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# THE EFFECTS OF ULTRASOUND AND ELECTROCOAGULATION ON REMOVAL OF MANGANESE FROM WASTEWATER

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### Abstract:

*In this paper the effect of ultrasound and electrocoagulation on the removal of manganese from synthetic wastewater was investigated. It was shown that 20 kHz ultrasound alone is not very efficient for manganese removal, but it enhances the efficiency of electrocoagulation. It was also shown that settling has a great effect on the electrocoagulation process because it stabilizes the flocs. A total of four laboratory-scale experiments were conducted, and the optimal experiment included 10 minutes of ultrasound and 10 minutes of electrocoagulation with iron electrodes. Under these conditions, 89.0 % of the manganese was removed with an operating cost of 0.13 EUR/mg Mn. Adding the cost of replacing the immersed ultrasonic probe, the total operating cost increased 3000-fold, making the immersed ultrasonic probe method unviable for large-scale application.*

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## 1 Introduction

Proper disposal of industrial wastewater, especially with regard to the presence of heavy metals, is necessary for reasons of public health, environmental protection, and social responsibility [1]. Manganese (Mn) is common in groundwater supplies but exceeding its suggested maximum levels (50 µg/L according to the Croatian regulations considering drinking water quality) can result with discolored water, water quality deterioration and increased water conditioning costs [2, 3]. Manganese also occurs in metallurgical wastewaters, and unlike other heavy metals, such as lead or chrome, it is only toxic in higher concentrations, but represents an increased risk for aquifer pollution [1]. Considering wastewater emissions, if discharged directly to surface water, the manganese limit is 2 mg/L, and 4 mg/L for public sewerage system [4].

Several techniques were employed for heavy metal removal, including adsorption, coagulation-flocculation, ion-exchange, chemical precipitation, biosorption, membrane filtration and flotation [5, 6]. Although these are considered as high efficiency technologies, there are several disadvantages such as operational problems, large amounts of settled sludge, high material and operating costs, some of them cannot be used as a standalone treatment, etc. [5, 6]. Thus, these disadvantages have created space for a development of other technologies, such as electrocoagulation (EC) and ultrasound (US).

EC involves the application of electric current through sacrificial electrodes to an aqueous solution [7]. Aluminium (Al) and iron (Fe) electrodes are most used [8]. It is an emerging technology in water and wastewater treatment that combines the advantages of coagulation, flotation (or settling) and electrochemistry [7]. EC is usually easy to operate, has high treatment efficiency and moderate to short operating time. On the other hand, there may be drawbacks such as electrode passivation and consumption, electrolyte addition due to the low electric conductivity of most solutions and sludge formation [9]. To eliminate these disadvantages, the combination of EC and other technologies, mostly advanced oxidation processes, has been studied by numerous authors [2, 9, 10, 11, 12].

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In general, US is a longitudinal wave with frequencies above 20 kHz. It belongs to the group of advanced oxidation processes and is usually combined with other processes such as ozonation and ultraviolet radiation [13, 14]. The main mechanism of US is related to ultrasonic cavitation, in which sound waves generate pressure fluctuations that lead to the formation, growth and implosion of microbubbles and the generation of hydroxyl radicals. It is a simple and versatile technology with generally low investment costs, but it is not very efficient as a sole treatment [13, 15].

Therefore, the aim of this research is to investigate the use of electrocoagulation and ultrasound to remove manganese.

## 2 The state of knowledge on manganese removal

The combination of EC and US has been studied by several authors, mostly for chemical oxygen demand (COD) and turbidity removal [16, 17]. All authors agree that the removal efficiencies were much higher with the combination of EC and US than with EC alone. Moreover, the removal efficiencies by US alone were very low [16].

So far, several researchers have studied the manganese removal by EC but, to the best of author's knowledge, there is no papers in which a combination of EC and US had been discussed.

Mores et al. [7] used EC for removal of total phosphorus and metals, including manganese, from slaughterhouse wastewater. Under the current density of 38.39 mA/cm<sup>2</sup> and pH 6.0, 65 % of manganese was removed in 60 min with Al electrodes. When Fe electrodes were used, 61 % of manganese was removed in 110 min under the same conditions. Pascu et al. [2] used photo-electrochemical method combined with EC for iron and manganese removal. The optimum current density and temperature were 3.4 mA/cm<sup>2</sup> and 25 °C, respectively. The removal efficiency was 55 % in 15 min. Further, the efficiency of photo-chemical and EC combination was compared with the composite and cellulose membranes adsorption. It was concluded that adsorption membrane method is simpler and has lower power consumption but is costly.

Mota et al. [1] studied the possibility of zinc, cadmium and manganese removal by EC using stainless steel electrodes. The best results were obtained at pH 10, 28 °C and 30 mA/cm<sup>2</sup> when about 90 % of each metal was removed from the solution within 20 min. Under these conditions, power consumption was 9 kWh/m<sup>3</sup>.

Salman [5] investigated manganese removal by combined EC/electroflotation technologies. In this experiment stainless steel electrodes were used. Under the optimal conditions (initial concentration 100 mg Mn/L, current density 4 mA/cm<sup>2</sup> and 120 min of electrolysis time) 99 % of manganese was removed. Current density and operation time were highlighted as the most significant parameters. Al Aji et al. [6] used iron electrodes for copper, zinc and manganese removal. The influence of current density, initial metal concentration and initial pH on heavy metal removal was investigated. It was determined that high current density and pH values are more suitable for metal removal. At current density of 25 mA/cm<sup>2</sup>, 72.6 % of manganese was removed.

## 3 Materials and methods

0.5 L of solution was prepared in a plastic container (11.5×11.5×9.5 cm). 12 mL of Imunn 44 (STADA) and 2 mL of Floradix (DIETPHARM) as dietary supplements were added to the potable water and mixed until the homogenous solution was reached (Figure 1). 2 g/L of NaCl was used as an electrolyte which, combined with added supplements, gives initial NaCl concentration of 4.69 ± 0.45 mg/L. The initial manganese concentration was 7.3 mg/L.



Figure 1. Initial solution in dark brown color containing 7.3 mg Mn/L.

Physical characteristics and concentrations of the initial solution are listed in Table 1. Manganese concentrations were measured using NANOCOLOR 500D (from Eutech) test 0-58 (manganese 10), while other parameters (pH, conductivity, TDS, NaCl, dissolved oxygen (DO), temperature, and electrical resistance) were measured using CyberScan PCD 650 multimeter (from Eutech).

Table 1. Physical characteristics and concentrations of the initial solution.

Parameter	Value
pH	$7.01 \pm 0.5$
Conductivity (mS)	$4.21 \pm 0.01$
TDS (mg/L)	$4.24 \pm 0.39$
NaCl (mg/L)	$4.69 \pm 0.45$
DO (mg/L)	$2.61 \pm 0.56$
Temperature (°C)	$25.9 \pm 1.8$
Electrical resistance ( $\Omega$ )	$118.3 \pm 10.5$
Mn (mg/L)	7.3

For the EC process (Figure 2), 4 Fe electrodes in monopolar parallel arrangement, are placed at 1 cm distance and immersed 2 cm in the solution. Total effective area of the electrodes was  $44.03 \text{ cm}^2$ . The container was placed on a magnetic stirrer RCT basic Pt1000 (IKA) and mixed at 150 rpm. Electrical current was set at 2 A. DF 1730 LCD (Goobay) power supply unit was used for voltage and current regulation. The duration of EC process was 10 min.

For the US process, Sonoplus HD 2200.2 (BANDELIN) ultrasonic homogeniser is used, Figure 2. An US transducer with titanium flat tip TT 13 ( $\text{\O} 13 \text{ mm}$ ) was immersed 1.5 cm into the solution. The US frequency was 20 kHz and with 50 % amplitude. The duration of the US process was 10 to 40 minutes.

In general, there were two experimental investigations. First one included EC process before US, and the second was in reverse order, US run before EC process (three separated experiments). The aim of this analysis was to determine whether the US would improve the efficiency of the EC. Therefore, in the first study, EC operated for 10 min, after which US continuously operated for 40 min. Samples were taken after 5 and 10 min of EC, and after 10, 20 and 40 min of US. Lastly, the solution was set to settle for 60 min.

When US operated before EC, three separated experiments were conducted. In the first one, US operated for 10 min, after which EC also operated for 10 min. The sample was then set to settle for 60 min. The samples for manganese concentrations were taken after 10 min of US, after 5 and 10 min of EC, and after settling. The second and the third experiment were the same as the first, only US operated for 20 and 40 min.



Figure 2. EC process with 4 Fe electrodes placed on a magnetic stirrer (left) and US homogenizer (right).

### 4 Results and discussion

Experimental results are shown in Table 2 and Figure 3. In experiment 1, when EC was used before US, after 5 min of EC the flocs started to settle slowly, and a thin layer of pure water was visible above the flocs. The manganese concentration decreased from initial 7.3 mg/L to 5.2 mg/L. But, after 10 min of EC, the amount of flocs increased and dispersed in the solution, so when the sample was taken, the photometer could not have read the concentration.

Then, the solution was treated with US. After 10 min manganese concentration decreased to 1.1 mg/L. It is assumed that this large decrease is a result of previous EC, and not the US. Still, US enhanced the settling process because right after the US treatment, a layer of clean water was again noticed above previously dispersed flocs. However, as it will be shown later, US itself is not a very effective method for manganese removal, but it enhances EC and settling processes when it is used as a prior treatment. To support the previous assumption, after additional 10 min of US the manganese concentration remained the same (1.1 mg/L). After a total of 40 min of US, the flocs began to break (melt), and the manganese concentration began to increase (1.8 mg/L). This resulted in an increase of the temperature (50.5 °C) and overmixing. Since the flocs were no longer firm, the concentration continued to increase through the settling phase, so the final concentration was risen to 2.5 mg Mn/L.

Table 2. Experimental results (after EC, US and 60 min of settling).

Experiment		Final manganese concentrations and percentage of removal	
No.	Description	mg Mn/L	[%]
1a	10 min EC + 10 min US	1.1	84.9
1b	10 min EC + 20 min US	1.1	84.9
1c	10 min EC + 40 min US	(1.8) 2.5	65.8
2	10 min US + 10 min EC	0.8	89.0
3	20 min US + 10 min EC	0.6	91.8
4	40 min US + 10 min EC	0.5	93.2

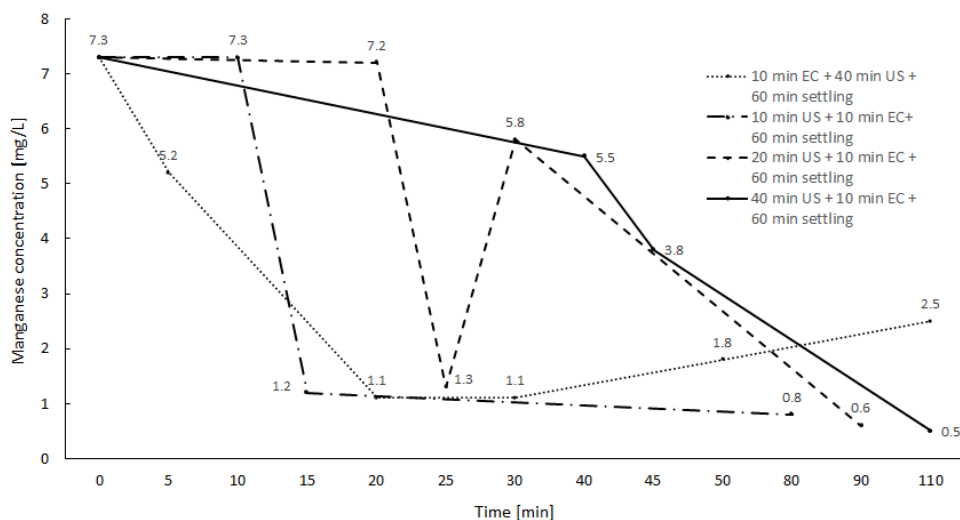


Figure 3. Manganese removal efficiency for experiments 1 (10 min EC + 40 min US), 2 (10 min US + 10 min EC), 3 (20 min US + 10 min EC) and 4 (40 min US + 10 min EC). All experiments include 60 min of settling at the end of the process.

In other experiments US operated before EC. After 10 and 20 min, experiments 2 and 3, US did not affect the reduction of manganese, but after 40 min, experiment 4, concentration decreased by 25 %.

In experiments 2 and 3, after 5 min of EC, the concentration decreased to 1.2 mg Mn/L and 1.3 mg Mn/L, respectively. Further, 10 min of EC caused the same problems as in experiment 1. Since the flocs dispersed, in experiment 2 manganese concentration again could not be read, and in experiment 3 the concentration increased to 5.8 mg Mn/L. This can be overcome by allowing the solution to settle for several minutes. The total manganese removal was 89.0% in experiment 2 and 91.8% in experiment 3. In experiment 4, after 40 min of US manganese concentration decreased by 25 %. The concentration also continued to decrease during EC, thus after 5 min of EC, concentration decreased by additional 23 % (totally 48 %), but after another 5 min, the sample was dispersed again, and the concentration could not have been determined.

Further, as it can be seen in Figure 4, the differences in color and settled quantities were noticed. When EC operated before US, the samples were brown (rust colored) from the beginning. This means that during EC  $\text{Fe}(\text{OH})_3$  hydroxides were formed and started to settle. But, when US operated before EC, the color stayed the same as the initial solution (light brown, Fig. 1) but turned green during the EC. This means that  $\text{Fe}(\text{OH})_2$  hydroxides were formed and since  $\text{Fe}^{2+}$  did not oxidize to  $\text{Fe}^{3+}$  the color stayed green till the end of the process.



Figure 4. Observed distinction in color and quantity of flocs (after 60 min of settling): EC process operated before US (left) and US operated before EC (right).

The type of hydroxide formed depends on the pH of the solution and DO. If the pH of the solution is above 2, rust-coloured  $\text{Fe}(\text{OH})_3$  forms and settles. The same is true when the pH is above 9, when greenish  $\text{Fe}(\text{OH})_2$  forms and settles [18]. The pH of the solution was measured during each phase of the experiments, and the above theory was confirmed, Figure 5. In all four cases, the pH increased during EC. In experiment 1, when EC was operated before US, the maximum pH was 8.4 and rusty brown  $\text{Fe}(\text{OH})_3$  formed. In experiments 2, 3 and 4, the pH increased to above 9 and greenish  $\text{Fe}(\text{OH})_2$  formed. Visually, the  $\text{Fe}(\text{OH})_3$  flakes were denser and occupied less volume.

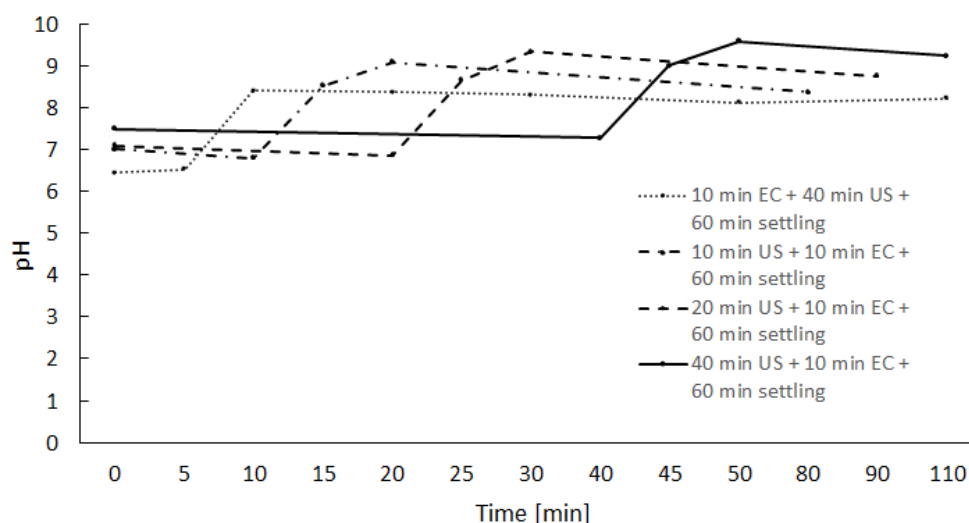


Figure 5. Measured pH for experiments 1 (10 min EC + 40 min US), 2 (10 min US + 10 min EC), 3 (20 min US + 10 min EC) and 4 (40 min US + 10 min EC). All experiments include 60 min of settling.

Furthermore,  $\text{Fe}^{2+}$  will also not oxidize at lower dissolved oxygen (DO) values [19]. In all cases DO decreased during first 5 min of EC, and stayed about the same during next 5 min when US operated before EC. But, when EC operated before US, DO increased strongly (from 0.29 mg/L to 1.96 mg/L) between 5 and 10 min of EC causing oxidation of  $\text{Fe}^{2+}$  to  $\text{Fe}^{3+}$ , Figure 6.

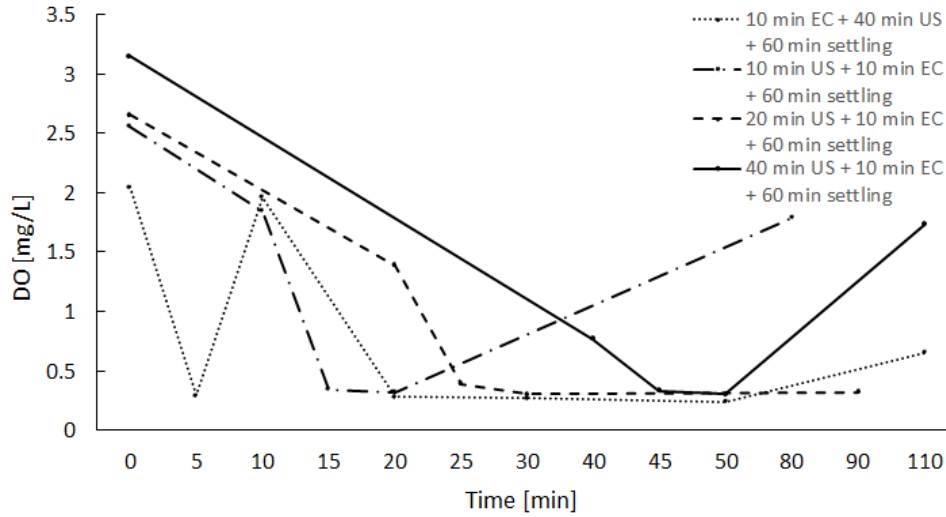


Figure 6. Measured DO for experiments 1 (10 min EC + 40 min US), 2 (10 min US + 10 min EC), 3 (20 min US + 10 min EC) and 4 (40 min US + 10 min EC). All experiments include 60 min of settling.

## 5 Operating cost analysis

The estimation of the operating costs (Tables 3 and 4) is given regarding the electricity used by electrodes, magnetic stirrer and US homogenizer, as well as the material (electrodes, US probe (horn)) according to [9]:

$$\text{Operat. cost} = a \cdot C_{\text{electricity}} + b \cdot C_{\text{material}} + c \cdot C_{\text{probe}} \quad (1)$$

where:  $C_{\text{electricity}}$  is electric energy consumption of 1  $\text{m}^3$  of treated water ( $\text{kWh}/\text{m}^3$ ), including energy consumption of electrodes, stirring mixer and US probe,  $C_{\text{material}}$  ( $\text{kg Fe}/\text{m}^3$ ) is the cost of the electrode material used in 1  $\text{m}^3$  of the treated water and  $C_{\text{probe}}$  ( $\text{pc.}/\text{m}^3$ ) is the cost of an US probe used in 1  $\text{m}^3$  of the treated water.  $a$  is the average electricity price (0.13 EUR/kWh) according to the national tariff models,  $b$  is the average market price of iron given as 0.073 EUR/kg and  $c$  is the average market price of an US probe TT 13 (66.54 EUR/pc.). Power meter and analyzer Emos P5821 was used to measure the energy consumption of a stirring mixer and US probe. 10 min of US consumes 0.011 kWh and 10 min of stirring consumes 0.002 kWh. The consumption of iron electrodes ( $C_{\text{electrodes}}$  ( $\text{kWh}/\text{m}^3$ )) is calculated according to the following expression [9]:

$$C_{\text{electricity}} = U \cdot I \cdot \frac{t_{\text{EC}}}{V} \quad (2)$$

where:  $U$  is the power supply voltage (V),  $I$  is the power supply current (A),  $t_{\text{EC}}$  the duration of the electrocoagulation process (h),  $V$  is the volume of the treated water ( $\text{m}^3$ ).

The electrode material consumption ( $\text{kg}/\text{m}^3$ ) is determined according to Faraday's law [9, 22]:

$$C_{\text{material}} = \frac{(I \cdot t_{\text{EC}} \cdot M_w)}{(z \cdot F \cdot V)} \quad (3)$$

where:  $M_w$  is molecular mass (55.85 (g/mol) for Fe),  $z$  number of electrons transferred (2 or 3 for Fe, depending on the oxidation state),  $F$  Faraday constant (96487 (C/mol)),  $t_{\text{EC}}$  the duration of the electrocoagulation process (s).

Since it is not specified in the product catalogue, a lifespan of an US probe is estimated. According to the BANDELIN product catalogue [20], titanium probe (type KE 76,  $\varnothing$  6 mm) with a small radiation surface can last approximately 6 hours. Also, Nedelcu et al. [21] tested a lifetime of the probe made from the same material as the one used in this study (Ti-6Al-4V), working at the same frequency (20 kHz), but with a larger diameter ( $\varnothing$  15.9 mm). It was found that the failure occurs after about 15 hours of tests. Therefore, a lifespan ( $T_{lifespan}$  (h)) of a TT 13 probe is estimated at 13 working hours. Accordingly, an US probe consumption (pc./m<sup>3</sup>) is determined by the following equation:

$$C_{probe} = N \cdot \frac{\left(\frac{t_{US}}{T_{lifespan}}\right)}{V} \quad (4)$$

where:  $N$  is the number of the probes used (pc.) and  $t_{US}$  is the duration of the US process (h).

Table 3. Operating costs per volume of the treated water (EUR/m<sup>3</sup>).

Experiment		a	C <sub>electricity</sub>	b	C <sub>material</sub>	c	C <sub>probe</sub>	Operating cost
No.	Description	EUR/kWh	kWh/m <sup>3</sup>	EUR/kg	kg/m <sup>3</sup>	EUR/pc.	pc./m <sup>3</sup>	EUR/m <sup>3</sup>
1a	10 min EC + 10 min US	0.13	37.07	0.073	1.85	66.54	25.64	1711.11
1b	10 min EC + 20 min US		59.07		1.85		51.28	3420.12
1c	10 min EC + 40 min US		103.07		1.85		102.56	6838.15
2	10 min US + 10 min EC		37.07		2.78		25.64	1711.18
3	20 min US + 10 min EC		59.07		2.78		51.28	3420.19
4	40 min US + 10 min EC		103.07		2.78		102.56	6838.22

Table 4. Operating costs per milligram of removed manganese (EUR/mg Mn).

Experiment		a	C <sub>electricity</sub>	b	C <sub>material</sub>	c	C <sub>probe</sub>	Operating cost
No.	Description	EUR/kWh	kWh/mg Mn	EUR/kg	kg/mg Mn	EUR/pc.	pc./mg Mn	EUR/mg Mn
1a	10 min EC + 10 min US	0.13	0.003	0.073	0.0001	66.54	0.002	0.14
1b	10 min EC + 20 min US		0.003		0.0001		0.004	0.28
1c	10 min EC + 40 min US		0.004		0.0002		0.011	0.71
2	10 min US + 10 min EC		0.003		0.0002		0.002	0.13
3	20 min US + 10 min EC		0.003		0.0002		0.004	0.26
4	40 min US + 10 min EC		0.003		0.0002		0.008	0.50

In addition to the calculation in EUR/m<sup>3</sup> (Table 3), the operating costs are expressed per milligram of manganese removed (EUR/mg Mn), as shown in Table 4. It should be noted that the calculation method is the same, but instead of the volume of water treated, the mass of manganese removed is considered. According to the results, experiment 2 has the lowest operating cost of 0.13 EUR/mg Mn, and second lowest when the results are expressed in EUR/m<sup>3</sup> (1711.18 EUR/m<sup>3</sup>). The operating costs expressed in volume are extremely high due



to the cost of the US probe, which must be replaced regularly. The replacement of the probe increases the operating costs up to 3000 times, which makes the US process with submerged probe unprofitable.

It is noted that the achieved values of electricity consumption are theoretical, specified by Faraday's law, and the real consumption can differ [22, 23]. Therefore, the real consumption of the electrode mass should be precisely determined by weighing the electrodes before and after each EC process. For comparison, Yoo [24] studied the combination of coagulation and ultrafiltration for the treatment of a raw water collected from the receiving well of a water treatment plant. This water contained 0.09 mg/L of manganese among other impurities. The operating cost of this coagulation/ultrafiltration combination were 0.3380 \$/m<sup>3</sup> including sludge disposal.

## 6 Conclusion

The results show that the combination of EC and US is a promising technology for manganese removal. In all experiments a large quantity of flocs was generated and settled after only 5 min of EC. But after 10 min, flocs dispersed in the volume and the sample of clear water could not be taken. Since generated flocs contain manganese, it is normal that its concentrations are higher or could not be read by a photometer. However, this can be overcome by letting the solution and flocs to set and stabilize for some time before taking a sample. It is also suggested that the settling time immediately after EC be taken into account. The shorter settling time is expected when using a lamella clarifier with the same efficiency, and it is recommended that this issue be analyzed in future studies.

The results also show that US is not efficient as a sole method for manganese removal, but its application before EC increases the removal efficiency. In addition, the application of US after EC may lead to floc degradation due to temperature increase and overmixing. In future studies, a water cooler may be used to control the temperature. Considering the national standards for wastewater discharge, all final concentrations achieved values lower than the limit (4 mg/L). Thus, this combination of EC and US can be used to treat the wastewater that contains manganese as a step before discharging effluent in the public sewerage system. It is also an alternative treatment method to established technologies, such as ion exchange or membrane filtration, already used in practice. 40 min of US and 10 min of EC (experiment 4) reduced manganese concentration to 0.5 mg/L and even though this experiment had the highest efficiency (93.2 %), the second experiment (10 min US and 10 min EC) is optimal because it has the best efficiency and cost ratio (89 % and 0.13 EUR/mg Mn).

The cost of replacing the immersed ultrasound probe increases the total operating cost up to 3000 times, making the immersed ultrasound process unprofitable for a large-scale application. Overall, better removal efficiency was also recognized in the quantity of generated sludge and supernatant turbidity. When US operated before EC, a higher efficiency was achieved for the same values of operative parameters, more sludge was generated, and the supernatant was clearer. It can be concluded that the type of generated hydroxide depends on the pH and DO. pH between 2 and 9 and higher DO values cause oxidation of Fe<sup>2+</sup> to Fe<sup>3+</sup>, thus brown (rust) color.

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