ARTIFICIAL BATIK WASTEWATER TREATMENT USING THE INTEGRATION METHOD OF ELECTROCOAGULATION AND ULTRASONIC

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SCHOOL OF CIVIL ENGINEERING
UNIVERSITI SAINS MALAYSIA
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by

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ABSTRAK

Industri tekstil adalah sebuah industri yang melopori penggunaan air dan punca pencemaran sumber air. Saban tahun, penggunaan pewarna fabrik mencapai ratusan tan di seluruh dunia, menghasilkan air buangan yang berkepekatan tinggi. Efluens tekstil dikenal mempunyai warna yang terang, mempunyai nilai COD dan BOD yang tinggi, kandungan logam berat yang tinggi serta bahan-bahan yang berpotensi membahayakan organisma hidup. Industri batik di Malaysia merupakan satu identiti budaya serta industri yang biasanya diusahakan secara kecil-kecilan atau sederhana. Ini bererti, efluens batik yang dihasilkan adalah sedikit jika dibandingkan dengan industri komersial. Kajian ini bertujuan menentukan penyingkiran warna dan penipisin bahan anod, oleh itu, kaedah elektropenggumpalan dipadankan dengan ultrasonik bagi meningkatkan kecekapan, kadar pergerakan bahan dan mengelak kepasifan elektrod. Rawatan ini dilakukan dengan mengambil kira tiga parameter operasi iaitu, jarak antara elektrod, jumlah voltan, dan tempoh rawatan dengan penggunaan elektrod aluminium. Sampel buatan yang disediakan adalah mengikut proses sebenar pembuatan batik. Ia terdiri daripada pewarna, sodium silikat, sodium karbonat, lilin parafin dan air paip. Elektrod dengan jarak 1cm, 2cm atau 3cm disambung kepada punca kuasa aliran terus yg diset pada voltan 5, 10, 15, 20 atau 25 volt. Rawatan dijalankan selama 25 minit di mana, setiap tempoh 5 minit, 10ml sampel diambil dan ditapis untuk analisis warna sebenar dan nyata serta peratus penyingkiran warna. Setiap elektrod yang berfungsi sebagai anod juga dibersihkan dan ditimbang sebelum dan selepas setiap rawatan untuk analisa pengurangan berat elektrod. Analisis menunjukkan peratus penyahwarnaan tertinggi adalah 92.6% dan penceraian 0.3254g berat anod pada 2cm, 25V, pada minit 25 juga menunjukan perubahan warna sampel ungu tua kepada biru muda.

ABSTRACT

The textile industry consumes the most water and pollutes the water source most. Annually, hundreds of tonnes of dyes, mostly synthetic dyes, are being used worldwide which resulted in the textile effluent that generally characterized as high strength. This is due to the strong colours, high concentration of COD and BOD, high level of heavy metals and any other compounds that most likely to be toxic and harmful to the living organism. On the other hand, the batik industry in Malaysia is a part of cultural identity and most likely to exist as small and medium enterprises (SMEs). This means the effluent produced is a lot less than commercialized textile production but also means the effluent does not undergo advanced treatment before released to the water bodies. In this study to determine the dye removal percentage of artificial batik wastewater and anode material consumption, electrocoagulation is integrated with ultrasonic to enhance efficiency, mass transport rate and avoid electrode passivation. This method is executed by considering three operating parameters: inter-electrode distance, amount of voltage and treatment time using aluminium electrodes. The artificial batik sample was prepared according to the batik making process, made of ingredients such as dyes, sodium silicate, sodium carbonate, paraffin wax and tap water. Electrodes at a distance of 1cm, 2cm, 3cm are put into the beaker and connected to the DC power supply at 5V, 10, 15V, 20, 25V. The set-up was running for 25 minutes. Every 5 min interval, a 10mL sample was taken out and filtered for true and apparent colour determination and colour removal percentage. Besides, electrodes used as an anode are cleaned and weighed before and after running the treatment. Time statistical analysis of the data obtained shows the highest removal of dyes is at 2cm, 25V at 25 min with a percentage of 92.6% and 0.3254g of loss in anode weight. The colour of the batik sample went from dark purple to a subtle light blue hue.

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V	Volt for voltage
°C	Degree Celsius
$	Money currency for United States Dollar
%	Percent, out of 100 percentage.
Pt-Co	Platinum-Cobalt, unit of colour
mg/L	Milligram per litre

LIST OF ABBREVIATIONS

BOD	Biochemical Oxygen Demand
COD	Chemical Oxygen Demand
SME	Small and Medium Enterprises
UV	Ultraviolet
DO	Dissolved Oxygen
HDPE	High Density Polyethylene
MYR	Malaysian Ringgit
TSS	Total Suspended Solid
TDS	Total Dissolved Solids
TKN	Total Kjeldahl Nitrogen
EC	Electrocoagulation
EC-US	Electrocoagulation-Ultrasonic
USM	Universiti Sains Malaysia
NaCl	Sodium Chloride
DC	Direct Current
APHA	American Public Health Association

CHAPTER 1

INTRODUCTION

1.1 Research Background

Textile industries are fit to be described as fast-growing and dynamic. In line with the fashion industry, which constantly ever-changing, the demand for textiles is endless. High demand for textile materials leads to the increase of textile factories which unfortunately brings an inversed proportion to the environment. The compositions of textile wastewater effluents vary depending on what fabric and product they are manufacturing. One of the major problems of textile effluent would be dyes as the dyeing process is the most crucial part of textile making. Various chemicals are added into the dyeing process to create a more stable molecular structure of dyes or to enhance the absorption of dyes to fabrics (Yaseen and Scholz, 2019). Moreover, the batik textile commonly produced to be full of vibrant colours resulted in high strength effluent.

Dated back to the 1700 s, ‘batik’ was invented and spread across continents. Centuries long worth of history certainly lands batik a prominent prestige among cultural art practitioners and lovers, which blooms into the paramount creative industry. ’batik’ is a name of wax resisting colour technique. Wax is applied to fabric in patterns to prevent colour absorption creating the motive of the clothes. The origin place of the technique was unclear; however, most believe it brought from the Indian subcontinent and ended up being greatly expressed in the Indonesian culture, specifically in Java. It was later spread to Indonesian archipelagos and the coastline of Malaysia. In Malaysia, batik textile is a part of national identity and an asset to the art and craft industry. Back then, the production of batik in Malaysia is majorly operated on a small scale in East Coast Malaysia (Kelantan, Terengganu and Pahang) in the 1960s. Gradually, the industry has grown into small and medium enterprises (SMEs) and some huge productions. It becomes known locally and globally through the rise of the fashion scene and contemporary touch. By 2004, the local industry was valued at MYR50 million with 70% of the production came from SMEs based in Kelantan and Terengganu.

Annually about a hundred thousand tonnes of dyes is used in textile industries. Dyes could be natural-based or synthetic. However, synthetic dyes are cheaper and easier to obtain resulting in massive usage of synthetic dyes in mass production. In most batik productions in Malaysia, Remazol dyes are used. It is a fibre reactive dye that is typically used to colour silk or wool as it can react to hydroxyl groups forming cellulosic linkages. In addition, Remazol dyes offer a wide range of brilliant hues at affordable prices. The dyes in batik wastewater are a mixture of dyes from various colour spectrums. To execute colour removal of batik effluent, electrocoagulation was deemed effective instead of other methods and this proven by countless studies that show most of the outcome of dyes removal efficiency exceeding 90% (Naje et al., 2017).

In textile wastewater treatment, some methods are already being applied either biologically or chemically. However, biological methods such as oxidation or ozonation have their pros and cons. Though it was great to remove COD, it was not able to entirely decolourize the wastewater and prone to the problem such as bulking. As for chemical methods such as chemical oxidation by ozone or combined UV radiation and ozone, it does work effectively however the cost was too high to bear. For lower energy consumption, smooth operation and cost-efficient, the integrated electrocoagulation-ultrasonic method was an option to consider (Moradi et al., 2021). Furthermore, the electrocoagulation does not have any additional coagulant added as it can be produced in-situ through the sacrificial anode. By combining ultrasound to electrocoagulation, it is said to be more efficient in increasing reaction rate and mass transport rate by cavitation phenomenon (Ritesh and Srivastava, 2020). Henceforth, in this project, the integrated method will be used to determine its effect on the decolourization of dyes in textile wastewater.

1.2 Problem Statement

Water pollution has been a source of concern around the world. This is because water is essential for any living organism. Even though the earth is surrounded by water, there are far fewer potable water sources than saline water. Furthermore, the seas are home to many species and a vital contributor to oxygen, implying that any pollution of water bodies is simply unjustifiable.

On the other hand, the textile industry is booming and is not showing any sign of reversal. Textile demand is ever-increasing, and so are textile manufacturers. The expansion of textile factories results in massive water consumption and the generation of textile wastewater. Untreated textile wastewater has strong colour characteristics, a high number of COD, high suspended solids, abundant heavy metals, etc. Batik industry is one of the textile industries which produces distinctive batik fabrics, full of exquisite colours. The dyes would be the most problematic aspect of the textile effluent. The dyeing process is the most important part of textile making. Various chemicals are added into the dyeing process to create the dyes or enhance dyes' absorption into fabrics. Untreated effluents are full of chemicals, which make them harder to degrade due to various molecular structures. Those effluents might be too carcinogenic and might as well be mutagenic. The dyes that pollute wastewater not only reduce the aesthetic value but also limit light penetration into the surface water, disrupting photosynthesis and collapsing the ecosystem.

Concerning the batik industry in Malaysia, specifically in Kelantan, the effluent from the small factory of batik houses is often released to the rivers or streams nearby. This may seem unfit and legally incorrect. However, the release of raw effluent to the nearby water bodies is unfortunately common. This is because society is not well-versed in the incoming harms that untreated wastewater can bring. Moreover, the application of advanced treatment of wastewater is still foreign and unfamiliar technology to the Malaysian. The field is yet to be delved into. In addition, these small-scale businesses do not have the capital to assemble a build-in system of wastewater treatment which often on the higher side of cost. Therefore, it is important to find simple and low-cost technology such as electrocoagulation.

In this research project, the main purpose is to treat batik wastewater focusing on dye removal through the integration method of electrocoagulation and ultrasonic. The treatment will consider the factors of electrodes distance, the amount of voltage supplied and treatment time. The initial characterization of the sample will be done before and after the treatment to observe any significant changes.

1.3 Objectives

1. To determine the efficiency of integration method of electrocoagulation and ultrasonic on the percentage of dye removal in textile wastewater.

2. To determine the factors influencing electrocoagulation-ultrasonic performance, ie treatment time, inter electrode distance, and voltage.

3. To calculate the anode material dissociation in terms of post-integrated treatment weight.

1.4 Scope of Work

- The batik wastewater sample used in this research is made to replicate real batik wastewater. Due to the COVID-19 situation, it is hard and presumptuous to obtain real batik wastewater from the local batik factory.
- The artificial wastewater is made of dyes, sodium silicate, sodium carbonate, paraffin wax and tap water to enhance the characteristics of high strength textile wastewater.
- The initial characterization of the artificial sample is taken in terms of the measurement of colour, pH, temperature, turbidity, COD, DO and salinity. Then the sample was kept in an HDPE container and stored in the cold room at 4 ℃ to minimize changes.
- The integration of electrocoagulation and ultrasonic is to observe the efficiency of dyes removal of batik wastewater.
- The aluminium plate is used as an electrode in electrocoagulation for both anode and cathode.
- The time-based statistical analysis will consider the effects of voltage supplied, inter-electrode distance, and reaction time upon the percentage of colour removal percentage and anode consumption at laboratory-scaled setting.

1.5 Thesis Outline

This thesis is arranged into a sequence of five major chapters. The chapters consist of introduction, literature review, research methodology, results and discussions, and conclusion and recommendations.

- Chapter 1 introduces the background of this study. It will state the problem statement, the objectives, and the scope of this study.
- Chapter 2 is the literature review of the topic of this study. This chapter gathers and discusses the information, also findings written by other scholars related to the topic.
- Chapter 3 highlights the research methodology. This chapter meticulously writes the flow of experimental work and methods used to achieve the objectives stated.
- Chapter 4 is the result and discussion. The obtained results are analysed and discussed with supporting findings of recent studies.
- Chapter 5 concludes the study objectives. Then, recommendations are given for improvement in further research.

CHAPTER 2

LITERATURE REVIEW

2.1 Textile Wastewater

Derived from the Latin word 'texere' which brought the meaning of to weave, textile is defined as woven fabrics or clothes made by hand or machines in huge numbers. Textile wastewater refers to the by-product of processes required to make the textile. Textiles have differed from the basic unit of fibre used to produce the fabric, henceforth differs it wastewater compositions. For instance, cellulosic, protein and synthetic are the main categories of fibre used. It is either extracted from plants, animals or man-made through technological advancement. The textile industries are massive and one of the earliest mass-produced down the history. The industries are in favour of the economy as the demand for fabrics fuels the factories growth. Furthermore, it is profitable and provides job opportunities either for skilled or unskilled workers. Besides, China valued the textile industry at $94.4 billion, leading the rank of textile exporter and manufacturer (Ananthashankar, 2013; Yaseen and Scholz, 2019). In 2015, the value of the world textile industry market come out at $667.5 billion with 54.6% is contributed by the Asia-Pacific region and Europe accounted for a further 20.6% of the market. By the year 2020, the global textile industry market is expected to hit $842.6 billion, an increase of 26.2% since the year 2015 (Sivaram et al., 2018).

Along with the increment of industry value, textile wastewater generation is expected to triple the current volume. This causes a huge concern regarding the chemical loads that its effluent carries to the environment. It is estimated that China, the United States, and the United Kingdom the leading textile manufacturer, annually discharge 26.0, 12.4 and 1.0 million tons of textile wastewater, respectively. As textile production involve various processing and chemical operations like sizing, resizing, scrubbing, mercerizing, decolorizing, dyeing, printing, and finishing, more water is consumed. Typical textile production generally consumes around 1.6 million litres of groundwater to produce 8000 kg of textile fabric per day, out of which 30–40% water is used in the dyeing process, 60–70% of washing stage and 10–50% of unused dyes are released into the aquatic resources along with the generated wastewater. The printing, bleaching, scouring and finishing processes in the textile industry also discharge a large volume of wastewater up to 1–10 million litres per day (Kishor et al., 2021).

Furthermore, more than 500 tons of dyes are also discharged into the effluent of the textile industry worldwide. Henceforth, about a hundred thousand tonnes of dyes are used in textile industries annually and 40% of globally used colourants contain organically bound chlorine, a known carcinogen. All the organic materials present in the wastewater from the textile industry are of great concern in water treatment because they react with many disinfectants, especially chlorine(Mia et al., 2019). Thus, the effluent is serious environmental pollution that can cause serious health hazards.

On the other hand, the batik industry is one of the prominent industries in Malaysia. Annually, it was valued at MYR370 up to MYR400 million according to (Ismail et al.,2013). The industry is expected to be growing as it readily approaches the global market through the fashion industry. The batik industry in Malaysia mostly operated in East Coast Malaysia as small and medium enterprises. It also commonly run as family-owned small businesses. Hence, most entrepreneur of batik production houses often oversees the issue of batik effluent. Most batik effluent is often drained untreated to the nearest drains or rivers. The wastewater has slightly different to the other textile wastewater due to batik techniques which use wax and resin, then coloured in vibrant colours. The batik wastewater contains dyes, wax, resin, sodium silicate and sodium carbonate. Dyes and chemical found in batik effluent is abundance due to unfixed dye and chemicals which loosen when going through its washing processes. Moreover, it is said about 10% – 15% of the total dyes and chemicals are released to the effluent. Those components were undoubtedly harmful to aquatic lives. (Subki, 2017)

2.2 Typical Characteristics of Textile Wastewater

Over the years, countless research had been done upon textile wastewater. However, the effluent of the textile industry may differ due to fabric materials and textile processes (Kishor et al., 2021). The table below shows the typical characteristics of textile effluent according to previous researchers.

Table 2.1: General Parameter of Textile Wastewater

Parameter	Unit	Range
Temp.	℃	21-60
pH	—	5.5-11.8
Colour	Pt-Co	50-2500
COD	mg/L	150-30000
BOD	mg/L	80-6000
TSS	mg/L	15-8000
TDS	mg/L	1500-6000
TKN	mg/L	70-80
Oil and Grease	mg/L	5-50
Chlorides	mg/L	200-6000
Sulphates	mg/L	500-1000
Zinc	mg/L	<10
Copper	mg/L	<10
Potassium	mg/L	30-50
Sodium	mg/L	400-7000

The table is summarised by (Yaseen and Scholz, 2019).

2.3 Pollutions by Textile Wastewater

Textile wastewater is loaded with pollutants such as dyes, metals, salts, surfactants, organic processing aids, sulphates, and formaldehyde. These pollutants are being added in stages during textile making processes. In addition, textile wastewater is characterized by the high dye content, high temperature, pH, BOD, COD, TDS, TS, SS, TOC, TSS, chlorides and phosphate etc(Mia et al., 2019; Yaseen and Scholz, 2019). Its effluent also contains sulphates, nitrates, electrical conductivity, turbidity, alkalinity, salts, acids, bases, mordants, surfactants, VOCs, chlorobenzenes, phenols, dioxin, bleaching, fixing, and finishing agents along with various metals like Cr, Cd, Pb, Sb, As, Cu, Ni and Zn etc (Hussain et al., 2019). The presence of an excessive inorganic pollutant in textile wastewater is an insurmountable problem. The existence of inorganic chemicals and soluble salts in textile effluent are harmful to the aquatic ecosystem even in a small amount. These cause oxygen depletion and lead organisms to experience chemical and biological changes (Maman et al., 2020). Plus, the suspended solids found in the effluent could also threaten marine lives as they can clog fish gills. In contrast, the presence of sulphate and phosphate in wastewater is non-toxic to aquatic lives at normal concentrations and only harmful at immense concentrations. However, to humans, sulphates could take effect even in a small amount. It can cause purgative effects and illness in the long run.

Many types of dyes used in the textile industry are not only disrupting the aesthetical value of receiving water bodies, but it has also proven to be toxic, carcinogenic, and mutagenic. In addition to the fact stated, around 20000 kg of highly toxic acidic dyes is discharged from each washing process of polyamide textiles and direct dyes are released from the washing stage of cotton manufacturing industries (Kishor et al., 2021). In addition, batik effluent often drained to the nearest water bodies without going through necessary treatment or following regulations set (Subki, 2017). According to Ananthashankar et al. lung and severe skin irritations, physical abnormalities, headache, and nausea can be observed on humans constantly exposed to textile dyes. It also stated by Lima et al, that benzidine, a carcinogen in textile effluent and Mathur et al found a mutagenic agent when testing seven common textile dyes and stated that, the only direct violet dye has a lower mutagenicity ratio than 2:0. Cremazoles dyes are highly toxic to any microorganism according to Morikawa et al. In addition, textile production workers were reported to be diagnosed with cancerous diseases after being exposed to textile dyes for a long time. Same cases with reactive dyes, it could trigger asthma, dermatitis, and nasal problems (Ananthashankar, 2013). On the other hand, the strong colours of textile effluent restrict the light penetration and highly toxic to aquatic lives. Henceforth, it inhibits photosynthesis and increases biological oxygen demand also eventually threaten aquatic lives (Yaseen and Scholz, 2019).

2.4 Electrocoagulation – Ultrasonic

To remove dyes in textile effluent, electrocoagulation is one of the most effective options which also reduces sludge generation. Electrocoagulation is the process that involves flocculation and floatation (Sadik, 2019). Pollutant removal in electrocoagulation can be simplified into three stages. Firstly, a coagulant is formed by an oxidation reaction at the anode. Then, ions and colloids in the electrolyte, which both opposite charges from anode dissolution destabilizes colloids, allowing them to coagulate. Lastly, sludge is formed from the aggregation of unstable materials(Moradi et al., 2021). Coagulant agents in electrocoagulation can be generated with the presence of a sacrificial anode due to its dissolution when electrical current is introduced to both cathode and anode (Sadik, 2019).

The ultrasonic waves are a sound frequency higher than human hearing. For ultrasonic, it is the usage of ultrasound waves to degrade pollutants in a process of advanced oxidation. However, the degradation of pollutants in ultrasonic are incomplete due to the limitation of range in frequency of ultrasonic waves being generated by ultrasonic power range. Therefore, it is favourable to integrate another method to optimize the ultrasonic method (Ritesh and Srivastava, 2020). It is also proved by researchers that ultrasonic-assisted processes in significantly shown higher efficiency (Özyonar et al., 2020).

By the integration of electrocoagulation – ultrasonic as pictured in Figure 2.1, it can overcome the weaknesses of both methods. During electrocoagulation, the passive film can form around the electrodes, causing higher power consumption with lower pollutant removal (Moradi et al., 2021). The acoustic stream of ultrasound offers electrodes cleaning which increases the chemical reaction rate and mass transfer rate of pollutants. The continuous synergy will also lower the degree of mineralization and passivation of electrodes, henceforth lengthen its life. Ultrasound also increases the formation of hydroxyl (OH) which reacts to pollutants(Emerick et al., 2020). To sum up the integration method, it creates the balance where the ultrasonic increase the mass transport rate to the electrodes which increase coagulation. The increase of reaction can be catered by an electrochemical cell which being added by ultrasonic waves (Ritesh and Srivastava, 2020). For instance, the electrocoagulation–ultrasonic method showed an impressive outcome of 100% colour removal when treating pulp and paper industry effluent (Asaithambi et al., 2017).

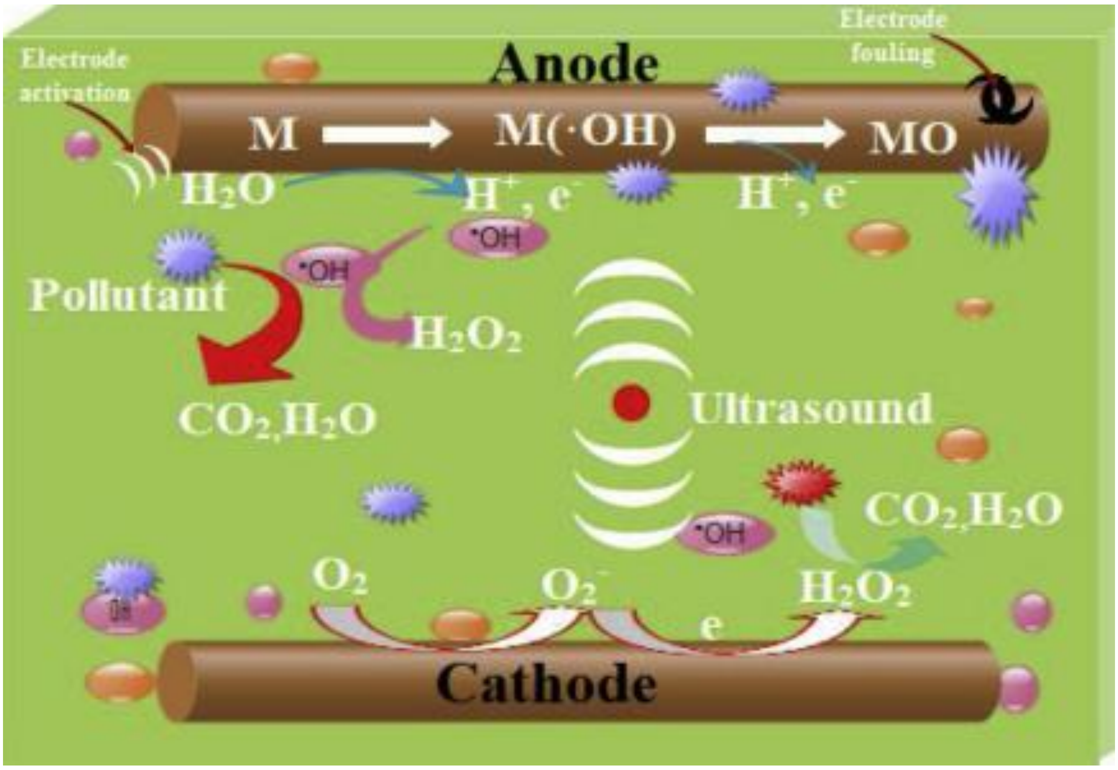

Figure 2.1 Illustration of Electrocoagulation-Ultrasonic process (Ritesh and Srivastava, 2020).

2.5 Electrodes

Electrodes played huge roles as a part of electrocoagulation. The use of material, surface area, arrangement, and combination of electrodes pairs in electrocoagulation can give a significant outcome to the process. Electrocoagulation does begin with the reaction at both electrodes. At the anode, metal is oxidized.

$$M \rightarrow M^{Z+} + Ze^{-} \quad (2.0)$$

M represent the type of metal used as an electrode, meanwhile, Z is the number of electrons that dissociates from anode per mole of metal.

For the cathode, reduction of water into hydrogen gas and hydroxyl anions occur.

$$2H_20 + 2e^{-} \rightarrow 2OH^{-} + H_2 + 2H^{+} + 2e^{-} \rightarrow H_2 \quad (2.1)$$

An integrated method of electrocoagulation and ultrasonic, the combination increases bubble buoyancy through cavitation and rise the coagulant content. This leads to more dissociation of metal ions from the anode (Moradi et al., 2021). Meanwhile, ultrasonic enhances the collisions between coagulants and pollutants in pollutant removal. Plus, the hydroxyl radical species electrolyte (NaCl) can be added in a small amount to increase the performance of the system by producing chlorine radical (Ahangarnokolaei et al., 2018).

The materials and combination of electrodes pair show a different outcome of each configuration. For instance, in Table 2.2, the study review electrocoagulation dyes removal efficiency with different electrodes configuration (Naje et al., 2017). It results show a significant difference between each combination of electrode pairs yet most of the outcome displayed a rather high amount removal efficiency percentage, which is around 90% for most cases.

Table 2.2: EC process used for removal of various types of dyes.

Dye	Current or current density	Time (min)	Anode-cathode	Removal efficiency (%)	Reference
Reactive Red 120	75 A/m2	60	Fe-Fe	95	Pirkarami and Olya (2014)
CI Reactive Blue 25	2.5 mA/cm2	90	Al-Al	97	Singh and Ramesh (2014)
Reactive Orange 16	20 mA/cm2	30	Fe-Fe	95	Alizadeh et al. (2014)
Brilliant Green	139 A/m2	30	Fe-Fe	93	Nandi and Patel (2013)
Acid Yellow 23	120 A/m2	60	Fe-Fe	93	Modirshahla et al. (2013)
Acid Red 73	80 A/cm2	90	Al-Al	95	Mahmoodi and Dalvand (2013)
CI Direct Blue 1 CI Fast Red B base CI Green G	1 A	20	Al-Al	88, 90, 92	Lambert et al. (2011)
Reactive Blue 19	30 mA/cm2	90	Fe-Fe	98	Khoomsab and Khummongkol (2013)
Reactive orange 84	130 A/m2	90	SS-SS, Fe-Fe	66, 76	Yuksel et al. (2013)
Acid red 131, Reactive yellow 86, Indanthrene blue RS, Basic GR 4, Reactive yellow 145	0.0625 A/m2	60	Al-Al	97	Khandegar and Saroha (2013a)
Reactive black B, Orange 3R, Yellow GR	0.0625 A/m2	60	Al-Al	98	Khandegar and Saroha (2013b)
Azo, Anthraquinone, Xanthene	0.3 A	90	Fe-Fe, Fe-SS	98	Wei et al. (2012)
Acid black 52, Acid yellow 220	40 A/m2	90	Al-Al	92, 94	Pajootan et al. (2012)
Levafix brilliant blue E-B	100 A/m2	120	Al-Al, Fe-Fe	95, 83	Akbal and Kuleyin (2011)

Acid, Reactive	4 mA/cm2	120	Fe-Carbon	95	Körbahti et al. (2011)
Disperse red	20.8 mA/cm2	90	Al-Al	95	Merzouk et al. (2011a)
Acid Brown 14	6.329 A/m2	60	Al-SS	Bench scale: 91 Pilot scale: 80	Parsa et al. (2011)
Reactive black 5	7.5 mA/cm2	60	Fe-SS	90	Patel et al. (2011)
Indigo carmine	54.45 mA/cm2	120	SS-SS	99	Secula et al. (2011)
Direct red 81	1.875 mA/cm2	60	Al-Al	98	Aoudj et al. (2010)
Crystal violet	28 A/m2	90	Fe-Al, Al-Fe	96, 98	Durango-Usuga et al. (2010)
Remazol red 3B	15 mA/cm2	90	Fe-Fe	97	Kobya et al. (2010)
Reactive blue 140, Direct red 1	40 A/m2	120	Fe-Fe, Al-Al	>95	Phalakornkule et al. (2010a)
Orange II	160 A/m2	90	Al-Al	94	Mollah et al. (2010)
Reactive black 5	4.575 mA/cm2	60	Al-Al	95	Şengil and Özacar (2009)
Direct red 23	30 A/m2	80	Fe-Fe, Al-Al	>94	Phalakornkule et al. (2009)
Acid Red 14	102 A/m2	60	Fe-SS	>91	Aleboyeh et al. (2008)
Levafix blue CA	35.5 mA/cm2	90	Fe-Fe	96	Körbahti and Tanyolaç (2008)
Acid yellow 23	112.5 A/m2	60	Al-SS	98	Daneshvar et al. (2007)
Basic red 46, Basic blue 3	80 A/m2	120	Fe-SS	98	Daneshvar et al. (2006)
Acid Yellow 36	127.8 A/m2	90	Fe-Fe	83	Kashefialasl et al. (2006)
Reactive Blue 19, Acid red 266, Disperse Yellow 218	1.6 A	120	Al-Graphite, Fe-Graphite	95	Yang and McGarrahan (2005)

Methylene blue, Eosin yellowish	16.1 mA/cm2	90	SS-SS	95, 75	Golder et al. (2005)
Acid red 14	80 A/m2	120	Fe-SS	93	Daneshvar et al. (2004)
Remazol red RB 133	15 mA/cm2	60	Al-Al	92	Can et al. (2003)

Source by (Naje et al., 2017)

It is noted, the aluminium electrode in electrocoagulation performs better at a wide range of pH, removal of dyes and COD (Özyonar et al., 2020; Yaseen and Scholz, 2019), resulted in the elimination of alcohol-based compounds with genotoxic effects (Naje et al., 2017).

Furthermore, as the electrodes undergo oxidation and reduction, the electrode change often observed in terms of electrode consumption. According to Faraday's Law, the quantity of chemical change produced by a current at an electrode-electrolyte boundary is proportional to the quantity of electricity used. It is also stated that the amounts of chemical changes produced by the same quantity of electricity in different substances are proportional to their equivalent weights. Following the theory, electrode consumption often determined in terms of energy or power consumption. It is calculated by the formulas:

$$SEC = \frac{UIt}{V.(C_0 - C)} \qquad (2.2)$$

SEC stands specific energy consumption in the unit of kWh/kg $dye_{removed}$, U is for voltage (V), I is for electric current (A), t is the treatment time (hr), V is wastewater volume (L) while C_0 and C is the initial and instant dye concentration (g/L).

$$m = \frac{ItM}{F.Z} \qquad (2.3)$$

Equation 1.4 is to calculate m, dissolved metal mass (g) for anode dissociation in Equation 1.5. Then I stand for electric current (A), t is electrolysis time (s), M is molar mass, F is the Faraday Constant (96,485 C/mol) and z is metal valence.

$$Anode\ Dissociation = \frac{m}{V.(C_0 - C)} \qquad (2.4)$$

2.6 Factors Affecting Considered:

For the integration method of electrocoagulation and ultrasonic, some factors are being taken into consideration to optimize the effect of this treatment method. Factors affecting considered are the distance between electrodes, amount of voltage supplied and lastly is the treatment time.

2.6.1 Electrodes Distance

The distance between electrodes during electrocoagulation is said to be affecting the performance of the electrodes. According to Ohm's Law, decreasing the distance lowers the resistivity hence increase greater current. Besides, lower resistivity means a better mass transport rate and efficient electrocoagulation (Ritesh and Srivastava, 2020). Furthermore, the farther inter-electrode distance requires a higher current to oxidise sacrificial anode (Changmai et al., 2020). However, a close distance of electrode can create a high density of electrostatic field which collisions rapidly occurs and prevent coagulation (Omwene et al., 2020). Therefore, the optimization of electrode distances is crucial and it is suggested by most that inter-electrode distance ranges between 1cm – 3cm (Dimoglo et al., 2019; Elazzouzi et al., 2017; Nawarkar and Salkar, 2019)

2.6.2 Voltage Supply

In this treatment method, the amount of voltage supplied by the DC power supply is considered. Electrical potential often associated with current density, the distribution of electrical current over the surface area. The higher voltage supplied resulted in higher current density. The direct current electric field-induced electrochemical reaction when introduced to electrodes (Sadik, 2019). It is stated that the amount of voltage supplied affects its thermodynamic conditions. If the voltage supply is high, the bubble density is high with smaller and more bubble produced (Bazrafshan et al., 2014). And it goes vice versa for the lower voltage supply. It also said the operating voltage has quite an influence on pollutant removal (Naraghi et al., 2018)

2.6.3 Treatment Time

Hypothetically, the treatment time is linearly proportional to dyes removal percentage. The longer the running time, the better the efficiency of the treatment until the electrode reaches its limiting capacity or the reaction rate has reached the equilibrium condition (Oden and Sari-Erkan, 2018). Many studies have shown that the reaction time is an important operating parameter to the electrocoagulation process (Oden and Sari-Erkan, 2018). It is also stated that the treatment time affected the removal efficiency as it determines the charge loading. With increasing time, the pollutant removal increases up to an optimum state then remain constant (Verma, 2017).

2.7 Advantages and Disadvantages

The integrated electrocoagulation – ultrasonic has its advantages and disadvantages.

Advantages

- Increase pollutant mass transport across electrodes (Ritesh and Srivastava, 2020)
- Eliminate electrode fouling (Ritesh and Srivastava, 2020)
- Eliminate passivation layer from the electrode surface (Moradi et al., 2021)
- In situ generation of the oxidizing agent without chemicals (Asaithambi et al., 2017)
- Reduce mineralization energy usage (Ritesh and Srivastava, 2020)
- Higher degradation rate (Özyonar et al., 2020)
- Energy efficient (Asaithambi et al., 2017)
- Safe operational condition (Tahreen et al., 2020)
- Manageable automation (Sadik, 2019)

Disadvantages

- Inefficient energy conversion (electrical – cavitation)
- High power density (Tahreen et al., 2020)
- High treatment cost (Verma, 2017)

2.8 Wastewater Standard

In Malaysia, the effluent generator must comply with Environmental Quality Act 1974 and subsidiary legislation act, Environmental Quality (Industrial Effluent)

Regulations 2009. Table 2.3 shows the acceptable conditions for discharge of industrial effluent or mixed effluent of standards A and B. Therefore, the batik effluents should comply with the scheduled characterization before draining out to any water bodies.

Table 2.3: Discharge of Industrial Effluent or Mixed Effluent of Standards A and B

Parameter		Unit	Standard	
			A	B
	(1)	(2)	(3)	(4)
(i)	Temperature	^{o}C	40	40
(ii)	pH Value	-	6.0-9.0	5.5-9.0
(iii)	BOD at $20^{o}C$	mg/L	20	50
(iv)	Suspended Solids	mg/L	50	100
(v)	Mercury	mg/L	0.005	0.05
(vi)	Cadmium	mg/L	0.01	0.02
(vii)	Chromium, Hexavalent	mg/L	0.05	0.05
(viii)	Chromium, Trivalent	mg/L	0.20	1.0
(ix)	Arsenic	mg/L	0.05	0.10
(x)	Cyanide	mg/L	0.05	0.10
(xi)	Lead	mg/L	0.10	0.5
(xii)	Copper	mg/L	0.20	1.0
(xiii)	Manganese	mg/L	0.20	1.0
(xiv)	Nickel	mg/L	0.20	1.0
(xv)	Tin	mg/L	0.20	1.0
(xvi)	Zinc	mg/L	2.0	2.0
(xvii)	Boron	mg/L	1.0	4.0
(xviii)	Iron (Fe)	mg/L	1.0	5.0
(xix)	Silver	mg/L	0.1	1.0
(xx)	Aluminium	mg/L	10	15
(xxi)	Selenium	mg/L	0.02	0.5
(xxii)	Barium	mg/L	1.0	2.0
(xxiii)	Fluoride	mg/L	2.0	5.0
(xxiv)	Formaldehyde	mg/L	1.0	2.0
(xxv)	Phenol	mg/L	0.001	1.0
(xxvi)	Free Chlorine	mg/L	1.0	2.0
(xxvii)	Sulphide	mg/L	0.50	0.50
(xxviii)	Oil and Grease	mg/L	1.0	10
(xxix)	Ammoniacal Nitrogen	mg/L	10	20
(xxx)	Colour	ADMI*	100	200
*ADMI-American Dye Manufacturers Institute				

This table is from the Malaysia Department of Environment (DOE) Environmental Quality Act 1974 and subsidiary legislation act, Environmental Quality (Industrial Effluent) Regulations 2009.

CHAPTER 3

RESEARCH METHODOLOGY

3.1 Introduction

The project revolves around textile wastewater treatment using the integration method of electrocoagulation and ultrasonic. It is to observe the colour removal of batik dyes while considering the affecting factors of electrode distances, the amount of voltage supplied and duration of treatment time. The sample of textile wastewater is an artificial sample of batik wastewater.

Batik wastewater commonly made up of dyes, soda ash, water glass, wax and tap water. The process of making artificial sample is done through the observation of the batik production process by local enterprises. The ingredients needed for the artificial sample were also supplied to local art & craft store. In this chapter, the materials and apparatus will be explained in detail. The ingredients were mixed all together and kept in a 10L HDPE bottle, then were stored in the cold room at 4 ℃. After the mixing, the sample was tested using a YSI multi-meter for the raw analysis. The tests for colour, COD and turbidity were done right after that.

Next, the wastewater sample is run through an electrocoagulation – ultrasonic setup. A 150mL sample poured into a 250mL beaker was set up with two aluminium electrodes connected to the power supply. Then, the beaker was placed into an ultrasonic cleaner filled with distilled water. Both the power supply and ultrasonic bath were run at the same time. A 10 mL of sample is taken at the 5 minutes time interval to calculate its percentage of dyes removal. In addition to that, the treated sample is observed post-treatment.

3.2 The Flowchart for Methodology

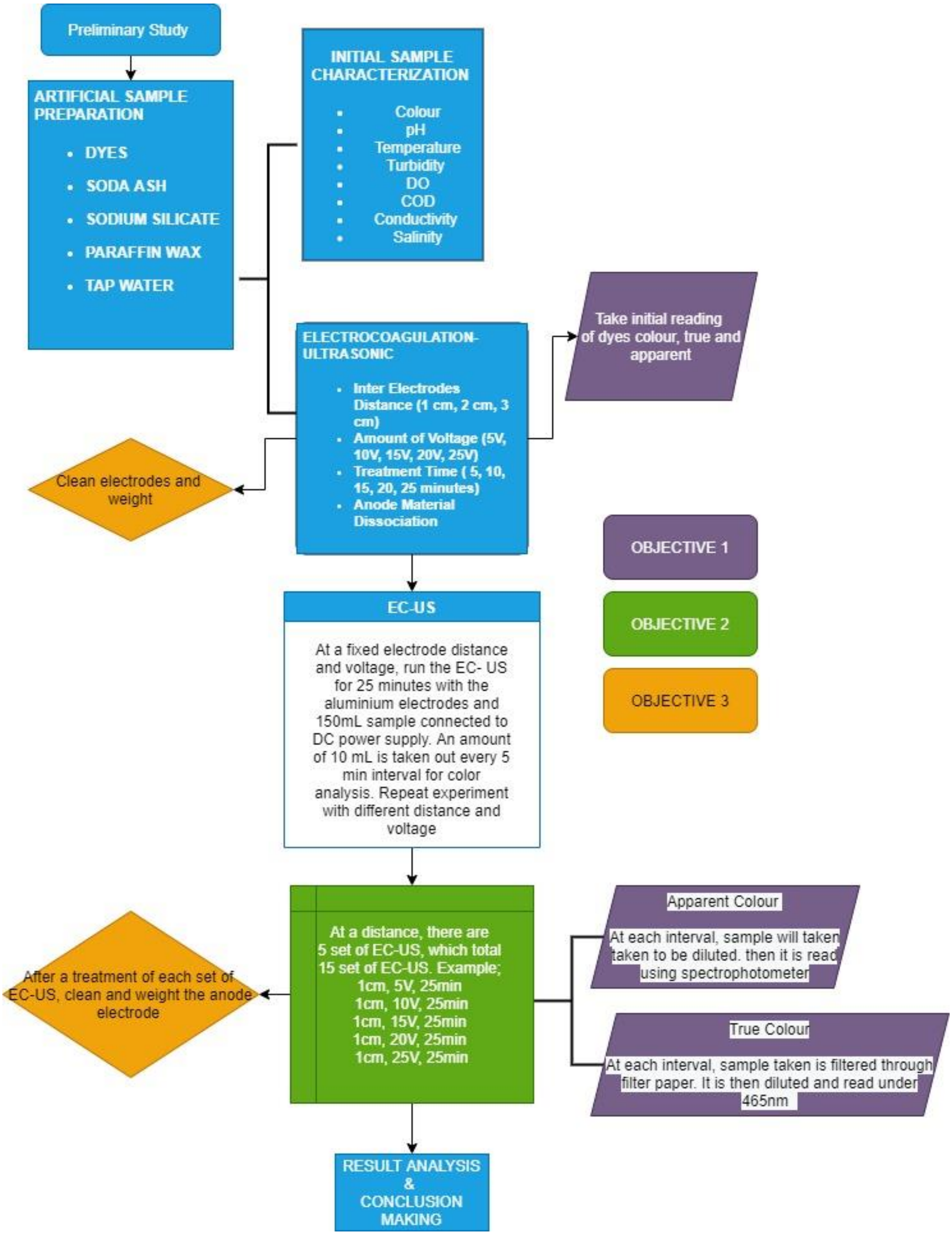

Figure 3.1 Flowchart of Research Methodology

3.3 Artificial Sample Preparation

The sample used in this study is artificial batik wastewater made of Remazol dyes, sodium silicate, sodium carbonate and resin mixed paraffin wax added into tap water. It was later stored in a 10L HDPE bottle and was refrigerated at 4 ℃. The process of making the artificial sample was executed through the information gathered on batik production processes. It is to simulate the real effluent from the batik factory.

For artificial batik wastewater,

- Remazol Dyes (Bottle Green, Blue KNR, Red 2G) = 75g
- Soda Ash = 600g
- Sodium Silicate = 500 mL
- Paraffin Wax (mixed with resin) = 19g
- Tap water = 10L

All the materials are thoroughly weighed and added to the sample bottle. As for the paraffin wax, it was heated in the oven lab for around 10 minutes at 110℃ to be melted down. The proportion of materials are based on the batik making process seen and explained through the mybatik.org website and the Department of Environment article for the Cleaner Production campaign (Masrom, 2012). It was also based on Subki, (2017) study on the traces of dyes that went to batik effluent which 10 – 15 % of the number of dyes used in batik production. Next, those materials were added to the bottle and were given a good shake to ensure everything was well combined. Then, take 1L from the sample into another 10L HDPE bottle, fill the container up to brim with distilled water. This is to dilute the sample to one-tenth to match the apparatus capability.

Later, the sample was tested using YSI multi-parameter probe as shown in Figure 3.2, then was stored in a cold room at 4 ℃ exclude the amount needed for the raw analysis.