ARTIFICIAL BARRIER AS TO ENHANCE REMOVAL OF *E.COLI* IN RIVERBANK FILTRATION

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ARTIFICIAL BARRIER AS TO ENHANCE REMOVAL OF *E.COLI* IN RIVERBANK FILTRATION

by

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LIST OF ABBREVATIONS

2D	Two dimension
3D	Three dimension
AKSB	Air Kelantan Sdn. Bhd.
ANOVA	Analysis of variance
APHA	American Public Health Association
BART	Biological activity reaction test
BET	Brunauer–Emmett–Teller
BOD	Biological oxygen demand
COD	Chemical oxygen demand
CV	Coefficient of variation
DF	Degree of Freedom
DO	Dissolved oxygen
DOE	Design of experiment
E. coli	*Escherichia coli*
EBCT	Empty bed contact time
FTIR	Fourier transform infrared
GAC	Granular activated carbon
HW	Horizontal well
ICP	Ion Conductive Plasma
ICS	Ion Chromatography System
IRB	Iron-reducing bacteria
IUPAC	International Union of Pure and Applied Chemistry
MLD	Million liters per day

MSDS	Material safety and data sheet
mTEC	Modified thermotolerant *E. coli*
MW	Monitoring well
NTU	Nephelometric turbidity unit
OFAT	One factor at one time
PSD	Particle size distribution
PW	Pumping well
RBF	Riverbank filtration
rpm	Rotation per minute
RW	River water
SD	Standard deviation
SEM	Scanning electron microscopy
SRB	Sulphate-reducing bacteria
SSE	Sum of squares not accounted by the fitted regression model
SSR	Sum squares due to regression
SST	Sum of squares
TCU	True colour unit
TDS	Total dissolve solid
UKKP	Unit kawalan dan keselamatan pekerja
US	United State
USD	United states dollar
USEPA	United States Environmental Protection Agency
USM	University sains Malaysia
UV	Ultraviolet
VOC	Volatile organic compound

VW	Vertical well
WHO	World Health Organization
WTP	Water treatment plant
XRD	X-ray Diffraction
XRF	X-ray fluorescence

LIST OF SYMBOLS

µm	Micrometer
µS/cm	micro-Siemens per centimeter
Ag^+	Silver
Br^-	Bromide
C_{10}	Breakthrough time
C_{50}	Ineffective time
C_{90}	Exhaustion time
Ca^{2+}	Calcium
CaO	Calcium oxide
C_b	Breakpoint
C_b	Concentration final
cc/g	Cubic centimeter per gram
CFU/mL	Colony forming unit per mililiter
C_i	Concentration final
Cl^-	Chloride
cm	Centimeter
cm/s	Centimeter per second
C_o	Concentration initial
C_o	Concentration initial
CO_2	Carbon dioxide
CO_3^{2-}	Carbonate
CU	Colour unit

F	Ratio of the mean regression sum of squares divided by the mean error sum of squares
F^-	Fluoride
Fe	Iron
Fe^{2+}	Ferrous
Fe_2O_3	ferric oxide
Fe^{3+}	Ferric
FeS_2	Iron sulphide
g	Gram
g/g	Gram/gram
H	Hydrogen
H_2O	Water
H_2S	Hydrogen sulphide
H_2SO_4	Sulphuric acid
HCO_3	Carbonic acid
HCO_3^{2-}	Bicarbonate
Hr	Hour
K	Kelvin
k	Permeability
K^+	Potassium
K_2O	Potassium oxide
KBr	Potassium bromide
K_{TH}	Thomas constant
k_{TH}	Thomas rate constant
k_{YN}	Rate velocity constant

Li^+	Lithium
m	Meter
m^2/g	Meter square per gram
m^3	Meter cubic
$m^3/m^2/hr$	Meter cubic meter square per hour
mg	miligram
mg/L	Milligram per liter
Mg^{2+}	Magnesium
MgO	magnesium peroxide
min	Minutes
ml	Milliliter
ml/min	Milliliter per minute
mm	Millimeter
Mn	Manganese
MPN/L	Most probable number per liter
N_2	Nitrogen gas
Na^+	Sodium
Na_2O	Sodium oxide
NaOH	Sodium hydroxide
NH_4^+	Ammonium
Ni^{2+}	Nickel
nm	Nanometer
N_o	Breakthrough capacity
NO^{2-}	Nitrate
NO^{3-}	Nitric

NTU	Nephelometric turbidity unit
O	Oxygen
ºC	Degree Celsius
p	Lack of fit
PACI	Poly Aluminium Chloride
PO^{4-}	Phosphate
ppm	Part per million
Prob	Probability that the null hypothesis for the full model is true
PtCo	Platinum Cobalt
q	Adsorption capacity
Q	Flowrate
q_0	Maximum solid-phase concentration
q_o	Maximum solid phase concentration
R^2	Coefficient of determination
s	Seconds
SiO_2	Silicon dioxide
SO_3	Sulfur trioxide
SO_4^{2-}	Sulphate
SO_4^{2-}	Sulphate
t_b	Breakpoint time
TiO_2	Titanium dioxide
V	Flowrate of water
Zn^{2+}	Zinc
µm	micronmeter
τ	Time in required for 50% adsorbate breakthrough

PENYARING BUATAN SEBAGAI PEMANGKIN PENYINGKIRAN *E. COLI* DI PENURASAN TEBING SUNGAI

ABSTRAK

Penurasan tebing sungai (RBF) adalah kaedah abstraksi air yang mempunyai pelbagai penghalang untuk menghilangkan banyak bahan pencemar. Bagaimanapun, beberapa tapak RBF melaporkan bahawa pelbagai penghalang ini mungkin tidak berkesan dalam keadaan tertentu. Kajian ini telah membuat pengesahan berkaitan hal itu dan dari hasil pemantauan data 18 bulan (2015-2017) di Lubok Buntar, Kedah, menunjukkan bahawa bahan pencemar yang tidak terdapat dalam tiub telaga wujud pada hari-hari hujan, dan kepekatan awal *E. coli* kebiasaannya tidak wujud pada hari-hari biasa. Untuk mencegah dan merawat abstraksi air supaya penyingkiran *E. coli* dapat dikekalkan dalam operasi untuk jangka masa panjang, kajian ini mencadangkan penyaring buatan untuk aplikasi di tapak RBF. Penyaring buatan adalah penyaring menegak yang mempunyai lapisan karbon aktif granular (GAC) dan zeolit berhampiran tiub telaga. Hasil kajian awal menunjukkan GAC dan zeolit sesuai di mana ia menyingkirkan 100% *E. coli* dalam keadaan berasid. Kajian ini memberi tumpuan kepada penyaring buatan skala makmal dengan menggunakan ujian lajur dan kaedah 'Mixture' dengan 'simplex lattice' digunakan untuk mengoptimumkan perkadaran media dalam menyingkirkan *E. coli*. Pada mulanya, tanah (Tanah A, B dan C) memberikan penyingkiran E. coli tertinggi dengan penghapusan 100%. Walau bagaimanapun, dari masa ke masa, penyingkiran *E. coli* telah menurun dengan ketara dan penggunaan penyaring buatan dengan tanah memberikan penyingkiran yang lebih konsisten berbanding dengan pemilihan tunggal tanah sahaja. Nisbah optimum bagi tanah tempatan A ialah 60% tanah tempatan, 16% GAC dan 24% zeolite. Tanah tempatan B adalah tanah setempat 74% dan zeolite 26%. Tanah tempatan C adalah tanah setempat 62%, GAC 14% dan zeolite 24%. Analisis menunjukkan bahawa model penjerapan untuk *E. coli* mengikuti model Thomas dan tidak Yoon-Nelson. Data optimum yang diperoleh daripada kaedah campuran juga membuktikan bahawa bahagian ini sesuai digunakan di bawah keadaan anaerobik pada kadar alir yang berbeza. Akhirnya, kajian ini menunjukkan keupayaan penyaring buatan meningkatkan keupayaan tanah alluvial untuk menghapus bahan cemar yang berkesan bagi aplikasi RBF sebagai langkah mitigasi. Penemuan ini menyokong keperluan proses penulinan berikutnya, yang dipanggil penyaring perlindungan kedua.

ARTIFICIAL BARRIER AS TO ENHANCE REMOVAL OF *E. COLI* IN RIVERBANK FILTRATION

ABSTRACT

Riverbank filtration (RBF) is a water abstraction method which has a multi-barrier to remove many pollutants. However, some RBF sites report that the multi-barrier may be not effective in certain circumstances. This study has made such related verification and from the monitoring result of 18 months data (2015-2017) at Lubok Buntar, sites in Kedah, showed that pollutants and *E. coli* that were not present in the wells of the tubes appeared on rainy days, and the initial concentration of *E. coli* was mostly absent in normal days. In order to mitigate and pre-treat the water abstraction intake so that the removal of *E. coli* can be sustained in a long term operation, this study suggested an artificial barrier for application at RBF sites. An artificial barrier is a vertical barrier which contain layer of granular activated carbon (GAC) and zeolite near the tube well. The preliminary results of GAC and zeolite to adsorb *E. coli* shows that both media suitable where it removed 100% of *E. coli* in acidic environment. This study focuses on a laboratory scale artificial barrier using a column test and The Mixture methodology concerning simplex lattice was used to optimize the media proportion in removing *E. coli*. Initially, soil (Soil A, B and C) gave the highest of *E. coli* removal with 100% eliminations. However, over time, the removal of *E. coli* has decreased significantly and the application of artificial barrier with soil provides a more consistent removal compared using solitary soil only selection. The optimum ratio for local soil A is 60% local soil, 16% GAC and 24 % zeolite. Local soil B is local soil 74% and zeolite 26%. Local soil C is local soil 62%, GAC 14% and zeolite 24%. The breakthrough analysis shows that the adsorption model for *E. coli* follow Thomas and not Yoon-Nelson model. The optimum data acquire from the mixture methodology also proved that this proportion is suitable to be applied under anaerobic condition at different flowrates. Finally, this research demonstrates the capability of artificial barrier to enhance the alluvial soil characteristics to eliminate contaminants which are effective for RBF application as mitigation measure. These findings support the need of subsequent purification processes, the so-called second protective barrier.

CHAPTER ONE

INTRODUCTION

1.1 Research Background

Globally, a shortage of potable water is an issue that is affecting many countries, including Malaysia. Some of the main reasons why this issue arises are climate change, deterioration of river water quality, unreliable water treatment systems, and increase in population which ultimately leads to poor human health. During dry weather conditions, further depletion of water occurs. Pertinently, climate changes make the drought season become longer and hotter than usual. The dam water becomes low, and the river water could dry up. The deterioration of river water quality in Malaysia has brought an impact to water treatment plants in terms of an increase in treatment cost and maintenance. Chemicals such as PACI, alum, and others have to be increased to treat the polluted river. Thus, water security in the water treatment plants is being doubted, and the treatment process may produce unreliable and unsafe water to the public instead. Utusan Malaysia reported on November 19, 2011 that, through annual laboratory tests conducted on water samples in Kelantan, the Ministry of Health detected heavy metals and harmful bacteria including *Escherichia coli* (*E. coli*) in the water samples from 2008 to 2010. More worryingly, *E. coli* was also found in the water supplied to homes by Air Kelantan Sdn. Bhd. (AKSB). Recently, the Berita Harian reported on October 21, 2018 that, water pipes that have been processed through a water treatment plant can be drinked as they meet Drinking Water standards. However, when it comes to pipes within ten kilometres before the water reaches the house, we do not know whether it is safe to drink. In this regard, Dr Lee said it is better for the public not to drink water directly from the tap but need to cook it first.

Being able to consume reliable and safe potable water is a basic human right (Ralph *et al.*, 2014). Therefore, finding a solution to these issues is highly desirable to improve the safety and reliability of potable water. In 2010, Malaysia began to embark on a new treatment technique, namely riverbank filtration (RBF) (Chew *et al.*, 2015). RBF is a method using groundwater that is expected to provide an alternative for water intake and untapped resources in Malaysia, which was first used at the Water Treatment Plant in Jeli, Kelantan, and Kuala Kangsar, Perak (Siti *et al.*, 2015). RBF is a natural system in which it involves the entry of river water into underground aquifers, caused by hydraulic gradients, whereby water retrieval is from collector wells located at banks at a certain distance from the river (Michael, 2006). Although it is still new in Malaysia, the RBF method has shown good results in reducing costs and maintenance operations in water treatment processes (Hasnul *et al.*, 2011). As a sustainable and natural treatment process that avoids or reduces the use of chemicals, and produces biologically stable water (reduces pathogenic microbes), the system also improves water quality by removing particles (turbidity and suspended solids), organic pollutants, microorganisms, heavy metals and nitrogen (Sharma and Amy, 2009). One previous experience in Germany showed that RBF provides a strong barrier for various pollutants, and can help to ease the temperature fluctuations and pollutant concentration peaks associated with spills into rivers (Schmidt *et al.*, 2003). It also replaces and supports other treatment processes, and reduces the overall costs of water treatment plants (Ray *et al.*, 2002). This is because, during RBF, the removal of sediments, organic and inorganic compounds, and pathogens take place during the first several metres from the river in what is known as the hyporheic zone, which results in groundwater that is cleaner than surface or river water. Thus, the overall costs of water treatment plants are reduced. Furthermore, the hyporheic zone usually presents reducing conditions due to high microbial activity that consumes oxygen in the water. Within this zone, there are important biochemical processes and redox reactions that affect groundwater quality (Lewandowski *et al.*, 2011). In general, every stage of RBF, from the river up until the abstraction well, has an environmental influence (temperature, natural disaster and flood).

However, some of the limitations of RBF include the invisible groundwater flow that makes it difficult to predict the transport of contaminants. A specific concern of RBF limitations is the hydrology and dynamics of the river and groundwater, which have different climate variations (drought and rainy seasons); thus, the groundwater level patterns result in significant fluctuations of contaminants in well stream loads. During dry seasons, minimum and ideal flow rates for pollutants are attached to the local soil. On the other hand, in rainy seasons, the rate of groundwater flow increases to a maximum level, and causes small particles and pollutants to absorb into the local soil where it encloses the flow along the groundwater flow, and initiates pollutants to enter the borehole. Hence, certain biological, inorganic and organic contaminations may exist in borehole water due to this. Moreover, since maximum groundwater flow rates occur frequently in Malaysia, this incident is predicted to often result in significant fluctuations of underground hydraulic conductivity of groundwater and the shock load of pollutants. A significant amount of pollutants may exist in borehole water due to this high hydraulic conductivity and local soil feature (Stephen *et al.*, 2003), which concludes that RBF is a natural treatment method that depends on natural behaviour. In general, the quality of RBF water is influenced by environmental conditions, where managing groundwater is important to ensure that water is aligned in compliance with government legislation, and environmental protection measures.

1.2 Problem Statement

Riverbank filtration (RBF) offers a naturally safe water source for public health as it can remove most contaminants in the river. However, it is worth mentioning that the RBF process is beneficial but has some limitations as it is influenced by natural behaviours. RBF may seem incapable of removing certain biological, inorganic and organic contaminations associated with limitations by hydrology and the dynamics of the rivers and groundwater (Schubert, 2002).

The shortage of water during drought season is currently an issue in Malaysia which cannot be ignored. Because of that, the usage and study of untapped resources such as groundwater have been initiated either by the Malaysian government or private institutions. The deterioration of the quality of water sources occurred as a result of the surrounding urbanisation, which introduced non-point source pollution. More than that, the impact of using high quantities of chemicals in treatments cannot be ignored as the chemicals will affect human health. Therefore, this transition in water treatment method is highly desirable to reduce the use of these harmful chemicals.

According to site monitoring data by Eckert and Teermann (2006), and Schubert (2002), while most raw waters already fulfilled the Drinking Water Standard, higher colony counts (coliform bacteria and *Escherichia coli*) were observed in the production wells following flood events (level of river water was high). The flood events caused an increase in the hydraulic conductivity in groundwater originating from infiltration (Marco, 2014), while the increase of hydraulic conductivity was attributed to the increase in the distance of pollutant transports such as *Escherichia coli* (*E. coli*), and *Cryptosporidium parvum* (Laura, 2007). In certain conditions, iron and manganese contents were higher because of the natural process of mineralisation. In addition, the problem of the ‘rotten egg’ smell in the water was due to the process of decomposition by sulphate-reducing bacteria (SRB) resulting in methane gas. However, this did not happen at all sites because it depended on well depth—deeper aquifers contained high organic matter and minerals, which resulted in more hydrogen sulfide gas when coming into contact with water.

The water abstracted from RBF usually has good turbidity, colour, and fewer micro-pollutants, owing to the simple water treatment process used, which is also cheap and has low maintenance costs (Marcela, 2012). However, some pollutants may not be removed by RBF, which may require post-treatment. There are various post-treatment methods used to treat the abstracted water from RBF, such as ozonisation, and ultrafiltration system. Ozone and ultrafiltration systems are used to oxidise or remove iron and manganese that are picked up in the aquifers. An activated carbon filter is used for adsorption and protection against more persistent contaminants (Schmidt *et al*., 2003: Chew *et al*., 2015). One disadvantage of the post-treatment, however, is during shock loads and clogging in RBF, a relatively high concentration of solubles will place loads on the treatment process at the filtration or activated carbon filter. Apart from that, small particles continue to flow along with water due to high pressure and, in certain circumstances, spillage from industrial accidents cause some major problems in the water treatment process. For example, since the late 1950s, water quality of major rivers in Europe has begun to deteriorate, and high waste water inputs have threatened the use of the bank filtrate (Sontheimer, 1991). Therefore, emergency protection measures for RBFs were taken to solve the problem, such as monitoring the activities of waterworks by water-industry associations, enforcement by authorities with industries, creating transborder housing programmes, and the closure of the industries themselves. However, though these efforts were viable, they had certain limitations, especially in terms of the nature of transboundary conflicts, and unpredictable natural disasters such as floods (Choudhury and Islam, 2015).

The load increase in the treatment process creates huge by-product wastes and costs, which is harmful to the environment and human beings. Hence, this requires a new management plan that is economical, efficient, and effective, that gives benefits to the operators, society, and environment. In addition, the spectacular spills, for example, the Sandoz accident on November 1, 1986 (Sontheimer, 1991) has highlighted the need for a barrier for sanitation measures and pollution control. In this study, the existence of RBF and artificial barriers is seen as an effective new purifying method to maintain safe water abstraction.

This (artificial barrier) pre-treatment or purifying method is to improve the effectiveness of RBF in removing pollutants during shock loads, and reduce the load placed on the water treatment process. Due to that, this study suggests an implementation of an artificial barrier for microorganisms in RBF so as to sustain the good water quality abstracted from the abstraction well. The microorganism that will be monitored during the experiments is *E. coli*, and it is preferred as an indicator because it is the WHO standard. The objective of this study is to improve the water quality produced in the water abstraction well for RBF using an artificial barrier to retain the removal of *E. coli* during a high load. The mentioned artificial barrier used in this study comprises of a mixture between granular activated carbon (GAC), and zeolite. GAC and zeolite are organic anti-microbial adsorbents, and are widely used as filter medias to remove pollutants such as heavy metals and microorganisms (Chojnacka *et al.*, 2004; Jocelyne *et al.*, 2012). Furthermore, because of their chemical and mechanical stabilities, high adsorption capacity, and high degree of surface reactivity, GAC and zeolite are considered as ideal adsorbents over other existing adsorbents.

1.3 Gap of Knowledge

Riverbank filtration (RBF) is a new approach in Malaysia which introduces natural treatment. It involves the inflow of river water to the underground aquifers, which is induced by the hydraulic gradient, but the efficiency of the system depends on the natural behaviour of the location. In order to design the RBF system in Malaysia, this study suggests to enhance its efficiency on the local soil structure of the site location with the application of an artificial barrier in the system. This application of the artificial barrier has not been applied at any other known RBF sites. The main criteria of focus in this research is the characteristics of the alluvial soil (local soil) structure after application of the artificial barrier (combination of GAC and zeolite) at the laboratory stage.

However, separation of GAC from the post-water treatment process may result in blocking of the sand filters, and loss of adsorbents. In fact, this drawback even creates a secondary pollution in the system. The applications of GAC and zeolite under aerobic conditions are not significant because the zeta potential of both materials are negative, unless they are prepared to adsorb metals and organic parameters simultaneously. Apart from that, zeolite particles tend to dissolve in solutions when left in the filtration matrix with mineral acid. The changes in the zeolite particles may cause them to be not suitable to be used again, and thus, result in wastages. Therefore, there is an essential gap of knowledge in the applicability of GAC and zeolite in the same filter media with alluvial soil in RBF.

Consequently, the applications of GAC and zeolite in RBF are not well explored. Besides that, the usage of these two substances in RBF are bound to differ due to anaerobic underground conditions, and sub-surface water flow rates that are influenced by the weather conditions. The applications of GAC and zeolite in groundwater would enhance the adsorptive properties towards effective parameter removals. By having hydrophobic and hydrophilic characteristics from cross-link processes in anaerobic and CO_2 conditions, the surface of the adsorbents are modified, and therefore the surface charge of the filter medias (artificial barrier) can be neutralised or reversed as their surface may change from hydrophobic to hydrophilic, and vice versa. The redox process underground will produce more CO_2, which results in the increase of groundwater pH to become acidic, which may change the hydrophobic or hydrophilic nature of the media.

In this work, the best ratio for local soil, GAC and zeolite is developed and tested for parameter removals through river water treatment by batch and fixed bed studies. The individual precursor's performance is determined before establishing optimal conditions for the mixture. By using Mixture in Design Expert, the optimal filter ratio with respect to *E. coli* removal can be obtained. The prepared ratio filter is characterised physically and chemically in order to determine its adsorptive characteristics. Furthermore, the optimal filter ratio is also further tested in fixed bed studies to obtain the breakthrough and suitable dynamic model. Dynamic adsorption models are utilised to understand the adsorption behaviour of the artificial barrier adsorbents. Finally, the spent filter ratio is continued to anaerobic (CO_2 partial pressure) studies to determine the most appropriate removal efficiency. As a final point, the data from the fixed bed studies is used in the filter ratio adsorbent for designing the filtration bed accordingly.

1.4 Research Objectives

The following are the objectives that this study seeks to achieve:

i. To characterize the pollutants (*E. coli*, iron, manganese, etc.) present at the Lubok Buntar riverbank tube well and Sungai Kerian.

ii. To determine the suitability of adsorbents (GAC and zeolite individually) to be applied as an artificial barrier in RBF via batch study, and permeability in relation to *E. coli* removal.

iii. To determine the optimal ratio of combination for soil with GAC and zeolite as an artificial barrier.

iv. To compare the effectiveness of using only soil, and a soil mixture with GAC and zeolite on *E. coli* removal at different water flow rates, based on breakthrough curve analysis, dynamic adsorption model, and the effects of anaerobic conditions (with partial pressure CO_2) on the soil and artificial barrier.

1.5 Organisation of Thesis

This dissertation is divided into the following chapters:

Chapter 1: Introduction

A brief introduction to the research work, problem statement, gap of knowledge, and research objectives is provided.

Chapter 2: Literature Review

The science of RBF in Malaysia, RBF water quality composition, RBF treatment, adsorbent materials, as well as the utilisation of the Mixture in Design Expert for the design parameters and optimisation are explained in this chapter.

Chapter 3: Methodology

This chapter presents the experimental designs and procedures for the batch studies, and fixed bed flow studies. In addition, the site location, sampling procedure, types and properties of the materials used, as well as filter adsorbent preparations are described here. Besides that, the descriptions of the method used to determine RBF water properties, operational variables, optimisation sequence using Mixture in Design Expert, and the dynamic adsorption model implemented in this study, followed by the effects of CO_2 partial pressure (environmental stress) are also included in this chapter.

Chapter 4: Results and Discussion

This chapter imparts the characterisation of RBF water and optimal filter ratio for the removal of *E. coli* from river water as obtained from the batch test and continuous flow studies using the artificial barrier adsorbents. The equations for the removal of *E. coli* in terms of its individual process parameters, and their interactions are presented and extensively discussed. Furthermore, the dynamic adsorption models, and the results of CO_2 effects obtained from the experiment are reported in this chapter. Lastly, the implementation of the dynamic adsorption models and fixed bed flow studies is performed in order to design the filtration bed for on-site application.

Chapter 5: Conclusion and Recommendations

The conclusion and recommendations based on the research findings are discussed, and future work prospects are also elaborated on in this chapter.

CHAPTER TWO

LITERATURE REVIEW

2.0 Introduction

This chapter consists of six sections. The first section discusses about the riverbank filtration (RBF) and its water quality components including pre-treatment options (2.1 - 2.5). The second section consists of environmental stress effect to RBF optimization (2.6). The third section consists of adsorbent material for artificial barrier including the types of adsorbent, their applications and characteristics (2.7 – 2.8). The fourth section discusses test methods, bench scale and fixed-bed flow studies. The process behaviours are also discussed in detail (2.9). Finally, in the sixth section, it extensively discusses the statistical analysis and dynamic adsorption models used in this study including the principles and application of Mixture design with Design expert Software (2.10).

2.1 Riverbank filtration

2.1.1 Application and basic principle

RBF post water treatment has been employed since the nineteenth century (Ray *et al.*, 2008). During RBF, river or lake water is extracted indirectly by drawing it through the subsurface prior to use as shown in Figure 2.1. The extraction is accomplished by using a well infiltration line either vertical or horizontal. The well is located at a short (below 30 m) to intermediate (up to 60 m) distance from the riverbank or lake (Eckert and Irmscher, 2006). During the extraction of water, the groundwater that is discharged into the river decreases and the groundwater table near the waterline may decrease (up to 1-15 m, depends on aquifer type of soil) below the river water level which later results in surface water entering the aquifer and flowing to the abstraction well. To ensure a satisfactory purification, the distance between the river and the extraction well should be such that the travel time exceeds 30 to 60 days. This will also ensure a satisfactory reduction of microbial pollutants (Huisman and Olsthoorn, 1983).

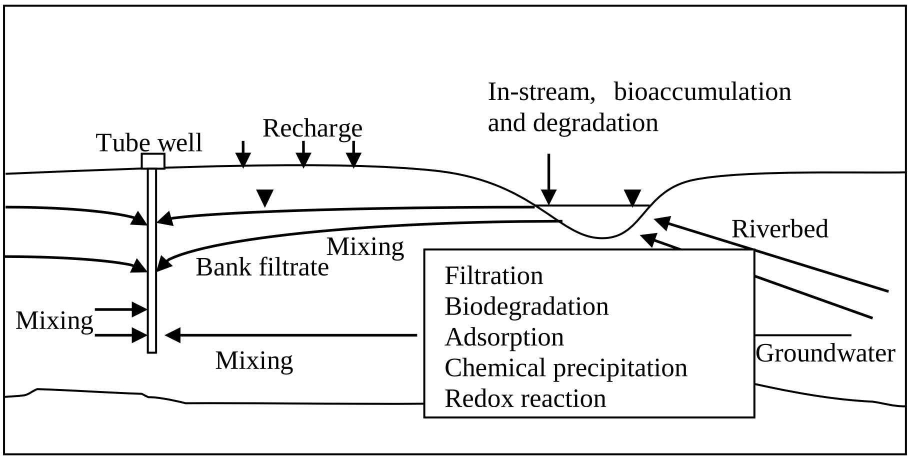


Figure 2.1: Scehmatic factor effect RBF system
(Sources: Hiscock and Grischek ,2002)

During infiltration and travel through the soil and aquifer sediments, surface water is subjected to a combination of physical, chemical as well as biological process of filtration. The top few centimeters of the riverbank materials formed is a screen or filter media that removes the suspended solids present in the water which acts as a physical process (Gutiérrez *et al*., 2017). Heavy metal, phosphorous and hydrophobic organic compounds present in the water are removed by adsorption into certain aquifer materials which acts as a chemical process. In the presence of biomass or when a particle becomes attached to the biofilm, the organic matter is further biodegrade by microorganism which acts as a biological process (firstly, under oxic conditions then later, under anoxic conditions). However, the water quality in most cases will improve by dilution of the source surface water with native groundwater (Gina, 2003).

In all that filtration process, there is a general agreement that the mechanisms where straining, adhesion, attachment, chemical adsorption, sedimentation and biological growth all operate to some extent according to the types of pollutants. Pollutant such as suspended solids is strained between soil pores. As for colour and COD, they are removed once a particle has been put in contact with the soil surface and this is known as a chemical adsorption mechanism. While phosphorus is removed by sedimentation mechanism. Meanwhile, to filter microbial, there are two potential mechanisms that can be used. Either by attachment cell to soil surface or by biological growth within the soil particles. Interception happens when particles are carried by one of the streamlines closest to the sand grain and a brushing effect occurs. The summary of mechanisms during infiltration process is shown in Table 2.1.

Table 2.1: Summary of mechanisms during infiltration process.
(modified from *Metcalf and Eddy, 2004*)

Mechanism	Description	Pollutants	References
Straining	Particles larger than the pore space of the filtering medium are straining out mechanically Particles smaller than the pore space are trapped within the filter by chance contact	Suspended solid	Juan *et al.*, 2017
Adhesion	Particles become attached to the surface of the filtering medium as they pass by. Because of the force of the flowing water, some material is sheared away before it becomes firmly attached and is pushed deeper into the filter bed.	DOC and microbial	Vasiliki and Robin, 2007
Attachement	The acquisition of cells from the bulk liquid by an existing biofilm	Microbial	Unger and Collins, 2006

Table 2.1 continue

Chemical adsorption	Once a particle has been brought in contact with the surface of the filtering medium or with other particles, either one of these mechanisms, chemical or physical adsorption or both, may be responsible for holding it there	Colour and COD	Weiyan *et al*., 2018
Sedimentation	The particles settle on the filtering medium within the filter	Phosphorus	Regnery *et al*., 2015
Biological growth	Biological growth within the filter will reduce the pore volume and may enhance the removal of particles with any of the above removal mechanisms by microbial degradation process.	Microbial & organic contaminats (pesticides, herbacides, odour compounds and pharmaceuticals)	Marcela, 2012

2.1.2 Advantages and disadvantages

RBF treatment is a sustainable natural treatment process which avoids or reduces the use of chemicals, and produces biologically stable water. The system improves water quality by removing particles (turbidity and suspended solid), organic pollutants, microorganism, heavy metals and nitrogen. It also helps to dampen the temperature fluctuation, allowing concentration to peak when it is associated with spills into a river or lake. This treatment process also replaces and supports the other treatment processes by providing a robust barrier for multiple contaminants and reduces the overall cost of water treatment (Ray *et al*.,2002).

The improvements made by RBF treatment also helps reduce the overall cost of water treatment by lowering the costs associated with the operation and maintenance of the primary treatment. This leads to low life-cycle costs when compared to treatments without the use of RBF treatment. It is proven that the use of RBF can reduce the treatment costs by 10-20% in comparison to traditional pretreatments. The major savings stem from a reduction in capital costs as well as a reduction in O&M expenditures (Stephen and Carollo, 2006). Some RBF sites show that its use can reduce up to 65% of capital costs and 45% of operational costs (Ismail, 2012). Overall, the ability of RBF to serve as a standalone pretreatment to primary treatments is dependent upon site-specific water quality and aquifer conditions.

One of the major consequences when applying an RBF system is the occurrence of clogging effects in the alluvial aquifer. A distinction is drawn between three different types of clogging. They are, mechanical clogging, biological clogging and chemical clogging. Mechanical clogging happens when suspended matter intrudes into the alluvial aquifer from the flow that leads towards the well and subsequently clogs the voids of the adjacent soil layers. While biological clogging refers to the effect when microorganisms form a biological film and thereby constrict the voids of the alluvial aquifer. Chemical clogging however, are described as the effect of a reduced hydraulic conductivity due to clogging by chemical precipitants. The precipitation of substances can emerge from a high level of biodegradable matter which causes changes in the redox-potential and pH level of the river water. Furthermore, the potential for chemical clogging is represented by the presence of iron, ammonia and nitrate concentrations, and the hardness of the water (Alexandra *et al*., 2007). The actual biochemical interactions that sustain the quality of the pumped bank filtrate depends on numerous factors, including aquifer mineralogy and the extent of the aquifer (Hiscock and Grischek, 2002). The geochemical context of river-aquifer transfers and their evolution is a reflection of interactions between biological and physiochemical mechanisms. In addition to influencing the chemistry, the bacterial activities may also affect the hydrodynamic parameters of the soil media (Doussan *et al.*, 1997).

2.1.3 Factor influence performance of RBF

The most important things to consider in an RBF design are the water quality (which will be discussed in section 2.3) and the capacity of water that can be abstracted. Therefore, in order to ensure water quality and capacity is enough, there are four basic important criteria that needs attention as it will affect the performance of the RBF. They are hydrogeological conditions, source water quality and mixing with native groundwater, distance of the well from the riverbank and spacing of wells as well as pumping rates, and sediment permeability. The effectiveness of an RBF in removing surface water contaminants depend largely on hydrogeological conditions. It is all about the soil microbiology, characteristic of the bank materials and streambed, as well as scouring characteristic (Sahoo *et al.*, 2005). In many countries, the alluvial soil aquifers are hydraulically connected to a water course. This would be the preferred sites for drinking water production (Doussan *et al.*, 1997).

In order to study more about RBF design, reviews of RBF designs from other countries are made and summarized in Table 2.2. There are two types of collector wells in an RBF. These are a horizontal well (HW) and a vertical well (VC) as in Figure 2.2. Some RBF sites applies both types of collector wells.

Table2.2: Summary of RBF design from other country

Country, river	**Soil type**	**Collector well depth (m)**	**Distance from river (m)**	**Capacity (MLD)**
China, Yellow River (Hu *et al.*, 2016)	Sand gravel, coarse sand and sand	VW: 65 HW: -	272	0.02
Germany, Elbe River (Grischek., 2003)	Fine sand and silt, medium sand and gravel	VW: 30-50 HW: 20	300	150
Netherland, Lek River (Hamann *et al.*, 2016)	Fluvial sand or fluvial gravel with sandy clay at base	VW: 20-40 HW: -	370-906	0.01
Korea, Nakdong River (Lee *et al.*, 2017)	Sand and gravel with several silt and clay	VW: 12.5 HW: -	150	10
Egypt, Nile River (Abdalla and Shamrukh, 2010)	Sand and gravel, little clay	VW: 60 HW: -	20-80	22
India, Kali River (Cady, 2011)	Brownish red silty loam	VW: 18-23 HW: -	29-79	0.8
Kentucky, Ohio River (Stephen-Hubbs *et al.*, 2003)	Sand and gravel with several silt and clay. Limestone layer at 40-48 depth.	VW: 30 HW: 512	20	80
US, Great Miami River (Sheets *et al.*, 2002)	Most sand, some gravel and laterally bounded by limestone	VW: 60 HW: -	50	57-61
Malaysia, Sungai Semerak (Chew *et al.*, 2015)	Gravelly sand	VW: 6-12 HW: -	10	25

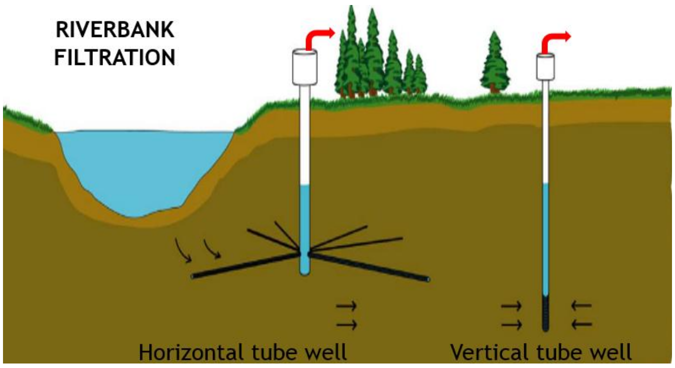


Figure 2.2: Schematic diagram for horizontal and vertical tube well

The depth and distance of the collector wells from the river is determined by the capacity of water that can be abstracted. A working RBF shows a decrease in RBF water levels when the distance of the well is further away from the riverbank. In addition to the decreasing RBF water level due to increment of distance, it is found that there is no cross flow of natural groundwater in which the well could abstract the river water (Shamrukh and Ahmed, 2011). Which means, although the well is deeper and reaches groundwater, more river water is needed consistently in order to abstract a huge capacity of water. This is proven in a pumping test where the results show that the water in the well (below 60 m) comes from the river water (Mohamad *et al.*, 2013).

However, if the well is short, the low-laying shallow aquifers are generally fragile, which can easily deplete due to anthropogenic activities and over exploitation of groundwater and agriculture. But the collector wells can be placed far from the river if the soil type is of sand and gravel, such as the RBF at Yellow River, China. In addition, the combination of HW and VW can maximise the water capacity such as the RBF at Elbe River, Germany. But in other cases, although they have a combination of HW and VW, clayey alluvial soil will limit the water capacity as seen at the RBF site at Lek River, Netherlands. At this RBF, it shows that the water capacity is only at 0.01 MLD in comparison to the RBF placed in clayey alluvial soil at Nakdong River, Korea which can abstract 10 MLD water capacity. The reason being, for clayey alluvial soil types, a collector well needs to be built near the riverbank and at a deeper depth. For example, the collector well at Nakdong River, Korea which abstracts 10 MLD at 150m distance from the river, but compare that to the collector well at Nile River, Egypt that has a capacity of 22 MLD.

The Kali River, India is a highly pollutant river which demands the RBF method to be used. However, the collector well can only abstract a mere 0.8 MLD of water due to the low transmissivity of brownish red silty loam alluvial soil. Hence, other than building wells nearer to rivers, limestones can be added to RBF sites with clayey alluvial soil to increase the transmisivity of the water, such as the RBF sites located at Ohio River, Kentucky and Great Miami River, US.

In Malaysia, the RBF site at Sungai Semerak (refer Figure 2.2) contains gravelly sand and a shallow vertical well collector type. According to data obtained from the monitoring wells, the shallow geology of the RBF area is related to the alluvial deposition from the river which usually consists of upper fine, medium, and lower fine sand layers (Lee *et al*., 2009). However, research also shows that there are some RBF sites that have silt or clay mixed with sand. And this exists at several depths in the layers (Water authority of Changwon City, 2003). The shallow collector well and its position nearer to the riverbank helps the RBF to avoid problems with iron and manganese. Hence, the largest capacity RBF site holds a water supply of 25 MLD. The actual biochemical interactions that sustain the quality of the pumped bank filtrate however, depends on numerous factors, which includes aquifer mineralogy and the extent of the aquifer (Hiscock and Grischek, 2002).

So, to manage and plan an efficient RBF design, the characterisation and understanding of the nature of the aquifer such as soil and rock types is crucial to elucidate their geochemical nature and its relation to abstracted water.

2.2 Riverbank filtration in Malaysia

The shortage of water during drought seasons is what brought about the application of RBF technologies in Malaysia namely in Kelantan and Perak. Studies of untapped resources such as groundwater were started either by the Malaysia government or by private institutions. The combination of water purification methods work by filtering river water through alluvial soil near the riverbank as pre-treatment. The river water is pumping out from an abstraction collector well which supplies to the water treatment systems known as RBF. This method is expected to provide enormous water supply and a new reliable water treatment method. In comparison, the quality of river water varies over time, while water gotten through RBF yields consistent high-quality drinkable water.

RBF technologies have begun to be extensively used in Malaysia as to optimise the water supply. Most RBFs in Malaysia are applied in the areas of Kelantan (Hasnul *et al*., 2011). The introduction of RBF in Malaysia began in 2010 at Jeli, Kelantan. The plant operations have demonstrated the success of combining RBF (as pre-treatment) with water treatment plant (as post-treatment), resulting in a reductions of water treatment costs where 1 m^3 of drinking water equals approximately USD 0.04. This is considered a competitive price for Malaysians (Chew *et al*., 2015). These findings should pave the way for other municipal authorities to introduce their own RBF systems. Figure 2.3 shows the RBF at Wakaf Bunut water treatment plant, Kelantan, Malaysia.

Figure 2.3: RBF at Wakaf Bunut water treatment plant, Kelantan, Malaysia
(Sources: Chew et al.,2015)

2.3 RBF tube well water characteristics

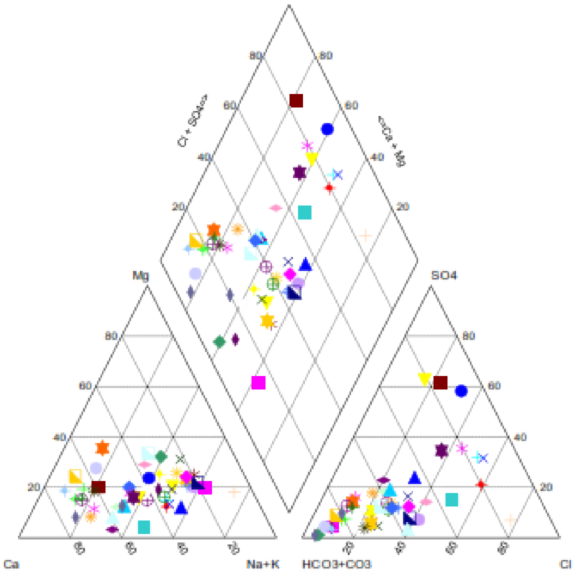
2.3.1 Hydrogeochemistry in RBF

It is important to understand the hydrochemistry of RBF when it comes to analysing the minerals in the water that effects its quality. The water in the abstracted tube well consists of both groundwater and river waters. Both groundwater and river water that moves along its flow paths in the saturated zone, will increase the total of dissolved solids and most major ions normally occur here. The major ion change in the following sequence results in the reaction shown in R1.

$$\mathrm{HCO_3}^- \rightarrow HCO_3^- + \mathrm{SO_4^{2-}} \rightarrow \mathrm{SO_4^{2-}} + \mathrm{HCO_{3-}} \rightarrow SO_4^{2-} + Cl^- \rightarrow Cl^- + SO_4^{2-} \rightarrow Cl^- \qquad \text{R1}$$
$$\text{Bicarbonate waters} \rightarrow \text{Sulphate waters} \rightarrow \text{Chloride waters}$$

Bicarbonate water occurs near the earth surface, while chloride water occurs in the deeper geological strata. Furthermore, water with high salinity have higher specific gravity and tends to occupy the lower strata. Hence, bicarbonate waters occur at shallow depth while sulphate waters is of transitional type (Ponce, 2012). Groundwater hydrochemistry concentrations can be plotted on several type of diagrams in order to create a visual image of the water quality. Piper diagrams are widely used to present and classify major ions for groundwater and summarise the main contrasts in hydrochemical composition between different water sources in a river basin (Zhang *et al.*, 2017).

Results of major ions analysis have been plotted as a Piper diagram using the Aquachem 5.1 software. The Piper diagram consists of two triangles and a diamond which is cation (Ca^{2+}, Mg^{2+}, Na^+ and K^+) and are plotted as a point on the left triangle while the anions (Cl^-, SO_4^{2-}, CO_3^{2-} and HCO_3^{2-}) are on the right as shown in Figure 2.4. A point on the diamond is plotted where two lines intersect. The values are calculated as percentages of cation or anions in equivalent per litre (Kehew, 2001). These diamond diagrams show the different types of water. The top quadrant represents calcium sulphate water (gypsum groundwater and mine drainage), while the left quadrant is calcium bicarbonate water (shallow fresh groundwater). The right quadrant shows sodium chloride water (marine and deep ancient groundwater) and the bottom quadrant is sodium bicarbonate water (deep groundwater influenced by ion exchange.



(a)

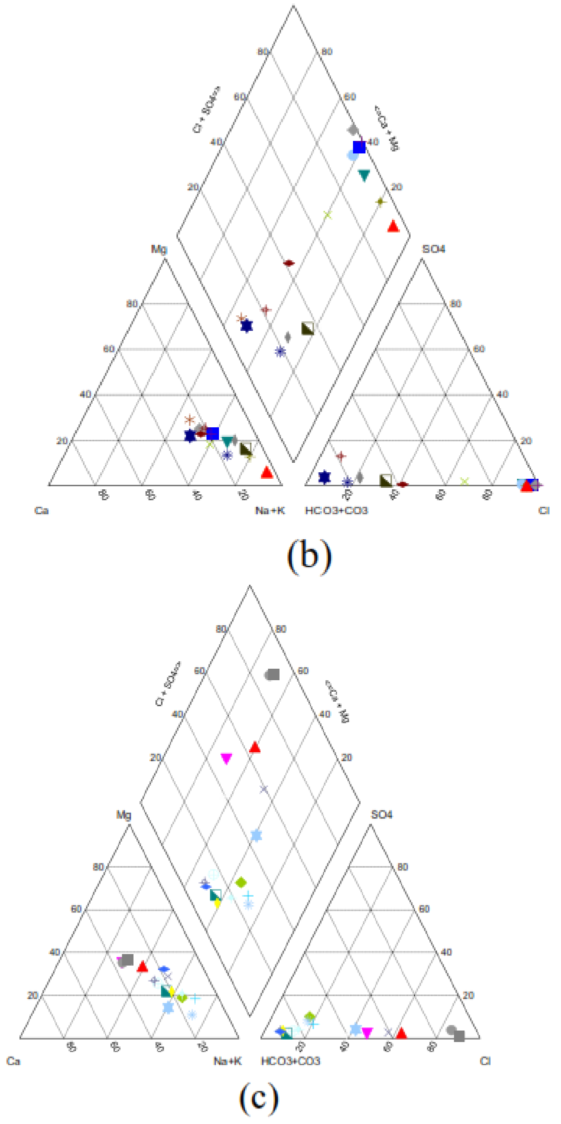


Figure 2.4: Guides for Piper Diagram in (a) shallow groundwater, (b) intermediate groundwater and (c) deep groundwater (Anuar *et al*., 2015)

Besides being able to show graphically the nature of the given sample, the Piper diagrams may also determine the relationship between the other samples (Sultan *et al*., 2009). It can be used to describe various hydrochemical processes, such as base cation exchange, cement pollution, mixing of natural waters, sulphate reduction, saline water (end-product water), and other related hydrochemical problems. In context of micro-organism, the hydrochemistry studies facilitates to expand the exploration of the bacterial diversity such as sulphate-reducing bacteria (SRB) and