

# ***Removal of Manganese from Toa Vaca Reservoir Raw Water using Electrocoagulation***

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**Abstract** — *The aim of this study was to investigate the efficiency of this technique in manganese removal from the raw water at the Toa Vaca Filter Plant. Preliminary runs with different electrode materials were performed in order to determine and select the best electrode pair / configuration for manganese removal. The effect of parameters such as processing time and charge loading were investigated for the remaining stages of this study using an aluminum / aluminum electrode pair which was the combination that gave the highest manganese removal (82.88 %) during preliminary stage. Results revealed that manganese removal efficiency increases with processing time and applied charge loading although voltage may also affect the electrocoagulation process. In addition, water turbidity was also impacted positively obtaining removal up to 92.89 %. Considering the obtained efficiency in the present study, electrocoagulation technique may be suggested as an effective alternative technique in manganese removal.*

**Key Terms** — *Charge Density, Electrode, Electrocoagulation. Manganese.*

## **INTRODUCTION**

This section provides an overview of manganese element regarding its origin and possible effect on human life. Toa Vaca Treatment Plant is also overviewed in this section as well as past researches on manganese removal and electrocoagulation process.

### **Manganese Element**

Manganese (Mn) is silver-gray metallic element naturally presented in many types of rocks. Weathering of manganese bearing materials is

known as the main natural source of this element in groundwater. The Environmental Protection Agency (EPA) established for manganese, which is an essential element in human diet, a Secondary Maximum Contaminant Level of 0.05 mg/L. This limit is based on aesthetic concerns only. Manganese solids may form deposits within pipes and break off as black particles that give water an unpleasant appearance, odor and taste. Even though, EPA has recently indicated that there is a health concern with high levels of manganese in drinking water Manganese may affect neurological and muscle in humans. A recent study suggested that higher levels of exposure of manganese in drinking water are associated with increased intellectual impairment and reduced intelligence quotients in school-age children [1]. While EPA is developing their health-based drinking water standard, some regional offices such as the Department of Environmental Service of New Hampshire has adopted an interim health based standard for manganese of 0.84 mg/L. In addition, water containing excessive amounts of manganese may cause the staining of plumbing fixtures or laundry [2].

### **Toa Vaca Treatment Plant**

Manganese usually present a though challenge to Water Treatment Plants including the Toa Vaca Filter Plant. This plant receives raw water with moderate manganese levels from the Toa Vaca reservoir located at the Villalba municipality in Puerto Rico. Manganese concentration in raw water was in the range of 0.220 – 0.454 mg/L during interval time of this study. The mentioned plant is designed to treat 7.5 MG of water daily. As a pre-oxidation stage, Potassium Permanganate (KMnO<sub>4</sub>) is injected in raw water prior to the

entrance of Toa Vaca Filter plant. These oxides and other sediments from the coagulation / flocculation process are then removed by ultrafiltration membranes resulting in a Manganese concentration at the effluent of approximately 0 mg/L.

### Manganese Removal

There are a variety of methods available to remove manganese from water. The most common technique in most circumstances is oxidation / filtration. Common oxidizing agents include chlorine, potassium permanganate, ozone and hydrogen peroxide. Manganese has also been treated by ion exchange, oxidation filters and reverse osmosis. Chlorine oxidation in conjunction with activated carbon has yielded manganese removal up to 92% [3]. Pre-oxidation with potassium permanganate has been reported to remove manganese up to 72% [4]. In addition, other technologies such as electrocoagulation could be seen as an alternative for several species removal including manganese.

Electrocoagulation is the process of destabilizing suspended, emulsified or dissolved contaminants in aqueous medium by introducing an electrical current (DC) into the medium. Figure 1 shows a schematic representation of the electrocoagulation process. Electrocoagulation systems have been in existence for many years [5] using a variety of anode and cathode geometries, including plates, balls, fluidized bed spheres, wire mesh, rods, and tubes. Researches from recent years have increased considerably focused on different species removal from water and wastewater. This technique is a complex process with a multitude of mechanisms operating synergistically to remove pollutants from the water in which electrical current provides electromotive force to drive the chemical reactions. When reactions are driven or forced, the elements or compounds will approach the most stable state. Generally, this state of stability produces a solid that is either less colloidal or less emulsified (or soluble) than the compound at equilibrium values.

As this occurs, the contaminants form hydrophobic entities precipitate and can easily be removed by a number of secondary separation techniques. Due to the induced current, electrocoagulation often neutralizes ion and particle charges allowing contaminants to precipitate. Most of these researches establish the charge density and electrode material as very important design factors for the electrocoagulation process [6]-[9].

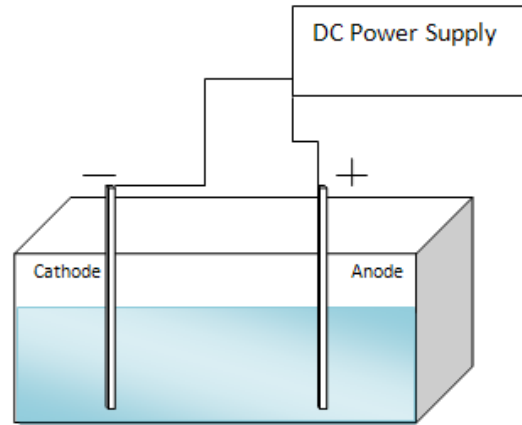


Figure 1  
Electrocoagulation Process Scheme

Manganese removal by electrocoagulation has been previously studied (Shafaei et. Al, 2010) obtaining removal percents approximately up to 99.% [10]-[11]. Employment of electrocoagulation for the removal of many other pollutants such as Arsenic have been studied in recent years. Recent researches have yielded Arsenic removal up to 98% using electrocoagulation with iron and /or aluminum electrodes [9], [12]. Nickel, copper, zinc and chromium have also been treated with electrocoagulation obtaining removal percents up to 98% [6], [13]. Reference [14] shows hardness removal using aluminum electrodes obtaining removal efficiency of 95.6%. In addition color removal and COD removal have also been documented to be removed by electrocoagulation [15]-[16]. In addition, contaminants such as heavy metals, BOD, Total Suspended Solids and Total Coliform could be easily removed by this technique. Thus, it is very important to continue developing this technique as an alternative to many other conventional treatment techniques.

## METHODOLOGY

This section provides an overview of the methodology used such as sample collection, procedure and analytical methods.

### Sample Collection

Water samples were taken from the Toa Vaca Reservoir raw water testing tap. A five (5) gallons container was previously washed with raw water from the same tap before taking the sample. The purpose of using a five (5) gallon container was only for convenience to avoid using multiple containers which would have lead to a thoroughly washing process. Water collected was immediately tested for Manganese concentration level.

### Procedure

Preliminary experiments were conducted at a constant processing time with different material combination as cathode and anode to determine the best possible combination for manganese removal. Combinations of iron/iron and aluminum/aluminum were included in the initially experimental runs as shown in Table 1. Anode and cathode electrodes were placed 3.25 inches (8.26 cm) apart in a rectangular container filled with 0.5 gallon of raw water from the Toa Vaca Reservoir. Direct current (DC) was provided using a variable DC power supply. Processing time (30 minutes) as well as voltage (5 V) and current (1.5 A) were set constant for all preliminary experimental runs shown in Table 1. However, charge density varied for some electrodes due to a different size and thus different submerged volume of some electrodes within the water. Zinc (Zn) and iron (Fe) electrode have an approximate equal volume of 5.0 cm<sup>3</sup> of which a total 1.67 cm<sup>3</sup> was submerged into raw water during the preliminary electrocoagulation experiments. Single titanium (Ti), nickel (Ni) and/or aluminum (Al) electrode have an approximate equal volume of 2.88 cm<sup>3</sup> of which 0.96 cm<sup>3</sup> was submerged into water during the preliminary electrocoagulation experiments. However, the total submerged volume for each of

these three electrodes (Ti, Ni, Al) was about 1.92 cm<sup>3</sup> since two electrodes were used due to compensate for a smaller size compared to Zn and Fe electrodes. Processes water was then filtered by gravity using 2 microns filter paper. Before each experiment, the electrodes and container were washed with isopropyl alcohol 70/30 (70% IPA, 30% Deionized water) to remove any scale. The real representation of the electrocoagulation cell used for this research purpose is shown in Figure 2.

An aluminum / aluminum configuration was chosen as the optimum pair for the continuation of the remaining research activities. Subsequent research included an evaluation of the electrode pair at a constant charge density varying only the processing time. The objective of evaluate the pair at a constant charge density but different processing time was to verify the removal of with respect to time. In addition, the pair was evaluated at different charge densities to determine the effect of manganese removal with respect to charge density.



**Figure 2**  
**Electrocoagulation Cell with Aluminum Electrodes**

### Analytical Method

Manganese concentration level was measured using Hach method 8149 (Manganese Low Range PAN method) adapted from previous work [17]-[18]. The PAN method is a highly sensitive and rapid procedure for detecting low levels of manganese. An ascorbic acid reagent is used initially to reduce all oxidized forms of manganese to Mn<sup>+2</sup>. An alkaline-cyanide reagent is added to

**Table 1**  
**Preliminary Experimental Runs with Different Electrode Pairs**

Experimental Run	Cathode / Anode	Processing Time (min)	Current (A)	Charge Density (C/cm <sup>3</sup> )	Final Mn (mg/L)	Removal %
Mn in Raw Water = 0.289 mg/L						
1	Zinc / Iron	30	1.5	1617	0.153	47.06
2	Iron / Iron	30	1.5	1617	0.109	62.28
3	Titanium / Iron	30	1.5	1617	0.099	65.74
4	Nickel / Iron	30	1.5	1617	0.116	59.86
5	Aluminum / Iron	30	1.5	1617	0.086	70.24
Mn in Raw Water = 0.237 mg/L						
6	Iron / Zinc	30	1.5	1617	0.202	14.77
7	Iron / Titanium	30	1.5	1406	0.044	81.43
8	Iron / Nickel	30	1.5	1406	0.059	75.11
9	Iron / Aluminum	30	1.5	1406	0.054	77.22
10	Aluminum / Aluminum	30	1.5	1406	0.042	82.88

mask any potential interference. PAN Indicator is then added to combine with the Mn<sup>2+</sup> to form an orange-colored complex. Turbidity and pH measurements were performed using a turbidity meter and a pH meter respectively.

## RESULTS AND DISCUSSION

This section provides an overview of manganese element, Toa Vaca Treatment Plant and past researches on manganese removal and electrocoagulation process.

### Comparison of Electrode Materials

In any electrochemical process, electrode material has significant effect on the treatment efficiency. Therefore, appropriate selection of the material is important. The electrode material for drinking water treatment should also be non-toxic to human health [9]. Hence Iron, Aluminum, Zinc, Nickel and Titanium were chosen as electrode material for preliminary experimental runs as these are non-toxic and readily available. Iron and Aluminum electrodes have been widely used in recent electrocoagulation researches [7], [9], [12]. In addition, manganese removal by electrocoagulation has been studied previously using Aluminium electrodes [10]-[11].

Preliminary runs 1 through 5 were performed on a different day to that of preliminary runs 6 through 10. Thus, the raw water manganese concentration was slightly different due to the

different sample collection date. Manganese concentration in raw water for runs 1 through 5 was approximately 0.289 mg/L while a concentration of 0.237 mg/L was found in raw water used for run 6 through 10. Results for preliminary runs shown in Table 1 could lead to several conclusions. It can be concluded as per results shown in Table 1 that a Zinc electrode (anode or cathode) is not effective for manganese removal. In general terms, manganese is best removed when iron is used as a cathode instead of anode. Even though, the aluminum / aluminum pair brought a manganese removal of 82.28 % which was the highest among the electrode pair tested during preliminary runs. The final manganese concentration obtained for preliminary run 10 was 0.042 mg/L which comply to the 0.05 mg/L Secondary Limit established by EPA.

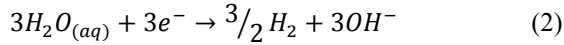
According to Figure 3, other electrode pair configurations such as iron / titanium and iron / aluminum (iron as cathodes in both cases) may provide very comparative manganese removal percent to that obtained for the aluminum / aluminum. Obtained manganese removal for the iron / titanium and iron / aluminum pairs were 81.43 % and 77.42% respectively. However, an aluminum / aluminum pair was chosen for the rest of research activities due it was the combination that brought better results and additionally electrode material is cheaper than others. Equations (1) through (4) have been suggested as the reactions

occurring at the electrocoagulation process using aluminum electrodes [7], [12]:

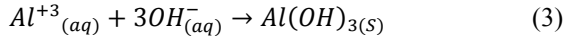
At the aluminum anode:



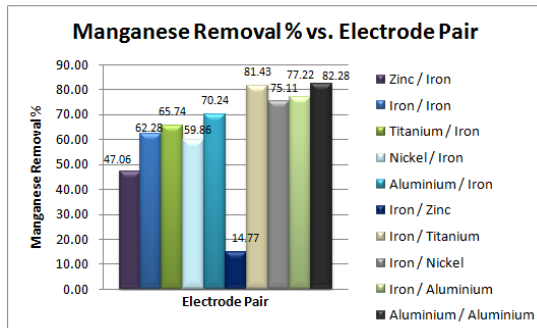
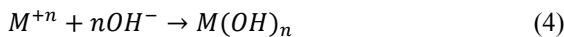
At the cathode:



At the solution:



Furthermore, the hydroxide ions formed induce the precipitation of metal ions as corresponding hydroxides and co-precipitation with aluminum hydroxides:



**Figure 3**  
Manganese Removal According Electrode Pair Configuration

Previous researches using aluminum electrodes have revealed manganese removal up to about 99% [10], [11]. Even though, those researches do not provide a realistic representation of manganese removal thru electrocoagulation process since those studies were performed using synthetic manganese polluted wastewater. It means that beside manganese, no other specie was competing for the aluminum and hydroxides ions produced from the electrocoagulation process. In addition, processed water in the mentioned researches was filtered using a 0.2 microns syringe filter.

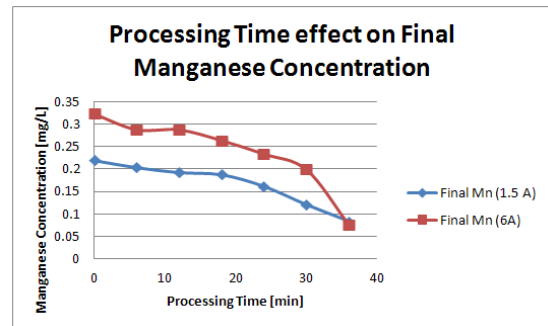
### Effect of Processing Time

The effectiveness and dependence of electrocoagulation process according to processing time it is well documented through recent electrocoagulation researches [7], [9]-[13]. The

efficiency of EC process depends directly on the amount of aluminum and hydroxide ions produced by the electrode dissolution which according to the Faraday's Law of electrolysis depends on time and current intensity. Faraday's Law of electrolysis is given by:

$$m = \frac{ItM}{zF} \quad (5)$$

where m is the amount of ion produced from electrocoagulation cell, I is the current intensity, M is the molecular weight of aluminum or hydroxide ion ( $g \text{ mol}^{-1}$ ), z is the number of electrons transferred in the reaction (3 for aluminum, 1 for hydroxide) and F is the Faraday constant ( $96,486 \text{ C mol}^{-1}$ ).



**Figure 4**  
Final Manganese Concentration Through Different Processing Time

Two different current intensities (amperes) were used to demonstrate the relationship and dependence of manganese removal with respect to time by electrocoagulation using aluminum electrodes. From both Figure 4 and Figure 5 it is appreciated that final manganese concentration decrease with increasing processing time. Table 2 shows results for experimental runs 11 – 22 at different processing time. As processing time increase, more  $Al^{+3}$  and  $OH^{-}$  ions are generated and consequently react to form various monomeric and polymeric species such as  $Al(OH)^{+2}$ ,  $Al(OH)_2^{+}$ ,  $Al_2(OH)_2^{+4}$ ,  $Al(OH)^{-4}$ ,  $Al_6(OH)_{15}^{+3}$ ,  $Al_7(OH)_{17}^{+4}$ ,  $Al_8(OH)_{20}^{+7}$ ,  $Al_{13}O_4(OH)_{24}^{+7}$ ,  $Al_{13}(OH)_{34}^{+5}$  which finally result in situ formation of gelatinous  $Al(OH)_3$ . All these species, specially aluminum hydroxide ( $Al(OH)_3$ ), have strong affinity for dispersed



**Table 2**  
**Experimental Runs at Different Processing Time Using Aluminum Electrode Pair**

Experimental Run	Processing Time (min)	Voltage (V)	Current (A)	Final Mn (mg/L)	Removal %
Mn in Raw Water = 0.220 mg/L					
11	6	5	1.5	0.204	7.27
12	12	5	1.5	0.193	12.27
13	18	5	1.5	0.188	14.55
14	24	5	1.5	0.162	26.36
15	30	5	1.5	0.121	45.00
16	36	5	1.5	0.084	61.82
Mn in Raw Water = 0.323 mg/L					
17	6	17	6	0.288	10.84
18	12	17	6	0.288	10.84
19	18	17	6	0.264	18.27
20	24	17	6	0.234	27.55
21	30	17	6	0.199	38.39
22	36	17	6	0.076	76.47

particles and act as absorbents and/or traps for metal ions eliminating them from water [12]-[13]. Figure 6 shows gelatinous flocs produced from electrocoagulation process.

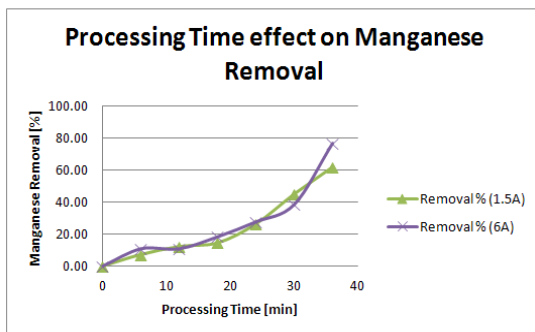
It is important to note that manganese removal percent obtained during this stage using the same process parameters of preliminary runs (Processing time = 30 min, Current Intensity = 1.4 A) was just 45%. This removal percent was too low compared to 82 % obtained during preliminary run 10. Several factors could have been caused such difference including electrodes wearing. In addition, chemical composition of the water may vary from day to day due to climate conditions (i.e. heavy rain) since water used is raw water the Toa Vaca reservoir. Thus, manganese reading results may be affected depending on the concentration of other species such as iron, magnesium and calcium [17].



**Figure 6**  
**Gelatinous Flocs from Electrocoagulation Process**

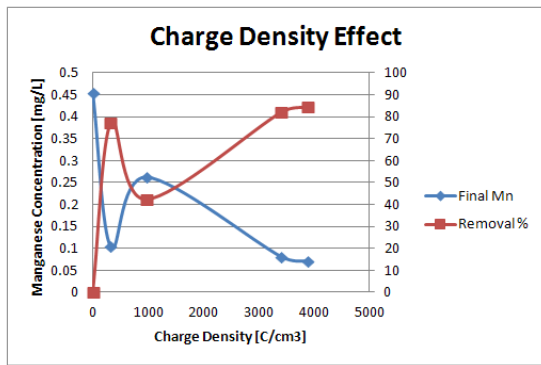
### Effect of Charge Density

As mentioned earlier the success of electrocoagulation depends greatly on the amount on aluminum and hydroxide ion that form in the solution. The submerged volume (or area) of the electrodes as well as the charge density is a limiting factor to the amount of those ions formed in the water to be treated. Thus, the electrocoagulation cell was modified to increase the submerged surface volume of aluminum electrodes in the raw water since only about a third of the electrodes volume was submerged in raw water during previous runs. The modification consisted in changing the rectangular container used in the previous (1 – 22) runs by a square container. The resulted submerged volume of a single aluminum electrode is 2.08 cm<sup>3</sup> and a total of 4.16 cm<sup>3</sup> for the two electrodes used.



**Figure 5**  
**Manganese Removal Percent Through Different Processing Time**

Charge density, which has been addressed to be a very important electrocoagulation parameter is defined as the electric charge (coulombs = current x time) per unit of volume of space [6], [8]. According to these previous researches, electrocoagulation efficiency tends to increase with at higher charge densities. This parameter could be adjusted with processing time or current intensity. Thus, experimental runs shown in Table 3 were performed at different current intensity to obtain different charge densities. The dependence of manganese removal of charge density is shown in Figure 7.



**Figure 7**  
Manganese Removal Percent Through Different Processing Time

Figure 7 evidenced, excluding experimental run 24, that manganese removal percent increased with an increase in charge density. This could also be interpreted as processing time required to reach certain manganese levels decreases with a charge loading increase. Similarly, manganese removal percent tends to increase (excluding experimental run 24) with charge density. Interesting data is revealed in Table 3 which shows that the experimental run 24 was performed at the smallest

voltage (5 V) during that research stage due to equipment constraints. Although voltage does not affect charge density, it has been reported to contribute strongly to electrocoagulation efficiency. Reference [6] reported a proportional relationship between arsenate removal and voltage applied during electrocoagulation process. In addition, applied voltage also affected chromium removal (Bazrafshan et. Al., 2008) using both iron and aluminum electrodes [7]. Thus, it is appropriate to concluded that the 5 V applied in experimental run 24 was fundamental in the results obtained for the final manganese concentration and its removal percent.

### Effect of Electrocoagulation on pH

According to Faraday Law of Electrolysis the number of aluminum and hydroxide ions are increased by current intensity. Thus, based on Faraday Law of Electrolysis and equation 2 it was expected higher pH values with an applied current increased. The possible effect on processed water pH due electrocoagulation was determined using experimental runs 23 – 26 which its raw water measured pH was about 7.83. As mentioned earlier, charge density through these experimental runs was increased by increasing applied current. The pH value for experimental run 23 remained constant at 7.83 basically due to the minimum charge density received controlled by a current of 0.5 A. However, subsequent pH values for the remaining runs increased with the charge density (or current) applied. Results of pH values are shown on Table 3.

**Table 3**  
Experimental Runs at Different Current Intensity Using Aluminum Electrode Pair

Experimental Run	Voltage (v)	Current (A)	Processing Time (min)	Charge Density (C/cm <sup>3</sup> )	Final [Mn] (mg/L)	Removal %	pH	Turbidity (NTU)
Mn in Raw Water = 0.454 mg/L, pH=7.83, NTU=2.84								
23	20	0.5	45	324.52	0.104	77.09	7.83	0.468
24	5	1.5	45	973.56	0.262	42.29	7.98	0.32
25	17	5.25	45	3407.45	0.081	82.16	8.36	0.268
26	24	6	45	3894.23	0.071	84.36	8.42	0.202

### Effect of Electrocoagulation on Turbidity

Colloidal particles stability in water is determined by its physicochemical properties. Colloids composed of similarly charged particles repel each other and consequently repulsive forces induce dispersion to remain stable. However, if the particles have little or no repulsive force then some instability mechanism will eventually take place e.g. flocculation, aggregation etc. In certain circumstances, the particles in a colloidal dispersion may adhere to one another and form aggregates of successively increasing size that may settle out to the bottom or precipitate to the top. In order to maintain electroneutrality, oppositely charged ions are attracted to the (charged) pollutant particles. The attraction of counter ions to a negatively charged pollutant forms an electric double layer divided into a Stern and diffuse layer. Within the diffuse layer there is a notional boundary inside which the ions and particles form a stable entity. When a particle moves (e.g. due to gravity), ions within the boundary move it. Those ions beyond the boundary stay with the bulk dispersant. The potential at this boundary known as zeta potential is illustrated on Figure 8 (taken from [www.substech.com](http://www.substech.com)). The magnitude of the zeta potential gives an indication of the potential stability of the colloidal system. If all the particles in suspension have a large negative or positive zeta potential then they will tend to repel each other and there is no tendency to flocculate. However, if the particles have low zeta potential values then there is no force to prevent the particles coming together and flocculating.

Effect of electrocoagulation in turbidity was studied during experimental runs 23 – 26; turbidity of raw water was 2.84 NTU. Figure 9 indicated that turbidity was positively impacted by applied charge loading obtaining removal up to 92.89 %. Reference [19] revealed that turbidity removal by electrocoagulation is a function of processing time and current and consequently charge loading. As mentioned in previous sections, the amount of aluminum and hydroxide ions increases with time

and applied charge loading, consequently the coagulant dose also increases  $Al(OH)_3$ . Recent researches have revealed that zeta potential of the colloidal system approach to zero (isoelectric point) up to a certain coagulant dose increases [20]-[21]. The isoelectric point is normally the point where the colloidal system is least stable and thus more susceptible to aggregate. That means that colloids are best removed at this point.

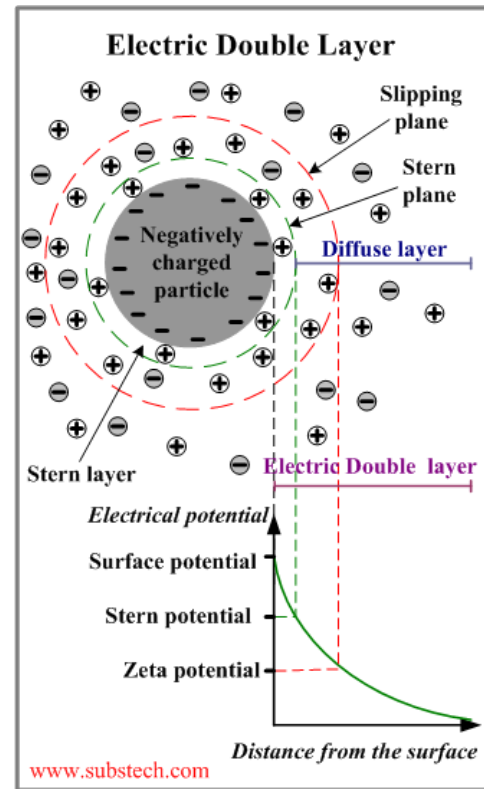


Figure 8  
Electric Double Layer

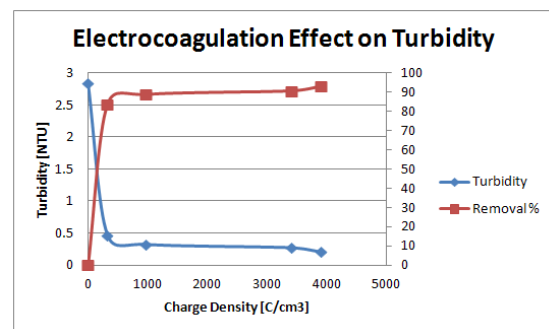


Figure 9  
Electrocoagulation Effect on Turbidity



## RESULTS AND DISCUSSION

Electrocoagulation process for manganese removal was evaluated initially using different electrode materials / pair configurations at a constant processing time with only a slight difference in charge loading due to respective electrodes dimensions. The configuration that best removed manganese was aluminum / aluminum which offered initially a removal percent of 82.88%. The raw water manganese concentration at that specific run was 0.289 mg/L while the final manganese concentration corresponding to that removal percent was 0.042 mg/L which met the secondary water drinking standard of 0.05 mg/L. Subsequent experimental runs which included processing time and charge loading variations, were performed using aluminum / aluminum pair.

Manganese removal increased with respect to processing time and charge loading. However, voltage applied could also be fundamental parameter for electrocoagulation process. The maximum removal obtained was 84.36 % at processing time of 45 minutes and a charge density of 38.94 C/m<sup>3</sup> (6 A) which belongs to experimental run 26. A higher removal percent would be expected considering that the charge loading used was considerably higher than that applied in experimental run 10 which was the one obtaining 82.88 % of manganese removal. However, a possible reason for obtaining a removal of just 84.36 % was the raw water manganese concentration. Manganese concentration in raw water at experimental run 26 was considerably greater than that of experimental run 10 (0.454 mg/L vs. 0.289 mg/L). Reference [10] reported a manganese removal efficiency reduction with an initial concentration increase. In addition, reference [6] reported heavy metals removal rate reduction at higher initial concentrations.

The results showed that electrocoagulation is a feasible process for removing manganese from aqueous solutions. The manganese ions are removed by direct reduction at the cathode surface as hydroxides by the hydroxyl ions formed at the

cathode via water electrolysis and by co-precipitation with the aluminum hydroxides. Additionally, electrocoagulation impacted positively the final turbidity obtaining a removal efficiency of approximately 92.89%. Nevertheless, further studies should be carried out to optimize the electrocoagulation process and confirm the practical feasibility of this treatment method for manganese removal and turbidity. The dependence of manganese removal on processing time, charge density and possibly voltage should be confirmed on future researches. It is important to determine in future researches the effect that raw water pH and other conditions such as electrode wearing have on manganese removal by electrocoagulation. In addition, it would be interesting to evaluate manganese removal efficiency by electrocoagulation researches using other pair configuration of electrode materials such as aluminum / titanium. Turbidity removal should be investigated using water with higher turbidities than the used in this study. It is clear that this electrocoagulation process is an alternative to chemical coagulation but however, feasibility of this process at a larger scale should continue to be studied.

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