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Durian husk wastes as low-cost adsorbent for physical pollutants removal: groundwater supply

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ABSTRACT

Durian peel can be the main contributor to agricultural wastes and becomes an environmental problem when it is discarded into the landfill site or even burning it. The average of entire durian fruit weight is about 255,353 MT for all over the country. The loading rate of landfills is increasing, especially with the massive amount and size of the durian waste that requires a larger space to dispose of. Therefore, it is certainly important to utilize durian husk as an adsorbent to improve water quality, especially groundwater source. Groundwater commonly has a higher hardness level than surface water. The higher hardness in water becomes a major concern, especially in every cleaning task. This research aimed to investigate the potential of durian husk in reducing water hardness, electrical conductivity (EC), and total dissolved solids (TDS). Durian husk was treated with NaOH solution to improve adsorption efficiency. Laboratory analyses such as the ethylenediaminetetraacetic acid (EDTA) titration method were performed for the water hardness, total dissolved solids, and conductivity concentrations respectively to test the performance of the removal efficiency before and after the treatments. As the results, the removal of hardness concentrations by durian husk has significantly dropped with dosage and settling time. However, not for TDS and EC concentrations removal, which it went sudden increased for a higher dosage. The novelty of this study is that it is the first-ever experiment using the real on-site samples in the field as for initial concentrations, different from other previous studies by lab-scale the synthetic hard water and most of them using the highest concentration of hardness which up to 700 mg/L of CaCO₃ as to find out the removal efficiency for water softening in water treatment using durian husk, compare to this study using a concentration that is too low within 300 mg/L and below.

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1. Introduction

Malaysia generates a large quantity of durian fruit per year, especially for commercializing. The durian produces in Malaysia is estimated at 320,164 MT in the year of 2013 [1]. The average of entire durian fruit weight is about 255,353 MT for all over the country, and the loading rate of landfills is increasing, especially with the massive amount and size of the durian waste that requires a larger space to dispose of [2,3]. Durian (*Durio zibethinus*) is among the famous fruit grown and consumed locally and exported, especially in South East Asia countries [4]. The fruit is

in ovoid nearly round-shaped with an average size weighing between 2 kg and 4.5 kg depending on their species as there approximately 30 different species of durian. The outer part of the durian skin normally weighs more than half of the total fruit weight. It is in green color to yellowish-brown, very thick and semi-woody with sharply pointed pyramidal thorns the edible part of the fruit that can only 15–30% of the entire mass of the fruit that can be eaten [5]. Approximately 70 – 85% of the fruit is discarded as waste and would be an environmental problem if not disposed of in a proper manner [6]. Durian peel can be seen as the primary contributor for agricultural wastes with increasing volume, with approximately reached 480 000 metric tonnes of durian wastes produced every year [7]. It will account a larger area for disposal

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area, especially in landfill sites, and with limited spaces exist nowadays that needed to decompose the waste. Therefore, it is undoubtedly essential to reduce this agricultural waste by reusing and utilizing them as low-cost water pollutant removal in water treatment. Generally, this agricultural waste was mostly discarded into landfill sites or even burning it [8]. Burning them is one way to hasten waste, but it can only instigate pollution to the environment [9].

Durian husk contains cellulose, hemicellulose, and lignin fiber, which makes it has the ability to trapping water pollutants and also due to their macro-porous nature and resistance to temperature, pH, mechanical stress and prolonged submersion in water [10]. Commonly, activated carbon is usually used in water treatment for pollutants removal as the adsorbent material. However, this type of water treatment seen as very expensive material especially for some developing countries that cannot afford to use it on a large scale [11]. Therefore, with this research, physical adsorbents using durian husk have been introduced as the removal of pollutants from wastewater and resources, and at the same time to minimize the agricultural waste in terms of optimizing them by reuse and utilization as the low-cost adsorbent in water treatment. A recent study by Ngabura et al. [12] which had optimized durian husk for biosorption of lead with the percentage biosorption efficiency ranged from 97.12 to 68.94% as a result of the effect of pH, bio-sorbent dose, temperature, initial metal ion concentration, and contact time in the removal of Pb (II).

It is important to note that in this research, an on-site water sample of groundwater was used for the before and after treatment of both adsorbents, using the real samples taken from the site and not based on lab-scale initial concentration as the previous studies have conducted [13,14]. In the meantime, groundwater can be highly contaminated by magnesium and calcium due to the dissolution of carbonate minerals in the rocks and soil properties of the geological formations. That makes the groundwater hardness higher compared to the other water resources [15]. Additionally, TDS, EC, and salinity are generally reflecting the mineral salts in the water and are influenced by major ions in groundwater [16]. The significant ions include calcium (Ca), magnesium (Mg), potassium (K), sodium (Na), bicarbonate (HCO_3), chloride (Cl), and sulfate (SO_4) [17]. Since water hardness categorized as major ions in water, it defined as the amount of dissolved calcium and magnesium in the water.

Water containing calcium carbonate at concentrations below 60 mg/L, which categorized as soft; 60–120 mg/L, moderately hard; 120–180 mg/L, hard; and more than 180 mg/L, very hard as referred to the WHO guideline. Hard water becomes a major concern in every cleaning task by households and health concerns [18]. On the other hand, the synthetic coagulant of aluminium sulphate (alum) that often used in water treatment technology for the removal of TDS and EC, was reported able to induce Alzheimer's and Parkinson's Neurological Disease [19]. Consequently, from this research, it is important to find a better and more organic compound to replace alum in order to treat the wastewater and resources. The aim of this research is to find out the potential of durian husk as a physical adsorbent in treating the hardness of groundwater by understanding the factors such as adsorbent dosage and settling time in terms of reducing total Hardness (TH), electroconductivity (EC), and total dissolved solids (TDS).

2. Research methodology

2.1. Materials and chemicals list

Mechanical wet and dry mill blender, laboratory jar test, 3 kg of durian peel, pellets of NaOH (sodium hydroxide, 98.5–100.5%,

AnalaR NORMAPUR), EDTA-disodium salt (ethylenediaminetetraacetic acid disodium salt-dihydrate, ChemAR), $\text{MgCl}_2 \cdot 6\text{H}_2\text{O}$ (magnesium chloride hexahydrate, $\geq 99.0\%$, Sigma-Aldrich), NH_4Cl (ammonium chloride, 99.998%, Sigma-Aldrich), $\text{C}_{20}\text{H}_{12}\text{N}_3\text{NaO}_7\text{S}$ (Eriochrome Black T, Sigma-Aldrich), Ammonia solution (25%, Merck), $\text{C}_2\text{H}_5\text{OH}$ (Ethyl Alcohol 95%, ChemAR), distilled water and groundwater sample.

2.2. Experimental work

Water samples were collected from groundwater wells located at the Kibagu Village, at the North Borneo Island of Sabah, Malaysia. The studied village is still using the untreated groundwater well for domestic water consumption as there still no proper pipe water supply installed and provided to the communities, although it is located in the main city area. The *in-situ* water quality parameters including the pH, conductivity (EC), total dissolved solids (TDS), and salinity were taken in the sampling sites for initial background concentrations. For the preparation of both adsorbents, the durian husk waste was cut into tiny pieces and ground with 2.0 L of the blender and filtered, washed, and rinsed with distilled water. It was then dried at 100 °C for 24 h in the oven to evaporate the remaining water molecules [7]. Then, durian husk was modified with 1 M NaOH for 24 h at room temperature with an agitation speed of 200 rpm jar test for durian husk modification. The purpose of modification of an adsorbent with alkaline treatment such as NaOH is to alter the surface functional groups, surface morphology, and textural properties of the adsorbent for improving its adsorptive selectivity capability on a certain adsorbate [20]. The size of the modified durian husk is undefined and amorphous due to forming elongated and fibrous structure after crushed with mechanical dry mill blender. The modified durian husk was then filtered and washed with distilled water to reduce its excess of NaOH, and finally dried at 100 °C for 24 h. A standard jar test unit equipped was used in this study [11]. Groundwater sample was used to treat by the modified durian husk adsorbent with initial mean concentrations of 320 mg/L. The modified adsorbent dosages used in this study were from a range of 0.5 to 2.5 g/L. The 1000 mL beaker was initially filled with groundwater samples and was placed in the slot of jar mixing stirrer and subsequently agitated. During this process, 0.5, 1, 1.5, 2, and 2.5 g/L of modified adsorbent were added to each beaker and the agitation process was conducted in two stages, 200 rpm for 3 min and 50 rpm for 30 min. Then settling time for 1 h done was met. After that, about 500 mL of water sample was collected and filtered with a filter paper to measure the physical properties of the treated water sample [6]. The batch experiments were repeated with settling time 6, 12, 18, 24 h for determines the saturation point with 1 g/L of physical adsorbents. Then, after settling time achieved, about 500 mL of water sample was collected and filtered with a filter paper. The collected treated water sample by adsorbents was stored in a polyethylene bottle for further hardness analysis.

2.3. Laboratory analysis

The analysis of EDTA titration was conducted for the hardness concentrations analysis, before and after treatment by the durian husk adsorbent. Fig. 1 shows a schematic diagram for a determination of TH by EDTA titration and TDS/EC parameter test. The method of the preparation for determination of water hardness which involved EDTA titration of 0.01 M EDTA by adding the stoichiometric amounts of 0.01 M EDTA and 0.01 M magnesium chloride, MgCl_2 [21]. A 0.5% (wt/vol) Eriochrome Black T indicator solution of ethanol was prepared. The $\text{NH}_3\text{-NH}_4\text{Cl}$ buffer solution was prepared by dissolve 3.2 g ammonium chloride, NH_4Cl in water, added by 29 mL concentrated ammonia, NH_3 then

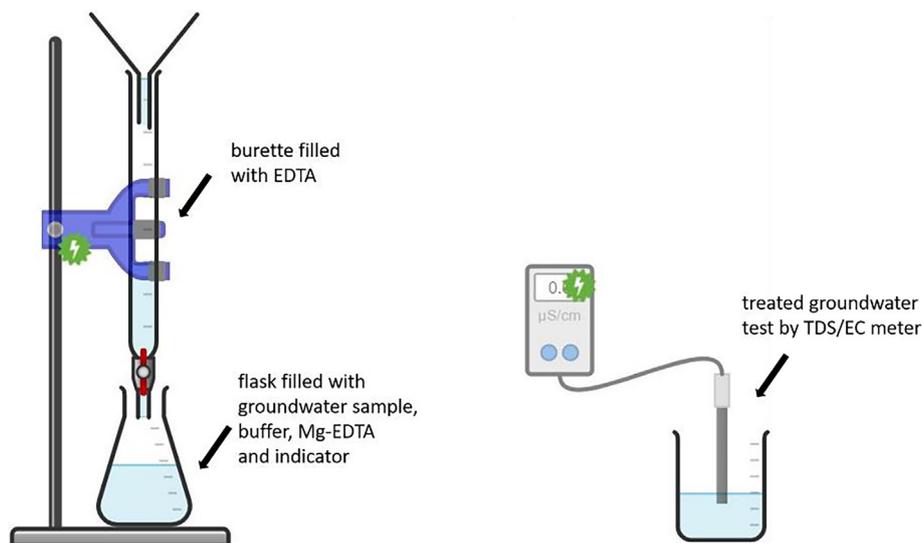


Fig. 1. Schematic diagram for the determination of total hardness by EDTA titration and TDS/EC test.

diluted for about 50 mL with distilled water. The buffer solution was then kept on a polyethylene bottle. Standard 0.01 M EDTA solution was prepared. The EDTA solution was prepared by drying about 1.5 g reagent-grade disodium EDTA dehydrate, $\text{Na}_2\text{H}_2\text{Y} \cdot 2\text{H}_2\text{O}$ in a 50 mL beaker at 80 °C for 2 h. Then it was cooled in a desiccator for 30 min and accurately weighted approximately 1.0 g. It was then transferred to a 250 mL volumetric flask. It was followed by adding about 2.0 mL distilled water and swirl periodically until the EDTA was dissolved. The solution was then allowed to stand overnight before use. Groundwater samples before treatment and the treated samples by adsorbents were prepared. The sample was added with 50 mL aliquot of a pipette to a 250 mL wide-mouth Erlenmeyer flask. Then it was added by 2 mL of the buffer solution, 0.5 mL of the Mg-EDTA solution, and 5 drops of the indicator solution. The solution was then titrated with 0.01 M EDTA until the color changes from wine red through purple to pure blue color. The reaction of color changes is slow, and titrant was added slowly and stirred thoroughly in the vicinity of the endpoint.

2.4. Total hardness calculation

Total hardness (mg/L) is determined by the ratio of volume EDTA, multiply by a factor of 1000 to the volume of a sample taken, shown in Eq. (1).

$$\text{TH}(\text{mg/L}) = \frac{\text{Volume of EDTA}(\text{mL}) \times 1000}{\text{Volume of sample taken}(\text{mL})} \quad (1)$$

2.5. Total hardness removal efficiency

Total hardness efficiency (R %) is defined as the ratio of the different TH concentration before and after adsorption ($C_i - C_f$) to the initial concentration of the adsorbate in aqueous solution (C_i), shown in Eq. (2).

$$R(\%) = \frac{(C_{\text{initial}} - C_{\text{final}})}{C_{\text{initial}}} \quad (2)$$

where; C_{initial} = initial concentration in mg/L and C_{final} = final concentration in mg/L.

3. Result and discussion

The removal efficiency of total hardness (TH) by durian husk, as shown in Fig. 2, it can be seen that it gradually increase from 7 to 18% with a significant rise of removal with the increase of the adsorbent dosage from 0.5 to 2.5 g/l by the durian husk. This could be due to the increase in the number of possible binding sites and surface area of the adsorbent for the removal of the pollutant [23]. At 2 g of adsorbent dosage, the removal efficiency of TH becomes almost constant that might due to the equilibration form of the active sites of the adsorbent for hardness ions [22]. It is also shown that the mean concentration of hardness (TH) has significantly reduced from the initial concentration of 320 mg/L before the treatment and decrease onto 261 mg/L of CaCO_3 , with the increased dosage of the modified durian husk. The uptake of TH shown decreases slowly at 2.5 g of durian husk due to the factor of aggregation particles, where at lower doses all the active binding sites were entirely exposed and filled, whilst only a small fraction of active sites are exposed at higher doses [23,24]. This can be explained by the fact that all adsorbents have a limited number of active sites, and at the certain higher concentrations the active sites will become saturated [23]. Thus, the increase in adsorbent dosage aggregation of particles take place, as a result the efficiency and TH uptake will decrease. The other reason could be associated with the adsorbent elements from the durian husk itself leaching out to the water samples with calcium carbonate that may consist of many constituents and properties such as metals, nutrients and other trace elements that may compete with each other for the binding sites during adsorption process [25]. Therefore, this might lower the percentage of removal efficiency in this research.

For the average total hardness (TH) and removal efficiency of durian husk by settling time are shown in Fig. 3. The settling time was performed to investigate the trend of the hardness removal efficiency as well as to determine the saturation point towards the TH removal after the rapid and slow mixing in the jar test. The removal efficiency of TH by durian husk was gradually increased from 9 to 25% with an increase of the settling time. The highest removal efficiency of hardness recorded 25.2% at 18 h of settling time and within 18 and 24 h the saturation point obtained as shown in Fig. 3. At 12 h, the removal efficiency of TH becomes constant might due to the active sites of adsorbent that are almost occupied with hardness ions after 12 h of settling time

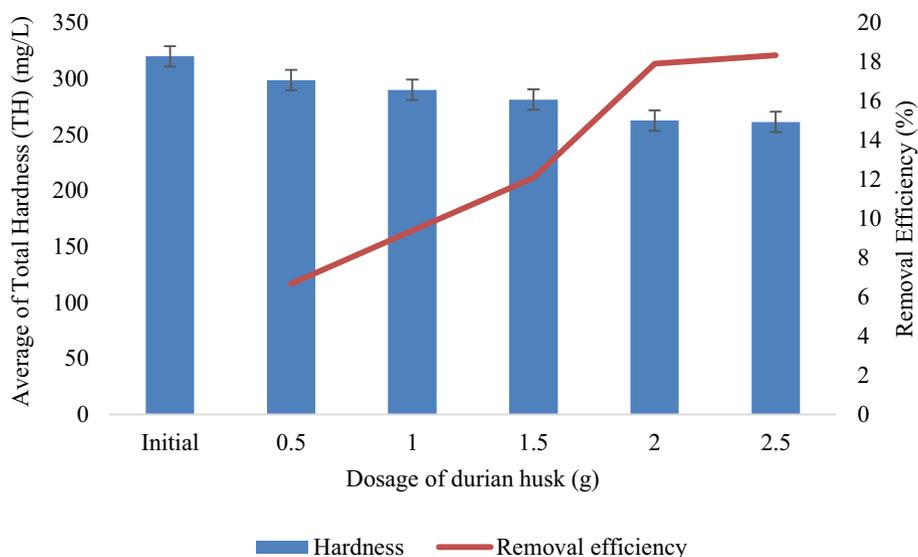


Fig. 2. Effect of adsorbent dosage on total hardness with its removal efficiency by modified durian husk without settling time.

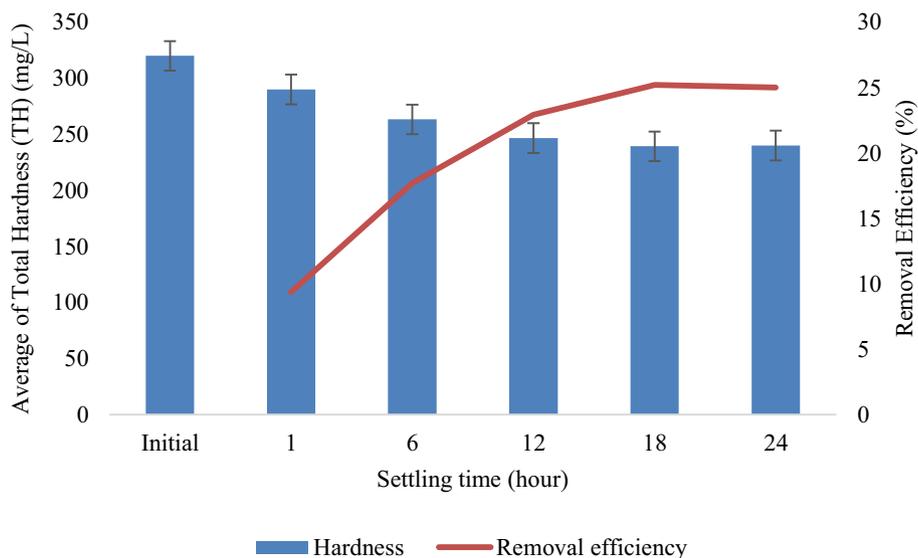


Fig. 3. Effect of settling time on total hardness with its removal efficiency by 1 g dosage of modified durian husk.

[22]. It is also shown that the mean concentration of hardness (TH) has significantly reduced from the initial concentration of 320 mg/L before the treatment and decrease onto 240 mg/L of CaCO_3 with the increase of the settling time. The removal of TH from the groundwater sample by durian husk shown significant with the settling time. The groundwater hardness reduced with settling time could be explained by ion exchange between functional groups such as carboxyl group as cation exchanger of durian husk and divalent metal cations of total hardness from groundwater sample, that makes the hardness particles to settled very slowly or maybe not at all that mutually repel each other between the surfaces force of the durian husk and the hardness particles over time [26,27].

From the result in Fig. 4 is shown that the mean concentrations of total dissolved solids (TDS) had reduced the initial concentration of 426 ppm before the treatment by durian husk adsorbent and decreased onto 424 ppm concentration at 1 g favorable dosage achieved. But then after the peak, a sudden increase in the TDS concentration observed during the second interval with more

dosage of adsorbent and longer settling time. The concentration of TDS shows a significant inconsistent trend for the different dosage of durian husk until the rest of the intervals, with lower fluctuations percentage of removal efficiency observed. While for electrical conductivity (EC) in Fig. 5, the mean concentrations have reduced from the initial concentrations of 676 $\mu\text{S}/\text{cm}$ before the treatment, onto 665 $\mu\text{S}/\text{cm}$ concentration at 1 g optimum dosage achieved. An unstable trend and EC fluctuations were observed throughout the settling time, with a sudden increase even for the first dosage of 0.5 g durian husk addition onto the removal of EC. Another dropped of concentrations at 2 g of dosage can be seen in this study and another significant rise onto 711 $\mu\text{S}/\text{cm}$ concentration at the end of the 2.5 g dosage. An inconsistent trend for both TDS and EC by the effect of the adsorbent dosage means there is no adsorption process occurs. The TDS is contained mainly of carbonates, bicarbonates, chlorides, sulfate, nitrates, calcium, magnesium, sodium, iron including manganese [23]. This will increase the number of active ions competing for the available binding sites and also the lack of active binding sites on the adsorbent at higher

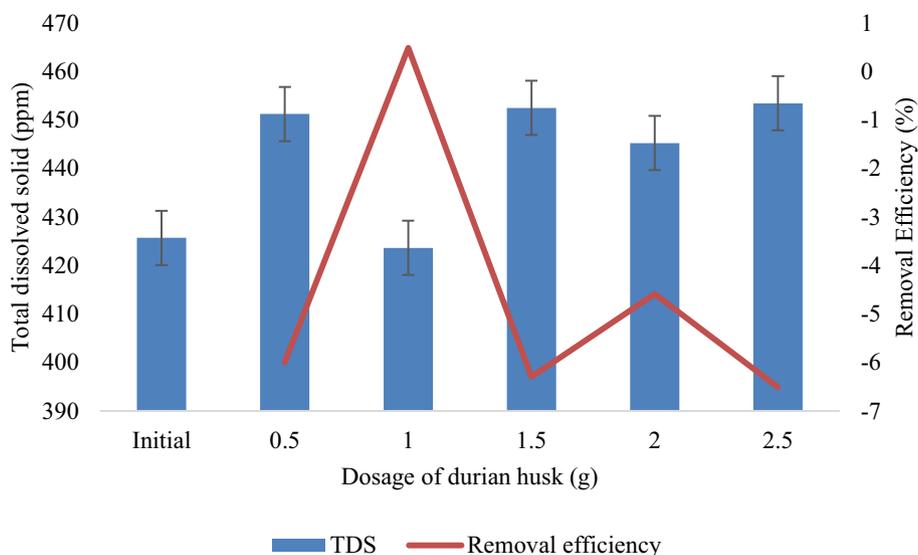


Fig. 4. Effect of adsorbent dosage on total dissolved solids and its removal efficiency by modified durian husk without settling time.

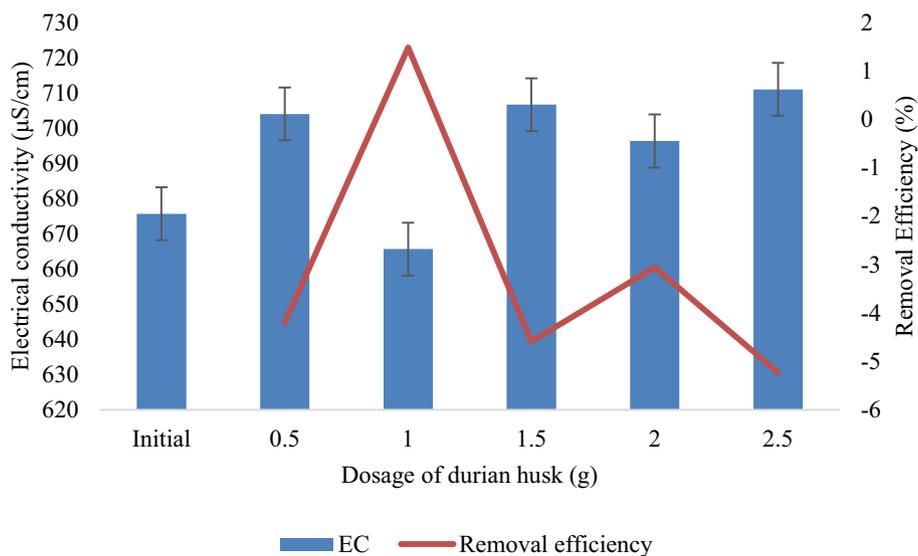


Fig. 5. Effect of adsorbent dosage on electrical conductivity and its removal efficiency by modified durian husk without settling time.

concentrations [28,29]. Therefore, more ions were left un-adsorbed in the solution at higher concentration levels [29].

Overall, the trend of the adsorbent's removal efficiency by durian husk was low for total dissolved solids (TDS) and conductivity (EC) in this research. Previous studies that were conducted by Lee et al. [2] and Abdullah et al. [8] found that durian husk adsorbent in their lab-scale study using synthetic water samples for removal managed to reduce the initial concentration of synthetic hard water from 709.33 reduced to 219 mg/L with increasing dosage of 0.5 to 2.5 g; TDS reduced by 1689.67 to 496 ppm; and EC also reduced by 1762.33 to 632.66 $\mu\text{S}/\text{cm}$. However, in their research was using the highest concentration of hardness which up to 700 mg/L of CaCO_3 as to find out the removal efficiency for water softening in water treatment by using the durian husk. Thus, the potential to reduce total hardness with a higher initial concentration of hardness is higher compared to lower concentration. Nevertheless, this research is the first one using the real or on-site water samples with a lower initial concentration of hardness, EC and TDS within 320 mg/L; 676 $\mu\text{S}/\text{cm}$; 426 ppm and below respectively,

compared to previous study using initial concentrations ranges between 700 mg/L to 1200 mg/L. It also is shown that the TDS and EC concentrations in this study increasing when the adsorbent dosage been added for the treatment of the water pollutant removal. This negative trend in Figs. 3 and 4 might occur due to the addition of sodium ions leaching from the durian husk itself that was modified with 1 M NaOH solutions that contribute to the elevated of TDS and EC when the dosage was increased. These cellulose, hemicellulose, and lignin fiber of durian husk contains methyl esters that modified with sodium hydroxide, thereby carboxylate ligands and sodium ions will be formed as products [30]. The modified methyl esters can be modified onto carboxylate ligands by treating the biomass with a base from the hydrolysis reaction such as sodium hydroxide that will increase the site-binding treatment ability and efficiency of the biomass [30].

The effect of settling time on the concentrations of total dissolved solids (TDS) is shown in Fig. 6, along with the effect of settling time on the concentrations of electrical conductivity (EC) in Fig. 7. Overall, the concentrations of TDS and EC before

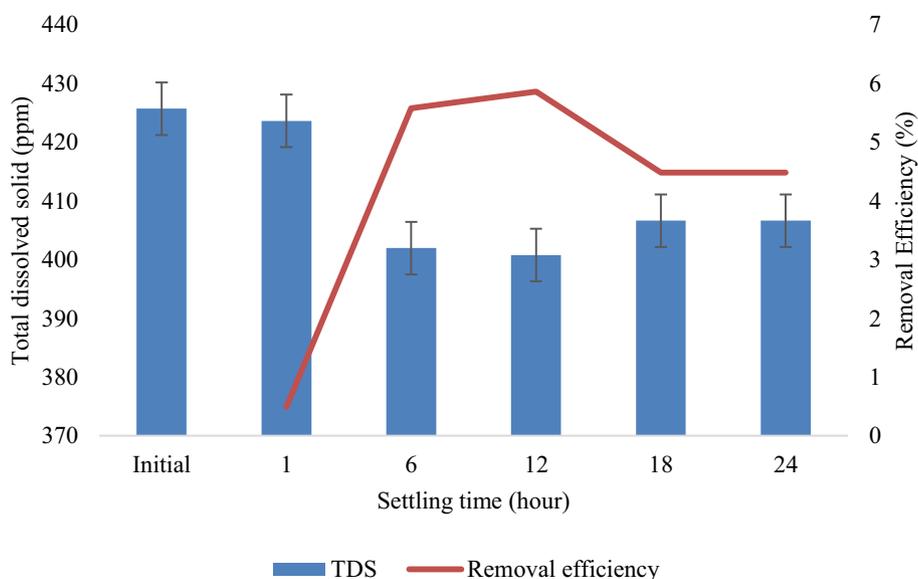


Fig. 6. Effect of settling time on total dissolved solids with its removal efficiency by 1 g dosage of modified durian husk.

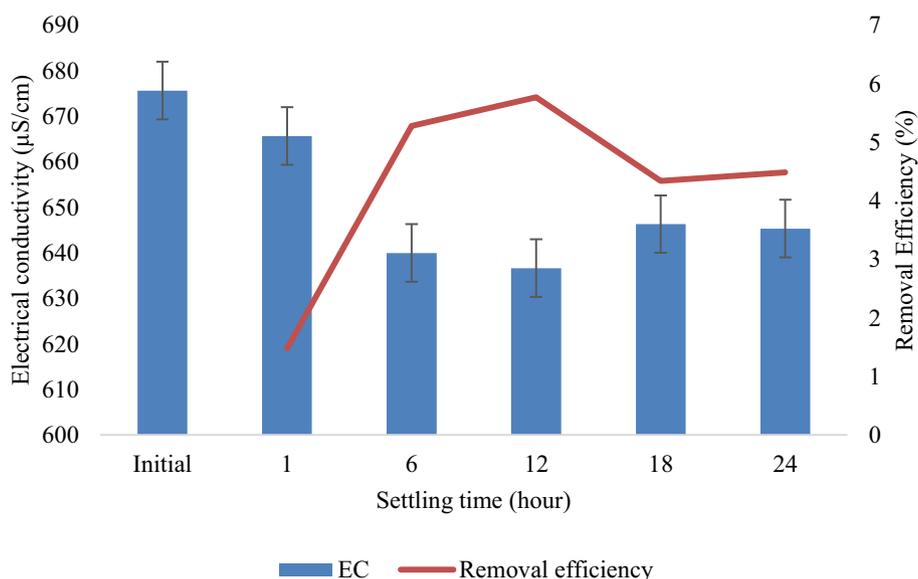


Fig. 7. Effect of settling time on electrical conductivity with its removal efficiency by 1 g dosage of modified durian husk.

and after the treatment of the groundwater samples by durian husk adsorbent are recorded at the ranges between 407 and 426 ppm of TDS; and 640–676 $\mu\text{S}/\text{cm}$ of EC respectively. It is also shown that the concentrations of TDS and EC have significantly reduced from the initial concentration of 426 ppm before the treatment and decrease onto 407 ppm of TDS; and from 676 $\mu\text{S}/\text{cm}$ reduced to 645 $\mu\text{S}/\text{cm}$ of EC after the treatment, with the increase settling time of the modified durian husk from 1 to 24 h. Additionally, it also found that there are small removal efficiency of TDS and EC towards the settling time effects, with the highest removal percentage recorded at 6% for both TDS and EC, both within the interval time 1 h to 6 h. In this research, the settling time shows a positive result in reducing TDS, EC, and hardness might be due to the ions such as magnesium and calcium from the water samples that eventually bind to the surface of the durian husk via ion exchange of the mechanism of adsorption [9,26,27].

Table 1 shows the comparison of total hardness (TH), total dissolved solids (TDS), and electroconductivity (EC) maximum removal from hard water by several types of adsorbents and its dosage. The initial TH, TDS, and EC of hard water and its removal efficiency by the previous study are highlighted in this work. This study is the first using initial concentration that is at the very low level within 300 mg/L compared to the previous studies using a higher concentration of hardness between 500 and 1200 mg/L of CaCO_3 . According to Argun & Dursun [31], chemical modification in adsorbents such as HCl, H_2SO_4 , HNO_3 , NaOH, and KOH was used to increase the proportion of active binding surfaces in the adsorbents [31]. They studied the adsorption capacities and ability on the wood-based adsorbents and found the maximum adsorption capacities of NaOH (98%), KOH (96%), HCl (85%), Fenton (94%), polymerization (94%), tetra ethylene glycol (97%), and diethyl ether (95%). They argued that the adsorption capacity of the individual adsorbent depends on the characteristics of the individual

Table 1

Comparison of the total hardness (TH), total dissolved solids (TDS), and electroconductivity (EC) maximum removal from hard water by the types of adsorbents and its dosage.

Adsorbent type	Durian husk	Peanut hull	<i>Moringa oleifera</i> seed pod husk	Cactus materials, <i>Opuntia</i> spp.
Modifying agent	NaOH	NaOH	KOH	Untreated
Factors affecting adsorption	Dosage			
Quantity of dosage, g	0.5 to 2.5	0.5 to 2.5	1 to 5	0.5 to 4.5
Initial TH, mg/L	320	1241	1214.8	547
Lowest TH achieved, mg/L	261	219	300	105.46
Highest removal efficiency TH, %	18	89.05	69	80.7
Initial TDS, ppm	426	2185.6	–	–
Lowest TDS achieved, ppm	423.6	496	–	–
Highest removal efficiency TDS, %	0.5	77.31	–	–
Initial EC, $\mu\text{S}/\text{cm}$	676	2355	–	–
Lowest EC achieved, $\mu\text{S}/\text{cm}$	665.7	632.66	–	–
Highest removal efficiency EC, %	1.5	73.14	–	–
Reference	This study	Fantasy [32]	Varada [33]	Mgombezi et al. [22]

Note: The symbol of “–” means that are not reported in the study.

adsorbent, the extent of chemical modification, and the concentration of adsorbate. The adsorbent, modifying agent, and adsorbate used in this study are durian husk, NaOH, and total hardness (CaCO_3) respectively.

4. Conclusion

In conclusion, from the level status of total hardness for ground-water after treatment still fell in hard water standard for drinking water quality as referred to WHO guidelines. However, both TDS and EC on groundwater after treatment were still classified as Class I within 1000 $\mu\text{S}/\text{cm}$ and 500 ppm respectively of national quality standard. Durian husk adsorbent in this research shown a significant increase in removal efficiency for TDS, EC, and total hardness concentrations with the effect of dosage and settling time, nonetheless, went sudden reduction when exposed to a higher dosage. The TH and TDS/EC results seem not to correlate with each other due to the TDS/EC interpreted as whole salt minerals or major ions while TH only figure out as Ca and Mg ions. The adsorption of other constituents such as nutrients or other metals and ions might occur during this experiment. Thus the removal efficiency of TH, TDS, and EC depicted as too low. The highest removal efficiency of hardness is recorded 25.2% by the effect of settling time at 1 g of adsorbent dosage. Yet, the percentage of removal efficiency by durian husk adsorbent overall was achieved lower in this study might be due to the initial concentration of hardness, TDS and EC are too low within 300 mg/L, 400 ppm and 600 $\mu\text{S}/\text{cm}$ using on-site samples compared to the previous study using the initial within 1200 mg/L, 1700 ppm and 1700 $\mu\text{S}/\text{cm}$. Higher concentrations will provide a relative abundance of dissolved elements and metals and solubility that will induce and increase the removal efficiency rate of the adsorbent, thus exposure rate of the adsorbents surface area and biding sites also higher and instigate. The mechanisms of durian husk adsorbent removal are complex and should be further examined for a better understanding.

CRedit authorship contribution statement

C.M. Payus: Conceptualization, Formal analysis, Methodology, Supervision, Writing - original draft. **M.A. Refdin:** Conceptualization, Formal analysis, Investigation, Methodology, Writing - original draft. **N.Z. Zahari:** Conceptualization, Formal analysis, Funding acquisition, Investigation, Methodology, Supervision, Writing - original draft. **A.B. Rimba:** Funding acquisition, Project administration. **M. Geetha:** Funding acquisition, Project administration. **C. Saroj:** Funding acquisition, Project administration. **A. Gasparatos:** Funding acquisition, Project administration.

K. Fukushi: Funding acquisition, Project administration. **P. Alvin Oliver:** Conceptualization.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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