



Effects of Biosorption Technique Using Various Fruit Waste Activated Carbon in Improving Chenderiang River Water Quality

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ABSTRACT

The release of heavy metals from industries and other human-made activities into waterways is the primary cause of environmental pollution. Heavy metals are very toxic and once ingested through the mouth and nose; they can accumulate in the organs inside the human body and poison the internal organs with serious illnesses and even can cause death. Hence, in order to determine the water quality, biosorption technique using low-cost, eco-friendly fruit or agricultural wastes i.e., banana and orange peel activated carbon were prepared and characterized using FT-IR spectroscopy and nitrogen adsorption-desorption analysis. The adsorbents were then used to treat and identify the physicochemical parameters of Chenderiang River water, Tapah, Perak, Malaysia. Physicochemical analysis of river water sample such as DO, BOD₅, pH, Temperature, EC, Turbidity, Hardness, Alkalinity, TSS, (PO₄)³⁻, AN, Cu, Cr, Fe, and Zn before and after treatment were measured using Multiprobe system, EC meter, Lamotte Smart3[®] Colorimeter, and AAS. The results showed that orange peel (OP) activated carbon is more effective than banana peel (BP) activated carbon in reducing most of the pollutants including heavy metals in Chenderiang river water. The findings of this study showed that the biosorption technique using fruit waste is a low-cost, biodegradable and eco-friendly alternative method to remove pollutants including heavy metals in water and indirectly can reduce municipal solid waste. Therefore, can reduce water pollution and ensure good water quality.

1. Introduction

Water pollution considered a global problem, and its control measures have become more and more important over the past few years. River water quality is necessary for determining the level of pollution, as 98% of the water used comes from rivers. Nevertheless, 70% of water resources are channeled into agriculture. Pollution of waterways is increasing because of the high concentration of

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existing pollutants. As a result, water “scarcity” increases as the availability of good quality water decreases, so water treatment costs also increase due to the pollutants in the water [1].

Municipal or industrial wastewater is treated by wastewater treatment plants that contain a high volume of organic matter and contaminants, including Cu, Cd, Fe and Zn ions. Even with very low concentrations of heavy metals, water can be polluted and become highly toxic. Heavy metals can enter a water supply from industrial and consumer waste, as well as acid rain, which causes heavy metals to be discharged from soils into streams, lakes, rivers, and groundwater [2]. However, municipal solid waste (MSW) is one of the main factors that have potential threats to the biological ecosystem in which involve interactions between living organisms and non-living organisms [3]. This is because of its mismanagement that has led to more serious environmental problems and affects the health of living organisms, including humans [4]. In Malaysia, a person from a rural region can produce MSW from about 0.5 kg to 0.8 kg, while people from an urban region produce about 1.9 kg per day [5]. This amount of waste is expected to increase as population growth leads to continued urbanization in Malaysia.

For example, water quality is mainly contaminated by toxic heavy metals that emanate from human activities such as industries, agriculture and river settlements [6,7]. A comparison of the presence of heavy metals in the sediment of the bed and water column was found to be extremely high in the sediment in the bed. They are noxious and accumulate permanently and can cause cancer. Cd and Pb ions may bioaccumulate in the body tissues of aquatic organisms and are also heavily absorbed in food chains [8]. Aquatic species are a source of food for people. As a result, they are readily carried to humans by food. The bioaccumulation and toxicity of heavy metal pollution are issues of concern around the world [9].

Various methods that have been employed in treating polluted wastewaters such as membrane filtration, solid-phase extraction, solvent extraction, reverse osmosis (RO), chemical co-precipitation, electrochemical precipitation, ion exchange, evaporation, chelation and adsorption [10]. Mainly, most of these methods eliminate metals incompletely and consume high operational costs. Removing toxic sludge and other secondary contaminants also entails additional costs [11]. In comparison with these methods, it is known that adsorption is one of the widely investigated methods for removing the contaminants [12]. This method is most efficient treatment method due to its low operating cost and low cost [13]. A number of low-cost materials were used as adsorbents to remove different metals from aqueous media by the adsorption method [11].

Nowadays adsorption or biosorption has become an effective alternative method in the removal of copper and also other metals when compared to existing technologies, because of its simplicity, low or zero cost, and its high ability in removing either organic or inorganic water pollutants. A large amount of organic waste from agriculture, wood and food industries can also be used to adsorb Cu^{2+} ions. Natural adsorbents had been proved to be more effective [14]. Furthermore, by using inexpensive, renewable sources of household or agricultural waste only needs minimal processing to make biosorbent which is a better and environmentally friendly alternative [15]. Among this, the agricultural waste adsorbent such as orange peel, lemon, mandarin, pomelo, banana peels, potato peel are very effective in adsorption of heavy metals [16-18]. Recent research also shows that agricultural waste is widely used in engineering applications to generate power and extract chemicals. It is noted that, the particle size of activated carbon (AC) derived from agricultural waste is finer, allowing it to access the surface more easily and increasing the rate of absorption [19].

Al Khusaibi *et al.*, [20] have reported that AC derived from the orange peel (OP) found to be more effective AC in removing heavy metals with the removal highest percentage of 50.1% compared to the banana peel (BP) activated where the removal percentage is found to be 44.1%. Both orange and banana peel mainly composed of carbohydrates which include hemicellulose and pectin that consists

of different functional groups which act as a bonding agent in the biosorption process and easily adsorb into the water sample to remove the pollutants [19]. However, Orange peel (OP) AC is more effective due to its fiber content in which consists of high hydroxyl radicals; hence it has more adsorption capacity towards heavy metals and other pollutants in the water.

In this present study, inexpensive AC from organic waste with no commercial value such as bananas and orange peel were used as an adsorbent to remove contaminants from the Chenderiang River water. The prepared carbonized adsorbents composed of more pores with high surface area. Thus, it increases the process of adsorption and mechanical strength. The opening of pores in adsorbents and its efficiency as an adsorbent is due to the use of an activating agent during the chemical activation process of producing activated carbon (AC) [21]. As a conclusion, using a carbonaceous adsorbent will reduce waste, lessen environmental barriers, and also provide value-added products.

2. Experimental Methodology

2.1 Preparation of the Adsorbent

Fruit wastes such as BP and OP were collected from a local fresh juice shop in Tapah, Perak. Fruit wastes such as BP and OP were collected in Tapah, Perak, from a local fresh juice shop. The BP and OP were properly cleaned with tap water, followed by distilled water, and then dried in the sun to remove dirt and other contaminants. To eliminate moisture, the dried BP and OP were trimmed into small pieces and dried again in a 90°C oven for 24 hours. The dry fragments were ground into powder and carbonised in a muffle furnace at 400°C for 4 hours (Figure 1). The carbonized BP and OP powders were separately soaked in 2M HCl (QR_{ec}, 37 %) solutions for 24 h to activate it. The AC powders were then dried for 12 h at 105°C in an oven. After cooling down the powder was kept in a respective bottle and labeled it as BP-AC and OP-AC for further application.



Fig. 1. Schematic diagram for preparation of banana and orange peel adsorbent

2.2 Water Sample Collection and Preservation

Chenderiang River (located in the district of Batang Padang, Tapah, Perak, Malaysia) water samples had been collected during a rainy season using high density polyethylene bottles from 10 cm below the water surface of the river. About 1.5 mL concentrated nitric acid, HNO₃ (QRèc, 65 %) were added to the water sample and then preserved in a cool icebox at 4°C. The purpose of adding nitric acid is to reduce the pH below than 2. The samples were than directly transported to the laboratory for further testing. The analysis was conducted immediately prior to avoid any physical and biochemical reactions to the water samples. The sample bottle was shaken vigorously before performed all the physicochemical analysis.

2.3 Physicochemical and Biological Parameters Measurements

Physicochemical and biological experiments were conducted to identify Chenderiang river water quality and its' level of pollution before and after treatment with banana and orange peel activated carbon. Initially, the river water sample (50 mL) was treated using 0.5 g of BP-AC and OP-AC respectively. The resultant solution was filtered through the filter paper. The pH, dissolved oxygen (DO) and temperature of the filtrated solution were measured using YSI Model 556 Multi probe system (MPS). The five days Biological Oxygen Demand (BOD₅) was determined from DO value. The BOD₅ value was calculated using Eq. (1).

$$\text{BOD}_5 \text{ (mg/l)} = \text{DO (1st day)} - \text{DO (5th day)} \quad (1)$$

The turbidity, hardness, alkalinity, phosphate and ammoniacal nitrogen were measured by Lamotte Smart3® Colorimeter (LaMotte Company, Chestertown, MD, USA). The electrical conductivity was measured using the EC meter. Heavy metals ions such as copper, chromium, iron, and zinc ions were analyzed based on standard analytical procedure using atomic absorption spectroscopy (AAS). Total suspended solids (TSS) were identified through the filtration process using filter paper.

3. Results and Discussion

3.1 Adsorbent Characterization

3.1.1 Infrared (IR) spectroscopy

The FT-IR results provide the qualitative information regarding the characteristic functional groups of the OP-AC and BP-AC (Figure 2). The highest absorption band at 3455 cm⁻¹ in the FT-IR of OP-AC is due to the O-H stretching mode of hydroxyl functional groups, whereas the absorption band at 2832 cm⁻¹ is due to C-H interaction with the OP-AC surface [22]. The presence of a C-O-C bond is shown by the absorption band at 1355 cm⁻¹ [23]. The C-C vibrations in aromatic rings cause the strong band at 1600-1580 cm⁻¹ [24]. However, for BP-AC, the larger peak about 3450 cm⁻¹ is due to the O-H stretching mode of hydroxyl functional groups, whereas the band around 2830 cm⁻¹ is due to C-H interactions on the carbon surface [25]. However, this peak is very small and overlapped with the larger peak of O-H stretch. The band at 1350 cm⁻¹ indicates the presence of Si-O-C bonds, arising from sp³ hybridized carbon.

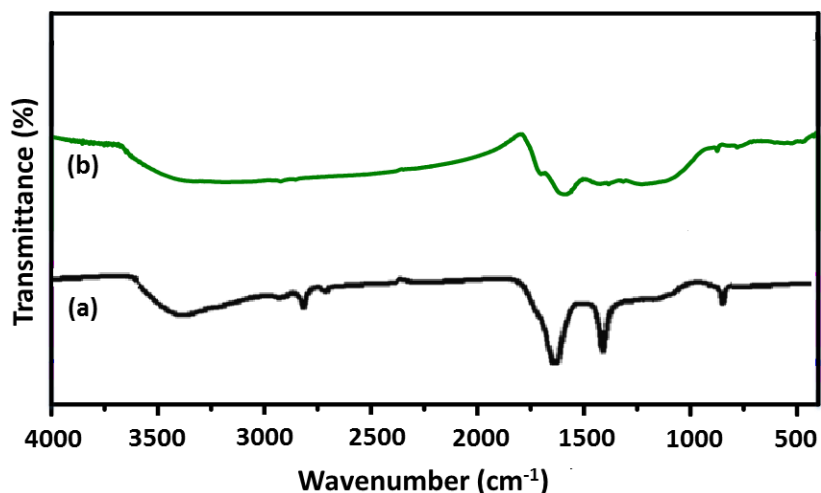


Fig. 2. FTIR spectra of (a) Orange-Peel Activated Carbon (OP-AC) and (b) Banana-Peel Activated Carbon (BP-AC)

3.1.2 Nitrogen adsorption-desorption analysis

Figure 3 shows the N₂ adsorption-desorption isotherms of the prepared OP and BP activated carbons. The OP-AC sample showed type IV isotherm with broader H3 hysteresis loop from P/P₀ = 0.45 to P/P₀ = 1, which indicates an increase in mesoporous adsorption of the OP-AC sample. The reason is that most of micropores would be burned off due to the high activation temperature and then the action of HCl acid on OP carbon essentially results in the formation of mesopores. As the temperature was raised, many small pores inside the carbon walls were gradually destroyed, as discovered by Zhu *et al.*, [26]. The BP-AC showed type I nitrogen isotherms, thus characterizing the microporous materials. Table 1 shows the pore characteristics of the BP-AC and OP-AC samples in detail. The BET surface area of OP-AC was 630.34 m²/g, which was higher than the BP-AC sample (460.02 m²/g), according to the results. This suggests that HCl activation enhances pore formation significantly during the activation phase.

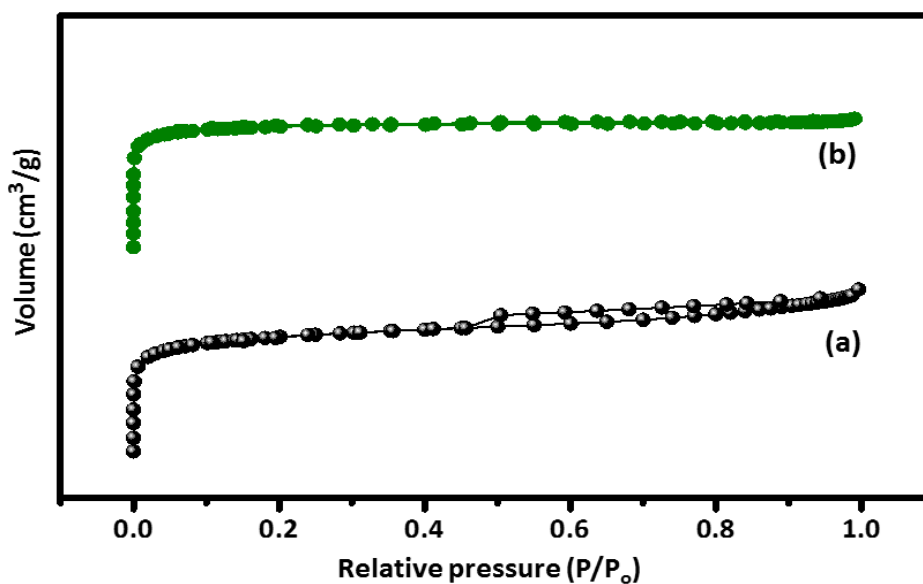


Fig. 3. Nitrogen sorption isotherms of (a) Orange-Peel Activated Carbon (OP-AC) and (b) Banana-Peel Activated Carbon (BP-AC)

Table 1
 Physico-chemical characterization of Orange-Peel and Banana-Peel Activated Carbon

Sample	S_{BET} ($m^2 g^{-1}$) ^a	V_{Total} ($cm^3 g^{-1}$) ^c	σ (nm) ^b
BP-AC	460.02	0.201	5.1
OP-AC	630.34	0.318	7.2
Sample	S_{BET} ($m^2 g^{-1}$) ^a	V_{Total} ($cm^3 g^{-1}$) ^c	σ (nm) ^b
BP-AC	460.02	0.201	5.1
OP-AC	630.34	0.318	7.2

^a Specific surface area

^b Mean pore size obtained from BJH model

^c Total pore volume measured at $P/P_0 = 0.994$

3.2 Physicochemical and Biological Parameters Studies

There is a total of 15 physicochemical and biological parameters of river water analyzed in this study including 4 heavy metals such as copper (Cu), iron (Fe), chromium (Cr) and zinc (Zn) and one inorganic ion: phosphate (PO_4^{3-}). The overall results of physicochemical parameters before and after treatment with BP-AC and OP-AC are presented in Table 2.

3.2.1 Temperature study

Temperature of river water is an important parameter because it influences the water chemistry and in overall it effects the biological activity and growth. The result of the temperature study of the Chenderiang river water is summarized in Table 2. The temperature of untreated river water sample was 9.96 °C and the temperature of river water sample after treated with BP-AC was 22.06 °C while the temperature of river water sample after been treated with OP-AC was 23.30 °C. The temperature of untreated river water was very low due to the measurement being taken after preserved and transfer to the laboratory. The small changes in the temperature for river water may reduce the quality of the stream and affects the aquatic organisms. Normally, warm water holds less dissolved oxygen as compared to cool water. Thus, it is noted that, higher temperature is not suitable for the river water since it may contribute to higher level of toxicity to aquatic life.

3.2.2 pH study

The pH of river water was obtained for untreated, treated with BP-AC and treated with OP-AC. The results are tabulated in Table 2. It is found that the pH of untreated river water was 6.41. The pH reading was slightly higher (6.50) for the water sample which is treated with BP-AC. As compared to BP-AC, the OP-AC treated water sample gives the pH reading of 6.39 which is close to untreated water sample. The pH value represents that the river water is acidic and these values were categorized as Class II based on (NWQS) for Malaysia. To have a suitable pH value that supports aquatic life, the range of pH should be from the range of 6.5 to 9. Thus, maintaining the pH of the river is very crucial since river water is the food source and also habitat for the aquatic organisms. Therefore, should ensure that aquatic ecosystem within this range because high and low pH can be disastrous in nature. Low pH is normally unsuitable for small fish and insects, and acidic water increases heavy metal leaching, which is harmful to fish.

Table 2
 Physico-chemical characterization of Orange-Peel and Banana-Peel Activated Carbon

Parameter	Before treatment	After treatment with BP-AC	After Treatment with OP-AC	Malaysian Environmental Standard Value
Temperature, °C	9.96	22.06	23.30	CLASS I (-) CLASS IIA (normal +2°C)
pH	6.41	6.50	6.39	CLASS II (6-7)
DO, mg/L	9.95	7.34	7.13	CLASS I (>7)
BOD ₅ , mg/L	2.35	1.01	1.54	CLASS II (1-3)
Ec, µS/cm	174.3	170.9	164.0	CLASS I AND IIA (1000 µS/cm)
Turbidity, FAU	28.59	5.02	14.00	CLASS I (5 FAU)
Hardness, ppm	16	4	12	CLASS I (-)
Alkalinity, mg/L	11.5	10	8	-
TSS, mg/L	0.011	0.046	0.004	CLASS I (< 25)
Phosphate, ppm	0.10	0.070	0.064	CLASS III (0.1 ppm)
AN ppm	1.00	0.8	0.64	CLASS III (0.3ppm-0.9ppm)

3.2.3 Dissolved oxygen study

The amount of oxygen dissolved in an aqueous solution is referred to as dissolved oxygen (DO), and an adequate supply of DO gas is essential for aquatic species' survival. The results in Table 2 shows that BP-AC has higher DO value (7.34 mg/L) as compared to OP-AC (7.13 mg/L). Yet, the untreated water has more DO (9.95 mg/L) as compared to the treated water with BP-AC and OP-AC. The difference in the value of dissolved oxygen is depending on several factors such as temperature, pressure, and salinity. Increased in temperature caused the reduction of DO value in the river water sample. Therefore, the results of DO value are directly influenced by temperature of river water sample. These results are well collaborated with the temperature study. It is indicated that, too much dissolved oxygen is unhealthy for the stream and this could be happened for the river which has more plants. Fortunately, this phenomenon is not occurred in the Chenderiang River.

3.2.4 Biological oxygen demand and electrical conductivity study

The BOD analysis for Chenderiang River water was tested and the results are summarized in Table 2. Basically, the BOD test is carried out to determine the level of the oxygen used by aerobic microbes to decompose the organic matter in the water. According to the results in Table 1, the water has good BOD₅ reading of 2.35 mg/L for the untreated water. However, the value has been reduced to 1.01 mg/L and 1.54 mg/L after treated with BP-AC and OP-AC respectively. On the basis of the findings, it can be concluded that, the value of BOD of BP-AC treated water is far better than the water treated with OP-AC. High BOD has the same effects as low dissolved oxygen, causing aquatic organisms to become stressed, choke, and die.

The water electrical conductivity depends on the concentration of ions (i.e., calcium, potassium, sulfate, chloride and nitrate) in the water. Both untreated and treated river water either with BP or OP-AC are still not exceed the standard value of electrical conductivity based on NWQS for Malaysia which is 1000 µS/cm. The electrical conductivity result (Table 2) indicates that the Chenderiang River is not polluted with highly concentrated ions since the readings are still within the natural level that will not cause any damage to the stream.

3.2.5 Turbidity and hardness study

The turbidity and hardness test were conducted for the river water and the results are presented in Table 2. Both the turbidity and hardness values were decreased after treated with OP and BP-AC. Based on the results, BP-AC is more effective than the OP-AC in reducing the turbidity and hardness of Chenderiang River water. The lower turbidity value after treated with BP-AC reveals that the BP-AC has adsorb more suspended solid present in the water as compared to OP-AC and thus the water appeared more clearer and less cloudiness. The hardness of water is tested and the results before and after treated with adsorbent shows that, BP-AC has a value of 4 ppm as compared to 12 ppm and 16 ppm for OP-AC and untreated water correspondingly. The low value of hardness represents by low compounds of calcium and magnesium, and other ions.

3.2.6 Alkalinity and total suspended solids study

The alkalinity and total suspended solid (TSS) tests are carried out for the Chenderiang River water and the results are summarized in Table 2. The alkalinity is measured to test the acidic-neutralizing capacity of the water., whereas the TSS is tested since it is a good indicator of other contaminants in water, particularly metals and organic nutrients that are carried on the surfaces of sediment in suspension. Based on the results, the untreated, OP-AC treated, and BP-AC treated water shows the alkalinity value of 11.5, 8 and 10 respectively. According to the alkalinity and TSS results, OP-AC is more effective than the BP-AC in reducing the alkalinity and TSS value.

3.2.7 Heavy metals study

Metal contamination is a significant issue in many rivers, particularly in developing countries. It's crucial to test the amount of heavy metal in river water to check if it's beyond the safe limit for drinking and cooking. Thus, four major heavy metal tests were conducted for Chenderiang River water. The results are tabulated in Table 3. The result shows that the zinc and chromium are more effective when treated with BP-AC while copper and iron are more effective when treated with OP-AC. There are also a few parameters that are classified under Class III such as ammoniacal nitrogen (AN) and phosphate (PO_4^{3-}) are classified under Class III and it does not safe for drinking and caused it required extensive treatment.

The heavy metal results further supported by the literature reported by Al Khusaibi *et al.*, [20] that the orange peel AC able to remove contaminants and heavy metals about 50.1% than the banana peels AC. BP-AC able to remove about 44.1%. These results due to its' fiber content which consists of high hydroxyl radicals [20]. It is noted that, the OP-AC has the highest potential in removing pollutants including heavy metals since it has high hydroxyl radicals. Thus, OP-AC has high adsorption capability that increases the binding of OP-AC with contaminants and heavy metals in the river water. According to the results, it can be concluded that, Chenderiang River water is not polluted and safe to be used for recreational purposes and also for agriculture.

Table 3

Heavy metal concentration (mg/L) in water sample of Chenderiang River and Malaysian Environmental Standard value (maximum permitted concentration in water (mg/L))

Parameter	Before treatment	After treatment with BP-AC	After Treatment with OP-AC	Malaysian Environmental Standard Value
Zn, mg/L	-0.001	-0.105	-0.067	CLASS I (-)
Cr, mg/L	-0.034	-0.035	-0.045	CLASS I (-)
Cu, mg/L	102.11	3.18	3.32	CLASS II (0.02 mg/L)
Fe, mg/L	1.466	-0.518	-0.503	CLASS I (-)

4. Conclusions

By having biosorption techniques using low-cost eco-friendly fruit/ agriculture waste: Banana (BP) and Orange peel (OP) AC able to remove pollutants including heavy metals to ensure good quality of river water which is safe for drinking. The use of OP-AC is more effective adsorbent than the use of BP-AC. The findings also added advantage in reducing municipal solid waste which mainly causes environmental pollution and gives harmful effects to human. As a future consideration, the river water will be collected weekly and treated using different concentrations of fruit peels containing BP and OP-AC. Additionally, research on identifying microbial contamination in river water will be conducted for a better assessment in the future work.

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