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REMOVAL OF METHANAL FROM AQUEOUS SOLUTION USING MICROWAVE INDUCED CARBON FROM *Coffea arabica* GROUNDS WASTE

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ABSTRACT

Activated carbon is a prominent adsorbent for removing hazardous chemicals, and abundant natural resources allow the production of activated carbon utilizing various sources of biomass. This study aims to investigate the capacity of carbon prepared from *Coffea arabica* grounds waste in adsorbing methanal. The carbon from used coffee grounds was activated using $ZnCl_2$ 30% in a microwave at 800 Watt for 10 minutes. The characteristics of activated carbon investigated include moisture content (1.47%), ash content (4.74%), and iodine number (803.7 mg/g). The study of methanal adsorption was carried out under the following conditions: adsorbent mass (gram) (0.5, 1, and 1.5), contact time (minute) (20, 50, and 80), methanal concentration (ppm) (5, 10, and 15), and pH (2, 5 and 8). The optimum conditions for adsorption were studied using a central composite design (CCD), and the data were analyzed using Minitab 16 software. The adsorption was optimally achieved at 1.50 grams of adsorbent, 58.18 minutes of contact time, 10.15 ppm of methanal concentration, and pH of 2. The percent of adsorption of methanal at optimum conditions was 54.31%. The adsorption of methanal by activated carbon from *Coffea arabica* ground waste fit best with the Freundlich isotherm model ($R^2 = 0.9918$), indicating multilayer adsorption.

Keywords: Activated Carbon, Coffee Grounds Waste, Adsorption, Methanal (CH_2O), Central Composite Design (CCD), Microwave Activation.

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INTRODUCTION

Methanal (CH_2O), also known as formaldehyde, is an organic compound classified as a contaminant both in the air and water. The compound is widely used in various industries such as cosmetics, detergents, synthetic resin manufacturing, fertilizers, explosives, preservatives, disinfectants in hospitals and industries, household products, glues, paper product coatings, fibreboards, and plywood.[1,2] Besides having several benefits in the industrial sector, methanal waste also harms the environment and humans. This compound is one of the most harmful organic components found in industrial waste. Methanal was classified as a Group I carcinogen (proven to be a human carcinogen) in 2004 by the International Agency for Research on Cancer (IARC) which can cause nasopharyngeal cancer and possibly leukaemia. United States Environmental Protection Agency (USEPA) also classified methanal as a toxic organic compound listed among 45 organic substances that raise concerns about environmental impacts. Therefore, it is necessary to minimize the impact caused by methanal as one of the pollutants in water.

Controlling chemical pollutants can be carried out in various ways, including recycling, reducing chemicals, or removing pollutants by adsorption with suitable adsorbents. Activated carbon (AC) is a commonly used material to adsorb chemical waste or pollutants. Currently, the use of natural waste (natural biowastes) as precursors to synthesizing carbon materials has been widely studied due to cost efficiency[3], renewability, and environmentally friendly. Various biomass from plants have been used as raw materials to produce AC including coconut shells[4], bamboo[5], and coffee grounds.[6-8]

Coffee is considered the most consumed beverage worldwide with more than 5 million tons annually[9], and Indonesia is among the top 4 coffee-producing countries. According to the United States Department of Agriculture (USDA), Indonesia is the fourth largest coffee-producing country after Brazil, Vietnam, and Colombia. Moreover, In Indonesia, several most prominent coffee-producing provinces including Aceh province, the fourth largest coffee-producing province after South Sumatra, Lampung, and North Sumatra,

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with a total production of 71,735 tons in 2020. No wonder why in Aceh province, there are more than tens of thousands of coffee shops available. Therefore, Aceh is nicknamed "The Land of a Thousand Coffee Stalls".[10,11]

The high consumption of coffee has resulted in many coffee grounds waste that Aceh produces every day. This coffee waste has not been widely used, so it is necessary to increase its added value through the use of coffee waste as a raw material for AC. However, there are very limited literatures that explores the adsorption of methanal in industrial waste using bio-sorbents from coffee grounds waste. However, previous research on methanal gas adsorption using bio sorbents from coffee grounds has been carried out by Boonamnuayvitaya. The bio-sorbent from coffee grounds was activated by using $ZnCl_2$ with a carbonation temperature of 600℃ resulting in the methanal adsorption capacity of 245 mg/g.[12] Furthermore, Laowachirasuwan also prepared AC from coffee grounds using several chemical activators ($ZnCl_2$, KOH, NaOH, H_3PO_4, and H_2SO_4), and the largest pore diameter was obtained from AC using $ZnCl_2$.[13]

Apart from chemical activation, carbon can also be activated physically or both. Combined activation usually generates AC with better adsorption capacity.[3,14,15] Physical activation aims to vaporize impurities and allow the pore to open wider due to heating. Heating usually requires more energy, so activation using a microwave could be an alternative. By using a microwave, we can save quite a lot of energy because the process only takes 10 minutes at most. Liu et al. prepared AC from starch that activated chemically and physically using KOH and microwave and obtained a larger BET surface area compared to the AC without microwave heating.[16] The preparation of coffee waste as AC that activated using $ZnCl_2$ and microwave has never been reported. Therefore, in this paper, we reported the preparation of coffee waste bio-sorbent activated using $ZnCl_2$ and microwave and its adsorption capacity against methanal.

EXPERIMENTAL

Preparation of AC

2 kg of collected coffee waste was washed using hot distilled water and filtered.[17] Furthermore, the coffee waste was dehydrated in an oven (Memmert, Germany) at 105℃ for five hours.[18,19] The dehydrated coffee waste was then carbonized using a muffle furnace (Barnstead Thermolyne, Type FB1400) at a temperature of 500℃ for 30 minutes.[7] After carbonization was finished, the charcoal was mashed and sieved using a 100 mesh sieve. The charcoal was subsequently activated using 30% $ZnCl_2$ (Merck, Germany) with a ratio of 3:1; activator: charcoal in a microwave (Rewez, Japan) with a power of 800 Watt for ten minutes.[10,13,16,20] The AC was then washed with hot distilled water (80℃) for 20 minutes, followed by washing using 0.1 N HCl (Sigma-Aldrich, Singapore) for 20 minutes and then was washed again using warm distilled water until a constant pH was obtained. The samples were re-dried using an oven at 105ºC for 4 hours until a constant weight was obtained.

Characterization of AC

The characteristics of AC including the physical appearance, water content, ash content, and adsorption capacity toward iodine were investigated. The topography, morphology, and chemical compositions of the AC were investigated using SEM-EDS (Shimadzu, Japan).

Moisture Content

Moisture content was investigated based on the procedure developed by Irmanto & Suyata.[21] 2 grams of AC was precisely weighted and heated at 105℃ for 3 hours and reweighted. The procedure was repeated until a constant weight was obtained. The moisture content was computed by using the following equation:[21,22]

$$\text{Water Content (\%)} = \frac{\text{Initial Weight of AC (g)} - \text{Final Weight of AC (g)}}{\text{Initial Weight of AC (g)}} \text{ x } 100\%$$

Ash Content

The ash content of AC was determined according to the work of Irmanto & Suyata.[21] 2 grams of AC was precisely weighed and placed in a porcelain tube (previously weighted and recorded). The AC was then heated for 4 hours in a 600℃ set *muffle furnace*, transferred to a desiccator, and the final weight was

determined. The procedure was replicated three times, and the ash content was determined using the following equation:[22]

$$\text{Ash Content (\%)} = \frac{\text{Final Weight of Ash (g)}}{\text{Initial Aeight of ash (g)}} \text{ x } 100\%$$

Iodine Number

The iodine adsorption of AC from the coffee ground was determined as follows: 0.5 gram of AC was added to a dark Erlenmeyer, and 50 mL of 0.1 N iodine solution was added, the Erlenmeyer was closed and shaken for 15 minutes. The solution was then filtered, 10 ml aliquot of the filtrate was poured into the Erlenmeyer and was titrated with $Na_2S_2O_3$ solution until the color changes to light yellow. After the color change occurs, 1 mL of the starch indicator was added to the filtrate and titrated again until the blue color disappears.[21] The procedure was repeated three times, and the iodine adsorption was calculated using the following formula:

$$\text{Adsorption Capacity} = \frac{\text{V } I_2 \text{ (mL)} - \frac{\text{V } Na_2S_2O_3 \text{ (mL) x Normality of } Na_2S_2O_3}{\text{Normality } Na_2S_2O_3}}{\text{Weight of AC}} \text{ x } 129.6 \text{ x df}$$

Solutions Preparation

A 1000 ppm methanal stock solution was prepared by dissolving 0.25 mL of 37% methanal in a 100 mL volumetric flask, and ethanol p.a (Merck, Germany) was added to the limit mark. Serial dilutions were performed to achieve the following concentrations 5, 10, 15, 20, and 25 mg L^{-1}. Whereas Nash reagent was prepared by dissolving 15 g of ammonium acetate (Sigma-Aldrich, Singapore), 0.3 mL acetic acid (Merck, Germany), and 0.2 mL acetylacetone (Merck, Germany) in a 100 mL volumetric flask, the volume was adjusted by adding aquadest.[23]

Study of Optimum Conditions for Methanal Adsorption

In this study, instead of the classical method, determination of the optimum conditions for methanal adsorption was carried out using the Response Surface Methodology with Central Composite Design (CCD) with statistical analysis. The optimum conditions were determined based on four independent variables: weight, contact time, adsorbate concentration, and adsorbate pH. Each variable was coded with X_1, X_2, X_3, and X_4, respectively. Furthermore, each variable was varied into three levels with the code -1 (lower level), 0 (center point), and 1 (upper level), as shown in Table-1. The number of runs is determined using the equation:

$$N = 2^n + 2n + n_c$$

Where: N = number of runs, n = number of factors, and n_c = number of central points.

The experiment was carried out by pouring 50 mL of methanal into 100 mL Erlenmeyer, and the bio-sorbent (each according to the concentration, mass, and pH as previously determined) was added to the Erlenmeyer. The pH was adjusted using 0.1 M HCl and 0.1 M NaOH[24]. The adsorption process was carried out at ambient temperature[25], closed, and with a stirring speed of 350 rpm[26] with a predetermined duration of contact time. All measurements of the optimum conditions were obtained based on analysis using UV-Vis mini 1240 spectrophotometer (Shimadzu, Kyoto, Japan).

Table-1: CCD Level Variables and Codes of Determining the Optimum Conditions for Methanal Adsorption

Level Code	Weight (g)	Contact Time (minute)	Concentration (mg L^{-1})	pH
	X_1	X_2	X_3	X_4
-1	0.5	20	5	2
0	1.0	50	10	5
1	1.5	80	15	8

The percentage of methanal adsorption for each run was calculated using the formula:

$$\%\ R_t = \frac{C_0 - C_t}{C_0} \text{ x } 100\%$$

Where, % R_t = percentage of adsorption, C_o = initial concentration (mg L^{-1}) , and C_t = final concentration (mg L^{-1})

The optimum conditions for each variable to obtain the highest predictive value for methanal adsorption is determined using the following equation[27]:

$$Y = b_0 + b_1X_1 + b_2X_2 + b_3X_3 + b_4X_4 + b_{12}X_1X_2 + b_{13}X_1X_3 + b_{14}X_1X_4 + b_{23}X_2X_3 + b_{24}X_2X_4 + b_{34}X_3X_4 + b_{11}X_1^2 + b_{22}X_2^2 + b_{33}X_3^2 + b_{44}X_4^2$$

Where, Y = predictive response of adsorption (%), b = regression coefficient, X = level code of each variable

Langmuir Isotherm and Freundlich Isotherm

The determination of the adsorption isotherm was carried out at the optimum conditions obtained by varying the methanal concentration. After the adsorption process was carried out, the absorbance of the solution was measured using UV-Vis. The Langmuir isotherm curve is obtained by plotting Ce (equilibrium concentration) against Ce/Qe (equilibrium concentration/amount of substance adsorbed per gram of bio-sorbent). Meanwhile, the Freundlich isotherm curve is obtained by plotting ln Ce against ln Qe.

RESULTS AND DISCUSSION

Characteristics of AC

Characterization of the AC includes yield, moisture content, ash content, iodine adsorption capacity, and SEM-EDS. SNI 06-3730-1995 was used as standard, and the results of characterization are shown in Table-2.

Table-2: Characteristics of AC according to Indonesian Standard (SNI 06-3730-1995)

Parameters	Values	
	Carbon	Activated Carbon
Yield (%)	21.84	19.42
Moisture content (%) (Max. 15)	5.80 ± 3.42	1.47 ± 0.31
Ash content (%) (Max. 10)	2.50 ± 0.70	4.73 ± 3.09
Iodine number (mg/g) (Min.750)	729.67 ± 51.81	803.70 ± 14.95

The yield of charcoal after activation was found to be lesser compared to the yield before the activation since the yield obtained is influenced by the amount of raw material and heating temperature both during carbonization and activation. Continued heating at the activation stage using a microwave and re-drying using an oven also causes decreasing yield.

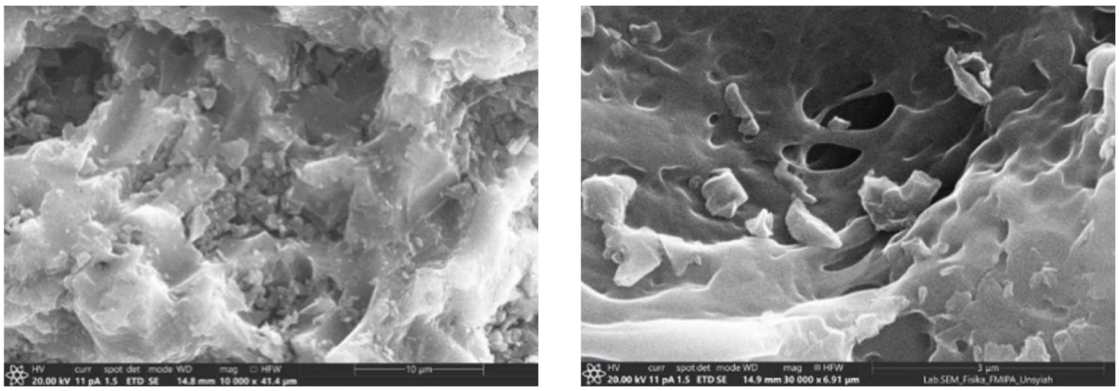
The moisture content test was carried out to determine the water content bound in the AC. The lower the water content, the lesser water that could cover the pores of the AC so that the adsorption can take place more optimally. The decrease of water content in the charcoal after activation was associated with the activation stage using microwave and $ZnCl_2$. Most water molecules were easily being removed during the microwave activating process due to their polarity.

Ash content test was carried out to determine the remaining mineral contained in the AC. The remaining content of this mineral can clog the pores of activated charcoal, so the lower the ash content in the AC, the greater the absorption capacity of activated charcoal would be occurred.[22]

The iodine adsorption test was carried out to determine the ability of activated charcoal to uptake small molecules. The size of the iodine adsorption capacity of AC indicates the number of micropores formed, so the more significant the iodine adsorption, the greater the number of micropores, and the larger the surface area.[22] The surface of both AC and charcoal before activation is depicted in Fig-1.

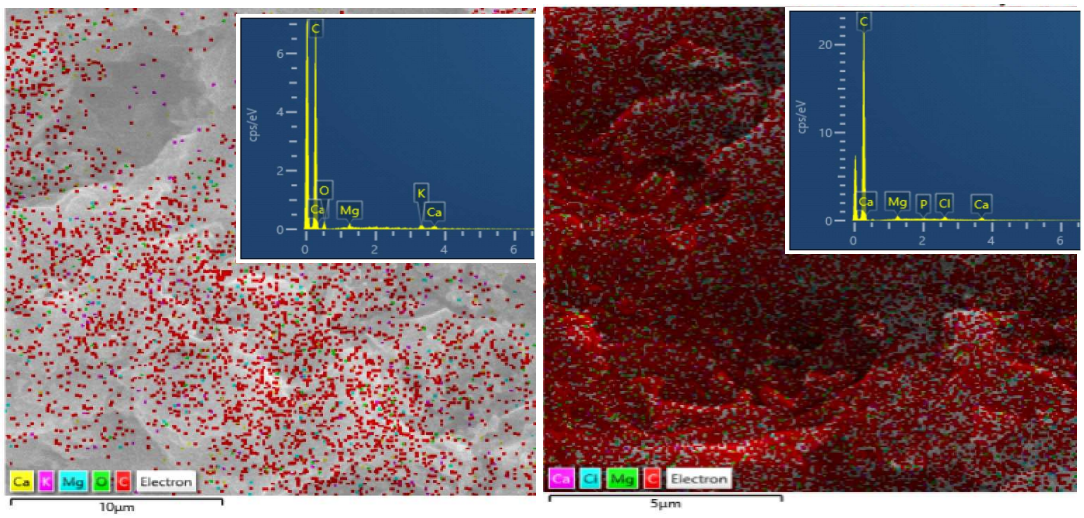
The morphology and surface of the AC were investigated before and after activation using SEM-EDS, and the micrographs are shown in Fig.-1. Based on the micrograph, the carbon before activation has an irregular surface, and the pore formed is not visible, while the carbon after activation has a higher number of pores and a more visible pore structure. The differences in carbon content before and after activation can also be observed from the results of EDS analysis in Fig.-2, Fig.-3. and Table-3. Based on these data, carbon from *Coffea arabica* grounds waste has significantly high carbon content both before and after activation, respectively 90.6% and 99.2%. After activation, carbon content has increased due to the activation process

with $ZnCl_2$ using a microwave. The activation was able to open the pores by removing unwanted atoms like oxygen and calcium.



a b

Fig.-1: SEM Micrographs of Carbon prepared from *Coffea arabica* grounds waste. (a) Before Activation and (b) After Activation



(a) (b)

Fig.-2: SEM-EDS MAP, Carbon Content and Elemental Distribution of Carbon prepared from *Coffea arabica* Grounds Waste. (a) Before Activation and (b) After Activation)

Table-3: Elemental Analysis results based on SEM-EDS Detection

Element	Atomic (%)	
	Before Activation	After Activation
C	90.6	99.2
O	8.7	-
Mg	0.3	0.3
K	0.2	-
Ca	0.2	0.2
Cl	-	0.2
P	-	0.1

Optimum Conditions for Methanal Adsorption

Determination of the optimum conditions for methanal adsorption was carried out using the CCD, and the data was analysed using Minitab 16 software. The CCD method was used to analyse the correlation

between variables and responses in order to determine the effect of each factor: weight (X_1), contact time (X_2), concentration (X_3), and pH (X_4) on the percentage of adsorption. The results of the experiment for determining the optimization of methanal adsorption using CCD are presented in Table-4.

Table-4: Experiment Results Determination of Optimization Using CCD

Run	Coded Variables				Variables				Adsorption (%)	
	X_1	X_2	X_3	X_4	Weight (g)	Contact time (minute)	Concentration (mg L^{-1})	pH	Experiment Value	Predicted Value
1	-1	-1	-1	-1	0.5	20	5	2	42.339	46.872
2	1	-1	-1	-1	1.5	20	5	2	48.772	40.984
3	-1	1	-1	-1	0.5	80	5	2	42.924	43.745
4	1	1	-1	-1	1.5	80	5	2	48.187	44.923
5	-1	-1	1	-1	0.5	20	15	2	26.979	32.706
6	1	-1	1	-1	1.5	20	15	2	36.725	36.612
7	-1	1	1	-1	0.5	80	15	2	43.158	39.081
8	1	1	1	-1	1.5	80	15	2	50.565	50.054
9	-1	-1	-1	1	0.5	20	5	8	39.415	36.002
10	1	-1	-1	1	1.5	20	5	8	21.871	28.408
11	-1	1	-1	1	0.5	80	5	8	24.795	27.368
12	1	1	-1	1	1.5	80	5	8	36.491	26.840
13	-1	-1	1	1	0.5	20	15	8	20.156	25.880
14	1	-1	1	1	1.5	20	15	8	32.827	28.081
15	-1	1	1	1	0.5	80	15	8	22.885	26.749
16	1	1	1	1	1.5	80	15	8	38.090	36.017
17	-1	0	0	0	0.5	50	10	5	66.491	50.739
18	1	0	0	0	1.5	50	10	5	30.819	52.428
19	0	-1	0	0	1	20	10	5	39.298	32.838
20	0	1	0	0	1	80	10	5	22.924	35.242
21	0	0	-1	0	1	50	5	5	22.456	32.110
22	0	0	1	0	1	50	15	5	33.411	29.615
23	0	0	0	-1	1	50	10	2	47.251	51.924
24	0	0	0	1	1	50	10	8	38.285	39.470
25	0	0	0	0	1	50	10	5	36.530	42.179
26	0	0	0	0	1	50	10	5	40.429	42.179
27	0	0	0	0	1	50	10	5	40.234	42.179
28	0	0	0	0	1	50	10	5	45.302	42.179
29	0	0	0	0	1	50	10	5	48.811	42.179
30	0	0	0	0	1	50	10	5	56.218	42.179
31	0	0	0	0	1	50	10	5	45.302	42.179

Table-5. Analysis of Varian (ANOVA) for Methanal Adsorption

Term	Coef.	SE Coef.	T	P-value
Constant	42.1792	3.066	13.755	0.000**
Weight	0.8447	2.436	0.347	0.733
Contact Time	1.2021	2.436	0.493	0.628
Concentration	-1.2476	2.436	-0.512	0.616
pH	-6.2270	2.436	-2.556	0.021*
Weight*Weight	9.4045	6.417	1.466	0.162
Contact Time*Contact Time	-8.1394	6.417	-1.268	0.223
Concentration*Concentration	-11.3168	6.417	-1.764	0.097
pH*pH	3.5176	6.417	0.548	0.591
Weight*Contact Time	1.7666	2.584	0.684	0.504
Weight*Concentration	2.4488	2.584	0.948	0.357
Weight*pH	-0.4264	2.584	-0.165	0.871

Contact Time*Concentration	2.3757	2.584	0.919	0.372
Contact Time*pH	-1.3767	2.584	-0.533	0.602
Concentration*pH	1.0112	2.584	0.391	0.701
Lack-of-Fit				0.073
S = 10.3370	PRESS = 7398.29			
R^2 = 52.34%	R^2 (predicted) = 0.00%		R^2 (adj) = 10.64%	

*P-*value* < 0.05 = significant

The percentage of experimental adsorption obtained was used for the analysis of variance (ANOVA), as presented in Table-5. The results show that all factors have a P-value higher than 0.05 except at pH (P-value = 0.021), so the overall model was considered not significant. This shows that weight, contact time, and concentration do not significantly affect the adsorption of methanal in the solution. However, based on the Lack-of-Fit test (regression model suitability test), the P-value is 0.073 (P-value > 0.05). This means that the second-order equation model in this study is significant and feasible to be used to predict the response to each variable.[10,28]

Based on Table-5, an analysis of the correlation between each factor and the percent adsorption of methanal was obtained. The regression equation obtained can be written as follows:

$$Y = 42.1792 + 0.8447*X_1 + 1.2021*X_2 – 1.2476*X_3 – 6.227*X_4 + 9.4045*X_1*X_1 – 8.1394*X_2*X_2 - 11.3168*X_3*X_3 + 3.5176*X_4*X_4 + 1.7666*X_1* X_2 + 2.4488*X_1*X_3 – 0.4264*X_1*X_4 + 2.3757*X_2*X_3 +1.3767*X_2*X_4 + 1.0112*X_3*X_4$$

Analysis of the correlation model resulted in a correlation value (R^2) of 0.5234. The R^2 value is not close to 1 but has reached 50% so that the feasibility of the resulting model from the data is low. The closer the R^2 value to 1, the better suitability to the experimental data.[10]

The relationship of each factor to the percent of adsorption is observed in the graph of surface plots and contour plots, as shown in Fig.-3. Surface plots are used to determine the three-dimensional relationship of the factors to the percent of adsorption, while contour plots are used to determine the two-dimensional relationship of the factors to the percent of adsorption. From both the surface and contour plots graphs, the optimum conditions for each variable were obtained.

The highest adsorption area was obtained for each interaction between the factors based on the contour plots graph. The intensity of the color density indicates the high and low percent of adsorption on the graph, where the darker color indicates the increasing percentage of adsorption. The interactions between contact time*weight, concentration*weight, concentration*contact time, pH*weight, pH*contact time, and pH*concentration showed similar values in the region of the highest percent adsorption. Based on the contour plots graph, it can be observed that the highest percent of adsorption area occurs in the contact time range of 40-70 minutes; weight 1.5 g; concentration of 7.5-12.5 mg L^{-1}; and pH 2. This result is coherent with the response optimization graph shown in Fig.-4.

The adsorption response optimization graph in Fig.-6. shows the optimum conditions obtained based on the prediction results for each factor: weight of 1.5 g; contact time of 58.18 minutes; concentration of 10.15 ppm; and pH of 2. Furthermore, the methanal adsorption process was carried out at optimum conditions to obtain the percent adsorption of methanal of the experimental results of 54.3%.

Adsorption Isotherms

The adsorption isotherm describes the relationship between the adsorbent (AC) and the adsorbate (methanal) in equilibrium. Two models of isotherms were used to examine the adsorption of methanal in this research as depicted by Fig.-5.

Based on the two isotherm curve figures, it can be observed that the experiment fits the Freundlich model concerning the value of the correlation coefficient (R^2) obtained. The isotherm with an R^2 value closer to 1 is the most suitable isotherm model for the experiment[29,30]. So, the isotherm model that best suits the adsorption process is the Freundlich isotherm with an R^2 value of 0.9918. This isotherm explains that the adsorption of methanal using AC prepared from *Coffea arabica* grounds waste occurs as a multilayer.

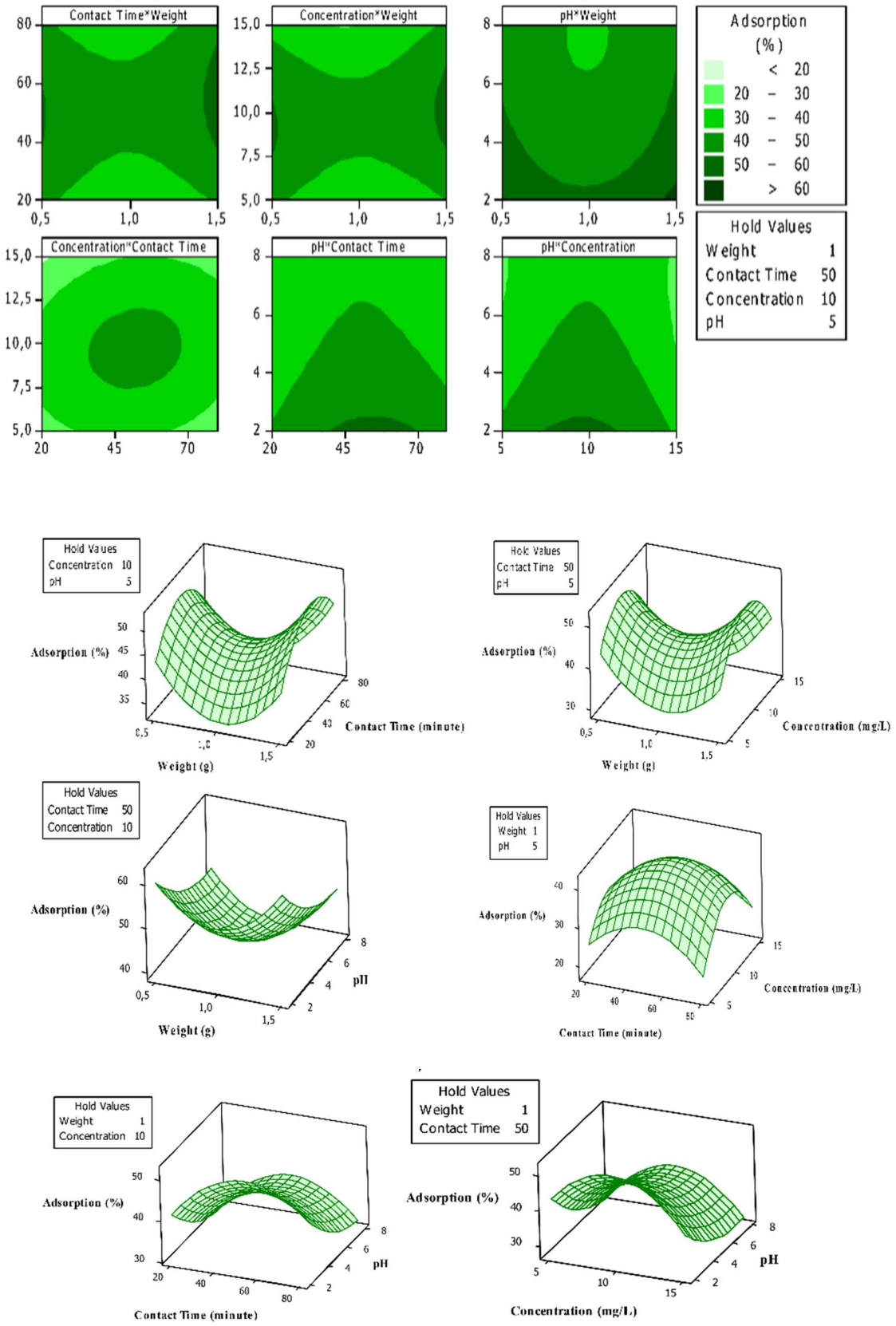


Fig.-3: Contour-plots and Surface-plots of Adsorption

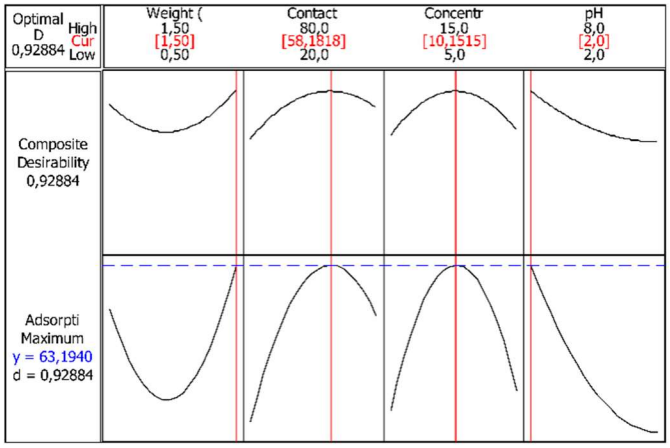


Fig.-4. Response Optimization Graph

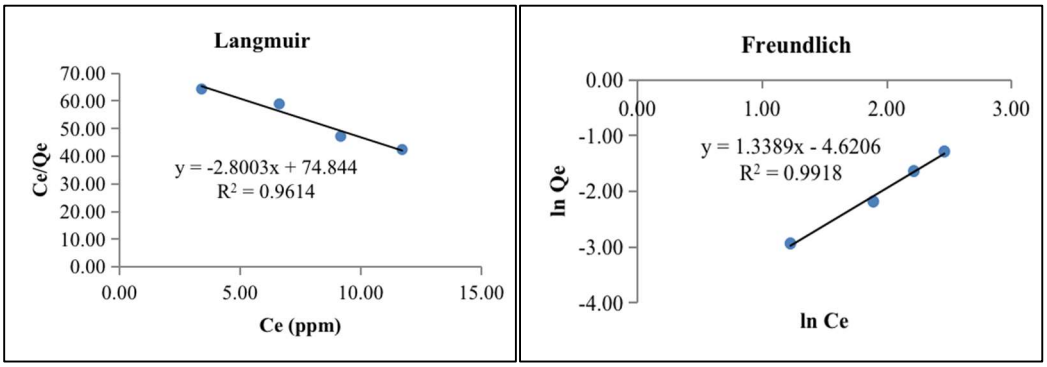


Fig.-5: Isotherm's Curve

CONCLUSION

Based on the research results, it was obtained that activated carbon from *Coffea arabica* grounds waste activated with $ZnCl_2$ using a microwave has a moisture content (1.47%), ash content (4.74%), and iodine adsorption (803.70 mg/g), which meets the standard of SNI 06-3730-1995. The adsorption of methanal was optimum at the weight of 1.5 g; 58.18 minutes of contact time; concentration of 10.15 mg L^{-1}; and pH=2. The adsorption percentage of methanal at optimum conditions was 54.31%. The factor that most influences the percent adsorption of methanal were the pH value of the environment. The methanal adsorption process fits the Freundlich isotherm model ($R^2 = 0.9918$), indicating that adsorption occurred in a multilayer mode.

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