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Sorption of Malachite Green from Aqueous Solution using *Typha australis* Leaves as a Low Cost Sorbent

Abdoulaye Demba N'diaye [1,2]*, Youssef Aoulad El Hadj Ali [3], Ould El Moustapha Abdallahi [4], Mohamed Abdallahi Bollahi [1], Mostafa Stitou [3], Mohamed Kankou [2] and Driss Fahmi [3]

[1] Laboratoire de Chimie, Service de Toxicologie et de Contrôle de Qualité, Institut National de Recherches en Santé Publique, Nouakchott, Mauritanie

[2] Unité de Recherche Eau, Pollution et Environnement, Département de Chimie, Faculté des Sciences et Technique, Université de Nouakchott Al Aasriya, Nouakchott, Mauritanie

[3] Laboratoire de L'Eau, les Etudes et les Analyses Environnementales, Département de Chimie, Faculté des Sciences, Université Abdelmalek Essadi, Tétouan, Maroc

[4] Institut Supérieur d'Enseignement Technologique, Département des Sciences et Technologies des Aliments, Laboratoire de Biotechnologie Alimentaire, Rosso, Mauritanie

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Abstract

Malachite Green (MG) is a carcinogenic and mutagenic dye which is harmful for human and animal cells; its discharge through wastewater creates major environmental problems. For this reason, we have used *Typha australis* leaves, an abundant and available plant along the Senegal River for removing MG from aqueous solution. The adsorption equilibrium isotherms of MG onto *Typha australis* leaves as low cost sorbent were studied and modeled. In order to determine the best fit isotherm, the experimental data were fitted to the two-parameter isotherms (Langmuir, Freundlich and Jovanovic) and three-parameter isotherms (Sips, Redlich – Peterson and Toth) by nonlinear method. The best fitting isotherm was found to be the Langmuir isotherm. The monolayer adsorption capacities were found to be 85.21 and 56.88 mg g^{-1} at 21.4 and 31.4 °C, respectively. This homogeneity was also confirmed by the constants of Sips and Redlich–Peterson isotherms. The present study showed that the *Typha australis* leaves can be effectively used as low cost sorbent for the removal of the MG from its aqueous solution.

Keywords: Malachite Green, *Typha australis*, Senegal River, Isotherms, Nonlinear

1 Introduction

Malachite Green (MG) is a cationic dye that has been used for dyeing wool, silk, paper, leather and cotton as well as a biocide and disinfectant [1-3]. It has become one of the most serious environmental issues to treat MG dye-contaminated wastewater because MG dye is toxic and create a serious hazard to the aquatic system and human health [4].

Many treatment methods have been developed to remove MG from aqueous solution. These include electrochemical degradation [5], photo-degradation [6] and photocatalytic degradation [7]. However, these processes are costly and cannot effectively be used to treat the wide range of dye wastewater. Adsorption is accepted as the most efficient technique for removing pollutants from aqueous solution among many other methods thanks to its characteristics such as simplicity of design, high efficiency and economic feasibility [8]. However, the adsorption of dyes onto activated carbons has attracted many researchers, but its high cost inhibits its application on a large scale [9]. In this reason, researchers have concentrated on finding alternative natural adsorbents to activated carbon. Natural adsorbents are preferred for their biodegradable, non-toxic nature, low commercial value and highly cost-effective nature. A number of low cost adsorbents are reported in the literature. These include Lignin [10], Humic acid [10], *Prosopis cineraria* [11], organomineral sorbent-iron humate (Janos and Smıdova, 2005) [12], de-oiled soya [13], citric acid modified rice straw [14], hen feathers [15], palm ash [16], bottom ash [17], Rattan Sawdust [18], natural zeolite [19], chlorella-based biomass [20], Sea Shell powder [21], chemically modified rice husk [22], Castor bean presscake [23], *Cerastoderma lamarcki* shell [24] and *Annona squmosa* seed [25]. However, sorption potential of most of these low cost sorbents is generally low.

The objective of our study were to investigate the potential of using *Typha australis* leaves, an abundant and available plant along the Senegal River as a low cost sorbent to remove MG from aqueous solutions, to model the equilibrium of the process. The

Corresponding author: Abdoulaye Demba N'diaye, (a**)** Laboratoire de Chimie, Service de Toxicologie et de Contrôle de Qualité, Institut National de Recherches en Santé Publique, Nouakchott, Mauritanie and (b) Unité de Recherche Eau, Pollution et Environnement, Département de Chimie, Faculté des Sciences et Technique, Université de Nouakchott Al Aasriya, Nouakchott, Mauritanie. E-mail: abdouldemba@yahoo.fr.

retention capacity of MG onto the *Typha australis* leaves is investigated with using the nonlinear two-parameter models (Langmuir, Freundlich and Jovanovic) and three-parameter models (Sips, Redlich-Peterson and Toth). So, the sorption parameters obtained using the *Typha australis* leaves as low cost sorbent will be compared with the ones presented in the literature.

2 Material and Methods

2.1 Preparation of adsorbate

The stock solution of MG was prepared by dissolving 1 g of MG in 1 L of distilled water. All working solutions of desired initial MG concentrations were prepared by diluting the stock solution with distilled water. Other concentrations are prepared by dilutions of the stock solution and used to develop the standard curves using the Spectrophotometer UV1800 Ray Leigh.

2.2 Collection, Preparation and Characterization of Typha australis

Typha australis leaves were collected from the south of Mauritania. The *Typha australis* leaves were washed thoroughly with distilled water to remove dirt. The biomass was then air dried for 3 days followed by drying in an oven at 105 °C for 24 h. The dried biomass was ground, sieved to obtain particle sizes below 0.5 mm and stored in a dessicator before use [26]. The physicochemical characteristics of the *Typha australis* leaf are reported by [27]. The content of C, H, N, S and O of the *Typha Australis* were measured by using an Element Analyzer CHNSO Flash 2000 EA 1112. The surface morphology of *Typha australis* leaves before and after adsorption of MG was observed using a Scanning Electron Microscope (SEM).

2.3 Adsorption experiments

Batch adsorption studies were carried out by contacting 0.2 g of *Typha australis* leaves with 100 mL of MG solution of known initial dye concentration in flasks at a constant agitation speed of 150 rpm at two different solution temperatures 21.4 and 31.4 °C. The contact was made for 24 h, which is more than sufficient time to reach equilibrium [28]. After 24 h, the MG solutions were separated from the adsorbent by centrifugation. The left out concentration in the supernatant solution was analyzed using a UV Spectrophotometer 1800 Ray Leigh. The adsorption uptake at equilibrium time, q_e, was expressed by equation (1):

$$q_e = \frac{(C_i - C_e)V}{m} \tag{1}$$

where q_e is the amount of MG adsorbed by *Typha australis* leaves adsorbent (mg g^{-1}), C_i is the initial liquid-phase concentrations of MG (mg L^{-1}), C_e is the liquid-phase concentration of MG (mg L^{-1}), V is the solution volume (L) and m is the mass of *Typha australis* leaves adsorbent used (g). All batch experiments were conducted in triplicate and the mean values are reported.

2.4 Equilibrium adsorption isotherms

Two parameter models (Langmuir, Freundlich and Jovanovic) and three parameter models (Sips, Redlich-Peterson and Toth) are used to analyze the experimental adsorption data. Applicability of these models to fit the experimental in predicting the mechanism of adsorption is accomplished using solver Excel. The relative parameters of each equation are obtained using Sum of the Squares of the Errors (SSE) and the coefficient of determination (R^2) between the calculated and the experimental data by nonlinear method. The SSE and R^2 values, by using the Solver Excel, are determined respectively by following equations (2) and (3):

$$\text{SSE} = \left(q_{\exp} - q_{\text{mod}}\right)^2 \tag{2}$$

$$R^2 = 100\left(1 - \frac{\left\| q_{\exp} - q_{\text{mod}} \right\|^2}{\left\| q_{\exp} - q_{\text{avr}} \right\|^2}\right) \tag{3}$$

where q_{exp} (mg g^{-1}) is equilibrium capacity from the experimental data, q_{avr} (mg g^{-1}) is equilibrium average capacity from the experimental data and q_{mod} (mg.g^{-1}) is equilibrium from model. So that $R^2 \leq 100$ – the closer the value is to 100, the more perfect is the fit. Langmuir isotherm is valid for monolayer adsorption onto a surface containing a finite number of identical sites [29]. The nonlinear Langmuir model can be expressed by non linear equation (4):

$$q_e = \frac{q_m K_L C_e}{1 + K_L C_e} \tag{4}$$

where q_e is the amount of MG adsorbed per unit mass of *Typha australis* leaves (mg g^{-1}), k_L is the Langmuir constant related to the adsorption capacity (L g^{-1}), C_e is the concentration of MG in the solution at equilibrium (mg L^{-1}), q_m is the maximum uptake per unit mass of *Typha australis* leaves (mg g^{-1}). The factor of separation of Langmuir, R_L, which is an essential factor characteristic of this isotherm is calculated by using the relation (5):

$$R_L = \frac{1}{(1 + k_L C_0)} \tag{5}$$

where C_0 is the higher initial concentration of MG and k_L is the Langmuir constant related to the adsorption capacity (L g^{-1}). The parameters indicate the shape of the isotherm as follows: R_L values indicate the type of isotherm. When $R_L = 1$ adsorption is linear; when $0 < R_L < 1$, it is favourable, when $R_L = 0$, it is irreversible, while to be unfavorable, while when $R_L > 1$, it is unfavorable. Freundlich model is commonly used to describe the adsorption characteristics for a heterogeneous surface [29]. The nonlinear representation of the Freundlich model is as in equation (6):

$$q_e = K_F C_e^{1/n} \tag{6}$$

where K_F (mg g^{-1}) (L mg^{-1})^n and 1/n are the Freundlich constants related to adsorption capacity and sorption intensity, respectively. Jovanovic adsorption isotherm is similar to that the Langmuir model with the approximation of monolayer localized adsorption without lateral interactions. The assumptions in this model are same in the Langmuir model in addition with the possibility of the some mechanical contacts between the adsorption and desorbing model [29]. The nonlinear Jovanovic model can be expressed by equation (7):

$$q_e = q_m\left(1 - e^{-K_J C_e}\right) \tag{7}$$

where q_m (mg g^{-1}) and K_J (L mg^{-1}) are Jovanovic constants related to the adsorption capacity and the rate of adsorption, respectively. Sips isotherm is a combination of the Langmuir and Freundlich isotherms, which represent systems for which one adsorbed molecule could occupy more than one adsorption site [30]. The nonlinear representation of the Sips model is as in equation (8):

$$q_e = q_m \frac{K_S C_e^{n_S}}{(1 + K_S C_e^{n_S}\ )} \tag{8}$$

where q_m the Sips maximum adsorption capacity (mg.g^{-1}), K_S the Sips equilibrium constant (L mg^{-1}) and n_S the Sips model exponent describing heterogeneity. Redlich–Peterson isotherm model combines elements from both the Langmuir and Freundlich equation and the mechanism of adsorption is a hybrid one and does not follow ideal monolayer adsorption. It is used as a compromise to improve the fit by Langmuir or Freundlich [30]. The nonlinear representation of the Redlich–Peterson model is as in equation (9):

$$q_e = \frac{K_{RP} C_e}{1 + \alpha_{RP} C_e^{n_{RP}}} \tag{9}$$

where K_{RP} (L g^{-1}) and α_{RP} (L mol^{-1}) are the Redlich-Peterson isotherm constants, while n_{RP} is the exponent, which lies between 0 and 1. Toth adsorption model is developed to describe the heterogeneous adsorption systems which satisfy both low and high end boundary of adsorbate concentration. This model is the modified form of Langmuir isotherm with the intension of rectifying the error between the experimental and predicted data [31]. The nonlinear representation of the Toth model is as in equation (10):

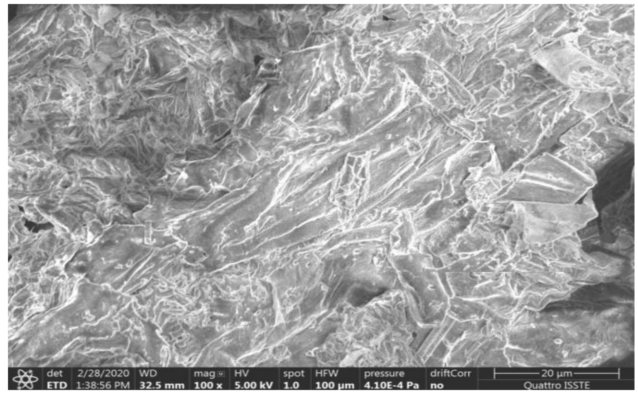
$$q_e = q_m \frac{C_e}{\left(1 + \alpha_T C_e\right)^{1/n}} \tag{10}$$

where q_m is the Toth maximum adsorption capacity (mg.g^{-1}), α_T is adsorptive potential constant (mg L^{-1}) and n Toth's heterogeneity factor.

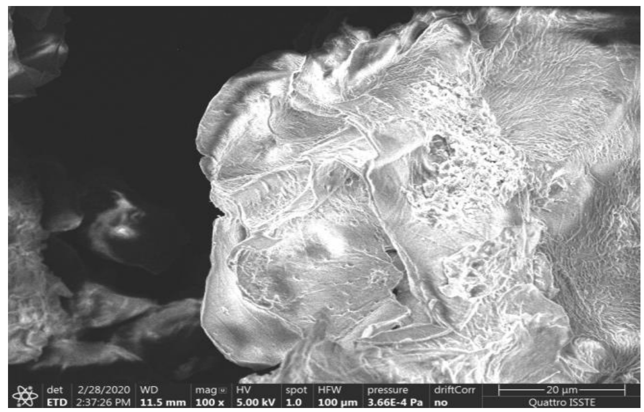
3 Results and Discussion

3.1 Characterization of Typha australis leaves

The outcome of the ultimate elemental analysis of *Typha australis* leaves indicates that oxygen (49.04 %) and carbon (43.93 %) are the major constituents of *Typha australis* leaves along with the quantifiable amount of hydrogen (5.87 %), nitrogen (0.88 %) and Sulfur (0.28 %). The surface morphology of *Typha australis* leaves adsorbent was evaluated according to the SEM images obtained in Figure 1. There is a change in surface morphology of the *Typha australis* leaves before (Figure 1(a)) and after the adsorption (Figure 1(b)) of MG.



(a)



(b)

Figure 1: Typical SEM micrograph of *Typha australis* leaves particle (100 magnification): (a) before MG adsorption and (b) after MG adsorption.

According to Figure 1(a), the *Typha australis* leaves before MG adsorption possesses uneven and irregular surface with considerable layers of rough heterogeneous pores which offers high possibility for dye molecules to be adsorbed [32]. Thus, the SEM image of *Typha australis* leaves after MG adsorption in Figure 1(b) reveals smoother surface features with apparent reduced pore structures, indicating the uptake and entrapment of MG molecules by the accessible pore vicinities of the *Typha australis* leaves surface. Figure 1(b) proves the engagement of MG onto *Typha australis* leaves and this may be related to the presence of carboxylic and hydroxyl groups within *Typha australis* leaves, as evidenced by the FTIR spectral reported by [27], which act as active sites for the adsorption of MG molecules.

3.2 Adsorption isotherm studies

The adsorption isotherm gives an idea of the equilibrium behavior of an MG–*Typha australis* leaves system. Figures 2 and 3 shows the experimental equilibrium data and the predicted theorical isotherms for the sorption of MG onto *Typha australis* leaves. The isotherm parameters, obtained using nonlinear method, are given in Table 1 for 21.4 and 31.4 °C. The values of 1/n, K_L and R_L are in between zero and one, indicating that the sorption of MG onto *Typha australis* leaves sorbent is favorable. The values of R^2 are compared, Langmuir isotherm are shown to have higher values than Freundlich and Jovanovic isotherms.

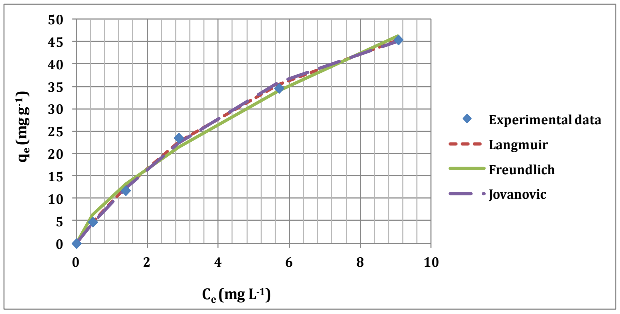


Figure 2: Langmuir, Freundlich and Jovanovic non linear for *Typha australis* leaves at 21.4 °C

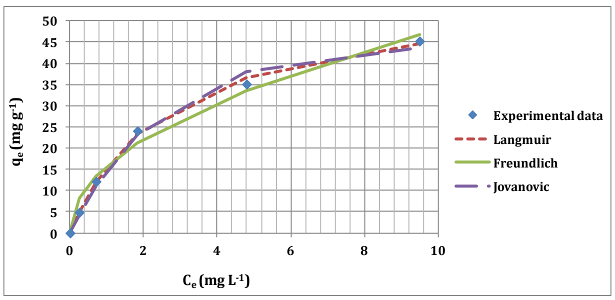


Figure 3: Langmuir, Freundlich and Jovanovic non linear for *Typha australis* leaves at 31.4 °C

Table 1: Two- parameter isotherm models for MG retention on the *Typha australis* leaves

Models	Parameters	21.4 °C	31.4 °C
Langmuir	q_m	85.21	56.88
	K_L	0.12	0.38
	R_L	0.077	0.026
	SSE	2.34	3.71
	R^2 (%)	99.85	99.76
Freundlich	1/n	0.67	0.48
	K_F	10.58	15.77
	SSE	9.57	25
	R^2 (%)	99.39	98.41
Jovanovic	q_m	56.38	44.69
	K_J	0.176	0.395
	SSE	3.04	13.94
	R^2 (%)	99.81	99.11

The lowest SSE values further confirmed the suitability of Langmuir model in describing the equilibrium data, suggesting the existence of monolayer adsorption of MG onto *Typha australis* leaves. This result is consistent with the literature where it is reported that the adsorption of MG using various adsorbents is well represented by Langmuir isotherm model [33-38].

The parameters for Sips, Redlich–Peterson and Toth isotherms, obtained using nonlinear method, at 21.4 and 31.4 °C are given in Table 2. The resulting curves of Sips, Redlich–Peterson and Toth parameters are compared to the experimental data at *Typha australis* leaves sorbent for MG removal in Figures 4 and 5. According to table 2, the results show that the experimental equilibrium data were best represented by the Sips, isotherm ($R^2 \geq 99.76$). The maximum adsorption capacities predicted by the Sips and Toth isotherms were lower than the Langmuir isotherm. Worth mentioning the maximum adsorption capacities obtained by Sips model, where q_m was equal to 31.72 and 20.75 mg g^{-1} at 21.4 and 31.4 °C, respectively. Values attained for *Typha australis* leaves are inferior to that reported by [38] for MG removal leaves of *Typha angustifolia* where q_m equal to 95.78, 83.53 and 83.07 mg g^{-1} at 25, 35 and 45 °C, respectively.

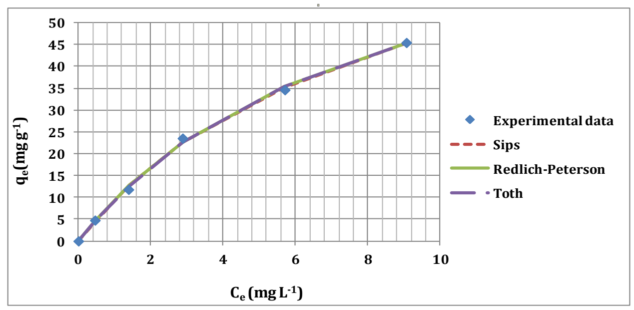


Figure 4: Sips, Redlich– Peterson and Toth non linear for *Typha australis* leaves at 21.4 °C

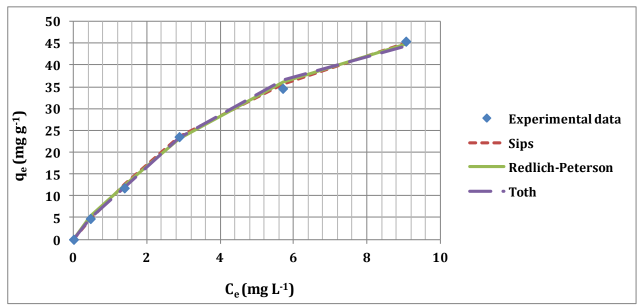


Figure 5: Sips, Redlich– Peterson and Toth non linear for *Typha australis* leaves at 31.4 °C

Table 2: Three- parameter isotherm models for MG retention on the *Typha australis* leaves

Models	Parameters	21.4 °C	31.4 °C
Sips	q_m	31.72	20.75
	K_S	0.55	1.79
	n_S	1.27	1.48
	SSE	2.17	1.06
	^{2}R (%)	99.87	99.94
Redlich-Peterson	K_{RP}	10.88	25.27
	α_{RP}	0.14	0.59
	n_{RP}	0.96	0.89
	SSE	2.32	2.36
	^{2}R(%)	99.85	99.85
Toth	q_m	1.33	1.94
	α_T	0.124	0.379
	n_T	0.125	0.091
	SSE	2.34	3.70
	^{2}R(%)	99.85	99.76

The Sips and Redlich–Peterson isotherm constants (n_S and n_{RP}) is nearly 1, this means that the equilibrium isotherm behaves as Langmuir, not as Freundlich isotherm. So, the application of the Langmuir, Sips and Redlich- Peterson isotherms showed that there was effective monolayer sorption and a homogeneous distribution of active sites on the surface of the *Typha australis* leaves sorbent. The monolayer adsorption capacities were found to be 85.21 and 56.88 mg g^{-1} at 21.4 and 31.4 °C, respectively. These results are comparable with those reported in the literature using other raw sorbents such as pineapple leaf powder [39], degreased coffee bean [40], Bivalve shell-Zea mays L husk leaf [41], Chitosan beads [42] and dead leaves of plane tree [43]. A comparison of q_m for MG dye using different sorbents which are previously reported, was performed and presented in Table 3. It was noted that *Typha australis* leaves had better sorption capacity in comparison to other sorbents listed here.

Table 3: Summary of previously published results for the removal of MG dye from aqueous medium

Adsorbents	q_m (mg g^{-1})	References
Lignin	31.2	[10]
Humic acid	6.4	
Rattan sawdust	22.4	[18]
Zeolite at 25 °C	23.94	[19]
Zeolite at 35 °C	25.14	
Chlorella biomass	18.4	[20]
Sea Shell powder	42	[21]
Activated Rice Husk	12	[22]
Castor bean presscake	37	[23]
Cerastoderma lamarcki shell	36	[24]
Annona squmosa seed	26	[25]
Diatomite at 25 °C	23.64	[37]
Diatomite at 35 °C	24.88	
Diatomite at 45 °C	27.10	
Typha australis at 21.4 °C	85.21	This study
Typha australis at 31.4 °C	56.88	This study

This, for instance, allows us to legitimately say that the *Typha australis* leaves is a better sorbent for the removal of MG. *Typha australis* leaves has a good sorption capacity and could be a reliable sorbent for the removal of MG.

4 Conclusions

The removal of MG dye using *Typha australis* leaves as sorbent was systematically investigated at 21.4 and 31.4 °C. The best fitting isotherm was found to be the Langmuir isotherm. The monolayer adsorption capacities were found to be 85.21 and 56.88 mg g^{-1} at 21.4 and 31.4 °C, respectively. This homogeneity was also confirmed by the constant of Sips isotherm. The findings from this study demonstrated that the use of *Typha australis* leaves as an alternative low cost sorbent for the removal MG from colored effluents is feasible. For future studies, the usability of *Typha australis* for dyes removal from real wastewater will be tested and as comparison, a fixed bed column will be employed to investigate the effect of reactor design.

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Competing interests

The authors declare that there is no conflict of interest that would prejudice the impartiality of this scientific work.

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