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Treatment of Simulated Carwash Wastewater by Electrocoagulation with Sonic Energy

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ABSTRACT

Oily carwash wastewater is a high organic and chemical wastewater. This paper targeted to investigate a treatment to decrease the water consumption and contaminants in car-washing stations. Electrocoagulation combined with ultrasonic energy (Sono-Electrocoagulation) was suggested so that the carwash wastewater is treated to be reused. The effect of both the voltage and time of treatment on the removal of COD, turbidity, conductivity, and total dissolved solids (TDS) were studied at constant initial pH 7 and electrode distance 2 cm. The results showed the best results of removal COD, turbidity, TDS, and reduce electrical conductivity is when the voltage was 30 V and a treatment time of 90 minutes.

Keywords: carwash, electrocoagulation, sonic energy, wastewater treatment.

معالجة مياه الصرف الصحي المحاكاة لغسيل السيارات بواسطة التخثر الكهربائي مع طاقة الموجات الفوق الصوتية

الخلاصة

مياه الصرف الصحي الزيتية لغسيل السيارات هي نوع من مياه الصرف الصحي ذات محتوى عضوي و كيميائي عالي. الهدف الاساسي لهذا البحث هو معالجة هذه المياه و اعادة استهلاكها و كذلك تقليل التلوث في محطات غسيل السيارات. اقترحت طريقه التخثر الكهربائي باستخدام الموجات فوق الصوتية لمعالجة هذه المياه. تاثير الفولتية و الزمن على ازالة COD، العكورة ، الموصلية و TDS درست عند اس هيدروجيني 7 و مسافة بين الاقطاب 2 سم. افضل النتائج التي اعطت افضل ازالة هي عند فولتية 30 فولت و زمن معالجة 90 دقيقة.

1. INTRODUCTION

Wastewater from car-washing stations usually contains contaminants such as oils, greases, oil in water emulsion, sand, surfactants, salts, in addition to the hydraulic fluid that is directly drained into the environment with no process of treatment. These contaminants are poisonous to the environment and humans, **Gönder, et al., 2017.**

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Soluble oil in carwash wastewaters are as oil in water emulsion, and it is contained in just one of the followings; the dispersed phase (oily layer), emulsifying agents from detergents and the external phase**, Mazumderm 2011.**

Oily wastewaters comprise an oil in water emulsions which are unstable or stable removable that hard to break. Consequently, there is an extraordinary interest in the improvement of advanced and cost-effective plus environmentally friendly treatment techniques for oily wastewaters to match strict environmental demands ever. This process is highly affected by oily wastewater contents. Thus, for oily wastewaters with high pollutants and oil content usually, one method of treatment or a combination of other treatment processes is required to ensure appropriate treatment results, **Mirjam Karhu, 2015.**

Electrochemical methods can break the emulsion. Electrocoagulation (EC) is a wastewater treatment method that includes electrochemical treatment processes with conventional chemical coagulation. In the EC process, an external direct current voltage is applied to add metal ions into wastewater which consequently produces coagulating species by hydrolyzed metal ions. The whole process is conducted in a batch or continuous mode electrolytic reactor in which a clarified liquid and a sludge are produced as basic products, **Marriaga-Cabrales, et al., 2014**.

Another method, such as the ultrasonic, could be combined with the EC process to improve the quality of water by increasing the rates of contaminants removal. EC process includes three consecutive steps**, Chaturvedi,2013:**

- Coagulants formation by electrolytic oxidation-reduction of the sacrificial electrodes.
- Destabilization of the pollutants, particulate suspension, and breaking of emulsions.
- Flocks formation by aggregation of the destabilized phase.

One of the metals used in such a process is aluminum. The anodic dissolution process includes the oxidation of aluminum into the aqueous solution as shown in the reaction given by equation (1) along with the oxidative dissociation of water as shown in the reaction of equation (2) [Barrera-Díaz et al., 2014].

$$Al \rightarrow Al^{+3} + 3e^{-} \quad (1)$$

$$2H_2O \rightarrow O_2(g) + 4H^{+} + 4e^{-} \quad (2)$$

Dissolved aluminum can participate in several chemical reactions. This is shown through the reactions represented in (Eqs. 3-7). Speciation of aluminum through electrocoagulation is very complex, **Cañizares et al., 2006 and Cañizares, et al., 2008.**

$$M(OH)^{-}_{4} + H^{+} \leftrightarrows M(OH)_3 + H_2O \quad (3)$$

$$M(OH)_3 + H^{+} \leftrightarrows M(OH)^{+}_{2} + H_2O \quad (4)$$

$$M(OH)^{+}_{2} + H^{+} \leftrightarrows M(OH)^{+2} + H_2O \quad (5)$$

$$M(OH)^{+2} + H^{+} \leftrightarrows M^{+3} + H_2O \quad (6)$$

$$M(OH)_{3(s)} \leftrightarrows M^{+3} + 3OH^{-} \quad (7)$$

The cathodic reaction is presented in equation (8), **Barrera-Díaz, et al., 2014**:

$$2H_2O + 2e^{-} \rightarrow H_{2(g)} + 2OH^{-}_{(aq)} \quad (8)$$

Three significant effects of this reaction on the electrocoagulation process are observed:

- It produces hydroxyl ions. These react with aluminum cations forming insoluble species and other coagulants (Eqs. 3 to 7)
- It produces hydrogen gas which contributes to the destabilization of colloidal particles consequently enhancing the flocculation process and
- It contributes to electro-flocculation that is floating of contaminants by their adhesion onto so small bubbles generated by the hydrogen evolution, **Casqueira, et al., 2006**.

Using ultrasound irradiation in the treatment of wastewater is expected to enhance significantly the effectiveness of the electrode processes happening in the cell, **Kovatcheva, et al., 1999,**

Kathiravan, et al., 2011, and Farooq, et al., 2002. Many favorable influences of utilizing ultrasound in the electrocoagulation process are the following, **Barrera-Díaz, et al., 2014**:

- The cohesive layer formed on the surfaces of the electrode as a result of electrode reactions products is destructed.
- The thickness of the diffuse part of the electrical double layer which is formed at the surface of the electrode is reduced.
- The ultrasound waves immediately activate the ions in the reaction zone at the electrodes.
- Defects in the crystal lattices of the surface of electrodes are generated to be more active.
- Increase in temperature at the surface of the electrode. This is due to the friction between the surfaces and the liquid.

In an electrocoagulation process for the treatment of wastewater, there is a gradual decrease in the current efficiency through the process of treatment. Consequently, the contaminants are often adsorbed onto the surface of the electrode. This will reduce the active sites on the surface of the electrode surface. As a result, a partial or complete poisoning of the electrode will occur. Efficient treatment requires that consistently both higher oxidation and reduction efficiency are maintained. Also, an electrocoagulation process is often limited by the mass transport in the system. Electrocoagulation with sonic energy has been given as an alternative to activate the surface of the electrode and enhance the efficiency of the mass transfer. This hybrid process has been used to treat contaminant in wastewater**, Feng, et al., 2016.**

Electrocoagulation with sonic energy was used to treat simulated oily carwash wastewater in this paper. Some parameters and their effects on the removal efficiency of COD, turbidity, TDS, and reducing the electrical conductivity were investigated. These included: voltage and treatment time at constant initial pH of 7. Besides, the mechanisms of this treatment method for oily carwash wastewater treatment were studied.

2. MATERIALS AND METHODS

2.1 Wastewater sample

Simulated oily carwash wastewater was prepared in this study. The prepared wastewater consisted of super motor oil multi-grade produced by Middle Refineries Company, **Al-Doura Refinery**, washing liquid (degreasing agent) produced by Al-Riyadh Company For Modern Sulphochemical Industry (Made in Jordan), and tap water. Tap water was used instead of distilled water to obtain a waste approximation of the real waste in terms of salt content and biological content. The content of washing liquid was; nonionic surfactants, sulphonic acid, sodium lauryl ether sulfate (SLES), thickeners, preservative, perfume, color, antibacterial mat, water.Q500 Sonicator device was used to create emulsions of oil in water this part was carried out in the department of renewable energy, Mistry of Science and Technology. The analysis of tap water has revealed that it contained 30 mg/L magnesium, 80 mg/L sulfate, total dissolved solids of 283 mg/L, COD of 5 mg/L and the pH was around (7). **Table 1** shows the characteristics of simulated oily carwash wastewater.

Table 1. Characteristics of simulated oily carwash wastewater.

Parameter	Value	Devises used in measure these parameter
pH	7.8	pH meter-ATC
COD, mg/L	2032	Lovibond MD 200, Germany
Turbidity, NTU	235	TurbiDirect Lovibond, TB 300 IR, Germany
Electrical conductivity, μS/cm	963.4818	SensoDirect Con 200 Lovibond
Total dissolved solids (TDS), mg/L	462	TDS meter (hold) TDS-3
Oil Content, mg/L	500	Handheld Oil-in-Water Meter

2.2 Experimental set-up and procedure

The experiments were carried out in hydrogen storage laboratory, Department of Renewable Energies, Ministry of Science and Technology. Electrocoagulation reactor was conducted in batch mode which had a volume of 2000 ml. It was positioned in an ultrasonic bath with a frequency of 40 kHz and power of 510 W, model WUC-D06H (WiseClean WUC Digital Ultrasonic Cleaner) with a capacity of 6 liters. Every electrode has a dimension of 120 mm×60 mm×2 mm. Four electrodes (two anodes and two cathodes) were used and the active area for electrodes was 432 cm^2. They are connected through the bipolar technique to a DC power supply (PS-305D) with a distance of 2 cm between each electrode fixed in the reactor. The bath of sonicator was filled with 5 liters of water. Different voltages (10, 20 and 30 V) were applied. All experiments were conducted at constant pH = 7 with 1800 ml of simulated wastewater. The pH value of wastewater was adjusted using 1 M NaOH and 1 M HCl. All experiments were carried out at laboratory temperature and atmospheric pressure. The solution was left for 60 min for sedimentation before sampling. The samples were also left for 15 min for further sedimentation before COD, turbidity, electrical conductivity, and TDS analyses. **Fig. 1** shows the photographic image of the present work.

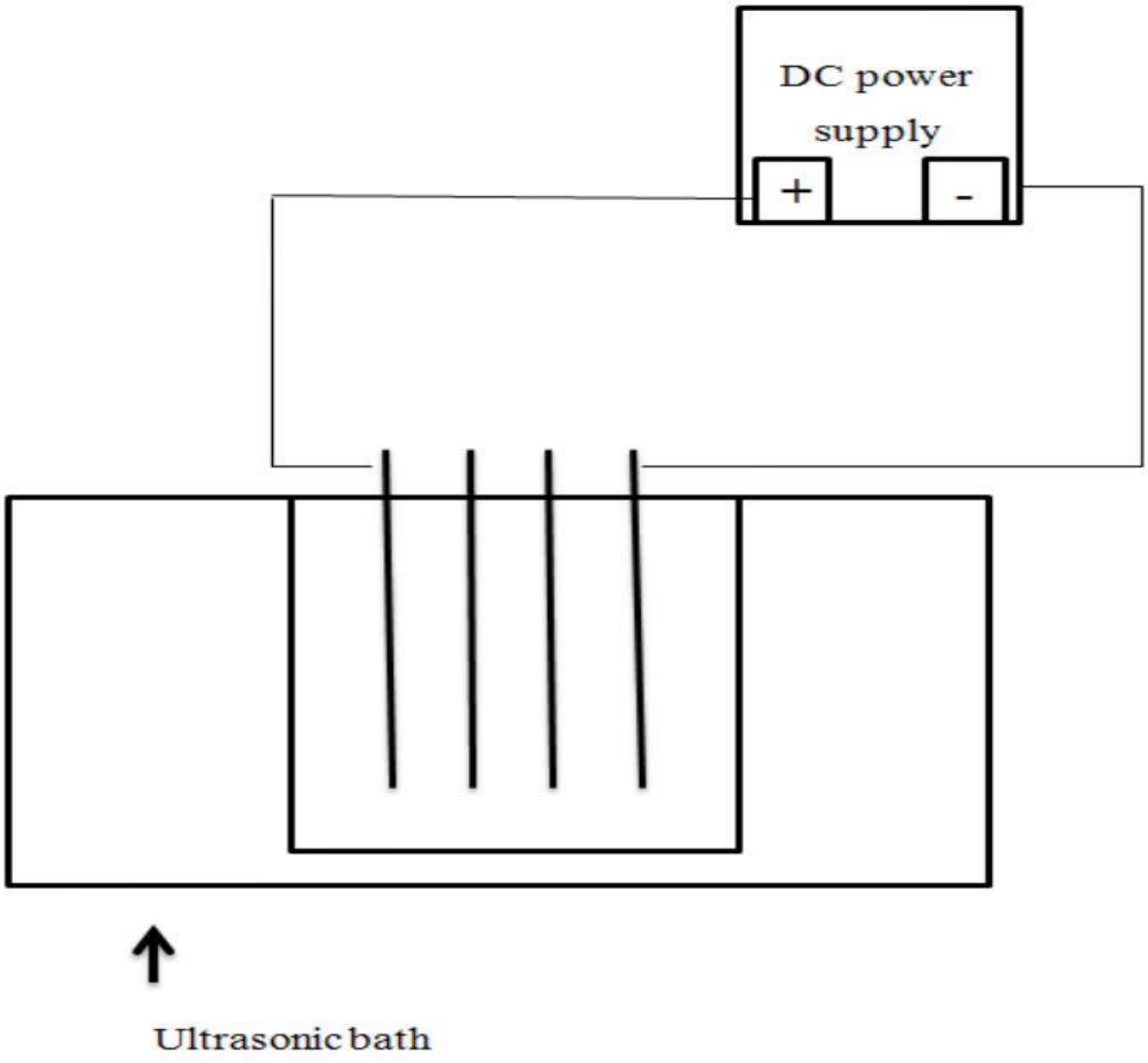

Figure 1.Schematic diagram of the experimental apparatus.

3. RESULTS AND DISCUSSION

3.1 Effect of voltage

One of the important parameters affecting the efficiency of removal in the electrocoagulation process is the voltage which can be controlled directly. Therefore, the applied voltage was varied to study its effect on the COD, turbidity, electrical conductivity and TDS at a constant initial pH 7 for the treatment time of 90 minutes.
Three voltages (10, 20, and 30 v) were used in this treatment. **Fig. 2** displays the effect of voltage on the removal of COD. COD decreased from an initial value of 2032 mg/L to 380 mg/L, 123 mg/L, and 91 mg/L for 10, 20, and 30v respectively.

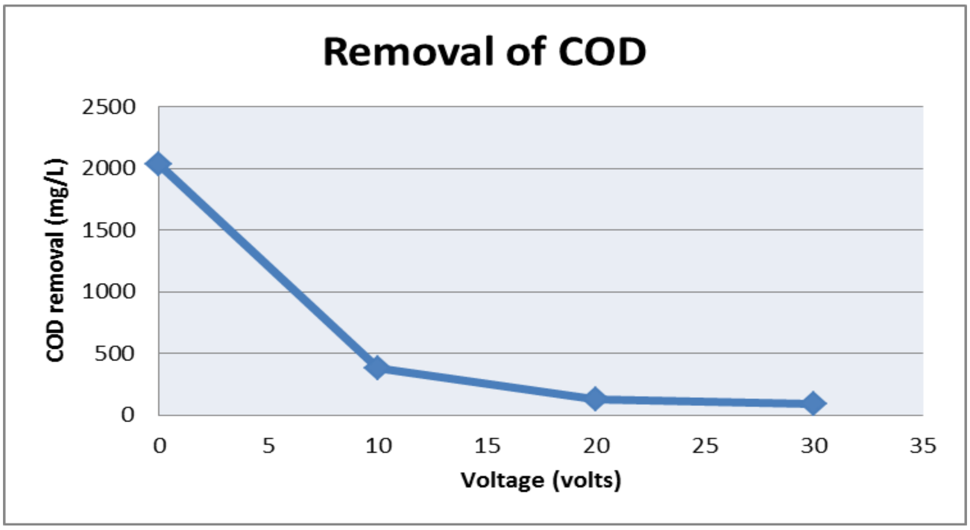

Figure 2. Effect of voltage on COD removal.

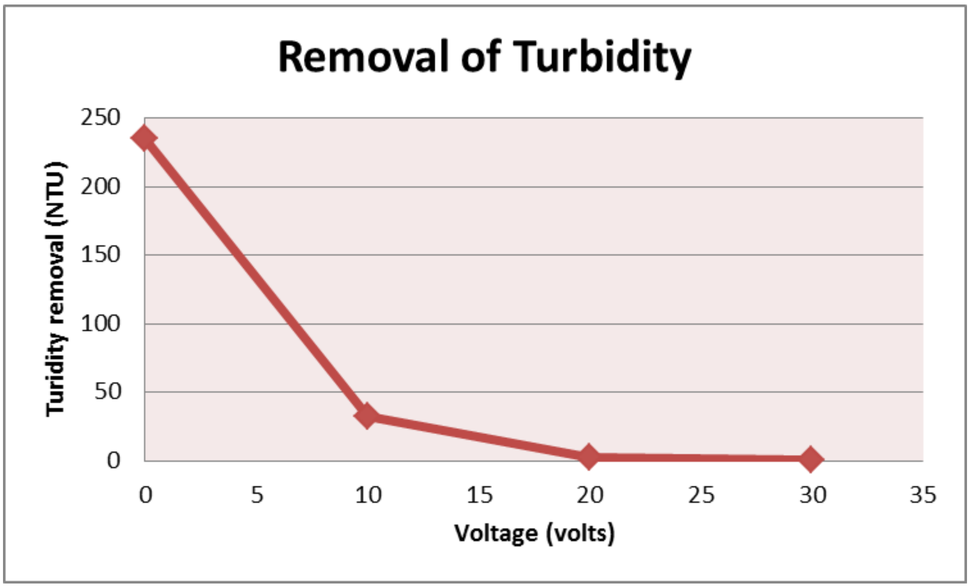

Figure 3. Effect of voltage on turbidity removal.

Fig. 4 shows the decrease in electrical conductivity from the initial value of 963.4818 μS/cm to 827.4534 μS/cm, 699.3015 μS/cm, and 663.6549 μS/cm respectively.

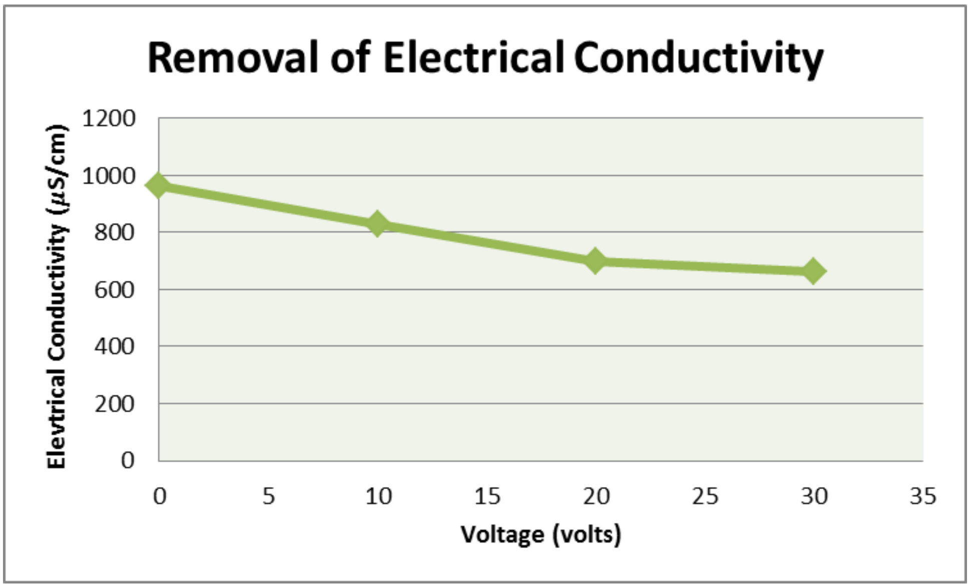

Figure 4: Effect of voltage on electrical conductivity.

Fig.5 shows the TDS change under the effect of voltage. The initial value of TDS decreased from 462 mg/L to 380 mg/L, 326 mg/L, and 300 mg/L respectively.

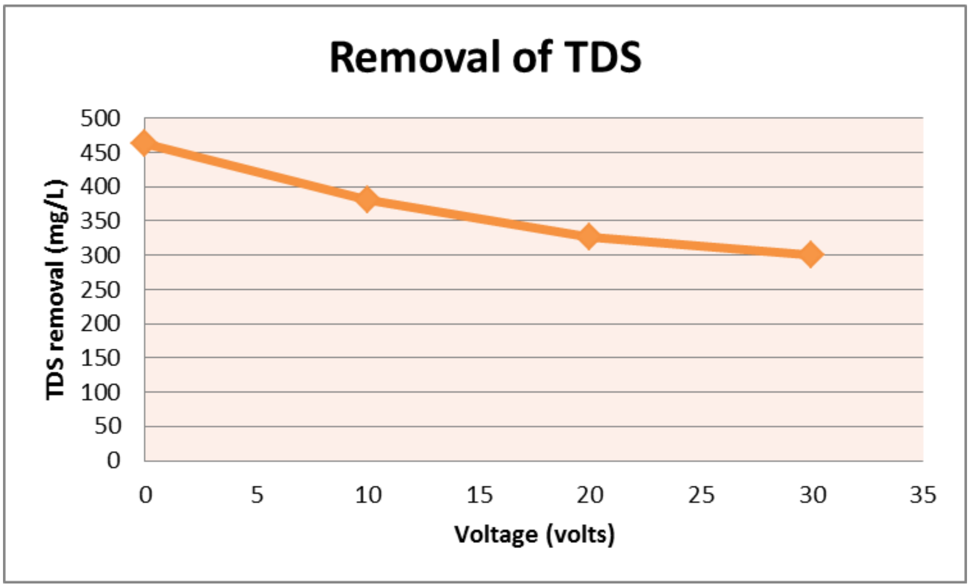

Figure 5. Effect of voltage on TDS removal.

The best results obtained at voltage 30v were the removal efficiency of COD, turbidity, and TDS were 95.52%, 99.7%, and 35.06% and reduced electrical conductivity to 31.12%.
An increase in the voltage from 10, 20 and 30 volts respectively will give rise to an increase in contaminant removal from the wastewater. Anode dissolution of the electrode increases as the voltage increases according to Faraday's law. More metal hydroxides are produced. Consequently, more sludge is produced.
Smaller bubbles produced during the dissolution of electrodes offer an additional surface area so that the denser and larger floc creation in the aqueous stream is attached. Consequently, the high separation of flocs was encountered, and that agree with, **Mahesh, et al., 2006**.

Figures 2 to 5 show the results of the effect of voltage after 90 minutes of treatment time on COD, turbidity, TDS, and electrical conductivity removal, these figures represent the relationship between voltage and remaining COD, TDS concentration and reduced turbidity and electrical conductivity at atmospheric pressure and room temperature. According to these figures, the COD, turbidity, TDS, and electrical conductivity are decreasing as the voltage of the process is increasing; therefore, the removal efficiency will be increased.

At lowest voltage, an only a small amount of the total mass of metal ions is present in the system which had been formed coagulant agent. The last with traps pollutants have been transported to the top of the reactor by flotation after long treatment time where fewer bubbles of oxygen and hydrogen were generated, and this resulted in a decrease in a mixing of the solution and pollutants uplift. Also, settling of the aggregated pollutants dominated the removal of COD, turbidity, TDS, and electrical conductivity at low voltage because of the fewer and small coagulant size produced that make these pollutants take a long time to settle.

At highest voltage, a larger amount of the total mass of metal ions presents in the system thus increasing the coagulant dosage in solution. Coagulant agent with traps pollutants has been transported to the top of the reactor by flotation after short treatment time. Hydrogen and oxygen gas bubbles were observed at high voltage especially for aluminum foil electrode where the bubble was large enough to pollutants uplift, and due to higher bubble density produced the removal of pollutants by flotation is increased.

3.2 Effect of treatment time

The effect of treatment time on the pollutant removal by electrocoagulation with sonic energy was studied under constant condition; initial pH 7 and voltage 30V. Three treatment times (30, 60, and 90 min) were applied to study the time and its effect of on removal efficiencies.

Fig. 6 shows COD removal under the effect of time. COD decreased from initial value 2032 mg/L to 214 mg/L, 120 mg/L, and 91 mg/L respectively.

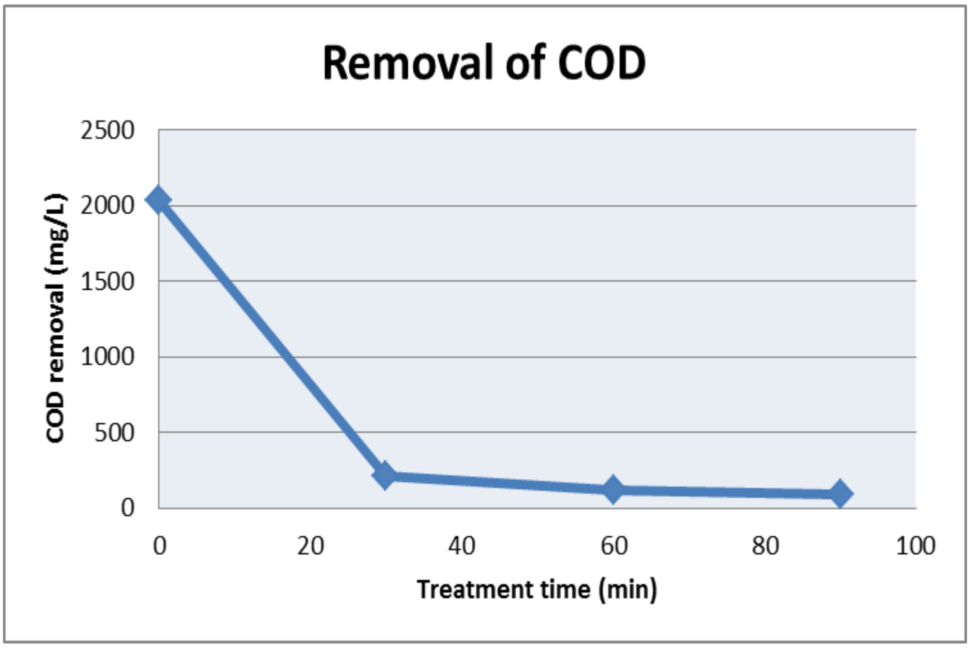

Figure 6. Effect of treatment time on COD removal.

Fig.7 shows decreasing in turbidity as time progresses. Turbidity decreased from 235 NTU to 29.2 NTU, 4.1 NTU, and 0.7 NTU.

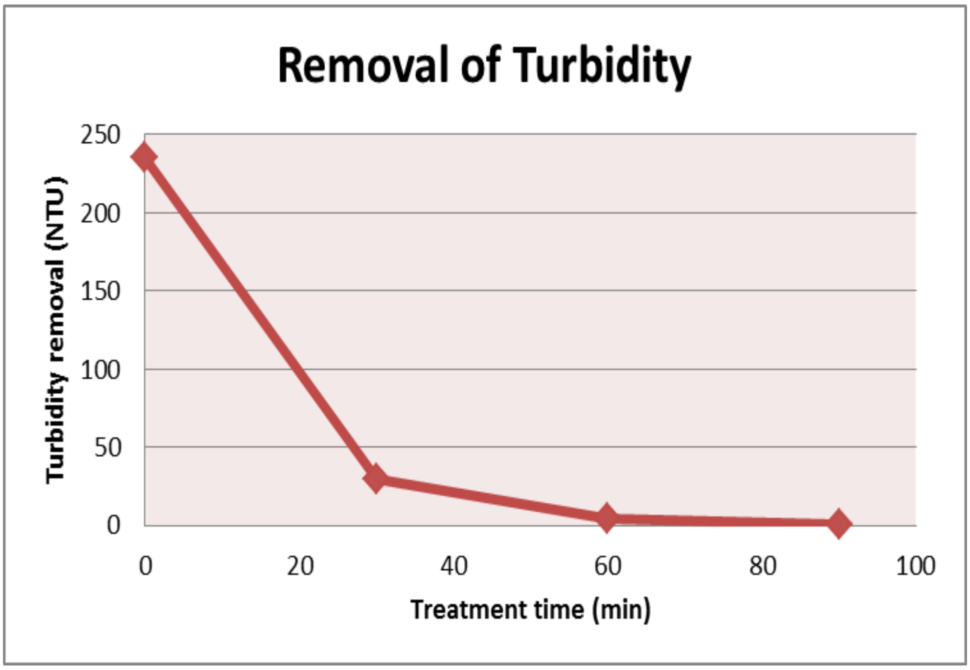

Figure 7. Effect of treatment time on turbidity removal.

Fig. 8 shows a decrease in electrical conductivity from 963.4818 μS/cm to 701.4697 μS/cm, 675.2012 μS/cm, and 663.6549 μS/cm respectively.

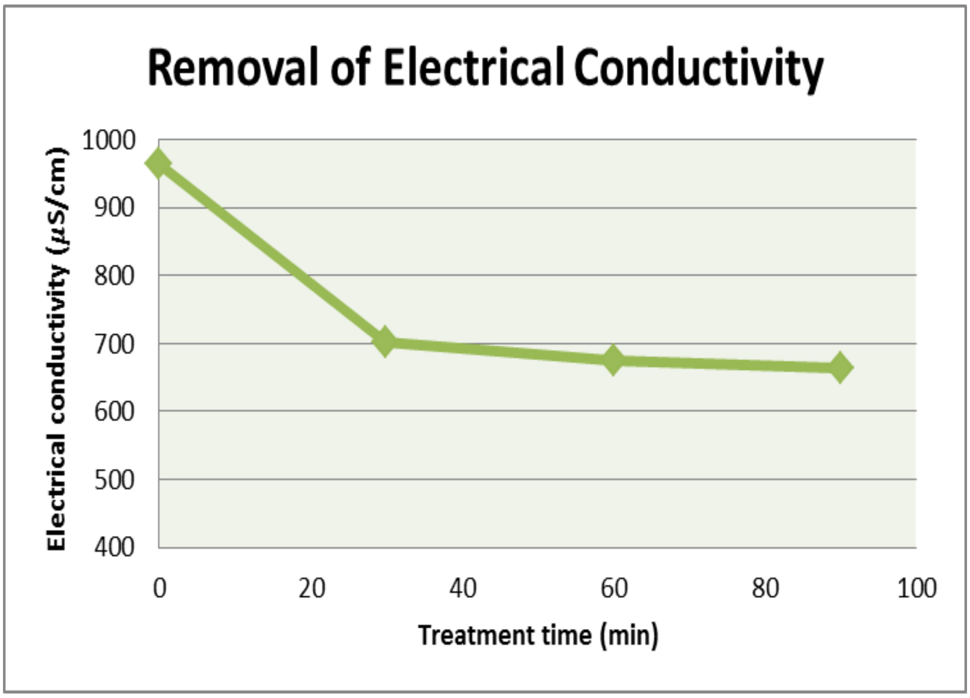

Figure 8. Effect of treatment time on electrical conductivity.

Fig.9 shows TDS removal from 462 mg/L to 379 mg/L, 320 mg/L, and 300 mg/L.

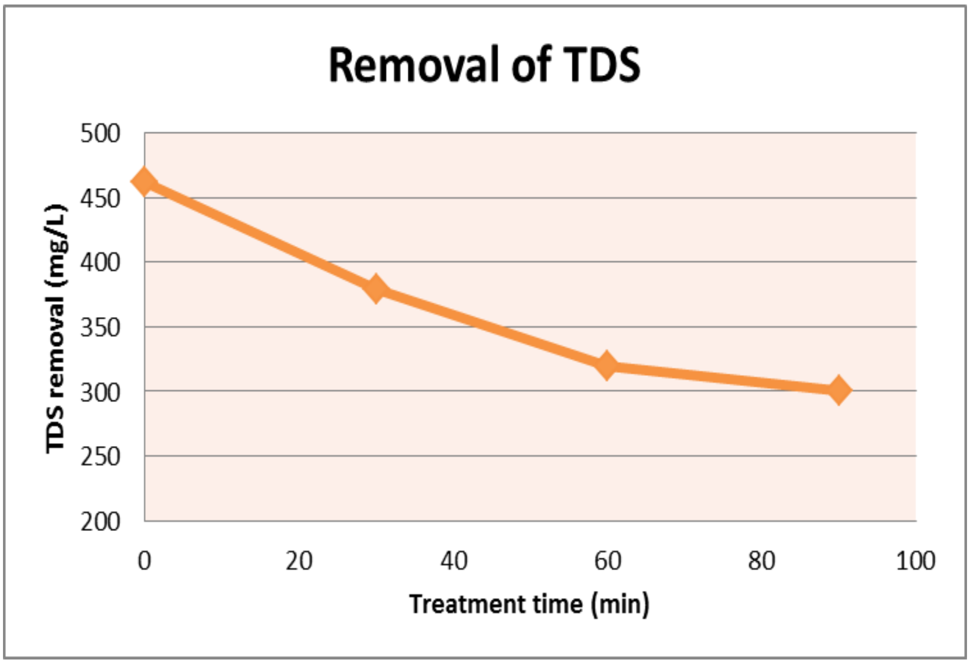

Figure 9. Effect of treatment time on TDS removal.

The best results were obtained at 90 minutes where the removal efficiency of COD, turbidity, and TDS was 95.52%, 99.7%, and 35.06% respectively.
The results show significantly increasing removal efficiencies when time progresses. At constant (30V), the longer the electrolysis time, the more $Al3^{+}$ dissolved from aluminum electrode plates, and the more aluminum hydroxide $Al(OH)_3$ was formed. Thus, the separation of contaminants is more and the sludge produced is also more. The removal curves leveled off with treatment time after 90 min. The result shows the removal of contaminants happens to a great extent within the first 30 minutes and completed in the remaining time of 60 minutes.

4. CONCLUSIONS

- The removal of COD, turbidity, electrical conductivity, and TDS by electrocoagulation coupled with sonic energy was acceptable at a voltage of 30 V, initial pH 7, and the time of treatment was 90 min.
- The ultrasound waves increased the mass transfer of species (rapid mixing).
- An impact to clean and reactivate the surface of the electrode and thus the there is a continuous renewal of the surface by ultrasound waves.

NOMENCLATURE

COD: Chemical Oxygen Demined
TDS: Total Dissolved Solids
EC: Electrocoagulation

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