



UNIVERSITI PUTRA MALAYSIA

***ENHANCEMENT OF PRIMARY TREATMENT PROCESS FOR
DOMESTIC WASTEWATER USING TANNIN-BASED COAGULANT***

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DOMESTIC WASTEWATER USING TANNIN-BASED COAGULANT**

By

YASIR TALIB HAMEED

**Thesis Submitted to the School of Graduate Studies, Universiti Putra Malaysia,
in Fulfillment of the Requirements for the Degree of Doctor of Philosophy**

November 2017

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Abstract of the thesis presented to the Senate of Universiti Putra Malaysia in fulfillment of the requirement for the Degree of Doctor of Philosophy

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Coagulation and flocculation as a pre-treatment before biological process is one of the options to enhance the treated water quality and drive possible savings in the construction and operation of treatment plants.

The common coagulants such as Al^{3+} and Fe^{3+} have been used extensively for long time. However, they are known to act as an additional burden to the environment. Furthermore, there is a public health risk from the use of Al^{3+} . Because of that, great efforts have been made to provide environmentally friendly alternatives to conventional coagulants and flocculants. One of these alternatives is a tannin-based coagulant and flocculant with the name Tanfloc.

The aim of this study was to improve the performance of a biofilm process by pre-treating the wastewater using Tanfloc and to study the effect of extended use of Tanfloc on the microbial community of the biofilm.

To achieve these objectives, a five-stage experiment was conducted. In the first stage, chemical characteristics of Tanfloc were determined using FTIR and EDX in addition to determination of Tanfloc biodegradability. Moreover, jar test experiments were conducted to compare the performance of Tanfloc to Polyaluminium chloride (PAC). In the second stage, a preliminary study was conducted on Tanfloc performance in a continuous flow experiment using only flocculation and sedimentation units. In the third stage, the biofilm unit in the continuous flow experiment was run and Tanfloc effects were evaluated on the three units. Flocculation process was evaluated by studying floc size and residual turbidity. Primary clarifier was evaluated by

determining the removal efficiencies. Finally, aeration tank was evaluated by studying treatment efficiency and dissolved oxygen level. When third stage has finished, results were analysed and they were not clear to show the effect of Tanfloc. Consequently, fourth stage using a smaller aeration tank has been decided to be conducted. In the fifth stage, the effect of Tanfloc on the biofilm community was investigated in a specific study of biofilm characteristics. Effect of Tanfloc on the percentage of bacterial genera was studied in addition to substrate concentration and dissolved oxygen.

The outcomes of the first stage showed that Tanfloc can compete with PAC as a flocculant. While Tanfloc achieved 85%, 60% and 64% removal efficiencies for TSS, BOD₅ and COD, the efficiencies were 64%, 55% and 55% for PAC. The improvement in floc size for Tanfloc compared to PAC improved turbidity removal, Tanfloc removed 70% of the turbidity within only 2 minutes, compared to 42% for PAC. The outcomes of the third and fourth stage showed that even at short flocculation time (7.5 min), Tanfloc showed a high potential to form big flocs with a size distribution of d (10), d (50) and d (90) of 18, 42 and 96 micron. Enhancement of the clarification process due to Tanfloc application was very clear and while the efficiency of TSS removal in the clarifier was only 4% at a flow of 18 L/min (HRT = 55.5 min), with Tanfloc it achieved a 60% efficiency. Even at a high flow of 26 L/min (HRT= 39 min), a removal efficiency of 31% was achieved when Tanfloc was applied. An enhancement in aeration tank performance was noticed due to Tanfloc's effect on reducing the organic load; the BOD₅ for the treated water dropped from the range of 24 – 50 to the range of 7–24 mg/L when Tanfloc was introduced. Moreover, the dissolved oxygen level in the aeration tank jumped almost to double the value when Tanfloc was introduced to the biological process. An interesting point in the results of the fifth stage is the ammonia nitrogen removal. In the experiment without Tanfloc, there was a complete inhibition of ammonia nitrogen removal at retention time of 4 hours, while Tanfloc produced a removal efficiency of around 70% of the ammonia nitrogen at the same retention time (4 hours). Biofilm community analysis showed a significant increment in the percentage of *Nitrosomonas* and *Nitrospira* genera in the biofilm cultured by flocculated water (3.33% and 7.8% respectively) compared to the biofilm cultured by raw wastewater (0.073% and 0.19 % respectively). This increase justified and confirmed the aforementioned improvement in ammonia nitrogen removal in the experiment with Tanfloc.

The aforementioned results suggest Tanfloc as a promising agent to enhance the performance of clarification and biological treatment units and consequently reduce the required volumes of treatment units and saving energy. In light of this enhancement, Tanfloc could be used to upgrade the existing treatment plants or design compact treatment units.

Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia sebagai memenuhi keperluan untuk Ijazah Doktor Falsafah

**PENAMBAHBAIKAN PROSES RAWATAN PRIMER UNTUK AIR SISA
DOMESTIK MENGGUNAKAN KOAGULAN BERASASKAN TANNIN**

Oleh

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Pembekuan dan pemberbukuan sebagai pra-rawatan dalam proses biologiikal ialah salah satu pilihan untuk menambah baik kualiti air terawat dan penjimatan dalam pembinaan dan operasi loji rawatan.

Koagulan konvensional seperti Al^{3+} dan Fe^{3+} telah digunakan secara meluas untuk jangka masa yang lama. Akan tetapi, unsur-unsur ini diketahui menyumbang kepada beban tambahan terhadap alam sekitar. Oleh kerana itu, banyak usaha telah dibuat untuk menyediakan alternatif kepada koagulan dan flokulan konvensional yang lebih mesra alam. Salah satu yang menjadi pilihan adalah koagulan dan flokulan berasaskan tannin di bawah nama Tanfloc.

Tujuan kajian ini ialah untuk meningkatkan prestasi proses biofilem melalui pra-rawatan air sisa dengan menggunakan Tanfloc dan juga mengkaji kesan lanjutan penggunaan Tanfloc terhadap komuniti mikrobial biofilem.

Untuk mencapai objektif-objektif ini, satu eksperimen lima peringkat telah dijalankan. Dalam peringkat pertama, ciri-ciri kimia untuk Tanfloc telah ditentukan melalui FTIR dan EDX sebagai tambahan kepada penentuan tahap biodegradasi Tanfloc. Tambahan pula, eksperimen ujian balang telah dijalankan untuk membandingkan kecekapan Tanfloc berbanding Polialuminium klorida (PAC). Dalam peringkat kedua, satu kajian awal telah dilakukan terhadap kecekapan Tanfloc dalam loji pandu dengan hanya menggunakan unit pemberbukuan dan pemendapan. Dalam peringkat ketiga, unit biofilem di dalam loji pandu telah dijalankan dan kesan-kesan Tanfloc terhadap loji pandu telah dinilai. Proses pemberbukuan telah dinilai melalui penelitian terhadap saiz

flok dan saki-baki kekeruhan. Penjernih primer telah dinilai berdasarkan kecekapan penyingkiran. Pada akhirnya, tangki pengudaraan telah dikaji melalui kecekapan rawatan dan paras oksigen terlarut. Apabila peringkat ketiga telah selesai, hasil-hasil kajian telah dianalisis dan ianya masih tidak jelas untuk menunjukkan kesan Tanfloc. Bertitik tolak daripada itu, peringkat keempat dengan menggunakan tangki pengudaraan yang lebih kecil telah diputuskan untuk dijalankan. Dalam peringkat kelima, kesan Tanfloc terhadap komuniti biofilem telah dikaji dalam eksperimen berskala kecil. Kesan Tanfloc terhadap peratusan genera bakteria telah dikaji sebagai tambahan kepada kepekatan substrat dan oksigen terlarut.

Hasil daripada peringkat pertama menunjukkan Tanfloc menyaingi PAC sebagai flokulan. Ketika Tanfloc mencapai 85%, 60% dan 64% kecekapan penyingkiran untuk TSS, BOD₅ dan COD, kecekapannya ialah 64%, 55% dan 55% untuk PAC. Peningkatan saiz flok dengan menggunakan Tanfloc berbanding PAC meningkatkan penyingkiran kekeruhan. Tanfloc menyingkirkan 70% daripada kekeruhan dalam masa hanya 2 minit, berbanding 33% untuk PAC. Hasil-hasil kajian peringkat ketiga dan keempat menunjukkan bahawa walaupun pada masa pemberbukan yang singkat (7.5 min), Tanfloc berpotensi besar untuk membentuk flok yang besar dengan taburan saiz d (10), d (50) dan d (90) dengan nilai 18, 42 dan 96 mikron. Penambahbaikan proses penjernihan melalui aplikasi Tanfloc adalah sangat jelas. Biarpun kecekapan penyingkiran TSS di dalam penjernih ialah hanya 4% pada kadar aliran 18 L/min (HRT = 55.5 min), 60% berjaya dicapai apabila Tanfloc digunakan. Walaupun pada kadar aliran yang tinggi pada 26 L/min (HRT = 39 min), 31% kecekapan penyingkiran telah dicapai apabila Tanfloc digunakan. Penambahbaikan dalam prestasi tangki pengudaraan telah dikesan hasil daripada kesan Tanfloc terhadap pengurangan beban organik, BOD₅ untuk air terawat jatuh daripada julat 24 – 50 kepada julat 7 – 24 mg/L apabila Tanfloc digunakan. Tambahan pula, paras oksigen terlarut di dalam tangki pengudaraan menginjak naik hampir dua kali ganda (ia mencecah had 6 mg/L) apabila Tanfloc digunakan dalam proses biologi. Apa yang menarik dalam hasil peringkat kelima ialah data dalam penyingkiran ammonia nitrogen. Dalam eksperimen tanpa Tanfloc, terdapat perencatan sepenuhnya terhadap penyingkiran ammonia pada masa tahanan 4 jam, sedangkan kecekapan penyingkiran ammonia nitrogen telah mencapai 70% pada masa tahanan yang sama (4 jam). Analisis komuniti biofilem menunjukkan kenaikan ketara dalam peratusan *Nitrosomonas* dan *Nitrospira* genera di dalam biofilem dibiakkan melalui air yang berbuku (masing-masing 3.33% dan 7.8%) berbanding biofilem dibiakkan melalui air sisa mentah (masing-masing 0.073% dan 0.19%). Kenaikan ini mewajarkan dan mengesahkan peningkatan dalam penyingkiran ammonia nitrogen dalam eksperimen dengan Tanfloc seperti dinyatakan di atas.

Hasil-hasil kajian yang dinyatakan di atas mencadangkan Tanfloc sebagai ejen berpotensi untuk meningkatkan prestasi unit penjernihan dan rawatan biologi serta mengurangkan isipadu yang diperlukan untuk unit rawatan dan menjimatkan tenaga. Berdasarkan penambahbaikan ini, Tanfloc boleh digunakan untuk menaik taraf loji rawatan sedia ada atau unit rawatan bereka bentuk kompak.

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“ IN THE NAME OF ALLAH THE MOST GRACIOUS MOST MERCIFUL”

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This thesis was submitted to the Senate of Universiti Putra Malaysia and has been accepted as fulfilment of the requirement for the degree of Doctor of Philosophy. The members of the Supervisory Committee were as follows:

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TABLE OF CONTENTS

	Page
ABSTRACT	i
ABSTRAK	iii
ACKNOWLEDGEMENTS	v
APPROVAL	vi
DECLARATION	viii
LIST OF TABLES	xiv
LIST OF FIGURES	xvii
LIST OF ABBREVIATIONS	xx
 CHAPTER	
1 INTRODUCTION	1
1.1 Introduction	1
1.2 Research Background	1
1.3 Problem Statement	2
1.4 Research Objectives:	3
1.5 Scope of the Study	4
1.6 Thesis layout	4
 2 LITERATURE REVIEW	6
2.1 Introduction	6
2.2 Domestic Wastewater Sources	6
2.3 Domestic Wastewater Characteristics	6
2.4 Effluent Standards	7
2.5 Wastewater Treatment Levels	8
2.6 Coagulation and Flocculation of Particles in Wastewater	9
2.7 Destabilization Mechanism	10
2.7.1 Charge Neutralization	10
2.7.2 Polymer Bridge Formation	11
2.7.3 Electrostatic Patch	13
2.7.4 Enmeshment in Sweep Flocc	13
2.8 Application of Coagulation and Flocculation in Water Treatment	14
2.8.1 Surface Water	14
2.8.2 Domestic Wastewater	16
2.8.2.1 Raw Domestic Wastewater	16
2.8.2.2 Treated Domestic Wastewater	17
2.8.2.3 Phosphate Removal from Domestic Wastewater	18
2.8.3 Algae Removal from Water and Algae Harvesting	20
2.8.4 Textile Wastewater	22
2.8.5 Food Industry Wastewater	22
2.8.6 Others	23
2.8.7 Types of Coagulants and Flocculants	31
2.8.7.1 Inorganic Coagulants and Flocculants	31

2.8.8	Natural Coagulants and Flocculants	31
2.9	Tanfloc as Natural Coagulant	40
2.10	Summary	44
3	GENERAL METHODOLOGY	45
3.1	Introduction	45
3.2	Research Design	45
3.3	Materials	48
3.3.1	Chemicals	48
3.3.2	Raw Water	48
3.3.3	Biofilm Carrier	49
3.3.4	The Continuous Flow Experiment (process (A))	49
3.3.4.1	Raw Water Tank	50
3.3.4.2	Dosing Pump	50
3.3.4.3	Coagulation Tank	51
3.3.4.4	Flocculation Tank	51
3.3.4.5	Primary Clarifier	51
3.3.4.6	Aeration Tank	51
3.3.4.7	Secondary Clarifier	52
3.3.5	The Continuous Flow Experiment (process (B))	52
3.3.6	Specific Study of Biofilm Characteristics	53
3.3.6.1	Holding Tank (200 L)	53
3.3.6.2	Storage Tanks	53
3.3.6.3	Peristaltic Pumps	54
3.3.6.4	Aeration Tanks	54
3.3.6.5	Holding Tanks (25 L)	54
3.4	Experiment Stages	55
3.4.1	First Stage (lab experiments)	55
3.4.2	Second Stage (preliminary study)	56
3.4.3	Third Stage (effect of Tanfloc on the performance of process (A) / aeration Tank is 2850 L)	56
3.4.3.1	Acclimatization steps of aeration tank	56
3.4.3.2	Sampling Points	57
3.4.3.3	Sampling Frequency	57
3.4.4	Fourth Stage (effect of Tanfloc on the performance of process (B) / aeration tank is 1250 L)	58
3.4.5	Fifth Stage (the effect of prolonged use of Tanfloc on the biofilm community / two aeration tanks each is 25 L)	58
3.4.5.1	Pretreatment of Wastewater before conveying to the Storage Tank	59
3.4.5.2	Wastewater Sampling	59
3.4.5.3	Biofilm Sampling	60
3.5	Analytical Methods	60
3.5.1	Chemical Components of Tanfloc	60
3.5.2	Biodegradability of Tanfloc	60
3.5.3	Wastewater Analysis	61
3.5.4	Sludge Volumetric Index (SVI)	61
3.5.5	Flocs Size Distribution	61

3.5.6	Zeta Potential	62
3.5.7	Estimation of Biomass	62
3.5.8	Metagenomic study using illumina next generation sequencing technology	62
4	RESULTS AND DISCUSSION	63
4.1	Characterization of Tanfloc	63
4.1.1	FT-IR Spectrum of Tanfloc.	63
4.1.2	Energy-Dispersive X-ray Spectroscopy (EDX) Analysis	65
4.1.3	Biodegradability of Tanfloc	65
4.2	Performance of Tanfloc (first stage)	66
4.2.1	The Best Dose and Mixing Condition	66
4.2.2	Floc Size and Settling Velocity	72
4.2.3	Sludge Volume Analysis	76
4.2.4	Tanfloc Effectiveness for Removing Pollutants from Domestic Wastewater	77
4.2.5	Effect of Cations Addition	78
4.2.6	Zeta Potential Measurement	79
4.3	Performance of Tanfloc (second stage)	81
4.4	Effect of Tanfloc on the Performance of process (A) (third stage / Aeration tank is 2850 L)	83
4.4.1	Flocculation Tank Evaluation	83
4.4.2	Primary Clarifier Evaluation	85
4.4.3	Aeration Tank Evaluation	89
4.4.3.1	Treatment Efficiency	89
4.4.3.2	Dissolved Oxygen Study	96
4.4.3.3	Sludge production	99
4.4.3.4	Estimation of Biomass	101
4.5	Evaluation of Aeration Tank of process B (fourth stage /aeration tank is 1250 L)	102
4.5.1	Treatment Efficiency	102
4.5.2	Dissolved Oxygen Study	107
4.5.3	Sludge Production	108
4.5.4	Estimation of Biomass	109
4.6	The Effect of the Extended Use of Tanfloc on the Biofilm Bacterial Community (fifth stage / two aeration tanks each is 25 L)	111
4.6.1	Biofilm community	111
4.6.2	The detected genera of AOB and NOB	116
4.6.3	The role of Tanfloc in creating the suitable environment for AOB	117
4.6.4	Biofilm Performance.	118
4.6.5	Biomass Estimation	125
4.7	Costing Study	125
4.7.1	Capital Cost	126
4.7.2	Operational Cost	127
4.8	Advantages and limitations of Tanfloc	128
4.9	Summary	129

5	CONCLUSIONS AND RECOMMENDATIONS	130
5.1	Conclusions	130
5.2	Recommendations	131
	REFERENCES	132
	APPENDICES	143
	BIODATA OF STUDENT	178
	LIST OF PUBLICATIONS	179



LIST OF TABLES

Table	Page
2.1 Typical composition of untreated domestic wastewater	7
2.2 Effluent standards in Malaysia	8
2.3 The most common uses of coagulation and flocculation process for different types of water and wastewater	25
2.4 Microorganism species and their exerted biocoagulants	38
2.5 Tanfloc applications	42
3.1 Experiment stages	46
3.2 Characteristics of wastewater produced in the hostel of Faculty of Engineering	48
3.3 Sequence of experiments	58
3.4 Flow rates and retention times investigated in the experiment	60
4.1 Functional group of Tanfloc	64
4.2 Biodegradability of Tanfloc	66
4.3 Coagulation rate of Tanfloc	68
4.4 Floc size distribution	72
4.5 Effect of floc size distribution on turbidity removal	75
4.6 SVI vs. dose of Tanfloc and PAC	77
4.7 Flocc size distribution	84
4.8 Residual turbidity in the beaker	84
4.9 Sludge volume index	85
4.10 Wastewater characteristics before and after primary clarifier	88
4.11 Removal efficiencies of pollutants in primary clarifier	88
4.12 Hydraulic retention time	89

4.13	Removal efficiencies of turbidity (NTU) in primary clarifier and secondary treatment (secondary clarifier after the aeration tank) in the third stage	90
4.14	Removal efficiencies of TSS (mg/L) in primary clarifier and secondary treatment (secondary clarifier after the aeration tank) in the third stage	90
4.15	Removal efficiencies of COD (mg/L) in primary clarifier and secondary treatment (secondary clarifier after the aeration tank) in the third stage	91
4.16	Removal efficiencies of BOD (mg/L) in primary clarifier and secondary treatment (secondary clarifier after the aeration tank) in the third stage	93
4.17	Removal efficiencies of NH ₃ -N (mg/L) in primary clarifier and secondary treatment (secondary clarifier after the aeration tank) in the third stage	94
4.18	Removal efficiencies of total phosphate (mg/L) in primary clarifier and secondary treatment (secondary clarifier after the aeration tank) in the third stage	95
4.19	pH measurements in primary clarifier and secondary treatment (secondary clarifier after the aeration tank) in the third stage	95
4.20	DO variation in aeration Tank in the third stage	97
4.21	Percentage allowable increment in BOD ₅ load to reach a DO level of 2 (mg/L) in the third stage	98
4.22	Treatment efficiency with one air pump on at 18 L/minute in the third stage	98
4.23	Volatile suspended solids concentration (VSS) for the effluent from aeration tank (mg/L) in the third stage	100
4.24	Weight of dry biomass on Cosmo balls in the third stage	101
4.25	Calculations of attached biomass in the third stage	101
4.26	Removal efficiencies of total turbidity (NTU) in primary clarifier and secondary treatment (secondary clarifier after aeration tank) in the fourth stage	103

4.27	Removal efficiencies of TSS (mg/L) in primary clarifier and secondary treatment (secondary clarifier after aeration tank) in the fourth stage	103
4.28	Removal efficiencies of COD (mg/L) in primary clarifier and secondary treatment (secondary clarifier after aeration tank) in the fourth stage	104
4.29	Removal efficiencies of BOD (mg/L) in primary clarifier and secondary treatment (secondary clarifier after aeration tank) in the fourth stage	105
4.30	Removal efficiencies of ammonia nitrogen (mg/L) in primary clarifier and secondary treatment (secondary clarifier after aeration tank) in the fourth stage	106
4.31	pH measurements in primary clarifier and secondary treatment (secondary clarifier after the aeration tank) in the fourth stage	106
4.32	DO variation in aeration Tank	108
4.33	Treatment efficiency with one pump on at 18 L/minute in the fourth stage	108
4.34	Volatile suspended solids concentration (VSS) and turbidity for the effluent of aeration tank in the fourth stage	109
4.35	Weight of dry biomass on cosmoballs in the fourth stage	110
4.36	Calculation of attached biomass in the fourth stage	110
4.37	Percentage of AOB and NOB in other experiments	117
4.38	Removal efficiencies of ammonia nitrogen (mg/L) in the fifth stage	119
4.39	DO level versus organic load	123
4.40	HRT for conventional and proposed treatment plants	126
4.41	Differences in operational cost between the conventional and proposed treatment plants	128

LIST OF FIGURES

Figure		Page
2.1	Types of flocculation	10
2.2	Illustration of charge neutralization and bridging mechanism	12
2.3	Electrostatic patch	13
2.4	Effect of continued addition of a coagulant (e.g. alum) on the destabilization and flocculation of colloidal particles	14
2.5	Probable chemical structure of Tanfloc	40
3.1	Research methodology	47
3.2	Cosmoballs	49
3.3	Process (A)	50
3.4	Process (B)	52
3.5	Set up of the specific study of biofilm characteristics	53
4.1	FT-IR spectrum of Tanfloc	63
4.2	FT-IR spectrum of (a) Chitosan, (b,c and d) modified Chitosan	64
4.3	Energy-dispersive X-ray spectroscopy (EDX) analysis	65
4.4	Effect of Tanfloc and PAC dose on residual turbidity	67
4.5	Effect of mixing time and speed on flocculation performance of Tanfloc	69
4.6	Effect of mixing duration and speed on flocculation performance of PAC.	70
4.7	Effect of prolonged mixing duration on the coagulation performance of PAC and Tanfloc	71
4.8	Comparison between the best doses of Tanfloc for different pollutants	72
4.9	Particle size distribution for the flocs of Tanfloc	73

4.10	Particle size distribution for the flocs of PAC	73
4.11	Effect of settling time on residual turbidity	74
4.12	Residual turbidity vs. depth of beaker	75
4.13	Effectiveness comparison between Tanfloc and PAC	78
4.14	Effect of cations addition on flocculation performance of Tanfloc.	79
4.15	Effect of Tanfloc dose on zeta potential measurements	81
4.16	Effectiveness comparison with and without Tanfloc	82
4.17	Contribution of primary clarifier in COD removal in the third stage	92
4.18	Contribution of primary clarifier in BOD removal in the third stage	93
4.19	Relationship between BOD load on DO level in the third stage	97
4.20	Effect of Tanfloc on the allowable increment of BOD ₅ load in the third stage	98
4.21	Comparison of removal efficiencies for the experiment without Tanfloc, with Tanfloc 100 % aeration capacity and with Tanfloc 50% aeration capacity in the third stage	99
4.22	Contribution of primary clarifier in COD removal in the fourth stage	104
4.23	Contribution of primary clarifier in BOD removal in the fourth stage	105
4.24	Comparison of removal efficiencies for the experiment without Tanfloc, with Tanfloc 100 % aeration capacity and with Tanfloc 50% aeration capacity in the fourth stage	109
4.25	Bacteria genera diversity in the biofilm sample in the experiment Without Tanfloc at 4 hours	112
4.26	Bacteria genera diversity in the biofilm sample in the experiment with Tanfloc at 4 hours	113
4.27	Bacteria genera diversity in the biofilm sample in the experiment Without Tanfloc at 2 hours	114

4.28	Bacteria genera diversity in the biofilm sample in the experiment with Tanfloc at 2 hours	115
4.29	Comparison between the percentage of Nitrosomonas and Nitrospira	116
4.30	Comparison of ammonia nitrogen removal efficiencies	120
4.31	Comparison of effluent NO ₃ level	120
4.32	pH drops during nitrification process	121
4.33	Comparison of influent BOD level	122
4.34	Comparison of influent COD level	122
4.35	Comparison of influent turbidity level	124
4.36	Comparison of influent suspended solids level	124
4.37	Illustration of conventional and proposed treatment plants	126

LIST OF ABBREVIATIONS

AOB	Ammonia Oxidizing Bacteria
BOD	Biochemical Oxygen Demand
COD	Chemical Oxygen Demand
C/N	Carbon / Nitrogen ratio
d (10)	The size that 10%, of total volume of flocs was below this value
d (50)	The size that 50%, of total volume of flocs was below this value
d (90)	The size that 90%, of total volume of flocs was below this value
DO	Dissolved Oxygen
DOC	Dissolved Organic Carbon
EDX	Energy-Dispersive X-ray Spectroscopy
FT-IR	Fourier-Transform Infrared Spectroscopy
HRT	Hydraulic Retention Time
KDa	Kilodalton
NGS	Next Generation Sequencing
NOB	Nitrite Oxidizing Bacteria
NTU	Nephelometric Turbidity Unit
OLR	Organic Loading Rate
PAC	Polyaluminium chloride
PCR	Polymerase Chain Reaction
PE	Population Equivalent
PFS	Polyferric sulphate
POME	Palm Oil Mill Effluent
QIIME	Quantitative Insights Into Microbial Ecology

SVI	Sludge Volume Index
TDS	Total Dissolved Solids
THMs	Trihalomethanes
TOC	Total Organic Carbon
TP	Total Phosphate
TSS	Total Suspended Solids
UV 254	Ultraviolet absorption at 254 nm
VSS	Volatile Suspended Solids
$\mu\text{ s/cm}$	Micro Siemens / cm

CHAPTER 1

INTRODUCTION

1.1 Introduction

This chapter introduces the background of wastewater treatment plants and common modifications that respond to changes in both water quality and quantity. The problem statement focuses on current interests and concerns about the treatment process; especially those related to the use of coagulation and flocculation processes, in addition to conventional and new materials utilized for that purpose. The objectives are determined and the scope of the study is elaborated upon in Chapter One.

1.2 Research Background

Water supply is one of the most important requirements of life. In human societies, most of the water supplied will eventually be converted into domestic wastewater. If this wastewater is not treated, it will accumulate and become anaerobic (due to a lack of dissolved oxygen), and consequently, it will be considered a terrible source of nuisance for the community.

For this reason, wastewater treatment is a main feature of urban areas. Treatment plants are comprised of sequencing steps of physical, chemical and biological processes that interact together to decrease wastewater pollution to a required level. Concerns about the treatment of wastewater started at the beginning of the last century. At that time, the objectives of treatment were limited to removing solids and biodegradable organics, and the elimination of pathogenic organisms. As the concerns about pollution and its effect on public health and the environment increased, the standards of treated water quality became more stringent and previous treatment processes were deemed to be insufficient to respond to these standards (Wang et al., 2015). Consequently, new processes and methods were introduced into this field (Fulazzaky et al., 2015; Leyva-Díaz et al., 2015a; Martín-Pascual et al., 2016; Wang et al., 2014).

One of the optional chemical processes, which are used in wastewater treatment plants to improve treatment efficiency, is the coagulation and flocculation process. Wastewater contains solids in a variety of size distributions. A certain proportion are colloids, which are, due to their small size, infeasible to be settled by gravity. As a practical solution, these colloids can be forced to agglomerate by coagulation and flocculation, in order to grow big enough to be removed physically (Suopajarvi et al., 2013). A variety of materials (inorganics and organics), show a superior potential to function as coagulants and flocculants (Aljuboori et al., 2013; Aljuboori et al., 2014; Choudhary et al., 2015; Liu et al., 2016).

However, the treatment of wastewater should not act as an additional source of pollution. In other words, sludge generated from the wastewater treatment process should be environmentally friendly as much as possible. A high percentage of the sludge produced from conventional treatment is biodegradable, and the rest is mainly dust, sand and other particles (Qasim, 1998). Regardless of the high requirement for funds and the necessary skills to manage this type of sludge, it is still feasible (Qasim, 1998); compared to the sludge produced from chemical treatments, like precipitation and coagulation, using metal ions (Choy et al., 2014; Lee et al., 2014).

1.3 Problem Statement

Construction of conventional sewage treatment plants is restricted by the availability of large required area, which is considered a major contributor to the high capital cost of the treatment plants. The operation of these plants is extremely energy intensive; for example, water and wastewater utilities consume about 2% according to Dotro et al. (2011) and 3% according to Spellman (2013) of the total amount of electricity produced in the United States. Upgrading the existing treatment plants to cater for the increasing population or to respond to new and more stringent standards is another challenge faced by the authorities due to the difficulties to provide the required land for the extension.

In order to accommodate these issues, several attempts have been conducted to develop and modify conventional treatment units or provide alternative methods with fewer requirements for construction, operation or land. Reducing the influent organic load to the biological unit is a possible approach to reduce the requirements for volume and oxygen supply for this unit. USEPA (2010) stated that oxygen requirement is a reflection of organic load. Practically, enhancing the sedimentation process is one of the alternatives to reduce the influent organic load to the biological unit.

Efficiently designed and operated primary sedimentation tanks should remove from 50 to 70% of the suspended solids and 25 – 40 % of the BOD. This anticipated efficiency is negatively affected by eddy currents formed by the inertia of the incoming fluid, wind induced circulation cells formed in uncovered tanks, thermal convection currents and thermal stratification in hot arid climate. Inclined plates and tube settlers are common modifications to enhance sedimentation process especially in the compact units with limited available space.

Enhancement of sedimentation process could be achieved by preceding the coagulation and flocculation process. Traditional chemical coagulants and flocculants are aluminium and ferrous salts. Several environmental and public health problems arise due to extended use of these conventional chemicals. From a medical point of view, aluminium residuals in alum treated water have been the centre of attention, as they are linked to serious health issues, such as Alzheimer (Lee et al., 2014). From an environmental point of view, a serious drawback of hydrolysing metal coagulants is

the production of large amounts of toxic sludge, which is non-biodegradable due to the nature of the coagulant. Moreover, 99% of alum sludge is made up of water and alum sludge is rather hard to dewater (Lee et al., 2014; Renault et al., 2009). Other drawbacks include large amounts are required for efficient flocculation, it is highly sensitive to pH, inefficient towards very fine particles, inefficient in cold water (especially Polyaluminium chloride), and finally, the presence of aluminium in water negatively affects the disinfection process (Choy et al., 2014; Lee et al., 2014).

Consequently, great efforts have been made to provide natural coagulants as a substitution for conventional inorganic coagulants (Abidin et al., 2013; Aljuboori et al., 2013; Aljuboori et al., 2014; Aljuboori et al., 2015; Amagloh and Benang, 2009; Beltrán-Heredia et al., 2010b; Beltrán-Heredia et al., 2012; Gong et al., 2008; Graham et al., 2008; Li et al., 2009; Lian et al., 2008; Xia et al., 2008a). Meanwhile, several natural coagulants are produced in commercial quantities; others are at the limits of lab scale production.

Introducing these natural coagulants to conventional treatment processes, in the hope of improving performance to accommodate higher flow (upgrade treatment plants for increased population), achieve better treatment efficiency (upgrade treatment plants for new stringent standards) and energy saving, have not been well investigated. For this reason, a tannin based agent (a natural coagulant) was used in this study to enhance the performance of sedimentation process, in the hope of improving the overall treatment efficiency.

1.4 Research Objectives:

This study aims to investigate the improvement of treatment process by introducing Tanfloc (a tannin based coagulant) as a pre-treatment for sewage treatment plants. The specific objectives of this study are as follows:

- (i) To assess the potential and effectiveness of applying Tanfloc in domestic wastewater treatment.
- (ii) To determine Tanfloc efficiency in a continuous flow experiment using flocculation and sedimentation units only.
- (iii) To investigate the effects of Tanfloc to reduce influent organic load to a biofilm treatment unit.
- (iv) To characterize the type of microbial community within a biofilm cultured in flocculated wastewater using Tanfloc.

1.5 Scope of the Study

The scope of this study extended to give more details about the characteristics and behaviour of Tanfloc. The investigation includes the chemical characterization of Tanfloc in addition to determination of the best dose and mixing conditions for both Tanfloc and polyaluminium chloride (PAC). The effect of Tanfloc on removal efficiency of pollutants from domestic wastewater was investigated. Furthermore, investigation of floc size and settling velocity of Tanfloc compared to PAC was conducted. The investigation extended to include the study of sludge produced for Tanfloc and PAC. The effect of cations on flocculation performance was evaluated. Moreover, zeta potential measurement was studied.

Preliminary study of Tanfloc performance in the flocculation and sedimentation units in continuous flow experiment was conducted to give more details about Tanfloc behaviour in continuous flow experiment. The experiment proceeded to study the effects of Tanfloc on the treatment process at different flow rates. To evaluate the flocculation process, floc size distribution, sludge volume index and residual turbidity were determined. However, clarifier performance was evaluated by the determination of influent and effluent concentration of COD, BOD, TSS, ammonia nitrogen, turbidity and total phosphate for five different flow rates with and without Tanfloc. Aeration tank performance was evaluated by the determination of COD, BOD, TSS, turbidity, and total phosphate for the influent and effluent for five different flow rates with and without Tanfloc. Moreover, the evaluation of aeration tank included dissolved oxygen study and estimation of secondary sludge production.

The effect of Tanfloc on the biofilm community was evaluated, biofilm samples were taken after the process had stabilized from the two identical reactors (with and without Tanfloc), and tested for illumina Next Generation Sequencing technology (NGS), which is used to identify the species of bacteria and their percentage in a biofilm community. Wastewater characteristics were determined in addition to dissolved oxygen level to have a detailed description of the entire scenario in which the biofilm was cultured.

1.6 Thesis layout

This thesis consists of five chapters. Chapter One explains the background of the study and the problem statement, and ends with stating the objectives and scope of the research. Literature review was covered in Chapter Two including coagulation, flocculation and biofilm treatment process as main topics in the review. In Chapter Three, the materials used in the experiments were listed and explained in details including chemicals and treatment process units. Moreover, details about preparation of samples for the analysis were explained, finally the methods and procedures of conducting the experiments and taking and analyzing the samples were covered also in Chapter Three. Chapter Four presents the results and discussion about Tanfloc and its effect on the sedimentation process and biofilm units, in addition to the effect of

Tanfloc on the biofilm bacterial community. Chapter Five wraps up the thesis with conclusions and recommendations for the future work.



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