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Fixed-bed adsorptive removal of metanil yellow from simulated wastewater in a fixed-bed column by nitric acid-treated-H₃PO₄-activated carbon (NATPAAC) from oil palm fruit mesocarpfibre

B. O. Isiuku^{1,*}, C. A. Ojike¹, O. U. Akakuru², F. C. Ibe¹

¹Department of Chemistry, Imo State University, P.M.B. 2000, Owerri, Nigeria ²Department of Pure and Applied Chemistry, University of Calabar, Calabar, Nigeria *E-mail address: obinnabisiuku@yahoo.com

*Tel: +2348035731300

ABSTRACT

We carried out fixed-bed column adsorption of metanil yellow from simulated wastewater on NATPAAC derived from oil palm fruit mesocarpfibre so as to determine the adsorption capacity, q_e , of the carbon under the effects of inlet concentration, C_o , carbon bed height, H and dye solution flow rate, Q. Our results indicate that the optimum q_e was 15.982 mg/g by C_o 25 mg/L, H 4.1cm and Q 8 mL/min. In the study, q_e was observed to decrease with increase in C_o and Q. The optimum bed height was 4.1cm. Our experimental data were modelled by applying Thomas and Yoon-Nelson kinetic models. Correlation coefficient, R^2 values (generally above 0.85) show that the two kinetic approaches provide an effective model of the experimental data. We conclude that oil palm fruit mesocarpfibre has potential as a precursor for production of carbon for acid-dye removal from wastewater.

Keywords: Adsorption capacity, activated carbon, fixed-bed column, kinetic models, metanil yellow

1. INTRODUCTION

Increasing awareness of water pollution and its far-reaching effects in recent times have prompted concerted efforts toward pollution abatement. A lot of wastewater treatments that are carried out in industries are focused toward treating water polluted with organic pollutants

due to the fact that organics are common waste constituents (Bello et al. 2012). A large fraction of these industrial effluents contain dyes (Aksu, 2005). Various aquatic lives are jeopardized since the discharge of the dyes effluents which are highly coloured wastewater to aquatic systems can block the penetration of sunlight and oxygen, thereby arresting photosynthesis of aquatic plants. Many of the dyes are carcinogenic (Crini 2006). Metanil yellow ([3-(4-anilinophnylazo)] benzene sulphonic acid sodium salt also called Acid Yellow 36) is an organic compound which constitutes one or more azo bonds (-N=N-) used in a number of industries such as dyeing, food, cosmetics, paper printing and textiles industries with the textiles industry a very large consumer. Metanil yellow is toxic if absorbed through the skin, respiratory and intestinal tract and may act as a skin, eye or respiratory irritant. It is harmful when swallowed or inhaled and may be carcinogenic under long time exposure (Jain et al., 2009; Sivashankar et al., 2013)

Biological or physicochemical techniques which include adsorption, oxidation-reduction, chemical coagulation, ozone treatment and membrane filtration are employed in removing waste dyes from polluted water (Pramanpol and Nitayapat, 2006)

Adsorption is the concentration of a substance at the surface. The adsorption at a surface or interface is largely as a result of binding forces between atoms, molecules and ions of the adsorbate on the surface, (Isiuku et al., 2014). Adsorption onto activated carbon has been found to be superior to other techniques of wastewater treatment because of its capability to adsorb a broad range of different types of adsorbates efficiently, and its simplicity of design, (Ahmad and Hameed, 2006). Adsorption is a well-established technique for the removal of low concentrations of organic pollutants from large volumes of portable water, process effluents, wastewater and aqueous solutions (Patel and Vashi, 2012)

Batch adsorption experiments are used easily in the laboratory for the treatment of small volumes of effluents, but less convenient to use on industrial scale, where large volumes of wastewater are continuously generated. Batch adsorption provides certain preliminary information which includes pH for maximum adsorption, maximum initial adsorbate concentration, particle size for optimum adsorption of adsorbate, approximate time for adsorption of adsorbate and the maximum adsorption capacity of the adsorbent. All these information are useful in packed-bed studies (Unuabonah et al., 2010)

For the removal of toxic organic compounds from wastewaters, packed-bed columns containing activated carbon are used successfully. In fixed-bed adsorption, the adsorbate is continuously in contact with a given quantity of fresh adsorbent, thus providing the required concentration gradients between adsorbent and adsorbate for adsorption (Unuabonah et al., 2010; Markovska et al., 2001). Fixed-bed adsorption of pollutants involves percolation of wastewater through a percolating material. In course of the flow of the wastewater through the percolator, the wastewater is purified by physicochemical processes. The design and theory of fixed-bed adsorption systems focuses on establishing the shape of the breakthrough curve and its velocity through the bed. Breakthrough and bed volumes are employed in the determination of the performance of a fixed-bed described using the concept of breakthrough curve (Unuabonah et al., 2010; Cheng and Wang, 2000).

Activated carbons are usually obtained from materials with high carbon content and possess a great adsorption capacity, which is mainly determined by their porous structure (Otero et al., 2003). The inherent nature of the starting material and the method and conditions employed for synthesizing carbon strongly affects the final pore size distribution and the adsorption properties of the activated carbons. Oil palm fruit mesocarpfibre is the

fibre obtained after expressing oil from the fruit mesocarp. For every tonne of fresh fruit bunches processed, 120kg of the fibre is produced. Oil palm fruit mesocarpfibre contains on a dry weight basis, approximately 40% cellulose, 21% lignin, 24% pentosan and 5% ash (Nwabanne and Igbokwe, 2012). Researches had been carried out in removing metanil yellow from aqueous solution by adsorption but work on removal of the dye by fixed-bed adsorption on oil palm fruit mesocarpfibre carbon is rare. The packed-bed adsorption of metanil yellow on oil palm fruit mesocarpfibre carbon was carried out to determine the adsorption capacity of the carbon under the influences of C_o, H and Q. Experimental data were modelled with Thomas and Yoon-Nelson kinetic models.

2. MATERIALS AND METHODS

2. 1. Adsorbate

Metanil yellow (Merck, Switzerland) also named 3-(4-anilinophenylazo) benzene sulphonic acid sodium salt, C.I. Acid Yellow 36, Tropaeoline G and Acid Leather Yellow R (Fig 1) used in this work, was purchased at Onitsha, Nigeria. The adsorbate was used without further treatment. The stock solution was prepared by dissolving 1g per litre solution with distilled water.

2. 2. Preparation of activated carbon

The method (Isiuku et. al., 2013; Ahmad and Hameed, 2010) was employed. The oil palm mesocarpfibre biomass collected at an oil mill at Umuapu, Ohaji, Imo State, Nigeria, was washed with distilled water to remove dirt and dust. After drying under sunlight for three days, the fibre was ground. The biomass was soaked in 20% H₃PO₄ at a ratio of 1 biomass: 3 H₃PO₄ by mass overnight. Excess acid was drained off and the biomass dried. The dry acid-treated fibre was carbonized at 500 °C for 5h, cooled and washed with distilled water to pH 6. The washed carbon was dried in a hot air oven at 110 °C for 2h. After drying and cooling the carbon was treated with 1 M HNO₃ solution overnight. Excess acid was decanted and the carbon was washed with distilled water to pH 6, dried at 110 °C, for 2h, cooled and sieved to get 0.420-0.841 mm particle sizes. The carbon was packed in an airtight plastic container.

2. 3. Adsorption Process

The adsorption column was a cylindrical Pyrex glass tube of measurement 30 cm in length and 1.8 cm internal diameter. The adsorption tube was plugged at the base with glass wool. Known mass of the carbon (equivalent to 2.1, 4.1 or 6.3 cm height) was introduced. The adsorbent was sealed with glass wool and the remaining space in the tube packed with glass beads. The glass beads ensured uniformity in the flow of the adsorbate. Different working concentrations of the dye obtained by diluting the stock solution with distilled water (25, 50 or 100 mg/L) were made to flow upward through the column at specific flow rates (8, 16 or 26 mL/min) for a given time at 29 °C with the help of a metering pump (Chem – Tech Pal No. 0-111.808). Samples of the effluent were collected at specific intervals for analysis using a UV-Visible spectrophotometer (Shimadzu UV-752, Japan) at λ_{max} 440 nm.

2. 4. Analysis of fixed-bed column data

The major characteristic of fixed-bed column adsorption is the history of effluent concentration (Nwabunne and Igbokwe, 2012). These concentrations – time curves are commonly called breakthrough curves, and the time at which the effluent concentration reaches the threshold value is called the breakthrough time. The breakthrough curves show the loading behaviour of the adsorbate to be removed from solution in a fixed-bed column and is usually expressed in terms of adsorbed adsorbate concentration C_{ad} , influent adsorbate concentration C_o , effluent adsorbate concentration, C_t , or normalized concentration (C_t/C_o), as a function of time or volume of effluent for a given bed height or adsorbent mass (Patel and Vashi, 2012; Ahmad and Hameed, 2006; Taty-Custodes et al., 2005; Aksu and Gönen, 2004). Effluent volume (V_{eff}) can be calculated from Eq. 1:

where: ttot is the total flow time (min).

The area under the breakthrough curve (A) obtained by integrating the adsorbed concentration $[C_{ad} (mg/L)]$ versus time (min) plot can be used to find the total adsorbed adsorbate quantity (maximum column capacity) (q_{tot}). For a given influent concentration and flow rate, q_{tot} is determined from Eq. 2:

$$q_{tot} = \frac{QA}{1000} = \frac{Q}{1000} \int_{t=0}^{t=t_{tot}} C_{ad} dt$$
 (2)

The total amount of dye sent to the column (m_{tot}) is determined using Eq. 3:

Total removal (%) of adsorbate (column performance) with respect to flow volume can also be found from the ratio of total adsorbed quantity of adsorbate (q_{tot}) to the total amount of adsorbate sent to the column (m_{tot}) Eq. 4:

$$Total\ removal\ (\%)\ =\ \frac{100q_{tot}}{m_{tot}}......(4)$$

Equilibrium uptake of the adsorbate q_e or column adsorption capacity at the end of total flow time is defined by Eq. 5: as the maximum column capacity per amount of adsorbent used x(g)

Un-adsorbed adsorbate concentration at equilibrium in the column, $C_e \, (mg/L)$ can be calculated from Eq. 6:

2. 5. Column adsorption modelling

A number of mathematical models have been developed for the evaluation of efficiency and applicability of the column models for large scale operations. The Thomas and Yoon-Nelson models were used to analyze the behaviour of adsorbent-adsorbate system in this work.

2. 5. 1. Thomas kinetic model

The Thomas model is one of the most general and widely used models in column performance simulation. The expression by Thomas for an adsorption column (Unuabonah et al., 2010; Fu and Viraraghavan, 2003) is given as Eq. 7:

$$\frac{C_t}{C_o} = \frac{1}{1 + \exp[K_{Th} (q_o x - C_o V_{eff})/Q]} \dots \dots \dots \dots (7)$$

where: C_t (mg/L) is the effluent adsorbate concentration at time t, q_o (mg/g) the maximum adsorption capacity of the adsorbent, x (g) mass of adsorbent used and K_{Th} (mL/mg min) the Thomas rate constant.

The value t (min) is expressed as Eq. 8:

The linearized form of Thomas model is expressed as Eq. 9:

The Thomas rate constant (or kinetic coefficient) K_{Th} and the maximum adsorbate adsorption capacity of the adsorbent q_o can be determined from the plot In [(C_o/C_t) - 1] versus t.

2. 5. 2. Yoon-Nelson model

The Yoon-Nelson model (Kundu and Gupta, 2007) is based on the assumption that the rate of decrease in the probability of adsorbate and the probability for a single component system is expressed as Eq. 10:

$$In\left[{\binom{C_t}{(C_o - C_t)}}\right] = K_{YN}t - \tau K_{YN} \dots \dots \dots (10)$$

where: K_{YN} (min⁻¹) is the Yoon-Nelson rate constant, t (min) sampling time and τ (min) the time required for 50% adsorbate breakthrough.

The calculation of theoretical breakthrough curves for a single-component system requires the determination of the parameters K_{YN} and τ for the adsorption from the slope and intercept respectively of a straight-line plot of In $[C_t/(C_o-C_t)]$ versus sampling time t.

The slope yields K_{YN} and the intercept - τK_{YN} . Based on the obtained value of τ the adsorption capacity, q_{oYN} can be calculated (Patel and Vashi, 2012) applying Eq. 11:

$$q_{oYN} = \frac{q_{tot}}{x} = \frac{C_o Q \tau}{1000 x}$$
 (11)

3. RESULTS AND DISCUSSION

3. 1. Effect of inlet dye solution concentration

The effect of C_o on the fixed-bed adsorption of metanil yellow on NATPAAC from oil palm fruit mesocarpfibre at fixed H and Q are shown in Fig. 2 where, it is seen that the higher the C_o , the quicker the adsorbent reached saturation. Table 1 shows that q_e decreased with increase in C_o . Total dye removal R (%) followed the same trend. The decrease in q_e and total dye removal from 15.982 mg/g for 25 mg/L to 7.14 mg/g for 50 mg/L was far less than from 7.14 mg/g for 50 mg/L to 1.57 mg/g for 100 mg/L. The reason for the trend was due to the fact that active sites on the adsorbent were saturated at higher adsorbate concentrations (Njoku et al., 2012).

3. 2. Effects of carbon bed height

The impact of removing metanil yellow from aqueous solution by carbon in a fixed-bed column by activated carbon from oil palm fruit mesocarpfibre at fixed C_0 25 mg/L and Q 8 mL/min at various H is shown in Fig. 3. The breakthrough curves for H 4.1 and 6.3 cm are virtually the same. The two beds could not reach saturation in 360min. Table 1 shows that equilibrium adsorption capacity and total dye removal were maximum for H 4.1cm. q_e and total removal increased from 9.407 to 15.982 mg/g and 8.04 – 27.32% respectively for H 2.1-4.1 cm but decreased from 15.982 to 5.97 mg/g and 27.32 – 15.39% respectively (H 4.1-6.3 cm) as H increased. The decrease may be due to the splitting effect of flux concentration gradient between adsorbate and adsorbent with increase in H (or carbon mass) causing a decrease in the amount of dye adsorbed onto unit mass of carbon (Pokordi and Kumar, 2006).

3. 3. Effect of flow rate

The effect of adsorbing metanil yellow on NATPAAC from oil palm fruit mesocarpfibre in a fixed-bed column at various Q 8, 16 and 26ml/min, C_0 25 mg/L and H 4.1cm is shown in Fig. 4. The figure shows that the higher the Q, the quicker the adsorbent reached saturation. Table 1 shows that q_e decreased with increase in Q; similarly, total dye removal decreased with increase in Q. This trend is attributed to the insufficient residence time of the solute in the column to allow for diffusion of the solute into the pores of the carbon (Salman and Hameed, 2011).

3. 4. Fixed-bed adsorption modelling

The time for breakthrough appearance and the shape of the breakthrough curve are important characteristics for determining the operation and dynamic response of an adsorption column. Again, successful design of an adsorption column requires prediction of the concentration-time profile from the breakthrough curve for the effluent from the column. The Thomas and Yoon-Nelson models were applied in this work.

3. 5. Thomas kinetic model

Experimental data at fixed C_o , H and Q were analysed with the Thomas model. Figs 5-7 show the ln $[(C_o/C_t)-1]$ versus time plots. Table 2 shows the Thomas parameters and correlation coefficient values. The table shows that q_o increased with increase in C_o , decrease in H and decrease in Q. The correlation coefficient values ($R^2>0.85$) show that experimental data fitted well to the Thomas model.

3. 6. Yoon-Nelson kinetic model

Yoon-Nelson model was applied to experimental data with respect to C_o , H and Q. A linear regression analysis was used on each set of experimental data to evaluate the Yoon-Nelson model parameters (see Figs 8-10). Table 3 shows these parameters and the respective R^2 values. The table shows that q_o increased with increase in C_o , decrease in H and decrease in H, and H showed the same trend with H0. The H0 values which are generally above 0.85, shows that experimental data fitted well to the Yoon-Nelson model.

4. CONCLUSION

Fixed-bed column adsorption on NATPAAC prepared from oil palm fruit mesocarpfibre was applied to remove metanil yellow from wastewater. The adsorption capacity of the carbon was evaluated at various C_o , H and Q. Experimental data showed optimum q_e as 15.982 mg/g. Thomas and Yoon-Nelson models simulated experimental data well based on R^2 values. Results show that the optimum C_o , H and Q were 25 mg/L, 4.1cm and 8mL/min respectively. The results also show that oil palm fruit mesocarpfibre activated carbon has potential as an adsorbent for acid dyes.

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World News of Natural Sciences 17 (2018) 157-172

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Table 1. Experimental data for fixed-bed column adsorption of metanil yellow from aqueous solution on NATPAAC from oil palm fruit mesocarpfibre at various process parameters.

Q (mg/L)	H (cm)	Q (ml/min)	q _{tot} mg/g	q _e mg/g	Total removal (%)
25	4.1	8	63.928	15.982	27.32
50	4.1	8	28.56	7.14	6.11
100	4.1	8	6.28	1.57	0.67
25	2.1	8	18.814	9.407	8.04
25	6.3	8	35.82	5.97	15.39
*25	4.1	8	44.18	11.045	61.36
*25	4.1	16	39.34	9.835	27.32
*25	4.1	26	38.284	9.571	16.39

^{*}Asorption was for 240 min

C _o (mg/L)	H (cm)	Q (ml/min)	K _{Th} ×10 ⁻⁴ (ml/min mg)	q _o (mg/g)	\mathbb{R}^2
25	4.1	8	1.88	0.944	0.9017
50	4.1	8	0.88	29.711	0.9012
100	4.1	8	0.25	324.06	0.8791
25	2.1	8	1.52	29.991	0.9121
25	4.1	8	2.12	7.029	0.8726
25	6.3	8	2.24	4.598	0.8549
*25	4.1	8	1.96	20.088	0.8782
*25	4.1	16	2.12	14.058	0.8549
*25	4.1	26	1.88	3.049	0.9019

^{*}Adsorption was run for 240min

Table 3. Yoon-Nelson model parameters for fixed-bed column adsorption of metanil yellow on NATPAAC from oil palm fruit mesocarpfibre at various adsorption conditions

C _o (mg/L)	H (cm)	Q (ml/min)	K _{YN} (min ⁻¹)	T (min)	q _{OYN} (mg/g)	\mathbb{R}^2
25	4.1	8	0.0053	18.06	0.903	0.8495
50	4.1	8	0.0044	297.25	29.725	0.9109
100	4.1	8	0.0027	1490.78	298.156	0.903
25	2.1	8	0.0038	299.61	29.961	0.9114
25	4.1	8	0.0053	140.51	7.026	0.8548
25	6.3	8	0.0056	138.07	4.602	0.8728
*25	4.1	8	0.0049	403.63	20.182	0.8844
*25	4.1	16	0.0053	140.509	14.051	0.8548
*25	4.1	26	0.0047	18.638	3.029	0.9025

^{*}Adsorption was run for 240 min

Fig. 1. Structure of metanil yellow

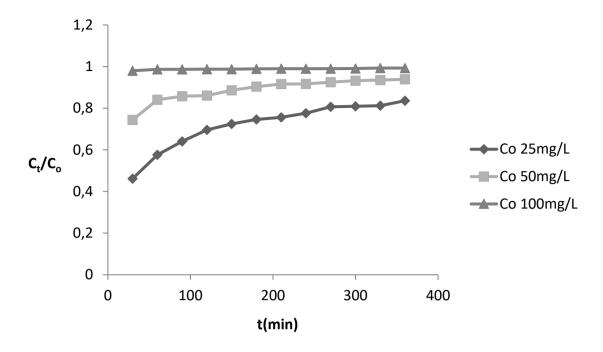


Fig. 2. Effect of dye concentration on the adsorption of metanil yellow on activated carbon from palm fibre

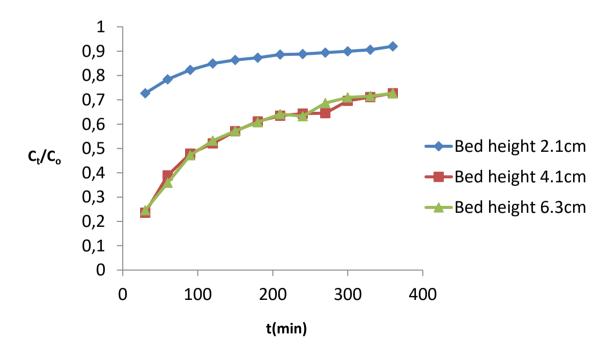


Fig. 3. Effect of bed height and the adsorption of metanil yellow on activated carbon from palm fibre

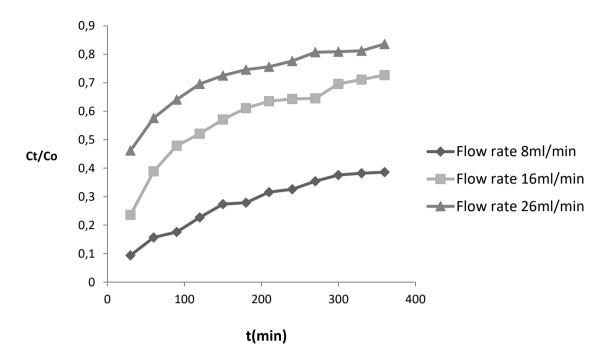


Fig. 4. Effect of flow rate on the adsorption of metanil yellow on activated carbon from palm fibre

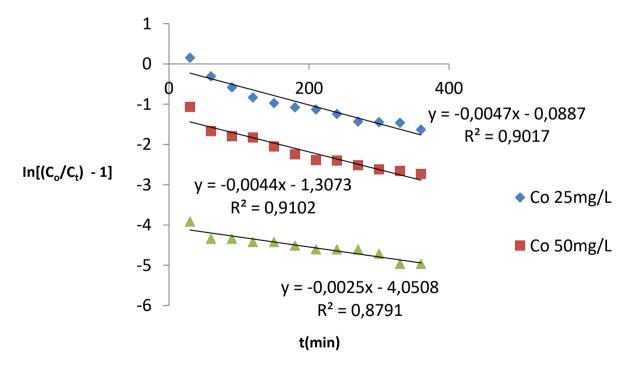


Fig. 5. Thomas model for the adsorption of metanil yellow on activated carbon from palm fibre at various initial concentrations.

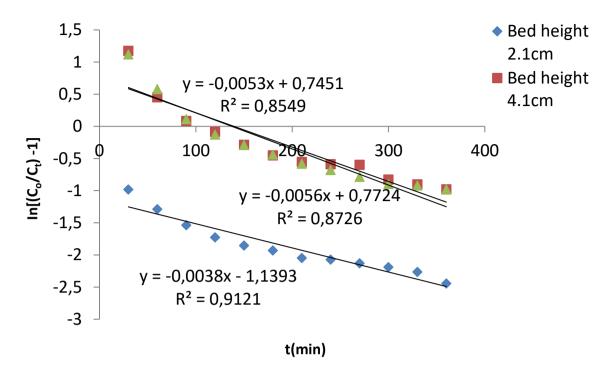


Fig. 6. Thomas model for the adsorption of metanil yellow on activated carbon from palm fibre at various bed height

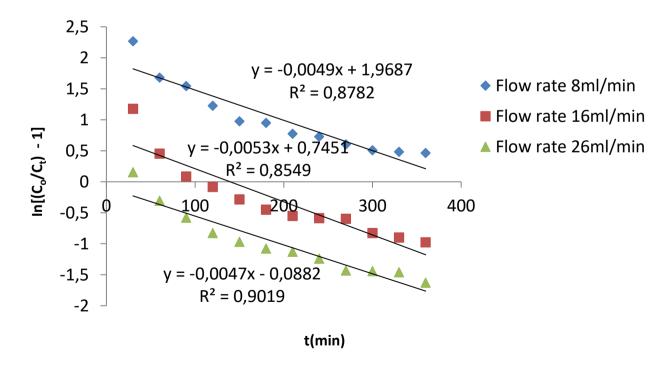


Fig. 7. Thomas model for the adsorption of metanil yellow on activated carbon from palm fibre at various flow rates.

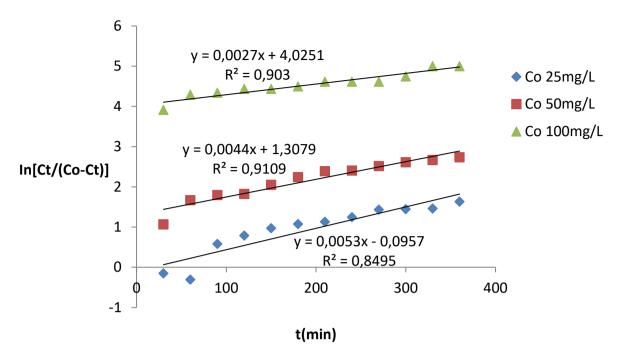


Fig. 8. Yoon-Nelson model plot for the adsorption of metanil yellow on palm fibre activated carbon at various initial concentrations.

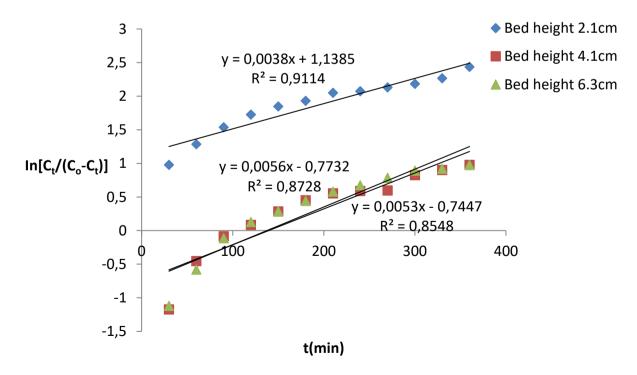


Fig. 9. Yoon-Nelson model plot for the adsorption of metanil yellow on activated palm firbe carbon at various bed heights.

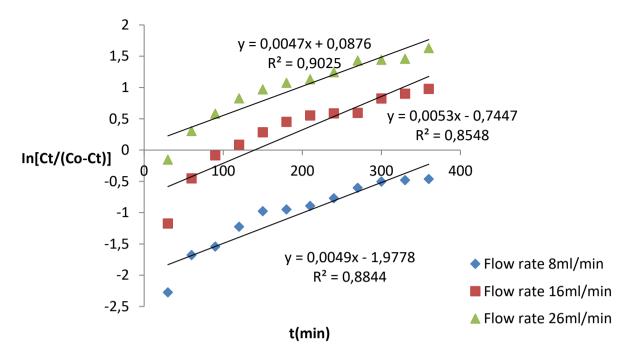


Fig. 10. Yoon-Nelson model plot for the adsorption of metanil yellow on activated palm fibre carbon at various flow rates.