An investigation of a mixture of coconut husk and rice husk as activated carbon for treatment of wastewater

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Abstract. Malaysia's population has rapidly developed every year, with considerable waste being produced. It caused a severe effect on the surroundings and reduced the quality of water on surface water bodies. Thus, significant improvement has been implemented to increase the quality of wastewater effluent and combat environmental issues. This research evaluates the activated carbon performance from rice husk and coconut husk to reduce the pollutants in wastewater effluent. The effluent was collected at a wastewater treatment plant in UiTM Dengkil. In this study, the characteristics of wastewater effluent were analysed. Later, the activated carbon (AC) was produced using the chemical agent of ZnCl2 divided into six batches of experiments. The experiment has been observed in two weeks, and the results have been examined for the percentage of nitrate-nitrogen, nitrite-nitrogen, ammonia-nitrogen, phosphorus, and E. Coli. For the batch experiment of AC in mixed coconut husk and rice husk, the percentage removal is 87%, 79%, 54%, 95%, and 100%, respectively. It has been found that the mixture of rice husk and coconut husk AC was significantly effective in removing all the pollutants. It can be deduced that the treatment using activated carbon has improved the water quality and fulfilled the limitation set in the Standard of Environmental Quality (Sewage), 2009.

Keywords: activated carbon; coconut husk; rice husk; wastewater

1. Introduction
Malaysia's population is rapidly increasing every year, around 32.73 million for the first quarter of 2020, rising by 0.6 per cent compared to the first quarter of 2019 [1]. Besides, the world's population will grow quickly in the next 30 years with more than double the value [2]. Thus, the effluent discharge from wastewater treatment will be reused to cater for water deficiency issues. This wastewater comes from various sources, which are industrial, commercial, agricultural, sewer inflow, surface runoff, and infiltration [3]. It can be denoted that this effluent must be treated before being released to the surface water bodies [4]. In the long term, wastewater reuse requires a proper system with best management practices to enhance the effluent's quality. Suitable, cost-effective and adaptation of greener technology need to be implemented in line with Sustainable Development Goals (SDGs) agenda [5].
In proportion to the SDGs 6 for clean water and sanitation, efficient treatment of the pollutants anticipated from the wastewater influent is critical. This ensures the availability and sustainable management of water and sanitation for all in 2030 [5]. Sewerage system and treatment output are essential to maintain surface water and groundwater [6]. Through this efficient technology, environmental and public health will be protected [7]. Along with SDGs 14 on life below water, improper wastewater treatment can cause environmental deterioration in rivers, lakes, and other water bodies, including the ocean [7]. Preserving and safeguarding the quality of water can conserve the ecosystem diversity.

Wastewater treatment is a method to remove the contaminants in the wastewater and ensure that the discharged water does not harm the environment, human and aquatic life. The wastewater treatment consists of various unit operations and must be designed according to several factors such as water characteristics, construction material, maintenance cost, wastewater flow rate and many more. The unit processes are categorised into primary, secondary, and tertiary wastewater treatment. Primary and secondary treatments are called conventional treatments [8].

For the tertiary treatment, different processes can be used, such as rapid sand filtration (RSF), slow sand filtration (SSF), microfiltration, coagulation, activated carbon, reverse osmosis, and additional disinfection [8, 11]. It can be denoted that tertiary treatment necessitates additional secondary or advanced techniques to reduce organics, turbidity, nitrogen, phosphorus, sulphate, metal, and pathogen [11]. The tertiary treatment can be emphasised as extra protection to enhance the quality of effluent wastewater in long term sustainable management.

Adsorption techniques using activated carbon (AC) can be considered one of the most efficient removing pollutants in wastewater [12], especially in tertiary treatment. The filtering process eliminates contaminants and any other impurities employing chemical adsorption [13]. Carbon refers to the raw material that undergoes the carbonisation process, while the active term is material in carbon stated undergoes activation process to the open-pore surface area to increase the absorption rate. There are three steps of the adsorption process, firstly substances adsorb to the outside of the carbon granules and then move into the carbon pores and later, they adsorb to the internal carbon wall [14]. In a developing country, activated carbon is widely used, and the demand becomes higher as this method improve water quality significantly. Local agricultural waste, livestock, and industrial by-products are potential sources for activated carbon materials [13]. Furthermore, on SDGs 11 for sustainable cities and communities, the waste materials accumulated from the solid waste management can be an alternative for the activated carbon components, which would later bring a safer environment to human living more inclusive and viable [5].

The production of activated carbon has been done using low cost and renewable materials, for instance, banana peel, orange peel, coconut shell [12], rice straw [15], rice husk [16], and coconut husk [17]. Moreover, abundant agricultural waste, especially rice husk and coconut husk, have created an undesirable problem in many developing countries. Rice husk is one of the paddy residues, and Malaysia is a prominent producer of paddy [18]. Hence, the coconut husk is waste from coconut that consists of 33 to 35% of husk. This much unwanted agricultural waste is usually being thrown away with no specific use [19]. Therefore, this study emphasises the combination of rice husk and coconut husk as activated carbon to treat the wastewater effluent. Here, the performance of activated carbon of rice husk, coconut husk, and the mixture of rice husk with coconut husk would be evaluated in treating the effluent coming from wastewater treatment plant of Universiti Teknologi MARA (UiTM), Dengkil, Malaysia. Different concentration of the AC was used to determine the percentage removal of pollutants after each experiment.

2. Methodology

2.1. Wastewater Sample Collection
The effluent samples were taken from the wastewater treatment plant (WWTP) outlet at UiTM Dengkil, Malaysia (see Figure 1). The samples were collected in April 2021. In situ testing was conducted to
determine pH, temperature, and dissolved oxygen [20]. Later, laboratory analysis was conducted to determine the values of biological oxygen demand (BOD), chemical oxygen demand (COD), total suspended solids (TSS), nitrate-nitrogen, nitrite-nitrogen, phosphorus, ammonia-nitrogen, and *E. coli* [20].

![Figure 1. Location of UiTM Dengkil, Malaysia](image)

2.2. *The activated carbon preparation.*
Rice and coconut husk were carbonised at 600°C for 1 hour in a furnace (see Figure 2). An aqueous solution of 1 L consisting of 100 grams of zinc chloride (ZnCl$_2$) was used. The char produced was then added to the aqueous solution of zinc chloride (see Figure 3). After 1 hour of proper mixing, the apparatus was allowed to cool. Later, the oven was switched on and set up at 105°C, and the blend materials were let to dry in the oven for 24 hours. After that, the activated product was then cooled at room temperature, and these materials were washed with distilled water to remove any undiluted residue of zinc chloride (ZnCl$_2$) (refer to Figure 4) [21].

![Figure 2. Coconut husk and rice rusk after burned in the furnace.](image)

![Figure 3. Material mixed with ZnCl$_2$.](image)
2.3. Experimental setup
The batch experimental setup is arranged to treat raw effluent samples and activated carbon from rice and coconut husk. At first, the materials consisted of rice husk, coconut husk, and a mixture of activated carbon from the rice husk with coconut husk were weighted in six different proportions. The variations of activated carbon with different rice husk and coconut husk concentrations were displayed in Table 1.

Table 1. Different weights of activated carbon for the batch experiment.

| Batch | Rice Husk (g) | Coconut Husk (g) | The total weight (g) |
|-------|---------------|------------------|---------------------|
| A     | 0             | 4.3              | 4.3                 |
| B     | 17.2          | 4.3              | 21.5                |
| C     | 4.3           | 0                | 4.3                 |
| D     | 8.6           | 0                | 8.6                 |
| E     | 12.9          | 0                | 12.9                |
| F     | 17.2          | 0                | 17.2                |

The six (6) beakers of 2 L were prepared and labelled as A, B, C, D, E and F, with 1 L of effluent wastewater samples were poured into each beaker. Then, activated carbon was added into every beaker based on its respective weight (see Table 1). The experiment was done using the jar test method [12]. The adsorption process occurs in the jar test experiment, which allowed the activated carbon to be mixed with the effluent samples. Then, all parameters were evaluated in triplicate every day for almost two weeks for nitrate-nitrogen, nitrite-nitrogen, phosphorus, ammonia-nitrogen, and *E. coli*.

3. Results and discussion

3.1. Characteristics of wastewater effluent from WWTP of UiTM, Dengkil
Table 2 shows the characteristics of untreated effluent from wastewater treatment plant (WWTP) at UiTM Dengkil compared to the Standard of Environmental Quality (Sewage), 2009 [22].
Table 2. Untreated effluent results of wastewater treatment plant of UiTM Dengkil compared to Standard of Environmental Quality (Sewage), 2009 [22].

| Parameter                        | Wastewater effluent | Standard of Environmental Quality (Sewage), 2009 |
|----------------------------------|---------------------|-------------------------------------------------|
| TSS (mg/l)                       | 8                   | 50                                              |
| COD (mg/l)                       | 0                   | 120                                             |
| BOD (mg/l)                       | 4.2                 | 20                                              |
| pH                               | 7.81                | 6 - 9                                           |
| Nitrate – nitrogen (mg/l)        | 1.11                | 10                                              |
| Nitrite – nitrogen (mg/l)        | 0.562               | -                                               |
| Ammoniacal – nitrogen (mg/l)     | 1.75                | 5                                               |
| Phosphorus (mg/l)                | 3.72                | 5                                               |
| E. coli (MPN)*                   | > 2419.6            | -                                               |

* MPN: Most probable number

Based on Table 2, all parameters were lower than the Standard of Environmental Quality (Sewage), 2009 [22]. The suspended solid was 8 mg/l which was lower than the standard of 50 mg/l. This result showed a low inorganic and organic matter concentration in the sample [23]. BOD was 4.2 mg/l that was smaller than 20 mg/l. High BOD value relates to the extreme amount of organic matter that the bacteria need to oxidise the pollutants, reducing the dissolved oxygen level in the wastewater. However, in this case of the effluent sample from WWTP of UiTM Dengkil, the BOD value is low, which a smaller quantity of pollutants existed in the wastewater. Here, the COD value was not detected, and this showed that the samples did not have oxidisable organic material. pH was at the value of 7.81, which was in the range of 6 to 9. Therefore, the sample is neither too acidic nor alkaline. The pH value must be within the limit to avoid harmful effects on aquatic life and the growth retardation of microorganisms [24]. The ammonia – nitrogen was 1.75 mg/l which was reduced than the standard (5 mg/l); the quantity of nitrite – nitrogen was 0.562 mg/l, and nitrate – nitrogen was 1.11 mg/l lower compared to 10 mg/l from the standard [22]. Thus, this emphasised that the nitrification and denitrification process occurred in the aeration tank in the WWTP.

Furthermore, phosphorus (3.72 mg/l) was lesser than 5 mg/l referred to the Standard of Environmental Quality (Sewage), 2009 [22]. However, it has been detected that E. coli was higher than 2419.6 MPN, indicating faecal contamination and risk of pathogen infection in the effluent wastewater samples. These E. coli bacteria were found in the environment, foods and intestines of people and animals. Most E. coli are harmless, but other serotypes can cause diarrhoea, urinary tract infections, respiratory illness, and other diseases. To sum up, the effluent wastewater from UiTM Dengkil can be considered high quality except for a significant quantity of E. coli bacteria.

The WWTP in UiTM Dengkil only consists of primary and secondary treatment, which is conventional methods. Therefore, it is crucial to evaluate the study using AC to enhance the quality of the effluent of the wastewater in UiTM Dengkil, as this effluent will be discharged to the Langat River and flows to the intake of Bukit Tampoi Water Treatment Plant (WTP), which is 10 km away from the campus site.

3.2. The batch experiment of activated carbon

The batch experiment consists of six (6) sets of beakers (refer to Table 1). The two sets of beakers contained activated carbon of coconut husk (4.3 g), coconut husk mixed with rice husk (21.5 g) labelled as A and B, respectively. Another four sets of beakers comprised activated carbon from rice husk with different concentrations of 4.3 g, 8.6 g, 12.9 g, and 17.2 g, labelled as C, D, E and F, separately. The batch experiments were observed for about two weeks. Day 0 indicated the data for the effluent of wastewater without activated carbon (see Table 2).
Figure 5. pH values after adsorption process.

According to the graph trend shown in Figure 5, the pH value of all six-batch of experiments can be seen decreasing linearly every two days. The raw data already achieved the neutral state and passed the standard [22]. The value drops after the adsorption process but remains within the standard [22] between pH 6 and 9. The coconut husk is an excellent material for activated carbon compared to rice husk as this substance accelerated in showing good results of pH value; as can be seen in Figure 5, Batch A consists of 4.3 g of coconut husk rapidly decline from pH 7.81 to pH 6 in 3 days despite the result shown for Batch C of 4.3 g of rice husk which shows the decrease to pH 6 in longer time (9 days). In addition, the different weights of rice husk of 17.2 g (F) affected the pH of the effluent sample from 7.81 falls to 6.54 while for 4.3 g rice husk (C) has dropped from 7.81 to 6.94 pH value. The rice and coconut husk mixture as activated carbon show a larger reduction from the actual value towards the nine-day incubation period. Based on Figure 5, the pH constantly decreases as the contact time of the AC with effluent samples increases.
Referring to Figure 6, batch experiments for C, D, E, and F give the same values as raw data of effluent wastewater at the value of 1.11 mg/l for the first day. Batch experiments for A and B have shown good reduction since each batch consists of coconut husk. It can be observed that the rice husk as activated carbon did not show good performance compared to the samples which contained coconut husk. However, it can be denoted that the value of samples comprised of activated carbon of rice husk gradually declines as more time is employed to the batch experiment. In batch B, the coconut husks aid the rice husk in reducing the nitrate – nitrogen value when both materials are mixed. In comparison between batch C, D, E and F, the batch experiment of F had the most efficient removal with 94% of nitrate – nitrogen reduction. However, activated carbon with coconut husk and rice husk mixture has achieved a high removal rate of 87% for nitrate – nitrogen in the batch experiment.
Figure 7. Nitrite - nitrogen vs Time

Figure 7 shows the value of nitrite - nitrogen of effluent wastewater sample which have gradually declined for all batch of the experiment except Batch E. Highest reduction for nitrite – nitrogen has been detected in Batch F until day 5. The removal of nitrite – nitrogen concentration for batch A, B, C, D, E and F is 41%, 79%, 66%, 60%, 31%, and 84%, respectively. From the observation, 17.2 g of rice husk for the batch F showing the most efficient rate with 84% of removal for nitrite - nitrogen. Hence, the additional weight of the AC has become the important factor for the highest removal of nitrite - nitrogen achieved in the experiment. The mixture of both materials (B) also attained a good removal rate which was 79% of nitrite – nitrogen reduction. Thus, this activated carbon can consider as an efficient method to improve the water quality of the effluent.
Figure 8. Ammonia - nitrogen values after the adsorption process.

Based on Table 2, the raw data for the effluent of wastewater was 1.75 mg/l; all ammonia - nitrogen results have greatly reduced after being treated with AC. Coconut husk and rice husk effectively remove ammonia – nitrogen from the wastewater samples, where each batch experiment achieved 90%, 54%, 70%, 66%, 93%, and 54%, respectively, towards the end of the incubation period. All removals were more than 50% removal and less than 5 mg/l, an acceptable value for wastewater effluent. The results indicated that, when contact time increases, the ammonia – nitrogen value will be lowered. Batch F showed less reduction in ammonia – nitrogen, which resulted in 0.80 mg/l after nine days. This contrasts with Batch E, which consists of 12.9 g rice husk reflected the most efficient activated carbon with 93% removal equivalent with 0.13 mg/l. Other than that, 4.3 g of coconut husk in Batch A gives a good reduction rate of 0.17 mg/l compared to 4.3 g of rice husk in Batch C (0.53 mg/l). Hence, 4.3 g of coconut husk AC in Batch A is sufficient to remove the same value of ammonia – nitrogen concentration compared to the high amount of rice husk AC (12.9g) needed in Batch E to achieve the highest removal rate (refer to Figure 8).
Figure 9. Phosphorus values after the adsorption process.

Figure 9 shows phosphorus reduction in batch experiments consisting of coconut husk and rice husk activated carbon. From the experimental results, Batch A and Batch B contained both coconut husks showed a significant decrease of 0.44 mg/l and 0.3 mg/l, respectively and continued to drop until 0.19 mg/l and 0.17 mg/l towards the end of the incubation period. This can be displayed that the coconut husk substance in AC contributed to a considerable reduction of phosphorus. The highest phosphorus removal was achieved in Batch B, consisting of 4.3 g of coconut husk and 17.2 g of rice husk with 95% removal. Thus, based on the results shown, the mixture of coconut husk and rice husk can be presumed to be an effective method for phosphorus removal and increased the water quality of effluent samples. Moreover, positive results can be achieved for all the batches which demonstrated declination patterns parallel to the increment of time given.
Figure 10. *E. coli* values after the adsorption process.

From Figure 10, it can be revealed that the AC from coconut husk and rice husk were the best material for removing *E. coli*. The raw effluent sample shows more than 2419.6 MPN. Nearly 100% of *E. coli* has been removed in most batches, with the highest shown in Batch E and F. It can be denoted that, as the weight of AC increases, the removal of pollutants becomes higher for rice husk AC. However, after being treated with AC, a significant reduction has been observed for all the batches.

Implementation of tertiary treatment is crucial to enhance the effluent quality of wastewater before being discharged to the water bodies. This can be reflected by the treatment of AC using coconut husk and rice husk. The nitrate–nitrogen and nitrite - nitrogen in mixed activated carbon produced 87% and 79% removal rates. For the pH and ammonia – nitrogen in mixed AC became less effective after two weeks incubation period where pH has declined to an acidic condition, and the removal rate of ammonia – nitrogen was 54%. However, good performance has been identified in the phosphorus and *E. Coli* removal rates, respectively, at 95% and 100%.

4. Conclusion

It has been deduced that the materials used for activated carbon can be substituted to low-cost material, are easy to get and have higher removal efficiency. The study on this reuse agricultural waste AC can sustain and protect the environment based on the results.

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