

Nanoparticles Based Adsorbent for Removal of Arsenic from Aqueous Solution

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Abstract: Arsenic is a metalloid which poses a risk on water quality, a severe health problem for human and serious impact on environment. Occurrence of arsenic in natural environment may be due to natural processes or due to anthropogenic activities. Removal of arsenic can be done by many different techniques like adsorption, precipitation, flotation, ion exchange etc. Among them adsorption is widely used for removal of heavy metals from water due to its simplicity; also it's cost effective. The aim of the study is to evaluate the potential of nanoadsorbent for arsenic removal. A number of potent adsorbent have been developed from metal, carbon and oxide based nanoparticle for enhancing the adsorption capacity and removal capacity of arsenic from aqueous solution. A short overview of nanoadsorbent for arsenic removal from aqueous solution has been discussed in this review article.

Key words: Arsenic, nanoparticle, adsorption, precipitation, ion exchange.

Introduction

Arsenic is an element that raises much concern from both environmental and human health standpoints (Saha et al., 1999). Occurrence of As in natural environments may be due to natural processes like weathering, biological and volcanic activities together with anthropogenic use. Humans may encounter arsenic in water from wells drilled into arsenic-rich ground strata or in water contaminated by industrial or agrochemical wastes (Hughes et al., 1988). Arsenic is a widely diffused metalloid, commonly found in all the environmental matrices. High levels of arsenic have been measured in many ground water worldwide as a consequence of either natural or anthropogenic sources. Thus treatment of As contaminated water with cheaper and improved technologies becomes necessity to safe drinking water to the community. Several physico-

chemical techniques have been developed for this purpose (Ng et al., 2004). The most commonly used technologies are: oxidation, co-precipitation followed by adsorption onto coagulated flocks, lime treatment, ion exchange, adsorption onto various solid media and membrane filtration (Choong et al., 2007). Among them, precipitation usually requires high chemical dosages and produces high volumes of sludge which needs proper treatment and disposal. Furthermore, the addition of chemicals to the water may negatively alter its quality in view of the use as drinking water source.

Ion exchange is highly efficient, provided that interfering ions are absent or at very low concentration. Membrane filtration, although capable of producing a very high quality permeates, recovers only a fraction of the treated water volume while the remaining fraction is wasted as concentrate stream. Extensive studies have been performed to determine performances and optimal

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operating conditions of numerous natural or commercial media. Nanotechnology shows effective and durable ways of water purification and it is more efficient than other technologies used for water purification.

Arsenic in the Environment

Arsenic, atomic number 33, is a trace element and is the 53rd most abundant of the 92 naturally occurring elements in the Earth's crust. While often referred to as a heavy metal in the environmental literature, it is not a metal but a metalloid with predominantly non-metallic character (Singh et al., 2007).

High arsenic level in drinking water affects the visual perception of children and chronic ingestion of inorganic arsenic causes multisystem adverse health effects. High dose of arsenic in drinking water causes characteristic skin manifestation, vascular disease including arteriosclerosis, peripheral vascular disease and ischemic heart disease (ISHD), renal disease, neurological effects, cardiovascular disease, chronic lung disease, cerebrovascular disease, reproductive effects and cancers of skin, lungs, liver, kidney and bladder. Increased exposure of arsenic is also associated with non-insulin dependent diabetes mellitus (Rahman et al., 1998; Wang et al., 2003). In medicinal exposure to arsenic, some arsenic containing drugs historically and presently are used to treat some diseases like syphilis, asthma, rheumatism, cough, pruritus and itching (Wong et al., 1998; Ko and Ko, 1999).

Drinking water is the primary and main route of exposure to arsenic. MCL is the standard concentration of arsenic in drinking water which is not hazardous, is set by the US EPA i.e. 10 µg/l (EPA, 2001) and the guideline value for concentration of arsenic in drinking water recommended by the WHO is also 10 µg/l (WHO, 1992). Millions of people are compelled to use the drinking water with higher arsenic level than MCL worldwide. West Bengal (India) and Bangladesh are the worst affected areas in the world from arsenicism. In West Bengal (India), the arsenic concentration in drinking water is about 60 to 3700 µg/l and about 40 million people are affected from it. In Bangladesh, more than 70-80 million people are at a risk of drinking contaminated water. The drinking water arsenic levels were ranged from non-detectable to 4700 µg/l (British Geological Survey, 2001).

In Chile, the arsenic level in drinking water was very high and about 750 to 800 µg/l, which caused several skin and lung diseases (Singh et al., 2007). In China, the concentration of arsenic in well water in the

affected areas was 50 µg/l to 2000 µg/l and about two million people in the affected area use the drinking well water containing arsenite more than common standard, which cause Raynaud's disease in the population. The patients of Raynaud's disease suffer from increasing the resistance of blood vessels and reduced the speed of blood flow in the extremities (Xia and Liu, 2004).

Various Techniques for Removal of Arsenic from Water

Various technologies have been developed to reduce the arsenic concentration in water such as adsorption, ion exchange and precipitation. Among these methods, adsorption technique seems to be a promising one for arsenic removal, because of ease of operation and selectivity. The efficiency of the technique depends on the nature of adsorbents. In Figure 1, the presently available technologies are summarized that can be used for the removal of arsenic.

Isotherm and Kinetic Equations

It is necessary to form the most appropriate adsorption equilibrium correlation in the attempt to discover innovative adsorbents to gain access to an ideal adsorption system (Srivastava et al., 2006). It is vital for consistent prediction of adsorption parameters and quantitative comparison of adsorbent behaviour for various adsorbent systems (or for varied experimental conditions). Adsorption isotherms, which are a common name of equilibrium relationships, are essential for optimization of the adsorption mechanism pathways, expression of the surface properties and capacities of adsorbents, and productive design of the adsorption

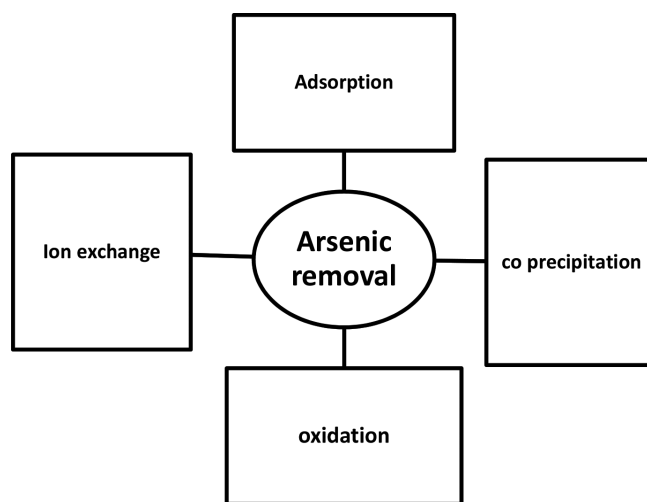


Figure 1: Various techniques for arsenic removal from water.

systems since they explain how pollutants interrelate with the adsorbent materials (Thompson et al., 2001). Explaining the phenomenon through which the preservation (or release) or mobility of a substance from the aqueous porous media or aquatic environments to a solid-phase at a persistent temperature and pH takes place, in broad-spectrum, an adsorption isotherm is an invaluable curve.

The mathematical association which establishes a significant role towards the modelling analysis, operational design and applicable practice of the adsorption systems is normally represented by plotting a graph between solid-phase and its residual concentration. (Chaker, 2008). When the concentration of the solute remains unchanged as a result of zero net transfer of solute adsorbed and desorbed from sorbent surface, a condition of equilibrium is achieved. These associations between the equilibrium concentration of the adsorbate in the solid and liquid phase at a persistent temperature are defined by the equilibrium sorption isotherms. Linear, favourable, strongly favourable, irreversible and unfavourable are some of the isotherm shapes that may form.

Understanding of the mechanism of adsorption, surface properties, along with the extent of affinity of the adsorbents is delivered by the physicochemical parameters accompanied by the fundamental thermodynamic suppositions (Bulut, 2008). In terms of three basic approaches, an extensive diversity of equilibrium isotherm models has been framed in the past. The first approach to be mentioned is kinetic consideration, while thermodynamics being the second one. A state of dynamic equilibrium with both adsorption and desorption rates in balance is an adsorption equilibrium and a framework of deriving numerous forms of adsorption isotherm models is given by thermodynamics (Jossens et al., 1978). The key idea in the generation of characteristic curve is generally given by the third approach which is potential theory. The alteration in the physical interpretation of the model parameters compels us to follow an interesting trend in the isotherm modelling which is the derivation in more than a single approach.

Adsorption Kinetics

The amount of the arsenic adsorbed onto the media was calculated based on the mass balance of arsenic between solid and liquid phases. The adsorption capacity of the adsorbent at equilibrium, Q_{ads} , which represents the mass of arsenic adsorbed per unit mass of adsorbent, was calculated by applying the following equation:

$$Q_{ads} = \frac{V(C_o - C_e)}{m}$$

where C_o and C_e are the initial and final (at equilibrium) arsenic concentration in solution, respectively, V is the solution volume and m is the mass of the adsorbent material (Chiavola et al., 2016).

Kinetic Equations

Kinetic studies are important for the prediction of optimum conditions in full-scale batch adsorption processes (Febrianto et al., 2009). Kinetic isotherm models provide information about adsorption mechanisms and rate determining steps such as mass transport or chemical reaction processes. Several kinetic models as pseudo-first and pseudo-second order (linear or non-linear forms), intraparticle diffusion and Elovich are available. The most prevalent are the pseudo-first and the pseudo-second order kinetic equations.

Langmuir Isotherm Model

Langmuir adsorption isotherm has been traditionally used to quantify the performance of different bio-sorbent. It was originally developed to describe gas-solid-phase adsorption onto activated carbon. In its formulation, this empirical model assumes monolayer sorption (the absorbable layer is one molecule in thickness), and adsorption will only occur at a finite (fixed) variety of definite localized sites, that are identical and equivalent, with no lateral interaction and steric hindrance between the adsorbed molecules, even on adjacent sites (Allena et al., 2004). Graphically, it is characterized by a plateau, an equilibrium saturation point where once a molecule occupies a site, no further adsorption can take place (Allena et al., 2004; Demirbasa and Kobya, 2008). Moreover, Langmuir theory has related rapid decrease of the intermolecular attractive forces to the rise of distance.

Freundlich Isotherm Model

Freundlich isotherm is the earliest isotherm model for describing the non-ideal and reversible adsorption. It can be applied to multilayer adsorption with non-uniform distribution of adsorption heat and affinities over the heterogeneous surface. It is widely applied in heterogeneous systems especially for organic compounds or highly interactive species on activated carbon and molecular sieves. The adsorption intensity or surface heterogeneity is measured by the slope ranges between 0 and 1. The intensity becomes more

heterogeneous as its value gets closer to zero, whereas, a value below unity implies chemisorptions process where $1/n$ above one is an indicative of cooperative adsorption (Foo and Hameed, 2010).

Redlich–Peterson Isotherm Model

Redlich–Peterson isotherm may be a hybrid isotherm that includes both Irving Langmuir and Freundlich isotherms that incorporate three parameters into an empirical equation (Prasad and Srivastava, 2009). The model includes a linear dependence on concentration within the numerator and a function in the denominator (Ng et al., 2002), to represent adsorption equilibria over a large concentration range, which will be applied either in homogeneous or heterogeneous systems because of its versatility (Morin-crini and Badot, 2008). The linearized and non-linearized equations for above isotherms model are listed in Table 1.

Nanoadsorbents

Nanoadsorbents have nanoscale pores, high selectivity, high surface area, high permeability, good mechanical stability and good thermal stability (Gangadhar and Maheshwari, 2012). Iron oxide based nanoparticle was synthesized and reported 45% efficient for arsenic removal (Feng et al., 2012). Biomimetically synthesized iron nanoparticle was also reported for arsenic removal (Prasad et al., 2014). Nanosized Mn oxides (MnO_x) exhibit an adsorptive performance superior to their bulk counterpart due to its polymorphic structures and better specific surface area (Luo et al., 2010). In the past decades, MnO_x had been exploited for adsorption of cationic or anionic pollutants from natural water sources, i.e., metal ions arsenate and phosphate (Trivedi and Axe, 1999). NCMO (nanostructured cerium incorporated manganese oxide) reported to remove arsenic from aqueous solution with efficiency of 38% (Gupta et al., 2011).

Activated carbons generally have high porosity, high surface area and prepared from readily available

carbonaceous precursors, such as coal, wood, coconut shells and agricultural wastes (Mauter and Elimelech, 2008; Patnukao et al., 2008; Selvi et al., 2001; Di Natale et al., 2008). The adsorption of As^{5+} on a granular activated carbon was done through an experimental study (Mauter and Elimelech, 2008). There are several more metal based nanoparticles like CeO (Luo et al., 2010), TiO_2 (Jing et al., 2009) and M-RGO (Magnetite-reduced graphite oxide (Chandra et al., 2010) which have been reported for arsenic removal from water. Some of the nanoparticles studied for arsenic removal are listed in Table 2.

Processes Involved for the Removal of Heavy Metal Ions

Adsorption, oxidation-reduction reactions, photocatalytic transformation and size exclusion are processes which might be increased or initiated by nanoparticles for the removal of toxic heavy metal ions from wastewater. Redox reactions are favoured for persistent inorganic pollutants to initiate the transformation of the ionic structure. For metals it is well known that a change of the redox conditions has a major influence on its toxicity (Chen and Mao, 2007; Aillon et al., 2009).

Regeneration and Reuse

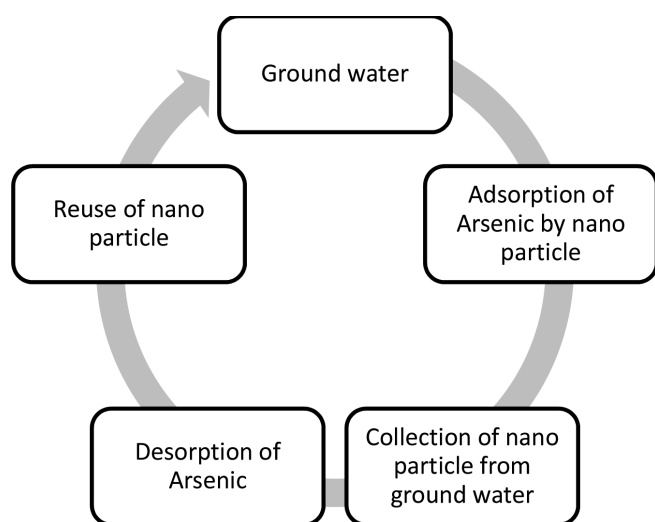
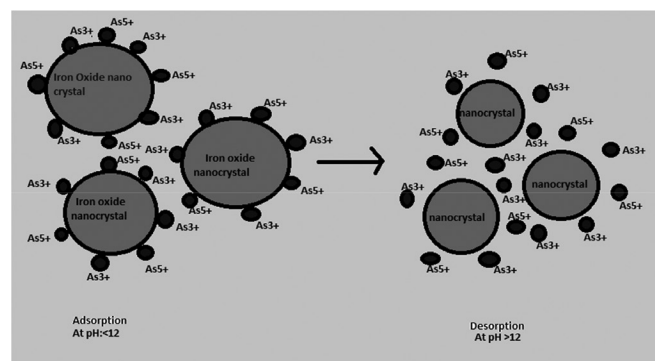
In cases where process economy dictates that immediate disposal isn't efficient, regeneration of the adsorbent looks to be the popular option. Many studies recommend that the utmost adsorption capability of metal-based nanoparticle adsorbents remains almost constant after several cycles of regeneration and reuse (Figure 2) (Tuutijärvi et al., 2012; Banerjee and Chen, 2007). Moreover, pH scale is taken into account as a vital factor in the desorption of metals from the adsorbents (Figure 3). This is in accordance with the results obtained (Tuutijärvi et al., 2012) regarding the desorption characteristics of arsenate and the recovery of the adsorbent maghemite ($-\text{Fe}_2\text{O}_3$) nanoparticle.

Table 1: Some isotherm model equations for linear and non-linear form

<i>Isotherm</i>	<i>Formula for non-linear form</i>	<i>Linear form</i>
Langmuir	$Q_e = \frac{Q_{eK_{LC_e}}}{1 + K_{LC_e}}$	$\frac{C_e}{Q_e} = \frac{1}{K_{LQ_m}} + \frac{1}{Q_m} C_e$
Freundlich	$Q_e = K_{FCE_n^1}$	$\ln Q_e = \ln K_{F+\frac{1}{n}} \ln C_e$
Redlich–Peterson	$Q_e = \frac{K_{RC_e}}{1 + a_{RC_e}^{\beta_s}}$	$\ln \left(K_R \frac{C_e}{Q_e} - 1 \right) = g \ln(C_e) + \ln(a_R)$

Table 2: Comparison between different nanoadsorbents

<i>Adsorbents</i>	<i>Surface area</i>	<i>Removal percentage</i>	<i>Condition</i>	<i>Reference</i>
Fe ₃ O ₄	178.48	45	Contact Time: 24 hr Temp: 298 K pH: 2-13	Feng et al., 2012
NCMO (Nanostructured cerium incorporated manganese oxide)	116.96	38	pH: 7 Contact Time: 3 hr Temp: 303 K	Gupta et al., 2011
CeO	72	NA	pH: 7 Temp: 298 K	Luo et al., 2010
Fe ₃ O ₄ -NCs (Nanocrystals)	300	99.2	Contact Time: 24 hr	Yavuz et al., 2006
TiO ₂ -NCs	329	NA	pH: 6 Temp: 291 K	Jing et al., 2009
Granular AC (Activated carbon)	950	NA	pH: 6 Temp: 291 K	Di Natale et al., 2008
M-RGO (Magnetite –reduced graphite oxide)	137-148	99.9	pH: 7 Contact Time: 120	Chandra et al., 2010

**Figure 2: Application of nanoparticle for water purification.****Figure 3: Adsorption and desorption of arsenic with differences in the pH.**

Conclusion

Nanomaterials have a number of specific physicochemical properties that make them notably attractive for wastewater purification. The property of getting higher specific surface area resulting in a better capability makes the nanoparticles one amongst the best adsorbents for the effective removal of heavy metals from waste water. Recent researches had indicated that use of nanomaterials as adsorbents may be a very helpful and powerful tool for the removal of metals like arsenic. These materials are capable to get rid of harmful metal ions even at low concentration i.e., up to ppb level also, with a very high selectivity and adsorption capability. These unique and specific properties of nanosorbents create them ideal materials for wastewater treatment technology.

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