# การพัฒนาถังปฏิกิริยารวมตะกอนทางไฟฟ้ารูปแบบใหม่สำหรับการกำจัดความขุ่นและสีในน้ำเสีย จากอุตสาหกรรมสิ่งทอ



## จุหาลงกรณ์มหาวิทยาลัย

บทคัดย่อและแฟ้มข้อมูลฉบับเต็มของวิทยานิพนธ์ตั้งแต่ปีการศึกษา 2554 ที่ให้บริการในคลังปัญญาจุฬาฯ (CUIR) เป็นแฟ้มข้อมูลของนิสิตเจ้าของวิทยานิพนธ์ ที่ส่งผ่านทางบัณฑิตวิทยาลัย

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### Development of Novel Electrocoagulation Reactor (ECR) for Turbidity Removal and Decolorization from Textile Industry Wastewater



A Thesis Submitted in Partial Fulfillment of the Requirements for the Degree of Master of Engineering Program in Environmental Engineering Department of Environmental Engineering Faculty of Engineering Chulalongkorn University Academic Year 2017 Copyright of Chulalongkorn University

Thesis Title	Development of Novel Electrocoagulation Reactor (ECR) for Turbidity Removal and Decolorization from Textile Industry Wastewater
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เพิ่งหัว ฮง : การพัฒนาถังปฏิกิริยารวมตะกอนทางไฟฟ้ารูปแบบใหม่สำหรับการกำจัดความขุ่น และสีในน้ำเสียจากอุตสาหกรรมสิ่งทอ (Development of Novel Electrocoagulation Reactor (ECR) for Turbidity Removal and Decolorization from Textile Industry Wastewater) อ.ที่ปรึกษาวิทยานิพนธ์หลัก: รศ. คร. พิสุทธิ์ เพียรมนกุล, อ.ที่ปรึกษา วิทยานิพนธ์ร่วม: คร. ณัฐวิญญ์ ชวเลิศพรศิยา, 166 หน้า.

งานวิจัยนี้มีวัตถุประสงค์เพื่อพัฒนาถังปฏิกิริยาสำหรับกำจัดความขุ่นและสีออกจากน้ำเสียจาก การฟอกย้อมด้วยกระบวนการรวมตะกอนด้วยไฟฟ้า (Electrocoagulation, EC) และการตกตะกอน โดย ้ทำการทดลองกับน้ำเสียสังเคราะห์ด้วยสีย้อมและเบนโทในท์ซึ่งเป็นตัวแทนของสีและความขุ่นในน้ำ ้ตามลำคับ และใช้อลูมิเนียมเป็นขั้วไฟฟ้า เพื่อหาสภาวะการเดินระบบที่เหมาะสม ได้แก่ วิธีการจัดเรียง ้ขั้วไฟฟ้า ระยะห่างระหว่างขั้ว และความหนาแน่นของกระแสไฟฟ้า ในถังปฏิกิริยารวมตะกอนด้วยไฟฟ้า (Electrocoagulation reactor, ECR) ขนาด 4 ลิตรที่พัฒนาขึ้นมา ผลการทดลองพบว่า สภาวะเดินระบบ ที่ดีที่สุด คือ การจัดเรียงขั้วไฟฟ้าแบบ Monopolar ให้มีระยะห่างระหว่างขั้ว 1.5 เซนติเมตร และใช้ความ เข้มข้นกระแสไฟฟ้าที่ 1.5 มิลลิแอมแปร์ต่อตารางเซนติเมตร เมื่อพิจารณาจากอัตราการสร้างแก๊สและ ้น้ำหนักขั้วไฟฟ้าที่สูญเสีย แต่เมื่อทดสอบการบำบัดน้ำเสียสังเคราะห์ พบว่าค่าความหนาแน่นกระแสไฟฟ้า ที่เหมาะสมจะเท่ากับ 4.5 มิลลิแอมแปร์ต่อตารางเซนติเมตร ซึ่งให้ประสิทธิภาพการกำจัดสีและความขุ่น เท่ากับ 95 เปอร์เซ็นต์ ภายใน 30 นาที อย่างไรก็ตาม เมื่อเปรียบเทียบกับกระบวนการ โกแอกกูเลชันที่มี การเติมสารเคมี พบว่าทั้งสองกระบวนการสามารถกำจัดความขุ่นและสีออกจากน้ำได้อย่างมีประสิทธิภาพ ภายใต้ความเข้มข้นที่แตกต่างกัน กล่าวคือ กระบวนการ โคแอกกูเลชันที่มีการเติมสารเคมีจะมี ประสิทธิภาพสูงเมื่อใช้กำจัดความขุ่นเริ่มต้นต่ำกว่า 50 NTU แต่กระบวนการ EC จะเหมาะกับความขุ่นที่ ้สูงกว่า นอกจากนี้ ยังสามารถเพิ่มประสิทธิภาพการบำบัคของถังปฏิกิริยาได้โดยการติดตั้งแผ่นกั้นภายใน ้ถังเพื่อเพิ่มเวลากักของน้ำในถัง โดยพบว่าอัตราการใหลของน้ำเสียที่เข้าส่ถังที่เหมาะสม คือ 2 ลิตรต่อนาที ซึ่งให้ประสิทธิภาพการกำจัดความข่นและสีย้อมเท่ากับ 94 เปอร์เซ็นต์ และ 96 เปอร์เซ็นต์ ตามลำดับ นอกจากนี้ ยังสามารถเพิ่มประสิทธิภาพการตกตะกอนเพื่อแยกความขุ่นได้เพิ่มเติมจากการติดตั้ง Tube settler ในส่วนการตกตะกอนของถัง โดยเมื่อวิเคราะห์รูปแบบการใหลของถังปฏิกิริยา ECR ที่มีการติด แผ่นกั้นด้วยการทดลอง Residence time distribution (RTD) พบว่า รูปแบบการใหลภายในถังมีลักษณะ เป็นการใหลภายในท่อ (Plug flow)

ภาควิชา	วิศวกรรมสิ่งแวคล้อม	ถายมือชื่อนิสิต
สาขาวิชา	วิศวกรรมสิ่งแวคล้อม	ลายมือชื่อ อ.ที่ปรึกษาหลัก
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KEYWORDS: ELECTROCOAGULATION PROCESS; REACTIVE DYE 135; RESIDENCE TIME DISTRIBUTION (RTD), TEXTILE INDUSTRY WASTEWATER, TURBIDITY REMOVAL

PENGHOUR HONG: Development of Novel Electrocoagulation Reactor (ECR) for Turbidity Removal and Decolorization from Textile Industry Wastewater. ADVISOR: ASSOC. PROF. PISUT PAINMANAKUL, Ph.D., CO-ADVISOR: NATTAWIN CHAWALOESPHONSIYA, Ph.D., 166 pp.

High concentration of dyes together with turbidity were frequently found in the effluents of textile industry wastewater, which are the toxic substances. To remove these contaminants, the combination between electrocoagulation process (EC) and separation has been proposed in this study. The objective of this present work is to design and evaluate the new electrocoagulation reactor (ECR) for treating dye and turbidity from synthetic textile wastewater. The optimization of electrode configuration and design parameter were examined with the batch column reactor for containing 4 liters of wastewater. The result showed that monopolar arrangement within the inner gap 1.5 cm and current density 1.5 mA/cm<sup>2</sup> were the optimal condition in terms of gas flow rate over electrode loss ratio (Qg/Loss). It was then studied the removal kinetic of the design parameters for ECR such as hydraulic retention time, optimal current density, and settling overflow rate. Removal kinetic was studied with initial concentration 250 NTU and 6,000 ADMI of turbidity and reactive dye, respectively. The optimal current density 4.5 mA/cm<sup>2</sup> was obtained with 25 to 30 minutes of EC electrolyze time in order to achieve the treatment efficiency 95% of both bentonite and dye. Moreover, the comparative analysis between electrocoagulation (EC) and chemical coagulation (CC) was examined. Both EC and CC are possibly used for removal turbidity and color, however, the economization will be subsequently studied. Estimation of the treatment cost through both processes was examined. It can be concluded that if there was only turbidity in wastewater with low concentration, about 50 NTU, conventional system is preferred. However, it is usually co-existed with color and high concentration. Therefore, EC is more suitable for treating turbidity and color from textile wastewater in terms of economic aspect for high concentration. ECR was finally designed in rectangular shape, which was combined between EC and separation process in continue operation. The internal flow pattern in ECR was studied by residence time distribution (RTD) method and the result showed that ECR tended to be plug flow reactor with homogenous baffles of 3 baffles (90% - 90%). ECR could remove turbidity and dye individually up to 95% with flow rate 3 LPM, and 97% with flow rate 1 LPM, respectively. For simultaneous pollutants, the results showed that removal efficiency can be achieved up to 94% (turbidity) and 96 % (color) with flow rate 2 LPM. Furthermore, the improvement by adding tube settler was studied and expressed that the 2 modules at the higher position could improve the treatment efficiency of turbidity about 10 - 15 % at 50 minutes. The design criteria of ECR for removal of turbidity and dye from textile wastewater was finally proposed.

Department: Environmental Engineering Field of Study: Environmental Engineering Academic Year: 2017

Student's Signature	-
Advisor's Signature	
Co-Advisor's Signature	

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# CHAPTER 1 INTRODUCTION

#### **1.1 General Context**

Water is considered as a renewable resource which cannot runs out, but the global still faced some problems when the demand of water quantity was increasing from year to year which consume by many purposes such as drinking, sanitation, irrigation, and industrial use. The rising demand of water quantity still continuous, so that there is a massive concern in recent years about water treatment and reuse requiring (Figure 1.1) From year to year, the amount of wastewater was increasing which generated by many factors and many different sources in the world that make citizen who live in some area suffered by wastewater.

Wastewater is mainly originated from the water usage of industrial, agriculture, residences, and commercial together with groundwater, surface water, and storm water. In fact, wastewater typically averages 99.94% water by weight and only a small amount 0.06% is actually waste material. Mostly, urban wastewater including domestic and industrial wastewater discharged to municipal sewage system contains color, odor, turbidity, temperature, biochemical oxygen demand (BOD), chemical oxygen demand (COD), total organic carbon (TOC), total oxygen demand (TOD), pathogens, organic pollutants, suspended solids (SS), nitrogen compounds, phosphorous compounds, heavy metals, oil and grease, etc. (ESCWA, 2010).

The industry is one sector which is considered as the main key for the development in the economic growth of the country. At the same time, it is also provided many pollutions to the environment. Due to different types of industry, environmental pollution is one of the major problems facing in the world (Paul et al., 2012). Rapid urbanization and industrialization lead to several problems in a collection, treatment, and disposal of effluents. Lack of treatment of domestic and industrial wastewater cause the serious health and environmental hazard in many cities, particularly in developing countries where 80-90% of urban wastewater is untreated or insufficiently treated before discharging. Even in developed countries wastewater is not universally treated.

A huge volume of untreated wastewater is directly dumped into water sources, threatening human health, ecosystems, biodiversity, food security, and the sustainability of water resources (Phani Madhavi et al., 2014). For instance, China is one of the most powerful countries have strong economies in the world, but it still faced the problem of water pollution. About one-third of the industrial wastewater and more than 90% of household sewage in China are released into rivers and lakes directly without proper treatment. It may cause 90% of underground water supply in 278 cities contaminated with high color, turbidity, COD, and other heavy mental substances in 2008 (M. Wang et al., 2008).

Most of the industrial effluents wastewater contains high color and turbidity as the contaminants. The discharge of untreated wastewaters from industries such as textile and dyeing operation, pulp and paper production, food processing, mining and coal processing operation, refinery, and slaughterhouse operation may add substantial coloration to water in the receiving the stream. The discharge of colored effluents into water bodies or onto land is an indicator of pollution. Recent reports suggested that several color causing substances are micro toxic to aquatic biota (Kadirvelu et al., 2003). The discharge of strongly colored wastewater from dyeing operations not only has unfavorable aesthetic effects, but such discharges can be carcinogenic, mutagenic and generally detrimental to our environment (Papić et al., 2004). Thus, a highly colored body of water could not sustain aquatic life, which could lead to the long-term impairment of the ecosystem.



*Figure 1.1.* Ratio of treated to untreated wastewater from 10 regions in the world (*Gadipelly et al., 2014*)

One million tons of more than ten thousand types of synthetic dyes and pigments are produced annually worldwide and are used highly in many various industrial fields, such as textile and leather industries, paper production, food processing, agricultural research, light-harvesting arrays, photo electrochemical cells, and hair coloring (Roa-Morales et al., 2014). The concentration of dye is different depending on the effluence from each industrial, mostly the high concentration of color is from textile, printing industrial because it has many processes(Chollom et al., 2015; Patel and Vashi, 2010). In fact, the concentration of true or synthetic dye from industrial mostly is very high ranging from 10 - 7,000 mg/l and some industrials are higher than this value (O'Neill et al., 1999). In a small industry in Asian country, the concentration of dye from the industry mostly range from 1 - 250 mg/l which is excessed the limitation of dye detection about 0.005 mg/l. In recent year the unit that considers as standard for the industry is American Dye Manufacturers' Institute (ADMI) unit.

Turbidity is one of the parameters that cause some problems to our environment and ecology system. High turbidity can minimize the aesthetic quality of lakes or streams and have a harmful impact on recreation and tourism. It can expand the cost of water treatment for drinking and food processing. It can injure fish and other aquatic life by decreasing food supplies, degrading spawning beds, and affecting gill function (MPCA, 2008). Normally, the concentration of turbidity also different and don't have a specific standard to show the value which allowed to discharge. Base on (Cerqueira et al., 2009; Lau et al., 2013; Mahmoud, 2009) reports that the concentration of turbidity range from 8 - 86 NTU which is over standard if we compare with Bureau Indian Standard (BIS) which allowed only 10 NTU (Elango et al., 2016).

#### **1.2 Problem Statement**

Many dye contaminants are toxic to fish and mammalian life, which obstruct the growth of microorganisms and affect flora and fauna. Besides that, aquatic life (aquatic plants, microorganisms, fish, and mammals) is demonstrated as toxic from several dyes and their decomposition derivatives. They are also carcinogenic in nature that can promote intestinal cancer and cerebral abnormalities in the fetus (Malik et al., 2014).

Effluent from industry wastewater contains a very high colorant and turbidity which is much higher than the limitation of standard. Therefore, the conventional methods are not the effective process to remove these substances in terms of time and economic. Many methods were applied to treat the high amount of color and turbidity from industrial in order to protect the environment and respect the standard.

The conventional process such as coagulation, flocculation, and sedimentation can use for treating the color and turbidity. However, the treatment efficiency is very low comparing to another modern technology processes. Zaleschi et al. (2012) proved that the electrocoagulation process (EC) can treat wastewater more efficient than conventional coagulant about 10 - 15 %. Furthermore, EC process has many advantages providing a simple, reliable and cost-effective method for the treatment of wastewater without any requirement for additional chemicals, and secondary pollution. It can reduce the amount of sludge, which may require to dispose in coagulation/flocculation in a conventional process. The EC technique uses a direct current source between metal or aluminum electrodes immersed in polluted water. The process creates a floc of metallic hydroxides or aluminum hydrates within the effluent to be cleaned, by electro-dissolution of soluble anodes. In recent years, EC has been successfully applied to decolonization of dye-containing solutions. It can treat the color from dye wastewater by coagulated of iron (II) and aluminum hydrates or hydroxides which produced from the sacrificial anode.

Likewise, the conventional system needs many processes to treat and more space for installation comparing to the electrocoagulation process. Additionally, many existed study was explored in a batch system but the continuous system is still limitation. So that, the development of combination between electrocoagulation and separation process together in a continue system need to study for responding in the industrial sector like on-site equipment in terms of treatment efficiency, compress system, and economic.

#### **1.3** Research Gap

Although many researchers and institution were figured out the phenomenon inside the electrocoagulation flotation process (ECF) and try to combine electrocoagulation with

another process including sedimentation, membrane, advanced oxidation, etc. It is still required the reactor development for better performance.

The combination of ECF process with sedimentation is still in limited literature. Even there are some existed researches, the improvement of its performance is still the main challenging. For instance, the study of wastewater effluent from textile industry by Ubale and Salkar (2017) using many electrodes of aluminum connected in series with monopolar as the baffle. It can remove the turbidity around 95 - 98 % and the color about 85 - 88 %. However, this process performed with many surfaces of the electrode plate and produce much sludge. It may difficult to conduct with real application in industry because it requires to check for all plates before operating and cleaning after operate. If they were not clean, the plates may consume high voltage and current density because the sludge covered the surfaces of plate as well as block the plate for generating the coagulant and bubble. Moreover, in terms of economic, many aluminum plates of electrocoagulation sets may require higher power consuming and replace the new plate frequently.

Another research used 4 plates of aluminum connect as the parallel of monopolar with no baffle. It is just divided into 2 blocks and it can remove turbidity from 85 - 88 % (Jafari Mansoorian et al., 2016). However, the last process is easier to operate than the first process because this process is not complicated to operate without the baffle. So it doesn't need to check and clean the baffle electrode plates before and after the operation, but the treatment efficiency in this research is less than the first research about 10% in terms of the retention time of water or wastewater that stay in the reactor to react with electrode plate shorter than the last process. Unlike, in terms of the economic, this process much save the money than the first process because the first process needs more baffles for replacement and electricity consumption. In short, the adjustment of the amount of baffles and type of baffles will be considered to study in terms of treatment performance of color and turbidity removal.

Even there are many research works concentrated on the development of combination between ECF and sedimentation process, different pollutants were interested. The gap of this study is the development of the newly combined reactor between ECF and Sedimentation process for effective removal efficiency of color and turbidity in industrial wastewater in terms of operation easiness, energy consumption, and removal efficiency.

#### 1.4 Hypothesis

The hypothesis of this work are:

- 1. The optimum condition and configuration of EC process could treat successfully of turbidity and dye in term of economization.
- 2. The newly developed electrocoagulation reactor which calls ECR could be effective to treat turbidity and dye from synthetic textile industry in a continuous system.

#### 1.5 Research Objectives

The main objective of this research is to develop and evaluated the combination process between electrocoagulation (EC) with settling process for dye and turbidity removal from the textile industry wastewater. Two main objectives are listed below:

- 1. To optimize the reactor configuration and operation condition of the combined reactor between electrocoagulation with sedimentation process.
- 2. To evaluate the treatment performance of the newly developed reactor on color and turbidity removal from synthetic wastewater of textile industry in continuous condition.

#### 1.6 Scopes of Work

In this research work, several scopes were studies as descript in the following points:

- Prepare synthetic wastewater as textile industry contaminated turbidity and color by using respective powder bentonite and powder reactive yellow dye with tap water.
- Examine the jar test for representing the chemical coagulation to identify the amount of alum dosage for turbidity and dye removal.

- Optimize the electrodes arrangement (i.e., monopolar and bipolar), electrode configuration (electrode gap), and current density in EC with tap water in terms of gas flowrate and electrode loss ratio (Qg/Loss) in the simple column reactor. Analysis the treatment kinetics for turbidity and color removal in terms of current density. The hydraulic retention time (HRT) of EC process and over flow rate (OFR) of settling process as the basic information for designing the combined reactor in continuous flow mode.
- Design the new continued electrocoagulation reactor (ECR) involving between EC and settling process from the previous results. It was then constructed for future evaluation experiment.
- Analysis the liquid flow pattern in the new reactor to obtain the optimal condition by using residence time distribution (RTD) in terms of probe position, baffle in the reactor, mixing with and without EC, and liquid flowrate.
- Evaluate the treatment performance in new reactor on dye removal with the varying liquid flowrate and current density. The optimum condition from dye treatment was then validated for turbidity treatment and both contaminants.
- Study the separation performance in the sedimentation part by installing the tube settler in terms of mount of tub sets and its position. The guideline of design criteria and operating condition of EC in batch and continuous conditions for treating dye and turbidity from textile industry was therefore proposed.

# CHAPTER 2 THEORY AND LITERATURE REVIEW

#### 2.1 Industrial Wastewater Characteristic

There are many industries which are generated turbidity, color, chemical oxygen demand (COD), total dissolved and suspended solids (TDS and TSS), and toxic heavy metal of their final effluent to the public sewage system (Kabdaşlı et al., 2012). The Figure 2.1 and Table 2.1 below will show the detail about the type of industrial around the world which are generated many pollutants to our environment. Wastewaters originating from the textile, cellulose, paper, chemical, tanning, food and cosmetic industries containing a high concentration of dyes are a hazardous source of natural environment contamination, which is troublesome in purification process due to a complex structure of dye molecules. Even small amounts of dye are undesirable as they color water making it look unaesthetic and disturb life processes in water. Most dyes do not undergo biodegradation, deteriorate light penetration into the water and inhibit photosynthesis, increase chemical and biological demand for oxygen (Wawrzkiewicz and Hubicki, 2015). For instance, the major sources of wastewater are generated by the textile wet processing industry from the washing of natural fibers, bleaching, dyeing, and finishing process of origin. Due to a vast variety of fibers, dyes, processing aids, and finishing products in use, these processes have a strong chemical complexity and diversity without fully processing conventional sewage treatment plant effluent.



Figure 2.1. Main type industry around the world (Kabdaşlı et al., 2012)

N°	Industry	Pollutants characteristics	Suggested treatment methods
1	Paper and pulp	<ul> <li>Strong color</li> <li>High BOD</li> <li>High COD/BOD ratio</li> <li>Highly alkaline</li> <li>High sodium content</li> </ul>	<ul> <li>Chemical recovery</li> <li>Lime treatment for color</li> <li>Biological treatment</li> </ul>
2	Tannery	<ul> <li>Strong color</li> <li>High salt content</li> <li>High BOD</li> <li>High dissolved solid</li> </ul>	<ul><li>Chemical treatment</li><li>Biological treatment</li></ul>
3	Textile (cotton)	<ul> <li>Highly alkaline</li> <li>High BOD, COD</li> <li>High suspended solid</li> <li>Strong color</li> </ul>	<ul><li>Chemical treatment</li><li>Biological treatment</li></ul>
4	Distillery and brewery	<ul><li>Strong color</li><li>High chloride, sulphate</li><li>Very high BOD</li></ul>	- Biological treatment
5	Pharmaceuticals	<ul> <li>High total solid</li> <li>High COD</li> <li>High COD/BOD ratio</li> <li>High acidic or alkaline</li> </ul>	<ul><li>Chemical treatment</li><li>Biological treatment</li></ul>
6	Oil refineries	<ul><li>Free and emulsified oil</li><li>High Turbidity</li></ul>	<ul><li>Oil separation</li><li>Chemical treatment</li><li>Biological treatment</li></ul>
7	Fertilizers	- High nitrogen content	- Biological treatment
8	Sugar	<ul><li>High BOD, volatile solid</li><li>Low pH</li></ul>	- Biological treatment
9	Dairy	<ul> <li>High dissolve solid</li> <li>High suspended solid</li> <li>High BOD</li> <li>Presence of oil and grease</li> </ul>	- Biological treatment

Table 2.1. Characteristic of industrial wastewater (Saumya, 2017)

#### 2.1.1 Turbidity

Turbidity is an optical determination of water clarity. Turbid water will appear cloudy, murky, or color which affects the physical look of the water. Suspended solids and dissolved color material reduce water clarity by creating an opaque, hazy or muddy appearance. Turbidity measurements are often used as an indicator of water quality based on clarity and estimated total suspended solids in water (Smith and Davies-Colley, 2002). The turbidity of water depended on the amount of light scattered by particles in the water column. The more particles that are present, the more light that will be scattered. As such, turbidity and total suspended solids are related (Gray et al., 2000).

Generally, the concentration of turbidity from industrial is various depended on type of industry. Mostly the average wastewater turbidity ranged from 16 to 660 NTU. Turbidity discharged from industrial are generated by dyes and pigments, binder, solvents and others inorganic substances and also caused by a wide variety of suspended materials as colloidal, sub colloidal (Mahmoud, 2009).

#### 2.1.2 Color

Synthetic dyes are hugely used in textile dyeing, paper, printing, color photography, pharmaceutics, cosmetics and other industries which know that it generated as a color of their final effluent to the waterway and water sources. According to (Marmion, 1991; Wawrzkiewicz and Hubicki, 2015) shown that in the Asia continent consumed an amount of dye than another continent in the world. Figure 2.2 shows the amount of dye usage from all continents in the world. Azo dyes are the vastest used dyes and represent over 60% of the total dyes (Fu and Viraraghavan, 2001). They are complex aromatic compounds with significant structural diversity. Their properties are enhanced to provide a high degree of chemical, biological and photocatalytic stability and resist breakdown due to time, exposure to sunlight, microorganisms, water, and soap; or resistant to degradation(Keharia et al., 2004). Approximately 10–15% of the dyes are released into the environment during manufacturing and usage (He et al., 2004; Spadaro et al., 1994). On the other hand, some of the dyes are harmful and when dye-containing wastes, it will cause a problem to an environmental (P. Verma and Madamwar, 2002).



Figure 2.2. World dye consumption by the regions in 2008 (Marmion, 1991; Wawrzkiewicz and Hubicki, 2015)

There are many kinds of synthetic dye which used in the industry or commercial such as direct dye, disperse dye, basic dye, the azoic dye, reactive dye, vat dye, and sulfur dye. These dyes were indicated their commercial name by the standard of a color index (C.I.) with their wavelength. The Figure 2.3 will show the percentage of different types of dye was discharged from industrial wastewater. Moreover, the types of dyes were classified based on their solubility and chemical properties which show in Table 2.2. The units of color have several depend on the methods that the researcher prefer to use, in those the unit which are usually see such as Abs, Pt/Co, mg/L, and ADMI. Nowadays most country popular use unit of ADMI. For the effluent of dye form industry were different base on type of industry, but normally their effluent in range 10mg/L to700 mg/L, and some represent in ADMI unit is about 1500 to 15000 ADM (O'Neill et al., 1999).



Figure 2.3. Percentage of non-fixed dye and auxiliaries which are discharged from the industrial wastewater effluent (Marmion, 1991; Wawrzkiewicz and Hubicki, 2015)

Class of dyes	Chemical character of dyes	Exemplary dyes	Kind of colored materials
Acid	Organic salts of sulfonic and carboxylic acids soluble in water	Methyl orange, Methyl red, Orange I, Orange II, and Congo red.	Protein fibers (mainly wool), polyamide fibers, paper, leather, food, cosmetics
Basic	Salts of organic bases soluble in water	Aniline yellow, Butter yellow, Methylene blue, and Malachite green	Wool, natural silk, polyester and polyacrylonitrile fibers
Direct	Salts of organic sulfonic acids soluble in water	Martius, yellow and Congo red	Cellulose fibers (cotton, linen), leather
Reactive	Salts of organic bases and acids soluble in water	Procion dye (2, 4, 6- tricholor and 1, 3, 5- triazine)	Cellulose and protein fibers
Mordant	Soluble in water, form complex with metals	Alizarin	Cellulose and protein fibers
Vat	Insoluble in water	Indigo, Tyrian purple, Benzanthrone	Cellulose fibers
Sulfur	Insoluble in water	N/A	Cellulose fibers, coloring agent in food
Dispersive	Insoluble or sparsely soluble in water	Celliton fast pink B, Celliton fast blue B	Synthetic mainly poliester, polyamide, polyacrylnitrile, acetic

Table 2.2. Technical classification of dye of industrial (Correia et al., 1994)

### 2.1.3 Summary จุฬาลงกรณิมหาวิทยาลัย

To sum up, the effluent from industries are different depends on the type of industry and their technology. Therefore, their effluent was a little bit far range from each other such as color is mostly in a range 10 mg/L to 700 mg/L which is in range of 1500 to 15000 ADMI and the turbidity is in a range 16 NTU to 3000 NTU. The common concentration of dye and turbidity which is normally used in the range 10 mg/L to 300 mg/L and 16 NTU to 660 NTU, respectively. For this research, the color and turbidity were selected to study with 50 mg/L, 100 mg/L, and 250 mg/L, 50 NTU, 250 NTU, and 500 NTU, respectively. The standard of discharge in Thailand of dye was 300 ADMI which is follow by Notification of Ministry of Industry regarding Industrial Effluent Standards B.E. 2560 (2017) and turbidity measure as total suspended solids (TSS)

which are not more than 50 mg/L due to parameter of turbidity don't have in Thai standard.

#### 2.2 Overall Treatment Techniques for Industrial Wastewater

There are three main types of treatment process such as a biological, physical, and chemical method which are commonly used in the world since the last until now. In those three main parts, there are many kinds of the conventional and advanced treatment process that can treat turbidity and colors from industrial wastewater such as coagulation/flocculation, electrocoagulation/ floatation, ion exchange, membrane, adsorption, and others. Table 2.3 summary the methods that commonly apply in the industries nowadays with their advantages and limitations.

#### 2.2.1 Biological methods for turbidity and color

The principal applications of the biological treatment methods are carbonaceous organic material representing in biochemical oxygen demand (BOD) removal, nitrification, denitrification, stabilization, and phosphorous removal. The biological process is well known classify by aerobic and anaerobic (Chan et al., 2009). Optimum condition for decolorization by microbial was expressed that anaerobic performance better than aerobic process. Anaerobic biological treatment methods use bacteria to decolorize azo dye solutions through cleavage of the azo bond, yielding aromatic amines as products. Aerobic bacteria have been described to oxidative decolorize many dyes from several classes, among which azo dyes always turned out to be the most recalcitrant compounds. Dye-degrading fungi find applications in bioreactors for the decolorization and degradation of azo dyes (Maier et al., 2004). Moreover, primarily involved in the bioremediation of textile effluents azo-reductase and laccase are cellular enzyme and bioreactor

#### a. Enzymatic decomposition

The character of enzymes and enzyme systems in microorganisms are appropriate for the decomposition of dyes have been extensively investigated. An effort has been dedicated to the separation, isolation, and testing of these enzymes. Exact knowledge of the enzymatic processes governing the decomposition of dyes is major in the environmental protection for both from theoretical and practical points of view.

The usage of enzyme preparations shows considerable benefits over the direct use of microorganisms but it still consumes a long time. Commercial enzyme preparations can be easily standardized and facilitate accurate dosage. The application is simple to remove dye and can be rapidly modified according to the character of the dye (Forgacs et al., 2004).

#### b. Bioreactor

Bioreactor processes could give better performance compared to conventional activated sludge processes. Anaerobic and aerobic conditions were explored by many researchers to determine the optimum conditions for dye removal. Several bioreactors or equipment are being investigated including sequencing batch bio-filter granular reactor (SBBGR), sequencing batch reactor (SBR), sequencing batch bio-film reactor, moving bed sequencing batch bio-film (MB-SBBR), and membrane aerated biofilm reactor (MABR) (Ahmad et al., 2015). For example, MABR has the ability to remove high Acid Orange 7 decoloration (98%) and COD removal in 6 hours at optimized conditions (J. Wang et al., 2012). A similarity of this, MBR also removes COD about 99% of high-strength wastewater with COD range from 6000 – 14,500 mg/L at HRT for 24 hours (Ahn et al., 2007).

# 2.2.2 Chemical methods for turbidity and color

The chemical treatments can be applied for removing substance through five points such as producing insoluble solids and gas, producing coagulation of a colloidal suspension, producing biological degradable substances from non-biodegradable, destroying or deactivating chelating agents, and producing non-objectionable substances that can be removed more easily (Elsheikh and Al-Hemaidi, 2012). Coagulants permit the colloidal particles to participate together by slow stirring. Some highly objectionable substances can be chemically oxidized to generate nonobjectionable substances, such as  $CO_2$  and water. The chemical methods detail below:

#### a. Chemical Precipitation

Chemical precipitation is a simple technique which uses chemical substances such as sulfides, hydroxides, and carbonates react with organic and inorganic pollutants present in wastewater to form insoluble precipitates. In general, additional chemicals can react with dissolved dye molecules to form insoluble precipitates and then dye can be removed by settle. The wastewater can be decanted to separate the sludge after settle and the most common chemical precipitation method for dye removal is hydroxide precipitation. G. Zhang et al. (2011) used the leaching solutions of white mud to remove Acid Orange II, Reactive Light Yellow K-6G, Reactive Bright Red K-2BP, and Direct Yellow was achieved removal efficiency more than 90% within 90 seconds which showed the fast kinetics of this system.

#### b. Coagulation-Flocculation

Coagulation and flocculation are traditional methods which are an effective part of drinking water treatment as well as wastewater treatment plant. The principle is destabilized the dispersed solid particles in water by reducing their surface charge and gather them to form larger particles. Coagulation and flocculation can be divided into three steps such as flash mixing, coagulation, and flocculation. Sedimentation is usually occurred afterward to remove the larger particles which settle down. Sometimes coagulant aids are also added to increase the treatment process. Lee et al. (2012) explored the significance of five parameters on Cibracon Red FN-R removal using an inorganic-organic composite polymer through flocculation system. The dosage of a composite polymer was the main factor for dye removal, followed by pH, dye concentration, agitation speed, and the least significant factor was agitation time.

#### c. Electrochemistry / Electrostatic process

The main difference between electrocoagulation and chemical coagulation is that coagulation used coagulant direct in the system such as alum, ferric chloride, etc., but electrocoagulation (EC) process require direct current in the system to dissolve mental ion from electrode plate at anode side as coagulants and generated gas bubble at cathodes side. More EC process doesn't have secondary pollution occurs (Kuokkanen et al., 2013). Nowadays, in the industrial sector was investigated on chemical coagulant

to be advancement than before and provided more efficiency. Many industries and researcher were studied on electrocoagulation/ electroflotation which is process that could quickly coagulate and float to remove the colloidal, turbidity and suspended particles then electro-oxidation oxidizes the remaining organics. The coupled process eliminates COD, BOD, color, turbidity, and coliforms in a practical HRT for 2 hours with more effective than conventional chemical coagulant (Linares-Hernández et al., 2010).

#### d. Oxidation processes

Oxidation processes are considered to challenge as methods in industrial wastewater treatment technology for removal of organic pollutants by using oxidizing agents that are not treated by a conventional process. There is much power oxidized such as hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>), UV, Fenton's reagent, ozone (O<sub>3</sub>) that use to convert organics to CO<sub>2</sub>, water, and completely oxidized states of other atoms that were part of the original organic pollutants, including sulfates and nitrates. (Oller et al., 2011). In Oxidation process, pH and catalysts play an important role. For instance, using of UV/H<sub>2</sub>O<sub>2</sub>/O<sub>3</sub> can remove 99% of COD with initial concentration about 930 mg/l and 95% of color that spent 90 minutes of the whole process to treat wastewater contained disperse dyestuff and pigments (Neamtu et al., 2002).

#### 2.2.3 Physical methods for turbidity and color

Physical treatment was depended on naturally occurring forces such as gravity, electrical attraction, and van der Waal forces, and physical barriers. The methods of physical treatment used in industry were sedimentation, flotation, adsorption, and barriers (barracks, screens, deep bed filters, membranes, electro-dialysis and ion exchange). Some process of physical treatment (sedimentation, flotation, and deep filtration) are not enough ability to treat wastes from the industrial sector, so it needs to combine with another process in order to help removal efficiency and time operation (Elsheikh and Al-Hemaidi, 2012).

#### a. Adsorption process

Adsorption is a surface phenomenon in which adsorbate molecules or ions (liquid or gas) is concentrated on the surface of a solid (adsorbent). The process can be classified as physisorption or chemisorption based on how the adsorbate species get adsorbed onto the adsorbent surface (Gupta, 2009). Several forces such as hydrogen bonding, electrostatic interaction, van der Waals forces, hydrophobic interaction applied to dyes molecules adsorbed on the surface of adsorbent in adsorption process (Kumar et al., 2014). The adsorption process has ability to removal of dyes from water and wastewater by using activated carbon, Nano particulate adsorbents, low cost adsorbents and other types of adsorbents. The adsorption process has an ability to remove dyes from water and wastewater by using activated carbon, Nano particulate adsorbents, low cost adsorbents, low-cost adsorbents and other types of adsorbents.

Adsorption on activated carbon is one among of adsorption process which is popular use for removal many compounds from wastes including pesticides (2, 4-D herbicides and carbamate insecticides), Dithiothreitol (DTT), chlorobenzene, and p-chlorobenzene sulfonic acid. This process is low cost because now some absorbents from agriculture by-products are renewable and inexpensive such as nut shells, wood, bone, peat, coconut shells (Amuda et al., 2007). According to M. Zhang et al. (2010) was studied about activated coke to treat wastewater from cafeteria which indicated that COD was removed about 91.6% and color until 90% that spent 6 hours. Moreover, other research also use adsorption process to treat many reactive such as (blue 2, red 2, yellow 2, and yellow 86) by mix adsorbents of ionically and chemically cross-linked chitosan beads (Chiou et al., 2004).

#### b. Ion exchange process

Ion exchange process is techniques, which can effectively remove dyes from aqueous solutions through strong interactions between charged dyes and functional groups on ion exchange resins. This process involves an exchange of ions to form strong bonds between solutes and resins, as the results achieving effective separation (Nemerow, 2010). The resins are available as anion exchangers or cation exchangers for separating solutes with different surface charges. A main disadvantage is treatment cost. Organic

solvents are very high cost, and the ion exchange method is not very effective for disperse dyes and reactive dye (Mishra and Tripathy, 1993).

#### c. Membrane Filtration

Membrane filtration is an advanced treatment technology for the removal of turbidity, color, COD and salinity from wastewater (Zheng et al., 2013). The procedure of membrane is wastewater through passing small pores of the filter. Any solutes which are larger than the pore size will be trapped and the solution after passing through the membranes is free from those solutes. The needed of removal trapped solutes form a layer of cake filter is also considered to ensure the smooth running of the filtration process. Membrane filtration can be classified based on the size of the pores on the membranes such as Ultrafiltration (UF), Nano filtration (NF), and Reverse Osmosis (RO). The performance of membranes is usually evaluated through rejection and permeates flux.

Process	Advantages	Disadvantages			
Biological process					
Aerobic and Anaerobic	<ul> <li>Cost-competitive option.</li> <li>Direct, disperse and basic dyes have high level of adsorption on to bioreactor and enzyme.</li> </ul>	<ul> <li>Dyes are toxic and very resistant to biodegradation.</li> <li>Acid and reactive dyes are highly water soluble and poor adsorption on to sludge</li> <li>Slow process</li> </ul>			
Chemical process					
Electrochemical	<ul> <li>Effective decolorization of soluble/insoluble dyes; reduction of COD possible.</li> <li>Not affected by presence of salt in wastewater</li> <li>Produces sludge with no hazard</li> </ul>	<ul> <li>Need to replace plate of electrode and some mental support at anode</li> <li>Require high energy and operating cost</li> </ul>			
Chemical oxidation	- Initiates and accelerates azo-bond cleavage	- Thermodynamic and kinetic limitations along with secondary pollution are associated with different oxidants.			

Table 2.3. Advantages and limitations methods of for dyes removal (Shah, 2014)
		<ul> <li>Not applicable for disperse dyes.</li> <li>Negligible mineralization possible, release of aromatic amines and additional contamination with chlorine (in case of NaOCl) is suspected</li> </ul>
Photo catalysis	- No sludge production, considerable reduction of COD, potential of solar light utilization	<ul> <li>Light penetration limitation, fouling of catalysts, and problem of fine catalyst separation from treat water</li> <li>Generation of hazardous by- product</li> </ul>
Fenton's reagent	<ul> <li>Effective decolorization of both soluble and insoluble dyes; applicable even with high suspended solid concentration.</li> </ul>	<ul> <li>Effective within narrow pH range of &lt;3.5; and involves sludge generation.</li> <li>Comparatively longer reaction time required</li> </ul>
Physico-chemical	process	
Ion exchange	<ul> <li>Adsorbent can be regenerated without loss, dye recovery conceptually possible</li> </ul>	- Ion exchange resins are dye- specific; regeneration is expensive; large-scale dye recovery cost-prohibitive
Coagulation	<ul> <li>Economically feasible; satisfactory removal of disperse, Sulphur and vat dyes</li> </ul>	<ul> <li>Removal depended on pH and produces large quantity of sludge.</li> <li>May not remove highly soluble dyes; unsatisfactory result with azo, reactive, acid and basic dyes</li> </ul>
Activated carbon adsorption	- Good removal of wide variety of dyes, namely, azo, reactive and acid dyes; especially suitable for basic dye	<ul> <li>Removal is dependent pH, unsatisfactory result for disperse, sulfur and vat dyes.</li> <li>Regeneration is expensive and involves adsorbent loss; necessitates costly disposal</li> </ul>
Membrane	- Capable of treating all dyes and high quality of treat effluent	<ul> <li>High pressure</li> <li>Very expensive</li> <li>Incapable of treating large volumes of wastewater</li> <li>Easily collaging</li> </ul>

# 2.2.4 Summary

To sum up there are many methods which are available for the industrial sector. Each method has advantages and disadvantages through their technology. The Conventional

Coagulation (CC) and Electrocoagulation (EC) are more commonly used due to their by-product is non-toxic, economic for a price of product and time operation, and especially is easy to operate. Hence, the coagulation and electrocoagulation were selected to study in this research to enhance the knowledge.

#### 2.3 Coagulation/ Flocculation

#### 2.3.1 Coagulation

The coagulation is a process which adds a chemical to water with the objective of destabilizing particles by reducing the forces acting to keep the particles apart after contacting with others (example: lower repulsion forces), so they aggregate or forming a precipitate that particles to be sweep particles from a solution or adsorb dissolved constituents. The significance of coagulation is precipitation of insoluble hydroxide forms (Al (OH) <sub>3</sub> or Fe (OH) <sub>3</sub>). There are four main methods such as compression of an Electric double layer, Adsorption and Charge Neutralization, Enmeshment in a precipitate (Sweep floc), and polymer bridging (A. K. Verma et al., 2012).

# a. Electro double layer compression (EDL)

This can be accomplished by addition of an indifferent electrolyte to increases the ionic strength of solution which has the effect of compressing the EDL. As the counter-ions are pushed closer to the surface the repulsion forces become easier to negate by van der Waals forces.

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#### b. Adsorption and Charge Neutralization

The neutralization will occur when the adding chemical coagulants by charge positive which met the charge of colloids are negative. This will reduce the net potential at any particular radical thus making the attraction forces more effective. Zeta Potential is likely to be reduced also. So that, the charge neutralization encourages floc to settle down via gravitation.

#### c. Enmeshment in a precipitate (Sweep floc)

When metal salts (Al  $_2(SO_4)$  3, FeCl<sub>3</sub>, MgCO<sub>3</sub>, and CaO) are added in sufficient quantities to exceed the solubility products of the metal hydroxide, oxide or, sometimes

carbonates a "sweep floc" will form. Colloids then become enmeshed in the settling sweep floc and be removed from the suspension. The precipitates of metal salts above are Al (OH) <sub>3</sub>, Fe (OH) <sub>3</sub>, Mg (OH) <sub>2</sub>, and CaCO<sub>3</sub>.

The primary metal salts used as coagulants are Al or Fe sulfate. Many equations of these mental salts will express below:

Primary Coagulants

$$Al_2(SO_4)_3 \rightarrow Al^{3+} + 3SO_4^{2-}$$
Eq. 2.1

$$\operatorname{FeCl}_3 \rightarrow \operatorname{Fe}^{3+} + 3\operatorname{Cl}$$
 Eq. 2.2

Aqueous Chemistry of Fe and Al

Al<sup>3+</sup>, Fe<sup>3+</sup> complex with OH<sup>-</sup> (ligand) 
$$\rightarrow$$
 Hydroxo-metal complex Eq. 2.3  
Al<sup>3+</sup> + OH<sup>-</sup>  $\rightarrow$  Al(OH)<sup>2+</sup>, Al<sub>8</sub> (OH)<sup>4+</sup> Al<sub>9</sub>, Al(OH)<sup>4-</sup>, .... Eq. 2.4  
d. Polymer bridging

In this case polymers, metal salt or synthetic organic types, specifically adsorbed to a surface, often charge neutralization occurs. Further, other parts of the polymer adsorbed to other colloids. The most popular of coagulants which used nowadays is Al<sub>2</sub> (SO<sub>4</sub>)<sub>3</sub>. 14H<sub>2</sub>O, Al<sub>2</sub> (SO<sub>4</sub>)<sub>3</sub>. 18H<sub>2</sub>O (Alum), FeCl<sub>3</sub>, FeCl<sub>3</sub> with lime, and FeSO<sub>4</sub>. 7H<sub>2</sub>O (copperas) (with lime).

Polymers are classified as three types such as group one is cationic polymers: polydialydimethyl ammonium (PDADMA, cat-floc). Group two is anionic polymers: polyacrylamide acid (PAA), hydrolyzed polyacrylamide (HPAM) and the last group is the polystyrene sulfate (PSS), and Non-ionic polymers: cellulose, gelatin and starch (natural non-ionic polymer).

#### 2.3.2 Flocculation

Flocculation is a process which is an aggregation of destabilized particles into larger masses that are easier to remove from water than the original particles. Flocculation occurred after coagulation where there is a slow machoism to enhance contact between destabilized coagulated particles and later settle down by the gravity. The frequency of collision is reverse proportion to shear rate. The floc can form large floc when low shear rate. On the other hand, the floc will break up when the intensity of shear rate is too high.

#### 2.3.3 Jar test

The jar test simulates the coagulation/flocculation process in a batch mode. A series of batch tests are run in which pH, coagulant type, coagulant dosage, and coagulant aid are varied to get the optimal dosage (lowest residual turbidity). An economic analysis is performed to select these parameters.

Jar tests generally are performed using 6 glasses of one-liter samples of the water or wastewater to be treated. Immediately after the coagulant is added the samples are "flash mixed" for approximately one minute with speed of rapid mixing in range (100-300 rpm), while flocculation takes much longer detention time, approximately 15-30 minute, for slow mixing (30-60 rpm). At the end of the flocculation period, the stirrers are turned off and the floc is allowed to settle for one-half hour (Golob et al., 2005). After this settling period, supernatant samples are drawn off from each sample and analyzed for turbidity and sometimes alkalinity and pH.

#### 2.4 Electrocoagulation/Flotation (ECF) Process

#### 2.4.1 Principle of ECF process

Electrocoagulation/ flotation is green technology for environmental since their sludge by-product is nonhazardous. The process is stabilized suspended emulsified or dissolved contaminants in an aqueous medium by introducing an electric current into a medium (Emamjomeh and Sivakumar, 2009). ECF process can treat wastewater with soluble or colloid pollutants such as color, dye from industrial wastewater by two electrodes sides which resent as an anode side and cathode side is known as sacrificial electrodes. Mostly typical of electrodes plate of electrocoagulation is aluminum (Al), Iron (Fe), and stainless steel was selected depending on the purpose of wastewater removal, which provided more efficiency with low material cost. The direct current (DC) supply is applied to electrodes in wastewater to produce aluminum ion and ferric ion while the other reaction generates oxygen and hydrogen bubble to help suspended or dissolved organic of dyes to move upward (Equation 2.5). The metal ion reacts with hydroxyl ions and turns into metal hydroxide, oxyhydroxides, and polymeric hydroxides which presented coagulants. The metal hydroxide destabilizes the dye stability to form floc and move upward due to buoyancy (P. K. Holt et al., 2002). The Figure 2.4 will be show the whole reaction of electrocoagulation/flotation process.

$$2H_2O \leftrightarrow 2H_2 + O_2$$
 Eq. 2.5

The following reaction in ECF will be occurred when anode with a mental (Al), others equation will be show as below:

• Anode:

Al 
$$\rightarrow$$
 Al  $^{3+}$  + 3 e Eq. 2.6  
2H<sub>2</sub>O  $\rightarrow$  O<sub>2</sub> + 4H + 4 e



Figure 2.4. The interrelation occurred in ECF process (P. K. Holt et al., 2002)

In solution

$$Al^{3+} + 3H_2O \iff Al(OH)_3 + 3H^+$$
 Eq. 2.8

$$Al^{3+} + OH^{-} \rightarrow Al(OH)_{3}$$
 Eq. 2.9

• Cathode:

$$2H_2O + 2e \rightarrow H_2 + 2OH$$
 Eq. 2.10

$$2H^{+} + 2e^{-} \rightarrow H_{2}$$
 Eq. 2.11

#### 2.4.2 Factors affecting ECF process

1

There are many parameters which affect the ECF process such as a type of electrode, current density, electrode arrangement, pH effect, temperature effect, and effect of HRT.

#### a. Type of electrode

The plate of an electrode is one of the main parameters which are considered. The majority of EC studies select aluminum (Al) and Iron (Fe) plate to treat color from industrial wastewater with high efficiency. The number of metal ions required to remove a number of pollutants, it is usually to use iron for wastewater treatment and aluminum for water treatment because iron is relatively cheaper. Nowadays the aluminum plates are also popular use in wastewater treatment either alone or in combination with iron plates due to the high coagulation efficiency of Al<sup>3+</sup> (Shen et al., 2003). When there are a significant amount of Ca<sup>2+</sup> or Mg<sup>2+</sup> ions present in water, so the cathode material is recommended to use stainless steel plate (Kul'skii et al., 1978).

#### b. Electrode arrangement

Electrodes may be arranged either in two different way including monopolar or bipolar. In a monopolar electrode arrangement of an electrocoagulation cell, each pair of sacrificial electrodes is internally connected with each other and has no interconnections with the outer electrodes (Mollah, Morkovsky, et al., 2004a). This arrangement of monopolar electrodes with cells in series is electrically similar to a single cell with many electrodes and interconnections. There is two connections in monopolar electrode such parallel and series. In series cell arrangement required a higher potential difference for a given current to flow because cells connected in series have higher resistance. In this arrangement, the same current flows through all the electrodes. On the other hand, in a parallel arrangement, the electric current is divided between all electrodes in relation to the resistance of the individual cells which require low voltage than series arrangement. Some authors have used bipolar electrodes (Mollah, Morkovsky, et al., 2004a) with cells connected in series, where the sacrificial electrodes are placed between two parallel electrodes without any electrical connection. Only the two outer plate electrodes are connected to the electric power source, with no interconnections between the sacrificial electrodes. This type of cell arrangement provides a simple set-up, which facilitates easy maintenance during operation. When an electric current is passed through the two electrodes, the neutral sides of the conductive plate will be transformed to charged sides, which have opposite charge compared with the Outer plate side beside it. The sacrificial electrodes, in this case, are also known as bipolar electrodes.

#### c. Electrode gap

Electrode gap is also the important parameter that we must to consider. Many research was investigated and raised that increased distance between each couple of anode and cathode electrodes leads to increased voltage. The advantage of the increasing electrode gap is that it helps to promote the time for coagulants with particles to attract each other and then aggregated. On other hands, a closer distance between electrodes is desirable because electrical resistance increases with an electrode gap distance the gap between anode and cathode was too closed. Moreover, the accumulated solid particles and gas bubbles between the anodes and cathodes caused a consequent higher electrical resistance (Malakootian et al., 2010).

#### d. Current density

Current density is one of the most critical operating parameters in electrocoagulation, having a significant effect on process efficiency (Linares-Hernández et al., 2009). This parameter dictates both the rate of electrochemical metal dosing to the water and the

density of electrolytic bubble production. Literature sources report a wide range of current densities mostly applied between 1-100 mA/cm<sup>2</sup> depending on the case study. The current density was varied depending on their application use, for high current densities are needed for separation processes involving flotation cells or large settling tanks, while small current densities are appropriate for electro-coagulators that are integrated with conventional sand and coal filters. Faraday's law was expressed by a relationship between an amount of aluminum produced to electrolysis time and current. The Faraday's law relates to a theoretical amount of aluminum or Iron ions (M) and hydroxyl ions generated in the reactor to the current flow (Equation 2.12).

$$m = \frac{J \times t \times M}{n \times F}$$
 Eq. 2.12

Where, *m* is the amount of metal dissolved in the solution (g of M/cm<sup>2</sup>) *J* is the current density (A/cm<sup>2</sup>), *t* is detention time (s), *M* is relative molar mass of the electrode concerned (g/mol), *n* is Number of electrons exchanged in the reaction (C/mol), *F* is Faraday's constant (96,487 A\*sec/C).

*e. pH* 

Coagulation is strongly depended on the pH of a solution, so pH of water or wastewater is an important parameter that effects current efficiency and solubility of metal hydroxides present in the solution. Moreover, the number of aluminum ions are highly produced at acidic and basic conditions than at neutral conditions.

The reaction of floc formation created by hydrolysis and polymerization which form the complex polymer compound by different pH is expressed in the chemical equations as follow:

$$Al^{3+} + H_2O \rightarrow Al(OH)^{2+} + H^+$$
Eq. 2.13

$$AI(OH)_2 + H_2O \rightarrow AI(OH)_2^+ + H^+$$
Eq. 2.14

$$Al(OH)_{2}^{+} + H_{2}O \rightarrow Al(OH)_{3} + H^{+}$$
Eq. 2.15

$$\operatorname{Al}(\operatorname{OH})_3 + \operatorname{H}_2 \operatorname{O} \rightarrow \operatorname{Al}(\operatorname{OH})_4^- + \operatorname{H}^+$$
 Eq. 2.16

Aluminum electrode dissolute into various forms of ions based on pH of the aqueous medium. At pH of 2 to 3 dissolution of aluminum electrode forms  $Al^{3+}$ , and  $Al(OH)^{2+}$ .  $Al_{13}O_4$  (a polymeric form of aluminum) was formed and precipitated as  $Al(OH)_3$  (an amorphous form of aluminum) at a pH of 4- 9. The predominant chemical species formed at pH 10.

#### f. Temperature

Water temperature also one parameter may affect to treatment efficiency in electrocoagulation. The dissolution of the aluminum electrode was founded that the current efficiency increased rapidly when water temperature is between 2– 30 degree Celsius. An aluminum oxide surface layer will decrease when the temperature was increasing. However, the treatment effect will decrease when the temperature highly rises above 60 degrees Celsius (Sahu et al., 2014).

#### 2.5 Development of Electrocoagulation Reactor (ECR)

Electrocoagulation reactors have been built in a number of configurations. Each system has its own set of advantages and disadvantages, among which are varying degrees of treatment ability. Normally the electrocoagulation units are used to replace chemical dosing system with more advantages of the electrolytic gases produced. Many designs of electrocoagulation reactors were developed from year to year but electrocoagulation reactor design is still largely empirical and heuristic. Therefore, it becomes very difficult to compare the performance of different reactor configurations (Figure 2.7). Reactor geometry affects several operational parameters, including reactor configuration, bubble path and size, flotation effectiveness, floc formation, fluid flow regime and mixing/settling characteristics (P. K. Holt et al., 2005).

#### **2.5.1** Optimization design of reactor configuration

The major difference between alternative designs is whether a reactor was constructed as a batch or a continuous system. It is clear that the vast of applications fall into the latter category, having a continuous feed of wastewater and operating under (pseudo) steady-state conditions. A key advantage for this reactor systems is that their coagulant requirements are fixed, a vital advantage in terms of both design and operation. In a continuous system, water to be treated is dosed with dissolved metal ions as it passes through the electrocoagulation cell.



Figure 2.5. The design of batch reactor of EC system (Eyvaz et al., 2014)

For instance, Figure 2.6 will be shown about the continue system of electrocoagulation process. On another hand, in Batch reactor applications is typically operate with a fixed wastewater volume per treatment cycle but hurt from the perceived disadvantage (from a design and operational standpoint) that conditions within the reactor change with time. The Figure 2.5.express the design of EC as bipolar connection.



Figure 2.6. Design of continue reactor of EC system (Pérez-Sicairos et al., 2011)



*Figure 2.7. Classification of electrocoagulation reactor system (P. K. Holt et al., 2005)* 

#### 2.5.2 Optimization design of separation process

The important parameter which plays a major role after wastewater pass through EC unit is separation process. The additional separation processes are reported in bold with relevant references grouped accordingly. Integrated units have two main pollutant removal paths by flotation and settling (P. Holt et al., 2001). In the EC unit, the aluminum cations are hydrolyzed to a form capable of aggregating the pollutant which is then removed from solution by sedimentation or flotation. Separation by settling is the more familiar option, with the fact that electrolytic gases are also being produced concurrently with the dosing process often viewed as an unnecessary operational complication (Mameri et al., 1998). The main different between pollutant removal by settling or flotation would seem to be the current density worked in the reactor. A low current generates a low bubble density, leading to a low upward momentum fluxconditions that encourage sedimentation over flotation. As the current is increased, so the bubble density increases resulting in a greater upwards momentum flux and thus more likely removal by flotation. There are many kinds of equipment which are available for commercial such as Tube and plate settler and micro bubble generator that use to improve on separation process in sedimentation tank (Figure 2.8).



Figure 2.8. Equipment of improvement separation: plate and tube settler (left), micro bubble generation (right)

#### 2.5.3 Optimization Design of Fluid Dynamic

Due to the performance of the EC process depends on the geometry of the EC reactor design, so fluid dynamics is another aspect that requires consideration. EC cells can be either horizontal or vertical, depending on the placement of the electrode plates. Moreover, there can be single or multiple channels, depending on the solution flow. Multiple channels have a simple flow arrangement, but the flow distribution can be non-uniform due to either channeling or dead zones (Okoth et al., 2008). According to Ubale and Salkar (2017), EC cell was designed as the baffle to make horizontal flow and H<sub>2</sub> micro-bubbles produced (Figure 2.9). Therefore, the micro-bubbles formed at electrodes, the liquid phase, and the solid particles flowed all together in this EC unit. The formation of H<sub>2</sub> micro-bubbles was large enough due to a large surface area of electrodes. In fact, this fostered a homogeneous dispersion of the gas phase and the liquid phase and further promoted floc removal by electroflotation in the settling tank.



Figure 2.9. Design of fluid dynamic with multi baffles (Ubale and Salkar, 2017)

#### **2.6** Residence Time Distribution (RTD)

The resident time distribution (RTD) is a method which is used to analyze the performance of the flow behavior in all kind reactor (batch, plug flow tubular, and mixed continuous tank). Moreover, RTD of a reactor is a characteristic of the mixing which appeared in the chemical reactor. The plug-flow reactor is no axial mixing inside and this omission is reflected in the RTD which is shown by this class of reactors. The CSTR is a kind of reactor which thoroughly mixed and possesses a far different kind of RTD than the plug-flow reactor. Not all RTDs are unique to a particular reactor type; markedly different reactors can show identical RTDs. Nevertheless, the RTD displayed by a given reactor yields distinctive clues to the type of mixing occurring within it and is one of the most instructive characterizations of the reactor.

#### 2.6.1 RTD measurement

The RTD is determined experimentally by injecting an inert chemical, molecule, or atom, called a tracer, into the reactor at some time t = 0 and then measuring the tracer concentration, C, in the effluent stream as a function of time. In addition to being a nonreactive species that is easily detectable, the tracer should have physical properties similar to those of the reacting mixture and be completely soluble in the mixture. It also should not adsorb on the walls or other surfaces in the reactor. The latter requirements are needed so that the tracer's behavior will honestly reflect that of the material flowing through the reactor. Colored and radioactive materials along with inert gases are the most common types of tracers. The two most used methods of injection are pulse input and step input.

#### a. Pulse input experiment

In a pulse input, an amount of tracer  $N_0$  is suddenly injected in one shot into the feed stream entering the reactor in as short a time as possible. Then measure the outlet concentration as a function of time. The relationship between concentration-time curves at the inlet and outlet of an arbitrary reactor are shown in Figure 2.10. The effluent concentration-time curve is referred to as the C curve in RTD analysis. We analyze the injection of a tracer pulse for a single-input and single-output system in which only flow (i.e., no dispersion) carries the tracer material across system boundaries. First, we choose an addition of time ( $\Delta t$ ) sufficiently small that the concentration of tracer, C(t), existing between time (t) and  $(t+\Delta t)$  is essentially the same. The amount of tracer material,  $\Delta N$ , leaving the reactor between times (t) and  $(t+\Delta t)$  is in equation 2.17,

$$\Delta N = C(t) \times v \times \Delta t$$
 Eq. 2.17

Where, *v* is the effluent volumetric flow rate,  $\Delta N$  is the amount of material exiting the reactor that has spent an amount of time between *t* and  $t+\Delta t$  in the reactor. For pulse injecton we define with equation 2.18 and equation 2.19.

$$E(t) = \frac{v \times C(t)}{N_0}$$
Eq. 2.18
$$\frac{\Delta N}{N_0} = E(t) \times \Delta t$$
Eq. 2.19

Where, the quantity E(t) is called the residence-time distribution function. It is the function that describes in a quantitative manner how much time different fluid elements have spent in the reactor.



Figure 2.10. The measurement of RTD (Fogler, 2004)

The volumetric flow rate v is usually constant, so we can define E(t) as equation 2.26:

$$E(t) = \frac{C(t)}{\int_0^\infty C(t) dt}$$
 Eq. 2.20

#### b. Step tracer experiment

The step tracer experiment will be conducted after having a knowledge from the Pulse input experiment and try to formulate a more general relationship between a time-varying tracer injection and the corresponding concentration in the effluent. The output concentration from a vessel is related to the input concentration without development by the convolution integral (Levenspiel and Levenspiel, 1972).

$$C_{out}(t) = \int_0^t C_{in}(t-t') \times E(t') \times dt$$
 Eq. 2.21

Because the inlet concentration is a constant with time,  $C_0$ , it can be taken outside the integral sign, that is equation 2.22

$$C_{out} = C_{in} \int_0^t E(t') \times dt'$$
 Eq. 2.22

This expression was then differentiate to obtain the RTD function E(t):as equation 2.23.

$$E(t) = \frac{d}{dt} \left[ \frac{C(t)}{C_0} \right]_{step}$$
 Eq. 2.23

The positive step is usually easier to carry out experimentally than the pulse test, and it has the additional advantage that the total amount of tracer in the feed over the period of the test does not have to be known as it does in the pulse test. One possible disadvantage of this technique is that it is sometimes difficult to control a constant tracer concentration in the feed. Obtaining the RTD from this test also involves differentiation of the data and presents an additional and probably more serious drawback to the technique, because differentiation of data can, on occasion, lead to large errors. A third problem lies with a large amount of tracer required for this test. If the tracer is very expensive, a pulse test is almost always used to minimize the cost.

#### 2.6.2 Mean and moments of residence time

From the previous section 2.7.1.a) treating ideal reactors, a parameter frequently used was the space-time or average residence time ( $\tau$ ), which was defined as being equal to V/v. It expresses that, in the absence of dispersion, and for constant volumetric flow (v = v0) no matter what RTD exists for a particular reactor, ideal or non-ideal, this nominal space-time,  $\tau$ , is equal to the mean residence time,  $t_m$ .

In a case with other variables described by distribution functions, the mean value of the variable is equal to the first moment of the RTD function, E (t). Thus, the mean residence time as shown below with equation 2,24.

$$t_m = \tau = \frac{\int_0^\infty t \times E(t) dt}{\int_0^\infty E(t) dt} = \int_0^\infty t \times E(t) dt$$
Eq. 2.24

It is very popular to compare RTDs by using their moments instead of trying to compare their entire distributions (Wen and Fan, 1975). For this purpose, three moments are normally used. The first is the mean residence time. The second moment commonly used is taken about the mean and is called the variance, or square of the standard deviation. It is defined by equation 2.25.

$$\sigma^{2} = \int_{0}^{\infty} (t - t_{m})^{2} \times E(t) \times dt$$
 Eq. 2.25

#### 2.7 Literature Review

#### 2.7.1 Turbidity and color removal technique

There are many methods which are possible for turbidity and dye removal from the industrial wastewater including chemical, physical, biological process. The combination of 2 processes of adsorption and ion exchange was studied to treat acid dye from industrial wastewater effective which consider on effected parameter such as dye/sorbent interaction, sorbent surface area, particle size, temperature, pH, and contact time (Slokar and Le Marechal, 1998; Son et al., 2001). Another method is oxidization process which is commonly used a chemical to decolorize such as chlorine, ozone,

Fenton's reagents, UV/peroxide, UV/ozone, or other oxidizing techniques or combinations. Swaminathan et al. (2003) chose Fenton's reaction for the decolorization of 3 commercial dyes including Red M5B, Blue MR, and H-acid which is normally used in chemical industries for the synthesis of direct, reactive and azo dyes. The process is successful to remove all of these dyes which needed 10-25 mg/l of Fe<sup>2+</sup> dose and 400–500 mg/l of H<sub>2</sub>O<sub>2</sub> dose at pH 3.0. Moreover, in this study, the EC is also appropriate methods which applied ion sacrificial anode as a coagulant to treat the color of orange II dye in the aqueous phase was effectively removed (> 98%). Further same authors presented the decolorization of dye solution containing Acid red 14 by EC and compared different electrode connections. They concluded that an EC cell with monopolar electrodes had higher color removal efficiency than an EC cell with bipolar electrodes (Daneshvar et al., 2004). Nowadays biological process treatment improved their ability to enhance the decolorization by many agents such as aerobic, anaerobic, fungi, algae, and bacteria. An investigation of (Anjaneyulu et al., 2005) raised that adsorption of 2 basic dyes viz. basic blue and malachite green and two acidic dyes fast red and Congo red by using biomass of fungi which basic blue, malachite green and fast red showed 98% removal and Congo red showed only 85% removal. Until now membrane technique was available on the commercial with many types including microfiltration (MF), NF, UF, and RO. Base on (Koyuncu, 2002) observed Nanofiltration membranes to be the best for the removal of reactive dye in dye-salt mixture. Color removal was greater than 99% and decreased with increasing salt concentration.

#### 2.7.2 Electrocoagulation/floatation process for industrial wastewater

There are many technologies were applied in industrial wastewater in thought the ECF process was selected to treat COD, color, turbidity, BOD, and TOC with high efficiency. Base on B Merzouk, Yakoubi, et al. (2011) chose EC for serving the textile wastewater which contains pure red dye solution (disperse and acid dyes), COD, TOC, and turbidity. The results show that the treatment is satisfied with a high level of color from the effluent. Another researcher was shown that EC with sacrificial electrodes can treat COD (95%), color (99%), and turbidity (99%) which is reused 4 times from the industrial wastewater (Meas et al., 2010). According to (Aoudj et al., 2010) prove that

decolorization of Direct Red 8 from synthetic wastewater which can be achieved at 98% under the optimum condition of current density 1.875 A/cm<sup>2</sup>, an inert-electrode distance of 1.5 cm, and sodium chloride (NaCl). Additionally, the achievement removal from textile industrial turbidity about 76.2 %, suspended solid 85.5 %, 88.9% BOD, 79.7% COD, and 93% color which treat by ECF with optimum condition of current density of 11.55 mA/cm<sup>2</sup>, electrode gap 1 cm, 300 mg/L of silica, pH of 7.6, and conductivity of 2.1 mS/cm with treatment time of 10 min (Belkacem Merzouk et al., 2010).

#### 2.7.3 Study of residual time distribution (RTD) in the ECR

The study of liquid flow is very important in order to know the flow mixing inside the new reactor and also the effect of the baffle on hydrodynamic by using RTD as the main parameter. In electrocoagulation reactor has many researchers investigated and try to develop by installing the baffle to improve flow mixing inside and reduce some dead zone and bypass. When the lesser dead zone and bypass is, the spread of molecule inside the reactor is better. According to Ubale and Salkar (2017) and Apshankar and Goel (2014) express that when they install the baffle, it is significant that it can make the flow from laminar to turbulent flow for increasing the catching of coagulant with pollutant.

#### 2.7.4 Reactor design of electrocoagulation process

Reactor design of EC was developed and more choices for a commercial and industrial sector. The monopolar as series and parallel and bipolar as series are the arrangement of electrode plate as one part of a design. Base on who investigate on three arrangements together to improve the capacity of treatment efficiency of the textile industry that can treat color and turbidity by aluminum and iron. Besides that, another researcher designed the ECR by varying the baffle inside to improve treatment performance. Moreover, the design as a continuous or batch system is also important for consideration. According to Kim et al. (2002) expressed that electrocoagulation as continue system to treat color which is generated by reactive dye and disperse dye from textile industry with efficiency of rectangular reactor (110 mm \* 100 mm \* 150 mm) with flow in range 50 to 200 ml/min and take electrode as baffle in order to make one

direct as tubular flow. Study by Zodi et al. (2009) showed that treatment of wastewater from textile by EC with sedimentation process in reactor of 2 litters with aluminum and iron rectangular plate 15 cm \* 7 cm \* 1.2 cm and current density in range of 50 to 200 A/m<sup>2</sup>, pH from 3 to 9, and time for settle is about 2 or 3 hours.

#### 2.8 Research Summary

Wastewater from industries is the most concern for worldwide especially for any countries which are poor technology. The most contaminants from effluence industries wastewater are color, turbidity, COD, and heavy metal. A color which is generated by synthetic dye and turbidity are the main pollutant of the effluent. Azo group is one of the most popular and the vast amount of usage around 50 - 60 % around the world. In the azo group, it has many types of dye such as the reactive dye, disperse dye, acid dye, vat dye, sulfur dye, basic dye, and other dyes. The type of reactive dye was select to study in this research because of it also has a huge amount uses in the textile industry about 20 - 50%.

There are many treatment methods was applied to remove effluent from industry which categorize in 3 main parts including phyco-chemical, chemical, and biological. In those, there are 2 technologies were select to study such as conventional coagulation and electrocoagulation process because of these technologies are well performance in terms of comparing advantages and disadvantages to another treatment methods in Table 2.3 above. Due to the effluent from EC is acceptable, which by-product is nontoxic, faster time, and easy to operate comparing to CC, hence, EC was selected to study.

Although there is much research on the Electrocoagulation process, it still has some gap and limitation on design reactor. In this research was investigate the reactor design by combine 2 processes together by EC with sedimentation process as continue system which calls electrocoagulation reactor (ECR). Inside of this ECR was explored by varying the acrylic baffle inside to make flow inside as one direction and make a turbulent flow which substitutes the electrode cell of EC before. At the same time RTD will be applied to measure the flow pattern inside the reactor and then DOE will help to apply to reduce some experiments of design new ECR. Moreover, the amount of EC sets and economic current also investigate too.

# CHAPTER 3 MATERIAL AND METHODS

#### 3.1 Study Overview

This research tries to develop a combination process between EC with sedimentation to treat color and turbidity from synthetic textile as industry wastewater. The overview of this research was classified into 7 parts such as (i) prepare and synthetic wastewater, (ii) explore removal efficiency of CC, (iii) investigate the optimal electrode configuration, (iv) study kinetic treatment in batch column, design and construct of rectangular reactor, (v) study the flow mixing inside the new reactor, (vi) evaluate treatment performance of ECR for turbidity and color removal, and (vii) improve separation part by adding tube settler and provide guideline of batch and continuous system of EC. The detail diagram of each system was shown in Figure 3.1.



Figure 3.1. Overview of framework

#### 3.2 Experimental Set-up

There are 2 reactors that use to study in this research. The explanation and description of each reactor described below:

• For first reactor, it is a batch acrylic column which contain volume 4 litters with diameter 13 cm and high 40 cm. Moreover, it has 1 set of electrode plate, which 1 set contains 4 electrodes plates. The size of each electrode plate was 20 \* 5\*0.2 cm that connect to Direct Current (DC) supply for screening the optimal electrode configuration, current density and some parameter related to reactor design including hydraulic retention time (HRT), and overflow rate (OFR). The reactor is shown in Figure 3.2. In the beginning, the water pump bring water to the batch column, and then the EC set was connected to the DC supply, after that put in the reactor. During operate EC process in batch column, the oxygen and hydrogen gas was collected by the gas soap film meter at the cap. The optimal electrode configuration was selected in terms of ratio between gas flowrate over loss. For another parameter such as kinetic, HRT, OFR, and treatment efficiency was measure the final concentration at the effluent.



Figure 3.2. Experimental Set-up for screening test in simple batch column reactor

• Another reactor is Electrocoagulation reactor (ECR) which has 2compartments. The first compartment is electrocoagulation process which contains 90 L with width 26 cm, length 100 cm, and height 40 cm include freeboard 0.5 cm that varies with vertical baffles and uses 4 sets of electrode plats. The second part is sedimentation process which contains 34 L with width 16 cm, length 70 cm, and average height 30 cm. Furthermore, in the second part also has one port which contains 17 L with width 16 cm, length 30 cm, and height 35 cm which has functioned as flocculation after electrocoagulation part.



Figure 3.3. Experimental Set-up with Electrocoagulation reactor (ECR): a) Top view, b) Perspective view.

Tube settler also present in separation part. The experimental set-up is shown both clearly in a top view and section view in Figure 3.3. At the beginning, the synthetic wastewater was filled in the tank, then the flow meter was used to control the liquid flowrate in the system. Each set of electrode plate was put inside the reactor and connect to the DC supply. The baffle has a function to reuse the bypass and dead zone inside reactor by making the turbulent flow. Tube settler also helps to improve the removal efficiency of sedimentation in case that sedimentation cannot remove the contaminant to reach the target. After finish the processed, sludge will throughout by 3 places which install for flush sludge out as the blue point of top view in Figure 3.4 (a).

## 3.3 Material and chemical reagents

#### 3.3.1 Apparatus

As mention above, simple column reactor was constructed with acrylic of thickness 0.5 cm. which has area  $133 \text{ cm}^2$  and contain volume 4 L. This reactor explore some parameter such as electrode arrangement (monopolar and bipolar), inner gap of electrode. The real installation of column reactor, inner gap and electrode plate shown in Figure 3.5 (a), (b) and (c), respectively.



Figure 3.5. a) Column reactor of 4 L with experiment, b) support of plate with inner gap of electrode, and c) type of electrode plate

 The acrylic rectangular of ECR reactor combine 2 processes together of electrocoagulation with separation process that has total volume 141 L. The study effect of baffle was investigated in this reactor with various length of baffle. The real installation of continue ECR reactor and their compartments show in Figure 3.6



Figure 3.6. a) Continue ECR reactor 141 L with experiment, b) Vertical baffle with 3 length, and c) Amount of EC sets

- 2. Pair of aluminums and irons electrode size with 5cm width, 20 cm height, and the length 0.2 cm. For one set of electrode has 4 plates, which all of the plate are the major role to produce the coagulant and generate the bubble.
- 3. Direct current (DC) is important role in the electrocoagulation process because it convert the AC to DC to supply the electricity to anode and cathode side with maximum capacity of (60V, 6A).
- 4. Submerge pump is necessary for the whole system which bring the synthetic wastewater into the reactor. The SONIC AP 4500 model was used in the system which has capacity 2500 litter per hour.
- 5. Plastic tank which has volume 90 liter for store the synthetic wastewater when operation start and receive the wastewater or sludge after operation stop.

#### 3.3.2 Analyze equipment

- Flow meter: is important to control the flow in and out of the system to get the balance of liquid flow which run as continuous mode in the turbidity and color removal and RTD measurement. The limitation of this equipment was 1 – 10 L/min.
- 2. Soap film meter: finding the best configuration of electrode condition in EC process to measure the gas flowrate. The range of equipment was 0 30 ml.
- 3. Jar test set: this experiment was performed to explore the optimal level of alum dosage by adding coagulants in chemical coagulation process (CC). The speed of machine stay range 0 300 rpm.
- 4. pH meter: is necessary for the process. It measure the pH, which is the parameter that can effect to the coagulation process when it is very low or high. METTLER-TOLEDO model was used in experiment. This machine stay in range 0 -14 which has accuracy about  $\pm$  0.01.
- 5. Turbidity meter: measure the turbid parameter during the finding the treatment efficiency of bentonite removal. The HI 98703 model was selected with limitation less than 1000 NTU.

 Spectrometer meter: find the color removal in batch column and ECR reactor to validate the treatment performance of the process. It use GENESYS 10S UV-Vis model with limitation in of wave length range 190 – 1100 nm and glass cuvette 10 mm

7. Conductivity meter: to study the resident time distribution (RTD) to represent the fluid flow pattern in reactor, tracer concentration of sodium chloride (NaCl) need to measure along the time. The electrical conductivity meter model LUTRON CD-4317SD with capacity range from 0 to 200 mS and accuracy level of  $\pm 2\%$  at 23  $\pm 5^{\circ}$ C is used to measure the electrical conductivity in both influent and effluent stream.

#### 3.3.3 Chemical reagents

There are several chemical below will be use in this experiment:

- Powder Bentonite: use as turbidity by synthetic the powder bentonite clay with tap water to get the concentration we want. The specification of bentonite was calcium base which has chemical composition SiO<sub>2</sub> = 70%, Al<sub>2</sub>O<sub>3</sub> = 14%, and Fe<sub>2</sub>O<sub>3</sub> = 1.9%, and also has the physical performance of passing rate (200 mesh %) = 95%.
- Powder Dye @ Reactive yellow 135dye (HE-6G) of commercial grade which use as color by synthetic the powder dye from commercial with tap water to get the concentration we want. The solubility of this dye around 80 – 85 % in water, which has molecular formula C<sub>52</sub>H<sub>30</sub>Cl<sub>6</sub>N<sub>18</sub>Na<sub>6</sub>O<sub>20</sub>S<sub>6</sub>.
- Tracer Sodium chloride (NaCl): use as tracer in residual time distribution (RTD) to find the hydraulic which NaCl has solubility around 95% in water at temperature 25 °C.
- Alum (Al<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub>.18H<sub>2</sub>O): use as coagulant dose in coagulation process which has specification such as Al<sub>2</sub>O<sub>3</sub> around 17% as powder form.

# 3.4 Analytical parameter

# 3.4.1 Gas flowrate HULALONGKORN UNIVERSITY

The gas flow rate was measured by a direct method which using the soap film meter and stopwatch. This parameter will be measured in the electrocoagulation process in the batch column reactor, which calculates by the equation 3.1 below:

$$Q_{g} = \frac{\Delta V}{\Delta t}$$
 Eq. 3.1

Where,  $Q_g$  is the gas flow rate (ml/s),  $\Delta V = V_2 - V_1$  is the different volume of gas (ml), and  $\Delta t = t_2 - t_1$  is the different time of gas move (s).

Electrodeless will use the direct method to measure the weight of electrode plate at both anode and cathode side before and after an experiment in the column reactor for electrocoagulation process which measures with six digits. One experiment for pretest takes time 15 min. The electrode loss formula show with equation 3.2 as below:

$$Electrode \ loss \ (\%) = \frac{Initial \ weight \ (g) - Final \ weight \ (g)}{Initial \ weight \ (g)} Eq. \ 3.2$$

#### 3.4.2 Kinetic modeling

Kinetic is the important parameter to show the reaction in the system such as reaction order, constant rate, and hydraulic retention time of reaction in the batch reactor. For this study kinetic observe with zero, first order and second order kinetic model were considered to describe for the removal rate (Al-Shannag et al., 2015). The EC batch process, the mass conservation of pollutants ion is show in equation 3.3:

$$-\frac{dc}{dt} = (-r_D)$$
 Eq. 3.3

Where,  $(r_D)$  is the removal rate in (ppm/min) and t is EC time in (min).

The first order model:

For the first order model  $(-r_D = k_1C)$ , the integration of equation 3.3 at the initial concentration  $C(0) = C_0$  gives with the equation 3.4 below:

$$C(t) = C_0 e^{-k_1 t}$$
 Eq. 3.4

Where,  $k_1$  is the first-order rate constant in (min<sup>-1</sup>).

• The second order model:

For the second order model  $(-r_D = k_2 C^2)$ , the time-dependent concentration is obtained as equation 3.5.

$$\frac{1}{C(t)} = \frac{1}{C_0} + k_2 t$$
 Eq. 3.5

Where,  $k_2$  is the second-order rate constant in (ppm<sup>-1</sup>min<sup>-1</sup>).

#### **3.4.3** Settling fraction removal

The setting test was test in the batch bubble column reactor to determine the settling fraction removal ( $R_T$ ). The percentage removal of each sample at any time and each depth of the column reactor. The friction removal at time  $t_c$  after find an over flowrate for different settling time ( $t_a$ ,  $t_b$ , and  $t_c$ ),  $v_0 = H / t_c$  should be equation 3.6 following:

$$R_T = R_C + \frac{H_2}{H} \times \left(R_D - R_C\right) + \frac{H_1}{H} \times \left(R_E - R_D\right)$$
Eq. 3.6

Where,  $H_2$ : The height that particle ( $R_D - R_C$ ) size settling at  $t_c$ . The various time was use such as  $t_a t_b$  and so on with the different fraction removal. The graph of overflow ate and friction removal will be construct in terms of depth and times as Figure 3.7.



Figure 3.7. Percentage removal graphic at certain depth and time

#### 3.4.4 Turbidity removal

The turbidity removal efficiency will study in 2 parts in the research. The first, removal efficiency was found and validated the optimal condition and configuration of electrode in column reactor. The second, the removal efficiency evaluate and validated the treatment performance of development of ECR reactor. The turbidity removal efficiency can calculate by comparing the initial and final of turbidity as NTU unit. The equation 3.7 express as below:

$$Turbidity \ removal \ (\%) = \frac{Initial \ turbidity \ - \ Final \ turbidity}{Initial \ turbidity} \times 100$$
Eq. 3.7

#### 3.4.5 Color removal

The color removal efficiency will study in 3 parts for this research work. The first, the removal efficiency was found and validated the condition and configuration of electrode in column reactor. The second, finding treatment performance in ECR as continuous system. Then the last part, to evaluate the treatment performance of development of ECR reactor. The color removal efficiency can calculate by comparing the initial and final of color from the spectrometer as absorbent (Abs.) which is convert to ADMI with a standard curve. The equation 3.8 express as below:

$$Color \ removal \ (\%) = \frac{Initial \ color \ - \ Final \ color}{Initial \ color} \times 100 \qquad \text{Eq. 3.8}$$

For measuring color removal in this was measure only absorbent (Abs.), then convert Abs to ADMI unit. The correlation between concentration (mg/L) to Abs and ADMI was show in the Figure 2 in appendix 1. For measuring Abs required wavelength 420 nm and ADMI require wavelength 720 nm. The Abs to ADMI was convert follow by this correlation in appendix1.

#### 3.4.6 Residual time distribution (RTD)

The objective of this study was used the RTD to know characteristic or behavior of the fluid dynamic in the ECR reactor during the operation process. The experiment started with plus injection method by adding the 220 mg/L of NaCl as a tracer. The conductivity profile is converted to RTD function (E-curve) by the equation 3.9 (Fogler, 2010)

$$E(t) = \frac{C(t)}{\int_{0}^{\infty} C(t)dt}$$
Eq. 3.9

Normally, E(t).dt is a fraction of the tracer that spend amount of time between time t and t+dt and C(t) is the concentration of tracer that is measured along the time t at an

exit stream. Due to the linear relationship between concentration and electrical conductivity of tracer in the system, C(t) can be obtained by converting from existed electrical conductivity data of experiment. The E(t) can be described the characteristic of reactor base on its aspect. The mean of residence time can be calculate from equation 3.10

Furthermore, it is possible to indicate that the reactor exist dead volume or circuiting by comparing between the first moments of distribution function called mean residence time,  $t_m$  as define in equation 3.10, with theoretical residence time  $\tau$ , that is the ratio of reactor volume and liquid flow rate ( $\tau = \nu/Q$ ).

After designed and constructed of ECR reactor as the continue mode already, the flow pattern and reactor types (plug flow, completely mix, or combine together between CSTR with PFR) of the non-ideal reactor were studied with the Residence time distribution (RTD) principle show in *Figure* 3.8. The RTD function in this research was select pulse injection method by injecting the tracer one time at the influent and then measure the conductivity along the time with the probe of conductivity meter at the effluent of the one at least. The sodium chloride (NaCl) was selected as the tracer, which was a popular tracer that many researchers used (Romphophak et al., 2016).

# 3.4.7 Power consumption and operation cost

The importance parameter economical in EC process are electrical energy consumption and current efficiency (Daneshvar et al., 2006) Electrical energy consumption was also calculated using the commonly equation.3.11.

$$E = U \times I \times t$$
 Eq. 3.11

Where, *E*: Electrical energy consumption with unit (kWh/kg <sub>dye removal</sub>), *U*: The cell voltage in volt (V), *I*: The current with Ampere (A), and *t*: The time of operation (hour).



Figure 3.8. Characteristics of reactors by RTD principle (a) RTD for near plug-flow reactor; (b) RTD for near perfectly mixed CSTR; (c) Packed-bed reactor with dead zones and channeling; (d) RTD for packed-bed reactor in (c); (e) tank reactor with short-circuiting flow (bypass); (f) RTD for tank reactor with channeling (bypassing or short circuiting) and a dead zone in which the tracer slowly diffuses (Fogler, 2010).

The current efficiency ( $\phi$ ) of EC process was calculated by the equation.3.12. Normally we use the equation 3.13 to show the dissolve of ion in the water but in this calculation was based on the comparison of experimental weight loss of iron electrodes ( $\Delta M(exp.)$ ) during EC with theoretical amount of ion dissolution ( $\Delta M(Theo.)$ ) as following:

$$\varphi = \frac{\Delta M (\exp.)}{\Delta M (Theo.)} \times 100$$
 Eq. 3.12

$$\Delta M (Theo.) = \frac{M \times I \times t}{n \times F}$$
 Eq. 3.13

Where, *M*: the molecular weight of the iron (g/mol), *n*: the number of electron moles, *F*: the Faraday constant (F = 96487C/mol). The specific electrical energy consumption (SEEC) was calculated as a function of iron electrodes weight consumption during EC process follow by the equation 3.14.

$$SEEC = \frac{n \times F \times U}{3.6 \times 10^3 \times M \times \varphi}$$
 Eq. 3.14

Therefore, the operating cost includes material cost focus on mainly electrodes, utility cost depend on mainly electrical energy, as well as labor, maintenance and other fixed costs. Energy and electrode material costs are taken in account as a vast cost item in calculation of the operating cost for economic study. According to Bayramoglu et al. (2004), equation 3.15 show about the calculation of the operating cost as kWh per kg of dye removed, where a is the electrical energy price and b is electrode material price and C <sub>energy</sub> and C <sub>electrode</sub>, are consumption quantities per kg of dye removed, which are obtained experimentally:

Operating 
$$\cos t = a \times C_{energy} + b \times C_{electrode}$$
 Eq. 3.15

#### 3.5 Experimental Procedure

This research was described in the series work as below. The experimental procedure was divide in 8 parts to represent the overall work including:

- Prepare and analyze of industry wastewater as the color and turbidity
- Conventional coagulant with Jar test
- Electrocoagulation (EC) study in the batch column reactor
- Analyze and design of continues rectangular ECR reactor, then the fluid dynamic characteristic inside the new reactor was studied.
- Fluid dynamic in ECR reactor

- Evaluate treatment efficiency of turbidity and color by continue ECR
- Improve the continuous ECR reactor with tube settler in case that effluent from sedimentation are not acceptable.

## 3.5.1 Prepare and analyze of industrial wastewater as color and turbidity

This section aims to prepare synthetic power bentonite (g) and reactive powder dye (g) mix with tap water as turbidity and color respectively. Synthetic wastewater was kept for future use to find the optimal electrode configuration and treatment efficiency. The process of synthetic dye was shown as the flowing step below:

a. Turbidity

Synthetic turbid water was prepared by adding bentonite in (g) into 1 L of tap water to create the standard curve between values of turbidity with concentration of bentonite. Then, the rapid mechanical in speed 300 rpm in 5 min and slow mixing agitator 40 rpm in 30 min will be need to make the bentonite suspension is a uniform dispersion of bentonite particles. After the agitation the bentonite turbid solution was allowed to settle for 35 minute. The resultant solution obtained upon filtration is the Stock solution to get the turbidity in 50, 250, and 500 NTU.

b. Color

# จุฬาลงกรณ์มหาวิทยาลัย

Synthetic of color was prepare as the wastewater from textile industry by adding power reactive dye of commercial grade (g) with 1L of tap water in order to get the standard curve for both absorbent and ADMI. The rapid mechanical mixing agitator will be need to make the solution homogenous in order to get the concentration the color 25, 50, and 100 mg/L, respectively.

# **3.5.2** Chemical coagulants process (CC)

Jar test methods was studied to figure out the optimal of coagulant dosage for chemical coagulant process. After find the optimal dosage of coagulant of aluminum sulfate  $(Al_2(SO_4)_3 \cdot 18H_2O, AR Grade)$ , then compare with aluminum that use as electrode plate of ECF process. In jar test process, the requirement of rapid mixing process took 1 minute 100 rpm and, then change to slow mixing at 40 rpm for 40 minutes. After that,

the wastewater was settled for 30 minutes. After finishing 30 minutes, the turbidity, color, and pH was measured. Figure 3.9 and Table 3.1 presented in detail the parameters and data required for this part.



Figure 3.9. Schematic of chemical coagulation part

Parameters	Name or Value		
Fixed Parameter			
Synthetic wastewater (color and turbidity respectively)	(25, 50, 100) mg/L & (50, 250, 500) NTU		
Wastewater	1 L		
Coagulant <b>วุฬาลง</b>	Aluminum sulfate		
Time settle CHULALO	30 minutes		
4 Studied Parameter			
Coagulant dose range	5 – 800 mg/L, 1 - 100 mg/L, respectively.		
<b>4</b> Respond parameter	•		

# 3.5.3 Electrocoagulation (EC) study in the batch column reactor

## a. Optimization of electrode configuration operation

Wastewater quality

This part was investigated with tap water to get the best electrode gap, electrode arrangement, and current density in terms of gas flowrate and electrode loss after 15

Optimal dosage with removal efficiency

min. The diagram of the whole process in this part was shown in Figure 3.10 and Table 3.2 and will be detail the necessary parameters.



Figure 3.10. Schematic of screening the electrode development part

Table 3.2.	Best configuration of electrode parameter

Parameters	Name or Value	
Fixed Parameter		
Amount of Aluminum electrode plate	4 plate	
Tap water	4 L	
Wastewater (Bentonite, dye)	250 NTU, and 50 mg/L	
Time operated <b>GHULALONGI</b>	30 – 90 min. <b>EST</b>	
4 Studied Parameter		
Electrode gap	1.0 cm, 1.5 cm, 2.0 cm	
Electrode arrangement	Monopolar and Bipolar	
Current density	1.5 mA/cm <sup>2</sup> , 2.0 mA/cm <sup>2</sup> , 2.5 mA/cm <sup>2</sup>	
<b>4</b> Respond parameter		
Electrode performance	Gas flowrate (ml/s)	
Electrode performance	Electrode loss (%)	
Wastewater quality	Turbidity and color removal	
# b. The simple kinetic study

Taking the optimal configuration of electrode from the last part to explore the optimal current density and kinetic rate constant, reaction order, HRT of reaction in the system of electrocoagulation process in terms of treatment efficiency (turbidity and color removal) along the time, as shown in Figure 3.11 and Table 3.3.



Figure 3.11. Schematic of study treatment performance and kinetic

Table 3.3.The study of kinetic and treatment performance with different currentdensity in batch column ECF reactor

Parameters	Name or Value	
+ Fixed Parameter	หาวิทยาลัย	
Optimal configuration of electrode	arrangement, gap	
Volume of synthetic wastewater	4 L	
Synthetic wastewater (color and turbidity respectively)	50 mg/L and 250 NTU	
Time sampling	Every 5 min.	
4 Studied Parameter		
Current density	1.5, 2.5, 3.5, 4.5, 5.5 mA/cm <sup>2</sup>	
Initial concentration (turbidity, dye)	1.5, 2.5, 3.5, 4.5, 5.5 mA/cm <sup>2</sup> (50, 250, and 500) NTU, (25, 50, and 100) mg/L, respectively.	
4 Respond parameter		
Wastewater quality	Turbidity & Color removal	
Kinetic performance	Reaction order, Constant, and HRT of reaction	

#### c. Settling test in batch column ECF reactor

This section will find the friction removal, overflow rate (OFR) of floc settle down or floating up by operate the settling test in the batch column ECF reactor after electrocoagulation process. There are 2 study of the settling, the first is taken the synthetic settle down without chemical and the second is take it settle down after EC process immediately. The sedimentation process consume time about 100 min with 6 ports for sampling from the top surface of water respectively. The detail show in the Figure 3.12 and Table 3.4.





Figure 3.12. Schematic of settling test performance

Table 3.4. Parameter measurement of sedimentation part

Parameters	Name or Value				
Fixed Parameter					
Volume wastewater	4 L				
Synthetic wastewater (color and turbidity respectively)	50 mg/L and 250 NTU				
Measurement period	Every 3 min				
4 Studied Parameter					
Time	30 min				
Depth from the top surface water	13, 18, 23, and 28 cm				
+ Respond parameter					
Settling performance	Friction removal ( $R_T$ %) of turbidity and color respectively.				
	Overflow rate (OFR)				

# 3.5.4 Analyze and design of continue rectangular ECR

After well study of batch column reactor in ECF, we get many data such as best configuration of electrode (gap, arrangement), optimal current density, kinetic performance in system (reaction rate, constant, and HRT), and OFR of floc settle. All of these parameters will be calculate in order to design and construct continue rectangular ECR (Figure 3.13).



Figure 3.13. The ideal on reactor design and modification for RTD

## 3.5.5 Fluid dynamic in the ECR reactor

The modify thing in reactor will start such as amount of baffle and baffle length. Then Residual time distribution (RTD) will be study to explore the best configuration of new ECR reactor by focus on four points. The first point was study the position of probe, the second points was studied about the effect of flow mixing after have baffle. The third points was focus on the effect when operated EC and without operated, and the last point was the effect of flowrate on the system. The detail diagram and parameter show as Figure 3.14 and Table 3.5.



Figure 3.14. Process of design and study in ECR reactor

Table 3.5. Parameter	• design for the	effect baffles
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Tuble 5.5. I drameter design for the effect baffies					
Parameters	Name or Value				
+ Fixed Parameter					
ECR reactor	141 L with tap water				
Tracer (NaCl) concentration ONGKOP	220 mg/L				
Flowrate	6 L/min.				
4 Studied Parameter					
Amount of baffles	0,1,2,3				
Baffles length	(50%, 70%, 90%) of total baffle length				
Probe position	6, 12, 18, and 24 cm				
Vary flowrate	1, 3, and 6 L/min				
4 Respond parameter					
Retention time distribution (RTD)	Flow pattern in reactor				
Best ECR configuration	Configuration and condition inside the reactor				

## 3.5.6 Study treatment of turbidity and color by continue ECR

In this part, there are four points to study. At the first point was focused on the effect of flowrate on the dye removal with initial concentration at 50 gm/L. The second point was that the effect of the varying the current density which started from 4.5, 9.0, and  $13.5 \text{ mA/cm}^2$ , respectively. The third point is validation of initial turbidity concentration about 250 NTU with optimal condition from dye removal. The last point is that combine together of turbidity and dye with 250NTU and 50 mg/L in the same time. The schematic and detail parameter will be list in *Figure* 3.15 and Table 3.6.



Figure 3.15. The overall process of electrocoagulation process

# 3.5.7 Improvement of the ECR

In this section, it is important when the removal efficiency form the separation process do not perform well. The improvement of ECR in the sedimentation port is installed tube or plate settler which help to take particle settle faster than normal by gravity. The design of tube settler including tube sets, location, and size (diameter, length, and angle) will be investigate. The detail diagram of whole process and improvement parameter was shown as *Figure* 3.16 and Table 3.7.



Figure 3.16. Diagram of improvement of continue ECR by tube settler system

Table 3.6.Parameter design i	n electrocoagulation part
_	

Parameters	Name or Value				
Fixed Parameter					
ECR reactor	141 L				
Initial Concentration of dye and turbidity	50 mg/L, and 250 NTU				
4 Studied Parameter					
Current density	4.5, 9, 13.5 mA/cm <sup>2</sup>				
Flowrate	1, 2, 3 L/min				
Contaminant condition	Dye, turbidity, and mix (dye + turbidity)				
Respond parameter					
<b>CHULALONG</b> Wastewater quality	Operation condition of single and mix of contaminant from synthetic of wastewater				

Parameters	Name or Value
Fixed Parameters	
ECR reactor	141 L
Contaminant of wastewater	Mix of dye with turbidity
Concentration	250 NTU and 6000 ADMI
Length of tube sets (1 set)	30 cm
4 Studied Parameter	
Design of tube settler	Angle, size, and length
Tube sets	1 and 2 modules
+ Respond parameter	
Wastewater quality	Improve turbidity, color removal
Design criteria of tub settler	Knowledge of design on improvement of separation process

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Table 3.7. Parameter improve in sedimentation part

# CHAPTER 4 RESULTS AND DISCUSSIONS

This chapter presented the results and discussion of each part in this work. There are two objectives of this present work including optimize the configuration and economize the new EC reactor for turbidity and color removal, and to evaluate the treatment performance of ECR in continue mode. The main results were listed below:

- Prepare and analysis synthetic wastewater
- Removal of turbidity and color by chemical coagulation (CC)
  - Turbidity and color removal by CC
  - Estimating Treatment Cost of Conventional Coagulation (CC)
- Optimization of EC Condition
  - Electrode configuration
  - Electrode arrangement
  - Current density
- Study the Kinetic Treatment in EC
  - Effect of initial concentration
  - Model prediction
  - Settling test
  - Cost estimation
  - Comparative analysis between EC and CC
- Design of ECR for Turbidity and Color Removal
  - Liquid Flow Pattern in ECR
    - Effect of probe position height
    - Effect of baffles on fluid flow in ECR
    - Effect of operation condition with and without EC Process
    - Effect of Liquid Flow rate in ECR
- Treatment Performance of ECR as Continuous Mode
  - Effect of flow rate on decolorization
  - Effect of current density on decolorization
  - Validation turbidity treatment
  - Effect of flow rate on turbidity removal
  - Simultaneous removal of turbidity and color
- Settling Performance through Tube Settler Addition
  - Design tube settler
  - Effect of tube settler quantity
  - Effect of position installation
  - Design criteria

## 4.1 Prepare and Analysis Synthetic Wastewater

This section aims to describe the physical and chemical parameters of synthetic reactive dye and bentonite clay as the textile industry wastewater. It was prepared by mixing dyes together with tap water under 300 rpm stirring for 5 minutes in order to get concentration 2500 ADMI, 6000 ADMI, and 15000 ADMI, respectively (Figure 4.1.b). For turbidity was synthesized from bentonite mixing with tap water under 300 rpm, 40 rpm for 5 minutes and 30 minutes, respectively. Then let it settles about 35 minutes in order to obtain the concentration 50, 250, and 500 NTU (Figure 4.1.a). Table 4.1 showed the detail characteristics of all parameters including color, turbidity, COD, BOD, TSS, TDS, pH, conductivity, and temperature. Based on this results shown that bentonite preparation for turbidity 1000 mg/L get only SS 200 mg/L, which expressed that 80% was omitted. Moreover, mostly of dye was dissolve in water around 90%.

The standard curve for these preparations of color, turbidity, and sodium chloride (NaCl) was used as the tracer for the residence time distribution (RTD) part, which provided in Appendix I. The next experiment focuses on treatment efficiency of turbidity and color removal by chemical coagulation (CC) by using aluminum sulfate or alum. Moreover, the cost treatment also shows in the same section.

Parameters	Unit	Unit Tap water (25 -		Bentonite clay (1 – 12 g/L)
рН	-	7.05	6.90 - 7.01	7.04 - 7.06
Conductivity	mS/cm	0.281	0.296 - 0.336	0.285 - 0.289
Temperature	°C	24.7	24.8 - 24.9	24.6 - 24.7
Color	ADMI	-	2,500 - 15,000	-
Turbidity	NTU	0.5	-	50 - 500
TSS	mg/L	3	6 – 22	193 – 2,563
TDS	mg/L	188	208 - 269	190 - 192

Table 4.1. Charateristc of yellow reactive dye 135 (HG-6) and Bentonite clay



Figure 4.1. The preparation of turbidity and color, a) synthetic of bentonite as turbidity, b) synthetic of reactive dye as color.

# 4.2 Removal of Turbidity and Color by Chemical Coagulation (CC)

Chemical coagulation (CC) was selected to treat the turbidity and color from the synthetic wastewater by choosing alum ( $Al_2(SO_4)_3 \cdot 18H_2O$ ). Jar test applied to find the optimal concentration alum dosage to remove the different concentration of turbidity and dye. The procedure of jar test is rapid mixing 100 rpm for 1 min, 40 rpm for 40 min, and let settle 30 min. (Black et al., 1957). After that, the sample also measures on the top clear for turbidity and color, respectively.

## 4.2.1 Turbidity and color removal by CC

The removal efficiency of the chemical coagulation process is mainly depended on pH, coagulant dose, and coagulation time (Khandegar and Saroha, 2013).

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a. Turbidity removal

The concentrations of aluminum sulfate were studied in range between 1 to 160 mg/L, however, the result showed only in the range up to 30 mg/L as the optimum zone. The result was illustrated in Figure 4.2. It can be observed that the residual turbidity decrease after increasing amount of coagulant due to the required amount of coagulant is much enough for destabilization mechanism (Duan and Gregory, 2003). Specifically, at the low concentration of turbidity, 50 NTU, the residual bentonite was rapidly decreased with low amount of alum, just approximate 1 mg/L. However, higher concentrations of turbidity, 250 and 500 NTU required higher amount of alum respective 10 and 25 mg/L. The addition, the residual turbidity decreased after adding

chemical coagulation, which it decreased from initial concentration 50, 250, and 500 NTU to 9.2, 11.2, and 14, respectively. After convert this turbidity value to TSS, it was equal to 2.77, 13.15, and 27.67 mg/L that are lower than Thai standard was not more than 50 mg/L. Based on the theory of coagulation/flocculation process, it can be explained due to the charge neutralization mechanism for low concentration, which the enough of coagulant can attach with surface of bentonite particles (Dalvand et al., 2017b). For higher concentration, it can be removed through two mechanisms including charge neutralization and sweep floc. At first stage, the Al<sup>3+</sup> coagulant reduced a charge of colloids which carried the negative charge, and then it was then formed a large floc as the particles increase in the system which is effective removed through flocculation process (Crittenden et al., 2012).



Figure 4.2. Residual turbidity removed by Jar test using alum with different concentration 50 NTU, 250 NTU, and 500 NTU

As mentioned that pH value was allowed fluctuate naturally without any adjustment, the initial pH was presented in the previous part. The final pH was then observed as shown in Appendix II. Based on the result, the pH value of the chemical coagulation ranged between 5 to 7, which is appropriate for destabilization mechanism, 4 to 9 (Duan and Gregory, 2003). This result is similar to the existed study, which worked on the chemical coagulation for dye removal using alum as the coagulant (Engelhardt, 2010). The pH is decreased after more alum was added due to alum gave H<sup>+</sup> ion in the system and resulted acidic solution as well as decreasing the pH. For concentration 50 NTU

showed that the initial pH was approximate 6.95, then it dropped with the increasing alum dosage until pH reached about 6.6. At same dosage ranges, 250 NTU decreased from 7.65 to 6.7, while 500 NTU dropped from 6.75 to 6.58. Besides that, alkalinity is also another main parameter for chemical coagulation performance. In this study, it was just determined from the tap water characteristic in Thailand to confirm that its level for coagulation process. Thailand tap water ranges the alkalinity from 300 to 400 mg/L (Lekskulchai, 2015). After calculation, the alkalinity for tap water is enough for alum requirement (Engelhardt, 2010). Table 2 of the Appendix presented how to estimate the require pH and alkalinity for alum in coagulation process.

#### b. Decolorization

For reactive dye removal, the concentrations of alum were varied from 10 mg/L to 800 mg/L. The decolorization result of three concentrations including 25, 50, and 100 mg/L was shown in Figure 4.3. For residual dye was decreased depending on the amount of alum dosage (B Merzouk, Gourich, et al., 2011). It means that higher alum dosage was added, the rapid decreasing of the residual dye was obtained. According to Figure 4.3, the initial dye concentration of 25 mg/L (2500 ADMI), 50 mg/L (6000 ADMI), and 100 mg/L (15000 ADMI) were required the optimal alum about 60, 120, and 180 mg/L, respectively. The residual dye concentration after adding the optimal alum dose were 257, 96, and 117 ADM from the initial concentration was 2500, 6000, and 15000 ADMI, respectively. These values are lower than the discharge effluent from industry in Thailand, 300 ADMI. Considering related to the theory of coagulation/ flocculation process, it can be explained that the decolorization was followed two main mechanisms including adsorption and neutralization of the dye molecules due to dye structure was a complex form with many resin that can catch the coagulant easily than bentonite structure (X. Zhang et al., 2012). This results was similar to other previous research which removed reactive dye by using ferric chloride as coagulant. It expressed that disperse and reactive dye was follow by adsorption and charge neutralization by measuring the zeta potential to prove. Kim et al. (2004) said that if the pH of the solution is below the IEP of the respective metal hydroxide, then positively charged polymers will predominate and the adsorption of these can destabilize the negatively charged colloids through charge neutralization.



Figure 4.3. The residual of dye removed by Jar test using alum with different concentration 2500 ADMI, 6000 ADMI, and 15000 ADMI.

The final pH after chemical coagulation was decreased from 7 to 5.5 as shown in Appendix II. This pH decreasing can be explained as the following. Alum was hydrolyzed by producing  $Al^{3+}$  and  $(SO_4)^{2-}$  ions, then,  $Al^{3+}$  ion reacted with alkaline agents presented in wastewater. So the generated insoluble sediment formed as  $Al(OH)_3$ . The consumption of hydroxide ions was responded by the alkalinity reduction, therefore, the pH of the wastewater was dropped (Dalvand et al., 2017a). The alkalinity consumes in system was also checked and it was in the range of 21 mg/L to 81 mg/L.

# 4.2.2 Estimating Treatment Cost of Conventional Coagulation (CC)

In chemical coagulation process (CC), the operating cost involve the cost related to the coagulant reagents, and mixing energy for coagulation and flocculation processes. In addition, if the coagulant was presented in solid form, it was necessary to consider on the mixing power for preparing the coagulant solution as liquid phase. The velocity gradient was used to calculate the power consumption, which the mixing impeller normally presents in range of 700 to 1200 s<sup>-1</sup>(Cañizares et al., 2009). For CC operating cost using alum coagulant was divided into two conditions i.e., industry and laboratory condition as the result was summarized in Table 4.2. At lab condition, the alum was \$US 29 per kg, while the industrial scale is just about \$US 0.70 per kg (B Merzouk,

Gourich, et al., 2011) obtaining from different commercial budgets in 2007 for largescale orders. Moreover, if the alum reagent presented in liquid form, the mixing energy is not necessary. Then, the energy consumption was considered to be negligible due to the coagulant solution pumping consumed less energy. To estimate the energy cost, the electricity cost was highly depended on the country. It is the key of the operating cost using CC. For this study, the cost of electricity was followed Thailand condition, \$ US 0.11 per kWh.

Init	tial		Alum cos	st	Mixing cost			Total
concen	tration	Kg/m <sup>3</sup>	\$U.S/Kg	\$US/m <sup>3</sup>	kWh/m <sup>3</sup>	\$US/kWh	\$US/m <sup>3</sup>	\$US/m <sup>3</sup>
For turl	oidity (N	TU)	Litteres	Im Sal				
	50	0.001		0.029				0.095
Lab	250	0.010	29	0.290	0.61	0.11	0.066	0.356
	500	0.025	2/11	0.725				0.791
y	50	0.001	1/3	0.001				0.067
ıdustr	250	0.010	0.70	0.007	0.61	0.11	0.066	0.073
Г	500	0.025	A	0.018				0.084
For dye	(ADMI)		S.		X			
	2500	0.060		1.740				1.806
Lab	6000	0.120	29	3.480	0.61	0.11	0.066	3.546
	15000	0.180	II AI ONG	5.220	NIVERS	ту		5.286
ry	2500	0.060		0.042				0.108
dust	6000	0.120	0.70	0.084	0.61	0.11	0.066	0.150
In	15000	0.180		0.126				0.192

Table 4.2. Operating cost of Chemical Coagulation process in lab and industry

# 4.2.3 Summary

For CC process, the alum was selected as coagulant this study. The initial concentration of turbidity and dye pollutants were 50, 250, 500 NTU and 2500, 6000, 15000 ADMI, respectively. After adding the alum into the synthetic wastewater, the results showed that chemical coagulation process can remove turbidity approximate 82% for initial concentration 50 NTU, 95% for initial concentration 250 NTU, and 97% for initial concentration 500 NTU by using the optimal alum dose 1 mg/L, 10 mg/L, and 25 mg/L,

respectively, to lower than 20 NTU as Thailand drinking water standard of Ministry of Natural Resources and Environment (2008). Moreover, the removal efficiency of decolorization was about 88% for 2,500 ADMI, 98% for 6,000 ADMI, and 99% for 15,000 ADMI using the optimal alum dose 60, 120, and 180 mg/L, respectively. The residual dye for every studied initial concentration was less than Thailand standard, 300 ADMI. The effluent pH for turbidity and dye removal was fluctuate in the range of 5 to 7, which was very suitable for destabilization mechanism. Furthermore, the treatment cost for turbidity removal of the initial turbidity concentration 50, 250, and 500 NTU were respective \$US 0.095, \$US 0.356, and \$US 0.791 for the laboratory scale, and respective \$US 0.067, \$US 0.073, and \$US 0.084 for industrial scale. Plus, the treatment cost for decolorization of initial dye 2500, 6000, and 15000 ADMI were respective \$US 1.806, \$US 3.546, and \$US 5.286 for the laboratory scale, and respective \$US 0.108, \$US 0.150, and \$US 0.192 for industrial scale.

#### 4.3 Optimization of EC Condition

This part aimed to optimize the electrocoagulation condition including electrode configuration, electrode arrangement, and current density. However, the electrode configuration such as gap, arrangement, and current density was initially studied as the screening analysis for briefly understanding the effective behaviors in terms of generated gas flow and electrode corrosive loss ratio (Qg/Loss). It was then fully analyzed in terms of turbidity and color removal efficiency. Electrode arrangement was studied between monopolar and bipolar, while the inner gap of every electrode was varies between 1.0 and 2.5 cm. In screening analysis, current density was studied in three level including 1.5, 2.0, and 2.5 mA/cm<sup>2</sup>.

#### 4.3.1 Electrode configuration

This section explored the best condition of the electrode including inner gap and the current density in monopolar and bipolar arrangement in terms of produced gas and electrode loss ratio (Qg/Loss). All the experiments were performed with tap water for 15 minutes. The Qg/Loss was observed at 4, 9, and 14 minutes. The results of various electrode conditions in monopolar and bipolar were illustrated in Figure 4.4 and Figure 4.5, respectively. Before starting to analysis the result, the meaning of the respond

parameter should be explained. The higher ratio represented high amount of gas generated with less amount of aluminum plate corrosion as electrode life period (Nawadol et al., 2017). According to Figure 4.4, it showed that the connection in monopolar with gap 1.5 cm and current density 1.5 mA/cm<sup>2</sup> provided better performance comparing to other conditions. The inner gap 1.5 cm with current density 1.5 mA/cm<sup>2</sup> of monopolar arrangement resulted the ratio up to 2.42, 2.65, and 2.88 mL/g.s at respective 4, 9, and 14 minutes operation time. The inner gap of the electrode could affect voltage in the system as can be known that voltage was the main parameter related to power consumption. When the gap space was large, higher voltage was required. Based on the result of Figure 4.5, it showed that the bipolar connection of inner gap 2.cm with current density 1.5 mA/cm<sup>2</sup> provided the highest ratio performance comparing to other conditions. The ratio performance of the optimum condition was 2.53, 2.67, and 2.84 mL/g.s at respective 4, 9, and 14 minutes.

In conclusion, the optimum electrode configuration in terms of gas flow and electrode loss ratio (Qg/Loss) in monopolar and bipolar were gap 1.5 cm with 1.5 mA/cm<sup>2</sup> of current density, and gap 2.cm with 1.5 mA/cm<sup>2</sup> of current density, respectively.



Figure 4.4. Ratio of gas flowrate over electrode loss of monopolar connection



Figure 4.5. The ratio of gas flowrate over electrode loss of bipolar connection

## 4.3.2 Electrode arrangement

After getting the maximal condition of mono- and bipolar connection from the last part, it was the examined with synthetic wastewater of turbidity 250 NTU and color 6000 ADMI. For monopolar, inner electrode gap is 1.5 cm with current density  $1.5 \text{ mA/cm}^2$ . For bipolar, inner electrode gap is 2.0 cm with current density 1.5 mA/cm<sup>2</sup>. The results of treatment efficiency of turbidity and color were shown in Figure 4.6 and Figure 4.7, respectively. Since the bipolar connection consumed high voltage than monopolar one for providing high temperature to the system, it can promote electrode plate to generate much coagulants and gas bubble more since first stage. However, it required longer time to heat the electrode plates for producing coagulant and hydrogen gas in the system in monopolar arrangement (Demirci et al., 2015). Finally, at a stable stage, the results of treatment efficiency for both arrangements were similar. From the results of Figure 4.6 and Figure 4.7, it showed that the performance of bipolar connection was better at the first stage, around 25 to 30 minutes for both turbidity and dye removal, but the treatment efficiency for both connections were very similar at the stable stage after 30 minutes. At 35 minutes treatment time, monopolar could remove turbidity and dye up to 95 % and 89%, respectively, while bipolar could treat up to 96% and 93% for turbidity and dye, respectively. The final pH in the system for both treatments stayed in the range of 6.5 to 8.5, which are very suitable for destabilization mechanism.



Figure 4.6. The removal efficiency of turbidity by monopolar and bipolar connection with maximal condition



*Figure 4.7. The removal efficiency of reactive dye by monopolar and bipolar connection with maximal condition* 

For selecting the optimal condition of electrode arrangements, many evaluate factors should be considered such as treatment efficiency, treatment cost, and volume of treated water. It was then analyzed in this study for selecting the optimum electrode arrangement for removal turbidity and color as the results shown in Table 4.3. From the cost estimation, the bipolar required approximate \$US 0.31 to remove one kilogram of dye from 1 m<sup>3</sup> of wastewater, while \$US 0.05 was consumed by monopolar one, which detail calculation was shown in the T*able* 4.4. From this result, the arrangement in monopolar required less the treatment cost more than 6 times comparing to the one by

bipolar. Based on the previous studies (Demirci et al., 2015; Duan and Gregory, 2003; Kobya et al., 2011), the removal efficiency of three different electrode connections including monopolar as parallel, monopolar as series, and bipolar was not notable different at the steady stage. However, the treatment costs were vastly different due to the high voltage for connections of bipolar than the monopolar as parallel one. Moreover, monopolar operation produced more volume of treated water as well as producing less sludge about 6% comparing to bipolar arrangement. In conclusion, both mono- and bipolar arrangement resulted in a similar performance in terms of treatment efficiency at certain electrolyze time; however, monopolar provided better performance in terms of the operation cost and treated water as the economic selection.

Electrode arrangement	Bipolar	Monopolar
Treatment cost (\$US/kg of dye)	0.31	0.05
Treated volume out of 4L ( <i>litter</i> )	3.35	3.54

Table 4.3. Treatment cost and treated water between monopolar and bipolar

In summary, the electrode arrangement in monopolar of aluminum plate with the internal gap 1.5 cm and  $1.5 \text{ mA/cm}^2$  of current density was selected as the economize condition for this study and design criteria for treating turbidity and color. Although the current density  $1.5 \text{ mA/cm}^2$  with gap 1.5 cm by monopolar connection was found as the optimal condition, it was just considered as the screening analysis in limited ranges and response variable. Therefore, the effect current density in the wide ranges in terms of treatment efficiency was studied.

## 4.3.3 Current density

Current density plays an important role in EC process. Higher current density provided to the system, much dissolve aluminum from electrode plates as coagulants with more gas bubble affected the growth floc was obtained (Chen, 2004; Mollah, Morkovsky, et al., 2004b). It can improve from lag stage to stabilizing stage much faster, however, it is also mainly related to the power consumption. In this study, it was then varied in 5 levels, 1.5, 2.5, 3.5, 4.5, and 5.5 mA/cm<sup>2</sup> for treating turbidity and color. The result of

turbidity removal was presented in *Figure* 4.8. It showed that the stabilizing stages of each current density were obtained at 50, 35, 30, 30, and 20 minutes for the current density 1.5, 2.5, 3.5, 4.5, and 5.5 mA/cm<sup>2</sup>, respectively. Based on the *Figure* 4.8 showed that the turbidity removal didn't change much at the first stage about 5 minutes of some current density including 1.5, 2.5, 3.5, and 4.5 mA/cm<sup>2</sup>, but the current density 5.5 mA/cm<sup>2</sup> was improve removal efficiency about double comparing to the smallest current density 1.5 mA/cm<sup>2</sup>. However, the current density 5.5 mA/cm<sup>2</sup> was not selected as the optimal condition since after 20 minutes, the treatment efficiency optimization and economization, current density 3.5 and 4.5 mA/cm<sup>2</sup> was selected. Not only turbidity removal was considered to select the current density, but the decolorization also studied by varying current density as the same condition of turbidity. Then, the current density was selected after got the optimal current density from each treatment efficiency condition of turbidity and color.



Figure 4.8. The different current density for turbidity removal of EC process



Figure 4.9. The different current density for dye removal of EC process

*Figure* 4.9 presented the result of color removal kinetic in various current densities. The result showed that the destabilizing stage of current density was more than 60, 60, 50, 30, and 20 minutes from the supplied current density of respective 1.5, 2.5, 3.5, 4.5, and 5.5 mA/cm<sup>2</sup>. At lag stage about 15 minutes, the decolorization of current density  $1.5 \text{ mA/cm}^2$  was double comparing to current density  $5.5 \text{ mA/cm}^2$ . The current density  $4.5 \text{ mA/cm}^2$  selected for optimal in terms of economic as well as its treatment efficiency was much closer to the current density  $5.5 \text{ mA/cm}^2$  from the lag stage until the stable stage. From both results, it can conclude that the current density of  $4.5 \text{ mA/cm}^2$  was the optimal condition and the operation time of steady stage was 30 minutes for both turbidity and color removal.

# 4.4 Study the Kinetic Treatment in EC

After completing the optimization of EC condition for removal turbidity and color from synthetic wastewater, the optimal condition including monopolar arrangement with inner gap 1.5 cm and current density 4.5 mA/cm<sup>2</sup> was obtained. The EC was applied to treat both turbidity and color about 30 minutes and let it settled down for 30 minutes in order to compare with chemical coagulation.

## 4.4.1 Effect of initial concentration

The result in *Figure* 4.10 showed that EC with current density 4.5 mA/cm<sup>2</sup> could treat turbidity to be lower than Thailand standard, which allowed to discharge about 20 NTU. The residual turbidity were 13.35, 11.20, and 11.80 NTU from 50, 250, and 500 NTU, respectively. After settle 30 minutes, the residual turbidity kept decreasing until 6.61, 5.60, and 4.34 NTU. It can prove that EC process needs time to settle after having mixing by a gas bubble with coagulant in the system. Moreover, the pH in the system increased along the time because of the hydrogen evolution. It was a fact that pH was very important in the coagulation process (Ching et al., 1994). The initial pH value was in range between 7.1 to 7.7 and the final pH stayed in the range of 8 to 8.2 show in Appendix III. Based on previous study, Abuzaid et al. (1998) raised that coagulation with aluminum ion occurred with many mechanisms at pH in the sweep floc coagulation mechanism. Therefore, the final pH increased in an electrochemical cell due to hydrogen evolution is advantageous from coagulation point-of-view.



Figure 4.10. The residual turbidity removed by EC process with different concentration 50 NTU, 250 NTU, and 500 NTU

Since dye as color pollutant was difficult to remove, it required more time for molecule or colloid to attach with coagulant in ordering to precipitate, as the theory of flocculation called sweep flocculation mechanism (Crittenden et al., 2012). Then, the precipitate captured by the Al(OH)<sub>3</sub> or polymeric aluminum hydroxide flocs and floated into a gel state (Bazrafshan et al., 2016). Figure 4.11 expressed that the residual dye after the EC process at optimal time, about 30 minutes, decreased to 56, 6.8, and 435 ADMI from the initial concentration 2500, 6000, and 15000 ADMI, respectively. After keeping it settles down 30 minutes, the residual dye still decreased to 34, 23, and 283 ADMI, which was lower than Thailand standard for discharging, about 300 ADMI. The initial pH in system stayed in the range of 7.1 to 7.3. The pH increased along the time until the final one ranging from 8.1 to 8.5 show in Appendix III, which was the proper pH for destabilization mechanisms. This result was similar to the previous researches reporting that the majority of textile wastewater collected from equalization tanks had pH in the range of 5.0 to 10.0, which was the optimum operation range in this present experiment (Lin and Peng, 1994).



Figure 4.11. The residual dye removed by EC process with different concentration 2500 ADMI, 6000 ADMI, and 15000 ADMI

The effect of different initial concentrations on the treatment performance was kinetically studied in this part. The initial bentonite and dye concentrations were examined in range of 50 to 500 NTU, and 2500 to 15000 ADMI, respectively. The results were shown in *Figure* 4.12. The result in *Figure* 4.12 (a) showed that the lag stage of the low concentration on turbidity removal was larger than the high concentration. The initial concentration of 50 NTU needed electrolysis time about 25

minutes to dissolve the aluminum ion into a system in order to improve the lag stage until reaching the stable stage, which provided removal efficiency around 74.67%. For 250 NTU and 500 NTU was required retention time only 20 minutes with removal efficiency 95.52 and 97.70%, respectively. Form *Figure* 4.12 (b) expressed that the low concentration of dye consumed less electrolysis time and provided higher treatment efficiency than the high concentration at the lag stage. Moreover, results were surprising at stable stage because the removal efficiency of three initial concentrations were very similar,  $98 \pm 1\%$  at stable state of 20 minutes.



Figure 4.12. The different of initial bentonite concentration show in (a) and dye (RY 135) concentration show in (b) on removal efficiency again related with operation time

Based on results from Figure 4.13 (a), it showed that the treatment performance increased with the initial concentration of bentonite. It can prove that the initial concentration of bentonite was much different from 50 NTU to 250 NTU because the adsorption capacity of flocs was not exhausted and the rate of turbidity removal was constant. Based on the Figure 4.12 (a) showed that the turbidity removal at the reactive stage around 10 minutes were similar, but the removal started hugely different from each other after stable stage. The additional, the coagulant will become the pollutants after the more aluminum ion was generated from the electrode plate, while particles has a little amount that can cause the coagulant was excess. However, it was nearly stable from concentration 250 NTU to 500 NTU due to the ability of flocs become exhausted and the amount of coagulant was proportion with amount of pollutants. For the results in Figure 4.13 (b), it expressed that the initial concentration of reactive dye from 2500

ADMI to 15000 ADMI were similar of treatment efficiency above 90%, but there was the optimal point at concentration 6000 ADMI. After check the significant effect, it can say that the increase of initial dye concentration was not statistical significant effect on decolorization capacity at the steady state due to the adsorption of flocs was exhausted. This result was similar with other previous research of decolorization by EC process (Kobya et al., 2011).



Figure 4.13. The different of initial bentonite concentration show in (a) and dye (RY 135) concentration show in (b) on removal efficiency with optimal  $j = 4.5 \text{ mA/cm}^2$  at operation time 30 min.

#### 4.4.2 Model prediction

This section focused on the kinetic order and kinetic constant rate (k) in the batch column reactor with initial turbidity and color concentration about 250 NTU and 6000 ADMI, respectively. For determine the simple kinetic of turbidity and dye removal, it was separated into two parts. For turbidity removal was followed the s-curve or sigmoid model because the removal efficiency had 3 phase including lag, reactive stage, and stable stage. At the lag stage, turbidity removal did not change much (Mongkolnauwarat, 2014). The nonlinear regression of sigmoid equation was expressed by equation 4.1, where Y is output, X is input,  $X_{50}$  is half of the input, and k is sigmoid steepness.

$$Y = \frac{100}{1 + e^{-k(x - x_{50})}}$$
 Eq. 4.1

The treatment efficiency of S-curve can derive as the equation 4.2. Where  $\eta$  is removal efficiency,  $t_{50}$  is the time of treatment at 50%.

$$\eta = \frac{\eta_{stable}}{1 + e^{-k(t - t_{s_0})}}$$
Eq. 4.2

From the results of *Figure* 4.14, it showed that the turbidity removal with current density of 1.5, 2.5, 3.5, 4.5, and 5.5 mA/cm<sup>2</sup> was almost fit with the s-curve followed the kinetic constant of respective 0.19, 0.30, 0.38, 0.45, and 0.54 and t<sub>50</sub> of respective 15.85, 12.27, 10.12, 9.20, and 6.98 minutes, respectively. The correlation between current density, kinetic constant rate, and t<sub>50</sub> was investigated as shown in Figure 4.15. It showed the correlation of current density with kinetic constant (k) as k = 0.085j + 0.0745, where j is the current density (mA/cm<sup>2</sup>) and k is the kinetic rate. Moreover, the other correlation between the current density with time and removal efficiency reached 50% (t<sub>50</sub>) can be expressed as  $t_{50} = -2.081j + 18.168$  where j is the current density (mA/cm<sup>2</sup>) and t<sub>50</sub> is the time that removal efficiency reached 50% (minute).

For kinetic of dye removal, the simple model including zero, first, and second order was applied. The results obtained to plot the concentration of dye [dye] along the time as zero-order, ln[dye] with operation time as first-order, and 1/ln[dye] with operation time as second order (Nwabanne and Obi, 2017). The results in *Figure* 4.16 proved that the second order was closer or fitter than zero- and first-order. The R-square in second order was higher than 0.9, which was higher than zero- and first-order was follow by equation 0.6, respectively. Therefore, the simple kinetic of second order was follow by equation

 $\frac{1}{C(t)} = \frac{1}{C_0} + k_2 t$ . The constant rates were 4.10<sup>-5</sup>, 4.10<sup>-5</sup>, 6.10<sup>-5</sup>, 7.10<sup>-5</sup>, and 8.10<sup>-5</sup> (1/mole

\* minutes or M<sup>-1</sup>. min<sup>-1</sup>) of the current density 1.5, 2.5, 3.5, 4.5, and 5.5 mA/cm<sup>2</sup>, respectively. After that, the correlation between current density with constant rate was constructed as k = 1.1j + 1.95, where j is the current density (mA/cm<sup>2</sup>) and k is the constant rate as shown in Figure 4.17.



Figure 4.14. The simple kinetic model follow by s-curve with initial turbidity concentration 250 NTU and different current density



*Figure 4.15. Correlation between current density with kinetic rate and t<sub>50</sub> for turbidity removal with initial concentration 250 NTU* 



Figure 4.16. Scatter plot for the rate constant in zero, first, and second-order with initial dye concentration 6000 ADMI and various current density



Figure 4.17. Correlation between current density with the kinetic constant rate for decolorization with initial concentration 6000 ADMI

## 4.4.3 Settling test

The settling test studied in a batch column containing 4 litters with total height of 33 cm. The four sampling ports was prepared for analysis in terms of turbidity removal, where the first, second, third, and fourth port were about 13, 18, 23, and 28 cm from the top of water surface as shown in Figure 4.18. In addition, the settling test was studied in three conditions including (i) bentonite removal without chemical, (ii) bentonite removal with chemical by EC process, and the third was dye removal with the chemical. For bentonite and dye chemical, the settling test studied after operated EC process with optimal time at 30 min. of initial concentration 250 NTU and 6000 ADMI, respectively. The setting test studied with total time 200 min. and 30 min. for without chemical and chemical, respectively. The sampling time measured every 3 min.



Figure 4.18. Sampling port for settling test in batch column reactor

The steps of calculation followed by Reynolds (1977). The results of settling test shown in Figure 4.19, which plotted the overflow rate with removal efficiency. The results have shown that the bentonite without chemical was low treatment efficiency comparing to others after EC process, which removal efficiency decreased from 70% to 24.7%, while the overflow was increased from 0.1 m/hr. to 1.36 m/hr. shown in Figure 4.19 (a). For bentonite after EC process, the removal efficiency was decreased from 90% to 25%, while the overflow was increased from 1.89 m/hr. to 12.92 m/hr. shown in Figure 4.19 (b), and for dye, the treatment efficiency was reduced 90% to 25.8

%, while the overflow increased from 1.54 m/hr. to 11.06 m/hr. shown in Figure 4.19 (c). We can observe that higher overflow provided lower treatment efficiency (Saret, 2015). Moreover, the results have shown that 2.1 m/hr. and 1.7 m/hr. of OFR obtained for standardly lower separating of turbidity and dye, respectively. This result was going to use as the design criteria of the separation process.



*Figure 4.19. The fraction removal with overflow in batch column reactor, which (a) bentonite without chemical, (b) bentonite after EC process, (c) dye after EC process* 

# 4.4.4 Cost estimation

As mentioned, the treatment cost through electrocoagulation process is mainly involved with material cost (electrodes), utility cost (electrical energy), as well as labor, and maintenance costs. For the main cost was considered on material cost and electrical energy cost, which are taken into account in various calculation steps. The electrodissolution of aluminum was estimated by the theory of Faraday's Law was much different from the experiment. So that, the correction ratio of an electro dissolution yield,  $\phi$  (%) occurred was more than 100% in all cases depending on the total aluminum concentration added.

For calculation of operating cost, the power consumption was determined by electric current, voltage, and time in the system and the electrode consumption obtained from the experimental. For cost of the electrode estimated from the previous research is \$US 1.80 (Ozyonar and Karagozoglu, 2011), and cost of the electrical energy in Thailand is \$US 0.11. After calculation, the results show that turbidity removal cost \$U.S 0.128, 0.129, and 0.129 respect the initial concentration 50, 250, and 500 NTU. For color removal, it cost \$U.S 0.129, 0.136, and 0.149, respect initial concentration 2500, 600, and 15000 ADMI. According to the results showed that the treatment cost of turbidity and color didn't change much. The summary of treatment cost by EC process shown in Table 4.4.

Initial concentration		Electrodes cost		Power consumption cost			Total	
		10 <sup>-3</sup> * Kg/m <sup>3</sup>	\$U.S/Kg	10 <sup>-3</sup> * \$US/m <sup>3</sup>	kWh/m <sup>3</sup>	\$US/kWh	\$US/m <sup>3</sup>	\$US/m <sup>3</sup>
For tur	bidity (N	TU)	Q	NN N MM	e e			
Σ	50	0.320		0.576	1.173		0.128	0.128
ıdustı	250	0.327	1.80	0.589	1.182	0.11	0.129	0.129
II	500	0.311		0.560	1.216		0.132	0.132
For dye (ADMI)								
ry	2500	0.332		0.598	1.171		0.127	0.129
dust	6000	0.343	1.80	0.617	1.234	0.11	0.136	0.136
In	15000	0.334		0.601	1.359		0.148	0.149

Table 4.4. The operating cost of Chemical Coagulation process in lab and industry

# 4.4.5 Comparative analysis between EC and CC

There are many factors should be considered to compare between EC with CC such as treatment efficiency, cost of treatment, operation time, and sludge production. In this section, it focused on treatment efficiency and operating cost. From the previous results of both CC and EC, the comparative analysis was summarized in Figure 4.20 and Figure 4.21 for respective turbidity and dye removal.

Figure 4.20 showed the result after EC operation with optimal electrolysis time, about 30 minutes and current density 4.5 mA/cm<sup>2</sup> before letting it settle 30 minutes in order to compare with CC. The results expressed that it could remove turbidity from the initial concentrations 50, 250, and 500 NTU about 87.47, 97.76, and 99.15 % by utilizing \$US 0.128, 0.129, and 0.132 per kg of turbidity in 1 m<sup>3</sup>, respectively. On the other hand, CC process could remove turbidity about 81.62, 95.36, and 97.20% from respective initial concentrations as mentioned by utilizing \$U.S 0.067, 0.073, and 0.084, respectively. At the optimal point, the treatment efficiency of turbidity by EC was higher than CC even the treatment cost of CC was lower. This results was comparable to previous work of (Ubale and Salkar, 2017). If considering on the cost of treatment as Figure 4.20, it can be found that the treatment of cost of EC was not much different between the low and high concentration, however, the treatment cost was liner increasing in CC process, which was effect in operation with higher concentration.



Figure 4.20. The removal efficiency with operating cost of conventional coagulation (CC) and electrocoagulation (EC) process of turbidity removal

Figure 4.21 expressed that the capability of EC on dye removal from the initial concentration 2500, 6000, and 15000 ADMI were 98.75%, 99.16%, and 98.01%, with treatment cost of \$US 0.129, .0.136, and 0.148, respectively. For CC process could remove dye from the mentioned initial concentration about 88.26, 98.15, and 99.21 % consuming the treatment cost about \$US 0.108, 0.150, and 0.192, respectively. Based on the results, it can be observed that EC process was better than CC at the low and

medium concentration of reactive dye, but high concentration presented that CC was better performance than EC since the capacity of adsorption of floc was exhausted. Moreover, the operating cost was liner increasing from the low to high concentration of dye by using CC process. In contrast, the treatment cost of dye by EC was not much change comparing CC process.

In conclusion, the EC process was more suitable for turbidity and dye removal from textile industry wastewater with economic aspect in terms of operation time, treatment efficiency, and operating cost for higher concentration than CC process.



Figure 4.21. The removal efficiency with operating cost of conventional coagulation (CC) and electrocoagulation (EC) process of decolorization

# 4.4.6 Summary

In summary, the optimum condition of the electrode was selected an inner electrode gap 1.5 cm and 2.0 cm with current density 1.5 mA/cm<sup>2</sup> for the respective monopolar and bipolar arrangement in terms electrode loss. The selected optimal condition from the last part for monopolar and bipolar connection was examined for turbidity and color removal. The result showed that the monopolar connection was chosen over bipolar in terms of operating cost and volume of treated water, which monopolar arrangement was higher than the 6 times and 6%, respectively, comparing to bipolar. Then, the effect of current density was studied with the optimal configuration of inner electrode gap 1.5 cm of monopolar arrangement. The results expressed that current density of 4.5

 $mA/cm^2$  was obtained as the optimal condition due to the removal efficiency of this current density was similar to the highest current density at the stable stage up to 90% for both turbidity and dye removal with electrolysis time about 20 and 30 minutes, respectively. After obtaining the optimal current density, the turbidity and dye removal was studied with different concentration. The results showed that after the optimal configuration and condition of EC process was applied, it can achieve 13.35, 11.20, and 11.80 NTU from 50, 250, and 500 NT U of turbidity removal with a hydraulic retention time (HRT) 30 minutes, and reach 6.61, 5.60, and 4.34 NTU after 30 minutes settle down. Moreover, dye removal was also operated with HRT 30 minutes and it could achieved 56, 34, and 435 ADMI from the initial concentration 2500, 6000, and 15000 ADMI, respectively. After let it settle for 30 minutes, the residual color was reached 34, 23, and 283 ADMI. The effect of initial concentration for both turbidity and dye was examined and it showed that there was an effect on turbidity removal efficiency at the low concentration of 50 NTU to 250 NTU, but it was not significant effect at a concentration from 250 NTU to 500 NTU. For dye removal, it was not significant effect on the removal efficiency even the initial concentration was increased since the ability of adsorption floc was exhausted. The kinetic study for bentonite and reactive dye with different current density was then analyzed. It presented that the bentonite followed the sigmoid or s-curve and reactive dye followed Pseudo-second-order model. The settling test was also studied, the optimum overflow of bentonite and dye were 2.1 m/hr and 1.7 m/hr, respectively. The operating cost of EC for turbidity and dye removal with different concentration showed that \$US 0.128, 0.129, and 0.132 were required to removed respective initial turbidity concentration 50, 250, and 500 NTU, while \$US 0.129, 0.136, and 0.148 required to remove initial dye concentration 2500, 6000, and 15000 ADMI, respectively. The comparative analysis between EC and CC was therefore studied. The result expressed that the EC process was selected over the CC process in terms of economic aspect as operation time, operating cost, treatment efficiency.

After getting all parameters in batch column reactor study such as electrode configuration, optimal current density, kinetic constant rate and order, HRT, OFR, removal efficiency, and operating cost, the design and evaluate the new

electrocoagulation reactor (ECR) in continue mode was performed. ECR was combined two processes of electrocoagulation and separation process for treating turbidity and dye from the textile industry wastewater.

# 4.5 Design of ECR for Turbidity and Color Removal

This section aimed to design and construct the new reactor, ECR for treating synthetic turbidity and color from textile industry, following the common design criteria of Kawamura (2000). There are two parts of this reactor, electrocoagulation and separation section which shown in Figure 3.3. The sedimentation tank was firstly calculated, then, the EC tank was designed after basic dimensions were used.

The step of calculation was shown below with the criteria in the T*a*ble 4.5 from the design book of Kawamura (2000). Since the overflow of settling performance from the previous part had two conditions for turbidity and dye, 2.1 m/hr and 1.7 m/hr, respectively. The 1.7 m/hr of dye contaminant resulting with removal efficiency 95% from the Figure 4.19 was selected because it can surly separate the both pollutants to the desired level. Another parameter was hydraulic retention time (HRT) or electrolysis time from EC in the batch column was 20 and 30 minutes for turbidity and dye removal, respectively. Therefore, 30 minutes was chosen for the design part of EC with the same reason of selecting OFR for sedimentation design. The flow rate was selected at 3 L/min in order to prevent the accumulated sludge in the EC port.

Parameters	Units	Range
Mean flow velocity	m/min	< 1.1
Detention time	hr	< 4.0
Surface loading	m/hr	1.25 - 2.5
Length/width ratio (L/W)	Minimum	4:1
Water depth/ length ratio (H/L)	Minimum	1:15

Table 4.5. The design criteria of Rectangular Sedimentation Tank (Kawamura, 2000)

Surface Area (A) = 
$$\frac{Q}{OFR} = \frac{3L/\min}{1.7 m/hr}$$
 = 0.11 m<sup>2</sup>

Since trial-and-error was used for varying the width (W), length (L), and height (H), the reaction dimension was obtained and check for the design criteria as Table 4.5,

Propose: 
$$W = 0.16 \text{ m}, L = 0.7 \text{ m}, \text{ and } H = 0.3 \text{ m}, \text{ Volume } (V) = 34 \text{ L}$$

Checking,

*Length* / *width* ratio = 
$$\frac{7}{1.6}$$
 = 4.4:1 > 4:1 **Ok!**

*Water depth / length ratio* =  $\frac{3}{7}$  = 1:2.3 > 1:15 **Ok!** 

Detention time 
$$(\tau) = \frac{34(L)}{3 \times 60(L/hr)} = 0.2 hr < 4.0 hr.$$
 Ok!

Mean velocity (v) = 
$$\frac{3 \times 10^{-3} (m^3 / \min.)}{0.16 \times 0.3 (m^2)} = 0.0625 \ m / \min. < 1.1 \ m / min \ Ok!$$

Re = 
$$\frac{v \times R_h}{V}$$
 = 73.70 < 2000 Ok!

Since the propose size of sedimentation tank was checked, and it passed the criteria. So that the W ~ 0.16 m and L ~ 0.7 m were designed and L ~ 0.3 m was considered for flocculation tank before entering the sedimentation tank. Therefore, the total L is 1.0 m with H ~ 0.30 m is the water depth from the water level to the middle of the tank and then plus freeboard 0.05m, as well as the slope at the bottom for sludge collection. The detail design and dimension with as front view in Figure 4.22. After designing the size of separation tank with the total length, L ~ 1.0 m, and H ~ 0.35 m plus freeboard 0.05 m. Then, the EC port was started to design, which followed the equation below.

 $Volume = Q \times \theta = 3L / \min \times 30 \min = 90L$ 

$$A = \frac{Volume}{L} = \frac{90 L}{10 dm} = 9 dm^2 \qquad \qquad W = \frac{A}{H} = \frac{9 dm^2}{3.5 dm} = 2.6 dm$$


Figure 4.22. The detail design and size of sedimentation tank with dimensions

Therefore, the total length of EC is 1.0 m, with width 0.26 m, and height 0.35 m plus freeboard 0.05 m. The detail design with dimensions of electrocoagulation tank was showed in Figure 4.23 and the solid purple line in the figure is the baffles and purple of dashed line to black line about 100 mm was the entry of the wastewater from EC to flocculation tank. After finishing the calculation, the volume of the electrocoagulation tank is 90 litters, plus flocculation tank is 17 litters, and sedimentation tank about 34 litters, while the total of the ECR is 141 litters. The completed reactor with their functions was shown in Figure 4.24.



Front View of EC tank

Figure 4.23. The detail design and size of electrocoagulation tank with dimensions



Figure 4.24. Design and constructed of ECR reactor a) in perspective view of plan and b) pilot reactor in lab

After constructing the novel ECR, the liquid flow pattern in the reactor was then studied in order to understand the liquid flow in the reactor such as plug flow reactor (PFR) or CSRT. The residence time distribution (RTD) was expressed as E(t) function, mean time ( $\tau$ ), and axial dispersion.

## 4.6 Liquid Flow Pattern in ECR

In this present work, the residence time distribution (RTD) experimental method was chosen for evaluating the flow pattern of the new designed ECR reactor. All experimental results represented as the E(t) curve called the exit-age distribution, which was defined in Eq. 3.9. The characteristic of E(t) basically indicates the type of flow pattern model of the particular reactor. For example, piston flow reactor (PFR), perfect mixing (CSTR), or non-ideal flow model. The detail of characteristic of reactors with

tracer shown in Figure 3.8. The mean residence time  $(t_m)$  of the experimental was calculated from E(t) curve, following Eq. 3.10. The mean residence time  $(t_m)$  was compared with the theoretical of liquid residence time ( $\tau$  = volume/liquid flow rate). This comparison can consider the actual problem within the particular reactor in term of mixing characteristic, included the liquid bypass or/and existed dead zone of the system. Moreover, the momentum variance ( $\sigma^2$ ) and the axial dispersion were also evaluated, these two parameters can indicate the spread of the distribution in the reactor. It means that the greater variance was, the distribution of molecules spread well (Fogler, 2010). The effect of various operating conditions on mixing behavior inside the system was studied, such as the position of detector' probe, the present of baffles in reactor, the presence or absence of DC supply, and the introducing of liquid flow.

## 4.6.1 Effect of probe position

To diagnose the mixing behavior at different positions inside reactor, four probe of conductivity meters were placed at 0.6, 1.2, 1.8, 2.4 decimeter respectively from the bottom of the reactor as shown in Figure 4.25. The experiment was operated with a constant liquid flow rate equal to 6 L/min. It was two times higher than the designed value, to reduce time operation and to avoid a problem of detection of conductivity value. Moreover, the conductivity of tracer in water stayed in ranged 0.00 - 2.00 mS/cm, by mixing 180 g of NaCl salt in 0.6L of tap water, which was rapidly injected to the system at inlet point. The conductivity value of each probes were recorded along the time, then converted to E(t) curve as following.



Front View of EC tank

*Figure 4.25. Different positions of conductivity meter probe at outlet of EC process to sedimentation tank* 



Figure 4.26. The E (t) curve of probe position at 0.6, 1.2, 1.8, and 2.4 dm from the bottom surface with flowrate 6 L/min

Figure 4.26 represented the plots of the exit-age distribution E(t) curve of different probe position. As the result, the started detection time of the probes were different, the lowest position was spent shortest time before detection. The duration time that the tracer traveled to firstly attach the probe were 2, 4, 8 and 14 min at different height position of 0.6, 1.2, 1.8, and 2.4 dm respectively. Due to, the different density between tracer (NaCl) and water, the injected tracer originally refers to the bottom of water and needs a period of time for dispersing to the top. It means that the different of probe position can make the mixing were dissimilar depending on the height of probe. The highest probe position, the longer detection time was.

Probe position [dm]	Mean time distribution (τ) [min.]
Theory	16.36
0.6	15.70
1.2	13.16
1.8	16.16
2.4	19.65

*Table 4.6. The mean time distribution*  $(\tau)$  *with different positions* 

Moreover, the mean residence time at each position were calculated and summarized in Table 4.6. After analyses, the correlation between probes positions with mean time distribution by statistic has shown that P-value was 0.277, which was higher than 0.05 (it was a confidential level about 95%). Base on this results, it was not a significant effect. Therefore, the different position of probe height was not affected to the mean residence time, although it can affect the mixing. So that, the height of probe position 0.6 and 1.8 cm were considered for study due to their results were similar to mean time of theoretical.

#### 4.6.2 Effect of EC compartment baffles

In this section, the effect of various aspect of presented baffles inside the system on flow mixing characteristic of reactor, were examined. Included, the number of baffle which was changed with 0, 1, 2 and 3 baffles, and the different length of baffle which was ranged 50%, 70%, and 90% which respectively corresponded to the total length of 1.3, 1.82 and 2.34 decimeters.



(a) Top View of EC tank (3 baffles with 50% length)







(c) Top View of EC tank (1 baffles with 90% length)

Figure 4.27. The total length of baffles and number of baffles in the ECR rector including 0, 1, 2, and 3 baffles with varies length 50%, 70%, and 90%.

All experiment operated same condition with liquid flow rate 6 L/min, and the concentration of tracer were recorded at both the outlet point of EC and the effluent of the ECR reactor. The total experiment was 40 units were performed with different location and length of baffles as shown in Figure 4.27.

Due to, there were many operating condition, it was easy to show only the plots of the special condition of the exit age E(t). Figure 4.28 shown that there were some fluctuated results, due to the detector probe was placed with wide open space at the outlet of the EC tank. E(t) function of the single baffle with different lengths (Figure 4.28 (a)) performed not much different for both characteristic and mixing behavior in the reactor. Figure 4.28 (b) shown the result of E(t) curve after adding one more baffle in the system. As the results, there was a bit change from the previous result of Figure 4.28(a). Event, the condition of the 90% - 90% baffle was significantly changed, the other two configurations were no significantly changed.



Figure 4.28. The *E*(*t*) curve of different baffle configuration with flow rate 6 L/min at outlet of EC tank a) single baffle with different length, b) homogenous of 2 baffles with different length, c) homogenous of 3 baffles with different length, d) alternate of 3 baffle with different length

Figure 4.28 (c) shown the results in case of presented three baffles at the same length. The result was similar to the result in Figure 4.28 (b). It was importantly noted that there was only the configuration 90% - 90% - 90% of baffle performed in different. It seems help to reduce the liquid bypass inside the system. Figure 4.28 (d) shown the result of E(t) curve, while the introduce of different lengths within three baffles. The characteristic curve fluctuated due to the effect of random baffles and also operated as the open system. It means that the mixing behavior of baffles has different three lengths was not equal in the reactor or sometime the tracer can come back. However, when we observed all curve in Figure 4.28 show that baffles can reduce bypass or short-cut in the system depending on the amount and length. Moreover, all curve shown that the reactor tended to plugged flow combine with completely mixing reactor (CSTR) (Levenspiel, 1999).

Based on the Figure 4.28 (a) shown that E(t) at the outlet of EC with one baffle of different length was similarity of the peak curve value which increased the signal around 2 - 7 min, 4 - 8 min, 6 - 11 min, and 12 - 20 min ordering with no baffle, one baffle with 50%, one baffle with 70%, and one baffle with 90 %, and then started to decreased until zero at the time about 50 min. Due to the baffle length of 90% was affected by the flow rate which made the tracer need longer time than others baffles length. The Figure 4.28 (b) shown that E(t) of homogenous 2 baffles increased the signal around 2 - 7 min., 5 - 8 min., 6 - 8 min., and 11 - 14 min., respective no baffle, one baffle with 50%, one baffle with 70%, and one baffle with 90 %, and then started to decreased until zero at the time about 50 min. The Figure 4.28 (c) and Figure 4.28 (d) can describe followed by the Figure 4.28 (a) and The Figure 4.28 (b).

In short, after put the baffle in the reactor shown that the probe detection needs time to detect the tracer exit longer than non-baffle and can help to reduce short-cut in the system. Moreover, the baffle length 90% was the effect a little or much depending on the baffle configuration.



Figure 4.29. The mean time distribution of 40 experiments at the outlet of EC tank

In addition, flow pattern can express as with the RTD experiment data could also use to determine the average time that substance resided inside the reactor. The comparison between mean residence time and design residence time could as well imply the flow behavior in the reactor. The design and experiment mean residence time and molecules spread shown in Figure 4.29 and Figure 4.30. From Figure 4.29 show that the mean time from the experiment was a little bit higher than the design time for theory. In those, the mean time who has baffled length with 90% was higher than other which is provided wastewater stayed a long time in the reactor and can minimize the short-cut in the rector well. Moreover, the dispersion in the reactor also calculates by covariance which relates meantime and variance shown in Figure 4.30. The value of covariance nearly 0 was the plug flow reactor and the value was approximately 1 was classified as the mixed reactor (Fogler, 2010). Base on the results in Figure 4.35 expressed that the value of dispersion was in the range 0.25 - 0.55 which was nearly 0 so that the ECR with and without was a plug flow behavior.



*Figure 4.30. The axial dispersion number in reactor of 40 experiments at outlet of EC* The effluent of reactor also studies to make sure the mechanism of flow in the reactor while we vary the baffle condition same as the outlet of EC tank, which plotted the E(t) curve, meantime, and dispersion.

Figure 4.31 shown that the aspect of E(t) at effluent of ECR was trended to the plug flow reactor model, which has smooth curve than the outlet of EC tank. Due to the whole process has a large volume and at the sedimentation tank was not obstacles that can effect flow rate, so that it can minimize the error of the result. The additional, at the effluent of ECR, was measured as the closed system. From the Figure 4.31 (a) shown that it was not a significant effect, even though the single baffle was added. For the Figure 4.31 (b) expressed that there has some change only baffle 90% - 90%, but the baffles 50% - 50% and 70% - 70% were not changed and similar results for the Figure 4.31 (c). Based on these results show that the 90% can help to reduce bypass in the reactor. For Figure 4.31 (d) were not clearly seen the difference between three baffles.



Figure 4.31. The E (t) curve of different baffle configuration with flowrate 6 L/min at effluent of ECR redactor a) single baffle with different length, b) homogenous of 2 baffles with different length, c) homogenous of 3 baffles with different length, d) alternate of 3 baffle with different length

Figure 4.32 shown that most baffle has meantime was higher than the non-baffle due to the baffle can help to keep the wastewater stay long in the reactor, especially for baffle length 90%. Therefore, we can conclude that having baffle can help reduced bypass and the dead zone in the ECR reactor. The dispersion inside the reactor shown in Figure 4.33 expressed that the co-variance for all experiments stayed in the range 0.2 - 0.55, which was nearly 0 and smaller than 1. So these results helped to prove the previous results stronger that this new reactor has plug flow behavior although having the baffle.

In conclusion, the presence of the baffle was increased the flow pattern behavior by reducing the short-cut or bypass in the reactor, which promoted the wastewater stay in reactor longer depending on the conditions of a baffle that we selected, but it didn't change the reactor behavior, finally, it was the plug flow reactor. This type was preferred use now for the idea of reactor design.



Figure 4.32. The mean time distribution of 40 experiments at effluent of ECR reactor



Figure 4.33. The axial dispersion number in reactor of 40 experiments at effluent of ECR reactor

After study the baffles configuration, it cannot say any baffles was better than others. It just known that the ECR was plug flow behavior, but the selecting started after the criteria was set. Before selecting the prefer baffles, some reasons was shown as below: - The easiness design: for real application in industry or other sectors, the easiness installation and design was preferred from the industry owner. So that the homogenous baffles was selected including (50%, 50% - 50%, 50%-50%-50%, 70%, 70%, 70%, 70%, 70%, 90%, 90%, 90%, and 90% - 90% - 90%). So that, the alternated baffle design was not select due to it was a complicated design.

- The comparison between 1 baffle (50%, 70%, and 90%) Vs 2 baffles (50% - 50%, 70% - 70%, and 90% - 90%) Vs 3 baffles (50% - 50% - 50%, 70% - 70% - 70%, and 90% - 90% - 90%): Base on the results from the Figure 4.29 and Figure 4.32 shown that the mean time distribution of 3 baffles with length 90% was higher than others which can reduced volume of dead zone and can promoted the wastewater to stayed in reactor longer for EC to treated it. The 3 baffles were 90% - 90% - 90% which has meantime about 25.36 min. and 24.79 min. at the outlet of EC and effluent of ECR reactor which was similar results with the optimal time from batch column reactor about 30 min. Moreover, the 3 baffles with 90% of baffle length can help reduce bypass from the non-baffle and others baffles.

- The detail of 3baffles with different lengths of liquid flow mechanisms shown in Figure 4.34 below:

• The baffle length 50% - 50% - 50%

Since Figure 4.34and Figure 4.32shown that the 50% shown that the 50% length of baffle was 18.47 min which was higher than non-baffle about 17.18 min. at the outlet of EC tank and 21.43 min. of 3 baffles with 50% length was higher than 19.89 min. of non-baffles at the effluent of ECR reactor Base on this results, we can be indicated that the 50% length was no effect on mean time ( $\tau$ ). Moreover, we can prove that the 50% length of baffle was not affected by the pathway of wastewater flow rate. The flow mechanisms of 50% length were expressed in Figure 4.34 (a).

• The baffle length 70% - 70% - 70%

It was a similar result from the baffle length. 50%. The mean time distribution of 3 baffles with 70% length was 18.94 min. was higher than non-baffle only 17.18 min. at

the outlet of EC, and the 21.77 min. of 3 baffles of 70% length was higher than nonbaffle only 19.89 min. at the effluent of ECR reactor. Therefore, the 3 baffles of 70% were not affected to wastewater flow pathways because the mean time of 70% length, 50% length, and non-baffle was close together. The flow mechanism of 70% length was shown in Figure 4.34 (b).

• The baffle length 90% - 90% - 90%

From the results of Figure 4.34and Figure 4.32 shown that the 90% of baffles has meantime distribution about 25.36 min. and 17.18 for non-baffles at the outlet of EC tank and then 24.79 min. of 3 baffles with 90% was higher than the non-baffle only 19.89 min. at the effluent of ECR reactor. Base on this results shown that the baffles length 90% has mean time higher than non-baffle, 70%, and 50% of baffles length. When the observation of mean time distribution carefully, it was shown that the baffles length of 90% was much higher than non-baffle about (5 -10 min) at both outlets of EC tank and effluent of ECR reactor. To sum up, the 90% of the baffle length was effect the flow pathway of wastewater by increasing the velocity which make flow was turbulent. The flow mechanism was shown in Figure 4.34 (c). According to the 3 criteria selections was shown that the 3 baffles with 90% of baffles length (90% - 90%) were chosen for future study.

#### 4.6.3 Effect of operation condition with and without EC

In this section, three baffles with same length of 90 % selected for evaluating effect of the operation with and without EC process on liquid flow behavior. While, the DC was supplied, it generated the small bubble from the cathode side and coagulant at the anode side. The concentration of the tracer were measured at both outlet of EC and effluent of ECR as shown in Figure 4.35.



(a) Top View of EC tank with 3 baffles and 50% lengths



(b) Top View of EC tank with 3 baffles and 70% lengths



(c) Top View of EC tank with 3 baffles and 90% lengths

Figure 4.34. The flow mechanism in EC tank with 3 baffles lengths



Top View of ECR with detail of probes position

Figure 4.35. The detail of probe position of 90% - 90% - 90% baffle configuration for the whole process



Figure 4.36. The E (t) curve of 3 baffles with length 90% - 90% - 90 by operated EC and without operated EC with flowrate 6 L/min at outlet of EC

Figure 4.36 represented the characteristic of E (t) curve was trended to the plug-flow model in case of the presence or absence the DC supply. It means even EC sets were installed, the behavior of reactor didn't change. However, it importantly noted that the tracer was detected faster after putting the EC sets which are 4 min to peak around 10 min, then decrease to lowest value after 45 min. In case, the absence of EC sets, the

conductivity meter firstly detected at 10 min to the peak about 23 min. after operation, then decrease to the lowest level after 45 min. as well. In addition, the operation with the EC sets could reduce bypass in reactor. It mean the presented bubble can enhance the mixing in the system compared to without EC sets. Table 4.7 shown that the mean time distribution of having EC sets were less than without EC sets about 1.3 times, it means that wastewater can stay in reactor shorter when having EC sets.

The main reason which made EC sets inside reactor reduces the meantime has 2 reasons including the theory of geometry and the mixing by a gas bubble. According to Gadd et al. (2010) proved that bauble generation from EC process can be enhancing the mixing inside the reactor so that the mean time could help to reduce the meantime. Another reason was a principle of oblique triangular (has angle  $90^{\circ}$ ) shown that hypotenuse was longer than other two lengths. The flow pathway of 90% - 90% - 90% baffles length without EC was similar to the hypotenuse of oblique triangular, it means that it was the highest length for flow to travel. In contrasted, when installed the EC sets in the reactor, it was not an obstacle to disturbed the flow pattern, but the bubble can help the flow of wastewater travel faster than without EC. Nevertheless, this results cannot apply to the other two monogamous baffles condition (50% - 50% - 50% and 70% - 70% - 70%) due to the flow pathway of these baffles configuration were straight line of a little bit curve not the hypotenuse which is the shortest or shorter pathway than 90% baffles length one. When having the mixing of the bubble in the middle of a pathway from EC process, it was an obstacle which can delay the meantime in the system. So that, the mean time of having EC sets was higher than the without EC sets one. The graphics and meantime will show in appendix IV.

Baffles condition (90% -90% - 90%)	Mean time (min.)
Mean time of Theory	15.00
With EC (w/EC) sets	19.39
Without EC (w/o EC) sets	25.36

*Table 4.7. Mean time distribution*  $(\tau)$  *with and without EC at EC outlet* 

From the result of Figure 4.37 shown that it was not much different on the E (t) curve for both condition with and without EC sets. Nevertheless, with EC sets also detected tracer concentration faster little than without EC sets. Base on T*a*ble 4.8 shown that the mean time from both was close to theory together, but the with EC sets has meantime lower than without EC about 1.03 times.

The validation of these results started with other four places of probe position ( $P_1$ ,  $P_2$ ,  $P_3$ , and  $P_0$ ) which pinpoint on the Figure 4.35. For  $P_1$ ,  $P_2$ , and  $P_3$  expressed as meantime ( $\tau$ ), but the  $P_0$  was a location to check the bypass or dead zone in the reactor.



Figure 4.37. The E (t) curve of 3 baffles with length 90% - 90% - 90 by operated EC and without operated EC with flowrate 6 L/min at ECR reactor

Table 4.8. The mean time distribution ( $\tau$ ) of having EC sets and without EC set at the effluent of ECR reactor

Baffles condition (90% -90% - 90%)	Mean time (min.)
Mean time of Theory	23.67
With EC (w/EC) sets	24.05
Without EC (w/o EC) sets	24.79

From the results in Table 4.9 shown that the mean time distribution from the experiment was higher than theory for both with and without EC sets. All positions of having EC

sets has meantime lower than without EC sets about 2.18, 1.47, and 3.02 min., respective of  $P_1$ ,  $P_2$ , and  $P_3$ . After validating this results, it can prove that having EC sets can improve the short-cut and dead zone in the reactor by the gas bubble which generated by EC process. Moreover, the last position was  $P_0$  which checked the residual conductivity or not after operated with EC process.

Table 4.9. The mean time distribution ( $\tau$ ) of having EC sets and without EC set at the  $P_1$ ,  $P_2$ , and  $P_3$  in the EC tank

	Mean time (min)					
Parameters	With EC sets (W/ EC)			Without EC sets (W/OEC)		
	<b>P</b> <sub>1</sub>	P <sub>2</sub>	P <sub>3</sub>	<b>P</b> <sub>1</sub>	P <sub>2</sub>	P 3
Mean time of Theory	3.75	7.50	11.25	3.75	7.50	11.25
Mean time of experiments	8.89	11.39	14.35	11.07	12.83	17.36



Figure 4.38. The conductivity value of  $P_3$ ,  $P_0$ , and outlet of EC tank with baffle length 90% of flow 6 L/min.

Base on the results from Figure 4.38 shown that the conductivity value of  $P_3$ ,  $P_0$ , and outlet of EC tank along the time. From Figure 4.38 (a) expressed that at 30 min. of without EC at  $P_0$  was higher than  $P_3$  and outlet of EC tank, which was a signal of having a dead zone, but form 90 min. until close operation, the signal was a similar value that expressed the signal of the non-dead zone. The addition, Figure 4.38 (b) shown that all

points of  $P_0$  from the having EC sets was higher than  $P_3$  and outlet of EC tank. Therefore, these results pointed that non-dead zone of point  $P_0$ .

### 4.6.4 Effect of Liquid Flow rate in ECR

Due to the presence of EC performed better than without EC process, then the effect of different liquid flow rate was studied. In this section, the flow rate ranged 1, 3 and 6 L/min were investigated. Figure 4.39 shown that the flow rate of 6 L/min. performed higher peak than case 3 and 1 L/min. about 2 and 4 times, respectively.



Figure 4.39. The effect of flowrate on liquid flow in range 1 L/min, 3 L/min., and 6 L/min. with baffles length of 90% (90% - 90% - 90%) operated by EC process

Figure 4.39 (a) shown that at flow 6L/min., half of the tracer was left from the reactor around 8 min. at the outlet of EC tank and around 15 min. from the effluent of ECR reactor (Figure 4.39 (b)). Moreover, at the flow rate 1 L/min. some of the tracer leave from the reactor less than 80 min and around 30 min. at flow rate 3 L/min. at the outlet of EC tank see in the Figure 4.39 (a). The tracer leave from reactor around 85 min at flow rate 1 L/min. and spend about 35 min. at flow rate 3 L/min. The flow rate of 3 L/min. was selected due to the optimal time from the batch column about 30 min. and from the Figure 4.39 expressed that it can reduce the bypass about 2 times from the 6 L/min.

In addition, the mean time ( $\tau$ ) of flow 1 L/min was 19.39 and 24.05 min. at the outlet of EC and effluent of ECR reactor was less than the design time about 30 mins at the EC tank and 47.33 min as shown in T*a*ble 4.10 for the whole process. Furthermore, the small flowrate about 1L/min. need much time for the liquid to stay in the reactor about 102.61 and 115.98 min. respective with at the outlet of EC tank and the effluent of ECR reactor which is much higher than design. For the flow, 3L/min with the best configuration of baffles was 41.17 min. which was higher than the design need time only 30 min. at the outlet of EC tank and 47.82 min. was similar to the design of the whole process about 47.33 min.

Table 4.10. The mean time distribution ( $\tau$ ) of different flowrate with 1 L/min., 3 L/min., and 6 L/min. at the outlet of EC tank and the effluent of ECR reactor

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D	At outlet of EC tank			At effluent of ECR reactor		
Parameters	6 L/min.	3 L/min.	1 L/min.	6 L/min.	3 L/min.	1 L/min.
Mean time of experiments	19.39	41.17	102.61	24.05	47.82	115.98

#### 4.6.5 Summary

In summary, the probe position with different height was effected on the curve behavior due to the mixing in the reactor, but it was not effected on the meantime distribution ( $\tau$ ) after checking the statistic of P-value shown that it was not significantly affected. For position 6 and 18 cm were detect mean time closer to the theory one. The baffle configuration of 3 baffles with length 90% (90% - 90% - 90%) was selected based on the criteria demanded including mechanism, easiness design, and reduce bypass or dead zone. After that, EC was operated and then comparing to without EC process. The results have shown that bubble which generated from EC process can help reduce bypass in reactor comparing to the without EC operation. From the best baffle configuration shown that flowrate 3 L/min. was optimal and fit with the design.

The next section focuses on the treatment efficiency in the continue system with the single of reactive dye and bentonite, then study the combine contaminates together after getting the optimal configuration and condition.

#### 4.7 Treatment Performance of ECR as Continuous Mode

In this section aimed to evaluate the treatment performance in ECR reactor as continue operation for decolorization and turbidity removal. The treatment performance of ECR was analyzed in systematic process. As it can be known that color is difficult to remove by coagulation process comparing to turbidity pollutant, then, the treatment of color was firstly examined through the variation of liquid flow. Liquid flow used this study was designed from electrolyze time of the kinetic study. Secondly, it will be studied through varying the current density for color removal as it is the most important factor of EC application. The optimum condition of color removal. Finally, it was considered the treatment performance for turbidity removal. Finally, it was considered the treatment condition for simultaneous removal of turbidity and color.

#### 4.7.1 Effect of flow rate on decolorization

For decolorization in continue operation of ECR with the optimal condition of baffle configuration was used to treat the initial concentration of dye 50 mg/L, equal 6000 ADMI. The optimal current density of 4.5 mA/cm<sup>2</sup> was used for a single set of EC. From the liquid flow analysis, 3 baffles (4 ports) with all 90% length (90% - 90% - 90%) with the flow rate 3 L/min was used to treat the color. It should be noted that every port was installed 1 set of EC. As be known that flow rate is one of the important parameters in continue reactor. Higher flow rate resulted less hydraulic retention time. However, if low flow rate was very low, higher liquid retention time was obtained as well as treatment operation. Therefore, the flow rate in this part was varied from 1, 2, and 3 L/min.



*Figure 4.40. Percentage of decolorization and residual dye along the time with different flow 1 L/min., 2 L/min., and 3 L/min. with current density 4.5 mA/cm<sup>2</sup>* 

Based on Figure 4.40, it showed that at early stage of treatment for every flow rate was fluctuate due to the initial volume that just filled in the reactor was left along the time with 142, 71, and 47 minutes from the design of the flow rate 1, 2, and 3 L/min, respectively. For flow rate 1 L/min, it required about 120 minutes operation time for letting the initial concentration of reactive dye reach the effluent of the tank, while flow 2 and 3 L/min required about 70 and 45 minutes, respectively. At the steady stage, the removal efficiency for dye removal were 78, 68, and 52% for the liquid flow rate 1, 2, 3 L/min, respectively. Dye concentration of every studied flow decreased along the time. At flow rate 3 L/min, it showed that the residual dye was 2800 to 2850 ADMI at the steady stead, while it was 1850 to 1900 ADMI for the flow 2 L/min and 1550 to 1600 ADMI for 1 L/min. Moreover, the initial pH in the system kept in range of 7.20 to 7.40 and slightly increase up to 7.40 to 7.60 as the final pH. These pH range was suitable for destabilize and sweep flocculent mechanism.

Figure 4.41 presented the summary of the effect of liquid flow rate on the color treatment. The removal efficiency was decreased after increasing the flow rate. It can be explained related to the resident time, higher flow rate resulted short residence time. In contrast, shorter flowrate can give higher retention time in the reactor that can provide a chance for untreated wastewater to stay long in reactor to attach the coagulant from electrode cell. The slower flow rate allowed the coagulant generated by EC at anode side and gas bubble at the cathode side, had enough chance for the dye molecules

to mix properly as well as enhance the rate of coagulation. Therefore, the flow rate 1 L/min. was selected for decolorization. This result was similar to other research (Mollah, Pathak, et al., 2004)



Figure 4.41.The effect of flowrate on reactive dye 135 removal with current density 4.5 mA/cm<sup>2</sup>

Even though the removal reached 78 %, the effluent also is still higher than standard. Therefore, the increasing of current density was considered in order to obtain better treatment efficiency.

## 4.7.2 Effect of current density on decolorization

The current density was played an important role for EC process. Since the optimal current density from the batch column was about 4.5 mA/cm<sup>2</sup>, it may inefficient after the operation of EC in ECR due to the operation volume different between batch column and ECR. For 1 port of ECR, only 1 EC set was installed due to the limitation of DC supply. Actually, the demand from the design required 2 sets per port.

In this section, the current density was increased 2 and 3 times with the liquid flow 1 L/minutes. The current density ranged at 4.5, 9.0, and 13.5 mA/cm<sup>2</sup> for the initial reactive dye concentration 6000 ADMI. Based on the Figure 4.42 (a) showed that the removal efficiency was significantly increased at current density 9.0 mA/cm<sup>2</sup> and 13.5 mA/cm<sup>2</sup>. At the current density, 4.5 mA/cm<sup>2</sup> could remove dye about 78% with the

residence time about 120 minutes, while the current density 9.0 mA/cm<sup>2</sup> and 13.5 mA/cm<sup>2</sup> could treat dye up to 95% (100 minutes) and 98% (70 minutes), respectively. Besides that, the Figure 4.42 (b) described that the current density 4.5 mA/cm<sup>2</sup> cannot reach the Thailand standard (300 ADMI), but at the same flow rate of 1 L/min, the current density 9.0 mA/cm<sup>2</sup> and 13.5 mA/cm<sup>2</sup> can remove dye to low than standard about 268, and 100 ADMI, respectively. Moreover, the initial pH in the system stayed in range 7.4 – 7.6 and after the experiment, it was 7.7 – 7.9, which is good for destabilization mechanism.



Figure 4.42. Percentage of decolorization and residual dye along the time with different current density 4.5 mA/cm<sup>2</sup>, 9.0 mA/cm<sup>2</sup>, and 13.5 mA/cm<sup>2</sup> with flow 1 L/min

Figure 4.43 illustrated that the removal efficiency of yellow reactive dye 135 depended on the current density, higher current density resulted better removal performance. At the same time, the electrode loss at the anode side also increased depending on the current density. According to the Faraday's equation, higher currents provided a significant amount of aluminum ions, which may trap the dye molecules and enhance the removal efficiency (Pletcher and Walsh, 2012). Besides that, when applied to high current density to the system it was increased a little or the treatment efficiency can decrease due to the reverse of coagulant. So that, the current density should be limited in order to avoid excessive oxygen evolution as well as to eliminate another adverse effect, like heat generation, but in this research, the current density 13.5 mA/ cm<sup>2</sup> was selected in order to discharge safety with standard allowable. The next section was validated the best condition with bentonite to prove that the best condition for decolorization was suitable for turbidity removal.



Figure 4.43.The effect of current density on reactive dye 135 removal with flowrate 1 L/min. and initial dye concentration 6000 ADMI

## 4.7.3 Validation turbidity treatment

Bentonite was used as turbidity in order to validate the best condition from the decolorization from the previous section. So that, the flowrate 1L/min with current density 13.5 mA/cm<sup>2</sup> of each set was used to treat the initial concentration of turbidity 250 NTU. The results of residual also compare with the Thailand standard about 20 NTU for the maximal of drinking water.

Base on Figure 4.44 described that the removal efficiency was increasing along the time and it can reach the 90 % removal at 30 min. until the stable point at 60 min. can treated contaminant about 97.12 %. Moreover, the residuals turbidity started to decrease from 250 NTU to the 7.04 NTU (TSS = 1.05 mg/L) at the steady stead, which was less than the Thailand standard allowable. Therefore, the best condition from the decolorization can treat the turbidity with initial concentration 250 NTU. Although the residual was less than the standard, the removal efficiency started decreased at the 130 min. upward. The addition, we observed that the sludge was high at the first 2 ports of reactors at EC process about 17.5 cm and 15.0 cm, respectively. Due to the high of sludge blanket at the first 2 comportments has sludge very high, which was effected to

the treatment efficiency and sludge collection problems when we want to recirculate the sludge. Therefore, the effect of flow rate on turbidity removal was studied.



*Figure 4.44. Percentage of turbidity removal and residual turbidity along the time with optimal condition of current density 13.5 mA/cm<sup>2</sup> with flow 1 L/min* 

## 4.7.4 Effect of flow rate on turbidity removal

After validating with turbidity removal by the optimal condition as flow rate 1 L/min and current density 13.5 mA/cm<sup>2</sup>, the study of the effective flow rate on turbidity removal was examined as considering on the accumulated sludge blanket at 1 L/min flow. The different flow rates were varied in range of 1, 2, and 3 L/min at the same current density of 13.5 mA/cm<sup>2</sup>.

From the Figure 4.45 showed that the removal efficiency of turbidity was similar for every studied flowrate. At 60 minutes, the removal efficiency reached 97.19, 96.18, and 94.46% as well as the residual turbidity 7.04, 9.55, and 13.85 NTU and TSS was equaled 1.05, 4.59, and 26.90 mg/L of the respective flowrate 1, 2, and 3 L/min. However, the removal efficiency reached up to get 95.32, 96.74, and 97.01% with residual turbidity of 11.65, 8.14, and 7.24 NTU and TSS was equaled 15.48, 2.05, and 1.32 mg/L, respectively, after 150 minutes. Based on this result, it showed that the treatment efficiency was comparable with 90% removal efficiency evening the flow rate was increasing. However, the accumulated sludge was decreased after higher flow

rate was supplied. Table 4.11 showed the visible sludge blanket at first and second port of EC compartment. It was reduced from 17.5 cm (port #1) and 15 cm (port #2) to 10 cm (port #1) and 12.5 cm (port #2) after increasing from the respective flowrate of 1 to 2 L/min, then, keep decreasing to 5 cm (port #1) and 10 cm (port #2) after increasing to 3L/min of the flowrate. In addition, when the flowrate increased from 1 to 2 and to 3 L/min, the treatment cost was increased from \$US 1.515 to 1.409, and to 1.344, respectively.

In conclusion, the amount of flow rate was not much influent on the turbidity removal. However, higher flow could reduce the amount of the accumulate sludge blanket at first and second port of EC tank. Therefore, if there is only turbidity pollutant was presented, the optimum flowrate 3 L/min was selected in terms of sludge collection ease and treatment cost. After studying each pollutant from the synthetic wastewater of reactive dye 135 as color and bentonite as turbidity, the combination of both contaminates together was necessary to study in order to optimize the treatment condition as well as evaluate the treatment performance.



*Figure 4.45. Percentage of turbidity removal and residual turbidity along the time with different flowrate of current density 13.5 mA/cm<sup>2</sup>* 

Flowmata	Sludge blank	et height (cm)	Cost of treatment	% R	
(L/min.)	Port 1 of EC tank	Port 2 of EC tank	(\$U.S)		
1	17.5	15	3.745	95.32	
2	10	12.5	3.488	96.74	
3	5	10	3.327	97.1	

*Table 4.11. The effect of different flowrate on removal efficiency, sludge blanket height, and treatment cost with current density 13.5 mA/cm<sup>2</sup> at 150 min.* 

## 4.7.5 Simultaneous removal of turbidity and color

The initial color concentration about 6000 ADMI was successfully treated with current density 13.5 mA/cm<sup>2</sup> and flow rate 1 L/min, while turbidity concentration about 250 NTU required the optimum flow rate 3 L/min. In order to select the operate condition for simultaneous removal of turbidity and color, it required to evaluate both different conditions with the current density 13.5 mA/cm<sup>2</sup> at flowrate between 1 to 3 L/min.



Figure 4.46. The removal efficiency and residual contaminants mixing between reactive dye with bentonite as turbidity and color with flowrate 3 L/min. and current density 13.5 mA/cm<sup>2</sup>

Flowrate	Sludge blanket height (cm)		<b>Removal efficiency (%)</b>	
(L/min)	Port 1 of EC tank	Port 2 of EC tank	Color	Turbidity
2	12.25	12.25	96.45	94.32
3	7.5	7.5	87.34	90.16

*Table 4.12. The effect of removal efficiency between liquid flowrate and sludge blanket height at 150 min.* 

Figure 4.46 showed that the flow rate 3 L/min could remove the simultaneous turbidity and color just about 90% with 80 minutes operation time as in the steady stage. In specific, color can be treated about 90.12%, it is about 90.36% for turbidity with the residual color 593 ADMI and 24.1 NTU. The experiment was operated until 150 minutes as the residual dye and turbidity were higher than standard level. Therefore, the flowrate 3 L/min with current density 13.5 mA/cm<sup>2</sup> may not appropriate for treating wastewater, which contained both contaminants simultaneously.

From this result, it can be observed that the operation with flowrate 3 L/min, the removal efficiency of turbidity was lower than the one with only single contaminant, specifically, about 1.08 time because the dye molecule adsorbed on the aluminum ion for creating the slurry sludge, which is another additional turbidity. Moreover, it was not enough coagulant and retention time for dye and bentonite attachment in order to settle down. As the result at 150 minutes, the treatment performance decreased for both color and turbidity after the steady stage. According to Romphophak et al. (2016), the turbidity and dye removal is mainly depended on two parameters i.e., flow rate and sludge blanket height. The flowrate and accumulated sludge blanket were related to each other as higher flowrate may minimize the accumulated sludge as well as lower flowrate may result higher sludge accumulation. That work also mentioned that the height of the sludge blanket was significant effect on the turbidity removal. As the cumulative sludge volume would be recirculated to the flocculation zone, it may increase the contact probability between the particles and enhance the agglomeration of destabilized particles for forming larger solid flocs (Kawamura, 2000; Sotirakou et al., 1999). In contrast, if the height of sludge blanket was too high, it reversed to decrease the removal efficiency. Therefore, the flow rate 2 L/min was selected to evaluate the treatment performance as operating condition.



Figure 4.47. The removal efficiency and residual mixing between reactive dye with bentonite as turbidity and color with flowrate 2 L/min. and current density 13.5 mA/cm<sup>2</sup>

Based on the Figure 4.47, the removal efficiency was reached to 90% for both turbidity and dye after 60 minutes operation as the stable stage. At 150 minutes, the dye removal reaches 96.45% with the residual color approximate 213 ADMI, which was lower than the standard level (300 ADMI), and 94.32% for turbidity removal with residual turbidity 14.2 NTU equal to TSS about 28.71 mg/L, which is lower than the standard as well (50 mg/L). It can be explained that after decreasing the liquid flow to 2 L/min, the removal efficiency was increased due to the higher hydraulic retention time was allowed for coagulant and particles attached to each other to form floc for settle down. In addition, the height of sludge blanket also increased after decreasing the flow rate to 2 L/min, which it may the sludge from bentonite and benefit for the treatment performance (Romphophak et al., 2016). However, as mentioned, higher sludge may not good for treatment and collection as well. The optimal sludge should be considered. The results of treatment efficiency with flowrate and sludge blanket showed in Table 4.12. Therefore the flow rate 2 L/min with current density 13.5 mA/cm<sup>2</sup> was selected as the optimal operation condition in terms of simultaneous removal efficiency and sludge production.

## 4.7.6 Summary

In conclusion, the effect of flow rate on color treatment was studied in the range of 1, 2, and 3 L/min with current density 4.5 mA/cm<sup>2</sup> and initial color concentration 6000 ADMI. The results showed that flow rate 1 L/min provided better performance as decolorization efficiency up to 78 %. Furthermore, the current density was studied in range of 4.5, 9.0, and 13.5 mA/cm<sup>2</sup> with 1 L/min of flowrate for decolorization. As the result, current density 13.5 mA/cm<sup>2</sup> resulted much better treatment performance comparing to other current density, about 98% removal efficiency. After that, the turbidity removal was used to validate the optimum condition with current density 13.5 mA/cm<sup>2</sup> and flow rate 1 L/min. The results showed that it also can treated the turbidity but it seem too much sludge blanket at the first and second port of EC tank, which may inefficient for sludge collection and may result on the sludge recirculate. The flowrate of 1, 2, and 3 L/min were studied for turbidity removal in the expected of reducing the sludge blanket in the first two EC ports. Since the treatment efficiency was similar together, above 90%, the flowrate 3 L/min was selected in order to reduce sludge blanket and treatment cost. Since flow 3 L/min could successfully treat the turbidity, hence, it was used to treat turbidity and color simultaneous. The result showed that this flow rate cannot treat both co-existing contaminants lower than the standard requirement. Decreasing flowrate to 2 L/min was considered to improve the treatment performance to be lower than the standard level. The result of flow rate 2 L/min resulted better efficiency than flowrate 3 L/min with the residual turbidity and dye were lower than Thailand standard level.

## 4.8 Settling Performance through Tube Settler Addition

The tube settler was considered to study in this section in order to evaluate as well as to enhance the settling performance of turbidity and color in the sedimentation compartment of ECR. Three different part was divided including the design of tube settler based on theory criteria, followed by study the effect of tube settler sets, and the last, study the effect of the position of tube settler sets.



Figure 4.48. The mechanism in the incline tube settler of turbidity and dye by flotation and sedimentation

The separation mechanism in tube settler of turbidity and dye was simply explained in Figure 4.48.

## 4.8.1 Design tube settler

The tube settler was calculated based on the design criteria of Kawamura (2000). The important parameter such as flowrate of wastewater, overflow of particles, angle of tube settler, and velocity of liquid in tube were used to identify the size of tube settler through trial-and-error method. The design criteria expressed in T*a*ble 4.13. The step of the calculation was provided in the following.

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## Table 4.13. The design criteria of Rectangular Sedimentation Tank

Parameters	Units	Range
Mean flow velocity	m/min	< 1.1
Detention time	hr	< 4.0
Surface loading	m/hr	1.25 - 2.5
Length/width ratio (L/W)	Minimum	4:1
Water depth/ length ratio (H/L)	Minimum	1:15

For the velocity of floc settle down was write as equation:

$$u = \frac{Q \times W}{A(H \times \cos \theta + W \times \cos^2 \theta)}$$

Since trial-and-error method was used for varying the width (W), length (L), height (H) and angle ( $\theta$ ) from the equation above in order to keep the criteria in range of T*a*ble 4.13.

Propose: 
$$W = 1,8 \text{ cm}, H = 7 \text{ cm}, L = 30 \text{ cm}, \theta = 45^{\circ}$$

Checking:

$$\frac{Q}{A} = \frac{(H \times \cos\theta + W \times \cos^2\theta) \times u}{W} = 55.25 \, dm/hr = 5.5 \, \text{m/hr} \qquad \text{OK!}$$
Since Q = 2 L/min, g = 9.81m/s<sup>2</sup>  

$$\Rightarrow A = \frac{120 L/hr}{55.25 \, dm/hr} = 2.17 \, \text{dm}^2$$

$$Q = A \times v \times \sin\theta \Rightarrow v = 78.1 \, dm/hr = 0.13 \, m/\min. < 0.15 \, m/\min. \qquad \text{OK!}$$

$$\Rightarrow R_h = \frac{W}{4} = 0.045 \, dm = 0.0045 \, m$$

$$\text{Re} = \frac{v \times R_h}{V} = 10.9 < 50 \qquad \text{OK!}$$

$$Fr = \frac{v^2}{g \times R_h} = 1.07 \times 10^{-4} > 10^{-5} \qquad \text{OK!}$$

 $HRT = 4.6 \min .> 4.0 \min .$  OK!

## 4.8.2 Effect of tube settler quantity

In this section, effect of tube settler was divided into 2 parts. Firstly, one module of tube settler was studied, followed by 2 modules of tube settler. For both conditions were studied in the same position (P1), which is about 8 cm from the water surface to the

tube settle and the sampling position (P1) was about 4 cm from water surface. The configuration of tube settler show in appendix VI. Then, the settling performance was analyzed and comparing to the uninstalling condition.

#### b. Single module

The first module of tube settler was presented its dimension of W ~ 1,8 cm, H ~ 7 cm, and L ~ 30 cm, for operating with initial concentration of turbidity and color about 250 NTU and 6000 ADMI, respectively. The result of one module (1M) tube settler at position P1 was shown in Figure 4.49. The result showed that the removal efficiency of turbidity and color were increased along the time and it can reached 93.78%, and 95.54% at 150 minutes, respectively, or presenting in residual concentration, turbidity and dye were decreased from 250 NTU to 14.2 NTU and 6000 ADMI to 213 ADMI, which are lower than Thailand standard.



Figure 4.49. The removal efficiency and residual mixing between reactive dye with bentonite as turbidity and color with flowrate 2 L/min. and current density 13.5 mA/cm<sup>2</sup> with tube settler 1 module and position 1

The comparison between without and with tube settler in one module and one position condition was shown in Figure 4.50, which can be expressed that it was similar results for turbidity removal due to the hydraulic retention time of 1 module not much effect for particles to aggregated each other. Moreover, the dye removal was not improved by tube settler due to the entrapped mechanism cannot occur and the dye solution was soluble in water, which presented as the small particles and difficult to remove by tube settler. Therefore, the removal efficiency of turbidity and dye before and after installing the tube settler with one module at position P1 condition was not improved the settling performance.



Figure 4.50. The comparison between Non tube settler with tube settler 1 module at position 1 in terms of color and turbidity removal efficiency

# c. Two modules \_\_\_\_\_ALONGKORN \_\_\_\_\_YERSITY

In this part, the size of tube settler was increased 2 times in length, which can be presented in the form of W ~ 1.8 cm, H ~ 7 cm, and L ~ 60 cm. The initial concentration of mixing contaminants was turbidity 250 NTU and 6000 ADMI. The residual and removal efficiency of contaminants were shown in Figure 4.51. The results expressed that the removal efficiency of turbidity and color were respective 94.66 and 96.53% at 150 minutes with residual turbidity and dye were 13.35 NTU and 208 ADMI. The residual of turbidity consider as TSS and dye was lower than Thailand standard, 50 mg/L (around 20 NTU) and 300 ADMI, respectively.



Figure 4.51. The removal efficiency and residual mixing between reactive dye with bentonite as turbidity and color with flowrate 2 L/min. and current density 13.5 mA/cm<sup>2</sup> with tube settler 2 module and position 1



Figure 4.52. The comparison between Non tube settler, with tube settler 1 modules at position 1, and with tube settler 2 modules at position 1 in terms of color and turbidity removal efficiency
Based on the results, Figure 4.52 showed that the removal efficiency of turbidity can be increased about 15 - 18 % at 25 - 30 minutes due to the hydraulic retention time (HRT) may enough to settle down as the floc particles can be aggregated together which was easy to form the floc. The turbidity removal was presented in similar results, about 90% at 60 minutes of the existed study (Chu and Ng, 2000). On the other hand, the color removal was similar to results of tube settler uninstallation, 1 module of the tube at position P1, and 2 module of position P1. It can be explained that dye particles were too small, which were difficult to remove. Therefore, the 2 modules of tube settler at position P1 was improved for turbidity removal at first 25 to 30 minutes, however, results of all conditions were very similar after 60 minutes operation, about 90 %. For dye removal, the 2 modules installation at position P1 were not affected by tube settler.

#### 4.8.3 Effect of position installation

The variation of the position of tube settler was then studied in this part. The position P1 was studied as last part where placed 8 cm from water surface and sampling point at 4 cm from surface water, while position P2 placed about 15 cm from the water surface with sampling was 11 cm from the water surface. The configuration of tube settler was shown in appendix VI.



Figure 4.53. The removal efficiency and residual mixing between reactive dye with bentonite as turbidity and color with flowrate 2 L/min. and current density 13.5 mA/cm<sup>2</sup> with tube settler 2 module and position 2



Figure 4.54. The comparison between Non tube settler, with tube settler 2 modules (2M) at position 1 (P1), and with tube settler 2 modules (2M) at position 2 (P2) in terms of color and turbidity removal efficiency

The initial concentration of turbidity and dye were 250 NTU and 6000 ADMI operating along the time with 2 modules of tube settler. Figure 4.53 presented that at 150 minutes, the removal efficiency of turbidity and dye were 94.02% and 95.46%, respectively. The residual turbidity was 14.95 NTU and the residual dye was 272 ADMI.

The comparison between without tube settler installation, 2 modules (2M) at position P1, and 2 modules (2M) at position P2. The results of Figure 4.54 expressed that the removal efficiency of turbidity at the position P2 was similar comparing to the one without the tube settling as well as similar to the one at position P1. In this situation, it can be described that the liquid velocity at position P2 was higher than position P1 since the position P2 installed at lower, and the time for traveling of the floc to the turbe was faster than P1, then, it may effect to the turbidity. In addition, the result of the turbidity at P2 was similar to no tube settler installation. Therefore, the velocity of both conditions may similar to each other for all three mentioned conditions. It means that the dye particles was difficult to remove by tube settler. Therefore, the tube settler of 2 modules (2M) did not improve the separation performance for both turbidity and dye. In conclusion, the results of turbidity were similar to the regular condition without tube settler, and was lower removal efficiency than P1 at first 50 minutes, but the results of dye removal were similar for every condition.

#### 4.8.4 Summary

The tube settler was studied with 2 main parts including amount of modules, and position of tube settler. The results showed that after adding one module (1M) of tube settler at the position P1, the removal efficiency of turbidity and dye was not improved. It may due to the HRT was not enough for floc to aggregate. Moreover, one more module of tube settler was added (2M) at the same position (P1) in order to increase the HRT in the system. The results showed that it increased the removal efficiency of turbidity but not effect on color removal. Moreover, the position of the tube settler was studied. Two positions with the same modules of two (2M) was studied. The results expressed that the varying position of the tube settler could not enhance the removal efficiency. In summary, one and two modules at both positions did not enhance the separation performance in sedimentation tank of ECR.

#### 4.8.5 Design criteria

Actually, this part is not only cover the settling performance section, summary all the result of this study was presented here. After completely analysis the results of both batch and continuous system of eletrocoagulation process for developing the new reactor (ECR) in the aimed to treat turbidity and color textile wastewater, the design criteria was summarily provided in T*a*ble 4.14

Parameters	Description				
Electrode configuration	• Monopolar connection was consume low voltage then bipolar connection at the first 40 min.				
	• Inner gap of electrode will be effect of the voltage in the system.				
	• Treatment cost of monopolar connection was cheaper and had ability to produce the volume of treated water more than bipolar.				
	• Type of electrode plate will be considered depending on the pollutants				
	• Optimal configuration of this study: aluminum plate with gap 1.5 cm and monopolar connection.				

Table 4.14. The design criteria of batch and continue parameters with the description

Current density (j)	<ul> <li>Current density plays important roles in EC process due to it can generated the coagulants and gas bubble in the system.</li> <li>More current density consumed high voltage and produced much sludge.</li> <li>The optimal current density of the system was 4.5 mA/cm<sup>2</sup> and 13.5 mA/cm<sup>2</sup> of batch and continuous system, respectively.</li> </ul>			
Kinetic	<ul> <li>The simple model (zero, first, and second order) was applied to the turbidity removal with current density as the main parameter. This model was not fit, but it was fitted with S-curve model.</li> <li>For dye removal was apply the simple model which current density as the main part. The second order was fit among zero and first order.</li> </ul>			
Hydraulic retention time (HRT)	<ul> <li>The longer of electrolysis time of EC process can provide the chance for floc to destabilize and aggregated to bigger floc form, the settle down.</li> <li>HRT was important parameter for design EC tank</li> <li>Optimal time at the steady state of turbidity and dye removal was similar results at 30 min.</li> </ul>			
Overflow (OFR)	<ul> <li>OFR was a parameter design for sedimentation tank</li> <li>The small particles have OFR smaller than the larger particle</li> <li>This research was measured the settling test of the 3 conditions of without chemical (whiteout EC process), bentonite, and reactive dye. The OFR dye was 1.7 m/hr.</li> </ul>			
Baffles	<ul> <li>The baffles can delay the HRT in the reactor</li> <li>Baffles can reduced the bypass in the system</li> <li>The gas bubbles from EC process at cathode side was effect to the shorter of baffles and it can improve the longer of baffles</li> <li>Baffles design was selected as a homogenous such as 90% sets (90% - 90% - 90%).</li> </ul>			
Tube settler	<ul> <li>Modules of tube settler was important for increasing the HRT</li> <li>The position of tube settler at the bottom was not improved comparing to the without tube, but the high position can treat turbidity well comparing to the low turbidity.</li> </ul>			

	• The light particles was not effected by tube settler due to its pretended to float up				
	• The results shown that 2 modules at position higher can improve turbidity about $10 - 15$ % at 30 min. comparing to the without tube settler.				
Scale from batch column to continuous system of ECR	• The amount of EC sets was important parameter to scale up related to the volume of wastewater				
	• Current density was played a main role for EC process which can determine the amount of EC sets and DC supply				
	• Flowrate was a main parameter which controlled HRT in the system				
	• The parameters design was found that 4 sets of EC, current density 13.5 mA/cm <sup>2</sup> , and the flowrate 1 L/min. and 3 L/min. of dye and turbidity removal, respectively.				
Comparison of EC and CC process	<ul> <li>The EC was performed better than CC at the high initial concentration of turbidity and dye in terms of treatment efficiency. For low concentration of turbidity and dye, the CC was performed better than EC process.</li> <li>Treatment cost of EC was lower than CC of dye</li> </ul>				
	removal but CC was higher than EC of turbidity removal				
	• Moreover, the sludge of EC was softer than CC process				

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# CHAPTER 5 CONCLUSION AND RECOMMENDATION

#### 5.1 CONCLUSION

The objectives of this study was developed the novel electrocoagulation reactor (ECR) as the continuous mode for treated the turbidity and dye from textile industry wastewater. The effect of several parameters in batch and continue reactor was evaluated. The obtained result can be conclude as the summary below:

> The batch column reactor of EC can removed turbidity and reactive dye reach to 95 % up by the optimal operation time, current density of monopolar arrangement with inner gap of electrode 1.5 cm. When the initial concentration was change of turbidity and dye, so that the removal efficiency changed a little bit for turbidity at low concentration, but the dye removal didn't change much.

> The design parameters in batch column reactor were detail in Table 5.1 with detail value and conditions:

Parameters design in batch column reactor	Range		
Electrode configuration	Monopolar		
Inner electrode configuration (cm)	1.5		
Electrolysis time (min.)	30		
Optimal current density (mA/cm <sup>2</sup> )	4.5		
Study the kinetic of bentonite and reactive dye	S-curve, Second order		
Overflow [OFR] (m/hr.)	1.7, 2.1		
Removal efficiency (%)	95		

Table 5.1. The design criteria of batch and continue parameters with the description

> The ECR reactor was design as the continuous mode which has 3 compartments to treat turbidity and dye removal from the textile industry wastewater. The first compartment was EC tank follow by flocculation tank, and then the sedimentation tank. For the first compartment, EC tank has the width (W) = 26 cm, height (H) = 35 cm with

freeboard 5 cm, and the length (L) = 100 cm. For the second compartment has width (W) = 16 cm, height (H) =35 cm with freeboard 5 cm, and the length (L) = 30 cm. The last lank was sedimentation tank which has width (W) = 16 cm, height (H) = 35 cm with freeboard 5 cm, and the length (L) = 70 cm.

➤ The fluid flow in reactor was studied by using residence time distribution (RTD) principle which has pulse injection methods which has solution of sodium chloride (NaCl) as a tracer. The baffles 90% - 90% - 90% was selected in terms of easy design as a homogenous baffles, increase residence time distribution in the system which can reduce bypass in the system, and the gas bubble in the system can help can reduce some dead zone in reactor. Moreover, the flowrate 3 L/min. was appropriated in the system which provided the best characteristic.

> After selected the best condition from the baffles, the treatment efficiency of ECR was studied with varying the flowrate and current density for turbidity and dye separately. The ECR reactor can treat the removal efficiency of turbidity and dye can reach the 95 % with flow 1 L/min. and 3 L/min., respectively. After the single contaminant was studied so that the combination of turbidity and dye was studied follow. The removal efficiency was studied in flow 2 L/min. can treated the turbidity and dye about 95 %, which the residual of turbidity and dye was less than standard about 20 NTU and 300 ADMI, respectively.

The tube settler was design with the width (W) = 1.8 cm, height (H) = 7 cm, length (L) = 30 cm, and the angle of tube was  $45^{\circ}$ . After put one module (1M) of tube settler in the sedimentation tank the removal of turbidity and dye was not improve. After one more tube added (2M) at high position (P1) of the top surface which can improve the turbidity removal about 10 - 15 % over without tube settler at the time about 50 min., but the result of removal efficiency was similar of HRT at the 70 min. On the other hand, the color removal was not improve since the dye particles was light and difficult to remove. The different position of tube settler was studied, the results shown that it was not effect from the without tube settler.

#### 5.2 **RECOMMENDATION**

After this research was studied in the batch column and continue ECR reactor to treated dye and turbidity from textile industry, some recommendations was for further research were conducted as some points below:

> The ratio of gas flowrate over loss in the optimization part was not suitable use for next study due to the loss of electrode also provide the advantage in EC process.

> The varying of initial concentration of turbidity and reactive dye should be studied with different range in ECR reactor.

> The varying the initial pH in the system should be considered in account because coagulation process is very important in the system which effect on destabilization mechanism.

 $\succ$  Chloride ion (Cl<sup>-</sup>) should be studied to reduce the high voltage in the system which is important parameter for treatment cost.

Since the organic sludge of turbidity can enhance treatment performance in the system, so that the height of organic sludge with recirculation should be investigated.

➢ ECR reactor should perform with real textile industry wastewater in order to validate the treatment performance due to the real wastewater has many parameter which can effect to the system.

➤ Many scale of ECR reactor should be performed for suitable use in the industry sectors.

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### **Appendix I: Characteristic and preparation of wastewater**

#### 1. Preparation of synthetic bentonite and yellow reactive dye

The synthetic of bentonite as the turbidity by plotted the bentonite concentration (mg/L) with the NTU which shown in the Figure 1, after that the synthetic of color was mix reactive dye with tap water. The plotting the dye concentration with Absorbent (Abs.) and ADMI, then made a correlation between Abs. with ADMI which shown in Figure 2. For absorbent, wavelength 420 nm was used and ADMI required wavelength 720 nm.



Figure 1. The standard curve of bentonite as turbidity and correlation between NTU and TSS.



*Figure 2. The standard curve of reactive dye from Abs. to ADMI by correlation with concentration follow as equation "ADMI = (161.82 \* ((Abs+0.0564)/0.0137)-717.08)"* 

## 2. The preparation of solution tracer

The sodium chloride (NaCl) was used as the tracer for residence time distribution (RTD) method which use the pulse injection technique. The standard curve of tracer was shown in Figure 3.



Figure 3. The standard curve of tracer (Nacl) which has correlation between concentrations of tracer with conduct.

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# **Appendix II: Treatment of turbidity and color by chemical coagulation (CC)**

## 1. Conditions of CC process

For CC process, the important parameters were pH and alkalinity which was effect to the destabilization process. The Table 1 detail the condition that allowed for CC.

Table 1. The pH ranges and alkalinity consumed for aluminum and iron coagulants

Coagulant	Empirical Formula	pH range	Alkalinity consumed	
Aluminum Sulfate	$Ab(SO_{1}) = 18H_{2}O_{1}$	Theory 5.5 to 7.8	0.45 mg/L for each 1	
	M2(504)3.101120	Typical 6.0 to 7.4	mg/L of alum	
Ferric Sulfate	Fe <sub>2</sub> SO <sub>4</sub> .9H <sub>2</sub> O	4 0 to 11 0	0.53 mg/L for each	
			mg/L of ferric sulfate	
Ferric Chloride	FeCl <sub>3</sub>	4 0 to 11 0	0.92 mg/L for each	
		1.0 10 11.0	mg/L of ferric chloride	

## 2. The treatment cost and produced water of CC process

The main parameters of treatment cost for CC were alum dosage and power consumption by mixing. The power consumption base on the equation:  $G = \sqrt{\frac{P}{\mu.V}}$ , where G is velocity gradient (s<sup>-1</sup>), P is power mixing (w),  $\mu$  is the viscosity (kg/m.s) and V is the volume of tank (m<sup>3</sup>). The treatment cost of CC showed in Table 2.

Types	Concentration.	Alum dose (mg/L)	Alum price (\$/kg)	Power mixing (kW.h/m <sup>3</sup> )	Energy price (\$/kWh)	Mixing Cost (\$/m <sup>3</sup> )	Total cost (\$/m <sup>3</sup> )
C	2500	60					0.108
CReactive dye (ADMI)	6000	120					0.150
	15,000	180	0.7	0.609	0.109	0.066	0.192
<b>C</b>	50	1					0.067
(NTU)	250	10					0.073
	500	25					0.084

Table 2. The calculation of treatment cost for turbidity and dye removal of CC

#### 3. The initial and final pH after CC process

The section show the initial and final pH of the system for turbidity and color removal which showed in Figure 1 and Figure 2 below.



Figure 1. The final pH of turbidity removal by Jar test along the alum dosage with different concentration 50 NTU, 250 NTU, and 500 NTU



Figure 2. The final pH of decolorization by Jar test along the alum dosage with different concentration 2500 ADMI, 6000 ADMI, and 15000 ADMI.

# **Appendix III: Electrocoagulation (EC) in simple batch column** reactor

### 1. The simple kinetic study

For kinetic of turbidity also studied with zero, first, and second order with follow as the figures below. The Figure 1 and Figure 2 also shown that the zero order not fit with this system due to the R-square was low. The second order was studied follow by the Figure 3 and R-square also was better, but the results also expressed that it was low some current density. Therefore, the zero, first, and second order was not fit to for the turbidity removal.



Figure 1. The zero order of turbidity was studied with different current density



Figure 2. The first order of turbidity was studied with different current density



*Figure 3.* The second order of turbidity was studied with different current density For dye removal was shown that the calibration of reactive dye follow the second order by plotting the concentration (ADMI) along the time which shown in Figure 4.

## 2. Volume of treated water

The Figure 5 shown the treated water from EC process after the final process minus the sludge from the system.



Figure 5. The volume of treated water of dye and turbidity in EC reactor



Figure 4. The validation of different current density with experiment and model

#### 3. The initial and final pH after EC process

The section show the initial and final pH of the system for turbidity and color removal which showed in Figure 6 and Figure 7 below.



Figure 6. The final pH of turbidity removal by EC process with different initial concentration 50 NTU, 250 NTU, and 500 NTU



Figure 7. The final pH of dye removal by EC process with different initial concentration 2500 ADMI, 6000 ADMI, and 15000 ADMI

# Appendix IV: Design and Evaluated of ECR reactor

# 1. Turbidity removal by ECR

The Figure 1, Figure 2, Figure 3, and Figure 4 illustrated the real application of ECR reactor on turbidity removal from the beginning until end process.



Figure 1. The ECR rector removal turbidity at the first stage at EC port



Figure 2. The ECR rector removal turbidity at the final stage at EC port



Figure 3. The ECR rector removal turbidity at the final stage at sedimentation port



Figure 4. Zooming in sedimentation port at the final stage of turbidity removal

## 2. Decolorization by ECR

The Figure 5, Figure 6, and Figure 7 illustrated the real application of ECR reactor on decolorization from the beginning until end process.



Figure 5. The ECR rector removal color at the first stage at EC port



Figure 6. The ECR rector removal color at the final stage at EC port



Figure 7. The ECR rector removal color at the final stage at sedimentation port



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# Appendix V: Residence time distribution (RTD) in new ECR reactor

#### 1. The effect of baffles on E (t) curve

The Figure 1 represent the baffles can affect the flow pattern in reactor at the outlet of EC tank and the effluent of ECR reactor. After that the Figure 2 also shown the effect of baffles at the effluent of ECR reactor. All condition of baffles about 40 experiment was operated with EC process. The operation was study with tap water by adding the tracer into the system then using conduct meter to measure the at 2 points were at outlet of EC process and the effluent of ECR. The steps of calculation was covert from conductivity to concentration of tracer and then convert to the E (t).



Figure 1. The varying of 40 baffles at the outlet of EC process



Figure 2.The varying of 40 baffles at the effluent of ECR reactor CHULALONGKORN UNIVERSITY

#### 2. The effect of operation with and without EC process

In these section shown the graphics E(t) of homogenous baffles 50% - 50% - 50% and 70% - 70% - 70% with both condition which shown in Figure 3 and Figure 4, respectively. More over the each section in EC tank also check such as P1, P2, and P3 which shown in Figure 5.



Figure 3. The effect of operation condition with and without EC process of 50% and 70% of total length at the outlet of EC process



*Figure 4.The effect of operation condition with and without EC process of 50% and 70% of total length at the effluent of ECR reactor*\



Figure 5.The effect of operation condition with and without EC process of 90% of total length at the position 1, 2 and 3

## 3. The real application of RTD method on ECR

All figure below represent the variable parameter such as probe position, baffles, and with and with EC operation.



Figure 6. The experiment of RTD methods on ECR rector without any conditions



Figure 7. The experiment of RTD methods on effect of probe position



Figure 8. The experiment of RTD methods on effect of baffles



Figure 9. The experiment of RTD methods on effect with and without EC process
## **Appendix VI: Improvement by Tube Settler**

## 1. Design and position of tube settler

The improvement of tube settler was studied with 2 conditions. The first condition, it was investigated the module of tube settler and the second, it was studied the position of tube settler. Figure 1 was shown one module of tube settler at position 1 compare to 2 modules of position 1 that showed in Figure 2. Moreover, The Figure 3 showed about the design of tube settler with 2 modules at position 2.



From top surface 4 cm to the sampling port and 8 cm to tube settler



From top surface 4 cm to the sampling port and 8 cm to tube settler

Figure 2. Tube settler design with 2 module at position 1



From top surface 11 cm to the sampling port and 15 cm to tube settler

Figure 3. Tube settler design with 2 module at position 2

## 2. The real application of tube settler on ECR

The detail appearance of tube settler in this work was shown in figures below.



Figure 4. The top view of tube settler installation with 2 modules



Figure 5. The perspective view of tube settler installation with 2 modules



Figure 6. The top view of tube settler while operated with color removal

## VITA

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