

Full Length Research Paper

Kinetic Sorption Study of Sucrose Osmotic Agent onto Granular Activated Carbon in Manipulated Osmosis Desalination System (MOD) Combined with a Solute Exchange Technique (SET)

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Abstract. The previous work elucidated that, the commercial granular activated carbon (GAC) with mesh (12-20) supplied by Sigma Aldrich Company in the UK has a capability to adsorb specific organic compounds such as sucrose from an aqueous solution. As well as other sucrose specifications such as; high solubility and non-toxicity, sucrose can be used as an osmotic agent in Manipulated Osmosis Desalination system (MOD) combined with a Solute Exchange Technique (SET). However, the sorption kinetic of sucrose onto granular activated carbon (mesh, 12-20) is investigated in this study. The sorption rates of sucrose (q_t) onto GAC within 8 hrs contact time using three different GAC sample doses (5, 10 and 30 g/l) at different shaking speed values (45, 100, 145 RPM) are determined. The sorption kinetic models: pseudo-first order, pseudo-second order, intra-particle diffusion and mass transfer were used for analyzing sucrose sorption by GAC. The finding showed that pseudo second order kinetic model was the most appropriate description of sucrose transport.

Keywords: GAC, adsorption, Kinetic, SET, MOD

1. INTRODUCTION

In the previous work, the commercial GAC (mesh 12-20) supplied by Sigma Aldrich Company in the UK exhibited a capability to adsorb specific organic compounds such as; sucrose, glucose and maltose from their aqueous solutions (Sami et al., 2014). This gave an indication that Solute Exchange Technique (SET) combined with a Manipulated Osmosis Desalination System (MOD) was reliable to be used. Interestingly, the MOD - SET combined system leads mainly to reduce energy consumption and accordingly reducing the cost of water produced. From the mechanistic viewpoint 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 and predicting the mechanism which is required for design purposes. For solid liquid sorption processes, 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 (Jr et al., 1974)

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

(b) Solute diffusion into the pores of the 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 sorption rate 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 the adsorbate, small particle sizes of adsorbent and a higher affinity of adsorbate for 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 low affinity of adsorbate for adsorbent (Jr et al. 1974). More interest in this work, the kinetics of sucrose removal will be studied. This is because sorption kinetics is an important parameter in water treatment, since sorption kinetics can be used to predict the rate of sucrose removal from aqueous solutions in the design of appropriate sorption treatment plants. The different kinetic models that will be used to analyses the kinetic data for sucrose sorption are, the pseudo-first-order kinetic model of Lagergren, Ho and McKay's Equation (pseudo-second-order), the intra particle diffusion model, and the mass transfer model.

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 and 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, Belhachemi et al., 2009 and El Nemer et al., 2008)

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

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:

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

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

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

Then equations (6 and 7) become:

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

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 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, the model for which is expressed as:

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

Where C , 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 q_t versus $t^{1/2}$ (Hameed et al., 2006). The intercept of the

plot reflects the boundary layer effect. The larger intercept, the greater contribution of 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 (Vadivelan et al., 2005, Augustine et al., 2004, Hameed et al., 2008 and Qadeer et al., 2005): origin. This indicates that intra-particle diffusion was not the only rate controlling step.

1.4. The mass transfer model

The mass transfer model can be expressed as bellow (Vadivelan et al., 2005, Augustine et al., 2004, Hameed et al., 2008, Qadeer et al., 2005, Kumar et al., 2005, Arivolis, 2008, Ho and Ofomaja, 2006, Ho and McKay, 1998, Dogan et al., 2004 and Goel et al., 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 slop of the plot respectively

2. THE EXPERIMENTAL WORKS

2.1. Equipments

A four digit 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 a water- bath shaker (Mickle Laboratory Eng. Co England) with variable speeds (shaking rate up to 200 rpm) at temperatures varied between 20-40°C. HPLC instrument (Varian 385-LC ELSD with Evaporative Light Scattering Detector Column and with mobile phase 80% acetonitrile, flow rate 3.0 ml/min) was used to determine the concentration of sucrose in this study.

2.2. Materials and reagents

Commercial granular activated carbon Darco with mesh (12-20) supplied by Sigma- Aldrich Company in the UK and 5 kg bags of sucrose powder compound with good purity ($\geq 99.5\%$) supplied by Tate & Lyle Company in the UK were used in this study. Also, analytical reagent grade of sodium hydroxide (Assay: 98.05 %) and laboratory reagent grade of 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 produced in CORA lab.

2.3. Methods and 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 sucrose concentration solution at different shaking speeds of 45, 100 and 145rpm for 8 hrs. These experiment sets were performed at an ambient temperature with a pH range 6-7. In each experiment, and at different intervals of time during the 8 hrs 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 followed in the second and third experiment sets using two GAC samples of 1 and 3g respectively. The sucrose concentrations in the filtered solutions were determined using HPLC equipment and all results of this study are presented in Figs.1-15 and in Tables.1 and 2.

3. RESULTS AND DISCUSSIONS

From Figs.1,2, and 3, It is clear, 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 stagnant with an increase in contact time when using 10 and 30g/l GAC sample doses. Moreover, by increasing the shaking speeds the higher sample dose showed more stability with a low adsorption rate, while for a low sorbent sample dose (5g/l) the sorption capacity increased with time for all shaking speed values used.

The Pseudo first order, pseudo second order, intra-particle diffusion and mass transfer kinetic models were 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, first order equation of Lagergren did not apply throughout the contact time and was generally applicable over initial (20-30min) sorption process time. As seen from

Table.1, the correlation coefficient was much higher in the pseudo second order rate equation compared with other models. From Table 2, it can be observed that the calculated equilibrium sorption capacity (q_e) at different shaking speeds and GAC samples doses for all models are not close to the experimental values except for pseudo second order. This gives an indication that, the pseudo second order model was the most appropriate model for the description of sucrose transfer. Also, from Table.2 and from Figs.4-15, it was observed that, generally, the intra-particle diffusion rate (K_{id}) decreased with increased GAC sample doses at different shaking speed values (45-145), while, it decreased with increased mixture

shaking speed values. However, in this model, the intercept values (c) which represent the boundary layer effect generally increased with increased GAC sample doses and shaking speed values. Furthermore, the line of plotting q_t vs $t^{1/2}$, has not passed through the origin, which indicates that intra-particle diffusion is not the only rate controlling step and some other mechanisms are also involved. From Table.2, it can also be seen that the parameter factor (D) is increased with increased GAC sample dose and shaking speed, while the mass transfer adsorption coefficient (K_o) in general decreased with increased GAC sample doses and shaking time.

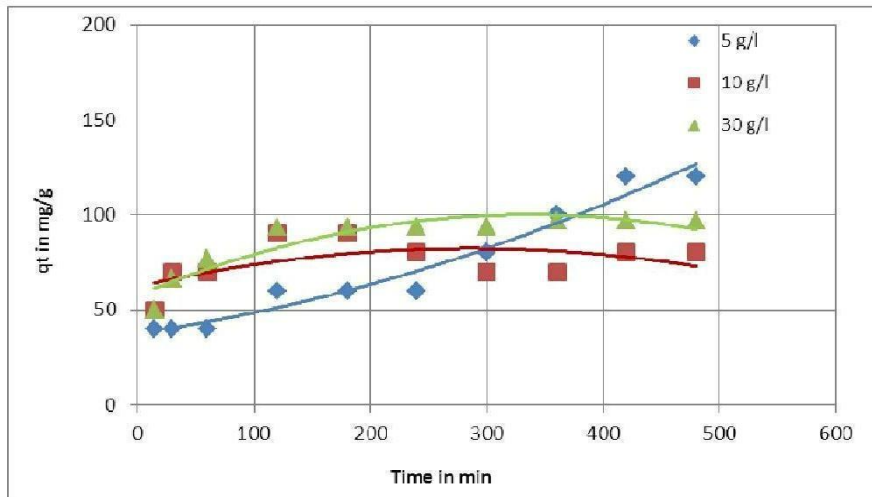


Fig. 1: Sorption capacity profile of sucrose with time onto different GAC sample doses when initial concentration solution is 6000mg/l and shaking speed 45 rpm

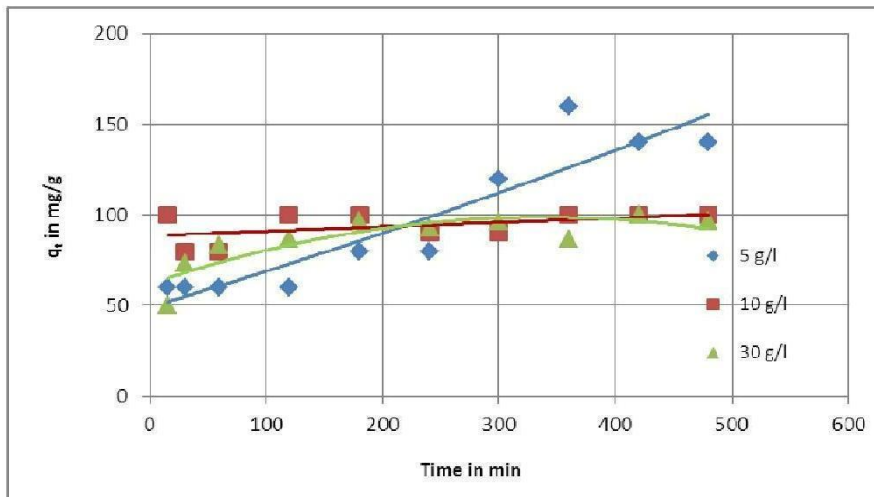


Fig. 2: Sorption capacity profile of sucrose with time onto different GAC sample doses when initial concentration solution is 6000 mg/l and shaking speed 100 rpm

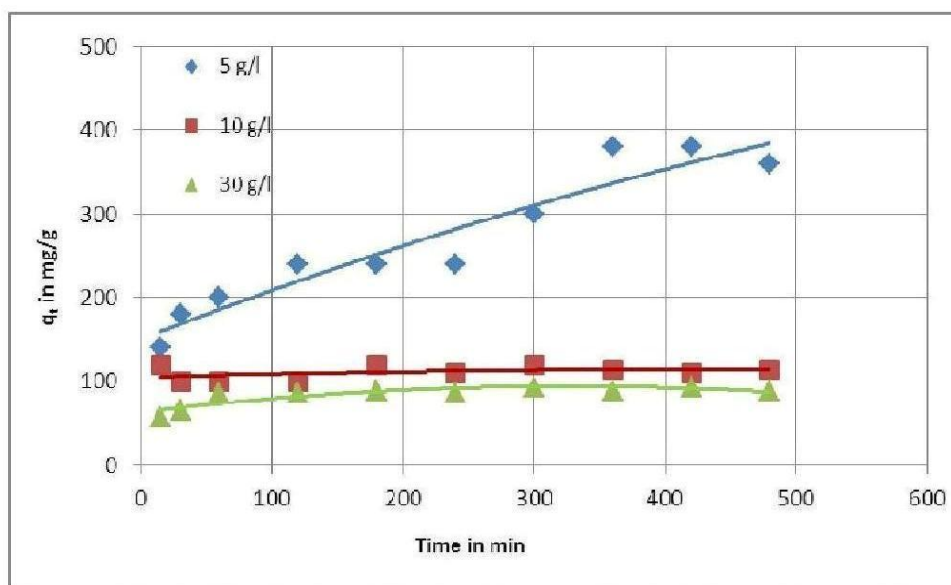


Fig. 3: Sorption capacity profile of sucrose with time onto different GAC sample doses when initial concentration solution is 6000 mg/l and shaking speed 145 rpm

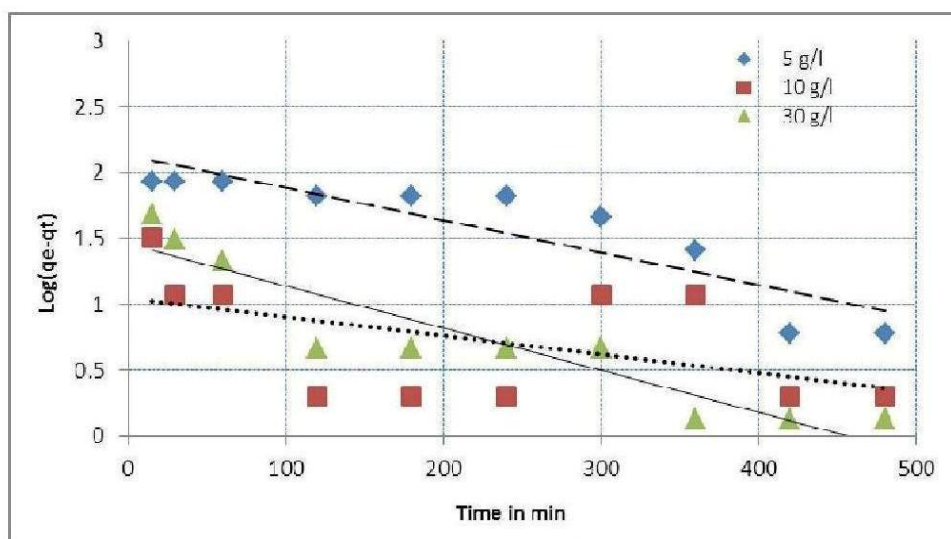


Fig. 4: Pseudo first order kinetic plot for sucrose sorption at different GAC sample doses when initial concentration solution is 6000mg/l and shaking speed 45 rpm

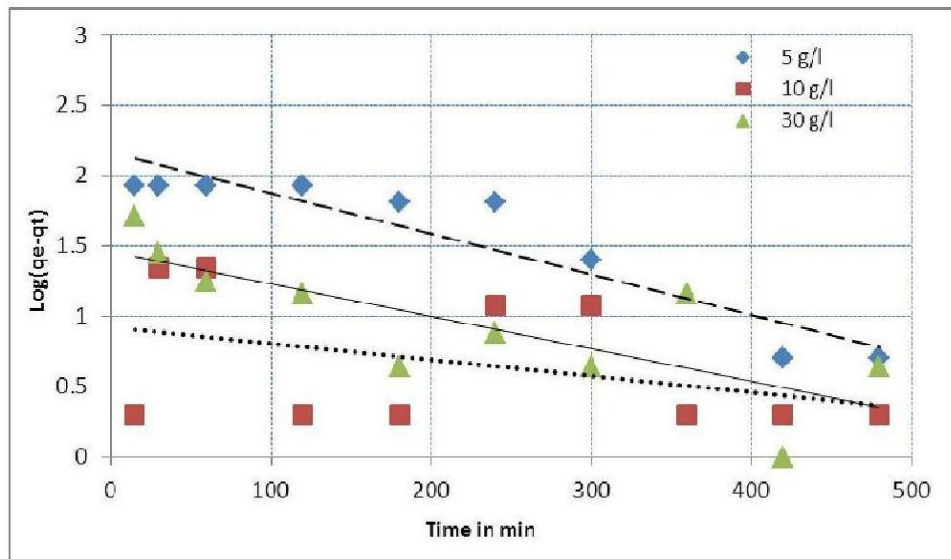


Fig. 5: Pseudo first order kinetic plot for sucrose adsorption at different GAC sample doses when initial concentration solution is 6000mg/l and shaking speed 100 rpm

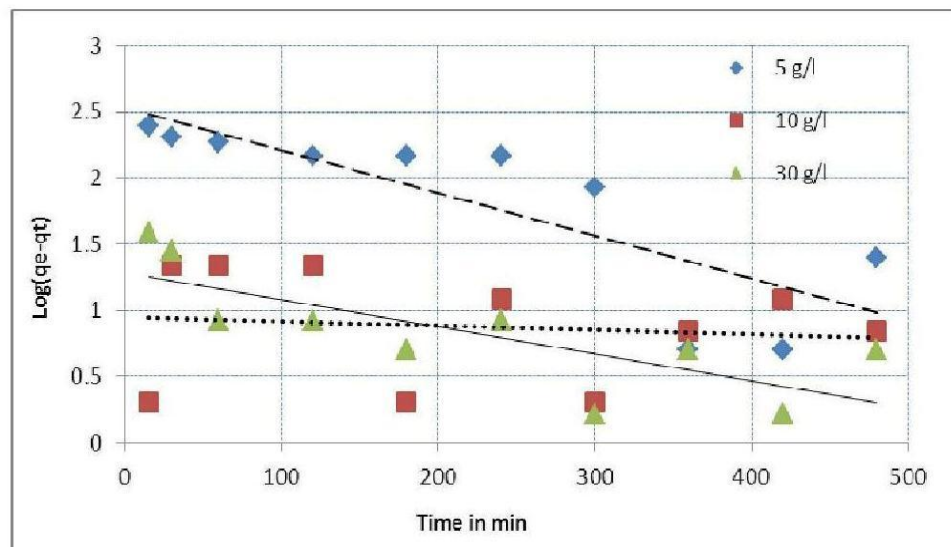


Fig. 6: Pseudo first order kinetic plot for sucrose sorption at different GAC sample doses when initial concentration solution is 6000mg/l and shaking speed 145 rpm

Pseudo second order model figures:

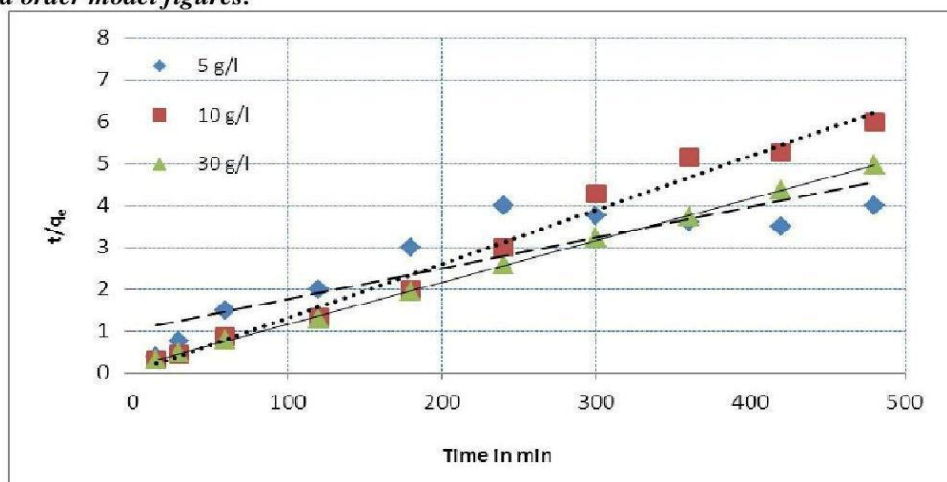


Fig.7: Pseudo second order kinetic plot for sucrose sorption at different GAC sample doses when initial concentration solution is 6000mg/l and shaking speed 45 rpm

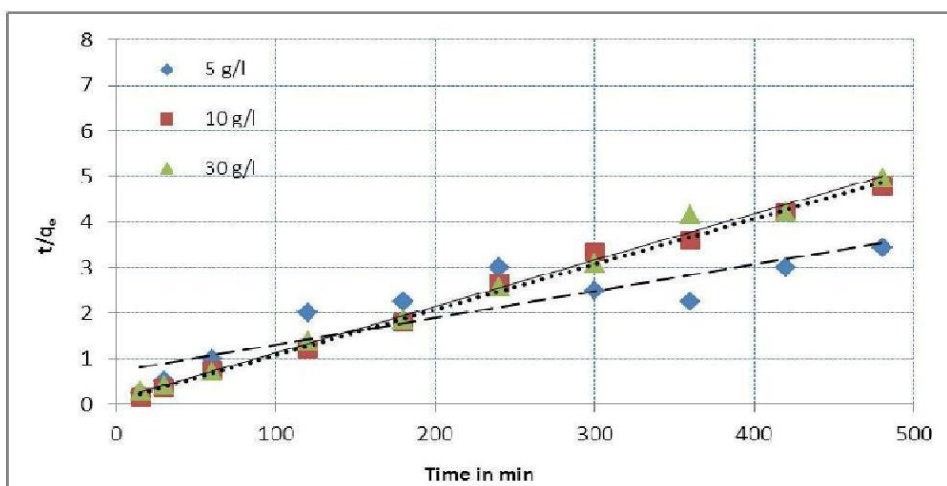


Fig. 8: Pseudo second order kinetic plot for sucrose sorption at different GAC sample doses when initial concentration solution is 6000mg/l and shaking speed 100 rpm

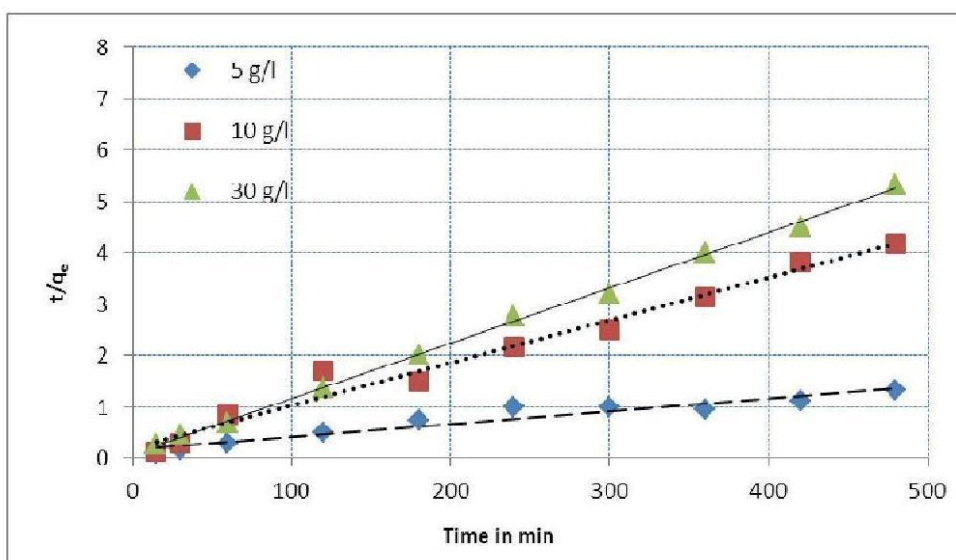


Fig. 9: Pseudo second order kinetic plot for sucrose sorption at different GAC sample doses when initial concentration solution is 6000mg/l and shaking speed 145 rpm

Intra- particle model figures:

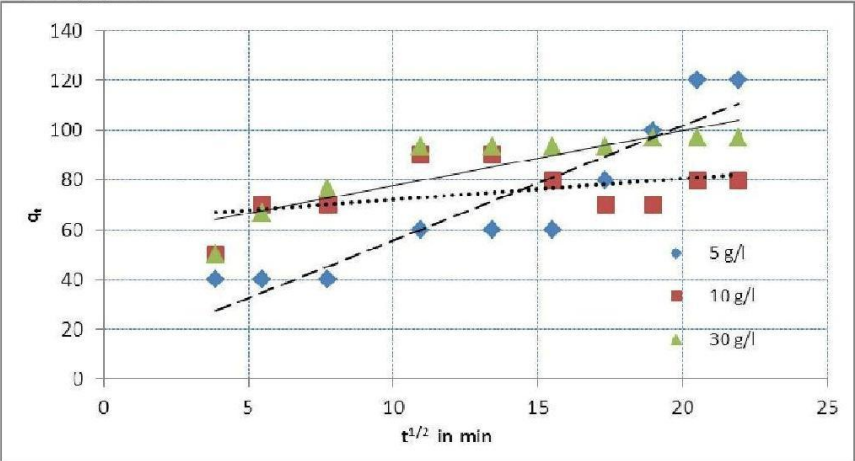


Fig. 10: Intra particle kinetic plot for sucrose sorption at different GAC sample doses when initial concentration solution is 6000mg/l and shaking speed 45 rpm

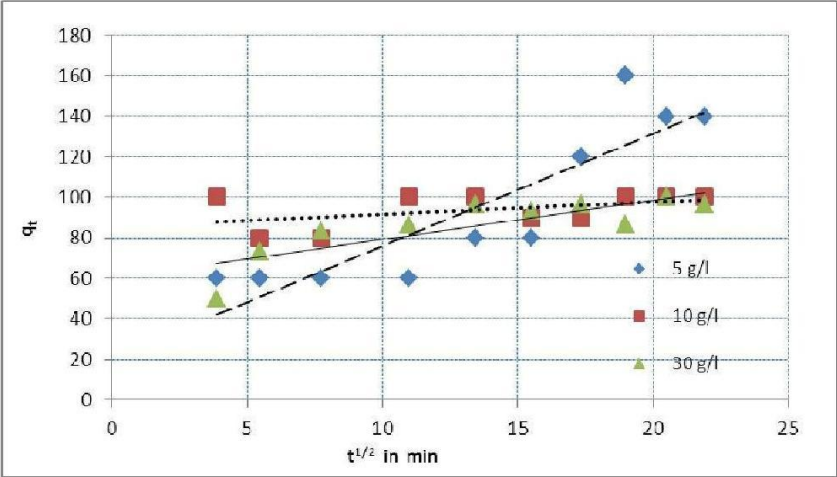


Fig. 11: Intra particle kinetic plot for sucrose sorption at different GAC sample doses when initial concentration solution is 6000mg/l and shaking speed 100 rpm

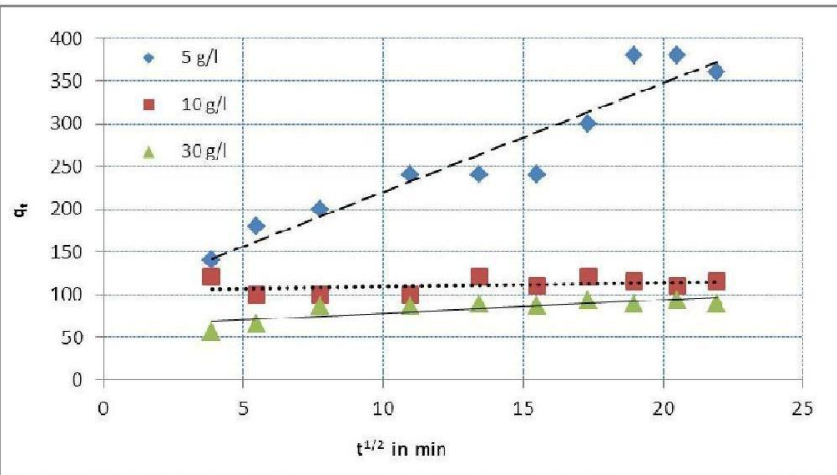


Fig. 12: Intra particle diffusion kinetic plot for sucrose sorption at different GAC sample doses when initial concentration solution is 6000mg/l and shaking speed 145 rpm

Mass transfer model figures:

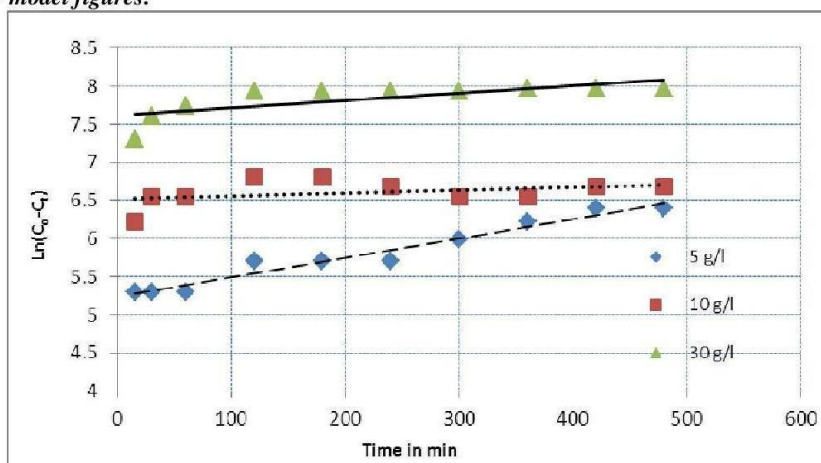


Fig. 13: Mass transfer kinetic plot for sucrose sorption at different GAC sample doses when initial concentration solution is 6000mg/l and shaking speed 45 rpm

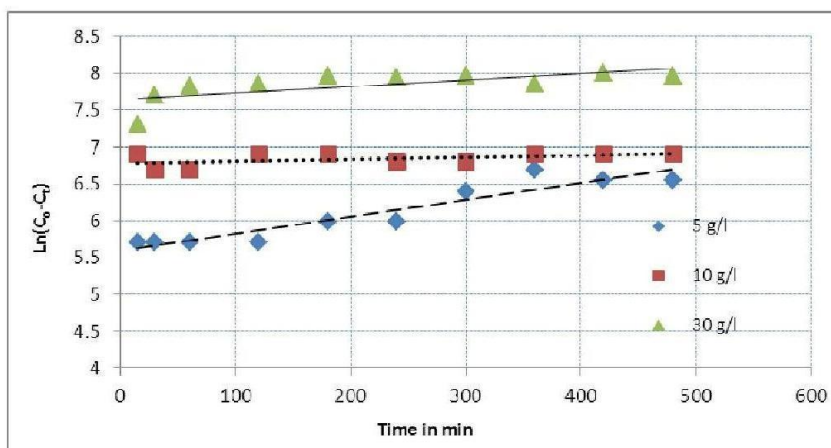


Fig. 14: Mass transfer kinetic plot for sucrose sorption at different GAC sample doses when initial concentration solution is 6000mg/l and shaking speed 100 rpm

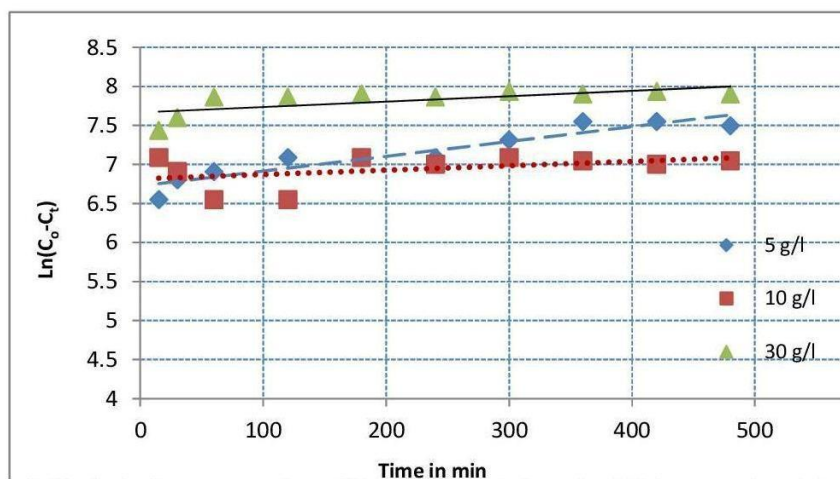


Fig. 15: Mass transfer kinetic plot for sucrose sorption at different GAC sample doses when initial concentration solution is 6000mg/l and shaking speed 145 rpm

Kinetic Sorption Study of Sucrose Osmotic Agent onto Granular Activated Carbon in Manipulated Osmosis Desalination System (MOD) Combined with a Solute Exchange Technique (SET)

Table1: kintec parameters for sucrose sorption at different GAC sample doses and shaking speeds (rpm) when initial concentration solution is 6000mg/L

Model	rpm	45 rpm		101 rpm		145 rpm	
		Eq.	R ²	Eq.	R ²	Eq.	R ²
Pseudo 1st order	5	$y = -0.0025x + 2.1299$	0.801	$y = -0.0029x + 2.1604$	0.863	$y = -0.0032x + 2.5295$	0.680
	10	$y = -0.0014x + 1.0424$	0.245	$y = -0.0012x + 0.9201$	0.162	$y = -0.0003x + 0.951$	0.016
	30	$y = -0.0032x + 1.4607$	0.862	$y = -0.0023x + 1.4575$	0.602	$y = -0.0021x + 1.2867$	0.594
Pseudo 2nd order	5	$y = 0.0073x + 1.0278$	0.785	$y = 0.0059x + 0.7266$	0.797	$y = 0.0025x + 0.177$	0.925
	10	$y = 0.0129x + 0.0074$	0.985	$y = 0.01x + 0.0884$	0.994	$y = 0.0083x + 0.1999$	0.974
	30	$y = 0.01x + 0.1455$	0.999	$y = 0.0102x + 0.1268$	0.993	$y = 0.0107x + 0.0925$	0.999
Intra-particle diffusion	5	$y = 4.6052x + 9.5277$	0.863	$y = 5.5473x + 20.747$	0.788	$y = 12.724x + 93.395$	0.900
	10	$y = 0.8516x + 63.447$	0.212	$y = 0.6069x + 85.768$	0.210	$y = 0.4745x + 104.56$	0.128
	30	$y = 2.1978x + 55.853$	0.762	$y = 1.9393x + 60.026$	0.668	$y = 1.5603x + 62.834$	0.657
Mass transfer	5	$y = 0.0026x + 5.2364$	0.958	$y = 0.0023x + 5.5898$	0.890	$y = 0.0019x + 6.7251$	0.887
	10	$y = 0.0004x + 6.5214$	0.147	$y = 0.0003x + 6.782$	0.235	$y = 0.0006x + 6.8149$	0.194
	30	$y = 0.001x + 7.6155$	0.554	$y = 0.0009x + 7.6531$	0.471	$y = 0.0007x + 7.669$	0.479

Table 2: kintec model parameters of sucrose sorption onto GAC

rpm	45 rpm				101 rpm				145 rpm			
GAC Dose s g/l					Pseudo 1st order							
	K1	qe exp	qe cal	R ²	K1	qe exp	qe cal	R ²	K1	qe exp	qe cal	R ²
5	5.76×10^{-3}	126	134.9	0.801	6.679×10^{-3}	145	144.7	0.863	7.370×10^{-3}	385	338.4	0.680
10	3.22×10^{-3}	82	11.03	0.245	2.764×10^{-3}	102	8.30	0.162	0.691×10^{-3}	122	8.933	0.016
30	7.37×10^{-3}	98	28.89	0.862	5.287×10^{-3}	101	28.67	0.602	2.533×10^{-3}	95	19.35	0.594
Pseudo 2nd order												
GAC g/l	K2	qe exp	qe cal	R ²	K2	qe exp	qe cal	R ²	K2	qe exp	qe cal	R ²
5	5.19×10^{-5}	126	137	0.785	4.791×10^{-5}	145	169.5	0.797	3.531×10^{-5}	385	400	0.925
10	22.49×10^{-3}	82	77.52	0.985	1.131×10^{-3}	102	100	0.994	0.345×10^{-3}	122	120.5	0.974
30	0.687×10^{-3}	98	100	1	0.831×10^{-4}	101	98.04	0.993	1.238×10^{-3}	95	93.46	0.999
Intra-particle diffusion model												
GAC g/l	K _{id}	c		R ²	K _{id}	c		R ²	K _{id}	c		R ²
5	4.605	9.53		0.863	5.547	20.75		0.788	12.724	93.4		0.900
10	0.852	63.45		0.212	0.607	85.77		0.210	0.475	104.6		0.128
30	2.198	55.85		0.762	1.940	60.03		0.668	1.550	6.8		0.657
Mass transfer kinetic model												
GAC g/l	K _o	D		R ²	K _o	D		R ²	K _o	D		R ²
5	2.6×10^{-3}	188.0		0.958	2.3×10^{-3}	267.7		0.890	1.9×10^{-3}	833.1		0.887
10	4×10^{-4}	679.5		0.147	3×10^{-4}	881.8		0.235	6×10^{-4}	911.3		0.194
30	1×10^{-3}	2029		0.554	0.9×10^{-3}	2107		0.471	0.7×10^{-3}	2140		0.479

4. CONCLUSIONS

At different shaking speeds, the sorption rate of glucose onto GAC was found to be rapid during the initial period of contact time and then became slow and stagnant with an increase in contact time when using 10 and 30g/l GAC sample doses. Also, with increasing shaking speeds the higher sample dose shows more stability with low sorption rate, while for a low sorbent dose (5g/l) the sorption capacity increased with time for all shaking speeds values used. The correlation coefficient was much higher in the pseudo second order rate equation compared with

other models. Also, the calculated equilibrium sorption capacities (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 is agrees well with experimental data values. Finally, it is recommended to test the kinetic mechanism of different organic compounds that can be used as osmotic agents in a MOD-SET combined system, so as to find out the best organic compound that can be used in this system.

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Kinetic Sorption Study of Sucrose Osmotic Agent onto Granular Activated Carbon in Manipulated Osmosis Desalination System (MOD) Combined with a Solute Exchange Technique (SET)



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