



Recent developments in materials used for the removal of metal ions from acid mine drainage

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Abstract

Acid mine drainage is the reaction of surface water with sub-surface water located on sulfur bearing rocks, resulting in sulfuric acid. These highly acidic conditions result in leaching of non-biodegradable heavy metals from rock which then accumulate in flora, posing a significant environmental hazard. Hence, reliable, cost effective remediation techniques are continuously sought after by researchers. A range of materials were examined as adsorbents in the extraction of heavy metal ions from acid mine drainage (AMD). However, these materials generally have moderate to poor adsorption capacities. To address this problem, researchers have recently turned to nano-sized materials to enhance the surface area of the adsorbent when in contact with the heavy metal solution. Lately, there have been developments in studying the surface chemistry of nano-engineered materials during adsorption, which involved alterations in the physical and chemical make-up of nanomaterials. The resultant surface engineered nanomaterials have been proven to show rapid adsorption rates and remarkable adsorption capacities for removal of a wide range of heavy metal contaminants in AMD compared to the unmodified nanomaterials. A brief overview of zeolites as adsorbents and the development of nanosorbents to modernly applied magnetic sorbents and ion imprinted polymers will be discussed. This work provides researchers with thorough insight into the adsorption mechanism and performance of nanosorbents, and finds common ground between the past, present and future of these versatile materials.

Keywords Physically and chemically modified materials · Acid mine drainage · Sorption capacity · Bio-sorbents

Introduction

Exposure of pyrite (FeS_2) to oxidative conditions leads to the formation of acidic conditions, resulting in acid mine drainage (AMD). Subsequently, acidic conditions result in leaching of heavy metals into aquatic systems creating an environmental hazard as heavy metals are non-biodegradable (Ge et al. 2012). Common hazardous heavy metals include, amongst others, Cr, Cd, Cu, Ni and U in various oxidation forms (Li et al. 2018a, b; Lu et al. 2018; Tavengwa et al. 201a). Heavy metals enter the food chain in a variety of ways, accumulating in living organisms with time (Archary et al. 2017). One of the mechanisms include the high affinity for sulfur which inhibit normal enzyme function by formation of covalent bonds with

sulfur groups in enzymes. Heavy metals are chemically bound by organic functions such as protein carboxylic acid ($-\text{COOH}$) and amino ($-\text{NH}_2$). Considering the toxic nature and non-biodegradable characteristics of heavy metals, removal of toxic metals from industrial effluents is essential for the safety of inhabitants.

Remediation of heavy metals in AMD by adsorption has shown to be a productive approach (Sahu et al. 2017; Castro et al. 2018). Generally, adsorption is an inexpensive and useful in comparison to other remediation methods which includes lime neutralization (Othman et al. 2017), carbonate neutralization (Galhardi and Bonotto 2015), ion exchange (Jafaripour and Ghataora 2015; Nordstrom et al. 2015) and metal precipitation (Jafaripour and Ghataora 2015). Some materials studied using this technique include zeolites. These materials and their modified forms have been shown to be very good adsorbents for heavy metals due to their desirable cation exchange properties (Nek-hunguni et al. 2017a; Mokgehle et al. 2019). However, zeolites were reported to have setbacks such as poor ion selectivity, for instance, Hui et al. (2005) reported on the

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poor adsorption performance of zeolite-4A in comparison to coal fly ash for removal of Cu^{2+} , Cr^{3+} , Zn^{2+} , Co^{2+} and Ni^{2+} from aqueous systems.

In an effort to enhance adsorption performance, researchers focused on nano-sized materials for extraction of heavy metals. One eye-capturing class of adsorbent includes magnetic nanoparticles and its composites; an example includes magnetic graphene oxide composites. Structurally, graphene has a honeycomb arrangement, which significantly improves its surface area. This has led to its various applications as an adsorbent (Cui et al. 2012; Ren et al. 2014; Vilela et al. 2016). Additionally, graphene's versatility allows for functionalization, particularly with carbonyls, hydroxyls and epoxy groups, by oxidation of natural graphite (Gupta et al. 2017; Méndez-Romero et al. 2019). Addition of these groups makes graphene oxide sheets hydrophilic and soluble in water (Gupta et al. 2017). However, these materials had setbacks such as poor recyclability, due to its high dispersion in water (Liu et al. 2017).

To address this problem, researchers then focused on introducing physical modifications to graphene oxide nanoparticles by incorporation of magnetic Fe_3O_4 nanoparticles (Gugushe et al. 2019). Iron oxide containing materials exhibit useful characteristics such as paramagnetism, biocompatibility, safety, cheap, good adsorption potential and simplicity in separation of the adsorbent from the solution. Despite these advantages, these materials have a tendency of agglomerating and decomposing under acidic conditions resulting in limited adsorption performances. Recently, researchers introduced some modifications which included the use of functional groups such as NH_2 or SH to stabilize Fe_3O_4 (Yang et al. 2016).

The use of conventional techniques for treatment of heavy metals due to AMD was reported by Aubé et al. (2003) and Dempsey and Jeon (2001). However, the major drawbacks of conventional remediation approaches range from high operation costs, significant maintenance requirements, poor selectivity, clogging of filtration media to requirements of substantial amounts of remediation materials. Hence, the need for highly selective, efficient and high surface area adsorbents arises. To the best of our knowledge, little is known of a review that summarizes the development of physically and chemically modified zeolites from unmodified zeolites and its utilization for treatment of heavy metal ions in AMD. In this review, an update on advances into usage of iron oxide and metal oxide containing nanomaterials from unmodified nanosorbent are discussed. The development of highly selective ion imprinted polymers (IIP) is also reported. A summary of the adsorption capacities of the nanomaterials for heavy metal ions is presented. Finally, this work highlights some challenges and future perspectives to further bring about attractive developments in this area of water treatment.

Zeolites and chemically modified zeolites as adsorbents for heavy metals in AMD

Of late, zeolites, as sorbents have caught the attention of most researchers. Chemically, zeolites have the formula $\text{M}_{e2/n} \cdot \text{O} \cdot \text{Al}_2\text{O}_3 \cdot x\text{SiO}_2 \cdot y\text{H}_2\text{O}$ (Mahdi et al. 2017), where M_e represents first or second group atoms, n represents atomic charge, x highlights the number of Si tetrahedra $2 \leq x \leq 10$, and y is the amount of H_2O molecules ($2 \leq y \leq 7$) (Jha and Singh 2016). The zeolite is composed of SiO_4 and AlO_4 tetrahedra forming a lattice of Si and Al centers. The network of SiO_4 and AlO_4 tetrahedra frequently undergo cation exchange where the Si (+4) is replaced by Al (+3) ions, resulting in a net negative charge (Jha et al. 2012; Jha and Singh 2016).

There is a wide literature coverage which demonstrated the application of zeolites in adsorption of heavy metals from AMD (Masukume et al. 2017; Nekhunguni et al. 2017a; Mokgehle et al. 2019; Znedelska et al. 2018; Buenaño et al. 2017; Shabalala and Basitere 2020). For instance, zeolites were evaluated by Turan and Ozgonenel (2013) as Cu (II) adsorbents through an adaptive neural fuzzy interface system where average adsorption performances were obtained. However, zeolites have often been overwhelmed by setbacks such as early exhaustion of adsorption sites and high expenses associated with the synthesis of the majority of the zeolites. To enhance the adsorption performance, researchers then focused on the utilization of modified or organic coated zeolites (Table 1). In addition to cation exchange, organic coated zeolites adsorb heavy metals through opposite charge based attraction. This enhancement is due to the presence of charged organic groups on the surface of the zeolite (Mokgehle et al. 2019). Though the inclusion of organic moieties improved the removal efficiencies of zeolites, these materials still exhibit moderate heavy metal removal efficiencies as a result of their low surface area in the sample solution. Hence, researchers have of late turned their interests to nano-sized materials as potential adsorbents for heavy metal remediation in AMD.

Nanosorbents

Adsorption is known to be an environmentally, ecologically and green method for the removal of toxic metal(s) from contaminated solution. Adsorption is a versatile technique, in the sense that the adsorbent can be restored and subsequently re-used without reducing the removal efficiency (Bai et al. 2017). A range of adsorbents for treatment of AMD have been used, which include high density

Table 1 Modified zeolites applied for adsorption of AMD associated heavy metals

Modified zeolite	Target metal(s)	Sorption capacity (mg g ⁻¹)	Removal efficiency (%)	Reference
Iron (hydr) oxide	As(V)	1.69	–	Nekhunguni et al. (2017a)
Oxalic acid	Cd (II)	–	46.68	Mokgehle et al. (2019)
Succinic acid	Cd (II)	–	46.57	Mokgehle et al. (2019)
Adipic acid	Cd (II)	–	46.64	Mokgehle et al. (2019)
DCMD	Fe (II)	62.11	–	Ryu et al. (2019)
DCMD	Al (II)	44.64	–	Ryu et al. (2019)
DCMD	Zn (II)	39.96	–	Ryu et al. (2019)
HFOMZ	U (VI)	0.24	–	Nekhunguni et al. (2017b)

*DCMD-submerged direct contact membrane distillation, HFOMZ-hydrous ferric oxide-modified zeolite

sludge (Sukati et al. 2018; Dlamini et al. 2019), alkali–barium–calcium (Mulopo and Motaung 2014; Akinwekomi et al. 2017), coal fly ash (Rios et al. 2008; Madzivire et al. 2019) and lignite (Mohan and Chander 2006; Karagüzel et al. 2020). Among the studied adsorbents is activated carbon (Hong et al. 2017), silica (Etale et al. 2016; Lakovleva and Sillanpää 2013) and ferric oxide (Dlamini et al. 2019). The major shortfall of activated carbon nanosorbents is that they are expensive to use and laborious to isolate from wastewater after application. Naturally derived materials such as banana peels have also been investigated as sorbents for the removal of heavy metal ions. For example, Castro et al. (2011) studied the use of banana peels (particle size 35–45 mm) for Cu adsorption from aqueous solution and obtained an adsorption capacity of 20 mg g⁻¹. However, the macroscale materials studied by Castro et al. (2011) had low surface area and were responsible for low adsorption performances as argued by Bakiya and Sudha (2012) and Ashok et al. (2010). As a result, researchers have now directed their attention to a unique class of materials in the form of nanomaterials. These materials permit for more active sites with increased surface area and can display novel properties which can be applied in the design of new heavy metal treatment methods and improvement of current ones (Bakiya and Sudha 2012). The surface chemistry of the adsorbent is an important parameter that determines extent of adsorption. For instance, the acicular goethite nanoparticles studied by Moreira et al. (2017) were found to be effective for the removal of As (V) from AMD and was attributed to its acidic point of zero charge $pH_{pzc} = 3.5$. This was further indicated in the same study that the net charge of the sorbent above $pH = 3.5$ was negative and facilitated adsorption of cationic heavy metals from AMD. However, the surface charge may not be the only determining factor, for example Rodríguez et al. (2020) evaluated graphene oxide and graphene oxide with zinc nanoparticles (GO–ZnO) nanocomposites with $pH_{pzc} = 3.98$ and $pH_{pzc} = 5.57$, respectively, and observed that the latter adsorbent had

a better adsorption capacity for Al and Cu. Theoretically, in the same study, it was expected that the GO nanocomposite would be a better sorbent for heavy metals due to its negatively charged surface at a relatively lower pH to GO–ZnO. However, functionalization with ZnO seemed to have played a greater role in adsorption under AMD simulated conditions. Another factor, pore size, was shown to play a role in influencing the selectivity of heavy metals in AMD. Etale et al. (2017) evaluated mesoporous silica nanoparticles and reported that possible configurational advantages of Mn (II) lead to its better adsorption than Cu (II). Similar trends on the influence of pore size were also reported by Nqombolo et al. (2019) and Dlamini et al. (2019). Table 2 shows the adsorption capacities of some recently studied nano-materials. The nanosorbents studied ranged from bio-materials such as banana peels, microorganisms (fungi) to inorganic carbon composites.

Iron oxide containing materials, maghemite nanosorbent was evaluated by Etale et al. (2016) in multicomponent system for the adsorption of heavy metals, The adsorption efficiencies in the same study was observed to increase in the following order $U < Mn < Cu$. Furthermore, in the same study, factors such as competition from H⁺ and changes in adsorption surface charge were reported to have an influence on the adsorption performance. Oyewo et al. (2018) reported on the application of nanosorbent banana peels for removal of lanthanides compounds from acid mine water. It was concluded from the study (Oyewo et al. 2018) that a high surface area and particle size reduction improved banana peels sorption capacity for removal of radioactive lanthanides from synthetic and real mine water. Another class of nanoparticles involving metal oxides include metal oxide nanoparticles (MONPs) and is desired for its high adsorption capacity (Masoumi et al. 2019; Xu et al. 2018). Moreover, in the majority of cases, the metal oxide nanoparticles' surfaces are amphoteric indicating that it can function as either electron donors or acceptors (Wang et al. 2019a, b). Another study on MONPs was conducted by Gugushe et al. (2019) where poor adsorption capacities were obtained. Probably

Table 2 Adsorption capacities of some studied nanoparticles for heavy metals in AMD

Nanoparticle	Target metal(s)	Sorption capacity (mg g ⁻¹)	Reference
Banana peels	La	47.8	Oyewo et al. (2018)
	Gd	52.6	
MWCNT-Fe ₂ O ₃ @Zeolite	As (III)	20.4	Gugushe et al. (2019)
	Hap (g-C ₃ N ₄)	230	
Fungi	Zn (II)	130	Beygli et al. (2019)
	Pb (II)	130	
	Mn(II)	260	
Maghemite	Ni (II)	97.7	Shakya et al. (2018)
	Cu (II)	108.7	
	Mn (II)	208.3	
ZSNZVI	U (III)	20.3	Etale et al. (2016)
	As (II)	12.84	
	Cd (II)	62.02	
HNO ₃ -treated MCN	Pb (II)	85.90	Li et al. (2018a, b)
	Ni (II)	17.86	
	U (VI)	87.38	
Br-PADAP	Cr (VI)	1.48	Mobasherpour et al. (2012)
OS	Cr (VI)	1.48	
OS-nZVI	Cr (VI)	2.54	
OS-nMG	Cr (VI)	4.11	Khamirchi et al. (2018)
MPTS mesoporous silica	Hg (I)	479	
			Vilardi et al. (2018)
			Vilardi et al. (2018)
			Yan et al. (2019)

Magnetic multi-walled carbon nanotubes@zeolite (MWCNT-Fe₂O₃@Zeolite), Hydroxyapatite modified graphitic carbon nitrite (Hap (g-C₃N₄), Zeolite-supported nanoscale zero-valent iron (ZSNZVI), Nitric acid treated multiwalled carbon nanotubes (HNO₃-treated (MCN)), 2-(5-Bromo-2-pyridylazo)-5-(diethylamino) phenol impregnated on multiwall carbon nanotubes (Br-PADAP MWCNTs), Olive stone-Nanozero-valent iron and carbon nanotubes composite (OS-ZVI), Calcined Magnesium Aluminium hydroxides—(Cal. MgAl OH)

problems such as agglomeration which leads the nanoparticles to have a reduction in its nano-intergrity, contributed to the inefficient adsorption performance (Gugushe et al. 2019).

The majority of the interactions between the nanomaterials and the adsorbent of interest involve electrostatic interaction. For example, below the point of zero charge of the nanosorbent, physical adsorption is dominant whereas above the point of zero charge electrostatic interaction becomes more prominent as shown in Fig. 1 (Nqombolo et al. 2019).

This claim is supported by the results obtained by Beygli et al. (2019) who also observed that adsorption of AMD derived heavy metals such as Cu²⁺, Zn²⁺ and Pb²⁺ onto hydroxyapatite modified graphitic carbon nitrite (Hap (g-C₃N₄)) could be re-used for up to 12 cycles. Similarly, Vilardi et al. (2018) observed that the olive stones coated with iron-based nanoparticles could be used for up to 5 times.

Magnetic multi-walled carbon nanotubes@zeolite (MWCNT-Fe₂O₃@Zeolite), Hydroxyapatite modified graphitic carbon nitrite (Hap (g-C₃N₄), Zeolite-supported nanoscale zero-valent iron (ZSNZVI), Nitric acid treated multiwalled carbon nanotubes (HNO₃-treated (MCN)), 2-(5-Bromo-2-pyridylazo)-5-(diethylamino)phenol impregnated on multiwall carbon nanotubes (Br-PADAP MWCNTs), Olive

stone-Nanozero-valent iron and carbon nanotubes composite (OS-ZVI), Calcined Magnesium Aluminium hydroxides—(Cal. MgAl OH).

Magnetic sorbents

Nanosized materials exhibiting magnetic properties have attracted interest among researchers. These novel class of materials exhibit desired properties such as high surface area and superparamagnetism, which result in high adsorption efficiency and high removal rate of contaminants and fast separation of adsorbent from contaminated solution using an external magnetic field which is a major setback of non-magnetic nanosorbent materials (Chowdhury and Yanful 2010). To circumvent this problem, researchers focused on altering the magnetic properties by incorporating iron oxides such as magnetite (Fe₃O₄) and maghemite (γ-Fe₂O₃) to improve separation from the solution and the adsorption capacity. Briso et al. (2018) reported on good separation of the magnetic coated Fe₃O₄ nanostructured calcium silicate hydrate (mag-NanoCSH) and manganese oxide (mag-MnO₂) from solution. It was noted that the presence of iron in the nanosorbent had a major role in the removal of As through formation of water insoluble and stable double iron

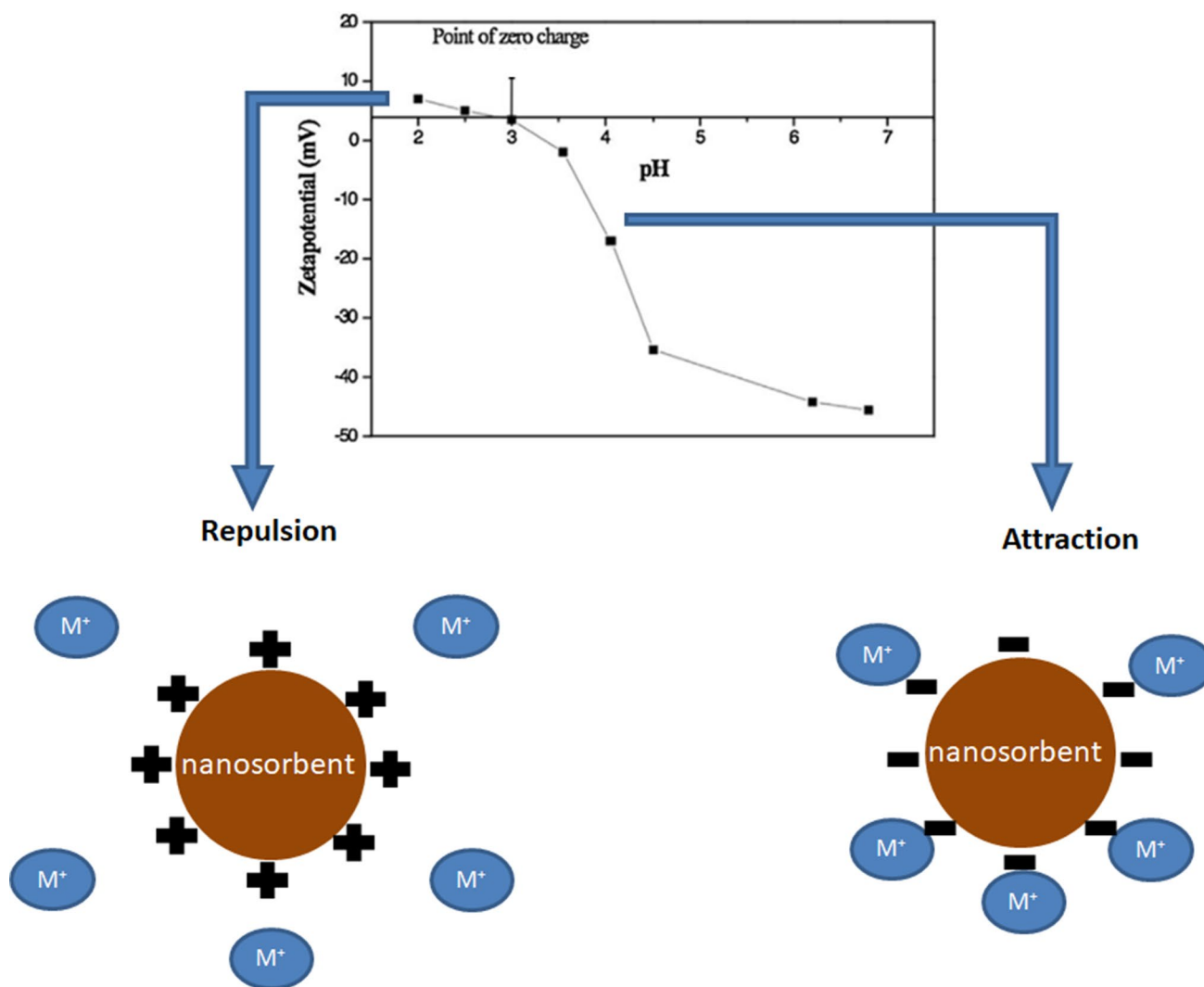


Fig. 1 Repulsion and attraction of a heavy metal (M^+) occurring below and above the point the point of zero charge on the surface of the nanosorbent, respectively

and calcium arsenate salts (Briso et al. 2018). In another study, Barrera et al. (2017) reported nanostructured calcium silicate hydrate, unmodified or modified with Fe (III), for removal of heavy metals such as Cd (II), Cu (II) and Zn (II) from acidic aqueous solutions. Some of the studied magnetic nanosorbents are shown in Table 3. Several studies on adsorption of highly toxic compounds such as Hg (II) and Cd (II) have been done using functionalized magnetic nanoparticles as seen with the study by Maia et al. (2019). The authors reported nanoparticles that L-cysteine functionalized oxyhydroxide (Cys- δ -FeOOH) had a greater adsorption capacity than δ -FeOOH to a lower point of zero charge for Cys- δ -FeOOH, permitting for greater adsorption of Hg (II). Additionally, one of the highest reported adsorption capacities is that for chitosan functionalized nanoparticles (CMNP) as studied by Christopher et al. (2017), which was also a

functionalized magnetic nano-adsorbent (Table 3). Multi-component studies were also evaluated for these magnetic sorbents, for instance the adsorption onto carboxylated magnetic iron oxide nanoparticles as studied by Xu et al. (2019) was shown to follow the order $Cd^{2+} > Cu^{2+} > Pb^{2+} > Ni^{2+}$. An additional feature of these compounds is their regeneration capability. Adsorption of heavy metals to the iron oxide coated sorbent is a pH dependant process, the used nanosorbent can be regenerated by adjusting the pH (Gugushe et al. 2019). Several studies indicated that even after the regeneration process, the adsorption capacity is maintained (Badruddoza et al. 2013; Gugushe et al. 2019).

Table 3 Adsorption capacities of some studied nanoparticles for heavy metals in AMD

Magnetic nanoparticle	Target metal(s)	Sorption capacity (mg g ⁻¹)	Reference
Acac-C@Fe ₃ O ₄	Hg (II)	98.0	Ma et al. (2019)
	Cr(III)	151	
Cu(tpa).GO	Cu (II)	180	Rahimi and Mohaghegh (2017)
	Cd(II)	95	
	Fe(II)	110	
CNZVI	Cd (II)	124	Lu et al. (2015)
Fe ₃ O ₄	As (III)	0.70	Sahu et al. (2017)
CMNP	Pb (II)	498	Christopher et al. (2017)
Fe ₃ O ₄ -CS@BT	Cr (VI)	62.1	Feng et al. (2019)
δ-FeOOH	Hg (II)	35.0	Maia et al. (2019)
Cys-δ-FeOOH	Hg (II)	217	
Fe ₃ O ₄ @EDTA	Ag (I)	71.0	Ghasemi et al. (2017)
	Cd (II)	169	
MNP-COOH	Pb (II)	177	Xu et al. (2019)
	Cu (II)	41.9	
	Cd (II)	58.2	
	Ni (II)	25.8	
Fe ₃ O ₄ @CMC@ZIF-8-OH	Rb (I)	109	Tian et al. (2019)
MAC/clay composite	Cu (II)	96.1	Le et al. (2019)
	Ni(II)	84.8	
Fe ₃ O ₄ @APS@AA-co-CA MNPs	Cd (II)	29.6	Ge et al. (2012)
	Zn (II)	43.4	
	Pb (II)	166	
	Cu (II)	126	
Tannic-Ni _{0.6} Fe _{2.4} O ₄	Pb (II)	139.8	Wang et al. (2019a, b)
	Cu (II)	70.7	
IO-NPs	Cu (II)	188	Ercan et al. (2019)
Fe ₃ O ₄ @SiO ₂ @PEI-NTDA	Pb (II)	285	Jia et al. (2019)

*Chitosan-stabilized nanozero-valent iron (CNVI), Magnetic Fe₃O₄-chitosan@bentonite (Fe₃O₄@BT), Superparamagnetic iron oxide (SPION), L-cysteine functionalized oxyhydroxide (Cys-δ-FeOOH), Acetylacetonate functionalized magnetic carbon microspheres (Acac-C@Fe₃O₄), Chitosan functionalized nanoparticles (CMNP), Carboxymethyl cellulose (CMC), Magnetic activated carbon nanoparticles (MACP), Fe₃O₄@SiO₂ nanoparticles with polyethylenimine and 1,4,5,8-naphthalenetetracarboxylic dianhydride, (Fe₃O₄@SiO₂@PEI-NTDA), Iron oxide nanoparticles (IO-NPs), Biogenic iron compound (FeCO₃/Fe₃O₄/Fe₃(PO₄)₂·8H₂O), magnetic activated carbon/clay composite (MAC/clay composite), Fe₃O₄ magnetic nanoparticles (MNPs) modified with 3-aminopropyltriethoxysilane (APS) and copolymers of acrylic acid (AA) and crotonic acid (CA) (Fe₃O₄@APS@AA-co-CA MNPs), Tannic-functionalized nickel-iron oxide (Tannic-Ni_{0.6}Fe_{2.4}O₄), Iron oxide nanoparticles (IO-NPs)

Ion imprinted polymers as adsorbents

Ion imprinted polymers (IIPs) are highly distinguished materials. Some of the attractive features of IIPs include thermal stability (Ren et al. 2008), the potential to segregate and preconcentrate the analyte from other competing ions (Darroudi et al. 2020). This has led to the substantial usage of IIPs in separation methods and membrane based technologies (Piletsky et al. 1999; Lu et al. 2018; Sun et al. 2017), this includes solid phase extraction (Hennion 1999; Zhu et al. 2019), the production of sensors (Prasad and Jauhari 2015; Bojdi et al. 2015) and as stationary phases in high

performance liquid chromatography (Bitas and Samanidou 2020). The design of IIPs is adopted from that of molecularly imprinted polymers (MIPs), the imprinted regions imitate the 'lock and key' mechanism of enzymes, in selective extraction or removal of the ion template(s) in the presence of competing ions. Other researchers evaluated eco-friendly IIPs through the inclusion of chitosan sorbent structure. Li et al. (2019) looked at mixed solutions of Pd (II) and Cd (II) for adsorption onto double-imprinted electrospun cross-linked chitosan nanofibers (Pd/Cd-DIECCNs) and reported adsorption capacities of 567 and 371 mg g⁻¹ for Pd (II) and Cd (II), respectively. Similarly IIPs bio-nanomaterials were

Table 4 Adsorption capacities of reported ion imprinted nanosorbents

Ion imprinted polymer(IIP)	Target metal(s)	Sorption capacity (mg g ⁻¹)	Reference
Pb-Cs IIP	Pb (II)	136	Gatabi et al. (2020)
NIP	Ni (II)	125	Kumar et al. (2019)
Pd/Cd-DIECCNs	Pd (II)	567	Li et al. (2019)
	Cd (II)	341	
Ag ⁺ - PHEMAC	Ag (I)	49.3	Şarkaya et al. (2019)
Fe(III)-IIP	Fe (III)	114	Zhu et al. (2019)
Pb-IIPs	Pb (II)	49.5	Balouch et al. (2019)
IIP-MAA	Co (II)	106	Yosuf et al. (2019)
Sm(III)-IIP	Sm (III)	14.9	Masoumi et al. (2019)
Co-IIP	Co (II)	95.4	Biswas et al. (2019)
Cu-IIP	Cd (II)	103	Biswas et al. (2019)
NiIMS1	Ni (II)	22.9	He et al. (2018)
NiIMS2	Ni (II)	20.8	He et al. (2018)
G-HQ-C IIPs	Cu(II)	111	Wang et al. (2019a, b)
Co(II)-IIP	Co (II)	175	Yuan et al. (2018)
IIMMs	Li (I)	27.1	Sun et al. (2017)
Cr(VI) IIP	Cr (VI)	56.1	Taghizadeh and Has-sanpour (2017)
IIP@SiO ₂ @Fe ₃ O ₄	As (V)	104.7	Sadani et al. (2020)

Pb-Cs (Chitosan lead ion imprinted polymer network), NIP (Nickel imprinted polymers), Pd-ATU (Palladium allyl thiourea), Pd/Cd-DIECCNs (Pb(II)-Cd(II) double-imprinted electrospun crosslinked chitosan nanofibers), Ag⁺-imprinted PHEMAC poly(hydroxyethylmethacrylate-N-methacryloyl-L-cysteine, Ni-SC (Ni(II) ions imprinted seaweed-chitosan), CA/CS IM (Cellulose acetate /Cellulose Chitosan imprinted membrane), NIP- Nickel ion imprinted polymer, Ni²⁺ imprinted mesoporous organosilica materials (NiIMS1) and MCM-41 (NiIMS2)), G-HQ-C IIPs (eco-friendly gelatin (G), 8-hydroxyquinoline (HQ) and chitosan (C) ion imprinted polymers), ion imprinted macroporous membranes (IIMMs) IIP@SiO₂@Fe₃O₄

reported by Xu et al. (2015) and Gatabi et al. (2020). Lately, further upgrades on chitosan containing IIPs have been made to better selectivity and adsorption efficiency (Pakdel and Peighambaroust 2018). Shakerian et al. (2016) evaluated chitosan based IIPs in solid-phase extraction of lead ions. Moreover, chitosan containing IIPs have also been described as useful scavengers of trace metals in conditions where matrix effects tend to limit the adsorption process (Zhang et al. 2015, 2019). Besides the advantages of IIPs, these materials also come with various constraints in particular; deeply placed imprinted cavities which make binding regions beyond reach; poor extraction of template bound analyte ions; reduced transfer of analyte ions, non-uniform binding spots and template bleeding. Hence, researchers, in an effort to improve adsorption and selectivity of target ions, focused on surface based IIPs (Jia et al. 2018; Masoumi et al. 2019; Tavengwa et al. 2015b). Similarly, Zheng et al. (2020) evaluated surface IIPs nanocrystals in adsorption of Dy (III) and achieved adsorption capacities of 22.57–34.03 mg g⁻¹. Not much, to date, is known on the regeneration of IIPs for adsorption of heavy metals, however a study by Sadani et al. (2020) evaluated the recyclability of IIP@SiO₂@Fe₃O₄ and reported that this material could be re-used for up to

four times for adsorption of As (V). More examples of the application of IIPs as heavy metal adsorbents are given in Table 4.

Pb-Cs (Chitosan lead ion imprinted polymer network), NIP (Nickel imprinted polymers), Pd-ATU (Palladium allyl thiourea), Pd/Cd-DIECCNs (Pb(II)-Cd(II) double-imprinted electrospun crosslinked chitosan nanofibers), Ag⁺-imprinted PHEMAC poly(hydroxyethylmethacrylate-N-methacryloyl-L-cysteine, Ni-SC (Ni(II) ions imprinted seaweed-chitosan), CA/CS IM (Cellulose acetate /Cellulose Chitosan imprinted membrane), NIP- Nickel ion imprinted polymer, Ni²⁺ imprinted mesoporous organosilica materials (NiIMS1) and MCM-41 (NiIMS2)), G-HQ-C IIPs (eco-friendly gelatin (G), 8-hydroxyquinoline (HQ) and chitosan (C) ion imprinted polymers), ion imprinted macroporous membranes (IIMMs) IIP@SiO₂@Fe₃O₄.

Conclusion

Over the past several decades, environmental laws set by WHO and other environmental monitoring bodies have become stricter, necessitating compliance with acceptable

concentrations of contaminants in water, particularly heavy metals. Hence, intensive research was undertaken to investigate suitable remediation methods for AMD derived heavy metals. Of the techniques studied, the adsorption technique was proven to be a popular technique due to its simplicity, affordability and reusability of the material. Typical desirable adsorbents such as nanosorbents exhibit high ion exchange capacity, high porosity, large surface area and adequate specific adsorption sites. Conventional materials, such as zeolites do not adequately satisfy the desired characteristics of typical adsorbents. This review paper provided a current perspective of an emerging class of nanosorbents. These included nanomaterials such as chemically modified iron oxide (Fe_2O_3) magnetic adsorbents which have shown to have superior adsorbent characteristics in comparison to unmodified nanomaterials, in that they permit for ease of separation of adsorbent from the analyte solution. Lately, a more versatile class of nanosorbents in ion imprinted polymers have attracted immense interest due to its high selectivity and its ability to preconcentrate heavy metal ions present in trace concentrations or from complex matrices.

Challenges and future perspectives

A major setback of nanomaterials is the tendency to form agglomerates in the aqueous solution through various surface forces, limiting the adsorption capacity. Hence, small scale nano-engineering studies investigating the surface chemistry of nanomaterial by incorporating function groups such as $-\text{OH}$, $-\text{NH}_2$ and $-\text{COOH}$ have been done with the aim of reducing the extent of agglomeration. Another limitation of nanomaterials particularly metal oxides, is their toxicity in the analyte solution. This implies that large scale application of metal oxide nanosorbents for treatment of heavy metals in AMD could result in cytotoxicity and DNA damage to plants and microorganisms. For instance, studies indicate that gram negative bacteria are susceptible to metal oxide nanomaterials such as ZnO (Dimapilis et al. 2018; Mirza et al. 2019). Additionally, 21 virulence genes of *Legionella pneumophila* were observed to down-regulate after exposure to CuO nanoparticles (Mou and Leung 2017). Furthermore, *Escherichia coli* when exposed to gold nanoparticles were found to down regulate the energy-metabolism and transcription (Cui et al. 2012). Hence, the toxicity of metal oxides towards the environment limits the application of these nanosorbents to the lab, which makes them impractical.

With regards to metal oxides, emphasis needs to be placed on the synthesis of nanomaterials through green techniques. Further, research needs to be directed at the recyclability of other types of nanomaterials, with the exception of magnetic nanoparticles. Magnetic nanoparticles have been proven to be effortlessly separated from water by inducing external

magnetic fields. More studies need to be done in exploring the surface chemistry of nanosorbents. The flexible surface structure of nanosorbents can pave the way for a host of various functional groups to be explored and incorporated onto nanomaterials. This in turn allows for potentially improved nanosorbents with good adsorption capacities, stability and recyclability.

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Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

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