Preparation of activated carbon from oil palm empty fruit bunch by physical activation for treatment of landfill leachate

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Abstract. The present work demonstrates the preparation of oil palm empty fruit bunch derived activated carbon (EFBAC) by air in a closed furnace for the adsorptive removal of COD, ammoniacal nitrogen and colour from landfill leachate. The removal of COD, ammoniacal nitrogen and colour were investigated with EFBAC activated at different activation temperatures and activation time. Yield of EFBAC at different activation temperatures and time were also examined. The experimental results manifested that increasing activation temperature increased COD removal but decreased colour removal. It has no significant removal on ammoniacal nitrogen. Increasing activation time increased COD and colour removal but has no significant removal on ammoniacal nitrogen. Furthermore, yield of EFBAC decreased with increasing activation temperature and time. The finding illustrated the applicability of EFBAC as a promising solution for pollution from landfill leachate due to its economic feasibility for real application.

1. Introduction

Municipal solid wastes have become a major issue globally. Increasing amount of municipal solid wastes occur from many factors such as growing of population, productivity, consumption habits, lifestyles, and resource use and also development of technologies [1]. Along with municipal solid wastes, landfill leachate is the real problem to the groundwater and also water course such as rivers and lakes.

Landfill leachate is known as a contaminated liquid effluent generated from rain water percolating through deposited municipal solid wastes. Typically, stabilized landfill leachate constitutes a high strength of ammoniacal nitrogen (>400 mg/L), moderate strength of chemical oxygen demand (COD) (5000 – 20000 mg/L) and also low BOD/COD ratio (< 0.1) [2]. Landfill leachate imposes negative impacts to humans as well as the environment as it may seep into groundwater and flow to the water course. Ammoniacal nitrogen is the main toxicant to the living organism. High concentration of untreated ammoniacal nitrogen can promote algae growth and decrease dissolved oxygen level through algae bloom (eutrophication). It is also toxic to aquatic lives [3].

There are various types of treatments that can be adopted to treat landfill leachate such as physical, chemical, or biological, such as adsorption, membrane filtration, chemical precipitation, air stripping, and reverse osmosis. Biological treatments are preferred if the landfill leachate is young due to their reliability, and cost-effectiveness [4]. However, most landfill leachate is considered stabilized leachate, therefore, biological treatment is not favourable. Adsorption is the most common treatment for stabilized
landfill leachate. The process is simple but the drawbacks include (i) the need of frequent regeneration, and (ii) high cost of production of adsorbent [2]. Hence, researchers all around the globe seek for renewable sources of activated carbon such as banana frond [5–6], coffee ground [7], durian peel [8], palm shell [9], tamarind fruit seed [10], sugarcane bagasse [11–13], orange peel [14], and rice husk [15]. However, no study has yet been found using palm oil empty fruit bunch (EFB) as precursor for the treatment of landfill leachate. Malaysia produced 19.92 million tonnes of crude palm oil in 2017 and it is estimated that approximately 19.92 million tonnes of EFB were generated as their waste [16]. The abundance of EFB attract researcher to explore the usage of EFB as a precursor of activated carbon [17–20]. Previously, EFB derived activated carbon focussed on removal of phenol and its derivatives, dyes and metals such as boron and iron.

The objective of this study is to examine the effect of preparation condition of EFB-based activated carbon by physical activation in the limited air environment for the treatment of the post-treatment landfill leachate. The effect of activation temperature and time were evaluated in terms of removal of COD, ammoniacal nitrogen, and colour.

2. Material and Methods

2.1. Landfill leachate
The leachate was collected from a municipal solid waste sanitary landfill, Tanjung Langsat Landfill Site, located at Pasir Gudang, Johor, Malaysia. The coordinates are 1°28'22"N 103°59'36"E. The samples were taken after the leachate undergoes anaerobic and aerobic treatment, before discharged into the nearby water course. The leachate samples were then stored in refrigerator at temperature of 4°C for preservation purpose. The characteristics of the leachate are presented in table 1. COD of leachate was measured according to ASTM D1252 (Test Method B—Micro COD by Sealed Digestion and Spectrometry). For ammoniacal nitrogen and colour, it was measured by using Hanna Multiparameter Photometer (HI 83099).

Table 1. Characteristic of leachate samples.

| Parameter               | Unit | Average | Min | Max |
|-------------------------|------|---------|-----|-----|
| COD                     | mg/L | 974     | 504 | 1279|
| BOD                     | mg/L | 77.1    | 63  | 102 |
| Suspended Solid         | mg/L | 213     | 180 | 280 |
| Ammoniacal Nitrogen     | mg/L | 129     | 117 | 160 |
| Colour                  | PCU  | 4042    | 2960| 7450|

2.2. Oil palm EFB
EFB was collected from Alaf Palm Oil Mill located in Kulai, Johor, Malaysia.

2.3. Preparation of EFB based activated carbon
EFB was washed and dried at 105°C for 24 hours. Then, it was ground into particles sized 1.0 – 2.0 mm. The precursor then underwent carbonization in a closed furnace at 450°C for 2 hours before activated at various temperatures (600 – 900°C) and various activation times (15 minutes – 4 hours) as in table 2. The activated carbon (EFBAC) produced was cooled down to room temperature overnight. The yield of the products were calculated. EFBAC underwent batch adsorption study by putting in 0.2 g of adsorbent in 30 mL of leachate and was shaken in isothermal shaker for 2 hours at 200 rpm. The removal efficiency of all parameters was calculated as follows,
Percentage removal = \frac{C_o - C_e}{C_o} \times 100\% \quad (1)

Where,
C_o = Initial concentration of parameters, mg/L
C_e = Final concentration of parameters, mg/L

Yield percentage of samples were measured by using equation as follows,

\text{Yield} = \frac{m_f}{m_i} \times 100\% \quad (2)

Where,
m_f = mass of activated carbon (after activation), g
m_i = mass of precursor (before activation), g

| Table 2. Preparation of EFBAC |
|-----------------------------|
| Sample | Activation Temperature (°C) | Activation Time (min) |
|--------|-----------------------------|-----------------------|
| 1      | 600                         | 15                    |
| 2      | 700                         | 15                    |
| 3      | 800                         | 15                    |
| 4      | 900                         | 15                    |
| 5      | 800                         | 30                    |
| 6      | 800                         | 45                    |
| 7      | 800                         | 60                    |
| 8      | 800                         | 90                    |
| 9      | 800                         | 120                   |
| 10     | 800                         | 180                   |
| 11     | 800                         | 240                   |

2.4. Characterization of EFBAC
EFB and EFBAC (Sample 11) were characterized by their textural properties by nitrogen adsorption using Micromeritics 3 Flex Surface Characterization analyzer to obtain surface area, pore volume, and pore size.

3. Results and Discussion
3.1. Characterization of EFBAC
Surface area, pore volume, and pore size are important characteristics of activated carbon as it can explain the removal performance of adsorbate. Nitrogen adsorption is a standard procedure for determination of porosity of the carbonaceous adsorbents. Detailed characteristics of the porosity of EFB and EFBAC are listed in table 3. From the data, it is evident that BET surface area, Langmuir area, and total pore volume of EFB were 1.1163 m²/g, 1.7836 m²/g, and 0.0006 cm³/g, for EFBAC 694.86 m²/g, 803.62 m²/g, and 0.3170 cm³/g respectively. It is noted that micropore area and micropore volume for EFBAC were 559.31 m²/g, and 0.2208 cm³/g, respectively. However, there was no significant difference on average pore size between EFB and EFBAC.

Increased surface area and pore volume were due to increasing amount of micropores. The EFBAC developed in this study was basically microporous since 69.6% of total pore volume was micropore volume. It was also noted that 80.5% of total surface area of EFBAC was micropore area. This phenomenon happens because at high temperature, low molecular weight volatiles are released from the
matrix structure, and generated a fragile structure, resulting in increasing pore size [20]. Insignificant difference on pore size between EFB and EFBAC may be due to the volatilization of low molecular weight volatiles which used the same route or pores that made the pores not widened but deepened in EFBAC. It was proven by data from this study that the pore size is almost similar but the pore volume increases as the EFB is activated into EFBAC.

Table 3. Textural properties of EFB and EFBAC.

| Parameter               | Unit       | EFB      | EFBAC    |
|-------------------------|------------|----------|----------|
| BET surface area        | m²/g       | 1.1163   | 694.86   |
| Langmuir surface area   | m²/g       | 1.7836   | 803.62   |
| Micropore Area          | m²/g       | N.A *    | 559.31   |
| External surface area   | m²/g       | 1.7260   | 135.54   |
| Total pore volume       | cm³/g      | 0.0006   | 0.3170   |
| Micropore volume        | cm³/g      | N.A *    | 0.2208   |
| Average pore size       | nm         | 1.896    | 1.837    |

*The micropore area and volume are below the detection limit of the instrument.

3.2. Yield of activated carbon from different activation condition

Yield, in activated carbon preparation, is the percentage final weight of activated carbon produced after activation, from its initial weight of raw material on a dry basis. Apart from removal performance, yield of the activated carbon is a relatively important information for the industry since it can predict the amount of activated carbon generated after activation. Figure 1 represents the yield percentage of EFBAC at different activation temperatures and also with reference to carbonized EFB. Increasing activation temperature reduces the yield of the EFBAC. This happens as increasing temperature will increase volatile releases, resulting in decreasing yield [21].

The effect of activation time on EFBAC yield is displayed in figure 2. It is apparent that the yield percentage decreases with increasing activation time. This phenomenon is due to more volatiles which might be released with longer activation time [21]. It is also suggested that the weight loss is due to the development of pore structure at longer activation time [22].

![Figure 1. Yield percentage vs activation temperature.](image1)

![Figure 2. Yield percentage vs activation time.](image2)

Table 4 lists a comparison of yield of activated carbon derived from different precursors under different activation conditions. In can be concluded that the activated carbon prepared in this work

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showed significant yield compared to other activated carbon by physical activation with 23.08\% yield. This finding can reduce the cost of producing activated carbon.

Table 4. Comparative evaluation of yield of various activated carbon by physical activation.

| Precursor        | Activator | Yield (%) | Reference |
|------------------|-----------|-----------|-----------|
| EFB              | Air       | 23.1      | Present study |
| Date stones      | Steam     | 28.4      | [23]      |
| Almond shell     | CO\(_2\)  | 25.0      | [24]      |
| Sugarcane bagasse| Steam     | 23.0      | [25]      |

3.3. Removal performances of activated carbon from different activation condition

The removal of COD, ammoniacal nitrogen, and colour by EFBAC synthesized at different temperatures with reference to EFB and also carbonized EFB were depicted as figure 3. The removal of COD ranged from 23\% to 81\%. The removal of ammoniacal nitrogen ranged from 16\% to 32\% and removal of colour ranged from 55\% to 74\%. It was observed that higher removal of COD by using EFBAC was activated at higher activation temperature, and the highest COD removal was by using EFBAC activated at 900\(^\circ\)C (81\%). For ammoniacal nitrogen, there was no significant effect of activation temperature towards removal of ammoniacal nitrogen. For colour, the removal was decreasing with increasing activation temperature.

Using EFBAC activated at high temperature gives high COD removal due to its highly developed surface area. According to Aishah [20], high activation temperature produces high surface area of activated carbon. High surface area of activated carbon gives more active sites to adsorb organics that contributes to COD. Apart from surface area, pore size also contributes to adsorption of COD from landfill leachate. It was reported in previous study that activated carbon derived from EFB has average pore diameter ranging from 1.2 to 16.4 nm [21, 26–28]. This pore size is suitable for adsorption of humic substances such as humic and fulvic acid since they are the main components that make landfill leachate highly non-biodegradable [29]. It was also reported that aquatic humic substances have particle sizes that vary from 0.9 to 6.6 nm in diameter [30]. This finding is supported by textural properties of EFBAC depicted in table 3.

In contrast, EFBAC activated at high temperature gives lower colour removal from landfill leachate which might be due to decreasing total pore volume. Closure of pores can occur at high temperature due to shrinkage of activated carbon [20]. Currently, there are no study yet to investigate correlation of activation temperature and pore volume on physically activated EFB. However, Tan et al. (2012) found that increasing carbonization temperature can increase pore volume but after 200\(^\circ\)C, pore volume decreases [31]. Although high surface area of activated carbon is achieved by using high activation temperature, the high temperature also ruptures pores that are already developed making the remaining pores to have less volume but higher in number. On the other hand, suspended solids in leachate may contribute to colour reading. The most plausible explanation is that suspended solid particle size in leachate is larger than pores developed in EFBAC which makes the removal to decrease with increasing activation temperature. This finding is in contrast with Aziz et al. (2007) where they found that colour in landfill leachate was mainly contributed by organic matters [32].

However, ammoniacal nitrogen removal was not significantly affected by activation temperature and time. This might be due to EFBAC has no functional group since it is physically activated. This is in agreement with Aziz et al. (2004) who concluded that the adsorption capacity of activated carbon is determined not only by its total surface area, but also by its internal porous structure and the presence of functional groups on the pore surface [33].
The optimum activation temperature of EFBAC was found at 800°C, where at activation temperature higher than 800°C has no significant increment of COD and ammoniacal nitrogen removal, and also decreased colour removal.

Removal of COD, ammoniacal nitrogen, and colour by using EFBAC activated at different activation times are shown in figure 4. It is observed that increasing activation time can increase the removal of COD and colour in leachate with maximum removal of 72.3% and 68.2%, respectively. However, there was no significant removal in ammoniacal nitrogen using EFBAC activated at different activation times (16% to 20% removal).

![Figure 3. Removal percentage of COD, ammoniacal nitrogen and colour vs activation temperature.](image1)

![Figure 4. Removal percentage of COD, ammoniacal nitrogen and colour vs activation time.](image2)

Increasing removal of COD and colour by increasing activation time might be due to increase in surface area. This may be due to the activator (air) which was penetrated deeper inside particle making the development of porosity higher at long activation time compared to the porosity of particles at short activation time [20]. High surface area gives more sites for adsorption to occur. However, insignificant amount of ammoniacal nitrogen removal with respect of activation time may be due to desorption of the small size of ammonia particle in leachate relative to pore size in EFBAC.

4. Conclusion

The potential of EFB based activated carbon by air was examined. The adsorption of COD, ammoniacal nitrogen, and colour by using EFBAC has been demonstrated. The experimental data show that high activation temperature and high activation time produce activated carbon that can adsorb significant amount of COD, ammoniacal nitrogen, and colour from landfill leachate. It also shows that significant yield percentage obtained by using physical activation hints its economic feasibility for real application.

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