufacture, tanneries, petroleum refining, metal plating, mining activities, smelting, dye manufacture, printing, paint manufacture, pesticides, and photographic industries, etc., are often detected with heavy metals like cadmium, copper, nickel, zinc, lead, mercury and chromium (Kadiravelu et al. 2001). The management of heavy metals is of special concern because of the recalcitrance and persistent nature of heavy metals in the environment (Fenglian and Qi 2011). Human health is affected by the presence of heavy metals at increased concentrations. Beyond certain limit such pollutants bring serious health risk to living organisms. Cd(II), Ni(II) and Cu(II) cause kidney damage, liver damage or Wilson disease and dermatitis or chronic asthma (Dal et al. 2006; Kurniawan et al. 2006). The effluent generated from the textile industry affects the biotic and the abiotic systems (Rangabhashiyam et al. 2013a). Dye affects the aquatic plants since it reduces sunlight transmission through water. In human beings the dyes may have a mutagenic or carcinogenic influence and cause a dysfunction of the liver, kidneys, reproductive system, brain and central nervous system (Dincer et al. 2007; Shen et al. 2009). Wastewater containing dyes from textile industries is difficult to be treated or disposed due to its synthetic origin and complex aromatic molecular structures (Sun and Yang 2003). Particularly from the textile industry, more than 1.5×10^8 m^3 of colored effluents are discharged annually (Ip et al. 2010). Phenolic compounds contaminants in wastewater originate from petroleum and petrochemical, phenol producing industries, biocides, coal conversion and other chemical processes (Hamadaoui and Naffrechoux 2007). Organic compounds are the potent contaminants and mostly are not suited for the direct biological treatment (Levec and Pintar 2007). The thousands of known phenolic compounds distributed in a high

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molecular diversity and are categorized on the basis of carbon atoms and structure (Harbourne 1989; Vermerris and Nicholson 2006; Crozier et al. 2006). Phenol even in minute quantity causes severe diseases like cancer, vomiting, paralysis, nausea, smoky colored urine, etc. US Environmental Protection Agency (EPA) regulation took stringent measures for lowering phenol content in the wastewater to <1 mg/L (Banat et al. 2000). The importance of preservation and improvement of water quality is increasing continuously since it is the basic need of life. The surface and ground waters are polluted at the many places of the world and are unhygienic for drinking purpose. The global population expected to reach up to 9.3 billion by the time period of 2050 and the world may face the immense problem on fresh water scarcity (UN 2011). Technologies available to control water pollution are filtration (Zouboulis et al. 2002), ion exchange (Bolto et al. 2002), advanced oxidation processes (Esplugas et al. 2002), coagulation (Tan et al. 2000), foam flotation (Mavros et al. 1994), solvent extraction (Lin and Juang 2002), adsorption (Faust and Aly 1987), electrolysis (Szpyrkowicz et al. 1995), But most of mentioned methods are restricted due to the high cost factor. The convenience, ease of operation method and simple design are the reason why adsorption processes are more appropriate for wastewater treatment. Adsorption can be used to remove wastewater containing pollutants at low concentration in an efficient manner (Selvaraju and Pushpavanam 2009).

Biosorption is an efficient, economical and eco friendly alternative for the removal of wastewaters pollutants. The biosorbent performance on pollutants removal depends primarily on their biochemical composition, in particular the functional groups properties in the cell wall constituents, such as peptidoglycan, and the role of functional groups, such as carboxyl, amine and phosphonate (Gadd 2009; Francesca et al. 2008). A number of biosorbents from algae, fungi, bacteria, and agricultural byproducts have been studied for the biosorption of heavy metals and dyes. In general, the raw form of the biosorbents has low adsorption capacities in comparison to commercial activated carbons and ion-exchange resins. Therefore, there is an increasing research focus to enhance the biosorption capacity of the biomass (Crini 2006; Wang and Chen 2009). The adsorption of ionic pollutants using biomass is a surface phenomenon, therefore biosorption takes place on the surface of the biomass (Vijayaraghavan and Yun 2008a). The foremost desirability of biosorption is high selectivity and efficiency, cost effectiveness, possible regeneration at low cost and availability of known process equipment (Won et al. 2006). The limitations associated with the biosorption process includes that the process is highly effective only in case of the treatment of dilute effluents and the biosorbents have shorter life time when compared to the conventional sorbents (Orhan et al. 2006; Gadd 2009). The characterictics feature of the biosorption includes (1) Passive and reversible process. (2) Utilizes dead biomass. (3) Pollutants are cellular surface bound. (4) No need of nutrients supplements. (5) Process occurs through single-stage. (6) The process rate is rapid. (7) Independent of cellular metabolism. (8) Free from toxicity risk. (9) Absence of cellular growth (Chojnacka 2010).

The main objective of this review is to provide the collective and recent information pertaining to the subject of bisorbents for decontaminating the wastewater. The review (1) discusses the dead biomass of bacteria, fungi, algae and their biosorption characteristics features of pollutant removal from wastewater; (2) describes the utilization of dead biomass of agricultural byproducts as biosorbent; (3) evaluate the biosorption capacity of various microbial and agricultural byproducts dead biomass towards wastewater pollutants removal. Therefore, the current review article guides for exploring novel biosorbents from the source of microbial and agricultural byproducts for the sequestration of wastewater pollutants.

Biosorption using microbial biomass

Biosorption is the characteristic feature of biomass to bind and concentrate selective molecules from aqueous solutions. The complex phenomenon of bioaccumulation is mainly based on active metabolic transport process. Conversely, biosorption by dead biomass is a passive process, based on the affinity between the biosorbent and sorbate (Bohumil 2007). Abundant research has been carried out in the field of biosorption. Even though several researchers worked on bioaccumulation (Costa and Leite 1990), it has been discovered that the dead biomass holds high sorbing potential, hence the current focus of the most researchers shifted to biosorption (Selatnia et al. 2004a, b, c; Yetis et al. 2000; Gupta et al. 2000; Yan and Viraraghavan 2000). Figure 1 outlines the biosorption of wastewater pollutants using non-living biomaterials. The biosorption and bioaccumulation are the two distinct processes. Bioaccumulation, defined as the pollutants uptake by living cells. Biosorption process differs from the bioaccumulation by the passive uptake of pollutants by non-living biological materials (Malik 2004). The biosorbent subjected to modification (either physical or chemical) or used directly in the raw form for the wastewater pollutant removal. But for the better pollutants removal, chemically modified biosorbent were employed (Rangabhashiyam et al. 2013b). Immobilizing nonliving biomass may enhance biosorption capacity, improves mechanical strength and ease the biosorbent separation after the pollutant removal process (Aksu and Gonen 2004; Aksu 2005).

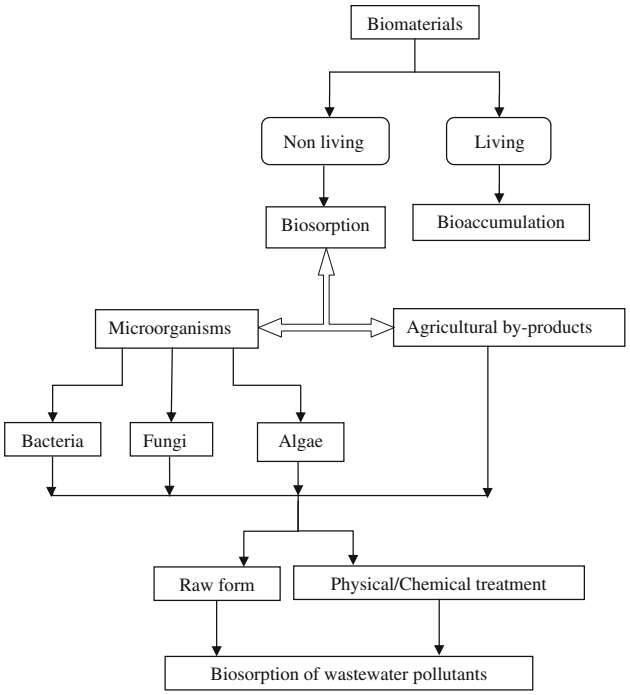

Fig. 1 Illustrates the biomaterial sources for the biosorption of wastewater pollutant

Algae

Algae are a diverse group of microorganisms distributed in water bodies and terrestrial environments, which varies from unicellular to multicellular organization. (Sisca et al. 2009). Algae possess good pollutants binding capacities due to the presence of polysaccharides, proteins or lipid on the surface of their cell walls containing some functional groups such as amino, hydroxyl, carboxyl and sulphate. In the industrial applications of wastewater treatment in order to reduce hazardous organic and inorganic pollutants biosorption often make use of dead biomass of algae, which does not require nutrients and can be exposed to environments of high toxicity (Crist et al. 1981). Microalgae in the dead form acts as the effective biosorbent since of its easier to cultivate, good yield, better efficiency and higher surface area (Azza et al. 2013).

Algal biosorption of metal

The removal of toxic heavy metals requires the biosorbent of high biosorption capacity, selectivity and good mechanical properties for the effective biosorption process. The basic biochemical constitution of algae shows enhanced metal removal performance. For example the cell wall constituents of brown algae, alginate and fucoidan are significant for heavy metal chelation (Davis et al. 2003). Algal strains like *Chlorella* sp., *Spirulina* sp., *Chaetophora elegans*, *Cladophora* sp. etc. have shown significant potential towards heavy metals removal in aqueous solution (Andrade et al. 2005; Vogel et al. 2010; Roula and Georgette 2012).

Oedogonium hatei was used as biosorbent for the removal of Ni(II) ions from aqueous solution. Batch adsorption study was carried out through untreated and treated algal biomass (with 0.1 M HCl). The maximum monolayer adsorption capacity from the Langmuir adsorption isotherm of untreated and acid-treated algae found as 40.9 and 44.2 mg g^{-1}, at contact time of 80 min, pH 5.0, algal dose 0.7 g L^{-1}, and temperature of 298 K respectively. The thermodynamic parameters reveal the biosorption was spontaneous and exothermic. Biosorbent regeneration was carried out using 0.1 M NaOH solution (Vinod et al. 2010). *Ulva lactuca*, a marine green algae used as a natural biosorbent for the removal of Cu(II), Zn(II), Cd(II) and Pb(II) at acidic conditions. Experiments were performed in glass columns and in the batch study the algae was suspended/fixed in an agar matrix. Langmuir model best fitted the adsorption isotherms in comparison with Freundlich models. The adsorption capacity increased with the increase of pH. The adsorption follows a pseudo second order kinetics model. The order of removal efficiencies of the biomaterial given as follows: fixed in columns >suspended in batch mode >fixed in agar. The adsorption rate of Cd(II) and Pb(II) were diminished when both metal added as binary solutions to the column (Maria et al. 2012). Biosorption of Cu(II) and Pb(II) on algal cells of *Chlamydomonas reinhardtii* was conducted in solutions containing 5×10^{-7} M of free metal at 303 K and pH 6. Dead algal cells showed higher biosorption than living cells for both metal ions. Removal efficiency of Pb(II) increased from 8 to 40 % for living cells and dead algal cells respectively. In case of Cu(II), the removal efficiency of dead algal cells was twice as high than living cells (55 vs. 28 %) (Roula and Georgette 2012). The investigation of raw and acid-treated *O. hatei* on Cr(VI) biosorption was carried out. The biosorption optimum conditions were 0.8 g L^{-1} biomass dose, 110 min contact time, 2.0 pH and 318 K temperature respectively. The biosorption capacities of the raw and acid-treated algae were found as 31 and 35.2 mg g^{-1} respectively. Thermodynamic parameters reveal the biosorption was spontaneous and endothermic under studied conditions. The pseudo first order kinetic model was better for describing the kinetic data (Gupta and Rastogi 2009). The absorption capacity of active and inactive biomass of *Microcystis novacekii* to remove Pb(II) was investigated. Study of inactive *M. novacekii* cells using infrared spectroscopy implies that the cell wall carboxyl and amide groups participate in Pb(II) biosorption. The maximum Pb(II) adsorbed of 70 mg g^{-1} was obtained. The biosorption of Pb(II) on inactive *M. novacekii* correlated well with the Langmuir equation in comparison to the

Freundlich isotherm equation (Rita et al. 2010). The evaluation on the biosorption of Cd(II), Ni(II) and Pb(II) ions was carried out by using both intact and pre-treated brown marine algae: *Cystoseira indica*, *Sargassum glaucescens*, *Nizimuddinia zanardini* and *Padina australis* treated with formaldehyde, glutaraldehyde, polyethylene imine, calcium chloride and hydrochloric acid. The maximum sorption capacity of formaldehyde treated *C. indica* for Cd(II) at an optimum pH of 5.5 was 19.42 mg g^{-1}, the maximum Ni(II) biosorption capacity at an optimum pH of 6.0 is 10.06 mg g^{-1} and the maximum sorption capacity for lead is observed on formaldehyde treated *N. zanardini* at an optimum pH of 5.5 was 51.83 mg g^{-1}. The maximum monolayer biosorption capacity for Cd(II), Ni(II) and Pb(II) was found as 19.56, 16.17 and 110.35 mg g^{-1}, respectively. The results were best fitted with Freundlich model among two parameter models and the Toth, Khan and Radke-Prausnitz models holded best fit among three parameter isotherm models for Cd(II), Ni(II) and Pb(II) respectively. The experimental data fitted well with the pseudo second order kinetic model (Mohammad et al. 2011). The potential of immobilized fresh water algae (in Ca-alginate) of *Scenedesmus quadricauda* studied to remove Cu(II), Zn(II) and Ni(II) ions from aqueous solutions using Ca-alginate beads as a control system was investigated. Biosorption of Cu(II), Zn(II) and Ni(II) ions on the immobilized algae indicated highest values at around pH 5.0. The maximum biosorption capacities of the immobilized algal biosorbents for Cu(II), Zn(II) and Ni(II) were 75.6, 55.2 and 30.4 mg g^{-1} respectively (Bayramoglu and Arica 2009). Methylated biomass of *Spirulina platensis* subjected for the removal of Cr(VI). Batch study carried out by varying of both Cr(VI) and methylated biomass concentrations showed that 2–4 g L^{-1} of biosorbent were able to remove Cr(VI) with efficiency $\geq$80 %, but for the higher Cr(VI) levels (43–50 mg L^{-1}) showed low removal efficiency. The Langmuir isotherm model described better the adsorption phenomenon than the Freundlich isotherm model (Elisabetta et al. 2010). An experimental design technique, the factorial design 3^3 was investigated for the biosorption of Cr(VI) by the brown seaweed *Sargassum muticum*. The three factors included were temperature, sorbent dosage and initial metal concentration at three distinctly different levels. An empirical model was developed and validated by means of ANOVA and optimized using response surface methodology. The optimization study showed 84 % as maximum Cr(VI) removal at the temperature of 323 K, 20 mg L^{-1} of metal concentration and a sorbent dosage of 2 g L^{-1}. The kinetics studies of biosorption of Cr(VI) with *S. muticum* followed pseudo second order. The best fitting equation for describing the isotherm profiles was the Langmuir model in comparison with Freundlich and Temkin (Yeslie et al. 2012). Batch biosorption studies were conducted in order to assess the effect of metal concentration and initial pH of solution on the biosorption of U(VI) and Pb(II) through chemical modified *C. indica*. The biosorption capacity of Pb(II) was enhanced by the increase of pH from 3.0 to 5.5 and maximum biosorption of U(VI) was occurred around initial pH 4.0. The single metal isotherm data at 298 K, were fitted on Langmuir equation and vacancy solution theory model using Flory–Huggins activity coefficients (Mohammad et al. 2013a, b).

Algae have been extensively studied based on the metal biosorption operating conditions. Most of the algal metal biosorption process has occurred at acidic pH, time period of <120 min, <1 g L^{-1} chemically treated algal dosage. The isotherm and kinetics studies of most of the algae for metal biosorption followed the Langmuir, Freundlich isotherm and pseudo second order kinetics. Algae have less removal capacity at high metal concentration. Most of the algal biosorbent found minimum recyclable of two times after desorption process. Algae can be used for the metal biosorption due to their ubiquitous occurrence in nature and different functional groups suits for the specific metal binding. A number of algal strains have been reviewed and most of the algae showed potential metal removal capacity in aqueous solution. Table 1 shows the metal biosorption capacities of different algae.

Algal biosorption of dye

Algae have been found as the suitable biosorbents because of their cheap availability, relatively high surface area and high binding affinity (Tien 2002; Sxatiroglu et al. 2002). The algal cells are mostly lined with micilaginous layers characterised by a significant biosorption capacity because of the presence of alginate (14–40 %) of the dry weight of the algal biomass (Lodeiro et al. 2005). Mostly microbial algal biomass towards dye biosorption has outperformed macroscopic materials like seaweeds mainly due to their cell wall constituents and functional groups distribustion (Vijayaraghavan and Yun 2008a).

The biosorption of acid orange II dye was examined by untreated and chemically modified *Stoechospermum marginatum*. The biosorption study carried out as a function of initial solution pH (2.0–10.0), initial dye concentration (30–90 mg L^{-1}), biomass dosage (0.2–2.2 g L^{-1}) contact time (5–60 min) and at constant temperature and agitation speed. The kinetic data were well described through pseudo second order model. The amine functional groups were primarily responsible for the biosorption of acid orange II dye. The propylamination modified biomass showed biosorption capacity of about two times than the untreated algal biomass (Masoud et al. 2012a, b). Activated carbons from the marine algae *U. lactuca* and *Systoceira stricta* were

Table 1 Algal biosorption capacities for metal

Algae	Metal	Adsorption capacities (mg g^{-1})	References
Spirulina platensis	Cd(II)	73.64	Abuzer and Huseyin (2011)
Spirulina platensis	Ni(II)	69.04	Abuzer and Huseyin (2011)
Cystoseira indica	Ur(VI)	318.15	Morteza et al. (2011)
Galaxaura oblongata	Cr(III)	105.2	Ibrahim (2011)
Corallina mediterranea	Co(II)	76.2	Ibrahim (2011)
Cladophora hutchinsiae	Se(IV)	74.9	Mustafa and Ahmet (2010)
Maugeotia genuflexa	As(III)	57.48	Ahmet et al. (2011)
Laminaria digitata	Cr(III)	42	Ingrid et al. (2012)
Anabaena sphaerica	Cd(II)	111.1	Azza et al. (2013)
Anabaena sphaerica	Pb(II)	121.95	Azza et al. (2013)
Lessonia nigrescens	As(V)	45.2	Hansen et al. (2006)
Cladophora fascicularis	Cu(II)	84.38	Deng et al. (2006)
Spirogyra species	Pb(II)	8.01	Gupta and Rastogi (2008a, b)
Laminaria japonica	Pb(II)	319.09	Luo et al. (2006)
Chlamydomonas reinhardtii	Pb(II)	96.3	Tuzun et al. (2005)

investigated for the biosorption of hazardous cationic dyes. Both algae were surface oxidised with phosphoric acid for 2 h and subsequently air activated at 873 K for 3 h. Biosorption capacities of 400 and 526 mg g^{-1} for malachite green and safranine O by the *S. stricta* and *U. lactuca* based activated carbons was obtained. The Langmuir and Freundlich isotherm better fitted the experimental data than the Temkin isotherms. Thermodynamic analysis confirmed the biosorption process as spontaneous and endothermic (Attouti et al. 2013). Chlorella-based biomass was used as a biosorbent for the removal of malachite green. The study data confirmed that the fast biosorption of cationic solute using the dead microalgae significantly depended on the initial malachite green concentration and algal loading. The biosorption system explained through the electrostatic interactions existed between the negatively charged biosorbent surface and the positively charged malachite green in the medium. The biosorption kinetics was well described by pseudo second order reaction model (Tsai and Chen 2010). Agitated type batch sorption carried out on the algal Spirogyra IO_2 for the biosorption of azo dye. The biosorption reaction and desorption studies showed the biosorption process as chemisorption type and reversible. The biosorption reaction followed pseudo second order type. Biosorption equilibrium data are fitted well with the Langmuir's adsorption isotherm. The biosorption reaction effective at low pH values (Venkata et al. 2008). Response surface methodology subjected to infer the biosorption characteristics of acid black 1 dye onto *Nizamuddin zanardini*, *S. glaucescens* and *S. marginatum*. The effects of three independent variables biomass dosage, dye concentration, and initial solution pH were considered for the biosorption study. A second-order polynomial model effectively described the effects of independent variables on the biosorption of acid black 1 dye. The Acid Black 1 dye removal of 99.27, 98.12 and 97.62 % were obtained for *N. zanardini*, *S. glaucescens* and *S. marginatum*, respectively (Masoud et al. 2012a, b). The potential ability of the algal *Spirogyra rhizopus* was investigated for the removal of AR 274 dye. Almost complete removal of AR 274 dye with concentration lower than 25 mg L^{-1} was done through *S. rhizopus* as the result of biocoagulation and biosorption (Ayla et al. 2006a, b). The biosorption of malachite green dye onto *S. quadricauda* and *Chlorella vulgaris* biomass was carried out using response surface methodology. The maximum malachite green dye removal efficiency of 73.49 and 91.61 % for *S. quadricauda* and *C. vulgaris* was obtained. The Fourier transform infrared spectroscopy analysis showed the distribution different functional groups on biosorbents surface, responsible for biosorption of malachite green dye. The experimental data fitted well with the pseudo second order model. The thermodynamic values indicated of biosorption process as spontaneous nature (Masoud et al. 2013). The biosorption of a malachite green oxalate by marine alga *Caulerpa racemosa* var. *cylindracea* was examined. Fourier transform infrared spectroscopy analysis confirmed the malachite green oxalate biosorption onto *C. racemosa* var. *cylindracea*. The Freundlich model better fitted the equilibrium data than Langmuir equation. The kinetic study of biosorption process followed the pseudo second order rate kinetics. The free energy changes were found as −7.078, −9.848 and −10.864 kJ mol^{-1} for 298, 308 and 318 K, respectively. Thermodynamic studies revealed the biosorption as spontaneous nature and adsorption type as physisorption (Zehra et al. 2009). The study dealt with the biosorption of methylene blue by means of dried *Ulothrix* sp. biomass. The biosorption equilibrium time was found as 30 min. The methylene blue biosorption onto *Ulothrix* sp. increased with increasing of pH, mixing rate and decreased with increasing of temperature, sorbent dosage. The biosorption kinetics followed the pseudo second order (Cetin et al. 2010). The acid black 1 removal with brown macroalgae *S. glaucescens* and *S. marginatum* was studied through batch system. The

biosorption study parameters are initial pH, contact time, initial dye concentration, biomass dosage, particle size of biosorbent and agitation speed. The equilibrium data best fitted with the Freundlich isotherm model. The kinetic data represented well with pseudo second order for the two biosorbents. Thermodynamic parameters showed acid black 1 biosorption was feasible and endothermic for *S. glaucescens* and *S. marginatum*. The acid black 1 removal capacity increased with the decrease in particle size of biosorbents. The agitation speed controls over acid black 1 biosorption capacity and was found 130 rpm as optimum agitation speed. Fourier transform infrared spectroscopy analysis confirmed the taking part of different functional groups like hydroxyl, carboxyl and amine groups in the biosorption process (Ehsan et al. 2012a, b). The acid black 1 Biosorption onto brown macroalgae, *Nizamuddina zanardinii* was investigated. The equilibrium data well described by the Freundlich model, and the biosorption capacity was 29.79 mg g^{-1}. The pseudo second order model was used as a successful model for the biosorption kinetics. The negative values of Gibbs free energy and positive value of enthalpy indicated the possibility of the biosorption process, the spontaneous nature of the biosorption. Decreased particle size of *N. zanardinii* showed increased acid black 1 biosorption (Alireza et al. 2013).

Algae have been broadly discussed under the various operating conditions for the dye biosorption. Most of the dye biosorption by algae occurred at acidic pH and the process is found significantly dependent on the initial dye concentration and biosorbent dosage. The isotherm and kinetics studies of most of the algae for dye removal followed the Langmuir, Freundlich isotherm and pseudo second order kinetics. Chemically modified algae have shown better dye removal capacity. Thermodynamic results of most part of the study revealed as spontaneous and endothermic. The Algae have been found to be as potential biosorbent because of their fast growth, availability, high surface area and high dye binding affinity. Most of the reviewed algal strains have showed effective dye removal capacity in aqueous solution. Table 2 illustrates the biosorption capacities of algae towards dye removal.

Table 2 Biosorption capacities of algae for dye removal

Algae	Dye	Adsorption capacity (mg g^{-1})	References
Stoechospermum marginatum	Acid Blue 25	22.2	Ehsan et al. (2012a, b)
Stoechospermum marginatum	Acid Orange 7	6.73	Ehsan et al. (2012a, b)
Stoechospermum marginatum	Acid Black 1	6.57	Ehsan et al. (2012a, b)
Spirulina platensis	FD&C red no. 40	295	Dotto et al. (2012)
Spirulina platensis	Acid blue 9	1450	Dotto et al. (2012)
Gelidium	Methylene blue	171	Vitor et al. (2007)
Caulerpa scalpelliformis	Basic yellow dye	27	Rathinam et al. (2007)
Spirogyra rhizopus	Acid Blue 290	1,356.6	Ayla et al. (2006a, b)
Spirogyra rhizopus	Acid Blue 324	367	Ayla et al. (2006a, b)
Azolla filiculoides	Acid red 88	109	Padmesh et al. (2005)
Azolla filiculoides	Acid green 3	133.5	Padmesh et al. (2005)
Azolla filiculoides	Acid orange 7	109.6	Padmesh et al. (2005)
Chlorella vulgaris	Remazol Black B	419.5	Zumriye and Sevilay (2005)
Caulerpa lentillifera	Astrazon Blue FGRL	49.26	Khanidtha and Prasert (2006)
Enteromorpha sp.	Methylene blue	274	Ncibi et al. (2009)
Cystoseira barbatula	Methylene blue	38.61	Caparkaya and Cavas (2008)

Algal biosorbent for phenolic compounds removal

Even though vast algal species were discovered but only a few algal species have been studied for organic pollutant biosorption. The algal biomass acts as the potential biosorbent for the removal of phenolic compounds. Hydrophobic interaction found to be the major role in algae-phenol interaction. Algal biosorption of phenolic compounds carried out using the driving force of hydrophobic and donor acceptor interactions (Rathinam et al. 2009).

Red algae, *Gracilaria verrucosa* was used as a potential biosorbent for the removal of phenoxyalkanoic acid herbicide 2,4-D. The biosorption capacity was found as 22.3 mg g^{-1}. In order to enhance the biosorption efficiency of *G. verrucosa*, the biosorbent was subjected to chemical modifications. The acid treated biomass, indicated a gradual 47 % for 2,4-D biosorption when compared with formaldehyde, alkali, and alcohol treatment. For 2,4-D biosorption, functional groups like carboxyl, hydroxyl, and amine were identified as the significant surface active groups (Ayca et al. 2012). The marine seaweeds *Macrocystis integrifolia Bory* and *Lessonia nigrescens Bory* were cross linked with $CaCl_2$ to improve their mechanical properties for the removal 2-nitrophenol and 2-chlorophenol. The maximum adsorption capacity was 97.37 and 71.28 mg g^{-1} for 2-nitrophenol using *Macrocystis integrifolia Bory and Lessonia nigrescens Bory*, whereas maximum adsorption capacity for 2-chlorophenol removal by

Macrocystis integrifolia Bory and *Lessonia nigrescens Bory* was 24.18 and 17.33 mg g^{-1} respectively (Abel et al. 2009). *Caulerpa scalpelliformis* was used for the biosorption of phenolic compounds. The study parameters included was the effects of initial pH of the solution, contact time, temperature, initial phenol concentration. Experimental data of modified green macro algae best fitted the Langmuir adsorption isotherm model. Biosorption kinetics follows pseudo second order model. The maximum biosorption capacity was found to be 20 mg g^{-1}. A Boyd plot confirmed the external mass transfer as the rate determining step of the biosorption process. The average effective diffusion coefficient was found as 1.44×10^{-9} cm^2 s^{-1} (Rathinam et al. 2009).

Chemically modified algae have shown better phenolic biosorption capacity than the unmodified form. The significant parameters for the biosorption study include pH, temperature, contact time, initial phenol concentration and algae dosage. After desorption, phenolic compounds should be subjected for further degradation process and the biosorbent can be subjected for recyle.

Fungi

Utilizing fungi as biosorbent have been extensively studied since fungal biomass available in large quantity from fermentation industries (Zhou and Kiff 1991).The fungal cell wall is made up of chitin, mannan, glucan, proteins and other polymers that possess carboxyl, hydroxyl, amino, amine phosphoryl and imidazole functional groups (Bowman and Free 2006). The dead fungal biomass holds significant biosorption capacity towards the toxic pollutants (Kumara et al. 2008).

Fungal biosorbent for metal removal

Fungal cell wall composed of different functional groups. Carboxyl, phosphate, amide, thiol and hydroxide functional groups are found to play a vital role in metal chelation (Yesim and Yucel 2002). Fungal species like *Aspergillus niger*, *Phanerochaete chrysosporium*, *Schizophyllum commune*, *Rhizopus arrhizus, Mucor* sp. and *Saccharomyces* sp. have been extensively subjected as a better biosorbent in the removal of metal ions as they are inexpensive and abundantly available (Yan and Viraraghavan 2003a, b; Veit et al. 2005; Kim et al. 2003; Say et al. 2001). Crucial parameters like pH, biosorbent modification methods, fungal species and dosage, metal species, contact time and initial metal concentration greatly influence the biosorption process (Fourest and Roux 1992).

Response surface methodology was applied for the biosorption of lead ion by *A. niger*. Lead biosorption capacity on dead *A. niger* fungal biomass was enhanced by NaOH pretreatment. Maximum biosorption capacity of the biomass for lead removal was found as 96.21 % (Malihe et al. 2008). The Cd(II) and Cu(II) biosorption by *Botrytis cinerea* was investigated with respect to initial pH, contact time and initial metal ion concentration in a batch system. Maximum biosorption capacities of Cd(II) and Cu(II) on *Botrytis cinerea* were found as 17.03 ± 0.76 and 9.23 ± 0.64 mg g^{-1}, respectively. Using 10 mM HCl solution the biosorbent was regenerated up to 96 % and utilized for five times in biosorption–desorption cycles consecutively. In comparison to the live biomass, physical and chemical pretreated biomass showed increased biosorption capacity (Tamer and Sibel 2005). Mechanism of Cr(VI) biosorption by *Termitomyces clypeatus* was analyzed. The biosorption efficiency enhanced with acid and pretreatment with $CaCl_2$. The surface of the biomass was acidic and was found as 7.75 mmol g^{-1}. The order of functional groups significance in chromium biosorption was given as follows: carboxyl > phosphates > lipids > sulfhydryl > amines (Lata et al. 2011). *Funalia trogii* in immobilized form was used for the biosorption of Hg(II), Cd(II) and Zn(II). The maximum biosorption capacities of the immobilized *Funalia trogii* for Hg(II), Cd(II) and Zn(II) were 403.2, 191.6 and 54.0 mg g^{-1} respectively. The equilibrium data exhibited good fit to the the Langmuir and the Freundlich models. The immobilized *Funalia trogii* system was regenerated by washing with 10 mM hydrochloride acid (Arica et al. 2004). Filamentous fungal biomass-loaded TiO_2 nanoparticles were used for the lead (II) biosorption. Lead ions were adsorbed on a biosorbent minicolumn at pH 4.0 followed by an elution step using 288 µL of 1.0 mol L^{-1} hydrochloric acid solution (Yasemin et al. 2010). *Aspergillus, Mortierella, Paecilomyces, Pythium*, *Rhizopus Penicillium,* and *Trichoderma* isolated from Andaman (India) serpentine soil. Maximum Co(II)-loading was achieved with *Mortierella* serpentine soil 403 biomass, removed 50 % 4.0 mM cobalt. The optimum pH and temperature for Co(II) biosorption was 7.0 and 303 K respectively. Freundlich adsorption isotherm showed better fits biosorption equilibrium data (Pal et al. 2006). The Pb(II) and Cu(II) biosorption using *Aspergillus flavus* was carried out. The maximum biosorption was 13.46 ± 0.99 mg g^{-1} for Pb(II) and 10.82 ± 1.46 mg g^{-1} for Cu(II). The equilibrium time was found as 2 h. Pretreatments of biomass by detergent, sodium hydroxide and dimethyl sulfoxide showed enhanced biosorption capacity than heat inactivated biomass. The equilibrium data fitted well the Freundlich isotherm model (Tamer and Sibel 2006). A kinetic model was developed for biosorption of copper using *A. niger*. Copper uptake by *A. niger* was confirmed as mass transfer driven process, required 30 min and attained 70 % adsorption efficiency. Formalin pretreated *A. niger* improved the uptake of copper ion (Mukhopadhyay et al. 2007).

Aspergillus and *Rhizopus* tested for their metal biosorption potential for chromium and cadmium. Maximum chromium and cadmium ions biosorption were found as 6 mM initial metal concentration. *Aspergillus* sp. 1 adsorbed 1.20 mg of chromium and 2.72 mg of cadmium per gram of biomass. *Rhizopus* sp adsorbed 4.33 mg of chromium and 2.72 mg of cadmium per g of biomass (Shaheen et al. 2007). *A. niger* was studied for the biosorption of thallium. The biosorption optimum pH was found to be between 4 and 5. The equilibrium time for biosorption was found as 6 h. The reaction kinetics well described by Lagergren's pseudo first order model and Ho's pseudo second order model. The biosorption followed the Freundlich isotherm model. In comparison with batch biosorption, column studies with iron oxide-coated immobilized fungal biomass showed lower biosorption capacities (John and Viraraghavan 2008). *Lentinus edodes* was used as an efficient biosorbent for cadmium biosorption. Cadmium uptake was higher in weak acid condition, biosorption was occurred when the pH value was greater than 2.5. Biosorption isothermal data was well represented by Freundlich model followed with the Langmuir and Temkin model (Guiqiu et al. 2008). *Pleurotus ostreatus* was immobilized on Amberlite XAD-4 for the biosorption of Cr(III), Cd(II) and Cu(II). Maximum adsorption of Cr(III), Cd(II) and Cu(II) ions was occurred in the pH range 4–5. Adsorbed metal ions desorbed using 1 M HCl. Even after 10 cycles of biosorption and desorption the variation of biosorption capacity was not more than 2.0 % (Sevgi and Munevver 2011). Cr(VI) ions biosorption with *A. niger, Aspergillus sydoni* and *Penicillium janthinellum* was investigated. The optimum pH was 2.0 for biosorption of Cr(VI) ions. Cr(VI) ions biosorption was 91.03 % with *A. niger* at biosorbent dosage of 0.6 g/50 mL, 87.95 and 86.61 % with *Aspergillus sydoni* and *Penicillium janthinellum* at 0.8 g/50 mL biosorbent dosage. Adsorption data was best fitted in both Freundlich and Langmuir isotherms model (Rajender et al. 2008). *Pleurotus mutilus* was tested as a biosorbent for the removal of iron(III)–cyanide complex ions. Biomass pretreatment with acetic acid slightly improved its biosorption efficiency. The biosorption kinetics followed both pseudo first order and pseudo second order models. Through Langmuir isotherm model the highest biosorption efficiency was found as 620 mg g^{-1} (Abdelmalek et al. 2009). *Mucor hiemalis* was subjected as the biosorbent for nickel removal in a batch system. The biosorption of Ni(II) increased with increase of pH, temperature, Ni(II) concentration and rotational speed, biosorption decreased with increase of biomass concentration. Biosorption equilibrium attained at 150 min. The biosorption data fitted well to both the Langmuir and the Freundlich isotherm models. The biosorbent regeneration efficiency was above 80 % (Kshama and Varsha 2011). *Mucor plumbeus*–alunite composite was tested for the biosorption of Pb(II). The Pb(II) biosorption capacity of composite biosorbent was higher than *Mucor plumbeus*. Langmuir isotherm better fitted the equilibrium data in comparison with the Freundlich and Dubinin–Radushkevich model. The intraparticle diffusion was played an important role in the first 40 and 50 min for *Mucor plumbeus* and *Mucor plumbeus*–alunite composite biosorbent (Tamer et al. 2013). *Tremella fuciformis* and *Auricularia polytricha* was studied as economical biosorbents for the removal of Cd(II), Cu(II), Pb(II) and Zn(II). The humid *Tremella fuciformis* showed the maximum biosorption capacity for the four metals in the multi-metal solutions. In comparison with pseudo first order model, the pseudo second order model well described the adsorption kinetics, representing a two-step biosorption process (Rong et al. 2010). A tropical white-rot basidiomycete, BDT- 14 (DSM 15396) was investigated for its Cr(VI) biosorption. The adsorption efficiency was 100 % when the initial Cr(VI) concentration was 100 mg l^{-1} with 1,000 mg biomass. But when Cr(VI) concentration was 500 mg l^{-1}, 47.5 % adsorption was only observed. The adsorption data was fitted well with the Langmuir and Freundlich isotherm models (Trivedi and Patel 2007). The potential of three fungi *Gliocladium viride* AI003, *Mucor* sp. HI33 and *A. niger* AH09, as a compatible/incompatible consortium for the cadmium biosorption from paddy water was carried out. Seven different combinations were studied as possible consortia. Maximum cadmium biosorption was found for the consortium of 48-hold *Mucor* sp. HI33 + 72-hold *Gliocladium viride* AI003 + 72-hold *A. niger* AH09. This consortium was given a maximum Cd biosorption of 99.98 % (Arifa and Humaira 2012).

Chemically modified fungus shows increased biosorption capacity towards metal removal. The isotherm and kinetics studies of most of the fungal metal biosorption followed the Langmuir, Freundlich isotherm and pseudo second order kinetics. Biosorbent regeneration carried out through desorption process and the biosorbent are subjected for good number of recycles. Fungi have a large capacity for the metal biosorption from aqueous solutions and even outperformed activated carbon. Fungal cell wall with numerous functional groups plays an important role in metal biosoption. Fungal species like *A. niger*, *Rhizopus* sp. and *Mucor* sp. shows potential metal biosorption. Table 3 represents the biosorption capacities of different fungi towards metal removal.

Fungal biosorbent for dye removal

Activated carbon acts as an effective adsorbent towards dyes removal in wastewater treatment with its large surface area and high load capacities. But the drawback associated with the activated carbon usage in dyes removal is its disposal problem (Xiong et al. 2010). Inactived fungal

Table 3 Adsorption capacities of fungi for the removal of metal

Fungi	Metal	Adsorption capacities (mg g^{-1})	References
Rhizopus arrihizus	U(VI)	112.2	Wang et al. (2010)
Trametes versicolor	Cu(II)	140.9	Venkata et al. (2011)
Candida tropicalis	Zr(IV)	179	Akhtar et al. (2008)
Mucor rouxii	Pb(II)	53.75	Yan and Viraraghavan (2003a, b)
Mucor rouxii	Zn(II)	53.85	Yan and Viraraghavan (2003a, b)
Aspergillus terreus	Cu(II)	15.24	Cordova et al. (2012)
Yarrowia lipolytica	Ni (II)	95.33	Nikhil et al. (2012)
Yarrowia lipolytica	Ni (II)	85.44	Nikhil et al. (2012)
Amanita rubescens	Pb(II)	38.4	Ahmet and Mustafa (2009)
Amanita rubescens	Cd(II)	27.3	Ahmet and Mustafa (2009)
Cladosporium cladosporioides	Cu(II)	7.74	Xiaona et al. (2009)
Gliomastix murorum	Cu(II)	9.01	Xiaona et al. (2009)
Bjerkandera sp.	Cu(II)	12.08	Xiaona et al. (2009)
Streptomyces rimosus	Al(III)	11.76	Amina et al. (2010)
Penicillium	Cu(II)	38.3	Xin et al. (2011)
Streptomyces lunalinharesii	Cd(II)	24.8	Diego et al. (2012)
Aspergillus niger	Cd(II)	10.14	Malihe et al. (2009)
Solanum nigrum	Cd(II)	247.5	Xiao et al. (2010)
Pleurotus ostreatus	Ni(II)	20.40	Amna et al. (2011)
Pleurotus ostreatus	Cr(VI)	10.75	Amna et al. (2011)
Rhizopus cohnii	Cd(II)	40.5	Luo et al. (2010)
Lactarius scrobiculatus	Pb(II)	56.2	Ruhan et al. (2009)
Lactarius scrobiculatus	Cd(II)	53.1	Ruhan et al. (2009)
Penicillium citrinum	Ur(IV)	127.3	Pang et al. (2011)
Clitopilus scyphoides	Cd(II)	200	Moussous et al. (2012)
Trametes versicolor	Pb(II)	208.3	Munagapati et al. (2011)
Trametes versicolor	Cd(II)	166.6	Munagapati et al. (2011)
Auricularia polytricha	Cd(II)	63.3	Haiwei et al. (2012)
Auricularia polytricha	Cu(II)	73.7	Haiwei et al. (2012)
Auricularia polytricha	Pb(II)	221	Haiwei et al. (2012)

mycelia have been used for dye biosorption. The dye binding mechanism by fungal biosorbent is based on the nature of dye, fungi type, biosorbent prepration and biosorption conditions (Casieri et al. 2008; Prigione et al. 2008a, b). Fungal species such as *Trametes versicolor*, *Candida tropicalis*, *Funalia trogii*, *Rhizopus stolonifer*, *A. niger*, *Ganoderma applanatum* etc. have been used as biosorbent for the elimination of dye from wastewater (Yesilada et al. 2002; Zeroual et al. 2006; Fu and Viraraghavan 2001; Matos et al. 2007; Qi et al. 2002; Pethkar et al. 2001).

In the batch system *Phanerocheate chrysosporium* was used for the removal of Reactive Blue 4. The biosorption equilibrium time was attained at 4 h. Maximum reactive blue 4 biosorption was observed at pH 3.0. At 600 mg L^{-1} dye concentration the biosorption was found as 132.5, 156.9, 147.6 and 81.1 mg g^{-1} for native, heat, acid and base treated biosorbent. The Freundlich and Temkin models well fitted the biosorption equilibrium data (Bayramoglu et al. 2006). Biosorption of reactive black 5 onto dried *Penicillium restrictum* was carried out. An optimum condition for biosorption was found as initial pH 1.0, equilibrium time 75 min and biomass concentration of 0.4 g dm^{-3} at 293 K. The maximum biosorption of reactive black 5 using *Penicillium restrictum* was obtained as 98.33 and 112.50 mg g^{-1} (Cansu et al. 2007). Immobilized *Trichoderma viride* showed enhanced biosorption of methylene blue by 30 % in comparison with free biomass. Biosorption kinetics followed the pseudo second order. The equilibrium data was fitted well the Langmuir adsorption models. Fourier transform infrared spectroscopy of *Trichoderma viride* showed amine, hydroxyl, carbonyl and amide bonds were involved biosorption (Asma et al. 2009a, b). *Agaricus bisporus* was used to remove acid red 44 dye. The highest acid red 44 biosorption was achieved at pH 2.0. Equilibrium time was attained at 30 min. The maximum monolayer biosorption capacity was found as 1.19×10^{-4} mol g^{-1}. Thermodynamic parameters indicated biosorption as spontaneous and endothermic in nature (Tamer et al. 2009). Biosorbents from *Fomes fomentarius* and *Phellinus igniarius* were subjected for the removal of methylene blue and rhodamine B. The methylene blue was found more adsorbable in comparison to rhodamine B. Molecular structure and ionic radius of methylene blue and rhodamine B were found for differences in biosorption. Biosorption of methylene blue increased but rhodamine B decreased as pH increased from 3 to 11. The presence of carboxylic and amino groups in rhodamine B made for its lower biosorption compared to methylene blue (Nityanand et al. 2006). *Penicillium chrysogenum* was used for the biosorption anionic dyes like acid orange 8, acid blue 45 and reactive orange 16. Biosorbent surface was modified with polyethylenimine, cross-linked with glutaraldehyde. The maximum biosorption capacity of the surface modified biosorbent for acid orange 8, acid blue 45 and reactive orange 16 was increased from 33, 18 and 25 mg g^{-1} to 352, 196 and 338 mg g^{-1}(Low

et al. 2008). *Cunninghamella elegans* was characterized in terms of adsorption isotherm and kinetics for the removal of acid blue 62, acid red 266 and acid yellow 49. Kinetics and equilibrium tests confirmed that the *Cunninghamella elegans* was more selective towards acid red 266 since the high negative charge density of functional group $-CF_3$ interact with $-NH_x$ active sites of the biosorbent (Russo et al. 2010). *Trichoderma harzianum* used for the biosorption of rhodamine 6G and erioglaucine. The Langmuir and Freundlich adsorption isotherms fitted well the equilibrium data. The second-order kinetics highly correlated with the biosorption data (Sadhasivam et al. 2009). *A. niger* and *Trichoderma* sp. was used as biosorbents for the removal of orange G. The maximum biosorption was occurred at pH 2. Experimental data was best fitted with Langmuir and Freundlich isotherms. The monolayer biosorption capacity was 0.48 mg g^{-1} for *A. niger* and 0.45 mg g^{-1} for *Trichoderma* sp. biomasses. The pseudo second order model fitted well for both the biosorbents (Arumugam and Nethaji 2011). Chemically modified *Aspergillus wentii* was used for biosorption studies of Safranin. The equilibrium data was analyzed through various parameters isotherms to understand the biosorption. Adsorption isotherms showed that the interaction of safranin with *Aspergillus wentii* surface was a localized monolayer biosorption. The order of experimental data fitted adsorption isotherm models was given as follows: Fritz–Schluender (five parameter) > Langmuir > Khan > Fritz–Schluender (four-parameter) > Temkin (Yasmin et al. 2011). *Paecilomyces* sp. was subjected for the biosorption of reactive yellow 85, reactive orange 12, for reactive black. Zeta potential showed an electrostatic interaction between the binding sites and dye anions. Fourier transform infrared spectroscopy showed that the functional groups like amine, hydroxyl, carbonyl and amide bonds were involved in biosorption. Equilibrium data of biosorption of reactive yellow 85 and reactive orange 12 dyestuffs fitted well to the Freundlich and Langmuir adsorption models (Ahmet et al. 2013).

Most of the fungal biosorption of dye occurs at the acidic pH. The isotherm of most of the fungal dye biosorption followed the Langmuir, Freundlich isotherm. *Aspergillus* sp. *Penicillium* sp. *Phanerocheate chrysosporium* are some suitable fungal biomass for the dye removal. The biosorption capacities of fungi for dye removal were shown in the Table 4.

Fungal biosorbent for phenolic compound removal

Researches in the biosorption of organic compounds with fungal biomass showed that enhanced phenolic removals were obtained with dead fungal biomass than the live biosorbent (Rao and Viraraghavan 2002). The immobilization of biosorbent has improved the mechanical strength, porosity characteristics, size and stability. Immobilized biosorbent also exhibits a better performance in packed bed column reactors because of the less clogging problem, ease for regeneration, biosorbent recycle and simple adsorption–desorption process (Zulfadhyl et al. 2001).

The biosorption of 2, 4-dichlorophenol was studied using *P. chrysosporium*. *P. chrysosporium* showed the highest biosorption capacity of 4.09 mg g^{-1} at pH 5.0, 2, 4-dichlorophenol concentration 50.48 mg l^{-1}, temperature 298 K. The Freundlich model better fitted the experimental data than the Langmuir model. The biosorption process followed pseudo second order kinetics. The second order kinetic constant was decreased with temperature increase. The biosorption activation energy was found as -16.95 kJ mol^{-1}. The thermodynamic study revealed biosorption exhibited exothermic and physisorption (Wu and Yu 2006). The biosorption of naphthalene, fluorene, phenanthrene and acenaphthene, pyrene by white-rot fungal

Table 4 Adsorption capacities of fungi for dye removal

Bacteria	Metal	Adsorption capacities (mg g^{-1})	References
Corynebacterium glutamicum	Ni(II)	111.4	Vijayaraghavan et al. (2008a)
Streptomyces rimosus	Ni(II)	32.6	Selatnia et al. (2004a, b, c)
Nostoc muscorum	Cr(VI)	22.92	Gupta and Rastogi (2008a, b)
Escherichia coli	Cd(II)	162.1	Fatthy (2011)
Escherichia coli	Zn(II)	137.9	Fatthy (2011)
Spirulina platensis	Hg(II)	428	Cain et al. (2008)
Aphanothece flocculosa	Hg(II)	456	Cain et al. (2008)
Micrococcus luteus	Pb(II)	1,965	Zully et al. (2012)
Micrococcus luteus	Cu(II)	408	Zully et al. (2012)
Bacillus sp.	Cr(VI)	17.8	Liu et al. (2008)
Bacillus subtilis	Cu(II)	100.70	Liu et al. (2013)
Geobacillus toebii	Cu(II)	41.5	Sadin et al. (2009)
Geobacillus thermoleovorans	Cu(II)	48.5	Sadin et al. (2009)
Corynebacterium glutamicum	Ag^+	52.5	Sneha et al. (2010)
Bacillus cereus	Pb(II)	22.1	Ferdag et al. (2011)
Bacillus pumilus	Pb(II)	28.06	Ferdag et al. (2011)
Acinetobacter junii	Cr(VI)	6.94	Madona et al. (2012)
Stenotrophomonas maltophilia	Cu(II)	26	Jinshao et al. (2013)
Bacillus licheniformis	Cr(VI)	69.4	Zhou et al. (2007)
Staphylococcus xylosus	Cr(VI)	143.0	Ziagova et al. (2007)

biomass fitted well with the Freundlich equation. Biosorption process involved simultaneous surface sorption, partitioning processes and chemical reactions respectively (Baoliang et al. 2010). The phenol biosorption was carried out using Ca-alginate immobilized *P. chrysosporium*. The phenol biosorption was increased with the increase of biosorption time and the biosorption equilibrium time was about 60 min. The biosorption process followed pseudo second order kinetics. The phenol removal was followed an anti-Langmuir behavior for biosorption. Therefore nonlinear estimation was suggested for the evaluation of biosorption equilibrium (Viktor et al. 2013). Sulfuric acid-treated *A. niger* was found as the most effective biosorbent for the removal of phenol. The phenol biosorption by pretreated *A. niger* was well represented through the Brunauer Emmet Teller model (Rao and Viraraghavan 2002). *P. chrysosporium* was immobilized on different matrices like Ca-alginate, Ca-alginate-polyvinyl alcohol and pectin for the biosorption of 2,4-dichlorophenol. Ca-alginate immobilized *P. chrysosporium* showed evidence of good adsorption efficiency 60 %. The biosorption data of 2,4-dichlorophenol was described well by the Langmuir and Freundlich isotherms. Desorption operation was done using distilled water as eluant and found 82.16 % efficiency. The adsorption/desorption cycles was demonstrated up to five cycles (Wu and Yu 2007). *A. niger* was studied for the biosorption of pentachlorophenol. Cetyltrimethylammonium bromide *A. niger* showed atmost removal of pentachlorophenol. The biosorption equilibrium time was found as 2 h. The biosorption kinetics was well described using pseudo second order model. Pentachlorophenol biosorption was found to be exothermic type and decreased with temperature increase. Biosorbent functional groups like carboxyl, amide and hydroxyl groups played a significant role in pentachlorophenol biosorption (Mathialagan and Viraraghavan 2009). *Rhizopus arrhizus* TISTR 3610 employed for the biosorption of nonylphenol. The biosorption data was correlated using the biosorption isotherm models and order was given as follows: Fritz–Schluender > Redlich–Peterson > Freundlich > Langmuir isotherms. The pseudo first order kinetics was best fitted with the experimental data. The diffusivity of nonylphenol in the *R. arrhizus*—chitosan beads was determined through the shrinking core model and the values were in the ranges of 2.3736×10^{-4}–1.8950×10^{-4} $cm^2\ s^{-1}$. Desorption was carried out by methanol (Weeranuch et al. 2009). *S. commune* was deployed for the biosorption of phenol, 2-chlorophenol and 4-chlorophenol. Through Langmuir model the maximum monolayer biosorption capacity of *S. commune* for phenol, 2-chlorophenol and 4-chlorophenol was found as 120, 178 and 244 mg g^{-1}. The equilibrium time was attained at 2 h. Kinetics of biosorption followed pseudo second order. The column regeneration was carried out up to three cycles. 0.1 M NaOH was used as eluant for the biosorbent regeneration (Nadavala and Kim 2011). The biosorption of methylene blue was carried out using *Aspergillus fumigatus*. Physical pre-treatment by autoclaving and adding SDS 1 mM, NaCl 1 mM to the methylene blue solutions highly improvement of methylene blue biosorption. The maximum biosorption capacity was 125 mg g^{-1}. The biosorption was favorable, physico-chemical in nature and fitted well with Langmuir and Freundlich models (Rawa and Samir 2012).

The immobilized form of fungal biomass shows enhanced phenolic compound biosorption. The isotherm of most of the fungal phenolic compound biosorption fitted the Langmuir and the Freundlich isotherm. Most of the fungal biosorption of phenolic compound carried out at the acidic pH condition. Desorption efficiency of the biosorbent evaluated though different solid/liquid ratios. After desorption, phenolic compounds should be subjected for further degradation process.

Bacteria

The cell wall of the bacteria acts as the primary component for interacting with pollutants like metal ions or dyes, where they are sorbed on the surface or within the cell wall (Beveridge and Murray 1976; Doyle et al. 1980). Cell wall functional groups play important roles in the biosorption process. The functional group includes carboxyl, amine, phosphonate and hydroxyl groups (Van et al. 1997). Bacterial biosorption has attracted attention of many researchers since they are extremely abundant available, versatile and constitute a significant fraction among the terrestrial living biomass (Mack et al. 2007). The potential of bacteria for the biosorption of various organic and inorganic wastewater pollutants have been well established (Aksu 2005; Volesky and Holan 1995).

Bacterial biosorbent for metal ion removal

A variety of naturally occurring bacteria exhibits high biosorption capacity towards the removal of metals. The negatively charged and abundantly distributed carboxyl groups in the bacterial cell wall actively involves in the binding of metal cations (Golab and Breitenbach 1995). Thermophilic bacteria able to grow at a temperature range of 45–80 °C. The membrane of such bacteria undergoes various adaptations for the optimal functioning at high temperatures. The thermophilic bacteria possess different metal biosorption mechanisms in comparison with the mesophilic species (Burnett et al. 2006; Hetzer et al. 2006).

The cadmium biosorption using NaOH treated *Streptomyces rimosus* was carried out in the batch study. The

external mass transfer was the controlling step of the overall biosorption process. The equilibrium data was better fitted by Langmuir isotherm model. Biosorption capacity was found as 63.3 mg g^{-1} (Selatnia et al. 2004a, b, c). *Bacillus thuringiensis* strain OSM29 was investigated for the biosorption towards cadmium, copper, lead chromium, and nickel. The optimum pH for nickel and chromium biosorption was 7, and for the biosorption of cadmium, copper and lead were 6. The biosorption by *B. thuringiensis* strain OSM29 was highest for nickel (94 %) followed by copper (91.8 %). Through Fourier transform infrared spectroscopy the surface chemical functional groups of *B. thuringiensis* strain OSM29 biomass were identified as amino, carboxyl, hydroxyl, and carbonyl groups (Mohammad et al. 2013a, b). Cr(VI) biosorption was carried out using alginate immobilized *Nostoc calcicola* HH-12 and *Chroococcus* sp. HH-11. The optimum biosorption conditions for the two strains are almost same (pH 3–4, contact time 30 min and initial chromium concentration of 20 mgl^{-1}). Based on the higher values of the Langmuir and Freundlich isotherm parameters, *Chroococcus* sp. HH-11 was found as an better biosorbent for the removal of Cr(VI) from wastewater (Kamra et al. 2007). *Bacillus subtilis* was immobilized on Amberlite XAD-4 for the biosorption of Cu(II) and Cd(II). The optimum pH values of 7.0 and 7.5 were obtained for the biosorption for Cu(II) and Cd(II) respectively. The procedure was applied to Cu(II) and Cd(II) determination in aqueous solutions of river and well water systems (Mehmet et al. 2007). The metal biosorption was investigated using *Geobacillus thermodenitrificans*. *Geobacillus thermodenitrificans* reduced the concentration of Fe(III)(91.31 %), Cr(III)(80.80 %), Co(II)(79.71 %), Cu(II)(57.14 %), Zn(II) (55.14 %), Cd(II)(49.02 %), Ag(II)(43.25 %) and Pb(II) (36.86 %) within the time period of 720 min. When *G. thermodenitrificans* was applied in the industrial waste water biosorption, metal ions concentrations was reduced up to 43.94 % for Fe(III), 39.2 % for Cr(III), 35.88 % for Cd(II), 18.22 % for Pb(II), 13.03 % for Cu(II), 11.43 % for Co(II), 9.02 % for Zn(II) and 7.65 % for Ag(I) within 120 min (Chatterjee et al. 2010). *Brevundimonas* sp. ZF12 with high 226Ra content made 50 % biosorption of 250 ppm cadmium. The biosorption equilibrium data was fitted well by the Langmuir isotherm. Kinetic studies followed pseudo second order model (Nasrin et al. 2011). Halobacterial strains isolated from saltern pan *Natronobacterium magadii*, *Natrononococcus occultus* and *Halobacterium sodomense* showed the maximum metal resistance activity against Ni(II), Al(III) and Hg(II) metals. The SEM–EDS analysis confirmed the metals biosorption was carried out at higher concentration of Ni(II) and Al(III) and at lower concentration of Hg(II) (Williams et al. 2012). Biosorption of Pb(II) and Cu(II) ions was studied through *Bacillus* sp. The maximum metal ions biosorption was observed at pH 3.0 ± 0.1 for Pb(II) and pH 5.0 ± 0.1 for Cu(II) ions. Equilibrium times for Pb(II) and Cu(II) ions was attained within 15 and 30 min respectively. The maximum biosorption of Pb(II) and Cu(II) ions was determined as 92.27 ± 1.17 mg g^{-1} at 250 mg l^{-1} concentration and 16.25 ± 1.64 mg g^{-1} at 200 mg l^{-1} concentration. The experimental data was fitted well to Langmuir isotherm model (Sibel et al. 2006). The Box–Behnken design matrix and response surface methodology was applied to analyze over the interactive effects of variables like pH, temperature and initial metal ions concentration (10.0–60.0 mg/L) for the biosorption of Cr(VI), Ni(II) and Zn(II) ions on immobilized *Bacillus brevis*. 17 experiments were conducted towards the construction of a quadratic model. Independent variables of significant value 0.0001 indicated the significance of these variables in the biosorption process. Values of "Prob > F" <0.0500 indicated model terms was significant for the biosorption of Cr(VI), Ni(II) and Zn(II). The experimental data fitted second order polynomial equation (Rajender et al. 2009). The biosorption of anionic $Sb(OH)_6^-$ by chemically modified *Microcystis* was examined and mechanisms were discussed. Acid treatment biosorbent showed enhanced biosorption than original biomass. The experimental data fitted with the Freundlich model. The zeta potential and Attenuated total reflectance-infrared spectroscopy analysis showed that the binding of $Sb(OH)_6^-$ on biosorbent occurred through electrostatic attraction and complexation. Further, the analysis confirmed the involvement of amino, carboxyl and hydroxyl groups in $Sb(OH)_6^-$ biosorption. Scanning electron microscopy–Energy-dispersive X-ray spectroscopy analyses indicated the biosorption process as highly heterogeneous (Fuhong et al. 2011). Biosorption of Zn(II) by *Acinetobacter* sp. was investigated in batch study. Optimum biosorption conditions were found at initial pH of 6, biosorbent dosage of 0.5 g L^{-1} and initial Zn(II) ion concentration of 100 mg L^{-1} and contact time of 90 min. The maximum biosorption capacity was found as 36 mg g^{-1}. The experimental data were better fitted with the Freundlich isotherm (Reza et al. 2013).

Chemically modified bacteria shows enhanced biosorption capacity towards metal removal. Baterial biosorption of metal mostly carried out with the pH of <7 and the equilibrium attained within <2 h. The isotherm and kinetics studies of most of the bacterial metal biosorption followed the Langmuir, Freundlich isotherm and pseudo second order kinetics. Bacteria in metal biosorption have attracted attention since bacteria are extremely abundant and versatile microorganism. The bacterial biosorption of various metal ions like Pb, Zn, Cu, Fe, Cr, Ni, U, Ag etc. were reported. Table 5 represents the maximum biosorption capacities of bacteria for metal removal.

Bacterial biosorbent for the dye removal

Most of the dye molecules exist as cations in solutions, the bacterial cell wall with carboxyl and other negatively charged groups will easily binds with the dye cations (Golab and Breitenbach 1995). The modification of the specific binding sites on the bacteria enhances the dye biosorption capacity by many multiples. The functional group of bacteria like carboxyl, phosphonate, sulfonate, amine and hydroxyl groups are mostly involved in the dye binding mechanisms (Vijayaraghavan and Yun 2007b; Won et al. 2004; Fu and Viraraghavan, 2002).

Binding mechanisms of reactive orange 16 was investigated by the protonated waste biomass of *Corynebacterium glutamicum*. The reactive orange 16 biosorption was reversible at <7 pH but irreversible under basic pH conditions. This was because of electrostatic interaction at acidic pH. The maximum biosorption was evaluated as 156.6 ± 6.2 and 64.0 ± 2.4 mg g^{-1} at pH 2 and 4 respectively. Fourier transform infrared spectroscopy and X-ray photoelectron spectroscopy analysis showed that the chemical bond existed between biomass surface and dye molecules under basic pH conditions (Won et al. 2009a, b). The reactive dye biosorption capacity was enhanced by cross-linking *Corynebacterium glutamicum* with polyethylenimine. The amine groups distributed in the cell wall of *Corynebacterium glutamicum* was electrostatically interacted with reactive dye anions. The protonation of the amine groups favoured reactive red 4 biosorption. Maximum biosorption capacity of crosslinked biosorbent was found as 485.1 mg g^{-1} when comparison to 171.9 mg g^{-1} of the raw *Corynebacterium glutamicum*. Desorption was successful at basic pH and the biosorbent was regenerated and reused over four cycles (Mao et al. 2009). *Corynebacterium glutamicum* was employed as a biosorbent for reactive black 5. The biosorbent was subjected to the pretreatment of HCl, H_2SO_4, HNO_3, NaOH, Na_2CO_3, $CaCl_2$ and NaCl. 0.1MHNO_3 pretreated biosorbent exhibited hightest biosorption of 195 mg g^{-1}. According to the Langmuir model maximum reactive black 5 biosorption of 419 mg g^{-1} was obtained. The kinetics followed pseudo second order model. Thermodynamic parameters revealed biosorption process as spontaneous and endothermic in nature (Vijayaraghavan and Yun 2007a). The biosorption ability of *Pseudomonas* sp. strain DY1 to adsorb acid black 172 was examined through kinetics and mechanisms involved in acid black 172 biosorption. Biosorption process followed pseudo second order model. Increased initial dye concentration significantly enhanceed acid black 172 biosorption by heat-treated biosorbent. Through the analysis of scanning electron microscopy, atomic force microscopy and transmission electron microscopy revealed that heat treatment of the biosorbent increased the cell wall permeability and denatured the intracellular proteins. The biosorption experiments on different celluluar components was confirmed that intracellular proteins played a significant role for the cause of enhanced biosorption of acid black 172 (Du et al. 2012). Polysulfone matrix immobilized *Corynebacterium glutamicum* was studied for the biosorption towards methylene blue. Neutral or alkaline pH favored methylene blue biosorption. The immobilized biosorbent was regenerated and reused up to three cycles. The packed column study exhibited good breakthrough curve with methylene blue biosorption of 124 mg g^{-1} biomass (Vijayaraghavan et al. 2008b). *Corynebacterium glutamicum* was treated with poly (amic acid) to enhance basic blue 3 biosorption. The grafting of poly(amic acid) on biosorbent increased the

Table 5 Adsorption capacities of bacteria for the removal of metal

Bacteria	Metal	Adsorption capacities (mg g^{-1})	References
Corynebacterium glutamicum	Ni(II)	111.4	Vijayaraghavan et al. (2008a)
Streptomyces rimosus	Ni(II)	32.6	Selatnia e al. (2004a, b, c)
Nostoc muscorum	Cr(VI)	22.92	Gupta and Rastogi (2008a, b)
Escherichia coli	Cd(II)	162.1	Fatthy (2011)
Escherichia coli	Zn(II)	137.9	Fatthy (2011)
Spirulina platensis	Hg(II)	428	Cain et al. (2008)
Aphanothece flocculosa	Hg(II)	456	Cain et al. (2008)
Micrococcus luteus	Pb(II)	1,965	Zully et al. (2012)
Micrococcus luteus	Cu(II)	408	Zully et al. (2012)
Bacillus sp.	Cr(VI)	17.8	Liu et al. (2008)
Bacillus subtilis	Cu(II)	100.70	Liu et al. (2013)
Geobacillus toebii	Cu(II)	41.5	Sadin et al. (2009)
Geobacillus thermoleovorans	Cu(II)	48.5	Sadin et al. (2009)
Corynebacterium glutamicum	Ag^+	52.5	Sneha et al. (2010)
Bacillus cereus	Pb(II)	22.1	Ferdag et al. (2011)
Bacillus pumilus	Pb(II)	28.06	Ferdag et al. (2011)
Acinetobacter junii	Cr(VI)	6.94	Madona et al. (2012)
Stenotrophomonas maltophilia	Cu(II)	26	Jinshao et al. (2013)
Bacillus licheniformis	Cr(VI)	69.4	Zhou et al. (2007)
Staphylococcus xylosus	Cr(VI)	143.0	Ziagova et al. (2007)

Table 6 Agricultural byproducts for the metal biosorption

Agricultural byproducts	Metal	Adsorption capacities (mg g^{-1})	References
Sugarcane bagasse	Cu(II)	9.48	Lu et al. (2012)
Coir pith	Cr(VI)	165.00	Parinda and Patip (2012)
Mungbean husk	Cd(II)	35.41	Asma et al. (2009a b)
Rice husk	Zn(II)	19.38	Shafey (2010)
Rice husk	Hg(II)	384.62	Shafey (2010)
Rice husk	As(V)	2.5	Pehlivan et al. (2013)
Sawdust	Zn(II)	47	Flaviane et al. (2010)
Sugarcane bagasse	Zn(II)	45	Flaviane et al. (2010)
Kenaf Fibres	Cu(II)	47.27	Hasfalina et al. (2012)
Almond green hull	Co(II)	45.5	Ahmadpour et al. (2009)
Corn Cob	Zn(II)	79.21	Achanai et al. (2012)
Date pits	Cu(II)	35.9	Mohammad et al. (2010)
Date pits	Cd(II)	39.5	Mohammad et al. (2010)
Orange peel	Cu(II)	50.25	Feng et al. (2010)
lemon peel	Co(II)	22	Bhatnagar et al. (2010)
Sawdust	Cr(VI)	41.5	Gupta and Babu (2009)
Orange peel	Pb(II)	476.1	Ningchuan (2011)
Orange peel	Cd(II)	293.3	Ningchuan (2011)
Orange peel	Ni(II)	162.6	Ningchuan (2011)
Banana peel	Pb(II)	2.18	Jamil et al. (2010)
Banana peel	Cd(II)	5.71	Jamil et al. (2010)
Sugarcane bagasse	Ni(II)	2	Aloma et al. (2012)

density of the carboxyl groups. The pH edge experiments conducted only within the range of 3–10 indicated electrostatic attraction was existed between carboxyl groups of biosorbent and basic blue 3. From the Langmuir model maximum removal of basic blue 3 was found as 173.6 mg g^{-1} at pH 9. The equilibrium time was found as 10 min (Won et al. 2009a, b). Through succination the amine groups of biosorbent *Corynebacterium glutamicum* modified to carboxyl and were examined towards methylene blue biosorption. Biosorption isotherm studies revealed that the succinated biosorbent showed better methylene blue biosorption. The methylene blue biosorption capacities were 207.3, 337.5 mg g^{-1} for raw and succinated biosorbent. Regeneration of succinated biosorbent was successful and five repeated cycles was performed well for over the methylene blue biosorption (Vijayaraghavan et al. 2008c). Polysulfone-immobilized *Corynebacterium glutamicum* was employed as biosorbent for the continuous removal of reactive black 5 using up-flow packed column. Thomas and Yoon–Nelson models well fitted the experimental data. Biosorbent was recycled for multiple cycles using 0.1 M NaOH (Vijayaraghavan and Yun 2008b).

Chemical modification of bacteria has brought out enhanced biosorption capacity towards dye removal.

Table 7 Agricultural byproducts for the biosorption of dye (Rangabhashiyam et al.2013b)

Agricultural byproducts	Dye	Adsorption capacities (mg g^{-1})	References
Neem leaf powder	Congo red	72	Bhattacharyya and Sharma (2004)
Teak tree bark	Methylene Blue	333.3	Patil et al. (2011)
Wheat straw	Basic Yellow 21	71.43	Hassanein and Koumanova (2010)
Hazlenut shell	Methylene Blue	76.9	Ferrero (2007)
Tree fern	Basic red 13	408	Ho et al. (2005)
Pine sawdust	Acid yellow 132	398.8	Ozacar and Sengil (2005)
Peanut hull	Methylene Blue	68.03	Gong et al. (2005)
Coir pith	Rhodamine-B	203.2	Namasivayam et al. (2001)
Rice husk	Indigo carmine	65.9	Lakshmi et al. (2009)
Banana pith	Acid brilliant blue	4.42	Namasivayam et al. (1998)
Orange peel	Malachite green	483.632	Kumar and Porkodi (2007)
Cotton waste	Safranine	875.00	Mckay et al. (1999)
Mango seed kernel	Methylene Blue	142.90	Kumar and Kumarana (2005)
Jack fruit peel	Basic blue 9	285.71	Hameed (2009)
Palm fruit bunch	Basic yellow 21	327.00	Nassar and Magdy (1997)
Jute fiber	Malachite green	136.586	Porkodi and Kumar (2007)
Oil palmshell	Methylene Blue	243.90	(Tan et al. 2008)
Vetiver roots	Methylene Blue	423.0	Altenor et al. (2009)
Brazilian pine-fruit shell	Remazol black B	446.2	Cardoso et al. (2011)
Coconut tree flower	Reactive red	181.90	Senthilkumaar et al. (2006)

Baterial biosorption of dye mostly carried out with the pH of <7 and the equilibrium time of <2 h. Biosorbent regeneration carried out through desorption process and the biosorbent are subjected for good number of recycles.

Agricultural byproducts as biosorbents

Agricultural byproducts are lignocellulosic materials that mainly composed of lignin, cellulose and hemicelluloses.

Basic constituents of agricultural byproducts are simple sugars, proteins, lipids, water, hydrocarbons and starch holds different functional groups like alcohols, aldehydes, carboxylates, phenols, ketones and ethers. Agricultural byproducts are better to employ in the biosorption of wastewater pollutants since they can be used without or with a minimum processing. Agricultural byproducts are cheap, available in plenty and renewable. These agricultural byproducts were used either in the raw form or through physical or chemical modification for pollutant biosorption (Ahmedna et al. 2000; Franca et al. 2009; Rangabhashiyam et al. 2013c).

Chemically modified agricultural byproducts have shown better heavy metal and dye biosorption compared to the unmodified form. Mostly acids and bases were used to modify agricultural byproducts. Such modifications bring out increased number of active binding sites and the introduction of new functional groups in the biosorbent which favours effective metal and dye removal. Tables 6 and 7 represent the different source of the agricultural byproducts for the biosorption of metal and dye.

Conclusions

Removal of pollutants from wastewater through biosorption has gained significant attention in the recent years since the process offers a number of advantages. Activated carbon use in developing countries is more problematic due to cost. The biosorbents from different dead microorganisms and agricultural byproducts act as the potential tool for the sequestration of wastewater pollutants. It is evident that most of the biosorbents employed for wastewater treatment have exhibited efficient sorption capacity. Therefore, such biosorbents play may a vital role especially in the developing and underdeveloped countries. However, the existing research gaps in biosorption using low cost adsorbents have been projected as follows: (1) employment of genetically engineered strains for biosorption; (2) isolation of potent strains from the industrial effluents for application in biosorption; (3) evaluation of mechanical strength of biosorbents for successful recycles; (4) development of appropriate modelling in order to further investigate biosorption; (5) studies on the combined use of dead microbial biomass and agricultural by-products; (6) consideration of adsorption/desorption process associated with minimum sludge generation; (7) investigation of competitive biosorption of different pollutants; (8) testing of biosorbent performance under different operational modes; (9) extension of batch biosorption to pilot scale in order to verify feasibility on commercial scale.

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