

Comparative Study of As (III) Removal Efficiency from Water by Electrocoagulation and Conventional Coagulation

Monica Ihoș*, Adina Negrea**, Lavinia Lupa**, Mihaela Ciopec**, P. Negrea**

* National Research and Development Institute for Industrial Ecology–ECOIND, P-ta Victoriei nr.2, 300006 Timisoara, Romania
Phone/Fax: ++40256220369, E-Mail: monica.ihos@rectorat.utt.ro

** "Politehnica" University of Timisoara, Faculty of Industrial Chemistry and Environmental Engineering, P-ta Victoriei No.2, 300006 Timisoara, Romania, Phone: ++40256404191, E-Mail: petru.negrea@chim.upt.ro

Abstract: The purpose of this paper was As (III) removal from simulated water by electrocoagulation (EC). The working conditions were: pH = 7, As (III) concentration 100 µg/L, and 0.01 M NaCl as supporting electrolyte. The sacrificial anode was made of aluminium and applied current densities were 25, 50 and 150 A/m² at various electrolysis times. The results of EC were compared with those obtained by conventional coagulation (CC). The coagulation reagent was AlK(SO₄)₂·12H₂O at the same concentration of Al³⁺ in solution as in EC experiments and working conditions: solutions of 100 µg/L As (III) and pH = 7. By comparison, As (III) was practically removed from solution by EC, while arsenic residual concentration was 63.08 mg/L for 604 mg/L of alum by CC.

Keywords: arsenic removal, conventional coagulation, electrocoagulation, water treatment

1. Introduction

Arsenic is a naturally occurring metalloid, which is widely distributed in nature. Recently, arsenic in drinking water attracted attention because some of the drinking water resources contain considerable concentrations of arsenic, which causes acute and chronic symptoms in many countries, especially in Bangladesh, China, Mongolia and Taiwan [1].

Because of As toxicity, the World Health Organisation has recommended an arsenic standard of 10 µg/L, while both the European Union and United States set a regulation limit of 10 µg/L for arsenic removal [2]. In Romania, the maximum allowed concentration of arsenic in drinking water is also 10 µg/L and it represents the limit allowed by the Law 458/2002 – Law concerning the drinking water quality.

Arsenic can be released into the environment through geological events, such as volcanic activity and erosion. Some industrial processes, such as mining, smelting and production of paints, metals, soaps, dyes, drugs, semi-conductors, pesticides and wood preservatives, may also release arsenic into the environment.

Arsenic in groundwater occurs in two oxidation states, As(III) (arsenite) and As(V) (arsenate). As (III) is more mobile in groundwater and 25-60 times more toxic than As(V) [3].

Various treatment methods have been applied to remove arsenic from drinking water. These methods involve: adsorption-coprecipitation using iron and aluminium salts; adsorption on activated alumina, activated carbon and activated bauxite; reverse osmosis; ion exchange and oxidation followed by filtration [4].

EC is an emerging water treatment technology and could be a good choice to remove As (III) from water: the amount of required chemicals is much lower, a smaller amount of sludge is produced, no mixing of chemical is required, coagulant dosing as well required overpotentials can be easily calculated and controlled, operating costs are much lower when compared with most of the conventional technologies [5]. The aim of this paper was to assess the efficiency removal of arsenic(III) from water by EC and to compare it with those achieved by CC.

2. Experimental

2.1. Solutions

2.1.1. Solution used in EC

Stock solution of 1000 mg/L As (III) was obtained by dissolving the appropriate quantity of As₂O₃ (Merck) in 2N NaOH, neutralising with 2 N H₂SO₄ and then diluting the solution up to 1 liter with distilled water. Working solution of 100 µg/L As (III) and pH = 7 were prepared freshly for each experiment by successive dilutions of stock solutions and adjusting the pH with NaOH.

The experiments were carried out at pH = 7 because most of arsenic contaminated ground waters have the pH in the range of 6 – 8.

The supporting electrolyte was 0.01 M Na Cl.

2.1.2. Solutions used in CC

Arsenic solutions were prepared according to section 2.1.1. Coagulant stock solution of 5g/L Al³⁺ and pH = 2 was prepared by dissolving the appropriate quantity of AlK(SO₄)₂·12H₂O (Reactivul Bucuresti) in distilled water and pH adjusting with H₂SO₄.

The pH was adjusted with 0.5 M Na_2CO_3 which was prepared by dissolving the appropriate quantity of Na_2CO_3 (Reactivul Bucuresti) in distilled water.

2.2. EC experiments

Arsenic removal by EC was carried out in a plexiglass cell with horizontal electrodes. The sacrificial anode was made of aluminium with an active surface area of 60 cm^2 . The cathode was in the shape of grid made up of stainless steel (3 mm diameter). The distance between the electrodes was 5 mm. The electrolysis set-up is shown in Fig.1.

Volumes of 500 ml solution of $100\text{ }\mu\text{g/L}$ As (III) were introduced in the cell, and the applied current densities were 50, 100 and 150 A/m^2 . Electrolysis durations were 5, 10, 15, 30 and respectively 60 minutes.

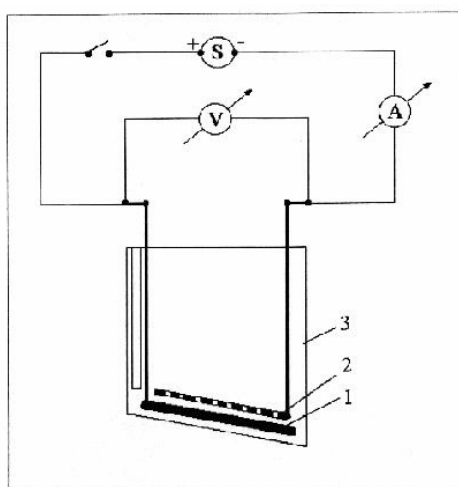


Figure 1. Batch experimental set-up for arsenic removal from water by EC in a cell with horizontal electrodes

1- anode; 2- cathode; 3 – electrolysis cell; A – ammeter; V – voltmeter; S – d.c. power supply

The electrolysed solutions were settled for 40 minutes and then samples were taken for measuring residual concentrations of arsenic and aluminium.

2.3. CC experiments

CC experiments were conducted using a jar test apparatus. The working conditions were: 500 ml solution of $100\text{ }\mu\text{g/L}$ As (III), $\text{pH} = 7$, time of slow stirring 15 minutes. Then the solutions were settled for 40 minutes and samples from solution (supernatant) were taken for measuring the residual concentrations of arsenic and aluminium.

2.4. Analytical methods

2.4.1. Arsenic determination

Arsenic in water samples was measured on a Varian SpectrAA atomic absorption spectrophotometer equipped

with with hydride system. Argon carried AsH_3 to a $900\text{ }^\circ\text{C}$ quartz cell where As was quantified at 197.2 nm . The detection limit for arsenic is $0.1\text{ }\mu\text{g/L}$.

2.4.1. Aluminium determination

Aluminium was determined spectroscopically with eriocromcyanin R. The measurements were carried out by using a Jasco V-530 spectrophotometer controlled by computer at a wavelength of 535 nm . The detection limit for aluminium is 0.01 mg/L .

3. Results and discussion

3.1. EC experiments

In EC (sacrificial anode aluminium) arsenic removal is governed by the formation of very active aluminium hydroxo-complexes. According to Faraday's law, the charge passed to the solution is directly proportional to amount of aluminium dissolved. This implies that arsenic removal by EC is due to the formation of arsenic – aluminium hydroxo-complexes.

Fig. 2 – 4 are show the residual arsenic concentration at different current densities versus charge passed to the solution. It can be seen the rapid removal of arsenic at the beginning of the process.

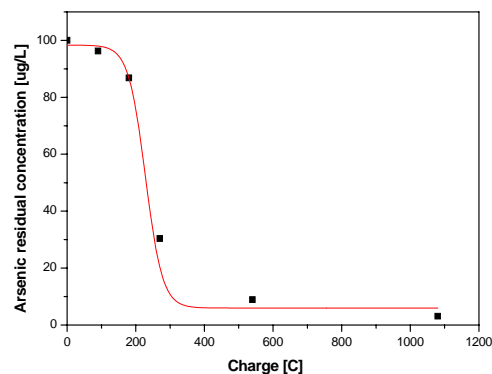


Figure 2. Dependence of arsenic residual concentration versus charge at 50 A/m^2

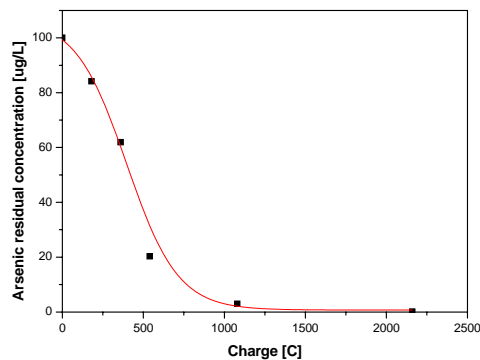


Figure 3. Dependence of arsenic residual concentration versus charge at 100 A/m^2

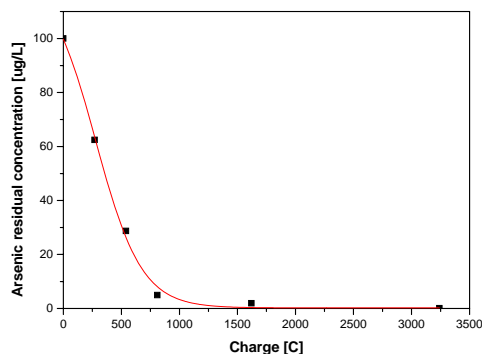


Figure 4. Dependence of arsenic residual concentration versus charge at 150 A/m²

One can also notice that the current density has not a significant effect on the final removal of arsenic from solution, but the removal is faster as the current density is higher. Thus, residual arsenic concentration at 3600 C is 3.08 µg/L at an applied current density of 50 A/m² and it is almost entirely removed from solution at 100 and 150 A/m².

In drinking water treatment, the electrodic material should not have toxic effects on human health. This is the reason for measuring the residual aluminium concentration. Results are presented in Table 1.

As it is shown in Table 1, aluminium did not exceed the limit detection of the method. Therefore, water contamination does not occur when aluminium is used as an electrode material for As removal by EC.

3.2. CC experiments

Coagulant doses were established in accordance with Al³⁺ quantities generated electrochemically. Results are shown in Table 2.

TABLE 1. Working conditions and aluminium residual concentrations for arsenic solution of 100 µg/L As(III) and pH=7

| Current density (A/m ²) | Time (min) | Cell voltage (V) | Aluminium residual concentration (mg/L) |
|-------------------------------------|------------|------------------|---|
| 50 | 5 | 2.3 | <0.01 |
| | 10 | 2.2 | <0.01 |
| | 15 | 2.2 | <0.01 |
| | 30 | 2.2 | <0.01 |
| | 60 | 2.1 | <0.01 |
| 100 | 5 | 4.1 | <0.01 |
| | 10 | 4.2 | <0.01 |
| | 15 | 4.1 | <0.01 |
| | 30 | 3.8 | <0.01 |
| | 60 | 3.6 | <0.01 |
| 150 | 5 | 5.4 | <0.01 |
| | 10 | 5.4 | <0.01 |
| | 15 | 5.4 | <0.01 |
| | 30 | 5.4 | <0.01 |
| | 60 | 5.0 | <0.01 |

TABLE 2. Coagulant dose, residual concentration of arsenic and aluminium

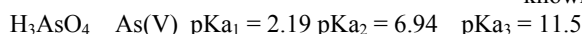
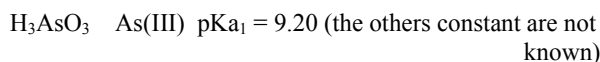
| Coagulant dose/ mg Al ³⁺ /L | Arsenic residual concentration/ µg/L | Aluminium residual concentration/ mg/L |
|--|--------------------------------------|--|
| 200 | 100 | <0.01 |
| 302 | 93.35 | <0.01 |
| 402 | 86.77 | <0.01 |
| 604 | 63.08 | <0.01 |

*Working conditions: 100 µg/L As (III) and pH=7

By comparison, it is obvious that EC is more effective for arsenic (III) removal than CC. In CC residual arsenic concentration decreased by 30 – 40 % while by EC As is almost entirely removed.

This behaviour is in accordance with the possibility of anodic oxidation and in situ generation of adsorbents in EC. Thus, As(III) is oxidised to As(V) during EC, and it is adsorbed onto generated metal hydroxides. Studies of arsenic removal from water by EC accomplished by Ratna et al [3] pointed out the oxidation of As(III) to As(V).

Both species As(III) and As(V) have significant differences in the chemical behaviour: the equilibrium constants of dissociation of the two oxidation state of arsenic are different and are reported below [6]:



Removal efficiency is much more lower for As (III) than for As(V). Actually, anionic species are often involved in the surface chemical reaction with the binding sites, but in pH range of most groundwaters (6 – 8), the anionic species of As(III), H₂AsO₃⁻, are present in relatively low concentrations.

In many cases, the sorbent surface is positively charged up to pH 7 and at this pH, As(III) species are uncharged and therefore can not be bounded to the surface by electrostatic reaction.

Also for CC, the water contamination with Al³⁺ do not occur. The determined residual aluminium concentration are under the limit detection of the method for all samples.

4. Conclusions

This paper deals with As(III) removal from waters by electrocoagulation. Removal efficiencies in electrocoagulation were compared with conventional coagulation.

The working solutions had 100 µg/L As(III) and pH = 7. The experiments were conducted at this value of pH because most of groundwaters have the pH in the range of 6 – 8.

In conventional coagulation, coagulant doses were established so that Al^{3+} concentration in solution was similar to Al^{3+} generated electrochemically.

The applied current densities were 50, 100 and 150 A/m^2 for electrolysis times of 5, 10, 15, 30 and 60 minutes. Under these circumstances, the residual arsenic concentrations were lower than 10 $\mu g/L$. For 60 minutes of electrolysis and applied current densities of 100 and 150 A/m^2 , arsenic is almost entirely removed from water, while only 30 – 40% of As(III) is removed by conventional coagulation.

Neither in electrocoagulation, nor in conventional coagulation contamination of water with aluminium occur.

References

1. Ming-Cheng Shih, *Desalination*, **2005**, 172, 85
2. W. Driehaus, R. Aguirre, *Water&Wastewater International*, **2005**, 20, 11
3. P. Radna Kumar, Sanjeev Chaudhari, Kartic C. Khilar, S.P. Mahajan, *Chemosphere*, **2004**, 55, 1245
4. T. Viraraghavan, K. S. Subramanian, J. A. Aruldoss, *Wat. Sci. Tech.*, **1999**, 40, 69
5. K. Rajeshwar, J. G. Ibanez, *Environmental Electrochemistry*, Academic Press, San Diego, **1997**, 410
6. S.D. Wilson, W.R. Kellz, T.R. Holm, J.L. Talbott, Arsenic Removal in *Water Treatment Facilities: Survey of Geochemical Factors and Pilot Plant Experiments*, **2005**, 1