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in raw water via coagulation and flocculation process

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via coagulation and flocculation process**

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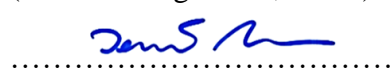
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บทคัดย่อ

การศึกษาในครั้งนี้มีวัตถุประสงค์เพื่อศึกษาประสิทธิภาพการกำจัดไมโครพลาสติกขนาด 103 ถึง 300 ไมโครเมตร ในน้ำดิบ ด้วยกระบวนการสร้างและรวมตะกอน โดยน้ำตัวอย่างที่ใช้ในการศึกษาเป็นน้ำดิบจากคลองประปา บริเวณเขตดอนเมือง จังหวัดกรุงเทพมหานคร ทำการตรวจวัดพารามิเตอร์เบื้องต้นของน้ำตัวอย่าง ได้แก่ ความกระด้างทั้งหมด สภาพความเป็นด่างทั้งหมด ค่าความเป็นกรด-ด่าง ความขุ่น และค่าการนำไฟฟ้า พบว่ามีค่า 115.72 mg/L as CaCO₃, 104.29 mg/L as CaCO₃, 7.4 ถึง 8.2, 20.40 ถึง 32 NTU และ 352 ถึง 565 µS/cm ตามลำดับ จากนั้น นำไมโครพลาสติกที่มีสารอินทรีย์เกาะบนพื้นผิวจากการแช่ในน้ำเสียโรงงานน้ำตาล จำนวน 20 ชั้น มาใส่ในน้ำดิบ เพื่อศึกษาค่าความเข้มข้นที่เหมาะสมที่สุดในการกำจัดไมโครพลาสติกด้วยกระบวนการสร้างและรวมตะกอน โดยการเติมสารส้มและสารโพลีอะคริลาไมด์ ชนิดประจุลบ ผลการศึกษา พบว่า สารส้มและสารโพลีอะคริลาไมด์ ชนิดประจุลบ ส่งผลให้ความเป็นกรด-ด่างลดลงเล็กน้อย และค่าการนำไฟฟ้าเพิ่มขึ้นเล็กน้อย ประสิทธิภาพสูงสุดของการกำจัดความขุ่นจากการเติมสารส้มอย่างเดียว จะต่ำกว่าชุดการทดลองที่มีการเติมสารส้มร่วมกับสารโพลีอะคริลาไมด์ชนิดประจุลบ (97.87±0.03% และ 98.25±0.04% ตามลำดับ) นอกจากนี้ การกำจัดไมโครพลาสติก ขนาด 103 ถึง 300 ไมโครเมตร ด้วยการเติมสารส้ม 37 มิลลิกรัมต่อลิตร จะมีประสิทธิภาพ 85.00±0.00% ในขณะที่การเติมสารส้ม 37 มิลลิกรัมต่อลิตร ร่วมกับสารโพลีอะคริลาไมด์ชนิดประจุลบ 0.04 มิลลิกรัมต่อลิตร จะได้ประสิทธิภาพมากกว่า 90.00±0.00%

คำสำคัญ: ไมโครพลาสติก, กระบวนการสร้างตะกอน, กระบวนการรวมตะกอน, น้ำดิบ, ประสิทธิภาพการกำจัด

Project Title	Removal efficiency of 103 to 300 micrometers microplastics in raw water via coagulation and flocculation process	
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Abstract

This study focused on finding removal efficiency of 103 to 300 μm microplastics (MPs) via coagulation and flocculation process. Water samples were collected from Khlong Prapa, Don Mueang, Bangkok, Thailand. Total hardness, total alkalinity, pH, turbidity, and conductivity in raw water were 115.72 mg/L as CaCO_3 , 104.29 mg/L as CaCO_3 , 7.4 to 8.2, 20.40 to 32 NTU, and 352 to 565 $\mu\text{S}/\text{cm}$ respectively. 20 MPs particles with organic matters on surface from immersing in sugar factory wastewater were then added in raw water to obtain optimum doses of alum and APAM for coagulation and flocculation processes. The results showed that alum and APAM affected in slightly decrease of pH and slightly increase of conductivity. The maximum turbidity removal efficiency by adding alum alone and adding alum with APAM were $97.87\pm 0.03\%$ and $98.25\pm 0.04\%$, respectively. In addition, the 103 to 300 μm MPs removal efficiency by adding 37 ppm alum was $85.00\pm 0.00\%$ whereas the removal efficiency by adding 37 ppm alum with 0.04 ppm APAM was more than $90.00\pm 0.00\%$.

Keywords: Microplastics, Coagulation, Flocculation, Removal efficiency, Raw water

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CONTENTS

	Page
ABSTRACT IN THAI.....	iv
ABSTRACT IN ENGLISH.....	v
ACKNOWLEDGEMENTS.....	vi
CONTENTS.....	vii
LIST OF TABLES	
LIST OF FIGURES	
CHAPTER I INTRODUCTION	
1.1 Background.....	1
1.2 Objective.....	2
1.3 Benefits.....	2
CHAPTER II THEORETICAL BACKGROUNDS AND LITERATURE REVIEW	
2.1 Plastics background.....	3
2.1.1 Plastic production process.....	3
2.1.2 Types of plastics.....	4
2.2 Microplastics (MPs) background.....	6
2.2.1 Types of MPs.....	6
2.2.2 Sources and routes of MPs.....	7
2.2.3 MPs contamination in aquatic environment.....	9
2.2.4 MPs contamination in water supply/drinking water.....	14
2.2.5 Effects of MPs in environmental system and human health.....	16
2.3 Water supply production processes.....	17
2.3.1 Coagulation and flocculation.....	17
2.3.2 Sedimentation.....	19
2.3.3 Filtration.....	19
2.3.4 Disinfection.....	19
2.4 Jar test.....	20
2.4.1 Definition.....	20

CONTENTS (continue)

	Page
2.4.2 Effecting factor.....	20
2.4.3 Optimum conditions.....	22
CHAPTER III MATERIALS AND METHODS	
3.1 Research Materials.....	24
3.1.1 Laboratory instruments.....	24
3.1.2 Chemical substances.....	24
3.1.3 Laboratory equipment and materials.....	25
3.2 Research methods.....	25
3.2.1 Water sample.....	28
3.2.2 Measurement of water quality.....	28
3.2.3 Quantification of MPs.....	29
3.2.4 Coagulation and flocculation procedure.....	30
3.2.5 MPs addition.....	31
3.2.6 Stereo microscope analysis.....	31
3.2.7 Statistical analysis.....	32
CHAPTER IV RESULTS AND DISCUSSION	
4.1 Characteristics of water sample.....	33
4.2 Effects of alum.....	33
4.2.1 Effects of alum on pH, conductivity, and turbidity.....	34
4.2.2 Effects of alum on MPs.....	35
4.3 Effects of APAM.....	37
4.3.1 Effects of APAM on pH, conductivity, and turbidity.....	37
4.3.2 Effects of APAM on MPs.....	38
CHAPTER V CONCLUSIONS AND RECOMMENDATIONS	
5.1 Conclusions.....	40
5.2 Recommendation.....	40
REFERENCES.....	41
APPENDIX A.....	47
APPENDIX B.....	52
APPENDIX C.....	55
AUTHOR'S BIOGRAPHY.....	57

LIST OF TABLES

	Page
Table 2.1 Types of plastic exist in current modern days.....	5
Table 2.2 MPs contamination in freshwater systems.....	11
Table 2.3 MPs contamination in marine systems.....	13
Table 2.4 MPs contamination in water supply/drinking water.....	15
Table 2.5 Advantages and disadvantages of coagulation and flocculation.....	18
Table 2.6 Optimum condition for jar test.....	23
Table 3.1 Measurement of the water sample quality.....	28
Table B.1 MPs removal efficiency by adding alum (narrow range concentration).....	53
Table B.2 MPs removal efficiency by adding APAM.....	54
Table C.1 Determine the optimum alum concentration.....	56
Table C.2 Determine the optimum APAM concentration.....	56

LIST OF FIGURES

	Page
Figure 2.1 Plastic products.....	3
Figure 2.2 Types of MPs.....	7
Figure 2.3 Routes for MPs contamination into aquatic environment.....	8
Figure 2.4 MPs contamination in food chain.....	16
Figure 2.5 Water treatment process.....	17
Figure 3.1 Water sampling point.....	26
Figure 3.2 Flow chart of experimental procedure.....	27
Figure 3.3 Laboratory instrument for measured.....	28
(A) pH.....	28
(B) Turbidity.....	28
(C) Conductivity.....	28
Figure 3.4 Coagulation and flocculation experiment.....	30
Figure 3.5 Stereo microscope.....	32
Figure 4.1 Effects of alum on turbidity removal efficiency.....	35
Figure 4.2 Effects of alum on MPs removal efficiency.....	36
Figure 4.3 Organic substances on MPs surface area.....	36
Figure 4.4 Effects of APAM on MPs removal efficiency and turbidity removal efficiency.....	38
Figure A.1 MPs particles.....	48
(A) Polypropylene.....	48
(B) Polyethylene.....	48
Figure A.2 Mechanically method for made MPs smaller.....	48
Figure A.3 MPs particles after mechanically method.....	49
Figure A.4 MPs particles after immersed in wastewater from sugar factories...	49
Figure A.5 Measurement of parameter.....	50
(A) pH.....	50
(B) Turbidity.....	50
(C) Conductivity.....	50
Figure A.6 Collection of suspension for MPs analysis with stereo microscope..	50
Figure A.7 MPs from stereo microscope analysis.....	51

CHAPTER I

INTRODUCTION

1.1 Background

Plastics are man-made synthetic organic polymer materials that are widely used in daily life and in manufacturing (Ma et al., 2019). The production of plastics is estimated 2.4 billion tons per year which makes up a considerable portion of the volume of waste produced over the world (Bazargan and McKay, 2012; Ma et al., 2019). If plastics break down by chemical, biological and physical processes to a size in range 100 nanometres to 5 millimetres (mm), they will be called microplastics (MPs) (Chain, 2016; Ng et al., 2018; Ma et al., 2019).

Nowadays, the whole world is paying attention and being concern of MPs pollution due to their properties, such as small size, non-biodegradable, resulting in easily contamination, distribution, and accumulation to the environment, especially in aquatic system. MPs in water are concerned that they can lead toxicity into food chain by adsorbing various toxic substances on the surface, such as polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs), heavy metals etc., which may cause health effects and the existence of various living things (Hoffman and Turner, 2015; Ma et al., 2019). MPs are not only detected in marine environment, but also detected in surface waters. There is few studies about MPs in surface water when compared to the marine environment (Di and Wang, 2018). MPs can contaminate in surface water from many ways, degraded plastic waste, atmospheric deposition, surface runoff, municipal sewage, and industrial effluent (World Health, 2019).

In Thailand, surface water is the main source that humans use for many aspects of life, especially raw water for water treatment system (Kruawal et al., 2005). And recently, the studies showed 0.001 mm to 5 mm MPs were detected in water supply (Kosuth et al., 2018; Pivokonsky et al., 2018; Eerkes-Medrano et al., 2019; Mintenig et al., 2019; Tong et al., 2020; Wang et al., 2020). Water treatment process is divided into 4 main processes: coagulation and flocculation, sedimentation, filtration, and disinfection (Metropolitan Waterworks Authority (MWA), 2010; Provincial Waterworks Authority (PWA), 2018). Unfortunately, each unit of water treatment system is not designed to treat MPs. However, there are some processes that have potential to improve the efficiency of MPs removal that is: coagulation and flocculation processes (Wang et al., 2020). The purpose of coagulation

and flocculation processes is turbidity reduction by adding an amount of coagulant (e.g., alum, polyaluminum chloride and ferric chloride) and coagulant aids (e.g., polymer) into the system for destroy colloidal stability and generate floc. Therefore, adjustment of an amount of coagulation and/or coagulant aids should affect the removal of some small particle such as MPs.

Hence, this study focuses on the removal efficiency of 103 to 300 μm MPs in raw water via coagulation and flocculation process, and evaluation of the optimum doses of aluminum sulfate (alum) and anionic polyacrylamide (APAM) for the treatment process.

1.2 Objective

To determine the removal efficiency of 103 to 300 μm MPs in raw water via coagulation and flocculation processes using alum and APAM as coagulant and coagulant aid.

1.3 Benefits

1.3.1 Knowing efficiency of MPs treatment in raw water via coagulation and flocculation process.

1.3.2 Improving the process of MPs treatment in water supply production system.

CHAPTER II

THEORETICAL BACKGROUNDS AND LITERATURE REVIEW

2.1 Plastics background

Plastics are organic synthetic or semi-synthetic materials which produce from cellulose, coal, natural gas, salt, crude oil, etc. Due to plastic properties, low density, non-conductive, heat resistance, corrosion resistance, and flexible, make it widely used in daily life and industrial applications. There are many forms of plastic products that are popularly used for human life such as packaging, building and construction materials, electronic devices, clothes, etc. (PlasticEurope, 2019).



Figure 2.1 Plastic products

(Department of International Trade Promotion (DITP), 2014)

2.1.1 Plastic production process

The raw materials for plastic production are material composed of structure with hydrocarbon chains (e.g., petroleum, crude oil, and natural gas). Fractional distillation of crude oil provides a mixture of hydrocarbon chains. And some of them are used as substrate in plastic production such as ethylene, and propylene. There are 2 main reaction of the production process: polymerization and polycondensation (National Metal and Materials Technology Center (MTEC), 2017; PlasticEurope, 2019).

2.1.1.1 Polymerization

Polymerization is the reaction that same monomers, such as ethylene and propylene, are linked together to form long polymer chains. This reaction

does not produce other polymers, there is only unit of substrate monomers (e.g., polyethylene, polypropylene, and polyvinyl chloride) (MTEC, 2017; PlasticEurope, 2019).

2.1.1.2 Polycondensation

Polycondensation is the reaction that more than 2 different monomers are linked together to form long polymer chains (e.g., nylon, and polyester (PEST)). This reaction provides the compounds with small molecule as by product, such as H₂O, HCl, and CH₃OH. (MTEC, 2017; PlasticEurope, 2019).

2.1.2 Types of plastics

Types of plastics are divided into 2 types, depending on thermal properties of plastics: thermoplastics and thermosetting (MTEC, 2017; PlasticEurope, 2019).

2.1.2.1 Thermoplastics

Thermoplastics are polymers, which the structure is linear or short branched chains. These types are softened on heating and then harden again on cooling without changing the chemical and physical properties. Polycarbonate (PC), polyethylene (PE), polyethylene terephthalate (PET), polytetrafluoroethylene (PTFE), polyvinyl chloride (PVC), polypropylene (PP), and polystyrene (PS) are the example of thermoplastics discover in daily life (MTEC, 2017; PlasticEurope, 2019).

2.1.2.2 Thermosetting

Thermosetting are polymer with network structure. Due to this polymer form, thermosetting has a permanent shape and heat resistance which cannot be recycled. There are many thermosetting find in daily life such as epoxide (EP), phenol-formaldehyde (PF), polyurethane (PUR), and unsaturated polyester resins (UP) (MTEC, 2017; PlasticEurope, 2019).

Moreover, a lot of plastics that are used over the world can be recycled. There are number symbols (NO.1 to 7) which use for divide types of plastics resulting in the easy plastic waste management. The symbol is usually found at the bottom of a plastic container (MTEC, 2017). Types of plastics with number symbol show in Table 2.1.

Table 2.1 Types of plastic exist in current modern days (waste4change, 2019; NON TOXIC REVOLUTION, 2020)








Type of plastic	Detail
 <p>NO.1 Polyethylene Terephthalate (PET or PETE)</p>	<ul style="list-style-type: none"> - Well known as a wrinkle-free fiber. - Generally used for food and drink packaging. - Considered as a carcinogen capable.
 <p>NO.2 High-Density Polyethylene (HDPE)</p>	<ul style="list-style-type: none"> - Very intense, stronger, thicker and more stable than PET. - Normally used as the grocery bag, milk bottles, juice container, detergent bottles, and medicine bottle. - Considered as a safer option for food and drinks use.
 <p>NO.3 Polyvinyl Chloride (PVC)</p>	<ul style="list-style-type: none"> - Normally used in toys, blister wrap, cling wrap, detergent bottles, loose-leaf binders, blood bags and medical tubing. - Considered as the most hazardous plastic. - Rarely accepted by recycling programs.
 <p>NO.4 Low-Density Polyethylene (LDPE)</p>	<ul style="list-style-type: none"> - Less intense, less crystalline and more flexible form of PE. - Mostly used for bags, plastic wraps, food containers, container lids, wire and cable covering. - Considered as a safer plastic option for food and drink use. - Quite difficult to be recycled.
 <p>NO.5 Polypropylene (PP)</p>	<ul style="list-style-type: none"> - Normally used for hot food containers. - Inflexible and more resistant to heat. - Considered a safer plastic option for food and drink use. - Considered to cause asthma and hormone disruption in human. - Unable to recycle.
 <p>NO.6 Polystyrene (PS)</p>	<ul style="list-style-type: none"> - Commonly used for food containers, egg cartons, disposable cups and bowls, packaging, and also bike helmet. - Considered as brain and nervous system toxicant. - Low recycling rate.

Table 2.1 Types of plastic exist in current modern days (continue) (waste4change, 2019; NON TOXIC REVOLUTION, 2020)

Types of plastic	Detail
 <p>NO.7 Other</p>	<ul style="list-style-type: none"> - All plastics other than those identified by number 1-6. - Most common plastic in this category is polycarbonate (PC). - Normally used for baby bottles, sippy cups, water bottles, water gallon, metal food can liner, ketchup container, and dental sealants. - Banned the use of PC for baby bottles and infant formula packaging. - Very low recycle rate quality.

2.2 Microplastics (MPs) background

MPs are the result from the fracture of plastic by chemical, biological and physical processes. Size of MPs is in the range of 100 nm (0.0001 mm) to 5 mm. If their size less than 100 nm, they will be called nanoplastics (Chain, 2016; Ng et al., 2018; Ma et al., 2019). MPs come from various sources, including from direct manufacture or from larger plastic debris that degrades into smaller pieces (National Ocean Service, 2020).

2.2.1 Types of MPs

MPs are divided into 2 types:

2.2.1.1 Primary MPs

Primary MPs are the MPs from direct manufacture that size is smaller than 5 mm such as microbeads, nurdles and fibers. This type is often found in health and beauty products: cleansers and toothpastes (National Ocean Service, 2020).

2.2.1.2 Secondary MPs

Secondary MPs are formed by breaking down of larger plastic particles, which caused by chemical, biological and physical processes into millions of smaller pieces. The example of secondary MPs discover in environment such as fragmented polystyrene that breaks off from foam boxes at the beach (Beaulieu, 2017).

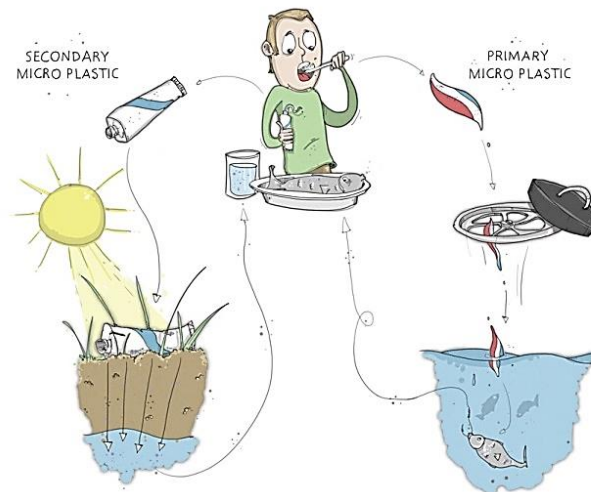


Figure 2.2 Types of MPs (Beaulieu, 2017)

2.2.2 Sources and routes of MPs

MPs were found in freshwater and ocean. There are many sources and routes of MPs to contaminate in aquatic system. Contamination of MPs in ocean is not only caused by dumping plastic waste into the sea, but also due to freshwater runoff with MPs contamination (World Health, 2019; Water-pollution.org.uk, 2020). Sources of MPs into freshwater included surface runoff, wastewater effluents and mishandled plastic wastes (World Health, 2019). Each of these and other potential sources are described below.

2.2.2.1 Surface runoff

Various types of land use response to human demands and driving the economy such as road construction that uses of road paint as a traffic sign, land transportation that causes wear and tear from rubber wheels to fall onto the road, agriculture that causes plastic waste from packaging. Other than, daily life activities such as washing clothes can cause plastic that are components of the clothes into the environment. These activities cause MP contamination to the ground and can be leached into the natural water source by surface runoff (World Health, 2019).

2.2.2.2 Wastewater effluent

Due to daily activities require humans to use cleaning agents to wash the body and clothes. Resulting in MPs ingredient in cleaning products, such as toothpaste, facial cleansing foam, are flushed down toilets and sinks, and inputs into sewage systems. If the wastewater treatment or other appropriate systems for collecting

and treating wastewater are ineffective, MPs can contaminate into natural water sources. Thus, management of wastewater treatment systems are considered as an important issue (World Health, 2019).

2.2.2.3 Industrial effluent

In many industries, MPs are used as raw materials for production which can lead MPs to contaminated with wastewater from the production process. If wastewater management is inefficient before release the wastewater into natural water sources, it may cause contamination of MPs in aquatic system (World Health, 2019).

2.2.2.4 Fragmentation and degradation of MPs

When plastic waste is dumped into the natural water source, it is transformed by physical, biological, and chemical process such as UV radiation, water temperature, water currents. Plastic waste is broke and degraded until it's size becomes MP and contaminate the aquatic environment (Ma et al., 2019; World Health, 2019).

2.2.2.5 Atmospheric deposition

Due to wet and dry deposition, precipitation and runoff can contribute MPs from air and land into the natural water source (World Health, 2019).



Figure 2.3 Routes for MPs contamination into aquatic environment

(World Health, 2019)

2.2.3 MPs contamination in aquatic environment

MPs are found in aquatic environment not only in marine but also freshwater (World Health, 2019; Water-pollution.org.uk, 2020). The MPs contamination situations in freshwater and marine systems can be summarized below.

2.2.3.1 MPs contamination in freshwater systems

Although, there are several studies about MPs contamination in freshwater systems over the world, the methods for water preparation and MPs measurement are not developed to the same standard. As a result, the results of the study cannot be compared directly (Damrongsiri and Chanpiwat, 2019). The results of case studies about MPs contamination in freshwater systems showed the differences amount, size and type of MPs in each study area. The results showed PE and PP were the dominant type with size ranged from 0.001 to 2 mm and the abundance of MPs ranged from 0.01 to 6,614 particles/L.

Wang et al. (2017) found MPs in surface water of 20 urban lakes and urban reaches of the Hanjiang River and Yangtze River of Wuhan. The abundance of MP ranged from 1.66 ± 6.39 to 8.93 ± 1.59 particles/L. More than 80% of MPs in number had a size of < 2 mm. The majority type of MPs was fibers. PET and PP were the dominant types of MPs, followed by PE, polyamide (Nylon), and PS respectively.

Di and Wang (2018) showed MPs pollution in surface waters from the Three Gorges Reservoir (TGR) was more serious in urban areas. The abundance of MPs ranged from 1.60 to 12.61 particles/L. The average abundance was 4.70 ± 2.82 particles/L. MPs with size < 0.5 mm were the most abundant in all water samples. The dominant shape of MPs is fibers, accounted for 28.6% to 90.5% of the total MPs. From identification of MPs by micro-Raman spectroscopy, PS was the most common type (38.5%). The two other MPs were PP and PE, which were responsible for 29.4 and 21 percent of MPs respectively.

Pivokonsky et al. (2018) found MPs in all water samples from raw water, provides for three water treatment plants (WTPs), all are located in urban areas. The average abundance of MPs ranged from 1473 ± 34 to 3605 ± 497 particles/L. The most plentiful size was < 0.01 mm, accounting for up to 95%. And the dominant shape of MPs was fragments. The majority of the MPs ($>70\%$) comprised of PET, PP, and PE.

Wang et al. (2018) presented an important source of MPs in two important lakes in the middle reaches of the Yangtze River is fishery activity. The

abundance ranges of MPs in Dongting Lake and Hong Lake were 0.90 to 2.80 and 1.25 to 4.65 particles/L, respectively. MPs with a size of < 2 mm were the dominant size. And the most common shape was fibers. PE, PP and PS were the main components of the identified particles.

Bordós et al. (2019) presented the detection of MPs in Central and Eastern European (CEE). Samples were taken from different types of fish ponds and natural water bodies. MPs in all water samples ranging from 0.003 to 0.03 particles/L. The average abundance was 0.01 ± 0.01 particles/L. The MPs size was less than 0.3 mm. PP was detected in almost all of the samples. The second most common material was PE, then PES and PS.

Di et al. (2019) investigated the contamination of MPs in surface water from Danjiangkou Reservoir. MPs were observed in water with abundances varying from 0.47 to 15.02 particles/L. The average abundance was 2.59 ± 3.88 particles/L. Particles with a size of < 2 mm were more frequently observed than other sizes. The major MPs shape was fibers. PP has the largest chemical composition of the identified MPs for 44.9%, whereas PS ranks second with 34.7%. Finally, 20.3% of PE was the composition of the identified MPs.

Wang et al. (2020) exhibited MPs contaminated in all raw water samples, supply for drinking water treatment plants (DWTPs), located in the Yangtze River Delta. The average abundance was 6614 ± 1132 particles/L. The most abundance size was 0.001 to 0.005 μm , accounting for 54.6 to 58.0% of the total MPs. And the majority of the types were PET, PE, and PP, respectively. Moreover, fibers were the most common shape, accounting for 53.9 to 73.9% of the total.

All of MPs contamination in freshwater systems are summarized in Table 2.2.

Table 2.2 MPs contamination in freshwater systems

Research area	Dominant abundance sizes of MPs (mm)	Average abundance of MPs in water (particles/L)	Dominant types of MPs	References
Urban lakes in Wuhan, China	< 2	1.67±6.40 to 8.39±1.59	PE > nylon > PS	Wang et al., 2017
Three Gorges Reservoir, China	< 0.5	4.70±2.82	PS > PP > PE	Di and Wang, 2018
Three drinking water treatment plants, Czech Republic	< 0.01	1473±34 to 3605±497	PET, PP, PE	Pivokonsky et al., 2018
Dongting Lake and Hong Lake, China	< 2	1.19 (Dongting Lake) 2.28 (Hong Lake)	PP, PE, PS	Wang et al., 2018
Carpathian basin, Hungary	< 0.3	0.01±0.01	PP > PE > PES	Bordós et al., 2019
Danjiangkou Reservoir, China	< 2	2.59±3.88	PP > PS > PE	Di et al., 2019
Drinking water treatment plants, China	0.001 to 0.005	6614±1132	PET > PE > PP	Wang et al., 2020

2.2.3.2 MPs contamination in marine systems

The results of case studies about MPs contamination in marine systems showed the different amount, size and type of MPs in each study area. The results showed PE and PP were the dominant type with size ranged from 0.1 to 15.98 mm and the abundance of MPs ranged from 0 to 2.43 particles/L.

Song et al. (2015) studied the presence of MPs in seawater, was collected near- and offshore of Geoje Island, South Korea. The average abundance was 0 to 0.3 particles/L. Fragments were the main MPs shape. The main size was < 1 mm. And EPS was the dominant MPs type.

Castillo et al. (2016) presented the evidence of the prevalence of MPs in the marine waters of Qatar's Exclusive Economic Zone (EEZ). The average abundance of MPs was 0.0007 particles/L. The main shapes of MPs were 0.13 to 1.82 mm granular and 0.15 to 15.98 mm fibrous shapes. The dominant types were PP, LDPE, PE, and PS etc.

Zheng et al. (2019) showed the presence of MPs in seawater of Jiaozhou Bay, China. The abundance of MPs in the bay seawater samples ranged between 0.02 to 0.12 particles/L. The average abundance of MPs was 0.05 ± 0.03 particles/L. The most common size of MPs in seawater samples was 1 to 1.99 mm. The dominant shape in all samples was fiber. The main types of MPs were PET, PP, and PE, accounting for 56.25%, 34.38%, and 3.13%, respectively.

Jiang et al. (2020) investigated the contamination of MPs in the Nordic Seas. In the area affected by the EastGreenland Current, the abundance of MPs was 1.19 ± 0.28 particles/L, whereas the cold basin affected by the Greenland Sea Gyre was 2.43 ± 0.84 particles/L. The most common size of both areas was 0.1 to 0.5 mm. And the main shape was fibers.

All of MPs contamination in marine systems are summarized in Table 2.3.

Table 2.3 MPs contamination in marine systems

Research area	Dominant abundance sizes of MPs (mm)	Average abundance of MPs in water (particles/L)	Dominant types of MPs	References
Geoje Island, South Korea	< 1	0 to 0.3	EPS	Song et al., 2015
Qatar's Exclusive Economic Zone, Qatar	0.13 to 1.82 (granular shape) 0.15 to 15.98 (fibrous shapes)	0.0007	PP, LDPE, and PE	Castillo et al., 2016
Jiaozhou Bay, China	1 to 1.99	0.05±0.03 particles/L	PET > PP > PE	Zheng et al., 2019
The Nordic Seas	0.1 to 0.5	1.19 ± 0.28 particles/L (The area affected by the EastGreenland Current) 2.43 ± 0.84 particles/L (The cold basin affected by the Greenland Sea Gyre)	-	Jiang et al., 2020

2.2.4 MPs contamination in water supply/drinking water

Due to the freshwater is the main source of raw water for water treatment system (Kruawal et al., 2005), the studies of MPs contamination in freshwater over the world can make concern about MPs contamination in water supply/drinking water.

Pivokonsky et al. (2018) found MPs in water samples from treated water, which effluent from three water treatment plants (WTPs), all are located in urban areas. The average abundance of MPs ranged from 338 ± 76 to 628 ± 28 particles/L. The most common size was 0.001 to 0.005 mm, comprising approximately 25 to 60% of the total. And the dominant shape of MPs was fragments. The majority of the MPs were PET, PP, and PE.

Strand et al. (2018) presented the contamination of MPs in drinking water from 17 sites around Denmark. The average abundance was 0.31 particles/L. The major size of MPs was > 0.10 mm. The predominant shape of MPs were fibers, accounting for 82 %. The most common types of MP particles detected in the samples were PET, PP, and PS.

Tong et al. (2020) presented the presence of MPs in tap water, taken at different cities of China. The abundances of MPs ranged from 0 to 1247 particles/L, with a mean concentration of 440 particles/L. The major size was < 0.05 mm. And fragments were the most abundant shape in most samples. In addition, PE was the most presence in all samples account for 26.8%, followed by PP (24.4%).

Wang et al. (2020) investigated the contamination of MPs in treated water, which effluent from drinking water treatment plants (DWTPs), located in the Yangtze River Delta. The average abundance was 930 ± 72 particles/L. The most abundance size was 0.001 to 0.005 mm, accounting for 84.4 to 86.7% of the total MPs. Moreover, fibers were the most common shape, accounting for 51.6 to 78.9% of the total. And the majority of the types were PET, PE, and PP, respectively.

All of MPs contamination in water supply/drinking water are summarized in Table 2.4.

Table 2.4 MPs contamination in water supply/drinking water

Research area	Dominant abundance sizes of MPs (mm)	Average abundance of MPs in water (particles/L)	Dominant types of MPs	References
Three drinking water treatment plants, Czech Republic	0.001 to 0.005	338±76 to 628±28	PET, PP, PE	Pivokonsky et al., 2018
Denmark	> 0.10	0.31	PET, PP, PS	Strand et al., 2018
China	< 0.05	440	PE > PP	Tong et al., 2020
Drinking water treatment plants, China	0.001 to 0.005	6614 ± 1132	PET > PE > PP	Wang et al., 2020

2.2.5 Effects of MPs in environmental system and human health

Because the size of MPs is small, so it is difficult to remove from environment. Including properties of MPs that are naturally biodegradable and take a long time to decompose, it is easily to contamination, distribution and accumulation in the environment. There is an extremely high risk that aquatic animals eat those plastic and MPs. Therefore, MPs are concerned to toxic in food chain because the small particle size lead to a large surface area and can absorb various toxic substances such as polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs), heavy metals (including Zn, Cu, Pb, Ag) etc., which may cause health effects and the existence of various living things (Ashton et al., 2010; Hoffman and Turner, 2015; Ma et al., 2019). After they uptake MPs, the feed efficiency and the growth rate of aquatic animals are decreased (Li et al., 2018; Ma et al., 2019). In addition, gene expression of aquatic species can change after taking MPs adsorbed organic pollutants (Ma et al., 2019; Rochman et al., 2014). As a result, human health can be seriously threatened through the food chain (Ma et al., 2019; Rochman et al., 2014).

In addition, the contamination of MPs in water supply/drinking water is not possible to draw any firm conclusions on toxicity related to MPs exposure through drinking-water, particularly for the smallest particles, but no reliable information suggests it's concern (World Health, 2019).



Figure 2.4 MPs contamination in food chain (Water-pollution.org.uk, 2020)

2.3 Water supply production processes

The raw water used in the water supply production process is from natural water sources. It is pumped up from the natural water source by a low-pressure pumping plant. The raw water, that can be used to produce water supply, must pass water quality standards, and must have a sufficient amount to continuously production. After that, raw water is brought through various production processes to get water supply that can be used for consumption (PWA, 2018). The processes for water supply production can be divided into 4 main processes:

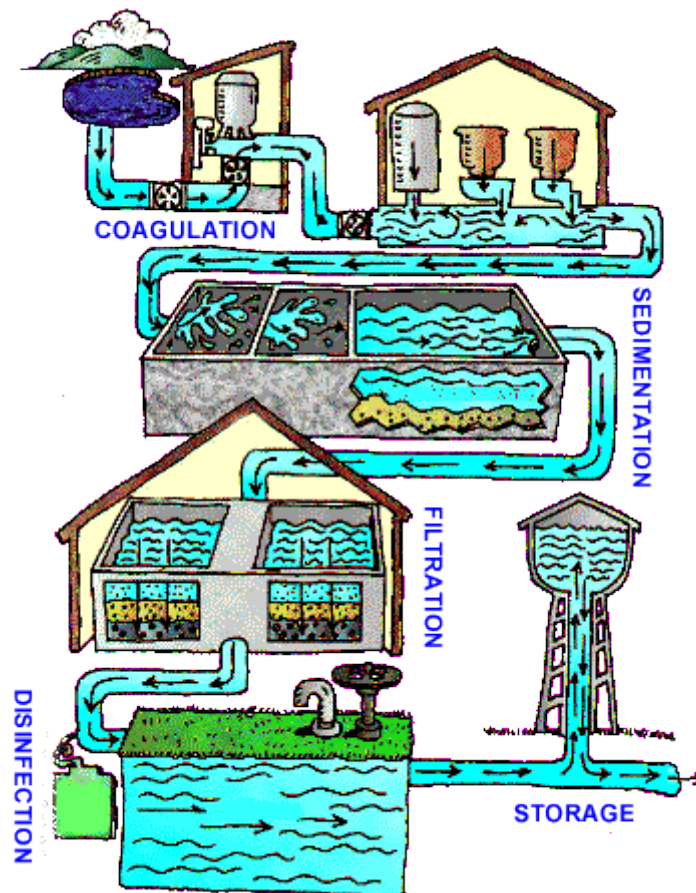


Figure 2.5 Water treatment process

(Center for Disease Control and Prevention (CDC), 2015)

2.3.1 Coagulation and flocculation

For improving raw water quality, raw water that has already been pumped is mixed with chemicals such as alum (aluminum sulfate, $\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$) and lime. Alum solution supports the sedimentation process to better than ever whereas lime solution inhibits the growth of algae in the water, and adjust the pH of raw water.

Occasionally, chlorine is added in order to kill germs that may mix with water in this initial step (PWA, 2018).

2.3.1.1 Coagulation principles

Coagulation is a process in which coagulant with the opposite charge to the colloidal particles, which neutralize the negative charges on colloidal surface, is added into the water. Coagulant is then rapidly mixed in water for breaking down the stability of the colloid resulting in forming larger flog and heavy enough to settle. The most common coagulants are aluminum sulfate (alum, $\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$), ferric sulfate and sodium aluminate. Coagulation is not affected by over-mixing, however, it can be incomplete step when lacking mixing occurs. Retention time in the rapid-mix chamber is 1 to 3 minutes (Mazille and Spuhler, 2019; PWA, 2018).

2.3.1.2 Flocculation principles

Following coagulation, flocculation is a gentle mixing stage, increases the opportunities for colloids to form larger blocks by stir water at a speed that does not cause too much turbulence until the particles break apart (slow mixing). The floc size continues to build through additional collisions and interaction with inorganic polymers formed by the coagulant or with polymers added, called coagulant aids or flocculant. Coagulant aids may be added during this step to help bridge, bind, and tighten the floc, and increase settling rate efficiency. Retention time in the slow-mix chamber is from 15 or 20 minutes to an hour or more (Mazille and Spuhler, 2019).

2.3.1.3 Advantages and disadvantages

Table 2.5 Advantages and disadvantages of coagulation and flocculation (Nathanson et al., 2019; Mazille & Spuhler, 2019)

Advantages	Disadvantages
Easy to use and cost effective.	Need chemicals.
Able to separate many suspended and dissolved compounds from water.	Need person who is expertise in system design and maintenance.
Increase filtration process efficiency.	Solids from the process may be toxic which needs to be treated with appropriate methods.

Table 2.5 Advantages and disadvantages of coagulation and flocculation (Nathanson et al., 2019; Mazille & Spuhler, 2019) (continue)

Advantages	Disadvantages
Able to use a variety of chemicals.	The treatment process takes quite a long time.
Low chemical cost.	

2.3.2 Sedimentation

In this step, the water from coagulation and flocculation step is brought to a large sedimentation tank. It removes the floc, which suspended in water, from water under the influence of gravity to the bottom of the tank. The clear water above will flow along the water receiving trough to the next step. Retention time for sedimentation process ranges from one hour up to two days. (PWA, 2018; Shrestha and Spuhler, 2020).

The shape of sedimentation tanks may be square or circle. The influent is released regularly into the tank while the clarified effluent is released from the tank via the weir with overflow. At the tank bottom, sediment that has been suspended in water will fall and become a cumulative layer, called sludge. Moreover, tank's surface area affects the efficiency of sedimentation tank more than its volume or depth (Nathanson et al., 2019).

2.3.3 Filtration

For filtration, it is the process to remove contaminants such as diseases, dust, chemicals etc. from water using different types of filters, coarse and fine sand is used in Thailand. This process will bring the clear water after the sedimentation process to flow through the filter. The filtered water will be very clear, but there is turbidity approximately 0.2-2.0 NTU. And the sand will be washed regularly to make the filter efficient. After that, the filtered water will flow into the disinfection system (PWA, 2018; Nathanson et al., 2019).

2.3.4 Disinfection

The method of disinfection in water supply production in Thailand uses chlorine or chlorine compounds, called chlorination. The filtered water is clear, but it may be contaminated with germs in water. Therefore, it is necessary to kill germs using

chlorine, which can kill germs as well. Proper dosages of chlorine at the treatment plant can cause taste or odor problems in treated water. However, the amount of chlorine in the water is controlled to a safe level for consumption. The water that has been mixed with chlorine is called "water supply". It can be used for consumption, and will be stored in a large tank called a clear water tank to manage the service further (PWA, 2018; Nathanson et al., 2019).

After disinfection step, the tap water that has already been produced must be analyzed and examined again by scientists for water quality control, and make sure that the water supply is safe for consumption. The final step of production is distribution of water supply by passing along the pipe line (PWA, 2018).

2.4 Jar test

2.4.1 Definition

Jar test is a process to determine the amount and concentration of suitable coagulant and coagulant aid for coagulation and flocculation in small amount of water. One set of jar test consists of many beakers, coagulant with the same volume but different concentration is added. At the end, the jar test results are used for full-scale coagulation and flocculation. The results will be selected from the concentration that leads to the minimum turbidity, called optimum dosage. However, the results of the full-scale production may not be consistent with the results of the jar test (Nathanson et al., 2019).

2.4.2 Effecting factor

2.4.2.1 Particle size and molecular weight

The smaller particle size provides the greater surface area per unit weight of colloid. Increases in the amount of coagulant used cannot be affected by decreases in particle size as a linear. Furthermore, the process performance can be affected by the molecular weight of colloid. Higher molecular weight results in better thickener activity although there is some case showed loss of activity when the molecular weight increases (Pillai, 2004).

2.4.2.2 Surface charge

The density of the surface charge of particles has a direct effect on coagulation process. The coagulant requirement increases to neutral the surface charges because the surface charge increases in density. However, reversal charge and inter-particle repulsion can be caused by the excessive amount of coagulant (Pillai, 2004).

2.4.2.3 Water properties

Dissolved species in water, such as Ca^{2+} , Mg^{2+} , Fe^{2+} and Fe^{3+} , help neutralize surface charge and can reduce the coagulant requirements. A greater number of ions, the higher of conductivity value. The coagulant requirement can be determined by the conductivity in water. In addition, the hardness in water can be determined by Ca^{2+} and Mg^{2+} . The demand of coagulant can be affected by the higher hardness results in the lower coagulant requirement (Pillai, 2004).

Moreover, the performance of the process may be caused by pH, temperature, and alkalinity. Destabilization of colloid is responsible to pH. The requirement of excessive amount of coagulant increases for lower pH to the optimal ranges (i.e., 6 to 7 for alum, 5.5 to 6.5 for iron) when the water has high alkalinity.

And water temperature can causes the viscosity of water. As a result, low temperature will decrease the precipitation and the hydrolysis kinetics (Environmental Protection Agency (EPA), 2017).

2.4.2.4 Polymer types

Polymer, a long chain hydrocarbon, is defined as flocculant or coagulant aid which added into raw water in flocculation process. Generally polymer is copolymers of acrylamide and sodium acrylate. Polymer plays an important roles for bridging flocs in order to form the bigger one, improves the sedimentation efficiency rate (Pillai, 2004).

2.4.2.5 Slurry solids

The distribution of polymer in the slurry can be caused by the higher solid concentration. Polymer has difficult uniform spread when the concentration of solid is higher. Due to the effect of smaller particle size, the decrease in particle size and the increase in solid concentration provide the increase of polymer requirement (Pillai, 2004).

2.4.2.6 Shear

The break apart of floc can be affected by the excessive shear force or mechanical action (i.e., over mixing). Therefore, floc distribution in slurry and floc shear must be balance (Pillai, 2004).

2.4.3 Optimum conditions

The conditions for coagulation and flocculation processes are determined, especially the amount and concentration of suitable coagulant and coagulant aid, resulting in the maximum of turbidity removal efficiency.

Ayekoe et al. (2017) showed the optimization of coagulation-flocculation conditions for removal turbidity in raw water from Agbô River, located in the south-east of Ivory Coast. Alum was use as coagulant. The optimum dose of alum was 110 mg/L, with pH 5. The removal efficiency for turbidity was 58.8%.

Samornkraisorakit (n.d.) presented the optimum doses of alum, 32 mg/L, for treat raw water in water supply production process. At pH ranged 6-7, the removal efficiency for turbidity was 96.6%.

Mittapalli (2017) studied the optimization of alum in jar test experiment. The raw water was collected from Lake Durgam Cheruvu, freshwater lake located in Rangareddy district, Telangana, India. The result showed 100 mg/L alum was the optimum dose, with pH 7. The highest turbidity removal efficiency was within 66-76 %.

All of optimum conditions for jar test are summarized in Table 2.6.

Table 2.6 Optimum conditions for jar test

Optimum doses of Alum (mg/L)	Optimum condition	Process condition				Sedimentation time (min)	Turbidity (NTU)		Removal efficiency (%)	References
		Coagulation		Flocculation			Raw water	Treated water		
		Mixing speed (rpm)	Mixing time (min)	Mixing speed (rpm)	Mixing time (min)					
110	pH 5	250	1	45	15	-	11.1	4.57	58.8	Ayekoe et al., 2017
32	pH ranged 6 to 7	200	1	40	20	10	93.8	3.15	96.6	Samornkraisorakit, n.d.
100	pH 7	161	2	25	30	120	~80	~20	66-76	Mittapalli, 2017

CHAPTER III

MATERIALS AND METHODS

3.1 Research Materials

3.1.1 Laboratory instruments

- 3.1.1.1 Jar tester
- 3.1.1.2 pH meter (Milwaukee Model pH55/pH66)
- 3.1.1.3 Turbidity meter (Hach Model 2100P)
- 3.1.1.4 Conductivity meter (Hach sension 156)
- 3.1.1.5 Stereo microscope (FMA050)
- 3.1.1.6 Hot plate
- 3.1.1.7 Laboratory fume hood
- 3.1.1.8 Drying oven
- 3.1.1.9 Air pump

3.1.2 Chemical substances

- 3.1.2.1 0.0005 ppm Aluminium sulfate (alum)
- 3.1.2.2 Anionic polyacrylamide (APAM)
- 3.1.2.3 Phenolphthalein
- 3.1.2.4 Methyl orange
- 3.1.2.5 Methyl red
- 3.1.2.6 95% Ethanol
- 3.1.2.7 0.02 N H₂SO₄
- 3.1.2.8 50% HCl
- 3.1.2.9 0.05 N Na₂CO₃
- 3.1.2.10 Conc. NH₄OH
- 3.1.2.11 NH₄Cl
- 3.1.2.12 Standard EDTA
- 3.1.2.13 Eriochrome Black T
- 3.1.2.14 Hydroxylamine hydrochloride
- 3.1.2.15 0.01 M Standard calcium Solution

3.1.3 Laboratory equipment and materials

- 3.1.3.1 100 μm , 300 μm , and 4 mm Stainless steel sieves
- 3.1.3.2 Polyethylene and polypropylene beads
- 3.1.3.3 Glass microfiber filters (GF/C \O 47 mm)
- 3.1.3.4 Watch glass
- 3.1.3.5 Standard Metal Forceps
- 3.1.3.6 50 mL, 150 mL, 250 mL, 500 mL, 1 L, and 2 L glass beakers
- 3.1.3.7 Analytical balance (precise to 0.1 mg)
- 3.1.3.8 Metal spatula
- 3.1.3.9 Stir bar
- 3.1.3.10 Retort stand
- 3.1.3.11 Aluminum foil
- 3.1.3.12 Squirt bottle containing distilled water
- 3.1.3.13 1 mL, 2 mL, 5mL, 25 mL, and 100 mL Pipettes
- 3.1.3.14 Pipettes bulb
- 3.1.3.15 100 mL, 200 mL, 250 mL, 1 L, and 2 L Volumetric flasks
- 3.1.3.16 Büchner funnel
- 3.1.3.17 Reagent bottles
- 3.1.3.18 Glass bottles
- 3.1.3.19 Stirring rod
- 3.1.3.20 Burette
- 3.1.3.21 15 mL and 250 mL Glass cylinder
- 3.1.3.22 250 mL Erlenmeyer flask
- 3.1.3.23 Glass funnel
- 3.1.3.24 Dropping Bottle, Amber
- 3.1.3.25 Dropper
- 3.1.3.26 Syringe
- 3.1.3.27 Desiccator

3.2 Research methods

The water samples were collected from Khlong Prapa, Don Mueang, Bangkok, Thailand (Figure 3.1). The sample was measured for pH, temperature, turbidity, conductivity, hardness and alkalinity. The optimum dose of alum for coagulation and flocculation processes were provided to remove turbidity, in case without MPs, from raw

water. Then, the same treatment processes were used to find out the removal efficiency of raw water with added MPs particles. All experiments were conducted three times. The flow chart of experimental procedure is shown in Figure 3.2.



Figure 3.1 Water sampling point

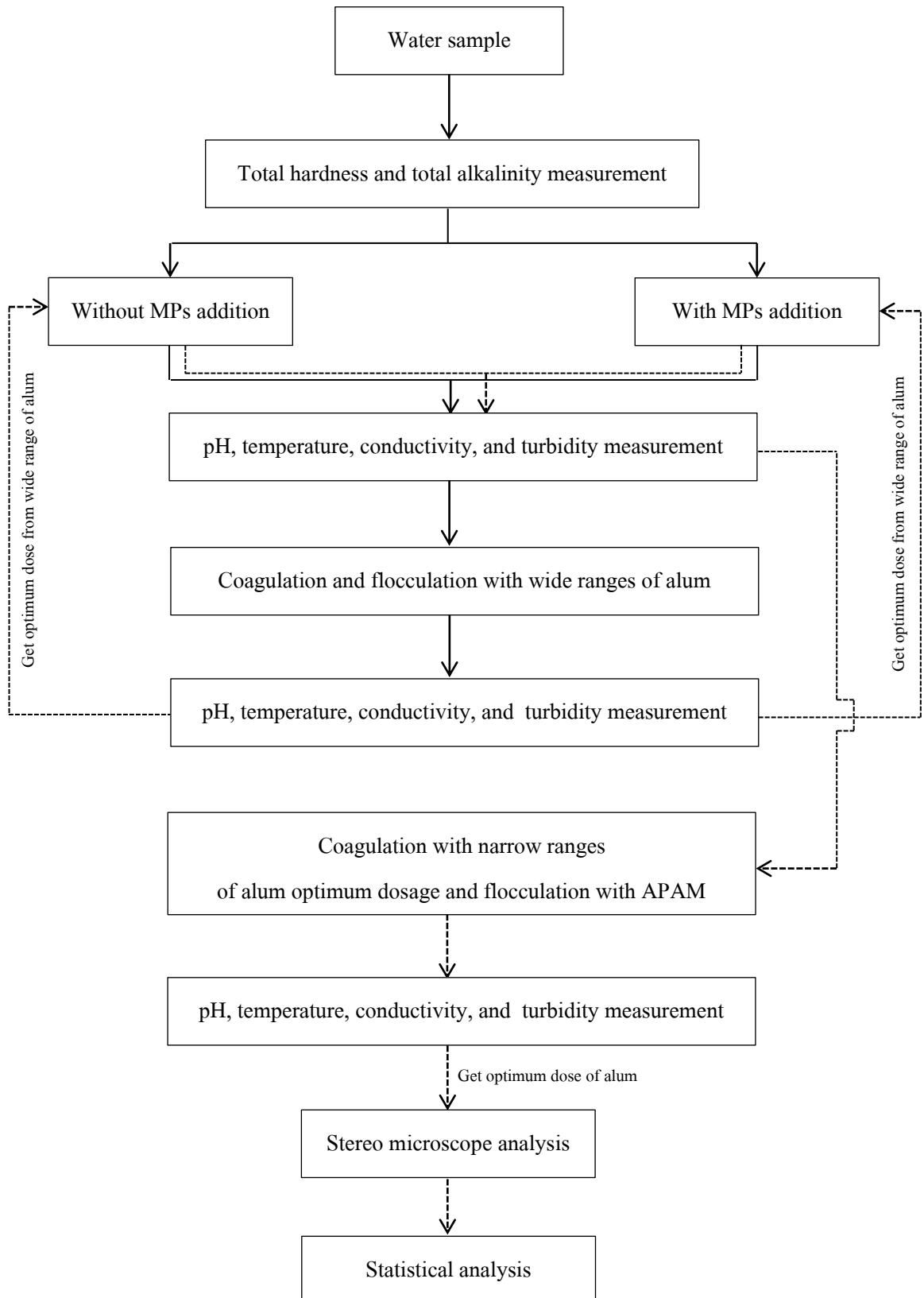


Figure 3.2 Flow chart of experimental procedure

Follow \longrightarrow for get optimum concentration from wide range of alum.

Follow \dashrightarrow for get optimum concentration from narrow range of alum.

3.2.1 Water sample

80 L of raw water was sieved at sampling site, using 100 μm , and 300 μm stainless steel sieves for MPs background analysis. Then, the 40 L water sampling was collected with stainless bucket from Khlong Prapa, Don Mueang, Bangkok, Thailand. The basic parameters for water production such as pH, temperature, turbidity, alkalinity, total hardness were measured. Then, MPs removal efficiency via coagulation and flocculation were approached.

3.2.2 Measurement of water quality

The standard methods to measure the water quality in this study are showed in Table 3.1.

Table 3.1 Measurement of the water sample quality (National Environmental Methods Index (NEMI); Tuntoollavest, 2008)

Parameter	Unit	Analytical Methods
Total alkalinity	mg/L as CaCO_3	Indicator method
Total hardness	mg/L as CaCO_3	EDTA titration method
pH	-	Electrometric method
Conductivity	$\mu\text{S}/\text{cm}$	Laboratory method
Turbidity	NTU	Nephelometric method



Figure 3.3 Laboratory instruments for measured (A) pH, (B) turbidity, and (C) conductivity.

3.2.3 Quantification of MPs

3.2.3.1 Collect MPs in raw water

- 1) Filtered the solid particles in the 80 L raw water through 100 and 300 μm meshes during the water sampling.
- 2) Collected the particles above sieve into beaker.
- 3) Rinsed sieve thoroughly using distilled water. Ensure all particles are well drained.
- 4) Covered beaker with aluminium foil.

3.2.3.2 Wet Peroxide Oxidation (WPO)

- 1) Added 20 mL of 0.05 M Fe (II) solution and 20 mL of hydrogen peroxide into the sample from the previous step.
- 2) Added stir bar into to the beaker and cover with a watch glass.
- 3) Left the solution at room temperature and mix without heat for 5 min in fume hood.
- 4) Heated the solution at 75 $^{\circ}\text{C}$ on a hotplate for 30 min.
- 5) Removed the beaker from the hot plate and add distilled water to slow the reaction until gas bubbles are observed at the solution surface.
- 6) Added 6 g of NaCl per 20 mL of sample.
- 7) Heated the solution until the NaCl dissolves.

3.2.3.3 Density separation

- 1) Transferred the solution from the previous step to the density separator.
- 2) Rinsed the beaker with NaCl solution to transfer all particles to the density separator.
- 3) Covered the density separator with aluminum foil and leave overnight.
- 4) Drained the settled solids from the density separator.
- 5) Added NaCl solution into the density separator until the solution level is the same as previous level.
- 6) Repeated steps 1) to 5) until the solution is clear and ensuring that separation occurs completely.

7) Took the solution from step 6) to filtrate using Büchner funnel with a PTFE 5 μm membrane filters.

3.2.3.4 Stereo microscope analysis

- 1) Took the filter with particles to stereo microscope.
- 2) Used 30X magnification to identify MPs.
- 3) Counted of MPs particles.

3.2.4 Coagulation and flocculation procedure

3.2.4.1 Optimum dose of alum investigation

- 1) Measured the initial pH, conductivity and turbidity the sample solution.
- 2) Added alum with different concentration i.e., 0, 5, 10, 15, 20, 25, 30, 35, 40, and 45 ppm. All concentrations were conducted 3 times.
- 3) Adjusted the mixing speed of jar test to 200 rpm for 1 min (rapid mixing).
- 4) Decreased the mixing speed to 40 rpm for 20 min (slow mixing).
- 5) Decreased the mixing speed to 0 rpm for 10 min (sedimentation).
- 6) Measured the final pH, conductivity and turbidity of the sample solution.

The optimum removal dose of alum was selected to repeat steps 1 to 6 with the narrow alum dose range.



Figure 3.4 Coagulation and flocculation experiment

3.2.4.2 Optimum dose of anionic PAM investigation

The optimum alum dose from step 1) was selected to find out the optimum anionic PAM as follow step below:

- 1) Measured the initial pH, temperature, conductivity and turbidity of the sample solution.
- 2) Added alum with optimum dose received from step 1).
- 3) Adjusted the mixing speed of jar tester to 200 rpm for 1 min (rapid mixing).
- 4) Added anionic PAM with different concentration i.e., 0.02, 0.04, and 0.06 ppm. All concentrations were conducted 3 times.
- 5) Decreased the mixing speed to 40 rpm for 20 min (slow mixing).
- 6) Decreased the mixing speed to 0 rpm for 10 min (sedimentation).
- 7) Measured the final pH, conductivity and turbidity of the sample solution.

3.2.5 MPs addition

The mixture of PE and PP particles were added in raw water to determine the treatment efficiency of MPs using coagulation and flocculation.

3.2.5.1 Made MPs smaller with mechanically method.

3.2.5.2 Soaked in wastewater from sugar industry for 1.5 months.

3.2.5.3 Sieved the particles with 100 and 300 μm meshes after 1.5 months.

3.2.5.4 Counted of MPs particles with stereo microscope, 20 particles per sample ($\text{NO}_{\text{before}}$).

3.2.5.5 Added the certain number of MPs particles into beaker.

3.2.5.6 Filled the beaker with 250 mL water sample and conducted the experiment according to 3.2.3.

3.2.6 Stereo microscope analysis

3.2.6.1 Removed the suspension with 25 mL syringe into glass bottle after step 3.2.4.

3.2.6.2 Filtered the solution by using Büchner funnel with a glass microfiber filters.

3.2.6.3 Took the filter with particles to stereo microscope.

3.2.6.4 Counted of MPs particles, after step 3.2.3, with stereo microscope

(NO_{after}).



Figure 3.5 Stereo microscope

3.2.7 Statistical analysis

Statistical analysis was performed using IBM SPSS Statistics 22 program and Microsoft Excel 2010. One-way ANOVA test, Dunnet's T3 test, Scheffe test, and Kruskal-Wallis test were performed to determine the optimum alum concentration and the optimum APAM concentration (significance level, 0.05).

CHAPTER IV

RESULTS AND DISCUSSION

This study focused on finding removal efficiency of 103 to 300 μm MPs via coagulation and flocculation process. Water samples were collected from Khlong Prapa, Don Mueang, Bangkok, Thailand. The sample was measured for total hardness, total alkalinity, pH, turbidity, and conductivity. The optimum doses of alum and APAM for coagulation and flocculation processes were provided to remove MPs. Other pollutants e.g., turbidity, pH and conductivity after treated were also measured. Moreover, current MPs in raw water were quantified before experiment. 80 L of raw water found 20 MPs particles, but only 250 mL of raw water was used in each sample. Therefore, we assumed that there were not MPs contamination in each sample. The results were presented and discussed in this chapter.

4.1 Characteristics of water sample

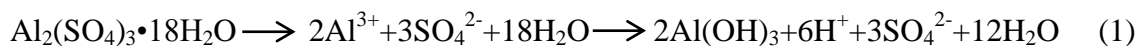
The physical properties of water sample were clear yellowish brown and had a slightly suspended sediment. Characteristics of water sample including total hardness and total alkalinity were measured by EDTA titration method and indicator method, respectively. Total hardness in water sample accounted for 115.72 mg/L as CaCO_3 which is considered as moderate (75–150 mg/L as CaCO_3) (Ministry of Industry, 1978). Typically, high hardness in water can cause clog in piping system (Budsareechai et al., 2016). Total alkalinity in water sample was 104.29 mg/L as CaCO_3 as in the recommended range (60-180 mg/L as CaCO_3) (The Association of Pool and Spa Professionals (APSP), 2014). Total alkalinity plays an important role in being a buffer that preventing the dramatically decrease in pH after the treatment process (Swenson and Baldwin, 1965).

4.2 Effects of alum

Alum with different concentration ranged from 0 to 45 ppm was added into water sample. The mixing speed of jar test was adjusted to 200 rpm for 1 min (rapid mixing), followed by 40 rpm for 20 min (slow mixing) and 0 rpm for 10 min (sedimentation), respectively. Finally, the suspension was collected to analysis.

4.2.1 Effects of alum on pH, conductivity, and turbidity

Parameters including pH, conductivity, and turbidity were measured before and after the experiment. The results showed that initial pH was ranged from 7.4 to 8.2 while the final pH was slightly decreased in ranged from 7.1 to 7.8. The decreasing in pH might be due to the effect of coagulant ($\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$) transforms to aluminium hydroxide, sulfate and hydrogen ions as shown in Eq. (1). The hydrogen ions react with the alkalinity in coagulation and flocculation processes resulting in the decrease of pH in treated water (Gebbie, 2006).



Conductivity before and after the experiment was ranged from 358 to 565 $\mu\text{S}/\text{cm}$ and 361 to 574 $\mu\text{S}/\text{cm}$. The slightly increase in conductivity after the treatment process might be due to the added alum which provided ionization as Eq. (1) resulting in the increase of Al^{3+} and SO_4^{2-} in treated water. It is according to the fact that the larger amount of electrolyte, the greater conductivity value (Munkwamdee et al., 2010).

One important role of coagulation and flocculation processes is turbidity removal. The initial turbidity was ranged from 20.40 to 32.00 NTU while the turbidity in treated water was dramatically decreased in ranged from 0.58 to 8.81 NTU (Figure 4.1). Moreover, Figure 4.1 shows the same removal efficiency trend between non-added MPs and added MPs set. The removal efficiency also initially increased when the concentration of alum was lower than 15 ppm, whereas it remained nearly stable when the concentration was higher than 15 ppm. The efficiency was slowly dropped when the concentration was higher than 35 ppm. The maximum turbidity removal efficiency was $97.87 \pm 0.03\%$ at alum 37 ppm.

This treatment process can remove turbidity from raw water with adsorption and charge neutralization mechanisms. Alum in raw water, which has positive charge, destroyed colloid stabilization, which has negative charge, resulting in neutralization of colloid. Colloid particles can gather together to become large and precipitated (PWA, 2015).

Sweep coagulation is one of the mechanisms used to remove turbidity with adding alum that can be adsorbed by colloid particles. As a result, colloid particles have more opportunity to link together until their weight is enough to settle resulting in lower level of turbidity in treated water (PWA, 2015).

In addition, Figure 4.1 shows the efficiency did not increase by increasing alum concentration in all cases, especially after 35 ppm. It is possible that excessive alum concentration might cause charge reversal resulting in re-stabilization of the suspended solids and decrease of turbidity removal efficiency.

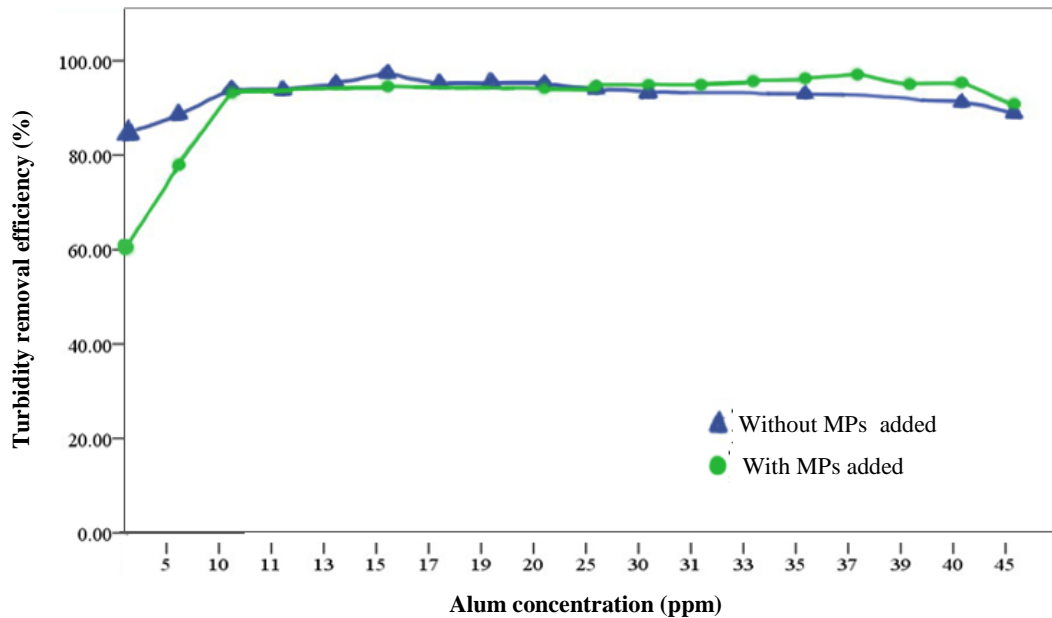


Figure 4.1 Effect of alum on turbidity removal efficiency

4.2.2 Effects of alum on MPs

20 MPs particles in size 103 to 300 μm were added into each water sample and alum with different concentration was then added to determine MPs removal efficiency. Initially, new MPs particles were used in this study, but they didn't suspend in water column. To solve this problem, MPs particles were then immersed in wastewater from sugar factories for approximately 1.5 months in order to organic matter adsorbed on the MPs particles surface, therefore MPs were heavy enough to be suspended in water. The result of MPs removal by alum illustrates in Figure 4.2. Figure 4.2 shows that the MPs removal efficiency increased when increased alum dose from 0 to 37 ppm and reached maximum $85.00 \pm 0.00\%$ at alum 37 ppm. The efficiency was dropped when alum more than 37 ppm. MPs removal efficiency in this work is higher than the study from Ma et al. (2019) which used coagulation by AlCl_3 to treat MPs in size <0.5 mm and found maximum removal efficiency of $36.89\% \pm 3.24\%$. However, that study used raw water as the water sample resulting in lower treatment efficiency.

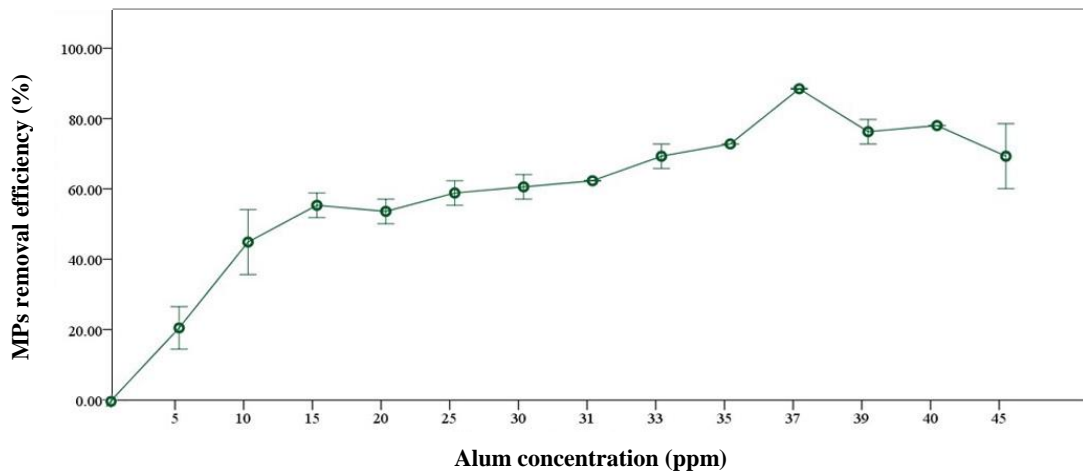


Figure 4.2 Effect of alum on MPs removal efficiency

Due to MPs used in this work appeared organic substances on surface area (Figure 4.3), these might react with the alum and provide flocs. The mechanism of MPs removal is the negative charge from organic matter deposited on the MPs surface being destabilized with positive charge of alum. As a result, it become neutral charges and has more opportunity to gather together into a larger particle, called floc, until their weight is enough to settle out of water (PWA, 2015). This mechanism is known as charge neutralization.

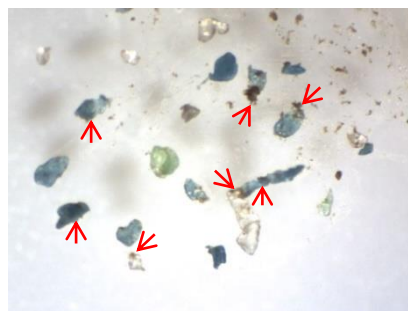


Figure 4.3 Organic substances on MPs surface area

Moreover, sweep coagulation mechanism which aluminum hydroxide settle down sweeping the MPs particles downward also occurs. Adding alum will produce white jelly-like crystals for colloidal particles to adsorb and forms flocs. As a result, their weight is enough to settle resulting in lower level of turbidity in treated water (PWA, 2015).

After reaching the optimum doses, excessive alum concentration might cause charge reversal resulting in re-stabilization of the suspended solids and decrease of MPs removal efficiency. Therefore, the MPs removal efficiency did not increase by

increasing alum concentration in all cases. In addition, added alum might only affect some areas on MPs surface where organic matter is attached, resulting in the decrease of MPs removal efficiency when compare to the turbidity removal efficiency.

The maximum MPs removal efficiency occurred at alum concentration 37 ppm, reached maximum $85.00\pm 0.00\%$. Therefore, 37 ppm was selected as an optimum alum concentration and used this concentration in the afterward experiment.

4.3 Effects of APAM

After the optimum alum concentration was discovered, the investigation of the optimum APAM concentration was conducted with optimum alum concentration. APAM with different concentration ranged from 0.00 to 0.06 ppm was added into water sample. The mixing speed of jar test was adjusted to 200 rpm for 1 min (rapid mixing), followed by 40 rpm for 20 min (slow mixing) and 0 rpm for 10 min (sedimentation), respectively. Finally, the suspension was collected to analysis.

4.3.1 Effects of APAM on pH, conductivity, and turbidity

Parameters including pH, conductivity, and turbidity were measured before and after the experiment. The results showed that pH was slightly decreased after the treatment process. The initial and final pH were ranged from 7.7 to 7.9 and 7.3 to 7.6, respectively.

Conductivity was slightly increased after the experiment. The initial conductivity was ranged from 352 $\mu\text{S}/\text{cm}$ to 360 $\mu\text{S}/\text{cm}$ while the final conductivity was ranged from 357 $\mu\text{S}/\text{cm}$ to 368 $\mu\text{S}/\text{cm}$. Aside from the effect of adding alum as Eq. (1), the slightly increase in conductivity after the treatment process might be due to the added APAM which provided negative charges into the water. It is according to the fact that the larger amount of electrolyte, the greater conductivity value (Munkwamdee et al., 2010).

For effects of APAM on turbidity, the results showed dramatically decrease of turbidity after the treatment process. The initial turbidity and final turbidity were ranged from 28.10 to 30.00 and 0.50 to 0.61 NTU, respectively. The removal efficiency was quite stable and reached maximum of $98.25\pm 0.04\%$ from APAM 0.06 ppm (as shown in Figure 4.4).

Adding alum in raw water destroyed colloid stabilization resulting in neutralization of colloid and colloid particles have more opportunity to link together. Then, adding APAM provided formation of a bridge among flocs called polymer bridging

mechanism that makes the bigger floc. Floccs link together until their weight enough to settle resulting in increased turbidity removal (PWA, 2015). When compared to adding alum alone, adding alum and APAM together provided more effective on turbidity treatment ($97.87\pm 0.03\%$ and $98.25\pm 0.04\%$, respectively).

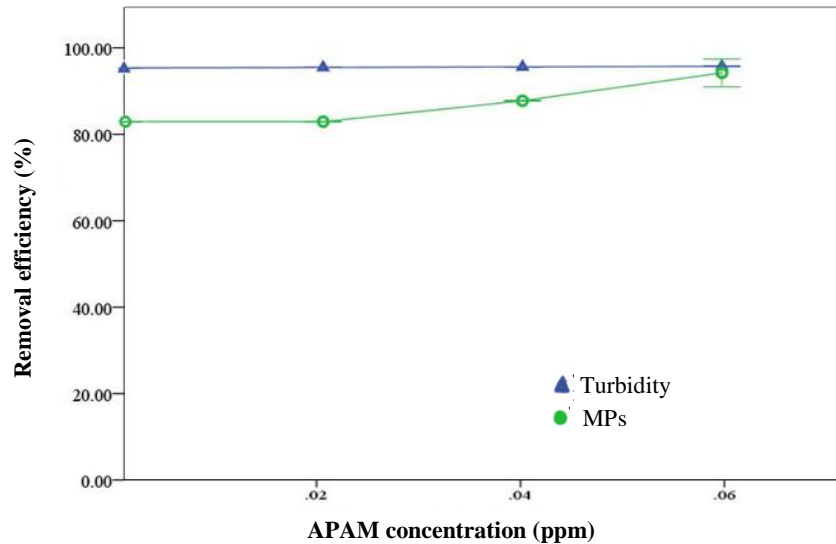


Figure 4.4 Effect of APAM on MPs removal efficiency and turbidity removal efficiency

4.3.2 Effects of APAM on MPs

In this section, 20 particles of 103 to 300 μm MPs particles were added into the water sample. Optimum alum concentration with different concentration of APAM i.e., 0.00, 0.02, 0.04, and 0.06 ppm was added into raw water to determine the ability of APAM for improved MPs removal efficiency. The results showed that adding of APAM with optimum alum concentration can enhance the MPs removal efficiency (Figure 4.4). The removal efficiency remained at $85.00\pm 0.00\%$ for APAM dose 0.00 to 0.02 ppm. Then, there was a steadily risen in removal efficiency between APAM dose 0.04 to 0.06 ppm. A number of MPs particles in treated water was 3.00 ± 0.00 , 3.00 ± 0.00 , 2.00 ± 0.00 , and 0.67 ± 0.33 particles/250 mL water. In another word, the MPs removal efficiencies were $85.00\pm 0.00\%$, $85.00\pm 0.00\%$, $90.00\pm 0.00\%$, and $96.67\pm 1.67\%$ for 0.00, 0.02, 0.04, and 0.06 ppm of APAM concentration, respectively. The maximum MPs removal efficiency made up $96.67\pm 1.67\%$ at 0.06 ppm APAM. This removal efficiency is higher than the study of Ma et al. (2019) which revealed that MPs in size <0.5 mm in raw water presented removal efficiency of $61.19\% \pm 3.67\%$ by 15 ppm PAM.

The mechanisms of MPs removal are charge neutralization and sweep coagulation by alum, and polymer bridging by APAM. MPs floc, which provided by alum,

can be affected by adding APAM. APAM can provide formation of a bridge among flocs that make the bigger floc. Flocs link together until their weight is enough to settle, resulting in increased MPs removal (PWA, 2015). When compared to adding alum alone, adding alum and APAM together provided more effective treatment on MPs (85.00±0.00% and 96.67±1.67%, respectively).

Note that Kruskal-Wallis test with a significance level of 0.05 showed no significant difference of MPs removal between APAM doses 0.04 and 0.06 ppm. This means it recommends using APAM 0.04 ppm, which has a removal efficiency of 90.00±0.00% in real situations.

CHAPTER V

CONCLUSIONS AND RECOMMENDATIONS

5.1 Conclusions

5.1.1 Characteristics of water sample i.e., pH, total hardness and total alkalinity were 115.72 mg/L as CaCO₃ and 104.29 mg/L as CaCO₃, respectively.

5.1.2 After the treatment process, alum and APAM affected the water parameters including slightly decrease of pH, slightly increase of conductivity, and dramatically decrease of turbidity. In addition, alum and APAM affected in dramatically decrease of MPs, especially optimum concentration of both alum and APAM.

5.1.3 The maximum removal efficiency of 103 to 300 µm MPs by adding alum was 85.00±0.00% at 37 ppm. And adding alum with APAM provided the maximum MPs removal efficiency made up 96.67±1.67% at 37 ppm alum with 0.06 ppm APAM.

5.1.4 Adding alum and APAM together provided more effective on turbidity and 103 to 300 µm MPs treatments compared to alum alone (the maximum turbidity removal efficiencies were 98.25±0.04% and 97.87±0.03%, respectively, and the maximum MPs removal efficiency were 96.67±1.67%, and 85.00±0.00, respectively).

5.1.5 The optimum alum and APAM concentration for the treatment of 103 to 300 µm MPs via coagulation and flocculation process were 37 ppm and 0.04 ppm, respectively. The average MPs removal efficiency of adding 37 ppm alum with 0.04 ppm APAM together accounted for 90.00±0.00%.

5.2 Recommendations

5.2.1 A container used to store water sample for MPs analysis should be glass or aluminium in order to prevent MPs contamination from the container. Due to lighter weight and cheaper price, aluminium container is more recommended.

5.2.2 Initial pH is one of the main parameters to control coagulation and flocculation processes, adjusting pH before experiment should be an option to improve MPs removal efficiency.

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APPENDIX A



Figure A.1 MPs particles (A) polypropylene and (B) polyethylene



Figure A.2 Mechanically method for made MPs smaller



Figure A.3 MPs particles after mechanically method



Figure A.4 MPs particles after immersed in wastewater from sugar factories

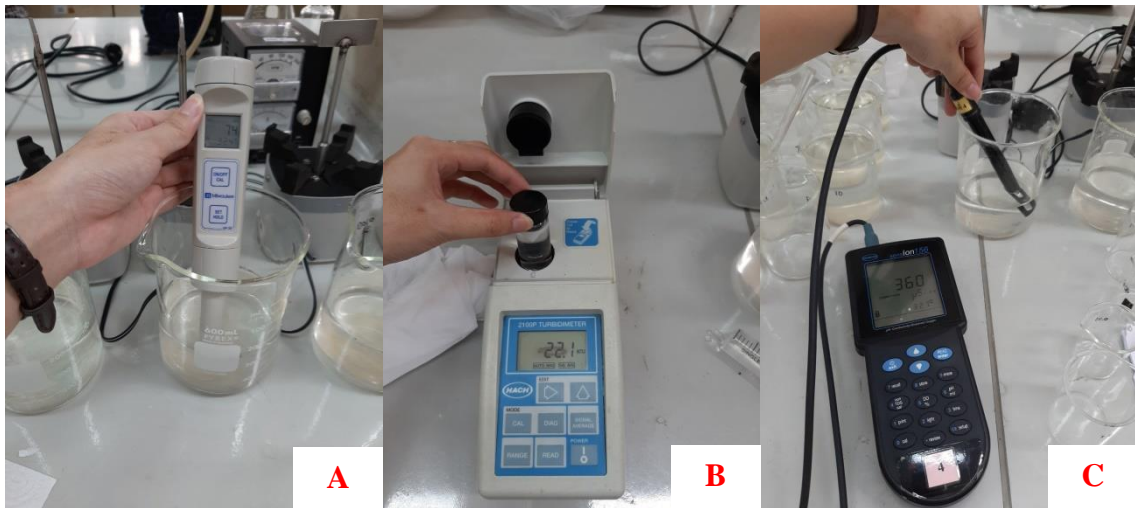


Figure A.5 Measurement of parameter (A) pH, (B) turbidity, and (C) conductivity



Figure A.6 Collection of suspension for MPs analysis with stereo microscope

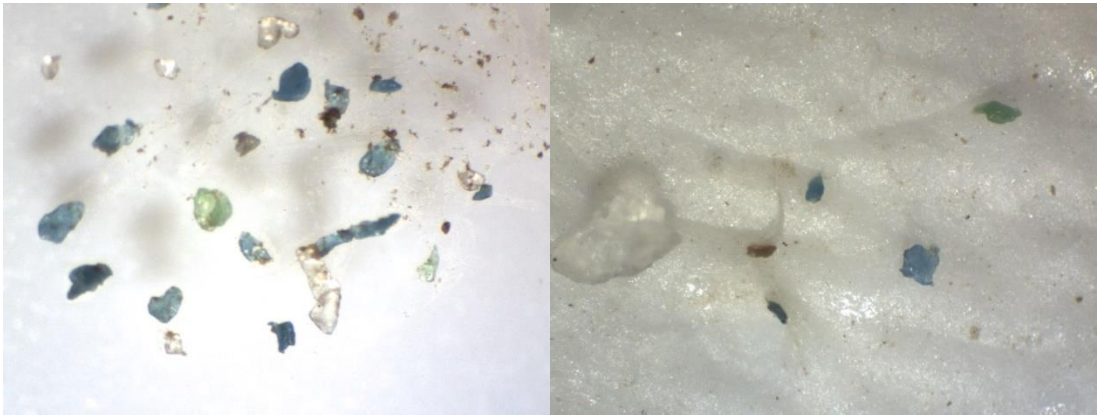


Figure A.7 MPs from stereo microscope analysis

APPENDIX B

Table B.1 MPs removal efficiency by adding alum (narrow range alum concentration)

Alum concentration (ppm)	Sample NO.	MPs particle NO. (particles/ 250 mL)	MPs removal efficiency (%)	Avg. MPs removal efficiency (%)	Std. Deviation	Std. Error	
0	1	20	20	0	0.00	0.00	0.00
	2		20	0			
	3		20	0			
31	1		8	60	60.00	0.00	0.00
	2		8	60			
	3		8	60			
33	1		7	65	66.67	2.89	1.67
	2		6	70			
	3		7	65			
35	1	6	70	70.00	0.00	0.00	
	2	6	70				
	3	6	70				
37	1	3	85	85.00	0.00	0.00	
	2	3	85				
	3	3	85				
39	1	5	75	73.33	2.89	1.67	
	2	5	75				
	3	6	70				

Table B.2 MPs removal efficiency by adding APAM

APAM concentration (ppm)	Sample NO.	MPs particle NO. (particles/ 250 mL)	MPs removal efficiency (%)	Avg. MPs removal efficiency (%)	Std. Deviation	Std. Error
0.00	1	20	3	85	85.00	0.00
	2		3	85		
	3		3	85		
0.02	1		3	85	85.00	0.00
	2		3	85		
	3		3	85		
0.04	1		2	90	90.00	0.00
	2		2	90		
	3		2	90		
0.06	1	1	95	96.67	2.89	
	2	0	100			
	3	1	95			

APPENDIX C

Table C.1 Determine the optimum alum concentration*

Sample 1 - Sample 2	Test Statistic	Std. Error	Std. Test Statistic	Sig.
31 - 33	-3.667	3.552	-1.032	0.302
31 - 35	-6.000	3.552	-1.689	0.091
31 - 39	-8.333	3.552	-2.346	0.019
31 - 37	-12.000	3.552	-3.378	0.001
33 - 35	-2.333	3.552	-0.657	0.511
33 - 39	-4.667	3.552	-1.314	0.189
33 - 37	-8.333	3.552	-2.346	0.019
35 - 39	-2.333	3.552	-0.657	0.511
35 - 37	-6.000	3.552	-1.689	0.091
39 - 37	3.667	3.552	1.032	0.302

Table C.2 Determine the optimum APAM concentration*

Sample 1 - Sample 2	Test Statistic	Std. Error	Std. Test Statistic	Sig.
0.00 - 0.02	0.000	2.730	0.000	1.000
0.00 - 0.04	-4.500	2.730	-1.648	0.099
0.00 - 0.06	-7.500	2.730	-2.747	0.006
0.02 - 0.04	-4.500	2.730	-1.648	0.099
0.02 - 0.06	-7.500	2.730	-2.747	0.006
0.04 - 0.06	-3.000	2.730	-1.099	0.272

*Each row tests the null hypothesis that the Sample 1 and Sample 2 distributions are the same. Asymptotic significances (2-sided tests) are displayed. The significance level is 0.05.

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