

Regular Article

Facile synthesis of novel calcined magnetic orange peel composites for efficient removal of arsenite through simultaneous oxidation and adsorption

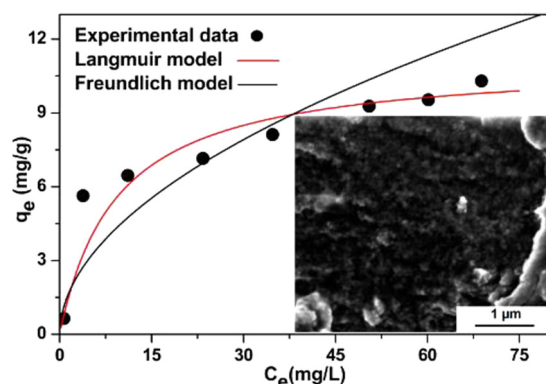


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GRAPHICAL ABSTRACT



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ABSTRACT

Increasing exposure to arsenic (As) contaminated ground water has become a global health hazard to humanity. Suitable adsorbent for As removal from water, especially for As(III) than As(V), is an urgent but still a challenging task. In this study, waste orange peel (OP) was modified with magnetic nanoparticles followed by calcination as a novel adsorbent and investigated for instantaneous oxidation and adsorption of As(III) from aqueous solutions. The batch adsorption experimental results showed that calcined magnetic orange peel composites (CMOPC) exhibited superior As(III) adsorption capacity (10.3 mg/g) as compared to similar cost effective adsorbents due to its high surface area, large pore size and greater numbers of active sites on its surface. The adsorption equilibrium data obeyed Langmuir model, and kinetic data was well described by the pseudo-second-order model. The adsorption mechanisms for As(III) might be involved ligand exchange of the hydroxyl in CMOPC to form Fe-O-As(III), and partial As(III) oxidation to As(V) followed by instantaneous adsorption on surface of CMOPC. The developed adsorbent has also demonstrated good anti-interference ability to co-existing ions, admirable regeneration ability and pronounced capacity in treating simulated real As(III) contaminated water. This study revealed that waste orange peel, after simple treatment could be used as a potential adsorbent for As(III) from aqueous solutions.

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1. Introduction

Arsenic has gained a considerable attention worldwide due to its toxic and hazardous effects. Millions of people in regions of Bangladesh, India, China, Pakistan, Argentina and Mexico are drinking water which contains arsenic concentrations much more than the safe limit (10 µg/L) defined by the WHO [1–6]. Consequently, some severe and chronic diseases, such as lungs, skin and bladder cancer have been reported in these regions [5,6]. Arsenic-bearing sulphide deposits, geothermal regions and industrial wastes are examples of some of the naturally occurring and anthropogenic sources of arsenic in environment [1].

Arsenic exists in four oxidation states (−3, 0, +3, +5), whereas the most prevalent arsenic species are arsenate (As (V)) and arsenite (As (III)). Arsenate is generally found under well-oxidized conditions, while arsenite is predominant under reduced conditions and near-neutral pH [7]. Arsenite is 60 times more toxic than arsenate and it is also more difficult to be removed due to its lower affinity towards different adsorbents [8,9]. Usually As(III) is oxidized to As(V) for remediation purposes before elimination [8], which increases operational cost and produces some secondary pollution problems. Therefore, it is necessary to synthesize a novel and economical material which can effectively remove As(III) from aqueous solutions by simultaneous oxidation and remediation of As(III).

In past, various techniques have been developed for arsenic remediation from wastewater such as membrane separation, precipitation, adsorption, reverse osmosis, chemical oxidation, and ion exchange [10]. Among all these available techniques, the adsorption of arsenic onto selected adsorbents is more effective method due to its simple operation, least waste generation and economic reliability [11]. In the meantime, various types of adsorbents, such as industrial wastes/by-products, agricultural products/by-products, biological materials, and mineral oxides have been developed with a key focus on high efficiency and low-cost [12–17]. Among these adsorbents, orange peels (OP), which are produced in large quantity from fresh juice shops/industries all over the world, are being considered as suitable low cost biomaterial for different pollutant removal from aqueous solutions [12,15–17], because its residue mainly consists of lignin, cellulose, hemicellulose, hydroxyl (−OH), carboxyl (−COOH), and amide (−NH) surface functional groups, which can play a vital role in removal of heavy metal ions from water [18]. On the other hand, a wide range of nanostructures with excellent adsorptive capacity have been investigated for adsorption of arsenic; among these nanostructures, iron (Fe) and its compounds have shown higher efficiency in arsenic remediation as compared to other oxides or hydroxides [19–26]. However, many of these adsorbents fail to remove arsenic in the presence of complex co-existing ions [26]. Some are likely to suffer from low synthesis yield, high cost, relatively low adsorption capacities or poor recyclability and reusability [21,24,25]. These problems drastically restrict their large scale production and application for adsorption purposes. Hence, it is highly required to develop effective adsorbents that exhibit good adsorption abilities for arsenic under different conditions and should be low cost. Furthermore, the calcination process has also been reported to influence the adsorption efficiencies of different adsorbent composites [21,27]. Thus, due to special characteristics of OP, Fe and calcination effects; the magnetic orange peel (MOP) and calcined magnetic orange peel composite (CMOPC) may offer a possible strategy for enhanced As(III) adsorption, but the related investigations are not reported yet.

In our present work, the objectives were to modify waste OP with magnetic nanoparticles followed by calcination under N₂ environment, and investigate their adsorption efficiencies for As

(III). The obtained novel adsorbent composite CMOPC was further evaluated for simultaneous oxidation and adsorption of As(III) from aqueous solutions under different operating parameters including initial pH, As(III) concentration, contact time and effects of co-existing ions in batch adsorption experiments. Moreover, current study focused on uncovering the possible adsorption mechanisms and practical application of CMOPC.

2. Experimental section

2.1. Materials

All the reagents, including FeSO₄·7H₂O, FeCl₃·6H₂O, NaOH, HCl and Na₃AsO₃, were analytical grades and used without any further purification. These chemicals were obtained from two dealers; (i) Shanghai Chemical Reagent Co., Ltd., China, and (ii) Shanghai Canspec Scientific Instruments Co., Ltd., Shanghai, China, whereas fresh OP were collected from nearby local market. Arsenite stock solution (1000 mg/L) was prepared by dissolving the weighted amount of Na₃AsO₃ in measured volume of de-ionized (DI) water. The stock solution was diluted in DI water to prepare fresh working solution of desired concentration of As(III) prior to experiments. NaOH and HCl were used to adjust the pH of solution as required. All glassware were cleaned with dilute nitric acid and repeatedly washed with DI water before each experiment.

2.2. Synthesis of adsorbents

Waste OP biomass materials were obtained from a local shop and washed with tap as well as DI water several times. After this, they were cut into small pieces and dried in a convection oven at 105 °C for 36 h. Then these dried OP were crushed by hammer and put through a sieve of 60 mesh size. Afterwards, washed again with DI water several times till the effluent were colorless and then dried at 60 °C for 72 h. Calcined orange peel (COP) was synthesized by thermal treatment of OP at 500 °C in a high temperature furnace for 1 h under nitrogen (N₂) flow.

MOP composites were synthesized by a simple chemical coprecipitation method. OP (5 g), FeSO₄·7H₂O (4 g) and FeCl₃·6H₂O (6 g) were transferred into a flask containing 100 mL of DI water. Furthermore, pH of the reaction medium was adjusted to 10 by adding proportionate amount of ammonia by drop wise and the reaction was held for 2 h at 50 °C. During the experiment, the mixture was also mechanically stirred. The MOP composites were collected by filtering and then washed with DI water for several times to remove unnecessary ions till the effluent had pH value of about 7. The filtrate was then dried at 50 °C for 36 h and finally stored for further use. Afterwards, the MOP composites were calcined at 500 and 1000 °C in a high temperature furnace for 1 h under nitrogen (N₂) flow to obtain other adsorbent composites, namely, CMOPC and CMOPC1.

2.3. Batch adsorption tests

Batch adsorption experiments were performed to obtain the equilibrium data in a series of 150 mL glass conical flask containing synthetic As(III) solutions kept at room temperature (25 ± 0.1 °C). The solution was mechanically shaken at 180 rpm in a rotary shaker. After the required reaction time, filtrates were taken out using syringes and filtered through a filter membrane (0.22 µm). These filtrates were analyzed for the remaining total arsenic concentration using inductively coupled plasma atomic emission spectroscopy (ICP-AES) or hydride generation atomic fluorescence spectroscopy (HG-AFS). The adsorption capacity (q_e) was calculated by using following equation:

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