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TECHNOLOGY DEVELOPMENT OF ADSORPTION CIGARETTE SMOKE USING MODIFIED ACTIVATED CARBON WITH MgO FROM WASTE BIOMASS OF DURIAN SHELL

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ABSTRACT

Activated carbon is one solution to overcome the problem of cigarette smoke which is very dangerous for health where *durian* shell waste is chosen as the base material. *Durian* shell waste was chosen because it contains high cellulose, lignin, and starch. Its production also reaches 746,805 thousand tons per year. *Durian* shell waste that had been activated by chemical activation with K_2CO_3 versus activated carbon is 1:1, 3:2, and 2:1, and by physical activation with N_2 was then modified with MgO with variations in concentrations of 0.5%, 1%, and 2% at 450^oC for 30 minutes. Activated carbon and modified activated carbon were then characterized by the Iod Number test, BET test, SEM test, and EDX test. The best non-modified activated carbon was the carbon which was chemical and physically activated at ratio 3:2 with results of 41.56% in yield, 399.44 mg/g in iodine numbers, and 694.13 m^2/g in surface area. While the best modified activated carbon was at MgO concentration of 2% with a yield of 97%, an iodine number of 625.70 mg/g, and a surface area of 1,029.90 m^2/g. The CO gas adsorption application, which is the component with the largest contribution in cigarette smoke, and cigarette smoke itself were tested using modified activated carbon. The results showed that 2% modified activated carbon was the best type of activated carbon to degrade CO in 3.89%/gram per minute with an adsorption ability of 0.215%. This activated carbon was also able to purify the air from the cigarette in 8.04%/gr activated carbon per minute with an adsorption ability of 0.87%.

Keywords: Activated carbon; Adsorption; Cigarette smoke; CO; Durian shell; MgO

1. INTRODUCTION

Cigarette smoke is a real threat to human life, from children to adults, as more than seven million people die each year due to exposure. The number consists of six million people who are active smokers and 890,000 people who are passive smokers (Yuliusman et al., 2017). Cigarette smoke consists of at least 250 harmful and deadly substances with 69 cancer-causing substances such as acetaldehyde, aromatic amine, arsenic, benzene, beryllium (toxic metal), 1,2-butadiene (dangerous gas), cadmium (toxic metal), polonium-210 (radioactive chemical elements), polycyclic aromatic hydrocarbons (PAHs), vinyl chloride, and more. Among these substances, the most dangerous and prevalent are pyridine, nicotine, tar, acetaldehyde, and carbon monoxide. These substances can cause chronic bronchitis, emphysema, constriction of blood vessels, pneumonia, and cancer (Yuliusman et al., 2015).

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In overcoming the problem of cigarette smoke, an adsorber becomes an effective substance to adsorb cigarette smoke. One of the adsorbers that has good adsorption ability is activated carbon. Activated carbon is a solid that results from heating it at high temperatures, which are maintained so that carbon does not experience oxidation and 85–95% pore carbon can be obtained. The source of the active carbon raw material is biomass which has a high percentage of cellulose, lignin, and starch content. One potential biomass waste that can be processed into activated carbon is *durian* shell because it has a cellulose element of around 50–60%, 5% lignin, and 5% starch (Yuliusman et al., 2018). *Durian* shell has a percentage of 60–75% of *durian* fruit (Yuliusman et al., 2017). The amount makes the potential of *durian* shell waste reach 597,444 thousand to 746,805 thousand tons based on 2015 *durian* fruit productions of 995.74 thousand tons (Tham et al., 2010).

The activated carbon from *durian* shell is chemically activated using K_2CO_3 and physically activated using N_2 in the reactor. K_2CO_3 was chosen because it is more environmentally friendly compared to other activators. Other than that, K_2CO_3 is a mineral. With the use of mineral material as an activator, the required activation time is relatively short so that more activated carbon is produced and the adsorption power of an adsorbate will be better. The presence of nitrogen gas flow in a physical activation prevents the presence of oxygen gas around the carbon which has the potential to rot excess carbon and damage the carbon's structure. It also removes all hydrocarbons and most of it remains as carbon. A carbon structure that is more damaged causes the structure to become more fragile and lighter than carbon from the carbonization process, which is still densely structured or less damaged. Adsorption on activated carbon can be extended back to the active surface with the addition of metal oxides, one of which is MgO. By using metal oxide, activated carbon has an active surface area of between 300–3,500 m^2/grams with an adsorption capacity of 25–100% for the weight of activated carbon (Hanafi, 2017).

The adsorption testing that was carried out refers to the study of Ibadurrahman (Ismail et al., 2010) who used a beam box measuring 20 cm × 10 cm × 18 cm made of acrylic glass as a representation of the state of space for cigarette smoke adsorption media. The test variables used were activated carbon, TiO_2, and modified TiO_2 activated carbon against pure CO gas pollutants, cigarette smoke, 10% metaldehyde, 37% methanol solution, and acetaldehyde. Based on the simplicity of the tool and the results of his research, pollutants could degrade up to 75–90% in 10 minutes. Looking at its highest potential, the research could be considered as a basis for conducting this study.

2. METHODS

This study has used some materials to finally complete the following methods with the below explanations.

2.1. Materials

The raw material that was used to make activated carbon was *durian* shell waste which was chemically activated by potassium carbonate (K_2CO_3) and physically activated by N_2. Magnesium oxide was used as a chemical for modifying the activated carbon. The concentration of MgO that was used is 0.5%, 1%, and 2% in 50 ml for 4-gram activated carbon. Carbon monoxide and cigarette smoke were used for an adsorption application with 600 ml volume in each trial.

2.2. Methods of Modifying Activated Carbon

The activated carbon was activated physically and chemically, then modified with MgO. Activated carbon that was modified with variations of MgO solution are 0.5%, 1%, and 2%. Four grams of activated carbon were added in 50 ml solution of each variation and stirred for about one hour. Then they were put in the furnace to impregnate the Mg inside activated carbon for

about 30 minutes at 450^{o}C.

2.3. Characterizations of Activated Carbon

Before doing an iodine number test, the activated carbon was dried in the oven at about 120^{o}C for one hour. This treatment was needed to make sure the activated carbon was dry and in the optimal condition. The 0.5 grams of activated carbon was then added to a 5 ml HCl and 25 ml iodine solution and was heated with a hotplate until it boiled. That solution was then stirred for about one hour to make sure it was homogenous. The carbon that was mixed by iodine solution was then filtrated using filter paper. The filtrated solution was then titrated by sodium thiosulfate ($Na_2S_2O_3$). The filtrate refers to iodine solution that is not adsorbed by activated carbon. During titration, when the color of solution became a pale yellow-color, a small amount of starch solution was added to clarify the endpoint of titration, which had a transparent color.

Aside from an iodine number test, an SEM (Scanning Electron Microscopy) test was also used to identify the activated carbon. SEM is a test that provides detailed high-resolution images of the activated carbon by rastering a focused electron beam across the surface and detecting a secondary or backscattered electron signal. The result from this test would indicate how clear and wide the surface of the activated carbon is. A clearer and wider magnification of the activated carbon indicates a better use in an adsorption application. The BET test (Brummer-Emmet-Teller) is also used to identify the surface area of an activated carbon. An Energy Dispersive X-Ray (EDX) test is also used to determine the elements that are contained in activated carbon (Armstrong et al., 2014).

2.4. Adsorption Test of Pure Carbon Monoxide Gas and Cigarette Smoke

After doing a characterization, the best result of activated carbon which had been chemically and physically activated is 3:2 (K_2CO_3: activated carbon). This variation had been modified by MgO and then been used in an adsorption test of pure CO gas and cigarette smoke by injecting the gas into a 600 ml adsorption box measuring 20×15×18 cm^3. The test that the gas contained in the adsorption box had been measured with GC-TCD as a quantitative test and GC-MS as a qualitative test (Tan et al., 2008).

3. RESULTS AND DISCUSSION

3.1. Results of Modified Activated Carbon

Activated carbon was produced by chemical and physical activation processes, then modified using variations of MgO 0.5%, 1%, and 2%. This variation was done to see the optimum condition of carbon obtained after undergoing modification. Modifications with MgO are useful as a co-impregnation of carbon, increasing the carbon's surface area, and strengthening the carbon's structure.

The homogeneous process aimed to allow MgO molecules to insert and fill up the space between the layers of the activated carbon structure. The inserted MgO could form a new activated carbon structure because it reacted with the remaining carbon and impurities resulting from the activation of physical chemistry. The reaction that occurred was characterized by the heat generated when mixing the active carbon with MgO (Cooper, 1994).

The carbon which was inserted by MgO was then heated at 450^{o}C for 30 minutes using a furnace. This heating process aimed to reactivate the carbon adsorption media, to stabilize the structure of the activated carbon layer, and to remove the water content and adhering impurities. MgO as a co-impregnation could form a new structure of activated carbon that was not yet solid since it was warmed up and caused the newly formed structure to become solid. The reaction that occurred between MgO and carbon was oxidation. The entire reaction was represented in Equation 1 as follows:

$$MgO + C \rightarrow Mg + CO \quad (1)$$

Based on the yield results obtained in Figure 1, the modification with the largest yield was in the 3:2 activation variation with a 0.5% MgO concentration and a value of 116%. This shows that under these conditions, a lot of Mg metal was inserted inside the carbon pores, making the carbon mass heavier than the initial mass. At large concentrations there is a decrease in yield because the carbon structure is not suitable and the Mg does not act as a binder but as a pore scraper so that the mass becomes smaller (Tan et al., 2008).

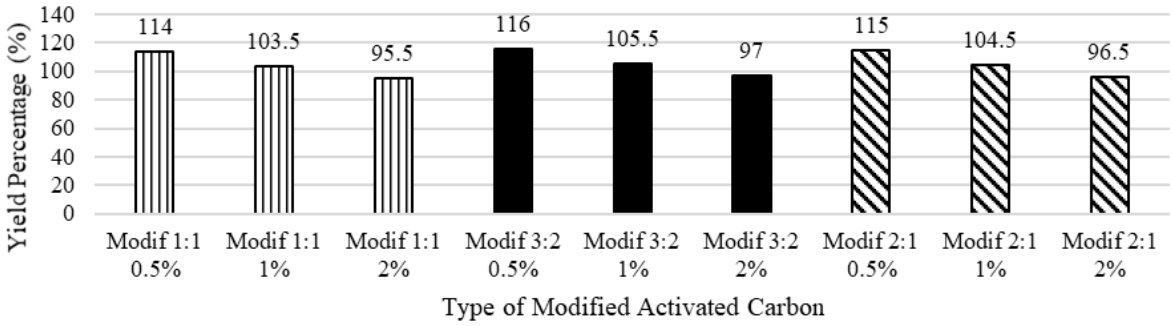

Figure 1 Yield of each modified activated carbon

3.2. Characterizations of Activated Carbon

The SEM test was carried out at the Bandung Institute of Technology laboratory with the JSM 6510 LA JEOL instrument for non-modified activated carbon samples as shown in Figure 2, as well as at the Jakarta State University laboratory with the SNE-4500M instrument for modified activated carbon samples as shown in Figure 3.

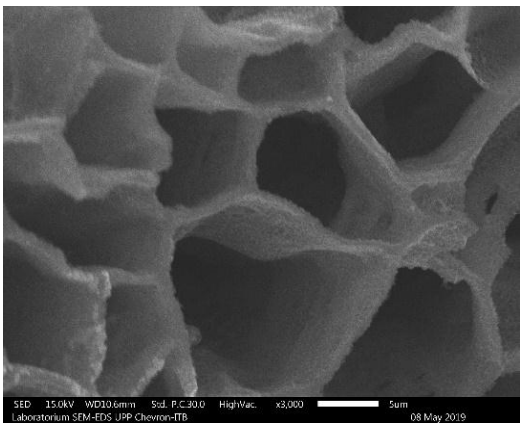

Figure 2 SEM result of non-modified activated carbon

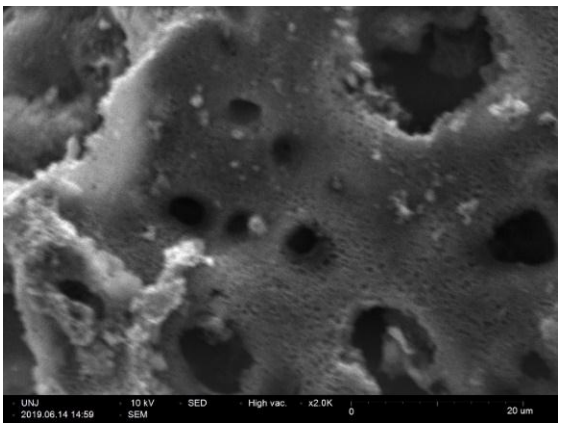

Figure 3 SEM result of modified activated carbon with 2% MgO

From Figure 2, it can be seen that the best type of non-modified activated carbon is chemical activation 3:2 because it has wider pores. This is because K_2CO_3 effectively erodes the impurities and breaks the hydrocarbon bonds. The amorphous structure is also clearer so that it can be useful in adsorption applications. Based on the results in Figure 2 and Figure 3, there are differences in the methods that lead to different results. In Figure 3, a magnification of 2,000 is carried out, while in Figure 3 it is 3,000. This difference in magnification is significant enough to influence the clarity of the amorphous structure. However, if examined more clearly, in Figure 3 there are many small pores that are bound so that the structure looks denser and stronger than carbon that is simply activated. This is because the Mg metal is inserted or meets the pore structure of the carbon so that the carbon structure looks strong with large diameter pores.

The EDX test aims to determine the composition of activated carbon. Table 1 shows the results of the EDX test of non-modified active carbon and modified activated carbon.

Table 1 EDX results of type of activated carbons

Elements	AC carbonized (%)	AC che-phys 1:1 (%)	AC che-phys 3:2 (%)	AC che-phys 2:1 (%)	AC modif 3:2 MgO 2% (%)
C	80.12	94.15	95.86	95.50	52.06
O	3.38	0.99	0.52	1.63	2.97
K	16.50	4.86	2.53	-	0.81
Cl	-	-	1.08	-	-
Mg	-	-	-	0.81	0.61
Si	-	-	-	0.61	-
Fe	-	-	-	1.41	-
B	-	-	-	-	43.86

From Table 1, it can be seen that the most dominant element in activated carbon in this study is carbon. The average carbon content contained in activated carbon in this study is 80–96%. This value is very high considering the Indonesia National Standard (SNI) requirements for activated carbon are a minimum of 65% of the total weight (Foo & Hameed, 2014). This shows that durian skin is potentially used as an activated carbon because it contains many carbon atoms.

Based on these results, it can also be seen that carbon from the chemical activation of 3:2 has a higher percentage of carbon than others. A high carbon content indicates that there will be more potential for the adsorbate to be bound to carbon/adsorbents. The carbon modification found that Boron is detected because it is present in the plant cell walls in the $B(OH)_3$ form and the activated carbon that is processed comes from a durian skin biomass, so this occurrence is very likely (Le Van & Luong Thi, 2014).

The adsorption power of iodized carbon has a correlation with the surface area of activated carbon. The greater the amount of iodine, the greater the ability of activated carbon to adsorb iodine (Iod). Iod is likened to an adsorbate for activated carbon, which is an adsorbent. Thus, the greater the value of the Iod amount, the greater the adsorption power of the adsorbent. The Iod amount is also closely related to the surface area where the greater the amount of Iod, the greater the surface area of activated carbon and activated carbon adsorption.

This relates to the BET test in determining the surface area of activated carbon. The greater the Iod amount, the greater the ability of activated carbon to adsorb iodine. The results of non-modified activated carbon that have the largest surface area are chemically 3:2 with 649.13 m^2/g. While in Figure 4, modified activated carbon reflects the best chemical and physical variation of 3:2 MgO 2%.

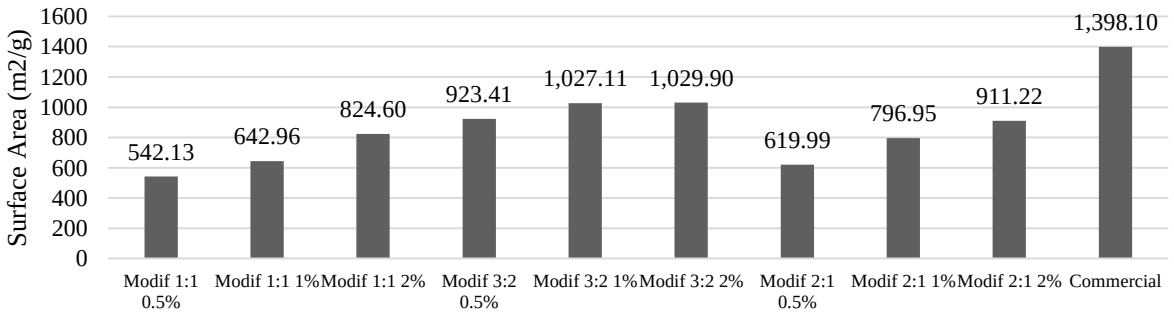

Figure 4 Surface area of modified activated carbon

The presence of MgO or Mg metal can cause carbon porosity to decrease. The distances between the pores will be greater as a result of the expansion of the pore of activated carbon and also the

opening of the pores closed by impurities. As a result, the number of pores produced will continue to increase so that more Iod is absorbed and the resulting Iod amount will be high. With the high Iod rate, surface area also increases.

3.3. Adsorption Test of Pure Carbon Monoxide Gas and Cigarette Smoke

The adsorption test was carried out using a 20×15×18 cm^3 adsorption box in a batch system. This box contained two trays with one gram of modified activated carbon in each. The amount of one gram of activated carbon was chosen so that the activated carbon would be spread evenly in the monolayer without overlap. If the amount was excessive, it was feared that the activated carbon that was accumulated would be unable to absorb the maximum amount of gas due to being covered by other carbons. Furthermore, 600 ml of gas was injected using a syringe. If simply calculated by using the ideal gas formulation with the assumption that the gas is at the same temperature and the number of moles gas entering is the same, then the pressure in the box obtained was around 1.1 atm. The pressure was not too different from the ambient condition. This was also adjusted to be the same as a realistic scenario containing a room full of cigarette smoke. However, the box space had to be pressurized as it is one of the factors that can cause adsorption (Linares-Solano et al., 2012).

One of the biggest components of cigarette smoke is CO gas. Therefore, pure CO is correlated with this experiment. Based on Figure 5, using GC-TCD as a quantitative instrument, modified activated carbon 3:2 MgO 0.5% could, on average, degrade the CO concentration per minute by 0.44% with a mass increase of 0.33 grams so that the adsorption capacity per minute was 1.33%/gram with an adsorption power of 0.169%. In the variation of 3:2 MgO 1%, it was found that the change in CO concentration per minute was an average of 0.5% with a mass increase of 0.24 grams so that the adsorption capacity per minute was 2.1%/gram with an adsorption power of 0.198%. Whereas on active carbon 3:2 MgO 2% it was found that the change in CO concentration per minute was an average of 0.55% with a mass increase of 0.14 grams with the ability to adsorb 3.89% per minute and the adsorption power was 0.215%.

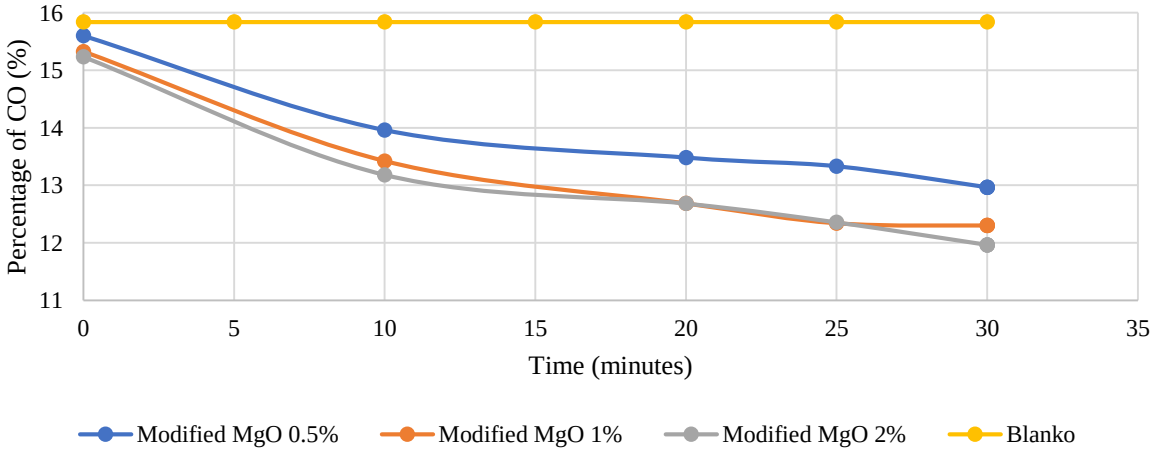

Figure 5 Adsorption of pure carbon monoxide

A slight increase in the mass indicates that the pore size of the activated carbon modification of 2% is much larger than the others, since CO is able to bind a lot but does not cause the final mass of carbon to increase. This shows that the greater the concentration of used modified compounds correlates with a greater absorption capacity. This is because the surface area is also getting bigger so that more adsorbates can be bound to the adsorbent pore.

In the air purification test of cigarette smoke adsorption, air concentration and cigarette smoke testing were carried out in GC-MS. The GC-MS data shows that the combined compounds that were detected in high amounts are alanine, carbon dioxide, oxalic acid, formic acid, and ethynyl

ester in percentages of 84.73%. In the tested samples of cigarette smoke, tar and nicotine should have been detected, but were unable to be with GC-MS. This is because of the lag of tar in the gas sampling bag since tar and nicotine will quickly condense at room temperature. Carbon monoxide was also not able to be detected because the reaction between CO and hydroxyl ions (HO*) in ambient air produces carbon dioxide (CO_2) and hydrogen atoms (H*) (Mopoung et al., 2015).

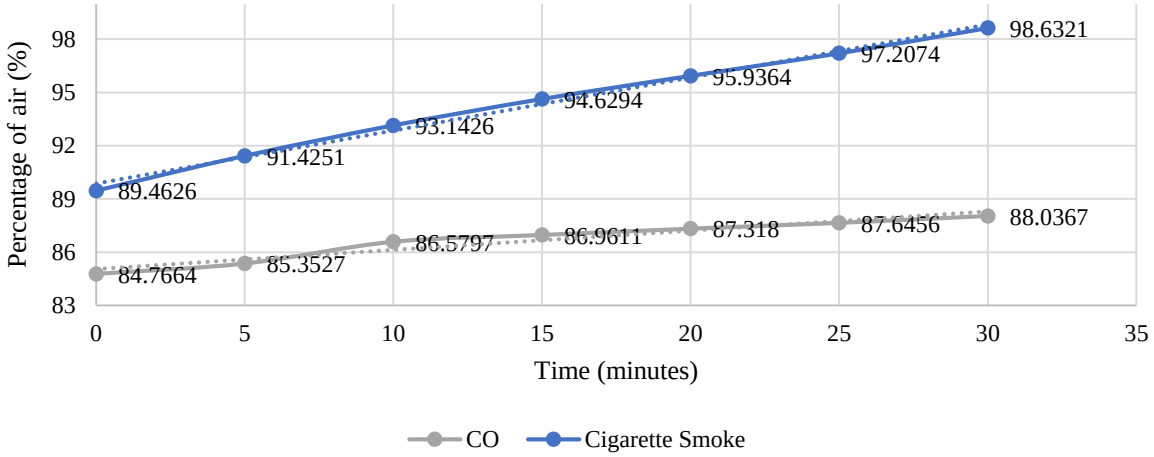

Figure 6 Comparison of air purity in cigarette smoke adsorption test and CO by using mod. carbon 2%

Because the GC-MS results did not produce conclusive quantitative data for each compound, a test using GC-TCD was conducted. Based on the results of the GC-MS, cigarette smoke has various compounds. As a result, the GC-TCD measurement focused on the purification of air from cigarette smoke instead of its degradation.

Based on Figure 6, it can be seen that activated carbon is able to purify the air more quickly in cigarette smoke with an ability of 1.53% per minute. This is because cigarette smoke has a variety of compounds so that the active side of the active pore is able to work optimally by adjusting the conditions of the active pore's side that is suitable for each compound.

Even after weighing the activated carbon, it was found that there was an increase in the activated carbon mass of 0.19 grams. This addition of 0.05 grams was heavier than the mass of activated carbon used to adsorb CO gas. This can occur because the types of compounds are also varied with different densities, so it is very possible that the mass of activated carbon will be heavier when adsorbing cigarette smoke. The ability of activated carbon at a 2% modification in absorbing cigarette smoke or purifying the air is 8.04%/gram per minute. Based on the formula for calculating an isothermal adsorption, the adsorption power is 0.87%.

4. CONCLUSION

The best non-modified activated carbon was the variation of 3:2 which had a yield of 41.56% with an Iod amount of 399.44 mg/g and a surface area of 694.13 m^2/g. The best modified activated carbon was activated carbon with a concentration of MgO of 2% which had a yield of 97% with an Iod amount of 625.70 mg/g and a surface area of 1,029.90 m^2/g. Activated carbon with a modification of 2% had the ability to degrade the best CO with an adsorption rate of 3.89%/gram per minute with an adsorption power of 0.215%. Modified 2% activated carbon had the ability to purify the air from the best cigarette smoke with an adsorption rate of 8.04%/gram per minute with an adsorption power of 0.87%.

5. ACKNOWLEDGEMENT

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