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Iron-oxidation processes in an electroflocculation (electrocoagulation) cell

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ABSTRACT

The processes of iron oxidation in an electroflocculation cell were investigated for a pH range of 5–9 and electric currents of 0.05–0.4 A (equivalent current densities of 8.6–69 A/m²). At all pH values and electric currents investigated, it was demonstrated and proven that for all practical purposes, the form of iron that dissolves from the anode is Fe^{2+} (ferrous). The difference between the amount of theoretical dissolution as calculated by Faraday's law and the amount of observed dissolved iron ions may indicate two phenomena in electrochemical cells. The first is possible dissolution of the anode even without the operation of an electric current; this led to higher theoretical dissolution rates at lower pH. The second is the participation of some of the electrons of the electric current in reactions other than anode dissolution makes in the local oxidation saturation level near the anode and did not affect iron-oxidation rates in the electroflocculation processes.

The oxidation rates of the dissolved Fe^{2+} (ferrous) to Fe^{3+} (ferric) ions in electroflocculation processes were strongly dependent on the pH and were similar to the known oxidation rates of iron in non-electrochemical cells.

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1. Introduction

1.1. General introduction

One of the modern water treatment technology that is currently drawing attention, both scientifically and for its practical aspects, is electroflocculation (electrocoagulation) [1–3]. This method presents an alternative to "conventional" flocculation with several advantages: easy operation, lower quantities of produced sludge, avoidance of chemical usage and, most importantly, no anions such as chloride and sulfate need to be added to the solution [4]. Unlike "conventional" flocculation in which the coagulants are added to the water as salts, in the electroflocculation process, the aluminum or iron ions used as the coagulants are added to the water by dissolving the anode in an electrochemical cell. These coagulant ions ultimately lead to aggregation of the original particles in the water, which are removed in a later sedimentation or filtration process. Using iron as the coagulant, a large number of studies [5–7] have demonstrated that electroflocculation is very efficient for removing particles from water. The present study on iron-oxidation processes in the electrochemical cell was initiated because some of the basic mechanisms remain unclear and several

questions arise from the theoretical description of the electroflocculation process in recent literature.

1.2. Scientific background

Modern electroflocculation literature commonly describes the following reactions near the anode and cathode:

Near the anode:

$$M_{(s)} \rightarrow M_{(aq)}^{+n} + ne^{-}$$
⁽¹⁾

According to water degradation some authors [5,6,8] also claim the presence of an oxygen-evolution reaction:

$$2H_2O \rightarrow O_{2(g)} + 4H^+_{(aq)} + 4e^-$$
 (2)

Near the cathode:

$$2H_2O + 2e^- \rightarrow H_{2(g)} + 2OH^-_{(a0)}$$
 (3)

These reactions explain the appearance of bubbles near the cathodes and anodes during the process and can also explain the concomitant rise in pH (dependent on solution alkalinity and electrochemical process intensity, i.e., the intensity of the electric current and operation time).

The connection between electrochemical process intensity and the amount of coagulants dissolved into the solution from the anode

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is described by Faraday's law:

$$w = \frac{ItM_{w}}{nF}$$

where *I* is the current intensity (A), *t* is operation time (s), M_w is molecular weight of the metal (g/mol), *F* is Faraday's constant (96,485 C/mol), *n* is the number of electrons involved in the reaction, and *w* is the quantity of metal dissolved (g).

Some authors added correction factors to Faraday's law such as current efficiency [17,10] or Faradic yield [18] to describe the gap between the theoretical and the experimental dissolution rates.

Faraday's law was verified and approved in the beginning of the 20th century by weighting the passivation layer that form on the cathode and not by direct measuring of the anode dissolution rates as can be done with modern apparatus.

Faraday's law, written without any correction factor, is valid only when all the electrons flowing in the system participate in the metal-dissolution reaction at the anode (reaction (1)). This law describes one of the electric-charge transfer mechanisms between solution and anode and without a correction factor it claims that this is the only mechanism occurring near the anode. Therefore, when using Faraday's law without a correction factor, there should not be any other reactions near the anode, such as the oxygenevolution reaction (reaction (2)).

While literature from the beginning of the 20th century [9] discusses the need to correct Faraday's law when there are more reactions near the anode, it is common to find in the modern literature description of the process by writing Faraday's law without any correction factor and also writing the two reactions (1) and (2), creating a contradiction in terms.

Over the last few years and for a number of reasons, there has been a tendency to use iron instead of aluminum as the coagulant ion. A scientific understanding of iron dissolution in electrochemical cells is much more complex, because of the two oxidation states of iron species that can exist in solution: Fe²⁺ (ferrous) and Fe³⁺ (ferric). While most authors assume that iron dissolves from the anode as Fe²⁺, this assumption still lacks direct evidence [10] and some researchers claim that the dissolved iron can be Fe³⁺ [8,2,11]. When studying the electroflocculation process, it is very important to clearly understand what form of iron is dissolved. Without this information, the number of electrons participating in each metal atom's dissolution reaction from the anode remains unknown and consequently, there is 33% uncertainty in the theoretical value of the iron-dissolution rate. Furthermore, Fe²⁺ and Fe³⁺ induce different hydrolysis and flocculation processes. Finally, Fe²⁺ oxidation to Fe^{3+} (reaction (4)) can occur in the process and this also needs to be taken into account.

$$4Fe^{2+} + O_2 + 2H_2O \to 4Fe^{3+} + 4OH^-$$
(4)

Matteson et al. [6] neglected the effect of reaction (4) by claiming that the presence of Fe^{3+} ions in equilibrium with Fe^{2+} is quite small. Other authors [12,13] claimed that reaction (4) should occur in the presence of oxygen but they did not mention the effect of solution pH value on the reaction rate.

Morgan and Lahav [14] demonstrated the strong dependence of the reaction rate of Fe^{2+} oxidation to Fe^{3+} on pH and oxygen saturation conditions in non-electrochemical processes. Under alkaline conditions, the Fe^{2+} will immediately oxidize to Fe^{3+} (pH 14–7.6). At lower pH, the rates will be much lower and below pH 4, the oxidation process becomes negligible.

While reaction (4) is not directly connected to the electrochemical process, it is affected by the oxygen saturation level (OSL) in the solution and, thus, by the importance of the oxygen-evolution reaction (reaction (2)) [10] and the redox potential of the solution. Both the oxygen state and the redox potential can change as a result of the electroflocculation process. Therefore, it is important to understand whether different iron-oxidation rates occur in electrochemical and non-electrochemical reactions, a question which was not answered before.

1.3. The research goals

The goal of this research was to study iron-oxidation processes of near neutrals water in the electroflocculation process. The first step in achieving this goal was to define what forms of iron (Fe^{2+} or Fe^{3+}) were dissolved from the anode for different electric currents and different pH values.

The second step was to define the relationship between the process intensity (operation time and electric current) and the dissolution rates at different pH values and to understand the deviations from the theoretical dissolution rates as calculated by Faraday's law.

The last step was examination of Fe²⁺ to Fe³⁺ oxidation rates (when Fe²⁺ is the iron species that is dissolved in the electrochemical cell) at different pH values. Those rates were compared with the rates that have been previously quoted in non-electrochemical cells.

2. Materials and methods

2.1. Methods

Solution: A 1-l solution was made up to represent tap water conditions with a conductivity of 1 mS/cm, obtained via the addition of different quantities of NaCl. To stabilize the pH, an organic buffer was added: 8, 3 and 2 g/l MOPS buffer (3-(N-morpholino)propanesulfonic acid; Roth Company) for pH 5, 6 and 7, respectively, and 1.5 g/l glycine (Roth Company) for pH 8 and 9. Organic buffers were used instead of the commonly used orthophosphate or carbonate buffers to avoid the possible formation of precipitates which could influence iron-dissolution rates.

Electrode: Before each test, the iron anode was immersed in an HCl solution for at least 30 min to remove the oxide layer. Thereafter, in order to clean the surface of iron particles, the electrode was rinsed, then immersed in distilled water for 10 min and re-rinsed with distilled water.

Three different experimental sets were designed in order to achieve the aforementioned research goals:

Experimental set 1: The aim of this set of experiments was to determine the type and quantity of iron dissolved under defined conditions (pH 5, 6, 7, 8, 9, and electric current 0.05, 0.1, 0.2, 0.4 A (corresponding to electric current densities of 8.6, 17.2, 34.5, 69 A/m^2). According to Faraday's law, the theoretical value of dissolved Fe²⁺ under each condition should be 3.5 mg/l. Hence operation time was inversely adjusted to the electric current (for current 0.05, 0.1, 0.2, 0.4 A, the time was 4, 2, 1, 0.5 min, respectively) to obtain this target value. Each test was performed at least three times for each condition (pH and electric current) and the results were averaged to determine the amount of iron dissolved in each experiment.

Since there was an indication of very rapid Fe^{2+}/Fe^{3+} oxidation rates at higher pH values, it was necessary to "freeze" the oxidation state of the iron immediately after its dissolution. This was done by adding 1,10-phenanthroline (100 mg/l) to the solution before starting the electrochemical process: 1,10-phenanthroline forms a red complex with Fe^{2+} ions but not with Fe^{3+} . Thus, if the Fe^{2+} ion is the type that dissolves, the 1,10-phenanthroline will complex it and prevent further oxidation.

The absorption of light (wavelength 510 nm) is linear for a concentration of Fe²⁺ between 0.5 and 4 mg/l. Thus, by adding 1,10-phenanthroline to the solution it was also possible to determine the

concentration of Fe^{2+} ions. In addition, at the beginning and end of each test, the total iron concentration, the oxygen concentration and redox potential were measured.

Experimental set 2: This set was designed to observe whether there is iron dissolution that is unbiased by the electric current and, if so, at which pH values this occurs. The results of this experimental set were expected to help explain the iron-dissolution rates investigated in experimental set 1. The same procedure was performed as in experimental set 1. The only difference was that no electrochemical reaction occurred as the electric current was zero. Each test was performed at least three times for each pH value and the results averaged to determine the amount of iron that dissolved in each condition.

Experimental set 3: The goal here was to understand the oxidation rates of Fe^{2+} to Fe^{3+} in the electrochemical process and their dependence on pH. The solution in these experiments was the same as in the two previous sets but without 1,10-phenanthroline, because its reaction with Fe^{2+} ions delays or prevents the oxidation process. An electric current of 0.2 A was applied for 1 min at different pH values (5, 6, 7, 8, 9), and then the Fe^{2+} concentration was measured using the phenanthroline method at fixed time intervals. The redox potential and oxygen values were also determined.

2.2. Materials

Electrochemical cell: A glass electrode spacer was designed to hold the electrodes at a fixed distance of 5 mm from each other. The cell was immersed in a 2-l glass beaker and a glass mechanical stirrer (without magnetic field, which could affect the experimental results) was placed in the middle of the cell. The velocity of the mechanical stirrer in all experiments was 250 rpm, which resulted in very fast mixing in the cell. The electric current was supplied by a DC power supply (Startron, maximum 30 V and 6.4 A).

Electrodes: The anode was made of iron (mild steel ST 37-2) and the cathode of stainless steel (in order to prevent the possibility of additional dissolution from the cathode [8,15]). The size of each electrode was 78 mm high \times 99 mm wide \times 2 mm deep. The effective size of the electrodes immersed in the solution was 58 cm². Each electrode had 28 holes in order to enable water flow in the cell and good mixing.

Measurements: The following parameters were measured during the experiments with suitable electrodes:

pH (Knick pH meter 765 calimatic), conductivity (WTW, cond 315), redox potential (WTW, pH meter pH 191, redox electrode (KCl 3 M, Schott, blue line 31 Rx)), oxygen (WTW, Oxi 323). Samples of 10 cm³ were taken from the solution for Fe²⁺ and total iron concentration measurements. The concentration of total iron was measured by flame AAS (Perkin-Elmer 1100B). Before analysis, each sample was acidified with a 1.4 M HNO₃ solution in order to prevent precipitation.

The concentration of ferrous iron (Fe²⁺) was measured using the 1,10-phenanthroline method [16]. The empirical iron concentration-absorption model was: $C_i = 5.0787Abs - 0.3717$, where C_i is the concentration of iron (mg/l), *Abs* is the light absorption (in wavelength of 510 nm).

3. Results

Tables 1 and 2 present the average concentration ratios between measured Fe^{2+} and total iron for various pH and electric current values, respectively, as determined in the first experimental set. At lower pH values (5 and 6), the Fe^{2+} -to-total Fe ratio was greater than 90% while at higher pH (7–9) the ratio was about 83%. At all electric currents, the Fe^{2+} -to-total Fe ratio was between 84 and 91%.

Table 1

Average Fe²⁺-to-total Fe ratio given in percents as a function of measured pH.

pН	Number of experiments	Average Fe ²⁺ :total Fe (%)	Standard deviation (%)
5	34	90	12.7
6	37	96	9.5
7	29	82	13.7
8	30	83	7.4
9	24	83	12.6

Table 2

Average Fe²⁺-to-total Fe ratio given in percents as a function of measured electric current.

Electric current (A)	Number of experiments	Average Fe ²⁺ :total Fe (%)	Standard deviation (%)
0.05	46	87	15
0.1	42	88	12
0.2	35	91	10
0.4	30	84	12

The concentrations of dissolved iron for experimental set 1 are shown in Fig. 1a–c. While for all of the experiments the theoretical value (according to Fe^{2+} dissolution) was 3.5 mg/l, the actual results showed more than twofold higher concentrations (>8 mg/l) at lower pH values (5, 6) and about half that concentration (1.7 mg/l) at pH 9 (Fig. 1b and c). Those lower dissolution rates had better matching to the theoretical value according to Fe^{3+} dissolution. Since at pH



Fig. 1. (a) The amount of total iron dissolved from the anode for each pH and electric current. Each point represents the average of three measurements. (b) The amount of total iron dissolved from the anode for various electric currents at pH 5–7. Each point represents the average of three measurements. (c) The amount of total iron dissolved from the anode for various electric currents at pH 8 and 9. Each point represents the average of three measurements.



Fig. 2. The change in oxygen saturation level (OSL) before and after electroflocculation at pH 9 for different currents.

values of 5, 6 and 7, the buffer was MOPS and at pH values of 8 and 9, the buffer was the same quantity of glycine (1.5 g/l), the results are shown in two separate illustrations (Fig. 1b and c).

For each electric current the voltage did not change during the electroflocculation process which can be related to the low electroflocculation operation time (maximum 4 min) and to the low electric current.

In all experiments, the oxygen saturation level (OSL) remained steady or decreased, but never increased, as can be seen in Fig. 2 for pH 9.

The high iron-dissolution rates observed at pH values of 5 and 6 raised the possibility that iron may be considerably dissolved, even without an electric current, at those pH values. This possibility was explored in the second experimental set (Fig. 3). In the absence of electric current, iron dissolution after 4.5 min at pH 5, 6 and 7 was significantly greater than at pH 8, while at pH 9 it was negligible. A comparison between Fe^{2+} ion concentration and total iron (data not shown) revealed that the dissolved iron was entirely in the form of Fe^{2+} at all pH values.

With no electric current activation, at lower pH (5–7), the high iron-dissolution rates were in accordance with high rates of oxygen consumption, while at higher pH (8, 9) the low iron dissolution was in accordance with lower rates of oxygen consumption (Fig. 4).

Studying the reaction rates of Fe²⁺ oxidation to Fe³⁺ at different pH values showed no oxidation at pH 5, moderate rates at pH 6 and very rapid oxidation at pH 7–9 (experimental set 3, Fig. 5).

4. Discussion

The results shown in Tables 1 and 2 represent the concentration ratios between Fe^{2+} ions that were dissolved and complexed by 1,10-phenanthroline molecules and total iron. Differences in these ratios may have resulted from some of the Fe^{2+} ions released from the anode being rapidly oxidized to Fe^{3+} , before the 1,10phenanthroline molecules could capture them. This would explain the decreasing trend in the Fe^{2+} -to-total Fe ratio at higher pH values, because of the higher tendency for Fe^{2+} to oxidize to Fe^{3+} (Fig. 5).



Fig. 4. The concentration of iron after 4.5 min of dissolution without electric current and the number of mol of oxygen consumed at different pH values.

Nevertheless, at all pH values and electric current intensities, more than 82% of the iron dissolved from the anode was in the form of Fe²⁺ (Tables 1 and 2). In a strict scenario, when 80% of the iron is Fe²⁺ and 20% Fe³⁺, the number of electrons induced (*n*) by Faraday's law should be 2.2, which would lead only to a deviation of 10% from the theoretical value for Fe²⁺ dissolution. In addition, the high ratio of Fe²⁺ to total Fe means that, regardless of pH values and electric currents, at least 80% of the iron in the solution will comply with the laws of the Fe²⁺ to Fe³⁺ oxidation reaction.

Consequently, it can be stated that if Fe³⁺ ions are also released from the anode, their quantity is negligible and for all *practical purposes*, it is the Fe²⁺ that dissolves under the experimental conditions (i.e. pH 5–9, electric current of 0.05–0.4 A). These findings reinforce the assumptions made by other investigators [11].

The cleaning of the electrode's passivation layer with acid before each experiments, the relatively low electroflocculation operation time and the observed linear relationships between the electric current and the voltage revealed that the overpotential effect was minimal and could be ignored.

Total iron dissolution is highly dependent on the pH value of the solution. At lower pH, the amount of iron dissolution can be more than twice its theoretical value calculated by Faraday's law (Fig. 1a) and in contrast to that law's logic, the results showed that at lower pH, as the current intensity increases, the amount of dissolved iron decreases (Fig. 1b). The high iron-dissolution rates at pH 5–7 without electric current (Fig. 3) imply the existence of an additional mechanism of iron dissolution from the anode, independent of the electrochemical process. Since all of the experiments were designed such that the amount of iron released according to Faraday's law would be equal, a lower electric current translates into a longer operating time, which leads to a higher amount of iron dissolution by a mechanism that is independent of the electrochemical process.



Fig. 3. Iron dissolution without electric current application. Each point represents the average of three measurements.



Fig. 5. Oxidation rates of Fe²⁺ to Fe³⁺ at pH values of 5–9.



Fig. 6. The direction of movement of electrons participating in the iron-dissolution reaction. (a) Dissolution without electric current and (b) electrochemical dissolution.

At pH 7, the amount of iron dissolution in the absence of any electric current (dissolution rates of 1.425 mg/min) is higher when both dissolution mechanisms (due to and independent of electric current; dissolution rates of 0.875 mg/min) act simultaneously (Figs. 1a and 3). While in both mechanisms, Fe²⁺ species dominate, they are differentiated by the direction of motion of the electrons participating in the dissolution. In electrochemical dissolution, the electric cycle wires, while in the case of no current, they enter the solution to balance its electric charge (Fig. 6). This leads to a non-linear additive relationship for the dual mechanisms (iron dissolution due to and independent of electric current) in predicting Fe²⁺ dissolution rates, which requires further investigation.

From Fig. 4, it can be seen that in the absence of electric current, the higher the amount of dissolved iron, the higher the depletion of oxygen. The high concentration of 1,10-phenanthroline molecules in solution prevents further oxidation of the dissolved Fe^{2+} to Fe^{3+} and oxygen depletion via this reaction. Hence, it can be concluded that the reaction of iron dissolution with no electric current is accompanied by oxygen depletion.

At higher pH values (Fig. 1a and c), the quantity of dissolved iron is almost half the amount calculated according to Faraday's law, meaning that reactions other than iron dissolution are occurring near the anode, such as oxygen evolution (reaction (2)). A change in local OSL near the anode due to such reactions may lead to a faster rate of Fe^{2+} oxidation to Fe^{3+} , since this oxidation reaction depends on the OSL [14].

The overall change in the OSL in solution during electroflocculation could result from three mechanisms:

- 1. A decrease in OSL due to oxidation of Fe²⁺ to Fe³⁺, this reaction played a negligible role in experimental sets 1 and 2 because of the appearance of a large quantity of 1,10-phenanthroline in solution (Tables 1 and 2).
- 2. A decrease in OSL due to the mechanism of iron dissolution independent of electric current this mechanism made a dominant contribution mostly at lower pH (Fig. 3).
- 3. An increase in OSL as result of the oxygen-evolution reaction (reaction (2)).

In all of the experimental sets, the OSL remained constant or decreased but never increased. At all pH values apart from pH 9, this could be attributed to a balance between the consumption of oxygen by the mechanism of iron dissolution which is independent of the electric current and the oxygen-evolution reaction. At pH 9, there was no dissolution in absence of electric current (Fig. 3) and the amount of iron dissolved was considerably less than that calculated using Faraday's law. In other words, there must be at least one reaction near the anode apart from the iron-dissolution reaction, such as an oxygen-evolution reaction. Consequently, the

small decrease in OSL at pH 9 (Fig. 2) for all currents studied (which may be attributed to the small quantity of Fe²⁺ to Fe³⁺ oxidation; Table 1) means that the reactions near the anode other than iron dissolution do not have a dramatic influence on the OSL in electroflocculation processes. The explanation for this could be twofold: (a) the reaction of oxygen evolution has no significant role [11] and other reactions, such as chlorine evolution, are dominant, or (b) the oxygen is formed as bubbles which escape the system with no chemical connection to the solution. Therefore, the Fe²⁺ to Fe³⁺ oxidation rates should not be faster during electroflocculation because of the oxygen-evolution reaction near the anode.

The oxidation rate of Fe²⁺ ions dissolved from the electrode to Fe³⁺ in solution with no 1,10-phenanthroline is strongly dependent on the pH value of the solution (Fig. 5, experimental set 3). Calculation of the oxidation rates based on the results revealed that, at pH 6, it will take 1 day for 95% of the Fe²⁺ ions to be oxidized, while at pH 5, there will not be any significant oxidation in the order of days. This general behavior is in accordance with the results of Morgan and Lahav [14] for aqueous solutions, except that their rates were slightly higher at pH 5 and 6.

Nevertheless, Faraday's law can predict the order of magnitude of the amount of iron dissolved, and this is its strength for practical purposes. However, its limits need to be understood. The deviation from Faraday's theoretical values may suggest other irondissolution phenomena and the existence of other reactions near the anode, which deserve further investigation.

5. Conclusions

Even though the iron-oxidation processes in an electroflocculation system for treating water are dependent on many parameters which were not considered in these experiments (such as the reaction of organic matter which can complex with the iron), the conclusions below illustrate several phenomena of the ironoxidation process in an electroflocculation cell and indicate the behavior of systems under real conditions:

- The type of iron released from the anode is Fe^{2+} for pH values of 5–9 and currents of 0.05–0.4 A (electric current densities of 8.6–69 A/m², voltages of 3–25 V).
- At lower pH values, a high dissolution rate can occur at the iron anode in the absence of an electric current. The type of iron that dissolves in this case is Fe²⁺, identified by determination of 1,10-phenanthroline molecules in the solution. In practice, when treating wastewater with electroflocculation, complexing agents must also be considered, as they can cause similar dissolution reactions with the iron anode. This phenomenon can decrease the life expectancy of the anode. It can also lead to a change in the operating time during the electroflocculation process that differs from the common current intensity-time multiplication represented by Faraday's law.
- The electrochemical dissolution of iron is different at different pH values. At higher pH, the real iron dissolution can be dramatically lower than the value calculated by Faraday's law, indicating that there are other reactions (apart from iron dissolution) that are occurring simultaneously near the anode.
- The OSL in the solution remains stable or decreases during the electroflocculation process.
- The oxidation rate of Fe²⁺ to Fe³⁺ in an electrochemical cell was slightly lower but nevertheless similar to the "normal" oxidation rate described by Morgan and Lahav [14] for an aqueous solution.
- In the presence of complexing compounds (1,10-phenanthroline in this study), there can be a significant delay in the oxidation of Fe²⁺ to Fe³⁺.

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