



M 2017

**U. PORTO**  
**FEUP** FACULDADE DE ENGENHARIA  
UNIVERSIDADE DO PORTO

# REMOVAL OF CONTAMINANTS FROM WATER BY ELECTROCOAGULATION

**JOÃO MIGUEL FERREIRA DE ALMEIDA E SILVA**  
DISSERTAÇÃO DE Mestrado APRESENTADA  
À FACULDADE DE ENGENHARIA DA UNIVERSIDADE DO PORTO EM  
ENGENHARIA QUÍMICA

# Integrated Master in Chemical Engineering

## *Removal of contaminants from water by electrocoagulation*

### Master Thesis

of

João Miguel Ferreira de Almeida e Silva

Developed within the Dissertation course

Held in

LSRE - LCM – Laboratory of Separation and Reaction Engineering - Laboratory of Catalysis and Materials



Supervisors: Doctor Ana Mafalda Ribeiro

Doctor Nuno Graça

Professor Alírio Rodrigues



Department of Chemical Engineering

February 2017

## Acknowledgments

No desenvolvimento desta tese foram várias as pessoas que contribuíram de uma forma ou de outra para a sua concretização.

Em primeiro lugar, quero agradecer ao LSRE-LCM e à equipa de orientadores por me terem acolhido e me terem dado a oportunidade de fazer parte deste projeto. Em especial agradeço à Ana Mafalda e ao Nuno Graça toda orientação, motivação, paciência e amizade.

A todos os professores e colegas que fizeram parte destes últimos anos, um obrigado pelos anos de aprendizagem e de crescimento quer a nível pessoal como profissional.

À minha família, em especial aos meus pais, agradeço todo o suporte e amor que sempre foram essenciais.

A todos aqueles que me são mais próximos, um gigante obrigado. A vossa presença e apoio incondicional foi fundamental para alcançar os meus objetivos e tornar-me a pessoa que sou hoje.

A todos, obrigado.

This work was financially supported by: Project POCI-01-0145-FEDER-006984 - Associate Laboratory LSRE-LCM funded by FEDER through COMPETE2020 - Programa Operacional Competitividade e Internacionalização (POCI) - and by national funds through FCT - Fundação para a Ciência e a Tecnologia; Project Inn-INDIGO/0003/2014 funded by FCT - Fundação para a Ciência e a Tecnologia.



## Abstract

The electrocoagulation process is a method used to treat wastewater which has gained increasing attention. This technology involves the production of coagulants by the oxidation of the anode. This method presents itself as an alternative to the use of chemical coagulants for the removal of pollutants thus exhibiting an advantage for communities with a better access to electricity than this type of chemicals.

In this work, several electrocoagulation tests were carried out using aluminum electrodes to determine the influence of different operating parameters such as the electric current, the initial concentration of the pollutant, the initial water pH and the arrangement of the electrodes. Removal of iron, fluoride and arsenic (V) was tested. Experiments were performed with only one contaminant, binary mixtures and a ternary mixture. The evolution of the concentration throughout the trials was determined as well as the total suspended solids and the particle size distribution at the end of each test.

It was observed that the removal improves with increasing electrical current for all contaminants. In the case of iron, higher pH values favor its removal, while for fluoride and arsenic (V) the opposite is observed.

In binary experiments, it was found that the presence of iron improves the removal of fluoride as well as the opposite. The same happened for mixtures with iron and arsenic (V). However, although the presence of arsenic (V) improves the removal of fluoride, the removal of arsenic (V) worsens with the presence of fluoride.

In the case of the experience with the ternary mixture it was found that the removal of the various contaminants was favored by the presence of the others.

**Keywords:** Electrocoagulation (EC), waterwater treatment, electrolysis, iron, fluoride, arsenic (V)

---

## Resumo

A eletrocoagulação é um método de tratamento de efluentes que tem ganho cada vez mais atenção. Esta tecnologia envolve a oxidação do ânodo para a produção de coagulante. Este método apresenta-se como uma alternativa ao uso de coagulantes químicos para a remoção de poluentes exibindo assim uma vantagem para comunidades com um acesso mais facilitado a eletricidade do que a este tipo de químicos.

Diversos ensaios de eletrocoagulação foram realizados recorrendo a elétrodos de alumínio para determinar a influência de diferentes parâmetros como a corrente elétrica, a concentração inicial do poluente, o pH inicial da água e o arranjo dos elétrodos. Foi testada a remoção de ferro, fluoreto e arsénio(V). Foram efetuadas experiências com apenas um contaminante, misturas binárias e uma mistura ternária. A evolução da concentração ao longo das experiências foi determinada, bem como os sólidos suspensos totais e a distribuição de tamanho das partículas no fim de cada ensaio.

Foi observado que a remoção melhora com o aumento da corrente elétrica para todos os contaminantes. No caso do ferro, valores de pH mais elevados favorecem a remoção, enquanto que para o fluoreto e para o arsénio (V) se observa o oposto.

Nas experiências binárias, constatou-se que a presença de ferro melhora a remoção do fluoreto tal como o oposto. O mesmo aconteceu para mistura com ferro e arsénio (V). No entanto, embora a presença do arsénio (V) tenha melhorado a remoção do fluoreto, a remoção de arsénio (V) piorou com a presença do fluoreto.

No caso da experiência com a mistura ternária verificou-se que a remoção dos vários contaminantes foi favorecida pela presença dos outros.

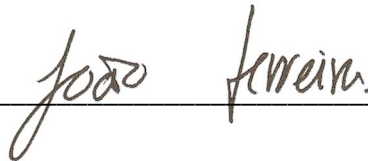
**Palavras-chave:** eletrocoagulação (EC), tratamento de efluentes, eletrólise, ferro, fluoreto, arsénio (V)

---

## Declaração

Declara, sob compromisso de honra, que este trabalho é original e que todas as contribuições não originais foram devidamente referenciadas com identificação da fonte.

Porto, 6 de fevereiro de 2017



---

*(João Miguel Ferreira de Almeida e Silva)*

---

# Table of Contents

<b>1</b>	<b>Introduction .....</b>	<b>1</b>
1.1	Motivation and Relevance .....	1
1.2	Outline .....	2
<b>2</b>	<b>State of the Art .....</b>	<b>3</b>
2.1	Electrocoagulation .....	3
2.1.1	History and current state of EC research .....	3
2.1.2	Principles of EC .....	3
2.1.2.1	Electrolytic reactions in an EC cell.....	4
2.1.2.2	Faraday's law .....	5
2.1.2.3	Energy consumption .....	5
2.1.3	Comparison of EC with conventional methods .....	5
2.1.3.1	Advantages of EC.....	5
2.1.3.2	Disadvantages of EC .....	6
2.2	Contaminants .....	7
2.2.1	Iron .....	7
2.2.2	Fluoride .....	7
2.2.3	Arsenic .....	8
<b>3</b>	<b>Materials and Methods.....</b>	<b>9</b>
3.1	Water treatment procedure .....	9
3.2	Analytical methods .....	10
3.2.1	Chemical analytics .....	10
3.2.2	Total suspended solids.....	10
3.2.3	Laser diffraction analytics .....	10
<b>4</b>	<b>Results and Discussion.....</b>	<b>11</b>
4.1	Iron.....	11
4.1.1	Influence of electric current .....	11
4.1.2	Influence of initial iron concentration .....	12
4.1.3	Influence of initial water pH.....	14
4.1.4	Influence of electrodes arrangement .....	16
4.2	Fluoride .....	18
4.2.1	Influence of the electric current.....	18
4.2.2	Influence of initial fluoride concentration .....	20
4.2.3	Influence of initial water pH.....	22
4.2.4	Influence of electrodes arrangement .....	24

<b>4.3</b>	<b>Arsenic (V)</b> .....	<b>26</b>
4.3.1	Influence of electric current .....	26
4.3.2	Influence of initial arsenic (V) concentration .....	28
4.3.3	Influence of initial water pH.....	30
4.3.4	Influence of electrodes arrangement .....	31
<b>4.4</b>	<b>Binary mixtures</b> .....	<b>33</b>
4.4.1	Influence of fluoride presence on the removal of iron .....	33
4.4.2	Influence of iron presence on the removal of fluoride .....	35
4.4.3	Influence of arsenic (V) presence on the removal of iron.....	37
4.4.4	Influence of iron presence on the removal of arsenic (V).....	39
4.4.5	Influence of arsenic (V) presence on the removal of fluoride.....	41
4.4.6	Influence of fluoride presence on the removal of arsenic (V).....	43
<b>4.5</b>	<b>Ternary mixtures</b> .....	<b>45</b>
<b>5</b>	<b>Conclusions</b> .....	<b>47</b>
<b>6</b>	<b>References</b> .....	<b>49</b>
<b>Appendix A1. Colloids theory</b> .....		<b>51</b>
A1.1	DLVO theory.....	51
<b>Appendix A2. Chemical analytics</b> .....		<b>53</b>
<b>Appendix A3. Results obtained during the EC treatment</b> .....		<b>55</b>
<b>A3.1</b>	<b>Iron</b> .....	<b>55</b>
A3.1.1	Concentration .....	55
A3.1.1.1	Influence of electric current .....	55
A3.1.1.2	Influence of initial iron concentration .....	55
A3.1.1.3	Influence of initial water pH .....	56
A3.1.1.4	Influence of electrodes arrangements .....	56
A3.1.2	Particles size .....	57
A3.1.2.1	Influence of electric current .....	57
A3.1.2.2	Influence of initial iron concentration .....	58
A3.1.2.3	Influence of initial water pH .....	59
A3.1.2.4	Influence of electrodes arrangements .....	60
<b>A3.2</b>	<b>Fluoride</b> .....	<b>62</b>
A3.2.1	Concentration .....	62
A3.2.1.1	Influence of electric current .....	62
A3.2.1.2	Influence of initial fluoride concentration .....	62
A3.2.1.3	Influence of initial water pH .....	62
A3.2.1.4	Influence of electrodes arrangements .....	63



A3.2.2	Particles size .....	64
A3.2.2.1	Influence of electric current .....	64
A3.2.2.2	Influence of initial fluoride concentration .....	65
A3.2.2.3	Influence of initial water pH .....	66
A3.2.2.4	Influence of electrodes arrangements .....	67
<b>A3.3</b>	<b>Arsenic (V) .....</b>	<b>69</b>
A3.3.1	Concentration .....	69
A3.3.1.1	Influence of electric current .....	69
A3.3.1.2	Influence of initial arsenic (V) concentration.....	69
A3.3.1.3	Influence of initial water pH .....	69
A3.3.1.4	Influence of electrodes arrangements .....	70
A3.3.2	Particles size .....	71
A3.3.2.1	Influence of electric current .....	71
A3.3.2.2	Influence of initial arsenic (V) concentration.....	72
A3.3.2.3	Influence of initial water pH .....	73
A3.3.2.4	Influence of electrodes arrangements .....	74
<b>A3.4</b>	<b>Binary mixtures.....</b>	<b>76</b>
A3.4.1	Concentration .....	76
A3.4.1.1	Influence of fluoride presence on the removal of iron .....	76
A3.4.1.2	Influence of iron presence on the removal of fluoride .....	77
A3.4.1.3	Influence of arsenic (V) presence on the removal of iron.....	78
A3.4.1.4	Influence of iron presence on the removal of arsenic (V).....	78
A3.4.1.5	Influence of arsenic (V) presence on the removal of fluoride.....	79
A3.4.1.6	Influence of fluoride presence on the removal of arsenic (V).....	79
A3.4.2	Particles size .....	80
A3.4.2.1	Influence of fluoride presence on the removal of iron .....	80
A3.4.2.2	Influence of iron presence on the removal of fluoride .....	81
A3.4.2.3	Influence of arsenic (V) presence on the removal of iron.....	82
A3.4.2.4	Influence of iron presence on the removal of arsenic (V).....	83
A3.4.2.5	Influence of arsenic (V) presence on the removal of fluoride.....	84
A3.4.2.6	Influence of fluoride presence on the removal of arsenic (V).....	85
<b>A3.5</b>	<b>Ternary mixtures.....</b>	<b>87</b>
A3.5.1	Concentration .....	87
A3.5.2	Particles size .....	88

# List of Figures

Figure 1 - Schematic diagram of a two-electrode EC cell (M. Y. A. Mollah et al., 2004).....	4
Figure 2 - (a) Experiment set-up: (1) EC reactor, (2) multimeter, (3) DC power supply; (b) EC reactor: (4) magnetic stirrer, (5) magnetic stirrer bar, (6) aluminium electrodes.....	9
Figure 3 - Effect of electric current on the removal of iron. Operating conditions - initial iron concentration: $25 \text{ mg}\cdot\text{L}^{-1}$ , initial water pH: 6 .....	11
Figure 4 - Effect of electric current on the particles formed while removing iron through EC treatment. Operating conditions - initial iron concentration: $25 \text{ mg}\cdot\text{L}^{-1}$ , initial water pH: 6 .....	12
Figure 5 - Effect of initial iron concentration on the removal of iron. Operating conditions - initial water pH: 6, electric current: 190 mA .....	13
Figure 6 - Effect of initial iron concentration on the particles formed while removing iron through EC treatment. Operating conditions - electric current: 190 mA, initial water pH: 6 .....	14
Figure 7 - Effect of initial water pH on the removal of iron. Operating conditions- initial iron concentration: $25 \text{ mg}\cdot\text{L}^{-1}$ , electric current: 190 mA.....	15
Figure 8 - Effect of initial water pH on the particles formed while removing iron through EC treatment. Operating conditions - electric current: 190 mA, initial iron concentration: $25 \text{ mg}\cdot\text{L}^{-1}$ .....	16
Figure 9 - (a) Parallel (b) Series monopolar (c) Series bipolar (Koby, Bayramoglu, & Eyvaz, 2007) .....	17
Figure 10 - Effect of different electrode arrangements on iron removal. Operating conditions - initial iron concentration: $25 \text{ mg}\cdot\text{L}^{-1}$ , electric current: 190 mA, initial water pH: 6.....	17
Figure 11 - Effect of different electrodes arrangements on the particles formed while removing iron through EC treatment. Operating conditions - electric current: 190 mA, initial water pH: 6, initial iron concentration: $25 \text{ mg}\cdot\text{L}^{-1}$ .....	18
Figure 12 - Effect of electric current on the removal of fluoride. Operating conditions - initial fluoride concentration: $15 \text{ mg}\cdot\text{L}^{-1}$ , initial water pH: 6.....	19
Figure 13 - Effect of electric current on the particles formed while removing fluoride through EC treatment. Operating conditions - initial fluoride concentration: $15 \text{ mg}\cdot\text{L}^{-1}$ , initial water pH: 6.	20
Figure 14 - Effect of initial fluoride concentration on the removal of fluoride. Operating conditions - initial water pH: 6, electric current: 190 mA .....	21

Figure 15 - Effect of initial fluoride concentration on the particles formed while removing fluoride through EC treatment. Operating conditions - electric current: 190 mA, initial water pH: 6 .....	22
Figure 16 - Effect of initial water pH on the removal of fluoride. Operating conditions - initial fluoride concentration: 15 mg·L <sup>-1</sup> , electric current: 190 mA .....	23
Figure 17 - Effect of initial water pH on the particles formed while removing fluoride through EC treatment. Operating conditions - electric current: 190 mA, initial fluoride concentration: 15 mg·L <sup>-1</sup> .....	24
Figure 18 - Effect of different electrode arrangements on fluoride removal. Operating conditions - initial fluoride concentration: 15 mg·L <sup>-1</sup> , electric current: 190 mA, initial water pH: 6 .....	25
Figure 19 - Effect of electrodes arrangements on the particles formed while removing fluoride through EC treatment. Operating conditions - initial water pH: 6, electric current: 190 mA, initial fluoride concentration: 15 mg·L <sup>-1</sup> .....	26
Figure 20 - Effect of electric current on the removal of arsenic (V). Operating conditions - initial concentration of arsenic (V): 3 mg·L <sup>-1</sup> , initial water pH: 6. ....	27
Figure 21 - Effect of electric current on the particles formed while removing arsenic (V) through EC treatment. Operating conditions - initial arsenic (V) concentration: 3 mg·L <sup>-1</sup> , initial water pH: 6 .....	28
Figure 22 - Effect of initial arsenic (V) concentration on the removal of arsenic (V). Operating conditions - initial water pH: 6, electric current: 190 mA .....	29
Figure 23 - Effect of initial arsenic (V) concentration on the particles formed while removing arsenic (V) through EC treatment. Operating conditions - electric current: 190 mA, initial water pH: 6 .....	29
Figure 24 - Effect of initial water pH on the removal of arsenic (V). Operating conditions - initial arsenic (V) concentration: 3 mg·L <sup>-1</sup> , electric current: 190 mA .....	30
Figure 25 - Effect of initial water pH on the particles formed while removing arsenic (V) through EC treatment. Operating conditions - electric current: 190 mA, initial arsenic (V) concentration: 3 mg·L <sup>-1</sup> .....	31
Figure 26 - Effect of different electrode arrangements on arsenic (V) removal. Operating conditions - initial arsenic (V) concentration: 3 mg·L <sup>-1</sup> , electric current: 190 mA, initial water pH: 6 .....	32
Figure 27 - Effect of electrodes arrangements on the particles formed while removing arsenic (V) through EC treatment. Operating conditions - electric current: 190 mA, initial arsenic (V) concentration: 3 mg·L <sup>-1</sup> , initial water pH: 6.....	33

Figure 28 - Effect of fluoride presence on iron removal. Operating conditions - initial iron concentration: $25 \text{ mg}\cdot\text{L}^{-1}$ , electric current: 190 mA, initial water pH: 6.....	34
Figure 29 - Effect of fluoride presence on the particles produced during the EC treatment of binary mixtures (iron + fluoride). Operating conditions - initial iron concentration: $25 \text{ mg}\cdot\text{L}^{-1}$ , electric current: 190 mA, initial water pH: 6 .....	35
Figure 30 - Effect of iron presence on fluoride removal. Operating conditions - initial fluoride concentration: $15 \text{ mg}\cdot\text{L}^{-1}$ , electric current: 190 mA, initial water pH: 6 .....	36
Figure 31 - Effect of iron presence on the particles produced during the EC treatment of binary mixtures (fluoride + iron). Operating conditions - initial fluoride concentration: $15 \text{ mg}\cdot\text{L}^{-1}$ , electric current: 190 mA, initial water pH: 6 .....	37
Figure 32 - Effect of arsenic (V) presence on iron removal. Operating conditions - initial iron concentration: $25 \text{ mg}\cdot\text{L}^{-1}$ , electric current: 190 mA, initial water pH: 6.....	38
Figure 33 - Effect of arsenic (V) presence on the particles produced during the EC treatment of binary mixtures (iron + arsenic (V)). Operating conditions - initial iron concentration: $25 \text{ mg}\cdot\text{L}^{-1}$ , electric current: 190 mA, initial water pH: 6 .....	39
Figure 34 - Effect of iron presence on arsenic (V) removal. Operating conditions - initial arsenic (V) concentration: $4 \text{ mg}\cdot\text{L}^{-1}$ , electric current: 190 mA, initial water pH: 6 .....	40
Figure 35 - Effect of iron presence on the particles produced during the EC treatment of binary mixtures (arsenic (V) + iron). Operating conditions - initial arsenic (V) concentration: $4 \text{ mg}\cdot\text{L}^{-1}$ , electric current: 190 mA, initial water pH: 6 .....	41
Figure 36 - Effect of arsenic (V) presence on fluoride removal. Operating conditions - initial fluoride concentration: $15 \text{ mg}\cdot\text{L}^{-1}$ , electric current: 190 mA, initial water pH: 7.....	42
Figure 37 - Effect of arsenic (V) presence on the particles produced during the EC treatment of binary mixtures (fluoride + arsenic (V)). Operating conditions - initial fluoride concentration: $15 \text{ mg}\cdot\text{L}^{-1}$ , electric current: 190 mA, initial water pH: 6 .....	43
Figure 38 - Effect of fluoride presence on arsenic (V) removal. Operating conditions - initial arsenic (V) concentration: $4 \text{ mg}\cdot\text{L}^{-1}$ , electric current: 190 mA, initial water pH: 7.....	44
Figure 39 - Effect of fluoride presence on the particles produced during the EC treatment of binary mixtures (arsenic (V) + fluoride). Operating conditions - initial iron concentration: $25 \text{ mg}\cdot\text{L}^{-1}$ , electric current: 190 mA, initial water pH: 6 .....	44
Figure 40 - Removal of contaminants on a ternary mixture (iron + fluoride + arsenic (V)). Operating conditions - initial water pH: 6, electric current: 190 mA, initial iron concentration: $20 \text{ mg}\cdot\text{L}^{-1}$ , initial fluoride concentration: $10 \text{ mg}\cdot\text{L}^{-1}$ , initial arsenic (V) concentration: $3 \text{ mg}\cdot\text{L}^{-1}$ .....	46

Figure 41 - Particles produced on a ternary mixture (iron + fluoride + arsenic (V)). Operating conditions - initial water pH: 6, electric current: 190 mA, initial iron concentration: 20 mg·L<sup>-1</sup>, initial fluoride concentration: 10 mg·L<sup>-1</sup>, initial arsenic (V) concentration: 3 mg·L<sup>-1</sup> ..... 46

Figure A1.1 - Structure of the electrical double layer (Pritchard et al., 2010) ..... 51

# List of Tables

Table 1 - Operating conditions of the experiments with different electric currents .....	11
Table 2 - Aluminium dissolved and energy consumed with different electric currents.....	12
Table 3 - Operating conditions for the experiments with different initial iron concentrations ...	13
Table 4 - Aluminium dissolved and energy consumed with different initial iron concentrations .	14
Table 5 - Operating conditions for the experiments with different pH values.....	15
Table 6 - Aluminium dissolved and energy consumed with different initial pH values .....	15
Table 7 - Operating conditions for the experiments with different electrode arrangements .....	17
Table 8 - Aluminium dissolved and energy consumed with different electrode arrangements....	18
Table 9 - Operating conditions of the experiments with different electric currents .....	19
Table 10 - Aluminium dissolved and energy consumed with different electric currents .....	19
Table 11 - Operating conditions for the experiments with different initial fluoride concentrations .....	20
Table 12 - Aluminium dissolved and energy consumed with different initial fluoride concentrations .....	21
Table 13 - Operating conditions for the experiments with different initial pH values .....	22
Table 14 - Aluminium dissolved and energy consumed with different initial pH values.....	23
Table 15 - Operating conditions for the experiments with different electrode arrangements....	24
Table 16 - Aluminium dissolved and energy consumed with different electrode arrangements ..	25
Table 17 - Operating conditions of the experiments with different electric currents .....	26
Table 18 - Aluminium dissolved and energy consumed with different electric currents .....	27
Table 19 - Operating conditions of the experiments with different arsenic (V) concentrations ..	28
Table 20 - Aluminium dissolved and energy consumed with different initial arsenic (V) concentrations .....	29
Table 21 - Operating conditions for the experiments with different initial pH values .....	30
Table 22 - Aluminium dissolved and energy consumed with different initial water pH .....	31
Table 23 - Operating conditions for the experiments with different electrode arrangements ....	32
Table 24 - Aluminium dissolved and energy consumed with different electrode arrangements ..	33

Table 25 - Operating conditions of the experiments with a binary mixture (iron + fluoride) .....	34
Table 26 - Aluminium dissolved and energy consumed with a binary mixture (iron + fluoride)...	35
Table 27 - Operating conditions of the experiments with a binary mixture (fluoride + iron) .....	36
Table 28 - Aluminium dissolved and energy consumed with a binary mixture (fluoride + iron)...	37
Table 29 - Operating conditions of the experiments with a binary mixture (iron + arsenic (V)) ..	38
Table 30 - Aluminium dissolved and energy consumed with a binary mixture (iron + arsenic (V))	38
Table 31 - Operating conditions of the experiments with a binary mixture (arsenic (V) + iron) ..	39
Table 32 - Aluminium dissolved and energy consumed with a binary mixture (arsenic (V) + iron)	40
Table 33 - Operating conditions of the experiments with a binary mixture (fluoride + arsenic (V)) .....	41
Table 34 - Aluminium dissolved and energy consumed with a binary mixture (fluoride + arsenic (V)) .....	42
Table 35 - Operating conditions of the experiments with a binary mixture (arsenic (V) + fluoride) .....	43
Table 36 - Aluminium dissolved and energy consumed with a binary mixture (arsenic (V) + fluoride) .....	44
Table 37 - Operating conditions of the experiments with a ternary mixture (iron + fluoride + arsenic (V)) .....	45
Table A2.1 - Spectroquant Prove 300 technical characteristics .....	53
Table A2.2 - Technical characteristics of the methods used to determine the contaminants concentrations .....	53
Table A3.1 - Results obtained for the iron concentration over time with different electric currents .....	55
Table A3.2 - Results obtained for the iron concentration over time with different initial iron concentrations .....	55
Table A3.3 - Results obtained for the iron concentration over time with different initial water pH .....	56
Table A3.4 - Results obtained for the iron concentration over time with different electrodes arrangements.....	56
Table A3.5 - Results obtained for the distribution of particle sizes with different electric currents .....	57

Table A3.6 - Results obtained for the distribution of particle sizes with different initial iron concentrations .....	58
Table A3.7 - Results obtained for the distribution of particle sizes with different initial water pH .....	59
Table A3.8 - Results obtained for the distribution of particle sizes with different electrodes arrangements.....	60
Table A3.9 - Results obtained for the fluoride concentration over time with different electric currents .....	62
Table A3.10 - Results obtained for the fluoride concentration over time with different initial fluoride concentrations .....	62
Table A3.11 - Results obtained for the fluoride concentration over time with different initial water pH .....	62
Table A3.12 - Results obtained for the fluoride concentration over time with different electrodes arrangements.....	63
Table A3.13 - Results obtained for the distribution of particle sizes with different electric currents .....	64
Table A3.14 - Results obtained for the distribution of particle sizes with different initial fluoride concentrations .....	65
Table A3.15 - Results obtained for the distribution of particle sizes with different initial water pH .....	66
Table A3.16 - Results obtained for the distribution of particle sizes with different electrodes arrangements.....	67
Table A3.17 - Results obtained for the arsenic (V) concentration over time with different electric currents .....	69
Table A3.18 - Results obtained for the arsenic (V) concentration over time with different initial arsenic (V) concentrations .....	69
Table A3.19 - Results obtained for the arsenic (V) concentration over time with different initial water pH .....	69
Table A3.20 - Results obtained for the arsenic (V) concentration over time with different electrodes arrangements .....	70
Table A3.21 - Results obtained for the distribution of particle sizes with different electric currents .....	71



Table A3.22 - Results obtained for the distribution of particle sizes with different initial arsenic (V) concentrations .....	72
Table A3.23 - Results obtained for the distribution of particle sizes with different initial water pH .....	73
Table A3.24 - Results obtained for the distribution of particle sizes with different electrodes arrangements.....	74
Table A3.25 - Results obtained for the iron concentration over time with different initial fluoride concentrations .....	76
Table A3.26 - Results obtained for the fluoride concentration over time with different initial iron concentrations .....	77
Table A3.27 - Results obtained for the iron concentration over time with different initial arsenic (V) concentrations .....	78
Table A3.28 - Results obtained for the arsenic (V) concentration over time with different initial iron concentrations .....	78
Table A3.29 - Results obtained for the fluoride concentration over time with different initial arsenic (V) concentrations .....	79
Table A3.30 - Results obtained for the arsenic (V) concentration over time with different initial fluoride concentrations .....	79
Table A3.31 - Results obtained for the distribution of particle sizes with a binary mixture (iron + fluoride) .....	80
Table A3.32 - Results obtained for the distribution of particle sizes with a binary mixture (fluoride + iron) .....	81
Table A3.33 - Results obtained for the distribution of particle sizes with a binary mixture (iron + arsenic (V)).....	82
Table A3.34 - Results obtained for the distribution of particle sizes with a binary mixture (arsenic (V) + iron) .....	83
Table A3.35 - Results obtained for the distribution of particle sizes with a binary mixture (fluoride + arsenic (V)).....	84
Table A3.36 - Results obtained for the distribution of particle sizes with a binary mixture (arsenic (V)+ fluoride) .....	85
Table A3.37 - Results obtained for the contaminants concentration over time with a ternary mixture (iron + fluoride + arsenic (V)).....	87

Table A3.38 - Results obtained for the distribution of particle sizes with a ternary mixture (iron + fluoride + arsenic (V)) ..... 88

# Notation and Glossary

## **Variables**

$V_{sample}$	Volume of sample filtered	L
$m_{metal}$	Mass of metal dissolved	g
$m_{particles}$	Mass of particles retained in the filter	mg
$F$	Faraday constant	$C \cdot mol^{-1}$
$I$	Electric current	A
$M$	Molar mass	$g \cdot mol^{-1}$
$P$	Electric power	W
$V$	Electric potential	V
$t$	Time	s
$z$	Valence number	

## **List of Acronyms**

AC	Alternating current
DC	Direct Current
DLVO	Derjagin, Landau, Verwey and Overbeek
EC	Electrocoagulation
EDL	Electrical double layer
EF	Electroflotation
TSS	Total suspended solids
USA	United States of America
USSR	Union of Soviet Socialist Republics
WHO	World Health Organization
SM	Series monopolar
SB	Series bipolar
S	Simple
P	Parallel

# 1 Introduction

## 1.1 Motivation and Relevance

There are vital factors for the existence of human life on the planet, being the access to clean water one of the most important. However, only 3% of all the water present on the planet is fresh water and the remaining fraction is salt water. Fresh water, in addition to being stored in different states, is unevenly distributed across the planet (Lohmann and Lorenz, 2000). Almost 800 million people have no access to safe drinking water and of these a big percentage live in rural areas (UNICEF, 2014).

At the same time, as billions of liters of wastewater are being produced, laws and regulations related to the environment are getting stricter. This creates an urgent need to develop new and more effective water treatment methods. This has led to an increasing interest in alternative processes such as electrocoagulation (EC), which has as advantage the fact that it is a method that does not depend on the addition of chemicals that could impact the environment. However, this method presents some down sides too such as the deposition of salts on the electrode surface which can lower the removal efficiency.

In this work, the electrocoagulation removal efficiency was tested for iron, fluoride, arsenic (V), binary and ternary mixtures. It was also an objective of this work to assess how operating parameters such as initial pH, electric current, contaminant initial concentration and electrode arrangement can affect the removal of the contaminants from the water and the particles produced during the electrocoagulation treatment.

## 1.2 Outline

This work is divided in five main parts.

In Chapter 1, the description and relevance of this work is presented.

In Chapter 2, State of the Art, a brief description of the electrocoagulation treatment and how it compares with similar separation methods is presented. A short description of each contaminant is also given in this chapter.

In Chapter 3, Materials and Methods, the experimental set-up and procedure is described. The analytical methods used for the determination of parameters such as the contaminants concentration, the total suspended solids (TSS) and the particles size distribution is also presented.

Chapter 4 reports the experimental results. First, the results obtained for each individual contaminant is presented by assessing the influence of each operating parameter on the contaminant removal by EC. Then, results for binary mixtures and a ternary mixture evaluating the effect of the present of other contaminants on the removal efficiency.

To finish, Chapter 5 presents the conclusions of this work and the suggestions for future work.

## 2 State of the Art

### 2.1 Electrocoagulation

#### 2.1.1 History and current state of EC research

The idea of treating water using electricity occurred for the first time in 1889 in the United Kingdom, and the electrocoagulation process with aluminium and iron electrodes was patented for the first time in the United States of America (USA) in 1909 (Harris, 1909). The EC was first applied on drinking water in 1946. However, due to the high investment needed and the expensive electricity supply, the EC remained unused in water and wastewater treatment until the 21<sup>st</sup> century. Nevertheless, extensive study in the USA and the former USSR accumulated a valuable amount of knowledge about this technology. In the past two decades, the increasing standard of drinking water and the more demanding environmental regulations regarding wastewater discharge allied with the decrease on the electricity price caused an increase of interest on EC (Chen, 2004; Holt et al., 2005; Mollah et al., 2001; Sillanpää, 2015, chap. 4).

Electrocoagulation combines the functions and advantages of chemical coagulation, flotation and electrochemistry in water and wastewater treatment. These technologies have decades of extensive research and development. However, the mechanism of interaction of those technologies which is present in an EC system is still not well known. Further research on the EC mechanism is needed to develop a better understanding of the technology as a whole (Holt et al., 2005).

#### 2.1.2 Principles of EC

As already mentioned, today EC is a commonly used method to treat water and wastewater. In an EC system, multiple electrochemical reactions occur simultaneously at the anodes and cathodes. At the same time, the electroflotation (EF) mechanism is promoted by the formation of gas bubbles through the cathodic reactions (Chen, 2004; Mollah et al., 2004; Sillanpää, 2015). Usually materials like aluminium, iron and stainless steel are used in the electrodes because of their availability, low price, non-toxicity and proven effectiveness (Chen, 2004; Kumar et al., 2004). However, other materials such as magnesium (Vasudevan et al., 2009a), zinc (Fajardo et al., 2015) and copper (Ali et al., 2013) were also tested. EC systems can consist of either one or multiple pairs of electrodes. In case they have multiple pairs they can be arranged in either series monopolar/bipolar or parallel (Mollah et al., 2001).

## 2.1.2.1 Electrolytic reactions in an EC cell

A simple EC reactor is made up of one anode and one cathode (Figure 1). When a potential is applied from an external power source, the anode metal undergoes oxidation while the cathode is subjected to reduction or reductive deposition of elemental metals.

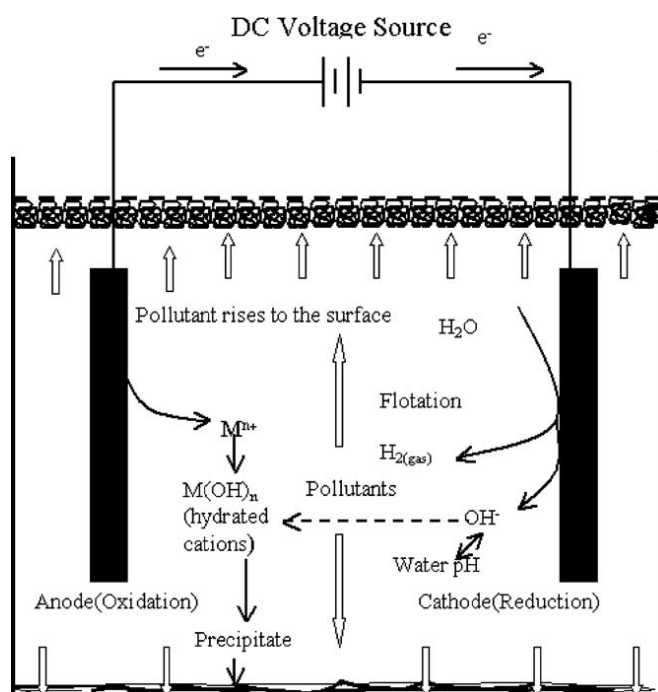
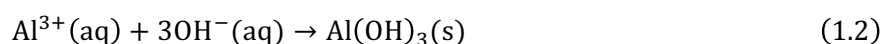


Figure 1 - Schematic diagram of a two-electrode EC cell (Mollah et al., 2004)

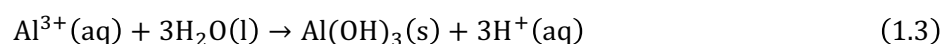
The electrocoagulation process involves the use of metallic hydroxide flocs to remove contaminants from water. These flocs are produced by electrodisolution of the metallic anode according to Equations (1.1)-(1.3) (Fajardo et al., 2014; Ghosh et al., 2008), given for the case of aluminium as it was the material used in this work.



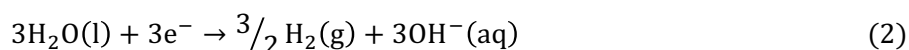
- In an alkaline solution:



- In an acidic solution:



At the aluminium cathode, hydrogen bubbles are produced according to Equation (2) (Ghosh et al., 2008).



These flocs remove the contaminants by adsorbing and precipitating dissolved contaminants (sweep floc) or by coalescing the colloidal contaminants and then removing them by electrolytic flotation. The EC process can be summarized in three steps (Mollah et al., 2001):

1. Formation of coagulants by electrolytic oxidation of the anode;
2. Destabilization of the contaminants, particulate suspension and breaking of emulsions;
3. Aggregation of destabilized phases to form flocs.

#### 2.1.2.2 Faraday's law

The theoretical mass of metal dissolved on the anode (g) can be predicted using Faraday's law (Equation (3)).

$$m_{metal} = \frac{ItM}{zF} \quad (3)$$

where  $I$  is the applied current in amperes (A),  $t$  is the treatment time in seconds (s),  $M$  is the molar mass of the anodic metal ( $\text{g}\cdot\text{mol}^{-1}$ ),  $z$  is the valence number of the ion of the substance ( $z_{Al} = 3$ ) and  $F$  is Faraday's constant ( $96485 \text{ C}\cdot\text{mol}^{-1}$ ). As Equation (3) shows, the amount of dissolved metal can be adjusted by regulating the applied current (Mollah et al., 2001; Picard et al., 2000; Sillanpää, 2015).

#### 2.1.2.3 Energy consumption

The energy consumed during the EC process can be estimated by calculating the integral of the electric power variation over time. The power can be determined according to Equation (4). Where  $P$  is the electric power in watt (W),  $V$  is the electric potential in volts (V) and  $I$  is the applied current in amperes (A)

$$P = V \cdot I \quad (4)$$

### 2.1.3 Comparison of EC with conventional methods

Other potential mechanisms for water and wastewater treatment are physical processes (e.g. filtration, screening and sedimentation/flotation), physiochemical processes (coagulation, adsorption and ion exchange), biological processes, membrane processes and disinfection methods (e.g. chlorination, ozonation or ultraviolet processes). Obviously, their efficiency depends on their application. Electrocoagulation can be seen as competition over these other methods. However, like any other technology, it has its advantages and disadvantages (Mollah et al., 2001).

#### 2.1.3.1 Advantages of EC

- EC equipment is simple and easy to operate. Furthermore, most of the problems encountered while running the process can be easily handled;
- Low start-up and operating costs;



- Wastewater treated by EC is transformed into palatable, clear, colourless and odourless water;
- Sludge formed by EC tends to settle quickly and to be easily de-watered, being mainly composed of metallic oxides/hydroxides. More importantly, it is a low sludge producing method;
- Flocs formed by EC are similar to chemical flocs. However, EC flocs are larger, contain less bounded water, are acid-resistant and more stable, being separated faster by filtration;
- Compared to chemical treatments, EC produces an effluent with less total dissolved solids (TDS). Therefore, the low TDS level contributes to a lower water recovery cost;
- The EC process has the ability to remove smallest ( $> 4 \mu\text{m}$ ) (Pritchard et al., 2010) colloidal particles since the applied electrical field set them in faster motion, facilitating the coagulation process;
- The EC process avoids the use of chemicals, so there is no problem of neutralizing excess of chemicals and no possibility of secondary pollution caused by addition of chemical substances as when chemical coagulation method is used;
- The gas bubbles produced can carry the pollutant to the top of the solution where it can be easily concentrated and removed;
- The electrolytic processes are controlled electrically with no moving parts, requiring less maintenance;
- The EC method can be used in rural areas where electricity is not available, since a solar panel unit may be sufficient to carry out the process.

#### 2.1.3.2 Disadvantages of EC

- As a result of oxidation, the 'sacrificial electrodes' are dissolved into the water streams and need to be regularly replaced;
- The use of electricity may be expensive;
- An impermeable oxide film may be formed on the cathode, leading to a loss of efficiency;
- It is required that the water to be treated has a high conductivity. Usually industrial wastewater fulfils this requirement. However, natural water and lightly polluted water does not;
- If the water to be treated contains chlorides, toxic chlorinated compounds may be formed. It is common to add sodium chloride in order to enhance the water conductivity, increasing this problem severity;

- Gelatinous hydroxide may solubilize in some cases.

## 2.2 Contaminants

### 2.2.1 Iron

Iron is the second most abundant metal on the earth's crust. As the iron ions  $\text{Fe}^{2+}$  and  $\text{Fe}^{3+}$  quickly combine with oxygen and sulphur containing compounds to form oxides, hydroxides, carbonates and sulphides, elemental iron is rarely found in nature. (WHO Recommendations, 2011).

Bad taste, discoloration, staining and high turbidity are some of the problems associated with iron. The World Health Organization (WHO) recommends that the iron contamination in drinking water should be less than  $0.3 \text{ mg}\cdot\text{L}^{-1}$  (WHO Recommendations, 2011). The European Commission directive states that the iron in water supplies should be less than  $0.2 \text{ mg}\cdot\text{L}^{-1}$  (The Council of the European Union, 1998).

Iron is an essential element in human nutrition with an estimate minimum daily requirement ranging from 10 to  $50 \text{ mg}\cdot\text{day}^{-1}$ . However, the average lethal dose of iron is  $200 - 250 \text{ mg}\cdot(\text{kg of body weight})^{-1}$ , but death has occurred following ingesting of doses as low as  $40 \text{ mg}\cdot(\text{kg of body weight})^{-1}$  (WHO Recommendations, 2011).

### 2.2.2 Fluoride

Fluoride is a high electronegative element with an extraordinary tendency to get attracted by cations like calcium. For this reason, the effect of fluoride on mineralized tissues (bone, teeth, etc.) has a clinical significance since these kinds of tissues have high amounts of calcium. Consequently, these tissues attract high amounts of fluoride that gets deposited as calcium-fluorapatite crystals. Bones are mainly composed of crystalline hydroxyapatite. Since fluorapatite is more stable than hydroxyapatite, the hydroxyl ions get substituted by fluoride ions. As the fluoride bounds in these tissues, only a small amount is excreted. This mechanism is called fluorosis.

While dental fluorosis mild form is characterized as white, opaque areas on the tooth surface, in severe form it is characterized has yellowish brown to black stains and severe pitting of the teeth. Skeletal fluorosis shows more severe symptoms that can be similar to arthritis or spondylitis. In an advanced stage, osteoporosis may be observed (Meenakshi and Maheshwari, 2006).

Both the WHO (WHO Recommendations, 2011) and the European Commission (The Council of the European Union, 1998) directives state that the maximum concentration of fluoride in drinking water should be  $1.5 \text{ mg}\cdot\text{L}^{-1}$ .

### 2.2.3 Arsenic

Arsenic is considered a carcinogen and its ingestion may affect the gastrointestinal tract, cardiac, vascular and central nervous system. It can provoke dermal lesions like hyperpigmentation and hypopigmentation (Farrell et al., 2001).

Two forms of arsenic can be found dissolved in water: inorganic arsenites (As (III)) and organic arsenates (As (V)). The inorganic form has a higher toxicity than the organic (Kingston et al., 1993). Both the WHO (WHO Recommendations, 2011) and the European Commission (The Council of the European Union, 1998) directives state that the maximum concentration of arsenic in drinking water should be  $0.01 \text{ mg}\cdot\text{L}^{-1}$ .

## 3 Materials and Methods

### 3.1 Water treatment procedure

The experiments were conducted in an acrylic reactor, using a volume of 3 L of deionized and filtered water. The electrodes were made of aluminium.

Before every experiment the electrodes were treated with a solution of hydrochloric acid (1 N) for 5 min and sanded to remove metal oxides that may have been deposited in their surface.

The water conductivity was adjusted with a sodium chloride solution ( $0.35 \text{ g}\cdot\text{mL}^{-1}$ ) and pH was adjusted with a sodium chloride solution (1 N) or with a hydrochloric acid solution (1 N), depending on the situation. To determine these conditions a phenomenal MU 6100 L (VWR) apparatus was used.

During the experiments a magnetic stirrer was used and set to 200 rpm. Samples were collected with a micropipette at the desired times and filtered. The electrodes were connected to a DC power supply and to a multimeter to control the desired current intensity.

The experiment set-up can be observed in Figure 2(a). A more detailed picture of the EC reactor can be observed in Figure 2(b). In this work the electrodes were always placed at a distance of 0.8 cm between each other.

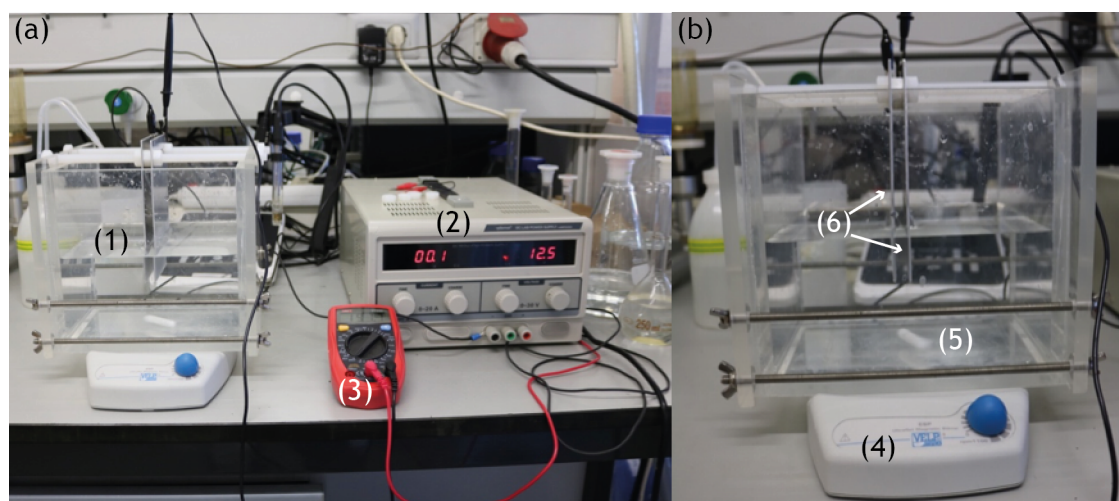


Figure 2 - (a) Experiment set-up: (1) EC reactor, (2) multimeter, (3) DC power supply; (b) EC reactor: (4) magnetic stirrer, (5) magnetic stirrer bar, (6) aluminium electrodes

## 3.2 Analytical methods

### 3.2.1 Chemical analytics

To determine the contaminants concentration a Spectroquant Prove 300 spectrophotometer (Merck Millipore) was employed. The measurements were conducted using a different Spectroquant kit for each element. Concentrations were determined by measuring the amount of light absorbed after it passed through the samples at a specific wavelength for each component.

The technical characteristics of the equipment and of the methods used to determine the contaminants concentrations can be found in Table A2.1 and Table A2.2, respectively.

### 3.2.2 Total suspended solids

To determine the total suspended solids (TSS), a 50 mL sample was vacuum filtered. The filter was then dried in an oven for 2 hours at 105°C. Every particle bigger than 2 µm got retained in the filter and this mass can be determined through the filter mass variation.

The TSS ( $\text{mg}\cdot\text{L}^{-1}$ ) can be calculated with Equation (5).

$$TSS = \frac{m_{particles}}{V_{sample}} \quad (5)$$

Where  $m_{particles}$  is the mass of particles retained in the filter (mg) and  $V_{sample}$  is the volume of the sample filtered (L).

### 3.2.3 Laser diffraction analytics

To determine the particle size distribution, laser diffraction measurements were performed using an LS 230 Laser Diffraction Particle Size Analyzer (Beckman Coulter). Particle size measurement using this equipment was based on the calculation of the angular variation in intensity of light scattered as a laser beam passed through a dispersed particulate sample. The characteristics of the equipment used are as follows:

- Range: 0.04 - 2000 µm;
- Repeatability: ≤ 1 % about mean size.
- Number of detectors: 132;
- Laser: solid state (750 nm).

## 4 Results and Discussion

### 4.1 Iron

#### 4.1.1 Influence of electric current

To determine the influence of the applied electric current ( $I$ ) on the removal of iron during the EC treatment, all experiments were conducted using an initial iron concentration of  $25 \text{ mg}\cdot\text{L}^{-1}$  and pH 6. Three different electric currents were tested (40 mA, 100 mA, 190 mA). The total anodic area was  $0.0076 \text{ m}^2$ . The operating conditions used are shown in Table 1.

Table 1 - Operating conditions of the experiments with different electric currents

I / mA	Start		End	
	pH	Conductivity / $\mu\text{S}\cdot\text{cm}^{-1}$	pH	Conductivity / $\mu\text{S}\cdot\text{cm}^{-1}$
40	6.4	363	6.0	340
100	6.1	356	5.6	308
190	6.3	345	6.3	335

The obtained results are provided in Table A3.1 and Figure 3.

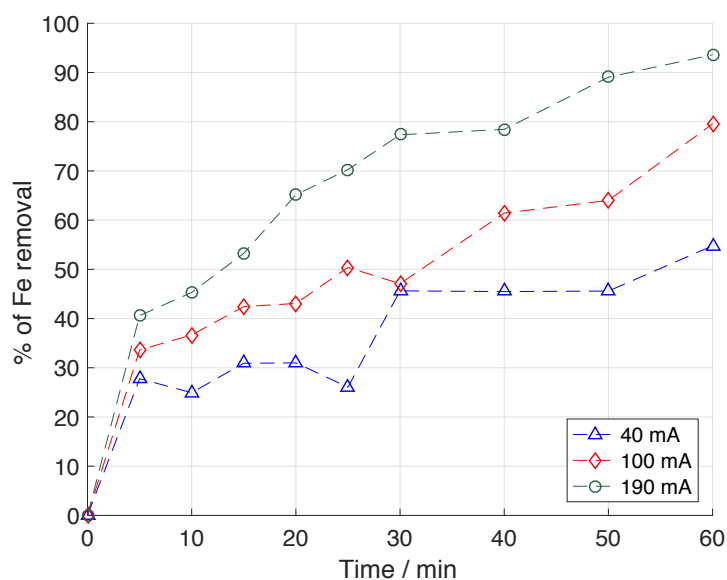


Figure 3 - Effect of electric current on the removal of iron. Operating conditions - initial iron concentration:  $25 \text{ mg}\cdot\text{L}^{-1}$ , initial water pH: 6

The electric current is one of the most important parameters in the EC process because, according to Faraday's law, the amount of aluminium dissolved into the water is proportional to the electric current. The obtained results are similar to the results reported by other authors

(Ghosh et al., 2008; Vasudevan et al., 2009a) which show that the amount of iron removed increases with the electric current.

Using Equations (3) and (4) it was possible to determine the theoretical amount of aluminium dissolved on the anode and the energy consumed on each experiment, respectively. The results are shown in Table 2.

Table 2 - Aluminium dissolved and energy consumed with different electric currents

I / mA	Aluminium dissolved / mg	Energy consumed / W·h
40	13.71	0.13
100	34.27	0.65
190	65.10	2.23

The results regarding the particle size distribution are shown in Table A3.5 and Figure 4.

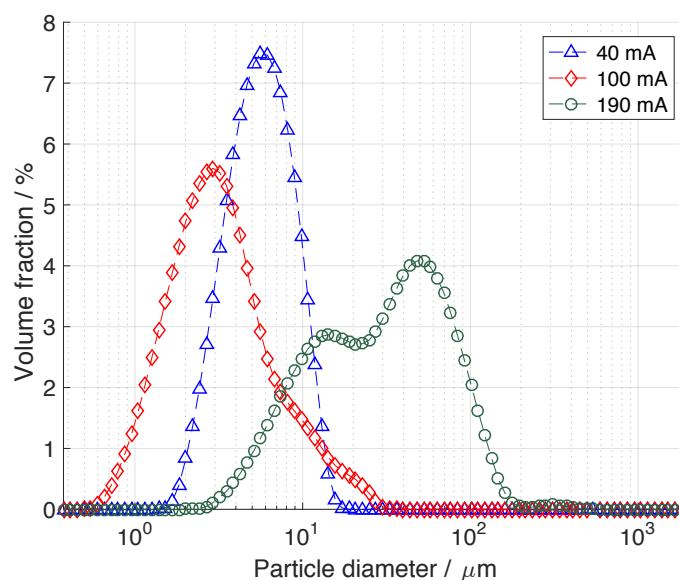


Figure 4 - Effect of electric current on the particles formed while removing iron through EC treatment. Operating conditions - initial iron concentration:  $25 \text{ mg}\cdot\text{L}^{-1}$ , initial water pH: 6

It seems that there is no direct tendency between the particles formed and the electric current applied, as the smallest particles are obtained with 100mA and the biggest with 190 mA. However, it is possible to see that higher currents achieve a wider range of diameters.

The TSS were determined at the end of each test. Values of  $23.0 \text{ mg}\cdot\text{L}^{-1}$ ,  $66.4 \text{ mg}\cdot\text{L}^{-1}$  and  $155.8 \text{ mg}\cdot\text{L}^{-1}$  were obtained respectively for the 40, 100 and 190 mA experiments.

#### 4.1.2 Influence of initial iron concentration

To determine the influence of the initial iron concentration present in the water on the removal of iron by the EC treatment, experiments were conducted using an electric current of 190 mA and

pH 6. Three different iron concentrations were tested ( $15 \text{ mg}\cdot\text{L}^{-1}$ ,  $20 \text{ mg}\cdot\text{L}^{-1}$ ,  $25 \text{ mg}\cdot\text{L}^{-1}$ ). The total anodic area was  $0.0076 \text{ m}^2$ . The operating conditions used are shown in Table 3.

The obtained results are provided in Table A3.2 and Figure 5.

Table 3 - Operating conditions for the experiments with different initial iron concentrations

[Fe] / $\text{mg}\cdot\text{L}^{-1}$	Start		End	
	pH	Conductivity / $\mu\text{S}\cdot\text{cm}^{-1}$	pH	Conductivity / $\mu\text{S}\cdot\text{cm}^{-1}$
15	6.1	351	6.2	290
20	5.9	346	6.5	284
25	6.3	345	6.3	335

The initial iron concentration has no relevant influence on the removal of iron. The only visible effect relies on the fact that for lower values of iron concentration, higher removal values are achieved faster but seem to converge after 50 min. The same behaviour was also observed by other authors (Ghosh et al., 2008).

The theoretical amount of aluminium dissolved on the anode and the energy consumed on each experiment were also calculated using Equations (3) and (4) respectively and the results are shown in Table 4.

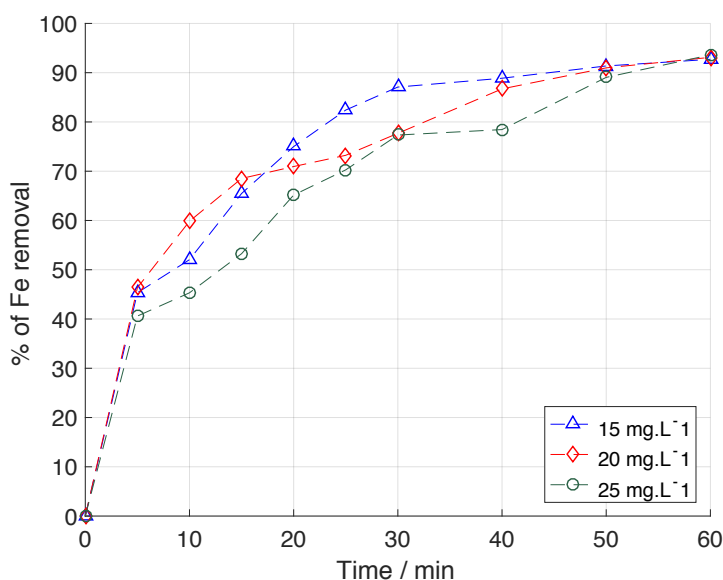


Figure 5 - Effect of initial iron concentration on the removal of iron. Operating conditions - initial water pH: 6, electric current: 190 mA

Figure 6 and Table A3.6 show the results of the particle size distribution. Besides the results for the three initial iron concentrations, the results for a blank test, e.g., the same conditions were used except for the presence of iron, are also shown.



Table 4 - Aluminium dissolved and energy consumed with different initial iron concentrations

[Fe] / mg·L <sup>-1</sup>	Aluminium dissolved / mg	Energy consumed / W·h
15	65.10	2.23
20	65.10	2.25
25	65.10	2.23

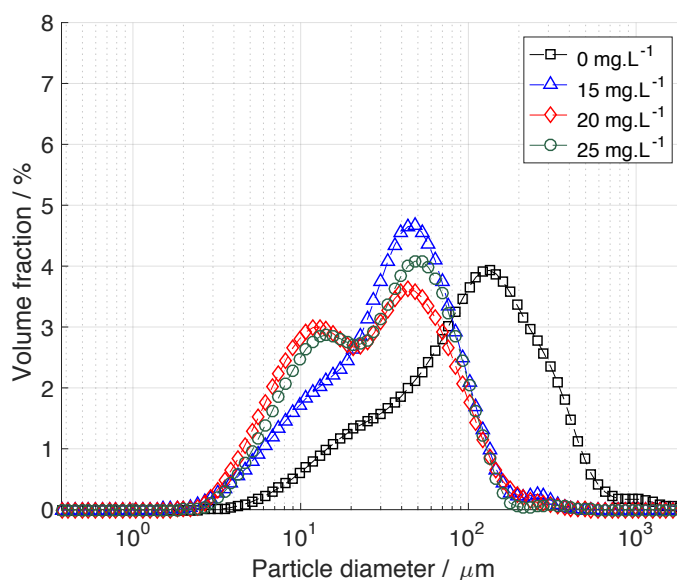


Figure 6 - Effect of initial iron concentration on the particles formed while removing iron through EC treatment.  
Operating conditions - electric current: 190 mA, initial water pH: 6

The obtained results prove that the experiments where iron was present achieve smaller particles than the one where no contaminant was present. However, the particle size does not seem to be significantly affected by the initial iron concentration.

The TSS were determined at the end of each test. Values of 25.8 mg·L<sup>-1</sup>, 128.2 mg·L<sup>-1</sup> and 155.8 mg·L<sup>-1</sup> were obtained respectively for the 15 mg·L<sup>-1</sup>, 20 mg·L<sup>-1</sup> and 25 mg·L<sup>-1</sup> experiments.

#### 4.1.3 Influence of initial water pH

To determine the influence of the initial water pH on the removal of iron by the EC treatment, experiments were conducted using an electric current of 190 mA and an initial iron concentration of 25 mg·L<sup>-1</sup>. Three different pH values were tested (3, 6, 10). The total anodic area was 0.0076 m<sup>2</sup>. The operating conditions used are shown in Table 5.

The obtained results are provided in Table A3.3 and Figure 7.

The removal of iron increases with the increase of the water initial pH. However, when adjusting the pH for the pH 10 experiment, it was observed that an orange precipitate formed as soon as the NaOH was added and that the pH did not begin to increase immediately. This happens because the ferrous ion hydrolyses to produce an array of mononuclear species Fe(OH)<sup>+</sup> to Fe(OH)<sub>4</sub><sup>-2</sup>

(Ghosh et al., 2008). For this reason, in the experiment with the alkaline solution the pH and TSS were also measured during the experiment.

The obtained results for the acidic and the neutral experiments show similarities to another published work (Vasudevan et al., 2009a).

Table 5 - Operating conditions for the experiments with different pH values

Initial pH	Start		End	
	pH	Conductivity / $\mu\text{S}\cdot\text{cm}^{-1}$	pH	Conductivity / $\mu\text{S}\cdot\text{cm}^{-1}$
3	3.3	354	4.8	198
6	6.3	345	6.3	335
10	10.1	367	8.9	295

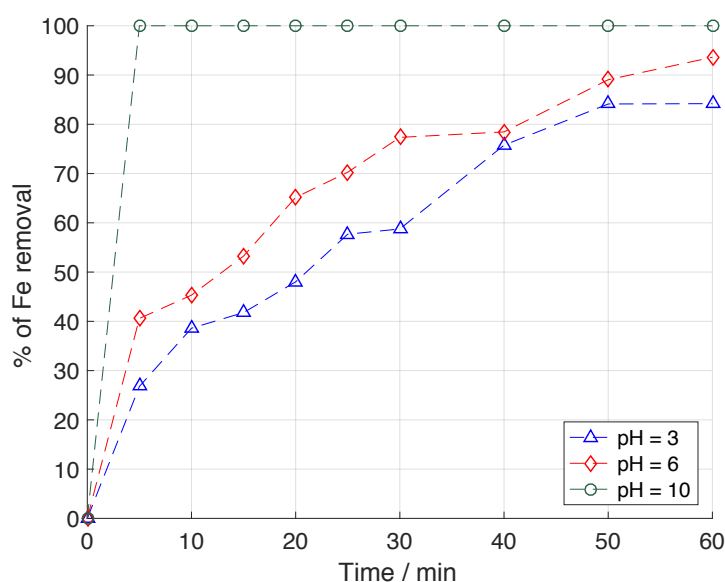


Figure 7 - Effect of initial water pH on the removal of iron. Operating conditions- initial iron concentration:  $25\text{ mg}\cdot\text{L}^{-1}$ , electric current: 190 mA

Using Equations (3) and (4) it was possible to determine the theoretical amount of aluminium dissolved on the anode and the energy consumed on each experiment, respectively. The results are shown in Table 6.

Table 6 - Aluminium dissolved and energy consumed with different initial pH values

Initial pH	Aluminium dissolved / mg	Energy consumed / W·h
3	65.10	3.14
6	65.10	2.23
10	65.10	2.91

The results of the particle size distribution are provided in Table A3.7 and Figure 8.

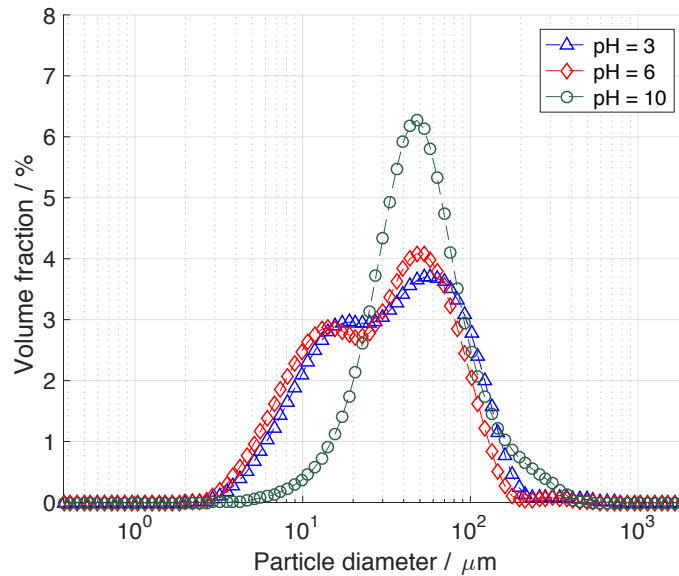


Figure 8 - Effect of initial water pH on the particles formed while removing iron through EC treatment. Operating conditions - electric current: 190 mA, initial iron concentration: 25 mg·L<sup>-1</sup>

The acidic and the neutral experiments show similar results. However, the alkaline trial shows a narrow range of particle sizes with a higher incidence on bigger particles than the other experiments. This may be due to the fact, previously reported, that the addition of NaOH created an iron precipitate.

The TSS were determined at the end of each test. Values of 15.2 mg·L<sup>-1</sup>, 66.4 mg·L<sup>-1</sup> and 92.4 mg·L<sup>-1</sup> were obtained respectively for the experiments with initial pH 3, 6 and 10.

#### 4.1.4 Influence of electrodes arrangement

To determine the influence of the different electrode arrangements on the removal of iron by the EC treatment, experiments were conducted with an electric current of 190 mA, an initial iron concentration of 25 mg·L<sup>-1</sup> and pH 6. Four different arrangements were tested: simple, series monopolar, series bipolar and parallel (Figure 9). The total anodic area for the parallel and series experiments was 0.0152 m<sup>2</sup> and the simple arrangement had a total anodic area of 0.0076 m<sup>2</sup>. The operating conditions used are shown in Table 7.

The obtained results are provided in Table A3.4 and Figure 10. The number of electrodes and their arrangements had no significant influence on the removal of iron from the water. The fact that the same electric current is applied in all tests, and therefore the same amount of aluminium is dissolved may justify these results.

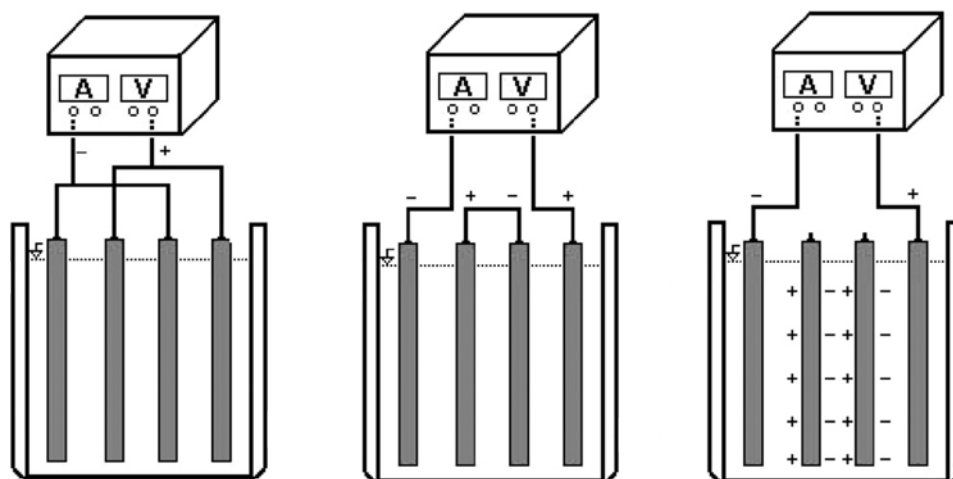


Figure 9 - (a) Parallel (b) Series monopolar (c) Series bipolar (Adaptado de Kobya, Bayramoglu, & Eyvaz, 2007)

Table 7 - Operating conditions for the experiments with different electrode arrangements

Arrangement	Start		End	
	pH	Conductivity / $\mu\text{S}\cdot\text{cm}^{-1}$	pH	Conductivity / $\mu\text{S}\cdot\text{cm}^{-1}$
Simple	6.3	345	6.3	335
Series monopolar	5.8	405	6.0	303
Series bipolar	6.2	352	6.9	303
Parallel	5.7	407	6.7	332

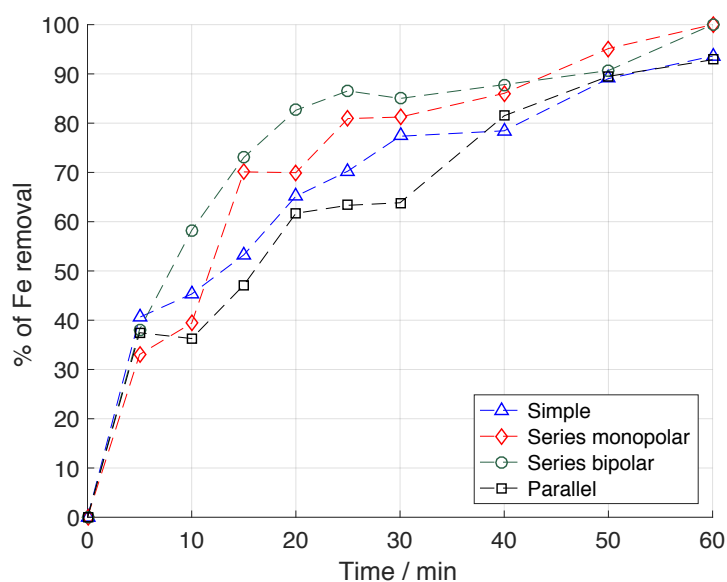


Figure 10 - Effect of different electrode arrangements on iron removal. Operating conditions - initial iron concentration:  $25\text{ mg}\cdot\text{L}^{-1}$ , electric current:  $190\text{ mA}$ , initial water pH: 6

Again, the theoretical amount of aluminium dissolved on the anode and the energy consumed on each experiment were calculated, and the results are shown in Table 8.

Table 8 - Aluminium dissolved and energy consumed with different electrode arrangements

Arrangement	Aluminium dissolved / mg	Energy consumed / W.h
Simple	65.10	2.23
Series monopolar	65.10	3.67
Series bipolar	65.10	3.38
Parallel	65.10	3.51

The results of the particle size distribution are given in Table A3.8 and Figure 11.

Both the simple and the series bipolar experiments achieved similar results. All the experiments present a wide range of particle sizes, however the parallel experiment stood out for its smaller particles and the series monopolar for its bigger particles.

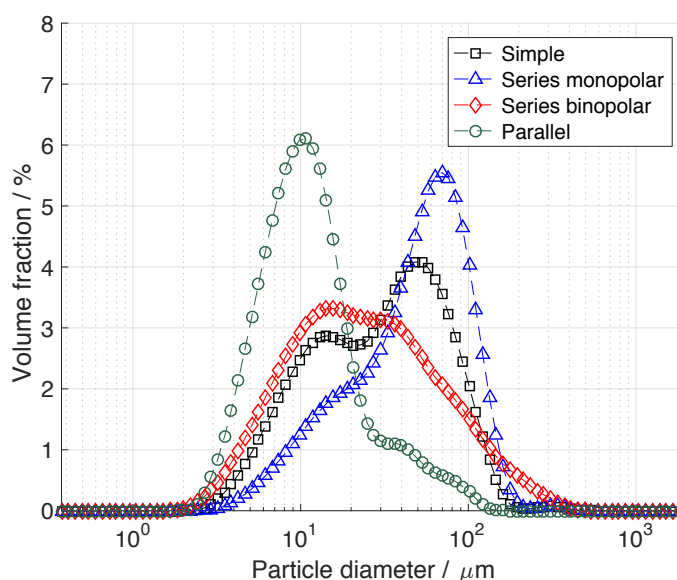


Figure 11 - Effect of different electrodes arrangements on the particles formed while removing iron through EC treatment. Operating conditions - electric current: 190 mA, initial water pH: 6, initial iron concentration: 25 mg·L<sup>-1</sup>

The TSS were determined at the end of each test. Values of 155.8 mg·L<sup>-1</sup>, 104.8 mg·L<sup>-1</sup>, 95.0 mg·L<sup>-1</sup> and 168.0 mg·L<sup>-1</sup> were obtained respectively for the simple, series monopolar, series bipolar and parallel experiments.

## 4.2 Fluoride

### 4.2.1 Influence of the electric current

The influence of the applied electric current ( $I$ ) on the removal of fluoride by the EC treatment was also assessed. All experiments were conducted using an initial fluoride concentration of 15 mg·L<sup>-1</sup> and pH 6. Three different electric currents were tested (40 mA, 100 mA, 190 mA). The total anodic area was 0.0076 m<sup>2</sup>. The operating conditions used are shown in Table 9.

The obtained results are provided in Table A3.9 and Figure 12.

Table 9 - Operating conditions of the experiments with different electric currents

I / mA	Start		End	
	pH	Conductivity / $\mu\text{S}\cdot\text{cm}^{-1}$	pH	Conductivity / $\mu\text{S}\cdot\text{cm}^{-1}$
40	6.3	376	8.6	367
100	6.3	374	8.5	369
190	6.6	370	8.7	361

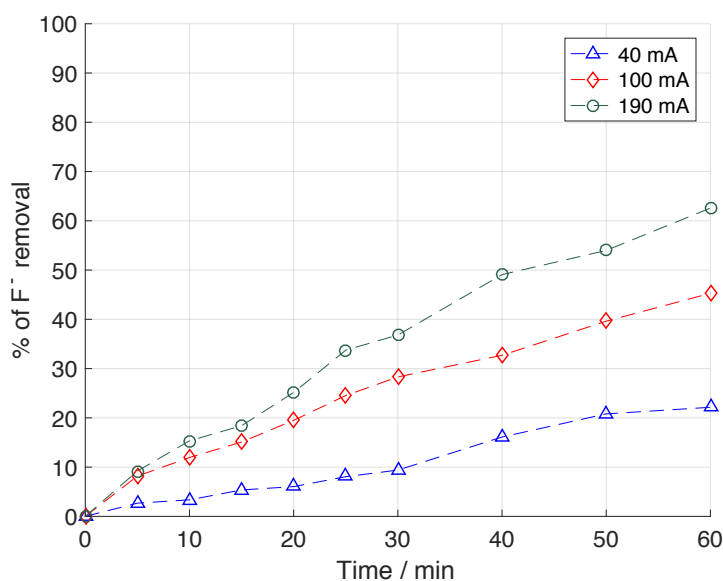


Figure 12 - Effect of electric current on the removal of fluoride. Operating conditions - initial fluoride concentration:  $15\text{ mg}\cdot\text{L}^{-1}$ , initial water pH: 6

The obtained results show that higher electric current attain quicker fluoride removal. These results are in accordance with what is expected since the amount of fluoride removed depends upon the  $\text{Al}^{3+}$  ions availability, which is related to the electric current and time. Other authors have also reported similar results (Emamjomeh et al., 2004; Eslami et al., 2014; Vasudevan et al., 2009b).

Using Equations (3) and (4) it was possible to determine the theoretical amount of aluminium dissolved on the anode and the energy consumed on each experiment, respectively. The results are shown in Table 10.

Table 10 - Aluminium dissolved and energy consumed with different electric currents

I / mA	Aluminium dissolved / mg	Energy consumed / W·h
40	13.71	0.13
100	34.27	0.65
190	65.10	2.23

The results of the particle size distribution are provided in Table A3.13 and Figure 13.

The experiments with 100 and 190 mA obtained similar results. However, the experiment with the lowest current produced smaller particles.

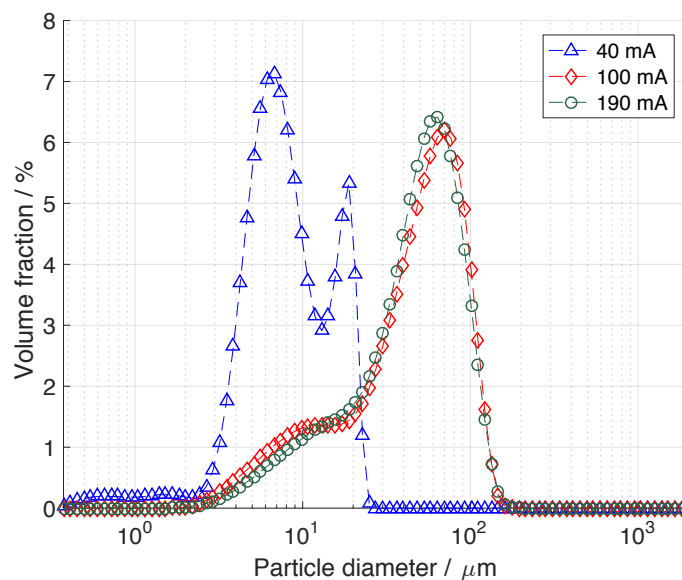


Figure 13 - Effect of electric current on the particles formed while removing fluoride through EC treatment.  
Operating conditions - initial fluoride concentration:  $15 \text{ mg}\cdot\text{L}^{-1}$ , initial water pH: 6

The TSS were determined at the end of each test. Values of  $15.4 \text{ mg}\cdot\text{L}^{-1}$ ,  $15.2$  and  $33.4 \text{ mg}\cdot\text{L}^{-1}$  were obtained respectively for the 40, 100 and 190 mA experiments.

#### 4.2.2 Influence of initial fluoride concentration

To determine the influence of the initial fluoride concentration present in the water on the removal of fluoride by the EC treatment, experiments were conducted using an electric current of 190 mA and pH 6. Three different fluoride concentrations were tested ( $5 \text{ mg}\cdot\text{L}^{-1}$ ,  $10 \text{ mg}\cdot\text{L}^{-1}$ ,  $15 \text{ mg}\cdot\text{L}^{-1}$ ). The total anodic area was  $0.0076 \text{ m}^2$ . The operating conditions used are shown in Table 11.

The obtained results can be seen in Table A3.10 and Figure 14.

Table 11 - Operating conditions for the experiments with different initial fluoride concentrations

[F <sup>-</sup> ] / $\text{mg}\cdot\text{L}^{-1}$	Start		End	
	pH	Conductivity / $\mu\text{S}\cdot\text{cm}^{-1}$	pH	Conductivity / $\mu\text{S}\cdot\text{cm}^{-1}$
5	6.6	350	8.9	345
10	6.8	357	8.7	351
15	6.3	370	8.6	361

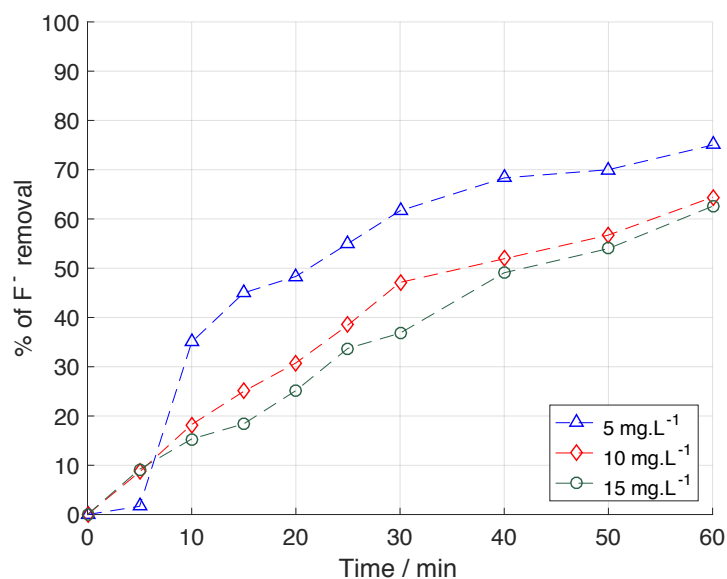


Figure 14 - Effect of initial fluoride concentration on the removal of fluoride. Operating conditions - initial water pH: 6, electric current: 190 mA

Higher concentrations of fluoride led to lower removal efficiencies. This happens because, with increasing fluoride concentration, the ratio of aqueous fluoride to available complexation sites increases. This leads to a competition for complexation sites that limits the rate of fluoride removal. Takdastan et al. (2014) and Vasudevan et al. (2009), on an Mg-Al-Zn alloy anode, observed similar results.

Using Equations (3) and (4) it was possible to determine the theoretical amount of aluminium dissolved on the anode and the energy consumed on each experiment, respectively. The results are shown in Table 12.

Table 12 - Aluminium dissolved and energy consumed with different initial fluoride concentrations

[F <sup>-</sup> ] / mg.L <sup>-1</sup>	Aluminium dissolved / mg	Energy consumed / W·h
5	65.10	2.23
10	65.10	2.25
15	65.10	2.23

The results of the particle size distribution are provided in Table A3.14 and Figure 15. Again, a blank test without fluoride was performed for comparison.

Every experiment with fluoride shows a similar curve and produced smaller particles than the trial without fluoride. However, the increasing initial fluoride concentration lead to a higher presence of bigger particles.



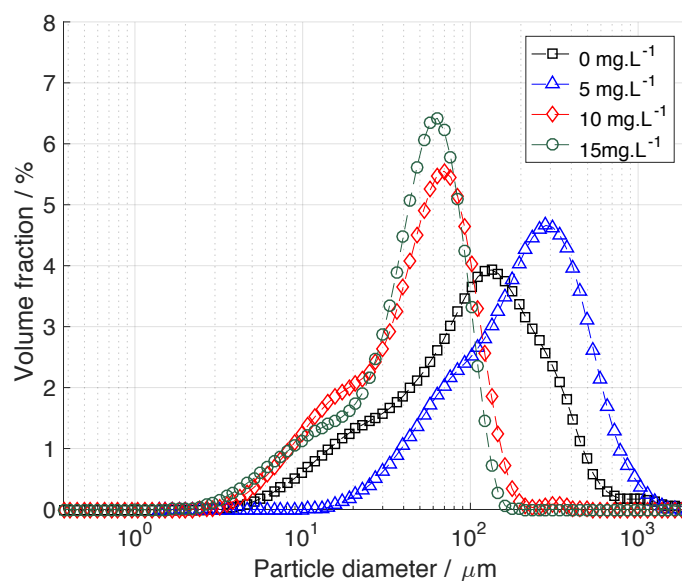


Figure 15 - Effect of initial fluoride concentration on the particles formed while removing fluoride through EC treatment. Operating conditions - electric current: 190 mA, initial water pH: 6

The TSS were determined at the end of each test. Values of  $59.4 \text{ mg}\cdot\text{L}^{-1}$ ,  $71.0 \text{ mg}\cdot\text{L}^{-1}$  and  $33.4 \text{ mg}\cdot\text{L}^{-1}$  were obtained respectively for the  $5 \text{ mg}\cdot\text{L}^{-1}$ ,  $10 \text{ mg}\cdot\text{L}^{-1}$  and  $15 \text{ mg}\cdot\text{L}^{-1}$  experiments.

#### 4.2.3 Influence of initial water pH

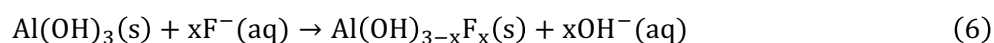
To determine the influence of the initial water pH on the removal of fluoride by the EC treatment, experiments were conducted using an electric current of 190 mA and an initial fluoride concentration of  $25 \text{ mg}\cdot\text{L}^{-1}$ . Three different pH values were tested (3, 6, 10). The total anodic area was  $0.0076 \text{ m}^2$ . The operating conditions used are shown in Table 13.

The obtained results are provided in Table A3.11 and Figure 16.

Table 13 - Operating conditions for the experiments with different initial pH values

Initial pH	Start		End	
	pH	Conductivity / $\mu\text{S}\cdot\text{cm}^{-1}$	pH	Conductivity / $\mu\text{S}\cdot\text{cm}^{-1}$
3	3.2	372	6.3	185
6	6.3	370	8.6	361
10	10.4	371	8.72	272

The obtained results show that the fluoride removal efficiency decreases with increasing pH. This happens because, when the  $\text{Al}(\text{OH})_3$  flocs adsorb the fluoride ions,  $\text{OH}^-$  is released (Equation (6) (Shen et al., 2003)).



For the experiment with an initial water pH of 10 there was a high initial  $\text{OH}^-$  concentration that may have affected the mechanism described in Equation (6) therefore decreasing the removal of fluoride in alkaline mediums.

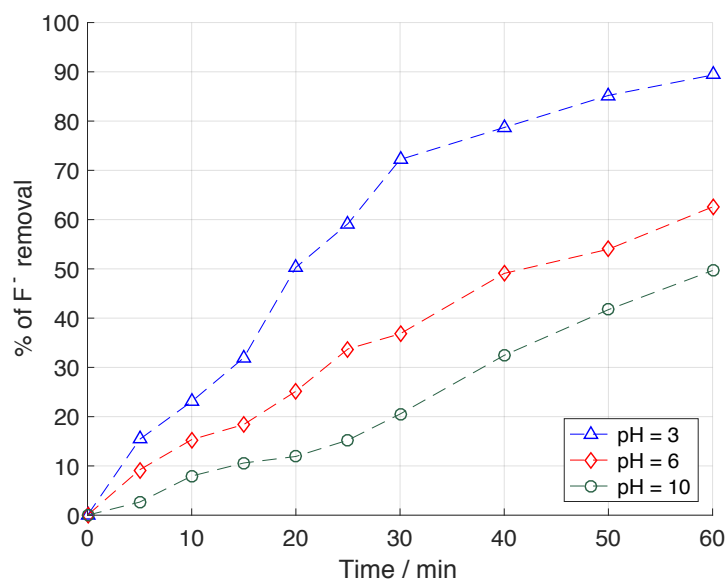


Figure 16 - Effect of initial water pH on the removal of fluoride. Operating conditions - initial fluoride concentration:  $15 \text{ mg}\cdot\text{L}^{-1}$ , electric current: 190 mA

Other authors have reported similar results in their experiments (Eslami et al., 2014; Kim et al., 2016; Vasudevan et al., 2009b).

The determined theoretical amount of aluminium dissolved on the anode and energy consumed on each experiment are shown in Table 14.

Table 14 - Aluminium dissolved and energy consumed with different initial pH values

Initial pH	Aluminium dissolved / mg	Energy consumed / W·h
3	65.10	3.14
6	65.10	2.23
10	65.10	2.91

The results of the particle size distribution are provided in Table A3.15 and Figure 17.

The initial water pH does not influence the size of the particles produced during the EC treatment. The only noticeable difference is a slightly higher volume fraction of particles with a bigger diameter.

The TSS were determined at the end of each test. Values of  $38.0 \text{ mg}\cdot\text{L}^{-1}$ ,  $33.4 \text{ mg}\cdot\text{L}^{-1}$  and  $14.4 \text{ mg}\cdot\text{L}^{-1}$  were obtained respectively for the initial pH of 3, 6 and 10.

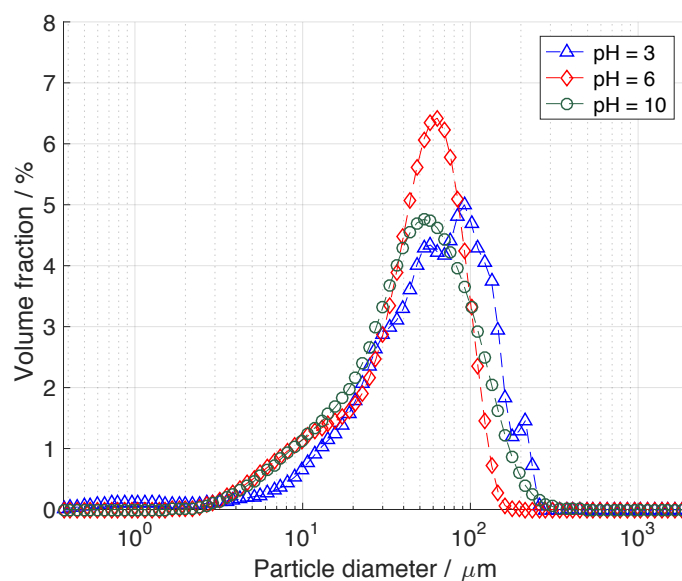


Figure 17 - Effect of initial water pH on the particles formed while removing fluoride through EC treatment.  
Operating conditions - electric current: 190 mA, initial fluoride concentration: 15 mg·L<sup>-1</sup>

#### 4.2.4 Influence of electrodes arrangement

The same four different arrangements were used to determine their influence on the removal of fluoride by EC: simple, series monopolar, series bipolar and parallel (Figure 9). The experiments were conducted with an electric current of 190 mA, an initial fluoride concentration of 15 mg·L<sup>-1</sup> and pH 6. The total anodic area for the parallel and series experiments was 0.0152 m<sup>2</sup> and the simple arrangement had a total anodic area of 0.0076 m<sup>2</sup>. The operating conditions used are shown in Table 15.

The obtained results are provided in Table A3.12 and Figure 18.

Table 15 - Operating conditions for the experiments with different electrode arrangements

Arrangement	Start		End	
	pH	Conductivity / $\mu\text{S}\cdot\text{cm}^{-1}$	pH	Conductivity / $\mu\text{S}\cdot\text{cm}^{-1}$
Simple	6.3	370	8.6	361
Series monopolar	6.6	409	8.8	400
Series bipolar	6.7	402	8.8	386
Parallel	6.7	408	8.8	403

As observed in the results with iron, once again, the number of electrodes and their arrangements had no significant influence on the removal of fluoride from the water.

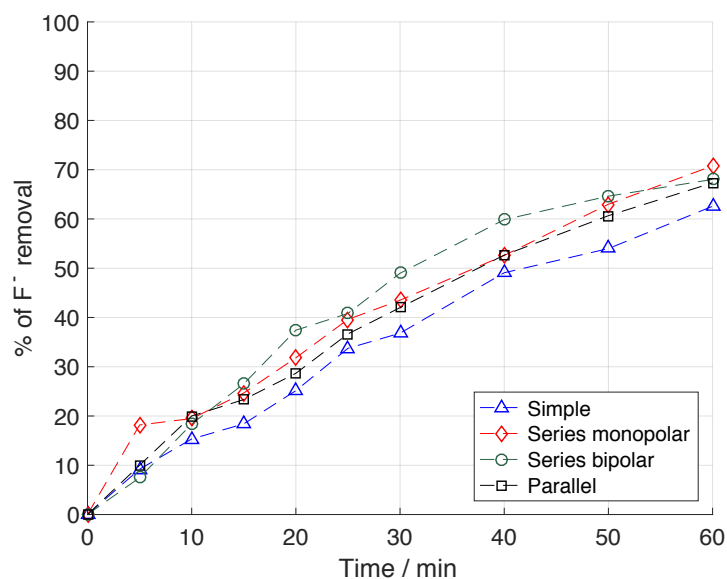


Figure 18 - Effect of different electrode arrangements on fluoride removal. Operating conditions - initial fluoride concentration:  $15 \text{ mg}\cdot\text{L}^{-1}$ , electric current: 190 mA, initial water pH: 6

Equations (3) and (4) were once more used to calculate the theoretical amount of aluminium dissolved on the anode and the energy consumed on each experiment, respectively. The results are shown in Table 16.

Table 16 - Aluminium dissolved and energy consumed with different electrode arrangements

Arrangement	Aluminium dissolved / mg	Energy consumed / W·h
Simple	65.10	2.23
Series monopolar	65.10	3.70
Series bipolar	65.10	3.51
Parallel	65.10	3.89

The results of the particle size distribution are provided in Table A3.16 and Figure 19.

The experiment with the parallel arrangement produced the smallest particles while the experiment with the simple arrangement produced the biggest particles. However, the experiments with the series arrangements produced a particle size distribution similar to the simple but with bigger volume fractions of smaller particles.

The TSS were determined at the end of each test. Values of  $33.4 \text{ mg}\cdot\text{L}^{-1}$ ,  $66.0 \text{ mg}\cdot\text{L}^{-1}$ ,  $30.0 \text{ mg}\cdot\text{L}^{-1}$  and  $38.0 \text{ mg}\cdot\text{L}^{-1}$  were obtained respectively for the simple, series monopolar, series bipolar and parallel experiments.

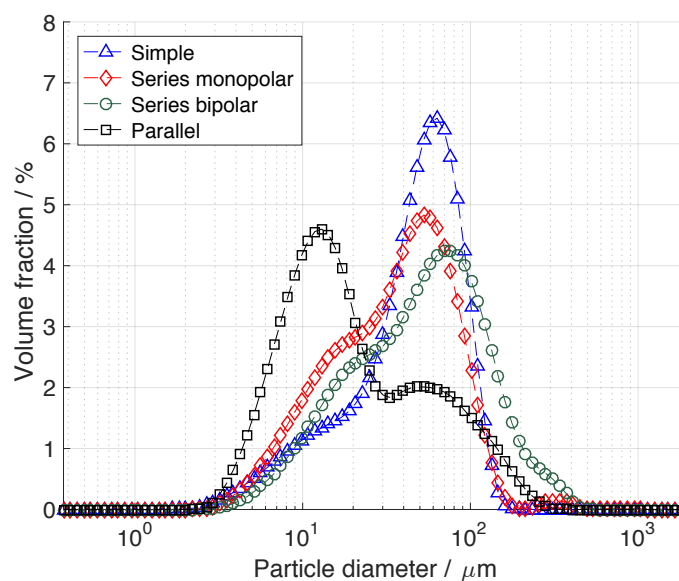


Figure 19 - Effect of electrodes arrangements on the particles formed while removing fluoride through EC treatment. Operating conditions - initial water pH: 6, electric current: 190 mA, initial fluoride concentration:  $15 \text{ mg}\cdot\text{L}^{-1}$

## 4.3 Arsenic (V)

### 4.3.1 Influence of electric current

To determine the influence of the applied electric current ( $I$ ) on the removal of arsenic (V) by EC experiments with three different electric currents were performed (40 mA, 100 mA, 190 mA). In these tests an initial arsenic (V) concentration of  $3 \text{ mg}\cdot\text{L}^{-1}$  and a pH 6 were employed. The total anodic area was  $0.0076 \text{ m}^2$ . The operating conditions used are shown in Table 17.

Table 17 - Operating conditions of the experiments with different electric currents

$I / \text{mA}$	Start		End	
	pH	Conductivity / $\mu\text{S}\cdot\text{cm}^{-1}$	pH	Conductivity / $\mu\text{S}\cdot\text{cm}^{-1}$
40	6.7	343	7.7	343
100	6.6	347	6.7	346
190	6.7	348	6.7	347

The obtained results are provided in Table A3.17 and Figure 20.

In the first 10 minutes the arsenic removal was proportional to the electric current. However, that difference faded over time and every experiment achieved total arsenic removal at around the same time. Similar results were reported by Kumar et al. (2004) in their experiments with iron electrodes.

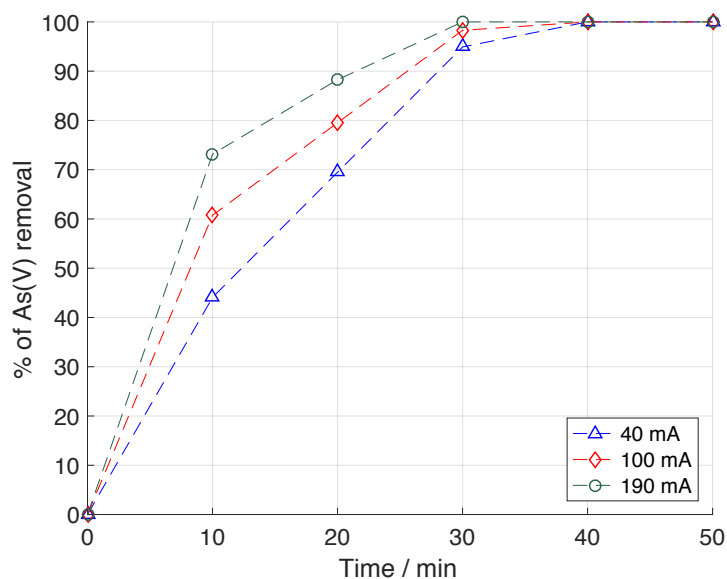


Figure 20 - Effect of electric current on the removal of arsenic (V). Operating conditions - initial concentration of arsenic (V):  $3 \text{ mg}\cdot\text{L}^{-1}$ , initial water pH: 6.

The determined theoretical amount of aluminium dissolved on the anode and energy consumed on each experiment calculated using Equations (3) and (4) are given in Table 18.

Table 18 - Aluminium dissolved and energy consumed with different electric currents

I / mA	Aluminium dissolved / mg	Energy consumed / W·h
40	11.42	0.10
100	28.55	0.56
190	54.25	1.84

The results of the particle size distribution are provided in Table A3.21 and Figure 21.

The experiment with 40 mA produced particles with low range of sizes and with a high volume fraction of small particles. While the other experiments produced a wide range of particle sizes but mostly bigger than the ones of the experiment with the lower electric current.

The TSS were determined at the end of each test. Values of  $12.8 \text{ mg}\cdot\text{L}^{-1}$ ,  $42.6 \text{ mg}\cdot\text{L}^{-1}$  and  $174.6 \text{ mg}\cdot\text{L}^{-1}$  were obtained respectively for the 40, 100 and 190 mA experiments.

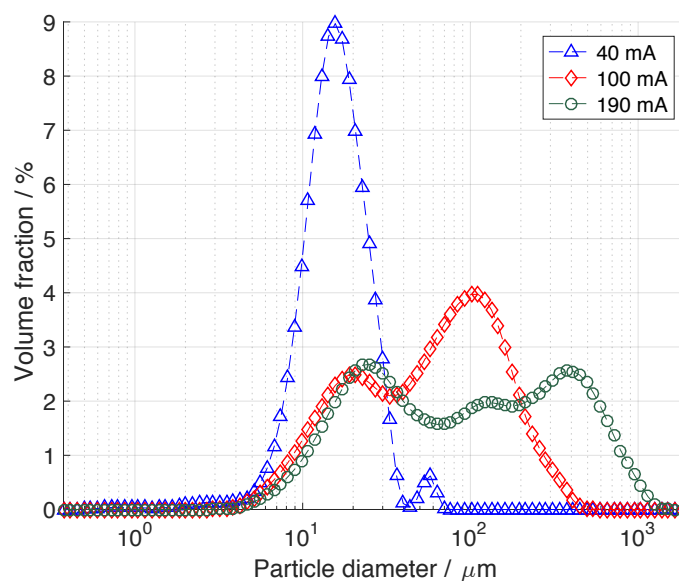


Figure 21 - Effect of electric current on the particles formed while removing arsenic (V) through EC treatment.  
Operating conditions - initial arsenic (V) concentration:  $3 \text{ mg}\cdot\text{L}^{-1}$ , initial water pH: 6

#### 4.3.2 Influence of initial arsenic (V) concentration

The influence of the initial arsenic (V) concentration on the removal of arsenic (V) by EC was determined by conducting experiments, with an electric current of 190 mA and pH 6. Three different arsenic (V) concentrations were tested ( $2 \text{ mg}\cdot\text{L}^{-1}$ ,  $3 \text{ mg}\cdot\text{L}^{-1}$ ,  $4 \text{ mg}\cdot\text{L}^{-1}$ ). The total anodic area was  $0.0076 \text{ m}^2$ . The operating conditions used are shown in Table 19.

The obtained results are provided in Table A3.18 and Figure 22.

Table 19 - Operating conditions of the experiments with different arsenic (V) concentrations

[As(V)] / $\text{mg}\cdot\text{L}^{-1}$	Start		End	
	pH	Conductivity / $\mu\text{S}\cdot\text{cm}^{-1}$	pH	Conductivity / $\mu\text{S}\cdot\text{cm}^{-1}$
2	6.7	358	6.8	358
3	6.7	348	6.7	348
4	6.6	350	7.0	347

The initial arsenic (V) concentration showed no significant effect on the removal of the pollutant since every experiment achieved total arsenic (V) removal at approximately the same time and very similar results between them.

Using Equations (3) and (4) it was possible to determine the theoretical amount of aluminium dissolved on the anode and the energy consumed on each experiment, respectively. The results are shown in Table 20.

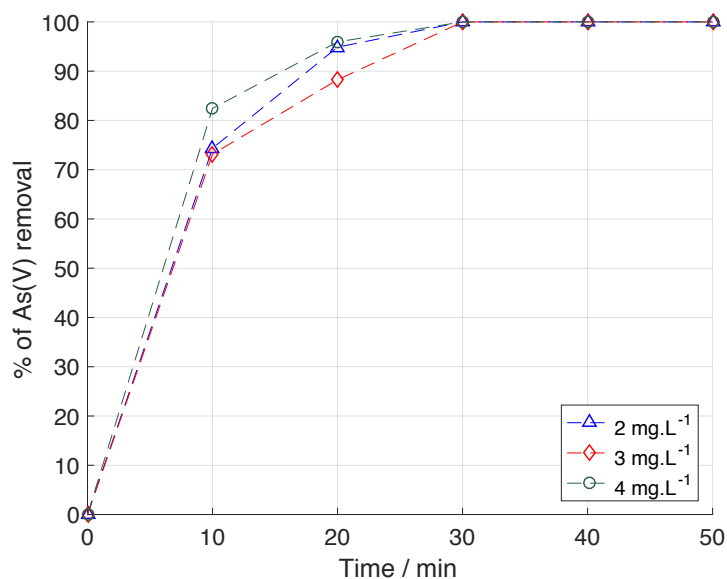


Figure 22 - Effect of initial arsenic (V) concentration on the removal of arsenic (V). Operating conditions - initial water pH: 6, electric current: 190 mA

Table 20 - Aluminium dissolved and energy consumed with different initial arsenic (V) concentrations

[As(V)] / mg·L <sup>-1</sup>	Aluminium dissolved / mg	Energy consumed / W·h
2	54.25	1.81
3	54.25	1.84
4	54.25	1.85

The results of the particle size distribution are provided in Table A3.22 and Figure 23, including the blank test.

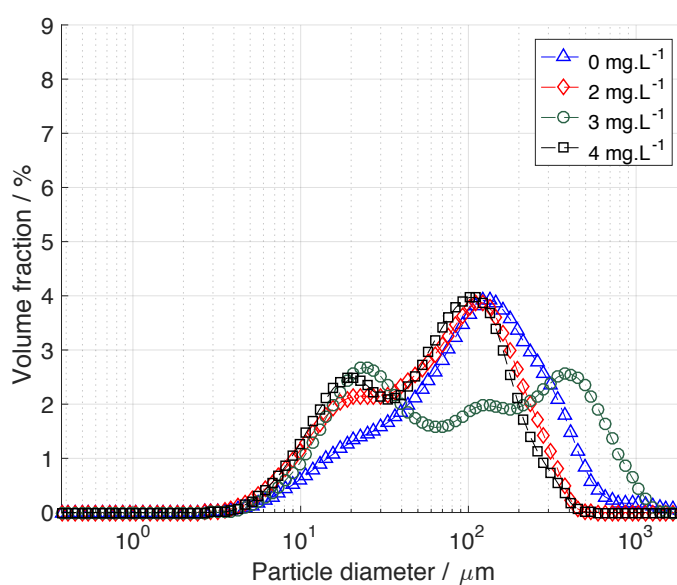


Figure 23 - Effect of initial arsenic (V) concentration on the particles formed while removing arsenic (V) through EC treatment. Operating conditions - electric current: 190 mA, initial water pH: 6



The four experiments produced similar particles with a wide range of sizes, the only prominent difference being the experiment with an initial concentration of  $2 \text{ mg}\cdot\text{L}^{-1}$  producing slightly bigger particles.

The TSS were determined at the end of each test. Values of  $79.0 \text{ mg}\cdot\text{L}^{-1}$ ,  $174.6 \text{ mg}\cdot\text{L}^{-1}$  and  $16.4 \text{ mg}\cdot\text{L}^{-1}$  were obtained respectively for the  $2 \text{ mg}\cdot\text{L}^{-1}$ ,  $3 \text{ mg}\cdot\text{L}^{-1}$  and  $4 \text{ mg}\cdot\text{L}^{-1}$  experiments.

#### 4.3.3 Influence of initial water pH

To determine the influence of the initial water pH on the removal of arsenic (V) by the EC treatment, experiments were conducted using an electric current of 190 mA and an initial arsenic (V) concentration of  $3 \text{ mg}\cdot\text{L}^{-1}$ . Three different pH values were tested (3, 6, 10). The total anodic area was  $0.0076 \text{ m}^2$ . The operating conditions used are shown in Table 21.

The obtained results are provided in Table A3.19 and Figure 24.

Table 21 - Operating conditions for the experiments with different initial pH values

Initial pH	Start		End	
	pH	Conductivity / $\mu\text{S}\cdot\text{cm}^{-1}$	pH	Conductivity / $\mu\text{S}\cdot\text{cm}^{-1}$
3	3.5	345	6.0	276
6	6.7	348	6.7	347
10	10.3	344	8.6	230

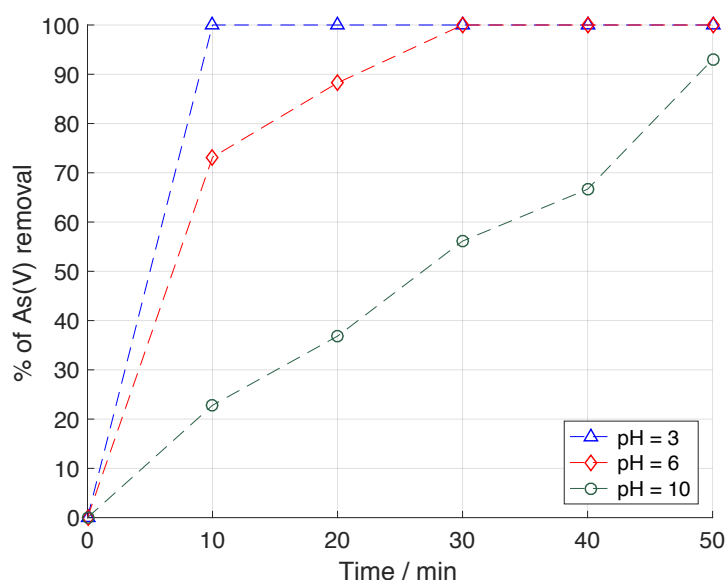


Figure 24 - Effect of initial water pH on the removal of arsenic (V). Operating conditions - initial arsenic (V) concentration:  $3 \text{ mg}\cdot\text{L}^{-1}$ , electric current: 190 mA

The arsenic (V) removal rate is slower when the initial water pH increases. Similar to the mechanism described in Equation (6), the adsorption of arsenic (V) in  $\text{Al}(\text{OH})_3$  flocs releases  $\text{OH}^-$  ions. This mechanism may be affected by the high concentration of  $\text{OH}^-$  in the alkaline medium

therefore affecting the arsenic (V) removal. Analogous results were reported by Wei Wan et al. (2011) in their work with iron electrodes. The mechanism for iron electrodes can be observed in Equation (7) (Can et al., 2012).

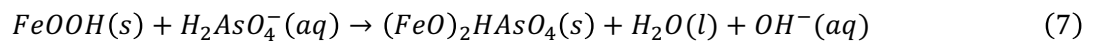


Table 22 reports the calculated theoretical amount of aluminium dissolved on the anode and the energy consumed on each experiment.

Table 22 - Aluminium dissolved and energy consumed with different initial water pH

Initial pH	Aluminium dissolved / mg	Energy consumed / W·h
3	54.25	2.30
6	54.25	1.82
10	54.25	2.48

The results of the particle size distribution are provided in Table A3.23 and Figure 25.

The experiments with initial alkaline and acidic mediums produced particles with a similar size distribution and with a lower range than the one with neutral medium.

Values of  $8.0 \text{ mg}\cdot\text{L}^{-1}$ ,  $174.6 \text{ mg}\cdot\text{L}^{-1}$  and  $15.0 \text{ mg}\cdot\text{L}^{-1}$  were obtained for the TSS respectively for the experiments with initial water pH of 3, 6 and 10.

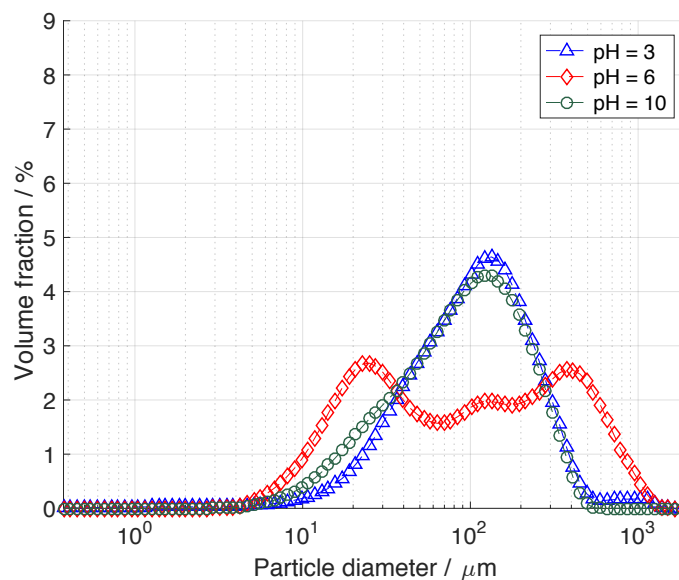


Figure 25 - Effect of initial water pH on the particles formed while removing arsenic (V) through EC treatment. Operating conditions - electric current: 190 mA, initial arsenic (V) concentration:  $3 \text{ mg}\cdot\text{L}^{-1}$

#### 4.3.4 Influence of electrodes arrangement

To determine the influence of the different electrode arrangements on the removal of arsenic (V) by the EC treatment, experiments were conducted with an electric current of 190 mA, an initial arsenic (V) concentration of  $3 \text{ mg}\cdot\text{L}^{-1}$  and pH 6. The four different arrangements presented

in Figure 9 were used (simple, series monopolar, series bipolar and parallel). The total anodic area for the parallel and series experiments was  $0.0152 \text{ m}^2$  and the simple arrangement had a total anodic area of  $0.0076 \text{ m}^2$ . The operating conditions used are shown in Table 23.

The obtained results are provided in Table A3.20 and Figure 26.

Table 23 - Operating conditions for the experiments with different electrode arrangements

Arrangement	Start		End	
	pH	Conductivity / $\mu\text{S}\cdot\text{cm}^{-1}$	pH	Conductivity / $\mu\text{S}\cdot\text{cm}^{-1}$
Simple	6.7	348	6.7	347
Series monopolar	6.7	409	7.6	406
Series bipolar	6.7	406	7.9	404
Parallel	6.6	402	7.7	400

The experiments with four electrodes achieved total arsenic (V) removal within the first 10 minutes, which is three times faster than the experiment with only two electrodes.

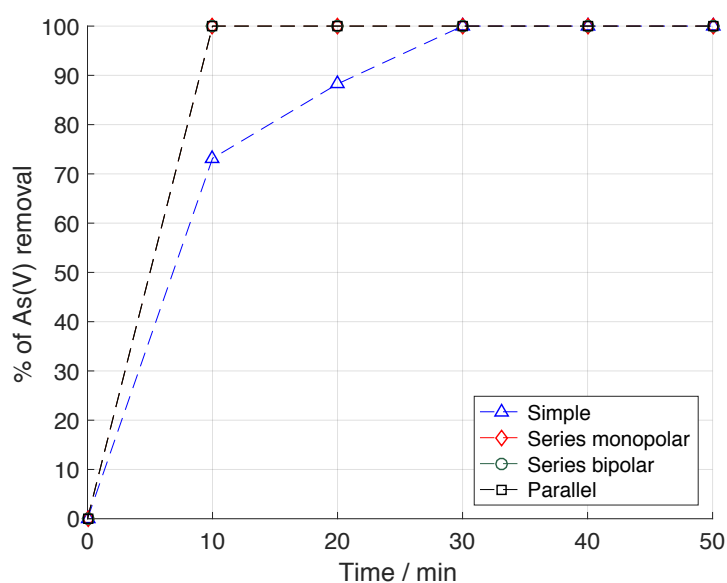


Figure 26 - Effect of different electrode arrangements on arsenic (V) removal. Operating conditions - initial arsenic (V) concentration:  $3 \text{ mg}\cdot\text{L}^{-1}$ , electric current: 190 mA, initial water pH: 6

Using Equations (3) and (4) it was possible to determine the theoretical amount of aluminium dissolved on the anode and the energy consumed on each experiment, respectively. The results are shown in Table 24.

Table 24 - Aluminium dissolved and energy consumed with different electrode arrangements

Arrangement	Aluminium dissolved / mg	Energy consumed / W·h
Simple	54.25	1.84
Series monopolar	54.25	1.54
Series bipolar	54.25	3.80
Parallel	54.25	2.52

The results of the particle size distribution are provided in Table A3.24 and Figure 27.

The results suggest that the electrode arrangements have small influence on the particles produced, as similar size distributions are obtained.

The TSS were determined at the end of each test. Values of  $174.6 \text{ mg}\cdot\text{L}^{-1}$ ,  $102.0 \text{ mg}\cdot\text{L}^{-1}$ ,  $73.8 \text{ mg}\cdot\text{L}^{-1}$  and  $67.8 \text{ mg}\cdot\text{L}^{-1}$  were obtained respectively for the simple, series monopolar, series bipolar and parallel.

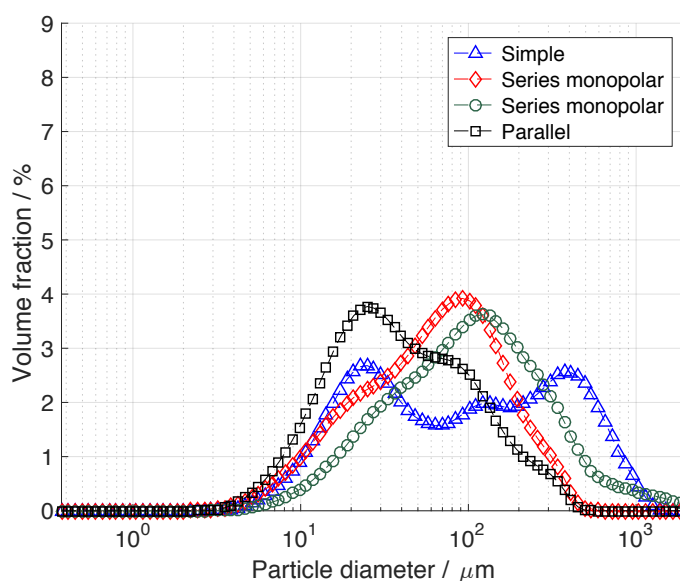


Figure 27 - Effect of electrodes arrangements on the particles formed while removing arsenic (V) through EC treatment. Operating conditions - electric current: 190 mA, initial arsenic (V) concentration:  $3 \text{ mg}\cdot\text{L}^{-1}$ , initial water pH: 6

## 4.4 Binary mixtures

### 4.4.1 Influence of fluoride presence on the removal of iron

The influence of the presence of fluoride on the removal of iron was assessed. For that experiments with an electric current of 190 mA, an initial iron concentration of  $25 \text{ mg}\cdot\text{L}^{-1}$  and pH 6 were conducted. Four different fluoride concentrations were tested ( $0 \text{ mg}\cdot\text{L}^{-1}$ ,  $5 \text{ mg}\cdot\text{L}^{-1}$ ,  $10 \text{ mg}\cdot\text{L}^{-1}$ ,  $15 \text{ mg}\cdot\text{L}^{-1}$ ). The total anodic area was  $0.0076 \text{ m}^2$ . The operating conditions used are shown in Table 25.

The results are provided in Table A3.25 and Figure 28.

Table 25 - Operating conditions of the experiments with a binary mixture (iron + fluoride)

[F <sup>-</sup> ] / mg·L <sup>-1</sup>	Start		End	
	pH	Conductivity / $\mu\text{S}\cdot\text{cm}^{-1}$	pH	Conductivity / $\mu\text{S}\cdot\text{cm}^{-1}$
0	6.3	345	6.3	335
5	6.0	344	5.1	264
10	5.9	342	6.7	257
15	6.1	349	7.2	266

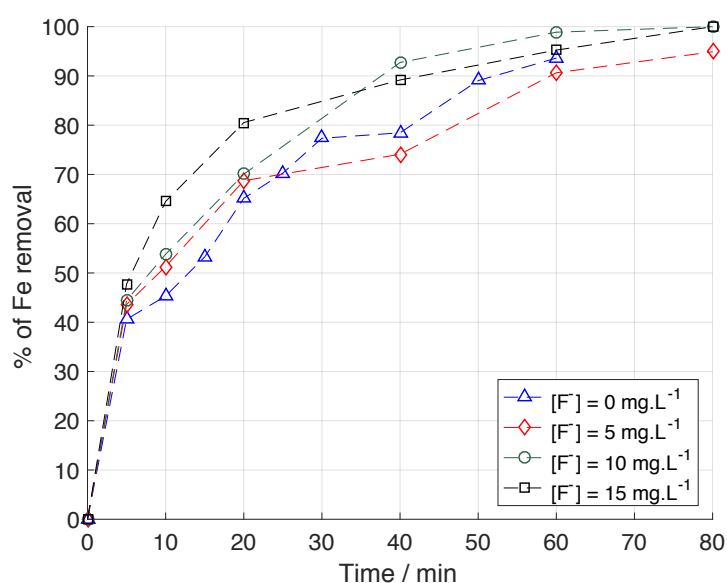


Figure 28 - Effect of fluoride presence on iron removal. Operating conditions - initial iron concentration: 25 mg·L<sup>-1</sup>, electric current: 190 mA, initial water pH: 6

The presence of fluoride causes no significant changes on the removal of iron since the experiments where it is present show similar results to the one where it is not.

Using Equations (3) and (4) it was possible to determine the theoretical amount of aluminium dissolved on the anode and the energy consumed on each experiment, respectively. The results are shown in Table 26. It should be noticed that the experiment without fluoride was conducted for 60 minutes while the other lasted 80 minutes.

Table 26 - Aluminium dissolved and energy consumed with a binary mixture (iron + fluoride)

[F <sup>-</sup> ] / mg·L <sup>-1</sup>	Aluminium dissolved / mg	Energy consumed / W·h
0	65.10	2.91
5	86.80	3.57
10	86.80	3.66
15	86.80	3.42

The results of the particle size distribution are provided in Table A3.31 and Figure 29.

The obtained results show that the presence of fluoride had no significant influence on the particles produced when compared to the experiment containing only iron.

The TSS were determined at the end of each test. Values of 155.8 mg·L<sup>-1</sup>, 98.0 mg·L<sup>-1</sup>, 138.0 mg·L<sup>-1</sup> and 141.6 mg·L<sup>-1</sup> were obtained respectively for the 0 mg·L<sup>-1</sup>, 5 mg·L<sup>-1</sup>, 10 mg·L<sup>-1</sup> and 15 mg·L<sup>-1</sup> experiments.

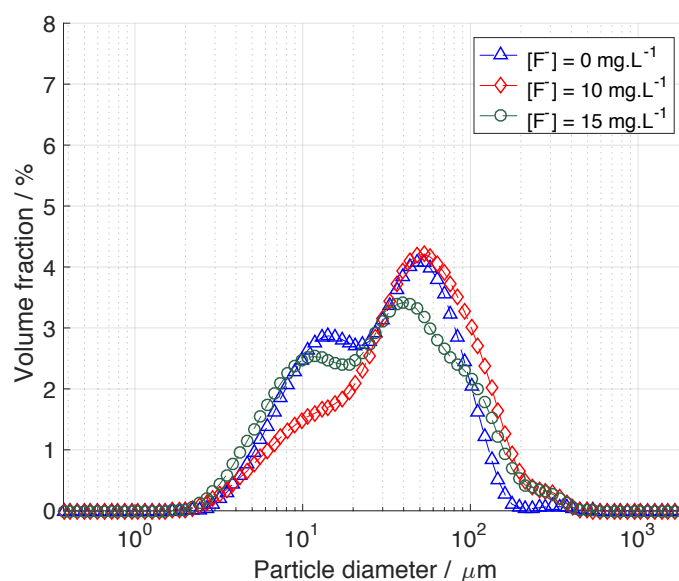


Figure 29 - Effect of fluoride presence on the particles produced during the EC treatment of binary mixtures (iron + fluoride). Operating conditions - initial iron concentration: 25 mg·L<sup>-1</sup>, electric current: 190 mA, initial water pH: 6

#### 4.4.2 Influence of iron presence on the removal of fluoride

To determine the influence of the presence of iron on the removal of fluoride experiments were conducted with an electric current of 190 mA, an initial fluoride concentration of 15 mg·L<sup>-1</sup> and pH 6. Five different iron concentrations were tested (0 mg·L<sup>-1</sup>, 5 mg·L<sup>-1</sup>, 10 mg·L<sup>-1</sup>, 15 mg·L<sup>-1</sup>, 25 mg·L<sup>-1</sup>). The total anodic area was 0.0076 m<sup>2</sup>. The operating conditions used are shown in Table 27.

Table 27 - Operating conditions of the experiments with a binary mixture (fluoride + iron)

[Fe] / mg·L <sup>-1</sup>	Start		End	
	pH	Conductivity / $\mu\text{S}\cdot\text{cm}^{-1}$	pH	Conductivity / $\mu\text{S}\cdot\text{cm}^{-1}$
0	6.3	370	8.6	361
5	5.9	342	8.1	312
10	5.9	342	8.1	314
15	6.0	344	8.0	291
25	6.1	349	7.2	266

The results are provided in Table A3.26 and Figure 30. It is observed that the removal of fluoride is not considerably affected by the presence of iron.

The theoretical amount of aluminium dissolved on the anode and the energy consumed on each experiment are reported in Table 28.

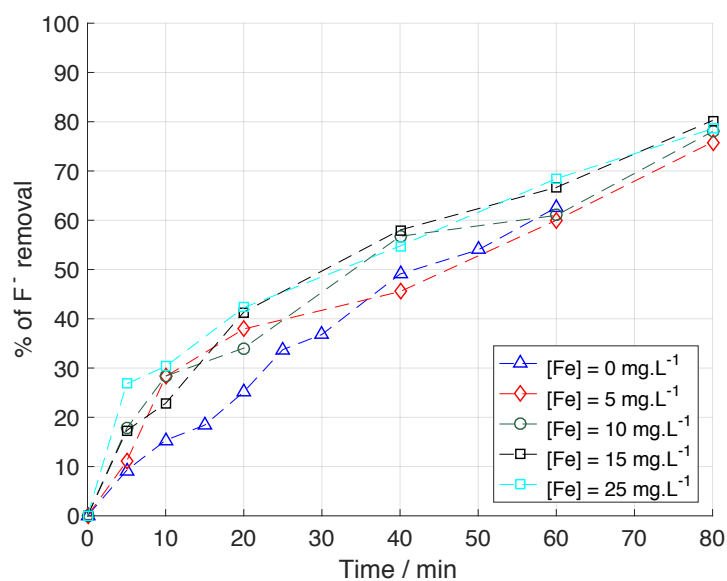


Figure 30 - Effect of iron presence on fluoride removal. Operating conditions - initial fluoride concentration: 15 mg·L<sup>-1</sup>, electric current: 190 mA, initial water pH: 6

Table 28 - Aluminium dissolved and energy consumed with a binary mixture (fluoride + iron)

[F <sup>-</sup> ] / mg·L <sup>-1</sup>	Aluminium dissolved / mg	Energy consumed / W·h
0	65.10	2.23
5	86.80	3.04
10	86.80	2.77
15	86.80	3.27
25	86.60	3.42

The results of the particle size distribution are provided in Table A3.32 and Figure 31.

The results show that the presence of iron affects the size of the particles obtained, with the presence of a wider range of smaller particle sizes. Also the shape of the obtained curve is different, showing more similarity with the results previously obtained for iron.

The TSS were determined at the end of each test. Values of 33.4 mg·L<sup>-1</sup>, 123.8 mg·L<sup>-1</sup>, 176.8 mg·L<sup>-1</sup>, 292.4 mg·L<sup>-1</sup> and 141.6 mg·L<sup>-1</sup> were obtained respectively for the 0 mg·L<sup>-1</sup>, 5 mg·L<sup>-1</sup>, 10 mg·L<sup>-1</sup>, 15 mg·L<sup>-1</sup> and 25 mg·L<sup>-1</sup> experiments.

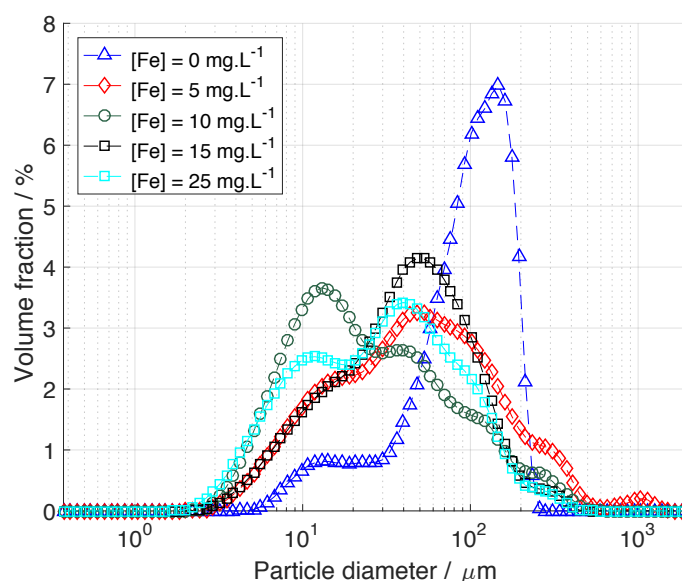


Figure 31 - Effect of iron presence on the particles produced during the EC treatment of binary mixtures (fluoride + iron). Operating conditions - initial fluoride concentration: 15 mg·L<sup>-1</sup>, electric current: 190 mA, initial water pH: 6

#### 4.4.3 Influence of arsenic (V) presence on the removal of iron

To determine the influence of the presence of arsenic (V) on the removal of iron, experiments were conducted with an electric current of 190 mA, an initial iron concentration of 25 mg·L<sup>-1</sup> and pH 6. Three different arsenic (V) concentrations were tested (0 mg·L<sup>-1</sup>, 2 mg·L<sup>-1</sup>, 4 mg·L<sup>-1</sup>). The total anodic area was 0.0076 m<sup>2</sup>. The operating conditions used are shown in Table 29.

The results are provided in Table A3.27 and Figure 32.



Table 29 - Operating conditions of the experiments with a binary mixture (iron + arsenic (V))

[As(V)] / mg·L <sup>-1</sup>	Start		End	
	pH	Conductivity / μS·cm <sup>-1</sup>	pH	Conductivity / μS·cm <sup>-1</sup>
0	6.3	345	6.3	335
2	5.8	344	4.6	302
4	6.2	344	4.9	292

The obtained results show that the presence of arsenic (V) had no major influence on the removal of iron during the EC treatment.

The results of the theoretical amount of aluminium dissolved on the anode and the energy consumed on each experiment are shown in Table 30.

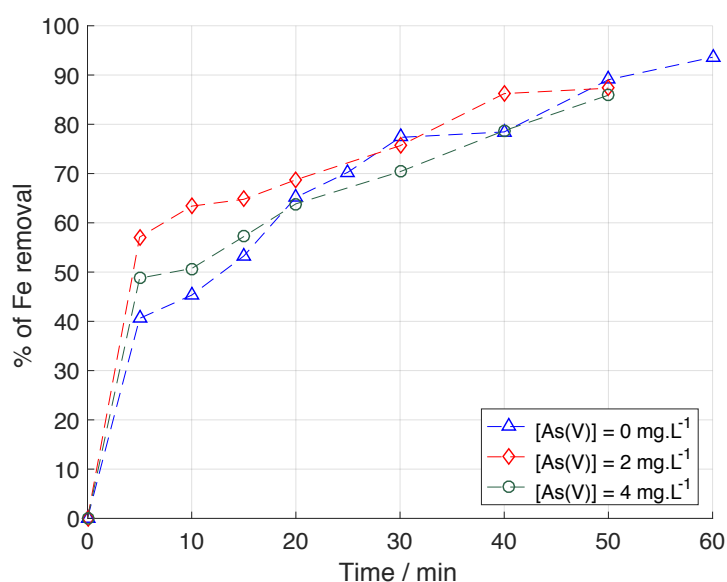


Figure 32 - Effect of arsenic (V) presence on iron removal. Operating conditions - initial iron concentration: 25 mg·L<sup>-1</sup>, electric current: 190 mA, initial water pH: 6

Table 30 - Aluminium dissolved and energy consumed with a binary mixture (iron + arsenic (V))

[As(V)] / mg·L <sup>-1</sup>	Aluminium dissolved / mg	Energy consumed / W·h
0	65.10	2.23
2	54.25	2.94
4	54.25	3.14

The results of the particle size distribution are provided in Table A3.33 and Figure 33.

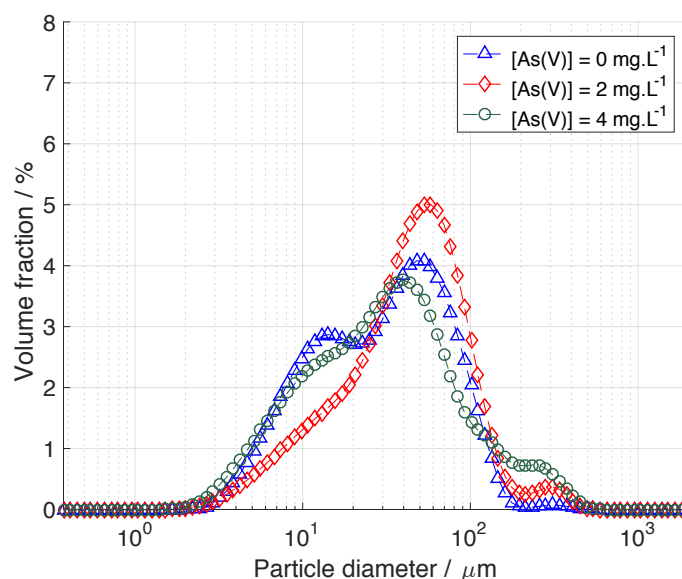


Figure 33 - Effect of arsenic (V) presence on the particles produced during the EC treatment of binary mixtures (iron + arsenic (V)). Operating conditions - initial iron concentration:  $25 \text{ mg}\cdot\text{L}^{-1}$ , electric current: 190 mA, initial water pH: 6

Similar curves were obtained showing that the presence of arsenic (V) does not affect significantly the particles produced.

TSS values of  $155.8 \text{ mg}\cdot\text{L}^{-1}$ ,  $88.4 \text{ mg}\cdot\text{L}^{-1}$  and  $53.4 \text{ mg}\cdot\text{L}^{-1}$  were obtained respectively for the  $0 \text{ mg}\cdot\text{L}^{-1}$ ,  $2 \text{ mg}\cdot\text{L}^{-1}$  and  $4 \text{ mg}\cdot\text{L}^{-1}$  experiments.

#### 4.4.4 Influence of iron presence on the removal of arsenic (V)

The presence of iron on the removal of arsenic (V) was also assessed. Experiments with an electric current of 190 mA, an initial arsenic (V) concentration of  $4 \text{ mg}\cdot\text{L}^{-1}$  and pH 6 were conducted. Three different iron concentrations were tested ( $0 \text{ mg}\cdot\text{L}^{-1}$ ,  $15 \text{ mg}\cdot\text{L}^{-1}$ ,  $25 \text{ mg}\cdot\text{L}^{-1}$ ). The total anodic area was  $0.0076 \text{ m}^2$ . The operating conditions used are shown in Table 31.

The results are provided in Table A3.28 and Figure 34.

Table 31 - Operating conditions of the experiments with a binary mixture (arsenic (V) + iron)

[Fe] / $\text{mg}\cdot\text{L}^{-1}$	Start		End	
	pH	Conductivity / $\mu\text{S}\cdot\text{cm}^{-1}$	pH	Conductivity / $\mu\text{S}\cdot\text{cm}^{-1}$
0	6.3	370	8.6	361
15	6.2	346	7.5	326
25	5.8	344	4.6	302

The obtained results show that the presence of iron allowed a faster removal of arsenic (V) achieving a total removal in less than 10 minutes, while for the experiment without iron, total arsenic (V) removal was only obtained at approximately 30 min.

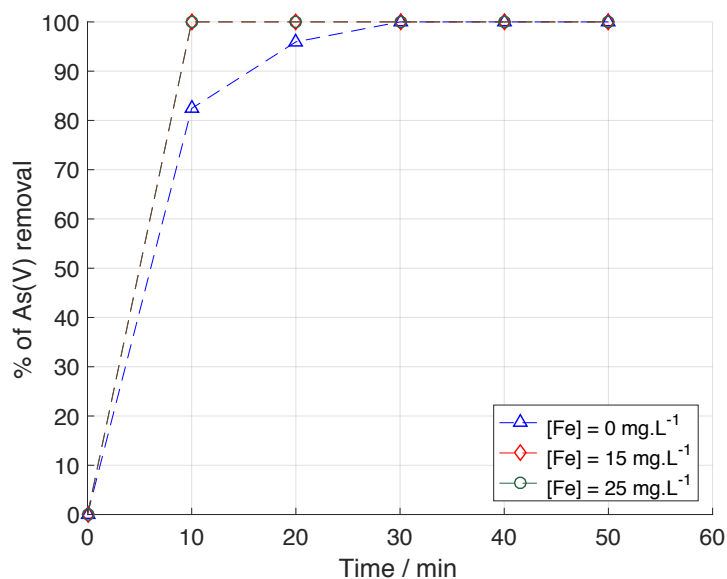


Figure 34 - Effect of iron presence on arsenic (V) removal. Operating conditions - initial arsenic (V) concentration:  $4 \text{ mg}\cdot\text{L}^{-1}$ , electric current: 190 mA, initial water pH: 6

Table 32 shows the calculated theoretical amount of aluminium dissolved on the anode and the energy consumed on each experiment.

Table 32 - Aluminium dissolved and energy consumed with a binary mixture (arsenic (V) + iron)

[Fe] / $\text{mg}\cdot\text{L}^{-1}$	Aluminium dissolved / mg	Energy consumed / W·h
0	65.10	1.84
15	54.25	2.76
25	54.25	3.14

The results of the particle size distribution are provided in Table A3.34.

The obtained results show that the size of the particles formed with and without the presence of iron are similar.

The TSS values determined at the end of each test were respectively  $16.4 \text{ mg}\cdot\text{L}^{-1}$ ,  $83.4 \text{ mg}\cdot\text{L}^{-1}$  and  $53.4 \text{ mg}\cdot\text{L}^{-1}$  for the experiments with  $0 \text{ mg}\cdot\text{L}^{-1}$ ,  $15 \text{ mg}\cdot\text{L}^{-1}$  and  $25 \text{ mg}\cdot\text{L}^{-1}$  of iron.

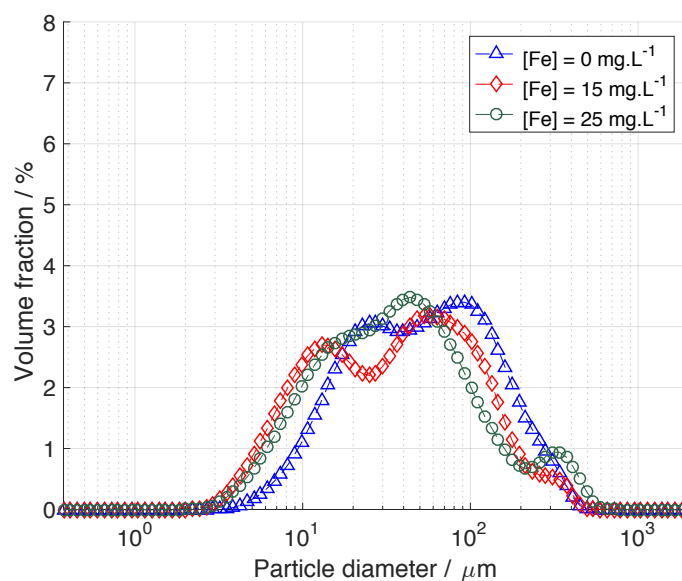


Figure 35 - Effect of iron presence on the particles produced during the EC treatment of binary mixtures (arsenic (V) + iron). Operating conditions - initial arsenic (V) concentration:  $4 \text{ mg}\cdot\text{L}^{-1}$ , electric current: 190 mA, initial water pH: 6

#### 4.4.5 Influence of arsenic (V) presence on the removal of fluoride

To determine the influence of the presence of arsenic (V) on the removal of fluoride experiments were conducted with an electric current of 190 mA, an initial fluoride concentration of  $15 \text{ mg}\cdot\text{L}^{-1}$  and pH 7. Three different arsenic (V) concentrations were tested ( $0 \text{ mg}\cdot\text{L}^{-1}$ ,  $2 \text{ mg}\cdot\text{L}^{-1}$ ,  $4 \text{ mg}\cdot\text{L}^{-1}$ ). The total anodic area was  $0.0076 \text{ m}^2$ . The operating conditions used are shown in Table 33.

The results are provided in Table A3.29 and Figure 36.

Table 33 - Operating conditions of the experiments with a binary mixture (fluoride + arsenic (V))

[As(V)] / $\text{mg}\cdot\text{L}^{-1}$	Start		End	
	pH	Conductivity / $\mu\text{S}\cdot\text{cm}^{-1}$	pH	Conductivity / $\mu\text{S}\cdot\text{cm}^{-1}$
0	6.3	370	8.6	361
2	7.2	346	8.5	341
4	7.0	344	8.4	340

The obtained results show that the presence of arsenic (V) had almost no influence on the removal of fluoride during the EC treatment.

The determined theoretical amount of aluminium dissolved on the anode and the energy consumed on each experiment are shown in Table 34.

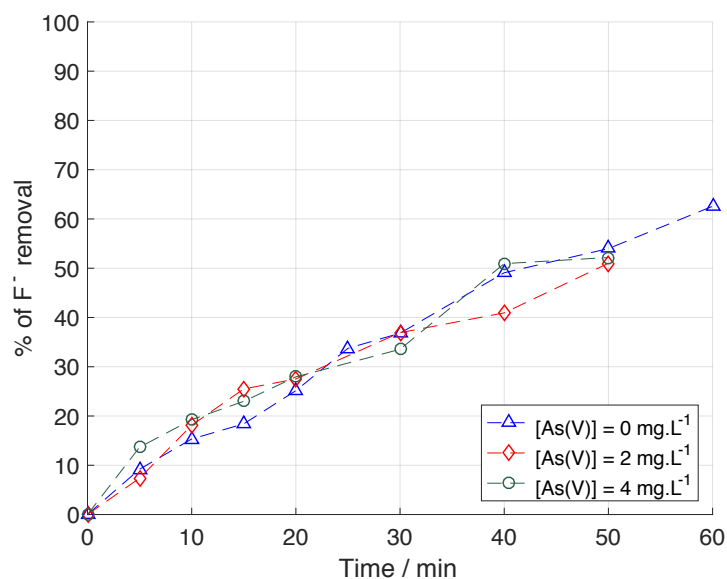


Figure 36 - Effect of arsenic (V) presence on fluoride removal. Operating conditions - initial fluoride concentration:  $15 \text{ mg}\cdot\text{L}^{-1}$ , electric current: 190 mA, initial water pH: 7

Table 34 - Aluminium dissolved and energy consumed with a binary mixture (fluoride + arsenic (V))

[As(V)] / $\text{mg}\cdot\text{L}^{-1}$	Aluminium dissolved / mg	Energy consumed / W·h
0	65.10	2.23
2	54.25	2.86
4	54.25	2.76

The results of the particle size distribution are provided in Table A3.35 and Figure 37.

The obtained results show that the range of particle sizes are similar between all the experiments. However, the increasing arsenic (V) initial concentration led to a decrease on the volume fraction of the bigger particles.

The TSS were determined at the end of each test. Values of  $33.4 \text{ mg}\cdot\text{L}^{-1}$ ,  $127.8 \text{ mg}\cdot\text{L}^{-1}$  and  $105.2 \text{ mg}\cdot\text{L}^{-1}$  were obtained respectively for the  $0 \text{ mg}\cdot\text{L}^{-1}$ ,  $2 \text{ mg}\cdot\text{L}^{-1}$  and  $4 \text{ mg}\cdot\text{L}^{-1}$  experiments.

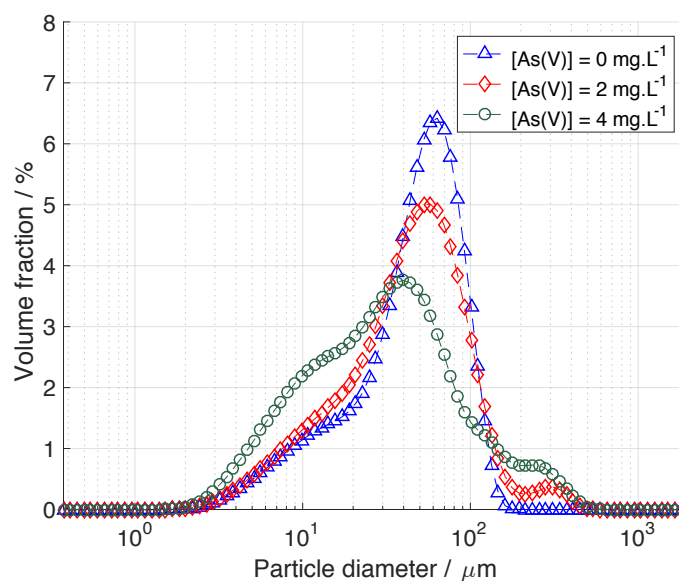


Figure 37 - Effect of arsenic (V) presence on the particles produced during the EC treatment of binary mixtures (fluoride + arsenic (V)). Operating conditions - initial fluoride concentration:  $15 \text{ mg}\cdot\text{L}^{-1}$ , electric current: 190 mA, initial water pH: 6

#### 4.4.6 Influence of fluoride presence on the removal of arsenic (V)

To determine the influence of the presence of fluoride on the removal of arsenic (V) experiments were conducted with an electric current of 190 mA, an initial arsenic (V) concentration of  $4 \text{ mg}\cdot\text{L}^{-1}$  and pH 7. Three different fluoride concentrations were tested ( $0 \text{ mg}\cdot\text{L}^{-1}$ ,  $5 \text{ mg}\cdot\text{L}^{-1}$ ,  $15 \text{ mg}\cdot\text{L}^{-1}$ ). The total anodic area was  $0.0076 \text{ m}^2$ . The operating conditions used are shown in Table 35.

The results are provided in Table A3.30 and Figure 38.

Table 35 - Operating conditions of the experiments with a binary mixture (arsenic (V) + fluoride)

[F <sup>-</sup> ] / $\text{mg}\cdot\text{L}^{-1}$	Start		End	
	pH	Conductivity / $\mu\text{S}\cdot\text{cm}^{-1}$	pH	Conductivity / $\mu\text{S}\cdot\text{cm}^{-1}$
0	6.6	350	7.0	347
5	7.2	347	8.3	346
15	7.0	344	8.4	340

The obtained results show that the fluoride presence affects the arsenic removal. As already discussed in 4.2.3 and 4.3.3, the removal mechanism of both contaminants releases  $\text{OH}^-$ . This fact can interfere with the adsorption mechanism leading to a decrease on the arsenic removal rate. Despite this influence, all the experiments reached a total arsenic removal at around the same time.

Using Equations (3) and (4), the theoretical amount of aluminium dissolved on the anode and the energy consumed on each experiment were calculated. The results are shown in Table 26.

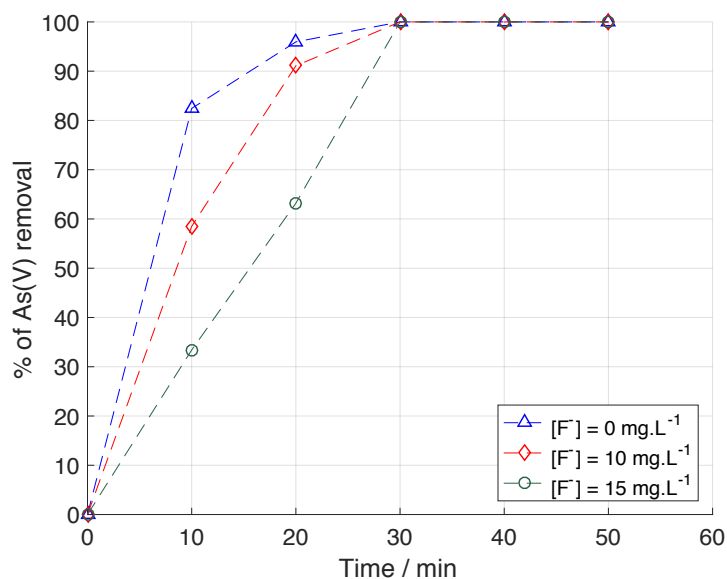


Figure 38 - Effect of fluoride presence on arsenic (V) removal. Operating conditions - initial arsenic (V) concentration:  $4 \text{ mg}\cdot\text{L}^{-1}$ , electric current: 190 mA, initial water pH: 7

Table 36 - Aluminium dissolved and energy consumed with a binary mixture (arsenic (V) + fluoride)

[F⁻] / $\text{mg}\cdot\text{L}^{-1}$	Aluminium dissolved / mg	Energy consumed / W·h
0	54.25	1.84
5	54.25	2.74
15	54.25	2.76

The results of the particle size distribution are provided in Table A3.36 and Figure 39.

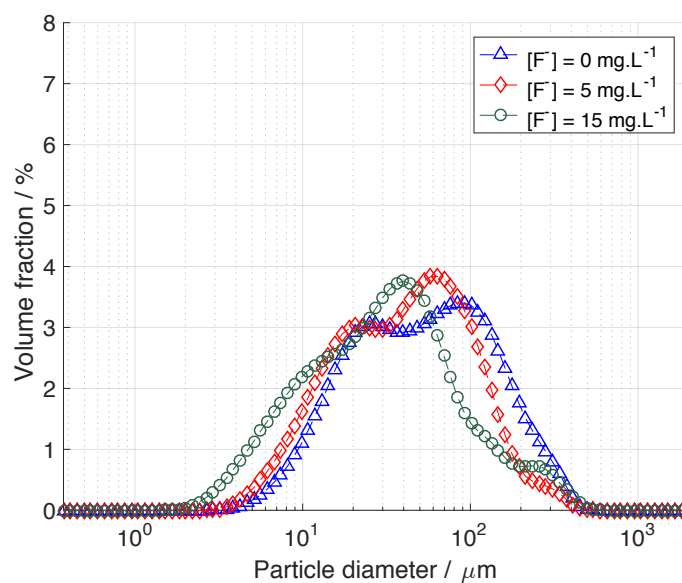


Figure 39 - Effect of fluoride presence on the particles produced during the EC treatment of binary mixtures (arsenic (V) + fluoride). Operating conditions - initial arsenic (V) concentration:  $25 \text{ mg}\cdot\text{L}^{-1}$ , electric current: 190 mA, initial water pH: 6

The obtained results show that there is no major difference between the particles formed in the presence of fluoride when compared to the experiment containing only arsenic (V).

The TSS were determined at the end of each test. Values of  $16.4 \text{ mg}\cdot\text{L}^{-1}$ ,  $131.6 \text{ mg}\cdot\text{L}^{-1}$  and  $105.2 \text{ mg}\cdot\text{L}^{-1}$  were obtained respectively for the  $0 \text{ mg}\cdot\text{L}^{-1}$ ,  $5 \text{ mg}\cdot\text{L}^{-1}$  and  $15 \text{ mg}\cdot\text{L}^{-1}$  experiments.

## 4.5 Ternary mixtures

To test the removal of contaminants on a ternary mixture an experiment was conducted with an initial iron concentration of  $20 \text{ mg}\cdot\text{L}^{-1}$ , an initial fluoride concentration of  $10 \text{ mg}\cdot\text{L}^{-1}$ , an initial arsenic (V) concentration of  $3 \text{ mg}\cdot\text{L}^{-1}$ , an electric current of 190 mA and pH 6. The total anodic area was  $0.0076 \text{ m}^2$ . The operating conditions used are shown in Table 37.

Table 37 - Operating conditions of the experiments with a ternary mixture (iron + fluoride + arsenic (V))

Start		End	
pH	Conductivity / $\mu\text{S}\cdot\text{cm}^{-1}$	pH	Conductivity / $\mu\text{S}\cdot\text{cm}^{-1}$
6.1	345	7.2	285

The obtained results are provided in Table A3.37 and Figure 40.

Comparing with the experiments where only one of the contaminants was present and using the same conditions, this experiment revealed that all contaminants were removed quicker. Furthermore, a more efficient removal of iron and fluoride was attained (higher removal percentages). After 50 min, the concentration of all contaminants was below the detection limit of the analytical methods.

The results of the particle size distribution are provided in Table A3.38 and Figure 41.

The obtained results are similar with those obtained with the experiment containing only iron. It also presents a similarity with the range of particle sizes produced by the experiment containing only fluoride, however it differs by having a smaller volume fraction of bigger particles. It differs from the experiment only containing arsenic (V) by exhibiting a range of smaller particles.

The TSS value of  $55.0 \text{ mg}\cdot\text{L}^{-1}$  was obtained at the end of the experiment.



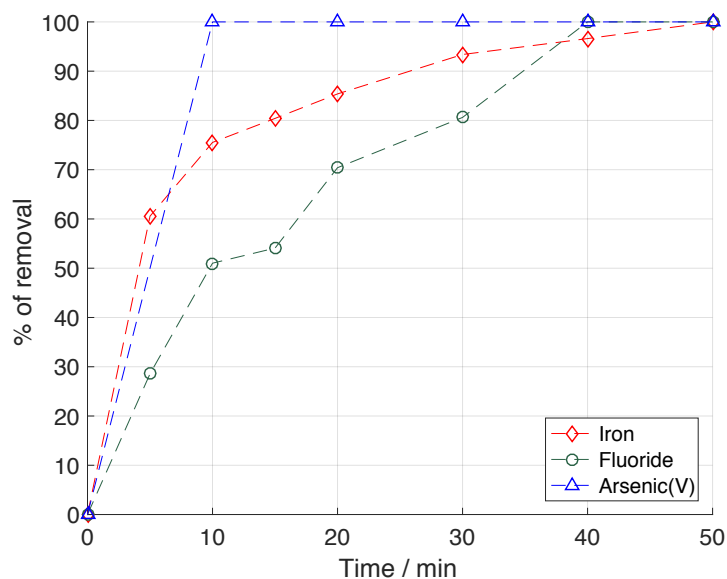


Figure 40 - Removal of contaminants on a ternary mixture (iron + fluoride + arsenic (V)). Operating conditions - initial water pH: 6, electric current: 190 mA, initial iron concentration:  $20 \text{ mg}\cdot\text{L}^{-1}$ , initial fluoride concentration:  $10 \text{ mg}\cdot\text{L}^{-1}$ , initial arsenic (V) concentration:  $3 \text{ mg}\cdot\text{L}^{-1}$

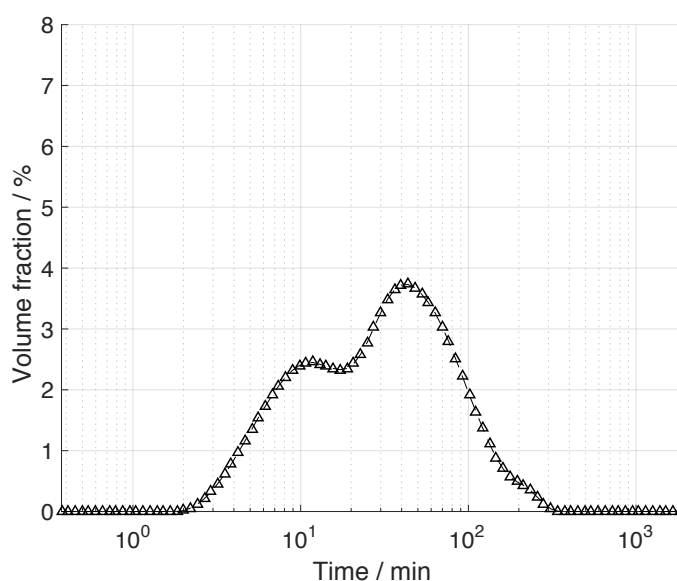


Figure 41 - Particles produced on a ternary mixture (iron + fluoride + arsenic (V)). Operating conditions - initial water pH: 6, electric current: 190 mA, initial iron concentration:  $20 \text{ mg}\cdot\text{L}^{-1}$ , initial fluoride concentration:  $10 \text{ mg}\cdot\text{L}^{-1}$ , initial arsenic (V) concentration:  $3 \text{ mg}\cdot\text{L}^{-1}$

Using Equations (3) and (4), the theoretical amount of aluminium dissolved on the anode and the energy consumed on each experiment were calculated. The results are shown in Table 38.

Table 38 - Aluminium dissolved and energy consumed with a ternary mixture (iron + arsenic (V) + fluoride)

Aluminium dissolved / mg	Energy consumed / W·h
65.10	3.57

## 5 Conclusions

With this work, it was possible to understand the influence of different operating parameters on the removal of different contaminants from water by electrocoagulation and on the particles produced during this process.

It was observed that the electric current applied can influence the rate at which the contaminant is removed. As based on Faraday's law, the number of flocs produced is proportional to the electric current applied and therefore higher electric currents lead to a higher production of flocs that remove the contaminants quicker. However, it was observed that, while testing this parameter for arsenic (V) removal, all experiments reached complete removal at the same time.

For the experiments where the initial contaminant concentration was the parameter analysed, it was observed that this parameter had no influence on the iron and arsenic (V) removal. However, on the fluoride trials, its removal was affected negatively by the increase of its initial concentration.

Experiments with different initial water pH resulted in different behaviours depending on the contaminant. While testing the influence of this parameter on the removal of iron, it was observed that the iron removal increased with the increase of the water initial pH. However, it was observed that in an alkaline medium the iron precipitates even before the EC process begins. In the experiments with fluoride and arsenic (V), it was observed that an increase of the initial water pH interferes with the removal of these contaminants due to an interference on the adsorption mechanism of these on the flocs due to higher  $\text{OH}^-$  concentration.

The number of electrodes and their arrangements showed no significant influence on the iron and on the fluoride experiments. However, a quicker removal of arsenic (V) was observed. As the electric current employed was the same in all tests, from the Faraday's law it is expected that the dissolved amount of aluminium was the same. However, the anodic area is bigger in the tests with four electrodes, which must be affecting the removal process. To verify if the size of the flocs formed with the different arrangements may be a possible explanation for the observed difference, it would be interesting to perform blank tests with these conditions.

The tests with binary mixtures showed that the only experiments for which some influence was observed were the iron presence improving the arsenic (V) removal and the fluoride presence decreasing the arsenic (V) removal.

On the experiment with a ternary mixture it was observed that all the contaminants were removed quicker than on the trials with only one of them present. However, for a better

understanding of the influence this kind of mixture might have on the removal of contaminants more trials should be conducted with different contaminant concentrations.

For future work, the same parameters could be tested while applying the EC process to remove other contaminants like copper, nickel, cadmium and lead. A continuous reactor could also be designed to assess the possibility of continuous operation of this process. It could also be interesting to repeat some experiments using alternating current (AC) to test its influence on the removal of contaminants and on the electrodes efficiency.

Overall, I consider that the balance of the work developed is highly positive. Despite some challenges that appeared along the way most of the planned work was concluded.

## 6 References

- Ali, I., Asim, M., Khan, T.A., 2013. Arsenite removal from water by electro-coagulation on zinc-zinc and copper-copper electrodes. *Int. J. Environ. Sci. Technol.* 10, 377-384. doi:10.1007/s13762-012-0113-z
- Bache, D., Gregory, R., 2007. *Flocs in Water Treatment*, 30th Ed. ed. International Water Association.
- Bratby, J., 2015. *Coagulation and Flocculation in Water and Wastewater Treatment*, Second. ed, Water Intelligence Online. International Water Association. doi:10.2166/9781780402321
- Can, B.Z., Boncukcuoglu, R., Yilmaz, A.E., Fil, B.A., 2012. Effect of some operational parameters on the arsenic removal by electrocoagulation using aluminum electrodes. *Energy Educ. Sci. Technol. Part A Energy Sci. Res.* 30, 611-622. doi:10.1186/2052-336X-12-95
- Chen, G., 2004. Electrochemical technologies in wastewater treatment. *Sep. Purif. Technol.* 38, 11-41. doi:10.1016/j.seppur.2003.10.006
- Emamjomeh, M., Sivakumar, M., Schafer, A., 2004. Fluoride removal by using a batch electrocoagulation reactor. *J. Environ. Manage.* 90, 143-152.
- Eslami, A., Takdastan, A., Emami Tabar, S., Neisi, A., 2014. Fluoride Removal From Drinking Water by Electrocoagulation Using Iron and Aluminum Electrodes. *Jundishapur J Heal. Sci* 6, 1-7. doi:10.5812/jjhs.21718
- Fajardo, A.S., Martins, R.C., Quinta-Ferreira, R.M., 2014. Treatment of a synthetic phenolic mixture by electrocoagulation using Al, Cu, Fe, Pb, and Zn as anode materials. *Ind. Eng. Chem. Res.* 53, 18339-18345. doi:10.1021/ie502575d
- Fajardo, A.S., Rodrigues, R.F., Martins, R.C., Castro, L.M., Quinta-Ferreira, R.M., 2015. Phenolic wastewaters treatment by electrocoagulation process using Zn anode. *Chem. Eng. J.* 275, 331-341. doi:10.1016/j.cej.2015.03.116
- Farrell, J., Wang, J., O'Day, P., Conklin, M., 2001. Electrochemical and Spectroscopic Study of Arsenate Removal from Water Using Zero-Valent Iron Media. *Environ. Sci. Technol.* 35, 2026-2032. doi:10.1021/es0016710
- Ghosh, D., Solanki, H., Purkait, M.K., 2008. Removal of Fe(II) from tap water by electrocoagulation technique. *J. Hazard. Mater.* 155, 135-143. doi:10.1016/j.jhazmat.2007.11.042
- Harris, J.T., 1909. *Process of preparing potable waters*. US 937210 A.
- Holt, P.K., Barton, G.W., Mitchell, C.A., 2005. The future for electrocoagulation as a localized water treatment technology. *Chemosphere* 59, 355-367. doi:10.1016/j.chemosphere.2004.10.023
- Kim, K.J., Baek, K., Ji, S., Cheong, Y., Yim, G., Jang, A., 2016. Study on electrocoagulation parameters (current density, pH, and electrode distance) for removal of fluoride from groundwater. *Environ. Earth Sci.* 75, 1-8. doi:10.1007/s12665-015-4832-6
- Kingston, R.L., Hall, S., Sioris, L., 1993. Clinical Observations and Medical Outcome in 149 Cases of Arsenate ant Killer Ingestion. *Clin. Toxicol.* 31, 581-591. doi:10.3109/15563659309025763
- Koby, M., Bayramoglu, M., Eyvaz, M., 2007. Techno-economical evaluation of electrocoagulation for the textile wastewater using different electrode connections. *J. Hazard. Mater.* 148, 311-318. doi:10.1016/j.jhazmat.2007.02.036
- Kumar, P.R., Chaudhari, S., Khilar, K.C., Mahajan, S.P., 2004. Removal of arsenic from water by electrocoagulation. *Chemosphere* 55, 1245-1252. doi:10.1016/j.chemosphere.2003.12.025
- Lohmann, G., Lorenz, S., 2000. On the hydrological cycle under paleoclimatic conditions as derived from AGCM simulations. *J. Geophys. Res. D Atmos.* 105, 17,417,436. doi:10.1029/2000jd900189
- Meenakshi, Maheshwari, R.C., 2006. Fluoride in drinking water and its removal. *J. Hazard. Mater.* 137, 456-463. doi:10.1016/j.jhazmat.2006.02.024
- Mollah, M.Y.A., Morkovsky, P., Gomes, J.A.G., Kesmez, M., Parga, J., Cocke, D.L., 2004. Fundamentals, present and future perspectives of electrocoagulation. *J. Hazard. Mater.* 114, 199-210. doi:10.1016/j.jhazmat.2004.08.009

- Mollah, M.Y., Schennach, R., Parga, J.R., Cocke, D.L., 2001. Electrocoagulation (EC)--science and applications. *J. Hazard. Mater.* 84, 29-41. doi:10.1016/S0304-3894(01)00176-5
- Ohshima, H., 2012. *Electrical Phenomena at Interfaces and Biointerfaces: Fundamentals and Applications in Nano-, Bio-, and Environmental Sciences*, 1st Ed. ed, Electrical Phenomena at Interfaces and Biointerfaces: Fundamentals and Applications in Nano-, Bio-, and Environmental Sciences. Wiley. doi:10.1002/9781118135440
- Picard, T., Cathalifaud-Feuillade, G., Mazet, M., Vandensteendam, C., Määttä, R., Tabakov, D., Koren, J., Syversen, U., Matteson, M.J., Dobson, R.L., Glenn, R.W., Kukunoor, N.S., Waits, W.H., Clayfield, E.J., Mameri, N., Yeddou, A., Lounici, H., Belhocine, D., Grib, H., Bariou, D., Lin, S.H., Shyu, C.T., Sun, M.C., Baklan, V.Y., Radygin, G. V., Gonopol'skii, S.N., Shuster, N.A., Rukitskii, A.I., Markodei, F. V., Khitrich, V.F., Ponomarenko, T.I., Szynekarczuk, J., Kan, J., Hassan, T.A., Donini, J.C., Averisyan, D., Tarkhanyan, A.S., Safaryan, L.N., Zolotukhin, I.A., Hosny, A.Y., Pouet, M.F., Grasmick, A., Vik, E.A., Carlson, D.A., Eikum, A.S., Gjessing, E.T., Radovan, C., Vlaicu, I., Ciorba, G.A., Adam-Stoia, M., Przhegorlinskii, V.I., Ivanishvili, A.I., Grebenyuk, V.D., Donini, J.C., Kan, J., Szynekarczuk, J., Hassan, T.A., Kar, K.L., Jiaqian, J., Dobolyi, E., 2000. Cathodic dissolution in the electrocoagulation process using aluminium electrodes. *J. Environ. Monit.* 2, 77-80. doi:10.1039/a908248d
- Pritchard, M., Craven, T., Mkandawire, T., Edmondson, A.S., O'Neill, J.G., 2010. A comparison between *Moringa oleifera* and chemical coagulants in the purification of drinking water - An alternative sustainable solution for developing countries. *Phys. Chem. Earth, Parts A/B/C* 35, 798-805. doi:10.1016/j.pce.2010.07.014
- Sawyer, C.N., McCarty, P.L., Parkin, G.F., 2016. *Chemistry for environmental engineering and science*, 5th Ed. ed. McGraw-Hill.
- Scholz, F., 2010. *Electroanalytical methods: Guide to experiments and applications*, 1st Ed. ed, *Electroanalytical Methods: Guide to Experiments and Applications*. Springer. doi:10.1007/978-3-642-02915-8
- Shen, F., Chen, X., Gao, P., Chen, G., 2003. Electrochemical removal of fluoride ions from industrial wastewater. *Chem. Eng. Sci.* 58, 987-993. doi:10.1016/S0009-2509(02)00639-5
- Sillanpää, M.E.T., 2015. *Natural organic matter in water: characterization and treatment methods*. International Water Association.
- The Council of the European Union, 1998. Council Directive 98/83/EC of 3 November 1998 on the quality of water intended for human consumption. *Off. J. Eur. Communities* L330, 32-54. doi:2004R0726 - v.7 of 05.06.2013
- UNICEF, 2014. UNICEF Tap Project [WWW Document]. URL <https://www.unicefusa.org/stories/mission/survival/water/tap-project/infographic-world-water-crisis/7556> (accessed 10.18.16).
- Vasudevan, S., 2012. Effects of alternating current (AC) and direct current (DC) in electrocoagulation process for the removal of iron from water. *Can. J. Chem. Eng.* 90, 1160-1169. doi:10.1002/cjce.20625
- Vasudevan, S., Jayaraj, J., Lakshmi, J., Sozhan, G., 2009a. Removal of iron from drinking water by electrocoagulation: Adsorption and kinetics studies. *Korean J. Chem. Eng.* 26, 1058-1064. doi:10.1007/s11814-009-0176-9
- Vasudevan, S., Lakshmi, J., Sozhan, G., 2009b. Studies on a Mg-Al-Zn alloy as an anode for the removal of fluoride from drinking water in an electrocoagulation process. *Clean - Soil, Air, Water* 37, 372-378. doi:10.1002/clen.200900031
- Wan, W., Pepping, T.J., Banerji, T., Chaudhari, S., Giammar, D.E., 2011. Effects of water chemistry on arsenic removal from drinking water by electrocoagulation. *Water Res.* 45, 384-392. doi:10.1016/j.watres.2010.08.016
- WHO Recommendations, 2011. *WHO Guidelines for Drinking Water Quality*, 4th Ed. ed, WHO Recommendations. World Health Organization, Geneva, Switzerland. doi:10.1016/S1462-0758(00)00006-6

## Appendix A1. Colloids theory

In wastewaters, most pollutants are colloidal particles. These particles are charged aggregates of atoms and molecules and they are very stable in water. Due to this stability, it is not easy to remove them from the water by filtration, sedimentation or flotation. The particles stability is also affected by the particles surface charges (Sawyer et al., 2016, chap. 7). For this reason, water treatment methods such as coagulation change the electrical properties of colloids to disrupt their stability.

### A1.1 DLVO theory

The DLVO theory, named after the work of Derjagin, Landau, Verwey and Overbeek, relates the distance between particles and the repulsive and attractive forces between them. London dispersion forces and van der Waals forces counteract the repulsive forces pulling the particles together and their intensity increases as the distance between the particles decreases.

The electrical double layer (EDL) has an important role in the origin of repulsive forces. The EDL consists of an inner region which includes water molecules and adsorbed ions and a diffuse region where the ions are distributed based on their influence on the electrical forces. The first theoretical model was developed in 1879 by Helmholtz which assumed the presence of a compact layer of ions in contact with a charged metal surface. Later, a new model by Gouy and Chapman described a diffuse double layer in which the accumulated ions extended to some distance from the solid surface. More recently, in 1924, it was suggested by Stern that the electrified solid-liquid interface included both Helmholtz and Gouy-Chapman models. It is also known that the EDL influences various interfacial electrical phenomena such as interactions between colloidal particles and electrokinetic phenomena (Ohshima, 2012, chap. 2; Sawyer et al., 2016, chap. 7; Scholz, 2010, chap. 1; Vasudevan, 2012).

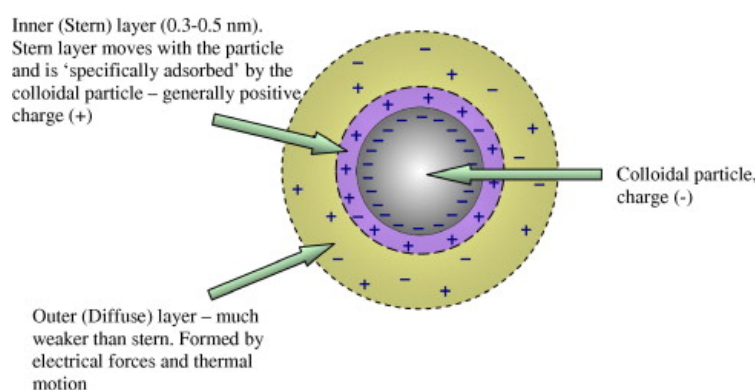


Figure A1.1 - Structure of the electrical double layer (Pritchard et al., 2010)

As shown in Figure A1.1 the inner region of the EDL is called fixed layer, or Stern layer, and it is where the particle surface binds ions to itself. The outer diffusion region is called the Gouy-Chapman layer and it is where ions can move due to diffusion. So, the EDL is formed by separating the charges on the particle surface by attracting oppositely charged ions to the surface and repelling ions with the same charge. Which means that when two colloidal particles become closer to each other their diffusion layers make them repel each other as the distance between them decreases (Bache and Gregory, 2007; Bratby, 2015, chap. 2; Sawyer et al., 2016, chap. 7).

Usually when two particles become closer the repulsive force is stronger than the attractive forces. Therefore, larger particles are not usually formed. However, if the attractive forces become dominant and the energy barrier is exceeded the particles become joined together (Bache and Gregory, 2007; Sawyer et al., 2016, chap. 7).

## Appendix A2. Chemical analytics

The characteristics of the Spectroquant Prove 300 spectrophotometer (Merck Millipore) are described in Table A2.1.

Table A2.1 - Spectroquant Prove 300 technical characteristics

Wavelength range	190 - 1100 nm
Spectral bandwidth	4 nm
Wavelength resolution	1 nm (scan 0.1 nm)
Wavelength reproducibility	± 0.2 nm
Wavelength accuracy	± 1 nm
Stray light	≤ 0.1 % transmission at 340 nm; ≤ 1 % transmission at 198 nm
Photometric range	± 3.0 Abs
Absorbance resolution	0.001 Abs
Absorbance reproducibility	± 0.003 at 1 absorbance between 200 nm and 900 nm

The characteristics of the methods used to determine the contaminants concentrations are provided in Table A2.2.

Table A2.2 - Technical characteristics of the methods used to determine the contaminants concentrations

	Iron test 114761 (Merck Millipore)	Fluoride Test 114598 (Merck Millipore)	Arsenic Test 101747 (Merck Millipore)
Measuring range (mg·L <sup>-1</sup> )	0.05 - 5.00	1.0 - 20.0	0.001 - 0.100
Standard deviation of the method (mg·L <sup>-1</sup> )	± 0.16	± 0.015	± 0.0017
Coefficient of variation of the method (%)	± 0.65	± 1.4	± 3.0
Confidence interval (mg·L <sup>-1</sup> )	± 0.04	± 0.04	± 0.004
Sensitivity: Absorbance 0.010 A corresponds to (mg·L <sup>-1</sup> )	0.02	0.02	0.001
Accuracy of a measurement value	Max. ± 0.07	Max. ± 1.2	Max. ± 0.006



## Appendix A3. Results obtained during the EC treatment

### A3.1 Iron

#### A3.1.1 Concentration

##### A3.1.1.1 Influence of electric current

Table A3.1 - Results obtained for the iron concentration over time with different electric currents

I / mA	Time / min	0	5	10	15	20	25	30	40	50	60
40	[Fe] / mg·L <sup>-1</sup>	26.10	18.85	19.59	18.03	18.01	19.30	14.19	14.23	14.20	11.80
	% of Fe removal	0.0	27.8	24.9	30.9	31.0	26.1	45.6	45.5	45.6	54.8
100	[Fe] / mg·L <sup>-1</sup>	25.31	16.80	16.03	14.57	14.41	12.56	13.39	9.76	9.11	5.17
	% of Fe removal	0	33.6	36.7	42.4	43.1	50.4	47.1	61.4	64.0	79.6
190	[Fe] / mg·L <sup>-1</sup>	25.20	14.97	13.78	11.78	8.78	7.49	5.71	5.43	2.76	1.61
	% of Fe removal	0	40.6	45.3	53.3	65.2	70.3	77.3	78.5	89.0	93.6

##### A3.1.1.2 Influence of initial iron concentration

Table A3.2 - Results obtained for the iron concentration over time with different initial iron concentrations

[Fe] / mg·L <sup>-1</sup>	Time / min	0	5	10	15	20	25	30	40	50	60
15	[Fe] / mg·L <sup>-1</sup>	15.64	8.55	7.50	5.40	3.91	2.75	2.02	1.74	1.35	1.14
	% of Fe removal	0.0	45.3	52.0	65.5	75.0	82.4	87.1	88.9	91.4	92.7
20	[Fe] / mg·L <sup>-1</sup>	20.62	11.06	8.28	6.49	5.99	5.52	4.60	2.74	1.86	1.42
	% of Fe removal	0.0	46.4	59.8	68.5	71.0	73.2	77.7	86.7	91.0	93.1
25	[Fe] / mg·L <sup>-1</sup>	25.20	14.97	13.78	11.78	8.78	7.49	5.71	5.43	2.76	1.61
	% of Fe removal	0.0	40.6	45.3	53.3	65.2	70.3	77.3	78.5	89.0	93.6

## A3.1.1.3 Influence of initial water pH

Table A3.3 - Results obtained for the iron concentration over time with different initial water pH

Initial pH	Time / min	0	5	10	15	20	25	30	40	50	60
3	[Fe] / mg·L <sup>-1</sup>	28.34	20.75	17.40	16.50	14.73	11.99	11.69	6.87	4.59	4.48
	% of Fe removal	0.0	26.8	38.6	41.8	48.0	57.7	58.8	75.8	84.1	84.2
6	[Fe] / mg·L <sup>-1</sup>	25.20	14.97	13.78	11.78	8.78	7.49	5.71	5.43	2.76	1.61
	% of Fe removal	0.0	40.6	45.3	53.3	65.2	70.3	77.3	78.5	89.0	93.6
10	[Fe] / mg·L <sup>-1</sup>	25.57	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	% of Fe removal	0.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0
	pH	10.2		9.3		9.4		9.3	8.9	8.9	9.0
	TSS / mg·L <sup>-1</sup>	2.0		5.6		15.8		27.6	48.8	53.8	69.6

## A3.1.1.4 Influence of electrodes arrangements

Table A3.4 - Results obtained for the iron concentration over time with different electrodes arrangements

Arrangement	Time / min	0	5	10	15	20	25	30	40	50	60
Simple	[Fe] / mg·L <sup>-1</sup>	25.20	14.97	13.78	11.78	8.78	7.49	5.71	5.43	2.76	1.61
	% of Fe removal	0.0	40.6	45.3	53.3	65.2	70.3	77.3	78.5	89.0	93.6
Series monopolar	[Fe] / mg·L <sup>-1</sup>	25.96	17.38	15.73	7.76	7.80	4.95	4.87	3.62	1.26	0.00
	% of Fe removal	0.0	33.1	39.4	70.1	70.0	81.0	81.2	86.1	95.2	100.0
Series bipolar	[Fe] / mg·L <sup>-1</sup>	25.96	16.06	10.84	7.00	4.51	3.49	3.89	3.16	2.42	0.00
	% of Fe removal	0.0	38.1	58.2	73.0	82.6	86.6	85.0	87.8	90.7	100.0
Parallel	[Fe] / mg·L <sup>-1</sup>	25.25	15.79	16.09	13.35	9.67	9.26	9.13	4.67	2.65	1.80
	% of Fe removal	0.0	37.5	36.3	47.1	61.7	63.3	63.8	81.5	89.5	92.87

### A3.1.2 Particles size

#### A3.1.2.1 Influence of electric current

Table A3.5 - Results obtained for the distribution of particle sizes with different electric currents

Particle diameter (µm)	Electric current (mA)						
	40	100	190				
	Volume fraction (%)						
0.375	0	0	0	11.83	1.94	5.53	2.76
0.412	0	0	0	12.99	2.03	5.31	2.84
0.452	0	0	0	14.26	2.12	4.96	2.87
0.496	0	0	0	15.65	2.21	4.5	2.85
0.545	0	0	0	17.18	2.3	3.96	2.81
0.598	0	0	0	18.86	2.42	3.41	2.75
0.657	0	0	0	20.7	2.57	2.91	2.71
0.721	0	0	0	22.73	2.76	2.48	2.72
0.791	0	0	0	24.95	3	2.15	2.78
0.869	0	0	0	27.38	3.27	1.92	2.92
0.953	0	0	0	30.07	3.55	1.75	3.13
1.047	0	0	0	33	3.81	1.61	3.37
1.149	0	0	0	36.24	4.03	1.48	3.62
1.261	0	0	0	39.77	4.16	1.34	3.84
1.385	0	0	0	43.66	4.21	1.18	4
1.52	0	0	0	47.93	4.16	1.01	4.08
1.669	0	0	0	52.63	4.02	0.85	4.08
1.832	0.0003	0.001	0	57.77	3.82	0.72	3.98
2.01	0.0032	0.013	0.00008	63.41	3.57	0.63	3.8
2.207	0.017	0.071	0.0016	69.62	3.3	0.56	3.55
2.423	0.047	0.2	0.012	76.43	3.01	0.49	3.22
2.66	0.091	0.39	0.045	83.9	2.74	0.39	2.85
2.92	0.14	0.62	0.11	92.09	2.47	0.26	2.45
3.206	0.21	0.9	0.2	101.1	2.22	0.13	2.04
3.519	0.29	1.23	0.3	111	1.98	0.038	1.62
3.862	0.39	1.61	0.43	121.8	1.74	0.0058	1.22
4.241	0.5	2.04	0.59	133.7	1.5	0.00028	0.84
4.656	0.62	2.49	0.77	146.8	1.25	0	0.5
5.111	0.74	2.95	0.96	161.2	1.02	0	0.26
5.611	0.88	3.42	1.17	176.8	0.8	0	0.11
6.158	1.02	3.88	1.39	194.2	0.62	0	0.055
6.761	1.16	4.32	1.62	213.2	0.5	0	0.036
7.421	1.31	4.73	1.85	234.1	0.42	0	0.038
8.147	1.45	5.08	2.07	256.8	0.39	0	0.05
8.944	1.58	5.35	2.28	282.1	0.38	0	0.064
9.819	1.71	5.54	2.47	309.6	0.39	0	0.072
10.78	1.83	5.6	2.64	339.8	0.37	0	0.068
				373.1	0.32	0	0.051
				409.6	0.24	0	0.028
				449.7	0.15	0	0.0097
				493.6	0.074	0	0.0017
				541.9	0.03	0	0.0001
				594.9	0.014	0	0
				653	0.012	0	0
				716.9	0.024	0	0
				786.9	0.035	0	0

863.9	0.034	0	0
948.2	0.011	0	0
1041	0.0017	0	0
1143	0	0	0
1255	0	0	0
1377	0	0	0
1512	0	0	0
1660	0	0	0
1822	0	0	0

## A3.1.2.2 Influence of initial iron concentration

Table A3.6 - Results obtained for the distribution of particle sizes with different initial iron concentrations

Particle diameter ( $\mu\text{m}$ )	Initial iron concentration ( $\text{mg}\cdot\text{L}^{-1}$ )			
	0	15	20	25
	Volume fraction (%)			
0.375	0	0	0	0
0.412	0	0	0	0
0.452	0	0	0	0
0.496	0	0	0	0
0.545	0	0	0	0
0.598	0	0	0	0
0.657	0	0	0	0
0.721	0	0	0	0
0.791	0	0	0	0
0.869	0	0	0	0
0.953	0	0	0	0
1.047	0	0	0	0
1.149	0	0	0	0
1.261	0	0	0	0
1.385	0	0	0	0
1.52	0	0.00001	0	0
1.669	0	0.00044	0	0
1.832	0	0.0039	0.00004	0
2.01	0	0.017	0.0013	0.00008
2.207	0	0.043	0.011	0.0016
2.423	0	0.08	0.046	0.012
2.66	0.00004	0.13	0.11	0.045
2.92	0.0008	0.18	0.21	0.11
3.206	0.0051	0.26	0.33	0.2
3.519	0.017	0.34	0.48	0.3
3.862	0.036	0.44	0.65	0.43
4.241	0.061	0.55	0.84	0.59
4.656	0.093	0.66	1.06	0.77

5.111	0.13	0.79	1.29	0.96
5.611	0.18	0.92	1.52	1.17
6.158	0.23	1.06	1.76	1.39
6.761	0.29	1.19	2.01	1.62
7.421	0.36	1.33	2.24	1.85
8.147	0.43	1.46	2.45	2.07
8.944	0.51	1.59	2.65	2.28
9.819	0.6	1.71	2.8	2.47
10.78	0.69	1.83	2.92	2.64
11.83	0.79	1.93	2.99	2.76
12.99	0.89	2.02	3	2.84
14.26	0.99	2.11	2.96	2.87
15.65	1.08	2.21	2.89	2.85
17.18	1.17	2.31	2.8	2.81
18.86	1.25	2.45	2.71	2.75
20.7	1.33	2.63	2.67	2.71
22.73	1.39	2.85	2.68	2.72
24.95	1.45	3.12	2.76	2.78
27.38	1.58	3.75	3.09	3.13
30.07	1.66	4.07	3.28	3.37
33	1.76	4.34	3.46	3.62
36.24	1.86	4.54	3.58	3.84
39.77	1.99	4.65	3.62	4
43.66	2.12	4.66	3.59	4.08
47.93	2.26	4.56	3.49	4.08
52.63	2.43	4.36	3.34	3.98
57.77	2.6	4.09	3.15	3.8
63.41	2.8	3.74	2.91	3.55
69.62	3.02	3.35	2.65	3.22
76.43	3.24	2.92	2.36	2.85
83.9	3.46	2.5	2.06	2.45
92.09	3.66	2.08	1.75	2.04
101.1	3.82	1.69	1.44	1.62
111	3.91	1.32	1.15	1.22
121.8	3.93	0.97	0.87	0.84
133.7	3.87	0.66	0.64	0.5
146.8	3.74	0.43	0.46	0.26
161.2	3.57	0.28	0.34	0.11
176.8	3.37	0.22	0.25	0.055
194.2	3.16	0.21	0.2	0.036
213.2	2.96	0.23	0.15	0.038
234.1	2.77	0.25	0.12	0.05
256.8	2.57	0.23	0.099	0.064
282.1	2.35	0.16	0.086	0.072
309.6	2.09	0.083	0.069	0.068
339.8	1.8	0.025	0.042	0.051

373.1	1.47	0.0037	0.017	0.028
409.6	1.13	0.00017	0.0032	0.0097
449.7	0.83	0	0.00024	0.0017
493.6	0.58	0	0	0.0001
541.9	0.41	0	0	0
594.9	0.29	0	0	0
653	0.22	0	0	0
716.9	0.18	0	0	0
786.9	0.17	0	0	0
863.9	0.17	0	0	0
948.2	0.17	0	0	0
1041	0.15	0	0	0
1143	0.12	0	0	0
1255	0.084	0	0	0
1377	0.067	0	0	0
1512	0.056	0	0	0
1660	0.046	0	0	0
1822	0	0	0	0

## A3.1.2.3 Influence of initial water pH

Table A3.7 - Results obtained for the distribution of particle sizes with different initial water pH

Particle diameter (µm)	Initial water pH		
	3	6	10
	Volume fraction (%)		
0.375	0	0	0
0.412	0	0	0
0.452	0	0	0
0.496	0	0	0
0.545	0	0	0
0.598	0	0	0
0.657	0	0	0
0.721	0	0	0
0.791	0	0	0
0.869	0	0	0
0.953	0	0	0
1.047	0	0	0
1.149	0	0	0
1.261	0	0	0
1.385	0	0	0
1.52	0	0	0
1.669	0	0	0
1.832	0	0.001	0
2.01	0	0.013	0
2.207	0.00017	0.071	0

2.423	0.0027	0.2	0
2.66	0.016	0.39	0
2.92	0.049	0.62	0.00002
3.206	0.1	0.9	0.00039
3.519	0.18	1.23	0.0025
3.862	0.27	1.61	0.0085
4.241	0.38	2.04	0.019
4.656	0.51	2.49	0.032
5.111	0.67	2.95	0.05
5.611	0.84	3.42	0.072
6.158	1.02	3.88	0.1
6.761	1.22	4.32	0.13
7.421	1.43	4.73	0.18
8.147	1.65	5.08	0.23
8.944	1.87	5.35	0.29
9.819	2.09	5.54	0.36
10.78	2.3	5.6	0.46
11.83	2.49	5.53	0.58
12.99	2.66	5.31	0.72
14.26	2.79	4.96	0.91
15.65	2.89	4.5	1.13
17.18	2.94	3.96	1.41
18.86	2.96	3.41	1.74
20.7	2.95	2.91	2.14
22.73	2.94	2.48	2.61
24.95	2.94	2.15	3.14
27.38	2.97	1.92	3.72
30.07	3.04	1.75	4.33
33	3.15	1.61	4.93
36.24	3.28	1.48	5.47
39.77	3.42	1.34	5.91
43.66	3.55	1.18	6.19
47.93	3.64	1.01	6.27
52.63	3.69	0.85	6.14
57.77	3.71	0.72	5.81
63.41	3.68	0.63	5.33
69.62	3.62	0.56	4.74
76.43	3.5	0.49	4.11
83.9	3.33	0.39	3.5
92.09	3.08	0.26	2.95
101.1	2.77	0.13	2.47
111	2.4	0.038	2.07
121.8	2	0.0058	1.73
133.7	1.57	0.00028	1.45
146.8	1.15	0	1.22
161.2	0.77	0	1.03

176.8	0.46	0	0.87
194.2	0.25	0	0.74
213.2	0.13	0	0.64
234.1	0.083	0	0.55
256.8	0.072	0	0.47
282.1	0.073	0	0.39
309.6	0.078	0	0.31
339.8	0.08	0	0.21
373.1	0.077	0	0.12
409.6	0.067	0	0.044
449.7	0.056	0	0.0083
493.6	0.048	0	0.00064
541.9	0.036	0	0
594.9	0.021	0	0
653	0.0054	0	0
716.9	0.00066	0	0
786.9	0	0	0
863.9	0	0	0
948.2	0	0	0
1041	0	0	0
1143	0	0	0
1255	0	0	0
1377	0	0	0
1512	0	0	0
1660	0	0	0
1822	0	0	0

## A3.1.2.4 Influence of electrodes arrangements

Table A3.8 - Results obtained for the distribution of particle sizes with different electrodes arrangements

Particle diameter (µm)	Electrodes arrangement			
	S	P	SM	SB
	Volume fraction (%)			
0.375	0	0	0	0
0.412	0	0	0	0
0.452	0	0	0	0
0.496	0	0	0	0
0.545	0	0	0	0
0.598	0	0	0	0
0.657	0	0	0	0
0.721	0	0	0	0
0.791	0	0	0	0
0.869	0	0	0	0
0.953	0	0	0	0

1.047	0	0	0	0
1.149	0	0	0	0
1.261	0	0	0	0
1.385	0	0	0	0
1.52	0	0	0	0
1.669	0	0	0	0.0004
1.832	0.001	0.00001	0	0.0056
2.01	0.013	0.002	0	0.029
2.207	0.071	0.022	0	0.077
2.423	0.2	0.11	0.0002	0.15
2.66	0.39	0.29	0.0027	0.23
2.92	0.62	0.55	0.015	0.34
3.206	0.9	0.85	0.042	0.47
3.519	1.23	1.22	0.084	0.62
3.862	1.61	1.65	0.13	0.79
4.241	2.04	2.13	0.2	0.98
4.656	2.49	2.65	0.27	1.19
5.111	2.95	3.18	0.36	1.4
5.611	3.42	3.72	0.46	1.63
6.158	3.88	4.25	0.57	1.86
6.761	4.32	4.76	0.69	2.09
7.421	4.73	5.22	0.82	2.31
8.147	5.08	5.61	0.95	2.53
8.944	5.35	5.9	1.1	2.73
9.819	5.54	6.08	1.24	2.92
10.78	5.6	6.1	1.38	3.07
11.83	5.53	5.95	1.52	3.19
12.99	5.31	5.61	1.64	3.27
14.26	4.96	5.1	1.76	3.31
15.65	4.5	4.45	1.85	3.32
17.18	3.96	3.73	1.93	3.29
18.86	3.41	3	2	3.25
20.7	2.91	2.34	2.07	3.21
22.73	2.48	1.81	2.15	3.17
24.95	2.15	1.44	2.26	3.15
21	1.92	1.23	2.42	3.14
27.38	1.75	1.14	2.63	3.13
30.07	1.61	1.11	2.91	3.11
33	1.48	1.11	3.26	3.07
36.24	1.34	1.08	3.65	2.98
39.77	1.18	1.01	4.07	2.85
43.66	1.01	0.9	4.5	2.69
47.93	0.85	0.79	4.91	2.51
52.63	0.72	0.69	5.25	2.34
57.77	0.63	0.62	5.48	2.19
63.41	0.56	0.57	5.55	2.07

69.62	0.49	0.53	5.45	1.94
76.43	0.39	0.48	5.14	1.81
83.9	0.26	0.4	4.65	1.67
92.09	0.13	0.31	4.02	1.51
101.1	0.038	0.2	3.3	1.34
111	0.0058	0.098	2.56	1.18
121.8	0.0003	0.033	1.85	1.02
133.7	0	0.0058	1.23	0.89
146.8	0	0.0004	0.71	0.77
161.2	0	0	0.35	0.67
176.8	0	0	0.13	0.58
194.2	0	0	0.05	0.5
213.2	0	0	0.033	0.41
234.1	0	0	0.045	0.33
256.8	0	0	0.07	0.26
282.1	0	0	0.089	0.21
309.6	0	0	0.084	0.14
339.8	0	0	0.054	0.074
373.1	0	0	0.023	0.025
409.6	0	0	0.0045	0.0044
449.7	0	0	0.0004	0.0003
493.6	0	0	0	0
541.9	0	0	0	0
594.9	0	0	0	0
653	0	0	0	0
716.9	0	0	0	0
786.9	0	0	0	0
863.9	0	0	0	0
948.2	0	0	0	0
1041	0	0	0	0
1143	0	0	0	0
1255	0	0	0	0
1377	0	0	0	0
1512	0	0	0	0
1660	0	0	0	0
1822	0	0	0	0

## A3.2 Fluoride

### A3.2.1 Concentration

#### A3.2.1.1 Influence of electric current

Table A3.9 - Results obtained for the fluoride concentration over time with different electric currents

I / mA	Time / min	0	5	10	15	20	25	30	40	50	60
40	[F <sup>-</sup> ] / mg·L <sup>-1</sup>	16.3	14.8	13.8	13.3	12.2	10.8	10.3	8.3	8.2	7.5
	% of F <sup>-</sup> removal	0.0	9.2	15.3	18.4	25.2	33.7	36.8	49.1	49.7	54.0
100	[F <sup>-</sup> ] / mg·L <sup>-1</sup>	15.9	14.6	14.0	13.5	12.8	12.0	11.4	10.7	9.6	8.7
	% of F <sup>-</sup> removal	0.0	8.2	11.9	15.1	19.5	24.5	28.3	32.7	39.6	45.3
190	[F <sup>-</sup> ] / mg·L <sup>-1</sup>	16.3	14.8	13.8	13.3	12.2	10.8	10.3	8.3	7.5	6.1
	% of F <sup>-</sup> removal	0.0	9.2	15.3	18.4	25.2	33.7	36.8	49.1	54.0	62.6

#### A3.2.1.2 Influence of initial fluoride concentration

Table A3.10 - Results obtained for the fluoride concentration over time with different initial fluoride concentrations

[F <sup>-</sup> ] / mg·L <sup>-1</sup>	Time / min	0	5	10	15	20	25	30	40	50	60
5	[F <sup>-</sup> ] / mg·L <sup>-1</sup>	6.0	5.9	3.9	3.3	3.1	2.7	2.3	1.9	1.8	1.5
	% of F <sup>-</sup> removal	0.0	1.7	35.0	45.0	48.3	55.0	61.7	68.3	70.0	75.0
10	[F <sup>-</sup> ] / mg·L <sup>-1</sup>	10.4	9.5	8.5	7.8	7.2	6.4	5.5	5.0	4.5	3.7
	% of F <sup>-</sup> removal	0.00	8.7	18.3	25.0	30.8	38.5	47.1	51.9	56.7	64.4
15	[F <sup>-</sup> ] / mg·L <sup>-1</sup>	16.3	14.8	13.8	13.3	12.2	10.8	10.3	8.3	7.5	6.1
	% of F <sup>-</sup> removal	0.00	9.2	15.3	18.4	25.2	33.7	36.8	49.1	54.0	62.6

#### A3.2.1.3 Influence of initial water pH

Table A3.11 - Results obtained for the fluoride concentration over time with different initial water pH

Initial pH	Time / min	0	5	10	15	20	25	30	40	50	60
3	[F <sup>-</sup> ] / mg·L <sup>-1</sup>	16.9	14.3	13.0	11.5	8.4	6.9	4.7	3.6	2.5	1.8
	% of F <sup>-</sup> removal	0.0	15.4	23.1	32.0	50.3	59.2	72.2	78.7	85.2	89.3
6	[F <sup>-</sup> ] / mg·L <sup>-1</sup>	16.3	14.8	13.8	13.3	12.2	10.8	10.3	8.3	7.5	6.1
	% of F <sup>-</sup> removal	0.00	9.2	15.3	18.4	25.2	33.7	36.8	49.1	54.0	62.6
10	[F <sup>-</sup> ] / mg·L <sup>-1</sup>	18.4	16.2	14.0	13.1	13.3	12.0	11.1	9.6	8.0	7.1
	% of F <sup>-</sup> removal	0.0	12.0	23.9	28.8	27.7	34.8	39.7	47.8	56.5	61.4



## A3.2.1.4 Influence of electrodes arrangements

Table A3.12 - Results obtained for the fluoride concentration over time with different electrodes arrangements

Arrangement	Time / min	0	5	10	15	20	25	30	40	50	60
Simple	[F <sup>-</sup> ] / mg·L <sup>-1</sup>	16.3	14.8	13.8	13.3	12.2	10.8	10.3	8.3	7.5	6.1
	% of F <sup>-</sup> removal	0.00	9.2	15.3	18.4	25.2	33.7	36.8	49.1	54.0	62.6
Series monopolar	[F <sup>-</sup> ] / mg·L <sup>-1</sup>	15.4	12.6	12.4	11.6	10.5	9.3	8.7	7.3	5.7	4.5
	% of F <sup>-</sup> removal	0.0	18.2	19.5	24.7	31.8	39.6	43.5	52.6	63.0	70.8
Series bipolar	[F <sup>-</sup> ] / mg·L <sup>-1</sup>	14.7	13.6	12.0	10.8	9.2	8.7	7.5	5.9	5.2	4.7
	% of F <sup>-</sup> removal	0.0	7.5	18.4	26.5	37.4	40.8	49.0	59.9	64.6	68.0
Parallel	[F <sup>-</sup> ] / mg·L <sup>-1</sup>	15.0	14.5	13.9	12.6	11.7	10.9	10.3	9.0	8.0	6.9
	% of F <sup>-</sup> removal	0.0	3.3	7.3	16.0	22.0	27.3	31.3	40.0	46.7	54.0

### A3.2.2 Particles size

#### A3.2.2.1 Influence of electric current

Table A3.13 - Results obtained for the distribution of particle sizes with different electric currents

Particle diameter (µm)	Electric current (mA)				3.15	1.36	1.28
	40	100	190				
	Volume fraction (%)						
0.375	0.04	0	0	11.83			
0.412	0.073	0	0	12.99	2.91	1.36	1.34
0.452	0.12	0	0	14.26	3.15	1.36	1.4
0.496	0.15	0	0	15.65	3.8	1.36	1.45
0.545	0.18	0	0	17.18	4.78	1.38	1.52
0.598	0.19	0	0	18.86	5.33	1.44	1.61
0.657	0.19	0	0	20.7	3.84	1.55	1.74
0.721	0.19	0	0	22.73	1.19	1.72	1.91
0.791	0.19	0	0	24.95	0.078	1.97	2.16
0.869	0.18	0	0	27.38	0	2.29	2.47
0.953	0.18	0	0	30.07	0	2.66	2.87
1.047	0.18	0	0	33	0	3.08	3.35
1.149	0.19	0	0	36.24	0	3.52	3.89
1.261	0.21	0	0	39.77	0	3.98	4.47
1.385	0.22	0	0	43.66	0	4.45	5.06
1.52	0.22	0	0	47.93	0	4.92	5.61
1.669	0.22	0.00012	0.00006	52.63	0	5.38	6.06
1.832	0.2	0.002	0.001	57.77	0	5.78	6.35
2.01	0.18	0.012	0.0064	63.41	0	6.08	6.42
2.207	0.17	0.036	0.021	69.62	0	6.2	6.22
2.423	0.22	0.075	0.044	76.43	0	6.07	5.77
2.66	0.35	0.13	0.075	83.9	0	5.65	5.1
2.92	0.62	0.18	0.11	92.09	0	4.91	4.25
3.206	1.08	0.26	0.16	101.1	0	3.9	3.31
3.519	1.76	0.34	0.22	111	0	2.75	2.34
3.862	2.65	0.43	0.28	121.8	0	1.61	1.45
4.241	3.69	0.53	0.35	133.7	0	0.71	0.73
4.656	4.77	0.63	0.43	146.8	0	0.21	0.27
5.111	5.78	0.73	0.51	161.2	0	0.033	0.062
5.611	6.57	0.83	0.6	176.8	0	0.0021	0.0072
6.158	7.04	0.93	0.69	194.2	0	0	0.00025
6.761	7.13	1.03	0.78	213.2	0	0	0
7.421	6.83	1.11	0.87	234.1	0	0	0
8.147	6.21	1.19	0.96	256.8	0	0	0
8.944	5.39	1.26	1.05	282.1	0	0	0
9.819	4.5	1.31	1.13	309.6	0	0	0
10.78	3.73	1.34	1.21	339.8	0	0	0
				373.1	0	0	0
				409.6	0	0	0
				449.7	0	0	0
				493.6	0	0	0
				541.9	0	0	0
				594.9	0	0	0
				653	0	0	0
				716.9	0	0	0
				786.9	0	0	0

863.9	0	0	0
948.2	0	0	0
1041	0	0	0
1143	0	0	0
1255	0	0	0
1377	0	0	0
1512	0	0	0
1660	0	0	0
1822	0	0	0

### A3.2.2.2 Influence of initial fluoride concentration

Table A3.14 - Results obtained for the distribution of particle sizes with different initial fluoride concentrations

Particle diameter ( $\mu\text{m}$ )	Initial fluoride concentration ( $\text{mg}\cdot\text{L}^{-1}$ )			
	0	5	10	15
	Volume fraction (%)			
0.375	0	0	0	0
0.412	0	0	0	0
0.452	0	0	0	0
0.496	0	0	0	0
0.545	0	0	0	0
0.598	0	0	0	0
0.657	0	0	0	0
0.721	0	0	0	0
0.791	0	0	0	0
0.869	0	0	0	0
0.953	0	0	0	0
1.047	0	0	0	0
1.149	0	0	0	0
1.261	0	0	0	0
1.385	0	0	0	0
1.52	0	0	0	0
1.669	0	0	0	0.0001
1.832	0	0	0	0.001
2.01	0	0	0	0.0064
2.207	0	0	0	0.021
2.423	0	0	0.0002	0.044
2.66	0	0	0.0027	0.075
2.92	0.0008	0.0002	0.015	0.11
3.206	0.0051	0.0026	0.042	0.16
3.519	0.017	0.014	0.084	0.22
3.862	0.036	0.037	0.13	0.28
4.241	0.061	0.073	0.2	0.35

4.656	0.093	0.12	0.27	0.43
5.111	0.13	0.17	0.36	0.51
5.611	0.18	0.24	0.46	0.6
6.158	0.23	0.31	0.57	0.69
6.761	0.29	0.4	0.69	0.78
7.421	0.36	0.5	0.82	0.87
8.147	0.43	0.62	0.95	0.96
8.944	0.51	0.75	1.1	1.05
9.819	0.6	0.89	1.24	1.13
10.78	0.69	1.04	1.38	1.21
11.83	0.79	1.2	1.52	1.28
12.99	0.89	1.37	1.64	1.34
14.26	0.99	1.54	1.76	1.4
15.65	1.08	1.71	1.85	1.45
17.18	1.17	1.88	1.93	1.52
18.86	1.25	2.02	2	1.61
20.7	1.33	2.16	2.07	1.74
22.73	1.39	2.28	2.15	1.91
24.95	1.45	2.39	2.26	2.16
27.38	1.58	2.51	2.42	2.47
30.07	1.66	2.65	2.63	2.87
33	1.76	2.81	2.91	3.35
36.24	1.86	3.01	3.26	3.89
39.77	1.99	3.24	3.65	4.47
43.66	2.12	3.49	4.07	5.06
47.93	2.26	3.76	4.5	5.61
52.63	2.43	4.02	4.91	6.06
57.77	2.6	4.26	5.25	6.35
63.41	2.8	4.46	5.48	6.42
69.62	3.02	4.6	5.55	6.22
76.43	3.24	4.66	5.45	5.77
83.9	3.46	4.63	5.14	5.1
92.09	3.66	4.51	4.65	4.25
101.1	3.82	4.29	4.02	3.31
111	3.91	3.97	3.3	2.34
121.8	3.93	3.57	2.56	1.45
133.7	3.87	3.11	1.85	0.73
146.8	3.74	2.62	1.23	0.27
161.2	3.57	2.13	0.71	0.062
176.8	3.37	1.68	0.35	0.0072
194.2	3.16	1.28	0.13	0.0003
213.2	2.96	0.96	0.05	0
234.1	2.77	0.71	0.033	0
256.8	2.57	0.52	0.045	0
282.1	2.35	0.37	0.07	0
309.6	2.09	0.25	0.089	0

339.8	1.8	0.13	0.084	0
373.1	1.47	0.05	0.054	0
409.6	1.13	0.0094	0.023	0
449.7	0.83	0.0007	0.0045	0
493.6	0.58	0	0.0004	0
541.9	0.41	0	0	0
594.9	0.29	0	0	0
653	0.22	0	0	0
716.9	0.18	0	0	0
786.9	0.17	0	0	0
863.9	0.17	0	0	0
948.2	0.17	0	0	0
1041	0.15	0	0	0
1143	0.12	0	0	0
1255	0.084	0	0	0
1377	0.067	0	0	0
1512	0.056	0	0	0
1660	0.046	0	0	0
1822	0	0	0	0

2.207	0.089	0.021	0.019
2.423	0.093	0.044	0.039
2.66	0.1	0.075	0.065
2.92	0.11	0.11	0.099
3.206	0.12	0.16	0.14
3.519	0.14	0.22	0.19
3.862	0.15	0.28	0.25
4.241	0.17	0.35	0.31
4.656	0.19	0.43	0.39
5.111	0.21	0.51	0.46
5.611	0.23	0.6	0.55
6.158	0.27	0.69	0.64
6.761	0.31	0.78	0.73
7.421	0.36	0.87	0.83
8.147	0.44	0.96	0.93
8.944	0.53	1.05	1.03
9.819	0.64	1.13	1.13
10.78	0.77	1.21	1.24
11.83	0.9	1.28	1.34
12.99	1.03	1.34	1.45
14.26	1.14	1.4	1.56
15.65	1.25	1.45	1.68
17.18	1.39	1.52	1.82
18.86	1.57	1.61	1.98
20.7	1.79	1.74	2.17
22.73	2.07	1.91	2.4
24.95	2.36	2.16	2.67
27.38	2.64	2.47	2.98
30.07	2.86	2.87	3.32
33	2.99	3.35	3.67
36.24	3.1	3.89	4
39.77	3.29	4.47	4.3
43.66	3.61	5.06	4.54
47.93	4.01	5.61	4.7
52.63	4.29	6.06	4.76
57.77	4.34	6.35	4.73
63.41	4.23	6.42	4.62
69.62	4.17	6.22	4.44
76.43	4.4	5.77	4.21
83.9	4.82	5.1	3.95
92.09	5	4.25	3.65
101.1	4.7	3.31	3.31
111	4.28	2.34	2.93
121.8	4.05	1.45	2.5
133.7	3.74	0.73	2.05
146.8	2.94	0.27	1.61

### A3.2.2.3 Influence of initial water pH

Table A3.15 - Results obtained for the distribution of particle sizes with different initial water pH

Particle diameter (µm)	Initial water pH		
	3	6	10
	Volume fraction (%)		
0.375	0.012	0	0
0.412	0.024	0	0
0.452	0.04	0	0
0.496	0.054	0	0
0.545	0.067	0	0
0.598	0.079	0	0
0.657	0.089	0	0
0.721	0.097	0	0
0.791	0.1	0	0
0.869	0.11	0	0
0.953	0.11	0	0
1.047	0.11	0	0
1.149	0.11	0	0
1.261	0.1	0	0
1.385	0.097	0	0
1.52	0.093	0	0
1.669	0.089	0.00006	0.000067
1.832	0.087	0.001	0.001
2.01	0.087	0.0064	0.0061

161.2	1.84	0.062	1.21	0.953	0	0	0	0
176.8	1.19	0.0072	0.87	1.047	0	0	0	0
194.2	1.28	0.00025	0.6	1.149	0	0	0	0
213.2	1.46	0	0.39	1.261	0	0	0	0
234.1	0.71	0	0.25	1.385	0	0	0	0
256.8	0.064	0	0.15	1.52	0	0	0	0
282.1	0	0	0.089	1.669	0.00006	0	0	0
309.6	0	0	0.048	1.832	0.001	0	0	0
339.8	0	0	0.021	2.01	0.0064	0	0	0
373.1	0	0	0.0064	2.207	0.021	0.0003	0.0002	0
409.6	0	0	0.0011	2.423	0.044	0.0051	0.003	0.0001
449.7	0	0	0.000095	2.66	0.075	0.033	0.017	0.0015
493.6	0	0	0	2.92	0.11	0.11	0.047	0.0091
541.9	0	0	0	3.206	0.16	0.25	0.095	0.028
594.9	0	0	0	3.519	0.22	0.43	0.16	0.058
653	0	0	0	3.862	0.28	0.65	0.23	0.098
716.9	0	0	0	4.241	0.35	0.91	0.32	0.15
786.9	0	0	0	4.656	0.43	1.21	0.44	0.21
863.9	0	0	0	5.111	0.51	1.55	0.56	0.29
948.2	0	0	0	5.611	0.6	1.92	0.71	0.38
1041	0	0	0	6.158	0.69	2.3	0.86	0.48
1143	0	0	0	6.761	0.78	2.7	1.03	0.59
1255	0	0	0	7.421	0.87	3.09	1.21	0.72
1377	0	0	0	8.147	0.96	3.48	1.4	0.86
1512	0	0	0	8.944	1.05	3.84	1.59	1.01
1660	0	0	0	9.819	1.13	4.16	1.79	1.17
1822	0	0	0	10.78	1.21	4.4	1.98	1.35
				11.83	1.28	4.56	2.17	1.53
				12.99	1.34	4.6	2.34	1.71
				14.26	1.4	4.51	2.49	1.89
				15.65	1.45	4.29	2.61	2.05
				17.18	1.52	3.95	2.7	2.2
				18.86	1.61	3.53	2.77	2.32
				20.7	1.74	3.07	2.83	2.41
				22.73	1.91	2.63	2.89	2.48
				24.95	2.16	2.27	2.98	2.54
				21	2.47	2.01	3.12	2.6
				27.38	2.87	1.88	3.33	2.68
				30.07	3.35	1.84	3.6	2.8
				33	3.89	1.88	3.91	2.95
				36.24	4.47	1.94	4.23	3.15
				39.77	5.06	1.99	4.52	3.37
				43.66	5.61	2.01	4.73	3.61
				47.93	6.06	2.01	4.83	3.83
				52.63	6.35	2	4.79	4.03
				57.77	6.42	1.97	4.62	4.17

#### A3.2.2.4 Influence of electrodes arrangements

Table A3.16 - Results obtained for the distribution of particle sizes with different electrodes arrangements

Particle diameter (µm)	Electrodes arrangement			
	S	P	SM	SB
	Volume fraction (%)			
0.375	0	0	0	0
0.412	0	0	0	0
0.452	0	0	0	0
0.496	0	0	0	0
0.545	0	0	0	0
0.598	0	0	0	0
0.657	0	0	0	0
0.721	0	0	0	0
0.791	0	0	0	0
0.869	0	0	0	0

63.41	6.22	1.93	4.32	4.25
69.62	5.77	1.86	3.9	4.25
76.43	5.1	1.76	3.41	4.17
83.9	4.25	1.63	2.85	4
92.09	3.31	1.5	2.28	3.75
101.1	2.34	1.38	1.72	3.42
111	1.45	1.25	1.21	3.04
121.8	0.73	1.12	0.77	2.62
133.7	0.27	0.97	0.43	2.18
146.8	0.062	0.8	0.19	1.77
161.2	0.0072	0.63	0.067	1.42
176.8	0.00025	0.47	0.023	1.13
194.2	0	0.33	0.022	0.92
213.2	0	0.22	0.045	0.77
234.1	0	0.13	0.086	0.67
256.8	0	0.065	0.12	0.59
282.1	0	0.022	0.14	0.5
309.6	0	0.0039	0.14	0.39
339.8	0	0.0003	0.12	0.26
373.1	0	0	0.082	0.13
409.6	0	0	0.046	0.042
449.7	0	0	0.022	0.0073
493.6	0	0	0.011	0.0005
541.9	0	0	0.0097	0
594.9	0	0	0.013	0
653	0	0	0.018	0
716.9	0	0	0.016	0
786.9	0	0	0.009	0
863.9	0	0	0.0022	0
948.2	0	0	0.0002	0
1041	0	0	0	0
1143	0	0	0	0
1255	0	0	0	0
1377	0	0	0	0
1512	0	0	0	0
1660	0	0	0	0
1822	0	0	0	0

## A3.3 Arsenic (V)

### A3.3.1 Concentration

#### A3.3.1.1 Influence of electric current

Table A3.17 - Results obtained for the arsenic (V) concentration over time with different electric currents

I / mA	Time / min	0	10	20	30	40	50
40	[As(V)] / mg·L <sup>-1</sup>	2.95	1.65	0.90	0.15	0.00	0.00
	% of As(V) removal	0.0	44.1	69.5	94.9	100.0	100.0
100	[As(V)] / mg·L <sup>-1</sup>	2.93	1.15	0.60	0.05	0.00	0.00
	% of As(V) removal	0.0	60.8	79.5	98.3	100.0	100.0
190	[As(V)] / mg·L <sup>-1</sup>	2.98	0.80	0.35	0.00	0.00	0.00
	% of As(V) removal	0.0	73.2	88.3	100.0	100.0	100.0

#### A3.3.1.2 Influence of initial arsenic (V) concentration

Table A3.18 - Results obtained for the arsenic (V) concentration over time with different initial arsenic (V) concentrations

[As(V)] / mg·L <sup>-1</sup>	Time / min	0	10	20	30	40	50
2	[As(V)] / mg·L <sup>-1</sup>	1.95	0.50	0.10	0.00	0.00	0.00
	% of As(V) removal	0.0	74.4	94.9	100.0	100.0	100.0
3	[As(V)] / mg·L <sup>-1</sup>	2.98	0.80	0.35	0.00	0.00	0.00
	% of As(V) removal	0.0	73.2	88.3	100.0	100.0	100.0
4	[As(V)] / mg·L <sup>-1</sup>	3.7	0.65	0.15	0.00	0.00	0.00
	% of As(V) removal	0.0	82.4	95.9	100.0	100.0	100.0

#### A3.3.1.3 Influence of initial water pH

Table A3.19 - Results obtained for the arsenic (V) concentration over time with different initial water pH

Initial pH	Time / min	0	10	20	30	40	50
3	[As(V)] / mg·L <sup>-1</sup>	2.6	0.00	0.00	0.00	0.00	0.00
	% of As(V) removal	0.0	100.0	100.0	100.0	100.0	100.0
6	[As(V)] / mg·L <sup>-1</sup>	2.98	0.8	0.35	0.00	0.00	0.00
	% of As(V) removal	0.0	73.2	88.3	100.0	100.0	100.0
10	[As(V)] / mg·L <sup>-1</sup>	2.85	2.20	1.80	1.25	0.95	0.20
	% of As(V) removal	0.0	22.8	36.8	56.1	66.7	93.0

## A3.3.1.4 Influence of electrodes arrangements

Table A3.20 - Results obtained for the arsenic (V) concentration over time with different electrodes arrangements

Arrangement	Time / min	0	10	20	30	40	50
Simple	[As(V)] / mg·L <sup>-1</sup>	2.98	0.80	0.35	0.00	0.00	0.00
	% of As(V) removal	0.0	73.2	88.3	100.0	100.0	100.0
Parallel	[As(V)] / mg·L <sup>-1</sup>	2.95	0.00	0.00	0.00	0.00	0.00
	% of As(V) removal	0.0	100.0	100.0	100.0	100.0	100.0
Series monopolar	[As(V)] / mg·L <sup>-1</sup>	2.80	0.00	0.00	0.00	0.00	0.00
	% of As(V) removal	0.0	100.0	100.0	100.0	100.0	100.0
Series bipolar	[As(V)] / mg·L <sup>-1</sup>	2.60	0.00	0.00	0.00	0.00	0.00
	% of As(V) removal	0.0	100.0	100.0	100.0	100.0	100.0



### A3.3.2 Particles size

#### A3.3.2.1 Influence of electric current

Table A3.21 - Results obtained for the distribution of particle sizes with different electric currents

Particle diameter (µm)	Electric current (mA)				6.93	1.69	1.3
	40	100	190				
	Volume fraction (%)						
0.375	0	0	0	11.83			
0.412	0	0	0	12.99	8	1.91	1.52
0.452	0	0	0	14.26	8.73	2.11	1.76
0.496	0.00048	0	0	15.65	8.97	2.29	1.99
0.545	0.0063	0	0	17.18	8.67	2.41	2.22
0.598	0.019	0	0	18.86	7.94	2.48	2.42
0.657	0.029	0	0	20.7	6.97	2.49	2.57
0.721	0.036	0	0	22.73	5.94	2.43	2.66
0.791	0.039	0	0	24.95	4.9	2.34	2.68
0.869	0.038	0	0	27.38	3.86	2.23	2.62
0.953	0.035	0	0	30.07	2.78	2.14	2.51
1.047	0.031	0	0	33	1.66	2.1	2.35
1.149	0.028	0	0	36.24	0.63	2.11	2.17
1.261	0.026	0	0	39.77	0.11	2.18	2
1.385	0.028	0	0	43.66	0.044	2.32	1.85
1.52	0.035	0	0	47.93	0.19	2.51	1.74
1.669	0.046	0	0	52.63	0.5	2.73	1.65
1.832	0.062	0	0	57.77	0.62	2.96	1.6
2.01	0.08	0	0	63.41	0.3	3.19	1.57
2.207	0.099	0	0	69.62	0.026	3.41	1.58
2.423	0.12	0	0	76.43	0	3.61	1.62
2.66	0.13	0	0	83.9	0	3.78	1.69
2.92	0.13	0.00007	0.00008	92.09	0	3.9	1.78
3.206	0.13	0.0015	0.0012	101.1	0	3.97	1.87
3.519	0.13	0.0099	0.0077	111	0	3.97	1.94
3.862	0.14	0.034	0.024	121.8	0	3.88	1.98
4.241	0.16	0.078	0.053	133.7	0	3.68	1.97
4.656	0.21	0.14	0.091	146.8	0	3.38	1.95
5.111	0.32	0.21	0.14	161.2	0	2.99	1.92
5.611	0.49	0.3	0.2	176.8	0	2.55	1.91
6.158	0.77	0.42	0.28	194.2	0	2.11	1.93
6.761	1.16	0.55	0.37	213.2	0	1.72	1.98
7.421	1.71	0.69	0.47	234.1	0	1.4	2.05
8.147	2.44	0.86	0.6	256.8	0	1.14	2.15
8.944	3.37	1.05	0.74	282.1	0	0.93	2.27
9.819	4.47	1.25	0.9	309.6	0	0.74	2.39
10.78	5.7	1.47	1.09	339.8	0	0.55	2.5
				373.1	0	0.35	2.56
				409.6	0	0.17	2.55
				449.7	0	0.055	2.48
				493.6	0	0.0092	2.35
				541.9	0	0.00065	2.15
				594.9	0	0	1.91
				653	0	0	1.64
				716.9	0	0	1.36
				786.9	0	0	1.09

863.9	0	0	0.86
948.2	0	0	0.64
1041	0	0	0.44
1143	0	0	0.24
1255	0	0	0.093
1377	0	0	0.018
1512	0	0	0.0015
1660	0	0	0
1822	0	0	0

### A3.3.2.2 Influence of initial arsenic (V) concentration

Table A3.22 - Results obtained for the distribution of particle sizes with different initial arsenic (V) concentrations

Particle diameter (µm)	Initial fluoride concentration (mg·L <sup>-1</sup> )			
	0	2	3	4
	Volume fraction (%)			
0.375	0	0	0	0
0.412	0	0	0	0
0.452	0	0	0	0
0.496	0	0	0	0
0.545	0	0	0	0
0.598	0	0	0	0
0.657	0	0	0	0
0.721	0	0	0	0
0.791	0	0	0	0
0.869	0	0	0	0
0.953	0	0	0	0
1.047	0	0	0	0
1.149	0	0	0	0
1.261	0	0	0	0
1.385	0	0	0	0
1.52	0	0	0	0
1.669	0	0	0	0
1.832	0	0	0	0
2.01	0	0	0	0
2.207	0	0	0	0
2.423	0	0	0	0
2.66	0.00004	0.0001	0	0
2.92	0.0008	0.0017	0.00008	0.00012
3.206	0.0051	0.01	0.0012	0.0018
3.519	0.017	0.032	0.0077	0.011
3.862	0.036	0.068	0.024	0.033
4.241	0.061	0.12	0.053	0.069

4.656	0.093	0.18	0.091	0.12
5.111	0.13	0.25	0.14	0.18
5.611	0.18	0.34	0.2	0.25
6.158	0.23	0.44	0.28	0.35
6.761	0.29	0.55	0.37	0.46
7.421	0.36	0.68	0.47	0.58
8.147	0.43	0.82	0.6	0.73
8.944	0.51	0.97	0.74	0.9
9.819	0.6	1.13	0.9	1.09
10.78	0.69	1.3	1.09	1.31
11.83	0.79	1.47	1.3	1.54
12.99	0.89	1.64	1.52	1.79
14.26	0.99	1.79	1.76	2.05
15.65	1.08	1.92	1.99	2.31
17.18	1.17	2.03	2.22	2.54
18.86	1.25	2.1	2.42	2.75
20.7	1.33	2.14	2.57	2.91
22.73	1.39	2.15	2.66	3.01
24.95	1.45	2.14	2.68	3.05
27.38	1.58	2.13	2.62	3.05
30.07	1.66	2.14	2.51	3.01
33	1.76	2.17	2.35	2.96
36.24	1.86	2.23	2.17	2.93
39.77	1.99	2.32	2	2.92
43.66	2.12	2.42	1.85	2.94
47.93	2.26	2.53	1.74	2.99
52.63	2.43	2.64	1.65	3.06
57.77	2.6	2.77	1.6	3.14
63.41	2.8	2.92	1.57	3.22
69.62	3.02	3.09	1.58	3.3
76.43	3.24	3.27	1.62	3.36
83.9	3.46	3.46	1.69	3.4
92.09	3.66	3.63	1.78	3.4
101.1	3.82	3.78	1.87	3.36
111	3.91	3.87	1.94	3.26
121.8	3.93	3.87	1.98	3.1
133.7	3.87	3.78	1.97	2.88
146.8	3.74	3.59	1.95	2.62
161.2	3.57	3.32	1.92	2.33
176.8	3.37	3	1.91	2.04
194.2	3.16	2.65	1.93	1.76
213.2	2.96	2.32	1.98	1.51
234.1	2.77	2	2.05	1.31
256.8	2.57	1.71	2.15	1.13
282.1	2.35	1.42	2.27	0.96
309.6	2.09	1.12	2.39	0.79

339.8	1.8	0.81	2.5	0.6
373.1	1.47	0.49	2.56	0.39
409.6	1.13	0.22	2.55	0.2
449.7	0.83	0.065	2.48	0.066
493.6	0.58	0.01	2.35	0.012
541.9	0.41	0.00061	2.15	0.00078
594.9	0.29	0	1.91	0
653	0.22	0	1.64	0
716.9	0.18	0	1.36	0
786.9	0.17	0	1.09	0
863.9	0.17	0	0.86	0
948.2	0.17	0	0.64	0
1041	0.15	0	0.44	0
1143	0.12	0	0.24	0
1255	0.084	0	0.093	0
1377	0.067	0	0.018	0
1512	0.056	0	0.0015	0
1660	0.046	0	0	0
1822	0	0	0	0

2.207	0.027	0	0
2.423	0.027	0	0
2.66	0.028	0	0
2.92	0.029	0.00008	0.000018
3.206	0.03	0.0012	0.00038
3.519	0.033	0.0077	0.0026
3.862	0.036	0.024	0.0091
4.241	0.041	0.053	0.021
4.656	0.047	0.091	0.037
5.111	0.054	0.14	0.057
5.611	0.063	0.2	0.082
6.158	0.075	0.28	0.11
6.761	0.088	0.37	0.15
7.421	0.1	0.47	0.2
8.147	0.12	0.6	0.25
8.944	0.15	0.74	0.31
9.819	0.18	0.9	0.38
10.78	0.21	1.09	0.47
11.83	0.25	1.3	0.56
12.99	0.31	1.52	0.67
14.26	0.37	1.76	0.8
15.65	0.46	1.99	0.93
17.18	0.55	2.22	1.07
18.86	0.67	2.42	1.22
20.7	0.81	2.57	1.37
22.73	0.97	2.66	1.51
24.95	1.15	2.68	1.65
27.38	1.35	2.62	1.78
30.07	1.57	2.51	1.91
33	1.79	2.35	2.04
36.24	2.02	2.17	2.17
39.77	2.25	2	2.32
43.66	2.47	1.85	2.49
47.93	2.68	1.74	2.67
52.63	2.88	1.65	2.86
57.77	3.08	1.6	3.05
63.41	3.26	1.57	3.25
69.62	3.46	1.58	3.46
76.43	3.66	1.62	3.66
83.9	3.88	1.69	3.85
92.09	4.1	1.78	4.02
101.1	4.31	1.87	4.16
111	4.5	1.94	4.26
121.8	4.61	1.98	4.3
133.7	4.64	1.97	4.29
146.8	4.57	1.95	4.2

### A3.3.2.3 Influence of initial water pH

Table A3.23 - Results obtained for the distribution of particle sizes with different initial water pH

Particle diameter (µm)	Initial water pH		
	3	6	10
	Volume fraction (%)		
0.375	0.0027	0	0
0.412	0.0049	0	0
0.452	0.0073	0	0
0.496	0.01	0	0
0.545	0.013	0	0
0.598	0.015	0	0
0.657	0.018	0	0
0.721	0.02	0	0
0.791	0.022	0	0
0.869	0.023	0	0
0.953	0.025	0	0
1.047	0.026	0	0
1.149	0.026	0	0
1.261	0.027	0	0
1.385	0.027	0	0
1.52	0.027	0	0
1.669	0.027	0	0
1.832	0.027	0	0
2.01	0.027	0	0

161.2	4.39	1.92	4.05	0.953	0	0	0	0
176.8	4.13	1.91	3.84	1.047	0	0	0	0
194.2	3.81	1.93	3.58	1.149	0	0	0	0
213.2	3.46	1.98	3.28	1.261	0	0	0	0
234.1	3.09	2.05	2.93	1.385	0	0	0	0
256.8	2.72	2.15	2.56	1.52	0	0	0	0
282.1	2.34	2.27	2.17	1.669	0	0	0	0
309.6	1.95	2.39	1.76	1.832	0	0	0	0
339.8	1.55	2.5	1.35	2.01	0	0	0	0
373.1	1.14	2.56	0.95	2.207	0	0	0.000001	0
409.6	0.77	2.55	0.57	2.423	0	0	0.00017	0
449.7	0.46	2.48	0.27	2.66	0	0.00033	0.002	0
493.6	0.27	2.35	0.084	2.92	0.00008	0.004	0.01	0.000057
541.9	0.17	2.15	0.014	3.206	0.0012	0.02	0.027	0.00079
594.9	0.14	1.91	0.00097	3.519	0.0077	0.054	0.053	0.0044
653	0.15	1.64	0	3.862	0.024	0.1	0.086	0.013
716.9	0.16	1.36	0	4.241	0.053	0.16	0.13	0.026
786.9	0.17	1.09	0	4.656	0.091	0.24	0.18	0.042
863.9	0.17	0.86	0	5.111	0.14	0.34	0.24	0.063
948.2	0.17	0.64	0	5.611	0.2	0.45	0.31	0.09
1041	0.18	0.44	0	6.158	0.28	0.58	0.4	0.12
1143	0.16	0.24	0	6.761	0.37	0.73	0.49	0.16
1255	0.097	0.093	0	7.421	0.47	0.9	0.59	0.2
1377	0.026	0.018	0	8.147	0.6	1.09	0.71	0.26
1512	0.0031	0.0015	0	8.944	0.74	1.31	0.83	0.32
1660	0	0	0	9.819	0.9	1.54	0.97	0.39
1822	0	0	0	10.78	1.09	1.8	1.11	0.47
				11.83	1.3	2.07	1.27	0.57
				12.99	1.52	2.36	1.42	0.68
				14.26	1.76	2.65	1.58	0.8
				15.65	1.99	2.94	1.73	0.93
				17.18	2.22	3.2	1.86	1.07
				18.86	2.42	3.42	1.98	1.22
				20.7	2.57	3.6	2.09	1.38
				22.73	2.66	3.72	2.17	1.53
				24.95	2.68	3.77	2.24	1.68
				21	2.62	3.74	2.31	1.81
				27.38	2.51	3.66	2.39	1.94
				30.07	2.35	3.53	2.48	2.05
				33	2.17	3.38	2.59	2.16
				36.24	2	3.23	2.72	2.26
				39.77	1.85	3.09	2.87	2.36
				43.66	1.74	2.98	3.04	2.46
				47.93	1.65	2.9	3.22	2.57
				52.63	1.6	2.85	3.39	2.69
				57.77	1.57	2.83	3.56	2.82

#### A3.3.2.4 Influence of electrodes arrangements

Table A3.24 - Results obtained for the distribution of particle sizes with different electrodes arrangements

Particle diameter (µm)	Electrodes arrangement			
	S	P	SM	SB
	Volume fraction (%)			
0.375	0	0	0	0
0.412	0	0	0	0
0.452	0	0	0	0
0.496	0	0	0	0
0.545	0	0	0	0
0.598	0	0	0	0
0.657	0	0	0	0
0.721	0	0	0	0
0.791	0	0	0	0
0.869	0	0	0	0

63.41	1.58	2.81	3.71	2.96
69.62	1.62	2.78	3.82	3.11
76.43	1.69	2.73	3.9	3.26
83.9	1.78	2.63	3.92	3.4
92.09	1.87	2.5	3.88	3.52
101.1	1.94	2.33	3.78	3.6
111	1.98	2.12	3.59	3.62
121.8	1.97	1.9	3.35	3.59
133.7	1.95	1.67	3.05	3.49
146.8	1.92	1.46	2.72	3.36
161.2	1.91	1.28	2.38	3.19
176.8	1.93	1.13	2.06	3.01
194.2	1.98	1.01	1.78	2.84
213.2	2.05	0.91	1.54	2.67
234.1	2.15	0.83	1.35	2.5
256.8	2.27	0.76	1.18	2.32
282.1	2.39	0.67	1.02	2.12
309.6	2.5	0.54	0.82	1.89
339.8	2.56	0.38	0.59	1.64
373.1	2.55	0.2	0.34	1.38
409.6	2.48	0.073	0.14	1.13
449.7	2.35	0.013	0.033	0.91
493.6	2.15	0.0009	0.0037	0.74
541.9	1.91	0	0.0001	0.62
594.9	1.64	0	0	0.54
653	1.36	0	0	0.48
716.9	1.09	0	0	0.45
786.9	0.86	0	0	0.41
863.9	0.64	0	0	0.38
948.2	0.44	0	0	0.34
1041	0.24	0	0	0.31
1143	0.093	0	0	0.28
1255	0.018	0	0	0.25
1377	0.0015	0	0	0.22
1512	0	0	0	0.18
1660	0	0	0	0.14
1822	0	0	0	0

## A3.4 Binary mixtures

### A3.4.1 Concentration

#### A3.4.1.1 Influence of fluoride presence on the removal of iron

Table A3.25 - Results obtained for the iron concentration over time with different initial fluoride concentrations

[F <sup>-</sup> ] / mg·L <sup>-1</sup>	Time / min	0	5	10	15	20	25	30	40	50	60	80
0	[Fe] / mg·L <sup>-1</sup>	25.20	14.97	13.78	11.78	8.78	7.49	5.71	5.43	2.76	1.61	
	% of Fe removal	0.0	40.6	45.3	53.3	65.2	70.3	77.3	78.5	89.0	93.6	
5	[Fe] / mg·L <sup>-1</sup>	25.10	14.16	12.22		7.85			6.50		2.35	1.28
	% of Fe removal	0.0	43.6	51.3		68.7			74.1		90.6	94.9
	[F <sup>-</sup> ] / mg·L <sup>-1</sup>	5.1	3.9	2.6		0.0			0.0		0.0	0.0
	% of F <sup>-</sup> removal	0.0	24.0	49.3		100.0			100.0		100.0	100.0
10	[Fe] / mg·L <sup>-1</sup>	24.87	13.82	11.49		7.45			1.81		0.28	0.00
	% of Fe removal	0.0	44.4	53.8		70.0			92.7		98.9	100.0
	[F <sup>-</sup> ] / mg·L <sup>-1</sup>	10.0	7.6	5.6		1.2			0.0		0.0	0.0
	% of F <sup>-</sup> removal	0.0	24.0	44.0		88.0			100.0		100.0	100.0
15	[Fe] / mg·L <sup>-1</sup>	23.93	12.5	8.49		4.66			2.59		1.14	0.00
	% of Fe removal	0.0	47.8	64.5		80.5			89.2		95.2	100.0
	[F <sup>-</sup> ] / mg·L <sup>-1</sup>	16.8	12.3	11.7		9.7			7.6		5.3	3.6
	% of F <sup>-</sup> removal	0.0	26.8	30.4		42.3			54.8		68.5	78.6

## A3.4.1.2 Influence of iron presence on the removal of fluoride

Table A3.26 - Results obtained for the fluoride concentration over time with different initial iron concentrations

[Fe] / mg·L <sup>-1</sup>	Time / min	0	5	10	15	20	25	30	40	50	60	80
0	[F] / mg·L <sup>-1</sup>	16.3	14.8	13.8	13.3	12.2	10.8	10.3	8.3	7.5	6.1	
	% of F removal	0.0	9.2	15.3	18.4	25.2	33.7	36.8	49.1	54.0	62.6	
5	[F] / mg·L <sup>-1</sup>	14.5	12.9	10.4		9.0			7.9		5.8	3.5
	% of F removal	0.0	11.0	28.3		37.9			45.5		60.0	75.9
	[Fe] / mg·L <sup>-1</sup>	6.84	1.20	0.65		0.00			0.00		0.00	0.00
	% of Fe removal	0.0	82.5	90.5		100.0			100.0		100.0	100.0
10	[F] / mg·L <sup>-1</sup>	14.1	11.6	10.1		9.3			6.1		5.5	3.1
	% of F removal	0.0	17.7	28.4		34.9			56.7		61.0	78.0
	[Fe] / mg·L <sup>-1</sup>	10.72	2.84	1.68		0.52			0.00		0.00	0.00
	% of Fe removal	0.0	73.5	84.3		95.1			100.0		100.0	100.0
15	[F] / mg·L <sup>-1</sup>	16.2	13.4	12.5		9.5			6.8		5.4	3.2
	% of F removal	0.0	17.3	22.8		41.4			58.0		66.7	80.2
	[Fe] / mg·L <sup>-1</sup>	16.59	6.50	3.18		0.97			0.00		0.00	0.00
	% of Fe removal	0.0	61.1	80.9		94.2			100.0		100.0	100.0
25	[F] / mg·L <sup>-1</sup>	16.8	12.3	11.7		9.7			7.6		5.3	3.6
	% of F removal	0.0	26.8	30.4		42.3			54.8		68.5	78.6
	[Fe] / mg·L <sup>-1</sup>	23.95	12.50	8.49		4.66			2.59		1.14	0.00
	% of Fe removal	0.0	47.8	64.5		80.5			89.2		95.2	100.0

## A3.4.1.3 Influence of arsenic (V) presence on the removal of iron

Table A3.27 - Results obtained for the iron concentration over time with different initial arsenic (V) concentrations

[As(V)] / mg·L <sup>-1</sup>	Time / min	0	5	10	15	20	25	30	40	50
0	[Fe] / mg·L <sup>-1</sup>	25.2	14.97	13.78	11.78	8.78	7.49	5.71	5.43	2.76
	% of Fe removal	0.0	40.6	45.3	53.3	65.2	70.3	77.3	78.5	89.0
2	[Fe] / mg·L <sup>-1</sup>	24.98	12.80	12.31	10.69	9.03		5.32	3.5	0.0
	% of Fe removal	0.0	48.8	50.7	57.2	63.9		78.7	85.9	0.0
	[As(V)] / mg·L <sup>-1</sup>	2.00		0.00		0.00		0.00	0.00	0.00
	% of As(V) removal	0.0		100.0		100.0		100.0	100.0	100.0
4	[Fe] / mg·L <sup>-1</sup>	24.47	10.50	8.96	8.62	7.65		3.37	3.10	0.00
	% of Fe removal	0.0	57.1	63.4	64.8	68.7		86.2	87.3	0.0
	[As(V)] / mg·L <sup>-1</sup>	3.90		0.00		0.00		0.00	0.00	0.00
	% of As(V) removal	0.0		100.0		100.0		100.0	100.0	100.0

## A3.4.1.4 Influence of iron presence on the removal of arsenic (V)

Table A3.28 - Results obtained for the arsenic (V) concentration over time with different initial iron concentrations

[Fe] / mg·L <sup>-1</sup>	Time / min	0	5	10	15	20	30	40	50
0	[As(V)] / mg·L <sup>-1</sup>	3.70		0.65		0.15	0.0	0.0	0.0
	% of As(V) removal	0.0		82.4		95.9	100.0	100.0	100.0
15	[As(V)] / mg·L <sup>-1</sup>	4.0		0.0		0.0	0.0	0.0	0.0
	% of As(V) removal	0.0		100.0		100.0	100.0	100.0	100.0
	[Fe] / mg·L <sup>-1</sup>	15.22	8.37	7.57	5.19	3.86	2.20	2.00	1.80
	% of Fe removal	0.0	45.0	50.3	65.9	74.6	85.5	86.9	88.4
25	[As(V)] / mg·L <sup>-1</sup>	3.9		0.0		0.0	0.0	0.0	0.0
	% of As(V) removal	0.0		100.0		100.0	100.0	100.0	100.0
	[Fe] / mg·L <sup>-1</sup>	24.47	10.50	8.96	8.62	7.65	5.96	3.37	3.10
	% of Fe removal	0.0	57.1	63.4	64.8	68.7	75.6	86.2	87.3



## A3.4.1.5 Influence of arsenic (V) presence on the removal of fluoride

Table A3.29 - Results obtained for the fluoride concentration over time with different initial arsenic (V) concentrations

[As(V)] / mg·L <sup>-1</sup>	Time / min	0	5	10	15	20	30	40	50
0	[F <sup>-</sup> ] / mg·L <sup>-1</sup>	16.3	14.8	13.8	13.3	12.2	10.8	10.3	8.3
	% of F <sup>-</sup> removal	0.0	9.2	15.3	18.4	25.2	33.7	36.8	49.1
2	[F <sup>-</sup> ] / mg·L <sup>-1</sup>	14.9	13.8	12.2	11.1	10.8	9.4	8.8	7.3
	% of F <sup>-</sup> removal	0.0	7.4	18.1	25.5	27.5	36.9	40.9	51.0
	[As(V)] / mg·L <sup>-1</sup>	1.85		0.00		0.00	0.00	0.00	0.00
	% of As(V) removal	0.0		100.0		100.0	100.0	100.0	100.0
4	[F <sup>-</sup> ] / mg·L <sup>-1</sup>	16.1	13.9	13.0	12.4	11.6	10.7	7.9	7.7
	% of F <sup>-</sup> removal	0.0	13.7	19.3	23.0	28.0	33.5	50.9	52.2
	[As(V)] / mg·L <sup>-1</sup>	3.59		2.39		1.32	0.00	0.00	0.00
	% of As(V) removal	0.0		33.4		63.2	100.0	100.0	100.0

## A3.4.1.6 Influence of fluoride presence on the removal of arsenic (V)

Table A3.30 - Results obtained for the arsenic (V) concentration over time with different initial fluoride concentrations

[F <sup>-</sup> ] / mg·L <sup>-1</sup>	Time / min	0	5	10	15	20	30	40	50
0	[As(V)] / mg·L <sup>-1</sup>	3.70		0.65		0.15	0.0	0.0	0.0
	% of As(V) removal	0.0		82.4		95.9	100.0	100.0	100.0
5	[As(V)] / mg·L <sup>-1</sup>	3.95		1.64		0.35	0.0	0.0	0.0
	% of As(V) removal	0.0		58.5		91.1	100.0	100.0	100.0
	[F <sup>-</sup> ] / mg·L <sup>-1</sup>	4.8	3.3	3.1	2.9	1.9	1.2	0.0	0.0
	% of F <sup>-</sup> removal	0.0	31.3	35.4	39.6	60.4	75.0	100.0	100.0
15	[As(V)] / mg·L <sup>-1</sup>	3.59		2.39		1.32	0.0	0.0	0.0
	% of As(V) removal	0.0		33.4		63.2	100.0	100.0	100.0
	[F <sup>-</sup> ] / mg·L <sup>-1</sup>	16.1	13.9	13.0	12.4	11.6	10.7	7.9	7.7
	% of F <sup>-</sup> removal	0.0	13.7	19.3	23.0	28.0	33.5	50.9	52.2

### A3.4.2 Particles size

#### A3.4.2.1 Influence of fluoride presence on the removal of iron

Table A3.31 - Results obtained for the distribution of particle sizes with a binary mixture (iron + fluoride)

Particle diameter (µm)	Initial fluoride concentration (mg·L <sup>-1</sup> )						
	0	10	15				
	Volume fraction (%)						
0.375	0	0	0	9.819	2.47	1.48	2.46
0.412	0	0	0	10.78	2.64	1.55	2.52
0.452	0	0	0	11.83	2.76	1.6	2.53
0.496	0	0	0	12.99	2.84	1.66	2.51
0.545	0	0	0	14.26	2.87	1.7	2.47
0.598	0	0	0	15.65	2.85	1.76	2.43
0.657	0	0	0	17.18	2.81	1.84	2.4
0.721	0	0	0	18.86	2.75	1.94	2.4
0.791	0	0	0	20.7	2.71	2.1	2.46
0.869	0	0	0	22.73	2.72	2.3	2.58
0.953	0	0	0	24.95	2.78	2.55	2.74
1.047	0	0	0	27.38	3.13	2.84	2.92
1.149	0	0	0	30.07	3.37	3.15	3.11
1.261	0	0	0	33	3.62	3.45	3.27
1.385	0	0	0	36.24	3.84	3.72	3.38
1.52	0	0	0	39.77	4	3.94	3.42
1.669	0	0.00019	0.00025	43.66	4.08	4.1	3.4
1.832	0	0.0026	0.0036	47.93	4.08	4.19	3.31
2.01	0.00008	0.014	0.021	52.63	3.98	4.21	3.17
2.207	0.0016	0.04	0.062	57.77	3.8	4.16	2.99
2.423	0.012	0.081	0.13	63.41	3.55	4.06	2.81
2.66	0.045	0.13	0.21	69.62	3.22	3.91	2.66
2.92	0.11	0.19	0.32	76.43	2.85	3.73	2.52
3.206	0.2	0.26	0.44	83.9	2.45	3.51	2.41
3.519	0.3	0.35	0.59	92.09	2.04	3.27	2.3
3.862	0.43	0.44	0.76	101.1	1.62	3.01	2.17
4.241	0.59	0.55	0.95	111	1.22	2.71	2
4.656	0.77	0.65	1.14	121.8	0.84	2.38	1.78
5.111	0.96	0.77	1.34	133.7	0.5	2.02	1.52
5.611	1.17	0.88	1.54	146.8	0.26	1.64	1.22
6.158	1.39	0.99	1.74	161.2	0.11	1.26	0.93
6.761	1.62	1.1	1.92	176.8	0.055	0.93	0.69
7.421	1.85	1.21	2.09	194.2	0.036	0.66	0.52
8.147	2.07	1.31	2.25	213.2	0.038	0.48	0.42
8.944	2.28	1.4	2.37	234.1	0.05	0.39	0.38
				256.8	0.064	0.35	0.35
				282.1	0.072	0.33	0.32
				309.6	0.068	0.3	0.27
				339.8	0.051	0.23	0.2
				373.1	0.028	0.13	0.12
				409.6	0.0097	0.048	0.055
				449.7	0.0017	0.0088	0.012
				493.6	0.0001	0.0006	0.0012
				541.9	0	0	0.000003
				594.9	0	0	0
				653	0	0	0

716.9	0	0	0
786.9	0	0	0
863.9	0	0	0
948.2	0	0	0
1041	0	0	0
1143	0	0	0
1255	0	0	0
1377	0	0	0
1512	0	0	0
1660	0	0	0
1822	0	0	0

#### A3.4.2.2 Influence of iron presence on the removal of fluoride

Table A3.32 - Results obtained for the distribution of particle sizes with a binary mixture (fluoride + iron)

Particle diameter (µm)	Initial iron concentration (mg·L <sup>-1</sup> )				
	0	5	10	15	25
	Volume fraction (%)				
0.375	0	0	0	0	0
0.412	0	0	0	0	0
0.452	0	0	0	0	0
0.496	0	0	0	0	0
0.545	0	0	0	0	0
0.598	0	0	0	0	0
0.657	0	0	0	0	0
0.721	0	0	0	0	0
0.791	0	0	0	0	0
0.869	0	0	0	0	0
0.953	0	0	0	0	0
1.047	0	0	0	0	0
1.149	0	0	0	0	0
1.261	0	0	0	0	0
1.385	0	0	0	0	0
1.52	0	0	0	0	0
1.669	0.0001	0	0	0	0.00025
1.832	0.001	0	0	0	0.0036
2.01	0.0064	0	0	0.0002	0.021
2.207	0.021	0.0004	0.0006	0.0025	0.062
2.423	0.044	0.0044	0.0084	0.014	0.13
2.66	0.075	0.022	0.046	0.042	0.21
2.92	0.11	0.059	0.13	0.086	0.32
3.206	0.16	0.12	0.25	0.14	0.44
3.519	0.22	0.18	0.4	0.21	0.59

3.862	0.28	0.27	0.58	0.3	0.76
4.241	0.35	0.37	0.79	0.39	0.95
4.656	0.43	0.48	1.04	0.51	1.14
5.111	0.51	0.61	1.31	0.63	1.34
5.611	0.6	0.76	1.59	0.76	1.54
6.158	0.69	0.9	1.89	0.9	1.74
6.761	0.78	1.06	2.19	1.04	1.92
7.421	0.87	1.22	2.49	1.19	2.09
8.147	0.96	1.38	2.79	1.33	2.25
8.944	1.05	1.54	3.06	1.48	2.37
9.819	1.13	1.69	3.29	1.61	2.46
10.78	1.21	1.83	3.48	1.74	2.52
11.83	1.28	1.95	3.61	1.85	2.53
12.99	1.34	2.04	3.66	1.95	2.51
14.26	1.4	2.11	3.63	2.04	2.47
15.65	1.45	2.16	3.53	2.12	2.43
17.18	1.52	2.18	3.37	2.2	2.4
18.86	1.61	2.2	3.17	2.3	2.4
20.7	1.74	2.23	2.97	2.42	2.46
22.73	1.91	2.28	2.79	2.57	2.58
24.95	2.16	2.37	2.66	2.77	2.74
27.38	2.47	2.49	2.6	3	2.92
30.07	2.87	2.65	2.58	3.26	3.11
33	3.35	2.83	2.61	3.51	3.27
36.24	3.89	3	2.64	3.75	3.38
39.77	4.47	3.14	2.64	3.95	3.42
43.66	5.06	3.23	2.6	4.08	3.4
47.93	5.61	3.26	2.51	4.14	3.31
52.63	6.06	3.24	2.37	4.14	3.17
57.77	6.35	3.19	2.22	4.07	2.99
63.41	6.42	3.13	2.06	3.96	2.81
69.62	6.22	3.06	1.91	3.8	2.66
76.43	5.77	3.01	1.79	3.61	2.52
83.9	5.1	2.95	1.69	3.39	2.41
92.09	4.25	2.87	1.62	3.13	2.3
101.1	3.31	2.77	1.57	2.84	2.17
111	2.34	2.64	1.53	2.52	2
121.8	1.45	2.47	1.47	2.17	1.78
133.7	0.73	2.27	1.35	1.8	1.52
146.8	0.27	2.04	1.18	1.44	1.22
161.2	0.062	1.79	0.99	1.1	0.93
176.8	0.0072	1.55	0.81	0.81	0.69
194.2	0.0003	1.35	0.7	0.59	0.52
213.2	0	1.21	0.65	0.46	0.42
234.1	0	1.12	0.63	0.39	0.38
256.8	0	1.08	0.62	0.36	0.35

282.1	0	1.05	0.57	0.35	0.32
309.6	0	0.99	0.48	0.32	0.27
339.8	0	0.87	0.37	0.25	0.2
373.1	0	0.69	0.27	0.15	0.12
409.6	0	0.47	0.17	0.058	0.055
449.7	0	0.27	0.081	0.011	0.012
493.6	0	0.14	0.025	0.0009	0.0012
541.9	0	0.076	0.0037	0	0
594.9	0	0.055	0.0002	0	0
653	0	0.059	0	0	0
716.9	0	0.075	0	0	0
786.9	0	0.1	0	0	0
863.9	0	0.13	0	0	0
948.2	0	0.17	0	0	0
1041	0	0.2	0	0	0
1143	0	0.18	0	0	0
1255	0	0.1	0	0	0
1377	0	0.027	0	0	0
1512	0	0.003	0	0	0
1660	0	0	0	0	0
1822	0	0	0	0	0

#### A3.4.2.3 Influence of arsenic (V) presence on the removal of iron

Table A3.33 - Results obtained for the distribution of particle sizes with a binary mixture (iron + arsenic (V))

Particle diameter ( $\mu\text{m}$ )	Initial arsenic (V) concentration ( $\text{mg}\cdot\text{L}^{-1}$ )		
	0	2	4
	Volume fraction (%)		
0.375	0	0	0
0.412	0	0	0
0.452	0	0	0
0.496	0	0	0
0.545	0	0	0
0.598	0	0	0
0.657	0	0	0
0.721	0	0	0
0.791	0	0	0
0.869	0	0	0
0.953	0	0	0
1.047	0	0	0
1.149	0	0	0
1.261	0	0	0
1.385	0	0	0

1.52	0	0	0.00007
1.669	0	0.00011	0.0014
1.832	0	0.0016	0.0099
2.01	0.00008	0.0091	0.035
2.207	0.0016	0.026	0.079
2.423	0.012	0.054	0.14
2.66	0.045	0.088	0.21
2.92	0.11	0.13	0.3
3.206	0.2	0.18	0.41
3.519	0.3	0.25	0.53
3.862	0.43	0.32	0.67
4.241	0.59	0.4	0.82
4.656	0.77	0.48	0.97
5.111	0.96	0.57	1.13
5.611	1.17	0.67	1.3
6.158	1.39	0.77	1.46
6.761	1.62	0.87	1.62
7.421	1.85	0.98	1.77
8.147	2.07	1.08	1.92
8.944	2.28	1.19	2.06
9.819	2.47	1.29	2.18
10.78	2.64	1.39	2.29
11.83	2.76	1.49	2.38
12.99	2.84	1.58	2.45
14.26	2.87	1.68	2.51
15.65	2.85	1.78	2.57
17.18	2.81	1.9	2.64
18.86	2.75	2.04	2.73
20.7	2.71	2.22	2.84
22.73	2.72	2.44	2.98
24.95	2.78	2.7	3.15
27.38	3.13	3.01	3.32
30.07	3.37	3.35	3.49
33	3.62	3.72	3.63
36.24	3.84	4.07	3.72
39.77	4	4.4	3.76
43.66	4.08	4.68	3.72
47.93	4.08	4.89	3.61
52.63	3.98	5	3.43
57.77	3.8	5.01	3.18
63.41	3.55	4.9	2.87
69.62	3.22	4.67	2.53
76.43	2.85	4.31	2.18
83.9	2.45	3.85	1.86
92.09	2.04	3.33	1.6
101.1	1.62	2.77	1.42

111	1.22	2.21	1.3
121.8	0.84	1.68	1.21
133.7	0.5	1.21	1.11
146.8	0.26	0.83	0.99
161.2	0.11	0.54	0.86
176.8	0.055	0.36	0.76
194.2	0.036	0.27	0.72
213.2	0.038	0.25	0.72
234.1	0.05	0.27	0.73
256.8	0.064	0.31	0.73
282.1	0.072	0.36	0.67
309.6	0.068	0.37	0.57
339.8	0.051	0.33	0.45
373.1	0.028	0.25	0.34
409.6	0.0097	0.14	0.23
449.7	0.0017	0.054	0.12
493.6	0.0001	0.01	0.042
541.9	0	0.00075	0.0073
594.9	0	0	0.00046
653	0	0	0
716.9	0	0	0
786.9	0	0	0
863.9	0	0	0
948.2	0	0	0
1041	0	0	0
1143	0	0	0
1255	0	0	0
1377	0	0	0
1512	0	0	0
1660	0	0	0
1822	0	0	0

#### A3.4.2.4 Influence of iron presence on the removal of arsenic (V)

Table A3.34 - Results obtained for the distribution of particle sizes with a binary mixture (arsenic (V) + iron)

Particle diameter (µm)	Initial iron concentration (mg·L <sup>-1</sup> )		
	0	15	25
	Volume fraction (%)		
0.375	0	0	0
0.412	0	0	0
0.452	0	0	0
0.496	0	0	0
0.545	0	0	0

0.598	0	0	0
0.657	0	0	0
0.721	0	0	0
0.791	0	0	0
0.869	0	0	0
0.953	0	0	0
1.047	0	0	0
1.149	0	0	0
1.261	0	0	0
1.385	0	0	0
1.52	0	0	0
1.669	0	0	0
1.832	0	0	0
2.01	0	0	0.000033
2.207	0	0.00045	0.00085
2.423	0	0.0059	0.0068
2.66	0	0.032	0.027
2.92	0.00012	0.088	0.068
3.206	0.0018	0.17	0.13
3.519	0.011	0.28	0.2
3.862	0.033	0.4	0.29
4.241	0.069	0.55	0.4
4.656	0.12	0.72	0.53
5.111	0.18	0.91	0.68
5.611	0.25	1.12	0.84
6.158	0.35	1.33	1.02
6.761	0.46	1.56	1.2
7.421	0.58	1.78	1.4
8.147	0.73	2	1.6
8.944	0.9	2.2	1.81
9.819	1.09	2.38	2.01
10.78	1.31	2.53	2.2
11.83	1.54	2.64	2.38
12.99	1.79	2.7	2.53
14.26	2.05	2.69	2.66
15.65	2.31	2.64	2.74
17.18	2.54	2.54	2.8
18.86	2.75	2.42	2.84
20.7	2.91	2.31	2.86
22.73	3.01	2.23	2.89
24.95	3.05	2.2	2.94
27.38	3.05	2.24	3.02
30.07	3.01	2.35	3.13
33	2.96	2.51	3.26
36.24	2.93	2.7	3.37
39.77	2.92	2.88	3.45

43.66	2.94	3.02	3.48
47.93	2.99	3.12	3.45
52.63	3.06	3.17	3.37
57.77	3.14	3.18	3.25
63.41	3.22	3.16	3.09
69.62	3.3	3.12	2.91
76.43	3.36	3.07	2.71
83.9	3.4	2.99	2.49
92.09	3.4	2.89	2.24
101.1	3.36	2.76	1.99
111	3.26	2.57	1.75
121.8	3.1	2.34	1.53
133.7	2.88	2.06	1.34
146.8	2.62	1.75	1.15
161.2	2.33	1.43	0.98
176.8	2.04	1.14	0.82
194.2	1.76	0.9	0.72
213.2	1.51	0.72	0.67
234.1	1.31	0.62	0.69
256.8	1.13	0.57	0.77
282.1	0.96	0.56	0.87
309.6	0.79	0.54	0.94
339.8	0.6	0.49	0.94
373.1	0.39	0.39	0.86
409.6	0.2	0.23	0.7
449.7	0.066	0.093	0.5
493.6	0.012	0.018	0.29
541.9	0.00078	0.0015	0.13
594.9	0	0	0.041
653	0	0	0.0068
716.9	0	0	0.00048
786.9	0	0	0
863.9	0	0	0
948.2	0	0	0
1041	0	0	0
1143	0	0	0
1255	0	0	0
1377	0	0	0
1512	0	0	0
1660	0	0	0
1822	0	0	0

#### A3.4.2.5 Influence of arsenic (V) presence on the removal of fluoride

Table A3.35 - Results obtained for the distribution of particle sizes with a binary mixture (fluoride + arsenic (V))

Particle diameter ( $\mu\text{m}$ )	Initial arsenic (V) concentration ( $\text{mg}\cdot\text{L}^{-1}$ )		
	0	2	4
	Volume fraction (%)		
0.375	0	0	0
0.412	0	0	0
0.452	0	0	0
0.496	0	0	0
0.545	0	0	0
0.598	0	0	0
0.657	0	0	0
0.721	0	0	0
0.791	0	0	0
0.869	0	0	0
0.953	0	0	0
1.047	0	0	0
1.149	0	0	0
1.261	0	0	0
1.385	0	0	0
1.52	0	0	0.00007
1.669	0.0001	0.00011	0.0014
1.832	0.001	0.0016	0.0099
2.01	0.0064	0.0091	0.035
2.207	0.021	0.026	0.079
2.423	0.044	0.054	0.14
2.66	0.075	0.088	0.21
2.92	0.11	0.13	0.3
3.206	0.16	0.18	0.41
3.519	0.22	0.25	0.53
3.862	0.28	0.32	0.67
4.241	0.35	0.4	0.82
4.656	0.43	0.48	0.97
5.111	0.51	0.57	1.13
5.611	0.6	0.67	1.3
6.158	0.69	0.77	1.46
6.761	0.78	0.87	1.62
7.421	0.87	0.98	1.77
8.147	0.96	1.08	1.92
8.944	1.05	1.19	2.06
9.819	1.13	1.29	2.18
10.78	1.21	1.39	2.29

11.83	1.28	1.49	2.38
12.99	1.34	1.58	2.45
14.26	1.4	1.68	2.51
15.65	1.45	1.78	2.57
17.18	1.52	1.9	2.64
18.86	1.61	2.04	2.73
20.7	1.74	2.22	2.84
22.73	1.91	2.44	2.98
24.95	2.16	2.7	3.15
27.38	2.47	3.01	3.32
30.07	2.87	3.35	3.49
33	3.35	3.72	3.63
36.24	3.89	4.07	3.72
39.77	4.47	4.4	3.76
43.66	5.06	4.68	3.72
47.93	5.61	4.89	3.61
52.63	6.06	5	3.43
57.77	6.35	5.01	3.18
63.41	6.42	4.9	2.87
69.62	6.22	4.67	2.53
76.43	5.77	4.31	2.18
83.9	5.1	3.85	1.86
92.09	4.25	3.33	1.6
101.1	3.31	2.77	1.42
111	2.34	2.21	1.3
121.8	1.45	1.68	1.21
133.7	0.73	1.21	1.11
146.8	0.27	0.83	0.99
161.2	0.062	0.54	0.86
176.8	0.0072	0.36	0.76
194.2	0.0003	0.27	0.72
213.2	0	0.25	0.72
234.1	0	0.27	0.73
256.8	0	0.31	0.73
282.1	0	0.36	0.67
309.6	0	0.37	0.57
339.8	0	0.33	0.45
373.1	0	0.25	0.34
409.6	0	0.14	0.23
449.7	0	0.054	0.12
493.6	0	0.01	0.042
541.9	0	0.00075	0.0073
594.9	0	0	0.00046
653	0	0	0
716.9	0	0	0
786.9	0	0	0

863.9	0	0	0
948.2	0	0	0
1041	0	0	0
1143	0	0	0
1255	0	0	0
1377	0	0	0
1512	0	0	0
1660	0	0	0
1822	0	0	0

#### A3.4.2.6 Influence of fluoride presence on the removal of arsenic (V)

Table A3.36 - Results obtained for the distribution of particle sizes with a binary mixture (arsenic (V)+ fluoride)

Particle diameter ( $\mu\text{m}$ )	Initial fluoride concentration ( $\text{mg}\cdot\text{L}^{-1}$ )		
	0	5	15
	Volume fraction (%)		
0.375	0	0	0
0.412	0	0	0
0.452	0	0	0
0.496	0	0	0
0.545	0	0	0
0.598	0	0	0
0.657	0	0	0
0.721	0	0	0
0.791	0	0	0
0.869	0	0	0
0.953	0	0	0
1.047	0	0	0
1.149	0	0	0
1.261	0	0	0
1.385	0	0	0
1.52	0	0	0.00007
1.669	0	0	0.0014
1.832	0	0	0.0099
2.01	0	0	0.035
2.207	0	0	0.079
2.423	0	0	0.14
2.66	0	0.0003	0.21
2.92	0.00012	0.0039	0.3
3.206	0.0018	0.021	0.41
3.519	0.011	0.057	0.53
3.862	0.033	0.11	0.67
4.241	0.069	0.18	0.82

4.656	0.12	0.27	0.97
5.111	0.18	0.37	1.13
5.611	0.25	0.5	1.3
6.158	0.35	0.64	1.46
6.761	0.46	0.8	1.62
7.421	0.58	0.98	1.77
8.147	0.73	1.17	1.92
8.944	0.9	1.39	2.06
9.819	1.09	1.62	2.18
10.78	1.31	1.85	2.29
11.83	1.54	2.1	2.38
12.99	1.79	2.33	2.45
14.26	2.05	2.55	2.51
15.65	2.31	2.74	2.57
17.18	2.54	2.89	2.64
18.86	2.75	2.98	2.73
20.7	2.91	3.03	2.84
22.73	3.01	3.02	2.98
24.95	3.05	3	3.15
27.38	3.05	2.97	3.32
30.07	3.01	2.97	3.49
33	2.96	3.03	3.63
36.24	2.93	3.13	3.72
39.77	2.92	3.29	3.76
43.66	2.94	3.46	3.72
47.93	2.99	3.63	3.61
52.63	3.06	3.76	3.43
57.77	3.14	3.83	3.18
63.41	3.22	3.84	2.87
69.62	3.3	3.79	2.53
76.43	3.36	3.67	2.18
83.9	3.4	3.5	1.86
92.09	3.4	3.28	1.6
101.1	3.36	3.01	1.42
111	3.26	2.69	1.3
121.8	3.1	2.34	1.21
133.7	2.88	1.97	1.11
146.8	2.62	1.58	0.99
161.2	2.33	1.23	0.86
176.8	2.04	0.93	0.76
194.2	1.76	0.71	0.72
213.2	1.51	0.56	0.72
234.1	1.31	0.48	0.73
256.8	1.13	0.43	0.73
282.1	0.96	0.4	0.67
309.6	0.79	0.35	0.57

339.8	0.6	0.28	0.45
373.1	0.39	0.18	0.34
409.6	0.2	0.085	0.23
449.7	0.066	0.025	0.12
493.6	0.012	0.0035	0.042
541.9	0.00078	0.00015	0.0073
594.9	0	0	0.00046
653	0	0	0
716.9	0	0	0
786.9	0	0	0
863.9	0	0	0
948.2	0	0	0
1041	0	0	0
1143	0	0	0
1255	0	0	0
1377	0	0	0
1512	0	0	0
1660	0	0	0
1822	0	0	0



## A3.5 Ternary mixtures

### A3.5.1 Concentration

Table A3.37 - Results obtained for the contaminants concentration over time with a ternary mixture (iron + fluoride + arsenic (V))

		Time / min	0	5	10	15	20	30	40	50
[Fe] / mg·L <sup>-1</sup>	[Fe] / mg·L <sup>-1</sup>	19.50	7.69	4.77	3.82	2.85	1.29	0.66	0.0	
	% of Fe removal	0.0	60.6	75.5	80.4	85.4	93.4	96.6	100.0	
[F] / mg·L <sup>-1</sup>	[F] / mg·L <sup>-1</sup>	9.8	7.0	4.8	4.5	2.9	1.9	0.0	0.0	
	% of F removal	0.0	28.6	51.0	54.1	70.4	80.6	100.0	100.0	
[As(V)] / mg·L <sup>-1</sup>	[As(V)] / mg·L <sup>-1</sup>	2.8		0.0		0.0	0.0	0.0	0.0	
	% of As(V) removal	0.0		100.0		100.0	100.0	100.0	100.0	

**A3.5.2 Particles size**

*Table A3.38 - Results  
obtained for the distribution of  
particle sizes with a ternary  
mixture (iron + fluoride +  
arsenic (V))*

Particle diameter (µm)	Volume Fraction / %
0.375	0
0.412	0
0.452	0
0.496	0
0.545	0
0.598	0
0.657	0
0.721	0
0.791	0
0.869	0
0.953	0
1.047	0
1.149	0
1.261	0
1.385	0
1.52	0
1.669	0.00013
1.832	0.0025
2.01	0.017
2.207	0.056
2.423	0.12
2.66	0.21
2.92	0.32
3.206	0.46
3.519	0.61
3.862	0.78
4.241	0.97
4.656	1.16
5.111	1.35
5.611	1.55
6.158	1.73
6.761	1.91
7.421	2.07
8.147	2.21
8.944	2.32
9.819	2.4
10.78	2.44

11.83	2.45	863.9	0
12.99	2.42	948.2	0
14.26	2.39	1041	0
15.65	2.35	1143	0
17.18	2.33	1255	0
18.86	2.35	1377	0
20.7	2.43	1512	0
22.73	2.58	1660	0
24.95	2.78	1822	0
27.38	3.03		
30.07	3.27		
33	3.49		
36.24	3.64		
39.77	3.72		
43.66	3.73		
47.93	3.68		
52.63	3.57		
57.77	3.43		
63.41	3.26		
69.62	3.04		
76.43	2.79		
83.9	2.51		
92.09	2.22		
101.1	1.92		
111	1.64		
121.8	1.37		
133.7	1.11		
146.8	0.88		
161.2	0.7		
176.8	0.57		
194.2	0.49		
213.2	0.43		
234.1	0.35		
256.8	0.24		
282.1	0.13		
309.6	0.044		
339.8	0.0078		
373.1	0.00057		
409.6	0		
449.7	0		
493.6	0		
541.9	0		
594.9	0		
653	0		
716.9	0		
786.9	0		