



Research article

Simultaneous arsenic and fluoride removal from synthetic and real groundwater by electrocoagulation process: Parametric and cost evaluation



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ARTICLE INFO

Article history:

Received 12 May 2016

Received in revised form

16 December 2016

Accepted 21 December 2016

Keywords:

Electrocoagulation

Synthetic groundwater

Real groundwater

Arsenic removal

Fluoride removal

Operating cost

ABSTRACT

Co-existence of arsenic and fluoride in groundwater has raised severe health issues to living being. Thus, the present research has been conducted for simultaneous removal of arsenic and fluoride from synthetic groundwater by using electrocoagulation process with aluminum electrode. Effects of initial pH, current density, run time, inter electrode distance and NaCl concentration over percentage removal of arsenic and fluoride as well as operating cost have been studied. The optimum experimental conditions are found to be initial pH: 7, current density: 10 A/m², run time: 95 min, inter electrode distance: 1 cm, NaCl concentration: 0.71 g/l for removal of 98.51% arsenic (initial concentration: 550 µg/l) and 88.33% fluoride (initial concentration: 12 mg/l). The concentration of arsenic and fluoride in treated water are found to be 8.19 µg/l and 1.4 mg/l, respectively, with an operating cost of 0.357 USD/m³ treated water. Pseudo first and second order kinetic model of individual and simultaneous arsenic and fluoride removal in electrocoagulation have also been studied. Produced sludge characterization studies also confirm the presence of arsenic in As(III) form, and fluoride in sludge. The present electrocoagulation process is able to reduce the arsenic and fluoride concentration of synthetic as well as real groundwater to below 10 µg/l and 1.5 mg/l, respectively, which are maximum contaminant level of these elements in drinking water according to WHO guidelines.

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1. Introduction

Co-existence of arsenic and fluoride in natural water has raised a severe health issue for many countries worldwide in recent years. A large populations in India, Bangladesh, Pakistan, Mexico, Mongolia, Argentina, etc. (Farooqi et al., 2007a, 2007b; He et al., 2009; Kumar et al., 2010; Chaurasia et al., 2012; Dutta, 2013; Reyes-Gómez et al., 2015; Gomez et al., 2009) are affecting due to various type of diseases related to arsenic and fluoride. Excess fluoride intake in body causes many diseases such as skeleton fluorosis, teeth disintegration etc. (Nell and Livanos, 1988; Brown et al., 1977). However, the fluoride concentration less than 1.5 mg/l (WHO, 2011) in drinking water is essential for strengthen of bone and teeth. In addition to that, arsenic concentration greater than 10 µg/l (WHO, 2011); is more severe threat to humans due to its inherent toxic and

carcinogenic nature, which appears in human body in the form skin, lung, liver and kidney cancer (Karim, 2000; Morales et al., 2000). It is reported that co-exposure of arsenic and fluoride can have more impact on the integrity of the genetic material of cells than the individual exposure (Rao and Tiwari, 2006). Co-exposure of these can also lead to both endemic fluorosis and arsenicosis (Alarcón-Herrera et al., 2013). Arsenic and fluoride are introduced in groundwater mainly through oxidative weathering and geochemical reactions (Banerjee et al., 2008; Meenakshi and Maheshwari, 2006). Anthropogenic sources, such as liquid waste of glass, metal, semiconductor, pesticides, mining etc. industries can also deep percolate inside the earth, and subsequently contaminate the groundwater (Lacasa et al., 2011; Ayoob and Gupta, 2006; Meenakshi and Maheshwari, 2006). Arsenic and fluoride both exist in the form of anion in water. In natural water, arsenic is predominantly present in the form of arsenite (As(III)) and arsenate (As(V)), the ratio of these arsenic species depends on the pH and redox potential (Luukkonen et al., 2016; Ungureanu et al., 2015). As(V) occurs in oxygenated water, while in anaerobic groundwater As(III) is normally available. As(III) species is much

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more toxic than As(V) (Koby et al., 2011). Very recently in 2015, high arsenic and fluoride concentration has been reported in the groundwater of Rajnandgaon district of Chhattisgarh in India (Patel et al., 2015). Hence, it is an urgent need to find a technology which can efficiently remove arsenite and fluoride both from groundwater.

From literature it is revealed that many approaches such as adsorption, chemical coagulation, membrane filtration, electrodiolysis, oxidation, biological methods etc. are applied for removal of arsenic and fluoride from contaminated water (Liu et al., 2012; Mohan and Pittman, 2007; Amor et al., 2001; Castel et al., 2000). All these technologies mentioned above suffer from various disadvantages; for examples the adsorption process is pH dependent, it requires pre-treatment step, high treatment time, and its contamination removal efficiency reduces after each regeneration cycle (Koby et al., 2011; Mohan and Pittman, 2007). Coagulation method requires a large amount of chemical, large area for treatment and produces a secondary sludge (Koby et al., 2011; Balasubramanian et al., 2009). High membrane and operating cost, skilled labour requirement, and concentrated sludge generation are major drawbacks associated with the membrane filtration technologies (Balasubramanian et al., 2009; Lacasa et al., 2011). Whereas, biological processes, are mainly used for treatment of industrial waste water and not for drinking water, as the processes require several types of pathogenic and non-pathogenic microorganisms like algae, yeast, fungi, bacteria etc. Moreover, their selectivity depends on type of contaminants which has to be removed (Veglio and Beolchini, 1997). High treatment time, large land area requirement, less design flexibility are additional drawbacks of biological processes (Veglio and Beolchini, 1997; Yadav et al., 2012).

In comparison to above conventional treatment methods, electrocoagulation process has drawn attention of researchers due to its high removal efficiency, easy to operate, cost effectiveness and possibility of complete automation of process (Rajeshwar et al., 1994; Chen, 2004; Mollah et al., 2004; Emanjomah and Sivakumar 2009). In electrocoagulation process insitu generated metal hydroxides act as coagulant in aqueous solution, which provides the active sites for adsorption of polluting species. Apart from adsorption, sweep coagulation, bridge coagulation, coprecipitation etc. also play a role for the removal of pollutants in electrocoagulation process. The electrochemical reactions using Al as electrode materials may be summarized as follows (Koby et al., 2011):

Reaction at anode



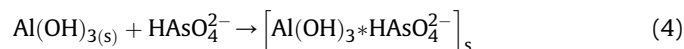
Reaction at cathode



Reaction in aqueous solution

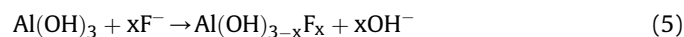


In electrocoagulation aluminum and hydroxides ion generated by reactions (1) and (2), react together and form various species of monomeric such as $\text{Al}(\text{OH})_2^+$, $\text{Al}_2(\text{OH})_2^{4+}$, $\text{Al}(\text{OH})_2^{2+}$, $\text{Al}(\text{OH})_4^-$ and polymeric species viz. $\text{Al}_6(\text{OH})_{15}^{3+}$, $\text{Al}_7(\text{OH})_{17}^{4+}$, $\text{Al}_8(\text{OH})_{20}^{4+}$, $\text{Al}_{13}(\text{OH})_{34}^{5+}$, $\text{Al}_{13}\text{O}_4(\text{OH})_{24}^{7+}$ (Ghosh et al., 2008; Rincón and La Motta, 2014). This species are further transformed into amorphous $\text{Al}(\text{OH})_{3(s)}$ according to complex precipitation kinetics (Behbahani et al., 2011). Furthermore, this $\text{Al}(\text{OH})_3$ complex is believed to adsorb arsenic (Koby et al., 2011; Flores et al., 2013) and fluoride (Hu et al., 2003).



The mechanism of fluoride removal in electrocoagulation is still not fully understood. This stems mainly from the fact that at least three mechanisms may compete (Zhu et al., 2007):

(a) Adsorption on aluminum hydroxide particles



(b) Coprecipitation



(c) Fluoride attachment to electrodes.

Many studies are available on removal of arsenic and fluoride from aqueous solution by applying electrocoagulation process (Ali et al., 2012; Flores et al., 2013; Lacasa et al., 2011; Song et al., 2014a; Ghosh et al., 2008; Zhu et al., 2007; Zuo et al., 2008; Zhao et al., 2009; Un et al., 2013). All these studies were conducted for removal of arsenic or fluoride separately. While, Zhao et al. (2011) had designed and used an integrated electro-oxidation and electrocoagulation process for simultaneous removal of arsenite and fluoride. Titanium based dimensionally stable anode with different combination of Fe and Al electrode were used in their research work. Cost of any treatment process is a significant factor associated with their implementation. Since, titanium based electrode was used, hence their cost of treatment was high. Although, no cost estimation study was performed by them. Moreover, initial concentrations of arsenite and fluoride were 1 mg/l and 4.5 mg/l, respectively. However, in real groundwater, the concentration of arsenic is found less (<500 µg/l) and fluoride concentration is found higher (>10 mg/l) (Nevárez et al., 2011; Aiuppa et al., 2000; Jadhav et al., 2015). Patel et al. (2015) also reported the presence of arsenic and fluoride in groundwater of Rajnandgaon district of Chhattisgarh, India, in which most of the sample contains arsenic concentration <575 µg/l and fluoride concentration >10 mg/l. According to the information available, no study has been performed for simultaneous removal of arsenic and fluoride in these range by electrocoagulation along with cost estimation.

In the present study, the lab experiments in batch mode have been performed at constant initial arsenic and fluoride concentration, to investigate the effects of initial pH, current density, run time, inter electrode distance and NaCl concentration on percentage removal of arsenic and fluoride as well as operating cost of treatment. Removal of arsenic and fluoride mainly depends on their initial concentration. Hence, variation of initial concentration on their removal efficiency has also been studied. Furthermore, the kinetic studies for individual and simultaneous arsenic and fluoride removal in electrocoagulation have also been performed to evaluate the reaction rate constants as well as interactions. To explore the mechanism of removal of arsenic and fluoride, produced sludge characterization has been conducted by Field emission scanning electron microscopy (FESEM), X-ray diffraction (XRD) and Fourier transform infrared spectroscopy (FTIR).

2. Materials and methods

2.1. Materials

All chemicals used in the present experiment were of analytical

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