Surface Water Treatment using Pomelo's Peel (Citrus Grandis) and Biosand Filter as Iron (Fe) Adsorption in Kangkar Senangar's River

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Abstract: ***Water is our most precious natural resource. Based on this study which was conducted in Kangkar Senangar, Johor, the majority of rural communities still drink superficial water that does not meet the required standard of quality, which can cause serious health problems. From the interview with the Village Head, it is also noted that they also do not have enough source of water supply. They only use groundwater as their water source to complete daily life tasks. The ability of Pomelo Peel is tested as natural adsorbent, to remove Iron (Fe) from aqueous solution by adsorption was investigated. Characterization of adsorbent was done by SEM and FTIR analyses to observe the surface morphology and functional groups available on the adsorbent. Adsorption was most efficient when 0.20g of adsorbent was used with 50ml of water sample that was taken from Kangkar Senagar's river. 0.20g was the optimum weight of pamelo peer powder with contact time of 20 minutes to change the quality of water sample. Based on the results, the higher the adsorbent dosages, the higher the adsorption removal of Fe. The highest adsorption removal of Fe was 80.84%. After the adsorption, the water sample was treated using Biosand Filtration (BSF). The function of BSF was to reduce the turbidity, change the colour of water sample, increase the pH and dissolve oxygen. The BSF filtered all the suspended matters in the water samples after the adsorption and produced clearer water.***

Index Terms: ***Pomelo peel, Adsorption, Iron (Fe), Biosand Filter, Water quality***

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I. INTRODUCTION

Clean and safe-drinking water is scarce. According to Michael, water supply has been recognized as a primary logistical challenge since the beginning of civilization and balancing water demand and supply has been a major concern of all human society of all times (Michael Chukwuma Obeta, 2015). Rural area has always been associated with the scarcity of clean water due to lack of a piping system provided by the government to distribute the treated water. Some of the areas do not have an adequate clean water supply from pump house because of the geographical area. Due to lack of treated water, the residents in the rural area take another initiative to use river or groundwater as their daily water supply (Aliamat, 2012). However, the quality of water from river and groundwater still does not meet Drinking Water Quality Standard set by Ministry of Health Malaysia.

The scope of this study was to cover the water necessity for the villagers in Kangkar Senangar. The location of the water sample was the surface water which a river located in front of the Village head's house, 30 kilometres from UTHM Pagoh. Water is a limited natural resource and a public good fundamental for life and health. The geographical location is the biggest contributing factor to this situation faced by the residents of Kangkar Senangar. The residents' houses are isolated and it causes the pipeline not being able to reach the houses. The government has spent a million Ringgit Malaysia to overcome this crisis for them to get treated water supply.

Based on the studies and investigations, the Kangkar Senangar's river was Contaminated by an accumulation of heavy metal Iron (Fe). The accumulation of Fe in the river is dangerous because it could give a health effect to the villagers. There are many methods for water treatment which include chemical and surface chemistry processes such as precipitation, biosorption, membrane process, ionic exchange, floatation and others (Saikaew, 2009). However, those techniques have their own inherent limitations such as less efficiency, costly disposal, sensitive operating condition and production of secondary sludge. So, one of the alternative methods to remove Fe in the water sample is by adsorption method using low-cost adsorbent which is agriculture or biomass waste that is also environmentally friendly (Chanmalee, 2016).

The Pomelo peel was selected as the adsorbent because of its characteristic and there was a pomelo plantation in Kangkar Senangar. Therefore, the objectives of this study were to characterize the surface morphology and chemical composition of Pomelo peels as adsorbent before and after adsorption process, determine optimize condition of adsorbent dosage by Pomelo peel as adsorbent and determine the maximum percentage of removal for Iron (Fe) in Kangkar Senangar's river under optimized conditions using adsorption and filtration methods.

II. LITERATURE REVIEW

Metal Contamination in a surface water

Through observations and studies, there were many polluted rivers that have a low environmental quality due to a large amount of untreated domestic and industrial waste that are released into the water bodies which indirectly increases the level of metal in river water (Khadse, Patni, Kelkar, & Devotta, 2008). Heavy metal can be defined as any metallic chemical elements that has a high density and poisonous at low concentration (Lenntech, 2017). Iron occurs naturally in soil, sediments and ground water and can be found in many types of rocks. Iron can be present in water in two forms; the soluble ferrous iron or the insoluble ferric iron. The ingestion of large quantities of iron can damage blood vessels, cause bloody vomitus/stool, and damage the liver and kidneys, and even cause death (Saskatchewan Health, 2018). Based on the studies and observations, agriculture was the source of soil and water contamination. The soil was contaminated by the accumulation of heavy metal that were produced by the land application of fertilizers and pesticides (Wuana & Okieimen, 2011). Iron is one of the most used metal in fertilizers and pesticides. When it is rainfall, the runoff of the soil will occur and large quantities of iron will enter the river especially in acidic conditions (Kakde, 2016).

Adsorption

Adsorption is a phase transfer process that is widely used in practice to remove substances from fluid phases (gases or liquids). It can also be observed as a natural process in different environmental compartments. The most general definition describes adsorption as an enrichment of chemical species from a fluid phase on the surface of a liquid or a solid. In water treatment, adsorption has been proved as an efficient removal process for a multiplicity of solutes (Worch, 2012). Molecules or ions are removed from the aqueous solution by adsorption onto solid surfaces.

The adsorbent dosage was one of the factors affecting the adsorption. This is because through an amount of adsorbent used in experiment, the capacity of adsorbent for a given initial concentration can be determined. The biomass provides binding sites for the adsorption of metal ions, and hence its concentration strongly affects the adsorption of metal ions from the solution. For a fixed metal initial concentration, increasing the adsorbent dose provides greater surface area and availability of more active sites, thus leading to the enhancement of metal ion uptake(Abdel-Ghani & El-Chaghaby, 2014). However, at a higher dosage, the ions adsorbed were higher due to the availability of more empty binding sites as compared to a lower dosage which had less binding sites to adsorb the same amount of metal ions in the adsorbate solution.

Pomelo Peels as Adsorbent

Pomelo (Citrus grandis) is the largest citrus fruit, the peel of which is a well-known agricultural waste. Disposal of pomelo peel after consumption is a serious environmental problem(Zhang G1, Sun Y1, Guo Y1, 2, Liu J1, Wu L1, 2018). Pomelo (Citrus grandis), a natural (non-hybrid) citrus fruit with a much thicker rind, is similar in appearance to a large grapefruit and widely cultivated in eastern and south-eastern China. Pomelo is native in Malaysia and has many benefits. The peel of the pomelo accounts for more than 50% of the total weights, tastes bitter, considered inedible, and thus is usually discarded. Pomelo peel, agricultural waste, was selected because of its high pectin content and the fact that it is an agricultural byproduct (Saikaew, 2009). Pectin has many functional group and this can be proved by FTIR analysis in result and discussion. Through these functional group, the PP can be used to bind the heavy metal which is Ferum in this experiment.

Biosand Filter

The biosand filter (BSF) is one example of a feasible technique to produce drinkable water in rural areas (Lindgren & Olivecrona, 2016). The biosand filter (BSF) is a simple household water treatment device which is an innovation on traditional slow sand water filters, specifically designed for intermittent or household use (Oxfam, 2008). The filter can be made easily because it is built using materials that are available everywhere in this world. The components of BSF are concrete or plastic container which contain selected prepared sand and gravel. When the water flows through the filter, the pathogen, iron, turbidity and manganese will be removed from drinking water by physical straining. A shallow layer of water sits atop the sand and the developing of biofilm (Schmutzdecke) will occur (CAWST, 2017).There are 4 process that occur in biofilm that can remove pathogen; mechanical trapping, predation, adsorption and dying.

III. METHODOLGY

Preparation of material

The adsorbent, pomelo (Citrus Grandis) was obtained from a fruit shop that was located in Pagoh, Johor. The water sample was taken from Kangkar Senangar's river. Iron Reagent was purchased at CST Arachem (M) Sdn Bhd, Kuala Lumpur. Instruments employed for the work include FTIR spectrophotometer (model 8400S), scanning electron microscopy (SEM) spectrophotometer, electric (platform shaker 20-880), DR6000, mottle P165 weighing balance, pH-meter-16, conical flask, HANNA Dissolved Oxygen meter, vacuum pump and sieves.

Preparation of adsorbents

The Pomelo peel was washed with deionized water which was prepared by the technique of reverse osmosis several times to remove heavy metals.

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The washed materials were cut into small pieces (1-2 centimetre; cm) then dried in a hot air oven at 60oC until they reached a constant weight, which was accomplished after 48 hours (hrs). During the final stage, the material was dried, ground and screened to a cut-off size of 150-212 micrometer (μm).

Preparation of adsorbate

The water samples were taken from Kangkar Senagar's river within a period of 4 weeks. The total of 4 samples were collected using grab sampling method. All of the samples were collected at different weeks in October 2018 and the samples were kept in 1500 mL plastic bottles, labelled with different codes and immediately stored at 4°C when transported back to the laboratory from sample collecting site.

Preparation of Biosand Filter

The types of material that were used in the biosand filtration include fine sand, separating gravel, drainage gravel and coconut coir. All the materials are available at Highway Laboratory, Faculty of Engineering Technology, University Tun Hussein Onn Malaysia, Pagoh, Johor. Below shows the figure of biosand filtration.

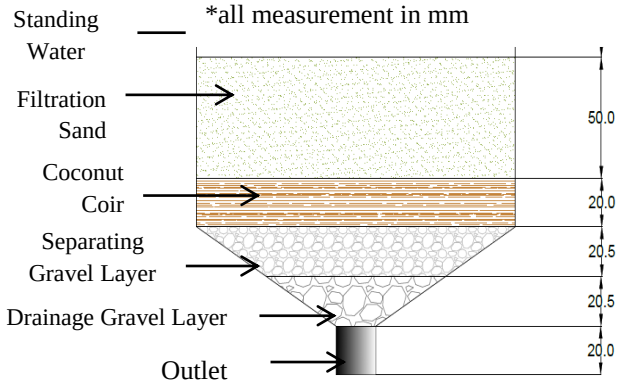

Fig. 1 The diagram of biosand filter

The components of a generic biosand filter were a plastic vessel filled with filter media and a gravel. The first modification made to the design in this study was the use of a plastic bucket with a height of 14 cm and a width of 16 cm and filter media at a height of 12 cm. The biosand filter with sand had six distinct sections. The first section was an inlet reservoir through which contaminated water is poured into the filter. A standing water level is the second section while the third section consists of fine sand with a particle size of ≤ 0.7 mm (0.03"), packed to a height of 5 cm. The fourth section consists of the coconut coir with a height of 2 cm. The fifth section consists of coarse sand with a particle size of 0.7 mm (0.03") - 6 mm (¼") as separating gravel layer and last section is gravel with a particle size of 6 mm (¼") - 12 mm (½") as drainage gravel layer.

Batch Adsorption Studies

Batch adsorption experiments were carried out in 250 mL flasks and the total volume of the reaction solution was kept at 50 mL. The flasks were shaken at 200 rpm for an equilibrium time of 20 min on a mechanical shaker. The effect of adsorbent dosage on the removal of Fe was studied with different adsorbent dosages (0.05, 0.06, 0.07, 0.08, 0.10 and 0.20 g) at a temperature of 27± 0.5 _C, optimum pH of 6.0 and shaken till equilibrium time. After the adsorption process, the water sample underwent a process of filtration by biosand filtration. Then, the concentration of Iron in the water that had been filtered was tested by DR6000 using Iron reagent. The percentage removal (R) of Fe was calculated using equation

$$\%R = \frac{C_o - C_e}{C_o} \times 100$$

Where Co and Ce are the initial concentration of Fe in water sample and final concentration of Fe after adsorption and filtration respectively (mg/L).

IV. RESULT & DISCUSSION

SEM Analysis

The characteristics in the microscopic appearance of the Pomelo Peels (PP) samples were observed using scanning electron microscopy (SEM). The morphology of the pomelo peels before the adsorption process is shown in Figure 2 (a) and (c). The surface of the pomelo peel's powder particle was rough, as shown in micrographs at low magnification through 20 μm and 10 μm. Under the magnification, a porous and honeycomb-like structure was clearly seen, which was beneficial for the adsorption of heavy metal. Such morphology was very attractive for adsorbent materials since the high surface area increased the adsorption sites on the pomelo peel's surface. This shows that the pores within the peels particles were highly heterogeneous. However, this was not the case after the adsorption. After the heavy metal adsorption, a significant change was observed in the structure of the peels. The surface morphology changed and became smoother. This is because the PP has bind the metal which was Ferum in the water sample. Based on the figure 2 (b) and (d), it shows that the empty spaces on the pomelo peel's surface after the adsorption decreased in comparison to before the adsorption.

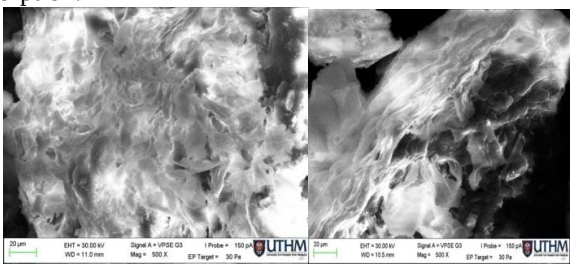

Fig. 2 SEM micrographs of PP: (a) The morphology of the pomelo peels before adsorption process under 20 μm , (b) The morphology of the pomelo peels after adsorption process under 20 μm

FTIR Analysis

The FTIR spectra of PP, Pomelo Peels is shown in the figure 3. The peaks were located at 3276, 2922, 1604, 1415, 1234 and 1016 cm-1 observed from figure 3 are characteristic of pomelo peels.

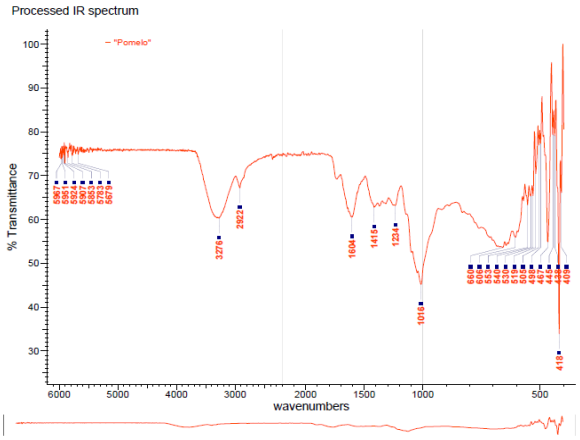

Fig. 3 FTIR spectra of a PP

Based on the result, it shows that the wide and intense peak located at 3276 cm-1 corresponded to the stretching vibrations of -OH group. The peak might contribute to the physically adsorbed water molecule or result from hydroxyl groups on the samples. The small peak located at 2922 cm-1 was the asymmetric and symmetric C-H stretching vibrations, and the weak peak located at 1415 cm-1 was the deformation mode of the C-H group. There were two observed peaks at 1604 cm-1 and 1016 cm-1 resulting from the stretching vibration of C=O and C=C, respectively, indicating the existence of aromatic groups. There was another wide peak that produced form PP which was located at 1234 cm-1 that could be described as C-O-C stretching vibration or C-C framework vibration. The peak that was located at 1415 cm-1 was resulted from the vibration of vinyl C-H and C-H plane bending vibration (Zhu et al., 2017).

Batch Adsorption Experiment

The effect of dose of adsorbent on the percentage removal of Fe ions was studied. The effects of pomelo peels powder dosages on the amount of Ferum was investigated by contacting 50 mL of surface water with different initial heavy metal concentrations. Each week produced different heavy metal concentrations because of rainfall. Rainfall was a major contributing factor in affecting the water quality. Ferum will affect water supply in industrial and consumer waste, or even from heavy rainfall, releasing metals into Kangkar Senangar's river. There were many palm oil plantations surrounding the river as there was a palm oil mill near the river. The effect of dosages was investigated by contacting 50 mL of water sample with different amounts of adsorbents (0.05,0.06,0.07,0.08,0.10 and 0.20 g) for a contact time of 20 min at a temperature of 27± 0.5 _C, a shaking speed of 200 rpm and optimum pH of 6.0. After equilibrium, the samples were allowed to settle for some time after which the supernatant solutions were collected and analyzed.

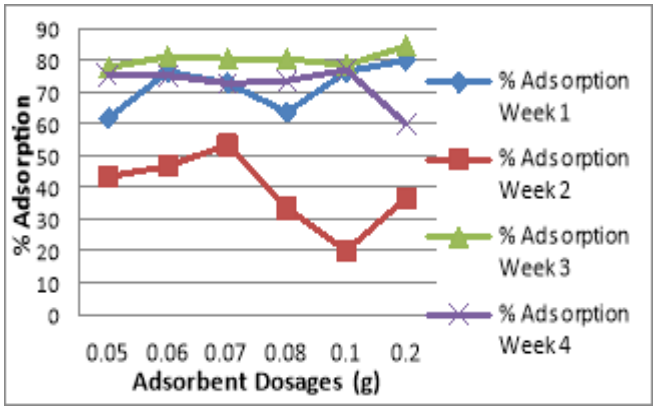

Fig. 4 The graph of effect of adsorbent dosage on the adsorption of Fe onto PP at room temperature; pH = 6.0; stirring; speed= 200 rpm and time =20 min

During week 1, the initial concentration of Fe was 0.55 mg/L. The adsorption increased as the amount of PP increased. The highest adsorption for week 1 was when 0.20 g of PP was added. The percentage adsorption was 80%. The lowest removal of Fe was when 0.05g of PP was added. The percentage adsorption was 61.82%. The weather condition in week 1 was rainy and windy. During week 2, the weather was sunny when the water sample was taken. The initial concentration during that week was only 0.30 mg/L which was the lowest compared to the other week. After the adsorption and the filtration processes, the adsorption of Fe decreased. The highest percentage adsorption was when 0.07 g amount of PP was added which was 53.33 % while the lowest was 20.00% when 0.10g PP was added.

During week 3, the initial concentration of Ferum was the highest compared to other weeks. This was affected by the rainy season during week 3. The initial concentration was 1.48 mg/L. The adsorption of Fe was the highest when the amount of 0.20g of adsorbent was added which was 84.46% after the adsorption and the filtration. The lowest adsorption was 77.70% that was produced from 0.05g of PP.

During week 4, the initial of concentration of Ferum was 1.17 mg/L. The highest percentage adsorption was when 0.10g of PP was added which was 76.92%. It was rainy during week 4. 59.83% was the lowest percentage removal of Fe when the largest amount of adsorbent of 0.20g was added. The result shows that the concentration level of the metal at each week was different based on the weathers condition. Based on the result analysis, the highest concentration of Ferum is during week 3 which was 1.48 mg/L while the lowest concentration was on week 2 which was 0.30 mg/L. Based on the graph, it shows that the PP and biosand filter can adsorb the Fe almost 85% of Fe in initial concentration. The increase in adsorption with adsorbent dosage can contribute to an increase in the adsorption surface and availability of more adsorption sites.

Turbidity

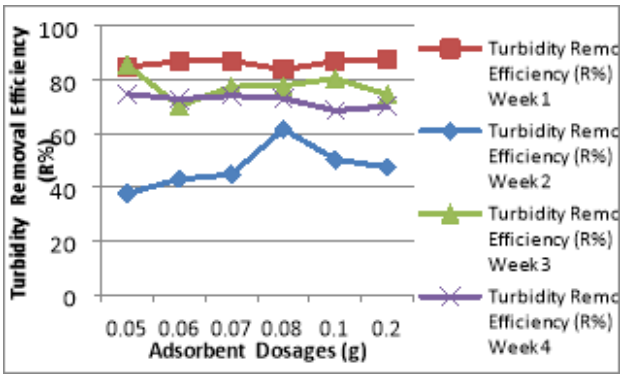

Fig. 5 The graph turbidity removal efficiency for 4 week

Based on the result, the highest turbidity removal efficiency is on week 1 which was 87.70% while the lowest turbidity removal was in week 2 which was 37.67% then followed by week 2 and week 3. During week 1, the initial turbidity was in the range of 75 NTU. This is due to the fact that the Kangkar Senangar's river was contaminated by the breaking down of the soil. The weather condition during that week was rainy. It was a heavy rainfall that caused the erosion to the soil. After the adsorption and the filtration, the turbidity decreased to 9.16 NTU from 75 NTU. This shows a great decline amount of turbidity. For week 1, as the PP dosages increased, the turbidity removal is increased. This shows that the PP had adsorbed the particle and contaminant that caused high turbidity.

After the adsorption process, the water sampling colour became yellow from brownish. This is because pomelo peel's powder is yellow when shake with water sample for 20 minutes. The water samples needed to be filtered because of the change in colors. After filtrating the sample using biosand filtration, the water sample became clear.

pH

Technically, pH is a measure of the activity of hydrogen ion (H+) and is reported as the reciprocal of the logarithm of the hydrogen ion activity (Oram, 2014). Below are the tables of initial pH and after the adsorption and the filtration.

Table. 1 The initial pH before adsorption and filtration

pH	Dosages					
	0.05	0.06	0.07	0.08	0.10	0.2
Week 1	6.17	6.11	6.14	6.13	6.18	6.09
Week 2	6.38	6.3-3	6.31	6.36	6.34	6.37
Week 3	6.52	6.56	6.54	6.58	6.55	6.55
Week 4	6.75	6.84	6.81	6.85	6.82	6.71

Table. 2 The value of pH after adsorption and filtration

pH	Dosages					
	0.05	0.06	0.07	0.08	0.10	0.2
Week 1	6.71	6.73	6.96	6.75	6.80	6.79
Week 2	6.78	6.78	6.72	6.70	6.68	6.64
Week 3	6.55	6.48	6.44	6.42	6.35	6.34
Week 4	7.15	7.10	7.13	7.14	7.02	7.05

Based on the table, s the initial pH values of Kangkar Senangar's river before the adsorption and the filtration was within the range of 6~6.5. For week 1 and 2, the initial pH values were below 6.5 which was acidic while for week 3 and 4, the initial pH values were above 6.5 which was a normal range of pH in surface water. According to Water Research Center, generally the water with a low pH value, which is below 6.5, could be acidic, soft and corrosive. This might be caused by the concentration of iron (Fe) in the river. Water with low level of pH could contain elevated level of toxic metals which can cause aesthetic problems such as metallic or sour taste (Oram, 2014). However, the value of pH had increased after the adsorption and the filtration. All four weeks showed chanfes in pH values. The pH value turned back to a normal range of pH in surface water except for the water sample in week 3. This might be caused by the high concentration of Fe during week 3.

Dissolved Oxygen

Dissolved Oxygen is the amount of gaseous oxygen that dissolves in the water. The oxygen enters the water by a direct absorption from the atmosphere, by a rapid movement or as a waste product of plant photosynthesis. The Kangkar Senangar's river was surrounded by a palm oil plantation. Below are the results of dissolved oxygen before and after the adsorption and the filtration.

Table. 3 The concentration of dissolved oxygen before adsorption and filtration

Dissolved Oxygen (mg/L)	Dosages					
	0.05	0.06	0.07	0.08	0.10	0.2
Week 1	8.34	8.31	8.32	8.33	8.34	8.33
Week 2	8.33	8.33	8.32	8.31	8.30	8.31
Week 3	7.53	8.23	8.21	8.09	8.17	8.11
Week 4	7.84	7.95	8.10	8.00	8.13	8.17

Table. 4 The concentration of dissolved oxygen after adsorption and filtration

Dissolved Oxygen (mg/L)	Dosages					
	0.05	0.06	0.07	0.08	0.10	0.2
Week 1	8.25	8.99	8.89	8.94	8.95	8.96
Week 2	7.95	7.77	7.47	7.48	7.22	7.32
Week 3	8.30	8.33	8.32	8.32	8.35	8.30
Week 4	8.31	8.28	8.29	8.23	8.26	8.23

Based on the results, the dissolved oxygen level before the adsorption and the filtration were within the range of 7.53 mg/L and 8.34 mg/L while for dissolved oxygen after the adsorption and the filtration, the values were within the range of 7.32 mg/L and 8.99 mg/L. This shows that the dissolved oxygen had increased after the adsorption and the filtration.

The factor that affected the dissolved oxygen was temperature. The temperature of the water sample after adsorption and filtration was much cooler than the initial temperature of the river. This shows that the oxygen dissolves more easily in cooler water than warmer water. Not only that, the volume of moving water also influenced the entrance of oxygen in the water sample. The water sample needed to be shake for 20 minutes in the adsorption process, which can conclude that as the water sample moving continuously, the volume of dissolved oxygen will be added into the water. Both result showed that the amount of dissolved oxygen was above 5.0 mg/L. According to Fundamental of Environmental Measurement,

the level of dissolved oxygen that drops below 5.0 mg/L can cause stress to aquatic life. The lower the oxygen level in the water that drops below 1-2 mg/L might result in a large fish kill (Measurements, 2016). The results show that the dissolved oxygen in the Kangkar Senangar's river was safe for the aquatic life to live.

V. CONCLUSION

In conclusion, the results obtained from this study revealed that Pomelo Peel (PP) is an effective low cost adsorbent for the removal of Iron (Fe) in the aqueous solutions. The adsorbent has a good adsorption capacity for Fe as the higher the dosages of PP, the higher the percentage of adsorption. The highest adsorption of Iron in this study was 84.46% which was almost 100%. This can be proved by SEM analysis which showed that empty spaces on the pomelo peel's surface after the adsorption decreased in comparison to before the adsorption. The surface morphology changed and became smoother. This was because the PP has bind the metal which was Ferum in the water sample. The binding of the metal can be proved by FTIR analysis as there is the pectin of PP that consists of a functional group.

However, biosand filtration also helps in the treatment process. As the water became yellow after the adsorption because of PP's colour, the biosand filtration make the water quality improved. The pH of the water increased and became a normal range of pH in surface water which was 6.5~8.5. The highest turbidity removal in this study was 87.70% during week 1 which shows a great high drop of turbidity level. Not only that, the value of dissolved oxygen increased after the adsorption and the filtration. The value of dissolved oxygen was above 5.0 mg/L which as safe for the aquatic life to live.

VI. RECOMMENDATIONS

There are several suggestions recommended to the next study for better result:

1. The adsorption studies can be tested by different factors affecting adsorption such as time, pH, temperature, Ferum concentration, organic compound, type of water sample.
2. Improvise the method of making adsorbent by making the pomelo peel as activation carbon.
3. Additional of adsorption isotherms to provide important models in the description of adsorption behavior.
4. "Swirl & Dump" the biosand filtration as the flow rate drop to an unacceptable level.
5. Reveal the reusability and efficiency of the adsorbent by doing more research and study about the pomelo peel's characteristic.

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