

# Simultaneous Removal of Turbidity and Humic Acid Using Electrocoagulation/Flotation Process in Aqua Solution

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## Abstract

In this study, the applicability of the Electrocoagulation/Flotation (ECF) process in batch operation was investigated for the simultaneous removal of turbidity and Humic acid (HA) using Fe and Al electrodes. The effects of solution pH (3 - 12), electrical potentials (10 - 30 V), initial turbidity concentration (300 - 1200 NTU), and reaction time (10 - 30 minutes) with or without HA were investigated in an attempt to achieve higher turbidity removal efficiency. The batch experimental results revealed that with initial turbidity of 300 NTU, at voltage of 30 V, after 30 minutes reaction times, and at pH values of 6 and 8, the ECF process for Fe and Al electrodes removed over 97% and 88% of turbidity, respectively. The percentage of turbidity removal from solution dropped with a decrease in voltages for both electrodes. The results displayed that the Fe-Fe electrode arrangement attained the highest performance for turbidity removal rate. As a result, ECF process was shown to be a very efficient, cost-effective, and promising process for efficient treatment of high turbid water. Regarding HA, the results showed that in ECF process over 67% and 43% of UV254 has been removed for Al and Fe electrodes, respectively at the optimum pH, 30 minutes reaction time and 30 V applied voltage. Thus, it can be considered that Fe and Al are the best electrodes for removing turbidity and HA, respectively.

**Keywords:** Water Pollution, Electrocoagulation/Flotation, Turbidity, Humic Acids

## 1. Introduction

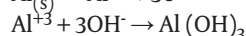
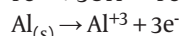
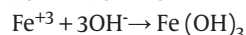
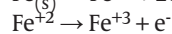
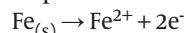
Surface water generally contains dissolved, suspended, and colloidal particles. Among water-contaminated parameters, colloidal particles have always been stable and, so removing colloidal particles is the first step of treating raw water (1). Due to limitation of conventional water treatment procedures, using coagulation and sedimentation, presedimentation and or roughing filter followed by coagulation sedimentation and filtration are commonly proposed for treating high water turbidity. However treating of high turbid water with these approaches imposes a challenging work due to the high operation and maintenance problems (2). Humic substance (HS) as a type of well-defined natural organic matter (NOM) is universally present in natural water. It not only affects the odor, color, and taste of water but also reacts with chlorine during drinking water treatment process and produce disinfection by products (DBPs) with potential adverse health problems (3-7). Hence, much attention has been focused on the removal of HS in water treatment.

Several methods are recently applied for removing HS and colloidal material each one alone from raw surface water such as chemical precipitation using synthetic polymers (5) or metal salts (4), membrane filtration (7), adsorption (6, 8), ozonation (9), and so on. However, there are only a few technologies reported for simultaneous removal

of turbidity and humic acid from contaminated water.

Coagulation/flocculation technologies have been widely applied in water treatment and are known for their capability of destabilizing and flocculating colloids and HS using inorganic metal salts, i.e. iron and aluminum as HS are large organic molecules that carry a negative charge (10-13). An alternative method to the coagulation/flocculation technology is ECF process in which coagulation and chemical precipitation are not conducted by delivering chemicals, called coagulants to the system, but via electrodes in the reactors. This procedure is simple in operation, proven, efficient, and feasible, especially for treating contaminated water in which the coagulating agents are generated by Al and or Fe electrodes (2-7). This treatment process is done by dissociation of electrodes and forming a range of coagulant agents and metal hydroxides as well as production of gas bubbles for easier removal of all pollutants.

The main electrode reactions occurring for Al and Fe in this process are as follows:



Recently, investigations have been justified on the treatment of water, especially for removing turbidity and HS using electrocoagulation process (5, 11). Among the electrocoagulation processes only a few research has been conducted on water treatment by Electrocoagulation/Flotation (ECF) process for removing turbidity (14-16). In addition our literature review revealed that few studies have been done on simultaneous removal of turbidity and Humic Acid HA using electrocoagulation technology (5) and there are no published articles about using EFC for removing of turbidity and HA.

Hence, the main objective of this study was to evaluate treatability of synthetically prepared water with turbidity and HA using electrocoagulation and ECF process and determining its effective parameters, i.e. different voltages, operating time, pH, and initial turbidity. During the experiments, the process was carried out with iron and aluminum electrodes separately.

## 2. Materials and Methods

### 2.1. Preparation of Reagents

The HA was obtained as a commercial reagent grade solid (Sigma Aldrich Chemical Co, USA). Stock solutions were prepared from 1 g of dry HA product dissolved into 1000 mL deionized water. In order to make water with expected turbidity (300, 500, 800, and 1200 NTU), bentonite powder (Sigma Aldrich Chemical Co.) was added into NaClO<sub>4</sub> solution (ionic strength = 0.01N). NaClO<sub>4</sub> solution was used in the preparation to provide background ionic strength in the distilled water.

### 2.2. Experimental Setup and Procedure

Figure 1 shows schematic diagram of ECF process in a batch mode consisted of a 6-L Plexiglas reactor (300 × 200 × 100 mm in dimensions) in which 3 anodes and 3 cathodes with dimensions of 200 × 20 × 2 mm made of iron or aluminum plates were connected to a DC power supply (ERAM TRONICS, 405 HD) providing voltages of 0 - 30 V. To enhance flotation, the air was pumped through a central valve into the bottom of reactor. ECF process was carried out at different voltages (10, 20, and 30 V), various pH values (3 to 12), and with different initial turbidities. In each run of experiments, 5500 mL of synthetic contaminated water containing different values of pH, turbidity, and HA was transferred into each of 6000 mL Plexiglas beakers with 6 plates used as coagulants sources.

The pH of synthetic water was adjusted with 0.1 mol/L H<sub>2</sub>SO<sub>4</sub> and 0.1 mol/L NaOH. Before ECF tests and in order to avoid any interference, all electrodes were washed with distilled water and then cleaned with 10% sodium hydroxide solution.

### 2.3. Analysis

Turbidity was measured by nephelometric method using each testing condition; the samples were taken every

5 minutes from 5 cm below the solution turbidimeter (2100 P, HACH Co) as described in the standard methods (17). HA value in collected water sample was determined using a UV spectrophotometer (DR 5000, HACH Co.) at an absorbance wavelength of 254 nm after filtrated through a membrane filter with the pore size of 0.45 μm. In this work, the removal of UV254 absorbance was used to evaluate the HA removal percentage. The relationship between UV254 absorbance and HA concentration is liner as shown in Figure 2, constituting a basis of conversion of UV254 to HA concentration. Also, the color absorbance was determined using spectrophotometer (DR 5000, AHCH Co.) at an absorbance wavelength of 400 nm (17). All experiments were carried out at room temperature and the average of 3 replicate experiments was reported. The turbidity and HA removal efficiency was calculated by Equation 1:

$$(1) \quad RE (\%) = \frac{(C_0 - C)}{C_0} \times 100$$

Where C<sub>0</sub> and C are water turbidity and HA before and after examination, respectively.

## 3. Results and Discussion

### 3.1. Effect of pH

The pH of the aqueous solution has a key role in ECF process, forming different metal hydroxide species, and removal mechanism of ions and pollutants (4, 7). For investigating the effect of initial solution pH range from 3 to 12 on turbidity removal efficiency in ECF process, experiments were performed at initial turbidity of 1200 NTU, applied voltage of 30V, separately with Al and Fe electrodes. Obtained results are shown in Figure 3. According to the data plotted in Figure 3 the removal efficiency of turbidity increased linearly from 54% at a pH of 3 to 97% with an increase of pH to 6 for Fe electrodes. It was observed that when the pH value increased from 7 to 12, the turbidity removal efficiency gradually decreased to 33% at pH 12.

This event can be explained by observing Fe electrode reaction in different pH values. In acidic solution (pH < 5), the major formed hydroxide is Fe(OH)<sub>2</sub> while in alkaline solution, Fe(OH)<sub>3</sub> dominates and in neutral solution there are combinations of Fe(OH)<sub>2</sub> and Fe(OH)<sub>3</sub>. Fe(OH)<sub>2</sub> and Fe(OH)<sub>3</sub> have high potential to produce floc, and high level of these hydroxides (in neutral pH) increase the removal efficiency of pollutants from aqueous solutions. Hence, the high removal efficiency was found in the pH of 6 for Fe electrodes.

Figure 3 shows that the maximum removal efficiency of turbidity (88%) was obtained for Al electrodes at pH values of 8 to 9. In this condition, the maximum turbidity removal efficiencies were 44% and 54% for initial pH value of 3 and 12, respectively.

This reaction might be explained by the aluminum species in the solution where cationic hydrolysis species such as Al<sup>3+</sup> and Al(OH)<sub>2</sub><sup>+</sup> predominate at pH < 4. When pH

increases and reach from 4 to 9, monomeric species such as  $\text{Al}(\text{OH})_2^{+2}$ ,  $\text{Al}(\text{OH})_2^{+4}$  and eventually polymeric species like  $\text{Al}_{13}(\text{OH})_{34}^{+5}$  and  $\text{Al}_6(\text{OH})_{15}^{+3}$  are generated which finally transform into precipitate amorphous  $\text{Al}(\text{OH})_3$  (7, 10). When the aqueous pH value is higher than 9, the  $\text{Al}(\text{OH})_4^-$  specie increases at the expense of  $\text{Al}(\text{OH})_3$  (4). Also, the result revealed that maximum and minimum turbidity removal efficiency was obtained at pH values of 6 and 12, respectively.

### 3.2. Effect of Different Voltages and Reaction Time

The amount of coagulating ions released from anode into the solution in an electrocoagulation reactor is strongly affected by the applied voltages, especially on the rate of turbidity removal (3-8, 18). Therefore, the influence of electrical potentials (10, 20, and 30 V) on the performance of ECF process was examined for removal of turbidity versus reaction time for Al and Fe electrodes. According to Figure 4, by increasing applied voltages from 10 to 30 V, speed and efficiency of removal process increased, while reaction time decreased in both Al and Fe electrodes. In the current study, applied voltage of 30 V in reaction time of 30 minutes produced the maximum turbidity removal efficiency and is the suggested applied voltage for other experiments. Other studies reported the same findings too (3-5). However, at the same applied voltage and reaction time, Fe electrodes achieved higher removal efficiency compared to Al ones. According to Figure 4, around 81%, 90%, and 97% of 300 NTU (as initial) turbidity was removed in 10, 20, and 30 V for Fe electrode, respectively, whereas 79%, 81%, and 87% turbidity removal were achieved for Al electrodes in 10, 20, and 30 V, respectively. It has been shown that at optimum pH value, applied voltage of 30 V, and 30 minutes operation time, maximum turbidity removal efficiencies of 97% and 86% are observed for Fe and Al electrodes, respectively.

### 3.3. Effect of Different Initial Turbidity

In coagulation process, the concentration of initial turbidity is one of the most important parameters which not only affects turbidity removal efficiency but also determine the coagulant dosages (13, 18, 19). On the other hand, different turbidity amounts might occur in different regions and different seasons. Hence, the effectiveness of ECF process was evaluated with Fe and Al electrodes and different initial turbidity ranges from 300 to 1200 NTU. According to Figure 5, with increase of initial turbidity, removal efficiency of turbidity decreases gradually for Fe and Al electrodes. This behavior may be attributed to insufficient amount of released iron and aluminum ions from main electrodes for the removal of higher initial turbidity which is in agreement with the results obtained from algae removal by ECF process with iron electrodes (5, 20) and  $\text{Fe}^{+2}$  ions using EC process.

In the current study, to optimize the effects of initial turbidity, the experiments for turbidity removal efficiency were carried out in a wide range of high initial turbidity from 300 to 1200 NTU at optimum pH and applied volt-

age for ECF process with Fe and Al electrodes (Figure 5). It has been shown that in initial turbidity of 300 NTU, the maximum removal efficiency was 38% in the reaction time of 5 minutes and 97% for 30 minutes with Fe electrodes. With increase of initial turbidity to 500, 800, and 1200 NTU, turbidity removal efficiency decreased and reached to 94%, 90%, and 87% in reaction time of 30 minutes, respectively.

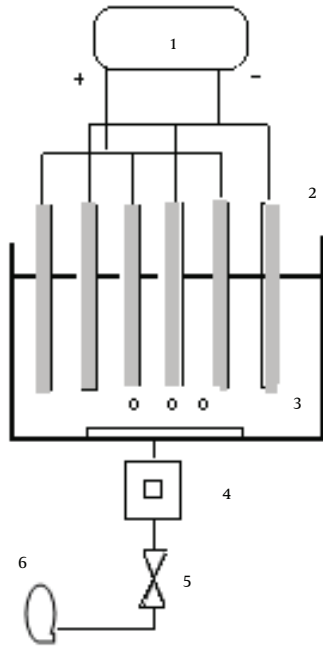
In addition, for Al electrodes, with increase of initial turbidity from 300 to 1200 NTU, the removal efficiency decreased gradually with the operation time. As it is shown, the efficiency of turbidity removal decreased from 87% at an initial turbidity of 300 NTU to a minimum of 75% at an initial turbidity of 1200 NTU and 30 minutes reaction time.

### 3.4. Effect of Different pH values and Initial Turbidities on Removing HA

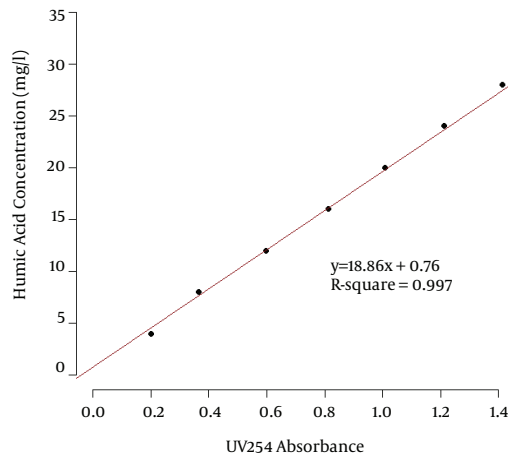
In order to investigate the effect of ECF on the HA removal, experiments were performed at initial HA of 30 mg/L, different pH values of 5 and 7, and various applied voltages of 10, 20, and 30 V. The results are shown in Figure 6 for Fe (6A) and Al (6B) electrodes. It could be seen from Figure 6 A that the HA removal efficiency in ECF process increased continuously with the increase of applied voltages from 10 to 30 V and could reach from 16% to 37% at an initial pH of 7. It can be seen from Figure 6 A that increased UV254 removal efficiency also occurs with an initial pH of 5 for Fe electrode. In this condition, the removal efficiency increased from 43% when initial pH adjusted from 7 to 5. Same results have been shown in EC process. HA removal rate increased, as applied voltage rose from 10 to 30 V for different initial pH values (Figure 6 B) for Al electrodes. It was found from Figure 6 B that the highest removal efficiency (67%) was observed in pH 5 and applied voltage of 30 V.

Among other factors, the initial pH and applied voltages play the main role for removing of HA in EC process (5, 6, 18). As shown in Figure 6 highest HA removal efficiency are achieved when the initial pH equals to 5 in applied voltage of 30 V for both Al and Fe electrodes used in ECF process. However, the variations of HA during examination indicated that the best removal of HA was achieved when Al used as electrodes. This may be due to removal of HA by charge neutralization, sweep flocs, and adsorption in EC technique (19, 21). When the pH of a solution increases from 3 to neutral medium, HA molecules take more negative charges and consequently need more positive charges to neutralize that. Also with the increase of pH, zeta potential values decrease when HA is available in the solution (5). On the other hand, since pH increases zeta potential, negative charge of HA equally increases. Therefore, this process is an intervention to the deprotonation of some functional groups in humic substance. Also, HA removal rate increased, as applied voltage rose for equal initial pH values. Based on the results shown in Figure 6, the influence of the current density is more pronounced in the initial periods of the reaction.

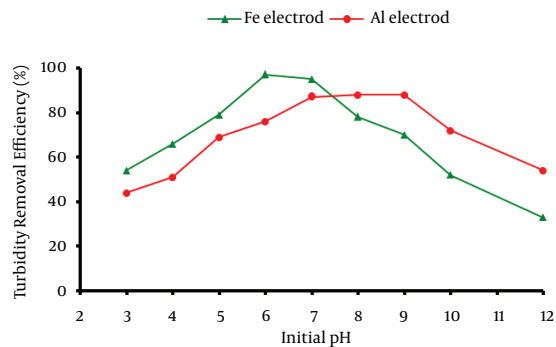
**Figure 1.** Schematic Diagram of the ECF Setup



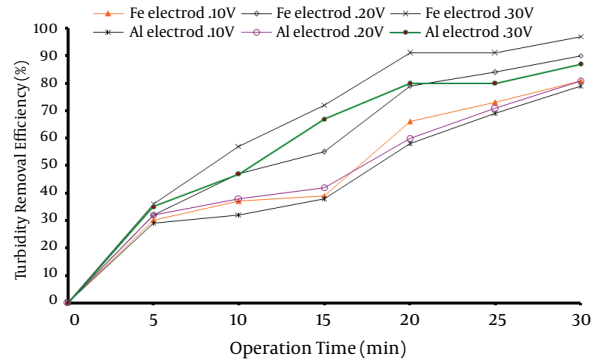
1. DC power supply; 2. Electrodes; 3. Air introducer; 4. Air flowmeter; 5. Air control valve; 6. Air pump.



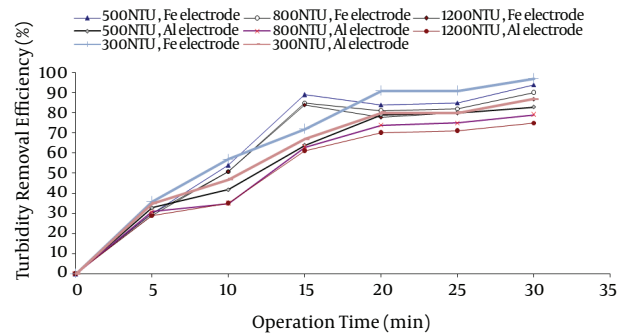
**Figure 2.** Liner Correlation of HA Concentration and UV254



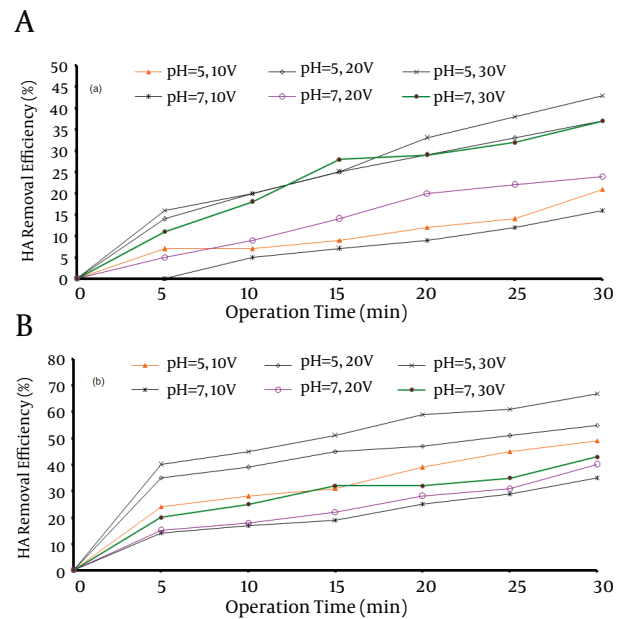
**Figure 3.** Removal Efficiency of Turbidity at Different pH Values (Initial Turbidity = 300 NTU, Applied Voltage = 30 V, and 30 Minutes Contact Time)



**Figure 4.** Removal Efficiency of Turbidity at Different Voltages, Optimum pH and Initial Turbidity of 300 NTU



**Figure 5.** Removal Efficiency of Turbidity at Different Initial Turbidities, Optimum pH and Applied Voltages



**Figure 6.** Removal Efficiency of HA at Different Initial pH Values and Electrodes for Fe (A) and Al (B), (Initial HA = 30 mg/L)

## 4. Conclusions

In the present study, the treatability of water containing high concentration of turbidity and humic acid was explored using ECF process with aluminum and iron electrodes. Effects of pH, applied voltages, reaction times, and initial concentration of turbidity were investigated. Based on the results of this study, ECF process could be used for removing high turbidity water (97% turbidity removal efficiency) at a voltage of 30 V, 30 minutes residence time, and pH 6 with Fe electrodes. In ECF process with Al electrodes, the highest turbidity removal efficiency was 88% which carried out at the pH range of 8, voltages of 30 V, and in 30 minutes residence time. The effectiveness of ECF process for Al and Fe electrodes strongly depends on initial pH value, applied voltages, and residence time. These results indicated that using aluminum electrodes can be more effective than iron electrodes for removing HA. However, the highest HA removal efficiency was achieved at pH 5 for Al and Fe electrodes.

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