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The Investigation of Thermodynamics Parameters and Adsorption Kinetic of The Maxilon Blue 5G Dye on Turkey Green Clay

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ABSTRACT: Maxilon Blue 5G dye and Turkey green clay minerals used in adsorption experiments are of great importance recently due to their biological and physicochemical properties. Therefore, kinetic parameters of the adsorption of maxilon blue 5G dye on green clay were investigated. Studies were performed under adsorption conditions such as pH (5-11), maxilon blue 5G dye concentration ($0.5.10^{-6}$ - 2.10^{-5} M) and temperature (298-328 K). Turkey green clay and maxilon blue 5G dye were characterized by BET, SEM and XRF. The most favorable conditions for the dye adsorption of clay mineral, which is the support substance, were found to be pH 11 and the temperature was 328 K. The kinetic datas obtained from experimental studies were investigated on three different kinetic models such as pseudo-first-order, pseudo-second-order and intraparticle diffusion and it was determined that the adsorption event was carried out more compatible with the second order equation. Thermodynamic functions such as activation enegy (Ea), enthalpy (ΔH), Gibbs free mechanism (ΔG) and entropy (ΔS) were calculated. According to the results obtained from the experimental datas, the interaction between green clay and maxilon blue 5G are a physical interaction and experimental processes of adsorption are endothermic. These results indicated that green clay can be used as an adsorbent for the adsorption of the maxilon blue 5G (MB-5G).

Keywords: Maxilon blue 5G, thermodynamics, green clay, adsorption

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INTRODUCTION

There is a significant amount of synthetic dyestuff in the wastewater of industrial enterprises such as paint, leather, plastic and paper (Chiou et al., 2004). Due to the nature of the paints, it is difficult to remove from waste water. Because the organic acids of the dyes are extremely resistant to heat, light conditions and oxidizing agents (Crini, 2006). Such fluids include a number of contaminating agents, including dissolved solids, toxic, acid or caustic substances and color (Yener et al., 2006). In the case of both volumetric and waste incineration, water pollution caused by the textile industry is more than the whole industry (Singh et al., 2003). Due to the nature of the colors, a very small amount of wastewater is noticeable and undesirable. Therefore it is defined as the first pollutant (Banat et al., 1996). Many methods are used to remove dyestuffs from wastewater, such as chemical oxidation applications, adsorption process, membrane filtration, biodegradation and separation method (Choy et al., 1999). The presence and intensity of the functional groups on the support material surface and the support material in adsorption studies are extremely important for the adsorption capacity (Yenisoy et al., 2004). In order to remove basic and acidic character dyes, alternative materials such as activated carbon, unburned carbon (Choy et al., 1999; Basibuyuk and Forster, 2003; Yenisoy et al., 2004; Wang and Li, 2005), silica material and montmorillonite clay (McKay et al., 1980; Wang et al., 2004), and other adsorbents such as orange fruit shells (Namasivayam et al., 1996), fruit bundle of palm tree (Ho and Mckay, 1998), bark of teak wood, cotton residues, sugar cane powder (McKay et al., 1999; Khattri and Singh, 1999), diatomite (Çalımlı et al., 2018), krill clay (Nas et al., 2017), polymer composite Films (Şen et al., 2018), Graphene Oxide-Chitosan (Savk et al., 2018a), Novel Chitosan-Based Nanocomposites (Savk et al., 2018b), Turkish Green Clay (Demirbaş et al., 2016) were utilized in many studies.

The characteristic of support material used in the process is extremely important (Ali, 2012; Ali et al., 2012) for the efficiency of adsorption studies. Due to its high surface area and adsorption capacity, activated carbon is the most preferred support material for adsorption studies (Ali, 2010; Auta and Hameed, 2011; Ali et al., 2012). But activated carbon is a very expensive support material despite its excellent adsorption capacity (Auta and Hameed, 2012). Clay minerals are inexpensive, suitable for ecosystems, and have simple-to-use properties (Rehman et al., 2013). In adsorption processes, the graphene-carbon nanotube composite is used extensively to remove methylene blue from the wastewater (Wang et al., 2014). In this study, the clay which is considered as a support agent for dye removing is really green. In the literature studies, green clay is an organic composite mineral composed of decomposed plant based wastes as seaweed and iron compounds and the illite, known as the French green clay for centuries, took this name from the rock quarries on the southern slopes of France. It has been evaluated in different areas such as medicine cleaning and detox. Recently, it has been found that green clay mineral has a curative effect on patients with ulcers (Williams et al., 2008; Anonymous, 2016). Williams et al. (2008) have found that clay minerals can provide an inexpensive treatment for skin infections due to the antibacterial properties of clay minerals. The green clay used in this study was obtained from the mineral rock beds in the Gürpınar district of Van. For this reason, the name was considered as Turkish green clay. The aim of this study was to investigate the adsorption kinetics of maxilon blue 5G on the Turkish green clay (TGC) under a series of physicochemical conditions. For adsorption characterization, it was detected with SEM (Scanning Electron Microscope) and BET (Brunauer, Emmet ve Teller) devices. In this

study, important parameters such as contact time, pH and temperature were investigated during the experiment. In addition, thermodynamic functions such as entropy, enthalpy and Gibbs free energy were examined.

MATERIALS AND METHODS

Materials

Clay mineral used in this study was obtained from the Gürpınar districts of Van and the characterization of the samples was evaluated. To determine the surface area and pore volume of TGC clay mineral, BET (micromeritics FlowSorb ll-2300) apparatus was used. The surface morphology of the samples used for the adsorption was determined with the SEM apparatus. MB-5G was obtained from Setaş Textile Co. (Bursa, Turkey). The chemical used in the study was analytical grade and the water used was evaluated by precision distillation in millimetric quality. The chemical composition and specific surface area of the Turkey green clay are shown in Table 1 and Table 2.

Table 1. Chemical composition of Turkey Green Clay

Constituent	Percentage present (%)
Mg	20,74
Al	9,20
Si	44,79
Fe	12,48
Ca	10,02
Others	2,77

Table 2. Some properties of Green Clay

Constituent	Values
Particle size (mesh)	325
pH	9.23
Specific surface areas m²/g	
Single point specific surface area	1.484e+01 m²/g
Multipoint specific surface areas	1.547e+01 m²/g

Experimental procedure

Adsorption studies were performed using mechanical mixer in the balloon glass. All colorant solution was prepared with ultra-pure water. The kinetic assays were carried out at a constant agitation rate of 600 rpm, at a concentration of 1.10^{-5} M dyestuff, under conditions of pH 7 and 298 K. As a result of preliminary studies, the dye concentration was 5.10^{-6} mol / L, 1.10^{-5} M and 2.10^{-5} M, and the effect of pH on the Turkey green clay of the dyestuff was analyzed from pH 5 to 11. The pH of the dyestuff solutions was adjusted by pH electrode from 0.05 N NaOH and 0.05 N HCl solutions. The effect of temperature on the adsorption studies was carried out at, 298 K, 308 K, 318 K and 328 K. 5 milliliters of sample were taken at a certain time interval and the samples were centrifuged at 3500 rpm for 6 minutes, and the UV-Vis spectrophotometer (Cary 1E UV-Vis spectrophotometer, Varian) was used to monitor the maximum absorbance wavelength changes of the obtained supernatant. The adsorbed amount of dyestuff at any time t, q_t, was calculated from the mass balance equation 1.

$$q_t = (Co-Ct)V/m \quad (1)$$

In the equation given herein, Co and Ct expressions indicate the concentration value at first and at any time. V and m denote the volume of the dye used and the mass of the green clay mineral (Alkan et al., 2008).

RESULTS AND DISCUSSION

The effect of contact and equilibrium times and dye concentration factor

Adsorption effect on TGC of maxilon blue 5G was studied at constant conditions such as a stirring time of 600 rpm, a specific contact time and pH 7. From Table 3, it was found that the amount of adsorbed maxilon blue 5G increased from 0.195 to 0.752 mmol g^{-1} for an increase of 0.5 x 10^{-5} M to 2.0 x 10^{-5} M at the initial maxilon blue 5G concentration. The time required to reach this concentration is about 60 minutes.

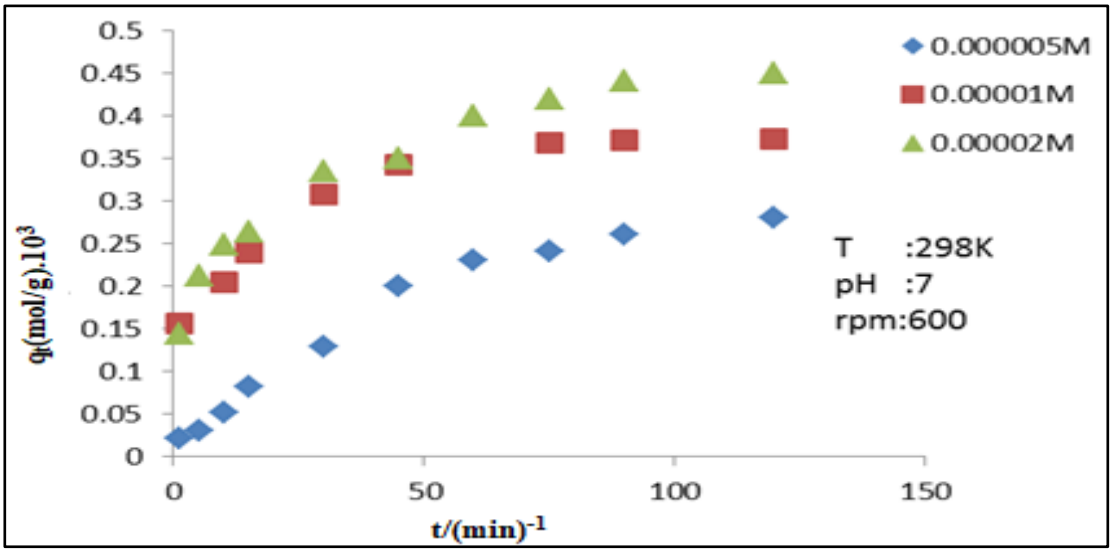

Figure.1. The effect of dyestuff concentration

Effect of solution pH and temperature

The adsorbent surfaces used in the adsorption studies have generally a negative and positive charge. The ions on these surfaces are affected because they are very sensitive to pH changes. In some cases, the net surface charge of the support material is zero. The pH value of the adsorbent considered for the support material is shown as pHpzc (the point of zero charge) where the net surface charge is zero (neutral) (Wang et al., 2014). Figure 2a shows the effect of changing the pH of 5 to 11 in MB-5G aqueous solutions under constant conditions such as temperature, initial dye concentration and clay amount. When Figure 2 is examined, the adsorbent (g) adsorbed per unit mass (mmol) is increased when the pH is increased from 5 to 11. This can be explained by the electrostatic attraction between the positively charged MB-5G and the negatively charged clay surface. One of the most important factors in adsorption studies is the temperature factor. In this study, adsorption kinetics of MB-5G on TGC clay mineral at 298, 308, 318 and 328 K temperature were investigated under initial MB-5G concentration conditions at 2.0x10^{-5} M at pH 7. The results showed that the increase in temperature showed that MB-5G increased the amount of adsorption on the clay mineral (Alkan et al., 2008; Nandi et al., 2009; Ghaedi et al., 2011) and the maximum adsorption capacity of MB-5G adsorbed onto the surface of TGC was determined at 328 K. In the results, we can say that the adsorption process is endothermic.

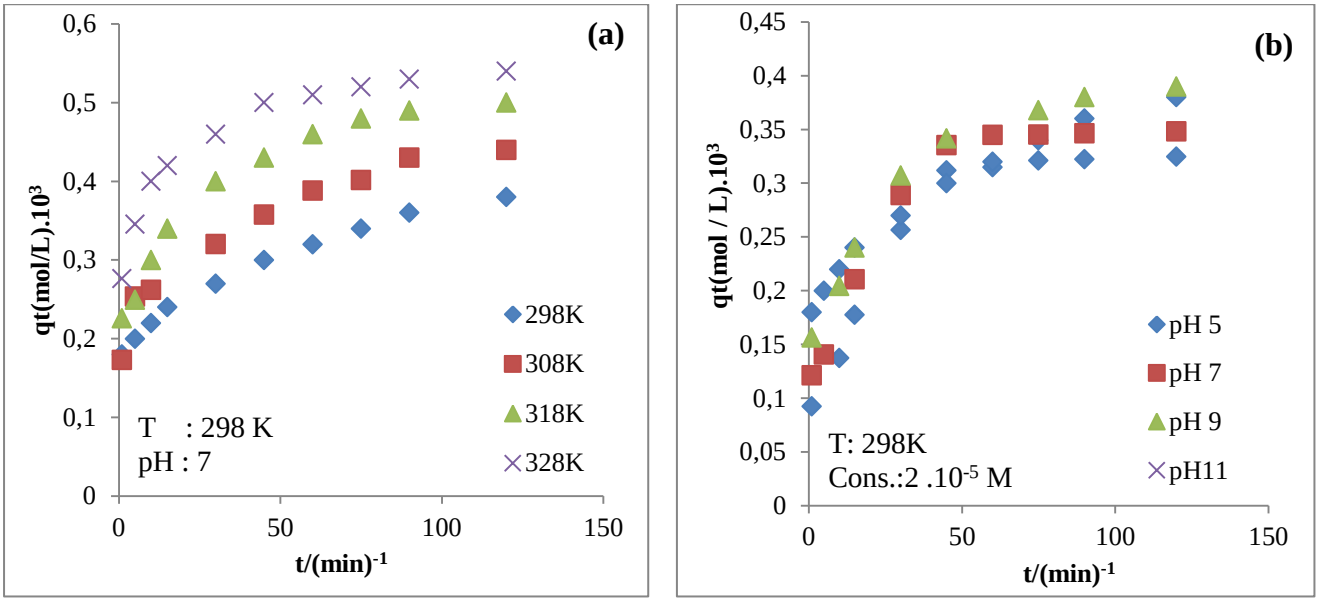

Figure 2. The effect of solution of pH(a) and temperature(b) to the adsorption rate of MB-5G on TGC

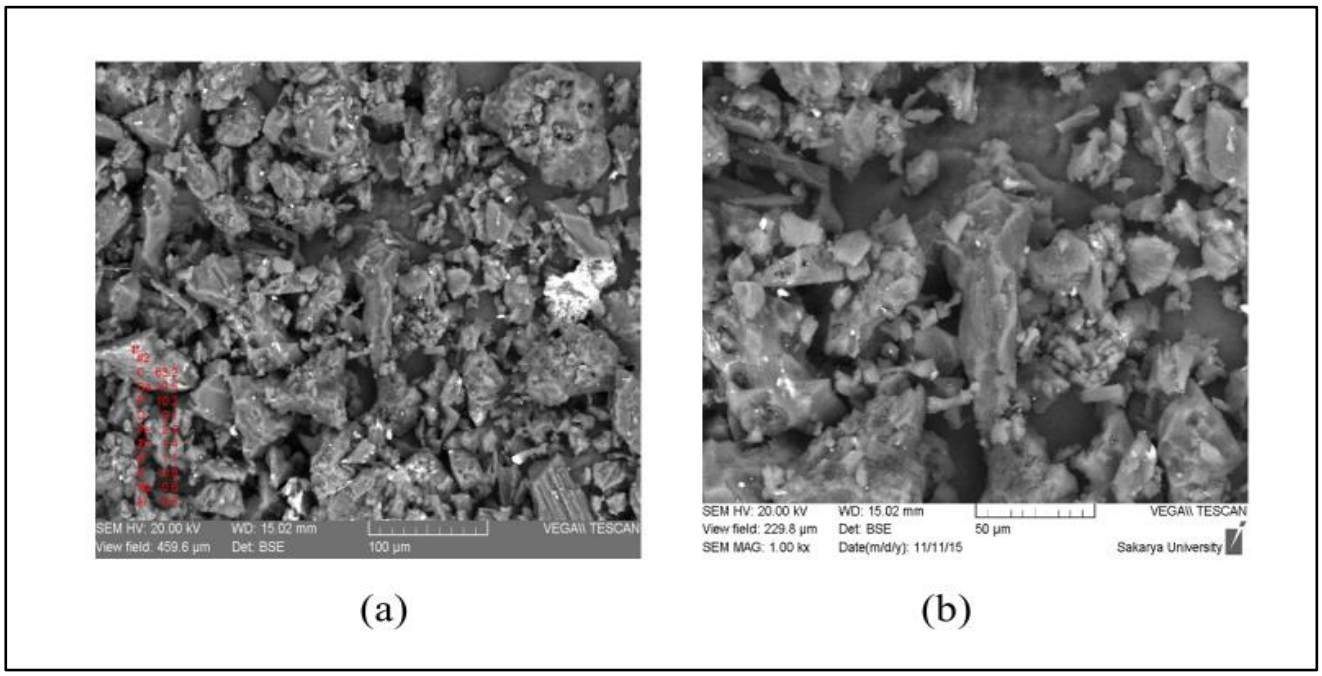

Figure 3. SEM microphotographs of (a) TGC, (b) MB-5G adsorbed by TGC after 120 min

Investigation of figures SEM

SEM images of Turkey green clay material (TGC) and the composite (TGC/MB-5G) consisted of pure maxilon blue 5G - green clay are given in Figure 3. Figure 3a shows SEM image of TGC. Some pores which have been formed because of air are seen on the surface. Figure 3b shows surface of composite (TGC/MB-5G). Bright and dark dots are seen on the surface of composite (TGC/MB-5G). These dots show both MB-5G and green clay. The surface of the adsorption agent appears brighter and smoother. This indicates that the material considered as support is coated with MB-5G.

Table 3. Kinetic datas calculated for adsorption

Parameters					Pseudo-second-order					
T/K	**Con. mol. L^{-1} 10^{-5}**	**pH**	**Stir. Speed rpm**	**Pseudo order R^2**	**q_e(cal.) first-g^{-1}**	**q_e(exp.) mg. g^{-1}**	**k_2(g) mg. min^{-1}**	**R^2**	**h(mol) min. g^{-1}**	**t**
298	0.2	7	600	0.92	0.39	0.40	0.362	0.99	0.140	6.06
308	0.2	7	600	0.9	0.42	0.44	0.420	0.99	0.160	5.03
318	0.2	7	600	0.94	0.49	0.50	0.491	0.99	0.195	4.12
328	0.2	7	600	0.98	0.50	0.49	0.637	0.99	0.383	2.41
298	0.05	7	600	0.94	0.19	0.20	0.322	0.98	0.068	9.7
298	0.1	7	600	0.96	0.39	0.40	0.362	0.99	0.140	6.06
298	0.2	7	600	0.85	0.54	0.57	0.106	0.96	0.066	12.5
298	0.2	5	600	0.97	0.36	0.38	0.423	0.98	0.156	5.46
298	0.2	7	600	0.93	0.38	0.39	0.916	0.99	0.335	3.14
298	0.2	9	600	0.98	0.39	0.40	0.362	0.99	0.140	6.06
298	0.2	11	600	0.97	0.39	0.41	0.368	0.99	0.144	6.13

Adsorption study kinetics

In order to understand the control mechanism in sorption studies. It is stated with experimental model which model is carried out with experimental datas. The pseudo first-order equation is generally expressed as follows (Lagergren, 1898; Weber et al., 1963; Hunter, 1999).

$$ln(q_e\text{-}q_t) = lnq_e\text{-}k_{it} \quad (2)$$

herein, the pseudo-first-order velocity constant is k_1. The datas are indicated in Table 3.

Pseudo - second - order equation

If the adsorption process is carried out by a second order mechanism, The so-called-second equation 3 is indicated below (Ho and McKay, 1998)

$$\frac{t}{qe} = \frac{1}{k_2 q_e^2} + \frac{1}{q_e} t \quad (3)$$

The initial adsorption rate (h) is expressed by equation 4.

$$h = k_2 q_e \quad (4)$$

herein, when we look k_2 and k_1 values in Table 3, it is understood that the adsorption mechanism is carried out by second order kinetic equations.

The initial ratio of intra-particle diffusion was obtained by the following equation 5 (Doğan et al., 2006).

$$q_t = k_{\text{int.}} t_{1/2} + C) \quad (5)$$

Herein, k_{int} is the intraparticle diffusion rate constant (mg(g $min^{-1/2}$)$^{-1}$ and is given in Table 4.

Thermodynamic parameters

In order to calculate the activation energy of the adsorbed MB-5G dyestuff on the support material in the study, arrhenius equation 6 was used.

$$lnk_2 = lnA - \frac{Ea}{Rg.T} \quad (6)$$

In the aforementioned equation, Ea, k_2 and A expressions respectively indicate the activation energy (J / mol), the rate constant (g / mol s) of the sorption and the Arrhenius factor. In order to calculate the Ea value, the Ea value of lnk versus 1 / T was found to be 11.8. From the activation energy, we can say that the working process is carried out under the conditions of physical adsorption (Laidler and Meiser, 1999). In the Eyring equation 7, the most important function values of thermodynamics such as entropy (ΔS), enthalpy (ΔH) and free Gibbs energy (ΔG) were obtained (Singh, 2000).

Table 4. Intra - particle diffusion data from adsorption studies

Mass transfer				Intra – particle diffusion			
T/K	**Conc. mol. L^{-1}. 10^{-5}**	**pH**	**Stirring Speed (rpm)**	**kint.$_1$ mg.g^{-1} min$^{-1/2}$**	**R_1^2**	**kint.$_2$ mg.g^{-1} min$^{-1/2}$**	**R_2^2**
298	0.2	7	600	0.003	0.95	0.012	0.98
308	0.2	7	600	0.023	0.98	0.093	0.94
318	0.2	7	600	0.044	0.97	0.0016	0.97
328	0.2	7	600	0.064	0.98	0.0056	0.78
298	0.05	7	600	0.024	0.96	0.004	0.92
298	0.1	7	600	0.003	0.95	0.012	0.98
298	0.2	7	600	0.041	0.98	0.0045	0.69
298	0.2	5	600	0.033	0.94	0.0011	0.99
298	0.2	9	600	0.035	0.98	0.0016	0.96
298	0.2	7	600	0.003	0.95	0.0012	0.98
298	0.2	11	600	0.0024	0.97	0.0027	0.95

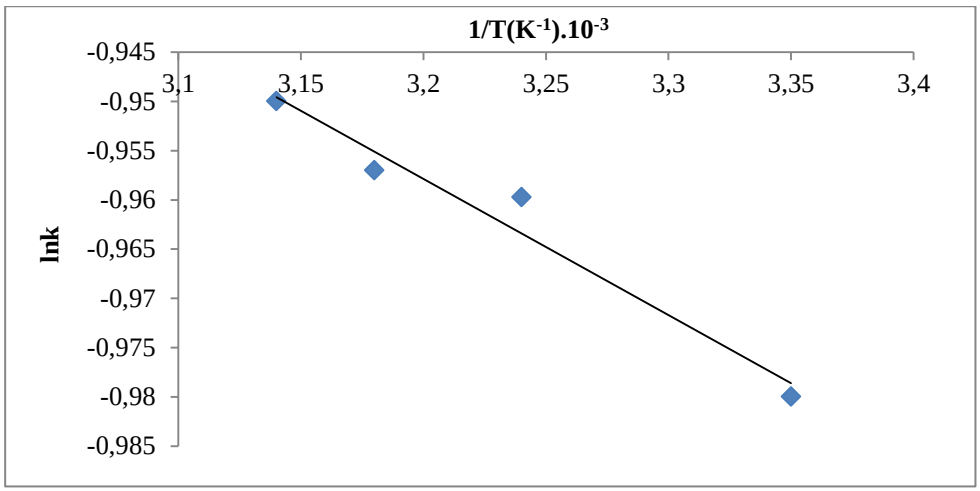

Figure 4. Arrhenius graph for adsorption of MB-5G in TGC.

$$ln(k_2/T) = ln\,(kb/h) + \frac{\Delta S}{Rg} - \frac{\Delta H}{RgT} \quad \text{..... (7)}$$

In the above equation, the expressions kb and h denote Boltzmann ($1.3807.10^{-23}$ J/K and Planck ($6.6261.10^{-34}$ J s) constants. The value of Standard enthalpy change was also found to be 15.2 k.J/mol. This indicates that the event is endothermic. The small amount of enthalpy change indicates a loose bond between the adsorbate molecules and the support surface (Mall and Upadhyay, 1995).

Table 5. Thermodynamic function datas obtained by adsorption of TGC surface with MB-5G

Parameter				
T/K	**ΔG (kJ/mol)**	**Ea (kJ/mol)**	**ΔH (kJ/mol)**	**ΔS (j/K.mol)**
298	64.96			
308	66.63	11.8	15.2	-167.3
318	68.3			
328	69.97			

CONCLUSION

Herein, we showed that Turkey green clay (TGC) can be used as a good support material for the adsortion of MB-5G. MB-5G concentration, temperature, pH and contact time were taken as kinetic parameters in the adsorption process. The amount of adsorption increased with the increase pH and temperature values. The experiments results showed the adsorption process are compatiple with the second order kinetic model. The positive value of Gibbs energy showed the adsorption process are not spontaneous, and the adsortion process are endothermic due to the small and positive value of enthalpy. The small enthalpy change indicates forming of a loose bond between the a MB-5G molecules and the support surface. The selected Turkey Green Clay as a carrier for the adsortion process has a high adsortion potentional because of an effective adsortion effect in aqueous medium.

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