

## Review Article

# Review on Advanced Nanomaterials to Remove Heavy Metals from Waste Water

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## Abstract

The pollution in water by heavy metals is a serious global issue and it becomes major threat for ecological system. Nano technology is an emerging field for various industrial applications. New materials with enhanced functionality are required to adsorb heavy metals from wastewater. This paper reviews, advanced nano-materials that have been used effectively to adsorb heavy metal ions. Different types of nano-materials are briefly discussed with their adsorption capacities. These nano-materials have been used to overcome conventional water filtration techniques. Efficiency of the heavy metal ion adsorption using nano-materials is very high while comparing conventional methods of filtration.

**Keywords:** Nano materials; Heavy metal; adsorption; waste water; efficiency.

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## Introduction

During the latest century, environmental pollution by heavy metals has become serious global problem. Heavy metal discharge into the environment is increasing day by day due to fastest development of industries such as metal plating facilities, mining operations, fertilizer industry, tannery industry, battery industry, paper industry, pesticides manufacturing industry, etc., [1, 2]. Heavy metals are major pollutants in marine, ground, fresh-water and even in treated effluent. Toxic heavy metals like chromium (Cr), mercury (Hg), arsenic (As), cadmium (Cd), lead (Pb), zinc (Zn), and copper (Cu) can cause severe threat to living organisms and agriculture because they are non-degradable and has capacity to accumulate in high concentrations in the living systems [3]. The toxicity of heavy metals depends on several factors including the dose, route of exposure as well as the genetics, gender, age and nutritional status of exposed individuals [4]. Heavy metals generally possess a specific density of more than 5 g/cm<sup>3</sup> and seriously affect the environment and living organisms [5]. **Table 1** reveals the maximum contaminant level of toxic metals. Once the heavy metals get deposited on the living organism, they directly cause many health effects including, cancer, organ damage, nervous system damage, development of autoimmunity, rheumatoid arthritis, damaging of the kidneys, circulatory system, and in extreme cases the deposition of heavy metals may cause death too [6].

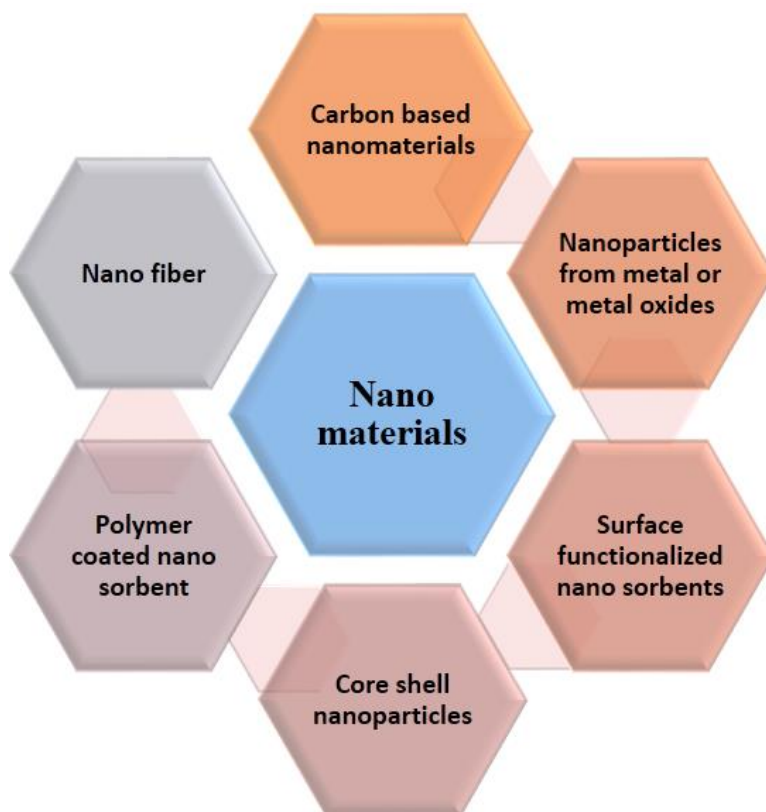
**Table 1** The Maximum Contamination Level standard and toxicity for the most hazardous heavy metals [11-13]

Heavy metal	Toxicities	MCL(mg/L)
Arsenic (Ar)	Leukemia, cancers, gene damage, anemia and neurobehavioural disorders.	0.050
Cadmium (Cd)	Human carcinogen, pulmonary adenocarcinomas, muscle cramps and abdominal pain.	0.01
Chromium (Cr)	Damage of reproductive system, carcinogenic, gene damage and skin allergic reactions.	0.05
Lead (Pb)	Damage of nervous system, reduced birth-weight, damage of kidneys and nervous system	0.006
Mercury (Hg)	Rheumatoid arthritis, and damage of kidneys, carcinogenic, and nervous system	0.00003

Metal toxicity has high impact on soil and ecosystems. Plants growing in metal-polluted sites exhibits growth reduction, altered cellular metabolism, lack of development, early maturation of plant. Some metals including copper, manganese, cobalt and zinc are considered to be essential for plant growth. These trace metals takes part in various functions such as redox reactions, electron transfer and structural functions in nucleic acid metabolism [7,8]. The other metals like Hg, Ag, Pb, Ni comes under non-essential elements, which are highly toxic in nature. These heavy metals can penetrate into ecological food chain through metal polluted water, soil and environment [9,10].

Nano-materials are promising candidate in removing heavy metal ions at low concentrations with high selectivity and adsorption capacity. Nano-materials can exhibits a number of physicochemical properties such as huge surface to volume ratio, high porosity, high aspect ratio and tunable pore volume [14]. Figure 1 shows the various

nanomaterials. The main advantage of using nano-materials, as opposed to conventional techniques is the need of less pressure to pass water across the filter [15]. Nowadays several conventional techniques have been used to remove heavy metals from wastewater such as chemical precipitation [16], ion-exchange [17], adsorption [18], membrane filtration [19], electrochemical treatment technologies [20], etc. Moreover, these methods also have advantages, disadvantages and limitations. Adsorption is based on sorption mechanism. It has certain merits over conventional methods such as low energy, less maintenance costs, the simplicity and the reliability. The efficiency of adsorbents depends on size of adsorbents and properties of adsorbents to remove the heavy metals. Membrane filtration processes relatively simple, easy to handle and requires low space. The disadvantages are high membrane cost and fouling [21, 22].



**Figure 1** Different type of nanomaterials for heavy metal ion adsorption

This article is an extensive review on usage of various nanomaterials in different forms and its effect on adsorption of heavy metals from wastewater. This review also covers materials like carbon-based nanomaterials, nanoparticles from metal or metal oxides, surface functionalized nanosorbents, core shell nanoparticles, polymer coated nanosorbents and nano fibre.

### **Carbon Based Nanomaterial**

Carbon based nanomaterials are used in many forms to adsorb heavy metal ions in recent decades, due to its cost effective, tunable properties and high sorption capacities. Materials include carbon nanotubes, graphene and fullerene are promising candidates to remove heavy metals. Table 2 shows the maximum adsorption efficiency of heavy metals on various nano adsorbents. The CNTs are effective candidates for the adsorption of divalent metal ions [23, 24]. Gaurav Bhanjana et al [25] have reported multi-walled carbon nanotubes (MWCNTs) were prepared by chemical vapour deposition (CVD) technique to remove cadmium metal ion and the results show that maximum adsorption capacity of 181.8 mg/g which has good removal efficiency comparing with single-walled carbon nanotubes (10.86 mg/g). Divyansh and Sanjay [26] proposed carbon nanotubes have tendency to remove  $Mn^{+7}$  ions. The result shows that the decrease in metal ion concentration is purely based on increase in time period. The initial 1 hour mixing time gives 20 % efficiency. After 5 hours mixing time, the  $Mn^{+7}$  concentration was brought down to 94 % efficiency.

Due to the further development to enhance the sorption capacities, CNTs were encapsulated with magnetic nanoparticles [27], coated with other metal ions [28], and functionalize with organic compounds [29]. Yue Liu et al [30], used multi walled carbon nanotubes functionalized with biomolecule L-cysteine to remove cadmium. The result

shows that adsorption is rapid and the entire quantitative adsorption of  $\text{Cd}^{2+}$  take place in only 60 min. Functionalized MWCNTs being encouraging as selective sorbent to successfully remove heavy metals in environmental and biological samples. Meanwhile, Krystyna and Michal [31] synthesized three different carbon materials: Activated carbon, carbon nanotubes, and carbon-encapsulated magnetic nanoparticles. The results reveals that carbon-encapsulated magnetic nanoparticles attain high sorption efficiency and large sorption capacities than the other two materials to remove heavy metals from aqueous solutions.

Graphene is another effective carbon material used as nanosorbent, due to its large surface area, high-speed electron mobility, high young's modulus, electrocatalytic activities, good thermal conductivity etc. Arthi Gopalakrishnan et al., [32] succeeded in removing heavy metal ions such as lead, nickel and chromium from pharma effluents using graphene-oxide(GO) nanosheets. Adsorption isotherm and kinetics shows that 70 mg GO removes all the heavy metal ions effectively with pH of 8 and with very low conductivity. Vishal et al., [33] synthesized graphene based composites ( $\text{ZnO-GO}$  and  $\text{TiO}_2\text{GO}$ ) to adsorb toxic metal ions Pb, Hg, Co, Cr and Cd. They suggested that  $\text{ZnO-GO}$  is good nanoadsorbent and are effective than  $\text{TiO}_2\text{-GO}$ .

## Nanoparticles from Metal or Metal Oxides

Metal oxide nanoparticles are used as adsorbent to remove heavy metal ions due to cost effective, non-toxic synthesis route, easy fabrication and environmentally friendly nature [34]. T. Sheela et al., [35] used ZnO nanoparticles to remove  $\text{Zn}^{2+}$ ,  $\text{Cd}^{2+}$  and  $\text{Hg}^{2+}$  ions from aqueous solutions. Adsorption capacity of ZnO has been studied based on effects of pH, initial concentration, and temperature and contact time. The Langmuir model showed significant effects and obtained maximum adsorption capacities of 714, 387 and 357mg/g for mercury, Cadmium and zinc ions respectively. Susan et al., also [36] prepared ZnO nanoparticles using zerumbone extract to remove Pb(II) ions. The adsorption kinetics models and the thermodynamics of adsorption proved that using of zerumbone extract achieved maximum adsorption efficiency of  $\text{Pb}^{2+}$ . It was found to be 19.65 mg/g under pH of 5 in aqueous solution.

Shokati Poursani et al., [37] achieved uniform size and shape of nano- $\text{TiO}_2$  particles by sol-gel method. Nano- $\text{TiO}_2$  particles have great potential to adsorb  $\text{Pb}^{2+}$  ions from aqueous solutions. After desorption process particles can be reused with high efficiency. Nan Zhang et al., [38] studied Nano- $\text{TiO}_2$  for effective removal of heavy metals and it can be used to adsorb heavy metals fast. The adsorption conditions were optimized including pH, adsorption condition and elution condition.

Xiaoming et al., [39] reported that manganese oxide adsorbed Cd(II) ions from aqueous solution. Langmuir adsorption model revealed maximum adsorption capacity 104.17 mg/g at pH 6.0 and 298 K. Based on surface complexation modeling, Mn oxide increases its ionic strength at pH < 5.0 because its negatively charged above pH 3.5 which could be helpful in adsorbing positively charged Cd(II) ions. Haipeng et al., [40] prepared different crystallographic phases of manganese oxides,  $\alpha$ ,  $\beta$ ,  $\gamma$ , and  $\alpha$ - $\text{MnO}_2$ . Among all those phases, column test revealed that  $\beta$ - $\text{MnO}_2$  has maximum adsorption capacity and durability.

Stietiya and Wang [41] investigated  $\text{Al}_2\text{O}_3$  nanoparticles adsorbing Zn and Cd ions. It can be significantly enhanced by the addition of  $\text{PO}_4$  or HA. Sadott Pacheco et al., [42] proposed alumina silica nanoparticles used as promising nano-adsorbent to remove Cd (II) metal ion. Maximum removal capacity achieved at pH of 6 in 30 min.

Magnetic nanoparticles themselves are used as the active component for removal of heavy metals from water because of its unique properties compared to their bulk form, such as superpara magnetism behavior, high dispersion degree and enhanced catalytic activity [43, 44]. Liliana Giraldo et al [45]., synthesized effective, convenient and low-cost  $\text{Fe}_3\text{O}_4$  nanoparticles by co-precipitation method. The result shows maximum adsorption capacity of 0.140 mmol/g<sup>-1</sup>, 0.160 mmol/g<sup>-1</sup>, 0.170 mmol/g<sup>-1</sup> and 0.180 mmol/g<sup>-1</sup> for manganese, zinc, copper and lead, respectively, under optimized conditions of pH and temperature. The  $\text{Fe}_3\text{O}_4$  nanoparticles creates significant impression of electrostatic attraction between metal ions due to hydrated ionic radius of the metal cations. S.A. Elfeky et al. [46]., obtained magnetic nanoparticles (MNPs) coated with CTAB to adsorb toxic Cr(VI) from wastewater. CTAB enhance the adsorption capacity of  $\text{Fe}_3\text{O}_4$  nanoparticles and the advantage of  $\text{Fe}_3\text{O}_4/\text{CTAB}$  is that low-cost and easy collection from the Cr(VI) contaminated wastewater.

Lisandra de Castro Alves et al. [47] achieved novel iron oxide nanoparticles using co precipitation method. The adsorption data exhibits good and fast adsorption capacity to remove toxic metal ions such as cadmium, copper and nickel. Wherein, cadmium attains the maximum adsorption capacity when compared to other metal ions. Lixia et al. [48] synthesized water-soluble  $\text{Fe}_3\text{O}_4$  nanoparticles using hydrothermal method. Experimental analysis showed that water-soluble  $\text{Fe}_3\text{O}_4$  NPs exhibited relatively high saturation magnetization (83.4 emu g<sup>-1</sup>) and efficiency of water-soluble  $\text{Fe}_3\text{O}_4$  NPs is higher than water-insoluble  $\text{Fe}_3\text{O}_4$  NPs for  $\text{Pb}^{2+}$  and  $\text{Cr}^{6+}$ . Jing Hu et al. [49] prepared various magnetic nanoparticles for the removal of Cr(VI) from electroplating waste water. Among them  $\text{MnFe}_2\text{O}_4$  magnetic nanoparticles can reach equilibrium within 1 h and adsorption efficiency is higher (99.5%) with the shortest adsorption time.

## Surface Functionalized Nano Sorbents

Surface functionalization of nano sorbents enhances or adsorbs specific metal ions from waste water [50]. Zhili Li et al [51], investigated surface-functionalized porous lignin (SFPL) to remove lead ions. Polyethylenimine is a grafting agent to link primary and secondary amine groups to make high-branched structure of SFPL. SFPL nanoparticles with BET surface area of 22.3 m<sup>2</sup>/g and large number of mesopores are effectively used as adsorbent. Rahele Rostamian et al [52] obtained silica nano hollow spheres and then functionalized it with the thiol group to remove toxic ions such as Hg(II), Pb(II) and Cd(II) from water samples. The adsorption isotherms indicates thiol functionalization enhances adsorption capacity towards the metal ions. Thiol-SNHS shows higher adsorption rates towards Hg<sup>2+</sup> ions compared to Pb<sup>2+</sup> and Cd<sup>2+</sup> ions.

Jerina Majeed et al [53]. Proposed cost effective ZnO nanoparticles functionalized by thiol group. Thiol functionalization on ZnO nanoparticles is highly efficient for sorptive removal of Pb<sup>2+</sup> and Hg<sup>2+</sup> from waste-water. Xiaodong Xin et al [54] obtained amine-functionalized mesoporous Fe<sub>3</sub>O<sub>4</sub> nanoparticles which is cost effective, environment friendly and has good reproducibility. Langmuir adsorption model shows maximum adsorption capacities for Pb<sup>2+</sup>, Cd<sup>2+</sup>, and Cu<sup>2+</sup> from 369.0 to 523.6 mg/g. AF- Fe<sub>3</sub>O<sub>4</sub> nanoparticles would be reusable and reduce water treatment expenses. Renu Verma et al., [55] synthesized arginine functionalized magnetic nanoparticles. The result shows the maximum adsorption capacity found to be Cu (II) 172.4, Co(II) 161.2, and Ni(II) 103.0 mg g<sup>-1</sup>. The adsorption capacity is based on amino groups on its surface. M. Anbia and S. Amirmahmoodi [56] prepared amino functionalized mesoporous silica SBA-15 materials to adsorb halophenols. Result has showed that functionalized mesoporous silica SBA-15 had maximum efficiency to remove 4-chlorophenol (CP), 4-bromophenol (BP) and 4-iodophenol (IP) compared to untreated SBA-15. The Langmuir and Freundlich adsorption isotherms reveals the maximum adsorption capacities, which indicates that the amino groups can attain maximum adsorption capacity due to potential charge association.

Kong Xiang et al [57] reported surface functionalized silica nanoparticles prepared by different silane coupling agents. The results show that amine and thiol can be successfully functionalized on silica surface. The sulfur functionalized silica nanoparticles exhibited good adsorption on Cu<sup>2+</sup> and Hg<sup>2+</sup> and amine functionalized silica nanoparticles attains maximum adsorption capacity towards Pb<sup>2+</sup> and Hg<sup>2+</sup>.

**Table 2** Maximum adsorption efficiency of different adsorbate on various nano materials

Adsorbents	Adsorbate	Maximum adsorption efficiency mg.g <sup>-1</sup>	Reference
Chemically modified PAN fibre (PAN-DR-EDTA)	Cu(II)	47.6	[58]
PAN <sub>MW</sub> Thio fibres	Hg(II), Cd(II)	322.8, 350.6	[59]
Thiol functionalized PAN nanofibre	Hg(II)	53.6	[60]
Thiol functionalized silica	Hg(II)	59	[61]
Amino functionalized silica nano holo sphere	Pb(II), Cd(II)	143.295, 49.52	[62]
Carbon nanotube	Cu(II), Pb(II)	3.49, 2.96	[63]
Graphene multilayer nanosheets	Co(II)	67	[64]
Chromium doped nickel oxide nanoparticles	Cd(II), Pb(II) and Cu(II)	5.243, 1.79 and 1.302	[65]
Chitosan coated magnetic nanoparticles	Cu(II)	60.606	[66]
Ni@Mg(OH) <sub>2</sub> core shell Nano composite	Zn(II), Cd(II) and Cu(II)	36.03, 45.02 and 40.49	[67]

## Core Shell Nanoparticles

Core shell nanoparticles are promising candidates in nano materials because it can exhibits two different properties [68]. Chunrong Ren et al [69] constructed amino-functionalized super paramagnetic CoFe<sub>2</sub>O<sub>4</sub>@SiO<sub>2</sub>-NH<sub>2</sub> core-shell nanospheres showing homogeneous and uniform morphology with a thickness of 35 nm. The first cycle of adsorption efficiency found to be Cd(II) 99.85%, Cu(II) 87.80% and Pb(II) 90.10%. Suyue Jin et al [70] synthesized amine-functionalized ferromagnetic Fe<sub>3</sub>O<sub>4</sub>-mesoporous silica (mSiO<sub>2</sub>) core-shell nanoparticles. Optimization of magnetic core size of the amine functionalized nanoparticles enhances the removal efficiency of heavy meal ions. The result shows the microstructures of the Fe<sub>3</sub>O<sub>4</sub>-mSiO<sub>2</sub> nanoparticles with core size 103 nm. Adjusting the core size influences the magnetic susceptibility of nano-adsorbents, more than super paramagnetic property. Optimization of core size enhanced adsorption of heavy metal ions in a shorter time.

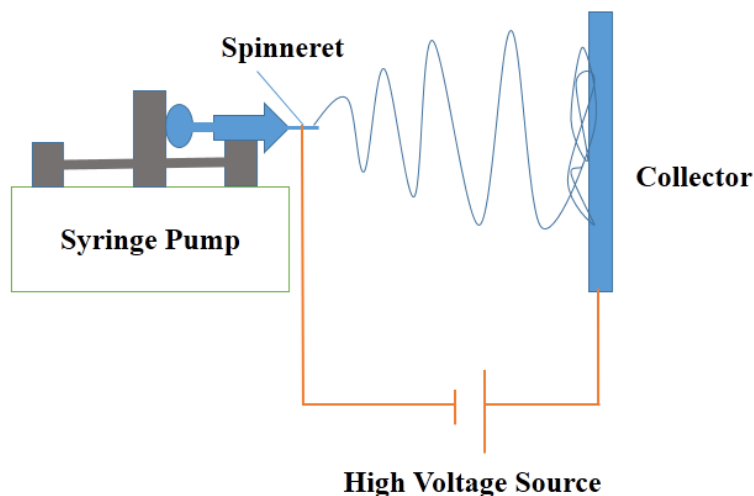
Shushan Hou et al [71] prepared magnetic mesoporous silica microspheres and functionalized with carboxyl groups to enhance adsorption performance. The silica coated magnetic microsphere adsorbent regenerated by acid treatment and reusability of the Fe<sub>3</sub>O<sub>4</sub>@SiO<sub>2</sub>@- mSiO<sub>2</sub>-COOH microspheres slightly decreases in sequential cycles. Na Wu and coworkers [72] synthesized mesoporous titania beads and alginate biopolymer used as template. The

maximum adsorption capacity reaches due to electrostatic adsorption of negatively charged  $\text{HCrO}_4^-$  absorbed by positively charged titania beads meanwhile Cr (VI) reduced to Cr (III) due to surface hydroxyl groups on the titania beads. Jiahua Zhu et al [73] magnetic graphene nanocomposites (MGNCs) were synthesized by thermo decomposition process. The core shell nanoparticles composed of crystalline iron core, iron oxide inner shell and amorphous silica outer shell. These are highly stable even when immersed in 1M HCl aqueous acid so it could attain maximum adsorption capacity at relatively lower pH value solutions. Siping Ji et al [74] reported that core-shelled  $\text{Fe}_3\text{O}_4@\text{C}$  hybrid nanoparticles via solvothermal method. The result shows the microsphere structure and the magnetic saturation value reached at 53 emu/g. The maximum adsorption efficiency is attained at 100% for  $\text{Pb}^{2+}$ , 99.2% for  $\text{Cd}^{2+}$ , 96.6% for  $\text{Cu}^{2+}$ , and 94.8% for  $\text{Cr}^{2+}$ , respectively.

### Polymer Coated Nano Sorbent

Ahmed fawzy el-kafrawy et al [75] prepared modified b-cyclodextrin coated magnetic particles to adsorb metal ions. It is pH dependent due to complexation nature of cyclodextrin. Gui-yin Li et al [76] synthesized  $\text{Fe}_3\text{O}_4$  nanoparticles coated with chitosan which showed higher adsorption of metal ions after 45 min. While synthesizing magnetized nanoparticles the super para magnetism and saturated magnetism was reached as  $21.5 \text{ emu g}^{-1}$ . Several polymers are used as a carrier to embedded magnetized nanoparticles such as PMMA, PEDOT, chitosan, alginate etc., Adil denizli et al [77] prepared mPMMA for the removal of selective metal ion. The chemically modified magnetic PMMA micro beads attains a maximum adsorption capacity of 201 mmol/g for Cu (II), 186 mmol/g for Pb (II), 162 mmol/g for Cd (II), and 150 mmol/g for Hg (II). Arameh Masoumi et al., [78] suggested that modified methyl methacrylate maleic anhydride (MMA) was synthesized by radical polymerization and analyzed the effect of various parameters such as pH, reaction time, adsorbent dosage and initial metal ion concentration. In the development of polymer-coated nanoparticles, core will be metal oxide or magnetic nano sorbents and shell will be a polymer layer. S. Nasirimoghaddam et al [79] developed chitosan coated magnetic nanoparticles for the removal of  $\text{Hg}^{2+}$  ions from industrial waste water. Chitosan-bound  $\text{Fe}_3\text{O}_4$  nanoparticles showed higher adsorption efficiency in aqueous solution.

### Nano Fibre



**Figure 2** Schematic diagram of electrospinning set up

Nanofibre spinning is a promising technique to develop high efficiency water filtration membranes [80]. Quan Feng et al [81] prepared blended Polyacrylonitrile/cellulose acetate (PAN/CA) composite nanofibrous membranes and it was chemically modified into amidoxime polyacrylonitrile/regenerate cellulose (AOPAN/RC) composite nanofibrous membranes. The modified composite membrane showed good reusability and desorption (~nearly 80%) after their first desorption. PAN polymer is widely used to prepare nanofibrous membrane and those fibres are chemically modified with various reagents such as ethylenediamine (EDA), ethylene glycol (EG) or diethylenetriamine (DTA) [82-84]. ZnO,  $\text{TiO}_2$  were embedded on nanofibrous membrane to enhance the rate of metal ion adsorption. Many chemical modifications of PAN nanofibre were attained such as Amidoxime PAN [85, 86], phosphorylated polyacrylonitrile-based (P-PAN) nanofibres [87] and thiol functionalized PAN [88]. Among them Amidoxime PAN widely used to improve the efficiency of nanofibre membrane [89-91].

In general, Fe (III), Cu (II), Cd (II), Zn (II), Ni (II), Ag + are adsorbed by all the above process. However, the rate of adsorption of individual metal ions varies depending on the polymers being selected, nanoparticles embedded on them and type of chemical modifications.

### Commercialization and practical utility

The review highlights the heavy metal ion removal efficiency of above-mentioned nano materials. In future, large amount of commercialized of nano materials or nano sorbents required to remove heavy metal from large water bodies such as rivers, lakes. Currently nano materials lagging the practical utility due to its scale up process and agglomeration of nano materials in large scale [92]. Researchers have been working to scale up large scale production and practical utility.

### Conclusion

In this review, an overview was given on the different types of nanomaterials and their maximum adsorption capacities and desorption rates. Nano materials gave significant contribution to adsorb heavy metals from waste water. The main advantages of nano-sorbents are that their properties can be modulated based on the adsorbate considered. Even though there are some challenges like fouling, reusability and reliability, these nanomaterials can still be considered as effective sorbents at the cost of high adsorption efficiencies. It is evident from literature survey of 90 articles proved that nanosorbents can effectively adsorb heavy metals from waste-water than conventional techniques.

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