

Full Length Research Paper

Kinetic Adsorption Study of Glucose Osmotic Agent onto Granular Activated Carbon in SET Technique

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Received 01 May 2014; Accepted 16 June 2014

Abstract. In the previous experimental work, the granular activated carbon (GAC) had shown a capability to adsorb glucose from its aqueous solution prior the regeneration step (RO or NF membrane) in a Manipulated Osmosis Desalination system (MOD) when using the glucose as an osmotic agent. In this paper, the kinetics adsorption of glucose onto commercial granular activated carbon (mesh, 12-20) is described. The glucose adsorption rate (q) onto GAC within an 8 hr contact time using three different GAC samples doses (5, 10 and 30 g/l) at different shaking speed values (45, 100, 145 rpm) is investigated. The adsorption kinetic models: pseud first order, pseud second order, intra-particle diffusion and mass transfer models are used for analyzing glucose sorption by GAC. The finding indicates that glucose sorption onto GAC is well represented by the Pseud second order kinetic model.

Keywords: GAC, glucose, adsorption, Kinetic, MOD

1. INTRODUCTION

In the first part of this work (Sami et al., 2014), the reliability of Solute Exchange technique (SET) to be used in the Manipulated Osmosis Desalination System (MOD) was examined. The reliability of this technique was investigated by examining the adsorption capacity of specific organic compounds such as glucose and maltose onto commercial granular activated carbon (mesh 12-20) manufactured by Sigma Aldrich Company in the UK. Glucose and maltose could be used as Osmotic Agents in MOD system as the previous study explained. The parameters affecting sorbate adsorption onto GAC are pH of the solution, the initial solution concentration, shaking time and speed, sorbent dose and temperature: these are also investigated experimentally. Additionally, the adsorption equilibrium isotherm was tested using the common isotherm modules; Langmuir and Freundlich. GAC exhibited a capability to adsorb glucose and maltose from their aqueous solutions. Also the glucose adsorption process was chemisorption and in a good agreement with the Freundlich isotherm model, while, the maltose adsorption process was a chemisorption and in a good agreement with Langmuir isotherm model. Based on

the findings in the first part of the study, the SET is sufficiently reliable to be used with an MOD system which leads mainly to reduce energy consumption and accordingly, a reduction in the cost of water produced

From the mechanistic viewpoint, in order to interpret the experimental data, prediction of the rate limiting step is an important factor to be considered in the sorption process. Both kinetic and equilibrium isotherm studies help to identify the sorption process, predicting the mechanism required for design purposes. For solid liquid sorption process, the solute transfer is usually characterized by either external mass transfer boundary layer diffusion or intraparticle diffusion or both. Generally, the sorption dynamics can be described by the following three consecutive steps (Walter and Weber, 2014).

(a) Transport of the solute from the bulk solution through the liquid film to the adsorbent exterior surface

(b) Solute diffusion into the pore of an adsorbent except for a small quantity of sorption on the external surface; parallel to this is the intraparticle transport mechanism of the surface diffusion.

(c) Sorption of solute on the interior surface of the pores and capillary spaces of the adsorbent.

The last step is considered to be an equilibrium reaction of the three steps. The third step is assumed to be rapid and considered to be negligible. The overall rate of sorption will be controlled by the slowest step which would be either film diffusion or pore diffusion. However, the controlling step may be distributed between intraparticle and external transport mechanisms. Whatever the case, the external diffusion will be involved in the sorption process. For design purposes, it is necessary to calculate the slowest step involved in the sorption process. For the sorption process, the external mass transfer controls the sorption process for the systems that have poor mixing, a dilute concentration of adsorbate, small particle sizes of adsorbent and a higher affinity with adsorbate than adsorbent. On the other hand, the intra-particle diffusion will control the sorption process for a system with good mixing, large particle sizes of adsorbent, high concentration of adsorbate, and lower affinity with adsorbate than adsorbent (Walter and Weber, 2014). The most common kinetic models that have been used in the kinetic adsorption studies and in this study are presented in the following sections. (Hameed et al., 2006).

1.1. The pseudo-first-order kinetic model of Lagergren

The Lagergren model for the sorption of liquid / solid system based on solid capacity can be expressed as follows (Hameed et al., 2006):

$$\frac{dq_t}{dt} = K_1(q_e - q_t) \quad (1)$$

Where: q_t : sorbed amount at time, t ; q_e : sorbed amount at equilibrium; K_1 : the rate constant of pseudo first order sorption process.

The integrated rate law, after applying the initial conditions of $q_t = 0$ at $t = 0$ is:

$$\log(q_e - q_t) = \log(q_e) - \frac{K_1}{2.303} t \quad (2)$$

Plot of $\log(q_e - q_t)$ versus t should give a linear relation from which K_1 and q_e can be determined from slope and intercept of plot respectively, a straight line for pseudo-first-order kinetics, which allows computation of the sorption rate constant (K_1). If the experimental results do not follow equation 1.1 and 1.2 they differ in two important aspects:

1- $K_1(q_e - q_t)$ is not represent the number of available sites

2- $\log(q_e)$ is not equal to the intercept of the plot of $\log(q_e - q_t)$ against (t) (Hameed et al., 2006).

1.2. Ho and McKay Equation (pseudo-second-order)

The pseudo-second-order chemisorptions kinetics may be expressed as ((Hameed et al., 2006, Ozkaya, 2006, Abia et al., 2006 and Belhachemi et al., 2009):

$$\frac{dq_t}{dt} = k_2(q_e - q_t)^2 \quad (3)$$

Where: K_2 : is the rate constant of sorption; By separating the variables in equation (3) gives:

$$\frac{dq_t}{(q_e - q_t)^2} = k_2 dt \quad (4)$$

Integrating this for boundary conditions $t = 0$ to $t = t$ and $q_t = 0$ to $q_t = q_t$ gives:

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

By integrate rate law, for a pseudo second-order-reaction Equation (4) can be rearranged to obtain:

$$q_t = \frac{1}{\left(\frac{1}{k_2 q_e}\right) + \left(\frac{t}{q_e}\right)} t \quad (6)$$

This has a linear form of:

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

If the initial sorption rate is

$$h = k_2 q_e^2 \quad (8)$$

$$q_t = \frac{t}{\left(\frac{1}{h}\right) + \left(\frac{t}{q_e}\right)} \quad (9)$$

And

$$\frac{t}{q_t} = \frac{1}{h} + \frac{1}{q_e} t \quad (10)$$

Plot of (t/q_t) and t of equation should give a linear relationship, from which q_e and k_2 , can be determined from slop and intercept of plot respectively (Hameed et al., 2006).

1.3. The intra particle diffusion model

In order to gain insight into the mechanisms and rate controlling steps affecting the kinetics of adsorption, the kinetic experimental results were fitted to the Weber's intra-particle diffusion (Hameed et al., 2006). The kinetic results were analysed by the intra-particle diffusion model to elucidate the diffusion mechanism, whose model is expressed as:

$$q_t = k_{id} t^{1/2} + C \quad (11)$$

Where, C_0 is the intercept and k_{id} is the intra-particle diffusion rate constant ($\text{mg/g h}^{1/2}$), which can be evaluated from the slope of the linear plot of qt versus $t^{1/2}$ (Hameed et al., 2006). The intercept of the plot reflects the boundary layer effect. The larger the intercept, the greater contribution of the surface sorption in the rate controlling step. If the regression of qt versus $t^{1/2}$ is linear and passes through the origin, then intra-particle diffusion is the sole rate-limiting step. However, the linear plots at each concentration did not pass through the (El Nemr et al., 2008, and Vadivlan et al., 2005): origin. This indicates that the intra-particle diffusion was not only rate controlling step.

1.4. The mass transfer model

The mass transfer model can be expressed as bellow (Vadivlan et al., 2005, Hameed et al., 2008, and Qadeer and Akhtars, 2005):

$$C_0 - C_t = D \exp(K_0 t) \quad (12)$$

Where: C_0 : is the initial ion concentration (mg/l); C_t : is the ion concentration at time, t ; t : is the contact time (shaking time) min; D : is the fitting parameter; K_0 : is a constant which is the mass transfer adsorption coefficient; A linearized form of equation: is:

$$\ln(C_0 - C_t) = \ln D + K_0 t \quad (13)$$

If the sorption of ion is depicted by the mass transfer model, then plot $\ln(C_0 - C_t)$ vs time (t) give a linear relationship from where $\ln D$ (is a measure of the apparent distribution ratio of adsorbed ion between the bulk solution and the adsorbent surface) and K_0 can be determined from the intercept and slope of the plot respectively.

The aim of this study is to investigate experimentally the kinetics adsorption of glucose onto commercial granular activated carbon (Darco type) with mesh (12-20). The glucose adsorption rate (qt) onto GAC within an 8 hr contact time using three different GAC samples doses at different shaking speed values is investigated.

2. THE EXPERIMENTAL WORK

2.1. Equipments

A four digits weight balance (Sartorius research type) was used in this investigation. For pH measurements, a pH- meter (Mettler-Toledo Company in UK) was used with a standard electrode. All experiments were carried out using water- bath shaker (Mickle Laboratory Eng. Co England) with variable speed (0-200 rpm) at temperature varied between 20-40°C. The concentrations of glucose were determined using HPLC instrument (Varian 385-LC ELSD with Evaporative Light Scattering Detector Column and with mobile phase 80% acetonitrile, flow rate 3.0 ml/min).

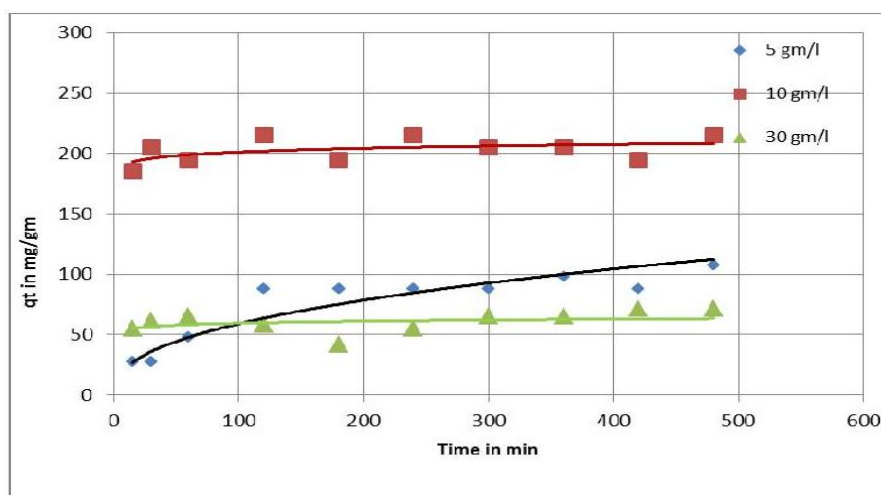


Fig.1: Adsorption capacity profile of glucose with time onto different GAC sample doses when initial concentration solution is 6000mg/l and shaking speed 45 rpm

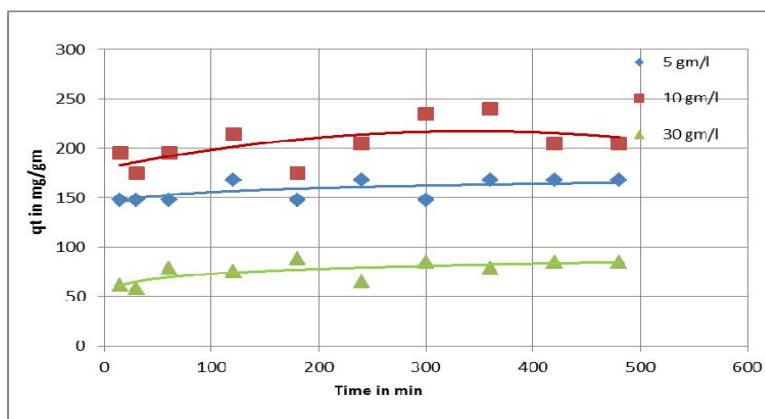


Fig. 2: Adsorption capacity profile of glucose with time onto different GAC sample doses when initial concentration solution is 6000 mg/l and shaking speed 100 rpm

2.2. Materials and reagents

Commercial granular activated carbon Darco with mesh (12-20) and D-(+) glucose compound with good purity ($\geq 99.5\%$) supplied by Sigma Aldrich company in the UK was used in this study. Also, analytical reagent grade sodium hydroxide (Assay: 98.05 %) and laboratory reagent grade hydrochloric acid solution (Assay: 35.38%) supplied by Fisher Scientific Co in the UK were used to alter the pH of solutions. All solutions and reagents were prepared by dissolving the weighed amount of these chemicals in deionized water.

2.3. Methods& Procedures

In this study, three kinetic experiments sets were carried out. In the first set, GAC samples of 0.5g were shaken separately with 100ml of 6000 mg/l glucose concentration solution at different shaking speeds of 45,100 and 145rpm for 8 hr. This experiments set was performed at an ambient temperature with a pH range

7-8. In each experiment, within suggested intervals of time during the 8 hr shaking time, one mixture was taken from the shaker and then the GAC sample was separated by filtration using Whitman filter paper grade-1 and filtrate solution samples were collected. The same procedure followed in the first set was repeated for the second and third experiments sets using two GAC samples of 1 and 3 g respectively. The glucose concentrations in the filtered solutions were determined using HPLC equipment and all the results of this study are presented in Figs.1-15 and in Tables.1 and 2.

3. RESULTS AND DISCUSSIONS

It is clear from Figs.1-2, and 3, that at different shaking speeds, the adsorption rate of glucose onto GAC was found to be rapid at the initial period of contact time and then became slow and stagnated with increased contact time. Moreover, by increasing the shaking speeds the higher sample dose showed more stability with a low adsorption rate.

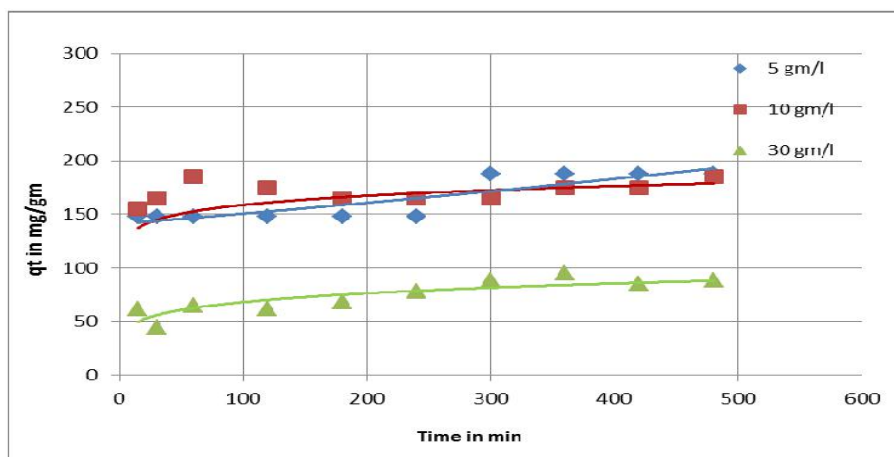


Fig. 3: Adsorption capacity profile of glucose with time onto different GAC sample doses when initial concentration solution is 6000 mg/l and shaking speed 145 rpm

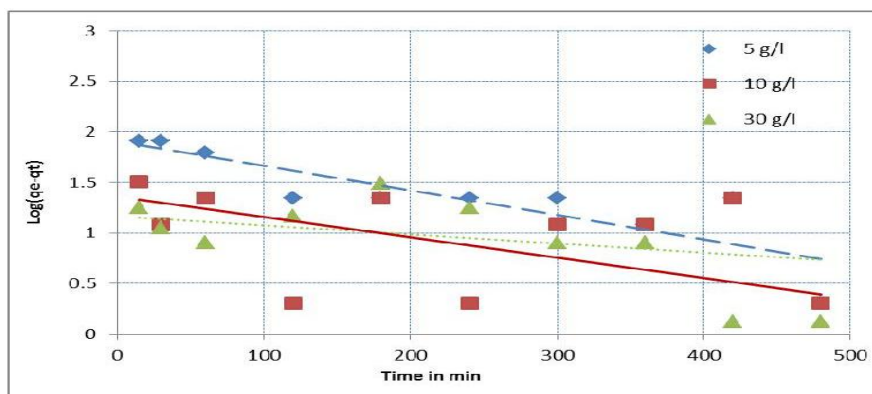


Fig. 4: Pseudo first order kinetic plot for glucose adsorption at different GAC sample doses when shaking speed 45 rpm and initial concentration solution is 6000mg/l

The Pseudo first order, pseudo second order, intra-particle diffusion and mass transfer kinetic models are employed in this work. The obtained parameters for all kinetic models are presented in Figs.4-15 and in Tables 1 and 2.

In most of the cases, the first order equation of Lagergren did not apply throughout the contact time and is generally applicable over the initial (20-30min) sorption. As seen from Table 1, the correlation coefficient was much higher in the pseudo second order rate equation compared with other models. Also, from this table, it was observed that calculated equilibrium sorption capacity (q_e) at different shaking speeds and GAC sample doses for all models were not close to the experimental values except for pseudo second order. This gives an indication that the pseudo second order model is agreed well with the experimental data values. From Table 2 it was observed that, the intra-

particle diffusion rate generally decreased with GAC sample doses increases at different shaking speed values (45-145), also, it decreased with mixture shaking speed increases. The value of intercept (c) which representing the boundary layer effect is generally increased with the GAC sample dose increases. While it decreased with shaking speed increases for all GAC sample doses used in this study. Furthermore, The line of plotting qt vs $t^{1/2}$, has not passed through the origin, which gives an indication that the intra-particle diffusion not only rate controlling step and that some other mechanisms are also involved. From Table 2, it can be seen that the parameter factor (D) increased with GAC sample dose and shaking speed increases, while the mass transfer adsorption coefficient (K_o) in general increased with GAC sample doses increases, whilst, it decreased with shaking time increases.

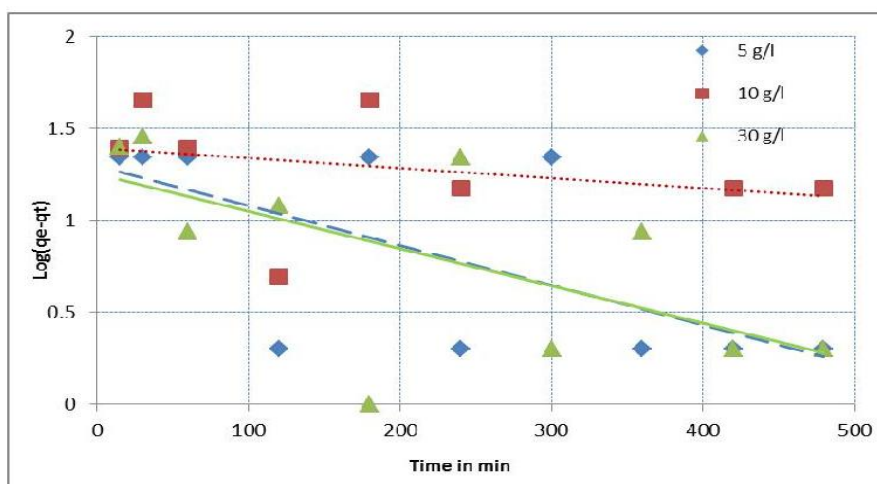


Fig. 5: Pseudo first order kinetic plot for glucose adsorption at different GAC sample doses when shaking speed 100 rpm and initial concentration solution is 6000mg/l

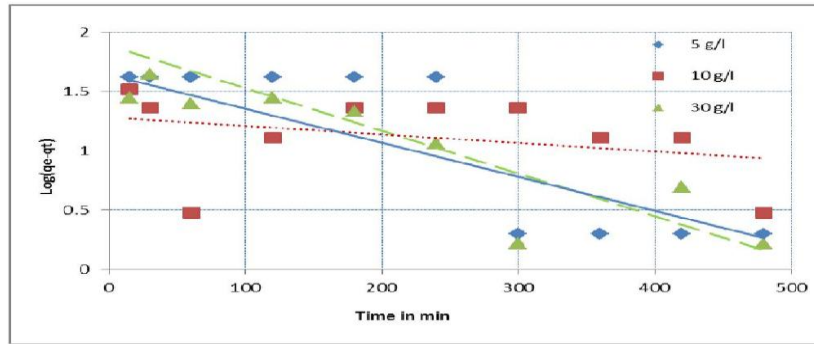


Fig. 6: Pseudo first order kinetic plot for glucose adsorption at different GAC sample doses when shaking speed 145 rpm and initial concentration solution is 6000mg/l

Pseudo Second order model figures

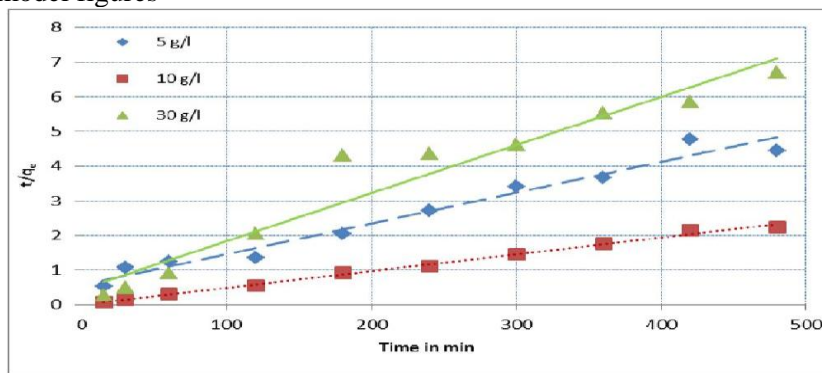


Fig. 7: Pseudo second order kinetic plot for glucose adsorption at different GAC sample doses when shaking speed 45 rpm and initial concentration solution is 6000mg/l

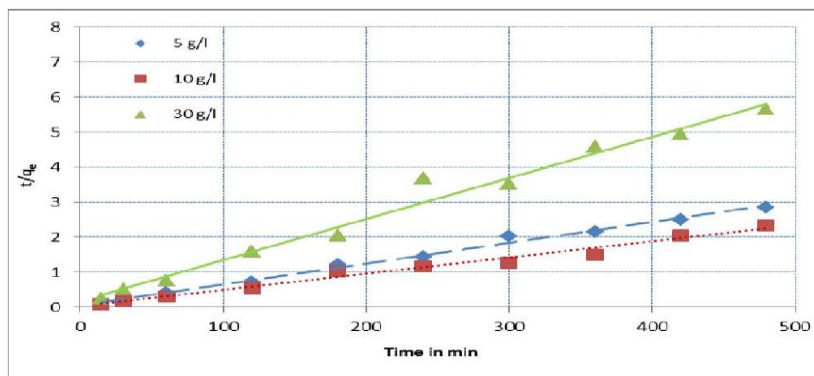


Fig. 8: Pseudo second order kinetic plot for glucose adsorption at different GAC sample doses when shaking speed 100 rpm and initial concentration solution is 6000mg/l

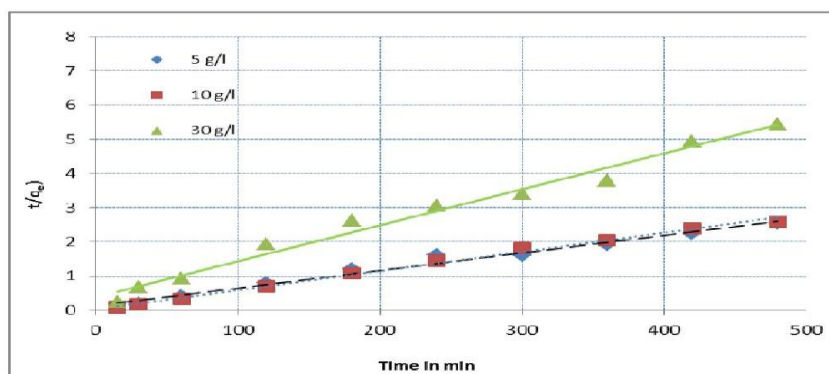


Fig. 9: Pseudo second order kinetic plot for glucose adsorption at different GAC sample doses when shaking speed 145 rpm and initial concentration solution is 6000mg/l

Intra particle model figures:

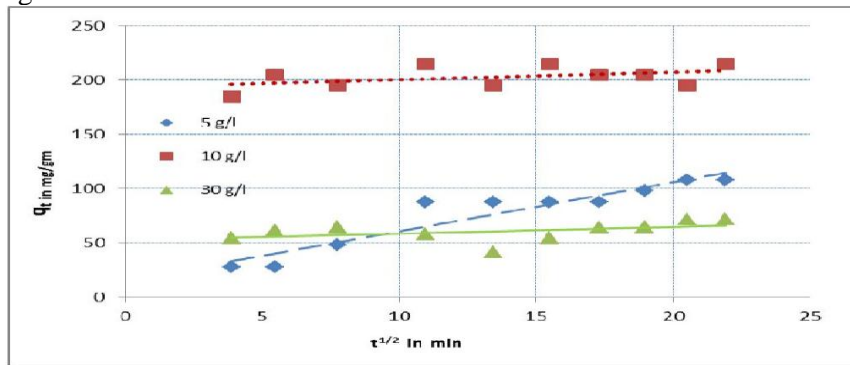


Fig. 10: Intra particle kinetic plot for glucose adsorption at different GAC sample doses when shaking speed 45 rpm and initial concentration solution is 6000mg/l

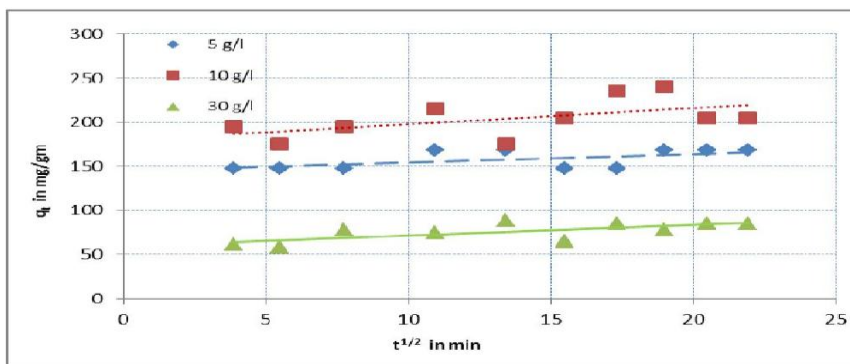


Fig.11.Intra particle kinetic plot for glucose adsorption at different GAC sample doses when shaking speed 100 rpm and initial concentration solution is 6000mg/l

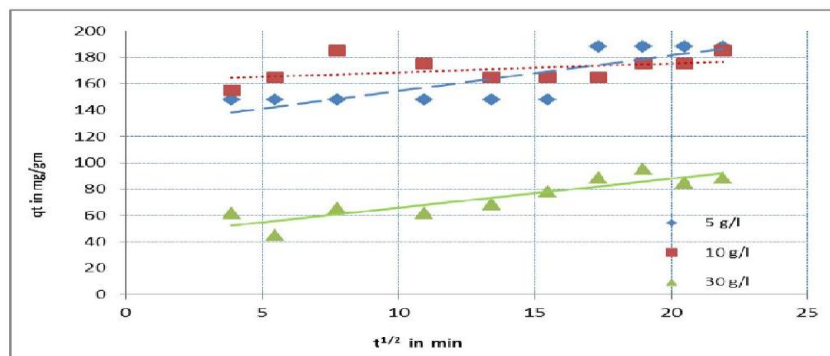


Fig. 12: Intra particle diffusion kinetic plot for glucose adsorption at different GAC sample doses when shaking speed 145 rpm and initial concentration solution is 6000mg/l

Mass transfer model figures:

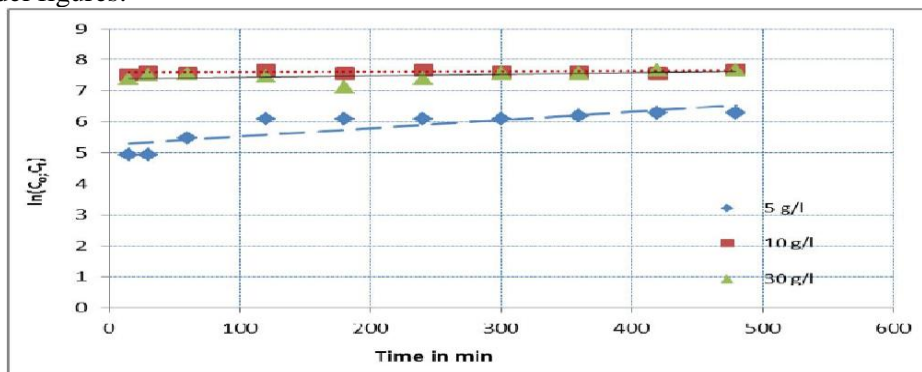


Fig. 13: Mass transfer kinetic plot for glucose adsorption at different GAC sample doses when shaking speed 45 rpm and initial concentration solution is 6000mg/l

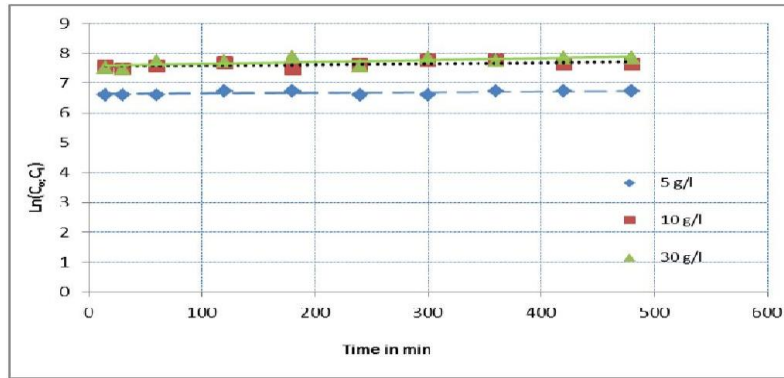


Fig. 14: Mass transfer kinetic plot for glucose adsorption at different GAC sample doses when shaking speed 100 rpm and initial concentration solution is 6000mg/l

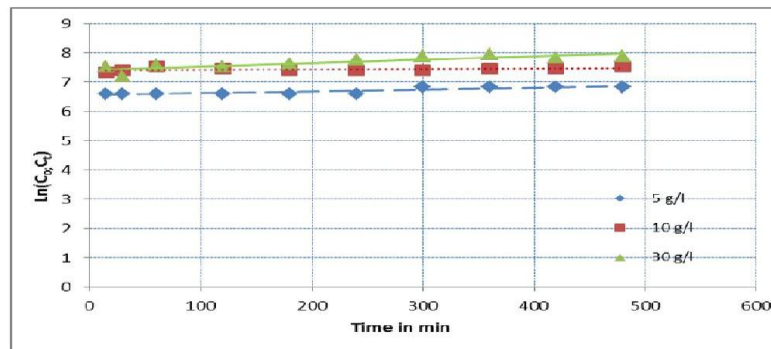


Fig.15.Mass transfer kinetic plot for glucose adsorption at different GAC sample doses when shaking speed 145 rpm and initial concentration solution is 6000mg/l

Table 1: kintec parameters for glucose adsorption at different; GAC sample doses and shaking speeds (rpm) when initial concentration solution is 6000mg/L

Model	gm/l	45 rpm		100 rpm		145 rpm	
		Eq.	R ²	Eq.	R ²	Eq.	R ²
Pseudo 1st order	5	y = -0.0024x + 1.9067	0.735	y = -0.0022x + 1.2983	0.430	y = -0.0036x + 1.8872	0.768
	10	y = -0.0009x + 1.1679	0.099	y = -0.0005x + 1.3962	0.095	<u>y = -0.0007x + 1.2844</u>	<u>0.105</u>
	30	y = -0.002x + 1.3626	0.502	y = -0.002x + 1.2536	0.396	y = -0.0029x + 1.6455	0.790
Pseudo 2nd order	5	y = 0.0089x + 0.5771	0.970	y = 0.0059x + 0.0484	0.993	y = 0.0051x + 0.1348	0.981
	10	y = 0.0048x + 0.0084	0.995	y = 0.0046x + 0.0272	0.981	<u>y = 0.0056x + 0.0391</u>	<u>0.995</u>
	30	y = 0.0139x + 0.4595	0.941	y = 0.0118x + 0.1648	0.980	y = 0.0105x + 0.3806	0.981
Intraparticle diffusion	5	y = 4.5488x + 15.293	0.889	y = 0.9826x + 144.67	0.351	y = 2.6796x + 127.65	0.682
	10	y = 0.7151x + 193.3	0.194	y = 1.859x + 179.28	0.299	y = 0.6707x + 161.9	0.195
	30	y = 0.6232x + 52.545	0.194	y = 1.1835x + 59.945	0.489	y = 2.2236x + 43.502	0.802
Mass transfer	5	y = 0.0026x + 5.2654	0.693	<u>y = 0.0002x + 6.6187</u>	<u>0.336</u>	y = 0.0007x + 6.5589	0.768
	10	y = 0.0001x + 7.5884	0.149	y = 0.0003x + 7.5448	0.275	<u>y = 0.0002x + 7.409</u>	<u>0.204</u>
	30	y = 0.0005x + 7.3996	0.228	y = 0.0006x + 7.5939	0.424	y = 0.0012x + 7.4143	0.748

4. CONCLUSSIONS

According to the experimental results of glucose sorption kinetic study onto GAC, the following points can be concluded:

(a) In this kinetic sorption study, the GAC higher sample dose showed more stability with low adsorption rate when increasing shaking speeds values.

(b) The correlation coefficient value obtained when applying the Pseudo second order rate equation was much higher compared with other models, Also, the calculated equilibrium sorption capacity (qe) at different shaking speeds and GAC samples doses for all models are not close to the experimental values except for the pseudo second order model. Thus the pseudo second order model agrees well with experimental data values.

Table 2: kintec model parameters of glucose adsorption onto GAC

rpm	45 rpm				100 rpm				145 rpm			
GAC Doses gm/l					Pseudo 1st order model							
	K1	q _e exp	q _e cal	R ²	K1	q _e exp	q _e cal	R ²	K1	q _e exp	q _e cal	R ²
5	5.527x10 ⁻³	110	80.67	0.735	5.067x10 ⁻³	170	19.87	0.430	8.291x10 ⁻³	190	77.12	0.768
10	2.073x10 ⁻³	217	14.72	0.099	1.152x10 ⁻³	220	24.9	0.095	1.612x10 ⁻³	188	19.25	0.105
30	4.606x10 ⁻³	73	23.05	0.532	4.606x10 ⁻³	87	17.93	0.396	6.679x10 ⁻³	90	44.21	0.790
GAC gm/l	Pseudo 2nd order model											
	K ₂	q _e exp	q _e cal	R ²	K ₂	q _e exp	q _e cal	R ²	K ₂	q _e exp	q _e cal	R ²
5	1.373x10 ⁻⁴	110	112.3	0.97	7.192x10 ⁻⁴	170	169.5	0.993	1.930x10 ⁻³	190	196.1	0.981
10	2.734x10 ⁻³	217	208.3	0.995	0.770x10 ⁻³	220	217.4	0.981	0.802x10 ⁻³	188	178.6	0.995
30	1.078x10 ⁻³	73	71.94	0.941	0.845x10 ⁻³	87	84.75	0.980	0.290x10 ⁻³	90	95.23	0.981
GAC gm/l	Intra-particle diffusion model											
	K _{id}	C		R ²	K _{id}	C		R ²	K _{id}	C		R ²
5	4.549	15.29		0.889	0.983	144.67		0.351	2.680	127.65		0.682
10	0.715	193.3		0.194	1.859	179.28		0.299	0.673	161.9		0.195
30	0.623	52.54		0.194	1.184	59.945		0.489	2.224	43.502		0.802
GAC gm/l	Mass transfer kinetic model											
	K _o	D		R ²	K _o	D		R ²	K _o	D		R ²
5	2.6x10 ⁻³	193.5		0.693	2x10 ⁻⁴	748.97		0.174	7x10 ⁻⁴	705.5		0.768
10	1x10 ⁻⁴	1975		0.149	3x10 ⁻⁴	1890		0.275	2x10 ⁻⁴	1651		0.204
30	5x10 ⁻⁴	1852		0.054	6x10 ⁻⁴	1986		0.424	1.2x10 ⁻³	1660		0.748

(c) It is recommended to exam the sorption kinetic of different sugar types such as; maltose and sucrose which can be used as osmotic agent in MOD-SET combined system onto same GAC type used in this study. Moreover, different commercial GAC and that prepared from agricultural by-products can be subjected to the sorption kinetic works so as to specify the most efficient GAC type can be used in MOD-SET system.

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Mr. Hameed B. Mahood, Ph.D student- University of Surrey- Department of Chemical Process Engineering, UK. He is specialist in Multiphase Flow and Heat transfer and their applications in water desalination and renewable energy.



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