Desalination and Water Treatment
www.deswater.com

doi: 10.5004/dwt.2017.21814

100 (2017) 223–230
December

Arsenic treatment from wells water by galvanostatic electrocoagulation method

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Received 11 July 2017; Accepted 10 December 2017

ABSTRACT

The removal of arsenic from simulated wells water was carried out by galvanostatic electrocoagulation (GEC) method using CT3 steel as anode material. The influences of several parameters such as current densities, electrolysis time, charge density and initial arsenic concentrations as well as the relationship of oxidation ratio between As(III) and Fe in GEC were studied. The optimum current density by GEC was 2 mA cm^{-2} at which the residual arsenic concentration in solution from initial arsenic concentrations of 251 ppb was smaller than WHO limit value (10 ppb) after 15 min. It could remove arsenic in wells water from concentration up to over 800 ppb and the residual arsenic concentration was less than 10 ppb. The maximal adsorption capacity was found 64.52 mg g^{-1}. The arsenic adsorption in GEC fitted well into the pseudo second-order model, where the equilibrium adsorption capacity decreased powerly with increase of current density, but, it increased linearly with the initial arsenic concentration. The oxidation ratio of As(III) to Fe in GEC decreased powerly and the dissolved ferric ion increased linearly with the electrolysis time. The oxidation ratio of As(III) to iron was significantly decreased until only iron was going on oxidized at the end of GEC process.

Keywords: Galvanostatic electrocoagulation; Simulation of arsenic contaminated wells water; Arsenic removal; CT3 steel electrode

1. Introduction

Arsenic contaminated groundwater has been a global problem because of the long and dangerous impact to community life [1–3]. The arsenic poisoning in along time (through drinking and food) affects human health seriously. It is known to cause troubles in the skin include pigmentation changes, skin lesion and hyperleratosis. It also can cause skin, bladder and lungs cancers [4–9]. In the world, about ten millions of people have got back and fallen nails, skin keratoses, skin cancer due to using arsenic contaminated drinking water (exceeding WHO standard for arsenic in the drinking water of 10 ppb) [10]. Many countries have found high level arsenic concentration in the ground water such as Canada, Alaska, Chile, Argentina, China, India, Thailand, Bangladesh [11]. Vietnam is one of the countries with high risk of arsenic poisoning, specially, in the Red River Delta and Mekong Delta [12,13]. Therefore, arsenic removal is a common concern of many researchers in the world [14–17].

In natural water, arsenic is usually in arsenite (As(III)) and arsenate (As(V)) forms, among them As(III) can be also oxidized to As(V) by hypochlorite using a noble metal oxide ($RuO_2/TiO_2/SnO_2$) coated titanium plate as anode [18]. Arsenate can be removed easily by adsorption method or coagulation with aluminum hydroxide and ferric hydroxide because these hydroxides have a strong affinity with dispersed/dissolved ions as well as the counter ions to cause coagulation/adsorption [19]. In order to get rid of arsenite, it is necessary to oxidize to arsenate then remove arsenate or denature adsorption materials to adsorb immediately.

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Nowadays, there are many methods to remove arsenic from water such as adsorption [20–26], electrocoagulation (EC) [27–28], coagulation [17,29] and membrane ones [30]. Among them the EC method is an optimal one because it is environment-friendly technology and it can remove thoroughly arsenic with low cost [31]. Moreover, this method does not require any supplementary addition of chemicals and expensive equipment. Materials as anode using for this method are usually aluminum, iron and magnesium [19,32–35], among them arsenic removal efficiency by iron electrode is lightly higher than that by the other ones.

In EC process, ferrous ion was released from anode and then oxidized to ferric ion leading to form ferric hydroxides [36]. On the other hand, As(III) might be oxidized to As(V) which exists as $HAsO_4^{2-}$ and $H_2AsO_4^-$ in ground water at pH from 7 to 8 [37] or only as $H_2AsO_4^-$ at pH from 3 to 7 [32]. It explains that As(V) adsorbed onto ferric hydroxides to form arsenic ferric hydroxide complexes which were separated from aqueous phase contributing to water treatment [1,8,38,39]. However, the relationship between oxidation of As(III) and iron during EC process needs more deep discussion.

This study focused on the GEC mode using CT3 steel as anode material for removing arsenic from simulated wells water containing different arsenic concentrations. The adsorption kinetics of GEC-arsenic removal and the oxidation relationship between As(III) and iron were also discussed.

2. Experimental

2.1. Preparation of wells water samples

In our study, wells water (pH = 6) without arsenic contaminant was chosen as bare electrolyte to which different As(III) (AR Grade, Merck, Germany) concentrations from 251 to 821 ppb were added for simulation of arsenic contaminated samples. The removal of arsenic was carried out by GEC mode.

2.2. Arsenic removal and detection methods

GEC process used three-electrode cell among them calomel electrode as reference, commercial grade stainless steel sheet and CT3 steel rod (diameter of 8 mm) as counter and working electrodes, respectively. Experiments were carried out on the electrochemical workstation unit IM6 (Zahner-Elektrik, Germany) at room temperature of 30°C. After this process the samples were stirred for 30 min before being filtered to separate aqueous phase. The arsenic concentration in samples before and after the treatment were determined by ICP-MS method on the Agilent 7700xICP-MS (USA).

2.3. Material characterization

The morphological structure of CT3 steel electrode before and after GEC process was characterized by scanning electron microscopy (SEM) on an equipment FE-SEM Hitachi S-4800 (Japan). The examination of arsenate ferric hydroxide in electrocoagulant was carried out by energy dispersive X-ray (EDX) analysis on JEOL JED-2300 equipment (Japan).

2.4. Some theories regarding to this study

Charge density (q) is the amount of charge delivered per liter of water treated (in CL^{-1}). It can be calculated from Eq. (1):

$$q = \frac{I \cdot t}{V} \tag{1}$$

where I is the passing current (in A), t is the electrolysis time (in s) and V is the solution volume (in L).

The quantity of ferric ion dissolved into solution during GEC process can be calculated from Faraday's law [40]:

$$m = \frac{I \cdot t \cdot M}{z \cdot F} \tag{2}$$

where m (g) is the quantity of ferric ion dissolved in solution, I (A) is the current passing the GEC cell, M is the relative molar mass of iron, z is the number of electron exchange and F (26.8 Ah) is the Faraday's constant.

The Langmuir isotherm model can be described as below [41]:

$$\frac{C}{N} = \frac{1}{N_{\max} K_L} + \frac{1}{N_{\max}} C \tag{3}$$

where N (mg g^{-1}) is the arsenic amount adsorbed onto ferric hydroxide (FH) in equilibrium), N_{max} (mg g^{-1}) is the maximum adsorption capacity corresponding to monolayer coverage on the surface of FH, C (mg L^{-1}) is the arsenic concentration in solution at equilibrium and K_L (L mg^{-1}) is the Langmuir constant.

The arsenic adsorption on the electrochemically dissolved FH can be determined by the second-order kinetic model Eq. (4) [42]:

$$N_t = \frac{C_{o,As} - C_{s,As}}{w} V \tag{4}$$

$$\frac{t}{N_t} = \frac{1}{k_2 N_e^2} + \frac{1}{N_e} t \tag{5}$$

where N_t (mg g^{-1}) is the amount of arsenic species adsorbed at time t (min), $C_{o,As}$ and $C_{s,As}$ are arsenic concentrations (mg L^{-1}) before and after the GEC, respectively, w (g) is the weight of FH as adsorbent, V (L) is the solution volume, k_2 (g mg^{-1} min^{-1}) is the rate of the pseudo second-order adsorption, N_e (mg g^{-1}) is the amount of arsenic species adsorbed at equilibrium. The slope and intercept were determined by the plot of (t/N_t) vs. t.

3. Results and discussion

3.1. Arsenic treatment

3.1.1. The effect of current density

Fig. 1 demonstrates the effect of current density on the ability of arsenic treatment by changing current density from 0.1 to 4 mA cm^{-2} for the same electrolysis time of 15 min. The result shows that the arsenic removal efficiency increased with the increase of current density. It rose quickly from 19.37 to 90.20%, when current density was used from 0.1 to 1 mA cm^{-2}, respectively. It received a value of 97.99% at 2 mA cm^{-2} and then slightly increased when current density continuously increased. It can be explained that if the current density bigger than 2 mA cm^{-2} is applied, a part of it may produce oxygen evolution at the anode electrode. It is therefore advisable to use a current density of 2 mA cm^{-2} to avoid wasting energy for such unwanted process.

3.1.2. The effect of electrolysis time

The dependence of arsenic removal on electrolysis time was investigated at different current densities. Fig. 2 shows that the higher current density applied, the bigger the kinetic curve's slope, among them the biggest one was at 4 mA cm^{-2} and the smallest one was at 0.5 mA cm^{-2}.

Fig. 3 demonstrates the data calculated from Fig. 2 indicating that the required time (t_{req}) of electrolysis for obtaining WHO limit (10 ppb) powerly depended on current density following Eq. (6) with R^2 of 0.9683.

$$y = 31.589x^{-0.825} \quad (6)$$

3.1.3. The effect of charge density

The influence of charge density on the arsenic treatment ability was investigated to find necessary charge passed per liter of water at different current densities. The obtained results in Fig. 4 show that the arsenic concentration $C_{s,As}$ in the GEC decreased rapidly until charge density increased to 10 C L^{-1}, however, after that it only decreased gradually. The WHO limit value (10 ppb) was surely obtained by applying charge density of over 25 C L^{-1} for all regarded current densities. However, the charge density used should be around 30 C L^{-1} for arsenic removal to avoid energy wasting due to evolution of oxygen gas.

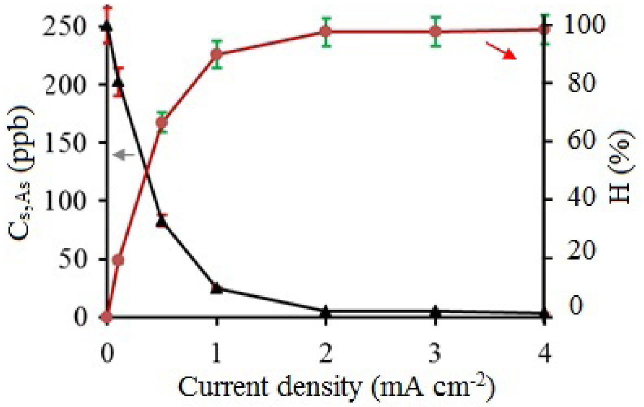

Fig. 1. Effect of current densities on the ability of arsenic treatment after electrolysis time of 15 min ($C_{0,As}$ = 251 ppb, pH = 6).

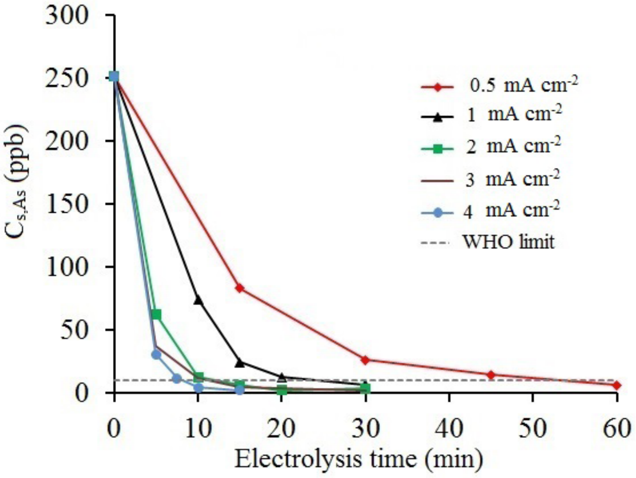

Fig. 2. Effect of electrolysis time on residual arsenic concentration at different current densities ($C_{0,As}$ = 251 ppb, pH =6).

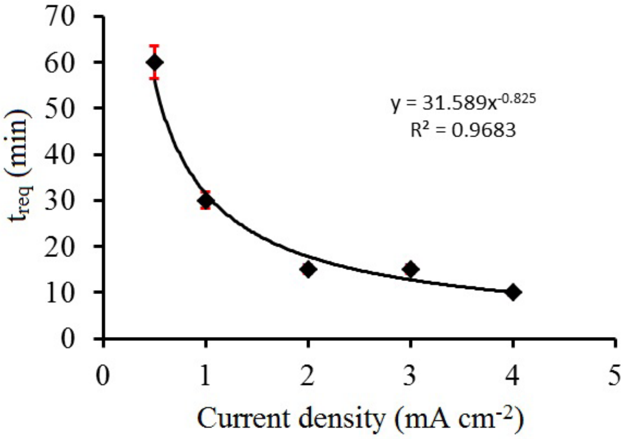

Fig. 3. Effect of current densities on required time for arsenic treatment to obtaining WHO limit (10 ppb), $C_{o,As}$ = 251 ppb, pH = 6.

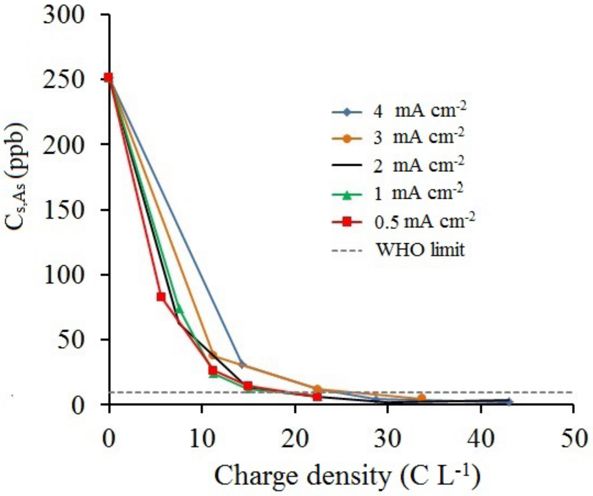

Fig. 4. Effect of charge density on the residual arsenic concentration in solution at different current densities (pH = 6).

3.1.4. The effect of initial arsenic concentration

Samples with different arsenic concentrations of 251, 410 and 821 ppb were carried out by using the constant current density of 2 mA cm^{-2} for arsenic removal. The results in Fig. 5 and Table 1 explain that the treatment process was powerly dependent not only on electrolysis time but also on initial arsenic concentration. The higher the $C_{0,As,}$ the longer the processing time needed to obtain WHO limit (10 ppb).

3.2. The oxidation relationship of As(III) and Fe in GEC process

Ferric ion can be formed directly from iron following reaction (7) or through ferrous ion according to (8) and (9) depending on anode potential and pH medium (4 < pH < 7) [43,44]:

$$\mathrm{Fe\ (s)} \rightarrow \mathrm{Fe^{3+}_{(aq)}} + 3e^{-} \tag{7}$$

$$\mathrm{Fe\ (s)} \rightarrow \mathrm{Fe^{2+}_{(aq)}} + 2e^{-} \tag{8}$$

$$\mathrm{Fe^{2+}_{(aq)}} \rightarrow \mathrm{Fe^{3+}_{(aq)}} + e^{-} \tag{9}$$

It suggests only FH formed at the end of GEC process following reaction (11) because the oxidation of ferrous ion is quickly changed to ferric ion following reaction (10) in bulk solution due to oxygen formed during electrolysis [32].

$$\mathrm{O_{2(g)}} + 4\mathrm{Fe^{2+}_{(aq)}} + 2\mathrm{H_2O_{(aq)}} \rightarrow 4\mathrm{Fe^{3+}_{(aq)}} + 4\mathrm{OH^{-}_{(aq)}} \tag{10}$$

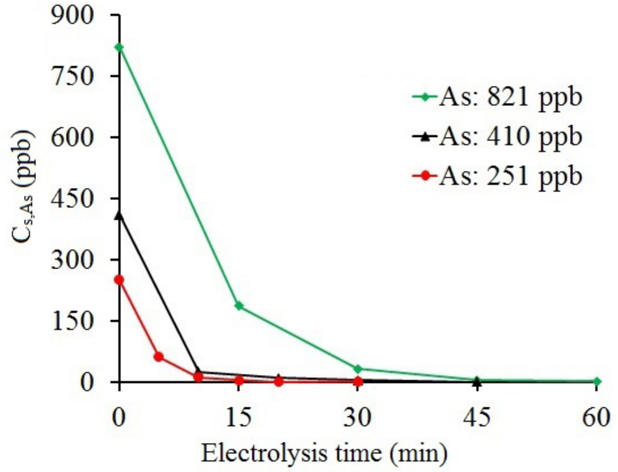

Fig. 5. Effect of initial arsenic concentration on treatment process at constant current density of 2 mA cm^{-2} (pH = 6).

Table 1
Effect of initial arsenic concentration on electrolysis time to obtain WHO limit (10 ppb)

$C_{0,As}$ (ppb)	$C_{s,As}$ (ppb)	t_{req} (min)	H (%)
251	3.34	15	98.67
410	6.41	30	98.44
821	4.42	45	99.46

$$\mathrm{Fe^{3+}_{(aq)}} + 3\mathrm{OH^{-}_{(aq)}} \rightarrow \mathrm{Fe(OH)_{3(s)}} \tag{11}$$

Arsenic that exists in form of H_3AsO_3 at pH under 7 could be oxidized to the form of As(V) as $H_2AsO_4^-$ following reaction (12) [45] during GEC process.

$$\mathrm{H_3AsO_{3\ (aq)}} + \mathrm{H_2O} \rightarrow \mathrm{H_2AsO^{-}_{4\ (aq)}} + 3\mathrm{H^{+}} + 2e^{-} \tag{12}$$

In fact, the amount of arsenic that is treated is the amount of As(III) oxidized to As(V) and the adsorption kinetics by GEC-arsenic removal can be discussed based on the following reaction (13):

$$\mathrm{Fe(OH)_{3(s)}} + \mathrm{H_2AsO^{-}_{4\ (aq)}} \rightarrow [\mathrm{Fe(OH)_3{\cdot}H_2AsO_4^{-}}]_{(s)} \tag{13}$$

According to reaction (13), one molar of iron can adsorb one molar of arsenic. However, according to Faraday's law [40], one third molar and half one of iron and As(III) can be oxidized at the same current passing through GEC cell, respectively. It explains that at the first minute in GEC process one sixth molar of arsenic is redundant in comparison with ferric ion until almost arsenic is removed from solution and at this moment there is only oxidation of iron to form ferric ion. Additionally, the removed arsenic amount that is the oxidized As(III) at the same time resulting to the ferric concentration can be calculated from experimental data which is presented in Figs. 6 and 7. It shows that the oxidation ratio of As(III) to iron significantly decreases until only iron is going on oxidized at the end of GEC process.

Fig. 7 explains that a powerly decrease of As(III) oxidized to As(V) was found at different initial arsenic concentrations, while ferric ion increased linearly with electrolysis time. It suggests that the time for arsenic removal processing by GEC shouldn't be longer than t_{req} (Table 1) corresponding to each initial arsenic concentration regarded above.

3.3. The adsorption model of arsenic in GEC cell

Fig. 8 was set up from the values calculated by Eqs. (2)–(4). It explains that the arsenic adsorption onto FH was fol-

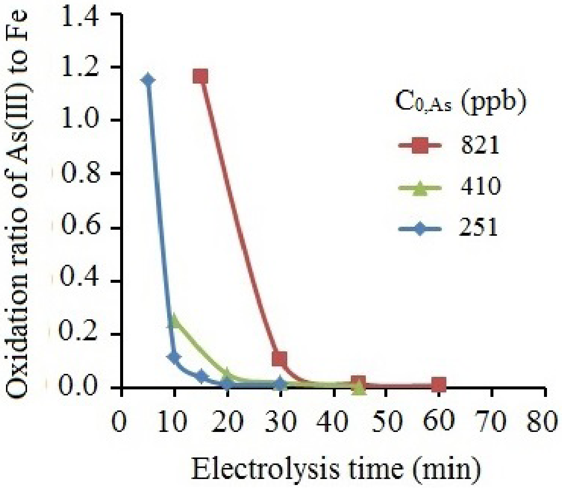

Fig. 6. The effect of initial arsenic concentration on oxidation ratio of As (III) to Fe in GEC cell (pH = 6, i = 2 mA cm^{-2}).

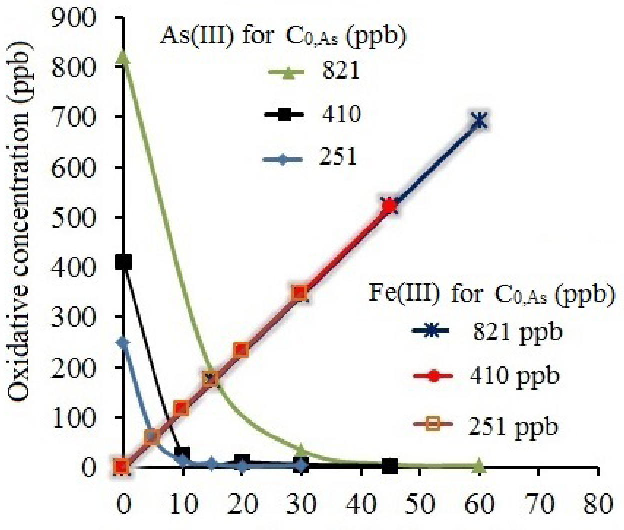

Fig. 7. The effect of initial arsenic concentration on oxidative ability of As(III) and Fe during GEC processing (pH = 6, i = 2 mA cm^{-2}).

Table 2
Comparison of Langmuir parameters of our study with some literatures by GEC at 2 mA cm^{-2}

q_{max} (mg g^{-1})	K_L (L mg^{-1})	R_L	R^2	H (%)	Ref.
25.387	0.0766	0.8555	0.9995	97.9	[19]
55.959	0.0171	0.9831	0.9995	98.4	[34]
30.844	0.0895	0.9889	0.9992	98.6	[35]
64.520	77.5192	0.0489	0.9951	98.0	This study

lowed the Langmuir's isotherm model because R^2 received a very high value (0.9951), which indicated that arsenic was adsorbed in the form of monolayer coverage on the surface of the adsorbent (FH) [46]. The maximum adsorption capacity of 64.52 mg g^{-1} and the Langmuir's constant of 77.5 L mg^{-1} for arsenic were found in our study (Table 2), which are much higher than those published in [19,34,35].

The dimensionless parameter (R_L) can be obtained by equation below:

$$R_L = \frac{1}{1+\left(K_L C_{0,As}\right)} \qquad (14)$$

where K_L is Langmuir's constant and $C_{0,As}$ is initial arsenic concentration. The parameter R_L given on Table 3 indicates that the arsenic adsorption was favourable because of $0 < R_L < 1$ [47].

3.4.Thermodynamic study

The thermodynamic parameter such as standard Gibbs free energy (kJ mol^{-1}) can be calculated from Langmuir's constant K_L, which is related to the energy of adsorption, following equation [44]:

$$\Delta G^0_{ads} = \; -RT\; ln\; K_L \qquad (15)$$

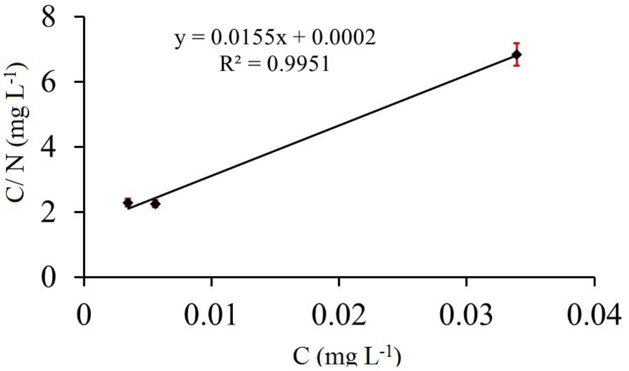

Fig. 8. Langmuir's isotherm model of arsenic adsorption onto ferric hydroxide in GEC cell (pH = 6, i = 2 mA cm^{-2}).

Table 3
Values of dimensionsless Langmuir's parameter for arsenic adsorption

$C_{0,As}$ (ppb)	$C_{s,As}$ (ppb)	R_L
251	3.45	0.0489
410	5.60	0.0305
821	33.90	0.0155

where R is the gas constant (8.314 J mol^{-1} K^{-1}) and T is the absolute temperature (K). The constant k_0 can be determined by plotting ln (C_s/C_e) vs. C_s (Fig. 9) and extrapolating C_s to zero [48], where C_e and C_s are the arsenic concentration in solution (mg L^{-1}) and in the solid-phase at equilibrium (mmol g^{-1}), respectively. The constant k_0 was 7.954 and ΔG^0_{ads} received a negative value (–10.96 kJ mol^{-1}) indicating that the arsenic adsorption on FH by GEC is a spontaneous process [46,51–53]. The enthalpy (ΔH^0_{ads}) and entropy (ΔS^0_{ads}) were calculated by using equations below [44]:

$$\ln\left(K_L\right) = k_0 - \left(\frac{\Delta H^0_{ads}}{\mathrm{RT}}\right) \qquad (16)$$

$$\Delta G^0_{ads} = \Delta H^0_{ads} - \Delta S^0_{ads} \qquad (17)$$

The positive standard enthalpy change of 1.43 J mol^{-1} in this study explains that the adsorption of arsenic by FH is lightly endothermic process. The positive standard entropy change of 0.036 J mol^{-1} K^{-1} indicating the affinity of the iron species towards arsenic ones [48].

3.5. The adsorption kinetics of GEC-arsenic removal

3.5.1. The effect of current density

The results presented in Fig. 10 and Table 4 show that the adsorption capacity at equilibrium (N_e) decreased powerly ($y = 18.984x^{-0.507}$; $R^2 = 0.8265$), while the rate constant (k_2) of the pseudo second-order kinetics for adsorption ($y = 0.0054x + 0.0005l$; $R^2 = 0.9697$) was very small and linearly increased, but very lightly, by raising current density passing through GEC cell.

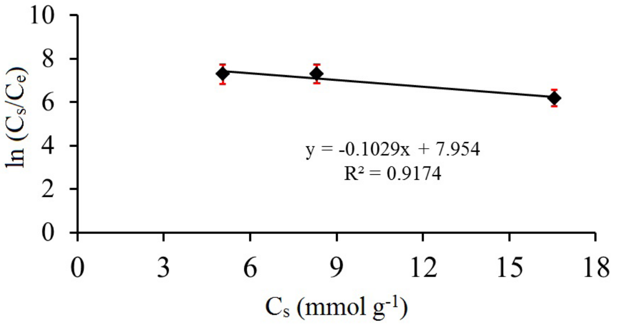

Fig. 9. Plot of ln (C_s/C_e) versus C_s (pH = 6, i = 2 mA cm^{-2}).

Table 4
The effect of current density on the adsorption kinetics by iron electrode in the GEC process (pH = 6, $C_{0,As}$ = 251 ppb)

i (mA cm^{-2})	k_2 (g mg^{-1} min^{-1})	N_e (mg g^{-1})	R^2
0.5	0.003	24.81	0.97
1.0	0.005	23.15	0.97
2.0	0.012	12.90	0.95
3.0	0.019	8.33	0.97
4.0	0.020	11.34	0.98

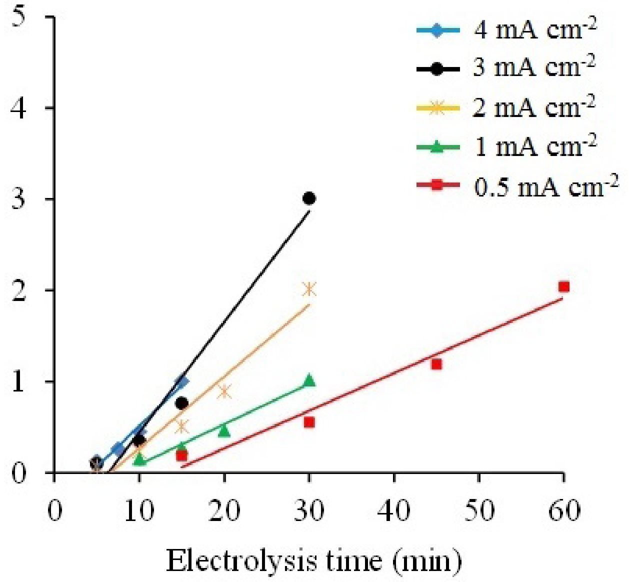

Fig. 10. Effect of current density on pseudo second-order plots for arsenic adsorption on ferric hydroxide from $C_{0,As}$ = 251 ppb at pH of 6.

3.5.2. *The effect of initial arsenic concentration*

The kinetics model fitted into the experimental data in Fig. 11 and Table 5 because of the high correlation coefficients (0.95–0.96). It demonstrates that the adsorption capacity at equilibrium increased linearly ($y = 0.0141x + 8.6652$; $R^2 = 0.9576$), while the rate constants of the pseudo second-order adsorption ($y = -6E\text{-}06x + 0.0093$; $R^2 = 0.9966$) were small and linearly reduced, but very lightly, by raising initial arsenic concentration in GEC cell.

3.6. *SEM analysis and EDX study*

The SEM images of the CT3 steel electrode before and after GEC process presented in Fig. 12 show that the surface of original electrode (Fig. 12a) was uniform, while its surface was roughly changed with a number of dents after GEC one (Fig. 12b). These dents were FH which were the results of anode electrode dissolution during GEC processing [35].

Fig. 13 shows that the presence of arsenate appears in the spectrum other than the principal elements Fe and O. It is an evidence that arsenate is adsorbed on FH. Other elements such as Al and Si detected in the electrocoagulant come from the impurities of anode electrode because of using CT3 steel.

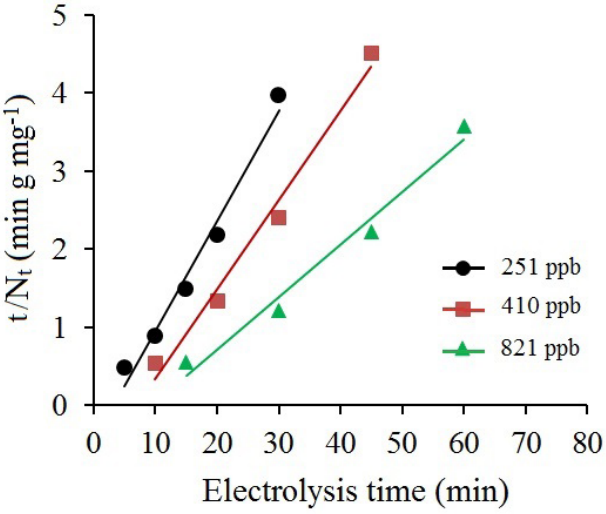

Fig. 11. Effect of initial arsenic concentrations on pseudo second-order plots for arsenic adsorption on ferric hydroxide at pH of 6 (i = 2 mA cm^{-2}).

Table 5
The effect of initial arsenic concentration on the adsorption kinetics by iron electrode in the EC process (pH = 6, i = 2 mA cm^{-2})

$C_{0,As}$ (mg L^{-1})	k_2 (g mg^{-1} min^{-1})	N_e (mg g^{-1})	R^2
251	0.0079	12.90	0.95
410	0.0068	13.46	0.96
821	0.0047	20.49	0.96

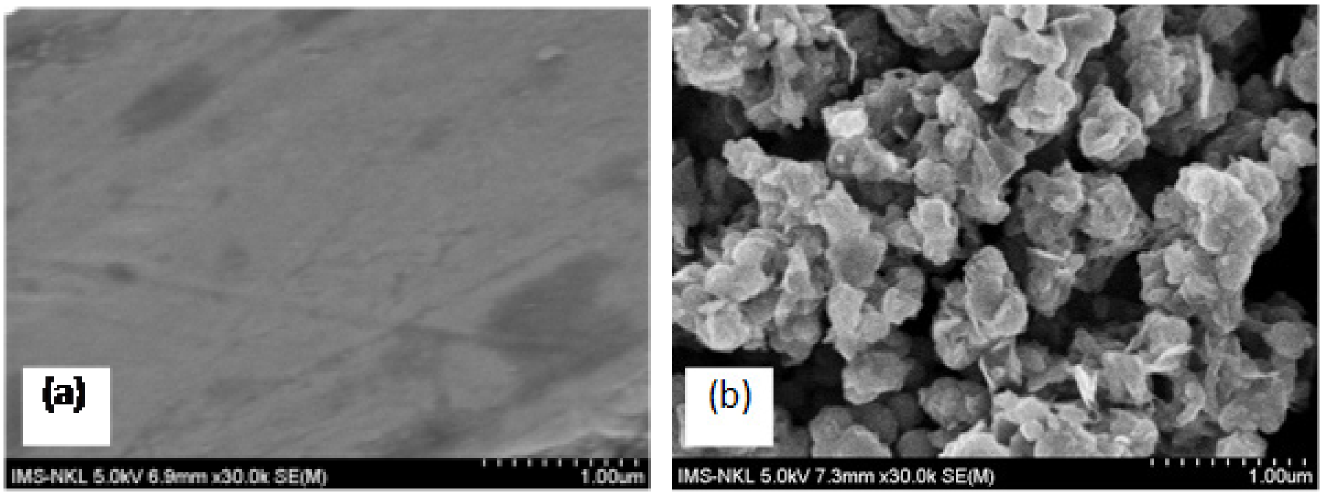

Fig. 12. SEM images of CT3 steel anode (a) before and (b) after treatment.

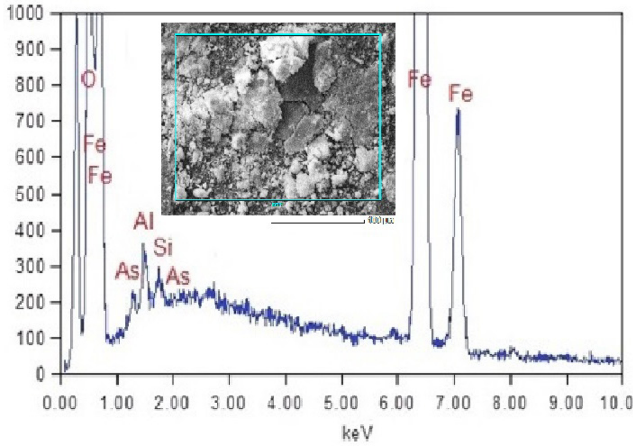

Fig. 13. EDX spectrum of arsenate-adsorbed ferric hydroxide.

4. Conclusion

The time required for electrolysis to reach WHO limit (10 ppb) for arsenic decreased when the GEC current density was raised following equation $y = 31.589x - 0.825$. The suitable GEC current and charge densities were 2 mA cm^{-2} and 30 C L^{-1}, respectively. The maximum adsorption capacity of arsenic was found 64.52 mg g^{-1}. The oxidation ratio of As(III) to Fe in GEC decreased powerly and the dissolved ferric ion increased linearly with the electrolysis time. The arsenic adsorption fitted into the second-order kinetic model, in which its rate constant increased lightly by raising current density passing through GEC, also decreased lightly by increasing initial arsenic concentration. The results in this study indicate that arsenic is spontaneously adsorbed onto ferric hydroxide generated by GEC, which is a lightly endothermic process and the affinity of the iron species towards arsenic ions. From the results of this study, it indicates that the CT3 steel material can be used for arsenic treatment from wells water by galvanostatic electrocoagulation.

Acknowledgements

This study was financially supported by Vietnam Academy of Science and Technology under code number VAST07.02/17-18.

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