Australian Journal of Basic and Applied Sciences
2018 December; 12(12): pages 83-89
DOI: 10.22587/ajbas.2018.12.12.14
Original paper
Journal home page: www.ajbasweb.com
AENSI Publications

Biosorption of Methylene Blue and Orange II on deactivated *lichen Parmotrema dilatatum* : Modeling and kinetic studies

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Received date: 15 November 2018, **Accepted date:** 20 December 2018, **Online date:** 31 December 2018

Abstract

The flowering of industrial activities is generally accompanied by a phenomenon of effluent discharges containing many dangerous chemical substances. These water pollutants include Methylene Blue (BM) and Orange II (OII), which can cause human health problems and disrupt the balance of the aquatic system. Several effluent treatment methods exist but remain expensive and therefore inaccessible for developing countries such as Côte d'Ivoire.
The present work aims at the adsorption removal of these two toxic dyes on inexpensive plant biomass in this case deactivated lichens.
The various parameters influencing the adsorption such as the contact time, the initial concentration of the solution and the temperature, were studied. The kinetic study has shown that the biosorption process of BM and OII suitably follows pseudo-order two kinetics. The adsorption isotherms have shown that biosorption of BM is best described by the Freundlich model and that of OII by the Langmuir model. The thermodynamic study of the lichen-dye system showed that the adsorption process of the two dyes is spontaneous, endothermic for BM and exothermic for OII. The results obtained suggested a process of chemisorption and physisorption.
These results show the possibility of treatment of colored effluents by lichens.

Key words: lichens, biosorption, Methylene Blue, Orange II.

INTRODUCTION

In most developing countries, particularly in Côte d'Ivoire, aquatic environment contamination by pollutants from the textile, agrochemical and pharmaceutical industries is widespread. Methylene Blue and Orange II are among these harmfull substances that are used in textile (cotton, wood, silk and paper) (Bhuiyan et al., 2017), pharmaceutical industry (Pérez-Ibarbia et al., 2016). Their release in aquatic system is undesirable, not only because of their color, but products resulting from degradation are toxic, carcinogenic or mutagenic (Croce et al., 2017). Indeed, these polluants can cause gastrointestinal irritation in humans with nausea, vomiting and diarrhea (Dulce et al., 2017; Jabs and Drutz, 2001). In plants, they affect negatively the process of photosynthesis (Tahir et al., 2016). Various methods such as electrochemical oxidation (Nidheesh et al., 2018), ozonation (Wijannarong et al., 2013), electrocoagulation (Pirkarami and Olya, 2017), photo process Fenton (Sohrabi et al., 2017), nanofiltration, reverse osmosis (Nataraj et al., 2009), etc. are used in order to remove these substances from wastewater. These methods are expensive and have revealed limitations in their use due to the production of toxic products.

Biosorption is an alternative technology for the removal of a wide range of pollutants from aquatic areas. This technology involves the use of natural or artificial adsorbents such as sawdust (Dulman and Cucu-Man, 2009), activated carbon (Baysal et al., 2018), clay (Kausar et al., 2018), peanut shell (Liu et al., 2018), nanofiber membranes (Mousavi et al., 2018) alumina, silica (Kannan et al., 2008), banana peel (Munagapati et al., 2018) and orange peel (Arami et al., 2005) all derived from biomass. This approach has several advantages including a low cost due to the abundance of biomass, high selectivity efficiency regeneration with high yields. Lichens are plants resulting from the symbiotic association between an algae and a mushroom. Because of their high capacity of accumulation and retention of a wide variety of pollutants, they are widely used as indicators both of air quality and the environment health (Chatenet and Botineau, 2001).

In the present study, it is a question of carrying out the adsorption of Methylene Blue and Orange II on deactivated lichen biomass. The influence of contact time, initial dye concentration and temperature on lichens adsorption capacity of was studied.

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1 MATERIALS AND METHODS

1.1. Sampling

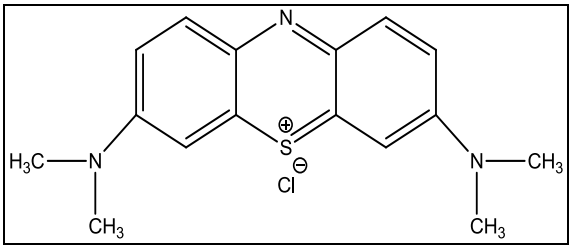
The thalli of the lichen Parmotrema dilatatum were collected in the forest reserve of the LAMTO ecological station (5.02 ° C West and 6.13 ° C North). This reserve is 174 km far from the city of Abidjan (Ivory Coast) and located on the Abidjan-Yamoussoukro axis, between Singrobo and Taabo, and stretches along the Bandama River. This space is a natural Park of 2500 ha, with a tropical climate. The average temperature per year is about 28.28 ° C. The annual precipitation is around 1194 mm and the rate of humidity is higher than 58%. Once the lichens were harvested, they were immediately placed in envelopes made with paper in order to avoid transpiration of the vegetal matter and then sent to the laboratory.

In laboratory, lichens are removed from their substrates and then cleaned manually to remove rubbish such as soil, leaves, dust or insects. Thalli were then softly well washed with distilled water. An appropriate quantity of lichens was placed in an oven at 80 ° C during 48 hours in order to deactivate them. The deactivated lichens were completely ground and sieved and the diameters of the particles those size belonging to the interval 125-250 µm is used in all experiments.

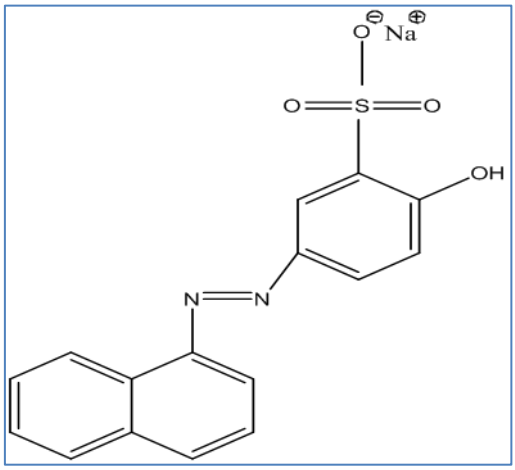
1.2. Dyes preparation

0.1g of Methylene Blue ($C_{16}H_{18}ClN_3S$, Cl 52015, molecular weight = 319.85g / mol, λmax = 664nm), and Orange II ($C_{16}H_{11}N_2OSO_3Na$, Cl 15510, molecular weight = 350.32 g / mol , λmax = 582 nm) are dissolved in a 2000 ml flask and then completed with distilled water in order to have an initial concentration of 50 mg/L for each colored solution. Dilution of the initial solutions permit to obtain the different desired concentrations.

The chemical structures of dyes are given in Figure 1.



(a)



(b)

Figure 1 : Chemical structures of BM (a) and OII (b)

1.3. Biosorption studies

The contact time study was carried out by stirring the mixture 5 to 200 min. The initial dye concentration (10 - 500 mg/L) effect for BM and (5 - 50 mg / L) for OII was also studied. The effect of temperature has been studied by varying temperature (20 - 60°C) to determine the thermodynamic parameters. The optical densities were read using a UV / visible spectrophotometer (WFJ-752).

The amount biosorbed dye per gram of biomass at equilibrum (mg.g-1) was calculated by using the following relationship (Jin et al., 2008) :

$$q_e = \frac{C_0 V_0 - C_e V}{m}$$

Where Co (mg/L) is the initial dye concentration, Ce (mg/L) the equilibrium dye concentration, Vo and V are respectively the initial and final volume (L) of the solution and m the quantity (mg) of used lichens.

1.4 Statistical analysis

All calculation was performed with Microsoft office Excel 2013 Professional (Microsoft Corporation, WA, and USA). Mean values and standard errors were determined from three individual measurements.

2. RESULTS AND DISCUSSION

2.1. Kinetic models of biosorption

In order to characterize adsorption kinetics of BM and OII on deactivated lichens, experimental kinetic data were applied to the kinetic model of pseudo-first order of Lagergren model (Lagergren, 1898) and pseudo-second order kinetics proposed by Ho and McKay (Ho and Mckay, 1999). These models are respectively described by the following relations (1) and (2) :

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$$\frac{dq}{dt} = k_1 \left(q_e - q_t \right) \quad (1)$$

$$\frac{dq}{dt} = k_2 \left(q_e - q_t \right)^2 \quad (2)$$

The Integration of equations (1) and (2) by respecting the boundary conditions which are :q_t=0 at t = 0 and q_t=q_t at t=t, leads to relations (3) and (4):

$$\ln\left(q_e - q_t \right) = \ln(q_e) - k_1 t \quad (3)$$

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{1}{q_e} t \quad (4)$$

Where k_1 (min-1) and k_2 (g.mg-1min-1) are respectively the pseudo-first and pseudo-second order constants and q_t the quantity of biosorbed dye at a given time t.

The kinetic parameters of pseudo-first order and pseudo-second order were calculated from the respective graph plots ln[(q_e- q_t)= f(t)] and t⁄q_t =f(t) (Fig. 2).

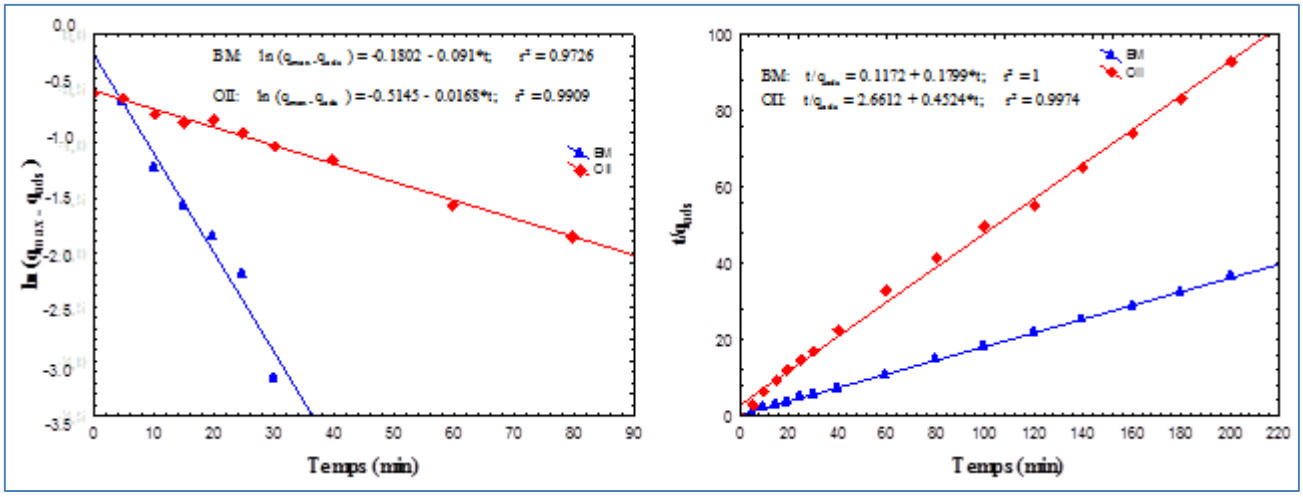

Figure 2 : Pseudo-first order (a) and pseudo-second order (b) models of BM and OII biosorption.

The results of the parameters calculation (k, q_e, r2) are presented in Table 1. The analysis of this table shows that the correlation coefficients r2 (0.9726, 0.9909) for the kinetic model of the pseudo-first order respectively for the BM and the OII, are relatively weaker than those of the pseudo-second order (1 ; 0.9974).

Table 1: Kinetic parameters (q_(e,exp) (mg/g), q_(e,th) (mg/g), k1(min-1) and k2 (g/mg.min) of pseudo-first order and pseudo-second order models (mean± standard deviation)

	Pseudo-first order				**Pseudo- second order**			
	$q_{e\,th}$	$q_{e\,exp}$	k_1	r^2	$q_{e\,th}$	$q_{e\,exp}$	k_2	r^2
BM	0.185 ± 0.003	5.533 ± 0.002	0.067 ± 0.003	0.887 ± 0.001	5.556 ± 0.003	5.533 ± 0.002	0.844 ± 0.001	1± 0.000
OII	0.649 ± 0.005	2.159 ± 0.003	0.014 ± 0.003	0.983 ± 0.002	2.244 ± 0.003	2.159 ± 0.003	0.053 ± 0.001	0.998± 0.001

In addition, the theoretical values (q_(e,th)) computed from the pseudo-second-order model showed good agreement with the experimental values (q_(e,exp)) compared to those of the pseudo-first order. Although the condition r2 ≥ 0.9 is satisfied for both models, the difference between the values (q_(e,th)) and (q_(e,exp)) allows to conclude that the biosorption of the BM and OII dyes on the deactivated lichen Parmotrema dilatatum is the kinetic of the pseudo -second order. These results are in agreement with those of Bentahar et al. (2017), and Jin et al. (2014) respectively in the adsorption of BM on powdered nut shells and OII on a zeolite coated with surfactant.

2.2. Adsorptions isotherms

Several adsorption isotherm models have been developed to describe these different interactions. In this study, Freundlich, Langmuir and Dubinin-Radushkevich (D-R) models were applied to the experimental data.

Langmuir isotherm involves monolayer adsorption on a surface containing homogeneous adsorption sites, without transmigration of adsorbates and without any interaction between the adsorbed molecules (Demirkiran, 2015). Once a site is occupied, no additional adsorption can take place, resulting in saturation of the surface (Kono and Kusumoto, 2015).

The Langmuir isotherm is translated by the following relation (Langmuir, 1918) :

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$$\frac{C_e}{q_e} = \frac{1}{q_{max} K_L} + \frac{C_e}{q_{max}} \quad (5)$$

Where q_max (mg/g) is the maximum dye biosorption corresponding to the saturation capacity and KL is the Langmuir constant.
In this work the application of experimental data to the Langmuir model is given in Figure 3.

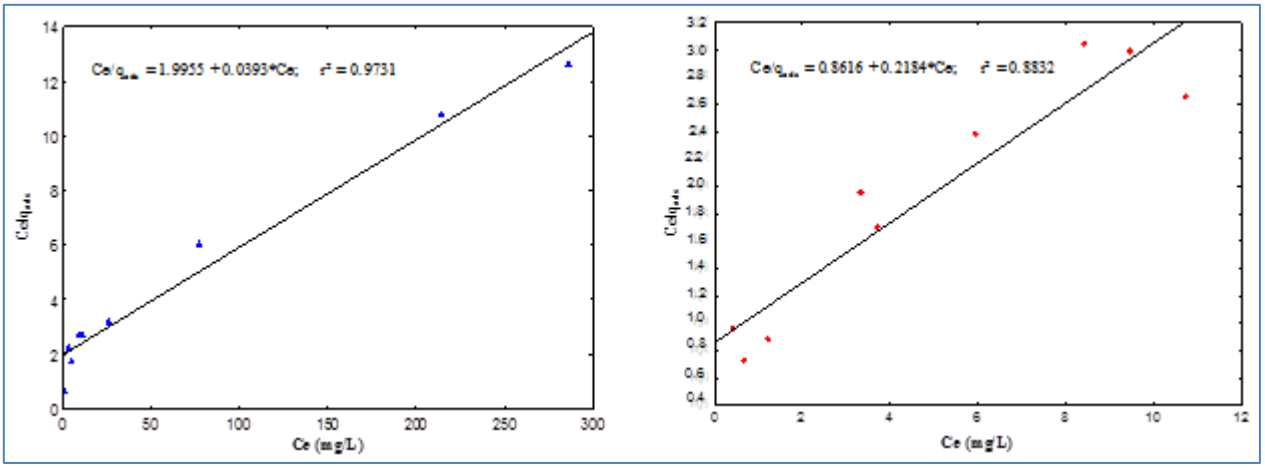

Figure 3 : Langmuir isotherm of adsorption of BM (a) and OII (b)

Freundlich model is based on the assumptions that adsorption is not limited to monolayer formation, hence a multilayer adsorption with active sites are not evenly distributed on the surface of the adsorbent (Mohamed et al., 2018).
The Freundlich adsorption isotherm can be expressed by the following relation (Freundlich, 1906):

$$\ln(q_e) = \ln(K_F) + \frac{1}{n}\ln(C_e) \quad (6)$$

Where KF (L/g) is the Freundlich constant linked to biosorption capacity and n is an empirical parameter that reports the intensity of biosorption and varies with the heterogeneity of the vegetal matter (Gorgulu Ari and Celik, 2013). The application of experimental data to the Freundlich model is given in Figure 4.

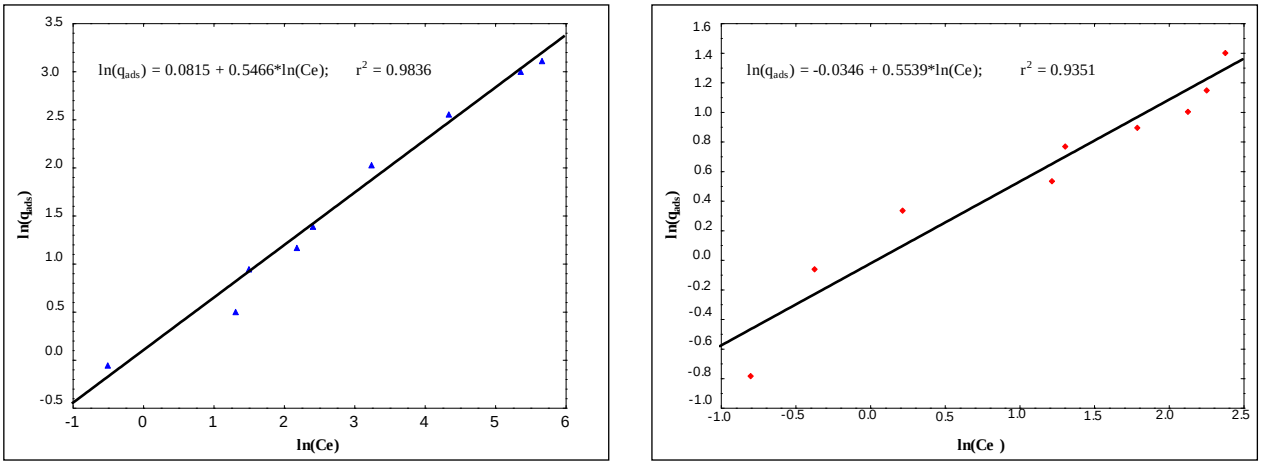

Figure 4 : Freundlich isotherm of adsorption of BM (a) and OII (b)

The Dubinin-Radushkevich isotherm is used to determine the physical or chemical nature of the biosorption processes of the dyes on the lichen Parmotrema dilatatum. The linear form of the D-R isothermal equation (Dubinin et al., 1947) is given by the following equation (7) :

$$\ln(q_e) = \ln(q_{max}) - \beta E^2 \quad (7)$$

Where q_e is the quantity of adsorbed dyes per gram of biomass at equilibrium (mol/L), q_max is the maximum biosorption capacity (mol/g), β is the activity coefficient related to the average energy of biosorption (mol2 / J2) and E is the potential of Polanyi defined by the following relation:
The average biosorption energy provides information about the biosorption mechanism. It is obtained by the relation given below :

$$\varepsilon\,(\text{kJ/mol}) = \frac{1}{\sqrt{(-2\beta)}} \quad (8)$$

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The application of the experimental data to the Freundlich model is given in Figure 5.

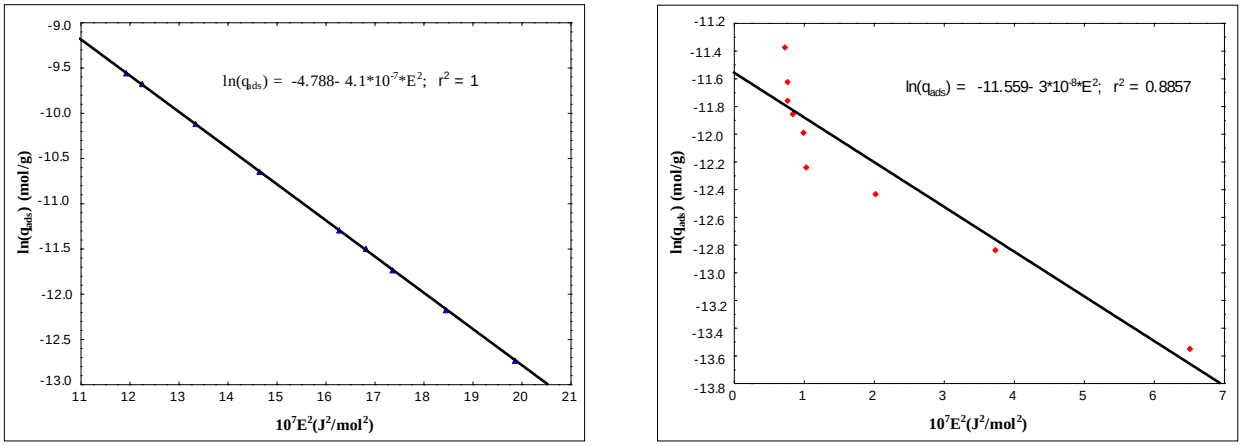

Figure 5 : Dubinin-Radushkevich adsorption isotherm of BM (a) and OII (b).

Constants derived from the model Langmuir, Freundlich and Dubinin-Radushkevich equations are gathered in Table 2.

Table 2 : BM and OII adsorption parameters (mean± standard deviation) on deactivated lichen according to Langmuir, Freundlich and Dubinin-Radushkevich (D-R) models.

	Langmuir			**Freunlich**			**Dubinin-Radushkevich**		
	Qmax(mg/g)	KL(L/mg)	r^2	1/n	KF(L/g)	r^2	Qmax(mol/g)	E(kJ/mol)	r^2
BM	25.445 ± 0.003	0.020 ±0.003	0.999 ± 0.00	0.547 ± 0.005	1.085 ± 0.003	0.984 ± 0.003	(8.329 ± 0.009)*10-3	1.118 ± 0.008	0.999 ± 0.001
OII	4.579 ± 0.002	0.254 ± 0.004	0.886 ± 0.004	0.554 ± 0.003	0.966 ± 0.005	0.935 ± 0.007	(9.549± 0.003)*10-6	4.082 ± 0.003	0.886 ± 0.002

The values of the constant 1 / n of Freundlich isotherm are 0.5466 and 0.5539 respectively for BM and OII. This result means that the adsorption of BM and OII on the deactivated lichen is favorable.
The correlation coefficients of Langmuir isotherm of Methylene Blue and Orange II are 0.9989 and 0.8857 respectively. For Freundlich isotherm, the coefficients are 0.9836 and 0.9351 respectively for BM and OII (Table 2). A comparison of these coefficients shows that biosorption of BM is well described by Langmuir model and that of OII by Freundlich model.
The calculated values of the mean energy of biosorption (Ɛ) are 1.118 and 4.082 kJ / mol respectively for BM and OII. These values are all less than 8 kJ / mol with high correlations. This result suggests that the adsorption of BM and OII dyes are all physisorption processes (Gorgulu Ari and Celik, 2013; Mohamed et al., 2018).

2. 3. Thermodynamic study
Thermodynamic parameters : Enthalpy ΔH °, Entropy ΔS ° and Free Energy ΔG ° were determined experimentally. The experimental results were applied to the following equations (9) and (10) (Elmoubarki et al., 2015):

$$\Delta Go = -RT\ln KD \qquad (9)$$

$$\ln(KD) = [\![\Delta S]\!]^0/R - [\![\Delta H]\!]^0/RT \qquad (10)$$

Where KD = qe/Ce is the distribution coefficient, R (8.314 J. mol-1.K-1) is the perfect gas constant, T (K) is the temperature of the solution.

The plot of the graph ln (KD) = f (1 / T) (Fig. 8) allowed to determine ΔH °, ΔS ° and ΔG°. The results are summarized in Table 3 below.

Table 3: Thermodynamic parameters (mean± standard deviation), for BM and OII adsorption.

	ΔG°(kJ/mol)		**ΔH°(kJ/mol)**		**ΔS°(J/mol)**	
T(K)	**BM**	**OII**	**BM**	**OII**	**BM**	**OII**
293	-70.439 ± 0.006	-3.021 ± 0.003				
303	-539.143 ± 0.005	-2.564 ± 0.004				
313	-602.858 ± 0.008	-1.858 ± 0.008				
323	-904.731 ± 0.007	-0.828 ± 0.004	8.752 ±0.012	-24.299 ± 0.007	30.213 ± 0.006	-72.180 ± 0.04
333	-1406.905 ± 0.022	-0.261 ± 0.006				

The values of Gibbs free energy variation (ΔG °) are all negative which means that the adsorption processes of BM and OII are spontaneous and therefore favorable, thus reflecting the affinity of the deactivated lichens for the cations and anions of dye (Vimonses et al., 2009; Pal et al., 2015).

The enthalpy of biosorption is -24.3 for OII. This negative value indicates the exothermic nature of the adsorption process which means that the adsorption of OII is favored by a decrease of the temperature (Toor and Jin, 2012).

The enthalpy of biosorption of BM is 2.57. The adsorption is endothermic and favorable by rising of temperature (Guyo et al., 2017; Güzel et al., 2015). Zhang et al have found the similar result (Zhang et al., 2013) which the positive value of [ΔH]^0 indicating the endothermic nature of adsorption of Methylene Blue on diatomite at different temperatures.

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The value of ⟦ΔS⟧^0 is 9.5 kJ / mol for the BM, this positive value suggests an increase in the disorder at the lichen/solution interface during adsorption (Ngulube et al., 2017). The negative value of OII indicates a decrease of desorder at the adsorbent / solution interface. This adsorption does not bring any change to the structure of the lichen Parmotrema dilatatum (Kalev and Toor, 2018; Li et al., 2017).

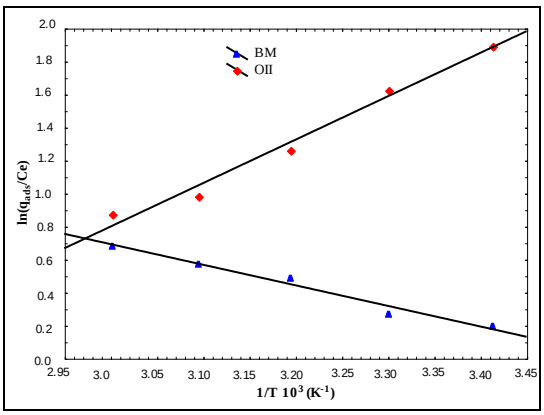

Figure 6 : Effect of temperature on sorption of BM and OII.

CONCLUSION

This study showed that lichen Parmotrema dilatatum deactivated can be used successfully as an effective adsorbent for the removal of Methylene Bleue and Orange II. The kinetics of biosorption of the two dyes is perfectly described by kinetics of pseudo-second order. The biosorption isotherm is well adjusted by Langmuir equation in the case of BM and Freundlich equation in the case of the OII. Thermodynamic studies have shown that adsorption is spontaneous for the two kinds of dyes, endothermic for BM and exothermic for OII and it leads to greater entropy in the case of BM and lower entropy in the case of OII.

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Citation: Kouassi Kouadio Dobi-Brice, *et al*., Biosorption of Methylene Blue and Orange II on deactivated *lichen Parmotrema dilatatum* : Modeling and kinetic studies. Australian Journal of Basic and Applied Sciences, 12(12): 83-89. DOI: 10.22587/ajbas.2018.12.12.14

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