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Adsorption of cationic dye from aqueous solution onto bentonite Maghnia

Fatiha Mahammedi*

[1]*Laboratory of Inorganic Chemistry and Environment, Abou Bakr Belkaid University 13000 Tlemcen, Algeria*

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Abstract

In this work, natural clay (BN) was studied for the removal of Basic dye from aqueous solution in batch system. The effects of initial VM concentration, contact time, solution temperature and solution pH on BN adsorption were investigated. The adsorbent was characterized by means of FTIR and XRD analysis. The equilibrium adsorption data were analyzed by Langmuir, Freundlich, and isotherm models. The pseudo-first-order kinetic model provided the best fit to the experimental data compared with pseudo-second-order kinetic adsorption models. The thermodynamic parameters such as, Gibbs free energy changes (ΔG°), standard enthalpy change (ΔH°) and standard entropy change (ΔS°) were determined. Adsorption of VM on BN is exothermic and spontaneous in nature.

Keywords:Aqueous, adsorbent, bentonite, methyl violet, wastewater

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*Corresponding author.
E-mail address:mahafati_2006@yahoo.fr

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1. Introduction

Aqueous effluents are well known as massive industrial wastewater. The presence of very low concentrations of these effluents are highly visible and undesirable and potentially inhibiting photosynthesis. The main pollution source of colored effluents comes from textiles, leather, printing, laundry, tannery, rubber, painting, etc., processes [1]. Untreated disposal of this colored water into receiving water body causes damage to aquatic life and also severe damage to the human health [2,3].

Methods and effluent treatment for dyes may be divided into three main categories namely physical, chemical and biological. Among them adsorption technology is generally considered to be an effective method for quickly lowering the concentration of dissolved dyes in an effluent [4]. Dyes can be effectively removed by adsorption process. Activated carbon [5,6], natural clays [7], modified clays [8,9], some industrial wastes and by-products [6] have been used as adsorbents for removal of organic compounds from waste waters. Naturally occurring clays have shown good results as an adsorbent for the removal of various metals [10,11], organic compounds [12] and various basic dyes [13].

Natural clay minerals have been the subject of many research studies because of their wide industrial applications. This is justified by their natural abundance in most continents of the world and at low cost, their high sorption properties and potential for ion exchange [14]. There are many types of clay material such as Bentonite, kaolinite, smectite, serpentine, vermiculite, and sepiolite [15]. These clays are produced by the decomposition of rock minerals [16].

Bentonite, one of the most common clays, has been used in a variety of industrial fields as a catalyst, filler in polymer, cosmetics, and pharmaceuticals, because of its high surface area and swelling property [17]. The natural Bentonite has limited adsorption capability, which restricts its practical applications [18]. Bentonite as one of the most extensively studied adsorbent material because it is naturally available, inexpensive and has good adsorption capacity mainly due to its physical and chemical stability, high superficial area and good cation exchange capacity [19 ,20]. The surface characteristics of bentonite are critical to its ability to attract various chemical species. However, bentonite from different parts of the world varies in mineralogy as this is affected by the geochemical conditions during its formation.

The objective of the present work is to study the adsorption characteristics of VM from aqueous solution using Algerian clay. The influences of adsorption conditions such as contact time, pH, initial concentration of VM and temperature effect were investigated. In addition, the evidence for physicochemical characteristics of Algerian clay obtained from X-ray diffraction (XRD)technique.

2. Experimental

Dye

Methyl violet (MV) labeled Merch (C.I.42535) was retained as a cationic dye and contains a secondary amino group (see figure 1). It has a maximum absorbency at wavelength 580nm on a UV visible spectrophotometer. The chemical formula and molecular mass of MV is $C_{24} H_{28} Cl N_3$ and 393.95g/mol, respectively.

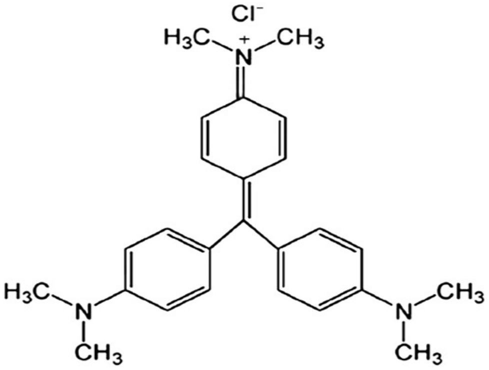

Figure 1. Chemical structure of the methyl violet dye

2.1. Adsorption procedure

Adsorption measurements were determined by batch experiments. The effect of contact time on the adsorption capacity of bentonite was studied in the range 1-120 min at an initial concentration of 100 mg/L. Adsorption kinetics was studied using an initial concentration of 100 mg/L with the adsorbent dosage of 0.1 g was contacted with 50 mL. Adsorption isotherms were studied at various initial concentrations of MV in the range of 100 - 600mg/L and the experiments were conducted at different constant temperatures in the range 10°C - 50°C. The amount of MV adsorbed per unit mass of bentonite was calculated by using the mass balance equation given in Equation (1) [21].

$$q_e = \frac{(C_o - C_e)V}{m} \qquad (1)$$

where qe is the maximum adsorption capacity in mg/g, C_o is the initial concentration and C_e is the concentration at equilibrium of MV solution in mg/L, V is the volume of the MV solution in mL and m is the mass of the bentonite in grams.

The sorption capacity at time t, qt (mg/g) was obtained as Equation (2) [22]:

$$q_t = \frac{(C_o - C_e)V}{m} \qquad (2)$$

Where C_o and C_t(mg/L) are the liquid phase concentrations of MV at initial and a given time t, V is the solution volume and m the mass bentonite (g).

3. Results and discussion

3.1 Characterization of bentonite

Figure 2 shows XRD pattern of bentonite from the pattern, we can conclude that quartz is the major constituent of bentonite.

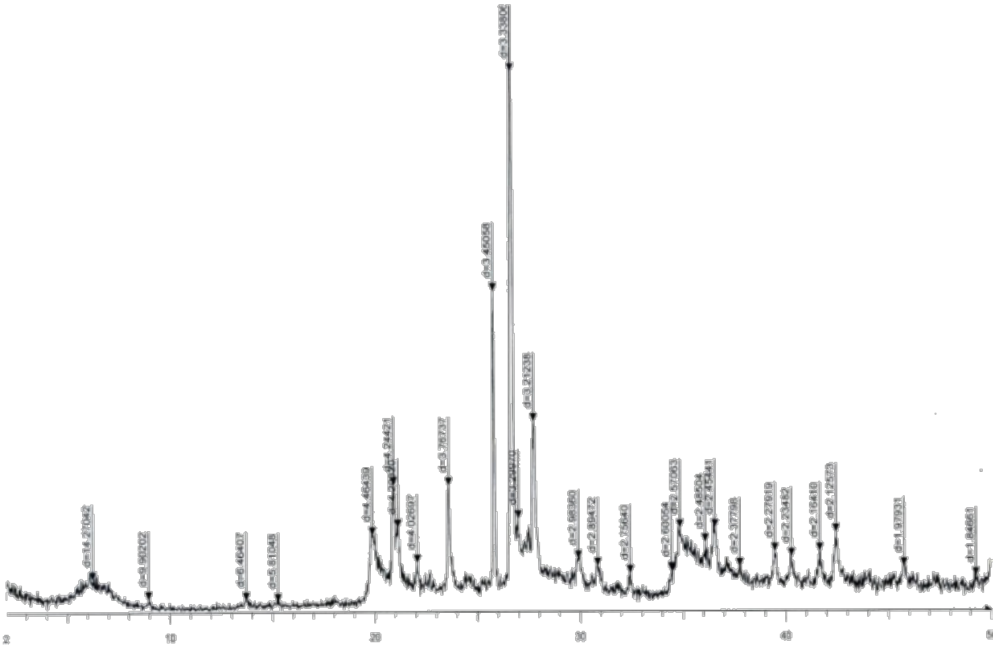

Figure 2. **RX** Diffractograms of Bentonite

3.2. Effect of contact time on adsorption

A series of experiments was performed to optimize the adsorption time at an initial dye concentration of 10−5 mol/L. The effect of contact time on the adsorption rate of dye is presented in Figure 3, the effect of contact time for the removal methyl violet dye by bentonite showed rapid adsorption of dye by increasing the contact time. This was caused by strong attractive forces between the dye molecules and the adsorbent; fast diffusion onto the external surface was followed by fast pore diffusion into the intra particle matrix to attain rapid equilibrium.

The influence of time is achieved at natural pH of the solution for an initial concentration of 100 mg/1, with a mass of clay of 0.1mg and at room temperature. Figure 4 shows the time course of adsorption equilibrium of methyl violet onto bentonite. The amount of dye adsorbed by adsorption on bentonite was found to be rapid at the initial period of contact time and then become slow and stagnant with increase in contact time; the mechanism of adsorbent removal can be described in migration of the dye molecule from the solution to the adsorbent particle and diffusion through the surface [23,24].

3.3. Effect of pH on Adsorption

The pH of a solution is of significance for its effect on the adsorbent, as well as on the adsorbate. The influence of pH on dye removal was determined by performing the adsorption experiments at different initial pH of the solution (1-12) at room temperature. The pH of the solution was adjusted with HCl (0.1 N) or NaOH (0.1 N) solution by using a HANNA 210 pHmeter equipped with a combined pH electrode. The adsorption of Methyl Violet onto Bentonite is highly dependent on pH of the solution. The Figure 4 shows that the adsorption increased with increasing pH. The removal dye is low in the acid pH region because the hydrogen ions neutralize the negatively charged clay surface thereby decreasing the adsorption of the positively charged cation because of reduction in the force of attraction between adsorbate and adsorbent. The removal of dye is more at higher pH, because the surface of used clay is negatively charged. Therefore, the electrostatic attractive force between the colorant dye, which has a positive charge, and the adsorbent surface increases, and consequently, the rate of dye adsorption increases, the highest dye removal was detected in pH 6. Similar results have been reported for the adsorption of Methylene blue on morocco clay [25-30].

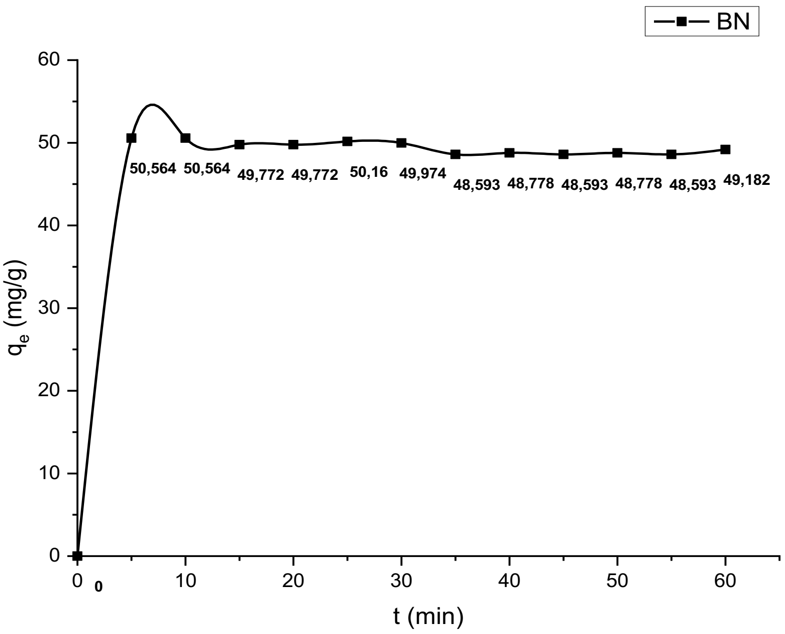

Figure 3. The Effect of Contact Time on Adsorption Methyl Violet onto Bentonite.

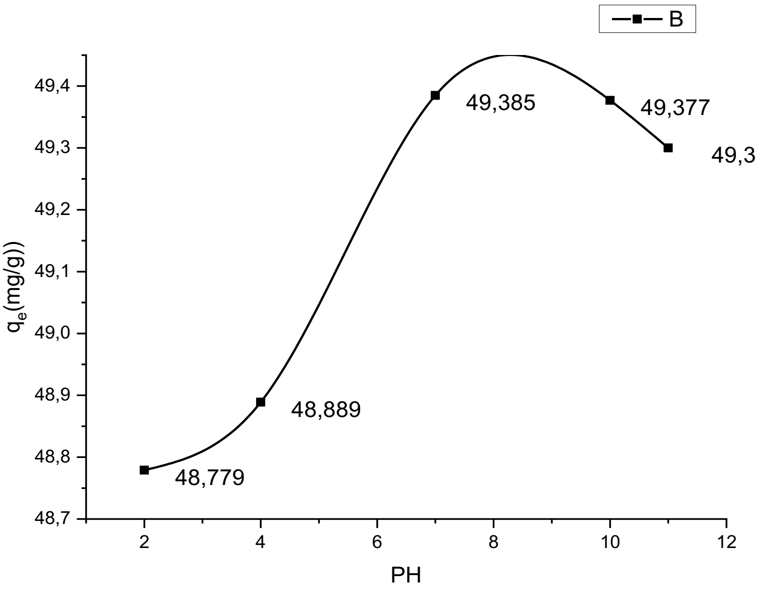

Figure 4. The Effect of pH on Adsorption Methyl Violet onto bentonite**.**

3.4. Effect of temperature on adsorption

The temperature has two major effects on the adsorption process. Increasing the temperature is known to increase the rate of diffusion of the adsorbate molecules across the external boundary layer and in the internal pores of the adsorbent particle, owing to the decrease in the viscosity of the solution. In addition, changing the temperature will change the equilibrium capacity of the adsorbent for a particular adsorbate. The results obtained are shown in Fig.5. It is seen that the amount of MV adsorbed on clay increases with temperature increase which is valid for an endothermic process The effect reported refers to the increase of the dye molecules mobility and that of the active sites number with temperature increase [31]. It is so because an increasing number of molecules can obtain the energy required for the interaction with the surface active sites. Furthermore, the temperature increase can bring about a swelling effect within the internal structure of the clay which in turn can enable the further penetration of the large dye molecules.

4. Adsorption kinetic study

In an attempt to present the kinetic equation representing adsorption of MV onto bentonite two kinds of kinetic models were used to test the experimental data. These are Lagergren-first order equation and second order equation. Lagergren-first-order equation is the most popular, kinetics equation (3). The form is :

$$\frac{dq}{dt} = k_1(q_e - q_t) \qquad (3)$$

After definite integration by applying the conditions :

$q_t =$ 0 at $t = 0$ and $q_t = q_t$ at $t = t$, Equation (4) becomes the following [32]:

$$ln(q_e - q_t) = lnq_e - k_t t \qquad (4)$$

where q_t (mg/g) is the amount is adsorption time t (min), K_1 the rate constant of the equation (min^{-1}) and q_e is the amount of adsorption equilibrium (mg/g). The adsorption rate constant $k1$, can be determined experimentally by plotting of ln (q_e –q_t) versus t (figure 6). The second –order equation is in the following form :

$$\frac{dq}{dt} = k_2(q_e - q_t)^2 \qquad (5)$$

After defined integration by applying the conditions $q_t = 0$ at $t = 0$ and $qt = qt$ at $t = t$, Equation (6) becomes the following [33].

$$\frac{t}{q_t} = \frac{1}{2kq_e^2} + \frac{1}{q_e}t \qquad (6)$$

The results show that the equilibrium adsorption capacity for dye increased with initial dye concentration. Furthermore, it can be seen from the coefficient of determination values (R^2=0.052) that the pseudo-first-order model was not suitable for describing the kinetic data.

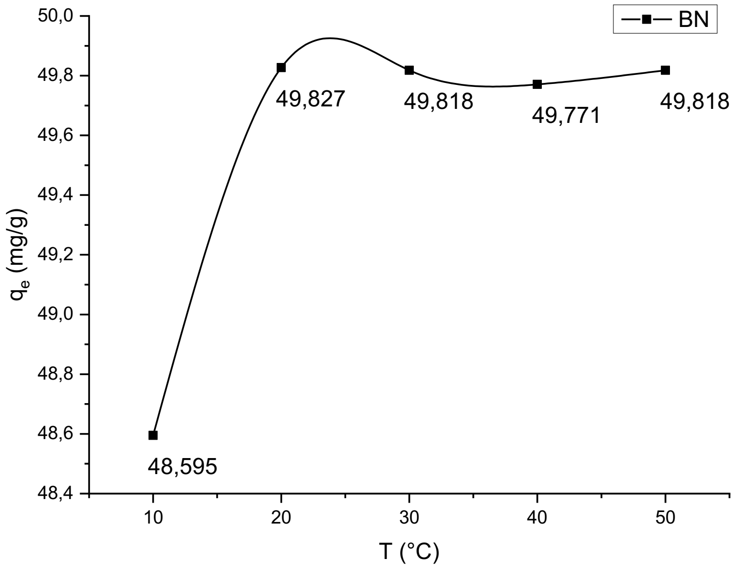

Figure 5. The Effect of Temperature on Adsorption Methyl Violet onto Bentonite.

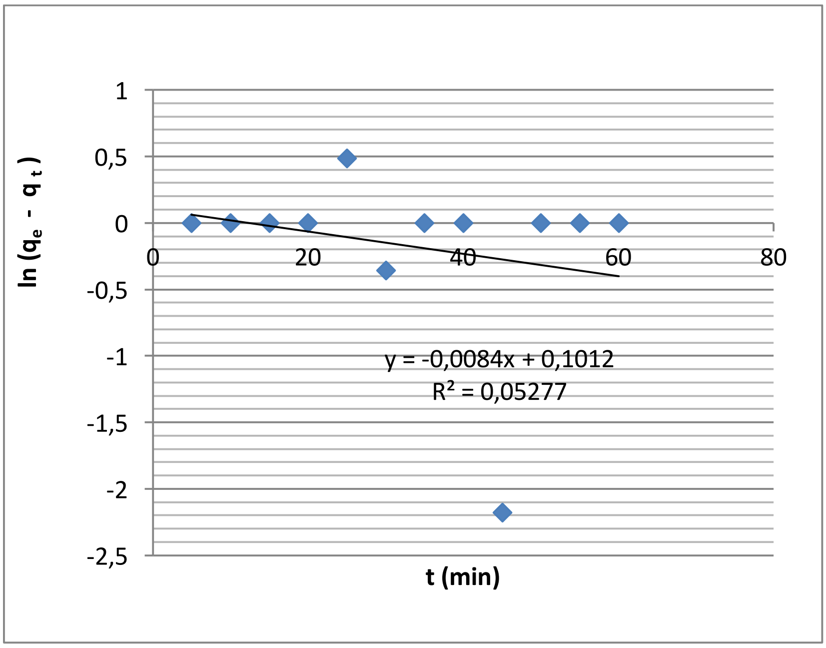

Figure 6. Pseudo -first-order of methyl violet Onto Bentonite.

The q_e value calculated from the pseudo-second-order model is in accordance with the experimental q_e value. The correlation coefficient R_2 of the linear plot is very high. The result is shown in Table 1. The value of kinetic constant and q_e indicates that the adsorption follows the pseudo-second order model (figure 7). Several studies found that the kinetics of adsorption of dyes on clay supports obey to the pseudo-second order [34,35].

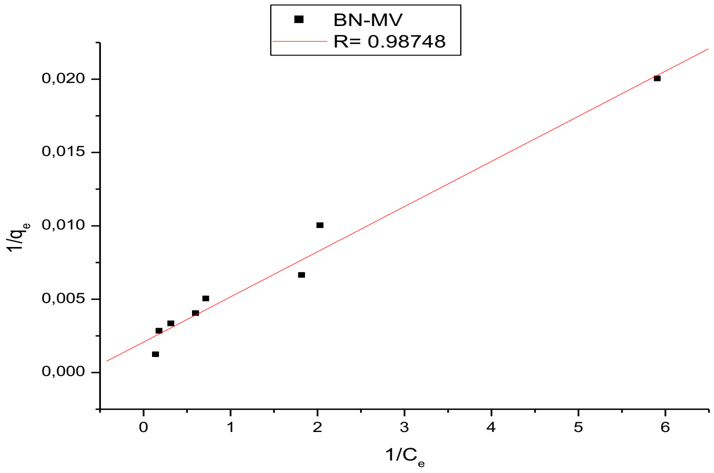

Figure 7. Pseudo-second-order of methyl violet onto Bentonite.

Table 1. Rate constants and coefficients of determination for the adsorption of methyl violet onto BN.

pseudo -first-order			pseudo-second-order		
K (min^{-1})	q_e (mg/g)	R^2	K_2 ($g.mg^{-1}min^{-1}$)	q_e (mg/g)	R^2
0.008	1.106	0.052	0.0292	48.57	0.999

5. Adsorption isotherm

Quantification of dye removal and the use of equilibrium models offer insight into the characterization of these solid surfaces especially on heterogeneity of the adsorbent surface. Two well-known and widely used models, the Freundlich and Langmuir isotherms, have been adopted to explicate dye-clay interaction. To account for the relevance of these two isotherm models, the values of Langmuir and Freundlich parameters correlation coefficients, R^2 at various temperatures together with their respective constants were obtained from the linear plots. The linearized Langmuir equation (7).

$$\frac{1}{q_e} = \frac{1}{q_{max}} + \frac{1}{q_{max}k_1} * \frac{1}{C_e} \quad (7)$$

The linearized Freundlich equation is given in equation (8).

$$lnq_e = Lnk_f + \frac{1}{nlnC_e} \quad (8)$$

The Freundlich constant, *n*, reflects both the average binding energy and the energetic heterogeneity of the sorbent binding sites [36]. The constant, K_F indicates adsorption capacity. As shown in Fig. 5, the plot of ln qe versus ln C_e is employed to determine the value of K_F and n from intercept and slope respectively. The values of q_{max}, k_L, k_f, and 1/n and the correlation coefficients for Langmuir (R^2) and for freundlich (R^2) models are given in table 2, and the linear fitting are plotted in figures 8 & 9. the correlation coefficient close to 0.98 for the Langmuir model indicate that the adsorption took place in the interlayer pores. Moreover, the values of 1/n for bentonite < 1, which indicates that adsorption processes are favorable and high adsorption took place.

The adsorption of the dye was characterized by the calculated Langmuir monolayer capacity value q_{max} = 480.79 mg/g, the value of the monolayer capacity q_{max} is indicated by a dashed line for comparison purposes. The frendlich isotherm was good enough for a model for the sorption study of dye solution. Therefore; the coefficient value for the determination of R^2 is 0.847for dye and was less than of the Langmuir isotherm.

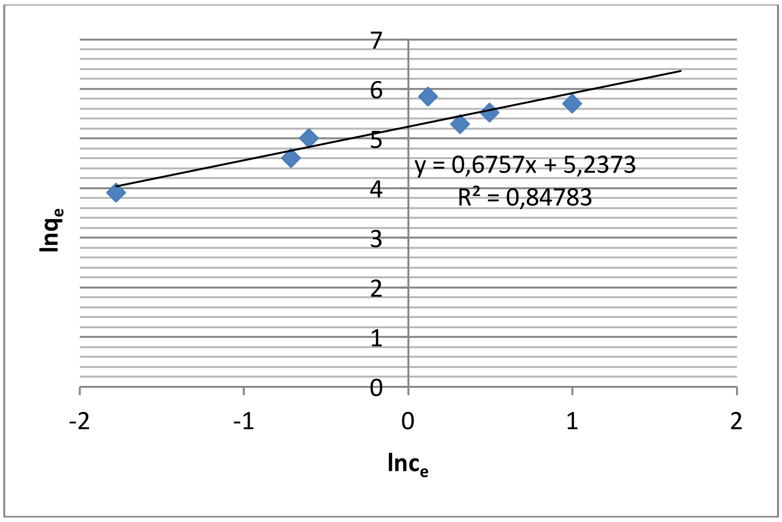

Figure 8. Freundlich isotherm for metyl violet adsorption on bentonite.

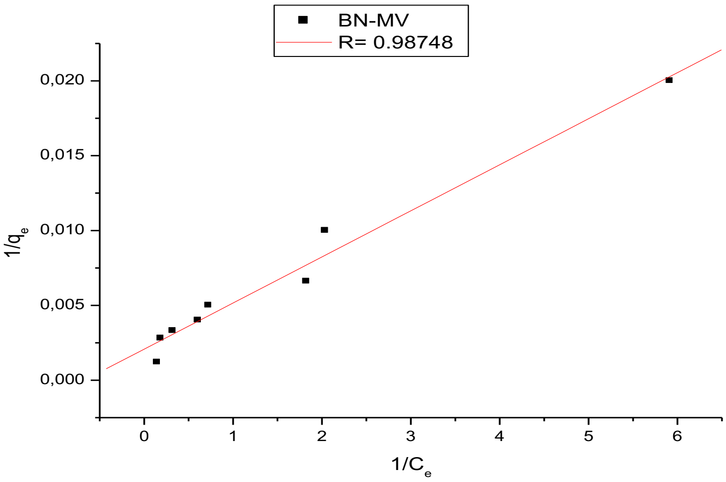

Figure 9. Langmuir isotherm for methyl violet adsorption on bentonite

Table 2. Equilibrium isotherm constants and coefficients of determination for the adsorption methyl violet onto BN.

Isotherm Langmuir				Isotherm de Freundlich		
q_{max}	K_L	R_L	R^2	1/n	K_F	R^2
480.79	0.675	0.0145	0.98	0.675	188.104	0.847

6. Thermodynamic Studies

Thermodynamic parameters including Gibbs free energy change (ΔG^o), enthalpy change (ΔH^o), and entropy change (ΔS^o) was calculated from the following (9):

$$\Delta G^{o} = -RT\ln k_{d} \tag{9}$$

$$\ln k_{d} = \frac{\Delta H^{o}}{RT} + \frac{\Delta S^{o}}{R} \tag{10}$$

Where R is the ideal gas constant (8.314 J.mol^{-1}.K^{-1}), T is the absolute temperature (Kelvin), and K_d is the distribution coefficient for adsorption.

Equation (8) known as the Van't Hoff equation, expresses a relationship between equilibrium constant and temperature. Plots of ln K_d versus 1/T should be a straight line as shown in Figure 10. The values of ΔH^{o} and ΔS^{o} were calculated from the slope and intercept of Van't Hoff plot, respectively. The thermodynamic parameters associated with the adsorption of MV onto bentonite are listed in Table 3.

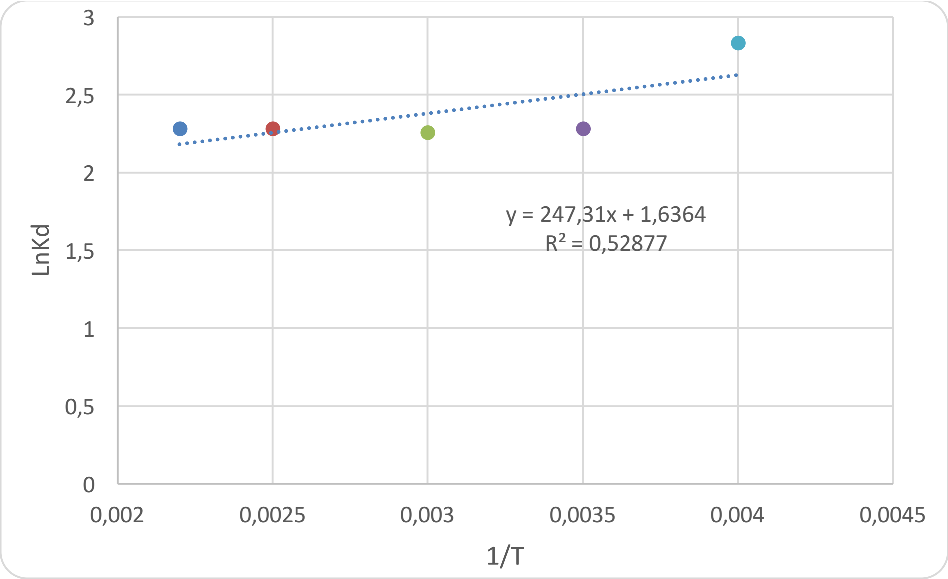

Figure 10. The plot of Vant-Hoff between Ln K_d VS 1/T on the adsorption of MV dye onto bentonite

Table 3. Values of thermodynamics parameters for MV adsorption onto Bentonite.

Temperature (K)	ΔG^{o} (kJ.mol^{-1})	ΔH^{o} (kJ.mol^{-1})	ΔS^{o} (kJ.mol^{-1}.K^{-1})
283	-6259.57	-6381.826	-0.432
293	-6255.25		
303	-6124.354		
313	-6120.034		
323	-6115.714		

According to the thermodynamic parameters represented in Table 3, we realized that the ΔH enthalpy of the system is negative so the adsorption process on bentonite is exothermic, the negative the value of ΔS^{o} shows the increased randomness at the solid-solution interface with some structural changes in the adsorbate and adsorbent and an affinity of the bentonite particles toward MV, the negative value of ΔG^{o} indicates that the adsorption is done through a spontaneous and favourable process [37].

Conclusion

The present study shows that the natural Algerian clay, abundant low-cost clay, can be used as sorbent for the removal of methyl violet. The results indicates that the adsorption follow the pseudo-second order model. Nearly 15 min of contact time are found to be sufficient for the adsorption to reach equilibrium. The results suggested that the adsorption capacity of organic compound colorant on raw clay adsorbent increased with increasing pH. By increasing the temperature, the q_e values continuously increased the negative value of ΔG^o indicates that the adsorption is done through a spontaneous and favourable process.

The Langmuir adsorption isotherm was found to have the best fit to the experimental data.

The results indicate bentonite is suitable as adsorbent material and it can be used as a cost-effective adsorbent for adsorption of MV from aqueous solutions.

According to experimental consequence. Parameters of thermodynamic as ΔH^o, ΔS^o and ΔG^o were computed, which demonstrated that the adsorption was exothermic nature and spontaneous.

Conflict of Interest

The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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