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Research paper

Modelling of Adsorption Kinetic and Equilibrium Isotherms of Hydrogen Sulfide onto Hydrogel Biochar Adsorbent

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Abstract

Every process produces byproducts, and it can be dangerous or not. Hydrogen sulfide (H_2S) is one of the dangerous byproducts produced. In order to prevent hydrogen sulfide from harming workers or residents, a control system is required. Currently, all industries have a control system such as an adsorption system, to control dangerous component such as H_2S. In order to optimize the adsorption system, research is required. Researching using laboratory method is very dangerous due to the harmful nature of H_2S. Thus, isotherm and kinetic model is used as an alternative method, as it doesn't involve H_2S usage in laboratory. Each isotherm model and kinetic model equation has different uses for adsorption process. The main objective of this experiment is to find the most suitable equation for adsorption of hydrogen sulfide. The isotherm models used were Langmuir, Freundlich and Elovich, while kinetic models used were Pseudo-First Order and Pseudo-Second Order. Suitability of equation was determined by correlation coefficient (R^2). The higher the R^2, the more suitable the equation is to process. After the result obtained, comparison was done, comparing R^2 of each model. It was found that the most suitable isotherm and kinetic model were Elovich model and Pseudo-Second Order model respectively. R^2 for Elovich model was 0.9686; the highest correlation coefficient among isotherms, while R^2 for Pseudo-Second Order model was 0.9284, highest among kinetic models.

Keywords: *Adsorption, Elovich, Freundlich, Hydrogen sulfide; Isotherm model; Kinetic model; Langmuir; Pseudo-First Order; Pseudo-Second Order.*

1. Introduction

The most toxic gas is carbon monoxide while the second is hydrogen sulfide (H_2S). H_2S can cause inhalational deaths. It is dangerous as its mechanism is unknown to people. It can be produced naturally from organic matter that decays [1]. This production of H_2S can be seen in the Petroleum production and refining, sewer and wastewater treatment and Agricultural silos [2]. Workers are exposed to H_2S by inhaling the gas. Effects depend on how much and how long is the workers exposed to the toxic gas. By referring to Occupational Safety and Health Act (OSHA), symptoms can identify when workers exposed to high concentrations of H_2S. The worst scenario is that workers die by rapid unconsciousness and stop of breath [3].

Hydrogen sulfide can be very dangerous but industry can take preventive actions to protect their employees and their building. It can be corrosive toward metals and concrete, thus affecting the mechanical strength of equipment [4],[5]. Commercialized process that is being used in industry is adsorption using activated carbon. Atoms, ions or molecules from adsorbate that can be in gas, liquid or dissolved solid adhere to a surface of the adsorbent is adsorption process, and this process occurs due to differences in concentration. There are many parameters in improving the adsorption rate, such as temperature, condition of adsorbent and adsorbent condition [6].

Activated carbon is one of the adsorbent's examples. It is a carbonaceous, possesses special surface properties, highly porous adsorptive medium due to high surface area, and has a complex structure composed of carbon atoms [7]. The pores in activated carbon are linked together with each other by chemical bond, to create a rigid skeleton of disordered layers of carbon atoms. Coconut shell, peat, lignite coal and empty fruit bunch are examples types of activated carbons. Each contains different carbon and different result will be produced if used as an adsorbent.

Every experiment must be done in laboratory scale so that the damage or side effect is in minimal amount. The experiment can be scaled up to industry level by using adsorption isotherm and adsorption kinetic. The data from adsorption isotherm is used in designing adsorption system, as it is referring to equilibrium relationship of H_2S distribution in the bulk gas stream and on the surface of adsorbent. The most common isotherms used in the analysis are Langmuir, Frendliuch and Temkin isotherms. Each isotherm contains different equation that differentiates from each other. Adsorption kinetic related to the adsorption rate onto surface of a unit. The rate is required in designing process adsorption system [8]. As a conclusion, both adsorption kinetic and isotherm are required in designing adsorption system.

Currently, the existing results from the developed model are only focusing on the relationship between H_2S and activated carbon. The developed model for adsorption isotherm and adsorption kinetic are still not applied to the relationship between H_2S and hydrogel biochar. This analysis will apply both adsorption isotherm and adsorption kinetic to the H_2S and hydrogel biochar. This ex-

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periment will provide the most suitable model for isotherm and kinetic model.

2. Methodology

2.1. Experimental Data

Lab scale experiment is needed to get raw data of adsorption process. The experiment used difference thickness of adsorbent as a manipulative variable, while concentration of hydrogen sulfide is kept constant. There were two main manipulative variables which were time and thickness of adsorbent. There should be only two manipulated parameters as the model was used for two parameters only.

2.2. Isotherm Model

Adsorption isotherms indicate how molecules subjected to adsorption distribute themselves between liquid and solid phases at equilibrium time [9]. It gives some insight to adsorption mechanism, surface properties and affinities of adsorbent. Adsorption mechanism is referring whether it is chemical adsorption or physical adsorption. The most common isotherm used is Langmuir model and Freundlich model. Due to the nature of adsorbate and adsorbent, Elovich model is used.

To investigate correlation coefficient (R^2), Langmuir, Freundlich and Elovich equations are used. These equations describe adsorption phenomena in gas-solid diffusion. By applying these equations, R^2 can be found. The Adsorption capacity of adsorbent can also be investigated. These equations are linearized to give straight form in graph, shown in Equations (1) to (6) [10].

Langmuir Model
(non-linearized)

$$q_e = \frac{Q_0 K_L C_e}{1 + K_L C_e} \tag{1}$$

Langmuir Model
(linearized)

$$\frac{C_e}{q_e} = \frac{1}{Q_0 K_L} + \left(\frac{1}{Q_0}\right) C_e \tag{2}$$

Freundlich Model
(linearized)

$$\log Q_e = K_f + \frac{1}{n} log C_e \tag{3}$$

Freundlich Model
(non-linearized)

$$Q_e = K_f C_e^{\frac{1}{n}} \tag{4}$$

Elovich Model
(non-linearized)

$$\frac{q_e}{q_m} = K_E C_e \left(\exp \frac{q_e}{q_m}\right) \tag{5}$$

Elovich Model
(linearized)

$$\ln \frac{q_e}{q_m} = \ln K_E q_m - \frac{q_e}{q_m} \tag{6}$$

2.3. Kinetic Model

Kinetic model is an empirical model neglecting mass transfer effects which can replace kinetic modeling to resemble the observed data. To find the most suitable kinetic model for this adsorption process, R^2 value is needed. R^2 can only be obtained by using straight line in graph, which is from the linearized equation. From straight line, slope and intercept value can be found. The adsorption rate also can be calculated from the slope and intercept value. Other than that, adsorption mechanism can now be explained by involving kinetic-based model that describe reaction order of adsorption systems based on solution concentration [11]. The non-linearized and linearized equations for Pseudo-First order and second order are given in Equations (7) to (10)

Pseudo-First Order
(non-linearized)

$$\frac{dq_t}{dt} = k_{p1} (q_e - q_t) \tag{7}$$

Pseudo-First Order
(linearized)

$$\log (q_e - q_t) = \log q_e - k_{p1} t \tag{8}$$

Pseudo-Second Order
(non-linearized)

$$\frac{dq_t}{dt} = k_{p2} (q_e - q_t)^2 \tag{9}$$

Pseudo-Second Order
(linearized)

$$\frac{t}{q_t} = \frac{1}{V_0} + \frac{1}{q_e} t \tag{10}$$

3. Results and Discussion

3.1. Langmuir Isotherm Model

Langmuir isotherm assumes that adsorption takes place at a specific surface that contains a finite number of adsorption sites. It is also known as homogenous adsorption, which constant enthalpy and sorption activation energy are extracted from each molecule. Fig. 1 to Fig. 3 are ~~is~~ the result of Langmuir isotherm models toward the experiment. Differences can be identified by looking at the equation of each manipulative. The data from experiment will substitute to the Freundlich equation to plot the graph.

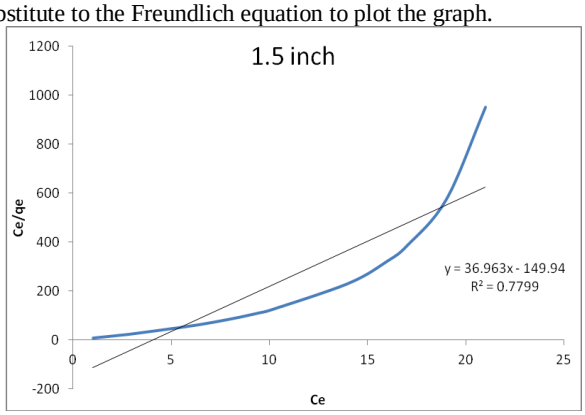


Fig. 1: Langmuir Isotherm Model for 1.5 inch of EFB-HBC

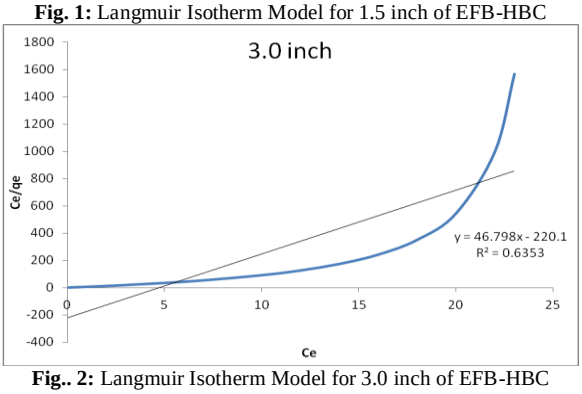


Fig.. 2: Langmuir Isotherm Model for 3.0 inch of EFB-HBC

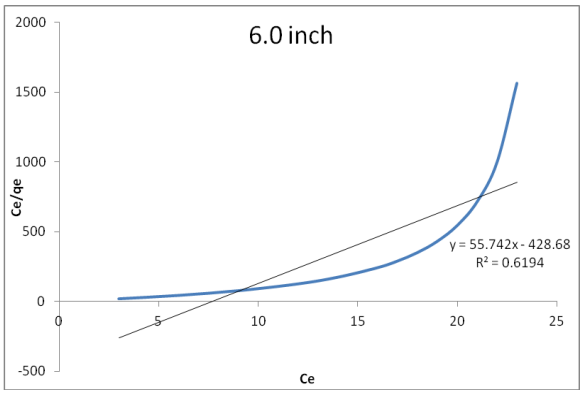


Fig. 3: Langmuir Isotherm Model for 6.0 inch of EFB-HBC

Table 1 represents data obtained from Fig. 1 to Fig. 3. Data are slope, intercept, R^2, b and Q_o value. This data can be obtained by making linear line.

Table 1: Langmuir Isotherm Model Parameters

Manipulative	Slope	Intercept	R^2	b	Q_o
1.5 inch	36.963	-149.94	0.7799	-4.0559	0.02705
3.0 inch	46.798	-220.10	0.6353	-4.7035	0.02137
6.0 inch	55.742	-428.68	0.6194	-7.6905	0.01794

Langmuir isotherm is designed to describe gas-solid phase adsorption. It also used to quantify and contrast adsorptive capacity of various adsorbents [10]. Adsorptive capacity of adsorbents can be seen in the value of b, in the equation, b is Langmuir constant that related to adsorption capacity ($mg.g^{-1}$). It correlated with variation of suitable area and porosity of adsorbent which implies that large surface area and pore volume will result in higher adsorption capacity [10]. From Table 1, the value of b increases as the thickness of the adsorbent increases. The negative value is due to value at negative section of axis. This proves the hypothesis is true. There is a drawback in increasing in thickness of adsorbent, the correlation coefficient decreases. Correlation coefficient (R^2) determined whether the isotherm is suitable for the process. Langmuir isotherm is classic isotherm. Classic isotherm is referring to isotherm that optimize in handling processes which involve 1:1 stoichiometry complex. Example for 1:1 stoichiometry complex is sorption site and a molecule. This experiment used multilayer adsorption. Thus, multilayer adsorption causes R^2 to decrease as the thickness of adsorbent increases.

3.2. Freundlich Isotherm Model

Freundlich isotherm assumes heterogeneous surface energies. As the value of slope approaches zero, the more heterogeneous it is. The range of slope is between zero to one. Slope is used to measure adsorption intensity or surface heterogeneity. It also can indicate cooperative adsorption, when the value of slope is more than one.

Fig. 4 to Fig. 6 is the result of Freundlich isotherm models toward the experiment. Differences can be identified by looking at equation of each manipulative. The data from experiment will substitute into Freundlich equation to plot the graph.

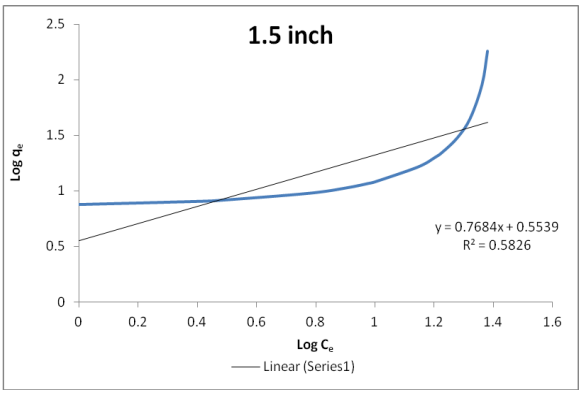


Fig. 4: Freundlich Isotherm Model for 1.5 inch of EFB-HBC

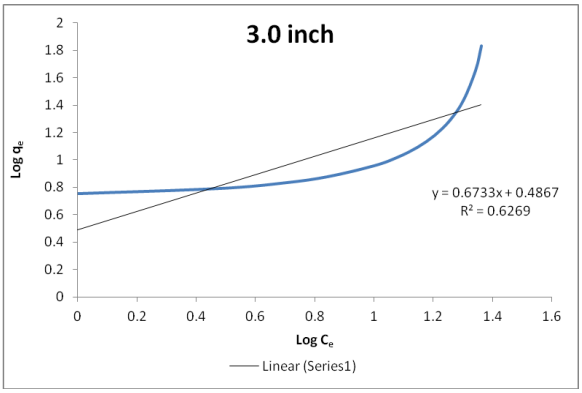


Fig. 5: Freundlich Isotherm Model for 3.0 inch of EFB-HBC

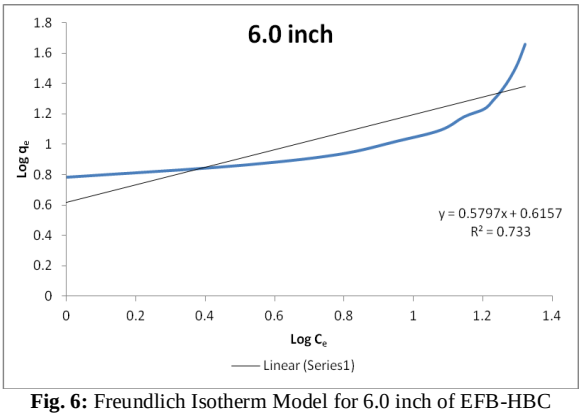


Fig. 6: Freundlich Isotherm Model for 6.0 inch of EFB-HBC

Table 2 represents data obtained from Fig. 4 to Fig. 6. Data obtained from the graphs are, slope, intercept, R^2, K_f and 1/n value. The data were extracted from a linearized equation.

Table 2: Freundlich Isotherm Model Parameters

Manipulative	Slope	Intercept	R^2	K_f	n
1.5 inch	0.7684	0.5539	0.5826	1.7400	1.3014
3.0 inch	0.6733	0.4867	0.6269	1.6269	1.4852
6.0 inch	0.5797	0.6157	0.7330	1.8510	1.7250

Freundlich isotherm is applicable to adsorption processes that occur on heterogonous surfaces [10]. It is used to describe adsorption characteristics for heterogenous surface [12]. The adsorption capacity can be seen in K_f value. K_f is an indicator for adsorption capacity while 1/n is a function of strength in adsorption process. Slope (1/n) value determined the type of adsorption of this process. When slope value is more than one, it indicates cooperative adsorption, while when slope value is less than one, it indicates normal adsorption. Based on Table 2, all slopes value is less than one, thus it is normal adsorption. The difference of normal adsorption and cooperative adsorption is movement of adsorbed. Cooperative adsorption causes adsorbed to track vertically and horizontally toward surface. Normal adsorption can only tract vertically towards surface.

Based on the raw data of experiment, K_f is supposed to increase as the thickness of adsorbent increase. As we can see in Fig. 8, the adsorbent, K_f value is less than adsorbent value in Fig. 7, for 1.5 inch, indicating that adsorption cannot occur properly at 3.0 inch of adsorbent. When thickness of adsorbent increases to 6.0 inch, K_f value increases higher than 1.5 inch. The inconsistent value of K_f is due to normal adsorption. Adsorption cannot fully occurred in constant rate, due to one motion only which is vertical motion. For the fitting error, correlation coefficient increases as the thickness of adsorbent increases. From Table 2, the correlation coefficient increases from 0.5826 to 0.7330. This indicates that Freundlich isotherm model can be used as thickness of adsorbent increases.

3.3. Elovich Isotherm Model

Elovich isotherm model is based on kinetic principle that assumed adsorption sites increases exponentially with adsorption [13]. This model also applies multilayer adsorption. Slope of this equation represents Elovich maximum adsorption capacity. The intercept value represents Elovich constant. Fig. 7 to Fig. 9 is the result of Elovich isotherm models toward the experiment. Differences can be identified by looking at the equation of each manipulative. The data from experiment will-substitute into Elovich equation to plot the graph.

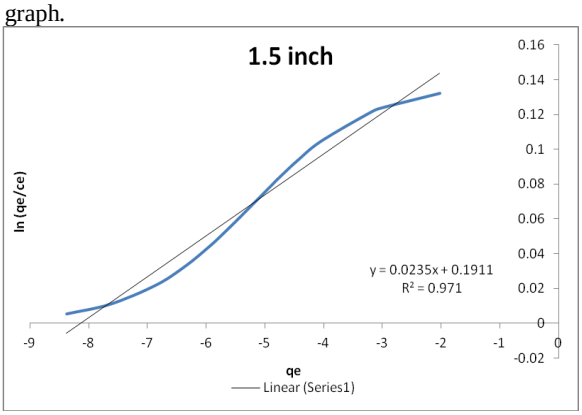


Fig. 7: Elovich Isotherm Model for 1.5 inch of EFB-HBC

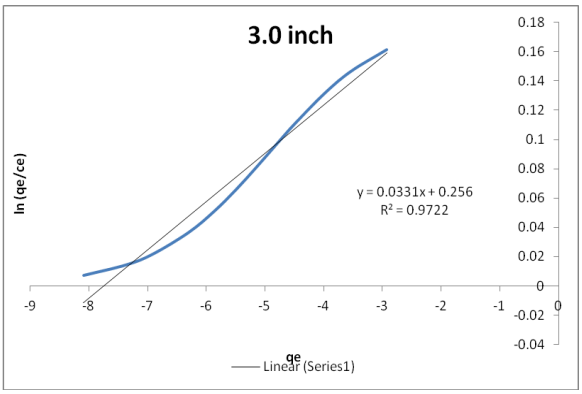


Fig. 8: Elovich Isotherm Model for 3.0 inch of EFB-HBC

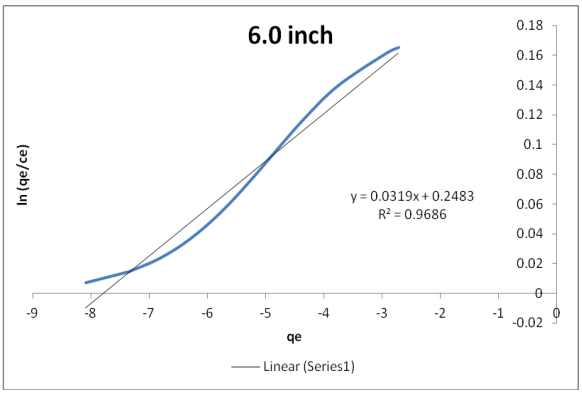


Fig. 9: Elovich Isotherm Model for 6.0 inch of EFB-HBC

Table 3 represents data obtained from Fig. 7 to Fig. 9. Data obtained from the graphs are, slope, intercept, R^2, K_e and q_e/q_m value. The data can be extracted from a linearized equation.

Table 3: Elovich Isotherm Model Parameters

Manipulative	Slope	Intercept	R^2	K_e
1.5 inch	0.0235	0.1911	0.9710	1.0238
3.0 inch	0.0331	0.2560	0.9722	1.0336
6.0 inch	0.0319	0.2483	0.9686	1.0324

In this Elovich model, it is assumed that adsorption sites increase as adsorption process and area of adsorbed molecules is added to adsorbing surface. The adsorption effective surface of this model is related exponentially with surface coverage. The equation was initially designed ~~it~~ to describe kinetics of chemisorptions of gas onto solids [10]

From Table 3, K_e is the maximum adsorption capacity. Theoretically, maximum adsorption capacity is directly proportional to the thickness of adsorbent. The difference of maximum adsorption capacity between 3.0 inch and 6.0 inch is not that big. Thus, maximum adsorption capacity increases as the thickness of adsorbent increases. For the fitting error, correlation coefficient (R^2) increases as the thickness of adsorbent increases. This means that the Elovich isotherm model can still be used even if the thickness above 6.0 inch.

3.4. Comparison of Isotherm Model

Table 4 is a summarization of data obtained from the graph of each of the isotherm model. The table shows which model is the most suitable to be used for adsorption process in this experimental setting.

Table 4: Comparison of Isotherm Model in term of Correlation Coefficient

	Isotherm Model					
	Langmuir		Freundlich		Elovich	
Correlation Coefficient (R^2)	1.5 inch	0.7799	1.5 inch	0.5826	1.5 inch	0.9710
	3.0 inch	0.6353	3.0 inch	0.6269	3.0 inch	0.9722
	6.0 inch	0.6194	6.0 inch	0.7330	6.0 inch	0.9686

By comparing the data in Table 4, the most suitable model is Elovich Isotherm Model. For each thickness variable, correlation coefficient of Elovich is the highest which are above 0.96.

Elovich model is suitable for chemisorptions process. Chemisorptions is a kind of adsorption that involve a chemical reaction between surface and adsorbate. From the chemical reaction, new chemical bonds are generated at adsorbant surface. All this occurs during adsorption process, in which this model was taken for calculation. In industry, every variables need to be taken as consideration to avoid worst case scenario. By using Elovich model, the industry takes precaution for adsorbent as this model includes

changes in adsorption site, in which other models did not considered.

Langmuir is not suitable to be used due to low R^2 value compare to Elovich. Even though, Langmuir model is suitable for gas-solid phase adsorption, but it assumes homogenous adsorption. Homogenous adsorption is a process in which enthalpy and sorption activation energy is constant while extracting from each molecule. When applying Langmuir model in real industry, only few elements can be constant, for example, pressure or temperature of the process. Activation energy and enthalpy for each molecule cannot be same throughout the process, thus making Langmuir model not valid.

Freundlich model is not suitable due to low R^2 value compare to other isotherm models. This model is not suitable for this experiment. This model focuses on heterogonous surfaces, which is non-uniform characteristic. In industry, adsorbent is uniform as it needs to fit into equipment. The characteristic of adsorbent is already pre-determined by industry. Thus, this model cannot be used as it does not predict accurate result.

3.5. Pseudo-First Order Model

According to Lagergren, first-order rate equation is presented as the kinetic process of liquid-solid phase adsorption of oxalic acid and malonic acid onto charcoal. It is believed to be the earliest model pertaining to adsorption rate based on the adsoption capacity. This model is widely used in industry to describe adsorption of pollutants from wastewater in different fields, such as adsorption of methylene blue from aqueous solution by broad bean peels and removal of malachite green from aqueous solutions using oil palm trunk fibre.

Fig. 10 to Fig. 12 is the result of Pseudo-First Order models toward the experiment. Differences can be identified by looking at the equation of each manipulative. The data from experiment will substitute into of Pseudo-First Order equation to plot the graph.

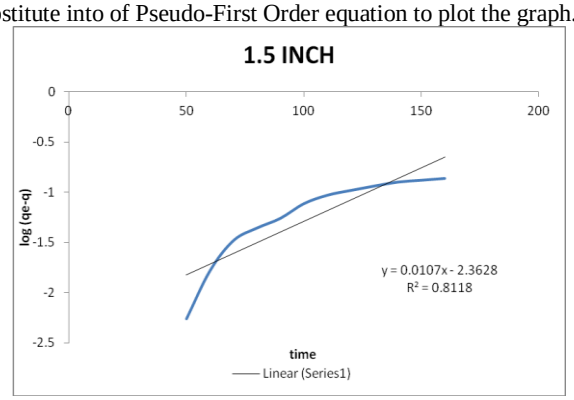


Fig. 10: Pseudo-First-Order Model for 1.5 inch of EFB-HBC

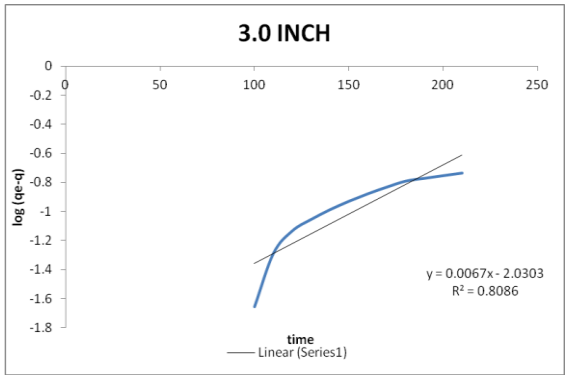


Fig. 11: Pseudo-First-Order Model for 3.0 inch of EFB-HBC

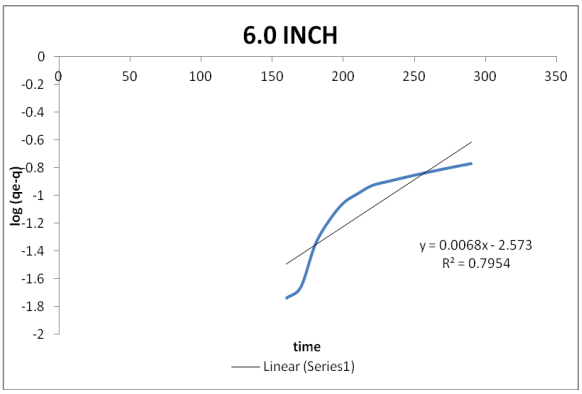


Fig. 12: Pseudo-First-Order Model for 6.0 inch of EFB-HBC

Table 5 represents data obtained from Fig. 10 to Fig. 12. Data obtained from the graphs are, slope, intercept, R^2, q_e and k_{p1} value. The data were extracted from a linearized equation.

Table 5: Pseudo-First-Order Model Parameters

Manipulative	Slope	Intercept	R^2	q_e	k_{p1} (1/s)
1.5 inch	0.0107	-2.3638	0.8118	0.0941	$6.6875*10^{-5}$
3.0 inch	0.0067	-2.0303	0.8086	0.1313	$3.1905*10^{-5}$
6.0 inch	0.0068	-2.5730	0.7954	0.0763	$2.0606*10^{-5}$

K_{p1} is the rate constant of pseudo-first order adsorption model, while q_e is the amount of hydrogen sulfide adsorbed at the time. The coefficient (R^2) decreases as the thickness of adsorbent increases. It shows that the model is unusable as the thickness increases. This model is used to describe adsorption of adsorbate from liquid phase [13].

The value of q_e, indicates that this model is unsuitable for this adsorption process. The value is inconsistent as the thickness of adsorbent increases. This will decrease the accuracy of this model. Thus, this model cannot be applied for real industry situation.

3.6. Pseudo-Second Order Model

The pseudo-second order kinetic model is based on assumption that chemisorptions is the rate-determining step [13]. If the adsorption process follows pseudo-second order kinetics, then the rate limiting step is chemical adsorption which involving valency forces through sharing or exchange of electrons between adsorbent and sorbate [9]. In earlier years, this model was used to describe adsorption reactions occurring in soil and soil minerals [14]. The model equation used describe fluoride adsorption onto acid-treated spent bleaching earth. It is also used to describe phosphamidon adsorption on an antimony (V) phosphate cation exchanger.

Fig. 13 to Fig. 15 are the result of Pseudo-Second Order models toward the experiment. Differences can be identified by looking at the equation of each manipulative. The data from experiment will substitute into of Pseudo-Second Order equation to plot the graph

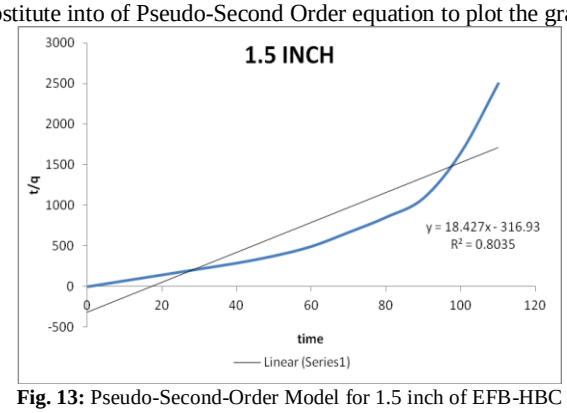


Fig. 13: Pseudo-Second-Order Model for 1.5 inch of EFB-HBC

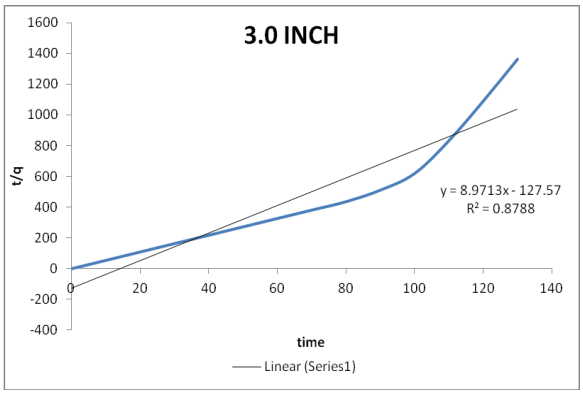


Fig. 14: Pseudo-Second-Order Model for 3.0 inch of EFB-HBC

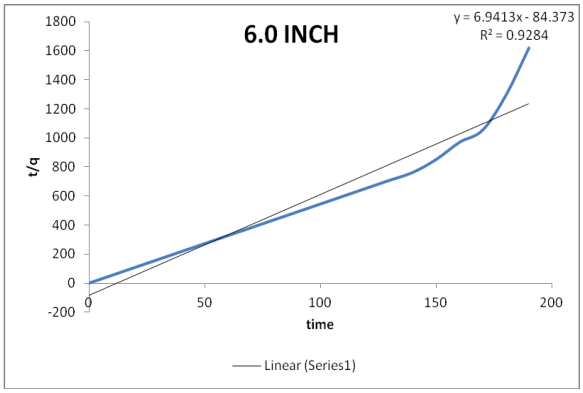


Fig. 15: Pseudo-Second-Order Model for 6.0 inch of EFB-HBC

Table 6 represents data obtained from Fig. 13 to Fig. 15. Data obtained from the graphs are, slope, intercept, R^2, q_e and k_{p2} value. The data can be extracted from a linearized equation.

Table 6: Pseudo-Second-Order Model Parameters

Manipulative	Slope	Intercept	R^2	q_e	k_{p2} (1/s)
1.5 inch	18.4270	-316.930	0.8035	0.0543	-1.0701
3.0 inch	8.9713	-127.570	0.8788	0.1115	-0.6305
6.0 inch	6.9413	-84.3730	0.9284	0.1441	-0.5708

K_{p2} is the rate constant of pseudo-second order adsorption model, while q_e is the amount of hydrogen sulfide adsorbed at the time. The correlation coefficient (R^2) increases as the thickness of adsorbent increases. The value of q_e increases showing an increase in adsorption rate of adsorbent. Both values are important in determining whether or not this model is useful. R^2 shows that industry can use this model to scale up their system. R^2 represents fitting toward process. Industry can estimate how much adsorption rate occurred at certain thickness of adsorbent by looking at the value of q_e. q_e is dependent on the concentration of adsorbate. Different concentration of adsorbate, will change adsorption rate of adsorbent.

3.7. Comparison of Adsorption Kinetic Model

Table 7 is a summarization of data obtained from the graph of each of the kinetic model. This table shows which model is the most suitable to be used for adsorption process in this experimental setting.

Table 7: Comparison of Isotherm Model in term of Correlation Coefficient

	Kinetic Model			
	Pseudo-First Order		Pseudo-Second Order	
R^2	1.5 inch	0.8118	1.5 inch	0.8035
	3.0 inch	0.8086	3.0 inch	0.8788
	6.0 inch	0.7954	6.0 inch	0.9284

By comparing data in Table 7, the most suitable model is Pseudo-Second Order model. For each thickness variable, correlation coefficient of Pseudo-Second Order is the highest. The pattern for correlation coefficient for Pseudo-Second Order had shown that as thickness of adsorbent increases, the correlation coefficient increases. This pattern proves that this model is suitable for adsorption system for scaling process.

Pseudo-First Order is not suitable is due to its requirement. The limitation of this model is that it describes adsorption of liquid and solid. For this experiment, the adsorbate are in gas form, thus it does not meet the requirement. In many adsorption processes, the pseudo-first order kinetics was found to be suitable for only the initial 20 to 30 minute of interaction time and not for the whole range of contact time [9]. In industry, the process will occurr continuously for a long duration of time. Thus, making this model is not suitable for this adsorption process.

Pseudo-Second Order shows that the adsorption process is chemical adsorption in which involve valency forces through sharing or exchange of electrons between adsorbent and metal ions [9]. The advantage of using this model is that q_e values has a small sensitivity to the influence of random experimental errors. This means that q_e does not has affect due to experimental errors. This advantage makes the model is suitable for industry use.

4. Conclusion

Hydrogen sulfide is second toxic gas in the world. It can be harmful substance even if the concentration of substances is low. Recently, many industries produce hydrogen sulfide as a waste product. Therefore they need adsorption system to adsorb hydrogen sulfide from waste stream. In order to optimize the adsorption system, isotherm and kinetic model is needed due to nature of hydrogen sulfide. Chemical kinetics is a study of rates of chemical processes while adsorption isotherms indicate how molecules subjected to adsorption distribute themselves between two phases at equilibrium time. Isotherm model used in this experiment are Langmuir, Freundlich and Elovich model, while kinetic model used in this experiment are Pseudo-First Order and Pseudo-Second Order. The results show that Elovich model is the most suitable model as it has the highest correlation coefficient compared to others. Pseudo-Second Order is the most suitable or compatible for adsorption system due to high value of correlation coefficient.

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