

# Adsorption Kinetics and Equilibrium Isotherms Modeling of H<sub>2</sub>S on Hydrogel Biochar and Biochar Adsorbent from Rice Husk



Ashadi Azwan Abd Rahman, Azil Bahari Alias, Muhamad Ariff Amir Hamzah, Mohammad Aidil Ali

**Abstract:** Hydrogen sulphide is a poisonous gas that is commonly found in swamps and areas of high volcanic activities. Due to the dangers and hazards that it may impose such as neurological disorders and miscarriages, continuous innovative attempts to remove the gas are in place. A study was conducted to synthesize an adsorbent that is made from activated rice husk biochar and also hydrogel biochar. This study is complementary to that study where the adsorption processes using the two adsorbents are mathematically modelled. Three parameters were studied which were the adsorbent mass, the gas flow rate, and the gas temperature. It was found that for rice husk-based activated biochar, for all three parameters, the adsorption processes could be mathematically represented using Thomas Model and Yoon-Nelson Model. Meanwhile, for rice husk-based hydrogel biochar, the adsorption process could be mathematically represented using Thomas and Yoon-Nelson Model for the effect of mass of adsorbent, and Adam-Bohart for the effect of gas flow rate and gas temperature. Although the coefficient of determination (R<sup>2</sup>) suggested that Thomas and Yoon-Nelson Model are more appropriate to be used to model the latter two parameters, because the exit concentration reached the point when it was greater than half the inlet concentration well before 0-th second, the linearly regressed equation became mathematically inconsistent with the isotherm models. Kinetic studies were also done, and it was found that the adsorption processes using the activated biochar fit both pseudo-first and pseudo-second order equation. This means that the adsorption processes using the activated biochar are both physisorption and chemisorption. Meanwhile, the adsorption processes using the hydrogel biochar fit only the pseudo-second order equation, suggesting that the adsorption process is chemisorption.

**Keywords :** Adsorption column, flow isotherm, hydrogen sulphide, kinetic modeling, mathematical modelling.

## I. INTRODUCTION

Hydrogen sulfide (H<sub>2</sub>S) is a gas that often a product of the anaerobic microbial breakdown of organic matter, which usually happens in swamps and sewers. Hydrogen sulfide is

also soluble in water, with an interpolated magnitude of 0.0880 mol/L at 29.5 °C [1]. Due to the harmful potential that it could bring and precedential detriments that it has caused, the removal of the gas has become a great interest in order to ensure within the safer limit.

One of the many ways it could be removed apart from the catalytic conversion is via adsorption. [2,3,4]. This study is complementary to a study done by Rahman et al (2019) which was the synthesis and characterization of rice-husk biochar and HBC adsorbents, and the process of adsorption of hydrogen sulphide onto the adsorbent [5]

This study investigates the adsorption kinetics of the adsorption process and the equilibrium isotherm that fits well into the adsorption process [6]. Adsorption isotherm has been used to mathematically model adsorption processes so that an adsorption process can be mathematically simulated without having the experiments to be conducted [7,8,9]

Mathematical modelling are also used to validate the experimental results. Hoegberg et al. (2002) conducted adsorption isotherm studies to determine the effect of pH and the presence of promoter on the adsorption of paracetamol onto activated charcoal [10]. Zheng et al. (2010) [11] also studied equilibrium isotherms and kinetic models on the adsorption of cadmium (II) onto modified corn stalk adsorbent and Azouaou et al. for adsorptions lead from aqueous solutions [12].

Therefore, this paper will investigate two things: the adsorption isotherm that is suitable to mathematically model the adsorption processes and the order of the kinetic equation to represent the adsorption processes.

## II. MATERIALS AND METHODS

The scope of the study is only focusing on the adsorptions of H<sub>2</sub>S onto rice husk biochar and hydrogel biochar.

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# Adsorption Kinetics and Equilibrium Isotherms Modeling of H<sub>2</sub>S on Hydrogel Biochar and Biochar Adsorbent From Rice Husk

The study flow covers starting from specified material, parameter of the study, equilibrium study and also kinetic study. Each of the item are further elaborate in section as below.

## A. Materials

The two adsorbents that are used are rice husk-based activated biochar and hydrogel biochar. The preparation of the biochar is following the method of Meri et al [13].

## B. Data Acquisition

Three parameters are studied in the equilibrium studies, which are the adsorbent mass, gas flow rate, and gas temperature. For each adsorbent, the adsorbent mass are set to be at 20 g, 30 g, and 40 g; the gas flow rate is set at 0.1 m<sup>3</sup>/h, 0.15 m<sup>3</sup>/h, and 0.2 m<sup>3</sup>/h; the gas temperature is set to be at 30 °C, 50 °C, and 70 °C.

The adsorption is done over a span of 25 minutes, where the inlet concentration is set at 25 ppm (34.87 mg/m<sup>3</sup>) of H<sub>2</sub>S carried by N<sub>2</sub>. The exit concentration is taken every minute.

For the effect of adsorbent mass, the gas flow rate is set constant at 0.1 m<sup>3</sup>/h and gas temperature of 30 °C; for the effect of gas flow rate, the adsorbent mass is set constant at 30 g and gas temperature at 30°C; and for the effect of gas temperature, the adsorbent mass is set at 30 g and the gas flow rate is set at 0.1 m<sup>3</sup>/h.

## C. Equilibrium Studies

The data acquired is used in the three equilibrium isotherms that are used for equilibrium studies, which are the Thomas Model, Yoon-Nelson Model, and Adam-Bohart Model. The equilibrium isotherms afore mentioned are shown below respectively in Equation (1) to (3):

$$\ln\left(\frac{C_o}{C_f} - 1\right) = \frac{k_{Th}q_o m}{F} - k_{Th}C_o t \quad (1)$$

$$\ln\left(\frac{C_f}{C_o - C_f}\right) = k_{YN}(t - \tau) \quad (2)$$

$$\ln\left(\frac{C_f}{C_o}\right) = k_{AB}C_o t - \frac{k_{AB}N_o V}{F} \quad (3)$$

## D. Kinetic Studies

Apart from equilibrium isotherms, the data acquired from the adsorption process is also used for kinetic studies. The data is used in two kinetic equations, which are the pseudo-first order kinetic equation, and the pseudo-second order kinetic equation which been used other researchers [14]. The following are the pseudo-first order kinetic equation and pseudo-second order kinetic equation respectively in Equations (4) and (5):

$$q_t = q_o e^{-kt} \quad (4)$$

$$q_t = \frac{k_2 q_e^2 t}{1 + k_2 q_e t} \quad (5)$$

The linearization of each model will yield the following equations in Equation (6) and (7) respectively for first and second order:

$$\ln q_t = \ln q_o - kt \quad (6)$$

$$\frac{t}{q_t} = \frac{t}{q_e} + \frac{1}{k_2 q_e^2} \quad (7)$$

The data is used to plot the isotherm models given above, and the kinetic equation whose line of best fit with the highest coefficient of correlation (R<sup>2</sup>) is chosen as the isotherm model that is the most suitable to mathematically model the adsorption process.

## III. RESULTS AND DISCUSSIONS

### A. Adsorption Process

The findings for the adsorption experiments for each parameter studied are shown in Fig. 1 to Fig. 6; where Fig. 1, Fig. 3 and Fig. 5 are for activated biochar adsorbent for effect of mass, gas flow and gas temperature respectively, while Fig. 2, Fig. 4 and Fig. 6 are for hydrogel biochar adsorbent, for the same three parameters mentioned above respectively.

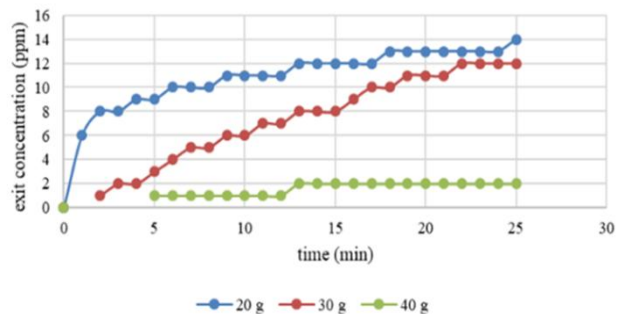


Fig. 1. Exit concentration profile for the effect of the mass of activated biochar adsorbent.

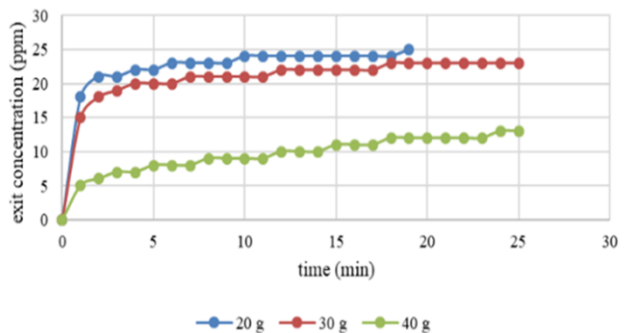


Fig. 2. Exit concentration profile for the effect of the mass of hydrogel biochar adsorbent.

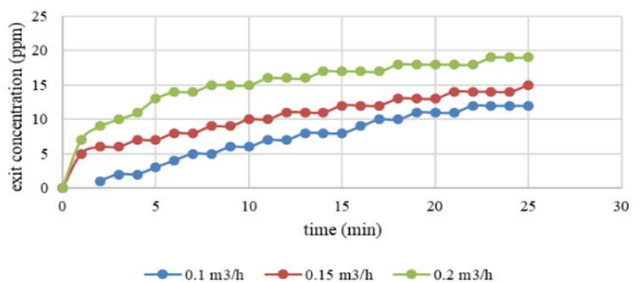


Fig. 3. Exit concentration profile for the effect of gas flow rate on activated biochar

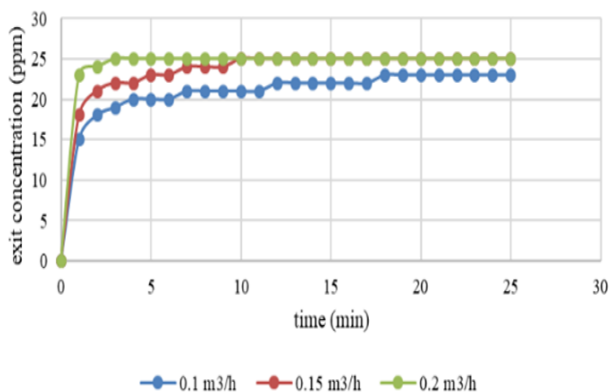


Fig. 4. Exit concentration profile for the effect of gas flow rate on hydrogel biochar

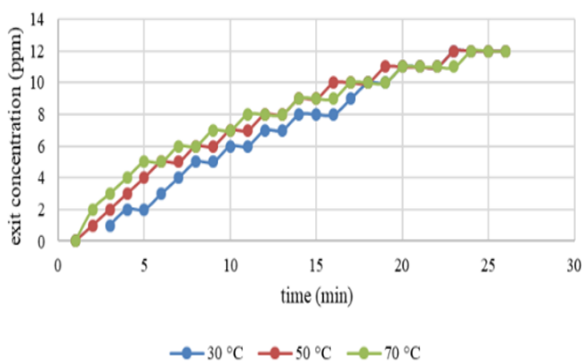


Fig. 5. Exit concentration profile for the effect of gas temperature on activated biochar.

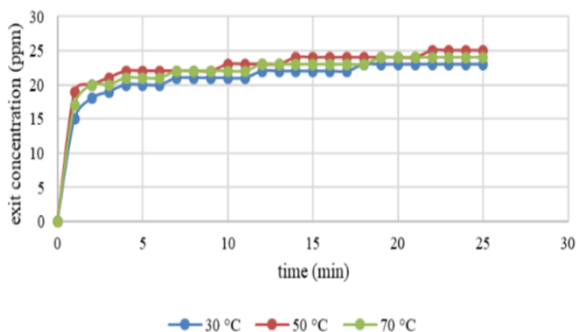


Fig. 6. Exit concentration profile for the effect of gas temperature on hydrogel biochar

**B. Equilibrium Studies**

The findings of the equilibrium studies for each parameter are summarized in the Table-I to Table-IV. The data presented in the tables show the suitability of isotherm model to represent the effect of the given parameter is decided based on how close the highest value of the  $R^2$  to unity is (which indicates higher correlation and reliability). This can be demonstrated via the effect of adsorbent mass for activated biochar, Thomas model and Yoon-Nelson model are chosen because the highest value of  $R^2$  is 0.8819 which is closer to 1 than Adam-Bohart whose highest value of  $R^2$  is 0.8234.

For the determination of isotherm parameters, the linear equation yielded from the data of the parameter value that yielded the highest  $R^2$  is chosen to calculate the isotherm parameters. The following table summarizes the calculation.

**Table-I: Summary of the effect of adsorbent mass**

Adsorbent	Isotherm Model	Mass (g)	$R^2$	Applicable
Activated biochar	Thomas	20	0.8726	Yes
		30	0.8819	
		40	0.7091	
	Yoon-Nelson	20	0.8726	Yes
		30	0.8819	
		40	0.7091	
Adam-Bohart	20	0.8234	No	
	30	0.8201		
	40	0.7091		
Hydrogel biochar	Thomas	20	0.8309	Yes
		30	0.887	
		40	0.9344	
	Yoon-Nelson	20	0.8309	Yes
		30	0.887	
		40	0.9344	
	Adam-Bohart	20	0.6804	No
		30	0.7092	
		40	0.9002	

**Table-II: Summary of the effect of gas flow rate**

Adsorbent	Isotherm Model	Flow rate (m <sup>3</sup> /h)	$R^2$	Applicable
Activated biochar	Thomas	0.10	0.8819	Yes
		0.15	0.9633	
		0.20	0.8722	
	Yoon-Nelson	0.10	0.8819	Yes
		0.15	0.9633	
		0.20	0.8722	
Adam-Bohart	0.10	0.8201	No	
	0.15	0.9280		
	0.20	0.7549		
Hydrogel biochar	Thomas	0.10	0.8870	Yes
		0.15	0.9278	
		0.20	1.0 (Invalid)	
	Yoon-Nelson	0.10	0.8870	Yes
		0.15	0.9278	
		0.20	1.0 (Invalid)	
Adam-Bohart	0.10	0.7029	No	
	0.15	0.7959		
	0.20	0.9999 (Invalid)		

**Table-III: Summary of the effect of gas temperature**

Adsorbent	Isotherm Model	Temperature (°C)	$R^2$	Applicable
Activated biochar	Thomas	30	0.8819	Yes
		50	0.8278	
		70	0.8806	
	Yoon-Nelson	30	0.8819	Yes
		50	0.8278	
		70	0.8806	
Adam-Bohart	30	0.8201	No	
	50	0.7534		
	70	0.8212		
Hydrogel biochar	Thomas	30	0.8922	Yes
		50	0.9086	
		70	0.9211	
	Yoon-Nelson	30	0.8922	Yes
		50	0.9086	
		70	0.9211	
	Adam-Bohart	30	0.7092	No
		50	0.8599	
		70	0.7634	

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From the study, it was found that the best isotherms to model the adsorption process are Thomas and Yoon-Nelson model, based on Equation (8):

$$y_{TH} = \ln\left(\frac{C_o}{C_f} - 1\right) = \ln\left(\frac{C_f}{C_o - C_f}\right)^{-1} = -\ln\left(\frac{C_f}{C_o - C_f}\right) = -y_{YN} \quad (8)$$

This indicates that any graph of Thomas model is the reflection of the graph of Yoon-Nelson model, thus the value of R<sup>2</sup> for Thomas model plots will always be equal to the value of R<sup>2</sup> for Yoon-Nelson model. That is why an adsorption process that is compatible with Thomas model will always be compatible with Yoon-Nelson model, vice versa. Also note however that for the effect of gas flow rate and gas temperature on the adsorption process by hydrogel biochar, the chosen isotherm was Adam-Bohart in spite the lower value of R<sup>2</sup>. This is because the linear equations generated and the general equation for Thomas and Yoon-Nelson model were mismatched.

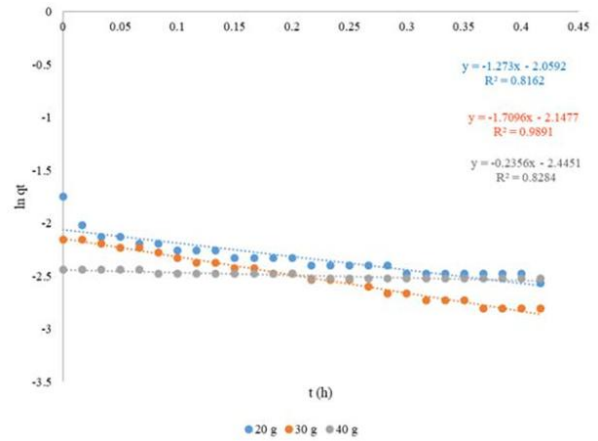
**Table-IV: Summary of the Isotherm Studies**

Adsorbent	Study Parameter	Best Isotherm Model	Best Para meter	Constants
Activated biochar	Mass	Thomas Yoon-Nelson	30g	$K_{TH} = 0.0033$ $q_o = 0.0422$ $K_{YN} = 0.1136$ $\tau = 22.0766$
	Gas Flow rate	Thomas Yoon-Nelson	0.15 m <sup>3</sup> /hr	$K_{TH} = 0.00196$ $q_o = 0.0520$ $K_{YN} = 0.0682$ $\tau = 17.9282$
	Temp	Thomas Yoon-Nelson	30°C	$K_{TH} = 0.0033$ $q_o = 0.0422$ $K_{YN} = 0.1136$ $\tau = 22.0766$
Hydrogel biochar	Mass	Thomas Yoon-Nelson	40g	$K_{TH} = 0.0015$ $q_o = 0.0312$ $K_{YN} = 0.0517$ $\tau = 21.7060$
	Gas Flow rate	Adam Bohart	0.15 m <sup>3</sup> /hr	$K_{AB} = 0.7972$
	Temp	Adam Bohart	70°C	$K_{AB} = 0.2610$

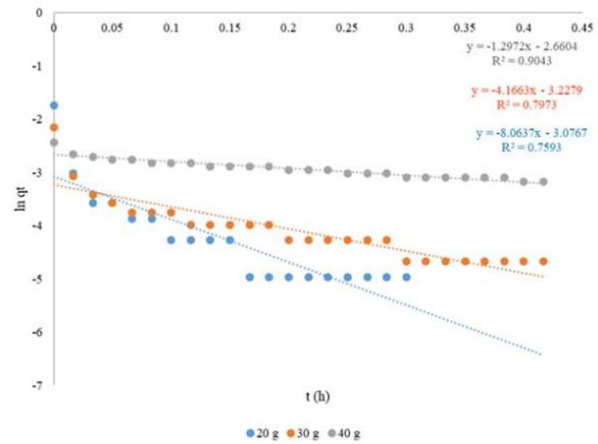
The reason to this was because the exit concentration reached more than half the inlet concentration before it reaches the first minute. This would cause the graph to fall below the x-axis, causing the y-intercept to be negative, subsequently causing the adsorption parameter to be negative.

### C. Kinetic Studies

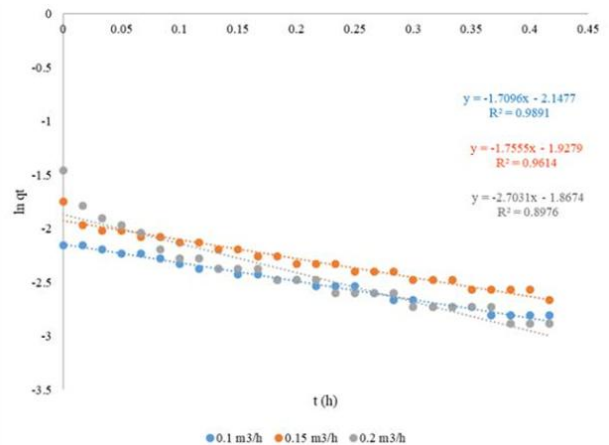
For pseudo-first order and second order kinetic equation, graphs of Ln qt vs t are plotted. The coefficient of correlation is then computed, and shown in Fig. 7 to Fig. 18 for all parameters.



**Fig. 7. Pseudo-first order kinetic plot for the effect of activated biochar adsorbent mass**



**Fig. 8. Pseudo-first order kinetic plot for the effect of hydrogel biochar adsorbent mass**



**Fig. 9. Pseudo-first order kinetic plot for the effect of gas flow rate on the adsorption by activated biochar**



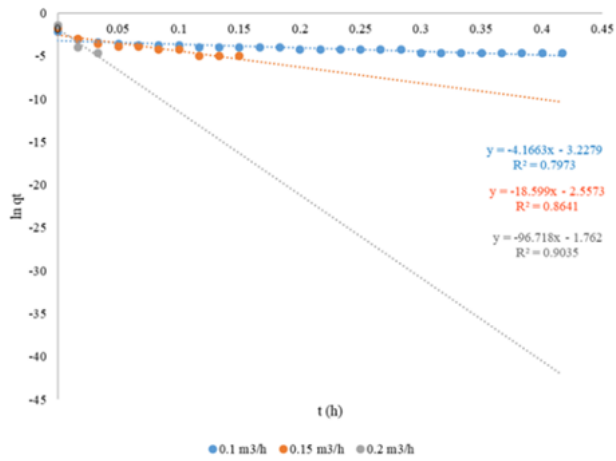


Fig. 10. Pseudo-first order kinetic plot for the effect of gas flow rate on the adsorption by hydrogel biochar

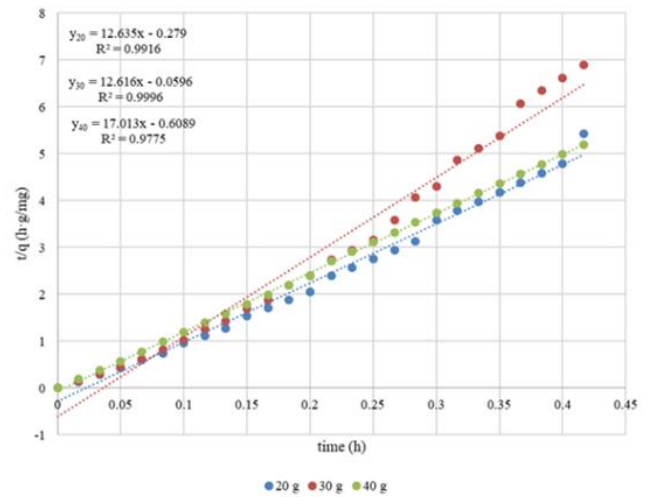


Fig. 13. Pseudo-second order kinetic plot for the effect of activated biochar adsorbent mass

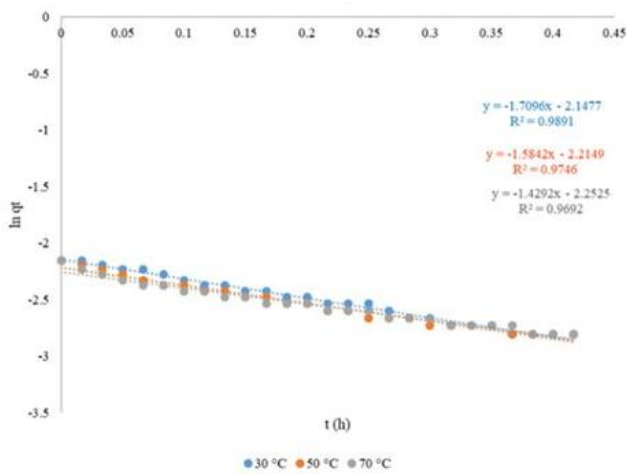


Fig. 11. Pseudo-first order kinetic plot for the effect of gas temperature on the adsorption by activated biochar

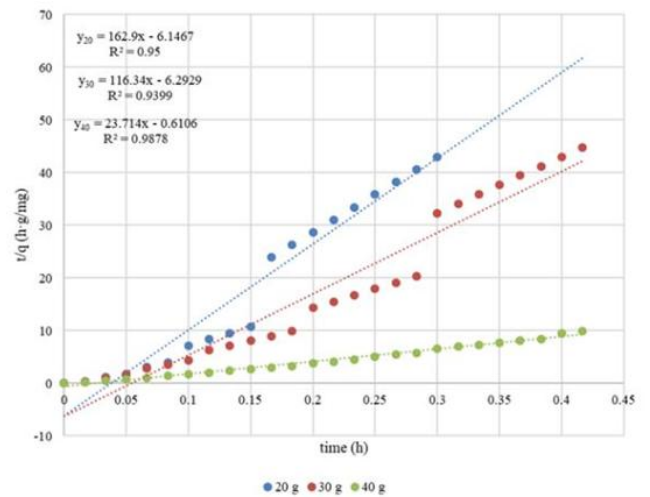


Fig. 14. Pseudo-second order kinetic plot for the effect of activated biochar adsorbent mass

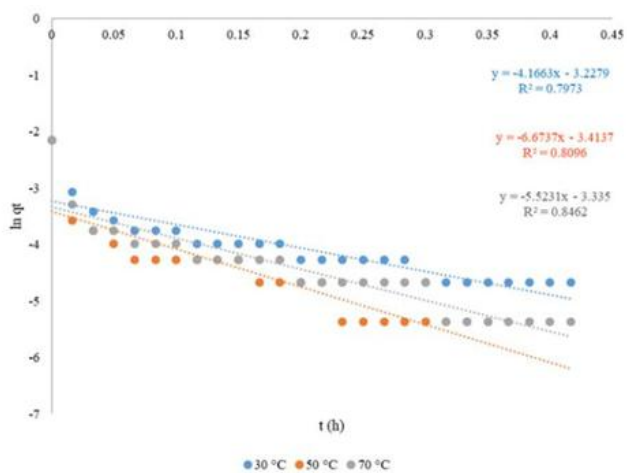


Fig. 12. Pseudo-first order kinetic plot for the effect of activated biochar adsorbent mass

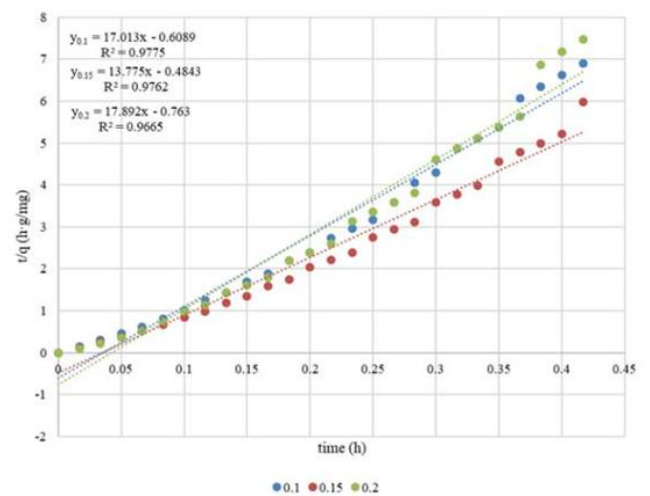
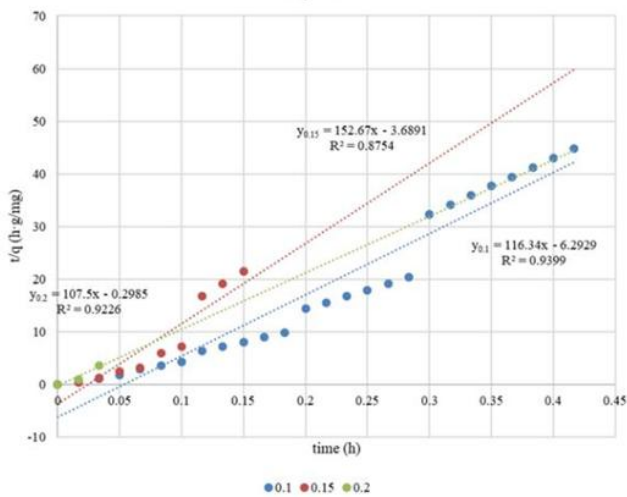
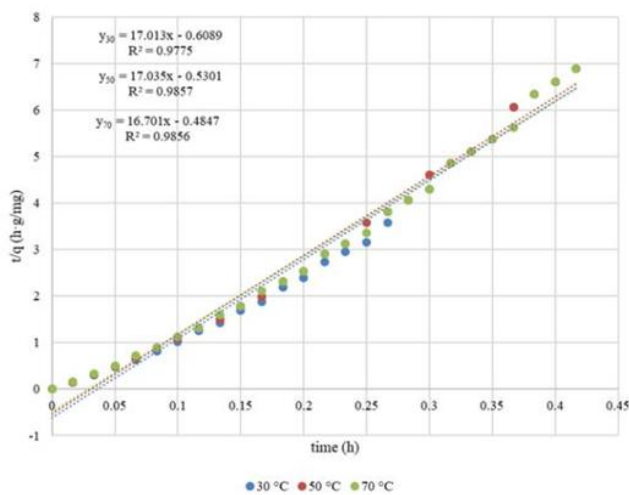


Fig. 15. Pseudo-second order kinetic plot for the effect of gas flow rate on the adsorption by activated biochar

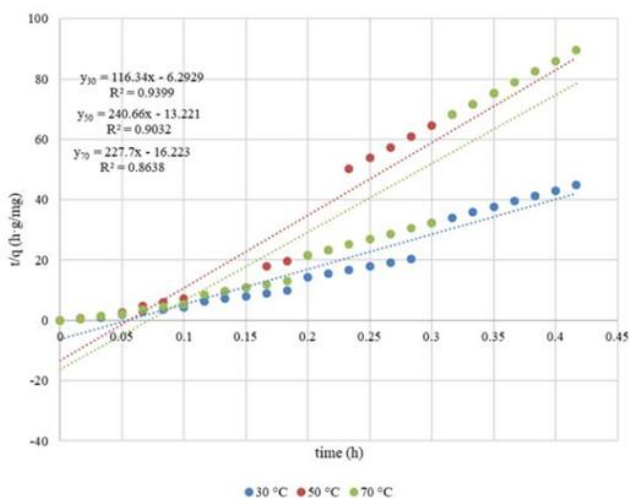
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**Fig. 16.** Pseudo-second order kinetic plot for the effect of gas flow rate on the adsorption by hydrogel biochar



**Fig. 17.** Pseudo-second order kinetic plot for the effect of gas temperature on the adsorption by activated biochar



**Fig. 18.** Pseudo-first order kinetic plot for the effect of gas flow rate on the adsorption by hydrogel biochar

From the summary of the twelve graphs shown above, as in Table-V and Table-VI, it was found that the adsorption processes using activated biochar fits well into both pseudo-first order kinetic equation and pseudo-second order kinetic equation. This indicates that the adsorption using activated biochar is both physisorption (first order) and chemisorption (second order). Meanwhile, for the adsorption processes using hydrogel biochar, it fit well into pseudo-second order kinetic equation.

Karaman (2016) from Al-Quds University explained that when the rate-limiting step of one particular adsorption process is the diffusion and is independent of the concentration of the adsorbate, it can be expressed using the first order equation [6]. Meanwhile, when there is a significant chemical reaction involved in the adsorption process and the kinetics corresponds to competitiveness of the adsorption and the magnitude of sorbate/sorbent ratio, it can be expressed using the second order equation [6].

**Table-V:** Summary of pseudo-first order kinetic equation

Adsorbent	Parameter		R <sup>2</sup>	Yes /No	Remarks
Activated Biochar	Adsorbent Mass (g)	20	0.8162	NO	Inconsistent
		30	0.9891		
		40	0.8284		
	Gas flow rate (m <sup>3</sup> /h)	0.10	0.9891	YES	R <sup>2</sup> >0.8
		0.15	0.9614		
		0.20	0.8976		
Gas temperature (°C)	30	0.9891	YES	R <sup>2</sup> >0.8	
	50	0.9746			
	70	0.9692			
Hydrogel Biochar	Adsorbent Mass (g)	20	0.7593	NO	Inconsistent
		30	0.7973		
		40	0.9043		

Gas flow rate (m <sup>3</sup> /h)	0.10	0.7973	NO	Inconsistent
	0.15	0.8641		
	0.20	0.9035		
Gas temperature (°C)	30	0.7973	NO	Inconsistent
	50	0.8096		
	70	0.8462		

Gas flow rate (m <sup>3</sup> /h)	40	0.9878	YES	R <sup>2</sup> >0.8
	0.10	0.9399		
	0.15	0.8754		
Gas temperature (°C)	0.20	0.9226	YES	R <sup>2</sup> >0.8
	30	0.9399		
	50	0.9302		
	70	0.8638		

The deduction from this statement is that both diffusion and competitive adsorption exist in the deduction from this statement is that both diffusion and competitive adsorption exist in the adsorption using activated biochar, while there was only competitive adsorption existing in the adsorption using hydrogel biochar. these phenomena may be due to difference in surface characteristics as well as physical characteristics of the adsorbents such as particle size, film thickness, porosity, etc.

**Table-VI: Summary of pseudo-second order kinetic equation**

Adsorbent	Parameter	R <sup>2</sup>	Yes /No	Remarks	
Activated Biochar	Adsorbent Mass (g)	20	0.9916	YES	R <sup>2</sup> >0.8
		30	0.9996		
		40	0.9775		
	Gas flow rate (m <sup>3</sup> /h)	0.10	0.9775	YES	R <sup>2</sup> >0.8
		0.15	0.9762		
		0.20	0.9665		
	Gas temperature (°C)	30	0.9775	YES	R <sup>2</sup> >0.8
		50	0.9857		
		70	0.9856		
Hydrogel Biochar	Adsorbent Mass (g)	20	0.9500	YES	R <sup>2</sup> >0.8
		30	0.9399		

**IV. CONCLUSIONS**

The three research objectives were successfully achieved. The best equilibrium isotherm has been identified which are Thomas Model and Yoon-Nelson model, and the kinetic modelling has been conducted. The exception applies to when the process encountered the situation when the exit concentration is greater than half the inlet concentration. At this point, Thomas Model and Yoon-Nelson Model became mathematically inconsistent with the linearly regressed data, and thus Adam-Bohart Model was used.

It was found that the adsorption process using activated biochar adsorbent is both chemisorption and physisorption due to the fact that they were effectively represented using both pseudo-first order kinetic equation and pseudo-second order kinetic equation respectively. Meanwhile, the adsorption process using hydrogel biochar is chemisorption because it only fits effectively into pseudo-second order kinetic equation.

**V. ACKNOWLEDGEMENT**

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