Electrocoagulation as a new and advanced technology for future challenges in the steel industry's water treatment plants

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Abstract

Water is a basic necessity of life, and it may seem inconceivable to imagine living without it. The environmental impact, together with social and economic impacts of traditional water treatment technologies in the steel industry plants and inevitable fact of water scarcity are leading forces driving a shift to a new paradigm in water treatments.

This research aimed to examine the effectiveness of an electrocoagulation (EC) process and a dissolved air flotation unit for removing various impurities in water from a steel manufacturing plant. Over the past decades EC has been accepted as an efficient and promising alternative technology in the field of water treatment. The EC technique is closely related to chemical coagulation, that involves the supply of coagulant ions by the application of electric current to a sacrificial anode (made of aluminum or iron) placed into a process tank. The main focus of the research is placed on total dissolved solids and conductivity of the treated water. In traditional systems, these are among the parameters that cannot be influenced by chemical dosing units. The specific goal was to understand parameters affecting efficiency of the EC process so to influence the total dissolved solids content and reduction of the residual inorganic and organic impurities, maintaining the necessary water balance.

Keywords: water savings, innovative solutions, green industry, recycling, electrochemical process

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1. INTRODUCTION

Industrial growth and the changes in manufacturing processes have resulted in the enhancement of the volume and complexity of water discharging to the environment. Many traditional and/or advanced and novel treatment processes have been modified and developed to eliminate or improve the quality of discharged water. Industrial effluents may also contain toxic pollutants, which have to be reduced or eliminated to protect the treatment plant in which they are treated, as well as the environment and public health at the end [1].

Steel industry is one of the main production technologies, which generates large amounts of wastewater. The key problems encountered for industrial wastewater discharges may be hydraulic overloads, temperature extremes and excessive amounts of oil, fats and grease as well as acidic or alkaline constituents, suspended solids, and various inorganic or organic contents [2-6]. Discharge of the steel industry wastewater into sewer systems without any treatment causes serious problems due to high potentials of toxicity causing a necessity for biological treatment [7].

The generated wastewater can be treated by different techniques which should allow either its reuse or its direct disposal into the sewage system. Some of these techniques are reverse osmosis carried out in plants with high energy consumption, or physical-chemical treatments that sometimes cannot achieve the satisfactory level of purification. [8]

Nowadays, the social concern about environmental impacts caused by industry is growing and new regulations demanding more strict environmental protection are being introduced. For this reason, the search for ''greener'' and more efficient methods for wastewater treatment is increasing [9].

Over the past decades, electrocoagulation (EC), sometimes also referred to as electroflocculation, has been accepted as an efficient and promising alternative technology in the field of water treatment. Interestingly, EC offers outstanding

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advantages including simple operation, lower sludge production, and simple automation. When EC operates at low current density, the operating cost of this process is often lower than that of the chemical coagulation process. In particular, EC offers more attractive properties for use in decentralized water treatments [10].

The EC methods use electrons as the main reagent in a heterogeneous reaction. When ionic constituents in wastewater are not sufficient, the use of supporting electrolyte is required to increase the ionic conductivity. The advantages of electrocoagulation include high particulate removal efficiency, a compact treatment facility, relatively low cost, does not require supplementary addition of chemicals, *etc.* [11].

The main advantage of this process is that the temperature control is not needed since EC is performed at ambient temperature. Electrochemical methods are simple, fast, inexpensive, easily operable and eco-friendly in nature. The main advantage is that water is purified, clear, colourless and odourless with low sludge production [12].

The EC technique is closely related to chemical coagulation, that involves the supply of coagulant ions (Al^{3+}, Fe^{3+}) by the application of an electric current to a sacrificial anode (made of aluminum or iron) placed into a process tank. The electric field allows greater suspension of solids compared to water purification processes that rely solely on chemicals. Thus, the electrocoagulation equipment, thanks to the created electric field and greater suspension of solids, is improving the coagulation process.

It proceeds through three stages: (i) coagulant formation by dissolution of metal ions of the anode, (ii) destabilization of pollutants, suspended particles and de-emulsification, and (iii) aggregation of unstable phases and floc-formation [12-15]. Destabilization of pollutants suspended particles, and de-emulsification mechanisms can be established through dispersed double-layer compression, ion neutralization species existing in water and wastewaters, leading to formation of flocs and sludge [15-16]. In this study, iron electrodes have been used in an EC process.

The mechanism of EC is extremely dependent on chemistry of the aqueous medium, especially its conductivity and total dissolved solids. Here the mechanism of generating ions by EC is explained with examples of iron, used as both the anode and cathode in this study. In an electrolytic system, iron is oxidized to produce iron hydroxide $Fe(OH)_n$ where n = 2 or 3. According to literature [17], two mechanisms have been proposed for the production of these species and we describe here one of the two (Equations (1) to (4)), while more details can be found in [17].

Reactions at the anode:

$$4Fe_{(s)} \rightarrow Fe_{aq}^{2+} + 8e^- \tag{1}$$

$$4Fe^{2+}_{(aq)} + 10H_2O + O_{2(g)} \rightarrow 4Fe(OH)_{3(s)} + 8H^+_{(aq)} \tag{2}$$

Reactions at the cathode:

$$8H^+_{(aq)} + 8e^- \rightarrow 4H_{2(g)} \tag{3}$$

Overall reaction is then:

$$4Fe_{(s)} + 10H_2O + O_{2(g)} \rightarrow Fe(OH)_{3(s)} + 4H_{2(g)} \tag{4}$$

From iron electrodes, iron ions are released into the solution through electrolytic oxidation of anode electrode and produce metal hydroxides after reacting with hydroxide monomer [11,18].

Hydrogen gas produced at the cathode during electrolysis cause flotation and better removal of pollutants [11,19].

In the steel industry, water is cooled in cooling towers and due to evaporation, salts and solids are created. The water composition determines the number of water cycles within the system (*NC*), before it is discharged, as per the Equation (5):

$$NC = \frac{TDS_{BD}}{TDS_{MW}} \tag{5}$$

where TDS_{BD} and TDS_{MW} represent total dissolved solids present in blow down (discharged) water and make up (raw, treated) water

In traditional systems, total dissolved solids and conductivity of the water, are one of the parameters that cannot be influenced by chemical dosing units.

The aim of this study was to investigate a pilot half-industrial system comprising an electrocoagulation process using rod-iron electrodes and a dissolved air flotation (DAF) unit. The specific goal was to understand parameters affecting efficiency of the EC process so to influence the total dissolved solids content and reduction of the residual inorganic and organic impurities. Such water quality improvement, any blow down from the system would be prevented leading to significant water savings.

2. RESEARCH METHODOLOGY

2. 1. Experimental setup

The studies were performed on a pilot plant that was in trial use in a steel manufacturing plant placed in North Italy and was composed of two main equipment pieces: an electrocoagulation unit and a dissolved air flotation (DAF) unit.

Inlet water was pumped from a scale pit at a flow rate of 10 $m^3 h^{-1}$ to a buffer tank, 1 m^3 in volume. From the buffer tank, a dedicated pump, placed inside the pilot system, pumped water at a flow rate of 1 $m^3 h^{-1}$ to the EC equipment, where minerals were removed. From this point, water was collected in a dedicated another buffer tank, from where water was pumped to the DAF unit. The finally treated water was returned into scale pit (Figure 1).

The EC equipment consists of oxidation cells, constructed of polypropylene, agitated by compressed air and supplied with an electric current rectifier with variable voltage/amperage for power supply. The configuration allows adjusting the number of electrodes and the distance between them, providing a tuning in the energy/electric ratio. There were 114 iron electrodes, each a rectangle 16×13 cm, spaced 0.5 cm apart.

The DAF unit is designed to treat the water coming from the EC equipment so to separate water from the sludge.

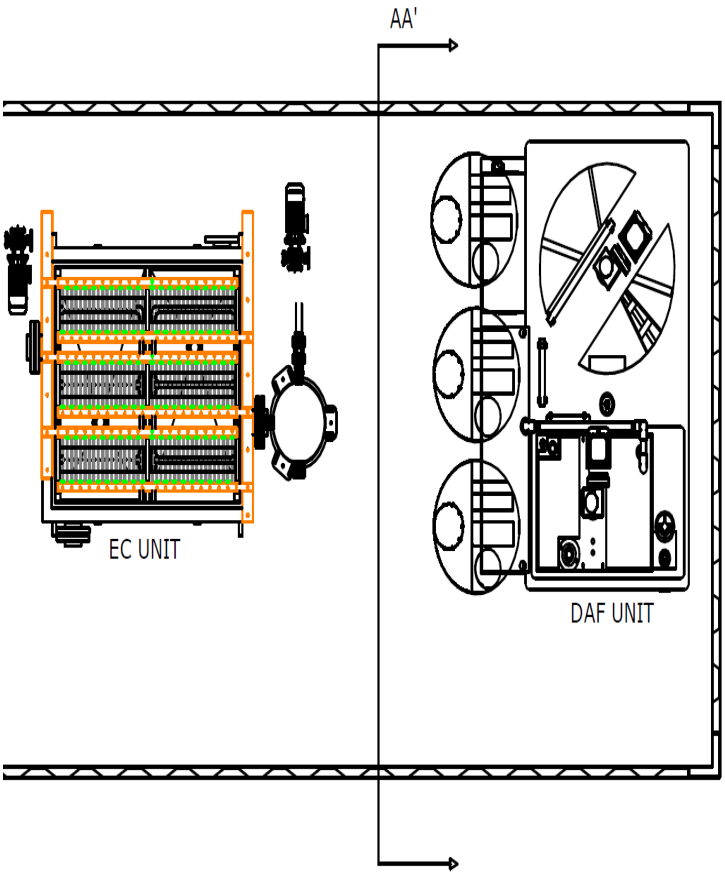

Figure 1. Schematic configuration of the pilot plant

2. 2. Analytical methods

Water was analyzed by standard methods as presented in Table 1.

Efficiency of the EC treatment was evaluated by analysing the removal of all parameters measured in Table 1, but the most important for the study was total dissolved solids (TDS).

Table 1. Analysed parameters in water and applied methods

Parameter	Method
Content of total dissolved solids, mg dm^{-3}	APAT CNR IRSA 2090 A Man 29 2003
Hardness, mg dm^{-3}	APAT CNR IRSA 2010 Man 29 2003
Total alkalinity, mg dm^{-3}	APAT CNR IRSA 2010 Man 29 2003
Content of chlorides, mg dm^{-3}	APAT CNR IRSA 4020 Man 29 2003
Content of fluorides, mg dm^{-3}	APAT CNR IRSA 4020 Man 29 2003
Content of sulfates, mg dm^{-3}	APAT CNR IRSA 4020 Man 29 2003
Total iron content, mg dm^{-3}	EPA 6020B 2014
Dissolved iron content, mg dm^{-3}	EPA 6020B 2014
Copper content, mg dm^{-3}	EPA 6020B 2014
Ammonia nitrogen content (as NH_4), mg dm^{-3}	UNI 11669:2017
Content of total suspended solids, mg dm^{-3}	APAT CNR IRSA 2090 B Man 29 2003
Oil and grease content, mg dm^{-3}	MI 30 REV.3 2018 (APAT 5160 A)
Silicium content, mg dm^{-3}	UNI EN ISO 11885:2009
Chemical oxygen demand, mg dm^{-3}	ISO 15705:2002

In research [20], the filtered cooling tower water was analyzed to determine the scale ions concentration. The analysis was performed using the APHA (2012) standard methods. Concentrations of the cations and anions were determined by using a UNICO SQ2800 UV/VIS spectrophotometer (Unico, NJ, USA) and a Dionex ICS-1000 ion chromatography system (Thermo Fisher Scientific, USA). Water pH was measured using a Fisherbrand™ FE150 pH meter (Fisher Scientific, Leicestershire, UK) and the conductivity was measured by a Laqua DS70 conductivity meter (Horiba Advanced Techno, Japan).

Via dosing pumps, anionic polymer flocculant solution VAFLOC 974 (Hangrui, China), was added continuously at a flowrate of 1 m^3 h^{-1} to the DAF unit for better segregation and separation of the particles.

3. RESULTS

Water quality analysis was conducted in a commercial pilot system in a steel manufacturing plant at three selected points:

- inlet point presenting raw water quality,
- intermediate point presenting water quality after the EC process,
- outlet point presenting water quality after the DAF unit.

Results of the measurements at the selected points are shown in Tables 2 to Table 4.

Table 2. Raw water quality at the inlet point over time

	Measurement date					
Parameters	21/03/23	22/03/23	23/03/23	27/03/23	28/03/23	29/03/23
Content of total dissolved solids, mg dm^{-3}	304	300	305	308	304	303
Hardness, mg dm^{-3}	150	162	170	154	160	150
Total alkalinity, mg dm^{-3}	80	90	87	100	90	70
Content of chlorides, mg dm^{-3}	42	43	45	46	48	46
Content of fluorides, mg dm^{-3}	0.17	0.16	0.16	0.16	0.17	0.15
Content of sulfates, mg dm^{-3}	88	89	90	94	92	89
Total iron content, mg dm^{-3}	7.2	14	9	5.1	13	9
Dissolved iron content, mg dm^{-3}	0.0084	0.0062	0.007	0.0052	0.007	0.0064
Copper content, mg dm^{-3}	0.2	0.29	0.3	0.14	0.3	0.24
Ammonia nitrogen content (as NH_4), mg dm^{-3}	0.034	0.02	0.02	0.02	0.02	0.02
Content of total suspended solids, mg dm^{-3}	72	112	105	70	115	102
Oil and grease content, mg dm^{-3}	2	2	2	12	2	6
Silicium content, mg dm^{-3}	2.255	2.108	2.3	1.76	2.1	1.9
Chemical oxygen demand, mg dm^{-3}	9	10	10	10	7	5

Performance of the studied pilot half-industrial system was monitored for 9 days, where the major focus was on understanding the influence on the total dissolved solids (TDS) removal, as that parameter can not be influenced with chemical dosing units.

When the system was put in the working mode, in the first week, parameters were changed day by day to understand the influence of each parameter on the TDS removal. On day 1 the EC system was set at 45 A and 5.1 m^2 kg s^{-3} A^{-1}, followed by 126 A and 8 m^2 kg s^{-3} A^{-1} on day 2, 85 A and 5.9 m^2 kg s^{-3} A^{-1} on day 3, while from day 4 to day 6 the system was set at 93 A and 6.3 m^2 kg s^{-3} A^{-1}.

Table 3. Water quality after the EC process at the intermediate point

	Measurement date			
Parameter	21/03/23	23/03/23	28/03/23	29/03/23
Content of total dissolved solids, mg dm^{-3}	246	212	296	268
Hardness, mg dm^{-3}	277.4	152	147	147
Total alkalinity, mg dm^{-3}	50	80	75	75
Content of chlorides, mg dm^{-3}	45	45	49	46
Content of fluorides, mg dm^{-3}	0.16	0.16	0.17	0.16
Content of sulfates, mg dm^{-3}	92	91	96	93
Total iron content, mg dm^{-3}	110	40	49	36
Dissolved iron content, mg dm^{-3}	1.5	0.036	5.1	0.012
Copper content, mg dm^{-3}	0.36	0.14	0.23	0.098
Ammonia nitrogen content (as NH_4), mg dm^{-3}	0.02	0.015	0.02	0.02
Content of total suspended solids, mg dm^{-3}	240	111	177	94
Oil and grease content, mg dm^{-3}	6	4	13	1,6
Silicium content, mg dm^{-3}	2.66	1.86	1.06	0.85

Water was analysed after the EC process to evaluate performance of this process as well.

Over the time, the effect of current density on various parameters is described above in text. With the increase in the current from 45 to 126 A the removal efficiency of total iron, hardness and total suspended solids (TSS) also increases. This is due to the higher number of ions produced on the electrodes promoting destabilization of the pollutant molecules. The removal percentage of major parameters (beside iron and hardness) maintains a plateau from current 85 to 126 A.

However, compared to the raw water, the total iron content after the EC process was increased due to the use of sacrificial electrodes made of iron.

Still, the most important parameter followed in this study, TDS, has shown a satisfactory removal trend, even after using the EC process, only (an average removal of about 16 % with a standard deviation of ~12 %).

Table 4. Water quality parameters after the DAF unit at the outlet point;

	Measurement date					
Parameters	21/03/23	22/03/23	23/03/23	27/03/23	28/03/23	29/03/23
Content of total dissolved solids, mg dm^{-3}	240	192	228	284	296	282
Hardness, mg dm^{-3}	160	146	16	151	156	153
Total alkalinity, mg dm^{-3}	80	80	1.8	75	100	75
Content of chlorides, mg dm^{-3}	42	43	43	46	46	45
Content of fluorides, mg dm^{-3}	0.15	0.16	0.16	0.17	0.17	0.17
Content of sulfates, mg dm^{-3}	88	89	90	93	95	95
Total iron content, mg dm^{-3}	4.4	16	26	6.7	1.5	5.8
Dissolved iron content, mg dm^{-3}	0.00019	0.033	0.0046	0.00576	0.0015	0.0016
Copper content, mg dm^{-3}	0.034	0.061	0.082	0.07	0.061	0.047
Ammonia nitrogen content (as NH_4), mg dm^{-3}	0.018	0.018	0.17	0.37	0.42	0.64
Content of total suspended solids, mg dm^{-3}	10	42	6	19	4	13
Oil and grease content, mg dm^{-3}	1.8	6	2	1.8	1.8	6
Silicium content, mg dm^{-3}	1.683	2.627	2.66	0.86	1.5	1.4
Content of total dissolved solids, mg dm^{-3}	6	5	6	7	4	4

At the outlet point, there was some increase in the NH_4, and this could be related to the usage of polymer flocculants that could have some quantity of the polyamide (which contains nitrogen).

Other parameters like chlorides, fluorides and sulfates were not influenced by the DAF unit operating conditions.

The TDS observed removal of around 20 to 30 % in the first several days after the DAF unit, and something less in the later period, but still almost at 10 %. The removal efficiencies were higher for copper, oil and greases, and TSS ranging between 81 to 50 %. Silicium was also significantly removed, achieving in some cases 50 % removal efficiency.

4. DISCUSSION

When designing a water treatment plant including the operational parameters it is crucial to know the raw water quality. Also, in order to evaluate the plant performance, it is necessary to monitor the inlet water quality and thus the water parameters were monitored at the moment of starting the trial of the pilot system.

Some of the key parameters affecting the outlet water quality and desirable performance on an EC process include current density, electrode material and electrode arrangement, inter-electrode distance, initial water composition and retention time. A key parameter, though, is the current density, which is the current applied per the effective electrode surface area. The current density determines the release rate of electrons as a consequence of the dissociation of metal ions from the electrode [21,22]. However, the range of current density applied varies widely for different types of the process water and its quality. Differences arise mostly due to variations in ionic interaction. The current if not applied correctly, can influence the removal of contamination. An excess of the current density, for example, can negatively affect the removal as it can enable secondary reactions. Thus, it is very important to make a pre-evaluation of the inlet water, to set the correct operation parameters.

In the present case, it is evident how various values of the current, have affected removal of the impurities.

Removal of total dissolved solids was shown to be the best at 126 A amounting to around 30 %, while on average during the complete study, removal was 17±13 %.

Ammonia nitrogen removal efficiency depends on the electrolysis time and the current density value. At both low current and electrolysis time, the ammonia nitrogen removal efficiency was lower.

The electrochemical method was used to investigate nitrite and ammonia removal from an aqueous solution in literature [23]. The results showed that removal was improved when electrolysis time in the cell was extended. The pH effect on nitrite removal was less significant than conductivity and current input. The iron electrode was unsuitable for removing nitrite due to its low removal efficiency. However insoluble electrodes such as a graphite anode and titanium dioxide cathode were appropriate for removing nitrite and ammonia in electrochemical cells. It was thought that there are two possible mechanisms involved in the process, electro-flotation and electro-oxidation, while it was concluded that more investigations need to be done [26].

Hardness removal was not successful probably due to non-optimized conditions applied during the investigation. A study in literature [24] has shown that the maximal removal efficiency at pH 7.0 and application of aluminum electrodes was achieved at the voltage of 6 m^2 kg s^{-3} A^{-1} and a reaction time of 60 min, amounting to 95.4 and 95.7 % for calcium and total hardness, respectively.

5. CONCLUSION

The electrocoagulation process presents an environmentally friendly process that can be applied for removal of various impurities. This study was conducted with the goal to examine the EC pilot system focusing particularly on TDS removal along with other water quality parameters. The system gave a positive answer for the purpose of the water treatment plants in the steel industry. The goal of this research was to decrease the TDS value so to show that by maintaining water impurities under necessary limits within water treatment plants, discharge of the water can be avoided. Still, along with some positive results, some parameters like hardness, chlorides, ammonia nitrogen and sulfates were not significantly removed. Further examination should be conducted to include a combination with some other applications (for example electro Fenton process) or by changing the electrode material and configuration of the EC process.

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Tehnologija elektrokoagulacije kao nova i napredna tehnologija za buduće izazove u postrojenjima za prečišćavanje vode u industriji čelika

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Izvod

Voda je osnovna životna potreba, i može se činiti nezamislivim zamisliti život bez nje. Uticaj na životnu sredinu, zajedno sa socijalnim i ekonomskim uticajem konvencionalnih i tradicionalnih tretmana vode u postrojenjima u industriji čelika i neizbežnom činjenicom nestašice vode, vode i pokreću prelazak na novu paradigmu u tretmanima vode. Ovo istraživanje je imalo za cilj da ispita efikasnost procesa elektrokoagulacije i jedinice za flotaciju rastvorenog vazduha za uklanjanje različitih nečistoća u vodi iz fabrike za proizvodnju čelika. Tokom proteklih decenija, elektrokoagulacija je prihvaćena kao efikasna i obećavajuća alternativna tehnologija u oblasti prečišćavanja vode. Elektrokoagulacija tehnika je usko povezana sa hemijskom koagulacijom, koja uključuje snabdevanje koagulantnih jona primenom električne struje na žrtvenu anodu (napravljenu od aluminijuma ili gvožđa) smeštenu u procesni rezervoar. Glavni fokus istraživanja je stavljen na ukupne rastvorene materije i provodljivost vode. U tradicionalnim sistemima, ukupne rastvorene materije i provodljivost vode su jedan od parametara na koje ne mogu uticati hemijske jedinice za doziranje. Specifični cilj je bio da se razumeju parametri koji utiču na efikasnost procesa elektrokoagulacije, tako da se utiče na ukupan sadržaj rastvorenih materija i smanjenje zaostalih neorganskih i organskih nečistoća, održavajući neophodnu ravnotežu vode.

Ključne reči: Ušteda vode, inovativna rešenja, zelena industrija, reciklaža, elektrohemijski proces