EFFECTIVENESS OF TRIHALOMETHANE (THM) PRECURSORS REMOVAL IN GROUNDWATER USING COAGULANT-DISINFECTANT

by

NUR IZYAN BINTI MUHAMAD JEFFRI

This dissertation is submitted to

UNIVERSITI SAINS MALAYSIA

as partial fulfillment of requirements for the degree of

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(ENVIRONMENTAL ENGINEERING)

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LIST OF ABBREVIATIONS

CDC	Centers for Disease Control and Prevention
$CHBr_3$	Bromoform (BF)
$CHCl_3$	Chloroform (CF)
$CHBrCl_2$	Bromodichloromethane (BDCM)
$CHClBr_2$	Dibromochloromethane (DBCM)
DBPFP	Disinfection by-products Formation Potential
DBPs	Disinfection by-products
DOC	Dissolved Organic Carbon
EPA	Environmental Protection Agency
$FeCl_3$	Ferric Chloride
$Fe_2(SO_4)_3$	Ferric Sulphate
HAAs	Haloacetic Acids
HPAC	High Efficient Composite Polyaluminium Chloride
HSE	Health Service Executive
IARC	International Agency for Research on Cancer
MOH	Ministry of Health
MW	Molecular Weight
NOM	Natural Organic Matter
PACl`	Polyaluminium Chloride
SUVA	Specific Ultra Violet Absorbance
THMs	Trihalomethanes
TTHMFP	Total Trihalomethane Formation Potential
TOC	Total Organic Carbon

UK	United Kingdom
USEPA	United States Environmental Protection Agency
UV_{254}	UV absorbance at 254nm
WHO	World Health Organization
WTP	Water Treatment Plant

ABSTRAK

Trihalometana (THM) merupakan salah satu produk sampingan disinfeksi (DBP) yang terbentuk hasil tindak balas bahan pembasmi kuman dengan bahan organik semulajadi dalam rawatan air minuman. Bahan organik semulajadi perlu dihapuskan melalui proses rawatan air. Oleh itu, kajian ini telah menggunakan serbuk komersial penggumpal (ferik sulfat) - disinfektan (kalsium hipoklorit) bagi merawat air dan ujian balang juga telah dijalankan untuk mengetahui keberkesanan penyingkiran bahan organik semulajadi di dalam air bumi dengan mengaplikasikan dua jenis bahan penggumpal yang berbeza dengan disinfektan. Dalam kajian ini, ferik sulfat $Fe_2(SO_4)_3$ dan ferik klorida ($FeCl_3$) telah digunakan sebagai bahan penggumpal. Sampel air telah diuji untuk beberapa parameter seperti tahap kekeruhan, kandungan karbon organik terlarut (DOC), UV_{254} and SUVA. Pertamanya air bumi telah dirawat menggunakan serbuk komersial penggumpal-disinfektan. Dengan penggunaan serbuk komersial ini, ia dapat mengurangkan tahap kekeruhan sebanyak 28.36% - 48.68% manakala kadar peratusan penyingkiran terhadap UV_{254} adalah di antara 12.12% - 34.11% di mana ia menunjukkan kadar peratusan lebih rendah berbanding dengan peratusan penyingkiran tahap kekeruhan di dalam sampel air. Selain itu, penggunaan serbuk komersial ini mendapati bahawa DOC menunjukkan kadar peratusan penyingkiran yang sangat rendah iaitu di antara 2.14% - 12.94%. Disebabkan peratusan penyingkiran yang rendah, masih berlakunya tindak balas di antara disinfektan dan bahan organik semulajadi yang menyebabkan terbentuknya THM di dalam air. Menurut Kementerian Kesihatan Malaysia, kloroform dan bromodiklorometana menunjukkan pembentukan THM melebihi kadar yang telah ditetapkan di mana masing-masing melebihi 0.200 mg/L dan 0.060 mg/L. Penggunaan penggumpal berasaskan ferik, telah menunjukkan

peratusan penyingkiran lebih tinggi jika dibandingkan dengan penggunaan serbuk komersial penggumpal-disinfektan. Ferik sulfat menunjukkan bahawa ia sesuai digunakan untuk tujuan penyingkiran kekeruhan di mana peratusannya mencapai 50.51% manakala ferik klorida pula lebih baik untuk tujuan penyingkiran UV_{254} di mana peratusannya mencapai sebanyak 80.76%. Di dalam kajian ini, terdapat beberapa kondisi pH dan kandungan penggumpal telah di uji. Di antara kesemua kondisi pH dan kandungan bahan penggumpal yang telah dikaji, di tahap pH 5.5, THM telah menunjukkan pembentukan paling rendah untuk kedua-dua jenis penggumpal jika dibandingkan dengan tahap kondisi pH yang lain tetapi masih melebihi tahap yang ditetapkan oleh Kementerian Kesihatan Malaysia. Kesan daripada lebihan pembentukan THM di dalam air akan memberi impak negatif kepada kesihatan manusia di mana mereka berpotensi menghidap penyakit kanser jika mereka mengambil atau menggunakan air tersebut dalam tempoh yang berterusan.

ABSTRACT

Trihalomethanes (THMs) is one of disinfection by-products (DBPs) groups and it will form due to the reaction between disinfectant and NOM in treated drinking water. Thus, as THM precursors, NOM needs to be removed through water treatment process. Therefore, this study applied commercialized coagulant-disinfectant powder, and carried out jar test applying two ferric-based coagulants, simultaneously with disinfectant, both to determine the effectiveness of THM precursors removal from groundwater. The coagulants applied in this study were, ferric sulphate and ferric chloride. Selected water quality parameter such as turbidity, DOC, UV_{254} and SUVA were tested for all water samples. For the first part, groundwater was treated by using commercialize coagulant-disinfectant powder. By using this commercialized powder, about 28.36% - 48.68% of turbidity has been removed while for UV_{254}, range of the percentage removal obtained was between 12.12% - 34.11% which was less than turbidity removal. Besides, it was found that, DOC had a very poor removal, only 2.14% - 12.94%. By using this commercialized powder, chloroform and bromodichloromethane were found to exceed the limit specified by Ministry of Health, greater than 0.200 mg/L and 0.060 mg/L, respectively. On the other hand, the use of ferric-based coagulant provided a higher percentage removal for all parameters compared to commercialize coagulant-disinfectant. Ferric sulphate showed to have a good turbidity removal, the highest at 50.51% while ferric chloride is great in UV_{254} removal, as the percentage removal obtained reached until 80.76%. From the various conditions of pH and coagulants dose tested, at pH 5.5, THMs formed less for both coagulants compared to other pH levels, though all those values still exceeded the limit. Previous study found that,

there will be adverse health effects to human which includes potential to get cancer disease due to long term use/consumption of water with elevated THM level.

CHAPTER 1

INTRODUCTION

1.1 Background

Water is a need in human daily life, however due to human activities; the clean water portion available is decreasing by day. It is important to maintain the quality of clean water for many purposes such as supplying drinking-water, production of food and water usage purposes (WHO, 2011). Human and living organisms will be the most affected if clean water supply keeps on depleting. Based on a previous research, high consumption of contaminated water was a major problem as it affected people with many diseases. In addition, to the quantity of water provided, it is also related to the quality of municipal drinking water (Wright et al., 2017).

Natural organic matter (NOM) exists in the environments, such as in soils, sediments, water and even at atmosphere where it consists of very complex mixtures of substances arised from an initial biological source (Beckett and Ranville, 2006). An increased amount of NOM in water may give poor effects on drinking water treatment (Matilainen et al., 2010). Furthermore, NOM is the major contributor as it can create a lot of problems such as odour, taste, colour, and bacteria regrowth in the distributions line that are used to distribute water to consumers (Yan et al., 2009). Besides, NOM or organic precursors may exist in different forms; either dissolved or particulate form, but in another recent studies found that NOM can also occur in an intermediate colloidal state (Beckett and Ranville, 2006).

In Malaysia, chlorination has been used as primary disinfectant to manage and reduce formation of pathogens in drinking water and water treatment plant (WTP) (Chang et al., 2010). Chlorine is known as a great oxidizing agent where it can destroys all complex organic molecules (HSE and EPA, 2011). Besides, chlorine also can be used as a primary disinfectant in water treatment process in order to maintain the quality of the water along the distribution line. Chlorinated drinking water will inactivate bacteria to prevent the spread of water borne diseases. However, chlorinated drinking water may form by-products compounds such as THMs which consist of four compounds, and is widely known that THMs is the most prevalent group formed during chlorination process (Hua and Reckhow, 2007).

THM is one of the chlorinated DBP groups which comprises of four compounds; chloroform ($CHCl_3$), bromoform ($CHBr_3$), bromodichloromethane ($CHBrCl_2$) and dibromochloromethane ($CHClBr_2$) (Kanchanamayoon, 2015). Furthermore, THMs were classified as dominant species of by-products and followed by HAAs (Rizzo et al., 2005). In Kanchanamayoon (2015), THMs was categorized as Cancer Group B which can cause cancer in laboratory animals (Kanchanamayoon, 2015). THMs also were reported in having bad effects on birth. In addition, these by-products were classified as carcinogenic group where bladder cancer was the common disease to be reported (Shafiee et al., 2012). In the study carried out by Shafiee et al. (2012), the results show that there was a positive correlation between exposure to disinfection by-products in drinking water and human bladder cancer. About 9% of all cases of bladder cancer were attributed to chlorinated drinking water and disinfection by-products. (Shafiee et al., 2012).

According to World Health Organization, chloroform is the dominant THM compound that forms generally in chlorinated water originating from surface water than in groundwater. This is due to high organic matter in surface water (WHO, 2005). In addition, amongst all four THMs compound, chloroform was able to form in high concentration and had strong correlation with mortality rate from bladder cancer while brain cancer was due to presence of brominated-THM levels (Chang et al., 2010). Therefore, many organizations have set certain regulations to limit the concentration of these THM compounds in drinking water due to the potential human health concerns of DBPs. THMs is categorized as carcinogenic group; specified by Environmental Protection Agency (EPA) (Chang et al., 2010). According to Ministry of Health, Malaysia, the maximum allowable limit for TTHMs in drinking water is 1 mg/L and it should not exceed the limit in order to have a safe-reliable drinking water.

In order to limit the occurrence of THMs in drinking water, organic precursors should be removed during water treatment; this is usually characterized by total organic carbon (TOC), dissolved organic carbon (DOC), UV absorbance at 254 nm wavelength (UV_{254}) or specific UV absorbance (SUVA) parameters (Rizzo et al., 2005). Referring to USEPA, percentage removal for total organic carbon to control THMs formation by enhanced coagulation involved pH control (Rizzo et al., 2005). In Hong et al. (2013), a study was conducted on factors affecting DBPs formation and the results obtained showed that THMs and other chlorinated DBPs formations significantly increased with high contact time, higher chlorine dosage and bromide levels, but less formation at high amount of nitrite contents.

1.2 Problem Statement

In Malaysia, chlorination has been applied as primary disinfectant but in the same time, it will form disinfection by-products in water especially THMs and haloacetic acids (HAAs). These DBPs are well known and are classified as carcinogenic group (Richardson et al., 2007). Previous research have been carried to remove/reduce DBPs in water by using alternative disinfectant such as chloramines, chlorine dioxide which resulted to a lower chlorinated DBPs formation (Hua and Reckhow, 2007).

According to Center for Disease Control and Prevention, there were cases of coagulant-disinfectant simultaneous application to treat water during disasters but it may cause a problem when disinfectant react with organic precursor that exist in the water and form elevated THM in water. A commercialized coagulant-disinfectant powder was introduced by the ‘P’ company to treat water in a very short time on small volumes of water (Crump et al., 2005). This can be applied especially when there are emergency cases such as natural disasters (flooding and tsunami). This powder has many benefits as many microorganisms can be reduced by using this powder such as removal of bacteria, viruses, parasites heavy metals and suspended organic matter in treated water (Crump et al., 2005). The ingredients of this powder are commonly used in municipal water treatment plants such as ferric sulfate, bentonite, sodium carbonate, chitosan, polyacrylamide, potassium permanganate and calcium hypochlorite (Reller et al., 2003).

Even though the effectiveness of this simultaneous coagulant-disinfectant was proved in improving the water quality in terms of clarity and microorganisms content, there was no previous study reported on the effects of the DBP formations.

There are possibilities that elevated DBPs will occur when using this method of treatment. Thus, there is a need to understand if it is indeed will elevate the DBP levels, what is the best conditions to control the DBP formation in order to satisfy the limits stated in the national drinking water guidelines. Therefore, in this study, the focus is to determine the effectiveness of this commercially available ferric sulphate-calcium hypochlorite (coagulant-disinfectant) in removing THM precursors in groundwater. In addition, this study also compares the use of different ferric-based coagulants, ferric sulphate and ferric chloride simultaneously with disinfectant in removing THM precursors.

1.3 Objectives

The objectives of this research are:

i. To determine the effectiveness of commercially available ferric sulphate-calcium hypochlorite (coagulant-disinfectant) in removing THM precursors.

ii. To compare THM precursors removal using different ferric-based coagulant, ferric sulphate and ferric chloride with disinfectant in removing THM precursors.

1.4 Scope of Work

This study focused on the effectiveness of using coagulant-disinfectant simultaneously for water treatment. Groundwater samples were collected at a borehole located in Universiti Sains Malaysia, Engineering Campus, for four times. This site is known from a previous study to contain high organic matter and it produced bad taste and odour. Water samples were tested for THMs concentration and also a few selected water quality parameters which were DOC, UV at 254 nm wavelength, and turbidity level. In addition, parameter pH and water temperature were also tested on site while the remaining three parameters were tested in the laboratory. The equipment used was YSI equipment (Model 556 MPS), Jar Test, UV Spectrophotometer (GENESYS 10S UV-VIS), Gas Chromatography Mass Spectrometry (GC-MS).

1.5 Dissertation Outline

This thesis contains five chapters. Chapter 1 consist of an introduction to the study, which is split up into background study, problem statement, objectives, scope of work, organization of the thesis and importance of the study.

Next in Chapter 2, the literature review of the study covers various subtopics such as groundwater characteristics, potential health effects of DBPs on human, previous occurrence of DBPs, previous study applying commercialized coagulant-disinfectant powder, principles of coagulation and flocculation, coagulant, THMs, NOM in source water, impact of water quality parameters on DBPs and disinfectant. This chapter explains more on previous study of DBPs and effectiveness of NOM removal in several countries. In Chapter 3, it explains the details of methodology of this study and about the details of field test and laboratory work which consist of water sampling, jar test, sample extraction and THM analysis.

Then, in Chapter 4, the results and discussion of the data collected was presented in table including elaborations. The discussions include the impact of each parameter in water after adding different coagulant. This chapter also explains the correlation of each parameter selected in order to determine the optimum results. Lastly, Chapter 5 contains the conclusion and recommendations that may be useful for future studies.

1.6 Importance and Benefit of Study

This study was carried out to determine the effectiveness in removing THM precursors by treating groundwater using commercially available ferric sulphate-calcium hypochlorite (coagulant-disinfectant). The purpose of using coagulant and disinfectant is to neutralize the charges on particles and deactivate or killing of pathogenic microorganisms respectively. This kind of product is used to treat water especially in rural area as it low cost and easy to bring.

Other than to determine the effectiveness, the comparison by using different coagulants (ferric chloride and ferric sulphate) was also applied in removing THM precursors. For this study, the precursors need to be removed as it will give major contribution to THM formation. THM is a carcinogenic group which gives adverse health effects to human as human need to consume more water in daily life.

By referring to Drinking Water Quality Standard stated by Ministry of Health, maximum allowable limit for TTHM levels is 1 mg/L. If it exceeds the limit given, there is high possibility for human to have serious disease such as cancer as THM were classified as carcinogenic group. Furthermore, it leads to a higher death rate.

CHAPTER 2

LITERATURE REVIEW

2.1 Groundwater Characteristics

Groundwater is defined as valued fresh water resources and it is used for drinking purposes all over the world (Chabukdhara et al., 2017). Groundwater also contains about more than half of the fresh water on earth. There are water quality parameters that are useful to be measured in order to determine groundwater characteristics which divided into three categories, chemical, physical and bacterial characteristics (Otieno et al., 2012).

Chemical characteristics of groundwater includes the determination of organic and inorganic constituent concentration, while physical is measuring the level of turbidity, colour, temperature, taste and odour (Otieno et al., 2012). Besides, physical analysis parameters for groundwater characteristics determination are shown in Table 2.1, where it stated that the undesirable effect produced towards the water quality parameters. For example, high content of heavy metals may resulted in poor taste and high turbidity (Otieno et al., 2012).

Furthermore, contamination of groundwater becomes a major issue in developing countries, as they lack in clean water (Chabukdhara et al., 2017). Over the past 50 years, people from South and Southeast Asia were shifted from consuming surface water to groundwater for drinking purposes and this has resulted in a reduction of people being affected by water borne pathogens (Bacquart et al., 2015).

Table 2.1: Drinking Water Quality
(Source from Otieno et al., 2012)

Parameters	Undesirable effect produced
A. Physical	
Colour	Discolouration odour
Odour	Taste
	Gastrointestinal
Total Solids	Irritation
Suspended Matter	Turbidity
	Gastrointestination
	Irritations
B. Chemicals	
pH	Taste, excessive scale formation
Calcium	Taste, corrosion in hot water system
Chloride	Mottling of teeth Disfiguring of skeletons
Fluoride	Excessive scale formation
Total Hardness	Taste odour
Mineral oil	Taste
Phenolics subs. Toxic	Toxic
C. Trace elements	
Copper	A stringent taste, discolouration, corrosion of pipe fittings and utensils
Cyanide	Toxic
Iron	Taste, discolouration constipation turbidity growth of bacteria
Lead	Toxic
Manganese	Taste, discolouration, turbidity, deposits in pipes
Zinc	A stringent taste
D. Pesticides	
DDT	Toxic
PCB	Toxic