MODELING OF ADSORPTION KINETIC AND EQUILIBRIUM ISOTHERMS OF HYDROGEN SULFIDE ONTO HYDROGEL BIOCHAR ADSORBENT

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***Abstract*—Every process produces byproduct, byproduct can be dangerous or not. Hydrogen sulfide is one of the dangerous byproduct produced. In order to prevent hydrogen sulfide to harm workers or residents, control system is required. Currently, all industry has a control system such as adsorption system, to control dangerous component such as hydrogen sulfide. In order to optimize adsorption system, research is required. Researching using laboratory method is very dangerous due to nature of hydrogen sulfide. Thus isotherm and kinetic model is used as alternative method, as it doesn't involve researching in laboratory. Each isotherm model and kinetic model equation has different uses for adsorption process. The isotherm models used are Langmuir, Freundlich and Elovich, while kinetic models used are Pseudo-First Order and Pseudo-Second Order. The main objective of this experiment is to find the most suitable equation for adsorption of hydrogen sulfide. Suitability of equation is determined by correlation coefficient (R^2). The higher correlation coefficient, the more suitable the equation is to process. After result obtained, the most suitable isotherm and kinetic model are Elovich model and Pseudo-Second Order model respectively. R^2 for Elovich model is 0.9686 while R^2 for Pseudo-Second Order model is 0.9284. By comparing R^2 of each model, it is shown that Elovich and Pseudo-Second Order model are the most suitable for this adsorption process due to highest correlation coefficient.**

Keywords—hydrogen sulfide, isotherm model, kinetic model, Langmuir, Freundlich, Elovich, Pseudo-First Order, Pseudo-Second Order, Adsorption.

I. INTRODUCTION

The most toxic gas is carbon monoxide while the second is hydrogen sulfide. The hydrogen sulfide can cause inhalational deaths. It is dangerous as mechanism is unknown to people. It can be produced naturally from organic matter that decays (Jiang et al., 2016). This production of hydrogen sulfide can be seen in the Petroleum production and refining, sewer and wastewater treatment and Agricultural silos (https ://www.osha.gov /SLTC/ hydrogensulfide/ hydrogensulfide_found.html). They are exposed to the hydrogen sulfide by inhaling the gas. Effects depend on how much and how long is the workers exposed to the hydrogen sulfide. By referring to Occupational Safety and Health Act (OSHA), symptoms can identify when workers exposed to high concentration. The worst scenario is that workers dead by rapid unconsciousness and stop breathing.

Hydrogen sulfide can be very dangerous but industry can take preventive action to protect their employees and their building. It can be corrosive toward metals and concrete. Commercialized process that 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. Atoms, ions or molecules adhere to a surface of adsorbent due to differences in concentration.There are many parameters in improving the adsorption rate, such as temperature, condition of adsorbent and adsorbent condition (Bajpai & Rajpoot, 1999).

Activated carbon is one of the adsorbent's examples. It is a carbonaceous, highly porous adsorptive medium that has a complex structure composed primarily that has complex structure composed primarily of carbon atoms. 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 produce 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 into 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 uses 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 requires in designing process adsorption system (Journal & Science, 2016). As conclusion, both adsorption kinetic and isotherm are required in designing adsorption system.

Currently the existing results from 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 experiment will provide the most suitable model for isotherm and kinetic model.

II. METHODOLOGY

A. Data Experiment

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

B. Isotherm Model

Adsorption isotherms indicate how molecules subjected to adsorption distribute themselves between liquid and solid phases

at equilibrium time (Aly, Graulet, Scales, & Hanley, 2014). 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 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. Adsorption capacity of adsorbent can also be found. These equations are linearized to give straight form in graph (Ayawei, Ebelegi, & Wankasi, 2017).

Langmuir Model (non-linearized)

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

Langmuir Model (linearized)

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

Freundlich Model (non-linearized)

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

Freundlich Model (linearized)

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

Elovich Model (non-linearized)

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

Elovich Model (linearized)

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

C. *Kinetic Model*

Kinetic model is an empirical model neglecting mass transfer effects, can replace kinetic modeling to resemble observe data. To find which the most suitable kinetic model for this adsorption process, R^2 value is needed. R^2 can only be found by using straight line in graph, which is from linearized equation. From straight line, slope and intercept value can be found. The adsorption rate also can be calculated from slope and intercept value. Other than that, adsorption mechanism now can be explained by involving kinetic-based model that describing reaction order of adsorption systems based on solution concentration (., Shukla, & Kisku, 2015).

Pseudo-First Order (non-linearized)

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

Pseudo-First Order (linearized)

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

Pseudo-Second Order (non-linearized)

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

Pseudo-Second Order (linearized)

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

III. RESULTS AND DISCUSSION

A. *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.

Figure 4.1 until figure 4.3 is the result of Langmuir isotherm models toward the experiment. Differences can be identified by looking at equation of each manipulative. The data from experiment will substitute to the Freundlich equation to plot the graph.

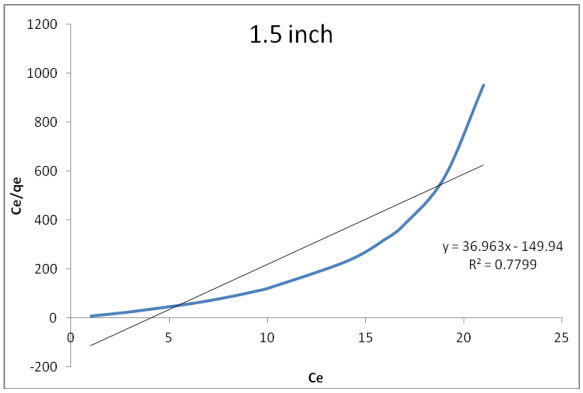

Figure 4.1: Langmuir Isotherm Model for 1.5 inch of EFB-HBC

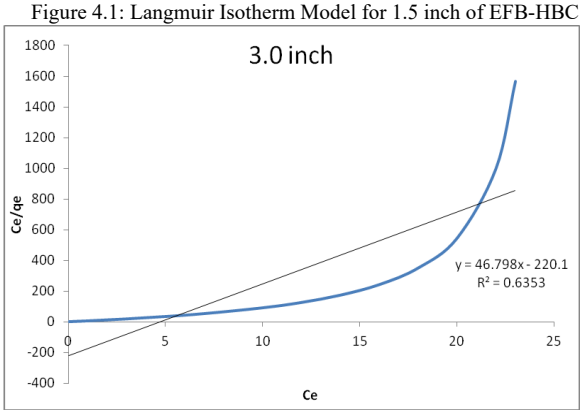

Figure 4.2: Langmuir Isotherm Model for 3.0 inch of EFB-HBC

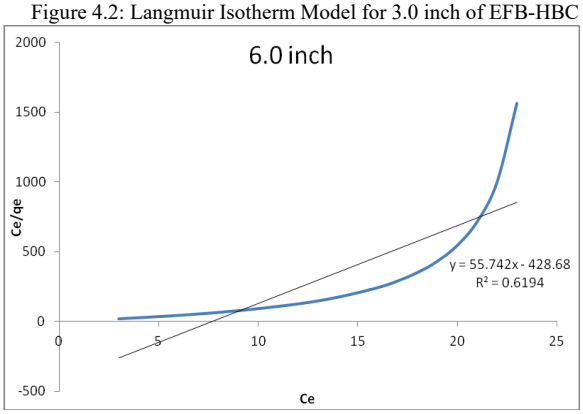

Figure 4.3: Langmuir Isotherm Model for 6.0 inch of EFB-HBC

Table 4.1 represents data obtained from figure 4.1 until 4.3. Data are slope, intercept, R^2, b and Q_o value. This data can be obtained by making linear line.

Table 4.1: 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 (Ayawei et al., 2017). Adsorptive capacity of adsorbents can be seen in the value of b, in 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 (Ayawei et al., 2017). From table 4.1, the value of b increases as the thickness of the adsorbent increases. The negative value is due to value is at negative section of axis. This proves the hypothesis is true. There is a con in increasing in thickness of adsorbent, the correlation coefficient decreases. Correlation coefficient (R^2) is what determined whether that isotherm is suitable or not for the process.

Langmuir isotherm is classic isotherm. Classic isotherm referring to it is isotherm that optimize in handling that process, that involving 1:1 stoichiometry complex. Example for 1:1 stoichiometry complex is sorption site and a molecule. This experiment use multilayer adsorption. Thus multilayer adsorption causes R^2 to decrease as thickness of adsorbent increases.

B. 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.

Figure 4.4 until figure 4.6 is the result of Freundlich isotherm models toward the experiment. 55Differences can be identified by looking at equation of each manipulative. The data from experiment will substitute into Freundlich equation to plot the graph.

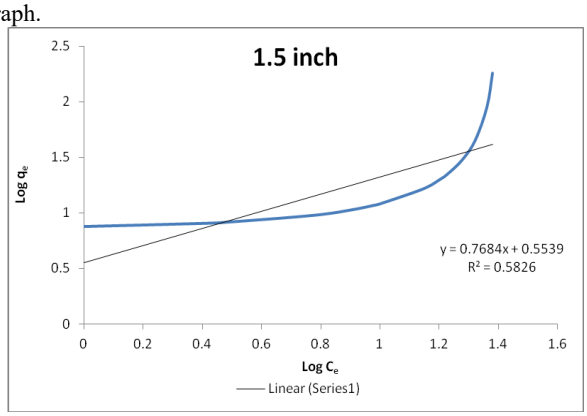

Figure 4.4: Freundlich Isotherm Model for 1.5 inch of EFB-HBC

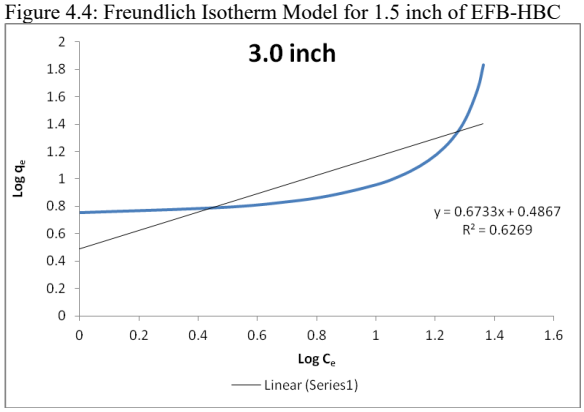

Figure 4.5: Freundlich Isotherm Model for 3.0 inch of EFB-HBC

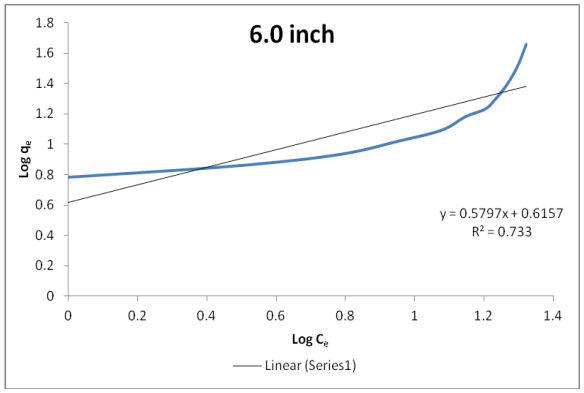

Figure 4.6: Freundlich Isotherm Model for 6.0 inch of EFB-HBC

Table 4.2 represents data obtained from figure 4.4 until 4.6. Data obtained from the graphs are, slope, intercept, R^2, K_f and 1/n value. The data can be extracted from a linearized equation.

Table 4.2: 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

Accoding to Journal of Chemistry, Freundlich isotherm is applicable to adsorption processes that occur on heterogonous surfaces (Ayawei et al., 2017). It is used to describe adsorption characteristics for heterogenous surface (A.O, 2012). The adsorption capacity can be seen in K_f value. K_f is indicator for adsorption capacity while 1/n is a function of strength in adsorption process.

Slope (1/n) value determined type of adsorption of this process. When slope value more than one, it indicates cooperative adsorption, while slope value less than one, it indicates normal adsorption. Based on table 4.2, 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 raw data of experiment, K_f is supposed to increase as thickness of adsorbent increase. As we can see that in 3.0 inch of adsorbent, K_f value is less than 1.5 inch of adsorbent value, which indicates adsorption cannot occur properly at 3.0 inch of adsorbent. Then thickness of adsorbent increases to 6.0 inch, K_f value increases higher than 1.5 inch value. 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 vertically motion.

For the fitting error, correlation coefficient increases as thickness of adsorbent increases. From table 4.2, correlation coefficient increases from 0.5826 to 0.7330, this indicates that freundlich isotherm model can be used as thickness of adsorbent increases.

C. Elovich Isotherm Model

Elovich isotherm model is based on kinetic principle that assumed adsorption sites increases exponentially with adsorption (Farouq & Yousef, 2015). This model also applies multilayer adsorption. Slope of this equation represents Elovich maximum adsorption capacity. The intercept value represents Elovich constant.

Figure 4.7 until figure 4.9 is the result of Elovich isotherm models toward the experiment. Differences can be identified by looking at equation of each manipulative. The data from experiment will substitute into Elovich equation to plot the graph.

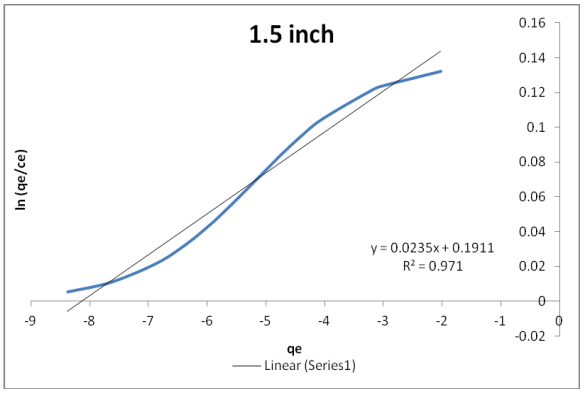

Figure 4.7: Elovich Isotherm Model for 1.5 inch of EFB-HBC

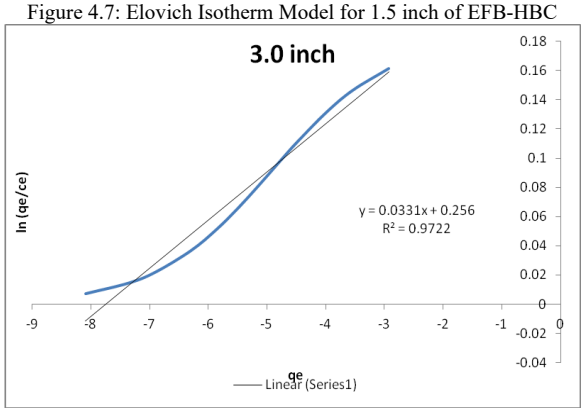

Figure 4.8: Elovich Isotherm Model for 3.0 inch of EFB-HBC

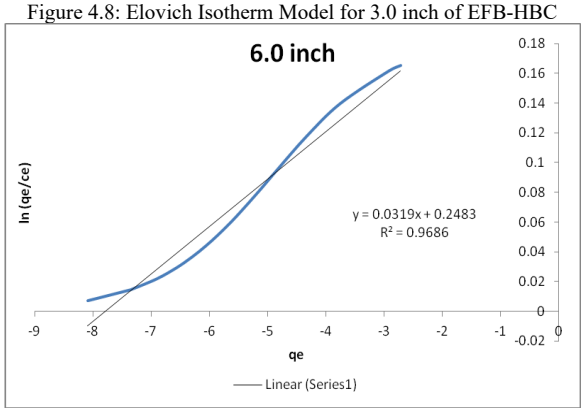

Figure 4.9: Elovich Isotherm Model for 6.0 inch of EFB-HBC

Table 4.3 represents data obtained from figure 4.7 until 4.9. 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 4.3: 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 ti describe kinetics of chemisorptions of gas onto solids (Ayawei et al., 2017).

From table 4.3, 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 thickness of adsorbent increases.

For the fitting error, correlation coefficient (R^2) increases as thickness of adsorbent increases. This means that the Elovich isotherm model can still be used even if the thickness above 6.0 inch.

D. Comparison of Isotherm Model

Table 4.4 is summarization of data obtained from the graph from each of the isotherm model. This table functions to show which model is the most suitable to use for adsorption process in this experiment setting.

Table 4.4: 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 compare data in table 4.4, the most suitable model is Elovich Isotherm Model. For each thickness variable, correlation coefficient of Elovich is the highest which are 0.96 above.
Elovich model is suitable for chemisorptions process. Chemisorptions is a kind of adsorption that involve chemical reaction between surface and adsorbate. From the chemical reaction, new chemical bonds are generated at adsorbant surface. All this things happened during adsorption process, which this model taken for calculated. 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 model didn't considered.

Langmuir is not suitable 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 process in which enthalpy and sorption activation energy is constant while extracting from each molecule. When applying Langmuir model in real industry, there little which element can be constant, for example, pressure or temperature of process. Activation energy and enthalpy for each molecule cannot be same throughout process, thus making Langmuir model is not valid.

Freundlich model is not suitable due to lowest R^2 value compare to other isotherm model. This model is not suitable for this experiment. This model is focusing 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.

E. Pseudo-First Order Model

According to Lagergren, first-order rate equation is presented as description of kinetic process of liquid-solid phase adsorption of oxalic acid and malonic acid onto charcoal (lagergren, 1898). It is believed to be earliest model pertaining to adsorption rate based on the adsoption capacity. This model 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 (Hameed and El-Khaiary, 2008a; 2008b; Tan et al., 2008).

Figure 4.10 until figure 4.12 is the result of Pseudo-First Order models toward the experiment. Differences can be identified by looking at equation of each manipulative. The data from experiment will substitute into of Pseudo-First Order equation to plot the graph.

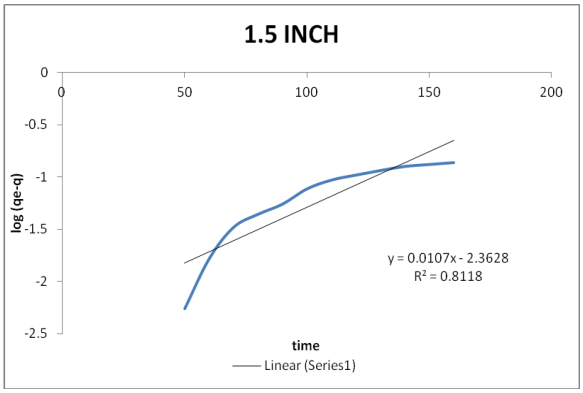

Figure 4.10: Pseudo-First-Order Model for 1.5 inch of EFB-HBC

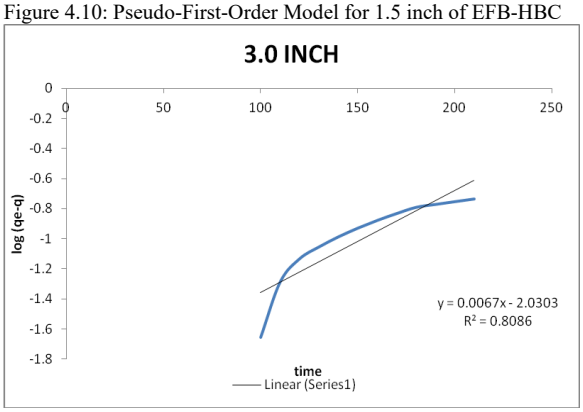

Figure 4.11: Pseudo-First-Order Model for 3.0 inch of EFB-HBC

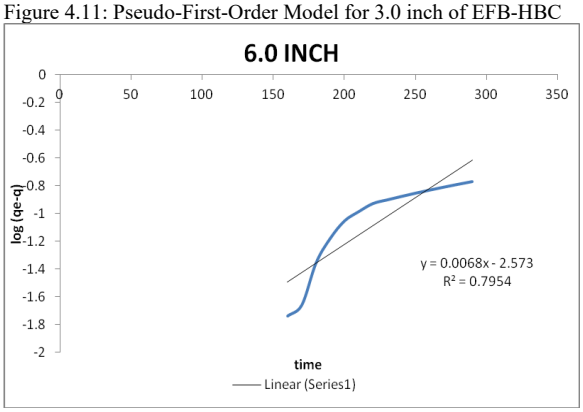

Figure 4.12: Pseudo-First-Order Model for 6.0 inch of EFB-HBC

Table 4.5 represents data obtained from figure 4.10 until 4.12. Data obtained from the graphs are, slope, intercept, R^2, q_e and k_{p1} value. The data can be extracted from a linearized equation.

Table 4.5: 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 time. The correlation coefficient (R^2) decreases as the thickness of adsorbent increases. It shows that the model is unusable as thickness increases. This is due to that this model is used to describe adsorption of adsorbate from liquid phase (Farouq & Yousef, 2015).

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

F. Pseudo-Second Order Model

The pseudo-second order kinetic model is based on assumption that chemisorptions is the rate-determining step (Farouq & Yousef, 2015). 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 (Aly et al., 2014).

In earlier years, this model used to describe adsorption reactions occurring in soil and soil minerals (Qiu et al., 2009). According to Mahramanliolu et al.,2002, the model equation used describe fluoride adsorption onto acid-treated spent bleaching earth. It also used to describe phosphamidon adsorption on an antimony (V) phosphate cation exchanger, said by Varshney et al., 1996..

Figure 4.13 until figure 4.15 is the result of Pseudo-Second Order models toward the experiment. Differences can be identified by looking at equation of each manipulative. The data from experiment will substitute into of Pseudo-Second Order equation to plot the graph.

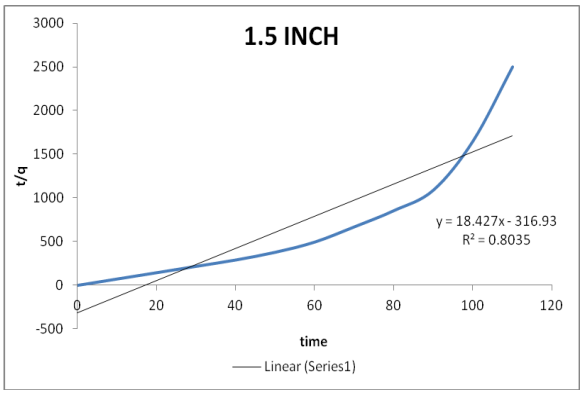

Figure 4.13: Pseudo-Second-Order Model for 1.5 inch of EFB-HBC

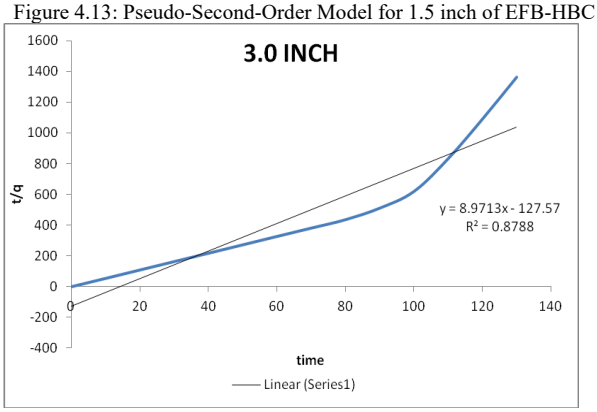

Figure 4.14: Pseudo-Second-Order Model for 3.0 inch of EFB-HBC

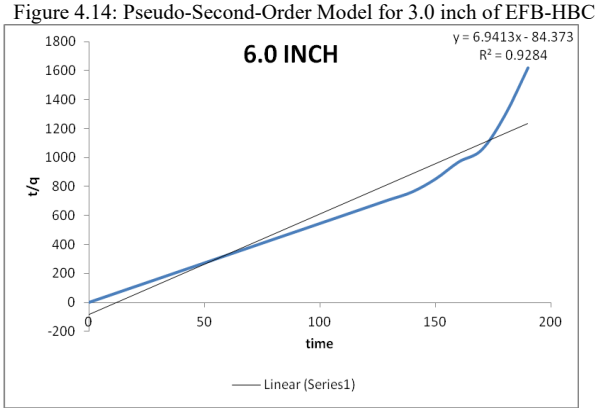

Figure 4.15: Pseudo-Second-Order Model for 6.0 inch of EFB-HBC

Table 4.6 represents data obtained from figure 4.13 until 4.15. 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 4.6: 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 time. The correlation coefficient (R^2) increases as the thickness of adsorbent increases. The value of q_e increases showing an increasing in adsorption rate of adsorbent. Both values are important in determining whether or not this model is useful or not. 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 the value of q_e. q_e is depend on the concentration of adsorbate. Different concentration of adsorbate, will change adsorption rate of adsorbent.

G. Comparison of Adsorption Kinetic Model

Table 4.7 is summarization of data obtained from the graph from each of the kinetic model. This table functions to show which model is the most suitable to use for adsorption process in this experiment setting.

Table 4.7: 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 compare data in table 4.7, 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 is describing for 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 initial 20 to 30 minute of interaction time and not for the whole range of contact time (Aly et al., 2014). In industry, the process will occurred 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(Aly et al., 2014). The advantage of using this model is that q_e values is has small sensitivity to the influence of random experimental errors. This means that q_e does not affect due to experimental errors. This advantage makes the model is suitable in using for industry.

IV. 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 industry produce hydrogen sulfide as waste product, they need adsorption system in order to adsorb hydrogen sulfide from waste stream. In order to optimize adsorption system, isotherm and kinetic model is needed due to nature of hydrogen sulfide. Chemical kinetics is 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 obtained are Elovich model is the most suitable due to correlation coefficient is the highest compare to other models. Pseudo-Second Order is the most suitable or compatible for adsorption system due to high value of correlation coefficient.

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