FORMALDEHYDE REMOVAL FROM SYNTHETIC CONTAMINATED AIR USING A BIOTRICKLING FILTER REACTOR

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ABSTRACT

Formaldehyde (FA) removal from contaminated air with biotrickling filter reactor (BTFR) has been extensively studied thus far, but the effects of different volumetric airflow rates, pH, temperature and nutrient concentration on FA removal efficiency needs to be verified to design better BTFRs with optimal operating conditions. The use of specific microorganisms to remove FA from the air still needs to be verified. This study used the laboratory scale BTFR with a volume of 3.319 L, operated at the different volumetric airflow rates to having an insight on the effect of gas retention time on FA removal efficiency. To determine the optimal retention time of contaminated airflow through the BTFR system, mathematical models were developed. The predominant microorganisms were identified as to investigate the microbiological diversity. Then the BTFR was optimized for temperature and pH as well as nitrogen and phosphorus concentration to achieve its maximum efficiency. The BTFR formaldehyde removal efficiencies of 99, 96 and 95% were verified for the volumetric airflow rates of 90, 291, 1512 L/h, respectively, and the optimal retention times of 141, 50 and 26 s were verified for the BTFR experiments operating at 90, 291 and 1512 L/h volumetric airflow rate, respectively. The optimization of the BTFR conditions for removing FA from a synthetic contaminated air stream assessed by Taghuchi method showed that the best efficiency of the BTFR could be at a neutral pH of 7, a temperature range of 40 - 45℃, an ammonium concentration of 0.5 mg/L and a phosphate concentration of 1 mg/L. The predominant bacteria attached on the surface of the supporting materials were identified as five bacterial colonies i.e., *Salmonella bongori*, *Salmonella choleraesuis subsp. arizonae, Salmonella typhimurium*, *Serratia entomophila* and *Serratia ficaria*. The logarithmic and linear models were proposed as an ideal approach for determining the optimal retention time, in order to make a significant contribution for future biotechnological developments and air quality improvement analysis.

ABSTRAK

Penyingkiran formaldehyde (FA) dari udara yang tercemar dengan menggunakan biotrickling filter reactor (BTFR) telah dikaji secara meluas sehinggi kini, tetapi kesan-kesan seperti isipadu kadaralir udara, pH, suhu dan kepekatan nutrien terhadap penyingkiran FA perlu disahkan untuk mereka bentuk BTFR yang terbaik dengan keadaan operasi yang optimum. Pengunaan mikroorganisma tertentu untuk penyingkiran FA dari udara perlu disahkan. Kajian ini menggunakan BTFR berskala makmal dengan isipadu 3.319 L, dioperasikan dengan isipadu kadaralir udara berbeza untuk mendapatkan penjelasan kesan masa tahanan gas terhadap keberkesanan penyingkiran FA. Model matematik telah dibangunkan untuk menentukan masa tahanan yang optimum bagi aliran udara yang tercemar melalui system BTFR. Mikroorganisma utama telah dikenalpasti untuk menyiasat kepelbagaian mikrobiologi. Kemudian, BTFR telah dioptimumkan terhadap suhu dan pH serta keperkatan nitrogen dan phosphorus untuk mencapai keberkesanan yang maksimum. Keberkesanan BTFR dalam penyingkiran formaldehyde adalah sebanyak 99, 96 and 95% telah disahkan dengan isipadu kadaralir udara masing-masin adalah 90, 291, 1512 L/h dan tempoh tahanan optimum adalah 141, 50 and 26 s untuk operasi isipadu kadaralir udara pada 90, 291 and 1512 L/h. Pengoptimuman keadaan BTFR untuk menyingkirkan FA dari aliran udara tercemar sintetik dinilai menggunakan kaedah Taghuchi menunjukkan keberkesanan terbaik bagi BTFR adalah pada pH neutral iaitu 7, suhu diantara 40 - 45℃, keperkatan ammonium pada 0.5 mg/L dan keperkatan phosphate pada 1 mg/L. Bakteria utama yang berada di atas permukaan bahan sokongan telah dikenalpasti terdiri daripada lima koloni bakteria iaitu *Salmonella bongori*, *Salmonella choleraesuis subsp. arizonae*, *Salmonella typhimurium*, *Serratia entomophila* and *Serratia ficaria*. Model logaritmik dan linear telah dicadangkan sebagai kaedah yang sesuai untuk menentukan tempoh tahanan optimum, seterusnya memberi sumbangan yang besar terhadap pembangunan bioteknologi di masa hadapan dan analisis peningkatan kualiti udara.

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LIST OF SYMBOLS

A	-	Constant coefficient (in s^{-1})
a	-	Constant coefficient (in s^{-1})
a_0	-	Surface area of the packing (in m^{-1})
B	-	pH of maximum activity
b	-	Constant coefficient (in s^{-1})
d	-	Constant coefficient (in mg)
C_{ads}	-	Concentration adsorbed (in mg/L)
C_{air}	-	Gas-phase pollutant concentration (in mg/L)
C_{bf}	-	Concentration of contaminant in the biofilm (in mg/L)
C_g	-	logarithmic average (in mg/L)
C_{in}	-	Inlet concentration (in mg/L)
C_{Nbf}	-	Concentration of carbon source in the biofilm (in mg/L)
C_{O2bf}	-	Concentration of oxygen in the biofilm (in mg/L)
C_{out}	-	Outlet concentration (mg/L)
D_{air}	-	Gas-phase diffusion constant (in g/L)
D_w	-	Molecular diffusion constant (in g/L)
E	-	Concentration of FA (in mg/L)
EC	-	Elimination capacity (in mg/L)
EC_{max}	-	Maximum elimination capacity (in mg/L)
f	-	Constant coefficient (in m)
g	-	Acceleration of gravity (m/s^2)
H	-	Henry’s Law constant for the contaminant (in L.atom/mol)
H	-	BTFR or BF height (in m)
H_T	-	Height of the tower (in m)
J_{ads}	-	Flux per unit surface area (in $mol/m^2.s$)
J_{bf}	-	Flux of contaminant per unit of surface area (in $mol/m^2.s$)

k	-	Maximum specific substrate utilization rate (in g substrate/g microorganism.d)
K_0	-	Constant
k_1	-	Kinetic constant (in h^{-1})
k_2	-	Kinetic constant (in m^3 mg^{-1})
K_{ads}	-	An empirically determined constant
K_{ads}	-	Rate constant
K_{air-bf}	-	Gas transfer coefficient
k_d	-	Bacteria decay rate (in g VSS/g VSS.d)
K_{inh}	-	Haldane constant
K_m	-	Saturate constant (in mg/L)
K_S	-	Monod half-saturation constant for carbon source (in mg/L)
K_{SN}	-	Monod half-saturation constant for nitrogen (in mg/L)
K_{SO2}	-	Monod half-saturation constant for oxygen (in mg/L)
L	-	Constant coefficient
L_{bf}	-	Thickness of the flowing water layer (in mm)
m	-	Constant coefficient (in m)
n	-	Constant coefficient (in m)
N_{Nbfi}	-	Concentration of nitrogen source in the biofilm (in mg/L)
Q_w	-	The water flow rate (in L/s)
R	-	Gas constant (in J / mol. K)
$r(c)$	-	Degradation rate at concentration c (mg/m^3 h)
R_e	-	Reynolds number (dimensionless)
Re	-	Removal efficiency (in %)
r_{max}	-	Maximum elimination capacity (in mg/L)
R_p	-	Particle radius (in mm)
r_{su}	-	Substrate degradation rate (in g/m^3.d)
S	-	Concentration of substrate after biodegradation process (mg/L)
S_0	-	Initial concentration of substrate (in mg/L)
S_c	-	Schmidt number
T	-	Temperature (in °C)
t	-	Time (in s)
V	-	Gas flow rate (in L/s)
W	-	Water content (in g/g)

x	-	Coordinate perpendicular to the biofilm surface
X	-	Microorganism concentration (in mg/L)
Y	-	Microorganism yield (in g/g)
ΔH	-	Heat of adsorption (in ℃)
θ	-	Gas retention time (in s)
θ_c	-	Microbial retention time (in day)
μ_w	-	Viscosity (in m/s)
ρ_w	-	Density (in Kg/m^3)
$[O_2]$	-	Oxygen concentration (in mg/L)
μ_{max}	-	Maximum specific bacterial rate (in g new cell/g cell.d)
β		Constant coefficient

LIST OF ABBREVIATIONS

AHTM	-	4-Amino-3-Hydrazino-5-Mercapto-4H-1,2,4-Triazole
BF	-	Biofilter
BS	-	Bioscrubber
BTFR	-	Biotrickling Filter Reactor
DNA	-	Deoxyribonucleic acid
DNPH	-	2,4-Dinitrophenylhydrazine
DOAS	-	Differential Optical Absorption Spectroscopy
EB	-	Ethyl benzene
EC	-	Elimination capacity
FA	-	Formaldehyde
FTIR	-	Fourier Transform Infrared Absorption
GAC	-	Granular Activated Carbon
GC	-	Gas Chromatography
HDF	-	High-Density Fiber Board
IDLS	-	Infrared Diode Laser Spectroscopy
LIFL	-	Laser Induced Fluorescence Spectroscopy
LOAEL	-	Lowest Observable Adverse Effect Level
MBTH	-	3-Methyl-2-Benzo Thiazolinone Hydrazone
MDF	-	Medium Density Fiber Board
MF	-	Melamine-Formaldehyde
MUF	-	Melamine-Urea-Formaldehyde
MUPF	-	Melamine-Urea-Phenol-Formaldehyde
NMR	-	Nuclear Magnetic Resonance
OSB	-	Oriented-Strand Board
PAS	-	Photoacoustic Spectroscopy
PTRMS	-	Proton-Transfer-Reaction Mass Spectrometry

SGR	-	Suspended Growth Reactor
SIFTMS	-	Selected Ion Flow Tube Mass Spectrometry
TDLS	-	Tunable Diode Laser Spectroscopy
UFR	-	Urea-Formaldehyde-Resin
VOC	-	Volatile Organic Compound
WHO	-	World Health Organization

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CHAPTER 1

INTRODUCTION

1.1 Background of study

Although technology has made the world a better place to live, the impact of technology generates a wide range of pollutants, if not well planned and controlled. Many pollutants present in wastewater, waste gases and solid wastes can negatively affect ecosystems and human health. The collection and treatment of waste gases are very difficult, with no economically effective method for treating them. Therefore, air pollution has become local as well as regional issues. Many VOCs present in air can travel hundreds of kilometers from their source point and can cause multiple human health and environmental problems on a national or international scale. Therefore, air pollutants must be extensively controlled and treated. Several chemical, physical and biological methods for treating polluted gas have been proposed. Hence, the biological treatment process is the best method, and can emerge as a cost effective and environmentally friendly technology in treating higher volume loading rates of gas streams with relatively lower concentration of the effluent pollutants (Groenestijn and van Kraakman, 2005; Devinny et al., 1999; Delhomenie and Heitz, 2005; Shareefdeen and Singh, 2005). Based on our literature survey on biological processes to treat polluted air, they are classified as (1) supporting materials, (2) mathematical modeling, (3) test of biodegradability on different toxic matter and (4) the use of different strains of microorganisms such as bacteria or fungi. This classification is shown in Figure 1.1.

Many previous studies prove that organic pollutants are degradable with the help of microorganisms (Barco et al., 2012). This property tempts researchers and industrial managers to direct the use of microorganisms for controlling pollutants present in industrial exhaust air. Removing odor from the polluted air of wastewater, composting and rendering plants was the initial application of biofilters. In recent years, biofiltration has been applied increasingly to the removal of organic matter such as VOCs from industrial polluted air. The use of small reactors for removal of pollutants from a large volume of air contaminated with a low concentration of organic matter makes biofiltration technology more cost effective when compared to other VOC control technologies such as carbon adsorption and incineration. In bioreactors, polluted air is passed from a wet environment containing supporting materials and microorganisms. Pollutants are adsorbed onto the surface of supporting materials and thus absorbed into the wet layer around the microorganisms. Then, the microorganisms can use absorbed pollutants as their carbon source. Biological oxidization of microorganisms can convert these pollutants into environmentally benign end products such as water and carbon dioxide. During biofiltration, the pollutants are completely biodegraded without the formation of aqueous effluents.

Traditionally, biofilter (BF) and biotrickling filter reactor (BTFR), bioscrubber (BS) and suspended-growth reactor (SGR) have been used to remove VOCs with concentrations lower than 10 g/m^3 (Shungang et al., 2011). Formaldehyde (FA) is one of the gaseous pollutants released from a wide number of industries. This compound is an extremely toxic and mutagen (Salthammer et al., 2010; Jonidi et al., 2009); therefore, the removal of FA from industrial polluted air is absolutely necessary. FA has also been identified as a suspected carcinogen by the World Health Organization (Jonidi et al., 2009). As FA can harm DNA, it has been classified as a mutagen (Kotzias et al., 2005). FA also has teratogenic effects on humans and animals. A teratogen is a drug or substance that is capable of interfering with the development of an embryo/fetus, which may lead to birth defects or some developmental malformations. FA at room temperature is in the form of gas due to its lower boiling point. At this condition, FA gas is highly flammable, reactive and colorless. FA can be easily absorbed in the respiratory and gastrointestinal systems of humans due to its high solubility in water (McGregor et al., 2006).

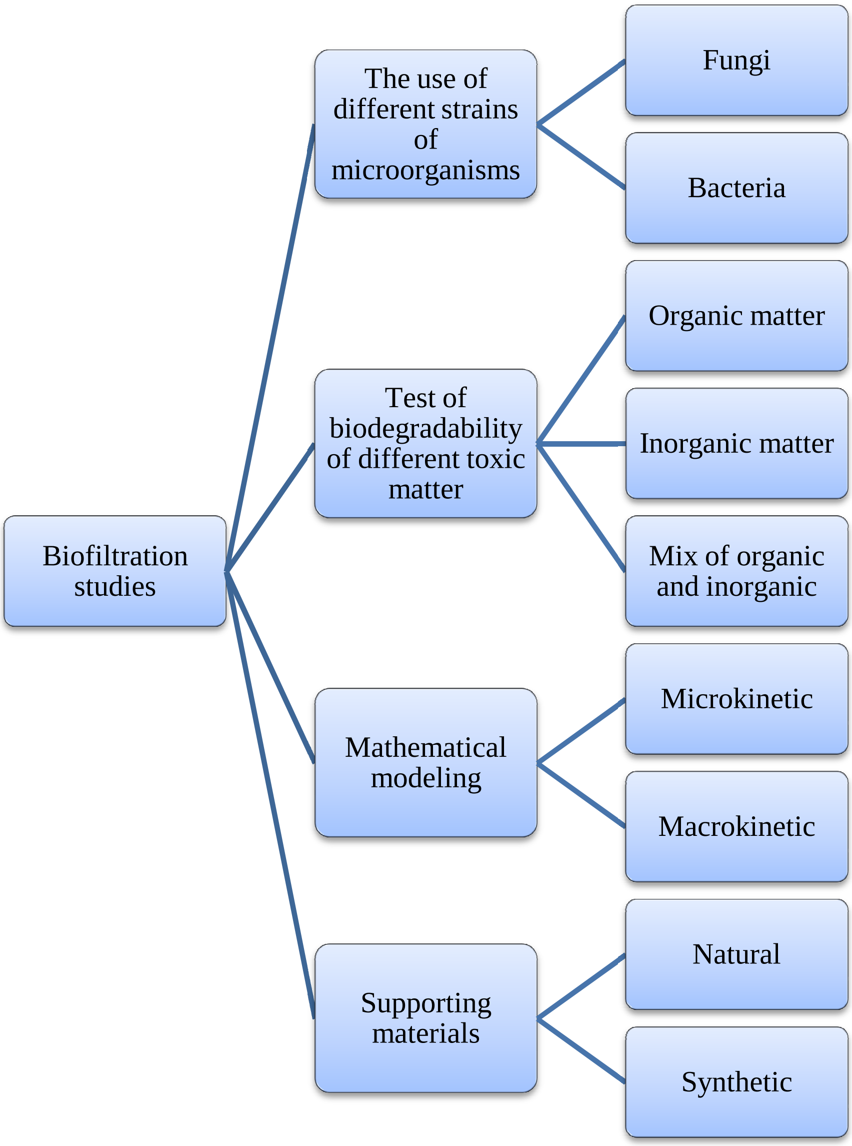

Figure 1.1 Topics of research in the biofiltration area

Tang et al. (2009) estimated that in 2006, 20 million tons of FA were released into the atmosphere worldwide. They also predicted that the amount of FA emission into the atmosphere increases year by year. Many petrochemical industries can be found worldwide, including Southeast Asian countries like Malaysia. Many studies exist regarding the removal of a wide range of pollutants from air by biofiltration technology. Each researcher has tried to find a novel method to develop this technology. Evaluation of biofiltration processes for the removal of various pollutants is one of the important areas undergoing research. Ramirez et al. (2008) used a biofilter to treat air contaminated with methanol and toluene. Biological removal of methanol in low concentrations is easy. However, biological removal of methanol in high concentrations, or when it is mixed with other compounds such as toluene, is complex and difficult. Ramirez et al. (2008) could determine the removal rate of these pollutants by a BTFR. Biodegradation rate is the speed of pollutant removal by microorganisms, measured by mg/L.s. This parameter is important for biofilter design in full-scale operation. Prado et al. (2008) demonstrated that low concentrations of dimethyl ether in air contaminated with FA and methanol have no significant effect on the removal rate of these pollutants in a biotrickling filter. However, moderate concentrations of this compound have a negative effect on the biodegradation of FA and methanol. Prado et al. (2008) demonstrated that many interactions could be found among different pollutants when biological methods are applied to treat polluted air. Therefore, before deciding to use biological methods to treat polluted air, the detection and identification of pollutants are very important. Hort et al. (2009) used a biological method to remove dimethyl sulfide and dimethyl disulfide from air, for which he used sewage sludge and yard waste compost as a biofilter bed. Hort was able to remove nearly 100% of these pollutants. Eldon et al. (2011) evaluated biological treatment of air contaminated with a mixture of VOCs from the fiberglass and composite manufacturing industries. They removed 100% of the acetone, toluene, and styrene in a biofilter inoculated with fungus. Such a study was very important due to the use of real polluted air in actual situation. Pratt et al. (2012) studied biofiltration of emitted methane from a dairy farm effluent pond. They achieved a removal capacity equal to 16 g/m^3h^1.

It is known from many studies that different supporting materials are being used for packing BFs and BTFRs. For example, Singh et al. (2006) used agro-waste as a biofilter supporting matter to remove toluene from air. A wide number of indigenous microorganisms can be found in agro-waste, which is why biofilters packed with agro-waste do not need microorganisms added as inoculum. This type of supporting material is inefficient in treating acidic pollutants such as H_2S, due to accumulation of acids around the microorganisms. In the near future, with increased release of pollutants into the atmosphere, using cheap supporting materials such as agro-waste can lead to finding cost-effective methods to control pollutants. Prado et al. (2004) used lava rock, perlite and activated carbon as supporting materials in a BTFR. They found that these supporting materials had only a small influence on the performance of the BTFR to remove a mixture of FA and methanol. This can result from the growth of a large number of microorganisms in the BTFR, creating enough capacity to remove a mixture of FA and methanol. In this condition, supporting materials do not have a significant impact on BTFR efficiency. However, the influence of different supporting materials on pollutant removal from high pollutant concentrations has been reported by other researchers, including Wan et al. (2011). Although ceramic particles are heavy and expensive, many report that such supporting materials are ideal for growing fungi (Prado et al., 2008; Wan et al., 2011; Shungang et al., 2011). Roshani et al. (2012) studied the effect of bed mixing and nutrient solution circulation on pressure drop in BTFR. They understood that circulation of nutrient solution can increase removal capacity up to 21 g s/m^3 bed height, but bed mixing had a negative impact on removal capacity of BTFR, and the removal capacity decreased up to 8 g s/m^3 bed height.

Fulazzaky et al. (2014d) reported that the removal of FA using either the chemical or physical method is still expensive, sometimes are not enough efficient. Researchers are looking at possible biological methods to remove the pollutants such as FA from the air to having an environmentally friendly method with low operational cost. Even though many industrial scales of BTFR treatment system have been proposed, the studies to having working knowledge on modeling the biofilter treatment process, optimum operational conditions for removing FA from the air, the types of microorganisms attached to the supporting materials and the mechanism of FA removal from the air are still limited. Based on above mentioned gaps this study were carried out to (1) to evaluate gas retention time effect on the performance of biotrickling filter reactor for treating air contaminated with formaldehyde; (2) optimize the operational conditions of biotrickling filter reactor for removing formaldehyde from a contaminated air such as pH, nitrogen concentration, phosphorus concentration and temperature; (3) to identify predominant bacteria attached on the supporting materials of biotrickling filter reactor; and (4) to verify the mechanisms of formaldehyde removal from contaminated air by a biotrickling filter reactor for describing biochemical complexity with maximum simplicity.

1.2 Problem statement

Although technology has made the world a better place to live, the impact of technology generates a wide range of pollutants, if not well planned and controlled. Many pollutants present in wastewater, waste gases and solid wastes can negatively affect ecosystems and human health. The collection and treatment of waste gases are very difficult, with no economically effective method for treating them. Therefore, air pollution has become local as well as regional issues. Many VOCs present in air can travel hundreds of kilometers from their source point and can cause multiple human health and environmental problems on a national or international scale. One such common air pollutant is FA, which is released from solid waste wood-burning sites, chipboard manufacturing plants, synthetic resin industries and several other chemical industries. FA is an organic compound with the formula CH_2O and has a dipolar resonance structure (Salthammer et al., 2010). FA can easily dissolve in water. It is a colorless, highly toxic, reactive, flammable gas at room temperature and slightly heavier than air. Therefore, FA is one of the most dangerous pollutants to the environment and human health, and has created potent mutagenic effects in humans and other organisms, both when acting alone and in combination with other mutagens (Gupta et al., 1982). Even though determination of global FA emission is quite difficult, an FA emission rate of about 20 million tons per year was predicted for the year 2006 (Tang et al., 2009). China is the largest producer and consumer of FA in the world (Tang et al., 2009). Various physical, chemical and biological methods have been developed for treating air contaminated with VOCs, such as activated carbon adsorption, chemical oxidation, and anaerobic and aerobic biological degradation (Mohammad et al., 2007; Prado et al., 2004; Wan et al., 2011; Li et al., 2012; Temtem et al., 2009; Kennes et al., 1998). Still, biological treatment processes are among the best methods, and can emerge as cost-effective. Most important, they are considered environmentally friendly technology.

Each type of research design (Dunn, 2012; Sheldon, 2012; Borole et al., 2011; Kumar et al., 2011) has its own standards for reliability and validity in treating VOCs. However, aspects not fully understood include determination of (1) FA biodegradation rate, (2) microorganisms' diversity, (3) the portion of FA removal to accumulate into biomass and to dilute into nutrient solution and (4) optimal gas retention time for eliminating FA in terms of removal efficiency of a biotrickling filter.

1.3 Objectives of the study

The aim of this study is the evaluation of FA removal from synthetic contaminated air stream (SCAS) using a BTFR with the objective as follows:

- To evaluate gas retention time effect on the formaldehyde removal efficiency of biotrickling filter reactor for treating air contaminated with formaldehyde by modeling the biofiltration process.
- To identify predominant bacteria attached on the supporting materials of biotrickling filter reactor at different parts of biofilter bed.
- To optimize the operational conditions of biotrickling filter reactor for removing formaldehyde from a contaminated air by assessing the operational parameters of pH, nitrogen concentration, phosphorus concentration and temperature affecting the performance of biotrickling filter reactor.
- To suggest the mechanisms of formaldehyde removal from contaminated air by a biotrickling filter reactor for imposing maximum possible simplicity of the model describing the data.

1.4 Research methodology

Following is a short discussion of the research methodology applied in this study. This study was conducted in 4 stages consisting of 16 steps. The stages of the study is shown in Figure 1.2 are as follows:

- Stage 1: Evaluation of the gas retention time effect on the performance of BTFR for treating air contaminated with formaldehyde (Evaluation of the gas retention time effect).
- Stage 2: Identification of predominant bacteria on the supporting materials of BTFR (Microorganisms Identification).
- Stage 3: Optimization of four environmental variables including pH, temperature, nitrogen and phosphorus concentrations to have the best efficiency of BTFR for the removal of formaldehyde from contaminated air.
- Stage 3: Verification of the formaldehyde removal mechanisms from contaminated air by a BTFR (Verification of the formaldehyde removal mechanisms).

1.4.1 Stage 1: Calculation of the optimal gas retention time

In Stage 1, firstly a strong literature review on biofiltration technology in Step 1 was carried out and the laboratory-scale BTFR was made in Step 2. The BTFR consists of a biofilter bed, using fragmented pieces of polyurethane pipe as supporting material, a nutrient solution tank, FA storage tank, compressor, peristaltic pump, flow meter, control air valve and a mechanical clock switch. This BTFR with 4 sampling ports monitored the FA content in the biofilter during the experimental period. Heights of 5, 15, 25 and 40 cm from the bottom of the biofilter bed were set up for Ports 1, 2, 3 and 4, respectively. The dimensions of the biofilter are 8 cm diameter, 66 cm height and 3.319 L volume. The percentage of void space in the biofilter is approximately 90%. In Step 3, the aerobic granular sludge from a municipal wastewater treatment plant of Isfahan city, Iran, was used as inoculum for the start-up of the BTFR system; one liter of activated sludge seeded the BTFR. To immobilize microorganisms, the activated sludge was circulated through the beds for approximately two weeks. In Step 4, after completing the immobilization process, biomass adaptation was performed. During the adaption phase, a mixture of glucose and FA was added to the nutrient solution as a carbon source. Starting with a low FA level, a high amount of glucose was added to the nutrient solution, giving an organic matter concentration of about 1 g/L of COD.

An increased FA concentration might decrease the glucose concentration for generation of the COD concentration with a similar organic matter fraction in the nutrient solution. After a 90-day of adaptation phase, FA was the sole carbon source being used to feed the BTFR. In Step 5, the SCAS, which can be ventilated using the air pressure generated by a compressor, was supplied from the FA storage tank to the biofilter. A gas meter was used to monitor the gas flow rate. The BTFR was conducted under different FA concentrations of SCAS. In each FA concentration, the BTFR was operated for 4 consecutive days until FA removal efficiency was achieved below 4% (steady-state). FA concentration in the biofilter bed at the position of Ports 1, 2, 3 and 4, air samples were collected daily during the experiment using a Champion Air Pump-AAP model vacuum pump. The experimental temperatures were monitored using a classical mercury thermometer at the influent, middle of the biofilter bed and effluent of the BTFR system. In Step 6, the empirical models were developed based on the experimental data obtained from the previous steps to determine the optimal gas retention time. Several mathematical models have been proposed to explain the removal of FA from contaminated air. The mathematical modeling of bacterial growth and process performance has led to the improved design and operation of bioreactor system development.

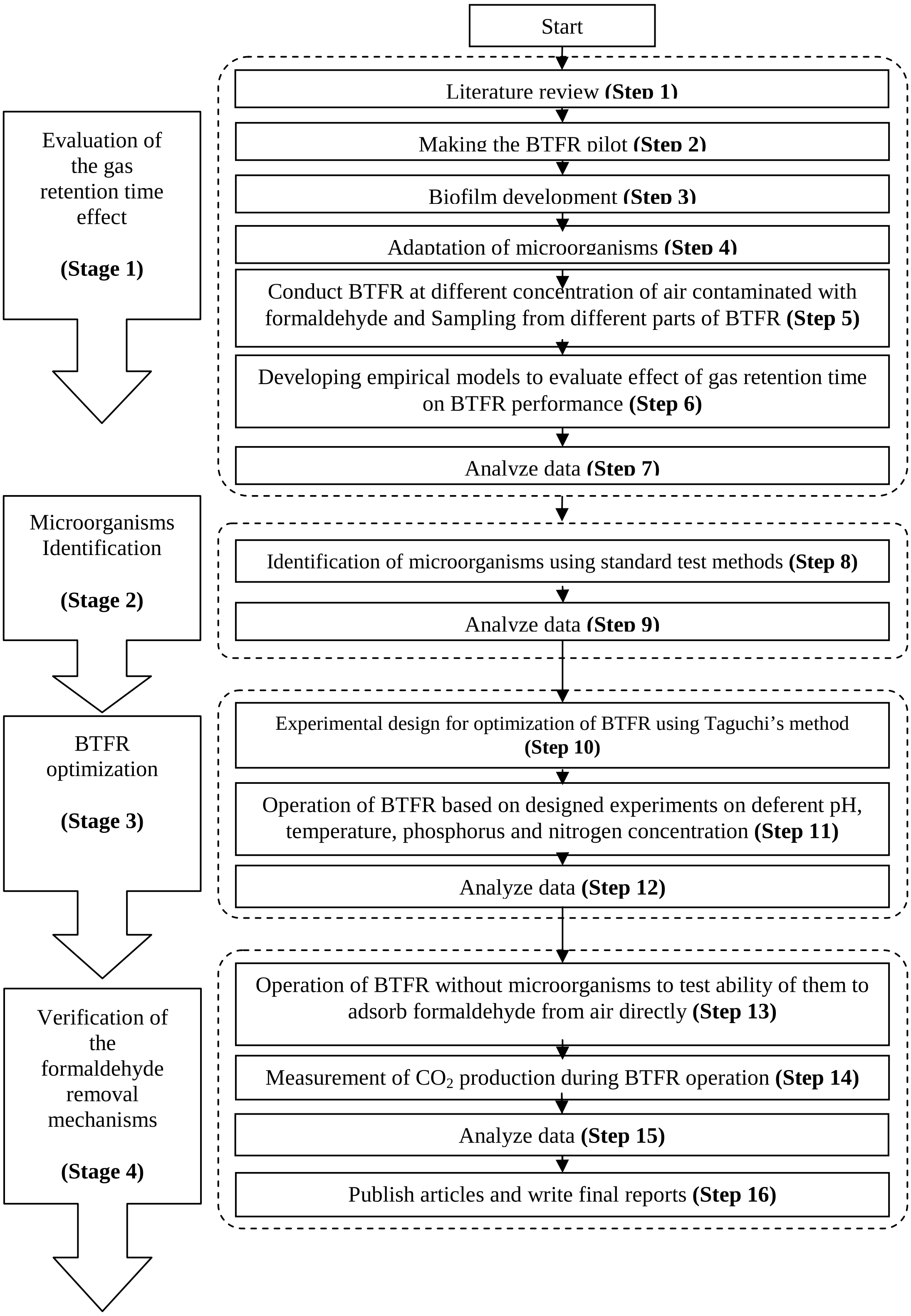

Figure 1.2 Methodology Flowchart

The need to operate on a day-to-day basis in practice requires further development of robust, integrated microfluidic systems. To find a micro kinetic model, using suitable software (Microsoft Excel, version 2008), all types of correlations between FA removal and gas retention time were evaluated. The best correlations obtained were selected to model the BTFR. Following the macro-balance equations of the BTFR system can be modeled to having the design parameters with three following approaches, such that: (1) a strong correlation between FA removal and gas retention time can be used to develop a logarithmic equation to calculate the optimum gas retention time, (2) the correlation between FA removal percentage and gas retention time can be used to develop a linear equation to calculate the efficiency of the BTFR system, and (3) the modified Monod equation was used to calculate the biomass concentration and the BTFR efficiency. The finding results of steps 1 to 6 were analyzed to having a new insight of body knowledge for air quality controlling system.

1.4.2 Stage 2: Microorganism identification

In Step 8, the biofilm samples were taken through each port of all parts of the BTFR. Subsequently, the microorganisms presented at the supporting materials were cultured onto nutrient agar medium. Then the predominant microorganisms were identified using conventional biochemical methods that consist of morphological identification and various biochemical tests, such that:

- Lactose test
- Indole test
- Urea hydrolysis test
- Motility test
- H_2S test
- KCN growth test
- Oxidase test
- Grams test
- Catalase test

- Lysine dicarboxylase test
- Methyl Red test
- Citrate test
- Pigment test
- Voges proskauer test
- Tattrate test
- Dulcitol test

The results of biochemical tests were analyzed in step 9 to understand the strains of unknown microorganisms attached at the supporting materials of the BTFR.

1.4.3 Stage 3: BTFR optimization

The literature review in Step 1 shows there are three methods for optimization of BTFR: (1) the full factorial design, (2) one factor at the time, and (3) the factorial experimental design. In this study, the factorial experimental design was selected to optimize the BTFR system. Factorial experimental design is a method in which multiple factors are changed at the same time. One of the most important methods of factorial experimental design is the Taguchi method. This method was developed by Genichi Taguchi (1990). This method was initially applied to improve manufactured quality. However, it has recently been used to optimize many processes in advertising, engineering, marketing and biotechnology (Taguchi, 1990). This method analyzes several different parameters with a small number of experiments. For instance, in a process with eight factors, in which each factor has three levels, the combination of those factors and levels can achieve up to 6561 experiments and are needed to examine of each combination when the full factorial method is used. The combination can be reduced to the only 18 experiments when the Taguchi method is used, which is only 3% of the full factorial design. The Taguchi method identifies key factors that have the greatest impact on the performance of a given process. Therefore, in this study the Taguchi method was used to optimize FA removal using the BTFR. In addition, both the methods of full factorial design and one factor at the time are labor-intensive and time consuming.

In this study, four operational parameters (i.e., pH, temperature, nitrogen and phosphorous concentration) were used as the selected factors (see Table 1-1). The Taguchi experimental design with four factors of a standard L_{16} orthogonal array (Table 1-2) was employed using Qualitek-4 (Nutek Inc.) software as Step 10.

Table 1.1 : Selected factors and their levels

Factors	**Levels**			
	Level (1)	**Level (2)**	**Level (3)**	**Level (4)**
pH	3	5	7	9
Nitrogen concentration (mg/L)	0.2	0.5	1	1.5
Phosphorous concentration (mg/L)	0.1	0.3	0.5	1
Temperature (°C)	22	26	40	45

In conducting the designed experiments, the operation of BTFR system (Step 11) was setup according to designed experiments. Each experiment was operated continuously until it can reach at a steady state and then the experimental results were analyzed in Step 12 using the dedicated Qualitek-4 software to understand the optimum operational condition of the BTFR system accounting for the parameter of pH, temperature as well as phosphorus and nitrogen concentration.

1.4.4 Stage 4: Understanding of FA removal mechanisms in the BTFR

In understanding the FA removal mechanisms in a BTFR system, it is a need to consider certain air pollutant removal mechanisms that has been published in the previous literatures, and the following assumptions have been made, such that: (1) FA is directly absorbed by microorganisms, (2) FA is absorbed and accumulated into nutrient solution because the existing microorganisms are not able of removing it, (3) Firstly, FA is absorbed into nutrient solution and then circulated through the top of biofilter bed to form biofilms at the supporting materials, Secondly, FA can be absorbed by microorganism from the biofilms, and (4) one part of FA present in SCAS can directly adsorb to biofilms and the other parts the FA that have been dissolved in nutrient solution can absorb to biofilms by trickling through the top of biofilter bed. In order to test the FA removal mechanisms in the BTFR system could be matched with one of the above assumptions, the FA concentrations were regularly monitored in nutrient solution and in SCAS during a period of 90 days with and without of microorganisms (Step 13). The production of CO_2 due to microbial metabolisms was determined by a digital analyzer for the operating BTFR of presented microorganisms (Step 14). The FA removal mechanisms can be selected from these above assumptions based on the experimental results analysis (Step 15). Formaldehyde in presence of oxygen can be converted to CO_2 and water. Therefore, if oxygen production can be detected in biofiltration bed, it means the microorganisms are removing FA. Operation and comparison of the BTFR with and without microorganisms can show whether microorganisms are able to remove formaldehyde from air or not. Also, monitoring of FA concentration into nutrient solution can show whether FA can be solved into nutrient solution or not.

1.5 Significance of the study

Polluted air contaminated with a wide range of pollutants is being released into atmosphere by industries. Many of these pollutants are dangerous to human health. Air quality is partially related to the control of pollutants in industrial exhaust air. During this study, a biological system was introduced to control a very dangerous pollutant named FA, which is often released into the atmosphere by various industries. Control of this pollutant is important, as it can cause many negative consequences to human health, including mutagenic, carcinogenic and teratogenic effects. Studies have shown that although physical and chemical methods are effective in removing FA from polluted air, they lack desirable economic impact and eco-friendly qualities. Therefore, results of this study can support or provide knowledge to industries selecting an economically and environmentally friendly system to remove FA from their exhaust air. This study will also help to enhance the living environment and may reduce the risk of human health threats worldwide if the proposed approach is put into practice in the near future. In addition, these results will be significant for industries that facing sustained high concentrations of FA vapor in their exhaust airs. Such concentrations are difficult to remove with physical and chemical methods. The developed models were found matched with the results obtained from the operations of the BTFR at different gas retention times. The use of the models could be useful to having an insight on the reliable design of the BTFR treatment process for removing FA from the air.

1.6 Scope of study

The removal of FA from a contaminated air in this study was performed using a laboratory-scale BTFR and conducted at the Laboratory of Institute of Environmental and Water Resource Management (IPASA), Universiti Teknologi Malaysia. The scope of this research study includes: (1) the calculation of the optimal gas retention time and the evaluation of the effect of gas retention time on the performance of BTFR system based on the development of the models to level of the system, (2) the identification of the predominant microorganisms attached on the supporting materials of the biofilter bed based on the biochemical tests, (3) the optimization of the BTFR system by using an experimental design method (Taguchi method), accounting for the parameters of pH, temperature, nitrogen and phosphorus concentration, and (4) the verification of the mechanisms of formaldehyde removal from SCAS in a BTFR system. The BTFR treatment process was only fed with SCAS to having better knowledge on FA affected the metabolisms of the adapted microorganisms in the filter bed.

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