# DEVELOPMENT OF GROUNDWATER FILTRATION SYSTEM USING MULTISERIES MEDIA

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A thesis submitted in fulfillment of the requirement for the award of the Doctor of Philosophy.

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### **DEDICATION**

I dedicate this thesis to my wonderful husband, Rashidan Ismail. Thank you for your encouragement and love that giving me strength to chase my

dreams.

This manuscript is heartly dedicated to my beloved children Adam Najmi, Nur Sarah 'Aisyah, Nur Farah Khadijah and Muhammad Amirul Amri who have been a constant source of support and encouragement during the challenges of graduate school and life,

To Mak Ton, Mak Biah and Ayah,

Thank you for your unconditional love and prayers throughout my life.



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### **ABSTRAK**

Air liat adalah masalah utama yang dihadapi oleh punca air bawah tanah. Ini berpunca dari kandungan Mg<sup>2+</sup> dan Ca2<sup>+</sup> yang tinggi. Objektif utama kajian ini adalah menghasilkan sistem rawatan air liat menggunakan pasir, zeolite dan RHAC sebagai media penapis. Media-media ini akan ditempatkan didalam turus yang berlainan dan akan disusun secara bersiri (MSMC). Tujuan utama bagi mengasingkan setiap media untuk memastikan masa luput yang tepat bagi setiap media dapat diramal. Ini adalah langkah bagi mengatasi masalah penggantian media yang sedia ada. RHAC dibangunkan dengan kaedah pengaktifan kimia menggunakan asid nitric berkepekatan 20% untuk rendaman yang bertindak sebagai agen pengaktifan. Suhu karbonisasi yang dipilih adalah pada 450°C. Pencirian bagi semua media dilakukan dengan menggunakan analisis FESEM, SEM-EDX, XRF, dan FTIR bagi rupa bentuk permukaan, komposisi unsur, dan kumpulan berfungsi pada media. Penjerapan paling efisien pada pH 6.5 untuk Mg<sup>2</sup> + dan Ca<sup>2</sup> + pada 60 minit masa sentuhan dengan suhu 30°C menggunakan air bawah tanah pada kepekatan 168 mg / L bagi  $Mg^{2+}$  dan 250 mg / L bagi  $Ca^{2-}$ <sup>+</sup> . Kajian FTIR mendapati bahawa kumpulan berfungsi pada permukaan media bersifat memudahkan penjerapan ion-ion logam. Data keseimbangan untuk Mg<sup>2</sup> <sup>+</sup> dan Ca<sup>2</sup> <sup>+</sup> paling sesuai dengan penjerapan Langmuir isoterm dengan kapasiti penjerapan maksimum masing – masing adalah pada 400 mg/g dan 302 mg/g. Model Pseudo - Second-order paling tepat menggambarkan data kinetik. Semua media berjaya menyingkirkan sehingga 99% dan 98% bagi ion metal Mg<sup>2 +</sup> dan Ca<sup>2 +</sup>. Proses penyaringan menggunakan turus penjerapan lajur dengan media yang disusun secara bersiri. Takat lupus adalah pada 200 minit untuk pasir, 100 minit untuk zeolitet dan 260 minit untuk RHAC. Kaedah terbukti berkesan pada kadar 90%. Jumlah isipadu air terawat sehingga semua kapasitas media berkeadaan tepu adalah 3.5L. Kaedah rawat dari kajian ini berjaya merawat air liat dan memenuhi tahap standard kriteria air minum yang disarankan oleh Kementerian Kesihatan Malaysia (KKM).

#### ABSTRACT

High concentration of Mg<sup>2+</sup> and Ca<sup>2+</sup> are caused of the groundwater hardness. This study developed a multi series media column as a filtration system for hard water treatment. Sand, zeolite, and rice husk activated carbon were used as filter media and placed individually in the column. The design of the column overcomes the issue of filter media replacing conditions. Rice husk activated carbon (RHAC) for MSMC media was developed using chemical activation methods with nitric acid (HNO<sub>3</sub>) as the impregnation agent. The chemical activation was adopted using 20% nitric acid in the impregnation process and 450°C for carbonization temperature. The characterization of all media was performed by means of FESEM, SEM-EDX, XRF, and FTIR analysis to observe the surface morphology, elemental composition, and functional groups available on the media. Sorption was most efficient at pH 6.5 for both Mg<sup>2+</sup> and Ca<sup>2+</sup> at contact time 60 minutes with temperature 30°C using 168mg/L Mg<sup>2+</sup> and 250mg/L Ca<sup>2+</sup> concentration in the groundwater. FTIR study revealed that functional groups on the media surface facilitate the adsorption of the metal ions. The equilibrium data for Mg<sup>2+</sup> and Ca<sup>2+</sup> were best fit to the Langmuir adsorption isotherm with maximum adsorption capacity of 400 mg/g and 302 mg/g, respectively, while the Pseudo – Second-order model best describes the kinetics data. MSMC successfully removed up to 99% and 98% respectively Mg<sup>2+</sup> and Ca<sup>2+</sup>. Breakthrough points were found at 200 minutes for sand, zeolite 100 minutes for zeolite and 260 minutes for RHAC. The combination of screening and adsorption process by combining these media, enhanced the treatment efficiency more than 90%. Total volume of treated water until all the capacity of the media is saturated was 3.5L. The treatment successfully reduces the water hardness and comply with the Ministry of Health (MOH) standard for drinking water supply.

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### LIST OF SYMBOL AND ABBREVIATION

C Solute/outlet concentration

 $C_{b}$ Effluent concentration

 $C_{ad}$ Different between inlet/initial and outlet

equilibrium concentration

The concentration of adsorbate desorbed  $C_{de}$ 

 $C_{e}$ Concentration of adsorbate at equilibrium

 $C_{i}$ Constant for Intra-particle diffusion model

 $C_t$ The concentration of adsorbate at the time, t

 $C_{0}$ Initial/inlet adsorbate concentration

 $K_F$ Adsorption or distribution coefficient for

Freundlich isotherm

The rate of adsorption for Langmuir isotherm  $K_{L}$ 

 $\mathbf{k}_1$ 

Adsorption rate constant for pseudo-second-order
Nitric Acid  $k_2$ 

HNO<sub>3</sub>

Constant for Freundlich isotherm n

Adsorption capacity for Langmuir isotherm  $Q_{o}$ 

Amount of adsorbate adsorbed per unit mass of

adsorbent at equilibrium

Amount of adsorbate adsorbed per unit mass of

adsorbent at the time, t

Calculated adsorption uptake at the time, t qt, cal

Experimental adsorption uptake at the time, t qt, exp

 $R_{L}$ Separation factor

 $\mathbb{R}^2$ Coefficient of determination

 $S_{BET}$ BET surface area

Т Time

MOH Ministry of Health

RH Rice Husk

RHAC Rice Husk Activated carbon Vmeso - Mesopore volume

 $V_T$  - Total pore volume

BTGW - Bukit Tembaga Groundwater



### **CHAPTER 1**

### INTRODUCTION

### 1.1 Research background

Water is a prime natural resource and a basic necessity for sustaining life on earth, which is essential for human and industrial development. Supplying adequate amount of potable water to the global population is a gigantic task in the wake of growing industrial and domestic needs. In the twenty-first century, environmental problem is one of the biggest issues all over the world. Water pollution is one of them. An increase in population leads to an increase in the demand for clean and safe water supply.

In Malaysia, one of the main sources of water supply for domestic use is from treated surface water which will distributed through a centralized piping system by municipal (Razak *et al.*, 2015). However, the current supply of clean water sources is steadily decreasing (Shahabudin, 2014). A few factors are contributing to the decrease of water such as water pollution, water governance which not maximize of alternative water resources and high cost of water filtration system (Halder, 2015; Sukereman, *et al.*, 2013; Shull, 2012; Tang, 2012). Due to this phenomenon, Ahmad (2014) in his research, found that groundwater is one of the best alternative sources for clean water supply.

The determination of groundwater quality around Peninsular, the description of the problems associated with the deteriorating quality of water as well as possible solutions to these issues in Malaysia as opined by Chappell *et al.* (2017). Groundwater always relates to water hardness problem (Sepehr *et al.*, 2013). Water hardness is classified as temporary hardness and permanent hardness. Temporary hardness is often caused by the presence of dissolved bicarbonate minerals in the water, such as calcium

bicarbonate and magnesium bicarbonate. Temporary hardness can be reduced by boiling the water or by adding lime (Calcium hydroxide), by boiling water the soluble salts of Mg (HCO<sub>3</sub>)<sub>2</sub> is converted to Mg (OH)<sub>2</sub> which is insoluble and hence gets precipitated and is removed. However permanent hardness of water cannot be treated by boiling the water. Permanent hardness is caused by dissolved sulphate, chloride and other ions like calcium sulphate or magnesium sulphate (Ochieng *et al.*, 2012).

The hardness of drinking water is determined mainly by its content of calcium (Ca<sup>2+</sup>) and Magnesium (Mg<sup>2+</sup>). Consequently, high levels of Ca<sup>2+</sup> and Mg<sup>2+</sup> also increase total dissolved solids (TDS) level in groundwater (Gomelya *et al.*,2016). Although Ca<sup>2+</sup> and Mg<sup>2+</sup> content in groundwater do not have any serious impact on health problem, it is still responsible for scaling the household appliances and reducing the cleaning performance of the detergents and soaps (Mahasti *et al.*, 2017). Furthermore, high magnesium concentrations in drinking water may induce a bitter taste (*Sepehr et al.*, 2013). The presence of permanent hardness in the groundwater had triggered researchers to find the best treatment method.

Recently there are a few methods for hard water treatment such as ion exchange, adsorption, filtration and precipitation using lime soda. There are advantages and disadvantages to all these methods. All this methods involved with complicated procedure and also generated of toxic sludge (Grassi *et al.*, 2012). Among the water treatment, adsorption process is one of the actions. Adsorption offers the advantages of good efficiency and promising good metal binding (Aji *et al.*, 2015).

Therefore, there is a need to develop an efficient, rapid, and environment-friendly method to remove Ca<sup>2+</sup> and Mg<sup>2+</sup>. Ions from groundwater which is suitable to be applied in any condition. According to Logsdon *et al.* (2012), environmentally friendly media that easy to handle and the media's capability to be regenerated are the main factors that should be focused on developing the water softening method.

### 1.2 Problem Statement

In Malaysia, the Ministry of Health (MOH) had developed a standard limit for water supply quality. Thus, any treatment methods performed on water sources, whether it is surface water or groundwater, must be based on the standard limit of water quality as set by MOH to ensure that the treated water is safe to be consumed. Nowadays,

groundwater is one of the choices for alternative drinking water sources in Malaysia (Shahabudin, 2012). Groundwater is rich in mineral and other natural organic matter (NOM), which must be treated to comply with MOH standards before consuming as drinking water or clean water supply.

According to Kura (2018), Malaysia's groundwater sources' main problems were hard water and turbidity. Turbidity is the cloudiness of water caused by various particles and is another critical parameter in drinking water analysis. It is also related to diseases' content - causing organisms in water, which may come from soil runoff (Rahmanian *et al.*, 2015). Hard water is formed when water percolates through deposits of limestone, or gypsum, primarily made up of calcium and magnesium carbonates, bicarbonates, and sulfates. Dissolved Ca<sup>2+</sup> and Mg<sup>2+</sup> are mainly responsible for most scaling in pipes and water heaters and cause numerous laundry, kitchen, and bath problems. Potential health problems associated with hard water were kidney stones, dermatitis, reproductive health issues, and pancreatic cancer (Kadir *et al.*, 2017). Reducing Ca<sup>2+</sup> and Mg<sup>2+</sup> in groundwater will overcome the issue of hardwater and groundwater can be used as alternative sources of clean water supply.

There have been many methods for hard water and turbidity treatment such as ion exchange (Gomelya, 2016), adsorption (Ma, 2012), filtration using membrane (Ferreira *et al.*, 2017), and precipitation using lime-soda (Evuti *et al.*, 2012). However, Grassi et al. (2012) showed that all of these methods were high in maintenance costs, creates secondary pollution with the generation of toxic sludge, and involved complicated procedure in the treatment. Therefore, research for an alternative treatment method is needed since there is an increase in groundwater demand.

Adsorption is a standard method for hard water treatment (Mahasti et al., 2017). Due to the high abundance of natural adsorbent, the method based on adsorption reaction is one of the most investigated and adopted. Furthermore, the adsorption process is more advantageous in terms of economy, design, working flexibility, effectiveness, efficiency, and high quality purified product (Kadir *et al.*,2017). Activated carbon, biological material, bone char, and composite materials are used as adsorbents precursors for water treatment and purification (Jiuhui,2008). Results from previous studies revealed that activated carbon proved to have magnificent removal capabilities for most NOM and metal in water (Ince, 2017; Deliyanni *et al.*, 2015; Rahman *et al.*,2014). In recent years, there has been a growing interest in the

production of activated carbons from agricultural by-products and residual wastes (Ahmad, 2015).

A wide variety of agricultural waste materials have been used for the preparation of activated carbon. Some studies have shown several cases whereby activated carbons prepared from agricultural wastes performed better than the commercially available activated carbons (Omookoro *et al.*, 2018). In Malaysia, rice husk was categorized as an abundant agricultural waste. In the meantime, there was an issue in managing rice husk due to an increase in rice production in Malaysia. Muda Agricultural Development Authority (MADA) reported that production of paddy in 2016 was 1,072,600 metric tonnes and in 2020 was 1,2257,400 metric tonnes (Ahmad *et al.*,2017). Transformation of rice husks into activated carbon can add to considerable economic value to a product which considered waste, and it will reduce the cost of waste disposal, and most importantly provides a potentially inexpensive alternative to the existing commercial activated carbons for numerous applications, including water treatment (Menya *et al.*, 2017). Previous studies also proved that rice husk activated carbon (RHAC) from agriculture waste present a comparable adsorptive characteristic (Hariprasad *et al.*, 2016: Masoud *et al.*, 2012; Akhtar *et al.*, 2006).

The process of manufacturing activated carbon was through chemical and physical activation (Mohamed, 2011). Chemical activation was proved to have more ability in developing the porosity of the carbon by impregnation process using selected chemical. As the application of activated carbon for groundwater treatment, nitric acid is more environmentally friendly and more favorable to be used in the impregnation process (Tran *et al.*,2018). Activated carbon texture characteristics and surface properties depend on the precursor and the method used in the preparation.

Tay *et al.*,(2016) had found that contact time, pH, temperature and metal concentration are the factors that will influence the adsorption capability of activated carbon. Therefore, this study has provided an optimum experimental design and the effect of batch adsorption parameter on the metal removal Ca<sup>2+</sup> and Mg<sup>2+</sup> from synthetic groundwater and actual groundwater. Determination of mechanism adsorption for Ca<sup>2+</sup> and Mg<sup>2+</sup> removal from the groundwater can be achieved for practical design and fundamental understanding of the adsorption systems (Somasundaram *et al.*, 2013).

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