

Adsorption of Malachite Green from Aqueous Solution using Plantain Stalk (*Musa paradisiaca*).

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ABSTRACT

The effectiveness of plantain stalk at removing malachite green dye from aqueous solution has been studied. The adsorption experiments were carried out as a function of contact time, different initial dye concentration and biosorbent dose. Kinetics and equilibrium data were obtained from batch experiments. Adsorption increases with increasing initial concentration of malachite dye but remains constant for adsorbent dose $\geq 0.3\text{g}$. The Freundlich isotherm gave the highest correlation coefficient ($R^2 = 0.9925$) followed by the Langmuir ($R^2 = 0.9765$), Temkin ($R^2 = 0.9552$) and Dubinin-Radushkevich ($R^2 = 0.8329$) isotherms respectively. The dye sorption processes was better described by the pseudo-second-order kinetic model. The results showed that plantain stalk has good potential for the removal of malachite green dye from aqueous solution.

(Keywords: biosorption, plantain stalk, malachite green, sorption isotherms)

INTRODUCTION

Malachite green as a dye is commonly used in the production of textile materials and paper products. The dye is also used in the aquaculture industry against fungi and parasitic infections in fish and other aquatic organisms due to its effectiveness, availability, and relatively low cost compared with other anti-fungal agents (Culp and Beland, 1996). However, Malachite green (MG) has been reported to be a suspected mutagen and teratogen (Culp and Beland, 1996).

Residues of MG and its reduced form, leucomalachite green was reportedly found in the serum, liver, kidney, muscles and other tissues of certain fish species, while causing organ damage, mutagenic, carcinogenic and developmental abnormalities in mammals (Srivastava *et al.*, 2004). Its use therefore needs to be regulated and controlled especially in developing countries like Nigeria.

Generally, the disposal of dyes in precious water resources has generated various treatment technologies, which ranges from physical (sedimentation, filtration, etc.), chemical (oxidation processes including use of free radicals, precipitation and electro-coagulation), physical-chemical (adsorption, ion-exchange, stripping, chemical oxidation and membrane separation), biological (aerobic, anaerobic, combined aerobic – anaerobic), acoustical, radiation and electrical processes (Gupta and Suhas, 2009). Adsorption appears to be one of the most prominent of these treatment methods because of its simplicity, efficiency, wide and social acceptability, economic and technical feasibility (Foo and Hammed, 2010). As stated by Gupta and Suhas (2009), the growing demand for efficient and low-cost treatment methods and the importance of adsorption has given rise to low-cost alternative adsorbents (LCAs).

Various LCAs have been used for the removal of malachite green from aqueous solution, ranging from aquatic microbes such as aerobic granules (Zheng *et al.*, 2005; Sun *et al.*, 2008) water algae (Kumar *et al.*, 2006), and microalgae (Daneshvar *et al.*, 2007), to biomasses such as activated carbon derived from rice husks (Guo *et al.*, 2003,

Rahman *et al.*, 2005), mesoporous tyre, *Terminalia catappa* Linn (Venkatraman *et al.*, 2011) groundnut shell waste (Malik *et al.*, 2007), treated ginger waste (Ahmad and Kumar, 2010), neem leaf powder (Khatod and Thakur, 2011), chitin hydrogels (Tang *et al.*, 2012), seashells, eggshells (Shamik and Papita, 2012), and odina wodier bark (Vijayakumaran and Arivoli, 2012). Others includes *Kapok hull* (Shabudeen 2011), poultry feathers (Beak *et al.*, 2009), chemically modified silica gel (Kushwaha *et al.*, 2010), and halloysite nanotubes (Kiani *et al.*, 2011).

Recently, Gupta *et al.* (2011) evaluated the kinetics and thermodynamics of malachite green adsorption on banana pseudo-stem fibers and concluded that the fiber is an effective adsorbent for the removal of MG dye from aqueous solution. Plantain and banana stalks are relatively abundant in major Nigerian markets with no specific usage but rather constituting pollution and waste disposal problems. Unlike plantain pseudostem and leaves, the use of plantain stalk as adsorbent for the removal of malachite green have not been reported in the literature.

In the present work, we report our evaluation of the feasibility of using plantain stalk for the removal of malachite green from aqueous solution. The biosorption equilibrium were modeled under the Langmuir, Freundlich, Temkin and Dubinin-Radushkevich isotherms while the sorption rates of adsorbent dosage and malachite green concentration with time were tested using the pseudo- first and second order kinetics, respectively.

Biosorption Isotherms

Isotherm studies are highly important in order to access a typical adsorption system and to ascertain the most appropriate equilibrium correlation (Srivastava *et al.*, 2006). They are also important for comparing biosorption performance, optimization, design and prediction purposes (Ho *et al.*, 2002). The biosorption of malachite green on plantain stalk was optimized by analyzing equilibrium curve using the following four isotherm models.

(a) **Langmuir Isotherm:** The Langmuir Isotherm, developed by Irving Langmuir in 1916 originally to describe the gas-solid phase adsorption onto activated carbon, is now extended and generally applied to liquid-solid

phase interaction. The model assumes in its formulation a monolayer adsorption at maximum capacity occurring through a uniform or equivalent surface of adsorbent without any interaction between the adsorbate molecules. The equation is express as follows:

$$q_e = q_m b C_e / (1 + b C_e) \quad (1)$$

where q_e (mg/g) and C_e (mg/L) is the concentration of the dye solution in the adsorbent and in solution at equilibrium respectively, q_m is the maximum sorption capacity (mg/g) and b is the Langmuir constant related to the affinity of the adsorbate for the adsorbent (L/mg). The linearized form of the equation is given as:

$$C_e / q_e = 1 / b q_m + C_e / q_m \quad (2)$$

A dimensionless separation factor R_L defined by Webber and Chakkravorti (1974) can be used to predict the affinity between adsorbate and adsorbent using the Langmuir parameter b . Thus:

$$R_L = 1 / (1 + b C_o) \quad (3)$$

where b refer to the Langmuir constant and C_o , the initial concentration of adsorbate. R_L is related to the nature of the biosorption as follows:

- $R_L < 1$ favorable
- $R_L > 1$ unfavorable
- $R_L = 1$ linear
- $R_L = 1$ irreversible

(b) **Freundlich isotherm:** The Freundlich isotherm (1906) relates the solute concentration on the adsorbent surface to the solute concentration in the liquid phase. The isotherm assumes that adsorption occur on a heterogeneous adsorbent surface (i.e., multilayer adsorption). Accordingly Freundlich proposed the equation:

$$q_e = K_f C_e^{1/n} \quad (4)$$

which when linearized becomes:

$$\ln q_e = \ln K_f + 1/n \ln C_e \quad (5)$$

Where K_f and $1/n$ are the Freundlich coefficients related to biosorption capacity and biosorption intensity, respectively.

(c) Tenkim Isotherm: The Tenkim Isotherm (1940) take into account the interaction between adsorbate and adsorbent and assumes a linear decrease in the heat of adsorption instead of a logarithm decrease. The isotherm is expressed as:

$$q_e = RT/b_T \ln (A_T C_e) \quad (6)$$

which on linearization gives:

$$q_e = (RT/b_T) \ln K_T + (RT/b_T) \ln C_e \quad (7)$$

(d) Dubinin-Radushkevich (D-R) Isotherm

The isotherm is represented as,

$$q_e = q_d \exp [-B_D \mathcal{E}^2] \quad (8)$$

where $\mathcal{E} = RT \ln(1 + 1/C_e)$

Where R, T, and C_e is the gas constant (8.314J/molK), absolute temperature (K) and adsorbate equilibrium concentration (mg/L) respectively. The equation showed that it is temperature dependent. The isotherm constant B_D is related to the free energy, E, per molecule of adsorbate (i.e. the energy required to remove a molecule from its location in the sorption space to the infinity) by the equation:

$$E = [1/(\sqrt{-2B_D})] \quad (9)$$

The isotherm is postulated to help determine whether the adsorption is physical in nature (physisorption) or chemical (chemisorption) depending on the value of E.

Sorption Kinetics

The kinetics of biosorption are important in predicting the rate of sorption for a given system and gaining insights into the mechanism of sorption reaction (Yuh – Shan, 2006). The pseudo-first order kinetic of Lagergren (1898) is given in the form:

$$d_q/d_t = k(q_e - q_t) \quad (10)$$

This on linearization gives:

$$\log(q_e - q_t) = \log q_e - (k_1/2.303)t \quad (11)$$

where q_e and q_t is the amount of dye sorbed per unit weight of sorbent at equilibrium and at time t (min) respectively, k_1 is the rate constant of the pseudo-first order sorption (min^{-1}). The value of the rate constant (k_1) is obtain from the linear plot of $\log(q_e - q_t)$ versus t. The pseudo-second order rate equation (Ho *et al.*, 2006) can be expressed as:

$$d_q/d_t = k_2(q_e - q_t)^2 \quad (12)$$

where k_2 is the equilibrium rate constant of the pseudo-second order sorption in g/mg min. Upon integration, the linear form of Equation (12) becomes:

$$t/q_t = 1/(k_2 q_e^2) + (1/q_e)t \quad (13)$$

The linear plot of t/q_t versus t give a linear plot of slope $1/q_e$ and intercept, $1/(k_2 q_e^2)$.

MATERIALS AND METHODS

Sample Collection and Preparation

The plantain stalk used was obtained from the plantain market Sabo Ilesha Osun state Nigeria. The plantain stalk was cut into pieces, washed with water and sun dried. The plantain stalk was then grinded into powder using a milling machine. The powdered plantain stalk was then sieved using a laboratory sieve. The coarse particle was washed with water several times to remove any attached dirt, color and other soluble impurities. The sample was dried in the oven at 50°C kept in a sealed polythene bag pending usage as adsorbent.

Dye Solution

Malachite green ($C_{23}H_{25}ClN_2$), the sorbate used in the present study, is a monovalent cationic dye. It is classified in dye classification as C.I. Basic green 4. It has a molecular weight of 364.9g/mol. The malachite green (Aldrich type) was used as supplied. The stock solution of 1000mg/l was prepared by dissolving an appropriate amount of MG in 1L of deionized water. The working solutions were prepared by diluting the stock solution with distilled water to give the required concentration namely 10, 20, 40, 80, and 100 mg/l.

Batch Biosorption Study

This was carried out within a concentration range of 10 – 100mg/L at natural pH. The fixed amount of adsorbent dosage (0.1g) was weighed into an amber bottle, 25ml of MG solution was added and the mixture was maintained at room temperature (27°C) in a thermostated water bath agitated at a constant speed of 120rpm.

After the optimum constant time, the solution was decanted from the sorbent and the residual concentration of MG was determined using a UV/Visible Spectrophotometer at a maximum wavelength of 617nm. The results were duplicated. The amount of dye biosorbed from solution was determined by difference and the mean value was calculated for each set of experiments. The biosorption capacity at each moment was calculated from the equation:

$$q_e = V(C_i - C_e) / m \quad (14)$$

where C_i and C_e refer to the initial and equilibrium concentration of dye (mg/L), respectively. V represents volume (L) of the solution and m , the dry weight (g) of the biosorbent used in the study.

Effect of Initial Concentration

Fixed sorbent dose (0.1 g) was added to 25ml of MG solution in an amber bottle at natural pH. The mixture was agitated at a speed of 120rpm at different time intervals (2, 5, 10, 20, 30, 60 and 180 minutes). The initial concentrations of dye solution tested were 10, 20, 40, 80, and 100mg/L.

Effect of Different Adsorbent Dosage

The sorbent dose effect was investigated at varying plantain stalk dosages (0.1, 0.2, 0.3, 0.4, and 0.5 g), and fixed sorbate concentration (100 mg/l of MG) and agitation speed (120 rpm) in a thermostatic water bath at fixed time intervals (2, 5, 10, 20, 30, 60, and 180 min).

RESULTS AND DISCUSSION

Effect of Initial Concentration

Figures 1 and 2 showed the time course of MG adsorption by the plantain stalk at different concentration and different adsorbent dosages,

respectively. The amount of MG dye adsorbed increases with increase in initial concentration per unit weight of adsorbent. Thus $q_e = f(C_e)$ i.e. the amount of dye adsorbed is a function of the concentration of the dye in solution. The higher the concentration, the higher the interaction between the dye and the biosorbent molecule and the lower the mass transfer resistance between the aqueous and solid phase (Maarof *et al.*, 2003). It was also observed that irrespective of the initial concentration of dye, the amount adsorbed increases for the first 30 minutes and then remains constant.

Effect of Adsorbent Dosage

There was a rapid adsorption in the first 30 minutes for adsorbent dosages 0.1g and 0.2g due to the presence of available sites on the sorbent. The rate however slows down and remains constant as the binding sites are used up. The situation was different for adsorbent dosages 0.3g, 0.4g and 0.5g as the amount adsorbed was constant throughout irrespective of the duration of adsorption. This implied that using lower adsorbent dosage will be more advantageous than higher ones for the same concentration of dye. It also indicates that molecules of the MG dye probably do not adsorb on each other, only a monolayer adsorption is formed.

Sorption Isotherms

Four isotherm models were used to analyze the equilibrium curve. The linear coefficient of determination, R^2 was used as an error function to evaluate the fitness of each isotherm equation to the experimental data obtained from the optimization process employed.

The Freundlich isotherm gave the highest correlation coefficient ($R^2 = 0.9925$), followed by the Langmuir Isotherm ($R^2 = 0.9765$). The value of the dimensionless Freundlich constant, $1/n$, which is a measure of the nature and strength of the sorption process showed that the bond energies increase with surface density of the biosorbent (Hubbe *et al.*, 2012). The sorption process showed a good fit to the Langmuir isotherm which suggests a finite adsorption capacity and energetically equivalent sites (Hubbe *et al.*, 2012). The maximum loading capacity estimated from Langmuir isotherm was 30.68mg/g.

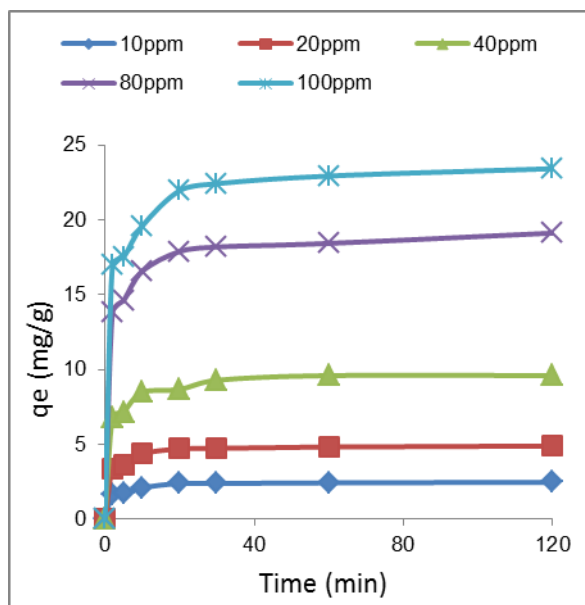


Figure 1: Time Course of MG Adsorption at Different Adsorbent Dosage.

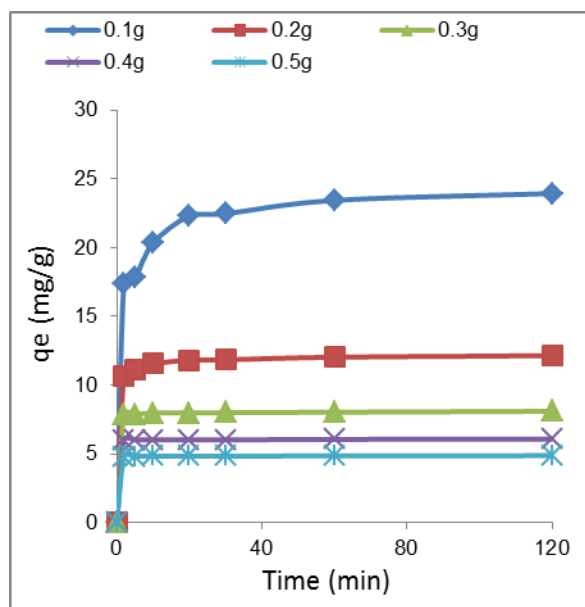


Figure 2: Time Course of MG Adsorption at Different Initial Dye Concentration.

The dimensionless separation factor, R_L , was estimated to be < 1 as shown in Table 2 indicating a favorable reaction for all the initial concentration. The result compare fairly well with that obtained for the removal of malachite green using odina wodier bark carbon (Vijayakumaran and Arivoli, 2012). The potential of the PS for the MG as seen from the Temkin isotherm showed a reasonable fit with a linear coefficient of determination, $R^2 = 0.9552$. However, the lower value of b_T is an indication of weak interaction.

The linear coefficient of determination for the Dubinin-Radushkevich isotherm was not very high. The isotherm constant B_D is related to the free energy, E , per molecule of adsorbate estimated to be $10^{-7} \text{ mol}^2 \text{ kJ}^2$. This constant gives a value of the mean free energy as 2.236 kJmol^{-1} which is an indication that the sorption is physical in nature. (Basar, 2006; Vijayaraghavan *et al.*, 2006; Monika *et al.*, 2009). It was also observed as contained in Table 1 that the value of q_D was half that of q_m . The reason for this may be due to the lower correlation coefficient obtained from the Dubinin-Radushkevich plot.

Sorption Kinetics

Pseudo-first-order: The data obtained were analyzed with the Lagergren pseudo-first order kinetic model. The plot of $\log (q_e - q_t)$ versus t gave the plot shown in Figures 3 and 4, corresponding to plantain stalk dose and malachite green dye concentration respectively. The values of k_1 and q_e were determined from the slope and intercept respectively as presented in Tables 3 and 4.

The result indicated that the data were not strongly correlated with R^2 ranging from 0.8507 to 0.9642 for the dose and 0.7961 to 0.9763 for the concentration. Also the theoretical values obtained from the equation did not yield reasonable values. Thus the rate of removal of MG from solution into the plantain stalk does not follow the pseudo-first-order equation. Indeed this is the general trend in most adsorption studies reported in the literature (Oladoja *et al.*, 2008; Gupta *et al.*, 2011; Tan *et al.*, 2012; Vijayakumaran and Arivoli, 2012).

Table 1: Isotherm Parameter for the Sorption of MG by Plantain Stalk.

Isotherm	Parameters	Values
Langmuir	q_m (mg/g)	30.68
	b (L/mg)	0.2945
	R^2	0.9765
Freundlich	$1/n$	0.628
	K_f (mg/g)	6.1892
	R^2	0.9925
Temkin	B	5.8066
	K_T L/mol	4.3791
	b_T KJ/mol	0.427
	R^2	0.9552
Dubinin-Radushkevich	$B_D \cdot 10 \exp^{-7 \text{mol}^2/\text{J}^2}$	1.0
	q_D mg/g	15.367
	E , KJ/mol	2.236
	R^2	0.8329

Table 2: Comparative R_L values of MG.

C_o	R_L^a	R_L^b
10	0.253	0.306
20	0.145	0.180
40	0.078	0.099
80	0.041	na
100	0.033	na

^a present study; ^b (Vijayakumaran1 and Arivoli 2012).
na = not available

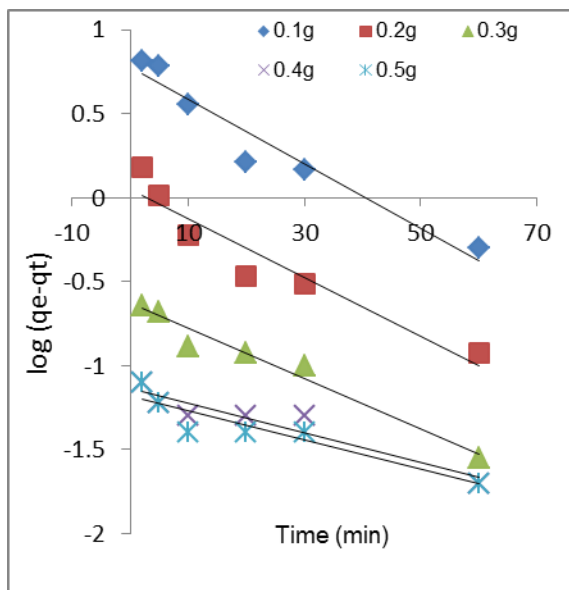


Figure 3: Pseudo-First Order Plot for Different Dose of PS.

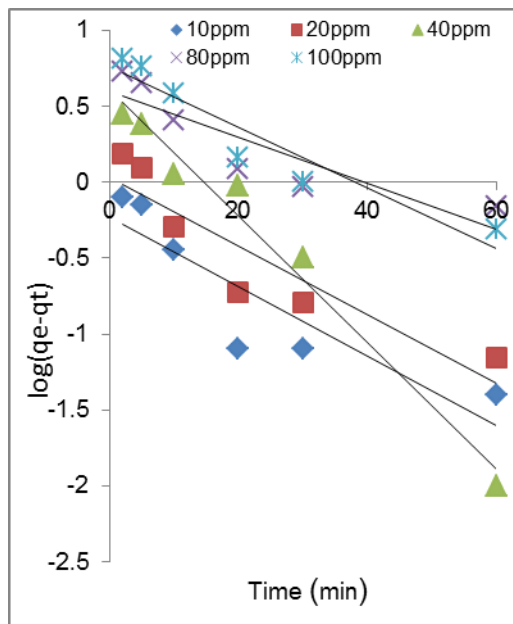


Figure 4: Pseudo-First Order Plot for Different Initial Concentration of MG.

Pseudo-second-order: The results obtained when the data was subjected to the *Pseudo-second-order* equation are shown in Figures 5 and 6 and the values tabulated in Tables 3 and 4 corresponding to the dosage and concentration respectively. The data applied showed very high correlation coefficients. The calculated values of q_e were also close to the experimental data. This confirms that the adsorption process followed the pseudo-second-order model.

CONCLUSION

The results of the present study revealed that plantain stalk has good potential as adsorbent for the treatment of malachite green in waste water. The equilibrium isotherm showed good fit to the Freundlich and Langmuir isotherms and both can be used to estimate the model parameter. The maximum loading capacity estimated from Langmuir isotherm was 30.68mg/g. The dimensionless separation factor, R_L , indicated a favorable reaction for all the initial concentration. The sorption process was found to follow the pseudo second order mechanism.

Table 3: Parameters for the effect of plantain stalk dose

Dose(g)	Pseudo-first-order equation			Pseudo -second-order equation			
	q_{e1}	k_1	R^2	q_{e2}	k_2	R^2	Experimental q_e
0.1	6.037	0.604	0.9390	24.39	0.586	0.999	23.43
0.2	1.117	0.223	0.9051	12.15	1.745	1.000	12.04
0.3	0.234	0.070	0.9642	8.13	4.393	1.000	8.04
0.4	0.074	0.029	0.8921	6.098	8.199	1.000	6.07
0.5	0.065	0.033	0.8507	4.90	7.599	1.000	4.87

Table 4: Parameters for the effect of MG concentration

Concentration (ppm)	Pseudo-first-order equation			Pseudo -second-order equation			
	q_{e1}	k_1	R^2	q_{e2}	k_2	R^2	Experimental q_e
10	0.587	0.053	0.7968	3.14	0.173	0.999	2.44
20	0.933	0.052	0.8437	4.95	0.780	1.000	4.83
40	4.056	0.096	0.9763	9.804	0.653	0.999	9.60
80	4.010	0.035	0.7961	19.61	0.622	0.999	18.43
100	5.724	0.046	0.9029	23.81	0.627	0.999	22.93

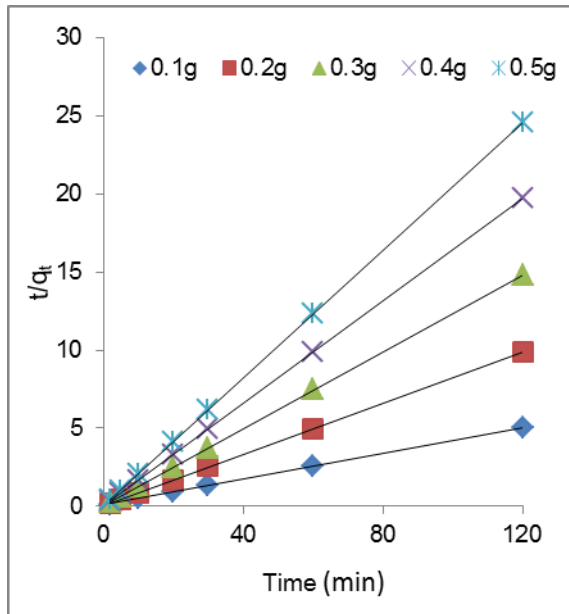


Figure 5: Pseudo-Second Order Plot for Different Dose of PS.

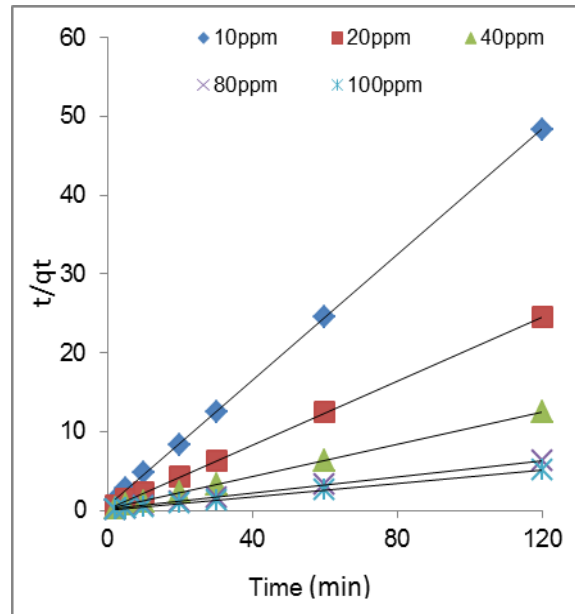


Figure 6: Pseudo-Second Order Plot for Different Initial Concentration of MG.

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