

Research Article

Assessment of Removal Methylene Blue and Green Dye in Water by Adsorption in Aqueous Solution by the Pan Bottom from a Mining Waste from Betaré-Oya

Jeanne Atchana¹ , Chrisdel Chancelice Ndjeumi^{2,*} , Moutsina Ngalda³

¹Department of Chemical Engineering, Higher Technical Teacher's Training Colleg, University of Douala, Douala, Cameroon

²Department of Environmental Sciences, National Advanced School of Engineering of Maroua, The University of Maroua, Maroua, Cameroon

³Department of Chemistry, Faculty of Sciences, The University of Maroua, Maroua, Cameroon

Abstract

This work focuses on the study of the adsorption of methylene blue (BM) and a green dye (CV) in aqueous solution by the pan bottom from a mining waste from Betaré-Oya. The aim is to recover mining waste as an adsorbent. The influence of physico-chemical parameters on the kinetics and isotherm of adsorption of these dyes on the pan bottom was determined. The UV/Visible spectroscopy technique was used for the analyses. The green dye was study at spectral wavelength 570 nm. Green. Adsorption equilibrium times are 15 minutes for these dyes. Experiments show that when the bottom mass of the pan increases, the quantities adsorbed decrease. In fact, it is the adsorption yields that increase from 60 to 80% for BM and from 30 to more than 90% for CV. Variations in pH showed greater adsorption of dyes in basic medium ($\text{pH} \geq 10$) which indicates that the dyes are cationic. It was also observed that the amounts of BM and CV per unit of pan bottom increase with increasing initial concentration. The experimental data was analyzed by isotherm models. The value of the constant n of the Freundlich model for BM is 1.53, indicating that adsorption is unfavorable, whereas in the case of CV, adsorption is moderate. Langmuir and Temkin models better describe MB adsorption. The mechanism of adsorption of BM and CV is of chemical type. Pseudo-first-order, pseudo-second-order, Elovich and intra-particle kinetic models have been applied on data and the most suitable is the pseudo-second-order kinetic model.

Keywords

Adsorption, Pan Bottom, Dye, Isotherm Models, Kinetic Modelling, Water Treatment

1. Introduction

Nowadays, due to population growth, industrialization, the use of fertilizers in agriculture, water is strongly polluted by various harmful contaminants (pesticides, heavy metals, dyes

etc.) [1]. In fact, the dyes used in the paper, cosmetics, food industry and even more in the textile industry [2] are one of the sources of water pollution. The presence of colors in the

*Corresponding author: nchrisdel@yahoo.fr (Chrisdel Chancelice Ndjeumi)

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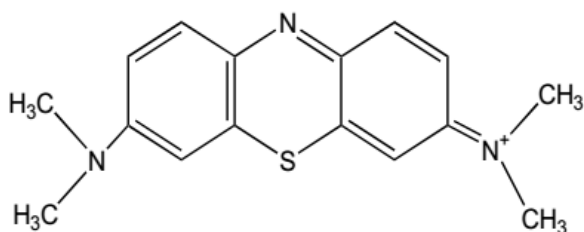
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environment negatively affects the aquatic ecosystem because it reduces the diffusion of light, thus inhibiting the photosynthesis of aquatic plants [3]. In humans, BM can cause irritation of the respiratory and intestinal routes, nausea, vomiting, diarrhea, headache, dizziness and blindness (irreversible lesion of the optic nerve) [4]. To overcome this situation, a wide variety of physical, chemical and biological techniques has been developed and tested in the treatment of effluents loaded with dyes. These processes include coagulation [5], flocculation [6], adsorption [7], electrocoagulation [8], precipitation, ion exchange, filtration on membrane [9]. Biodegradation does not allow satisfactory elimination of textile dyes because of the presence of aromatic nuclei in their molecules [10]. The elimination of adsorption colors on solid supports is a less expensive process, simple to implement, effective in low concentrations, and offers the possibility of regenerating adsorbent. The objective of this work is to reduce or even eliminate the content of methylene blue and green dye in aqueous solution by the background from the mining rejections of the locality of "B é á r é o y a" while valuing this material. More specifically we will study the influence of the parameters favoring the adsorption of these dyes as well as some adsorption isotherms and kinetic models.

2. Material and Methods

2.1. Material

The adsorbent material used here is the main waste (sludge) from mining activities (pan bottom). It was sampled in the East-Cameroon region, in the department of Lom and Djerem and more precisely in the district of B é á r é Oya. The bottom of the pan is obtained by successive washing of the sludge collected in the mining area using a sieve with a mesh of less than 1 mm and the coarse particles remaining above the sieve are recovered, dried with oven at 105 °C until total evaporation of the water and obtaining a constant mass, then ground and stored in jars to serve as an adsorbent.



Scheme 1. Molecular structure of MB.

Methylene blue, whose structure is shown in Figure 1, was obtained in the chemistry laboratory of the Faculty of Sciences of the University of Maroua. On the other hand, the "green dye" widely used by the dyers of the city of Maroua

was bought at the local market and an absorption spectroscopy was carried out there with the JENWAY 7310 spectrometer brand spectrophotometer to obtain the spectrum and thus have its absorption wavelength. All solutions were prepared with distilled water.

2.2. Experimental Protocols

The adsorption experiments were carried out in batch mode. In 100 ml bottles, 20 ml of solution (BM or CV) of concentrations 18 mg/L and 0.1 g of adsorbent are introduced. The flasks were shaken in an isothermal water bath shaker at 120 rpm at room temperature until equilibrium was reached. After decantation and filtration, the equilibrium dye concentrations in the solution were measured at 665 nm and 570 nm for BM and CV, respectively, using a UV-visible spectrophotometer. The residual amount of dye after adsorption is determined by interpolation on the calibration curve. Dye solutions at various concentrations of 3, 6, 9, 12, 15 and 18 ppm are prepared by gradually diluting a stock solution in distilled water. The reading of the optical densities of these solutions made it possible to draw the calibration curve ($OD = f(C)$) where C is the concentration of the dye solution (mg/l) and DO the optical density.

The residual quantities of dyes expressed in milligrams per gram of adsorbent are calculated by applying the relationship below:

$$Q_t = \frac{C_0 - C_t}{m} \times V$$

C_0 and C_t are respectively the initial concentrations and at time t (mg.L^{-1}).

V (en L) is the volume of adsorbate solution

m (en g), the mass at the bottom of the pan.

The adsorption rate (R) is also calculated from the following relationship

$$\%R = \frac{C_0 - C_t}{C_0}$$

2.3. Influence of Physico-Chemical Parameters on the Adsorption of Methylene Blue and Green Dye by the Bottom of the Pan

The influence of physico-chemical parameters on the adsorption kinetics were carried out under specific conditions. In particular the contact time (2 to 60 min) to determine the equilibrium state of saturation of the support by the substrate, the mass of adsorbent (0.1 to 0.7 g) to deduce the mass corresponding to an optimal adsorption, the pH of the medium (2 to 12), the initial concentration of dye (3 to 18 mg/L).

Each experiment was carried out in triplicate. The obtained data were analyzed by using Microsoft Excel 2007 and statistics plus software.

3. Results and Discussion

3.1. Characterisation of Green Dye

The absorption spectrum of the green dye is shown in Figure 1. This spectrum exhibits an absorption band that ranges from 570-610nm with a peak to maximum at the wavelength of 570nm. It is at this wavelength that the rest of the work was carried out.

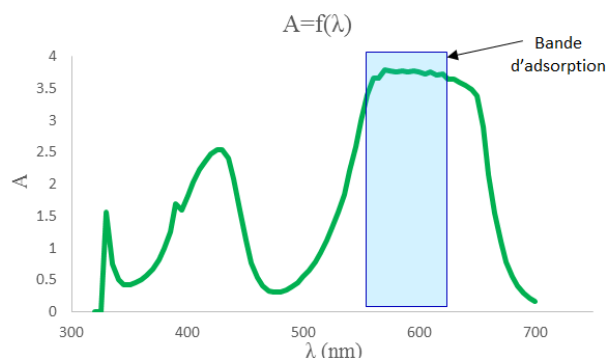


Figure 1. Absorption spectrum of CV.

3.2. Effect of Some Parameters onto BM and CV Adsorption

3.2.1. Influence of the Contact Time

The adsorption of BM and of the green dye (CV) in aqueous solution of identical initial concentrations of 18 ppm is carried out on a mass of 0.1 g of pan bottom. The results shown in figure 2 represent the variation in the quantity of the various dyes adsorbed per unit of pan bottom mass as a function of time.

The curves show an increasing adsorption of BM and CV during the first minutes until saturation of the adsorbent. Equilibrium is reached at almost 10 min for each dye. It is noted that the amount of adsorbed green dye increases until 15 min (5.32 mg/g) then a sort of release is observed with adsorption stability at 25 min (4.60 mg/g). On the other hand, the quantity of adsorbed BM (3.45 mg/g) no longer varies after 15 min.

The increasing adsorption phase during the first minutes corresponds to the progressive fixation of the BM or CV molecules on the adsorption sites on the bottom of the pan the formation of the plateau is the result of the saturation of the adsorbent, a sign of reaching the adsorption equilibrium. Similar results (15-25 min) at this contact time were obtained

by Dali [11] when he purified and characterized Algerian attapulgite by application to methylene blue adsorption.

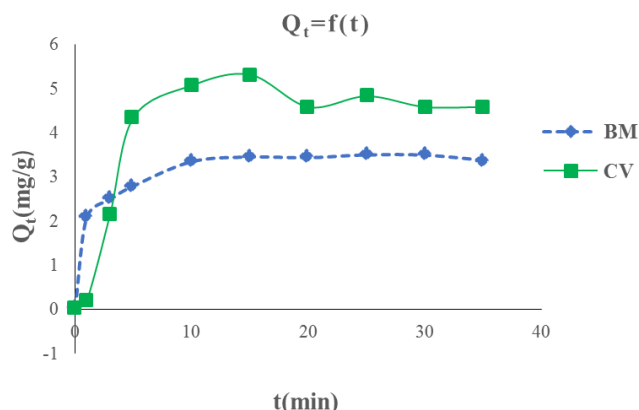


Figure 2. Influence of contact time on the adsorption of BM and CV.

In the rest of our manipulations, the contact times of 25 min and 15 min are retained respectively for the BM and the green dye.

3.2.2. Influence of the Mass of the Adsorbent

The results of varying the mass of adsorbent (0.1 to 0.7 g) on the adsorption of dyes are shown in Figure 3. At low adsorbent masses ($m \leq 0.25$ g), the BM is the most adsorbed (more than 60%), but beyond 0.3 g of adsorbent, the CV is more adsorbed than the BM. It is observed that whatever the dye, the quantity of adsorbed dye decreases with the increase in the mass of adsorbent (figure 3a). This seems incomprehensible, since increasing the amount of adsorbent would normally increase the number of adsorption sites. Looking at the adsorbed proportions as shown in Figure 3b, we see a gradual increase in adsorbed percentages with increasing adsorbent mass. In fact, the number of available adsorption sites increases with increasing amount of adsorbent which therefore results in an increase in the amount of adsorbed dye. Although the percentage adsorption increases with increasing adsorbent dose, the amount adsorbed per unit mass decreases. This result is similar to that of Mouthe et al. [12] who observed the same adsorption phenomena of crystal violet on biosorbents. Similar results were also observed by [13] when it adsorbed methylene blue and tartrazine in aqueous solution by raw clay and boboyo bridged clay and by Bouacherine when it removed specific pollutants by adsorption on activated carbon and treated and untreated clay [14].

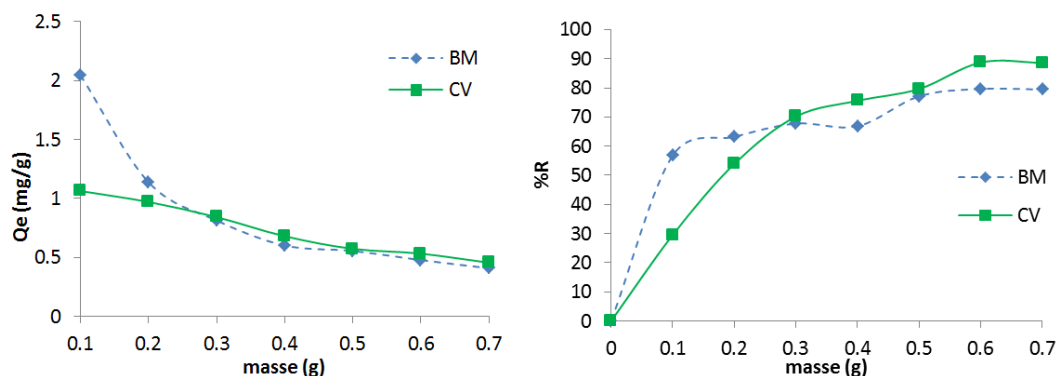


Figure 3. (a) Influence of the bottom mass of the pan on the amount of molecules of BM and green dye absorbed at equilibrium (b) Variation in BM and CV removal rate as a function of pan bottom mass.

3.2.3. Influence of pH

The pH effect was studied in the pH range from 2 to 12 and the results obtained are shown in Figure 4.

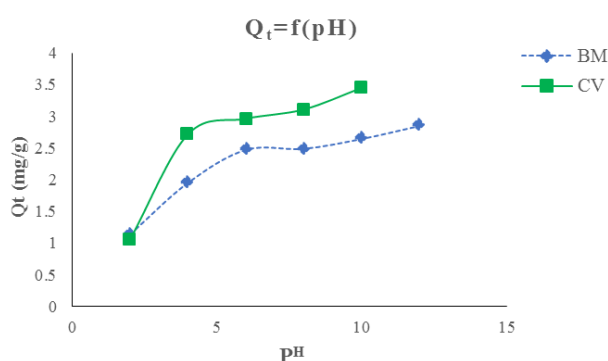


Figure 4. Influence of pH on the amount of BM and green dye adsorbed.

From figure 4, we note an increase in the quantity of dyes adsorbed when passing from the acidic medium (pH=2) to the basic medium (pH=12 for the BM) and (pH=10 for the CV). In fact, the adsorbed quantities of BM and of CV pass respectively from 1.063 (pH=2) to 2.851 (pH=12) for BM and 1.063 (pH=2) to 3.454 (pH=10) for CV.

This shows that these dyes are more easily adsorbed in basic medium. Work on BM has shown that it is a basic dye with regard to the behavior of adsorption, we can say that the pan bottom surfaces would be negatively charged. There is therefore an electrostatic interaction between the bottom surface of the pan and our dyes. This result is similar to that of Zied when he adsorbed magnetic latex [15].

3.2.4. Influence of the Initial Concentration of Dyes

The solutions of different concentrations are adjusted to pH=12 and to pH=10 respectively for BM and CV; The results obtained are represented by figure 5:

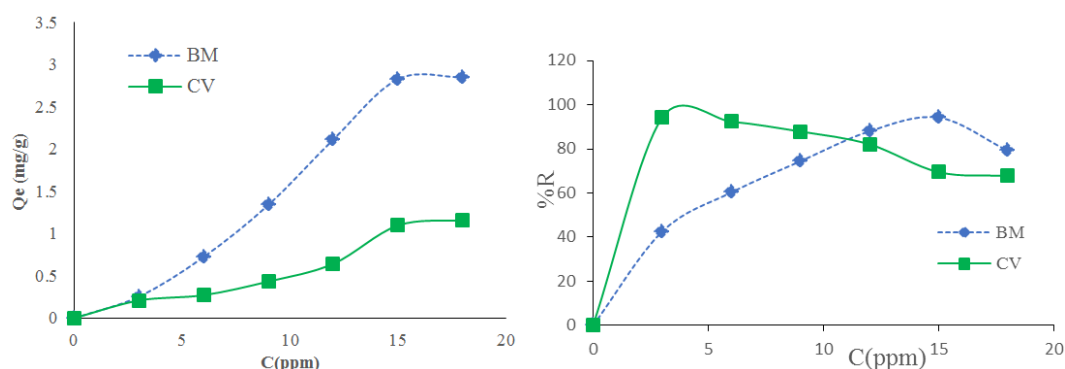


Figure 5. (a) Influence of the initial concentration on the adsorbed amount of BM and green dye (b) Variation of the removal rate of BM and of the green dye according to the initial concentration.

According to the results (figure 5a), the quantity of adsorbed dye increases with the increase in the initial concentrations of the dyes until reaching a level for high initial

concentration (15 ppm) which means that the capacity of maximum adsorption of the bottom of the pan is reached beyond 15 ppm of initial concentration. Looking at

the level of the adsorption rates (figure 5b), for the CV, we note that the more the initial concentration increases, the more the adsorption rate decreases, ranging from 95% at 3 ppm to less than 70% at more than 15 ppm. On the other hand, BM is better adsorbed (94%) at high concentrations (≥ 15 ppm). These results corroborate that of Emna when it adsorbed the anionic dye by natural clay [16].

3.3. Adsorption Kinetics Models

In order to elucidate the mechanism of adsorption of BM and green dye molecules on the bottom of the pan, the experimental data were compared by pseudo first order, pseudo second order, Elovich, and intra-particle diffusion models. The results are presented in Table 1.

Table 1. Adsorption Kinetic parameters models.

Models	Pseudo-premier order			Pseudo-second order			Elovich			Intra-particle diffusion		
Parameters	$K_1 (\text{min}^{-1})$	$Q_m (\text{mg/g})$	R^2	$K_2 (\text{g/min.g})$	$Q_e (\text{mg/g})$	R^2	B	A	R^2	C ($\text{m}^2.\text{S}^{-1}$)	$K_{int} (\text{mg.L.min}^{1/2})$	R^2
BM	0,20	0,69	0,43	0,45	3,52	0,99	1,45	5,08	0,70	1,28	0,46	0,71
CV	0,15	0,02	0,25	3,55	0,20	0,98	20,2	0,12	0,77	0,04	0,03	0,69

It is apparent from Table 1 above that the pseudo-first-order, Elovich and intra-particle models cannot be applied to explain the adsorption of BM and CV because of their low correlation coefficients which are all less than 0.90. However, the correlation coefficient calculated for the pseudo-second-order kinetic model describes well the experimental results of the adsorption of BM and CV by the bottom of the pan.

We also note that the values of the calculated theoretical adsorbed quantities (3.52 and 0.20mg/g) of the pseudo-second order kinetic model are very close to experimentally adsorbed quantities (3.50 and 0.213mg/g). This model reflects the existence of strong interactions between the pan bottom surface and the dyes. We can thus affirm that we are in the presence of chemisorption [17]. Indeed, the pseudo second-order kinetic model is based on the assumption that, in the liquid phase, adsorption takes place in two stages: the first being the diffusion of the green adsorbate on the surface and the second, the adsorbent interaction adsorbate. So the adsorption of BM and CV is done in two steps.

Pseudo-second-order kinetic models show that the adsorption takes place in multilayers. The phenomena operating in

this model confirm chemisorption [18].

These results are corroborated by several studies which have shown that the adsorption kinetics of dyes on mineral materials obey the pseudo-second order kinetic model [19, 20].

The low values of the intra-particle diffusion constants show that the diffusion through the pores of the adsorbents takes place slowly. Moreover, the value of the correlation coefficients of this model are lower than those of the pseudo-second order model, it can be concluded that diffusion is not the limiting step for this adsorption mechanism. These results are in agreement with those of Belaid and Kacha, on the kinetic and thermodynamic study of the adsorption of dyes [21, 22].

3.4. Equilibrium Isotherm Models

The study of adsorption isotherms aims to determine the mechanisms involved in the process of adsorption of the two dyes, namely BM and CV, by the pan bottom.

Table 2. Correlation coefficients and constants of Langmuir, Freundlich, Temkin and D-K-R for the adsorption of BM and CV.

Models	Langmuir				Freundlich			Temkin			D-K-R			
Parameters	R_L	$K_L 10^{-3}$	$Q_m \text{ mg/g}$	R^2	K_f	N	R^2	K_T	B	R^2	E (KJ/mol)	$B 10^{-7}$	$Q_m (\text{mg/g})$	R^2
BM	1	3,87	51,81	0,999	0,15	1,53	0,7	0,67	1,03	0,93	790,57	8	54,52	0,24
CV	1	2,81	45,45	0,998	0,023	0,98	0,999	1,46	0,57	0,96	1290,99	3	9,77	0,75

With regard to this table 2, the correlation coefficients of the adsorption models are greater than 0.90 for the Langmuir and Temkin models for the adsorption of BM on the pan bottom. But in the case of the CV, the Langmuir, Freundlich and Temkin models present acceptable values of the correlation coefficients, that is to say greater than 0.90 with the bottom of the pan.

The results obtained show that the model of Langmuir and Temkin better describes the phenomenon of adsorption at the bottom surface of the pan for the case of BM, on the other hand for the case of CV, it is the model of Langmuir, Freundlich and Temkin. which better describe the phenomenon of adsorption because of their higher correlation coefficient.

The Langmuir and Freundlich model for the adsorption of BM and green dye, which shows a good affinity with the process involved, is explained by the fact that the surface of an adsorbent solid is heterogeneous, so there is a multilayer with the interaction between adsorbed molecules. Similar results were obtained when studying the adsorption of carmoisine and crystal violet in aqueous solution by Boboyo clay [23].

The separation factor or equilibrium parameter RL is essential to describe the phenomenon of adsorption from the Langmuir isotherm model. The value of this parameter is equal to 1 for each of our dyes tested, this means that the adsorption isotherms of BM and CV are linear [24].

The value of the constant n of the Freundlich model for BM is equal to 1.53, a value greater than 1 thus indicating that the adsorption is weak, whereas in the case of CV, the adsorption is moderate because of the value of n which gives 0.98, a value between 0.5 and 1. Thus we can say that there is a good affinity between our bottom of the pan and the CV, on the other hand for the BM, it is weak. These are almost similar to that of GAINEUNBO when he adsorbed methylene blue and methyl orange in aqueous solution by activated carbon from the hulls of vitex doniana [25].

The Freundlich constant (K_f) values are essential to describe the power of an adsorbent with respect to its adsorbate in an adsorption phenomenon. We find that these K_f values for the two dyes (BM and CV) are very low (0.15 for BM and 0.023 for CV), these values mean that the adsorption of BM and CV by the pan bottom is not important.

The adsorption energy value determined from the Dubinin-Radushkevich model gives an indication of the type of adsorption. The adsorption energy values E are equal to $790.57 \text{ KJ.mol}^{-1}$ for the BM and $1290.99 \text{ KJ.mol}^{-1}$ for the green dye. These experimental values are all greater than 80 KJ.mol^{-1} , which indicates that the adsorption of BM and green dye on the bottom of the pan is of a chemical type [26].

4. Conclusion

The study of the adsorption of dyes by the pan bottom is a contribution to the treatment of textile effluents. The objective of this work was to valorize the pan bottom from mining

waste as an adsorbent for BM and CV. The influence of certain physicochemical parameters on the adsorption such as: the pH, the concentration of the substrate, the mass of the adsorbent has been highlighted.

It emerges from the study of the adsorption of BM and CV by the pan bottom that:

1. The adsorption of BM and CV molecules by the pan bottom is rapid and equilibrium is reached at 15 minutes. The CV has much more affinity with the bottom of the pan and is therefore more adsorbed (90%) than the BM (80%).
2. The pseudo-second-order kinetic model is the model that best suits the retention of BM and CV molecules; this implies, according to the hypothesis, that adsorption takes place in two phases: the diffusion of the adsorbate towards the surface of the adsorbent, followed by the adsorbent-adsorbate interaction.
3. The increase in the initial concentration of BM and CV leads to an increase in adsorption. A decrease in the amount adsorbed at equilibrium was observed with increasing pan bottom mass.

The best correlations were obtained with the Langmuir and Temkin models for BM. While the models that best describe CV adsorption are Langmuir, Freundlich and Temkin models. The value of the constant n of the Freundlich model for BM is 1.53 thus indicating that the adsorption is weak, whereas in the case of CV, the adsorption is moderate because the coefficient n is less than 1. Thus, we can say that there is a good affinity between our pan bottom and the CV molecules, on the other hand for the BM, it is weak.

Abbreviations

BM	Methylene Blue
CV	Green Dye

Conflicts of Interest

The authors declare no conflicts of interest.

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