Removal of Antibiotics and Heavy Metal from Water by Electrocoagulation Technology

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ABSTRACT

The release of heavy metals and antibiotics into the environment is a major concern. It affects both human and aquatic lives. A promising cost-effective and highefficiency water treatment process is electrocoagulation (EC). In this study, EC was adopted to treat nickel (Ni) and tetracycline (TC) synthetic wastewater solutions using iron and aluminium electrode materials separately. Two different electrode materials (aluminium and iron) were investigated and compared. The results showed that removal efficiency is favorable at neutral and basic condition with increasing stirring speed and decreasing inter electrode distance. After optimum conditions, removal percentage of Ni solution by iron electrodes reached more than 99% within 60 min with optimum conditions (10 g.L-1 initial concentration, pH 7, 1 cm gap, 1.12 g NaCl, 9 V and 400 rpm mixing speed), while more than 96% removal of TC (15 mg.L-1) with the same electrode within 5 min with pH 9, 1 cm gap, 2.24 g NaCl, 9 V and 250 rpm mixing speed. On the other hand, %removal of TC with Al electrode reached 99.9% within 5 min reaction time (15mg.L-1 initial concentration, pH 7, 1 cm gap, 0.56g NaCl, 4.5 V and 400 rpm mixing speed). The operating costs and electrical energy consumed to remove Ni and TC by Fe were (1.4 \$.m-3, 27.9 Kwh) and (0.95 \$.m-3, 18.9 Kwh) respectively. In contrast, 0.26 \$.m-3 and 5.18 Kwh were the operating cost and electrical energy consumed to remove TC via Al electrode respectively. EC operating parameters in this study at optinium conditions showed that the treatment is extremely effective.

Keywords: Nickel; Tetracycline; Electrocoagulation; FTIR analysis

ÖZ

Ağır metallerin ve antibiyotiklerin çevreye salınımı büyük bir endişe kaynağıdır. Hem insan hem de su yaşamını etkiler. Umut verici, uygun maliyetli ve yüksek verimli bir su arıtma işlemi elektrokoagülasyondur (EC). Bu çalışmada, demir ve alüminyum elektrot malzemeleri ayrı ayrı kullanılarak nikel (Ni) ve tetrasiklin (TC) sentetik atık su çözeltilerinin arıtılması için EC benimsenmiştir. İki farklı elektrot malzemesi (alüminyum ve demir) incelenmiş ve karşılaştırılmıştır. Sonuçlar, artan karıştırma hızı ve azalan elektrotlar arası mesafe ile nötr ve bazik durumda giderme verimliliğinin olumlu olduğunu göstermiştir. Optimum koşullardan sonra, optimum koşullarda (10 gL-1 başlangıç konsantrasyonu, pH 7, 1 cm boşluk, 1.12 g NaCl, 9 V ve 400 rpm karıştırma hızı) demir elektrotlar tarafından Ni çözeltisinin uzaklaştırma yüzdesi 60 dakika içinde %99'un üzerine çıkmıştır. pH 9, 1 cm boşluk, 2.24 g NaCl, 9 V ve 250 rpm karıştırma hızı ile aynı elektrot ile 5 dakika içerisinde TC (15 mg.L-1) %96'dan fazla giderilirken. Öte yandan, Al elektrotlu TC'nin % uzaklaştırılması, 5 dakikalık reaksiyon süresi (15mg.L-1 başlangıç konsantrasyonu, pH 7, 1 cm boşluk, 0.56g NaCl, 4.5 V ve 400 rpm karıştırma hızı) içerisinde %99.9'a ulaştı. Ni ve TC'yi Fe ile uzaklaştırmak için tüketilen işletme maliyetleri ve elektrik enerjisi sırasıyla (1.4 \$.m-3, 27.9 Kwh) ve (0.95 \$.m-3, 18.9 Kwh) idi. Buna karşılık, 0,26 \$.m-3 ve 5,18 Kwh, sırasıyla Al elektrot yoluyla TC'yi çıkarmak için tüketilen işletme maliyeti ve elektrik enerjisiydi. Optinium koşullarında yapılan bu çalışmada EC çalışma parametreleri, tedavinin son derece etkili olduğunu göstermiştir.

Anahtar Kelimeler: Nikel; tetrasiklin; Elektrokoagülasyon; FTIR analizi

DEDICATION

I dedicate this work to God and my family for their immerse love and care.

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INTRODUCTION

1.1 Water pollution

Water pollution is a major concern among many nations, it affects our environment and reduces the life extent of living organisms. Therefore, resources of water are highly polluted due to fast-growing industries, reckless control of essential resources, and migration to municipal areas [1]–[3]. Most of these wastewaters from industries being untreated before being discharged into the environment. However, water treatment is important for all living organisms. Human beings need water in its natural or clean form for metabolism and other body-related functions [1].

Several types of water pollutants exist in the environment from different sources such as hospitals and pharmaceutical companies, household or sewage release, agricultural and irrigation runoff, manufacturing industries, and health care clinics for both humans and animals [4]. These pollutants will affect marine life by increasing the amount of toxic compounds and heavy metals in water, therefore, they may react with dissolved oxygen (DO) and reduce it. A low amount of DO affects aquatic marine and plant growth [2]. Moreover, some anaerobic organisms can be spread in the environment like cholera, this bacterial cause dysentery, and diarrhoea if it inters the human body. Akpan and Ajayi studied the effect of pollutants (oil spills and petrochemical activities) on human health and the ecosystem of Niger Delta Nigeria and how it affected human health and the ecosystem through oil spills and petrochemical activities in the area [5]. Hence it is important to reduce the concentration of the pollutant before discharging it to the environment.

1.1.1 Heavy metal pollution

One of the most dangerous pollutants that exist in the environment is heavy metals for instance zinc, lead, cadmium, arsenic, nickel, copper, and silver [6,7]. Heavy metals accumulate in their chemical structure or combine with some other metals, thus remained difficult to eliminate from water. They are toxic, non-degradable, and non-decomposable that released daily into our water bodies and the environment, more than 40% of water sources have been polluted by them through the mining process, discharge from automobiles, coal combustion, fertilizer and textile industries, and metal plating activities [8]–[10]. They can change the physical, chemical, and biological properties of the water that caused serious problems in humans like destroy kidneys, lungs, and oesophagus [11].

Living organism metabolic activities need a tiny amount of most heavy metals to avoid or reduce disease [12]. However large amount can lead to serious health problems [13]. Heavy metal accumulations affect the aquatic lives in most organs by causing trouble in the immune system and the growth of the organisms [14]. To reduce or control the accumulation of heavy metals in the environment, policy and regulation from World Health Organization (WHO), Environmental protection agencies, European Union and various national regulatory bodies mandated treating wastewater before the last disposal [15], with this effect, there are standards placed for various types of pollutants [16].

One of the most essential reach metals is Nickel, a few of it is essential for human and aquatic lives because it initiates some enzymes in the body. However, it is toxic and carcinogenic when the level of accumulation is beyond the World Health Organization (WHO) limit (0.1mg/L) [17], [18]. Nickel accumulation in the body may be responsible for some adverse human effects such as heart and kidney diseases, respiratory tract, and lung fibrosis [19]. It is detected in soil, rocks, and wastewater from electroplating, mining, textile batteries, and galvanization industries [18].

1.1.2 Antibiotics pollution

Another pollutant that has been detected in the environment is antibiotics; they are chemotherapeutic or antimicrobial agents which restrain microorganism growth [20]. Currently, there are more than 250 recorded antibiotics in medicine and veterinary clinic [21]. Penicillin is the first production antibiotic from Penicillium fungi [20]. Therefore antibiotics are manufactured by chemical synthesis or natural source modification [20]. They are grouped based on their chemical structure or mechanism of action [20]. Antibiotics are applied in different types such as livestock farming to increase yield productivity and promote feed. Also, it treat pathogen diseases in humans due to its antibacterial activities [21].

Most antibiotics do not absorb completely by the human and animal systems, therefore, they are excreted by living organisms to the environment [22]. Additionally, waste from households, wastewater treatment plants, hospital and pharmaceutical companies, chemical producing industries, agricultural waste, or animal husbandry activities are the major source of antibiotics pollution [20] and the discarding of unused medical prescriptions. Reports revealed that tetracycline (TC) is an antibiotic used to enhance animal growth as well as used to treat acne and many other related diseases [22], [23]. The unabsorbed or not metabolized TC is released into the environment through urine or faeces, the amount of TC detected are at trace level (2ng/L to 60µg/L) in surface or drinking water [24].

1.2 Wastewater removal technologies

To remove pollutants from wastewater, several methods have been reported and used, each one has its advantages and disadvantages individually. Some of the methods have been mentioned and discussed below:

Figure 1.1: Some examples of treatment techniques used in wastewater.

1.2.1 Adsorption technology

The adsorption technique has been used to treat different types of pollutants. It works in the principle of mass transfer within the liquid or gas phase and adsorbent. The adsorbing material is adsorbent while the adsorbed substance is called adsorbate. It involves two processes, chemisorption and physisorption processes. The commonly used adsorptive material is activated carbon because of its high porous properties and larger surface area to which pollutants may adsorb. Moreover, Garcia and Giraldo reported adsorption of nickel ion from wastewater using

activated carbon coconut shells and obtained 67.56 mg.g⁻¹ at pH 5.8 for 10 min following Langmuir isotherm. It has great performance due to a large number of pores [25]. The adsorption technique treats water in a short time and is cost-effective [23]. The effectiveness and efficiency of pollutants removal are directly proportional to the type of adsorbent used.

1.2.2 Coagulation method

Coagulation is a chemical water treatment technology that removes solids from water by manipulating the electrostatic charges of suspended particles in water. Coagulation method is a primary step in wastewater treatment. It involves treating water by adding a coagulant such as iron chloride, aluminium sulfate or polymers. The positive charge of these coagulants neutralize the negative charges of the suspended pollutant in the wastewater and the particles bond together or coagulate as a result of this process [26]. Larger bound particles, known as flocs, are heavy and sink to the bottom of the water, where they are filtered.

1.2.3 Membrane separation process

This process allows water solution to pass through a semi-permeable material or membrane at high pressure to separate specific material from water solution. It is governed by two models, (a) solution-diffusion model i.e. transport by diffusion mechanism and, (b) hydrodynamic model i.e. transport through pores. The membrane separation process entails four types; reverse osmosis (RO), nanofiltration (NF), ultrafiltration (UF), and microfiltration (MF). These different types of membrane processes are the same in mechanism but differ in pore size, applied pressure, and membrane permeability [27]. This process has high removal efficiency, is easy to operate, is environmentally friendly, and can use to recover valuable components. It also produces low sludge and no chemical requirement. It requires

high energy, capital intensive and most time membrane fouling is highly reported. Algureiri and Abdulmajeed treated and obtained 98.5% removal efficiency from nickel industrial wastewater by reverse osmosis separation [28].

1.2.4 Biological treatment

This involves breaking down waste in wastewater by microorganisms. It is economical and effective than most chemical processes. It involves using aerobic or anaerobic processes. The aerobic process uses oxygen; the oxygen is introduced into the wastewater by a mechanical device such as a compressor or air blower while anaerobic uses bacteria. The biological process is a rapid (fast) process and environmentally friendly. It produces biogas (methane) as a by-product [29].

1.2.5 Electrocoagulation

Electrocoagulation is a type of electrochemical water treatment utilized in a range of industries. The process involves the destabilization of contaminant particles or ions such heavy metals, and colloids in water using an electrical charge. The electrochemical technique traditionally involves electrolytically oxidizing a sacrificial anode by a DC or AC power source to release metal ions that act as coagulants, destabilize impurities, and break emulsions. This coagulation produces flocculants, which float to the surface and are easily removed. The electrocoagulation technique is cheap, fast, and easy to operate. It requires no chemical with less sludge formation [27]. Although the electrochemical process is complicated, it is well-understood. It is divided into three stages, each of which includes chemical and physical phenomena.

Stage 1: Electrolytic Oxidation

Electrolytic oxidation of the sacrificial anode (usually iron or aluminum) which produces either oxides, oxyhydroxides or hydroxides (such as aluminum hydroxide or a polymeric hydroxide when aluminum hydrolyzes), which are good coagulants.

Stage 2: Destabilization of Contaminants

The metallic ions generated by the electrochemical dissolution of the sacrificial anode materials neutralize the charges of ionic species present in water and wastewaters, reducing electrostatic interparticle repulsion and causing coagulation.

Stage 3: Aggregation of the destabilized phases to produce flocs

The coagulants provide active surfaces for the adsorption of contaminating species, resulting in the formation of floc. Colloidal particles that remain in the aqueous media are entrapped and bridged by these flocs. At the anode, electrolyzed water creates little oxygen bubbles, and at the cathode, hydrogen bubbles. The bubbles pull flocculated particles to the surface and float them.

In Chapter 2 of the thesis, the detailed process, principle, and factors impacting electrocoagulation are discussed.

1.3 Aim and objectives of this thesis

This study aimed to treat synthetic water contaminated with nickel (Ni) ions and tetracycline (TC) antibiotic individually by using an electrocoagulation process with different electrodes (Al and Fe) under different treatment conditions such as initial pH, initial concentration, time, voltage, amount of electrolyte, the distance between electrodes and speed of rotating to find the optimum pollutant removal conditions.

Chapter 2

LITERATURE REVIEW

2.1 Theory of electrocoagulation

Electrochemical processes such as electrocoagulation, electroflotation, and electrooxidation have been developed to remove or minimize pollutants from wastewater industrial and laboratory scales [27]. The first country that used electric current to treat water was the UK in 1889 [30]. Afterwards, several researchers have reported the successful application of EC process to treat wastewater. In 1946, EC was first applied on an industrial scale in the USA [31]. Particularly, it has been widely used to treat wastewater from industries such as pulp and paper mill, oil refinery, textile, petroleum refining and mining, metal processing, pharmaceutical and hospital, restaurant and food processing, chemical mechanical polishing, bakery, colour and dye processing industries [32]. Furthermore, EC has grown fast because of its numerous advantages over other methods used in wastewater treatment for instance [33], [34], such as;

- EC does not use chemicals; therefore, it is environmentally friendly due to no secondary pollution caused by added chemical substances.
- EC technique is simple and easy to operate.
- It requires low current that can be operated by green energy such as solar and fuel cells, and windmills.
- It requires low current that can be operated by green energy such as solar and fuel cells, and windmills.
- The main reagent for this process is an electron generated in situ by electrolytic oxidation of the anodic material (Fe or Al), which removes the harmful material via the coagulation method.
- Flocs produced by EC are more stable and be separated by filtration.
- EC form odourless, colourless, and clear water at the end of the treatment.
- It forms valuable substances such as hydrogen gas.
- It forms low sludge accumulation that has consists mainly of metallic oxides or/and hydroxide.
- It can be scaled up to an industrialized scale.

On the other hand, this process has disadvantages [33], [34] such as;

- A lack of systematic reactor design for EC.
- The plate can passivate over time and need maintenance.

2.2 Principle and mechanism of EC technology

Electrocoagulation (EC) is a complex method that uses consumable metal plates to provide ions within the wastewater solution. The principle of EC followed the electrolysis of water expressed by Micheal Faraday [32]. Electrocoagulation is an electrochemical process that uses metal ions such as aluminium and iron to remove pollutants from contaminated water by acting as a destabilizing agent. The use of electricity and sacrificial electrodes combine with the pollutant in the wastewater stream producing insoluble oxides and hydroxide (floc) which can be simply removed to form clear water. Particularly, the electrodes are made up of different metals such as aluminium or iron connected in parallel, serial monopolar, or bipolar configuration, and the electrolyte is the wastewater solution. The reaction is set up with a stirrer to uniformly stir the suspension medium. A coagulant is produced in situ by oxidizing the metal from the anode and liberating hydrogen gas and hydroxide ion at the cathode. During the process, oxidation and reduction reactions are accomplished by positive and negative sides respectively. In the electrochemical reaction, the ions produced by the sacrificial anode (Fe or Al) ions in the wastewater neutralize the charges of the pollutant's particles and form a coagulant. The pollutants are removed either by blending or fusing the colloidal matter followed by electroflotation or by chemical reaction then precipitation. Furthermore, applied electric flow changes the qualities or properties of the water and the colloidal pollutant.

Chemical reactions at the electrode comprise metal cations disappearance at the anode and liberate hydrogen gas and hydroxide ions at the cathode which almost rapidly hydrolyze to polymeric metal hydroxide. These polymeric hydroxides are excellent coagulating agents. Herein, oxidation (at the anode) and reduction (at the cathode) reactions occurred at the electrodes when an electric current is connected or applied to the electrolyte solution [35]. The solid oxides, hydroxides and oxyhydroxides provide active surfaces for the adsorption of the polluting species.

EC process involves three continuous steps[33];

- 1. Formation of the coagulating agent by electrolytic oxidation of the sacrificial anode, ions produced decrease the electrostatic inter-particle repulsion thereby making the van der Waals attraction to predominates, hence affecting coagulation.
- 2. Destabilization of the pollutants, particulate or emulsions breaking. this second step can be summarize as; the diffuse double layer compresses around the charged species by interacting with the ions produced from sacrificial

anode oxidation. also, there will be charge neutralization of the ionic species being in wastewater by counter ions generated by step one.

3. Aggregation of the destabilized states to form flocs. the floc made as an effect of coagulation forms a sludge coating that captures and joins colloidal particles settling in the aqueous medium.

Figure 2.1: Interaction and mechanism electrocoagulation process [27].

The amount of metal deposited is dependent on the quantity of electricity passed through the electrolytic solution. A simple relationship between the amount od metal deposited and molecular weight of the electrodes can be obtained from Faraday's law;

$$
m = \frac{Mlt}{zF} \tag{1}
$$

Where M: is the atomic weight of the metal electron (26.98g/mol and 55.85g/mol for Al and Fe respectively),

- I: is the current in ampere.
- t: is time in the second.

F: is the Faradays constant (96486C/mol).

z: is the no of electron transfer.

Faraday law predicted cleaning anode plate per each experiment, the concentration of dissolved coagulant increases linearly with the amount of electric current dissolved Fe or Al undergoes hydrolysis to form mono, di, and poly complexes hence reduce the pH of the wastewater.

2.2.1 Description of reactions when iron anode is used

Iron anode produces iron hydroxide, $Fe(OH)_n$, which n depends on the oxidation state of iron, n can be divalent (2) or trivalent (3).

When $n = 2$, iron ion is Fe(II).

At the anode (oxidation):
$$
Fe_{(s)}^2 \rightarrow Fe_{(aq)}^{2+} + 2e^-
$$
 (2)

While hydroxide ion and hydrogen gas being liberated at the cathode.

Cathode (reduction):
$$
2H_2O_{(l)} + 2e^- \rightarrow 2OH_{(aq)}^- + H_{2(g)}
$$
 (3)

That produces the overall reaction equation:

$$
Fe_{(s)} + 2H_2O_{(l)} \to Fe(OH)_2 + H_{2(g)}
$$
\n⁽⁴⁾

In contrast, when $n = 3$, iron is oxidized to Fe(III) or Fe(II).

At the anode (oxidation);

$$
Fe_{(s)} \to Fe_{(aq)}^{3+} + 3e^-
$$
 (5)

$$
4Fe_{(aq)}^{2+} + 10H_2O_{(l)} + O_{2(g)} \rightarrow 4Fe(OH)_{3(s)} + 8H_{(aq)}^{+}
$$
\n⁽⁶⁾

While cathode (reduction);

$$
8H^+_{(aq)} + 8e^- \to 4H^-_{(2g)}
$$
 (7)

The overall reaction is;

$$
4Fe_{(aq)} + 10H_2O_{(l)} + O_{2(g)} \rightarrow 4Fe(OH)_{3(s)} + 4H_{2(g)}.
$$
\n(8)

Different iron hydroxide complexes can be formed in equations 3 and 5, OH can cause pH increase. Iron (III) present higher charge density than iron (II) because of the higher charge valence of the coagulant [36].

2.2.2 Reactions when aluminum anode is used

Many authors have published EC with an aluminum electrode, which dissociates at the anode to form a trivalent ion.

At the anode (oxidation);

$$
Al_{(s)} \to Al_{(aq)}^{3+} + 3e^- \tag{9}
$$

While at the cathode (reduction), OH^{\cdot} ion and H₂ gas is formed as follow:

$$
3H_2O_{(l)} + 3e^- \to 3OH_{(aq)}^- + \frac{3}{2}H_{2(g)}
$$
\n⁽¹⁰⁾

The overall reaction;

$$
Al_{(s)} + 3H_2O_{(l)} \to Al(OH)_{3(s)} + \frac{3}{2}H_{2(g)}.
$$
\n(11)

Different monomeric species will form such as $Al(OH)^{2+}$, $Al(OH)^{+}$ $_2^+$, Al(OH)₃, and $\text{Al}(\text{OH})$ ⁻ $\frac{1}{4}$ can be formed depending on the pH of the medium.

2.3 Parameter affecting EC process

According to the design and several literature reports, many parameters affect the EC process and efficiency, which includes; electrode materials, current density, process time, the distance between electrodes and many others that discussed below;

2.3.1 Effect of electrode material

The type of electrode materials used in the EC process is important. It determines the metal dissolution, percentage removal efficiency, and the type of coagulant formed [37]. Multivalent metals typically Fe and Al electrodes are highly used to enhance

the pollutant coagulation. They have higher electric multiple-layer compression differences. They are cheap, readily available, and have a high rate of anodic dissolution [37]. Al (III) and Fe (III) is a strong coagulant compared to Fe (II) because of higher positive charges [38].

2.3.2 Effect of current density

The rate of electrode dissolution, floc formation, hydrogen gas $(H₂$ gas) liberation, water reaction, and kinetics taking place in EC have been controlled by current density. It gives information about the amount of energy that has been consumed. Also, direct current is mostly used than alternating current. In the EC process oxidation reaction at the anode surface forms a stable oxide layer which generates a passivation effect to the sacrificial anode and promotes corrosion. Moreover, the passivation effect affects the electrode resistance and operational cost thereby decreasing the removal efficiency [37]. On the other hand, coagulation production can be calculated using the Faraday law of electrolysis when treatment time and current are known. Also, current density affects the rate of OH-ion formation at the cathode because of the chemical dissolution of the cathode [39]. For instance, when the current density in the process increases, the removal efficiency will increase too, same observation was recorded by A. Feryal and S. carnci using Fe-Fe electrode pairs to remove nickel from metal plating wastewater [40]. The authors observed an increase in the percent removal from 71.3 to 99.7% for a current density of 2.5 to 10 $mA.cm⁻²$ for 30 min electrolysis time.

2.3.3 Effect of electrolysis time

Electrolysis time determines the effectiveness and efficiency of pollutant removal. When the electrolysis time increases, the removal efficiency will also increase to the maximum value and stabilize even with an increase in treatment time [35]. Baazrafshan [41] reported 98.84, 97.95, and 97.75% removal of chemical oxygen demand (COD), biological oxygen demand (BOD), and total suspended solid (TSS) respectively by aluminium electrodes within 60 min $(5 - 60)$ min time interval) and 60 V from dairy wastewater while Esfandyari [42] obtained 92% removal efficiency of COD from hospital effluent within 50 min by aluminium and iron as an anode and a cathode electrode respectively for 10, 30, and 50 min treatment time interval.

2.3.4 Effect of distance between the electrodes

Similarly, to other parameters that affect the EC process, inter-electrode distance is playing an important role. However, it depends on the electric conductivity of the wastewater, the distance between electrodes affects the per cent removal of pollutants. Small distance consumes high energy and produced metal hydroxides collide with each other due to great electrostatic attraction, thereby having a high removal percentage [43], [44]. However far electrode distance provides enough time for produced metal hydroxide to form a floc by decreasing the electrostatic force and the pollutant removal efficiency [44].

2.3.5 Effect of stirring speed

The speed of stirring is one of the other factors that affect the EC by improving the mobility of the generated ions. Notably, the higher removal efficiency of pollutants is achieved by high optimum speed as observed in Khaled's work on cadmium removal using aluminium electrodes by electrocoagulation process [45]. The authors reported the effect of stirring speed at 0, 300, 450 and 600 rpm, the efficiencies were 95% within 30 min and 300 rpm, 99,5% within 60 min and 450 rpm. And finally, removal efficiency decreased by 600 rpm because much stirring breaks down the flocs. In contrast, below and beyond the optimum value of mixing speed the percent removal of pollutants will decrease.

2.3.6 Effect of initial concentration

The initial concentration of the pollutant affects the removal efficiency of the wastewater. Particularly, when the concentration of pollutant increases in the wastewater, the percent removal will decrease because the metal OH floc produced in the solution is unable to settle large molecules of pollutant [43], [46].

2.3.7 Electrode arrangement

Electrode connection affects the cost of operations, energy consumption, and removal performance of wastewater. The electrodes were arranged either in monopolar in parallel connection (MP-P) (Figure 2.3a), monopolar in a serial connection (MP-S) (Figure 2.3b), or bipolar in serial configuration (BP-S) (Figure 2.3c). All the anodes and cathodes are connected to the external power supply in a monopolar serial connection. For instance, in a monopolar parallel configuration, the outside anode and cathode are attached to the external DC circuit; hence the inner anode and cathode are connected singly from the outer electrodes. Herein, the inner electrodes known as sacrificial electrodes limit the passivation of the cathode and loss of the anode. On the other hand, in the bipolar system, the outer electrodes are combined with the external DC source while the innermost electrodes are not connected to any power circuit. The nearby site of the innermost electrodes becomes polarized and conducts a charge opposite to the charge of the adjacent electrode [33].

Figure 2.2: Monopolar electrode in parallel connection, b) Monopolar electrode in serial configuration and c) Bipolar connection [34].

2.3.8 Effect of power supply

There are two main connected sources of power in EC technology AC (Alternating Current) and DC (Alternating Current). In particular, DC supply causes oxide layer disposition on the cathode and oxidation or loss quantity at the anode. The oxide layer diminishes the effectiveness of EC by decreasing the movement of the current of the anode and cathode [42]. ,In contrast, AC supply counters cathode passivation and increase the removal performance.

2.3.9 Effect of initial pH

The last parameter is an effect of initial pH; initial pH is a vital factor in the electrochemical process [47]. It affects wastewater conductivity, electrode dissolution, metal hydroxide floc, and zero potential of the colloidal particles [48].

Higher efficiency can be optimum at acidic, neutral or basic media. Initial pH of the solution changes during the electrocoagulation process. Abdel [49] carried out an electrocoagulation process for cadmium removal using two aluminium electrodes as anode and cathode. The authors checked the percent removal by effecting of initial pH at 4, 7, and 10 and recorded 39.25, 88.75, and 95.5% respectively at 10 min treatment time.

2.4 Challenges of the electrocoagulation technology

EC technology is a good water treatment technology; with numerous advantages, it also has some setbacks that reduce its performance.

Firstly, the low sludge generated may include very toxic materials such as viruses, bacteria, toxic heavy metals, oil and grease, inorganic and organic substances [50]. The quality of sludge produced depends on which pollutants have been treated. Secondly, energy is also a challenge in an electrocoagulation process. The cost of treatment depends solemnly on the cost of electricity. However, electrocoagulation technology is not operational with great electricity consumption in rural cities or several developing countries due to electricity concerns. It will influence the maintenance and operation value and reduce profit venture. Thirdly, the sacrificial anode demand to exchange periodically [33]. and finally, Electrocoagulation technology faces hard competition from well-understood water treatment methods like adsorption, membrane separation, and biotechnology due to the kinetics of the method and mathematical modelling is not adequately understood.

2.5 Technology advancement in EC

Recently there are advancements in the EC method to increase performance and reduce cost.

2.5.1 Operation and control

Operation and control of parameters of EC dynamic is a major area of concern. The impermeable oxide film formed on the electrode creates electrode passivation leading to low removal efficiency of the wastewater. The application of an AC source has been employed to resolve the DC electrodes passivation difficulties. Gene and Bakiric [51] discovered that power was saved through pulsed voltage on emulsified oils by the electrocoagulation process.

2.5.2 Combining EC technology with established systems

Integrating EC with other removal methods is one of the fresh advances in EC. It will lead to great achievement. Good results were gained by using EC as a pretreatment measure to minimize the pollutants quantities before another treatment move. Bazrafshan [41] treated real textile wastewater with combined three technologies, chemical coagulation, EC, and adsorption. The author used poly aluminium chloride 'PAC' as a chemical coagulant to obtain 44.5% loss for dye, 40% for COD, 34% BOD, and 23.7% TSS. It is accompanied by 98.6, 93.1, and 88.8% dye removal in EC bipolar configuration employing four aluminium electrodes. In the end, the author reached 99.9, 98, and 94.2% for dyes, COD, and BOD respectively by adsorption process using pistachio nutshell ash.

2.5.3 Cell design and process enhancement

EC reactors have been improved in recent years to achieve greater performance and high-grade removal efficiency. Kian and Othman [52] studied the impact of integrating magnetic field using AlNiCo of 0.16 T magnetic strength with EC to advance the removal efficiency of the pollutant. The AlNiCo has placed under the reactor the pollutants removal at its best conditions surpasses 92% as compared to 85.99% without the use of the magnetic field.

2.6 Applications of EC technology

Many different pollutants were removed by the EC process such as;

2.6.1 Oil and refinery wastewater

Wastewater from these industries comprises have high levels of chemicals, COD, BOD, dissolved solids, aliphatic and aromatic hydrocarbons. It is released by petrochemical industries, petroleum factories, soap-making factories, and eateries. Tezcan [53] treated vegetable oil refinery wastewater by electrocoagulation process using an aluminium electrode. The authors at optimum conditions obtained 98.9% COD removal at 90 min within the energy consumption of 42 Kw.h/kg.

2.6.2 Pulp and paper industry wastewater

The pollutant is regularly blackish uwith high BOD, COD, suspended solids, and a high volume of organic particles. Wastewater from the pulp and paper industries adds significantly to environmental contamination. Khansorthong and Hanson[54] studied the effluent from the pulp and paper production with Fe electrode in a batch method. They recorded a 77 and 91% elimination of COD and percent colour removal respectively with a predicted treatment cost of 0.29 USD/m^3 .

2.6.3 Food industry wastewater

Effluents are released from different sources. They are extremely non-toxic and biodegradable. Food effluents are defined by high BOD, COD, and suspended solids. The contaminant depends solemnly on the sector. Asslim [55] reported 86, 99, 89, and 90% reduction in BOD, oil and grease, TSS, and turbidity respectively from slaughterhouse wastewater using mild steel and Al.

2.6.4 Dye and textile wastewater

Wastewater contains various types of dyes and organics with high stability and has resistance to chemical and biological degeneration. Dye-containing wastewater limits the penetration of sunlight. Merzouk [56] reported at 7.6 pH and 10 min treatment time with aluminum electrodes and 1 cm inter-electrode distance, 93% decrease in percent color, 88.9% in BOD, 85.5% in suspended solid, 79.7% in COD, and 76.2% in turbidity.

2.6.5 Heavy metals containing wastewater

Heavy metal contaminants have been a major problem to humans and the environment. EC one of the best technique that used to exclude heavy metals from wastewater. Vasudevan [57] studied wastewater containing arsenic using aluminium alloy and stainless steel as anode and cathode respectively and recorded 98.4% removal efficiency at pH 7 and 0.2 A/dm^2 current density.

Chapter 3

METHODOLOGY

3.1 Materials

Nickel nitrate hexahydrate $(Ni(NO₃)₂·6H₂O, 290.79 g/mol)$, NaCl (58.44 g/mol), HCl (37%) and NaOH (>99%) were purchased from Merck KGaA, Darmstadt Germany. Tetracycline hydrochloride (yellow colour, $C_{22}H_{25}C1N_2O_8$, 480.9g/mol >98% purity was purchased from Mustafa nevzat). Two aluminium and iron electrodes were purchased from a mechanical workshop in Famagusta North Cyprus costing 0.05\$ and 0.012\$ for each electrode respectively.

3.1.1 Chemicals Preparation

50 g.L⁻¹ and 100 mg.L⁻¹ stock solutions were prepared by dissolving 50 g and 0.01 g of nickel nitrate hexahydrate (Ni) and tetracycline hydrochloride (TC) respectively in a 1000 mL volumetric flask. Various concentrations were made for calibration curve by serial dilution for both Ni and TC solutions.

3.1.2 Analytical methods

The absorbance measurement of Ni and TC solutions were determined by UV-Vis spectrophotometer (UV-1201V, Japan Shimadzu) at 395 and 365 nm absorption peaks respectively. The iron and aluminium hydroxide coagulants produced called sludge were characterized by a Vertex 70 FTIR spectrophotometer. The pH and conductivity of the wastewater were monitored by a pH meter (InoLab pH/Cond720, Germany). DC power supply (MH03819, Frederiksen Denmark) was used to regulate the current and voltage of the system.

3.2 Experimental setup

3.2.1 Electrodes pretreatment

Two aluminium electrodes with dimensions (Length, Width, and Thickness) 16 cm x 2.7 cm x 0.2 cm and two iron electrodes with 16 cm x2.3 cm x0.2 cm were used in the experiments individually. The electrodes were polished by sandpaper to eliminate dust and get rid of the oxide coating on the surface of the electrodes then immersed in 0.1 M HCl for 1 min to restrict likely electrode passivation due to oxide deposit. After that they were rinsed with distilled water, dried and drained for the experiment. Before and after each experiment, the electrodes were cleaned and measured to determine the magnitude of electrode loss during the process.

Parameter	varying values
Treatment time (min)	$5 - 60$
pH range	$3, 5, 7, 9, \& 11$
Voltage (volt)	$4.5, 9, \& 12$
Electrolyte in gram	$0.56, 1.12, \& 2.24$
Ni concentration (g/L)	5, 10 & 15
TC concentration in mg/L	5, 10, & 15
Stirring speed (rpm)	100, 250, & 400

Table3.1: List of parameters with different ranges used

3.2.2 Experimental procedures

All the EC experiment was conducted in a 200 mL container as shown in Figure 3.1 with two plates attached to DC circuit, one serving as an anode and other as a cathode under consistent uniform stirring. The electrodes were immersed at 2.5 cm depth inside the solution connected to the DC power supply at laboratory temperature. NaCl used as a supporting electrolyte was added before each experimental process.

This experimental setup applied to three (3) treatment processes.

- 1. Nickel ion pollution using two iron electrodes.
- 2. TC pollution using two aluminium electrodes.
- 3. TC pollution using two iron electrodes.

After each treatment, the treated wastewater was filtered with filter paper. The filtrate was analyzed with a UV-Vis spectrophotometer while the sludge was dried in an oven at 40ºC and characterized by an FTIR spectrophotometer.

3.3 Experimental analysis

3.3.1 Test performed

The percentage removal of the treated sample was determined by scanning through the UV-Vis spectrophotometer. It was calculated with a corresponding wavelength of 395 and 365 nm for nickel ion and TTC respectively as follow;

Figure 3.1: (a) Laboratory set-up, (b) Nickel solution synthetic wastewater, (c) TC synthetic wastewater solution, and (d) wastewater after treatment.

$$
\%Removal = \frac{c_f - c_0}{c_f} \times 100\% \tag{13}
$$

where C_0 and C_f are the initial and final concentrations respectively.

Aluminium and iron weight were measured before and after every experiment to determine the weight loss. The experimental weight loss was calculated using the equation below;

$$
M_{experimental} = M_{before\ EC} - M_{after\ EC}
$$
 (14)

The electrode efficiency was determined as follow;

$$
E_{efficiency} = \frac{M_{experimental}}{M_{theoretical}} \times 100\%
$$
\n(15)

However, the mass of aluminium or iron was calculated theoretically by using the equation below;

$$
M_{theoretical} = \frac{M}{nF} \tag{16}
$$

Where M is the molar mass of Al or Fe (29.98 g/mol, 55.85 g/mol), J is current, t is the treatment time, n is the amount of transferred electrons (3 and 2 for Al and Fe respectively), and F is the Faraday constant (96486 C/mol).

3.3.2 Calibration curve

Calibration curves were performed for each stock solution prepared by serial dilution.

Figure 3.2: Calibration curve for nickel solution.

Figure 3.3: Calibration curve for TC solution.

3.3.3 pH determination

Before the EC, the initial pH of the solution is measured and adjusted to the desired value with 0.1 M HCl and 0.1 M NaOH. During the experimental process, the treated sample was collected every five minutes until 60 min to check the absorbance of the residual pollutant.

3.3.4 Sludge characterization

Reduced sludge produced during the EC process is one of the main advantages of EC over other treatment technology. At the end of the treatment time for each process, the treated wastewater was filtered and analyzed by UV-Vis spectrophotometer analysis. Dried sludge characterized by Fourier Transform Infrared Spectroscopy (FTIR) . Aluminium electrode produced sludge with amorphous aluminium hydroxide and aluminium oxyhydroxide while sludge with iron electrode contains crystalline or poorly crystalline phases. Vasaleven [58] used XRD and EDS to analyse the sludge produced from fluoride removal by the EC method with an aluminium electrode.

CHAPTER 4

RESULTS AND DISCUSSION

4.1 FTIR analysis

FTIR of $Ni(NO₃)$, $6H₂O$ (Ni) and the sludge at different pH from the EC process were analyzed as shown in Figure 4.1. At different pH, two major peaks appeared around 600 and 488 cm^{-1} referred to Fe-O (octahedral) and Ni-O (tetrahedral) respectively [59]. Meanwhile, the adsorption process of nickel solution occurred on the nanoparticles at all pH, peaks around 3300 , 1645 and 1341 cm⁻¹ in $Ni(NO₃)$ ₂.6H₂O correlated with O-H stretching, N=O stretching and vibration of nitrate asymmetric respectively [60]. These bands appeared in the sludge with different intensities, the same observation was noticed by Yesica et al [61].

On the other hand, TC can form a complex compound with metal, FTIR of TC and complex compounds are shown in Figure 4.2 at different pH. Peak around 3300 cm- $¹$ referred to O-H and N-H stretching, this band become wider which might be due to</sup> formation of $\text{Al}(\text{OH})_2^+$ and $\text{Al}(\text{OH})_3$ at low pH's while at high pH's polymer of Al might be formed such as AlO₄Al₁₂(OH)₂₄(H₂O)₁₂⁷⁺ and Al₃₀O₈(OH)₅₆(H₂O)_{2.4}¹⁸⁺ [62]. However, bands of interest appeared between 1650 to 1400 cm^{-1} referred to as C=O and C=C in TC, after the treatment process the band range changed due to complex formation between carbonyl group in TC with Al ion, same results were observed in Abdulghani et al [63]. The authors found that TC formed a complex compound based on the C=O with Au and Hg ions.

Figure 4.1: FTIR spectra of nickel solution (Ni) and sledge that formed at different pH.

Figure 4.2: FTIR spectra of TC and sledge that formed at different pH.

4.2 Effect of initial pH

Initial pH is an important parameter influencing the performance of the EC process. Herein, the removal efficiency of nickel and TC pollutants from wastewater were investigated under varying pH from 3 to 9 with Fe electrode and from 3 to 11 with Fe and Al electrodes individually for Ni and TC solutions respectively. Here 10 g.L^{-1} and 15 mg. L^{-1} of Ni and TC solutions were used with Fe electrode respectively, while 15 mg.L⁻¹ of TC solution was used with Al electrode, 1.12 g of NaCl was used as an electrolyte for 60 min. As shown in Figure 4.3a the performance of the Fe electrode with Ni is high at pH 7 with 77% removal because of adsorption of nitrate ion on the falcon that formed from Ni and Fe with oxygen. Meanwhile, the lowest removal of the nitrate ion (39%) at pH 3 due to the transformation of nitrate ion from $NO₃$ ⁻ to N₂O in acidic media while in basic media $NO₃$ ⁻ will transform to N₂. The same results were observed by To et al [64].

Figure 4.3: Effect of initial pH $(3, 5, 7, 9, \text{ and } 11)$ of (a) Ni solution (10 g.L-1) with Fe, (b) TC solution (15mg.L-1) with Fe and Al respectively.

On the other hand, the performance of the EC process to remove TC with both electrodes is high at all pH but remarkable removal at pH 9 and 7 with Fe and Al electrodes respectively (Figure 4.3b). Particularly, the removal efficiency of TC increased from 63% at pH 3 to 96.72, 96.51, 97.98 and 96.72% at pH 5, 7, 9 and 11 respectively after 60 min contact time with Fe electrode. While the % removal of TC by Al electrode reached more than 98% at all pH. The removal trend is attributed to the complex formation between TC with metals then adsorbed on metal hydroxide. A similar observation has been reported elsewhere by Gen Yoshida [65]. These results attributed to pH-dependent the distribution of TC; when the pH is less than $pKa₁$ (3.32) the cationic form of TC will participate in the removal process via adsorption on metal hydroxide. Between pKa_1 and pKa_2 (3.32 and 7.78), TC will exist in zwitterionic form and react with metals to form a complex then precipitate. Finally, beyond pKa_2 (7.72) completely deprotonation of TC will occur and an anionic charge will appear allowing the complex to form then be adsorbed [60].

4.3 Effect of initial concentration

The effect of initial pollutant concentration on the removal efficiency of Ni and TC solutions in the concentration range 5-15 g.L⁻¹ and 5-15 mg.L⁻¹ respectively. Figure 4.4a shows as Ni solution concentration increased from 5 to 10 g.L⁻¹, the percent removal increased from 54.8 to 77.4% then decreased to 54% when the concentration increased to 15 $g.L^{-1}$. In contrast, the percent removal of TC increased from 75.8 to 98.7% and from 98.5 to 99.9% when the concentration of TC increased from 5 to 15 mg. L^{-1} with Fe and Al electrodes respectively. The observed trend is due to the metal hydroxide flocs produced is adequate to settle the massive quantity of contaminant molecules at greater initial concentration[39]. The optimum

concentrations that was used farther more are 10 g.L⁻¹ and 15 mg.L⁻¹ for Ni and TC solution respectively.

4.4 Effect of applied voltage with time

The applied voltage is an operating parameter that directly affects the performance of the EC process. Generally, the time required to achieve the study state condition decreased when applied voltage increased. Figure 4.5 shows the effect of voltage Fe and Al electrodes individually. Figure 4.5a shows the removal of nickel solution. 4.5, 9 and 12 V within 60 min for Ni removal using Fe electrode and TC removal by was rapid where more than 50% was removed after 40 min by 4.5 V and 20 min for both

Figure 4.4: Effect of initial concentration of (a) Ni solution (5, 10 and 15 g.L-1) with Fe (b) TC solution (15 mg.L-1) with Fe and Al separately.

9 and 12 V. Note that Ni solutions removal rates reached equilibrium 77% (4.5v), 86% (9v), and 92% (12v) after 60, 40 and 50 min respectively. On the other hand, the removal of TC solutions $(15 \text{ mg} \cdot \text{L}^{-1})$ were rapid with Fe and Al electrodes within 60 min at different voltage as illustrated in Figure 4.5b and 4.5c. Clearly, from Figure 4.5b, the percent removal of TC pollutant with Fe electrode reached more than 80, 89 and 91% within 5 min and stabilized with more than 99.3% within 50, 60 and 30 min at a different applied voltage (4.5, 9, and 12 V) respectively. While removal of TC by Al electrode reached 98, 88 and 97% within 5 min and stabilized with almost 100% after 15, 55 and 45 min with different voltage (4.5, 9, and 12 V) respectively.

The observed increasing trend in the percent removal of Ni and TC solutions are likely due to the increase in the amount of Fe and Al ions that are released from anodes during the EC process. Therefore, the amount of $NiFe₂O₃$, Fe(OH)₃ and $Al(OH)$ ₃ particles also increased in the solutions which lead to more surface area for pollutants adsorption. Moreover, bubbles of H_2 gas generation rate increases and the size of the bubbles decreases with increasing the applied voltage from 4.5 to 12 V where both of them participated in the EC removal process same observation reported by Bazrafshan et al [66]. For the following experiments, the optimized voltage and time were determined as 9 V and 40 min for Ni removal by Fe electrode, 4.5 V and 40 min for TC removal by the same electrode and finally, 4.5 V with 20 min for TC removal by Al electrode.

Figure 4.5: Effect of applied voltage (4.5, 9 and 12 V) on (a) removal of Ni solution $(10 g.L-1)$ by Fe, (b) and (c) are removal of TC solution $(15 mg.L-1)$ by Fe and Al.

4.5 Effect of supporting electrolyte

The effects of supporting electrolytes on the removal of nickel and TC solutions by EC process were carried out by sodium chloride using different amounts 0.56, 1.12, and 2.24 g while keeping other parameters constant. Chloride ions diminish electrode passivation which results in increasing the elimination efficiency. Figure 4.6a shows that when the amount of NaCl increased from 0.56 to 1.12 g, the % removal increased from 64.8 to 86.3% due to an increase in solution conductivity [67]. However, when the amount of NaCl increased to 2.24 g, the % removal of Ni solution decreased to 68.4% most probably because of the competition between the positive charge ions (Ni^{2+} and Na^{+}) and negative charge ions (NO_3^- and Cl⁻) to form the nanoparticles and the adsorption process. García and his co-authors found the same results when they used sodium chloride as an electrolyte [68]. ,In contrast, the percentage removal of TC pollutant was increased when the electrolyte amount increased from 0.56 to 2.24 through 1.12 g of NaCl. Figure 4.6b revealed that when the amount of NaCl increased from 0.56 (98.5%) to 1.12 and 2.24g, the percentage removal increased to around 99.9% with Fe electrode, while with Al electrode (Figure 4.6c) no significant change was observed in percentage removal of TC (99.91%) with changing the amount of electrolyte from 0.56 to 1.12 and 2.24 g.

Figure 4.6: the effect of supporting electrodes NaCl (0.56, 1.12 and 2.24 g) on (a) removal of Ni solution (10 g.L-1) by Fe electrode, (b) and (c) are removal of TC solution (15 mg.L-1) by Fe and Al respectively.

4.6 Effect of electrode distance

The effect of inter-electrode distance (gap) was carried out in this study from 1 to 5 cm to investigate this parameter in the removal of Ni and TC solutions by different electrodes. %removal of pollutants is illustrated in Figure 4.7. As expected, the removal efficiency of Ni and TC solutions increased with decreasing the gap between the electrodes. When the gap decreased from 3.5 to 1 cm removal efficiency of Ni solution increased from 60% to 99.7% by Fe electrode after 60 min, while TC removal increased from 90% to 99.3% when the gap decreased from 5 cm to 2 and 1 cm within 40 min with the same electrode. When Al electrodes were used, the percent removal of TC solution was increased from 90.5 to 99.9% within 5 min.

The explanation of these results is due to the decrease in resistance through the pollutants solutions between the electrodes which lead to an increase in the rate of metal dissolution (Fe and Al) and forming ions,, therefore, increase in the percent removal of Ni and TC solutions. Furthermore, decreasing the distance between the electrodes could enhance the flotation process by decreasing the generated hydrogen bubbles in a narrow space which results in higher removal efficiencies. The same results were reported by Zeboudji et al when they used Fe electrodes to remove boron [69].

Figure 4.7: Effect of inter-electrode distance (gap) (1, 2, 3.5 and 5 cm) on a) removal of Ni solution (10 g.L-1) by Fe, b) and c) removal of TC solution (15 mg.L-1) by Fe and Al.

4.7 Effect of stirring speed

The effect of stirring speed at 100, 250, and 400 rpm was investigated to determine the optimum speed for nickel and TC elimination by the EC process. As shown in figure 4.12, at 60 min, the removal of nickel solution was reached to more than 99% with 100 and 400 rpm with low speed, the fast mixing time affected the removal efficiency. While 86.3% was removed with 250 rpm.

TC removal with Fe-Fe pairs in Figure 4.13a increased from 83 to 99.8% when the speed increased from 100 rpm to 400 rpm for 40 min respectively. However, with aluminium electrodes, % removal slightly raised from 99.8-99.9% with the same mixing speeds within 20 min. As observed generally in this study, an increase in stirring speed leads to higher percentage removal of pollutants. This can be explained by the fast hydrolysis of water and formation of $NiFe₂O₄$ nanoparticles $Fe(OH)$ ₃ and Al(OH)₃ that worked as adsorbents to adsorb nitrate ions and TC. B. Khaled and B. Wided [45] reported the same results with the removal of cadmium from synthetic and phosphate manufacturing wastewater.

Figure 4.8: Effect of stirring speed (100, 250 and 400 rpm) on a) removal of Ni solution by Fe (10 g.L-1), b) and c) removal of TC solution (15 mg) by Fe and Al respectively.

Chapter 5

CONCLUSION

EC system was carried out to eliminate nickel and TC from synthetic wastewater solutions using iron and aluminium electrodes individually. The treated wastewater was analyzed by UV-vis and the sludge was characterized by FTIR. The treatment process was subjected to various conditions, significant results obtained and observed trends are;

- This study was able to achieve 99.8% efficient removal for nickel using iron electrode at neutral pH (pH 7) within 60 min in the presence 1.12g NaCl electrolyte, 1cm inter-electrode distance and 10g/L pollutant initial concentration.
- Furthermore, 99.3% removal efficiency for TC was achieved using iron electrodes at basic pH (pH 9) within 40 min at 2.24g NaCl electrolyte, 15mg/L TC concentration, and 1cm anode-cathode distance. However, with aluminium electrode, at pH 7 (neutral pH), 0.56g NaCl electrolyte, 15mg/L TC initial concentration, and 1cm inter-electrode distance. The TC efficient removal was 99.9% within 20 min.
- Octahedral Fe-O (600c-1) and tetrahedral Ni-O (488cm-1) were observed during the FTIR of $Ni(NO₃)₂$. $6H₂O$ (Ni) at pH 3-9. Adsorption process occurred on the nanoparticles at all pH, also peaks around 3300, 1645 and 1341 cm⁻¹ in correlated with O-H stretching, N=O stretching and vibration of nitrate asymmetric respectively.
- FTIR of TC shows a peak around 3300 which is referred to as O-H and N-H stretching and 1650-1400cm-1 correlated to as C=O and C=C.
- The optimum pH in this study is pH 7 for nickel using iron electrodes and TC with aluminium electrodes separately and $pH 9$ for TC using iron electrodes.
- EC is an easy and less expensive treatment as compared to other methods. As expected, the treatment costs to remove TC and Ni by Fe were 0.95 \$.m-3 and 1.4 \$.m-3 respectively. However 0.26 \$.m-3 was the treatment cost to remove TC via Al electrodes.

Therefore EC method is the best method for water and wastewater treatment because it is effective and efficient having more than 99% removal efficiency in this study. Thus, using AC as a power source or EC in combination with other treatment methods can be recommended for future applications. AC source reduces the passivation of the electrodes thereby increasing the performance efficiency. These two recommendations will have a higher efficient removal of nickel and TC.

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