



## Application of LECA modified with Fenton in arsenite and arsenate removal as an adsorbent

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### ABSTRACT

This paper describes modification of LECA (light expanded clay aggregate) with Fenton reagent and determines removal of arsenite and arsenate ions from an aqueous solution. The influence of pH, contact time and  $\text{Fe}^{2+}/\text{H}_2\text{O}_2$  on modification were investigated and adsorption characteristics were examined before and after modification. Isotherm studies were carried out by studying the effects of various parameters such as pH, contact time, adsorbent concentration and initial concentration of arsenite and arsenate. The equilibrium adsorption data were fitted to Langmuir and Freundlich adsorption models, and the model parameters were evaluated. One gram of LECA is found enough to remove 96% and 99% of the As (III) and As (V) from an aqueous solution of  $150 \mu\text{g L}^{-1}$  at pH 2 and 4 respectively with a shaking time of 60 min. Column adsorption results indicated that no arsenic was found in the effluent solution up to about 10 and 18 bed volumes of As (III) and As (V), respectively.

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### 1. Introduction

Water, the most vital element of life, polluted with arsenic is a global human health hazard. Millions of the populations worldwide are exposed to arsenic-contaminated drinking water. Arsenic is commonly distributed in nature and released into the environment through natural sources, industrial processes and agriculture usage [1]. Arsenic contamination is a serious problem in the developing countries, West Bengal (India), Bangladesh, Mexico, and the Red river delta of Vietnam, and is also a persistent problem in China and Thailand [2–5]. In Asia, the arsenic problems are intensified by the additional arsenic consumed in rice, the primary food source, cooked in arsenic-contaminated water. Arsenic also has been accumulating in paddy soil, resulting in direct contamination of rice grain. Rice contributes to an estimated 30–60% of the dietary intake of arsenic in polluted regions [6]. Arsenic is classified as a human carcinogen (cancer of the bladder, lungs, skin, kidney, liver and prostate) by the International Agency for Research on Cancer and the National Research Council [7–9]. The World Health Organization (WHO) recommended a maximal level of  $50 \mu\text{g L}^{-1}$  of arsenic in drinking water since 1963. With increasing evidence showing a link between exposure to low arsenic concentrations and cancer risk, the WHO lowered the recommendation level to  $10 \mu\text{g L}^{-1}$  in 1992 [10]. Because arsenic at low concentrations in drinking water causes severe health effects, removal of arsenic has become increasingly important.

Conventionally, there are several methods for arsenic removal. These methods include coagulation and flocculation [11], precipitation [12], co-precipitation [13], ion-exchange [14], reverse osmosis [15], and microfiltration [16]. Alternative methods like ozone oxidation, bioremediation [17] and membrane processes [18], are also used in the removal of arsenic. Most current used methods have high maintenance cost and require relatively expensive mineral adsorbents which offset performance and efficiency advantages. Therefore, adsorption of metals from an aqueous solution has received more attention due to its high application efficiency and low cost. Cost is an important parameter for comparing the sorbent materials. In general, a sorbent can be presumed as “low cost” if it requires little processing, is plentiful in nature, or is a by-product or waste material of industry. Light expanded clay aggregate (LECA), is an aggregate made of expanded clay which is produced in rotary kiln at a temperature of about  $1200^\circ\text{C}$ . LECA consists of small, lightweight, bloated particles of burnt clay. The surface charge of the soils is predominantly positive charge at low pH and is negative charge at high pH [19]. Arsenic can occur in the environment in several oxidation states ( $-3$ ,  $0$ ,  $+3$  and  $+5$ ) but in natural water is mostly found in inorganic form as oxyanions of trivalent arsenite (As (III)) or pentavalent arsenate (As (V)). Removal of anionic metals and metalloids such as arsenate and arsenite may require modification of negative charge sorbent to specifically bind anions or neutral molecules. Iron oxides have been reported to be effective for metal ion removal. Iron oxide-coated sand [20–22], iron oxide-coated polystyrene, iron oxide-coated alginate (biopolymer) [23], and iron modified activated carbon [24] were used for arsenic removal and results were positive. All the above mentioned methods clearly express that iron

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coated materials are an efficient adsorbent for arsenic. Different types of sands, alumina, zeolite, red mud, etc. have been coated with iron salts. Hague et al. reported Fe (III) has a strong affinity toward inorganic arsenic species and creates favorable adsorption sites for arsenic throughout the various adsorption mechanisms such as mono and bidentate surface complex formation, ligand exchange and electrostatic attraction [25]. Argun et al. modified pine tree bark with Fenton's reagent and used it as a natural adsorbent for Cd (II) and Pb (II) removal; they reported an increased adsorption capacity of the functional groups with Fenton modification [26].

The Fenton reaction has several important advantages such as a short reaction time, iron and H<sub>2</sub>O<sub>2</sub> are economic and non-toxic, and the process is easily to run and control [27]. Consequently, this study is designed to: (1) develop a new sorbent from LECA by modification by Fenton reagent, (2) conduct batch tests to examine arsenite and arsenate adsorption using the new sorbent, and (3) conduct column studies to investigate the arsenite and arsenate characteristics of modified LECA as granular type under an optimum condition of batch examination.

## 2. Materials and methods

### 2.1. Adsorbent and chemicals

The activated LECA with Fenton's reagent (10,000 mg L<sup>-1</sup> H<sub>2</sub>O<sub>2</sub> + 100 mg L<sup>-1</sup> Fe<sup>2+</sup>) was used in this study. The amount of H<sub>2</sub>O<sub>2</sub> in the reagent was fixed but Fe<sup>2+</sup> was added to the reagent until the proper ratio of Fe<sup>2+</sup>/H<sub>2</sub>O<sub>2</sub> was reached. All chemicals used in this study were analytical grade without further purification and all stock solutions were prepared with doubly-distilled water. Glass materials and plastic bottles were washed with doubly-distilled water and exposed overnight to a 5% nitric acid solution. Various arsenite and arsenate solutions (10–300 µg L<sup>-1</sup>) for batch and column experiments were prepared from arsenic salt (NaAsO<sub>2</sub> and Na<sub>2</sub>HAsO<sub>4</sub>·7H<sub>2</sub>O respectively). Stock solutions (1000 ppm) of As (III) and As (V) were prepared by dissolving appropriate amounts of NaAsO<sub>2</sub> and Na<sub>2</sub>HAsO<sub>4</sub>·7H<sub>2</sub>O respectively, in doubly-distilled water. Solutions of required lower concentrations were prepared by diluting the stock solutions. Fenton's reagent was prepared from FeSO<sub>4</sub>·7H<sub>2</sub>O and hydrogen peroxide (H<sub>2</sub>O<sub>2</sub> 30% W/W). Sodium hydroxide or hydrochloric acid was used for adjusting the pH.

### 2.2. Instruments and characterization of the adsorbent

A scanning electron microscope (SEM) was used to examine the morphology of the surface of the adsorbent. The chemical and physical properties of LECA are listed in Table 1. All experiments run according to ASTM [28]. X-ray fluorescence analysis was carried out to characterize the LECA. LECA pH was measured according to ASTM (D4972-01, standard test method for pH of soils).

### 2.3. Adsorbent preparation

The adsorbent of LECA was washed with doubly-distilled water and dried in an oven at 80–85 °C for 2 h. The adsorbent was milled and placed into US standard 50 meshes in accordance with the ASTM [28]. An accurately weighed and oven dried 25 g of LECA separately was poured into 500 mL conical flask containing 250 mL Fenton's reagent solutions and stirred with 200 rpm at pH 2, 3, 4 and 5; at each pH, different Fe<sup>2+</sup>/H<sub>2</sub>O<sub>2</sub> ratios of 0.01, 0.02, 0.04, and 0.06 and contact time of 30–90 min, were used for all modification experiments. Afterward, the solution was filtered and LECA was taken and washed for several times with doubly-distilled water. Then the adsorbent was oven dried at 85 °C for 2 h. 150 µg L<sup>-1</sup> arsenic solution (As (III) or As (V)) was contacted with 3.5 g L<sup>-1</sup> of oven dried modified LECA at a speed of 150 rpm for 1 h in a platform shaker (CFL3005, Germany) at pH 7 separately. The arsenic solutions were filtered through 0.45-µm

**Table 1**  
Sorbent characterization.

Constituent	Natural sorbent % by weight	Modified sorbent % by weight
Al <sub>2</sub> O <sub>3</sub>	16.57	20.32
SiO <sub>2</sub>	66.06	55.33
CaO	2.46	0.5
Fe <sub>2</sub> O <sub>3</sub>	7.1	17.9
K <sub>2</sub> O	2.69	2.53
MgO	1.99	1.73
MnO <sub>2</sub>	0.09	0.03
Na <sub>2</sub> O	0.69	0.14
P <sub>2</sub> O <sub>5</sub>	0.21	0.07
TiO <sub>2</sub>	0.78	0.51
Loss on ignition	1.36	0.94
Moisture	0.03	0.04
Water solubles	0.696	0.94
Acid solubles	3.25	3.68
Total ash	99.73	99.98
Volatile matter	0.27	0.02
pH	8.4	3.5
Iodine number (mg/g)	528	532

membrane filters after settling and it was then analyzed using inductively coupled plasma-atomic emission spectrometry (ICP-AES), (Ultima 2C, French).

### 2.4. Batch experiments

Batch equilibrium adsorption isotherm studies were conducted with aqueous solutions of As (III) and As (V) with an initial concentration of 10 to 300 µg L<sup>-1</sup>. Equilibrium isotherm measurements were carried out by keeping the solution volume (200 mL) and the amount of the modified LECA constant and varying the concentration of As (III) or As (V) in 250 mL Erlenmeyer flasks. The pH of the solutions was adjusted by adding 1.0 mol L<sup>-1</sup> sodium hydroxide or 1.0 mol L<sup>-1</sup> hydrochloric acid solutions. The pH was measured using a pH meter (Metrohm 827 pH lab, Switzerland). The batch experiments were carried out at a constant temperature (20 ± 3 °C) in a platform shaker (CFL, 3005, Germany). The equilibration (shaking) time was 60 min at an agitation speed of 150 rpm. After the isothermal equilibration, the sorbent was separated by filtration with 0.45-µm membrane filter. The filtrate was analyzed for metals. The amount of the metal adsorbed (µg) per unit mass of LECA (g), q<sub>e</sub>, was obtained by mass balance using Eq. (1).

$$q_e = \frac{(C_i - C_e)V}{m} \quad (1)$$

where C<sub>i</sub> and C<sub>e</sub> are the initial and the equilibrium concentrations of the metal ion (µg L<sup>-1</sup>), m is the dry mass of LECA (g), and V is the volume of the solution (L).

### 2.5. Sorption in a fixed bed column reactor

Generally, an adsorbent for column adsorption of arsenic should assemble several requirements: (1) granular type; (2) efficient for removal of both As (V) and As (III); (3) high capacity, selectivity, and rate of adsorption; (4) high physical strength (not disintegrating) in water; (5) able to be regenerated if required; and (6) low cost [29]. Granular iron modified LECA was used as a fixed bed. Dynamic flow adsorption experiments were conducted in a glass column of about 4.3 cm internal diameter and 30 cm length. The bed volume of the column was 435 cm<sup>3</sup>. The feed water used was a synthetic solution containing arsenite and arsenate with an initial concentration of 150 µg L<sup>-1</sup>. The pH was adjusted to 2 and 4 for arsenite and arsenate respectively and the influent solution was pumped from the reservoir into the column by a submersible pump (JP-064, China), in down-flow

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