



Kinetic equilibrium and thermodynamic study of arsenic removal from water using alumina supported iron nano particles



Ayushi Jain, Madhu Agarwal*

Department of Chemical Engineering, Malviya National Institute of Technology, 302017 Jaipur, India

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ABSTRACT

In the present study attempt was made to synthesize iron nanoparticles in the presence of alumina (Al-nZVI) by sodium borohydride reduction process. The composite adsorbent has been characterized using various analytical techniques such as scanning electron microscopy (SEM), Fourier transform infrared spectroscopy (FTIR), and transmission electron microscopy (TEM). The batch adsorption experiment has been carried out to study the effect of different parameters. The maximum removal of As(III) was obtained at optimal condition of pH 7.0 and dose = 1 g/L has been observed to be 99.9% with contact time of 60 min temperature 50C and initial concentration 10 mg/L. With the synthesized adsorbent residual As(III) was below permissible limit (10 µg/L). The adsorption data fitted that pseudo second order kinetics and followed Langmuir isotherm model with maximum adsorption capacity of 15.50 mg/g. Thermodynamic study reveals the endothermic nature of adsorption. Alumina has been chosen as support due to easy separation after treatment; provide mechanical strength and ability to withstand wider operating conditions. The size of iron nanoparticles synthesized lie within 40–100 nm range.

1. Introduction

Arsenic is abundant in environment covering almost 0.00005% of earth crust [1]. The source of arsenic in water includes both natural (volcanic emissions, weathering and dissolution from minerals containing arsenic, biological activities,) and anthropogenic activities [2–4]. Arsenic in environment mainly exist in four oxidation states namely arsenite (As(III)), arsenate (As(V)), arsenic (As(O)) and arsine (As(-III)) [5]. Among them, arsenite is highly toxic [6] and mobile [7,8] in nature as compared to arsenate therefore required greater attention for its removal. For efficient arsenic removal, chemical oxidation of arsenite (As³⁺) to arsenate (As⁵⁺) has often been suggested [9]. Arsenic contamination in groundwater has been reported worldwide [10]. To address the problem of arsenic contamination in ground water WHO has set the permissible limit to 0.01 mg/L. Long term consumption of arsenic contaminated water and food stuff may result in lung cancer, reproductive disorder, skin problems, skin cancer [11].

Development always favored easy to handle techniques therefore, adsorption is more preferred over other techniques due to its simplicity of operation and low implementation cost [12]. Different adsorbent has been reported for application of arsenic removal red mud [13], activated carbon [14], ZnO [15], activated carbon, polymer resins [16,17] etc. Nowadays zero valent iron nanoparticles has been identified as the

potential adsorbent for removal of chlorinated solvents, organochlorine pesticides, polychlorinated biphenyls, organic dyes, and inorganic pollutants, heavy metal ions, tetracycline [18–21] to its higher surface to volume ratio, higher reactivity, small size and high adsorption capacity [22]. Oxidation of ZVI provide potential adsorption sites for removal of As(III) and As(V). Nano-scale iron particles represent a new generation of environmental remediation technologies that could provide cost-effective solutions to some of the most challenging environmental cleanup problems. Nano-scale iron particles have large surface areas and high surface reactivity. Equally important, they also provide enormous flexibility for in situ applications. The use of zero valent iron nanoparticles (nZVI) for groundwater remediation is restricted due to its tendency of agglomeration, instability and difficulty in separation. Therefore to overcome this problem it is required to load nZVI on proper support to provide it mechanical strength. Lot of study has been carried out on dispersing nZVI on different supports such as bentonite, kaolinite, resin and zeolite as shown in Table 1. In the present study alumina has been chosen as the support for nZVI due to its stability in different geochemical conditions, highly porous and active in nature, high surface area. Thus dispersion of nZVI on alumina prevents its aggregation and enhances the ease of separation. Both alumina and iron nanoparticles individually remove heavy metals efficiently as reported by different authors [23–26]. It is difficult to remove zero valent iron

* Corresponding author.

E-mail addresses: ayushijain070693@gmail.com (A. Jain), madhunaresh@gmail.com (M. Agarwal).

Nomenclature			
V	Volume of the adsorbate	b_T	Temkin isotherm constant
R_L	Separation factor	K_F	Freundlich isotherm constant
C_i	The concentration before adsorption	E	Energy per molecule of adsorbate
W	The weight in gram of the adsorbent	A_T	Equilibrium binding constant (L/g)
C_e	The equilibrium concentration of adsorbate (mg/L)	B	Constant related to heat of adsorption (J/mol)
K_L	Energy of adsorption (Langmuir constant)	R	Universal gas constant (8.314 (J/mol/K))
q_t	Amount of adsorbed onto the adsorbent at time t	C	The intercept
n	Adsorption intensity	ϵ	D–R isotherm constant
T	Temperature at 298 K	X_m	Theoretical isotherm saturation capacity (mg/g)
k_{pd}	Intra-particle diffusion rate constant ($\text{mg g}^{-1} \text{min}^{-1}$)	K_{DR}	D–R isotherm constant (mol^2/kJ^2)
		q_e	The amount of metal adsorbed per gram of the adsorbent at equilibrium (mg/g)

Table 1
List of nZVI loaded support used for removal of different pollutants.

Adsorbent	Pollutant	Adsorption capacity	Initial concentration	References
nZVI-Al	Cu^{2+}	1.5 mmol/g	10–500 mg/L	[18]
nZVI-Kaolinite	Cu^{2+} and Co^{2+}	23 & 32 mg/g	1–500 mg/L	[27]
nZVI-bentonite	Co^{2+}	–	100 mg/L	[28]
nZVI-graphene	Uranium	8173 U mg/g	714 mg/L	[29]
nZVI-graphene	Trichloronitromethane	–	60 mg/L	[30]
nZVI-kaolinite	Pb^{2+}	–	500 mg/L	[31]
nZVI-graphite oxide	As(III) & As(V)	35.83 mg/g & 29.04 mg/g	Below 8 and 3 mg/L	[32]
nZVI-bentonite	Chromium (VI)	–	50 mg/L	[33]

nanoparticles after treatment due to nano size, the possibility of its loss in treated water may lead health problems. The hybridization of alumina with iron nanoparticles causes easy separation and prevents agglomeration iron nanoparticles. For large scale operation continuous removal of impurities from water is essential, packed adsorbents bed is used for this purpose. However, it is difficult to operate packed bed of nanoparticles due to very low porosity. Therefore, the hybridized alumina with iron nanoparticles can be a solution for it. nZVI loaded on alumina was synthesized to eliminate arsenic present in water. As(III) adsorption increased with pH and maximum adsorption was obtained at pH 7.0. Zero-valent iron synthesized has core-shell structure. To best of our knowledge this was the first time Al-nZVI has been used for arsenic removal.

The objective of the present study was to synthesize the zero valent iron nanoparticles dispersed over alumina (Al-nZVI). The synthesized adsorbent was characterized by different analytical techniques: scanning electron microscopy (SEM), transmission electron microscopy (TEM) and fourier transform infrared spectroscopy (FTIR). The composite adsorbent has been used for arsenic removal at different operating conditions such as time, temperature, adsorbent dose etc. Finally the residual arsenic concentration has been tested by Atomic Adsorption Spectroscopy (AAS).

2. Materials and methods

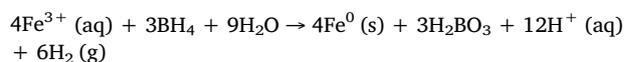
2.1. Materials

Reagent grade sodium arsenite (NaAsO_2), ferric chloride ($\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$), ethanol ($\text{C}_2\text{H}_5\text{OH}$), sodium borohydride (NaBH_4), hydrochloric acid (HCl), sodium hydroxide (NaOH), have been purchased from Merck Limited, Mumbai for the study. Stock solution (100 mg/L) of As (III) was prepared by adding 0.17320 g of sodium arsenite (NaAsO_2) in 1000 ml of deionized water. pH of prepared solutions was maintained by adding 0.1 M HCl and 0.1 M NaOH. All the required

solutions were prepared with deionized water for synthesis, dilution and adsorption study.

2.2. Preparation of Al-nZVI

The composite adsorbent (Al-nZVI) was synthesized by reduction of ferric chloride [34] on alumina particles suspended in the ethanol using sodium borohydride as reducing agent according to following reaction [35].



Two gram of $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ was dissolved in ethanol-water (225 ml ethanol + 25 ml water). Subsequently, 6 g of alumina powder were added to the solution and mixed with magnetic stirrer at 600 rpm for half hour. Sodium borohydride (NaBH_4) solution was prepared by dissolving 2 g of NaBH_4 in 100 ml of distilled water. Sodium borohydride was used as the reducing agent. The borohydride solution was added drop wise from burette to the iron-alumina mixture under continuous stirring. As borohydride solution was added to the mixture tiny black particles were appeared. Al-nZVI particles were centrifuged, washed with ethanol and filtered using Whatman filter paper. The sample was dried in oven over 75 °C and stored in desiccator under ambient condition for further study.

2.3. Characterization techniques

The synthesized adsorbent was characterized using different analytical techniques such as FTIR, SEM, TEM etc. The synthesized Al-nZVI and alumina were subjected to FTIR analysis using infrared spectrophotometer ranging from 500 cm^{-1} to 4000 cm^{-1} to confirm the presence of characteristic peaks of bonds. Surface morphology of the synthesized composite adsorbent was analyzed by scanning electron

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