OPTIMISATION OF COAGULATION PROCESS IN WATER TREATMENT PLANT USING STATISTICAL APPROACH

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A thesis submitted in fulfilment of the
requirements for the award of the degree of
Doctor of Philosophy (Civil Engineering)

Faculty of Civil Engineering
Universiti Teknologi Malaysia

JUNE 2016

I declare that this thesis entitled "*Optimisation Of Coagulation Process In Water Treatment Plant Using Statistical Approach*" is the result of my own research except as cited in the references. This thesis has not been accepted for any degree and is not concurrently submitted in candidature of any other degree.

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Love, Thoughts and Du’a to
my late mak (Raja Arbaiah) and abah (Hj Zainal Abideen);
my beloved wife, Dzurina;
my children, Iffah, Harraz, Asyraf and Ammar;
my lovely makcik (Hjh Jamaliah) and mama (Hjh Rokiah).

ACKNOWLEDGMENTS

In the name of Allah, the Most Gracious, the Most Merciful. All the praises and thanks are to Allah, the Lord of the *Alamin*. May His peace and blessings be upon His messenger, Prophet Muhammad *Sallahu Alaihi Wassalam*, his family and his companions.

First and foremost, I would like to express my gratefulness to Allah *Azza wa Jalla* for giving me the ability and strength to complete my doctorate study. I sincerely extend gratitude and appreciation to my main supervisor, Professor Dr. Azmi bin Aris, for very many reasons, especially for his guidance, advice, patience and words of wisdom. His confidence in me and his undivided support given over countless days and nights, have been very important for me in completing this long and tumultuous task. My thanks also go to Associate Professor Dr. Zaiton binti Abdul Majid and Associate Professor Dr. Fadhilah binti Yusof for serving as co-supervisors. Their help and commitment in the discussion and interpretation of results presented in this thesis are immensely invaluable.

I would also like to take this opportunity to thank my parents and my parents-in-law for their love and support throughout my life. My wholehearted love and thanks are also dedicated to my beloved wife, Dzurina; the apples of my eye, Iffah, Harraz, Asyraf and Ammar; for their continuous du'a, sacrifice and undivided support throughout this study. My beloved friends all over the world and especially in Johor, whom I have considered to be my brothers and sisters, have helped me a lot, physically and emotionally. They, as well as all family members deserve my utmost thanks too.

My appreciation also goes to Syarikat Air Johor Holdings for providing fund and permitting me to carry out my study at the Bandar Tenggara, Sungai Gembut and Sri Gading Water Treatment Plants. I would also like to thank the Ministry of Higher Education (MOHE) for generously awarding me the scholarship. Last but not least, I would like to thank Universiti Teknologi Malaysia, and especially the Faculty of Civil Engineering and Faculty of Science, for allowing me to use their laboratories, as well as to all who contributed directly or indirectly in accomplishing this thesis.

ABSTRACT

The typical jar test practice at water treatment plants to determine the optimum coagulation conditions is statistically inappropriate as it is based on one-factor-at-a-time (OFAT) approach. This approach, which does not cover the whole experimental space, could miss the actual optimum values results in higher cost and less effective coagulation performance. Additionally, the jar test exercise is time consuming and is therefore conducted once a day or when it is really needed. This study carried out a long-term comparison between the use of Response Surface Method (RSM) and the traditional (a modified OFAT) jar test in optimising the coagulation process. The study was carried out at Bandar Tenggara WTP (BTWTP), Sungai Gembut WTP (SGWTP) and Sri Gading WTP (SRGWTP) as these WTPs have different raw water characteristics. Characterisation of natural organic matters (NOMs) in terms of dissolved organic carbon (DOC) and UV_{254} and their removal via coagulation process were explored. The relationship between the raw water characteristics and coagulation optimum conditions were also developed. The results showed that the RSM and traditional approaches acquired almost identical optimum coagulation conditions at BTWTP and SGWTP. At SRGWTP, RSM technique was found to be better than the traditional method as the coagulant (alum) and flocculant (polymer) optimum concentrations produced by the RSM technique were respectively 50% and 20% of the optimum values obtained by the traditional method. The raw waters of BTWTP and SGWTP were identified as hydrophilic, non-humic, with low molecular mass of NOMs. At optimum coagulation conditions (based on turbidity removal), the average DOC and UV_{254} removals were about 11% and 70%, respectively. The models predicting optimum pH and coagulant dosing from raw water quality data were formulated from the RSM and traditional approach and was found to be reliable. From the models, it was found that NH_3–N in the raw water at BTWTP and turbidity, Mn and NH_3–N in the raw water at SGWTP did not affect the optimum coagulation conditions. The detail optimisation model derivation procedures were also developed.

ABSTRAK

Ujian balang yang biasa digunakan untuk menentukan keadaan optimum koagulasi adalah tidak tepat secara statistik kerana ia menggunakan pendekatan satu-faktor-dalam-satu-masa (OFAT). Pendekatan yang tidak merangkumi keseluruhan ruang eksperimen ini, mungkin terlepas di dalam mendapatkan nilai optimum sebenar yang menyebabkan peningkatan kos serta menghasilkan prestasi koagulasi yang kurang efektif. Tambahan lagi, pelaksanaan ujian balang agak mengambil masa yang panjang dan oleh itu, ia hanya dilakukan sehari sekali atau apabila berkeperluan. Kajian ini membuat perbandingan jangka panjang di antara penggunaan ujian balang berasaskan Kaedah Permukaan Maklumbalas (RSM) dengan ujian balang berasaskan kaedah tradisional (OFAT Terubahsuai) di dalam mengoptimumkan proses koagulasi. Kajian dilaksanakan di Loji Olahan Air Bandar Tenggara (BTWTP), Sungai Gembut (SGWTP) dan Sri Gading (SRGWTP) kerana loji-loji ini mempunyai ciri-ciri air mentah yang berbeza. Pencirian bahan organik semulajadi (NOM) dalam bentuk karbon organik terlarut (DOC) dan UV_{254} serta penyingkirannya melalui proses koagulasi juga diterokai. Hubungan di antara ciri-ciri air mentah dan keadaan optimum koagulasi juga dihasilkan. Berdasarkan pemerhatian, kaedah RSM dan tradisional yang dijalankan di BTWTP dan SGWTP memberikan keadaan optimum yang hampir serupa. Di SRGWTP, kaedah RSM yang dilaksanakan didapati lebih baik daripada kaedah tradisional kerana dos optimum koagulan (alum) dan flokulan (polimer) yang diperoleh daripada kaedah RSM adalah masing-masing 50% dan 20% daripada dos optimum yang diperoleh melalui kaedah tradisional. Air mentah dari BTWTP dan SGWTP dikenalpasti mengandungi NOM yang hidrofilik, bukan humik dan rendah jisim molekulnya. Pada keadaan koagulasi yang optimum (berasaskan penyingkiran kekeruhan), purata penyingkiran DOC dan UV_{254} adalah masing-masing sebanyak kira-kira 11% dan 70%. Model-model yang dijanakan untuk meramalkan hubungan di antara dos koagulan dan pH optimum dengan data kualiti air mentah melalui pendekatan RSM dan tradisional, didapati boleh dipercayai. Menerusi model-model ini, kandungan NH_3–N di dalam air mentah di BTWTP dan kekeruhan, Mn dan NH_3–N di dalam air mentah di SGWTP ini tidak memberi kesan terhadap keadaan optimum koagulasi loji terbabit. Perincian tentang prosedur penerbitan model pengoptimuman juga dihasilkan di dalam kajian ini.

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LIST OF ABBREVIATIONS

BBD	– Box Behnken Design
BTWTP	– Bandar Tenggara WTP
CCD	– Central Composite Design
COD	– Chemical Oxygen Demand
CRD	– Completely Randomised
DOC	– Dissolved Organic Carbon
DoE	– Design of Experiment
FCD	– Face-centred Central Composite Design
FCS	– Ferric Chloride Sludge
G	– Velocity Gradient
IC	– Inorganic Carbon
nC_{60}	– nanoscale colloidal product of carbon fullerene
NOM	– Natural Organic Matters
OFAT	– One-Factor-at-A-Time
PAC	– Polyaluminium chloride
PACS	– Polyaluminium chloride Sludge
RCBD	– Randomised Complete Block
RMSE	– Root Mean Square Error
RSM	– Respond Surface Method
SGWTP	– Sungai Gembut Water Treatment Plant
SRGWTP	– Sri Gading Water Treatment Plant
SUVA	– Specific Ultra-Violet Absorbance
SVI	– Sludge Volume Index
TC	– Total Carbon
TOC	– Total Organic Carbon

TSS – Total Suspended Solids

USEPA – United States of America Environmental Protection Agency

UV_{254} – Ultra-violet 254 nm

WTP – Water Treatment Plant

LIST OF SYMBOLS

α – constants associated with raw water quality

β – constants associated with raw water quality

γ – constants associated with raw water quality

X1 – pH

X2 – turbidity

X3 – colour (apparent colour)

X4 – Iron (Fe)

X5 – Aluminium (Al)

X6 – temperature

X7 – Manganese (Mn)

X8 – Ammonium (NH_3–N)

X9 – Total Dissolved Solids (TDS)

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CHAPTER 1

INTRODUCTION

1.1 Preamble

The presence of colloids and natural organic matters (NOM) in raw water sources excessively can cause a lot of problems. During intensive raining period, excessive soil erosion occurs and this increases the concentration of colloids and subsequently the raw water surface turbidity will multiply considerably. Having high turbidity for a raw water source may disturb the operation at the water treatment plant (WTP) and can be very costly (Chang and Liao, 2012). A huge amount of chemicals and coagulants must be used to ensure the turbidity is removed until it reaches the allowable turbidity for drinking water of less than 5 NTU outlined by the National Standard for Drinking Water Quality (NSDWQ) (Ministry of Health, 2004). Large pre-sedimentation basins may need to be constructed prior to the upstream of a WTP to remove the colloids. High concentration of sediments trapped in these basins needs to be treated too (Lin *et al.*, 2008).

The NOM is known to cause colour (Jiang and Graham, 1998; Fabris *et al.*, 2008; Matilainen *et al.*, 2010), unpleasant taste and odour problems in surface water (Matilainen *et al.*, 2010). It also acts as substrate for microbial and causes bacterial regrowth problem (Yan *et al.*, 2008b; Liang and Ma, 2009) and able to block membranes and activated carbon pores for adsorption site hence reducing adsorption efficiency (Fabris *et al.*, 2008; Matilainen *et al.*, 2010) in water supplies. More importantly, they have been reported to be one of the precursors for disinfection by-products (DBPs) (Amirtharajah and O'Melia, 1990; McGhee, 1991; Edzwald, 1993; Iriarte-Velasco *et al.*, 2007b) that may cause cancer to human (Bob and Walker, 2001; Abdullah *et al.*, 2003; Matilainen *et al.*, 2010; Chow *et al.*, 2011; Fooladvand *et al.*, 2011; Gan *et al.*, 2013).

Looking at the adverse impacts of colloids and NOM, it is therefore essential to remove these impurities. Coagulation is known to be an effective method to remove the colloidal particles of high and intermediate molecular weight that result in high turbidity (Lin *et al.*, 2008; AlMubaddal *et al.*, 2009) and NOM (Fabris *et al.*, 2008). It is an established agglomeration process to transform small particles into larger aggregates (flocs), which involves addition of chemicals. These chemicals will destabilise kinetically stable suspensions such as dissolved and colloid impurities hence producing larger flocs that can be removed at the clarification or filtration stage (Jiang and Graham, 1998; Gao *et al.*, 2002).

The most widely used chemical in this process is aluminium (Al) based coagulant. Though Al based coagulant is known to be effective in coagulation process, it raises serious concerns due to the increase of residual Al in treated water. Among the problems associated with the presence of Al in potable water are it may increase the turbidity owing to the formation of Al precipitates, raise the pressure of water distribution network, inhibit to the effect of disinfection and damage human's nervous system (Jiao *et al.*, 2015). Medical reports state that aluminium may induce Alzheimer's disease too (Zouboulis *et al.*, 2004; Kimura *et al.*, 2013).

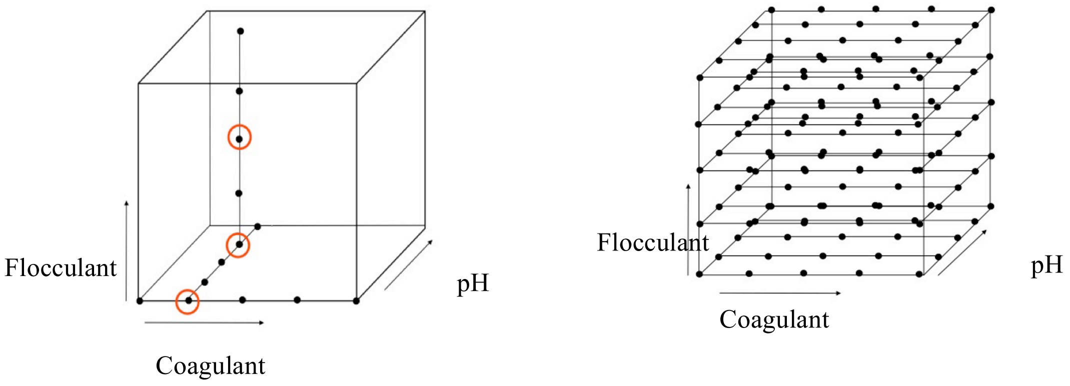
Therefore, coagulation process must be carried out in an optimum way as it has direct impact on the reliability of the treatment plant operations and the final water quality. It also has significant contribution to the operational cost of the treatment plant. Any Al concentrations in treated water detected to be greater than 0.3 mg/L shows a lack of optimisation in coagulation process of the conventional water treatment method (Qaiyum *et al.*, 2011). The effectiveness of the process is highly dependent on several factors including pH of the operation and dosage of coagulant and coagulant aids.

1.2 Problem Statement

Over the years, jar test is the most common method to determine the best pH and dosage of the chemicals used in the coagulation process. It is normally carried out at the WTP once or twice a day or whenever any significant changes occurred in the raw water due to many factors including the change in the weather conditions and contaminant intrusion from the urbanization activities. Since the raw water quality parameters are continuously changing over time resulting the optimum doses of pH and chemical should be changing as well, it is insufficient to carry out the jar test at the said rate above.

In the current jar test approach, the WTP operator determines the best pH and chemical dosages by systematically changing the level of the factors, that are pH and dosage, one at a time while holding the level of other factors constant. The level of that factor which results in the best response (for example, lowest turbidity value) is then selected and used in subsequent tests which continue in the same manner on other factors. While the approach, termed as One-Factor-at-A-Time (OFAT) (Figure 1.1 (a)) is rather straightforward, it suffers from shortcomings which may lead to a wrong conclusion. It is time-consuming and does not fully explore the space of possible solutions and hence, it is incapable of identifying the interaction effects resulted from the factors being considered (Montgomery, 2005). Due to these reasons, the current jar test approach could have missed the actual best pH and dosage that are possibly hidden in the experimental space not covered by the OFAT approach.

To overcome this problem, a better method to acquire the best pH and chemical dosage is to adopt an experimental approach as shown in Figure 1.1 (b). In this approach, all levels of factors are altered systematically (one-by-one) to ensure that the experimental space is fully covered. The best response could not possibly be missed through this approach. Nonetheless, it is very time-consuming and costly.



(a) (b)

Figure 1.1: Design of Experiment; (a) OFAT Approach and (b) All Factors Varied Systematically Approach

Response surface methodology (RSM) jar test has been proposed to overcome these problems. The RSM is a statistical technique for designing experiment and involves relatively small number of experiments. It is able to build mathematical models from the outcome of the experiments and search for the optimum conditions for desirable factors and responses. With RSM, the interaction between factors and responses can also be determined (Montgomery, 2005).

RSM has been used in many optimisation experiments (Wang *et al.*, 2007, Pinzi *et al.*, 2010; Moradi and Ghanbari, 2014). Ahmad *et al.* (2005) managed to explain the effect and interaction between coagulant dose, flocculant dose and pH when treating palm oil mill effluent (POME) through coagulation-flocculation process incorporated with membrane separation technology. Wang *et al.* (2007) optimised the coagulation-flocculation process to achieve minimum turbidity and sludge volume index (SVI) for paper-recycling wastewater treatment. Pinzi *et al.* (2010) used RSM to optimise the transesterification reaction for several types of vegetable oils. Moradi and Ghanbari (2014) applied RSM technique to treat landfill leachate and successfully obtained the optimum conditions for an integrated coagulation-fenton process in removing the chemical oxygen demand, colour and total suspended solids of the leachate. Despite the application of RSM in many experimental studies, its usage in optimising coagulation conditions through jar test in water treatment is apparently uncommon.

The raw water quality at any WTP keeps changing very frequently but the jar test is only carried out once or twice a day. Therefore, the optimum chemical doses obtained through the jar test are only valid for the period of sampling. It is impractical to carry out the jar test whenever the quality of the water changes, as it is very time-consuming. Therefore, there is a need to develop statistical models relating the historical data of a WTP's raw water quality parameters and the best coagulation conditions acquired through jar test. Once the statistical models are established, the optimum chemical doses may be known almost instantaneously by plugging the raw water parameters data into the models. The generated statistical models may also be used to monitor or check whether the chemical doses used in the plant are at optimum condition. The steps taken to produce the statistical models has got to be properly recorded and verified so that a standard procedure to develop the statistical models relating raw water quality parameters and optimum doses of chemical used for any WTP may be established.

Even though the negative effects of NOM present in water and how coagulation process is able to remove NOM are widely known, no maximum concentration limit for NOM is stated in the National Standard for Drinking Water Quality (NSDWQ) (Ministry of Health, 2004). It is believed that due to this fact, NOM are hardly analysed at any WTPs in Malaysia even though simple technique such as the analysis of dissolved organic carbon (DOC) and the ultra-violet 254 nm absorbance (UV_{254}) may be applied to characterise the NOM present in the WTP's raw water. Hence, the performance of coagulation process carried out daily at any WTP in removing NOM has never been explored by most, if not all WTPs in Malaysia, unlike turbidity that is clearly spelt out in the NSDWQ.

1.3 Objectives of Study

The objectives are as follows:

i) To carry out a long-term comparison between the optimum coagulation conditions obtained from the traditional (an adjusted OFAT) jar test with the one obtained from the RSM jar test for different sources of raw water

ii) To develop statistical models that correlate the relationship between raw water quality parameters normally analysed at WTP and the best coagulation conditions in terms of pH, chemical dosing and residual aluminium concentration.

iii) To characterise the NOM present in the raw water source in terms of its affinity towards water as well as to quantify the removal of NOM based on typical traditional jar test carried out at the WTP.

iv) To establish a standard procedure to obtain statistical model that correlates optimum coagulation conditions and raw water quality parameters for any WTP.

1.4 Scope of Study

The coagulation process in this study focussed solely on the water treatment process. The research was carried out for three Syarikat Air Johor Holdings Sdn. Bhd. (SAJ) WTPs that have different characteristics of raw water. These WTPs were Bandar Tenggara WTP (high turbidity water), Sungai Gembut WTP (yellowish-orange coloured acidic water) and Sri Gading WTP (low turbidity acidic water). The traditional and RSM jar tests were carried out at the WTPs by the operators on a daily basis for a minimum of six months and at Universiti Teknologi Malaysia (UTM) on weekly basis. Raw water quality parameters analysed daily at these WTPs were pH, turbidity, colour (apparent colour), ammonia nitrogen (NH_3-N), iron (Fe), aluminium (Al), manganese (Mn) and other parameters that suited the requirement of the WTPs. Parameters analysed for the jar tests were coagulant dose (and coagulant aid dose for Sri Gading WTP), pH (during initial setting for the jar test and after flocs settlement), water turbidity and residual aluminium after conducting the jar tests. The NOM present at the WTP was characterised by their specific ultra-violet absorbance (SUVA) values through the analysis of the dissolved organic carbon (DOC) and the ultra-violet 254 nm absorbance (UV_{254}). Design Expert Version 7.1 and Statistical Package for the Social Sciences Version 10.0 (SPSS) softwares were used to assist the results analyses.

1.5 Significance of Study

The study is important as it develops a more systematic approach to overcome the shortcomings of the traditional jar test typically carried out at most WTPs in Malaysia. In Malaysia, most WTPs have adopted an adjusted OFAT jar test whereby only the coagulant dosing is systematically altered and not the coagulant aid nor the pH of the coagulation. This is because these WTPs are more concerned on the amount of coagulant used as compared to any other chemicals since its usage is considerably higher than the others. The adjusted OFAT jar test has definitely neglected the determination of optimum values of pH and coagulant aid and hence it will not lead to the actual best values for these entities. As for the optimum coagulant, the jar test does not fully explore the experimental space of possible solutions and hence, it may miss the actual optimum value too. The wastage of chemicals used in the coagulation process at these WTPs is unavoidable.

The RSM jar test experiment is implemented in this study to overcome the stated problems earlier as RSM has been proven to be very successful in optimisation experiments (Wang *et al.*, 2007, Pinzi *et al.*, 2010; Moradi and Ghanbari, 2014). RSM involves small number of experiments, builds mathematical models from the outcome of the experiments and searches the optimum conditions for desirable factors and responses. With RSM, the interaction between factors and responses can also be determined (Montgomery, 2005). This study also eases the determination of optimum pH and chemicals dosage of coagulation process in producing acceptable water quality through the establishment of statistical models relating the raw water quality parameters and the jar test optimum conditions.

NOM characteristics present in the raw waters for two WTPs in Johor in terms of their affinity towards water and their interaction with coagulation process will be determined conducted by the traditional jar test at these WTPs. This study also determines the characteristics of the NOM present in the raw water for several WTPs in terms of their affinity towards water and how it interacts with the coagulation process carried out by the traditional jar test at these WTPs. Finally, this study establishes a standard procedure to obtain statistical model that correlates optimum coagulation conditions and their raw water quality parameters for any WTP.

1.6 Thesis Outline

This thesis is divided into six chapters. Chapter 1 is an introduction chapter. It contains the problem statement, objectives, scope and the significance of the study as well as the thesis outline. Chapter 2 consists of the literature review on coagulation process, jar test, NOM, design of experiment (DoE) and RSM topics. Chapter 3 describes the research methodology in detail followed by Chapter 4. In Chapter 4, the analysis of water characteristics, long-term comparison on the traditional and RSM jar tests, the generation of regression models relating the optimum jar test conditions with the raw water quality parameters and NOM removal through traditional jar test are related. Chapter 5 relates the standard procedure obtained from this study to acquire statistical model that correlates optimum coagulation conditions and their raw water quality parameters for any WTP. Chapter 6, the final chapter of the thesis, presents the conclusions and recommendations for future study.

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