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Removal of anionic dye from aqueous solution by magnesium silicate gel

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ABSTRACT

Mesoporous magnesium silicate gel was successfully synthesized and characterized by N_2 adsorption/desorption technique and FT-IR. The adsorption experiments of anionic dye (weak acid red 2R) onto prepared magnesium silicate gel were studied by varying mass of adsorbent, dye concentration, pH, temperature, and contact time. The results showed magnesium silicate gel had high surface area, $277.37\,\mathrm{m^2\,g^{-1}}$. The adsorption capacity increases with increasing initial dye concentration, adsorption temperature, and contact time. The experimental data were applied to three adsorption kinetic models and the results indicated that the adsorption behavior was described very well by the second-order kinetics model with the high correlation coefficients ($R^2 > 0.99$). Intra-particle diffusion was performed in three different stages. The activated energy obtained was $127.6\,\mathrm{kJ\,mol^{-1}}$ and the enthalpy, entropy, and standard free energy were calculated and given that ΔH^o was $80.1\,\mathrm{kJ\,mol^{-1}}$, ΔS^o was $300.1\,\mathrm{J\,mol^{-1}}$, and ΔG^o were negative values at the different temperatures. The adsorption process is rapid and physisorption in nature. Therefore, magnesium silicate gel would be used in the industrial wastewater treatment as a potential adsorbent.

Keywords: Magnesium silicate gel; Mesoporous adsorbent; Adsorption; Anionic dye; Kinetics study

1. Introduction

Dyes are extensively applied in dyeing processes such as textile, food, clothes, and paper [1,2]. Over 7×10^5 tons of synthetic dyes are produced annually worldwide [3]. It is said that 10–15% dyes are wasted in the dyeing processes. The waste dyes cause the serious pollution problem, for example, decreasing water transparency, ruining fish life, even damaging the biology balance. Recently, considerable attention has been paid to dyes wastewater by researchers and environmentalists. Many treatments of the removal of dyes from effluents have been reported such as chemical coagulation [4,5], photodegradation [6,7], biodegradation [8], and physical adsorption [9,10]. Among these methods, adsorption technique is considered as a great potential based on various adsorbents with some advantaged properties like high capacity, environmental friendliness, ease of use and without second-pollution. Activated carbon [11,12] with high surface area and porosity can be widely used in the removal of organic contaminant, metal ions, and dyes, but the high cost

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confined its usage to some extent in practical application. So, other alternative adsorbents with low cost and high efficiency have been investigated.

The surface of the natural silicate minerals (sepiolite, clays, zeolite, montmorillonite, etc.) has a plenty of silanol group (≡Si−OH) and magnesium hydroxide (Mg–OH) activated groups [13], which indicates that there are many activated sites on the surface of silicate minerals. Components of the MgO–SiO_2 system are easily obtained through the reaction with each other, yielding magnesium silicate. Many magnesium silicates have been synthesized and applied in eliminating dyes and heavy metal ions from wastewater solution decreasing acid value in frying oil and biodiesel and removing dyes, K^+ in preparation of polyether [14–19].

Anionic dyes are mostly used in wool, nylon, and silk fields, and the removal of the anionic dyes from the wastewater is also important for these industries. But many investigations have been devoted to remove the cationic dyes from the solution onto natural silica adsorbents [20–22] and as-prepared magnesium silicate adsorbents [17,23,24], however, a few papers were involved the removal of anionic dyes using sepiolite [25–29].

In this paper, this work aims to obtain a low-cost solid adsorbent, magnesium silicate gel, and first used it to investigate the adsorption properties of anionic dye, weak acid red 2R (WAR-2R), from aqueous solution. Many experiments were carried out to evaluate the adsorption effect of WAR-2R dye onto magnesium silicate gel from aqueous solution as the function of mass of adsorbent, initial dye concentration, solution pH, temperature, and contact time. The experimental data were analyzed using three adsorption kinetics models: first-order kinetic equation, second-order kinetic equation, and intra-particle diffusion. The thermodynamic parameters were calculated under different temperatures. The general approach would be applied in the industrial wastewater treatment.

2. Marerials and methods

2.1. Materials

All chemical reagents were of analytical grade purity and used as received without further purification.

2.2. Synthesis of mesoporous magnesium silicate gel

Magnesium chloride hexahydrate and sodium silicate nonahydrate (Na_2O/SiO_2 module = 1) were mixed with Mg/Si molar ratio as 3:1 at room temperature. The white precipitate occurred immediately. Keep the precipitate stirring for 5 h, then it was allowed to stand for overnight. The precipitate was filtered without further washing. The resulting precipitate (as-magnesium silicate gel) was dried in an air oven at 110℃ for 10 h and then allowed to cool naturally.

2.3. Characterization

The surface area and pore-size distribution of as-magnesium silicate gel were determined by N_2 adsorption/desorption analysis. The surface area was evaluated by Brunauer-Emmett-Teller (BET) and the pore-size distribution was determined by Barrett-Joyner-Halenda methods. The functional groups on the surface of adsorbent were analyzed by the FT-IR with the wavelength range 400–4,000 cm^{-1}.

2.4. Adsorption test

In the dye adsorption process, WAR-2R was chosen as a dye model. WAR-2R was dried at 110℃ for 2 h before using. All WAR-2R solution was prepared with distilled water. Adsorption studies were preformed under different conditions of mass of adsorbent, initial dye concentration, solution pH, temperature, and contact time with regular mass of magnesium silicate gel and some concentration of dye aqueous solution 50 mL in a series of flasks.

The effect of pH was investigated through adjusting the pH of dye solution by the addition of 0.1 mol L^{-1} NaOH or HCl solution, and pH data was observed by using a pHS-3C meter equipped with a combined pH electrode. The pH-meter was standardized with NBS before measurement. The effect of temperature was examined at four different solution temperatures (i.e. 293, 298, 303, and 308 K) and contact time. The thermostatic shake bath was used to keep the temperature constant. The adsorbent was dispersed homogeneously in the WAR-2R solution by shaking for a specified time. Then the solid and liquid was separated by filter. The concentration in the residual solution was measured by using a UV–Vis spectrophotometer at $\lambda = 504$ nm wavelength, which is corresponding to the WAR-2R maximum adsorption wavelength. The concentrations in solution before and after adsorption were calculated. The adsorbed amount of WAR-2R, q_t, was determined at time t and calculated from the following mass balance equation:

$$q_t = \frac{(C_0 - C_t)V}{m} \tag{1}$$

where q_t is the adsorption capacity, mg g^{-1}; C_0 is the WAR-2R initial concentration, mg L^{-1}; C_t is the concentration of the solution at time t, mg L^{-1}; V is the volume of the dye solution, L; m is the mass of adsorbent, g.

3. Results and discussion

3.1. Characterization of adsorbent

3.1.1. Surface area and pore-size distribution analysis

N_2 adsorption/desorption isotherms and pore-size distribution are determined. The isotherms can be categorized as type IV with a hysteresis loop in the relative pressure range 0–1.0. The hysteresis loop indicates magnesium silicate gel possessed a mesoporous structure. The adsorption quantity of nitrogen gradually rose when the relative pressure was under 0.8. Then, the amount of nitrogen adsorbed sharply, increasing with the relative pressure growing from 0.8 to 1.0 and reached the maximum value as 250 cm^3 g^{-1} (at $p/p_0 = 1$). The special surface area calculated from the BET was 277.37 m^2 g^{-1} and the single point adsorption total pore volume was 0.243 cm^3 g^{-1}. The pore-size distribution obtained from adsorption isotherm pointed a narrow distribution centered at 2–4 nm, which agreed with the average pore width as 3.50 nm (given by the instrument). Therefore, the N_2 adsorption/desorption isotherm established the possibility that synthetic magnesium silicate gel can be used as a considerable adsorbent in practical application [30].

3.1.2. FT-IR spectra

The FT-IR spectrums of magnesium silicate gel, with the adsorption before and after, are analyzed. Before adsorption, the broad band located in 3463 cm^{-1} is a strong hydrogen bonded (O–H) stretching adsorption and a sharp peak near 1,650 cm^{-1} is attributed to hydroxyl bending of zeolitic water [31]. The strong peak at 1,040 cm^{-1} with shoulder at approximately 1,006 cm^{-1} is ascribed to Si–O stretch vibration. A weak peak at 671 cm^{-1} with shoulder at about 649 cm^{-1} explains the Si–O bending vibration, while Mg–O vibration occurred at 464 cm^{-1} [32]. After adsorption, some new peaks appeared at 1,300–1,500 cm^{-1} and some original peaks became weak which indicated the function force existed between the surface of magnesium silicate gel and WAR2R.

3.2. Adsorption studies

3.2.1. Effect of mass of adsorbent

To determine the relationship of mass of adsorbent and adsorption capacity, different amount of magnesium silicate gel was chosen to investigate the effect in adsorption process. The experimental result is given in Fig. 1, which showed the plot of adsorption capacity with mass of adsorbent in 50 mL dye solution. The adsorption capacity reached to the maximum value as 240 mg g^{-1} when mass of magnesium silicate gel was 0.025 g, then decreased sharply to 100 mg g^{-1} with mass of adsorbent increasing to 0.1 g. Furthermore, the adsorption capacity diminished gradually to 50 mg g^{-1} when mass of magnesium silicate gel rose to 0.2 g. Therefore, the changing of adsorption capacity of WAR-2R onto the different mass of magnesium silicate gel can be interpreted that mass of adsorbent has an important influence on the adsorption of WAR-2R. 0.1 g, as mass of adsorbent, was determined in the following experiments.

3.2.2. Effect of initial concentration

The concentration of adsorbates is a dominating driving force in the adsorption process. Therefore, we studied the effect of initial dye concentration on the adsorption capacity of WAR-2R onto magnesium silicate gel. Experiments were performed as the function of contact time and the plots of adsorption capacity of WAR-2R onto magnesium silicate gel were shown in Fig. 2. As seen, the adsorption equilibrium was attained in 6 h and the adsorption capacity of WAR-2R onto magnesium silicate gel increased with increasing WAR-2R concentration. The adsorption equilibrium amount increased from 45 to 145 mg g^{-1}, when the dye concentration increased from 100 to 300 mg L^{-1}. The performance agreed with the conclusion: initial concentration provides a significant driving force to overcome the mass transfer resistance of the dye between the solutions and adsorbents surface, therefore, the high concentration was favorable

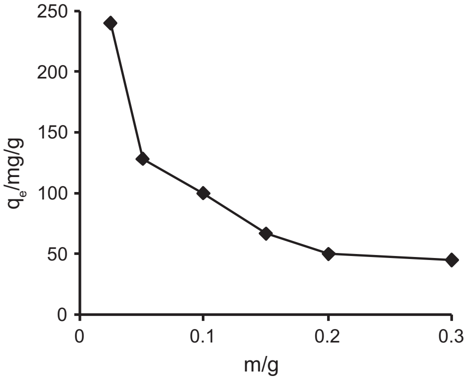

Fig. 1. The effect of mass of magnesium silicate gel on the removal of WAR-2R.

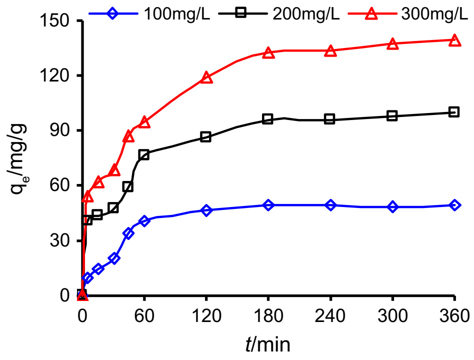

Fig. 2. The effect of dye concentration on the removal of WAR-2R as the function of time.

for the adsorption process [33]. Moreover, the adsorption capacity of dye also increased with the increase in contact time.

3.2.3. Effect of pH

The pH variation can influence the surface charge of the adsorbent. In other words, the concentration of H^+ and OH^- in solution can affect the adsorption of adsorbates onto adsorbents. Consequently, it is necessary to study the effect of pH in the adsorption process. The WAR-2R solution pH was adjusted to the desired value (pH 3–10) using either 0.1 mol L^{-1} HCl or NaOH solution. The plot of the adsorption capacity of WAR-2R onto magnesium silicate gel with solution pH was shown in Fig. 3. The adsorption capacity gradually decreased, then increased smoothly as the pH increases from 3 to 10. The slightly constant adsorption capacity was obtained under acidic and basic conditions. Furthermore, the experimental data also shown that the total adsorption capacity was changed by 1.5 mg g^{-1}. Maybe it can be explained that the adsorption process relied on the solution external diffusion and pH has little influence on the adsorption of WAR-2R onto magnesium silicate gel. Therefore, in the series experiments, pH was kept without adding HCl and NaOH.

3.2.4. Effect of ionic strength

The plot of adsorption capacity of WAR-2R onto magnesium silicate gel vs. the different NaCl concentration was given in Fig. 4. It can be seen that the adsorption capacity of WAR-2R decreased gradually from 98 to 68 mg g^{-1}, as NaCl concentration increases from 0 to 0.4 mol L^{-1}. Maybe it was attributed that NaCl prevented the dye diffusion to surface of magnesium silicate gel and the electrostatic interaction between surface and dye molecule became difficult and weak [22].

3.2.5. Effect of temperature

The plots of the adsorption capacity of WAR-2R onto magnesium silicate gel vs. different solution temperature and contact time were shown in Fig. 5. The experimental results demonstrated the removal of WAR-2R onto magnesium silicate gel was fast in the early 30 min and the adsorption capacity achieved much more than 90 mg g^{-1}. Then, the amount of WAR-2R adsorbed increased smoothly and the adsorption attained equilibrium in 120 min. Moreover, the experimental data also indicated that the adsorption capacity of WAR-2R onto magnesium silicate gel had increased slightly with the increase in adsorption temperature, while the finial equilibrium adsorption capacity was nearly equal.

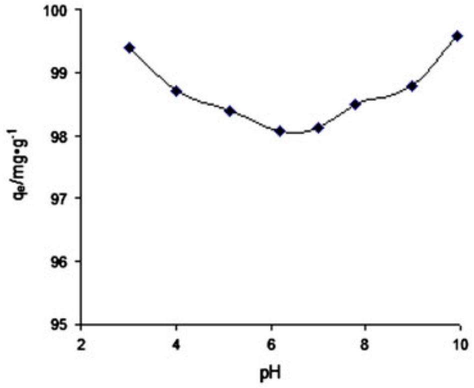

Fig. 3. The effect of pH on the removal of WAR-2R.

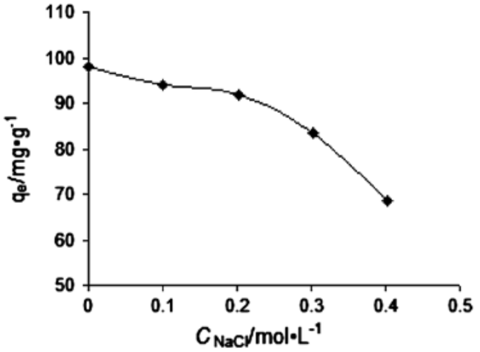

Fig. 4. The effect of NaCl concentration on the removal of WAR-2R.

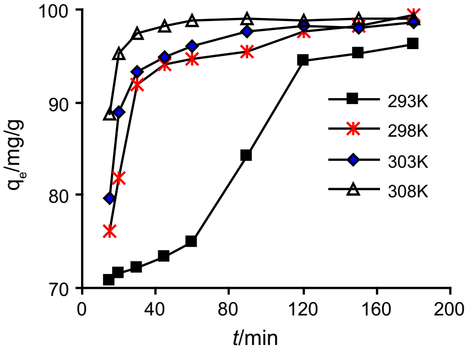

Fig. 5. The effect of temperature on the removal of WAR-2R as the function of time.

3.3. Adsorption kinetics

Adsorption rate was an important parameter to determine the efficiency of adsorbents in practical application. Understanding the adsorption mechanism of dyes on magnesium silicate gel surface is essential for an effective removal of dyes from aqueous solution. Therefore, adsorption mechanism can be preliminarily conducted from the kinetic models and kinetics adsorption parameters.

Three kinetics models were used to evaluate the experimental data and adsorption process.

The first-order kinetics equation [34] is presented as (2)

$$\ln(q_e - q_t) = \ln q_e - k_1 t \tag{2}$$

where q_e is the amount of WAR-2R adsorbed at equilibrium, mg g^{-1}, q_t is the amount of WAR-2R adsorbed at time t, min; k_1 is the first-order rate constant of adsorption, min^{-1}, which can be determined from the slopes of the plots $\ln(q_e - q_t)$ vs. t.

The second-order kinetics equation [35] is given as: (3)

$$\frac{t}{q_t} = 1/(k_2 q_e^2) + t/q_e \tag{3}$$

where k_2 is the second-order rate constant of adsorption, g mg^{-1} min^{-1} and can be obtained from the slopes and intercepts of plots t/q_t vs. t.

Intra-particle diffusion equation [36] can be written as: (4)

$$q_t = k_p t^{0.5} + C \tag{4}$$

where k_p is the intra-particle diffusion rate constant of adsorption, g·mg$^{-0.5}$·min^{-1} and can be get from the slopes of the plots q_t vs. $t^{0.5}$. C is intercept which indicates boundary layer thickness.

The experimental data were applied to the first-order kinetics model, the second-order kinetics model and intra-particle diffusion, respectively, and the corresponding kinetics parameters for adsorption of WAR-2R were calculated from Eqs. (2)–(4).

The experimental data deviated the straight lines and the correlation coefficient (R^2) values were very low, indicating the adsorption process did not follow the first-order kinetics model (Fig. 6(A)), The corresponding parameters were given in Table 1.

The correlation coefficient values (R^2) for the second-order kinetics model are higher than 0.99 with the plots t/q_t vs. time t fitting straight lines and the calculated $q_{e,cal}$ value agreed with the experimental $q_{e,exp}$ value well. Therefore, the adsorption process of WAR-2R onto magnesium silicate gel can be described by the second-order kinetics model very well (Fig. 6 (B)). Rate constant k_2 increased from 7.4×10^{-4} to 110×10^{-4} g mol^{-1} min^{-1} with increasing the adsorption temperature from 293 to 308 K. R^2, q_e, and k_2 are listed in Table 1.

For the porous adsorbents, the adsorbates transported to the surface of solid phase are controlled through intra-particle diffusion/transport process; therefore, it is necessary to investigate the intra-particle diffusion. The curves of intra-particle diffusion were given in Fig. 7. The plots q_t vs. $t^{0.5}$ did not pass through the origin and the curves demonstrated three different intercepting lines. This means that intra-particle diffusion was not the unique rate-limiting step. The curves elucidated multilinearity, which indicated three rate steps occurred. The first stage is determined by the diffusion of adsorbate transporting to the external surface of solid phase through the solution, which is a rapid adsorption process within the first 30 min; the second stage is a gradual adsorption stage, in which intra-particle diffusion was rate limiting; the third stage is the final equilibrium attainment[37]. The correlation coefficient R^2 and k_p obtained from the slopes are given in Table 2.

3.4. Thermodynamics

The second-order kinetics equation gives the defined rate constant k_2, so the activation energy E_a can be calculated from the following Arrhenius type relationship with k_2 as (5) [38], while the other thermodynamic parameters, standard free energy (ΔG^o), enthalpy (ΔH^o), and entropy (ΔS^o) can be obtained from Eqs. (6)-(8)[29].

$$\ln k_2 = \ln A - \frac{E_a}{RT} \tag{5}$$

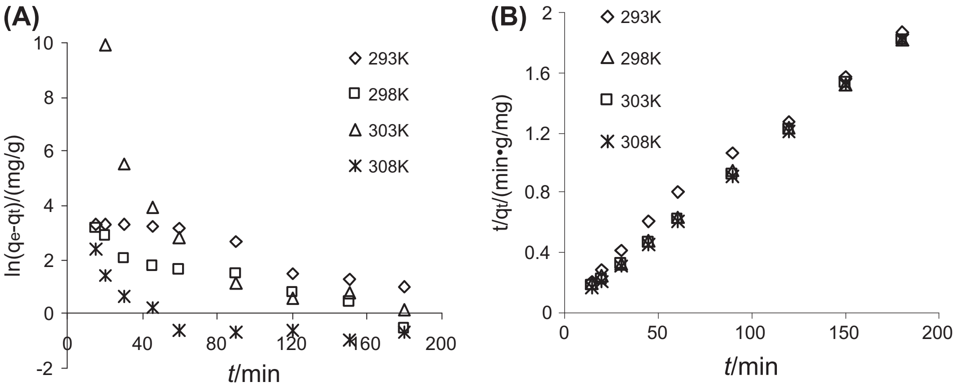

Fig. 6. First-order kinetic equation (A) and second-order kinetic equation (B) for adsorption of WAR-2R onto magnesium silicate gel.

Table 1
First-order kinetics and second-order kinetics values calculated for WAR-2R adsorption onto magnesium silicate gel

T	First-order		Second-order			
$/K$	k_1/min^{-1}	R^2	$k_2/\text{g mg}^{-1}\,\text{min}^{-1}$	$q_{e,\text{exp}}/\text{mg g}^{-1}$	$q_{e,\text{cal}}/\text{mg g}^{-1}$	R^2
293	0.0159	0.937	7.4×10^{-4}	96.22	102	0.992
298	0.0167	0.874	24×10^{-4}	99.9	101	0.999
303	0.0951	0.557	36×10^{-4}	99.84	100	1.000
308	0.0227	0.549	110×10^{-4}	99.46	100	0.999

$$K_C = C_A/C_S \tag{6}$$

$$\Delta G^\circ = -RT \ln K_C \tag{7}$$

$$\ln K_C = \frac{\Delta S^\circ}{R} - \frac{\Delta H^\circ}{RT} \tag{8}$$

where A is the Arrhenius factor; R is the gas constant; K_C is the equilibrium constant; C_A is the amount of dye adsorbed on the adsorbent of the solution at equilibrium, mg L^{-1}; and C_S is the equilibrium concentration of the dye in solution, mg L^{-1}. The slope from the linear plot of lnk_2 vs. $1/T$ for the removal of WAR-2R onto magnesium silicate gave the activation energy E_a and the value was 127.6 kJ mol^{-1}. The slope and intercept of the plot of lnK_C vs. $1/T$ gave enthalpy ΔH^o as 80.1 kJ·mol^{-1} and entropy ΔS^o as 300.1 J·mol^{-1}. The standard free energy ΔG^o calculated were −7.21, −10.1, −10.9, and −12.0 1 kJ mol^{-1}at different solution temperature (293, 298, 303, and 308 K).

ΔH^o was calculated as 8.01 kJ mol^{-1}, which indicate the interactions between adsorbent surface and adsorbates are a physical interaction and the adsorption is an endothermic process, and the adsorption capacity would increase with increasing solution temperature [39]. As known, the range of the change of standard free energy for a physisorption process is −20 to 0 kJ mol^{-1}, while for the chemisorption process the scale is −80 to −400 kJ mol^{-1} [40]. The results obtained in this study proved that the adsorption reaction was physisorption in nature.

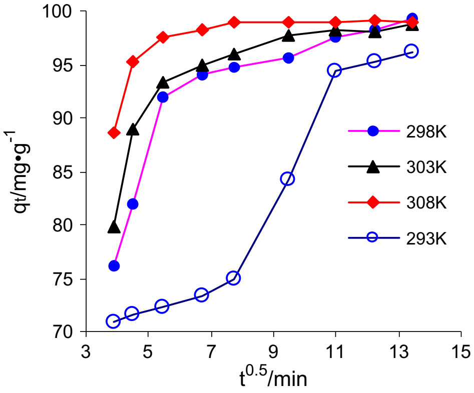

Fig. 7. Intra-particle diffusion plots for adsorption of WAR-2R on magnesium silicate gel.

Table 2
Intra-particle diffusion values calculated for WAR-2R adsorption onto magnesium silicate gel

T	Intra-particle diffusion (k_p)					
/K	k_{p-1}/$g\,mg^{-0.5}\,min^{-1}$	R^2	k_{p-2}/$g\,mg^{-0.5}\,min^{-1}$	R^2	k_{p-3}/$g\,mg^{-0.5}\,min^{-1}$	R^2
293	0.976	0.983	6.08	0.995	0.738	0.994
298	9.95	0.999	0.851	0.902	0.693	0.982
303	8.02	0.885	1.12	0.990	0.211	0.727
308	5.17	0.836	0.469	0.960	0.0243	0.108

4. Conclusion

We prepared a new mesoporous solid adsorbent, magnesium silicate gel, and the surface area was calculated as $277.37\,m^2\,g^{-1}$ by N_2 adsorption/desorption isotherms, the single point adsorption total pore volume value as $0.243\,cm^3\,g^{-1}$, N_2 adsorbed amount as $250\,cm^3\,g^{-1}$ and the narrow pore-size distribution centered at 2–4 nm. The adsorption effect was investigated and the results proved that the adsorption equilibrium capacity of WAR-2R onto magnesium silicate gel decreased from 240 to $50\,mg\,g^{-1}$ with mass of magnesium silicate gel increasing from 0.025 to 0.2 g and that increased from 45 to $145\,mg\,g^{-1}$ with dye concentration increasing from 100 to $300\,mg\,L^{-1}$. The pH effects indicated a slightly constant adsorption capacity under the acidic and basic solution (pH 3–10). The experimental data were followed by the second-order kinetics models. The rate constant k_2 increased with the increase in the adsorption temperature and q_e calculated values agreed with the experimental q_e values. The intra-particle diffusion curves showed three stage with first instantaneous adsorption process, second gradual adsorption process, and third equilibrium process, which indicate the intra-particle diffusion is not the unique rate-limiting step. The kinetics and thermodynamics parameters elucidate the adsorption process which is a rapid physisorption in nature. Therefore, the results would be favorable for practical application in wastewater treatment for the removal of dye.

Abbreviations

WAD-2R — weak Acid Red 2R (Acid Red 151), C. I. 26900
NBS — normal buffer solution
q_t — adsorption capacity at time t, $mg\,g^{-1}$
C_0 — initial dye concentration in aqueous solution, $mg\,L^{-1}$
C_t — dye concentration of the aqueous solution at time t, $mg\,L^{-1}$
V — volume of the dye solution, L
m — mass of adsorbent, g
q_e — adsorption capacity at equilibrium, $mg\,g^{-1}$
k_1 — first-order rate constant of adsorption, min^{-1}
k_2 — second-order rate constant of adsorption, $g\,mg^{-1}\,min^{-1}$
k_p — intra-particle diffusion rate constant of adsorption, $g\,mg^{-0.5}\,min^{-1}$
R^2 — correlation coefficient
T — temperature, K
A — arrhenius factor
R — gas constant, $8.314\,J\,mol^{-1}\,K^{-1}$
K_C — equilibrium constant
C_A — amount of dye adsorbed on the adsorbent of the solution at equilibrium, $mg\,L^{-1}$
C_S — equilibrium concentration of the dye in solution, $mg\,L^{-1}$

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