



Adsorption of arsenic by activated carbon, calcium alginate and their composite beads



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ABSTRACT

The present investigation deals with preparation of three different adsorbent materials namely; potassium hydroxide activated carbon based apricot stone (C), calcium alginate beads (G) and calcium alginate/activated carbon composite beads (GC) were used for the removal of arsenic. The prepared adsorbent materials were characterized by scanning electron microscope (SEM), Fourier transform infrared spectroscopy (FTIR), N₂-adsorption at –196 °C and point of zero charge. From the obtained results, it was found that the porosity, surface area and total pore volume of the adsorbent material C > GC > G respectively, however, the G adsorbent has more acidic function group than the other adsorbents. The influence of pH, time, temperature and initial concentration of arsenic(V) were studied and optimized. GC exhibits the maximum As(V) adsorption (66.7 mg/g at 30 °C). The adsorption of arsenic ions was observed to follow pseudo-second order mechanism as well as the thermodynamic parameters confirm also the endothermic spontaneous and a physisorption process.

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1. Introduction

Arsenic contamination in drinking ground water caused by industrial effluents and some agricultural activities is a worldwide problem. Arsenic is potentially toxic to humans as chronic effects for a long times intake food and drinking water with arsenic contamination. The chronic effects include skin diseases, cancer, neurological, etc. [1–3]. It is a slow poisoning which can affect the human health [2]. Arsenic occurrence in the environment can be due to human activities such as mining, pesticides use, smelting of non-ferrous metals, burning of fossil fuels and timber treatment. In the industrial sector, arsenic is mainly used as a wood preservative and hence, it can be used in industry of dyes, pigments and paints manufactures. It is also used in glass-preparation, and mentors manufacturing [4]. Arsenic concentration in the rural areas averaged between 0.6 and 0.9 mg/L, and between 3.2 and 5.6 mg/L

for rivers influenced by industrial discharges. A very small quantity of arsenic is used in human and animal medical treatments as well as care products, and it is added in many food supplement products also [5].

New and very old technologies used for removing arsenic from contaminated water have been developed, including oxidation, precipitation, coagulation and filtration, adsorption, ion exchange and membrane filtration like reverse osmosis and nanotechnology. Each of the new and old technology processes has their advantages and disadvantages point of view which make the choice or select a suitable processing [6]. Comparing between all the advantages and disadvantages of above mentioned processes, the adsorption technique is one of the most popular methods for arsenic removal from aqueous solutions, and wastewater, and is now considered as highly efficient and low cost method for waste water treatment. The degree of adsorption of arsenic is critically dependent on speciation, which in turn depends on solution conditions. The species which predominate under different conditions are well represented by Eh-pH diagrams. Many factors may effect on the adsorption of arsenic, which in turn depends on conditions of solution like concentrations and pH [7].

In the development of efficient arsenic adsorbents, a huge range of different substances have been used by a number of researchers [8,9]. Aluminium, iron, titanium and a small proportion magnesium based substance have been shown very high efficiency removal

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capacity towards arsenic. Activated alumina, granular $\text{Fe}(\text{OH})_3$ and granular TiO_2 exist as commercial arsenic adsorbents based on the metal, metal oxides and/or hydroxides. The preparation of various metal oxides and hydroxides, the development of coated materials or even three dimension structured components have also been used for adsorption of arsenic from wastewater [9,10].

Sodium alginate (sodium salt) is natural product carbohydrate polymer consisting of α -L-guluronate and β -D-mannuronate residues, and it has biocompatibility, biodegradability and water solubility [11]. The bio-sorbents like alginic acid originating from brown seaweeds have high affinity towards divalent cations [12]. Each divalent metal ion binds to two carboxyl groups on adjacent alginate molecules [12]. Pretreatment or doping of alginic acid with cations like Ba^{2+} or Fe^{2+} allows interaction of anionic species with the alginate moiety. Earlier, for the removal of anionic arsenic from water, calcium alginate (CA) beads were placed in a column to form a fixed bed and were treated with hydrous ferric oxide [12]. Calcium alginate has been widely used to immobilize activated carbon (C) [13], carbon nanotubes (C-NT) [14] TiO_2 nanoparticles [15], and magnetite nanoparticles [16] to generate new adsorbents materials to remove metals, heavy metals and pigments/dyes from aqueous wastewater.

In the present work, three different adsorbent materials potassium hydroxide activated carbon from apricot stone, calcium alginate beads, and calcium alginate/activated carbon composite beads were used for removal of arsenic. The prepared materials were characterized by Fourier transformed infrared spectroscopy (FTIR), N_2 -adsorption at -196°C and scanning electron microscopy (SEM). Kinetic studies of adsorption of arsenic from solutions by using three different adsorbent materials were studied.

2. Materials and methods

2.1. Materials

Sodium arsenate heptahydrate ($\text{Na}_2\text{HAsO}_4 \cdot 7\text{H}_2\text{O}$), calcium chloride, potassium hydroxide, sodium alginate, sodium chloride, hydrochloric acid, were purchased from Sigma–Aldrich, USA and El-Nasr for pharmaceutical and chemical industrial Co., Egypt). All chemicals were used without further purification. Deionized water was used for preparation of all the required solution.

2.2. Adsorbents preparation

2.2.1. Preparation of KOH-activated carbon (C)

Apricot stone collected from apricot obtained from Egyptian local markets to be used as precursor. The activated carbon was prepared according to the previous work [17]. Briefly, “apricot stone collected and washed several time with pure water to remove the impurities and dried at 120°C for 24 h, then grinding and sieved to use particles with 2–4 mm. Carbonization of dried apricot stone at 600°C for 4 h in presence of nitrogen gas using a stainless steel reactor (60 cm \times 4 cm) at a rate of $10^\circ\text{C}/\text{min}$ up to 650°C followed with cooling at room temperature. Potassium hydroxide activated carbon was prepared by using high concentration of potassium hydroxide, the carbonized sample was impregnated into certain weight of potassium hydroxide (ratio 3:1 of potassium hydroxide: carbonized sample), in distilled water for 24 h at room temperature, followed by drying at 120°C and finally activated in tubular muffle at 750°C for 4 h, and then cool down at room temperature, washed several time with distilled water till neutralized filtrate and dried at 110°C ”.

2.2.2. Synthesis of calcium alginate beads (G)

Calcium alginate beads were prepared by using sodium alginate treated with calcium chloride as described in details in previous

work [17]. Briefly, “solution of 1% (w/v) sodium alginate added drop-by-drop to 3% (w/v) calcium chloride, the so obtained water insoluble calcium alginate beads. Calcium alginate beads washed several time with distilled water to remove unreacted calcium chloride. The washed beads were dried at 70°C for 24 h”.

2.2.3. Synthesis of calcium alginate/activated carbon composite beads (GC)

Calcium alginate/activated carbon composite beads (GC) were prepared by ionic gelation method [17–20]. Two grams of powdered activated carbon (C) was dispersed in 100 mL of deionized water and mixed with clear viscous 200 mL (1% wt/v) solution of sodium alginate. The mixture was stirred for 2 h. Once the mixture was homogenous, it was added stepwise from 100 mL burette into aqueous solution of calcium chloride and the resulting composite beads were collected and treated as the last prepared G beads.

2.3. Characterization of solid adsorbents C, G, and GC

Fourier transform infrared spectra (FTIR) were recorded on a Mattson 5000 FTIR spectrometer in the range between 4000 and 500 cm^{-1} . Point of zero charge (pH_{PZC}) of solid adsorbents was carried out by: initially, 50 mL of 0.01 M NaCl solutions were put into several closed Erlenmeyer flasks. The pH within each flask was adjusted to a value between 2 and 12 by hydrochloric acid (0.1 M) or sodium hydroxide (0.1 M) solutions, and then a portion of the sample (0.20 g) was added to each flask, the flasks were agitated for 48 h and the final pH was then measured. The pH_{PZC} is the point where $\text{pH}_{\text{final}} - \text{pH}_{\text{initial}} = \text{zero}$ [21]. SEM was used to study the surface morphology of the prepared materials model Quanta 250 FEG (Field Emission Gun) attached with EDX unit (Energy Dispersive X-ray Analyses), with accelerating voltage 30 KV, magnification $14\times$ up to 1,000,000 and resolution for Gun. Nitrogen adsorption at -196°C (out gassing was carried out at room temperature) using NOVA 2000 gas sorption analyzer (Quantachrome Corporation, USA) system was used to determine specific surface area (S_{BET} , m^2/g), total pore volume (V_T , mL/g), and average pore radius (r , nm).

2.4. Adsorption procedure

The adsorption of arsenic was measured like previous work [17] with some modification. “Aqueous solutions of certain concentration of adsorbate were shaken in Stoppard Pyrex bottles of 100 mL capacity with 0.1 g of adsorbent (C, G, or GC) for 10 h. The supernatant was filtered, where the equilibrium concentration of arsenic(V) was measured using 211 Acusys Atomic Absorption Spectrophotometer–Buck Scientific, USA. The adsorbed amount at equilibrium, q_e (mg/g) was calculated by”:

$$q_e = \frac{(C_0 - C_e)V}{W} \quad (1)$$

where C_0 and C_e are the initial and equilibrium concentration (mg/L) of adsorbate solution, respectively; W is the weight (g) of adsorbent used and V is the volume of working solution (L).

Effect of pH was measured by carrying out the adsorption process at different pH values ranged between 2 and 12. Effect of time was carried out by contacting 0.1 g of adsorbent with 100 mL bottles of 75 mg/L of arsenic, and determines the concentration of adsorbate after recorded time intervals, the adsorption capacity at time t , q_t (mg/g) was calculated using:

$$q_t = \frac{(C_0 - C_t)V}{W} \quad (2)$$

where, C_t (mg/L) is the liquid-phase concentration of adsorbate at time t . Effect of temperature was examined at two different temperature 20 and 30°C at pH of 7.5.

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