

Preparation of Activated Carbon from Banana Peel Waste as Adsorbent for Motor Vehicle Exhaust Emissions

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Abstract. Air pollution caused by motor vehicle exhaust emissions in the form of harmful gases becomes a problem for the health of living things in the surrounding environment. The alternative way to reduce those emissions is by utilizing agricultural waste as activated carbon (AC). AC can adsorb those emissions due to its porous and high surface area. AC was made of banana peel waste that contains lignocellulose and has considerable amount because of banana processing industrialization. AC was made through dehydration, carbonization at 350°C furnace without or with N₂ gases of 0.15 NL/minute for 1 hour, then chemical activation using various concentrations of H₂SO₄ solution at 85°C for 1 hour. Characterization of activated carbon was done by iodine number test. The result shows that carbon chemical activation by 6 N of H₂SO₄ gave better result of iodine number than the lower concentration, obtaining 428 mg/g and overall yield of 41.68%. The result was even better on physical-and-chemical activation with same concentration of H₂SO₄, obtaining 617 mg/g with surface area of 614 m²/g and overall yield of 56.40%.

1 Introduction

Air pollution is one of serious problems that can be seen in everyday life. It refers to contamination of atmosphere by harmful gases that are dangerous for human health or surrounding environment. There are many cause of air pollution in this world, but one of the main cause that widely recognised to be a large source of air pollution globally comes from transportation sector [1]. In transportation, almost all motor vehicles emit exhaust emission gases from the combustion process of liquid fossil fuel.

A complete combustion of the fossil fuel produces carbon dioxide (CO₂) and water vapour (H₂O) as motor vehicle exhaust emissions. However, if combustion is incomplete, a small fraction of the fuel is oxidized only as carbon monoxide (CO) and some volatile hydrocarbons in the form of vapour also emitted. In addition, fuels that are composed of hydrocarbons mixture may also contain some impurities and sulphur is one of those impurities. When sulphur is oxidized on combustion, it mostly turns into sulphur dioxide (SO₂) and sometimes to sulphate that can assist nucleation particles in exhaust. Another exhaust emission gas is nitric oxide (NO) that comes from oxidation of atmospheric nitrogen at high combustion temperatures together with nitrogen dioxide (NO₂) in small quantities.

One of ways to remove pollutants in exhaust gases that known and often used is by using adsorbents. Many forms of adsorbents used in industry, such as silica gel, alumina, zeolite, bauxite, and activated carbon [2]. Activated carbon is the most frequently used adsorbent

in industry and has displaced many other materials in solvent recovery systems due to the highly adsorption capability [3]. As adsorbent, activated carbon can adsorb a large amount of pollutants due to its large pore volume [4].

Activated carbon can be made using organic matters that contain lignocellulose in the form of cellulose, hemicellulose, and lignin as raw material which can be found in agricultural wastes [5]. Agricultural wastes such as coconut shell, pistachio shells, bamboo, and waste coffee beans are the examples. One of agricultural wastes that can be found commonly in everyday life is banana peel. In 2014 among the tropical fruits, banana was the most commonly eaten fruit in the world [6]. Banana peels become a solid waste of banana processing during production of foods such as banana chips, fried bananas, and unripe banana biomass in human society [7]. Banana cultivation and industrialization generates a considerable amount of waste that is rich in cellulose and starch, which makes this waste is potential as a raw material to make activated carbon [8-9]. Therefore, this paper aims to prepare activated carbon made from banana peel waste as an adsorbent that has potential to be used to adsorb exhaust emission gases emitted from motor vehicle.

There are several processes to make agricultural waste turns into activated carbon, such as dehydration, carbonization, and activation. The activation process can be a chemical activation or physical activation and is used to get larger surface area of activated carbon. Various chemical activating agent can be used in chemical activation process, such as ZnCl₂, KOH,

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H₂SO₄, H₃PO₄, HNO₃, etc. [10]. Recently, various types of raw material, chemical activating agent, and physical activation method in the making process of activated carbon are studied [11-13].

One of recent studies had made activated carbons from banana peel waste for adsorbed natural gas application with the use of ZnCl₂ as the activating agent [14]. The production of activated carbons from banana peel waste in this study are based on that study. However, there are differences in the application and the chemical activating agent. Activated carbons in this study are made for adsorbed motor vehicle exhaust emission application.

The chemical activating agent selected in this study is sulphuric acid (H₂SO₄). It is known that the use of H₂SO₄ as an activating agent is more favoured than ZnCl₂ due to problems of environmental contamination with zinc (Zn) compounds [15]. The use of H₂SO₄ as chemical activating agent can develop porous structure by degrading the amorphous domains of cellulosic materials and carbon framework aromatization [16].

In this study, the concentration of H₂SO₄ used in chemical activation process was varied to get how much effect of H₂SO₄ concentration on the surface area of activated carbon produced. It is also known that physical and chemical activation both can be used in the making process of activated carbon instead of using chemical or physical activation only [17]. Therefore, in this study there was also addition of physical activation by using nitrogen (N₂) gas stream at high temperature before chemical activation process. Either chemical activated only and physical-chemical activated carbon are studied to get how much differences of activated carbon surface area made from banana peel waste by using those two type activation methods.

2 Material and methods

2.1 Material

There were several materials used in this study. The raw material used to make activated carbon was peel of ripe Kepok banana. Chemical activating agent that used in chemical activation process was H₂SO₄ solution with various concentrations. The concentrations of H₂SO₄ solution used were 1 N, 2 N, 4 N, and 6 N. Nitrogen gas was used at one of the treatments variation of the sample at carbonization process. Distilled water was used to rinse activated carbon until the pH is in neutral range.

2.2 Dehydration of banana peels

Before dehydration process, banana peels waste were cleaned and separated from any other trash that might mixed together when banana peels waste were collected. The cleaned banana peels waste then were drying in an oven with 105°C temperature until dried. The purpose of this step is to reduce amount of water, which is contained in banana peel. After the banana peels were dried, banana peels were mashed with a mortar and pestle to reduce size of dried banana peels.

2.3 Carbonization of dried banana peels

The amount of size-reduced dried banana peels were divided into two parts. The first part was carbonized at furnace without stream of N₂ gases and the other part is carbonized at reactor with stream of N₂ gases. The flow of N₂ gases was set at 0.15 NL/min. Carbonization process for both parts was carried out at 350°C for 1 hour. The carbonization process that took place in the absence of nitrogen gas stream produces carbon that is not yet activated. While the one of carbonization process that was carried out in high temperature and with stream of N₂ gases produces carbon that is physical activated. For both types carbon produced after carbonization, the carbons were mashed again with a mortar and pestle then were sieved with 30-mesh sieve to uniform size of carbons produced.

2.4 Chemical activation of carbon

Both types of carbon were broken down into four parts. Each part was immersed in a H₂SO₄ solution at different concentration separately. The ratio of the volume of H₂SO₄ solution to the carbon mass used at all concentration tested was 3 mL of H₂SO₄ solution for each 1 g carbon used. The mixture of carbon and H₂SO₄ solution then were heated at 85°C for 1 hour. After chemical activation was completed, the carbon is chemical activated.

2.5 pH neutralization of activated carbon

After chemical activation, there is a change in pH of activated carbon caused by the use of chemical activating agent. In order to neutralize the pH of activated carbon, the activated carbon was rinsed by distilled water. The activated carbon was rinsed repeatedly until the pH is close to neutral. For each rinsing, the distilled water that added to the activated carbon had been stirred using a stirrer for 10 minutes. After stirring, the mixture was filtered using filter paper and a vacuum filter. When pH of activated carbon had been close to neutral, the rinsing was stopped and the wet activated carbon was dried in an oven at 110°C for 1 hour. After drying, the activated carbon was stored for the iodine test.

2.6 Iodine number test of activated carbon

Iodine number test is used to measuring adsorption capability of activated carbon in an iodine solution. Iodine number measured how much iodine (mg) that can be adsorbed by each gram of activated carbon used. The mixture of carbon and iodine solution was filtrated using filter paper and the filtrate was titrated by sodium thiosulfate (Na₂S₂O₃). The filtrate refers to iodine solution that is not adsorbed by activated carbon. During titration, a little amount of starch solution was added to clarify the endpoint of titration. The titration was ended when the mixture that was originally pale yellow-

coloured changed into transparent. The equation for calculating the iodine number is as follows.

$$IN = \frac{(V_B - V_T) \times N_T \times M_I \times \frac{V_I}{V_B}}{m_{AC}} \quad (1)$$

where:

- IN Iodine number (mg/g)
- V_B Volume of iodine to be titrated (mL)
- V_T Volume of sodium thiosulphate used (mL)
- N_T Normality of sodium thiosulfate (N)
- M_I Iodine molar mass (g/mole)
- V_I Initial volume of iodine (mL)
- m_{AC} Mass of activated carbon (g)

The iodine number obtained was then used to projecting the surface area of the activated carbon using the equation as follows [18].

$$S_I = \frac{IN \times 10^{-3}}{M_I} \times N \times \omega_I \quad (2)$$

where:

- S_I Surface area based on iodine number (m²/g)
- IN Iodine number (mg/g)
- 10⁻³ Conversion factor (g/mg)
- M_I Iodine molecular mass = 126.92 g/mole
- N Avogadro constant = 6.023 × 10²³ mol⁻¹
- ω_I Hypothetical surface area per iodine atom at dense packing (m²)

3 Results and discussion

3.1 Dehydration of banana peels

Dehydration process of banana peels lasts for 24 hours in the oven. There are changes in banana peels condition before and after dehydration. Before had been dehydrated, banana peels were yellow-coloured, mushy, and not rather light. However, after had been dehydrated, banana peels were black-coloured, hard, and lighter. This was happened because there is reduction in water content of banana peels. The mushy condition of banana peels means there is still a high water content in banana peels, but banana peels become hard and lighter as a result of water content removal from banana peels. High temperature of oven makes waters contained in banana peels are evaporated into the air and leave from banana peels.

3.2 Carbonization of dried banana peels

The carbonization results for both types of carbonization treatments can be seen in Table 1.

Table 1. Carbon yields as carbonization result

Carbonization Type	Initial Mass (g)	Final Mass (g)	Yield (%)
Without N ₂ gas stream	9.00	3.99	44.33
	12.00	5.68	47.33
	15.00	7.39	49.27
With N ₂ gas stream at 0.15 NL/minute	30.00	19.76	65.87
	40.00	26.58	66.45

As can be seen in the Table 1, the carbon final mass produced by the carbonization process without nitrogen gas stream was not as large as the carbon final mass produced by the carbonization process accompanied by the nitrogen gas stream. The average carbon yield that produced from carbonization without nitrogen gas stream was 47.39%. Meanwhile, the average carbon yield that produced from carbonization in the presence of nitrogen gas stream was 66.20%. The presence of a nitrogen gas stream prevents the presence of oxygen gas around the carbon that has the potential to decompose carbon excessively and damage the carbon structure. The more damaged the carbon structure cause the carbon structure to become more fragile and lighter than carbon that is still solid structured or less damaged. In the absence of an inert gas stream like nitrogen around the carbon, the oxygen gas in the air is easier to fill in the free space around the carbon and to decompose carbon in excess.

3.3 Chemical activation of carbon

The chemical activation results for both not yet activated and physical activated carbon are shown in Table 2.

Table 2. Mass increase of chemical activated carbon

Carbon Feed Type	H ₂ SO ₄ Concentration (N)	Initial Mass (g)	Final Mass (g)	Mass Increase (%)
Not physical activated	1	2.00	2.10	5.00
	2	2.00	2.30	15.00
	4	2.00	2.83	41.50
		2.00	3.64	82.00
	6	20.00	52.40	162.00
	Physical activated	1	2.00	2.13
2		2.00	2.32	16.00
4		2.00	2.84	42.00
		2.00	3.94	97.00
6		20.00	60.20	201.00

Based on Table 2, the chemical activation process was made carbon mass increased. In a carbon immersion with a chemical activating agent, the chemical activating agent came in between the hexagonal layers and opened the closed surface subsequently [19]. Therefore, when mixing of carbon and H₂SO₄ solution is being done, ions from H₂SO₄ come between layers of carbon so the H₂SO₄ solution mass causes the total mass of carbon increased. The increase of H₂SO₄ concentration causes more ions from H₂SO₄ come in carbon so the mass increase is larger than the smaller one.

The increase of mass on physical activated feed carbon was larger than the not physical activated one. This was because the physical activated feed carbon tends to has stronger and more solid structure than the not physical activated one, so there is also more of carbon layers that ions from H₂SO₄ can come in between. The initial carbon mass difference that was used for chemical activation also affects the final mass of activated carbon, because by the same temperature and heating procedure, the heat load that must be transferred to carbon is different. The less amount of carbon used, the better heat transferred to the carbon so that more volatile substances in the mixture of carbon and H₂SO₄ solution released into the air.

3.4 pH neutralization of activated carbon

3.4.1 Rinse of activated carbon

The rinse of activated carbon aims to neutralize pH of activated carbon because of adding of chemical solution during activated carbon production. The rinse of activated carbon also used for removing other contaminants that still remain on the previous steps. In this study, H₂SO₄ solution was used as the chemical activating agent so it made the initial pH of the activated carbon produced was acidic. The rinse of activated carbon was done by using distilled water repeatedly until pH of activated carbon close to be 7 or neutral. However, the pH of distilled water was only six, so the pH of activated carbon after repeatedly rinse can only make up to six. To increase pH of activated carbon, additional base chemical solution is required. However, it is not be done in this study to prevents any other effect that can be occurred to activated carbon characteristic.

The first of rinsing process resulted a filtrate that was dirty and yellow in colour. It showed that in first rinse of activated carbon, the pH of filtered activated carbon was still low or acidic. When the rinse was being done repeatedly, the filtrate slowly became cleaner and more clear or transparent. The colour change shows that pH of the rinsed activated carbon is being closer to be neutral. The ions from H₂SO₄ that make activated carbon acidic are carried together with distilled water during the filtering process. However, during that process some part of ions from H₂SO₄ still remain in activated carbon and not carried together by distilled water. It makes the rinsing process to neutralize pH of activated carbon can not be done only once, but must done repeatedly.

The required distilled water volume to neutralize the activated carbon was vary on the use of different H₂SO₄ concentration at chemical activation process. The effect of H₂SO₄ concentration used at chemical activation on required distilled water volume for chemical activated carbon is shown in Figure 1. The chemical activated carbon used was the small amount type of carbon (the carbon that around 2 g before chemical activation)

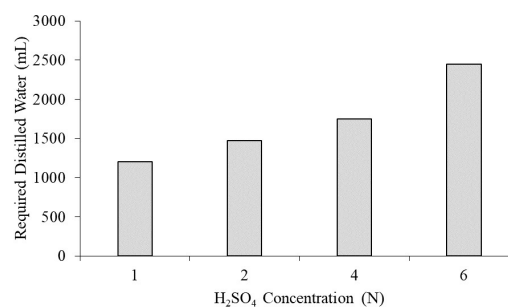


Fig. 1. Effect of H₂SO₄ concentration used on required distilled water volume for activated carbon rinsing (small amount)

In the Figure 1, the type of activated carbon that used higher concentration of H₂SO₄ solution required more distilled water to neutralize the activated carbon. Higher concentration of H₂SO₄ solution used makes more of remaining ions from H₂SO₄ in between carbon layers. Therefore, the carbon that activated with higher concentration of H₂SO₄ solution has lower initial pH than the lower one. To increase pH of activated carbon with lower initial pH required more of distilled water.

The comparison of required distilled water volume for chemical activated and physical-chemical activated carbon in big amount (the carbon that around 20 g before chemical activation) is shown in Table 3. Distilled water around 800 mL was used each rinse of carbon in big amount.

Table 3. Required distilled water volume in activated carbon rinsing (big amount)

Carbon Type	Initial Mass (g)	Required Distilled Water (mL)	Number of Washes (times)
Chemical activated H ₂ SO ₄ 6 N	52.40	5,650	7
Physical-chemical activated H ₂ SO ₄ 6 N	60.20	8,800	11

From Table 3, distilled water to neutralize physical-chemical activated carbon was more required than the chemical activated one. The physical-chemical activated carbon has stronger and more solid structure than chemical activated one that makes more ions from H₂SO₄ come in between carbon layers. Therefore, the initial pH of physical-chemical activated carbon was lower than chemical activated one so more distilled water is required to increase pH of the activated carbon during rinsing.

3.4.2 Drying of wet activated carbon

After the rinsing process, the wet activated carbon was filtered with filter paper then dried in oven. During the drying process, water content in activated carbon was decreasing until the activated carbon became dried. Table 4 shows result of dried activated carbons yield.

Table 4. Yield of activated carbon after rinsing and drying process

Carbon Type	H ₂ SO ₄ Concentration (N)	Mass Before Rinsing (g)	Mass After Drying (g)	Yield (%)
Chemical activated	1	2.10	1.29	61.43
	2	2.30	1.35	58.70
	4	2.83	1.35	47.70
		3.64	1.45	39.84
	6	52.40	17.59	33.56
Physical-chemical activated	1	2.13	1.55	72.77
	2	2.32	1.30	56.03
	4	2.84	1.46	51.05
		3.94	1.41	35.79
	6	60.20	17.04	28.30

From Table 4, activated carbons that activated with higher H₂SO₄ concentration had lower yields than the lower one. It was because activated carbons that more acidic are also lost more ions from H₂SO₄ during the rinsing process that make the yields decreased more. It is also known from Table 4 that the carbons final mass not became the initial carbons mass before chemical activation (neither 2 g or 20 g). It was because there are some contaminants in activated carbon that removed by H₂SO₄ solution and there are some parts of activated carbon that cannot be retrieved due to sticking on the filter paper.

3.5 Iodine number test of activated carbon

3.5.1 Iodine number results

Figure 2 shows the effect of various H₂SO₄ concentrations used at chemical activation to iodine number of activated carbons produced.

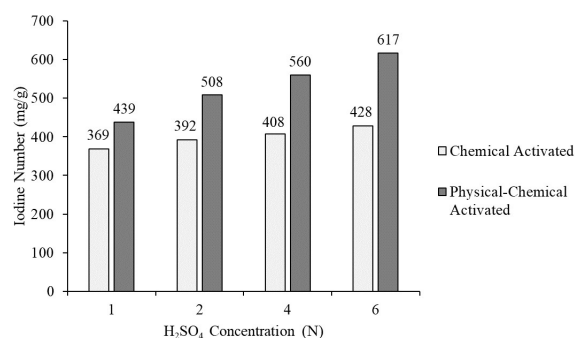


Fig. 2. Effect of H₂SO₄ concentration on iodine number of activated carbon

Iodine number was calculated by using Equation 1. Iodine number test on not yet activated carbon made from banana peel in this study was 362 mg/g. After going through the activation process, the iodine number was increased. The increase of iodine number varies as a result of various H₂SO₄ concentrations used as chemical activating agent. The greatest iodine number in chemical activated only carbon found with use of H₂SO₄ 6 N, which was 428 mg/g. Meanwhile, the greatest iodine number in physical-chemical activated carbon found with use of H₂SO₄ 6 N, which was 617 mg/g. According to Figure 2, higher H₂SO₄ concentration used at chemical activation results higher iodine adsorption capacity. Adsorption capability of activated carbon is stronger correspondingly with the increase of chemical activating agent concentration [20].

H₂SO₄ solution acts as dehydration agent, where H₂SO₄ solution releases water molecules when elimination of oxygen and hydrogen atoms occurs from carbon materials. This is caused further pores formation of the carbon. Higher H₂SO₄ concentration used means more ions from H₂SO₄ bind impurities that cover carbon pores and make carbon pores open.

Meanwhile, the availability of physical activation before chemical activation makes the iodine number was greater than chemical activated only carbon. This was because the nitrogen gas used in physical activation help to opens the pores of carbon that are still closed. Nitrogen gas stream acts to develop porous structures in carbon by removing tars and volatile substances on carbon during carbonization process. In addition, physical activation by inert gas stream such as nitrogen also helps to prevent occurrence of excessive oxidation due to presence of oxygen gas that damages carbon pore structures.

3.5.2 Surface area projection

The obtained iodine number results then used to project the surface area of activated carbons by using Equation 2. Figure 3 shows the effect of H₂SO₄ concentrations used at chemical activation on surface area of activated carbon.

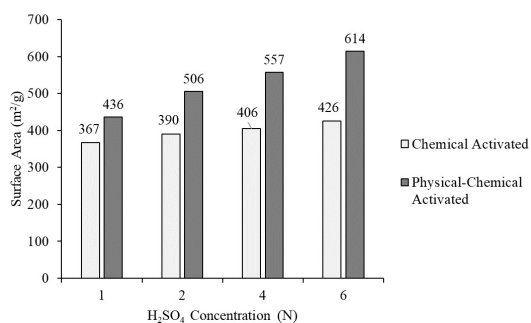


Fig. 3. Effect of H₂SO₄ concentration on surface area of activated carbon

The surface area of not yet activated carbon made from banana peel in this study was obtained at 360 m²/g. As can be seen in Figure 3, the chemical activation process with H₂SO₄ solution increases the surface area of the activated carbons. The increase in surface area was also proportional to the additional presence of physical activation and increased concentration of H₂SO₄ solution used as well as in the case of increased iodine numbers. This means the amount of iodine that can be adsorbed activated carbon on iodine number can represent how much activated carbon adsorption capability in the form of surface area data.

Based on Figure 3, it was also known the type of activated carbon that had the best surface area based on activation method and H₂SO₄ concentration used. The best type of activated carbon from the chemical activation only method was with the use of H₂SO₄ 6 N, which was 426 m²/g. Meanwhile, the best type of activated carbon from the combined physical-chemical activation method was with the use of H₂SO₄ 6 N, which was 614 m²/g.

Recently, there is a study of pollutants removal in diesel engines by using activated carbon [21]. In that study, some of activated carbons studied had surface area about 450-500 m²/g and can efficiently remove about 45-48% of pollutants in exhaust gases from diesel engines. The activated carbons produced in this study had higher and lower surface area than activated carbons tested from that study. Therefore, the activated carbons made in this study could also reduce pollutant gases content around that much and are well prepared to be used as adsorbent for motor vehicle exhaust emission.

4 Conclusion

It can be concluded from this study that activated carbons made from banana peel are well prepared to be used as adsorbent for motor vehicle exhaust emission. The greatest activated carbon surface area based on chemical concentration used in chemical only activation method was obtained on the use of H₂SO₄ 6 N, which was 426 m²/g. Therefore, the increase of H₂SO₄ concentration used at chemical activation process is increasing the surface area of activated carbon. However, the activated carbon surface area was greater in physical-chemical activation method than the chemical only activation on the same concentration,

which was 614 m²/g. It makes that activated carbon had the greatest surface area in this study. It shows physical-chemical activation method was better than chemical only activation method. The additional physical activation method in the making process of activated carbon helps to remove impurities that cover carbon pores and increase the activated carbon surface area.

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