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Removal of Heavy Metal Ions from Water using Beta-Cyclodextrin: A Review

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Abstract – *Host-guest behavior of beta-cyclodextrin results in the formation of inclusion complexes which aids in removal of heavy metal ions from water. This paper exclusively deals with heavy metal ions removal from water. We have discussed about the variety of modifications of Beta-Cyclodetxrin that have been researched and discovered till date. All the information in this paper is extracted from various papers written so far by honored scientists and researchers in this field. Future Prospects and Recyclability are also briefly described here.*

Key Words: Cyclodextrin, Water pollution, modifications, Crosslinking, Adsorption, Metal ions.

1. INTRODUCTION

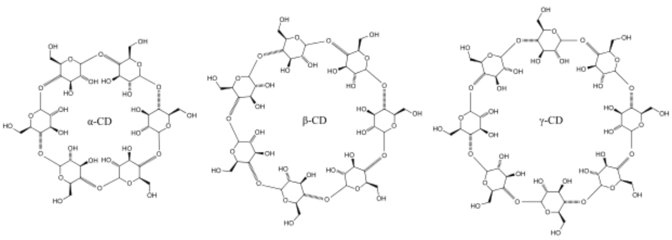
Industries these days are all concerned about mass production and they pay little or no heed to the harm caused by their manufacturing processes and also by their waste products to the environment. It is a common practice for industries like mining, steel and thermal power plants, print and paint industries, fertilizer plants, textile dyes etc. to release their byproducts into the water bodies. The frequent constituents of these industrial wastes are organic dyes, heavy metal ions amongst many other toxic substances. This paper discusses about the variety of modifications of beta-Cyclodextrin (Beta-CD) which have proven to be useful in the removal of heavy metal ions from water. Activated Carbon has been used for this purpose for several decades. However Activated Carbon has some disadvantages like low selectivity, regeneration problems, and had slow, non-specific uptake. To overcome those disadvantages Beta-Cyclodextrin has been brought into the picture. Beta-CD adsorbs heavy metal ions from waste water and form inclusion complexes. Hydrophilic exterior and hydrophobic interior of beta-CD contributes to the formation of complex with heavy metal ions. Researches showed that beta-CD and its derivatives had higher efficiency in absorbing, separating and detecting heavy metal ions indicating promising applications for removal of heavy metal ions from wastewater.

In this review article, various Modifications of Beta-cyclodextrin are explained based on previous researches.

2. ABOUT BETA-CYCLODEXTRIN

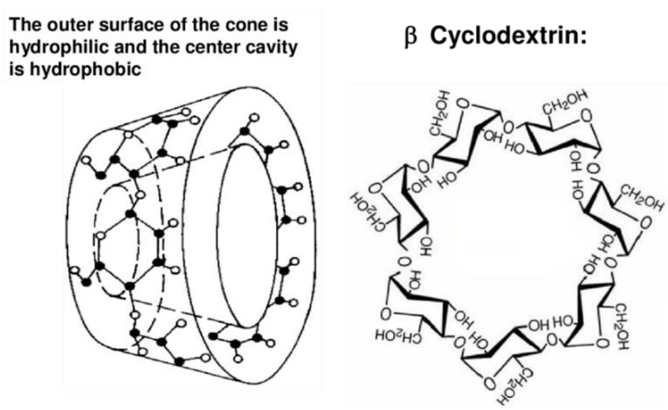
Enzymatic degradation of starch by bacteria leads to the formations of naturally formed cyclic oligosaccharides using which are called Cyclodextrin (CD). Chemical or Enzymatic modifications of Beta-CD derivatives can be made, in order to vary the chemical or physical properties of the CD molecule like its chemical stability, thermal properties, solubility, complex forming capacities and other properties too.

Cyclodextrins are found in 3 forms namely α-, β- and γ-CDs which are torus like macro-rings which are made up of 6,7 & 8 glucopyranose units which are doughnut or wreath-shaped truncated cone. They are homogeneous, crystalline and non – hygroscopic.



Types of Cyclodextrin, Source: Wikipedia

Beta-CDs are not soluble in most organic solvents, but the solubility in some solvent/water mixtures may change. More often, CDs are seen as empty capsules of some molecular size. CDs can form inclusion complexes which have different types of hydrophobic guest molecules wherein one of the molecule acts host which includes totally or partly the guest molecules by means of physical or chemical forces.



Structure of Beta-cyclodextrin, Source: Haynes, Robert. (2009). Using cyclodextrin to stabilize and control colloidal micro-stickies to improve paper machine runnability. 2. 1268-1298.

When one molecule of the guest and one molecule of the host i.e., CD try to come in contact with each other by forming complexes, then that phenomenon is called complexation. Inclusion complexes are formed when a water molecule is being substituted by a less polar guest where these interactions are favored by the guest with the hydrophobic cavity of the host. Herein, Enthalpy and Entropy play major roles during their respective changes. Driving forces for the complexations can be due to the following reasons:

a) The substitution of the energetically unfavored polar – apolar interactions from the more favored polar- apolar interaction along with the polar-polar interactions.

b) Cyclodextrin rings strain a bit release on complexation.

c) Various interactions like the hydrogen bond between the host and guest and even the Van der Waals interactions.

3. MODIFICATIONS

3.1. Simply Cross-Linked Beta-CD

Beta-CD is extremely soluble in water just like its alpha and gamma counterparts. Because of its solubility it cannot be directly used for water purification. Henceforth the modification of beta-CD is necessary. The most commonly used approach is to crosslink the beta-CD using suitable reagents. Crosslinking is referred to the copolymerization between the hydroxyl of CD and the di-functional groups or multi-functional groups of a compound for example: epoxide. Epichlorohydrin (EPI) is the most common crosslinking agent for bulk polymerization. This results in the formation of branched beta-cyclodextrin polymers with poor solubility in water. If the degree of cross-linking is sufficiently high, the resulting polymer becomes insoluble in water and henceforth making them favorable for use in water treatment.

Preparation of the polymers is simple, but the mechanical strength and chemical stability is poor. Especially in the chromatographic separation, the low efficiency restricts the application of the above mentioned polymers and chemical stability is also poor.

Xianshi Zhu et al., 2008 reported 95% of adsorption capacity for cobalt when Epichlorohydrin is used as the crosslinking agent for beta-CD.

Malefetse et. al., 2009 reported the use of ionic liquids with Beta-CD for the simultaneous quenching of these pollutants from the drinking water. Crosslinking agents used were hexamethylene diisocyanate (HDI) and toluene diisocyanate (TDI) which are bi-functional linkers and the ionic liquids which delivered better results and maximum adsorption capacities were mono-6-methylimidazolium tosylate and mono-6-pyridinium tosylate. This polymer could adsorb around 100% of Chromium.

Raoov et al., 2013 reported the adsorption of arsenic using a macroporous material such as beta-Cyclodextrin-ionic liquid polymer (CD-ILP) which was first synthesized by functionalized beta-cyclodextrin (CD) with 1-benzylimidazole (BIM) to form mono functionalized CD (beta-CD-BIMOTs) and was further polymerized using a toluene diisocyanate (TDI) linker to form insoluble CD-ILP (beta-CD-BIMOTs-TDI). The resultant adsorption capacity was found to be 95% which is a promising value.

Cross-linking of beta-CD with Epichlorohydrin was reported in 2 research papers by Sikder et. al., wherein he used this cross-linking agent to know the adsorptivity of cadmium and chromium in his paper published in the year 2013 and of copper in his paper published in 2014.

Zhao et al., 2015 synthesized an EDTA-cross-linked Beta-cyclodextrin (EDTA-Beta-CD) bifunctional adsorbent, which was fabricated by an easy and green approach through the polycondensation reaction of beta-cyclodextrin with EDTA as cross-linker, for simultaneous adsorption of metals and dyes from aqueous solution. The CD units are covalently joined by repeating EDTA linkers. The cavities of CDs are responsible for the capture of the dye molecules, forming inclusion complexes. The EDTA-groups are expected to act not only as cross-linkers but also as chelating sites for metal ions. The steric effect of EDTA moieties and CD cavities could endow the network-structure polymer more advantage to bind organic compounds on the network by physical sorption.

Liu et al., 2015 tried to modify Beta-CD with silica gel by using Epichlorohydrin as the crosslinking agent and it was seen that the adsorption of uranium was 6.45 mg/g which is a pretty low value to be of any commercial use.

Crini et al., 2016 reported 92% adsorptive capacities for metals like Co, Ni, and Zn in this research study where he cross-linked 1,2,3,4-butanetetracarboxylic acid (BTCA) with beta-CD.

Junyong et al., 2017 reported that the rigid structure of tetrafluoroterephthalonitrile linked with beta-CD would generate a high-surface-area and porous polymer of beta-CD. Thus the adsorption capacity and adsorption rate would be largely improved. At different pH values, the adsorption was affected by electrostatic interaction and distribution of HMs species. This study proved that the beta-CD polymer could be developed into a kind of effective adsorbent for heavy metals. It was also observed that the removal efficiency decreased over time but still remained at a high level, therefore making beta-CD polymer suitable for regeneration. Out of which he found that when beta-CD was cross linked with tetrafluoroterephthalonitrile, maximum adsorption capacity was found in lead followed by copper and finally cadmium.

Haitao et al., 2017 synthesized EDTA-Beta-CD modified with Fe-Al bimetallic hydroxides (FA) which exhibits an adsorption capacity of 6.64 mg/g for arsenic which is however not a high value as compared to its counterparts.

Lang Lin et al., 2017 synthesized magnetic beta-cyclodextrin inclusion complex with phosphonobutanetricarboxylic acid (MNP-CD-PBTCA) for the removal of Hg^{2+}. PBTCA included in CD showed stronger electron-donating ability due to the existence of phosphoryl, hydroxyl, and carboxyl group, which was proved by the data of quantum chemical calculation. Nanosized Fe_3O_4 is able to provide a giant loading area for CD due to its huge specific area and consequently include more PBTCA; simultaneously, other organic phosphoric acids except PBTCA could also be included in CD to achieve special adsorption. Hence, Lang Lin et al., 2017 used the Core–shell structure and morphology of beta-cyclodextrin-modified magnetic nanoparticle inclusion complexes with PBTCA (MNP-CD-PBTCA) to adsorb metal ions like Hg and achieve an adsorption capacity like 77.59 mg/g.

Commercial feasibility is also a factor which needs to considered and henceforth Sun et al., 2018 used Schwertmannite which can be biosynthesized with beta-CD. Schwertmannite has an advantage of being readily available across the globe.

Chen et al., 2019, tried to cross link beta-CD with Polydopamine (PDA). PDA is advantageous because of its excellent biocompatibility, strong adhesion, hydrogen bonding and metal chelation by catechol and abundant functional groups. Rich amine and catechol groups on the surface of PDA provide active adsorptive sites for heavy metals. The modification of PDA with beta-CD had an extreme performance improvement on the adsorptive capacities for the targeted pollutants.

A series of beta-CD crosslinked polymeric adsorbents were created by the copolymerization of methacrylic-modified beta-CD (MCD) and 1-vinylimidazole (VI) under different monomer feeding ratios .The resultant adsorbent was labeled PCD-VI by Qin et al., 2019.

On reacting Carboxymethyl-beta-cyclodextrin polymer with zirconium under mild reaction conditions resulted in Zr/CM-beta-CD which was used to remove metal ions like cadmium and copper. The above cross linked polymer was synthesized by Tang et al., 2019.

Epichlorohydrin cross linked beta-CD when loaded onto the surface of zeolite and vermiculite significantly increased the adsorption of lead and cadmium (Zheng et al., 2019). The adsorptivites of resultant polymer was 126.35 mg/g and 68.35 mg/g for lead and cadmium respectively.

A new polymer has been synthesized through polymerization of beta-CD in the presence of Epichlorohydrin and Thiourea in order to adsorb heavy metal ions like lead and cadmium. This polymer was synthesized by Nejad et al., 2020. The prepared beta-CD-ECH-TU was applied for the removal of toxic metals and gave excellent adsorptivities like 285.714 mg/g for lead and 126.58 mg/g for cadmium.

Since radioactive pollutants are becoming a big problem to our environment and as their uses are increasing day by day, which is why Wang et al., 2015, tried to focus on these pollutants and used attapulgite as an adsorbent to help B-CD adsorb the pollutants. Attapulgite is a hydrated magnesium aluminum phyllosilicate, which exists in nature as a fibrous clay mineral. It has a rough surface, resulting in a relatively high surface area and moderate cation exchange capacity. With increasing beta-CD amount in the composites, more oxygen-containing functional groups are available to form surface complexes with Eu(III) and thereby results in more Eu(III) adsorption to the beta-CD/magnetic attapulgite composites. Increase in beta-CD also leads to an increase in the removal percentage of Eu(III).

3.2. Nano-adsorbents

The breakthrough developments in nanotechnology has led to the discovery of nanoadsorbents which have large surface area, accessible active sites and short diffusion paths. These nanoadsorbents include carbon based adsorbents; graphene, carbon nanotubes and their composites which have been reported to possess a great potential of removing heavy metal ions from water. Graphene; an atom-thick single layer of carbon atoms in a closely packed honeycomb hexagonal two-dimensional lattice, is the basic building block for graphitic materials such as graphene oxide, fullerene, carbon nanotubes (CNT) and other carbon materials (Geim & Novoselov, 2007). Ying-Xia Ma et al., 2018 successfully fabricated beta-cyclodextrin modified magnetic graphene oxide (beta-CD/MGO) nanohybrids using simple one-step reverse phase co-precipitation method. The synthesized adsorbent showed an excellent adsorption capacity of 279.21, 51.29 mg/g for Pb(II) and Cu(II) respectively at 298K.

Graphene oxide (GO), as a derivative of graphene, has not only large specific surface area but also abundant oxygen-containing functional groups on the edges and basal planes, such as hydroxyl, carboxyl and epoxy groups, which makes GO to be well dispersed in water and easy to be modified. However, it is inconvenient to separate from the aqueous solution. Fe_3O_4 nanoparticles with relatively low cost, large specific surface area and convenient separation by an external magnetic field have been anchored on the GO-based materials surface and applied in various wastewater treatment. However, Fe_3O_4 nanoparticles anchored on the GO-based materials surface are highly susceptible to oxidation when they are exposed to the atmosphere. To overcome the drawback, it is necessary to use beta-CD functionalized magnetic graphene oxide composites as adsorbents. Beta-CD has abundant hydrophilic primary hydroxyl groups on its surface. Therefore, iron(III) and iron(II) cations in the solution could be anchored on the hydrophilic functional groups of GO and beta-CD, and then in situ formed Fe_3O_4 nanoparticles at these particular sites in the presence of NaOH, which prevented the agglomeration of pulverized Fe_3O_4 and avoided the stack of GO. Simultaneously, GO provided a large contact surface for these nanoparticles Fe_3O_4, which could prevent the detachment of Fe_3O_4 nanoparticles. In addition, beta-CD played a protection function to prevent the oxidation of Fe_3O_4 nanoparticles exposed in the air. Moreover, the beta-CD/MGO nanohybrids could be easily separated from the liquid phase system by an external magnetic field during the process of preparation.

Nyairo et al.,2018 synthesized GO conjugated with methyl-beta-cyclodextrin (GO-m-beta-CD) and studied its adsorption capacity towards Cu(II) and Pb(II). GO-m-beta-CD had an extremely high adsorption capacity of 312.5mg/g for Pb and a reasonable value of 83.3 mg/g for Cu. These values are significantly higher than beta-cyclodextrin modified magnetic graphene oxide (beta-CD/MGO) nanohybrids [Ying-Xia Ma et al., 2018]. Moreover, GO-m-beta-CD can be used repeatedly up to five times with minimal loss of their initial adsorption capacity. The beta-CDsGO@Fe_3O_4 nanoparticles exhibited relatively high adsorption capacities of 99.51 mg/g and 100.23 mg/g for As(V) and As(III) respectively as compared to other adsorbents available (A. Santhana et al.,2017).

GO modified with magnetic cyclodextrin has a reasonable adsorption capacity of 120 mg/g for Cr(VI) since it possess the unique properties of GO (large surface area and good mechanical properties) and magnetic cyclodextrin (strong acid-resistance, superparamagnetism and high adsorption capability) (Lulu Fan et al.,2012).

A novel beta-CD polymer named CD-E-MGO, obtained by grafting beta-CD onto MGO through ethylenediamine, was also applied as a sorbent to remove Cr(VI) from aqueous solution. However the synthesized polymer had a poor affinity for Cr(VI) with an adsorption capacity of 68.41 mg/g at 303K (Hui Wang et al.,2014).

Zongxue Yu et al.,2007 developed beta-cyclodextrin with gamma-(2,3-epoxypropoxy) propyl trimethoxysilane (GPTMS) functionalized GO which had a maximum adsorption of 44.795 mg/g for Cu(II) at 318 K. However, this value is not at par with the adsorption capacity of beta-cyclodextrin modified magnetic graphene oxide (Beta-CD/MGO) nanohybrids (Ying-Xia Ma et al., 2018). Beta-Cyclodextrin grafted graphene oxide (GO)(beta-CD-GO) was found to have a potential application as an adsorbent for the removal of Cd(II) with an excellent adsorption capacity of 117.07 mg/g. Huiling Zheng et al.,2018 found out that beta-cyclodextrin decorated graphene oxides (b-CD-GO) has maximum adsorption capacity of 149.56 mg/g for Pb(II).

SONG WenCheng et al.,2014 synthesized cyclodextrin-modified graphene oxide nanosheets (denoted as CD/GO) by an in-situ polymerization method and was characterized by Fourier transform-infrared spectroscopy, X-ray photoelectron spectroscopy, Raman spectroscopy and potentiometric acid-base titration. The characterization results indicated that CD was successfully grafted onto GO surfaces by forming a chemical bond. The work highlighted the simultaneous sorption of U(VI) and organic pollutants from aqueous solutions and discovered an enhancement of simultaneous sorption of U(VI) and HA (humic acid) onto CD/GO which was governed by electrostatic interaction and formation of ternary cation-anion surface complexes. CD/GO synthesized had an adsorption capacity of 97.3 mg/g in the absence of Humic Acid and 103.9 mg/g in the presence of HA for U(VI). The presence of HA both enhanced U(VI) sorption at low pH and reduced U(VI) sorption at high pH, whereas the presence of U(VI) enhanced HA sorption. The surface-adsorbed HA acts as a bridge between U(VI) and CD/GO and forms strong surface complexes with U(VI).

Carboxymethyl-cyclodextrin (CM-CD) has the ability to complex heavy metals such as cadmium, nickel, strontium and mercury through the interactions between the metal ions and –COOH functional groups. A.Z.M.Badruddoza et al.,2011 synthesized a nano-adsorbent for the adsorption of metal ions by surface modification of Fe_3O_4 nanoparticles with CM—CD. The high surface area to volume ratio of the Fe_3O_4 nanoparticles and magnetic properties together with the adsorption capabilities of CM--CD through complex formation facilitate the removal of heavy metals from wastewater. The maximum adsorption capacity of CMCD-MNPs for Cu(II) was determined to be 47.2 mg/g, which is considerably higher than that of some other reported magnetic adsorbents. Both FTIR and XPS analyses clearly reveal that the oxygen atoms on CMCD-MNPs are the main binding sites for Cu(II) to form surface complexes.

Abdolmaleki et al.,2015 fabricated novel nano-adsorbent, triazinyl-beta-cyclodextrin modified magnetic nanoparticles (T-beta-CD–MNPs) for the removal of heavy metal ions from aqueous solutions by reacting mono-chlorotriazinyl-beta-cyclodextrin (T-beta-CD) with magnetite Fe_3O_4 nanoparticles (MNPs) via the nucleophilic substitution of the chloride leaving group of T-beta-CD by the attacking hydroxyl group of the MNPs. T-beta-CD–MNPs exhibit excellent removal ability for heavy metal ions (Pb^{2+}, Cu^{2+}, Zn^{2+} and Co^{2+}). T-beta-CD-MNPs had maximum adsorption capacities of 105.38, 58.44, 51.30 and 33.33 mg/g for Pb^{2+}, Cu^{2+}, Zn^{2+} and Co^{2+} respectively which is larger than MNPs that had been modified by other natural compounds. It was observed that superparamagnetic magnetite (Fe_3O_4) nanoparticles coated with CM--CD polymer(carboxymethyl—cyclodextrin) has maximum uptake capacities for Pb^{2+}, Cd^{2+} and Ni^{2+} as 64.50, 27.70 and 13.20 mg/g respectively.

Magnetite nanoparticles (MNPs) modified with hydroxyapatite (HA) and beta-CD (Fe_3O_4@HA-CD) had maximum adsorption capacities for Cd^{2+} and Cu^{2+} as 100.00 and 66.66 mg/g, respectively, henceforth can be recognized as potential candidates for heavy metal adsorption applications. Zhiqiang Guo et al.,2015 grafted beta-CD on Fe_3O_4 magnetic nanoparticles via the chemical co-precipitation method to prepare Fe_3O_4@CD magnetic composites (MCs) which has excellent ability to adsorb Eu(III) from aqueous solution which is sufficiently higher than the prevailing adsorbents available. Beta-cyclodextrin coated magnetic nanoparticles as novel adsorbents can effectively be used to remove As(III) from aqueous solution because the modification of Fe_3O_4 by CM-beta CD enhances the arsenic adsorption. The maximum monolayer adsorption capacity of CM-beta-CDMNPs was 12.33 mg/g at room temperature which is very less compared to the adsorption capacity of the beta-CDsGO@Fe_3O_4 nanoparticles.

Beta-cyclodextrin chemically grafted onto halloysite nanotube/iron oxides (HNT/ iron oxides) forms a novel magnetic CD/HNT/iron oxide composite having an excellent adsorption capacity for U(VI) which is considerably higher than that of some other reported materials until when A. S. Helal et al.,2017 used APTES (3-Aminopropyltriethoxysilane) to modify and functionalize magnetic nanoparticles with succinyl-beta-cyclodextrin, to give succinyl-beta-cyclodextrin-APTES@Fe_2O_3, a nanocomposite material which has a remarkable potential for the recovery of uranium from aqueous solution. It is highly selective for Uranium(VI), is stable in acid solution and has a long service life. The maximum sorption capacity for uranyl ions is close to 286 mg/g at pH 6 which is a significantly high value. Linear beta-cyclodextrin polymer functionalized multiwalled carbon nanotubes (MWCNTs-CDP) had adsorption capacity of 89.54 mg/g at 323.15K and 66.16 mg/g at 293.15 K for U(VI) indicating that adsorption capacity increases with temperature.

Abu Zayed Md Badruddoza et al.,2017 developed a novel multifunctional sorbent, cyclodextrin−IL (ionic liquid) polymer-functionalized magnetic nanoparticles (Fe3O4-CDI-IL MNPs) which had the ability to adsorb Cr(VI) from industrial waste water. The primary mechanisms for the removal of Cr(VI) were electrostatic interactions and ion exchange. The magnetic iron oxide core rendered this sorbent magnetically responsive, thereby facilitating their separation and regeneration from the complex aqueous solutions. In addition, the synthesized sorbent maintained the high removal efficiency after 10 cycles of sorption−desorption regeneration tests. Overall, the chemical stability, efficient sorption performance, easy separability, and excellent recyclability of these magnetic nanocomposites make them excellent candidates as solid-phase extraction materials for water treatment of coexisting toxic pollutants.

The magnetic composite of beta-cyclodextrin grafted onto multiwalled carbon nanotubes/iron oxides (denoted as MWCNTs/iron oxides/CD) was synthesized using the plasma-induced grafting technique and was developed for the removal of inorganic and organic pollutants from aqueous solutions. Plasma-induced grafting treatment is a useful method for the introduction of functional groups onto material surfaces without altering material bulk properties. It is an environmentally friendly technique for modifying the surface properties of materials because large amounts of chemicals are not needed in grafting processes. The MWCNTs/iron oxides/CD material has very high adsorption capacities in the removal of Pb(II) and MWCNTs/iron oxides/CD can be separated and recovered from solution by magnetic separation. The decline in efficiency was not more than 5% after reuse five times, which indicates that MWCNTs/iron oxides/CD has a good reusability (Hu et al., 2010).

3.3 Bio Adsorbents

Jamil Rima et al., 2013 polymerized beta-Cyclodextrin polyurethane with beetroot fibers (Bio-polymer), which was further applied for removal of organic and inorganic contaminants from wastewater. Several functional groups like hydroxyl, carboxylic, aldehyde, ketone, C=N were observed in the FTIR spectra of the fibers.

For the removal of Pb^{2+} from aqueous solution, Hai-tao Zhao et al., 2018 synthesized beta-cyclodextrin functionalized biochars based adsorbent using epichlorohydrin as cross-linking agent. Pb^{2+} was mainly removed by means of chemical process for functionalized palm biochar, whereas for functionalized rice straw biochar both physical and chemical means were used.

Youyi Xia et al., 2007 modified novel cellulosic fibers with Beta-cyclodextrin (CFEC) for adsorption of Cu (II) and neutral red from their aqueous solutions. Modified cellulosic fibers exhibited higher Cu (II) adsorption, and Cu (II) ion uptake values of 6.24 mg/g were obtained at 293 °C, as against no adsorption for unmodified cellulosic fibers.

Beta-cyclodextrin–chitosan modified walnut shell biochars (Beta-CCWB) was synthesized by Xixian Huang et al., 2015 and used as a low-cost adsorbent for the removal of Cr (VI) from aqueous solutions. Amino and carboxyl groups were the major functional groups responsible for Cr (VI) removal. Removal efficiency of Cr (VI) by Beta-CCWB (about 93%) was higher than pristine biochar by about 27%.

To reduce the amount of Zn^{2+} in industrial waste water, Jiaxin Liu et al., 2020 modified beta-cyclodextrin (beta-CD) in order to obtain a beta-cyclodextrin polymer (beta-CDP) by crosslinking with chitosan (CTS) molecular chain. The adsorption amount and adsorption rates of Zn^{2+} using CTS/beta-CDP complex was 97.70 mg/g and 78.92% respectively.

For the removal of Cr(VI) and Cu(II) from contaminated surface and groundwater, Md. Tajuddin Sikder et al., 2014 synthesized chitosan –Fe^0- nanoparticle–carboxymethyl beta-cyclodextrin beads. The synthesized CS–NZVI–CM-beta-CD beads exhibited combined effects of highly dispersed NZVI (nanozero-valent iron) particles and highly functional CS–CM-beta-CD.

Di Wu et al., 2018 synthesized EDTA modified beta-cyclodextrin/chitosan (CDCS-EDTA) by a two-step method which was further applied for the removal of Pb (II) and anionic dye acid red (AR) from aqueous solution. The adsorption mechanism involved chelation between EDTA-groups and Pb (II) ions; and electrostatic attraction between Pb^{2+} and -OH and -NH_2 of adsorbent.

Microwave induced copolymerization of beta-Cyclodextrin (beta-CD) and chitosan (Ch) carried out by A.K. Sharma, et al., 2010 resulted in Ch-g-beta-CD copolymer (without any radical initiator or catalyst) which was applied for removal of Pb (II) from synthetic solutions.

To enhance the adsorption efficacy of Cr (VI) from aqueous solution, Xue-LianWang et al., 2019 synthesized non-toxic chitosan-based composite, namely beta-cyclodextrin/chitosan/hexamethylenetetramine (beta-CD-CS@HMTA). Maximum removal capacity of Cr (VI) reached 333.8 mg/g which was superior to most of the CS derivative adsorbents.

Beta-CD units and CS-EDTA are covalently joined by pentafluoropyridine (PFP), to obtain the porous beta-CD polymer using 2-methyltetrahydrofuran as solvent. Formation of porous polymer P-CDEC was verified with solid-state 13C-NMR. Host-guest inclusion and chelating effect were the main phenomena responsible for interaction between adsorbent and solutes. (Tingting Yu et al., 2013)

Alberto Rubin Pedrazzo et al., 2019 synthesized safe and sustainable nanosponges (NSs) by reacting beta-CD and linecaps (LC) with citric acid in water, using sodium hypophosphite monohydrate catalyst. The obtained NSs were compared with NSs prepared by crosslinking beta-CD and LC with pyromellitic dianhydride (PMDA).

Feiping Zhao et al., 2017 fabricated a bio-derived chitosan-EDTA-beta-cyclodextrin (CS-ED-CD) trifunctional adsorbent via a facile and green one-pot synthesis method using EDTA as a cross-linker. Introduction of chitosan enhanced the loading of beta-CD and improved the yield of the water-insoluble adsorbent. CS-ED-CD exhibited better removal efficiency than EDTA-CD for both metals and organic pollutants.

Arsenic ions are common heavy metal contaminants existing widely in wastewater. J. T. Tsiepe et al., 2017 developed a bio-nanocomposite using chitosan (CS), beta cyclodextrin (beta-CD) and iron oxide nanoparticle (Fe_3O_4) for the removal of As (III) from aqueous solutions. The stability of the beta-CD–CS blends was improved after the incorporation of Fe_3O_4 nanoparticles. It was observed that the adsorption capacity of the beta-CD–CS–Fe_3O_4 nanocomposite reached a maximum at pH 9 with 93% removal and a minimum at pH 10.5 with 76% removal.

A novel biodegradable beta-cyclodextrin-based hydrogel (CAM) synthesized by Zhanhua Huang et al., was applied for the removal of Cd^{2+}, Pb^{2+} and Cu^{2+} from aqueous solutions. The adsorption capacities of the metal ions occurred in the order Pb^{2+} (210.6 mg/g)> Cu^{2+} (116.41 mg/g)> Cd^{2+} (98.88 mg/g). CAM hydrogel was found to be biodegradable by Gloeophyllum trabeum, and the degradation efficiency was found to be 79.4% even after 21 days.

Yanmei Zhoua et al., 2011 studied the adsorption behaviour on a novel modified cellulose containing beta-cyclodextrin and quaternary ammonium groups. When the equilibrium concentration of Cr (VI) was changed in the range from 0 to 48.3mg/L, the adsorption quantity of Cr (VI) was found to increase sharply. Maximum adsorption capacity of chromium (VI) reached 61.05mg/g.

Many researches exhibited that biochar could serve as a potential environmental adsorbent to eliminate contaminants from soil and water systems. Luhua Jiang et al., 2017 explored biochar modification to enhance its ability to adsorb hexavalent chromium from aqueous solution. Ramie stem biomass was pyrolyzed and then treated with beta-cyclodextrin/poly (L-glutamic acid). Sorption performance of CGA-biochar was found to be better than that of pristine biochar.

3.4 Membrane Based

Feyisayo V. Adams et al., 2012 developed nanofiltration membranes by incorporating beta-CD polyurethane into polysulfone (PSf) as a novel mixed matrix composite for water treatment using modified phase inversion technique in order to test selectivity for Cd^{2+} ions. Addition of beta-CD polyurethane in the composite membrane enhanced the thermal stability of the membrane and also improved the hydrophilicity of PSf membranes with 5% addition.

In order to separate Pb(II), Zn(II), and Cu(II) ions from aqueous solutions by means of transport across polymer inclusion membranes (PIM), Cezary A. Kozlowski et al., 2008 synthesized hydrophobic beta-cyclodextrin (beta-CD) polymers by crosslinking beta-CD with 2-(1-docosenyl)succinic anhydride. The linear relation of initial flux values vs. metal ions concentration for Polymer inclusion PIM with beta-CD polymer as a carrier showed "fixed-site" mechanism of transport.

Electrospun nanofibers have been widely used for adsorption to treat wastewater, owing to the presence of various functional groups such as amido, carboxyl, hydroxyl, carbonyl, and amidoxime. Xiao Guan et al., 2019 fabricated Hordein and hordein/ N,N′-methylene bisacrylamide (MBA) nanofibers by electrospinning technique and then further modified it with beta-CD. The as-prepared membranes were then utilized for Cu (II) adsorption.

Cezary A. Kozlowski et al., 2005 developed hydrophobic beta-cyclodextrin (beta-CD) polymers for separation of Cu (II), Co (II), Ni (II) and Zn (II) by transport across polymer inclusion membranes (PIM) via cross-linking reaction of beta-CD with alkenyl (nonenyl and dodecenyl) succinic anhydride derivatives in anhydrous N,N-dimethylformamide in the presence of NaH. The selectivity of PIM process decreased in the order: Cu (II) > Co (II) > Ni (II) > Zn (II). Recovery factor of Cr (VI) was over 80%.

3.5 Unmodified Beta-CD

Md. Shariful Islam, 2016 tested arsenic (As) removal from contaminated water by using alpha-, beta- and gamma-cyclodextrin polymers (CDP). Conventional As removal techniques include coagulation followed by precipitation, membrane separation, ion exchange, phytoremediation and adsorption. However, adsorption is mostly used owing to its favourable economics, low installation and maintenance cost. Alpha-CDP reached maximum As removal of 86% at pH 6.8 within 30 minutes.

For the removal of dyes and Cu ions from aqueous solutions, Yaoyue Li et al., 2019 functionalized a filter paper with beta-cyclodextrin and citric acid via esterification reaction. Filter paper served as a typical cellulose membrane. After modification, the functional groups on the surface of the filter paper could greatly improve the adsorption efficiency of the material.

4. RECYCLABILITY

The removal of heavy metal ions from wastewater can be achieved using beta-cyclodextrin polymers that are crosslinked, organically modified or supported with other polymers. But in order to make them cost-effective, the polymers have to be re-used several times. Reusability serves as one of the significant features of advanced adsorbents for feasible and practical application.

Regeneration studies of several adsorbents revealed that alkalis could serve as efficient desorbing agents for desorption of heavy metals by chemically modified adsorbents. Acids could be used for efficient desorption of bio-adsorbents whereas chelating agents catered to the needs of biomass desorption.

Regeneration studies of several CD polymers revealed the effectiveness of polymers in the removal of contaminants after a number of cycles of use. Repetitive absorption experiments were carried out using the same polymer. Each cycle was conducted by flushing standard pollutants through the polymer-packed column. The removal efficiency was calculated by examining the amount of pollutant present in the filtrate, and the polymer was regenerated by washing with the desired solvent (eluent) and clean water. Polymers were able to maintain their effectiveness even after several cycles. In general, the polymers did disintegrate after several cycles, and hence there was a need to add strengthening components such as nanotubes.

It has been observed that regeneration studies have been carried out by several researchers by utilization of appropriate eluents. For the removal of Ni^{2+} from industrial wastewater, Tingting Yu et al., 2018 synthesized porous polymers (P-CDEC) by joining beta-CD and EDTA modified chitosan (CS-EDTA). Regenerated P-CDEC exhibited almost no decrease in removal efficiency (from 98% to 95% for Ni (II)) and more than 90% of the regeneration efficiencies were retained after the fifth cycle.

Beta-CD-crosslinked polymer micro-column developed by Ying Gu at al., 2015 was reproduced 6 times and its adsorption efficiency for Cr (III)-PAR was found to be greater than 90%.

Amir Abdolmaleki et al., 2015 analyzed the recyclability of T-beta-CD-MNPs for removal of Pb^{2+} ions using HNO_3 solution (0.01 M). It was noticed that even though the removal efficiency towards Pb^{2+} was slightly reduced during the later cycles, it was still found to be higher than 92% in the final cycle.

Cell-g-GMA-Beta-CDN+- type adsorbent had excellent ability of reuse and regeneration. The adsorbent could be reused 5 times and the removal efficiency of Cr (VI) remained the same with 0.5 mg/L sodium hydroxide as the desorption agent. (Yanmei Zhou et al., 2011)

Weiwei Huang et al., 2017 carried out regeneration experiments for Cu^{2+} loaded adsorbents using 0.5 M HCl solution. The adsorbent retained 90.48% of its adsorption capacity toward Cu^{2+} after five cycles. These results demonstrated the good stability and recyclability of CA-beta-CD in practical applications.

In accordance with several studies carried out by researchers it could be interpreted that many adsorbents could be reused effectively after regeneration.

5. FUTURE PROSPECTS

Development of efficient water purification techniques became essential due to the presence of organic and inorganic pollutants which posed a major threat to human health even when present at extremely low concentrations.

Activated carbon is used as an adsorbent in several water purification systems nowadays owing to its ability to capture organic pollutants and heavy metal ions. However, it exhibited low selectivity, regeneration problems, and had slow, non-specific uptake.

In order to overcome the drawbacks of activated carbon, the chemistry of host-guest complexes was utilized. Beta-cyclodextrin (beta-CD), a 7-unit glucose macrocycle with a hydrophilic exterior and relatively hydrophobic interior was rendered capable of complexation with heavy metal ions. Researches showed that beta-CD and its derivatives had higher efficiency in absorbing, separating and detecting heavy metal ions indicating promising applications for removal of heavy metal ions from wastewater. Owing to the high-water solubility of beta-CD, in order to efficiently bind metal ions, it has to be chemically modified by cross-linking beta-CD with cross-linkers or by grafting beta-CD via chemical linkers onto insoluble supports such as carbon nanotubes, plasticized membranes, etc.

Apart from water purification, cyclodextrins find a wide range of applications.

Cyclodextrins can be used to protect vitamins and pharmaceuticals containing easily oxidizable double bonds, such as prostaglandins, against oxidation. They also have the ability to catalyze certain chemical reactions. Styrene sulfonate polymerization was accelerated in the presence of cyclodextrin, and the polymers had a higher molecular weight. Beta-cyclodextrins could be used to produce stationary phase media for HPLC separations. Beta-cyclodextrin and methyl-beta-cyclodextrin could remove cholesterol from cultured cells.

Being derived from starch, beta-CD has high bioavailability and thus it is a good candidate for sustainable, inexpensive chemical adsorbents. Recyclability studies carried out by several researchers revealed that such adsorbents could be reused effectively after regeneration. Hence, beta-CD that was modified by organic adsorbents, crosslinking agents and immobilization onto supports appeared to be highly beneficial for the elimination of heavy metal ions from wastewater and industrial discharge.

6. CONCLUSION

It can be confidently concluded that Beta-CD offers a promising future in the field of water purification. Although the researches are still in the preliminary stage, Beta-CD has showed a huge potential for being replaced by the conventional water purification techniques such as those with activated carbon. A variety of researches are being carried across the globe by prominent scientists and it is only a matter of time that the Beta-CD based adsorbents will be available for commercial use and become a widely preferred choice for the purpose of removal of heavy metal ions from water bodies.

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