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Removal of Iron in Groundwater Using Marble Column Filter

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Abstract: Water shortage during dry season has become a big issue affecting the daily activities of the public as well as the world's attention. The potential of groundwater as a secondary water source can ease the situation of water shortage since the current water supply comes from surface water. However, groundwater pollution is inevitable because of human induced and natural occurrence. Therefore, this project studies to overcome excessive iron from the groundwater using marble in pebble size based on previous studies of calcium carbonate and sand size is in this study. Both sizes are compared to investigate the highest efficiency in removing iron without chemical use. The filtration is run with 4 different flow rates which are 0.017 ls^{-1}, 0.011 ls^{-1}, 0.008 ls^{-1} and 0.007 ls^{-1}. The highest removal percentage of iron, 93.83% is for the sand size marble column filter when the flow rate is 0.007 ls^{-1}. The marble is possible to be a cheap and effective filter media for iron removal in development of groundwater pre-treatment.

Keywords: Groundwater, iron removal, marble filter, marble media size, flowrate

1. Introduction

Groundwater is a supplementary source of freshwater in Malaysia which is cleaner than surface water, although it only consists of less than 10% of the public water supply source in the nation [1]. Being as the alternative source of water, the importance of groundwater is getting more significant as the available source of surface water is insufficient to supply the demand of public water usage. Prolonged dry seasons, unmatched water supply, growing population with increasing demand of water and lack of river basin management had become factors that causes water crisis in Malaysia [2]. Instead of depending on single source of surface water, the alternative source which is groundwater could help to solve the crisis of water shortage, yet the research and investigation on groundwater source need to be strengthened.

The groundwater is pumped from an aquifer under the ground and it is generally less contaminated compared to surface water yet contaminants such as heavy metals, nitrates, volatile organic compounds and pathogens had threatened the quality of groundwater. Iron is the heavy metal chosen to be treated as in the collected sample groundwater because it is a dominant heavy metal that had exceed threshold limit for potable water standards set by United States Environmental Protection Agency (EPA). This project will study about removal of iron by adsorption on marble and the efficient particle size of marble for iron removal.

Groundwater is an important source of freshwater which requires less treatment than surface water. Although groundwater is less contaminated, it is still susceptible to contaminants and it requires suitable water treatment to remove contaminants such as heavy metals, aromatic hydrocarbons, metals, volatile organic carbons, nitrates,

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microorganisms etc. in the water. Many technologies are established to treat groundwater and the cost for water treatment is biggest concern for the design and implementation of a groundwater treatment system.

In conventional water treatment plant, a high capital cost is spent on reverse osmosis (RO) filtration system treatment to decrease dissolved minerals besides removing taste, color and odor-producing organic compounds in the water. The water treated by RO filtration system is suitable for household use as RO membrane can remove virtually all microorganism in water [3]. Besides RO filtration system, activated carbon (AC) could be a potential material used in water treatment system to remove contaminants just like the RO membranes, yet the capital cost is estimated to be much lowered [4].

Heavy metal contamination in water source has many negative impacts on human health. Heavy metal is a term assigned to metals and metalloids that having high atomic density five times greater than water molecules, or at density higher than 4000 kgm^{-3}[5]. Heavy metals such as iron, manganese, lead, cadmium, arsenic and mercury are commonly found in groundwater. These heavy metals came from natural dissolution of minerals and human activities. Although heavy metals such as iron, manganese, selenium and chromium are essential micronutrients in human body, the daily intake of these elements are recommended under certain limits in concern of keeping a healthy life.

Iron is a key micronutrient for a good metabolism as it is an essential component to build proteins and enzymes in human body. U.S. Food and Nutrition Board have established a guideline on upper limit of 45 mg iron intake per day is a safe limit to most group of people but somehow the iron in water below 3 ppm will not bring health risk to the consumers as claimed by U.S. Food and Nutrition Board [6].

Iron is one of the most common elements on Earth crust and accounts for over 5 percent of the Earth's crust by weight. When an aquifer contacts with rock rich in iron minerals, the solids dissolve and will release iron into the groundwater. At concentration of 0.3 ppm is the threshold limit of iron in treated water following international standard of US EPA. Iron exists in dissolved state or ferrous ion under low oxygen level, particularly in deep aquifers or at low pH. The ions exist as ferric state when oxygen level is greater than 1 – 2 ppm or when the pH condition is acidic. Most of the treatment plant applied oxidation treatment method on soluble iron by adding oxidizing agent like potassium permanganate (IV) $KMnO_4$ into raw water and iron will precipitate for physical filtration.

Physical adsorption is a common method adapted to remove metal contaminants in raw water. The common media for physical adsorption includes activated carbon, sand, rice hulls and some other rock adsorbents such as limestone.

Limestone, which carries similar chemical properties as marble is widely used as material for water treatment throughout the world. It is used in acid mine drainage (AMD) treatment to increase pH of AMD, also used to neutralize acidity in industrial wastewater [7]. There are few physicochemical factors which control the effectiveness of water treatment using limestone such as co-precipitation of metal ions or adsorption, redox processes, pH fluctuation and dilution of groundwater by mixing with limestone [8]. In this study, the major factor in controlling the efficiency of iron removal is the adsorption of iron on marble. Different sizes of marble are used as filter media in a column filter which could determine the efficient of iron removal using different sizes marble chips.

This research project is a study on removal of soluble iron from groundwater using marble as a media by physical filtration. High purity marble and limestone which mainly composed of calcite $CaCO_3$ (67-90% CaO) has a potential to be a cheap and effective material to treat heavy metals in water as shown from previous studies [9].

2. Methodology

2.1 Groundwater Sampling

Sampling of groundwater is important to obtain a representative sample of groundwater with contamination concentration. Therefore, sampling method of 'EPA standard operating procedure for the well-volume method for collecting a groundwater sample from monitoring wells for site characterization' is followed in this step. Groundwater is sampled in USM from groundwater well located at School of Civil Engineering.

The groundwater is pumped out from aquifer and allowed it to contact with open air for oxidation of iron (II) under aerobic condition where in this sample ferrous sulfide FeS is formed. The FeS solid will be treated by using a column filter which applying physical filtration of metals on marbles.

The groundwater is sampled by using a groundwater sampler with probe. Before the groundwater is sampled, the oxidized groundwater is discharged from the well using a water pump for a duration of 3 hours with flowing rate of 2.5 ls^{-1} as following the EPA standard operating procedure for the well-volume method for collecting a groundwater sample from monitoring wells for site characterization [10].

The sample is quickly preserved with 3% of 3.0 M nitric acid, HNO_3 to avoid oxidation of sample. Each sample tube consists of 48.5 ml of fresh groundwater and 1.5 ml of 3.0 M HNO_3. 50 ml per sample was taken in total. These samples were kept in ice coolant box before transferring it into refrigerator at 4^oC for keeping the microorganism alive.

Besides contaminant concentration in groundwater, some other properties of groundwater are also good indicator for quality of groundwater. The fresh groundwater sample is also tested for temperature, total dissolved solid, salinity, dissolved oxygen, pH, nitrate concentration and conductivity using YSI Professional Plus Handheld Multiparameter. The readings of multiparameter are taken for 5 times to get average readings.

2.2 Filter Media Preparation

Crushing is a step to reduce size of marble into smaller sizes. The marble sample consists of cobble, very coarse gravel, coarse gravel, medium gravel and fine gravel as according to Wentworth grain size classification [11]. The filter media required are in Wentworth scale grain of very coarse grain (1 – 2 mm), coarse grain (0.6 – 1 mm), coarse-medium grain (0.4 – 0.6 mm) and medium grain (0.3 – 0.4 mm) for sand size. Meanwhile, for pebble grain size consisted of medium (10 – 14 mm), medium-fine (6 – 10 mm), fine (4 – 6 mm) and very fine grain (2 – 4 mm). Size reduction is required to obtain marble in size required for the filter media.

Cone crusher is used to break the marble into smaller particles less than 4 mm. The cone crusher is cleaned by air gun to remove dust on surfaces of the crusher before starting to crush marble. The 'set' of crushers is adjusted until desired size of products are obtained. The crushed marbles are collected before proceeding to screening.

Screening is the step to obtain marbles in desired sizes. The crushed marbles were screened using Gilson Screen Shaker according to ISO 565/3310 screen trays with aperture sizes of 14.00 mm, 10.00 mm, 6.7 mm, 4.25 mm, 2.12 mm, 1.15 mm, 0.825 mm, 0.600 mm, 0.425 mm and 0.300 mm. The trays were stacked in the machine with finest sieve followed by increasing coarser sieves with the coarsest on top. The locking screws were adjusted to lock the trays while screening. Screening was stopped after the particles were effectively screened.

2.3 Column filter installation

In this study, marble taken from Zantat Sdn. Bhd. in Simpang Pulai, Perak was used as filter media in the column water filter. There are two sets of marble column filter which consists of a set of pebble size marbles as filter media and another set is sand size marbles as filter media. The column filter has 4 columns in total, each column had an outlet valve at the lower and the input groundwater is entered from the top as shown in Fig. 1. The container of each column is made of composite glass with width × length of 34.5 cm × 33.5 cm. The outlet valve at lower part of each column with diameter of 1.5 cm was used to remove treated water during filtration process. This outlet valve is adjustable to control the flow rate.

Fig. 1 - Side view of marble column filter which arranged in increasing grain size from bottom to top column

The water sample will be pumped from ground level to water tank at highest part of the marble column filter. The water sample will exit the water tank via an outlet at lower part of the water tank under controlled rate. The water sample is flowed from top column to lowest column with free flow rate. The water sample exit from the bottom column is collected for water quality analysis.

2.4 Marble Placement and Cleaning

The screened marbles of each size range are placed into the containers flatly until the height, h of 9 cm from the bottom of container. The outlet of the column filter is covered by geotextile to prevent marble in sand size losses during filtration. There are 4 columns in the pebble size marble column filter where the larger size is sorted on the top and the smaller size at the bottom. Start from the top with size 10 – 14 mm and followed by 6 – 10 mm, 4 – 6 mm, and 2 – 4 mm.

Similarly, the set of sand size column filter is also consisted of 4 columns and sorted starting from the top with size 1 – 2 mm, 0.6 – 1 mm, 0.4 – 0.6 mm and lastly 0.3 – 0.4 mm. For flushing process of filter media, pipe water is flowed for a 15 minutes per column filter to flush through the marbles in the container which could clean the marbles' surfaces.

2.5 Filtration

The groundwater is filtered from top column to bottom column in sequence of decreasing grain size marbles. There are 4 flow rates used to determine optimum flow rate for contaminant removal using the marble column filter. The flow rates selected are 0.007, 0.008, 0.011, and 0.017 ls^{-1}, which are 60, 90, 120 and 150 s for per litre of groundwater. The outlet valve of each column is adjusted to control the output flow rate. The input and output water samples of the marble column filter are collected for ICP-OES analysis and also analyzed using handheld multiparameter.

The treated groundwater sample is collected at the outlet valve of column. A set of samples consists of input and output groundwater sample. For each marble column filter, there are 4 different flow rates used for the filtration. Therefore, there are 8 sets of samples collected in total.

These samples are preserved with 3% of HNO_3 which is in ratio of 1.5 ml HNO_3 to 48.5 ml of groundwater in 50 ml sampler bottle. The sample is then filtered to reduce the turbidity of water sample. The fresh sample is kept in refrigerator for storage before conducting ICP-OES test. Then, ICP-OES test is carried out to check iron (Fe) as the main element in this research and other side elements such as silver (Ag), aluminum (Al), boron (B), barium (Ba), bismuth (Bi), calcium (Ca), cadmium (Cd), cobalt (Co), chromium (Cr), copper (Cu), gallium (Ga), indium (In), potassium (K), lithium (Li), magnesium (Mg), manganese (Mn), sodium (Na), nickel (Ni), lead (Pb), strontium (Sr), titanium (Ti), and zinc (Zn) in the water sample. A multi-element standard solution is used to determine the concentration of iron elements in ICP-OES test.

3. Results and Discussion

Based on the ICP-OES test result, the most abundant count of element is magnesium (Mg), sodium (Na), potassium (K) and calcium (Ca) yet these elements are not heavy metals. The most abundant heavy metal in the groundwater sample is iron (Fe) which has mean concentration of 2.267 ppm, the concentration of other heavy metals is low in compare to the concentration of iron as can be seen in Fig. 2. Therefore, the iron in groundwater is worth to be studied for its removal.

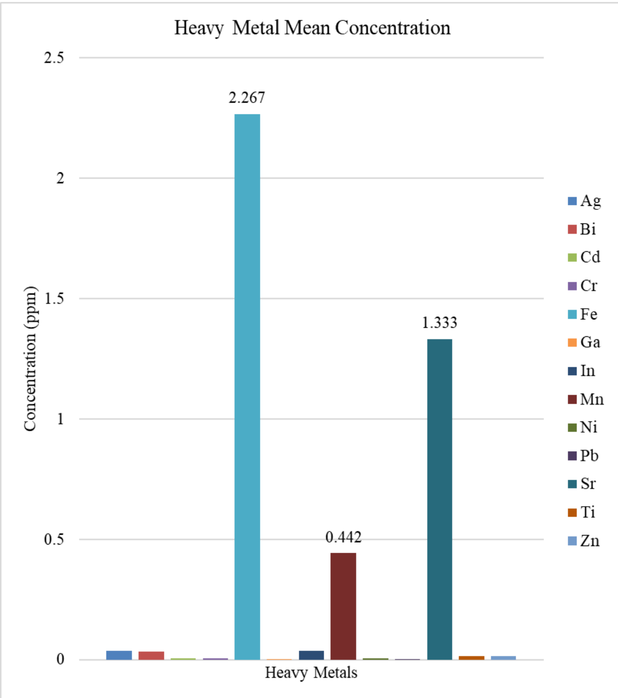

Fig. 2 - The graph of heavy metal mean concentration in the groundwater sample

3.1 Sand size marble filtration

Sand size marble column filter is used to treat the groundwater by using four different flow rates: 0.017 ls^{-1}, 0.011 ls^{-1}, 0.008 ls^{-1} and 0.007 ls^{-1}. The filter media is backwashed after it ran for a sample. The heavy metal with highest removal percentage is iron, Fe where among these 4 different flow rates, 99.06% to 99.35% of Fe is removed from the groundwater as shown in Fig. 3. The flow rate with highest iron removal is 0.007 ls^{-1}. This could be due to longer retention time of groundwater in the filter had given higher removal of iron oxide on the marble surfaces.

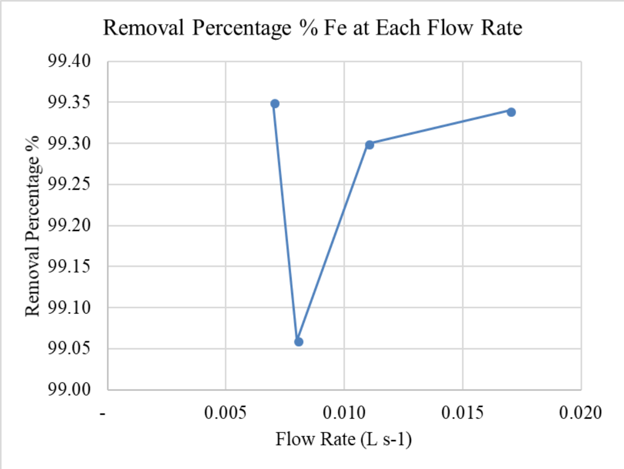

Fig. 3 - Removal percentage of iron from groundwater at each flow rate using sand size marble column filter

The sodium, Na in the groundwater is also removed for more than 95.15%, which means the salinity of the groundwater will decrease after filtration by the sand size marble column filter. The salinity measured using YSI handheld parameter had shown a decreased salinity from 5.15 ppt to 0.13 ppt and increased dissolved oxygen (DO) level for the output water sample from 0.07 ppm to 3.12 ppm. The lower DO content in water, the higher microbial contaminants content in the water.

The iron concentration in the groundwater before treatment is about 2 ppm and after the filtration, the concentration of iron decreased to 0.1 – 0.2 ppm. From the results of as shown in Fig. 3, the difference of removal percentage among 4 different flow rates is not significant and it has no general trend. The flow rate of 0.007 ls^{-1} and 0.017 ls^{-1} shows a difference in removal percentage of 0.01% only. It could be related to the difference of room temperature during the experiment.

As a summary, the removal of iron using sand size marble column filter has removal percentage higher than 99.06% and highest at 99.35%. The change in flow rate does not show a trend for better removal percentage of iron. The lowest removal percentage of iron is at flow rate of 0.008 ls^{-1}. The highest removal percentage of iron is at flow rate of 0.007 ls^{-1}.

3.2 Pebble size marble filtration

The pebble size marble column filter is also to treat the groundwater by using 4 different flow rates: 0.017 ls^{-1}, 0.011 ls^{-1}, 0.008 ls^{-1} and 0.007 ls^{-1}. The iron, Fe is removed from the water sample at highest removal percentage among the heavy metals.

The pebbles had obvious dark stains left on the marble surface after running the experiment for half an hour as can be seen in Fig. 4. The stain turns to rusty dark brown color after leaving in the air overnight. It shows the contaminants are retained on the marble surface, yet the composition of these stains is not identified.

Fig. 4 - Pebbles with black stains on the marble surface after filtration

The concentration of iron reduced to 0.15 – 0.20 ppm after filtrated by pebble size marble column filter where its initial concentration of iron is 2.4 – 2.6 ppm. Among the 4 different flow rates, 0.007 ls^{-1} shows highest percentage of iron removed from the water sample where 93.83 % of iron is removed as shown in Fig. 5. The lowest removal percentage for iron is 92.18 % at flow rate of 0.008 ls^{-1}. The difference of removal percentage among 4 different flow rates is not significant and it has no general trend.

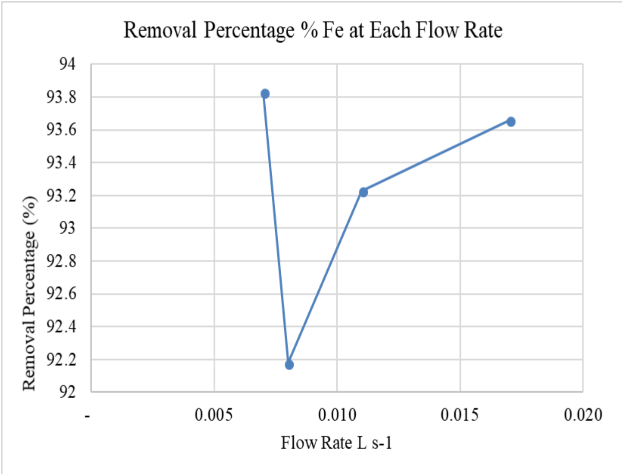

Fig. 5 - Removal percentage of iron from groundwater at different flow rate using pebble size marble column filter

As a summary, the removal of iron using pebble size marble column filter has removal percentage higher than 92.18% and highest at 93.83%. The change in flow rate does not show a trend for more effective removal percentage of iron. The lowest removal percentage of iron is at flow rate of 0.008 ls^{-1}. The highest removal percentage of iron is at flow rate of 0.007 ls^{-1}.

3.3 Comparison of Sand Size and Pebble Size Marble Column Filter Removal Percentage

The sand size marble column filter has shown better removal of iron from the groundwater at four of the different flowrates. After above 99% of iron is removed, there are only 0.01 ppm to 0.02 ppm of iron is left in the groundwater. It is already fulfilling the threshold limit of iron content in drinking water standard as set by EPA.

Based on the experimental results in Fig. 6, flow rate of 0.007 ls^{-1} shows the highest percentage of iron removed during filtration. It shows that the higher the retention time, the more contaminants are removed. Among the four-different flow rate, the optimum flow rate for iron removal percentage is 0.007 ls^{-1}. Although the longer retention time will have better removal of contaminant, it is difficult to have flow rate lower than 0.007 ls^{-1} due to restriction of the outlet valve to be adjusted.

The results had reported 0.007 ls^{-1} as the most effective flow rate for iron removal in the groundwater. As seen from the results, pebble size marble is less effective in contaminant removal compare to sand size marble. The highest removal percentage of iron is for the sand size marble column filter when the flow rate is 0.007 Ls-1. The initial concentration of iron in the groundwater is 1.999 ppm but end up with 0.013 ppm of iron left in the water after filtrated. The marble is possible to be a cheap and effective filter media for iron removal in development of groundwater pre-treatment process in USM as the results show high removal for iron so as other element in the groundwater.

4. Summary

Based on the analysis, the characterization of groundwater using ICP-OES test has shown the highest concentration of heavy metal is iron with the mean concentration of 2.267 mg/L. The other minor heavy metals are bismuth, cadmium, chromium, indium, manganese, nickel, lead, strontium and titanium.

The groundwater sampled from the monitoring well is saline water with salinity of 4.76% of salinity as measured with YSI handheld multiparameter. The pH of the groundwater is 7.19, which is slightly alkaline. The dissolved oxygen (DO) level in the groundwater is 0.11 mg/L which is very low, even at 10 mg/L the aquatic life is put under stress.

The marble has no porous surface. The removal of metals happens on the marble surfaces where contaminants are retained there. Therefore, the greater the surface area of marble is expected to have higher removal percentage of iron from the groundwater. The smaller size marble, which has sand grain size is possible to be better removal of metals compare to pebble size marble.

From the sand size marble column filter, the flowrate of 0.007 ls^{-1} removes the greatest amount of iron from the groundwater. The initial concentration of iron in the filter input is 1.999 ppm and it decrease to 0.013 ppm after filtered by sand size marble column filter, 99.35% of iron is removed. Using the pebble size marble column filter, the flowrate of 0.007 ls^{-1} has removed the most abundant iron from the water sample. The removal percentage is 93.83% with initial concentration of 2.528 ppm decreased to 0.156 ppm after treated.

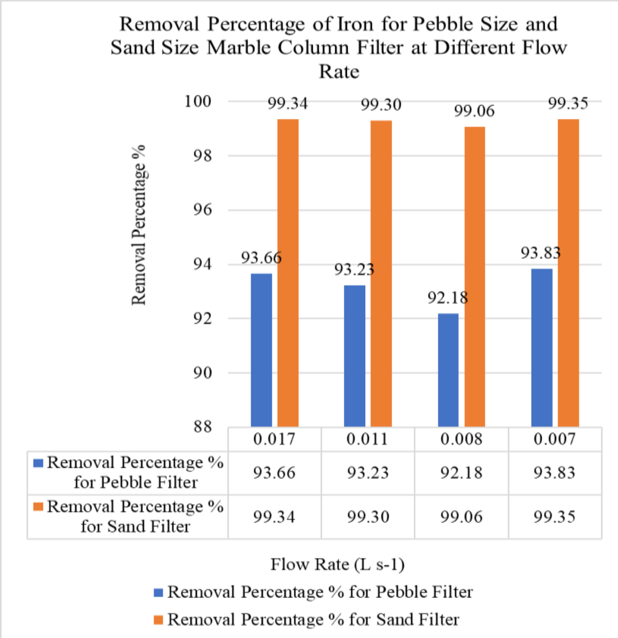

Fig. 6 - Removal percentage of iron for pebble size and sand size marble column filter at different flow rate

The sand size marble column filter has higher removal percentage of contaminants from the groundwater compare to the pebble size marble column filter. The sand size marble column filter has iron removal percentage above 99% disregard the flow rate, whereas pebble size marble column filter has iron removal percentage above 95% only. The sand size marble column filter is more effective as a physicochemical filter media. Both sand size marble column filter and pebble size marble column filter has most effective iron removal percentage with the flow rate of 0.007 ls^{-1}. The longer retention time of the groundwater in the filter, the increase amount of iron oxide and other contaminants to be removed.

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