REMOVAL OF NATURAL ORGANIC MATTER FROM AQUEOUS SOLUTIONS BY ELECTROCOAGULATION

ASKARI M.[1], ALIMOHAMMADI M., DEHGHANI M.H.[2] and EMAMJOMEH M.M.[3]

[1]Department of Environmental Engineering, Graduate School of the Environment and Energy, Islamic Azad University, Science and Research Branch, Tehran, Iran, [2]Department of Environmental Health Engineering, School of Public Health, Tehran University of Medical Sciences, Tehran, Iran, [3]Department of Environmental Health, School of Health, Qazvin University of Medical Sciences, Qazvin, Iran
E-mail: m_alimohammadi@tums.ac.ir

ABSTRACT

Natural organic matter (NOM) affects some qualitative parameters of water such as color. In addition, it can deteriorate the performance of water treatment process including coagulation, adsorption, and membranes. NOM also reacts with chlorine in the chlorination process and may form disinfection by-products. The present study was carried out in laboratory-scale in a batch system using a cylinder shape reactor with effective volume of 2 l. The initial NOM concentrations during the study period were 10, 25, and 50 mg/l. After specific time intervals, samples were taken from the reactor and filtered. Finally, the NOM removal according to total organic carbon (TOC) content of the samples that were analyzed with a TOC analyzer. The results showed that the highest NOM removal efficiency for three initial concentrations 10, 25, and 50 mg/l were 91, 94, and 82%, respectively. These removal efficiencies were obtained at pH 7, contact time of 20 min, and electrical current of 0.1 A. The electrical energy consumption was 0.08, 0.06, and 0.03 kWh/m^3, respectively. In this study, the application of electrocoagulation (EC) treatment method using combined Al and Fe electrode was examined to remove NOM from aqueous solution. Based on the obtained results, the EC can be used as an effective method for removing NOM from aqueous solution.

Keywords: Electrocoagulation, Natural Organic Matter, Bipolar and Monopolar, Aluminum, Iron

Introduction

Among water pollutants, organic pollutants are important due to their high quantities and ranges in water resources, an elevated concentration, specific properties, and incomplete removal by conventional water treatment plants.[1] Natural organic matter (NOM) is a complex mixture of different organic compounds originating from both natural and anthropogenic sources and is present in all water bodies.[2] The presence of NOM not only affects some water quality parameters such as color,[3] but also interferes with the performance of treatment processes such as coagulation,[4] adsorption,[2] and membranes.[2,5] It also has negative effects on the distribution system.[6] In addition, if chlorine is used in the disinfection process, NOM reacts with chlorine and may form disinfection by-products.[7,8] There are many methods such as chemical coagulation and sedimentation, oxidation, adsorption, ion exchange, and filtration using various membranes to remove NOM from aqueous environments.[9]

Electrocoagulation (EC) process is an environmentally-friendly method. This method has some advantages: no need for chemicals addition;[10] requires simple equipment and less space for installation; simple operation;[11] no need for pH adjustment; low retention time. Furthermore, it produces sludge with low water content in comparison with chemical coagulation,[12]. In addition, this process has lower effluent total dissolved solids compared with chemical treatment methods, and can remove the smallest colloidal particles.[13,14] This can be achieved by establishing an electrical current between the electrodes. Subsequently, the sacrificial anode corrodes due to the applied current while the simultaneous evolution of hydrogen at the cathode allows for pollutant removal by flotation.[15]

In the present study, the application of an EC treatment technique using combined iron and aluminum electrodes was studied at laborator scale to remove NOM compounds.

Materials and Methods

A cylindrical shape reactor with effective volume of 2 l was used to conduct the experiments. Four metal plates (2 anodes and 2 cathodes), which made of aluminum and iron were used as the electrodes. The dimensions of each electrode and the distance between the electrodes within the reactor were 110 × 110 mm and 0.5 cm, respectively.The synthetic samples with the initial NOM concentrations of 10, 25, and 50 mg/l were prepared by dissolving the desired amount of humic acid in deionized water. Initial pH of the samples was adjusted to 3, 7, and 10. The electrodes were connected as bipolar and monopolar to a direct current power supply using a wire (Figure 1a and b). A magnetic stirrer was used to mix the solution within the reactor during the treatment. When the predetermined time for the treatment (5, 10, 15, and 20) was up, the samples were taken about 25 cc from the reactor. After settling time, the samples were filtered through a 0.45 pore size filter and analyzed. The total organic carbon (TOC) of samples was measured before and after of each experimental run using a TOC analyzers of TOC-VCSH (Shimadzu, Japan). Also pH of The samples, and applied voltage and current were measured using a pH meter (Metrohm) and an ampere-meter (Sunwa), respectively. In addition, electrical conductivity and turbidity of the samples were measured using a conductivity-meter and a turbidity-meter (HACH) before and after the EC treatment, respectively.

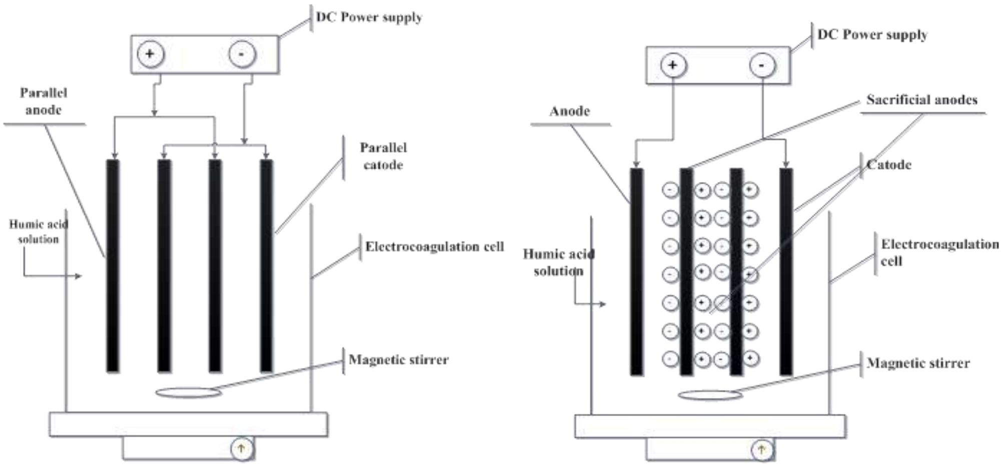

Figure 1. The electrode arrangement inside the reactor: bipolar (a) and monopolar (b)

Results and Discussion

Effect of reaction time on NOM removal

Figure 2 shows the effects of electrical current and reaction time on the performance of EC for the removal of NOM at neutral pH and the highest NOM concentration. As shown in figure 2, NOM removal efficiencies increased with increase in the reaction time. For example, at the applied current of 0.1, 0.3, and 0.5 ampere and reaction time of 20 min, NOM removal efficiencies were 82, 98, and 100%, respectively. Reaction or treatment time could decrease economic costs as well as will increase the applicability of the process.[15] Koparal *et al*. investigated the removal of humic acid by an EC process and reported that with increasing the reaction time and the energy consumption, the generation of suspended matter and removal efficiency increased until a certain time; afterwards the removal efficiency and suspended particle growth decreased.[16] Tezcan *et al*. evaluated the performance of the EC to treat the wastewater from a vegetable oil factory. The author stated that chemical oxygen demand removal efficiency was 98.9% for a reaction time of 90 min.[13] In the present study, NOM removal efficiency increased with increase in reaction time. The reason for this increase is the low production of aluminum and iron ions at the anode and hydroxide ions and hydrogen gas at the cathode during initial minutes of the reaction. Therefore, the formation of iron and aluminum hydroxides ions decreases because of a low contact between the metal ions formed at the anode and the hydroxides ions formed at the cathode. Consequently, the production of flocs and removal efficiency is low. With the increase in contact time, the concentration of iron and aluminum ions formed in the solution as well as the amount of hydroxides ions increases; thus the possibility of floc formation and removal efficiency increases.[17,18]

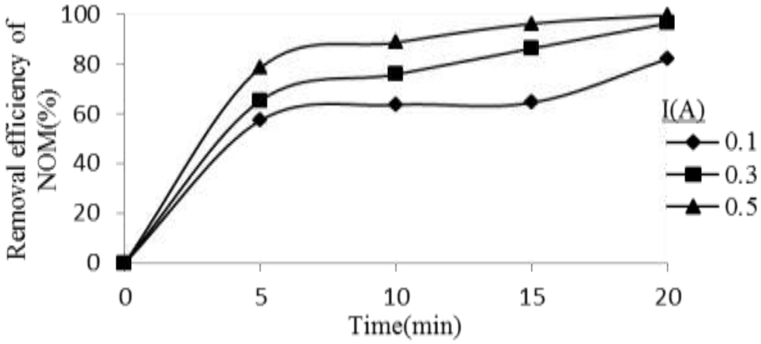

Figure 2. The relationships between natural organic matter (NOM) removal efficiencies with the current changes and reaction time (conditions: neutral pH, initial NOM concentration of 50 mg/l)

Effect of pH on NOM removal

NOM removal efficiencies for the initial NOM concentration of 50 mg/l at pH values of 3, 7, and 10 were 89, 82, and 10 %, respectively (Figure 3).The effect of initial pH can be attributed to the solubility of metal hydroxides formed in the solution.[19] In our study, which iron and aluminum electrodes were used together as the electrodes, pH 7 was obtained as the optimum pH because of the presence of insoluble $Al(OH)_{3(s)}$ and $Fe(OH)_{3(s)}$ and their high ability in removing NOM from the solution. In addition, at pH 7, when using iron electrodes, insoluble $Fe\ (OH)_{2(s)}$ and $Fe\ (OH)_{3(s)}$ were both present in the solution. Therefore, the presence of these species particularly $Fe\ (OH)_{3(s)}$ accelerates humic acid removal.[20] $Al(OH)^{4-}$ ion was the predominant species at the pH of 10; and, therefore, due to its high solubility and having negative charge were not able to remove NOM loads.[21] Ho *et al*.[22-26] used iron electrodes to remove humic acid from the aqueous solutions. They reported that the optimum pH range for those electrodes was 2-6. In another study, Heidmann *et al*.[27] investigated the performance of the EC system with Fe- and Al-electrodes for removal of Ni, Cu and Cr from a galvanic wastewater, and the highest removal efficiency was achieved at the pH values higher than 5.

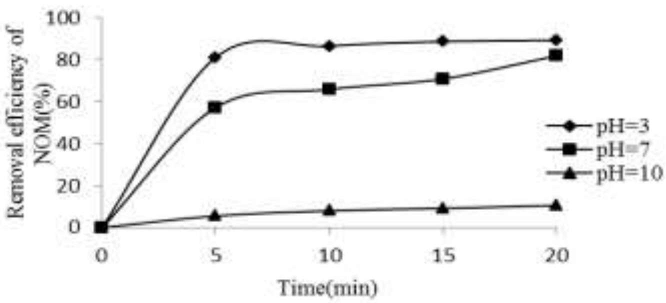

Figure 3: The effect of initial pH on NOM removal efficiency as a function of reaction time (condition: the applied current= 0.1 A, NOM concentration= 50 mg/l)

Effect on initial NOM concentration on NOM removal efficiency

NOM removal efficiencies at initial NOM concentrations of 10, 25, and 50 mg/l for optimum condition are depicted in figure 4. As shown in the figure at initial pH of 7 and reaction time of 20 min, for initial NOM concentrations of 10 mg/l (EC 279 µs/cm) and 25 mg/l (EC 417 µs/cm), NOM removal efficiency were about 88% and 92%, respectively. The corresponding value for NOM concentration of 50 mg/l (EC 808 µs/cm) was 82%.The results of the present study indicated that NOM removal efficiency decreased with increase in the initial NOM concentration. Since the limited iron and aluminum ions are produced at a constant current and reaction time, the removal efficiency decreases as initial concentration increases.[28] The results of this study are consistent with the results obtained from the study of Gomes *et al*.[29] in which an EC process using a combination of iron and aluminum electrodes was evaluated to remove arsenic pollutant.

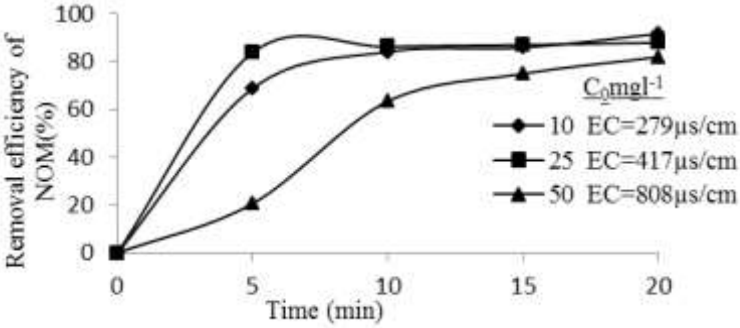

Figure 4. The effect of initial natural organic matter (NOM) concentration on NOM removal efficiencies as a function of reaction time and electrical conductivity (EC) (pH = 7, current = 0.1 A)

Effect of initial NOM concentration on energy consumption

As shown in figure 5, the maximum electrical energy consumption at optimum condition was observed for initial NOM concentration of 10 mg/l with the value of 0.08 kWh/m^3 of the solution. The minimum electrical energy consumption was detected for initial concentrationof50mg/lwith the value of 0.037 kWh/m^3 of the solution. Electrical conductivity depends on the initial concentration of organic compounds.[23,30]

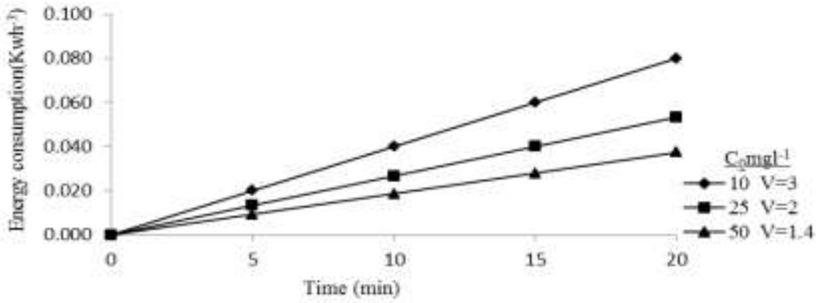

Figure 5. The effects of initial concentrations on the electrical energy consumptionatoptimum condition (pH = 7 and current = 0.1 A)

As a result, during an EC treatment system and at a constant current, by increasing the concentration and consequently electrical conductivity of the solution the amount of potential and energy consumption decreases.

Comparison of NOM removal efficiency for bipolar and monopolar electrodes arrangement at the optimum condition

In this study, due to the use of lower concentrations of such compounds, no significant difference was observed for both bipolar and monopolar arrangements of the electrodes (Fig 6). Asselin *et al*.[31] performed a study to remove organic pollutants by an EC process. It was noted that the higher electrical energy consumption of bipolar connection than monoplar connection was due to the potential difference between the electrodes. In addition, they noted that electrical resistivity generated between the electrodes in bipolar EC system is higher than that for monopolar one. This increases the electrical potential and subsequently organic pollutants removal rate. Our results are consistent with the results of Asselin *et al*.[31]

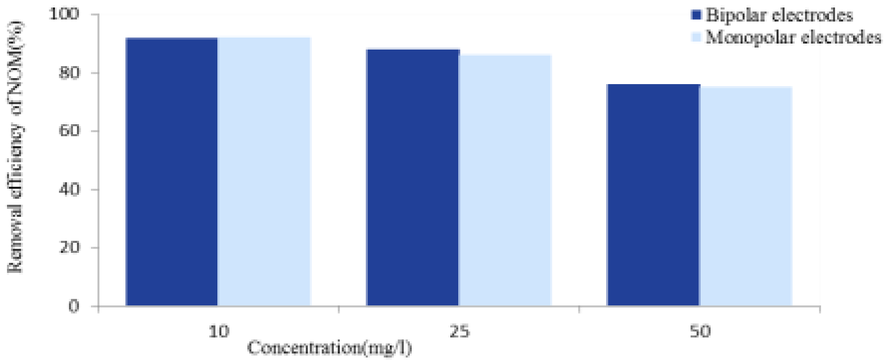

Figure 6. Natural organic matter (NOM) removal efficiencies for both bipolar and monopolar electrodes connection at optimum condition (pH = 7, current = 0.1 A, and reaction time = 20 min)

Variations of pH, turbidity, and electrical conductivity during the process

The variations in the turbidity, electrical conductivity, and pH at optimum condition are presented in table 1. It is obvious from table 1 that pH values for bipolar and monopolar connections of the electrodes remain in the neutral range. However, the turbidity and electrical conductivity decrease for both electrode connections. In many studies, researchers always agree on this principle that pH parameter has a moderate variation during an EC system. It is also mentioned that pH variations depend on the electrode material and initial pH. This is an important subject because according to the final pH, after the treatment process the effluent can be discharged into water bodies such as rivers, lakes, and seas without any pH adjustment.[15] The standard of pH for discharging effluent into water bodies is in the range of 6-9.[32] In this study, the final pH after treatment process was in the standard range and it did not have any problem for discharging into the environment.

Table 1. Changes in turbidity, conductivity and pH optimum conditions for bipolar and monopolar electrodes

	Experiments					
Electrode type	Electrode connection or arrangement (Aluminum/Iron)					
	Bipolar 1	Bipolar 2	Bipolar 3	Monopolar 1	Monopolar 2	Monopolar 3
Current intensity (A)	0.1	0.1	0.1	0.1	0.1	0.1
NOM concentration (mg/l)	10.0	25.0	50.0	10.0	25.0	50.0
Treatment time (min)	20.0	20.0	20.0	20.0	20.0	20.0
Initial pH	7.0	7.0	7.0	7.0	7.0	7.0
Final pH	7.7	7.5	7.7	7.4	7.6	8.0
Initial conductivity (µs/cm)	279	417.0	808.0	279.0	417.0	808
Final conductivity (µs/cm)	179.1	380.0	743.0	184.0	375.0	734
Initial turbidity (NTU)	5.6	10.6	20.8	5.6	10.6	20.8
Final turbidity (NTU)	0.5	1.0	3.5	0.1	0.1	2.5

NOM: Natural organic matter; NTU: Nephelometric turbidity units

Conclusion

The results of this study showed that the EC process by using of iron and aluminum as the electrodes can remove high concentrations of NOM. Therefore, the EC process with Al-and Fe- electrodes can be used as an effective and promising method for treatment of water containing contaminants such as NOM. Additionally, its high removal efficiency allowing the discharge of the effluent into the environment.

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