

Removal of mercury by adsorption: a review

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Abstract Due to natural and production activities, mercury contamination has become one of the major environmental problems over the world. Mercury contamination is a serious threat to human health. Among the existing technologies available for mercury pollution control, the adsorption process can get excellent separation effects and has been further studied. This review is attempted to cover a wide range of adsorbents that were developed for the removal of mercury from the year 2011. Various adsorbents, including the latest adsorbents, are presented along with highlighting and discussing the key advancements on their preparation, modification technologies, and strategies. By comparing their adsorption capacities, it is evident from the literature survey that some adsorbents have shown excellent potential for the removal of mercury. However, there is still a need to develop novel, efficient adsorbents with low cost, high stability, and easy production and manufacture for practical utility.

Keywords Adsorption · Adsorbents · Mercury · Removal · Properties

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Introduction

Mercury (Hg) is a naturally occurring element. It is widely used in electricity generation and industrial production such as chlor-alkali, plastics, metallurgy, and electronics. The two main oxidation states of Hg are +1 [Hg(I)] and +2 [Hg(II)] (Pervukhina et al. 1999). Mercury sulfide (HgS) is clinical used in Chinese and Indian traditional drugs, such as cinnabar and “Rasasindura,” which might have little or no therapeutic effects because it may not react with any cell receptors due to its insolubility in aqueous solution (Kamath et al. 2012). Mercury chloride (HgCl₂) was widely used in producing polyvinyl chloride. A higher oxidation state for Hg is +4 [Hg(IV)], which has been detected only under exceptional conditions, e.g., Hg(IV) fluoride (HgF₄) (Wang et al. 2007). Another highly toxic form of Hg(II), methylmercury [MeHg, (CH₃Hg⁺)X[−]], is a bioaccumulative environmental toxicant which can be enriched in fish, shellfish, and animals that eat fish (Choi et al. 2008).

Due to its volatility, persistence, and bioaccumulation, Hg has been considered as one of the most toxic metals which can affect the health of human beings. Long-term exposure to large amounts of Hg would harm the human brain, heart, kidneys, lungs, and even the immune system (Hsiao et al. 2011). Exposure to Hg in the womb or fed with seafood containing MeHg may harm the developing nervous system of unborn babies and young children and reduce their IQs (Yau et al. 2014). Elemental (metallic) Hg [Hg⁰] adsorbed through the lungs can cause health effects such as tremors, muscle atrophy, headaches, and so on when it is breathed as a vapor. However, the amount of Hg discharged into the environment is increasing continuously. Carbonaceous fuel combustion such as coal-fired plants and incinerators has contributed a lot of the current global Hg emissions. Up to now, Hg can be found in water, rocks, and soil. Additionally, trace amounts of

Hg can even be found in the air (Heaven et al. 2000; Xue et al. 2013). High Hg concentrations in the range of Hg-associated neurochemical effects have been found in the brain tissue of Arctic toothed whales (Kamath et al. 2012).

The adverse effects from food and water mercury contamination are difficult to measure. However, over 100,000 kg of Hg was generated by human activities and discharged into the environment annually. Most of the Hg in both gaseous and liquid forms is from the coal-fired power industry (Singh et al. 2011). and Hg^0 removal from flue gas has a tremendous effect on human daily life (Zheng et al. 2012). Hg^0 , HgCl_2 , which is also called Hg(II), and MeHg have already made their way into the overall environment (Henneberry et al. 2011) and have brought great harm to the environment and human health.

Due to its bioaccumulative properties and potential risk to human health, the Hg-containing wastewater and flue gas have attracted a great global concern. Therefore, efficient methods must be developed to remove them from the environment (Liu et al. 2014d). Adsorption separation is one of the most facile, convenient, and practical technologies among the existing Hg control technologies. In the past 4 years, we have seen significant progress of Hg removal in adsorptive separation technology and materials science. The latest developed adsorbents for Hg removal are reviewed in detail in this paper.

Adsorbent development

Carbon-based adsorbents

Activated carbons (ACs)

Due to its very big internal surface (between 500 and 1500 $\text{m}^2 \text{g}^{-1}$), AC is produced as an ideal adsorbent for removal of contaminants. AC has also been used for Hg removal for years by the researchers. Both the pore structure and chemical property of AC would affect its adsorption properties (Asasian et al. 2012; Lu et al. 2014; Zabihi et al. 2011). and the chemical modification seems to be one of the most efficient ways to improve its adsorption efficiencies (Asasian and Kaghazchi 2013a; Kim et al. 2011; Wahby et al. 2011). For example, incorporation of sulfur group to an adsorbent could improve its Hg adsorption capacities (Asasian and Kaghazchi 2013b; Hsi and Chen 2012; Morris et al. 2012). Derived from the pyrolysis, activation, and sulfurization of waste tires, the prepared AC possessed an adsorptive capacity of 5.236 $\mu\text{g g}^{-1}$ for vapor-phase HgCl_2 (Ie et al. 2012). If ACs were impregnated with several sulfur species such as elemental sulfur (S^0), aqueous-phase sodium sulfide (Na_2S), or hydrogen sulfide (H_2S), various sulfur forms such as sulfide, sulfoxide, elemental sulfur, sulfone, thiophene, and sulfate would be introduced (Ie et al. 2013). In comparison with the unmodified ACs, these

sulfur-impregnated ACs exhibited higher Hg adsorption capacities (Feng et al. 2006a, b; Li et al. 2013c). Besides the sulfurization methods mentioned above, treatment of ACs by aqueous solutions of Na_2S and H_2SO_4 is also feasible (Abdelouahab-Reddam et al. 2014). Also, the newly emerged interactions between the introduced -S- group and Hg(II) are considered to favor the high adsorption efficiencies of modified ACs (Fig. 1).

The physical properties of the adsorbents such as specific surface area, pore volume, pore size, and pore number could affect their adsorption capacities. However, sulfurization of AC will reduce the number of micropores, which would restrain its adsorption capacity. To retain the porous structure of AC after modification, simultaneous sulfurization/activation of ACs was developed, and the modified ACs showed an increased Hg(II) adsorption of 1637 $\text{m}^2 \text{g}^{-1}$ due to the higher specific surface area (ShamsiJazeyi and Kaghazchi 2014).

Besides the sulfur-containing functional groups, the introduction of other functional groups such as $-\text{NH}_2$, $-\text{NHCOCH}_3$, $-\text{Br}$, $-\text{Cl}$, $-\text{I}$, $-\text{C}=\text{N}$, $-\text{OH}$, and so on onto the surface of ACs is also responsible for Hg(II) adsorption (Hsi et al. 2013; Reddy et al. 2014). Various impregnating substances such as NaI, CuCl_2 , CuBr_2 , and FeCl_3 were applied to graft halogen functional groups onto the adsorbents (Zhang et al. 2012b). The Hg(II) removal efficiencies increased in the order of $\text{AC} < \text{Cl-impregnated AC} < \text{Br-impregnated AC} < \text{I-impregnated AC}$, indicating that the electron-donating ability of the introduced functional groups may help increase the adsorption capacities of the adsorbents (De et al. 2013).

It is really easy to collect magnetic materials by providing a magnetic environment, and magnetic AC (MAC) has found its application prospect in the adsorption field (Faulconer et al. 2012). However, as-prepared MAC is limited in practical use for removal of Hg(II) due to its poor gradient magnetic separation properties (Okamoto et al. 2011). The introduction of functional groups onto MAC was carried out, and the adsorption capacities of two kinds of modified MACs, oxidized MAC (ox-MAC) and thiol radical decorated MAC (SH-MAC), were improved (Miura and Tachibana 2014).

The resource consumption for production of ACs is huge, and the production may produce waste containing pollution to the environment at the same time. The synthesis of ACs from biomass materials through more straightforward, more

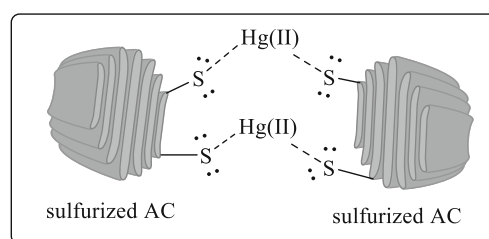


Fig. 1 The possible interactions between sulfurized AC and Hg(II)

economically viable, and greener approach is expected. Coconut shell was used to produce AC (Hu et al. 2014), and the adsorption capacity could also be improved by impregnating it with sulfur (Hsi et al. 2014).

Activated carbon fibers (ACFs)

Due to their larger surface area and lighter than that of ACs, ACFs are considered as efficient and alternative adsorbents for removal of pollution or contaminants to achieve environmental remediation. However, the comprehensive research on ACFs in removing Hg was less. Recently, dimethylsulfoxide (DMSO), disodium tetrasulfide (Na_2S_4), sodium hydrosulfide (NaSH), and sulfur vapor were used as sulfur sources to modify ACFs, and the prepared sulfur-impregnated ACFs (ACFs-SH) showed higher Hg adsorption capacities than that of pristine ACFs, demonstrating once again that sulfur impregnation is beneficial to increasing the Hg removal capacity of the adsorbents (Yao et al. 2014b).

Mesoporous carbons (MCs)

Due to its porous structure and high specific surface area, MC has been widely applied in adsorption and removal of metal ions from aqueous solutions. To maintain the high surface area and mesoporous structure of hexagonal ordered mesoporous carbon (OMC) CMK-3 and introduce functional groups, 2-amino-5-mercapto-1,3,4-thiadiazole (AMT) groups were used as the modification material, and the prepared AMT-OCMK-3 showed high adsorption capacity of 450.45 mg g^{-1} for Hg(II) (Anbia and Dehghan 2014).

The utilization of zinc oxide (ZnO) or copper (Cu) nanoparticles as efficient adsorbents for the removal of metal ions is considered viable. The diffusion or accumulation of metal ions onto the surface of ZnO or Cu nanoparticles has made important contributions to the adsorption. The ZnO-modified CMK-3 was found to adsorb Hg(II) with high removal efficiency (Zolfaghari et al. 2011). Taking advantage of the chemical affinity between Cu particles and Hg, Cu-coated porous carbon (Cu/PC) was developed for removal of Hg^0 , and the surface oxidation level of Cu particles and total Cu content could affect the Hg^0 removal efficiencies (Kim et al. 2012).

Carbon nanotubes (CNTs)

Due to the large particle size of the conventional adsorbents, the massive mass transport resistance reduces their adsorption capacities. To overcome this limitation, the use of nanoscale materials as adsorbents has attracted considerable attention in recent years (Habuda-Stanic and Nujic 2015; Yao et al. 2014a). Owing to their extremely small size and large specific surface area, nanoscale adsorbents have shown many unusual physical and chemical properties. The unique properties

include huge specific surface area, strong adsorption ability, high stability, and high mechanical strength. Due to these outstanding characteristics, various carbon-based nanoparticles (CBNPs) including CNTs and graphene have been used for the removal and adsorption of heavy metal ions (Yu et al. 2014c, 2015).

The introduction of functional groups onto CNTs could enhance their adsorption capacities (Yu et al. 2014b). For example, chitosan (CTS)-functionalized CNT beads, including oxidized multiwalled CNT (ox-MWCNT)-impregnated CTS beads, MWCNT-impregnated CTS beads, and single-walled CNT-impregnated CTS beads, all exhibited higher adsorption capacity toward Hg(II) than that of pristine CNTs (Shawky et al. 2012). Amino- and thiol-functionalized MWCNTs exhibited high adsorption capability for Hg(II) due to the introduction of the electron-donating groups (Hadavifar et al. 2014; Shadbad et al. 2011), and thiol-containing SWCNT (SWCNT-SH) showed threefold higher adsorption capacity than that of pristine SWCNT and fourfold higher adsorption capacity than that of AC (Bandaru et al. 2013).

Graphene and graphene oxide (GO)

As a new member of carbon-based nanomaterials, GO has attracted great attention in the adsorption field due to its huge specific surface area and the plentiful surface oxygen functional groups (Yu et al. 2015). To improve its adsorption performance, the introduction of CTS onto GO was carried out, and the CTS-nanofilled GO (GO/CTS) showed a high maximum adsorption capacity of 381 mg g^{-1} than that (187 mg g^{-1}) of the as-prepared GO (Kyzas et al. 2014). Sulfur/reduced graphene oxide (SRGO) nanohybrid also demonstrated a fast and efficient Hg(II) removal and excellent reusability (Thakur et al. 2013).

Because of the low production cost, abundant functional groups, diverse shapes and structures, strong adsorption ability, and easy processability, conducting polymers have a wide range of attractive applications in adsorption. The integration of conducting polymers with graphene might be an effective way for developing novel adsorbents. Combined with the advantages of both polyaniline (PANI) and reduced graphene oxide (RGO), the built PANI-RGO composites showed a tremendous adsorption capacity of 1000 mg g^{-1} for Hg(II), which was much higher than that (515.46 mg g^{-1}) of PANI (Li et al. 2013b).

Magnetic materials

Iron oxides

Iron oxides, such as maghemite and magnetite, possess some good characteristics such as low toxicity and price, high specific surface area, being magnetic, and easy modification of

surface. Taking advantage of the high affinity of the sulfur-containing functional groups for Hg(II), the dithiocarbamate group-modified iron magnetite particles were developed as a new adsorbent for removal of Hg(II) (Figueira et al. 2011).

The assembly of multifunctional materials has attracted increasing interest in recent years, and the sterically stabilized core-shell nanoparticles could offer some significant advantages for adsorption due to the extended hydrophilic shells. Using superparamagnetic Fe₃O₄ as the core and hydrophilic silica molecular sieve as the shell, the core-shell structured Fe₃O₄@SiO₂ nanocomposites exhibited high selectivity toward Hg(II) (Jing-po et al. 2014; Tavares et al. 2013) and so did the thiol group-modified Fe₃O₄@SiO₂ (Fe₃O₄@SiO₂-SH) nanoparticles (Hakami et al. 2012; Zhang et al. 2013), pyrene-modified Fe₃O₄@SiO₂ nanoparticles (Li et al. 2012a), and mercapto group-modified core-shell structured nanomagnetic Fe₃O₄ polymers (SH-Fe₃O₄-NMPs) (Pan et al. 2012a, b).

Although the modification often benefits the adsorption, multifunctionalization is sometimes redundant and absolutely useless due to the reduced specific surface area caused by the grafted components. For example, CTS-crosslinked magnetic Fe₃O₄ (CTS-Fe₃O₄) nanoparticles possessed higher adsorption capacity for Hg(II) than that of the raw nanoparticles (Katok et al. 2013) and so did 2-mercaptobenzothiazole-modified magnetic Fe₃O₄ (2-MBTZ-Fe₃O₄) (Parham et al. 2012). However, the adsorption capacity of phenylthiourea (PTU) resin-modified CTS-Fe₃O₄ (CTS-Fe₃O₄-PTU) is greatly decreased (Monier and Abdel-Latif 2012).

To obtain magnetic adsorbents with high efficiency, special efforts were devoted to constructing doped materials. Recently, Zn_{0.46}Fe_{2.54}O₄, a novel Zn-doped biomagnetite nanostructured material, was developed. For its easy operation in aqueous separation process, the 3-mercaptopropyl trimethoxysilane (MPTMS)-modified Zn_{0.46}Fe_{2.54}O₄ has prospects for Hg(II) removal in real applications due to its high adsorption capacity of 416 mg g⁻¹ for Hg(II) (He et al. 2012).

Cobalt ferrite

Cobalt ferrite (CoFe₂O₄), another kind of magnetic material, can be facilely recovered and reused. Taking advantage of the excellent characteristics of CoFe₂O₄ and RGO, magnetic cobalt ferrite (CoFe₂O₄-RGO) nanocomposite was developed. Also, the high adsorption capacity of 157.9 mg g⁻¹ makes it more attractive than other magnetic adsorbents (Zhang et al. 2014b).

Nonmetal and other metal adsorbents

Metals and metal oxides

Due to its structural and electronic properties, gold (Au) is in favor of Hg adsorption (Lim et al. 2012). Colloidal gold

nanoparticles (Au NPs) have been employed for the rapid removal of Hg(II) from aqueous solutions (Ojea-Jimenez et al. 2012). However, the high price of Au-related materials limits their potential practical applications. Therefore, adding other cheap materials to Au adsorbents might be a good choice. For example, aluminum oxide (Al₂O₃)-mixed Au NPs exhibited relatively low price besides its high removal efficiency (Lo et al. 2012).

Cheap adsorbents have gotten more and more attention in recent years. Due to the excellent hydrothermal and mechanical stability, copper oxide (CuO), copper chloride (CuCl₂), and carrier (such as neutral Al₂O₃, artificial zeolite, and AC)-impregnated CuCl₂ were used as adsorbents for the removal of Hg⁰ (Du et al. 2014). Hierarchical mesoporous CuO nanoflakes (HCMP-CuO-NFs) showed an enormous adsorption capacity of 1767.97 mg g⁻¹ for Hg(II) due to their very huge specific surface area (9.72 m² g) and total pore volume (mesopores with pore sizes of 9.35 nm; 0.023 cm³ g) (Kumar et al. 2014).

Material nanocrystallization is also beneficial in the increase in the adsorbents' specific surface areas and adsorption capacities. For example, zinc oxide (ZnO) nanoparticles were used for Hg(II) removal with a maximum adsorption capacity of 714 mg g⁻¹ (Sheela et al. 2012). TiO₂ nanocrystals could be employed as efficient adsorbents for both Hg⁰ (Tsai et al. 2011) and Hg(II) removal (Dou and Chen 2011; Ghasemi et al. 2012).

Mixed metal oxides, metal-impregnated oxides, and metal salts

Besides specific surface area and particle size, the affinity of an adsorbent for a solute is also crucial. A synergistic adsorption effect would be formed by constructing mixing metal oxides, metal-impregnated oxides, and metal salts, which provides a new way for making novel adsorbents. An environmentally benign chemical, potassium ferrate (K₂FeO₄), has given a convincing result in removing heavy metals from water and wastewater through oxidation and coagulation. The mixing of sodium chlorite (NaClO₂) and K₂FeO₄ solution would increase its removal efficiency for Hg⁰ (Zhang et al. 2014c). Hydrotalcite-type compounds/layered double hydroxides and their chemically modified derivatives such as (Mg_{3-x}M_x)AlCO₃ (M = Cu or Zn) were developed for Hg removal under medium high temperature conditions, providing an access to design novel high efficient adsorbents (Yu et al. 2014a).

A hybrid of different materials can help to improve their adsorption performance. For example, Hg adsorption on pure SnO₂ particles could be negligible, and Hg adsorption on MnO_x would also decrease significantly with an increase of the contact temperature. A newly developed binary metal oxide, Sn-Mn oxide, showed high Hg adsorption capacity at not

only low temperature but also high temperature (Xie et al. 2014). A novel Ce-Ti adsorbent, CeO₂ supported on TiO₂, also exhibited higher Hg⁰ removal efficiency than that of sulfur-impregnated AC (Hou et al. 2014; Zhou et al. 2013a). By incorporation of Ce-Mn into Ti-pillared-clay (Ti-PILC), the prepared Ce-Mn/Ti-PILC was an excellent Hg⁰ adsorbent (He et al. 2014). Despite the lower specific surface areas and total pore volumes after metal loading, the Hg⁰ adsorption capacities of Cu/AC or Ni/AC hybrid materials were also higher than those of the as-prepared material, and the high activity of nickel was thought to be beneficial in Hg⁰ adsorption (Bae et al. 2014).

Nonmetals, nonmetal oxides, and salts

Due to its excellent thermal and mechanical stability, silica gel is always used as a supporter material for chelating adsorbents. By introducing organic moieties which own some active donor atoms or groups such as O, N, and S onto the skeleton of silica gel, the modified solid silica gel would possess many extraordinary adsorption properties for Hg. For example, sulfur-functionalized silica materials exhibited higher adsorption capacity for Hg(II) than that of pristine silica without sulfur groups (Arsuaga et al. 2014; Saman et al. 2014). Using bis(triethoxysilylpropyl) tetrasulfide (BTESPT) and MPTMS as sulfur ligands, the sulfur-functionalized silica gels (BTESPT-SGs and MPTMS-SGs) showed increased adsorption capacities (Johari et al. 2014a, b). Therefore, it is worthwhile to construct a chelating matrix by introduction of multifunctional organic molecules onto the adsorbents. 4-Amino-5-methyl-1,2,4-triazole-3(4H)-thion-modified silica aerogel exhibited a maximum adsorption capacity of 142.85 mg g⁻¹, which was eight times more than that (17.24 mg g⁻¹) of silica aerogel (Tadayon et al. 2013). Also, 1-(pyridin-3-yl) imine-modified silica gel also exhibited high adsorption capacities due to the bidentate ligand of N atoms (Fig. 2) (Toubi et al. 2013). A novel chelating resin silica gel supported by aminophosphonic acids also possessed high adsorption capacity (Wang et al. 2012) and so did the vinyl imidazole (VIM)-modified silica (Sun et al. 2013). Thiol, amino, and amide groups could be simultaneously introduced onto silica by reacting SBA-15 with L-cysteine (L-cys), and the L-cys-SBA-15 nanocomposite (Fig. 3) showed higher Hg adsorption capacity and faster adsorption rate (Li et al. 2014). In addition, polymer grafting modification, hybrid processing, and nanocrystallization are all beneficial to improving the adsorption capacities of the silica-based adsorbents (Perez-Quintanilla and Sierra 2014). For example, poly(aniline-co-o-aminophenol) (PAOA)/mesoporous silica SBA-15 (PAOA/SBA-15) exhibited a maximum adsorption capacity of over 400 mg g⁻¹ toward Hg(II) (Tang et al. 2011).

Due to its tunable mesopore sizes and high surface area, a mesoporous molecular sieve was used as a promising

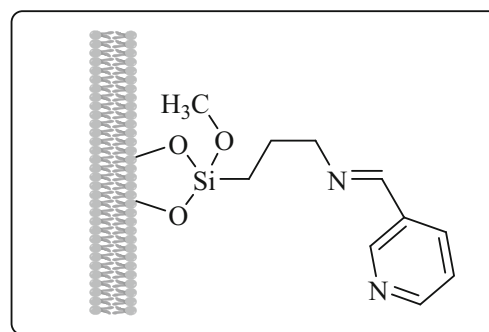


Fig. 2 The schematic drawing of 1-(pyridin-3-yl) imine-modified silica gel

adsorbent. Mesoporous silica (MS) is an ideal adsorbent for Hg removal because of its high surface area and biocompatibility. Owing to its hierarchical structure and hexagonal arrangement of one-dimensional mesopores, Mobil composition of matter No. 41 (MCM-41), a mesoporous or nanoporous silica, has been widely used in Hg(II) removal (Aboufazeli et al. 2013). Its adsorption capability could also be improved by ZnCl₂ modification (ZnCl₂-MCM-41) (Raji and Pakizeh 2013, 2014). The chelation between the -Cl groups of ZnCl₂-MCM-41 and Hg(II) species (Fig. 4) might make great contributions toward adsorption (Raji and Pakizeh 2014).

Besides affecting the adsorption efficiency, surface modification also affects the selectivity of the adsorbents. MPTMS, 3-aminopropyl-trimethoxysilane (APTES), and n-(2-aminoethyl)-3-aminopropyl-trimethoxysilane (AEAPTMS)-modified diatom silica microparticles exhibited higher maximum adsorption capacities of 185.2, 131.7, and 169.5 mg g⁻¹, respectively (Yu et al. 2012a). Amidoxime (AO) group-modified silica gel showed selective adsorption ability for Hg(II) (Chen et al. 2012). Also, 1-(3-carboxyphenyl)-2-thiourea (CPTU)-modified mesoporous silica showed both high efficiency and excellent selectivity for Hg(II) (Zhang et al. 2012c). Recently, a regenerable and highly selective core-shell structure composite, thymine (T) and T-rich DNA-functionalized magnetic mesoporous silica (Fe₃O₄@nSiO₂@mSiO₂-T-TRDNA), was developed for the simultaneous detection and removal of Hg(II) (He et al.

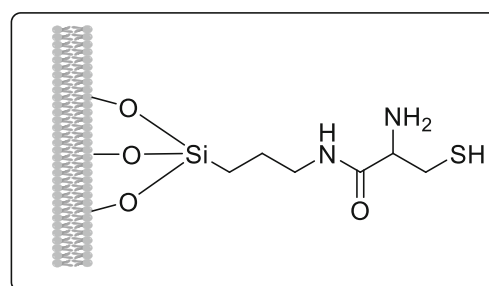


Fig. 3 The schematic drawing of L-cysteine-functionalized mesoporous silica

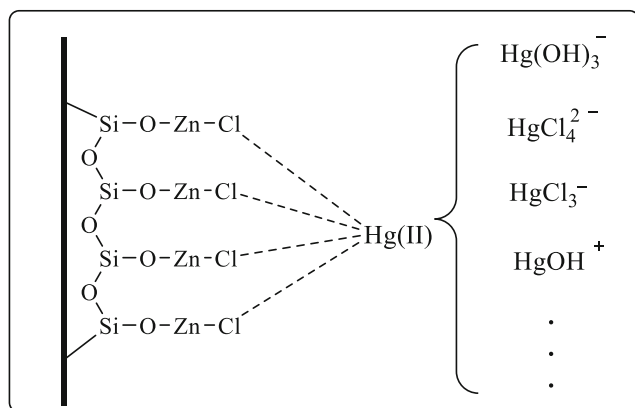


Fig. 4 Proposed interactions between Hg(II) species and ZnCl₂-MCM-41

2013). Through forming ion-pair interactions with Hg(II), quaternary ammonium-functionalized magnetic mesoporous silica exhibited fast Hg(II) removal rate (Liu and Du 2011). Such a rapid demercuration with high Hg(II) removal efficiency provided some new ideas for constructing hybrid nanostructures for efficient Hg removal.

Polymers

Conventional polymers

Polymeric materials have been used as one of the most promising and effective adsorbents for the removal of toxic metals from wastewater, and their application in Hg removal is becoming increasingly important. Polyacrylonitrile (PAN) is an important polymer in the adsorption field. However, PAN fibers exhibited low adsorption capacity for the removal of heavy metals, including Hg(II) and Hg⁰. A new chelating resin, PAN-2-aminothiazole (PAN-AT), possessed higher adsorption capability for Hg(II) (Xiong et al. 2013) and so did hydroxylamine hydrochloride (NH₂OH·HCl, AO·HCl)-modified PAN (Zong et al. 2012). Recently, a copolymer prepared from bromoacetyl bromide (BrCOCH₂Br)-modified corn starch (CS-Br), PAN, and AO (AO PAN-CS-Br) exhibited an enormous maximum adsorption capacity of 826.43 mg g⁻¹ for Hg(II) (Xu et al. 2014). Owing to the unlimited efforts made by the researchers, PAN-based resins have already shown great potential in Hg adsorption and removal technologies.

Due to its plenty of imine and amine groups that can chelate cationic metal ions, PANI was widely used as an efficient adsorbent for adsorption and removal of heavy metals. Humic acid (HA)-grafted PANI (HA-PANI) showed a high adsorption capacity for Hg(II) (Li et al. 2011a), and the introduction of HA changed the morphology of PANI and prevented its aggregation; therefore, the adsorption properties of modified PANI were improved.

Diethylenetriamine-bridged polysilsesquioxanes (DETA-b-PMPSQs) showed higher adsorption capacity for Hg(II) than as-prepared PMPSQs, indicating once again that electron donor atoms are beneficial for Hg(II) adsorption (Sun et al. 2014a). In addition, selective adsorption could be realized by grafting special functional groups onto polymer adsorbents. For example, thiol-functionalized PMPSQs (PMPSQs-SH) could selectively adsorb Hg(II) from binary ion systems in the presence of some coexistent ions such as Mn(II), Cu(II), Pb(II), Co(II), and Ni(II) (Niu et al. 2014) and so did 3-amino-5-methylisoxazole (AMI)-modified chloromethylated polystyrene (PS) beads (Xiong et al. 2012) and 2-pyridinethiol-grafted PS (Tan et al. 2012). Modification of chelating PS-ethylene diamine tetraacetic acid (EDTA) resins using metal (Fe, Al, and Zr) oxides could increase the adsorption efficiencies, accelerate the adsorption rate, and improve the adsorption selectivity toward Hg(II) (Li et al. 2012b). Moreover, PS-AMI also exhibited high maximum adsorption capacity for Hg(II) at 298 K. A novel chelating resin, which was based on chloromethylated copolymer of styrene-divinylbenzene (CMPS) with dicyandiamide (DCDA), exhibited an enormous adsorption capacity of up to 1077 mg g⁻¹ due to its cyanoguanidine moiety (Ma et al. 2011). A higher maximum adsorption capacity of 1163.8 mg g⁻¹ was obtained by bis-8-oxyquinoline-terminated open-chain crown ether-modified PS (Sun et al. 2012).

One of the most common synthetic polymers, poly(ethylene terephthalate) (PET), does not have adsorption ability toward anions or cations. However, a newly developed thiosemicarbazide-modified PET could remove Hg(II) from water quickly and efficiently, indicating that certain desirable properties could be introduced to PET by further functionalization (Monier and Abdel-Latif 2013).

A novel photo-crosslinkable nanofiber, uracil-functionalized poly[1-(4-vinylbenzyl uracil)] (PVBu), was converted into a covalent network nanofiber by exposure to UV light. This PVBu nanofiber could selectively remove Hg(II) with a high maximum adsorption capacity (Wang et al. 2013b). Due to the introduced amine functional groups, acrylic acid copolymerized polypropylene fiber, a newly developed chelating fiber, showed an astounding high adsorption capacity of 1144.86 mg g⁻¹ for Hg(II) (Ma et al. 2012).

Selectivity is always a challenge in adsorption technologies. 4-Vinyl pyridine-modified poly(ethylene terephthalate) (4-VP-PET) fiber exhibited high adsorption selectivity for Hg(II) (Bozkaya et al. 2012). Due to the adsorption for selectivity of bentonite (BT) toward Hg(II), CTS-poly(vinyl alcohol)/BT (CTS-PVA/BT) nanocomposites possessed excellent adsorption selectivity for Hg(II) (Wang et al. 2014c). Selective adsorption of Hg(II) could also be carried out by hydrolyzed acrylamide (AAM)-grafted polyethylene terephthalate (PET) (AAM-PET) films (Rahman et al. 2014). three-dimensional structured CTS-PVA hydrogel (Wang et al. 2013a), thymine

(T)-grafted PVA-polyethyleneimine (PEI) (T-g-PVA-PEI) nanofibers (Huang et al. 2014). and sulfonated crosslinked PEI CPEI (SCPEI) (Saad et al. 2012).

Dendrimers

Hyperbranched polymers, named dendrimers, have received significant attention for Hg removal due to their terminal surface functional groups and specific structures in contrast with the conventional linear polymers. Poly(amidoamine) (PAMAM) dendrimers, a family of highly branched well-defined polymers, have been widely used in sewage treatment (Sun et al. 2011a, 2014b). Due to their unique topological structures and their surface amino groups which are especially apt to bind metal ions, chitosan (CTS)-functionalized PAMAM dendrimers of generation 1.0, 2.0, and 3.0 showed high maximum adsorption capacities of 398.41, 259.07, and 259.07 mg g⁻¹ for Hg(II) at 35 °C, respectively (Ma et al. 2009).

Biomass materials

The removal of heavy metals by natural adsorbents, especially biomass materials, has become a hot area of research for several years. To develop efficient adsorbents with low cost, strong affinity, and high adsorption capacity and are ecofriendly, biomass materials such as bamboo strips (Siddiqui et al. 2011), starch (Huang et al. 2011), rambai tree leaves (Sen et al. 2011), cellulose (Zhou et al. 2013b), Lemna (Li et al. 2011b), lignin (Lv et al. 2012), kenaf fibers (Macias-Garcia et al. 2012), bamboo charcoal (Tan et al. 2012), guar gum (GG), cork stoppers (CSTs) (Lopes et al. 2014), CTS, lignocellulosic fiber (LCF) (Johari et al. 2014c), and polysaccharide were used. Other agricultural by-products such as almond shell (Khaloo et al. 2012), mandarin peel (Husein 2013), buckwheat hulls (Wang et al. 2013c), wheat straw (Zhang et al. 2014a), barley husk (Naser-Azizi et al. 2013), rice husk ash (RHA) (Ghorbani et al. 2011), banana stem (BS) (Mullassery et al. 2014), rice husk (Rocha et al. 2013, 2014b), mulberry twig chars (Shu et al. 2013), Sargassum muticum algal (Carro et al. 2013), adula leaves (ALP) (Aslam et al. 2013), and rice straw (Song et al. 2013) are all good choices. For example, rice husks were extremely efficient in the removal of Hg(II) with lower concentrations (less than 500 µg L⁻¹) from river water (Rocha et al. 2014a, b), and potassium permanganate (KMnO₄)-modified calcium (CaO)/rice husk could effectively remove Hg⁰ (Zhao et al. 2012).

Being associated with ion-dipole interactions which are stronger than hydrogen bonding, the hydroxyl, carboxyl, and methoxyl groups presented in alkali lignin (AL) are very important functional groups (Lopes et al. 2014). Therefore, lignin has been widely used as a promising material for adsorption and removal of Hg(II). The mercapto-functionalized

alkali lignin (AL-SH) showed more excellent affinity and higher adsorption capability for Hg(II) than that of AL (Zhou et al. 2014).

Besides improving their stability, chemical modification is also beneficial to increase the removal efficiency of biomass-based adsorbents due to the introduced additional useful functional groups (Kushwaha and Sudhakar 2011; Thakur et al. 2014). Amine and thiol moieties cellulose (Donia et al. 2014), mercapto-grafted rice straw (RS-GM) (Song et al. 2013), glycidyl methacrylate (GMA) and diethylenetriamine-modified cellulose (Donia et al. 2013), and formaldehyde polymerized BS (FPBS) (Mullassery et al. 2014) all exhibited higher maximum adsorption capacity for Hg(II) than that of the pristine adsorbents.

Recycled solid waste materials

Due to their porous structure and large specific surface area, recent researches have shown that some recycled solid waste materials such as sawdust (SD) (Hashem et al. 2011), fly ash (Liu et al. 2013; Song et al. 2014), sewage sludge (Liu et al. 2014a), mussel shell ash (Seco-Reigosa et al. 2013), lignite semi-coke (SC) (Zhang et al. 2012a), malt spent rootlets (MSR) (Anagnostopoulos et al. 2012), and even animal manure (Zhang et al. 2014b) can be used in sewage treatment, especially in heavy metal adsorption and removal. In general, the major influence factors for the Hg adsorption capability of fly ash are its average pore size, followed by its surface area. The application of solid waste materials in the area of Hg(II) adsorption could not only fulfill the production of novel adsorbents but could also realize the use of waste and save useful natural resources.

Halogen functional groups have a special and strong affinity for Hg. By halogenation of fly ashes with different agents such as calcium chloride (CaCl₂), HBr, and calcium bromide (CaBr₂) and HBr, the treated fly ash showed greater Hg adsorption efficiency 6.7 times than that of unmodified fly ash (Song et al. 2014). Specially, selective adsorption of Hg⁰, particulate Hg (p-Hg), and Hg(II) with higher efficiency was realized by hydrogen bromide (HBr)-functionalized fly ash (Song et al. 2014; Zhong et al. 2014). In addition, the incompletely burned carbon (UBC) present in fly ash is also an important factor for Hg removal due to its huge specific surface area which is similar to that of graphene (Xu et al. 2013). The predicated high adsorption energy for Hg on the simulated UBC surface is close to 289 kJ/mol, and the adsorption might occur as a chemical process (Li and Maroto-Valer 2012).

Although untreated synthetic zeolite (Na-X and Na-P1) derived from coal fly ash had negligible Hg adsorption capacity, its adsorption capacity for Hg⁰ could be markedly improved after impregnation with silver (Wdowin et al. 2014) or zeolite A precursors (Liu et al. 2013). A waste mixture

containing shell ash, sewage sludge, and wood ash exhibited higher adsorption capacity than that of any single one, indicating that the mixture could be employed as a cheap and efficient adsorbent (Pena-Rodriguez et al. 2013; Seco-Reigosa et al. 2013).

Bacterial species

Due to the presence of various organic and inorganic complexing ligands on the surface, cells may bind with Hg species to realize adsorption. The adsorption of Hg(II), HgCl(OH), and Hg(OH)₂ using nonmetabolizing cells of *Bacillus subtilis*, *Shewanella oneidensis* MR-1, and *Geobacter sulfurreducens* in both chloride-free and chloride-bearing systems was achieved (Dunham-Cheatham et al. 2014). *Paecilomyces catenulannulatus* also exhibited high adsorption capacity for Hg(II) (Li et al. 2013a).

Due to the bonding interactions between Hg(II) and the thiol functional groups on the surface of cells, Hg(II) adsorption and methylation on the cell surface of *G. sulfurreducens* PCA were feasible (Hu et al. 2013). Also, high MeHg removal efficiency was possible by a *Pseudomonas putida* V1 strain (Cabral et al. 2013). Due to its higher adsorption capacity for Hg(II), *Bacillus mucilaginosus* might be a promising biosorbent in Hg species removal (Mo and Lian 2011).

Three-dimensional structures

Hydrogel

Polymer-crosslinked chains are formed through several intermolecular forces such as physical, ionic, or covalent interactions, and the prepared hydrogel could be used as a highly efficient adsorbent for Hg species (Elisseff 2008). Besides chelation, electrostatic interaction also plays an important role in the adsorption of Hg(II). Due to its three-dimensional network structure and plenty of functional groups such as -NH₂, -NHCOCH₃, -C=N, and -OH, CTS/poly(vinyl alcohol) (CTS-PVA), a newly developed hydrogel, has shown superior adsorption properties for Hg(II) (Wang et al. 2014b). Its adsorption capacities reached up to 697.70 mg g⁻¹ at 20 °C and 950.62 mg g⁻¹ at 60 °C, respectively. A newly developed hydrogel, CTS-grafted polyacrylamide (PAA) (CTS-g-PAA) semi-interpenetrating polymer network (IPN) hydrogel, exhibiting extremely higher maximum adsorption capacity of 2001.8 mg g⁻¹ toward Hg(II) than that of any existing adsorbents, is miraculous (Saber-Samandari and Gazi 2013). Specially, CTS-g-PAA could be regenerated without losing its original activity and stability.

Cage structures

Cage structures are also attractive because polyhedral cage-based frameworks could provide strong adsorption interactions for metal ions. Besides its high selectivity for Hg(II) and MeHg, the thiol-rich polyhedral oligomeric silsesquioxane (POSS-SH) also possessed high adsorption capacities (Wang et al. 2014a). Meanwhile, the cage type and adsorption sites can affect the adsorption performance of cage molecules. Due to its cage geometry, 1,4,7,10,13,16,21,24-octaazabicyclo[8.8.8]hexacosane [H₆Aza222] (Fig. 5) could be used for selective binding of Hg (Redko et al. 2006). Polymeric material-grafted H₆Aza222 showed both higher removal efficiency toward Hg(II) (e.g., HgCl₂ or Na₂HgI₄) and more easily regenerable properties (Redko et al. 2014). The surface nanocrystallization of H₆Aza222 polymer would increase its selectivity and affinity for Hg; therefore, it could be used as a regenerable ultrahigh affinity adsorbent (Tadayon et al. 2013).

Other materials

Zeolites

Zeolite Y, β-zeolite, and mordenite were used for Hg(II) removal, and the adsorption capacity of β-zeolite was found to be the highest (Murthy et al. 2013). Encapsulating superparamagnetic Fe₃O₄ nanocrystals into a hierarchical mesoporous zeolite and the magnetic self-assembled zeolite clusters showed adsorption selectivity toward Hg(II) (Yin et al. 2012).

Clays

Natural clay is mainly composed of silica, alumina, iron, calcium, and magnesium oxides (Guerra et al. 2012). As a type of natural layered material with abundant mesopores, clay possesses good thermal stability and high specific surface area, making them useful adsorbents for the removal of heavy metals from wastewater (Kumar et al. 2012). Naturally, the Hg(II) adsorption capacity of black soil was greater than that

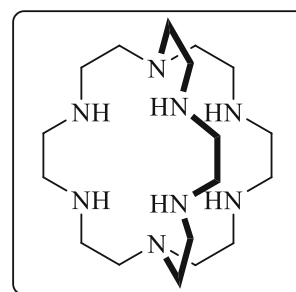


Fig. 5 Chemical structure of H₆Aza222

of red soil, and the concentration of an additive, chloride ion (Cl^-), had a great effect on the adsorption (Zhang et al. 2012d). Natural clay, aluminum pillared clay (Eloussaief et al. 2013), and Apatite II (TM) (Oliva et al. 2011) possessed relatively lower adsorption capacity for $\text{Hg}(\text{II})$. By introduction of halogen functional groups onto the adsorbents, the obtained KI- and KBr-modified clay (KI-clay and KBr-clay) showed enhanced adsorption capacities for Hg^0 (Cai et al. 2014).

As one kind of impure clay which mostly consists of montmorillonite, bentonite could be employed as an efficient adsorbent (Fernandez-Nava et al. 2011). Due to the emerging electrostatic interactions by the protonated amines, 4-aminoantipyrine-modified bentonite (Wang et al. 2011), trioctylamine (TOA)-modified sodium montmorillonite (Kumar et al. 2012), 3-aminopropyltriethoxysilane (APTES)-modified natural attapulgite (ATP) (ATP-APTES) (Cui et al. 2013), and PANI-modified ATP (PANI/ATP) (Cui et al. 2012) all showed increased maximum adsorption capacities toward $\text{Hg}(\text{II})$ and so did glycidyl methacrylate (GMA) and $\text{N,N}'$ -methylene bis-acrylamide (MBA) copolymer and tetraethylenepentamine (TEP)-modified kaolinite mineral (Donia et al. 2011).

Ionic liquids (ILs)

Most of the existing adsorbents either are costly or may release by-products that are harmful to the environment. The development of novel adsorbents with high efficiency, low pollution, and low cost for Hg species removal is thus to be expected. As promising functional materials, ILs have attracted great attention recently. Fe-based ionic liquids, 1-butyl-3-methylimidazolium halide ($[\text{bmim}]\text{X}$, $\text{X} = \text{Cl}$ or Br) (Fig. 6)-impregnated $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$, including ($[\text{bmim}]\text{FeXCl}_3$) and their analogs ($[\text{bmim}]\text{X}$), showed good Hg^0 removal efficiencies. Trioctylmethylammonium thiosalicylate (TOMATS), a task-specific IL containing $-\text{SH}$ groups, was built on a carrier of palm shell AC (PSAC), and the TOMATS-PSAC exhibited a high adsorption capacity for $\text{Hg}(\text{II})$ at pH 8 (Abu Ismaiel et al. 2013).

Synthetic organic adsorbents

Synthetic, organic, thiol-containing bidentate chelate molecules could strongly bind heavy metals to realize high

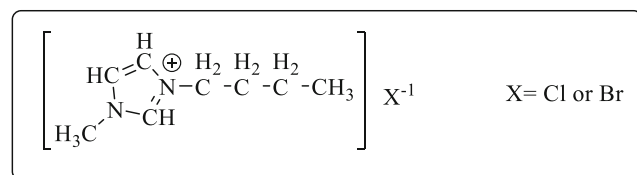


Fig. 6 Chemical structure of $[\text{bmim}]\text{X}$

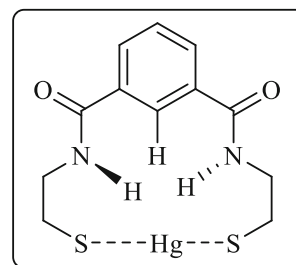


Fig. 7 Chemical structure of $\text{BD}(\text{S})\text{-Hg}$

adsorption capacities (Zhao et al. 2013a, b). Due to the high chelating energy, 3-benzenediamidoethanethiolate $[\text{BD}(\text{S})]$ would combine with $\text{Hg}(\text{II})$ to form a chelate of $\text{BD}(\text{S})\text{-Hg}$ (Fig. 7). If sulfur groups were replaced by selenium (Se) or tellurium (Te), the chelating strength of $\text{BD}(\text{X})$ ($\text{X} = \text{Se}$ or Te) complexes for $\text{Hg}(\text{II})$ would increase, and $\text{Hg}(\text{II})$ removal is thus predicted to form the $\text{BD}(\text{X})\text{-Hg}$ complex (Zou et al. 2013).

Membranes

Membranes always have a place in the field of adsorption. To remove $\text{Hg}(\text{II})$ from drinking water and industrial effluents, various membranes were developed in recent years. Epichlorohydrin-modified crosslinked CTS (CTS-ECH) membrane matrix (Rabelo et al. 2012), agarose gel trapped bovine serum albumin (BSA)-Au NPs (Au NPs@BSA/AGM) (Lee et al. 2012), and a mixed-matrix membrane made from the dispersion of thiol-functionalized metal oxide-coated carbon (MOCC) in the polysulfone (PSF) matrix (MOCC-PSF) (Meeks et al. 2013) all showed high maximum adsorption capacities for $\text{Hg}(\text{II})$. Specially, PVA-modified CTS-based ceramic ultrafiltration membrane exhibited almost 100 % removal for $500 \mu\text{g L}^{-1}$ of $\text{Hg}(\text{II})$ through a polymer-enhanced ultrafiltration (PEUF) technique (Jana et al. 2011).

Emulsion liquid membranes (ELM) were also widely applied in separation technologies. To overcome the hindrance in the application of ELM at an industrial scale, great efforts have been made to get a desired level of stability. A stable emulsion was developed for the removal of Hg from aqueous solution (Gupta et al. 2013). The removal of ultralow levels of Hg species from an aqueous solution is a challenge. An oligothymonucleic acid (OTA)-modified polyethylene (PE) film was developed, which could selectively remove $\text{Hg}(\text{II})$ of sub-ppb level from an aqueous solution (Yu et al. 2012b).

Adsorption mechanisms

The relationship between material structure and its adsorption ability

To provide molecular-level understanding of the effects of chemical functional groups on Hg adsorption, systematic

theoretical studies were performed. The mechanisms of adsorption of Hg^0 , HgCl , and HgCl_2 molecules onto ZnO surface were investigated by density functional theory (DFT) (Ling et al. 2014). Hg^0 is physically adsorbed onto ZnO surface at the Zn atom, and HgCl_2 can be adsorbed in molecular mode. However, HgCl is difficult to be adsorbed and exists in dissociative mode. The formation of oxidized Hg complexes (e.g., $[\text{HgX}]^+$, $[\text{HgX}_2]$, and $[\text{HgX}_4]_2$; X = Cl, Br, or I) might contribute to halogen-containing group-functionalized adsorbents in removing Hg^0 ; thus, their high Hg^0 adsorption performance is predictable (Yao et al. 2013).

To better understand the surface electronic structural changes upon adsorption, the adsorption of Hg^0 on CuO (110) surface was also investigated using first-principles quantum mechanical methods (Xiang et al. 2012). Hg^0 was found to bind weakly to the O-terminated CuO (110) surface and strongly on the Cu-terminated CuO (110) surface; therefore, the significant overlap between the d-state of Hg and the s-states of Cu might be beneficial to the strong Hg and Cu-terminated CuO (110) interactions. Also, Hg adsorption capacity of sulfur-impregnated adsorbent increased along with its decreased specific surface area; thus, the interaction of the lone pairs of electrons of sulfide groups might be associated with the higher adsorbent's Hg adsorption capability (Hu et al. 2013). The zigzag and armchair edges of functionalized carbonaceous adsorbents have a huge impact on Hg adsorption: the embedding of halogen atom can increase the adsorption activities, and the introduced phenol and carboxyl functional groups are beneficial for physisorption of Hg^0 and weaken Hg(II) adsorption (Liu et al. 2011).

As an electronic acceptor, Hg(II) would receive significant electrons transferred from the functional groups of adsorbents to form coordination bonds (Sun et al. 2011b). Therefore, functional groups such as sulfur, thiosemicarbazide, amino, sulfamine, AO, and so on could be applied as chelating media to modify the adsorbents. To form multidentate chelates, chelating adsorbents containing multityped functional groups that are capable of coordinating to Hg(II) are expected. Sulfur- and AO-containing bifunctional silica gel-based hybrid materials exhibited high adsorption capability for Hg(II) (Fig. 8) (Qu et

al. 2013) and so did thiosemicarbazide-modified PET (Monier and Abdel-Latif 2013). The amino group is likely to form new structures such as imino bonds with Hg(II) , which is one of the key factors that makes contribution to the higher adsorption capacity of amino group-functionalized adsorbents (Vieira et al. 2011). and selective and high efficient adsorption for Hg(II) could be achieved (Qi et al. 2013). An efficient Hg-specific adsorbent, AO group-modified PAN-grafted PS (PAN-g-PS), can selective remove Hg(II) from aqueous solutions (Zong et al. 2011).

Adsorption capacity contrast and isotherm and kinetic studies

A summary of the adsorption capacities of various adsorbents for Hg removal from aqueous solutions under optimum conditions is presented in Table 1. It can be seen from Table 1 and the “**Absorbent development**” section of this review that carbon material-based composites, NFs, and hydrogels have shown the most promising prospects in Hg removal. Tables 2 represents the applicable isotherm and kinetic models for Hg removal by various adsorbents, and the pseudo-second-order kinetic model and Freundlich and/or Langmuir isotherm models were found to fit well with the experimental data.

Future research directions and conclusions

Due to its high toxicity and bioaccumulation, the existence of Hg species in the environment is always a threat to human beings. In order to control Hg pollution, scientists have put great efforts and enthusiasm on the adsorption technologies in the past few decades. Based on a systematic review of the literature from 2011 to 2014, the following concluding remarks can be made:

1. Many factors of the adsorbents such as specific surface area, pore size distribution, polarity, and the introduced functional groups would affect their adsorption efficiencies. To develop novel adsorbents with high adsorption

Fig. 8 The proposed interactions of sulfur/amidoxime- (a) and thiosemicarbazide- (b) modified adsorbents with Hg(II) (Monier and Abdel-Latif 2013; Qu et al. 2013)

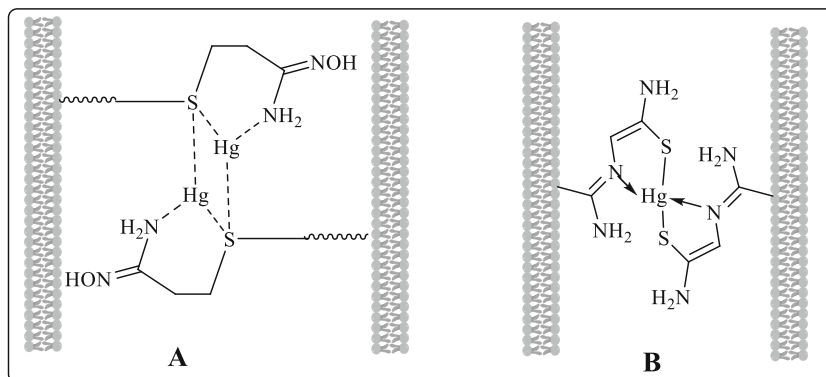


Table 1 Adsorption of Hg by various adsorbents

Type of Hg	Type of adsorbent	Optimal adsorption conditions	The maximum adsorption capacity	Reference
Hg(II)	Sulfurized ACs	Not provided	Enhanced adsorption abilities than AC	Abdelouhab-Reddam et al. (2014). Feng et al. (2006a, b), Ie et al. (2012, 2013), Li et al. (2013c)
Hg(II)	Concurrent sulfurized/activated ACs	Not provided	293 mg g ⁻¹	Shamsilazeyi and Kaghazchi (2014)
Hg ⁰	Functionalized ACs	At 25 °C and mercury concentration of 230 µg m ⁻³	AC<Cl-impregnated AC<Br-impregnated AC<I-impregnated AC	De et al. (2013)
Hg(II)	MAC	With a 3:1 for C/Fe	92.5 % sorbent recovery and 96.3 % Hg removal	Faulconer et al. (2012)
Hg(II)	Sulfurized MAC	Not provided	38.3 mg g ⁻¹	Okamoto et al. (2011)
Hg(II)	Ox-MAC and SH-MAC	Not provided	34.5 mg g ⁻¹ for Ox-MAC and 39.4 mg g ⁻¹ for SH-MAC	Miura and Tachibana (2014)
Hg ⁰	AC from biomass material and sulfurized AC	Hg ⁰ concentration of 10 µg Nm ⁻³ , at 70 °C	Sulfur introduction causes enhancements in Hg ⁰ adsorption; up to 97 % removal	Hsi et al. (2013, 2014). Hu et al. (2014)
Hg ⁰	ACFs-SH	Hg ⁰ concentration of ~21 ng mL ⁻¹	11~15 mg g ⁻¹	Yao et al. (2014b)
Hg(II)	AMT-OCMK-3	0.5 g L ⁻¹ of adsorbent and 10 mg L ⁻¹ mixed solutions, at pH 6	450.45 mg g ⁻¹	Anbia and Dehghan (2014)
Hg(II)	ZnO-modified CMK-3	The agitation time of 120 min, the initial C _{Hg(II)} of 10 mg L ⁻¹ , 35 °C, pH 6	99 % removal	Zolfaghari et al. (2011)
Hg(II)	Zeolite Y, β-zeolite, and mordenite	Not provided	β-Zeolite was found to be the highest	Murthy et al. (2013)
Hg(II)	Fe ₃ O ₄ nanocrystals encapsulated hierarchical mesoporous zeolite	Not provided	Selective adsorption of Hg(II)	Yin et al. (2012)
Hg(II)	MWCNTs	At pH 6.5; equilibrated for 24 h at 27 °C, with initial concentration of 50 mg L ⁻¹	90 % removal	Shadbad et al. (2011)
Hg(II)	Amino- and thiol-functionalized MWCNTs	At pH 6, 25 °C and 200 rpm agitation speed; adsorbent dose of 400 mg L ⁻¹	84.66 mg g ⁻¹	Hadavifar et al. (2014)
Hg(II)	SWCNT-SH	At pH 5.0; at an initial concentration of 0.4 mg mL ⁻¹	131 mg g ⁻¹	Bandaru et al. (2013)
Hg(II)	ox-MWCNT (or MWCNT, or SWCNT)-impregnated CTS	At pH=4, contact time=40 min, and temperature=70 °C	183.2, 167.5, and 172.7 mg g ⁻¹ , respectively	Shawky et al. (2012)
Hg(II)	GO and GO/CS	25 °C, pH=4	381 and 187 mg g ⁻¹ , respectively	Kyzas et al. (2014)
Hg(II)	SRGO nanohybrid	At neutral pH under ambient conditions	90 % removal	Thakur et al. (2013)
Hg(II)	Dithiocarbamate group-modified iron magnetite Fe ₃ O ₄	Initial C _{Hg(II)} of 50 mg L ⁻¹ , at 21 °C	206 mg g ⁻¹	Figueira et al. (2011)
Hg(II)	Fe ₃ O ₄ @SiO ₂	Not provided	Hg(II) concentration reached a plateau at 0.0025 mg L ⁻¹	Jing-po et al. (2014). Tavares et al. (2013)
Hg(II)	Fe ₃ O ₄ @SiO ₂ -SH	At pH 6.5	From 148.8 to 207.7 mg g ⁻¹	Hakami et al. (2012). Zhang et al. (2013)
Hg(II)	SH-Fe ₃ O ₄ -NMPs	Not provided	256.4 mg g ⁻¹	Pan et al. (2012a, b)
Hg(II)	CTS-Fe ₃ O ₄	At 20 °C and pH=2.7~7	152 mg g ⁻¹	Katok et al. (2013)
Hg(II)	CTS-Fe ₃ O ₄ -PTU	At pH=5.0	135 mg g ⁻¹	Monier and Abdel-Latif (2012)
Hg(II)	2-MBTZ-Fe ₃ O ₄	At pH=9.0	98.6 % removal	Parham et al. (2012)
Hg(II)	Zn _{0.46} Fe _{2.54} O ₄	At neutral pH, 5 min of contact time	416 mg g ⁻¹	He et al. (2012)

Table 1 (continued)

Type of Hg	Type of adsorbent	Optimal adsorption conditions	The maximum adsorption capacity	Reference
Hg(II)	CoFe ₂ O ₄	At pH 4.6 and 25 °C	157.9 mg g ⁻¹	Zhang et al. (2014b)
Hg(II)	Au NPs mixed Al ₂ O ₃	Flow rate of 2.5 mL min ⁻¹	97 % removal	Lo et al. (2012)
Hg ⁰	AC-impregnated CuCl ₂	At 333 K; 5 min; 20 ppm HCl of flue gas	90 % removal	Du et al. (2014)
Hg(II)	ZnO nanoparticles	At pH 5.5	714 mg g ⁻¹	Sheela et al. (2012)
Hg(II)	Sulfur-functionalized silica	At pH 5.8–8.2	47.50 mg g ⁻¹	Arsuaga et al. (2014), Saman et al. (2014)
Hg(II)	POSS xerogel	At pH 2.7–7	95 mg g ⁻¹	Katok et al. (2013)
Hg(II)	BTESPT-SGs and MPTMS-SGs	Not provided	93.32 and 102.37 mg g ⁻¹ , respectively	Johari et al. (2014a, b)
Hg(II)	4-Amino-5-methyl-1,2,4-triazole-3(4H)-thion-modified silica	At pH 6.0–7.5	142.85 mg g ⁻¹	Toubi et al. (2013)
Hg(II)	Aminophosphonic acid-supported silica gel	At pH=5.0	303.03 mg g ⁻¹	Wang et al. (2012)
Hg(II)	VIM-modified silica	At pH=5.0	355.9 mg g ⁻¹	Sun et al. (2013)
Hg(II)	PAOA/SBA-15	C ₀ =100 mg L ⁻¹ , ionic strength is 0.2 M NaNO ₃ , sorbent dosage is 0.1 g L ⁻¹ and pH=5.5	433.55 mg g ⁻¹	Tang et al. (2011)
Hg(II)	L-cys-SBA-15	At room temperature; Hg(II) adsorption time <5 min	429 mg g ⁻¹	Li et al. (2014)
Hg(II)	MPTMS, APTES, and AEAPTMS-modified diatom silica microparticles	At pH=6	185.2, 131.7, and 169.5 mg g ⁻¹ , respectively	Yu et al. (2012a)
Hg(II)	PAN-AT	At pH=6.5	454.9 mg g ⁻¹	Xiong et al. (2013)
Hg(II)	AO-modified PAN	At pH=2.0	567 mg g ⁻¹	Zong et al. (2012)
Hg(II)	AO PAN-CS-Br	At pH=4.0	826.43 mg g ⁻¹	Xu et al. (2014)
Hg(II)	HA-PANI	C ₀ =500 mg L ⁻¹ ; pH=5.0	671 mg g ⁻¹	Li et al. (2011a)
Hg(II)	PS-AMI	At 298 K	335.6 mg g ⁻¹	Xiong et al. (2012)
Hg(II)	Bis-8-oxyquinoline-terminated open-chain crown ether-modified PS	Not provided	1163.8 mg g ⁻¹	Sun et al. (2012)
Hg(II)	Thiosemicarbazide-modified PET	At pH=5.0	120 mg g ⁻¹	Monier and Abdel-Latif (2013)
Hg(II)	PVBU nanofiber	At pH=7.0	543.9 mg g ⁻¹	Wang et al. (2013b)
Hg(II)	Acrylic acid copolymerized polypropylene fiber	Not provided	1144.86 mg g ⁻¹	Ma et al. (2012)
Hg(II)	4-VP-PET	At pH=3.0	137.18 mg g ⁻¹	Bozkaya et al. (2012)
Hg(II)	CTS-PVA hydrogel	At pH=5.50	585.90 mg g ⁻¹	Wang et al. (2013a)
Hg(II)	CTS-functionalized generation 1.0, 2.0, and 3.0 of PAMAM	At 35 °C	398.41, 259.07, and 259.07 mg g ⁻¹ , respectively	Ma et al. (2009)
Hg(II)	CST	C ₀ =500 µg L ⁻¹ ; pH=8.1	94 % removal	Lopes et al. (2014)
Hg(II)	GG-cl-CH=N-(CH ₂) ₆ -N=CH-GG	At 20 °C; pH=5.0; C ₀ =25 ppm	41.13 mg g ⁻¹	Thakur et al. (2014)
Hg(II)	Amine- and thiol-modified cellulose	Not provided	7.2 and 38 mmol g ⁻¹ , respectively	Donia et al. (2014)
Hg(II)	RS-GM and RS-Raw	C ₀ =50 mg L ⁻¹ ; pH 9.0	161.30 and 103.1 mg g ⁻¹ , respectively	Song et al. (2013)
Hg(II)	GMA and diethylenetriamine-modified cellulose	Not provided	440 mg g ⁻¹	Donia et al. (2013)

Table 1 (continued)

Type of Hg	Type of adsorbent	Optimal adsorption conditions	The maximum adsorption capacity	Reference
Hg(II)	FPBS	$C_0=25 \text{ mg L}^{-1}$; pH=7.0	132.25 mg g^{-1}	Mullaseery et al. (2014)
Hg(II)	UBC	Not provided	288.632 kJ/mol	Li and Maroto-Valer (2012)
Hg(II)	LCF wastes of coconut	Not provided	144.4 mg g^{-1}	Johari et al. (2014c)
Hg(II)	Buckwheat hulls	At 35 °C; pH=5.0	243.90 mg g^{-1}	Wang et al. (2013c)
Hg(II)	<i>Paecilomyces catenulannulatus</i>	At pH=7.0	140.85 mg g^{-1}	Li et al. (2013a)
Hg(II)	<i>Pseudomonas putida</i> V1 strain	At pH in the range 4.0–6.0; at 21–25 °C	Over 80 % removal	Cabral et al. (2013)
Hg(II)	<i>Bacillus mucilaginosus</i>	Not provided	393 mg L^{-1}	Mo and Lian (2011)
Hg(II)	CTS-PVA	At 60 °C	950.62 mg g^{-1}	Wang et al. (2014b)
Hg(II)	CTS-g-PAA	At pH=6.0	2001.8 mg g^{-1}	Saber-Samandari and Gazi (2013)
Hg(II)	POSS-SH	At pH=6.0	12.90 mg g^{-1}	Wang et al. (2014a)
MeHg	POSS-SH	At pH=3.2	46.73 mg g^{-1}	Wang et al. (2014a)
Hg(II)	Clay and pillared clay	$C_0=75 \text{ mg L}^{-1}$ at pH=2	9.70 and 49.75 mg g^{-1} , respectively	Eloussaief et al. (2013)
Hg(II)	Apatite II (TM)	At pH=4.0	38 mg g^{-1}	Oliva et al. (2011)
Hg(II)	4-Aminoantipyrine-modified bentonite	$C_0=10 \text{ mg L}^{-1}$; pH=3.0	52.9 mg g^{-1}	Wang et al. (2011)
Hg(II)	TOA-modified sodium montmorillonite	At pH range of 3–11	140.84 mg g^{-1}	Kumar et al. (2012)
Hg(II)	ATP and ATP-APTES	At around pH 5–9	5 and 90 mg g^{-1} , respectively	Cui et al. (2013)
Hg(II)	PANI/ATP	At pH=8	800 mg g^{-1}	Cui et al. (2012)
Hg(II)	TOMATS-PSAC	Not provided	83.33 mg g^{-1}	Abu Ismaiel et al. (2013)
Hg(II)	CTS-ECH	$C_0=150 \text{ mg L}^{-1}$	682 mg g^{-1}	Rabelo et al. (2012)
Hg(II)	MOCC and MOCC-PSF	$C_0=500 \text{ } \mu\text{g L}^{-1}$	60 and 200 mg g^{-1} , respectively	Meeks et al. (2013)
Hg(II)	PEUF	pH 8.0, 47 °C, $C_{K_2FeO_4}=0.25 \text{ mmol L}^{-1}$	100 % removal	Jana et al. (2011)
Hg ⁰	K ₂ FeO ₄	Not provided	81.4 % removal	Zhao et al. (2014)
Hg(II)	Polypyrrole-CTS	pH 4.0, 20 °C, initial Hg(II) concentration of 200 mg L^{-1}	40 mg g^{-1}	Salahi et al. (2014)
Hg(II)	AL-SH	pH 2.0, initial Hg(II) concentration of 0.178 M, 25 °C	101.2 mg g^{-1}	Zhou et al. (2014)
Hg(II)	Hybrid nanosilica	pH 4.6, 25 °C	134 mg g^{-1}	Perez-Quintanilla and Sierra (2014)
Hg(II)	CoFe ₂ O ₄ -RGO	pH 8, initial Hg(II) concentration of 1000 and 1250 mM	157.9 mg g^{-1}	Zhang et al. (2014b)
Hg(II)	FeS	pH 6, 30 °C, 40 min, initial Hg(II) concentration of 100 mg L^{-1}	99 % removal	Han et al. (2014)
Hg(II)	ALP	pH 6.0, 35 °C	107.5 mg g^{-1}	Aslam et al. (2013)
Hg(II)	<i>Vibrio</i> sp. cells	pH 6.0–7.0, initial Hg(II) concentration of 200–400 mg L^{-1}	193.2 mg g^{-1}	Jafari and Cheraghi (2014)
Hg(II)	HCMP-CuO-NFs		1767.97 mg g^{-1}	Kumar et al. (2014)

Table 2 Adsorption isotherm and kinetic studies of Hg(II) removal from aqueous solution

Type of adsorbent	Applicable isotherm models	Applicable kinetic models	Reference
ZnO nanoparticles	The Langmuir model is better	Pseudo-second-order is better	Sheela et al. (2012)
TOMATS-PSAC	The Langmuir model is better	Pseudo-second-order is better	Abu Ismaiel et al. (2013)
AL-SH	Langmuir and Temkin better than Freundlich	Not provided	Zhou et al. (2014)
CoFe ₂ O ₄ -RGO	Langmuir is better than Freundlich	Pseudo-second-order is better	Zhang et al. (2014b)
Wheat straw, swine manure	Langmuir and Freundlich	Not provided	Zhang et al. (2014a)
AO PAN-CS-Br	Langmuir is better than Freundlich	Pseudo-second-order is better	Xu et al. (2014)
GG-cl-CH=N-(CH ₂) ₆ -N=CH-GG	Freundlich is better than Langmuir	Pseudo-second-order is better	Thakur et al. (2014)
ZnCl ₂ -MCM-41	Not provided	Pseudo-second-order is better	Raji and Pakizeh (2014)
AAm-PET films	The Langmuir model is better	Pseudo-second-order is better	Rahman et al. (2014)
PMPSQs-SH	The Langmuir model is better	Pseudo-second-order is better	Niu et al. (2014)
FPBS	The Freundlich model is better	Pseudo-second-order is better	Mullassery et al. (2014)
GO/CS	The Langmuir model is better	Pseudo-second-order is better	Kyzas et al. (2014)
LCF and LCF-NaOH	Langmuir model is better	Pseudo-second-order is better	Johari et al. (2014c)
ALP	The Langmuir model is better	Pseudo-second-order is better	Aslam et al. (2013)
SRGO	The Langmuir model is better	Pseudo-second-order is better	Thakur et al. (2013)
TiO ₂ nanocrystals	Freundlich is better than Langmuir	Pseudo-second-order is better	Ghasemi et al. (2012)
MSR	The Langmuir model is better	Pseudo-second-order is better	Anagnostopoulos et al. (2012)
AMT-OCMK-3	The Langmuir model is better	Not provided	Abdelouhab-Reddam et al. (2014)
BTESPT-SGs and MPTMS-SGs	The Langmuir model is better	Pseudo-second-order is better	Johari et al. (2014b)
HCMP-CuO-NFs	Fitted with both the Freundlich and Langmuir models	Pseudo-second-order is better	Kumar et al. (2014)

efficiencies, surface nanocrystallization (Singh and Kumary 2011) and modification of the materials are necessary. A newly developed nanoscale adsorbent, *pumice-nanoscale zero-valent iron* (P-NZVI), showed a very high adsorption ability for Hg(II) (Liu et al. 2014c). Oligonucleotide-modified magnetic silica sphere @Au nanoparticles (MSS@Au NPs) have the potential of effectively removing Hg(II) from surrounding solutions (Liu et al. 2014b) and so does thiol-functionalized MSS (Song et al. 2011).

2. Adsorbents with excellent properties such as high selectivity, high capacity, regenerability, and stability are expected. For example, newly developed FPBS could be easily regenerated by 0.1 M HCl (Mullassery et al. 2014). aerogel (Standeker et al. 2011). and xerogel (Singh and Singh 2011) possessing very high adsorption capacities.
3. To reduce the overall impact of the adsorption process on the environment, novel Hg treatment technologies with minimal generation of contaminated waste are expected. Ion exchange membrane bioreactor (IEMB) process might be a good choice (Oehmen et al. 2014). Hollow fiber-supported liquid membrane (HFSLM) can also provide a new concept in Hg removal (Lothongkum et al. 2011).
4. Being abundantly available and cheap, recycled solid waste materials could be reused as low-cost bioadsorbents for Hg(II) removal (Johari et al. 2014c).
5. Both the chemical structure and physical property of an adsorbent play important roles in its Hg adsorption capability. The relationship of the adsorbent's properties (including particle morphology, surface area, elemental composition, etc.) to adsorption is expected to clarify.
6. The adsorption mechanism for Hg species removal is still not well explained. Computer simulation of the interactions between the adsorbents and the Hg species still needs to be investigated deeply.

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