SIMULTANEOUS FLUORIDE AND ARSENIC REMOVAL FROM GROUNDWATER BY USING ALUMINUM SACRIFICIAL ANODE

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Abstract

The simultaneous removal of fluoride and arsenic in groundwater collected from three deep wells situated in Western part of Romania was carried out by electrocoagulation (EC) with aluminium sacrificial anode. The current density was of 156, 480 and 780 A/m^2, respectively and the electrolysis time 60 min. The removal efficiency of fluoride and arsenic was determined based on their residual concentration. Also, the specific energy consumption was calculated and it was in the range of 0.14 - 1.40 kWh/m^3.

Introduction

Groundwater is one of the most important sources of drinking water in the world. The most significant inorganic pollutants in the groundwater that have harmful effects on human health are fluoride and arsenic. However it should be noted that fluoride has beneficial effects on human health in the range of 0.5 - 1 mg/l in drinking water, and above 1.5 mg/l represents a risk of dental fluorosis and progressively higher concentrations lead to increasing risks of skeletal fluorosis. Also, the guideline value for arsenic is 10 µg/L [1].

Many people in the world are subjected to drinking water that contains concomitant fluoride and arsenic. The effects following the concomitant exposure to fluoride and arsenic need comprehensive studies because in the human body these pollutants may function independently or can act as synergistic or antagonistic to one another [2]. Salgado-Bustamante et al. have studied the effect of exposure to fluoride and arsenic on the pattern of expression of apoptosis and inflammatory genes by immune cell. The results show that the combined exposure to arsenic and fluoride has a different effect on gene expression than the exposure to arsenic or fluoride alone [3].

Research was carried out in order to develop technologies that are able to remove simultaneously fluoride and arsenic: adsorption [4-9] and electrochemical methods [10, 11].

In the last years electrocoagulation (EC) has been considered as water treatment for drinking purposes because of electrochemical methods advantages such as versatility, energy efficiency, easy operation, automation and environmental compatibility.

The purpose of this paper was to remove simultaneously the fluoride and arsenic in groundwater from the West of Romania by EC with aluminium as sacrificial anode.

Experimental

The groundwater was collected from three deep wells situated in Western part of Romania. The characteristics of groundwater are presented in Table 1.

The working solutions were prepared from the groundwater by adjusting the pH to 7 with NaOH p.a., adding NaCl p.a.so that its concentration was 0.01 M. Also, NaF p.a. was added so that the concentration of fluoride was 5.08 and 10 mg/L, respectively.

The EC experiments were carried out in a plexiglass cell with horizontal electrodes. The sacrificial anode was made of aluminium with an active surface area of 78.4 cm^2. The cathode was a wire mesh grid made up of 3 mm diameter stainless steel wires. The distance between the electrodes was 5 mm.

Volumes of 500 ml working solutions were introduced in the cell, and the current density was of 156, 480 and 780 A/m^3, respectively. Electrolysis duration was 60 minutes and samples were taken at every 10 minutes.

The fluoride concentration was determined by using a Thermo Scientific Orion fluoride ion selective electrode. TISAB II solution was used as a buffer to maintain the pH and background ion concentrations. The detection limit for fluoride is 0.02 mg/L. Arsenic concentration was measured on a Varian SpectrAA atomic absorption spectrophotometer equipped with hydride system. Argon carried AsH_3 to a 900 ˚C quartz cell where arsenic was quantified at 197.2 nm. The detection limit for arsenic is 0.1 µg/L.

Table 1. Characteristics of groundwater

Parameter	**Unit of measurement**	**Groundwater**		
		1	**2**	**3**
Depth	m	110	250	300
Turbidity	NTU	2.47	0.01	0.02
Conductivity	µS/cm	780	750	540
pH	pH units	7.6	7.9	7.9
Ammonia	mg/L	0.291	0.519	2.31
Nitrate	mg/L	< 0.074	< 0.074	< 0.074
Nitrite	mg/L	< 0.026	< 0.026	< 0.026
Chemical oxygen demand (COD-Mn)	mgO$_2$/L	< 1.6	< 1.6	< 1.6
Hardness	German degree	9.1	2.9	6.4
Chloride	mg/L	58.1	41.1	20.6
Sulphate	mg/L	5.40	4.58	4.39
Iron	mg/L	0.880	0.210	0.208
Manganese	mg/L	0.355	0.089	0.118
Arsenic	µg/L	71.6	98.4	99.5
Fluoride	mg/L	0.093	0.539	0.017

Results and discussion

Figures 1-3 show the fluoride removal efficiency versus electrolysis time at 156, 480 and 780 A/m^2, respectively. Regardless of fluoride concentration in the groundwater, as the electrolysis time and the current density increased, the fluoride removal efficiency increased. This finding is in accordance with the fact that in EC (sacrificial anode aluminium) the removal of the pollutants is managed by aluminium hydroxide yielding in solution. According to Faraday's Law, the amount of aluminium hydroxide is directly proportional to the charge passed into solution. The pollutants removal occurs by formation of pollutant-aluminium hydroxo-complexes. As the amount of aluminium hydroxide is larger the adsorbed pollutant amount will be larger.

The results listed in Table 2 show that simultaneously with fluoride removal, the arsenic was efficiently removed too. The arsenic residual concentration in the groundwater collected from the three deep wells after 60 min of electrolysis and 780 A/m^2, regardless of fluoride/arsenic mass ratio in the untreated groundwater is under 10 µg/L.

In Table 3 specific energy consumption for 60 min of electrolysis and the applied current density in the EC are presented. The values are in accordance with those reported in the literature [10].

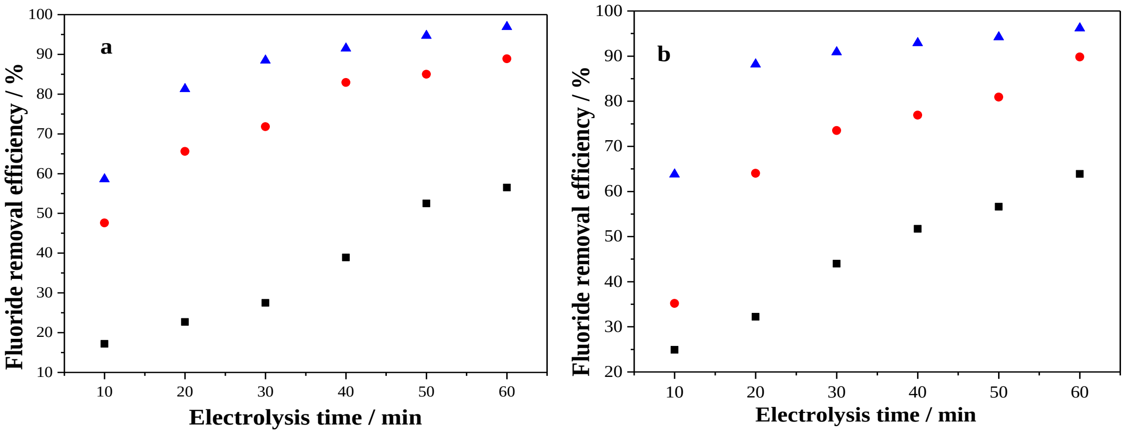

Figure 1. Fluoride removal efficiency versus electrolysis time for groundwater 1 current density: (■) - 156 A/m^3; (●) – 480 A/m^3; (▲) – 780 A/m^3; pH_{ini}=7; conc$_{ini}$: **a.** 10 mg/L F^-; **b.** 5.08 mg/L F^-

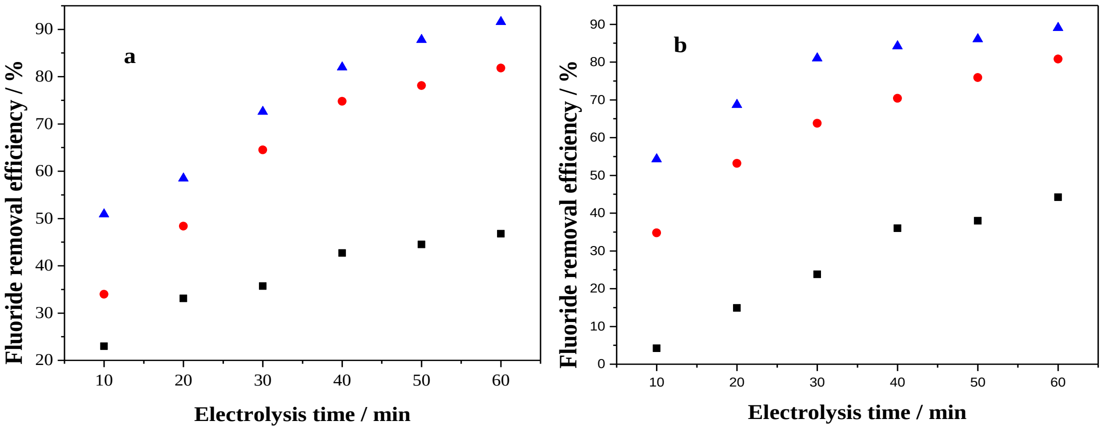

Figure 2. Fluoride removal efficiency versus electrolysis time for groundwater 2 current density: (■) - 156 A/m^3; (●) – 480 A/m^3; (▲) – 780 A/m^3; pH_{ini}=7; conc$_{ini}$: **a.** 10 mg/L F^-; **b.** 5.08 mg/L F^-

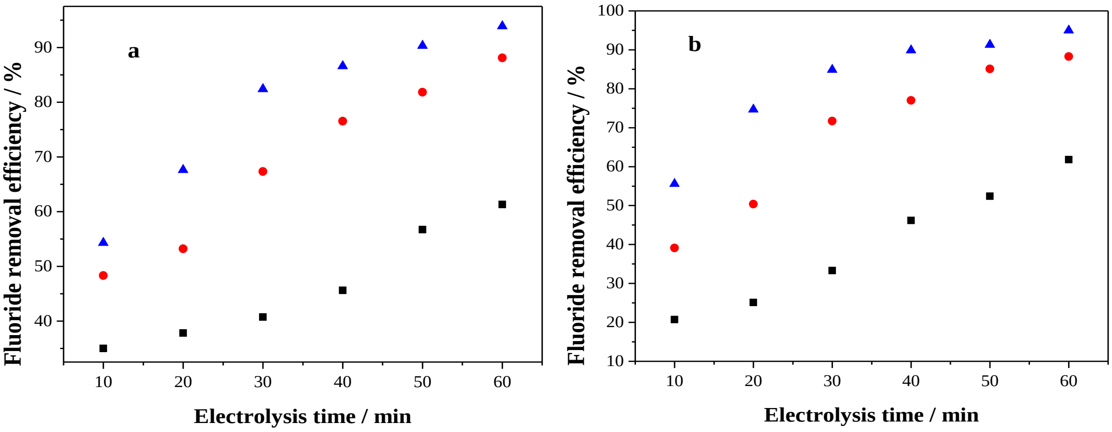

Figure 3. Fluoride removal efficiency versus electrolysis time for groundwater 3 current density: (■) - 156 A/m^3; (●) – 480 A/m^3; (▲) – 780 A/m^3; pH_{ini}=7; conc$_{ini}$: **a.** 10 mg/L F^-; **b.** 5.08 mg/L F^-

Table 2. Arsenic concentration in treated groundwater for 60 min of electrolysis and current density of 780 A/m^3

Groundwater	Fluoride/arsenic mass ratio in untreated groundwater	Arsenic concentration in untreated groundwater/ µg/L	Arsenic concentration in treated groundwater/ µg/L	Arsenic removal efficiency / %
1	140	71.56	< 0.1	> 99.9
2	102	98.36	7.39	92.5
3	101	99.47	2.07	97.9
1	71	71.56	2.75	96.2
2	52	98.36	1.10	98.9
3	51	99.47	1.69	98.3

Table 3. Specific energy consumption for 60 min of electrolysis

Current density / A/m^3	Cell voltage / V	Current intensity / A	Treated groundwater / L	Specific energy consumption / kWh/m^3
156	0.9	0.08	0.5	0.14
480	1.4	0.24		0.67
780	1.8	0.39		1.40

Conclusion

EC proved to be effective in simultaneous removal of fluoride and arsenic from the groundwater collected from three deep wells situated in Western part of Romania regardless of the fluoride/arsenic mass ratio. The residual concentration of fluoride and arsenic in treated groundwater were under the threshold limits stipulated in Law 458/2002 concerning the drinking water quality: 1.2 mg/L and10 µg/L, respectively. The removal efficiency of fluoride was in the range of 90-97% and of arsenic 93-99.9% for 60 min of electrolysis and current density of 780 A/m^3 regardless of where the groundwater was collected from.

Acknowledgements

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