Preparation of Iron Nanoparticles and Composites for Arsenic Removal: An Updated Review

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ABSTRACT

Due to advancement and increasing development in industrialization, heavy metals especially Arsenic (As) may cause an environmental threat because of continuous release of effluents in ground water. Metallic As is hazardous in nature and have severe harmful outcomes on human, aquatic animals, plants and environment. As cause severe lethal impacts on the healthy human as well as environment after appearing into the chain of food. As is one of major cancer causing agent in humans. Though, development of new technology like nanotechnology gives hope for better techniques for As removal from waste water. Preparation of unique, novel and low cost of nanomaterials for environmental applications, detection of pollutant and other uses has drew attention for further considerable. On this note, zero valent iron and iron oxide nanoparticles are observed as the suitable materials for the As adsorption from waste water or ground water. Electrical, ionic interaction, mechanical and physiochemical properties play key role in nanoparticle fabrication and control in desirable morphology. Iron oxide nanoparticles can also be used as catalyst, drug delivery carriers and contrasting agent. Different categories of iron oxide nanoparticles desired shape or topography, and size can be prepared by using different methodology such as sol-gel, co-precipitation, solvothermal reactions and iron oxide composites. Iron oxide nanoparticles have previously shown its efficiency, diversity and reusability in several areas including bio-imaging, drug or gene delivery, catalytic properties, immobilization of industrial important enzymes and removal of dye, phenol and toxic compounds. Present review is dedicated on the preparation of iron oxide nanoparticles and its composites for As metal removal.

KEY WORDS: NANOPARTICLES, IRON, IRON OXIDE, COMPOSITES, ARSENIC, ENVIRONMENT.

INTRODUCTION

Arsenic occurs as in oxides form within dirt, dregs, aqueous solution, and ground water in several part of the globe (Nurmesniemi et al., 2010, Park et al., 2011). Naturally, arsenic occurs more than different 200 distinctive mineral structures. Arsenic appears around in the form of arsenates (60%), sulphides (20%) and sulphosalts, and consequently arsenide, arsenite, silicates and elemental arsenic are the left over 20% (McCarty et al., 2011, Chiban et al., 2012). The well-known Aresenic compounds that naturally occur are arsenite (As(III)) and arsenate (As(V)) (Drewniak et al., 2012). As(V) is the prevalent type of As existing under oxidized surroundings and present as oxyanions of arsenic acid, while As(III) occurs as arsenious acid under mild reducing environments (Fig.1) (Sinha et al., 2013, Campbell et al., 2014, Podgorski, et al., 2020).

Arsenic and related compounds have been recognized as potent carcinogen as per guideline of the International Agency for Cancer Research (IARC) (Karagas et al., 2015).

Hydrothermal activity associated with granitic magma intrusion and orogenesis shapes the primary arsenic containing mineral arsenopyrite (Schindler et al.,2016). Due to the natural geological distribution in the bedrock, the presence of As in natural source water may appear (Bondu et al., 2016). It primarily presents as an inorganic as well as organic type in drinking H_2O, particularly in surface water. If there is a higher concentration of sulphide ions is present in water, and it forms precipitate with arsenic in a reduction state (Bibi et al., 2017). The permissible limit of As in drinking water is 10 mg/L as per recommendation of World Health Organization (WHO) (Chen et al.,2017, Adimalla, et al.,2020).

Figure 1: Graphical representation of Geochemical Cycle of As in Environment (Hao et al., 2018)
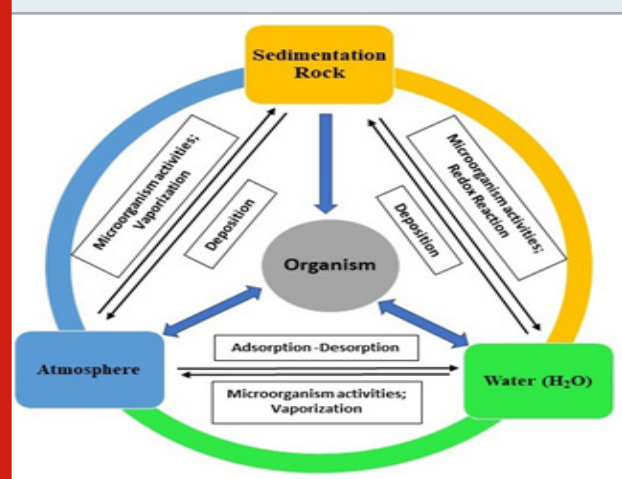
Natural phenomena like minerals dissolution due to weathering, activity of different types of microbes, and complexing with organic materials may release arsenic into the aquatic environments (Tabelin et al., 2018). On the other hand, arsenic pollution in soils and surface water may result from human caused activities such as mineral mining and metallurgy productions, fuel burning and, the use of arsenic based pesticides (Zhang et al., 2018, Rahman et al.,2019). There are several techniques including process of oxidation, co-precipitation with other materials, ion-exchange, membrane filtration processes and adsorption via using matrix have been practiced to treating polluted groundwater or surface water (Karthikeyan et al., 2019).

Adsorption on matrix is a purely surface phenomenon which relied on the interaction among the adsorbent and adsorbate (Ravi et al., 2019, Sirajudheen et al., 2020). Robust particle such as nanomaterials with smaller size, high volume to surface area ratio, and enormous number of pores, can significantly decrease the concentration of arsenic from aqueous solution (Ahmad et al., 2020). The matrix applied for the removal of arsenic must be cost effective, simple to formulate, and abundant availability in nature, (Hasan et al., 2020). This review highlights the removal of As (III) and As (V) from H_2O by adsorption process using iron nanoparticles and its composites.

Synthesis of Iron Oxides NPs and Their composites: Currently, Iron NPs and their composites of different shapes and sizes is already applied successfully in several areas including living tissue imaging, farming applications and environmental applications such as removal of dyes, toxic compounds and toxic metals like mercury, lead, arsenic etc. (Tucek et al., 2014, Gangadhar, et al., 2020). The importance of magnetic nanoparticles for several of applications is because of their significant properties including stability in different conditions, biological compatibility, easy to prepare and process and reusability (Bohara et al.,2016). Usually, there are two oxidation states of iron i.e. Fe^{2+} and Fe^{3+} present in iron oxides, possessing four and five unpaired electrons in 3d sub shell, respectively. Nassar (2012) claimed several categories of iron oxides for example hematite, maghemite, magnetite, and wustite, oxides/hydroxides of Fe such as goethite, akagneite, lepidocrocite, feroxyhyte, and hydroxides of iron viz. ferrihydrite and bernalite (Figure 2.). A brief outline of the different approaches used for the synthesis of iron nanoparticles or composite for arsenic removal is given below:

Sol–gel deposition: Sol- gel deposition is a one of the most used process for the preparation of nanostructured porous membranes, nanoscaled layers and coatings can be prepared via the production of Sol-gel (Nisticò et al., 2017). The initivial step involves a polymerization process that creates a suspension of colloid, or "sol," of separate, homogenous dispersion of fine particles which is held in suspension after adding by the surfactant (Alehosseini et al., 2020). Further, the sol may be treated to remove the suspended particles, like casting or spin-coating on a substrate. It is changed into a gel by chemical reactions so as to restrict the surfactant from making network of bound particles in the solution, which may lead to produce a class of superpolymer, a huge molecule in the form of 3D or, on the surface, a 2D complex i.e. the "gel." Sol-gel thin film deposition process offers numerous benefits including processing at low temperature and effortless processing. Many researchers like Puscasu et al., (2016), Demirci et al., (2018), Yilmaz, et al., (2020) synthesized difference (Fe_2O_3) particles by sol-gel method.

Co-precipitation: The co-precipitation technique is most probable method for synthesizing magnetic nanoparticles due to easy to prepare and proficient chemical method (Bhateria et al., 2019). Fe_3O_4 is typically prepared by stoichiometric ratio of mixing of Fe2+ and Fe3+ solution in water (Sundar et al., 2020). The Fe_3O_4 is precipitated and predicted range of pH between 8 to 14. The structure and topography of the nanoparticles can be controlled by changing the concentration of respective salts, pH of aqueous solution, strength of ions and temperature (Díaz-Amaya et al., 2020).

Hydrothermal and Solvo–thermal synthesis: Hydrothermal process enables the solvents to heat up in a tightly packed container (bomb, autoclave, etc.) to reach a temperature beyond their boiling point (BP) (Biswas et al., 2017). When reaction takes place under definite

conditions of temperature and pressure called as Solvo-thermal treatment and when H_2O is applied as solvent acknowledged as hydrothermal (Guo et al., 2019). When water attained above the critical temperature and pressure is stated as supercritical and, as a liquid, exhibits the properties of both liquid and gas. In addition, to obtain hollow iron oxide NPs, the hydrothermal and solvo thermal way is a simple and traditional process, (Ounacer et al., 2020). In a standard process, reagents are mixed together and continuous mixing by the help of stirrer with ferric salt as the iron supply, which gives homogeneous mixture is further moved to a steel autoclave lined with Teflon and heat sealed for 8-24 h at about 200°C. Organic solvent is used in a reaction mixture in its place of water in the solvothermal phase. Normally, hydrothermal process for synthesizing Fe3O4 using salt of ferrous, ferric and sodium hydroxide with a molar ratio of 1:2:8 to an autoclave and heat treatment at elevated temperatures (Bhateria et al., 2019).

Magnetic Iron nanocomposites: Several substances such as Ag, Si or Au may be applied to coat the external surface of Fe_3O_4 nanoparticles (Salem et al., 2019). These coatings on the external surface of the magnetic nanoparticle provide covalent binding positions for a specific ligand and also enhance the stability of the nanocomposites. The composition of magnetic nanoparticles is such a way where they have an outer shell which possess inorganic material while iron oxide is composing the inside center. For example, the alteration of electrolytes and pH is occupied by silica coating and therefore leads to a superior degree of robustness of nanoparticles (Hurtado et al., 2020). Magnetic nanoparticles are doped with other metal(s) and other polymer(s) to form nanocomposites. Doping of iron oxides can be achieved by using oxidizing metallic agent such as manganese leading to precipitation and hydrolysis-precipitation processes. Bimetallic such as Fe-Mn, Fe-Ce and Fe-Cu and its oxides are insufficient patterns of metal doped iron oxides (Ma et al.,2020). These composites are very significant to prevent the step carried out in the pre-oxidation of arsenic using oxidants. The improved composites efficiently removed arsenic via oxidation and adsorption process (Priyadarshni et al., 2020).

Figure 2: Illustration of Synthesis of IO NPs and its Composites using different methods (Ravi et al., 2020)

Removal of As (III) and As (V): Iron based compounds for example hematite (α-Fe_2O_3), goethite (Mineral of the diaspore group, consisting of Fe^{3+} oxide-hydroxide), iron oxide coated nanomaterials and ferric hydroxide the ideal set of materials for As removal due to the minimum leaching of adsorbed As from the use adsorbent (Ghanizadeh et al., 2010). Iron nanoparticles possess magnetic properties which helps in smooth isolation of iron nanoparticles from H_2O using powerful magnet (Gadad et al., 2014). Furthermore, Iron nanoparticles, zero-valent nanoparticles have also shown in different research papers for removal of metallic arsenic from H2O and industrial wastes or discharge to prove its efficacy for purification of water and environmental sustainability (Figure 3.) (Mosaferi et al., 2014).

Figure 3: Mechanistic representation of Arsenic removal using IO NPs under aerobic condition (Hao et al., 2018)
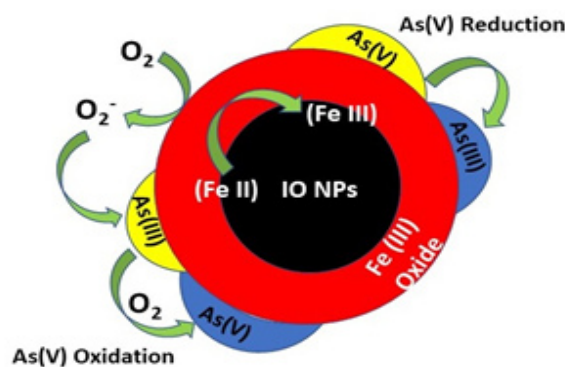
Interaction of arsenate with iron oxides nanoparticles are established by the establishment of inner part of sphere and to a lower degree by weaker ionic exchange reactions phenomena. Luther et al., (2012) synthesized the Nanophase IO and shown its efficiency for the removal of As(III) and As(V). Maximum binding observed by the IO (Fe2O3 and Fe_3O_4) NPs were found 1.25 (mg/g), 8.19 (mg/g) for As(III) and 4.6 (mg/g), 6.7 (mg/g) for As(V) respectively at incubation period 1 hr. Graphene carbon nanotube-IO have shown significant absorption capacity for removal from As from dirty water because of high surface area to volume ratio and open porous structure (Vadahanambi et al., 2013, Mamaril et al., 2020).

Aredes et al., (2013) revealed that all natural IO NPs adsorbed arsenic at pH ranges from 4-11. Raul et al., (2014) synthesized IOH nanoflower and application in removal of As(III) (475 µg/g) from water. Bhowmick et al., (2014) fabricated the Mt-nZVI which shown very impressive result at pH 7.0 on both As(III) and As(V) 59.9 and 45.5 mg/g for As(III) and As(V) respectively. Devi et al., (2014) used IO coated sand for the As (III) removal from drinking water. Qi et al., (2015) reported that bimetallic oxide NPs like (Fe-Mn) remove 39.1 mg/g and 54.2 mg/g of As(V) and As (III) respectively. Composite γ-Fe_2O_3@CTF (Leus et al., 2018) shows excellent removal for both form of arsenic (As(III) (198.0 mg g^{-1}) and As(V) (102.3 mg g^{-1}). Mishra et al., (2019) formulated aero gel

based cerium doped IO for the As(III) removal and the maximum efficacy was 263 mg/g. Recently Dong et al., (2020) have formulated cellulosed nanocrystal IO and used this for the removal As(III) and As(V) adsorption.

Which shows at pH levels of 7 and 3 CN/IO removed 13.866 mg/g and 15.712 mg/g of As(III) and As (V) from H_2O respectively. Table I. shows synthesis of iron nanoparticles and its composites for As removal.

Table I. Removal of Arsenic As (III) and As (V) by Iron NPs and its composite

S.N.	Nanoparticle Matrix	Isotherm	pH	Removal of Arsenic		Ref.
				As (III) (mg/g)	As (V) (mg/g)	
1	Bare NZVI	Freundlich	7	3.5	-	Kanel et al.,2005
2	Bare NZVI	Langumir	7	-	38.2	Yuan et al., 2006
3	γ-Fe_2O_3	Langumir	7	-	2.9	Park et al., 2009
4	α-Fe_2O_3	-	7	95	47	Tang et al.,2011
5	Fe_2O_3– γ-Fe_2O_3 nanoparticles	Langumir	2.0	3.69	3.71	Chowdhury et al.,2011
6	γ-Fe_2O_3	Langumir	3-11	74.83	105.25	Lin et al. 2012
7	γ-Fe_2O_3	Langumir	6.6	47	95	Prucek et al., 2013
8	B-FeOOH/GONs	Langumir	-	77.50	45.70	Ming et al. 2015
9	Fe-NN/BFs	-	7	70.22	93.94	Wei et al., 2019
10	IO gel	Langumir	7	35.75	-	Otero-González et al., 2020
11	RH+IO	Langmuir	7	82	-	Pillai et al., 2020
12	BT-FeN	Langmuir	7	-	18.98	
13	OL-FeNP	Langmuir	7	-	32.05	
14	GT-FeNP	Langmuir	7	-	13.70	Kamat et al., 2020
15	PL-FeNP	Langmuir	7	-	11.65	
16	EL-FeNP	Langmuir	7	-	39.84	
17	HFOR	Langmuir	5-7	41.6	71.5	Liu et al., 2020
18	Iron Oxide Composite	Langmuir	6	83.84	-	Ramasubbu et al., 2020

CONCLUSION

The present scenario of nanotechnology related with preparation and application of iron nanoparticle in the light of As removal has been outlined and reviewed. Iron oxide have exclusive magnetic and physiochemical characteristics which could be harness to use in environmental applications. Nanostructure iron oxide materials have outstanding capacity for the get rid of arsenic contaminants from water. Presently, this is similarly significant to find new ideas for enhancing the stability and biocompatibility of iron nanoparticles and composites to serve the purpose of environmental applications. In addition, iron oxide nanoparticles and its composites are observed to be the very good absorbents for arsenic removal.

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