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A review on iron oxide-based nanomaterials towards the adsorption Pb^{+2} ion from aqueous media

Juhi Rath[1], Preetilata Dwivedy[1], Bikramdev Madhi[1], Jayashree Das[1], Barsa Rani Patel[1]

[1*]Department of Chemistry, GIET University, Gunupur-765022, Rayagada, Odisha, India

Abstract—**All living body requires water for their essential needs. In the present era water is contaminated due to various types of pollutants like heavy metals, industries etc. Among all the heavy metals, Lead (Pb^{+2}) ions are a major trouble to the environment and human health too for their toxic nature. This investigation aims to eliminate the noxious Pb^{+2} ions from aqueous media as it is a prime importance to clean the waste water. Researchers used various metal oxide-based nanomaterials for the removal of Pb^{+2} ion using different removal methods. Most of them have used adsorption method for this heavy metal remove from waste water due to cost effective, zero toxic byproducts and reusability. In this paper we have studied about iron oxide-based nanomaterials used as adsorbent modified with polymer, metal oxide, bimetal oxide, activated carbon prepared from different bio-waste. Moreover, this review also compares various parameters of the adsorbents such as pH, contact time, temperature, adsorbent dosage, initial concentration, removal efficiency which were reported by other researchers. This review paper briefly discusses about the various type isotherm along with their maximum adsorption capacity (q_{max}) and kinetic model of the adsorbents in the literature study. The future scope of the toxic heavy metal removal was also specified.**

***Keywords*—Adsorption, Pb^{+2} removal, iron oxide-based nanomaterials**

I.INTRODUCTION

In our ecosystem a lot of basic requirements are there to live a perfect life, Water is one of them. But, in the present era water is getting polluted day by day, which have a very negative impact not only on environment, but also on human health[1]–[3]. Water plays the key role in our life as it is used for many purposes such as industrial use, domestic use, textile use, agricultural purpose and many more[4]. In today's world water resources are getting polluted every day because of global warming, deforestation, industrial and agricultural uses etc. Thus, it should be our priority to remove those hazardous wastes from water[5]. From many literatures study we knew that, the major water pollutants were organic dyes, surfactants, harmful pathogens, heavy metals and pharmaceutical wastes, etc[6]. Amongst all, heavy metals are usually considered to be very dangerous to mankind and ecosystem, due to their high density and high molar mass[7]–[9]. Generally, there are various toxic heavy metals present in polluted water bodies such as Pb^{+2}, As^{+3}, Cr^{+6}, Hg^{+2}[3], [10]. Among all these hazardous heavy metals Pb^{+2} is more toxic towards water system due to its amphoteric nature. Many researchers studied that this Pb^{+2} easily transported through soil and water so that it very badly affects the water body[11]. Pb^{+2} in water causes health issues like anemia, cancer, kidney damage, renal failure, hypertension like dangerous diseases[12]. Higher concentration of Pb^{+2} ions also decreases the male fertility and affects the blood, brain and placenta of pregnant women[13]. Due to this death causing nature of Pb^{+2} ions it should be removed from water bodies and many researchers also worked up on this. There are a lot of advanced method for removal of Pb^{+2} ion from contaminated water such as ion exchange[14], coagulation[15], membrane filtration[16], adsorption[17], ultrafiltration[13], neutralization[18], solvent extraction[19] and chemical precipitation[20]. Out of all these methods most of the researchers adopted the adsorption method as compared to other methods, because it is cost effective, environmental friendliness and works under a wide pH range. To remove Pb^{+2} ions from water the adsorption process required an efficient adsorbent. There are many adsorbents used in Pb^{+2} removals like iron oxide, activated carbon, graphene oxide, but many authors largely used the iron oxide adsorbent in the adsorption process. From the literature study we knew that the iron oxide has higher surface area, high biocompatibility, low cost and quality of magnetic properties so that Pb^{+2} ions were easily eliminated by applying external field. Again, majority of researchers used this iron oxide modified with different materials to enhance the adsorption capacity of the iron oxide. Examples of some modified iron oxide nano-adsorbents are Lingamdinne et al. produced Fe_3O_4 nanomaterials (T- Fe_3O_4) from bio-waste using tangerine peel extract[21], Khazaei et al. produced graphene oxide – Fe_3O_4 nanomaterials (GO- Fe_3O_4-SiO_2)[22], Hashemi et al. synthesized polyaniline-Fe_3O_4-silverdiethyldithiocarbamate (PANI-F-S)[23], Sarojini et al. synthesized Polypyrrole-iron oxide-seaweed nanomaterials (PPy-Fe_3O_4-SW)[24],Zhang et al. prepared magnetic biochar-

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$MnFe_2O_4$ (BC/FM)[25], Alsuhybani et al. produced Fe_3O_4-BDC-AGPA[26], Pelalak et al. prepared graphene oxide (GO) with magnetic oak wood ash (Ash/GO/Fe_3O_4) nonmaterial's[27],Sadeghi et al. manufactured cetyltrimethylammonium bromide and polyaniline polymer-Fe_3O_4[28], Nodeh et al. prepared methyltrimethoxysilane-Fe_2O_3 (Fe_2O_3-MTNOS)[29], Keshvardoostchokami et al. synthesized chitosan-iron oxide nanomaterials (Chitosan-FeO)[30]. In our paper we described different parameters such as initial Pb^{+2} concentrations, pH, time, temperature, adsorbent dosages, which were studied by different researchers. Again, we reported the isotherm model followed by different adsorbents and also studied the maximum adsorption capacity values for different adsorbents described by all authors. Moreover, we studied the different kinetics data followed by different adsorbents in this paper

II. DETAILED STUDY OF IRON-BASED NANOCOMPOSITE FOR REMOVAL OF LEAD HEAVY METAL

Khazaei et al. (2016)[22] studied about the removal of Pb^{+2} ion from waste water. In their experimental study they screening that Pb^{+2} ion was removed by adsorption process where graphene oxide – Fe_3O_4 nanomaterials has been used as an adsorbent. They prepared Fe_3O_4 nanomaterials and modify it with graphene oxide (GO). To analyzed the surface morphology and functional groups present on the iron oxide nanomaterials (GO-Fe_3O_4) they studied it under AFM, UV-Visible, FT-IR, SEM spectroscopy. From this research it was investigated that more than 75% of Pb+2 ions were removed at optimum pH of solution (3.5- 8.5), contact time of 2-30 mins, and initial concentration is 0.5-5 mg/L. This study was well fitted on sips adsorption isotherm and maximum adsorption efficiency was 598.4 mg/g. From kinetic study it was observed that it followed double exponential kinetic equation.

Keshvardoostchokami et al. (2017)[30] synthesized a bio adsorbent modified with nanomaterials of iron oxide for the effective removal of Pb^{+2} ion from noxious water body. In their experimental study they showed that Pb^{+2} ion removed by adsorption process where chitosan-iron oxide nanomaterials (Chitosan- FeO) used as an adsorbent. From the surface morphology study of the synthesized nanomaterials under FTIR, XRD, FE-SEM techniques it was found that the adsorption capacity of this nanomaterial was very high. This technique effectively removed 99.95% of Pb^{+2} ion at optimum pH 3 in room temperature (25ºC), contact time 30 min, initial concentration 10 mg/dm^3 and adsorbent dosage $20dm^3$. This study followed Freundlich adsorption isotherm and maxim adsorption capacity of Chitosan- FeO was 11.69 mg/g. pseudo second order kinetics. This adsorption study also followed pseudo second order reaction. These Chitosan- FeO nanocomposites also remove Cd^{+2} and Ni^{+2} ions from aqueous media. This Chitosan- FeO bio adsorbent can also recyclable and have no negative effect on environment.

Sadeghi et al. (2018)[28] modified magnetic nanomaterials (Fe_3O_4) with cetyltrimethylammonium bromide and polyaniline polymer for the absorptive removal of Pb^{+2} ions from the toxic water body. They studied that this adsorbent was effective towards industrial waste water treatment and they examined it under BET technique to know the exact size of the nanomaterials. The maximum Pb^{+2} ion removed at an optimum pH 9.3 at room temperature, contact time 60 mins, initial concentration 50 mg/L and adsorbent dosage 3mg. This literature study was well described by Langmuir adsorption isotherm and the maximum Pb^{+2} ions removal efficiency was obtained as 111.11 mg/g. This study is well explained by pseudo second order kinetics.

Alsuhybani et al. (2019)[26] conducted an experiment to prepare amino-guanidinopentanoic acid (AGPA) and further modified it with benzene dicarboxylic coated magnetic nanomaterials (Fe_3O_4- BDC) to make Fe_3O_4 -BDC-AGPA. For the morphological and structural study of this magnetic nanomaterial they studied it under Fourier-transform infrared spectroscopy (FT-IR), X-ray diffraction (XRD), transmission electron microscopy (TEM) etc. The maximum amount of Pb^{+2} was removed at a pH 5.6, contact time 180 mins, initial concentration 300 mg/l and adsorbent dosage 20 mg. 92% of Pb^{+2} was removed by this process. This study is well fitted Langmuir adsorption isotherm and maximum adsorption efficiency 157 mg/g. From the kinetic study it is found that this experiment followed pseudo second order kinetics.

Zhang et al. (2019)[25] studied a procedure to prepare biochar from corn straw which was easily available in market. After that they modified magnetic biochar- $MnFe_2O_4$ (BC/FM) and decorated the nanoparticles with two functional groups (-NH_2 and -COOH) for making it more effective towards heavy metals removal. For the study of surface morphology, structure and size of the modified nanomaterials they characterized it using different type of technique like SEM- EDX, XRD, VSM and BET. A effective 99% removal of Pb^{+2} was happened by this process at optimum pH 5 at temperature 25ºC – 45ºC. This literature study was well described by Langmuir adsorption isotherm and maximum adsorption capacity (q_{max}) of this experiment is 154.94 mg/g, contact time 300 minutes. From the literature study it is found that followed by sips kinetic model. Other than Pb^{+2}, and Cd^{+2} ions is also removed by this adsorption method.

Hashemi et al. (2019)[23] experimented about the removal of hazardous Pb^{+2} ion from the water bodies. In their experimental study they have used polyaniline-Fe_3O_4-silverdiethyldithiocarbamate (PANI-F-S)as an adsorbent and 99% of Pb^{+2} ion removed by this process. PANI-F-S also have some effective actions towards the microbes like fungi and bacteria in order to make the water drinkable. For the morphological and structural study of PANI-F-S they studied it under EDAX and XRD spectra. PANI-F-S is a very effective adsorbent as it can remove the heavy metals as well as work against the microorganisms. From this experimental study it is studied that 99% of Pb^{+2} ions were removed at optimum pH of the solution 5, contact time 25 min and initial concentration is 45 μg m/L. Other the Pb^{+2} it also removes some other heavy metals from waste water such as As^{+3}, As^{+5}, Hg^{+2}.

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Table-1

Adsorbent	Method of Pb^{+2} removal	pH	Adsorbent Dosage	Contact Time	Temperature	Initial concentration	Reference
T-Fe_3O_4	Adsorption	4.5	0.625 mg/L	95 min	25ºC	32.5 g/L	[21]
Magnetic GO - SiO_2	Adsorption	3.5-8.5	1-60 mg/L	2-30 min		0.5-5 mg/L	[22]
Magnetic biochar- $MnFe_2O_4$	Adsorption	5		300 min	25º C- 45 º C		[25]
Polyaniline-Fe_3O_4-silver diethyidithiocarbamate (PANI-F-S)	Adsorption	6		25 min	5º C- 50 º C		[23]
polypyrrole-Fe_3O_4- seaweed (PPy -Fe_3O_4 -SW)	Adsorption	5	10 mg	20 min	40ºC	100 mg/L	[24]
Fe_3O_4 -BDC -AGPA	Adsorption	5.6	20 mg	180 min	25ºC	300 mg/L	[26]
Ash/GO/ Fe_3O_4	Adsorption	6	1 g/L	60 min	25ºC	10 mg/L	[27]
cetyltrimethylammonium bromide and polyaniline polymer- Fe_3O_4	Adsorption	9.3	3 mg	50 min	25ºC	50 mg/L	[28]
Spherical Fe_2O_3- MTMOS (methyltrimethoxysilane)	Adsorption	5	150 mg	105 min	25ºC	100 mg/L	[29]
Chitosan- FeO	Adsorption	3	20×10^6 mg	30 min	25ºC	10 mg/L	[30]

The different parameters value (Initial Pb^{+2} concentration, Dosage, pH, Contact time, Temperature) for the removal of Pb^{+2}

Table-2

Comparison of different isotherm, kinetic, removal efficiency and q_{max} of different iron oxide based nano composite

Adsorbent	Isotherm	Kinetics	Removal efficiency (%)	q_{max}	Reference
T-Fe_3O_4	Langmuir	Pseudo second order	95%	100 mg/g	[21]
Magnetic GO - SiO_2	Sips	Double exponential	>75%	598.4 mg/g	[22]
Magnetic biochar- $MnFe_2O_4$	Sips	Pseudo second order			[25]
Polyaniline-Fe_3O_4-silver diethyidithiocarbamate (PANI-F-S)	Langmuir	Pseudo second order	99%	40.3 mg/g	[23]
polypyrrole-Fe_3O_4- seaweed (PPy -Fe_3O_4 -SW)	Langmuir	Pseudo second order	97.25%	333.33 mg/g	[24]
Fe_3O_4 -BDC -AGPA	Langmuir	Pseudo second order	92%	157 mg/g	[26]
Ash/GO/ Fe_3O_4	Langmuir	Pseudo second order	99.67%	47.16 mg/g	[27]
cetyltrimethylammonium bromide and polyaniline polymer- Fe_3O_4	Langmuir	Pseudo second order		111.11 mg/g	[28]
Spherical Fe_2O_3- MTMOS (methyltrimethoxysilane)	Freundlich	Pseudo second order	>80%		[29]
Chitosan- FeO	Freundlich	Pseudo second order	99.95%	11.69 mg/g	[30]

From this literature study, it was found that 95%of Pb^{+2} ions were removed at optimum pH of 4.5, contact time of 95 min, initial concentration of 32.5 g/L and the adsorbent dosage of 0.625 mg/L. This adsorption method followed Langmuir adsorption isotherm along with maximum adsorption capacity of 100 mg/g. From kinetics study it was found that it followed pseudo second order kinetic model.

Nodeh et al. (2020)[29] reported an adsorption study for the removal of harmful Pb^{+2} ions from the toxic aqueous media. They synthesized methyltrimethoxysilane and doped it with Fe_2O_3nanomaterials (Fe_2O_3 -MTMOS) to make it an effective adsorbent. To study about the surface morphology of this synthesized nanomaterials they examined it under various technique such as X-ray diffraction (XRD), Fourier-transform infrared spectroscopy (FT-IR), Field emission scanning electron microscope (FESEM) From this study they described that more than 80% of Pb^{+2} ions removed at optimum pH 5, contact time 105 min, initial concentration 100 mg/L and adsorbent dosage 150 mg. From this study it was known that this it followed Freundlich adsorption isotherm and the pseudo second order kinetics.

Pelalak et al. (2021)[27] studied an experiment about the removal of Pb^{+2} from noxious aqueous media. In their study they modified an adsorbent using graphene oxide (GO) with magnetic oak wood ash (Ash/GO/Fe_3O_4) nanomaterials. To analyze the surface morphology, structure, size of the nanomaterials they studied it under TEM, FESEM, EDX mapping, FTIR, XRD etc. instrument technique. From this literature study, it is determined that a maximum amount (99.67%) of Pb^{+2} removed at an optimum pH 6 at room temperature, contact time 60 min, initial concentration 10 mg/L and the the adsorbent dosage 1g/L. This adsorption study was well described by Langmuir adsorption isotherm and maximum adsorption efficiency 47.16 mg/g. From the kinetic study it is explained that this experiment followed pseudo second order kinetics. Other than Pb^{+2} ion this adsorbent also can remove toxic Cd^{+2} ion from aqueous media and this adsorbent can also be used repeatedly.

Sarojini et al. (2021)[24] studied to synthesize an effective adsorbent for adsorptive removal of deadly Pb^{+2} ions from the aqueous media. They have synthesized Polypyrrole- iron oxide-seaweed nanomaterials (PPy-Fe_3O_4-SW) for the removal of heavy metal as it is reusable and can remove 97.25% of Pb^{+2} ions from waste water. The morphological structure of the synthesized nanomaterials was examined under SEM, XRD, TEM, EDS and FTIR instrument technique to know its structure, size and surface morphology. The maximum amount of Pb^{+2} was removed at optimum of pH 5 and temperature 40^{o} C. The initial concentration, adsorbent dosage, temperature, and pH of adsorption were investigated 100 mg/L, 10mg, 40^{o}C, 97.25 % and pH 5 respectively. PPy-Fe_3O_4-SW nanomaterials is a unique and effective adsorbent as it has high removal efficiency. This study was well explained by Langmuir adsorption isotherm and maximum adsorption efficiency (q_{max}) of this study was 333.33 mg/g. From the above literature study, it is established that it followed pseudo second order kinetics.

Above mentioned adsorbent made up of different functionalization on iron oxide-based nanomaterials. According to this, the adsorption parameters and also their adsorption isotherm, kinetics models with their adsorption capacities and removal efficiency of nanomaterials are mentioned in **Table 1**and **Table 2**.

III. Conclusion

Many investigations have been carried out for effective elimination of Pb^{+2} ions from aqueous media. From various literature studies it has been revealed that Pb^{+2} ions is very toxic and carcinogenic in nature. Due to this many researchers showed interest for removal of Pb^{+2} ion from the aqueous solution via adsorption process because of its environmentally friendly nature and zero production of toxic byproducts. In recent era, many researchers adopted iron-based materials for removal of Pb^{+2} due to its good stability, high adsorption capacity, nontoxicity, low cost and large surface area. Here, to enhance the adsorption capacity of iron oxide, many researchers modified with metal oxide, bimetal oxide, polymer, tangerine peel etc. From adsorption study the removal of Pb^{+2} ions was studied by different parameters pH, adsorbent dosage, initial concentration, contact time and temperature. In our review, we elucidate the comparison between adsorbent which mentioned on the above literature studies. From this study it was found that most of the modified adsorbent adsorbed at acidic pH and among of them two adsorbents were adsorbed at basic pH i.e., 8.5-9.3. Similarly, the optimum temperature of most of the adsorbent were adsorb at 25 °C. In this review we interpret isotherm and kinetic model. From the isotherm study, we get the information that all adsorbent followed Langmuir, Freundlich, and Sips isotherm and among them most of the adsorbent followed Langmuir adsorption isotherm and the maximum adsorption capacity of Magnetic GO - SiO_2 was 598.4 mg/g, polypyrrole-Fe_3O_4- seaweed was 333.33 mg/g and of Fe_2O_4 -BDC-AGPA was 157 mg/g. From kinetics study, it was confirmed that most of the adsorbent followed Pseudo second order model. Again, in this review we differentiate removal efficiency of different iron-based materials and highest removal efficiency was found to be 99.95 % for Chitosan- FeO. Furthermore, this review will assist to specify which iron-based materials is favorable for Pb^{+2} removal and help in revealing the whole picture of iron-based materials for further application in the aqueous media as well as environment.

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